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## **Gasoline ether oxygenate occurrence in Europe, and a review of their fate and transport characteristics in the environment**

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# **Gasoline ether oxygenate occurrence in Europe, and a review of their fate and transport characteristics in the environment**

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## ABSTRACT

Ether oxygenates are added to certain gasoline (petrol) formulations to improve combustion efficiency and to increase the octane rating. In this report the term gasoline ether oxygenates (GEO) refers collectively to methyl tertiary butyl ether (MTBE), ethyl tertiary butyl ether (ETBE), tertiary amyl methyl ether (TAME), di-isopropyl ether (DIPE), tertiary amyl ethyl ether (TAE), tertiary hexyl methyl ether (THxME), and tertiary hexyl ethyl ether (THxEE), as well as the associated tertiary butyl alcohol (TBA).

This report presents newly collated data on the production capacities and use of MTBE, ETBE, TAME, DIPE and TBA in 30 countries (27 EU countries and Croatia, Norway and Switzerland) to inform continued and effective environmental management practices for GEO by CONCAWE members. The report comprises data on gasoline use in Europe that were provided by CONCAWE and obtained from the European Commission. Furthermore Société Générale de Surveillance (SGS) provided detailed analytical data (more than 1,200 sampling campaigns) on the GEO composition of gasoline in European countries in the period 2000-2010.

Another major aspect of this report is the investigation of GEO distribution in groundwater, drinking water, surface water, runoff water, precipitation (rain/snow) and air in the European environment. Apart from the general sources of literature for the study, local environmental authorities and institutes in the 30 European countries have been contacted for additional information.

Finally, a review of the international literature on GEO natural attenuation processes was undertaken with a focus on international reports and peer-reviewed scientific publications to give an overview on the known fate, transport and degradation mechanisms of GEO in the subsurface, to inform risk-management strategies that may rely on natural attenuation processes. The literature reveals that all GEO compounds used in fuels are highly water soluble and weakly retarded by aquifer materials, but are biodegradable under favourable environmental conditions, and volatilise from liquid (gasoline or water) to vapour phase. Consequently natural attenuation processes are expected to decrease GEO concentrations in the environment and, with appropriate site-specific evidence, may be incorporated into risk assessment / management strategies at GEO release sites.

## KEYWORDS

Gasoline ether oxygenates (GEO), methyl tertiary butyl ether (MTBE), ethyl tertiary butyl ether (ETBE), tertiary amyl methyl ether (TAME), di-isopropyl ether (DIPE), tertiary amyl ethyl ether (TAE), tertiary hexyl methyl ether (THxME), and tertiary hexyl ethyl ether (THxEE), tertiary butyl alcohol (TBA), GEO use, GEO occurrence in Europe, GEO transport characteristics in the subsurface, biodegradation.

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## SUMMARY

Ether oxygenates are added to certain gasoline (petrol) formulations to improve combustion efficiency and to increase the octane rating. In this report, the term gasoline ether oxygenates (GEO) includes methyl tertiary butyl ether (MTBE), ethyl tertiary butyl ether (ETBE), tertiary amyl methyl ether (TAME), di-isopropyl ether (DIPE), tertiary amyl ethyl ether (TAE), tertiary hexyl methyl ether (THxME), and tertiary hexyl ethyl ether (THxEE), as well as the associated tertiary butyl alcohol (TBA).

This technical report gives an overview of European GEO production capacity, the occurrence of GEO in the European environment and a review of the fate and transport characteristics of GEO in the subsurface.

Data was collected on the production and use of MTBE, ETBE, TAME, DIPE and TBA in 30 countries (27 EU countries and Croatia, Norway and Switzerland) to inform the debate on the environmental management issues relating to GEO. Furthermore, data on gasoline production and use in Europe were provided by CONCAWE and obtained from the European Commission. Data on GEO composition of 6 different gasoline types in EU 27, Norway, Switzerland and Turkey in the period 2000 to 2009 was obtained from Société Générale de Surveillance (SGS) and analysed (1,239 sampling events for MTBE, ETBE and TBA and 650 sampling events for TAME and DIPE). The report shows how the use of GEO has changed from MTBE to ETBE in certain countries, and onwards towards a lesser use of GEO (presumably in favour of direct ethanol blending) in certain markets.

Another major aspect was the investigation of GEO distribution in groundwater, drinking water, surface water, runoff water, precipitation (rain/snow) and air. Apart from the general sources of literature for the study, local environmental authorities and institutes in the 30 countries have been contacted for additional information. As a result, information from 11 countries was obtained for different environmental compartments, while in 19 countries no specific data on GEO in the environment were available.

The research found that, with the exception of localised point-source spill and release events, GEO are either not present in the European environment, or are not detected at concentrations that could give rise to either taste and odour concerns, or to ecological or human health impacts. GEO can be detected in some natural waters and in the European air using sophisticated analytical techniques, but generally at concentrations at least 1 order of magnitude lower than the relevant taste and odour thresholds, and 5 to 8 orders of magnitude lower concentration than the relevant health criteria. Most water samples in regional water quality surveys confirm the absence of GEO.

When GEO are accidentally released into the subsurface, they have the potential to cause impact due to their high aqueous solubility and low taste and odour thresholds. Furthermore, GEO are weakly adsorbed to aquifer and soil minerals and are generally less easily biodegraded than other constituents in gasoline; GEO plumes in groundwater, for example, tend to be larger and more persistent than BTEX plumes. However, it is now generally accepted that, after an initial acclimation period, GEO will biodegrade in the subsurface environment, albeit often at a lower rate than the BTEX compounds.

A review of the international literature was undertaken with a focus on international reports and scientific publications to give an overview on the known fate, transport and degradation mechanisms of GEO in the subsurface, to inform risk-management strategies that may rely on natural attenuation processes. It can be concluded that at sites without nearby receptors, monitored natural attenuation (MNA) of GEO contamination is a potentially viable risk management strategy, either in isolation, or as part of a remediation treatment train approach.

## **1. INTRODUCTION**

### **1.1. PURPOSE AND OBJECTIVES OF DOCUMENT**

Ether oxygenates are added to certain gasoline (petrol) formulations to improve combustion efficiency and increase the octane rating. In this report, the term gasoline ether oxygenates (GEO) collectively refers to methyl tertiary butyl ether (MTBE), ethyl tertiary butyl ether (ETBE), tertiary amyl methyl ether (TAME), di-isopropyl ether (DIPE), tertiary amyl ethyl ether (TAEE), tertiary hexyl methyl ether (THxME), and tertiary hexyl ethyl ether (THxEE), as well as the associated tertiary butyl alcohol (TBA).

While the replacement of lead-compounds with GEO has provided demonstrable air quality and public health benefits (e.g. Wei-Te et al 2010), it has also generated concerns regarding risks to the water environment, as GEO are generally more soluble and less biodegradable than petroleum hydrocarbon compounds. GEO also have low taste and odour thresholds which can make water unpalatable at concentrations orders of magnitude lower than the relevant health criteria.

This report presents newly collected data on the production and use of MTBE, ETBE, TAME, DIPE and TBA in 30 countries in Europe (27 EU countries and Croatia, Norway and Switzerland), to inform the debate on the environmental management issues relating to GEO. The report comprises data on gasoline use in Europe that were provided by CONCAWE and obtained from the European Commission. Société Générale de Surveillance (SGS) provided detailed analytical data (more than 1,200 sampling campaigns) on the GEO composition of gasoline in European countries in the period 2000-2010.

Data are reported on the GEO-distribution in groundwater, drinking water, surface water, runoff water, precipitation (rain/snow) and air. Apart from the general resources of literature for the study, local environmental authorities and institutes in 30 countries in Europe (27 EU countries and Croatia, Norway and Switzerland) have been contacted for additional information

Finally, a review of the international literature was undertaken with a focus on international reports and scientific publications to give an overview on the known fate, transport and degradation mechanisms of the GEO, to inform risk-management strategies that may rely on natural attenuation processes.

### **1.2. GENERAL INTRODUCTION ON THE HISTORICAL PRODUCTION AND USE OF GASOLINE ETHERS IN EUROPE**

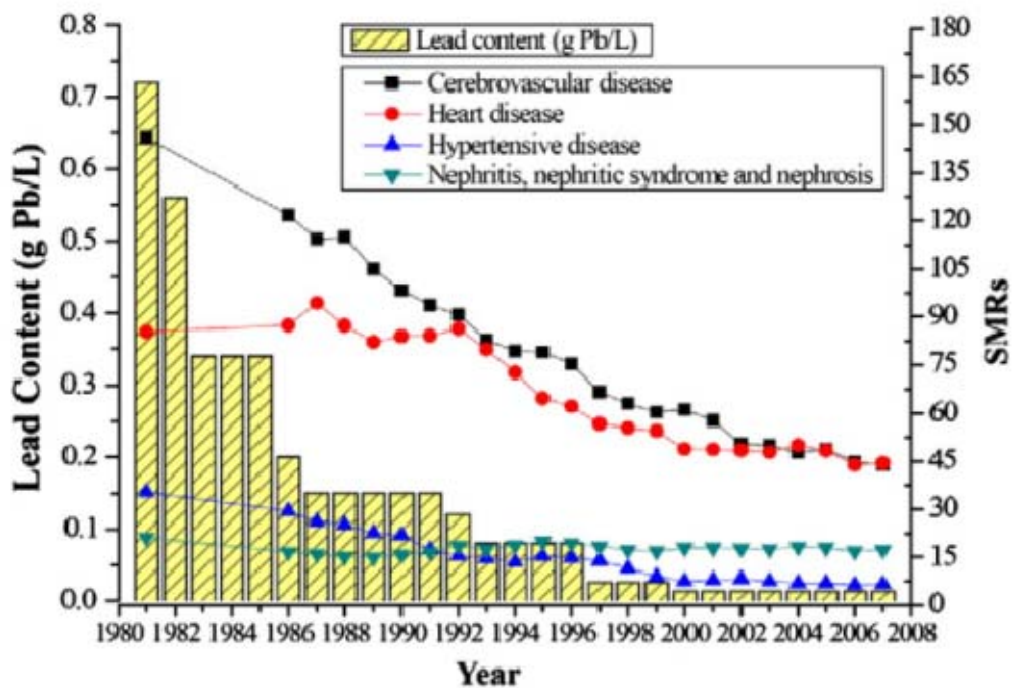
GEO were developed in the 1970s as octane enhancers to improve combustion efficiency, reduce vehicle exhaust emissions and improve air quality, and as a replacement for anti-knock additives like tetra-ethyl lead (TEL). Alkyl lead compounds were used in petrol as an octane booster since the 1920s and with the worldwide increase of petrol consumption, exhaust emissions from (leaded) petrol engine vehicles caused the largest lead exposure in comparison with other sources at that time (Wei-Te et al, 2010).

During the 1970s, the health impacts associated with lead emissions from vehicles became an issue of widespread concern. Several studies demonstrated that lead

emissions derived from the use of leaded-petrol resulted in significant effects on the environment and especially on human health (effects on the central nervous system, haemoglobin-synthesis, reproductive system and psychological and neurobehavioral functions). As a result of these issues alkyl lead compounds were replaced during the 1980s and had been removed from most European gasoline by the 1990s.

With the use of GEO as an octane booster, the largest lead exposure was cut off within one decade. This resulted in a significant improvement of public health. The recent study by Wei-Te et al (2010) provides a statistical evaluation of the effect of the “petrol-lead phase-out program” in Taiwan on human health. A major result is that there is a dramatic decrease in the mean blood lead level, from approx. 20 µg/dl in the leaded petrol phase to 3 µg/dl or lower in the unleaded petrol phase. Another important effect is the reduction of the standardized mortality ratio for several diseases, as shown in **Figure 1.1**. Thus, GEO were a key factor for the significant improvements in human health, due to the replacement of alkyl lead compounds in petrol.

**Figure 1.1** Trend of standardized mortality ratio of several diseases correlated with the reduction of lead compounds within petrol in Taiwan. (after Wei-Te et al, 2010)



Replacement of TEL was also necessary for modern engines with three-way catalytic converters due to the fact that lead causes a coating on the catalyst's surface, effectively disabling it. Many brands of gasoline sold today in Europe and around the world have some concentration of oxygenates to enhance octane rating and reduce atmospheric emissions. In the long term it is expected that gasoline consumption in the EU will stabilise or decrease.

Oxygenates are produced from a variety of feedstocks. Methanol, derived primarily from natural gas, is one feedstock used in the production of MTBE. Ethanol is derived primarily by fermenting maize (corn) and other agricultural biomass and is used directly as an additive or as a feedstock for the production of ETBE. Isobutylene is the other common feedstock used in both MTBE and ETBE production, and is also derived from natural gas or as a by-product of petroleum refining. Most of the production facilities that produce oxygenates can move between ETBE and MTBE production without significant difficulty.

In Europe, production of MTBE started in Italy in 1973. The MTBE market has increased strongly since the 1990s. More than 98% of the MTBE produced is used for gasoline blending (EU, 2002). MTBE has been added to gasoline blends since the second half of the 1970s, initially at relatively low concentration (2 – 5 w/w%) to boost the octane rating of unleaded premium or high performance grades. In later periods, MTBE has been added at higher concentrations (11 – 15%-vol) in North America and parts of Europe to promote more efficient combustion of the gasoline with benefits for air quality. ETBE was first used in France in 1992; today it is widely used and manufactured in most major gasoline markets in the EU.

The maximum permissible concentration of ether oxygenates in gasoline used in the EU, expressed in Directive 98/70/EC as “Ethers containing 5 or more carbon atoms per molecule”, is 15% by volume. The EU Biofuel Directive sets requirements for the biofuel contribution in gasoline. The required biofuel concentration of gasoline was 2% in 2005, rising to 5.75% in 2010, and may even rise to 8% by 2020. This is likely to further increase the market share of ETBE relative to MTBE, as ETBE made from bio-ethanol is considered a biofuel.

### 1.3. PHYSICAL-CHEMICAL PROPERTIES OF GEO

The GEO described in this report belong to a group of different ethers and alcohols. They are listed in **Table 1.1**.

**Table 1.1 Gasoline ether oxygenate (GEO)**

| <b>Methyl ethers</b>            | <b>Ethyl ethers</b>  |
|---------------------------------|--|
| MTBE (methyl tert butyl ether)  | ETBE (ethyl tert butyl ether)                                      |
| TAME (tert amyl methyl ether)   | TAEЕ (tert amyl ethyl ether)                                       |
| THxME (tert-hexyl methyl ether) | THxEE (tert-hexyl ethyl ether)                                     |
|                                 |  |
| <b>Propyl ethers</b>            | <b>Alcohol oxygenates</b>  |
| DIPE (diisopropyl ether)        | TBA (tert butyl alcohol)   |
|                                 | methanol, ethanol, butanol (not considered further in this report) |

**Table 1.2** summarizes the physical-chemical properties of GEO, focussing on properties that influence their distribution, transport and fate in the environment:

**Table 1.2** Physical-chemical properties of GEO compounds<sup>1</sup>

|   | MTBE                         | TBA                          | ETBE                         | TAME                        | DIPE                         | TAAE                      | THxME <sup>4</sup>        | THxEE                      | Benzene                  |
|---|------------------------------|------------------------------|------------------------------|-----------------------------|------------------------------|---------------------------|---------------------------|----------------------------|--------------------------|
| CAS No.                                     | 1634-04-4                    | 75-65-0                      | 627-92-3                     | 994-05-8                    | 108-20-3                     | 919-94-8                  | -                         | -                          | 71-43-2                  |
| Molecular weight (g/mol)                    | 88.15 <sup>1</sup>           | 74.12 <sup>1</sup>           | 102.18 <sup>1</sup>          | 102.18 <sup>1</sup>         | 102.18 <sup>1</sup>          | 116.23 <sup>3</sup>       | 116.2 <sup>3</sup>        | 130.2 <sup>3</sup>         | 78.11 <sup>1</sup>       |
| Melting point (°C)                          | -109 <sup>1</sup>            | 25.5 <sup>1</sup>            | -94 <sup>1</sup>             | -                           | -85.5 <sup>1</sup>           | -68.41 <sup>3</sup>       | -80.2 <sup>3</sup>        | -67.5 <sup>3</sup>         | 5.5 <sup>1</sup>         |
| Boiling point (°C)                          | 55.2 <sup>1</sup>            | 82.2 <sup>1</sup>            | 71 <sup>1</sup>              | 85-86 <sup>1</sup>          | 68.5 <sup>1</sup>            | 102 <sup>3</sup>          | 92.6 <sup>3</sup>         | 117 <sup>3</sup>           | 80.1 <sup>1</sup>        |
| Density (kg/l) @ 25°C                       | 0.741 <sup>1</sup>           | 0.786 <sup>1</sup>           | 0.752 <sup>1</sup>           | 0.764 <sup>1</sup>          | 0.724 <sup>1</sup>           | 0.772 <sup>3</sup>        | 0.777 <sup>3</sup>        | 0.778 <sup>3</sup>         | 0.879 <sup>1</sup>       |
| Vapour pressure (Pa) (variable Temperature) | 17600 – 60500 <sup>2</sup>   | 5413 – 5852 <sup>2</sup>     | 11965 – 16500 <sup>2</sup>   | 10112 – 13263 <sup>2</sup>  | 17290 – 19830 <sup>2</sup>   | 5240 <sup>3</sup> 25°C    | 7740 <sup>3</sup> 25°C    | 2819 <sup>3</sup> 25°C     | 12692 <sup>5</sup> 25°C  |
| Water solubility (g/l)                      | 43 – 51 @ 25°C <sup>2</sup>  | 1E+3 @ 25°C <sup>2</sup>     | 12 @ 20°C <sup>2</sup>       | 20 @ 25°C <sup>2</sup>      | 9 @ 20°C <sup>2</sup>        | 4.5 <sup>3</sup> No Temp. | 7.5 <sup>3</sup> No Temp. | 2.45 <sup>3</sup> No Temp. | 1.8 <sup>1</sup>         |
| Henry's law constant @ 25°C (-)             | 0.0216 – 0.0555 <sup>2</sup> | 0.0004 – 0.0006 <sup>2</sup> | 0.0564 – 0.0972 <sup>2</sup> | 0.054 – 0.1096 <sup>2</sup> | 0.0774 – 0.1050 <sup>2</sup> | 0.778 <sup>3</sup>        | 0.141 <sup>3</sup>        | 0.189 <sup>3</sup>         | 0.221 <sup>5</sup>       |
| log KOW (-)                                 | 0.94 – 1.6 <sup>2</sup>      | 0.35 – 0.73 <sup>2</sup>     | 1.74 – 1.92 <sup>2</sup>     | 1.55 – 1.95 <sup>2</sup>    | 1.52 – 1.88 <sup>2</sup>     | 2.41 <sup>3</sup>         | 2.34 <sup>3</sup>         | 2.83 <sup>3</sup>          | 2.13 <sup>5</sup>        |
| log KOC (-)                                 | 0.02 – 1.1 <sup>2</sup>      | 0.41 <sup>2</sup>            | 0.94 – 1.57 <sup>2</sup>     | 1.62 <sup>2</sup>           | 1.02 <sup>2</sup>            | 1.3 <sup>3</sup>          | 1.2 <sup>3</sup>          | 1.5 <sup>3</sup>           | 1.56 – 2.15 <sup>5</sup> |

1 = <http://chemfinder.cambridgesoft.com>  
 2 = Grathwohl et al 2005  
 3 = ChemSpider  
 4 = Synonym MTHxE in: Snelling et al 2007  
 5 = Interagency Assessment of Oxygenate Fuels (In ITRC 2005)

<sup>1</sup> The properties of Benzene are included as a reference gasoline component.

In addition, the physical-chemical properties for methyl-tert-octyl ether are documented in Snelling et al 2007 (molecular weight: 144 g/mol, solubility in water at 25°C: 900 mg/l; log K<sub>oc</sub> at 25°C: 2.5).

Initially, all the GEO mentioned in the introduction were additives in unleaded gasoline. However, today these oxygenates are substantial components and although, incorrectly, also ethanol is often included in these, as in may be added up to 2.7 % by oxygen weight. The other alcohol that is often mistakenly included is TBA that is never added deliberately, but may also occur in both gasoline and in the environment concurrently, due to other reasons:

- TBA is a widely used solvent and intermediate in industrial processes and is also used in the manufacturing of MTBE (Eweis et al, 2007)
- TBA is an impurity in commercial MTBE (Schmidt et al, 2004; USEPA, 2005)
- TBA has been documented as an intermediate or transformation product of MTBE and ETBE biodegradation (Schmidt et al, 2004)
- TBA may be detected in groundwater samples as a result of hydrolysis of MTBE to TBA during sample preservation or analysis (O'Reilly et al, 2001; Lin et al, 2003)
- TBA may also be produced by the degradation of isobutane or isopentane (Hyman et al, 2007)

Consequently, TBA is included in this report due to its close association with MTBE and ETBE.

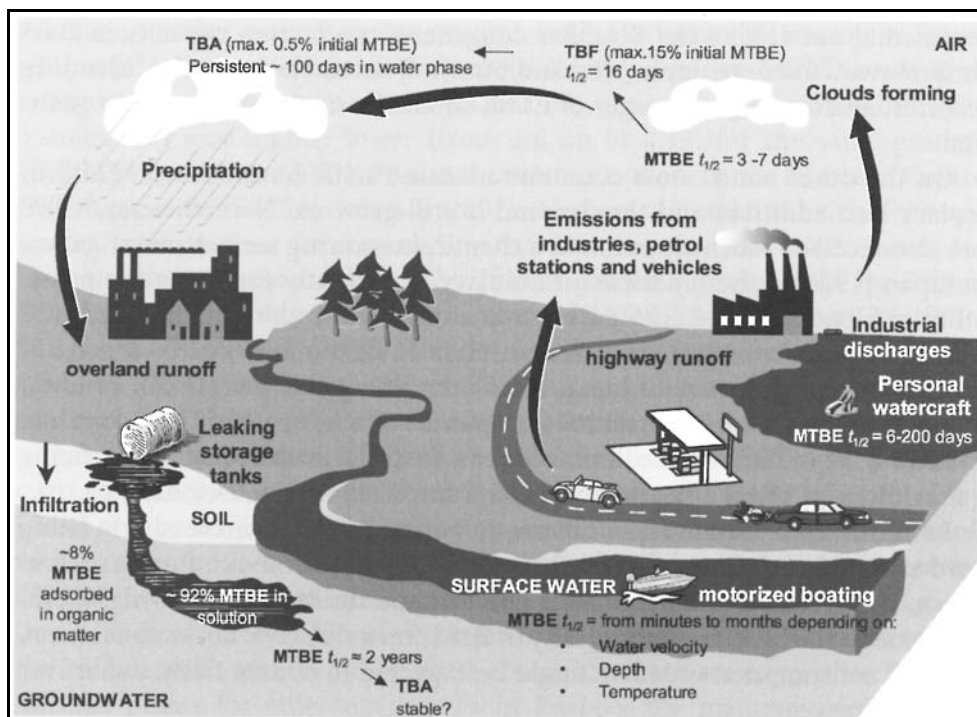
#### 1.4. FATE OF GEO IN THE ENVIRONMENT

This section provides a general overview of the fate of GEO in the environment and their major pathways for release and migration. A detailed description of the fate and transport of GEO in the subsurface in relation to their specific properties is given in Chapter 4.

Rosell et al (2007a) provide an illustrative conceptual model of the fate of MTBE in the environment and the major pathways between the different compartments (as shown in **Figure 1.2**). This conceptual model can also be adopted to illustrate the emission and transport pathways for the other GEO.

Due to the physical and chemical properties of GEO (see above section 1.3), these substances partition easily from gasoline into water and vapour phases, resulting in the potential for a wide distribution in the environment. The water pathway has a key role, due to slower biodegradation and limited retardation processes, and the general high mobility of GEO in water, compared with the air path.

**Figure 1.2** Illustrative conceptual model of the behaviour and fate of MTBE in the environment (Rosell et al, 2007a)



Air

GEO in the atmosphere originate mainly from industrial emissions, fugitive emissions from retail filling stations and the exhaust emissions from gasoline powered vehicles due to incomplete combustion or reduced efficiency of the catalytic converter during the cold run period of vehicles.

GEO are degraded by photo-induced hydroxyl radicals, with a relatively short half-life in the atmosphere, acting as an important sink for GEO. Rosell et al (2007) and Achten et al (2002a) reported a half-life for MTBE of 3 to 7 days in the air, mainly related to the concentration of hydroxyl radicals. Photolysis or the presence of other radicals is less relevant for the degradation of GEO in the air.

Precipitation

With regard to the presence of GEO in the air, these substances are also present in rain and snow. Studies by Achten et al (2001) and Kolb & Püttmann (2006a) report a strong relationship between the distribution of GEO and land use (rural areas vs. urban/industrial areas). In addition, seasonal effects must be considered (e.g. winter increase in urban areas).

With respect to the GEO-load in runoff water, especially in urban areas, this originates predominantly from direct vehicle emission uptake during a precipitation event (approx. 80%). Atmospheric transport mechanisms contribute only a minor portion (approx. 20%) to the runoff load in urban areas (Achten et al, 2001 and Rosell et al, 2007a).



### Surface water

Precipitation and road-runoff, especially from urban areas, lead to a background GEO emission to rivers and lakes. An additional GEO source is the emission from waste water treatment plants – also linked to drained precipitation and partly waste water from sites with GEO usage. Similar to the distribution effects observed for precipitation, higher GEO-loads are detected in urban rather than rural areas (Achten et al, 2002b).

Higher concentrations of GEO in rivers and lakes usually originate from spills, industrial discharges or illegal dumping of tank washings from tanker ships (Rosell et al, 2007a). An additional source of GEO, especially in lakes and recreational areas, is watercraft that can generate a significant and seasonal contribution (Rosell et al, 2007a). This source is particularly associated with the release of un-burnt GEO from two-stroke engines (e.g. outboard motors).

The major elimination mechanism of GEO from surface water is volatilisation over the air-water interface, which depends strongly on the water surface area, water depth, temperature and wind speed. Pankow et al (1996) calculated the potential of GEO for volatilisation from surface water for different alkyl ethers and TBA. They concluded that the volatilisation for ethers is comparable to that of BTEX (partly a bit slower for shallow fast flowing waters, but still fast enough to act as an effective mechanism for mass reduction of GEO). A much lower loss from surface water was observed for TBA.

The findings of Pankow et al (1996) were supported by Arp et al (2004a, b). They report DIPE as the easiest component among the GEO to volatilise from open water at environmental conditions, followed by EBTE and MTBE. They reported lower transport kinetics for TBA.

Rosell et al (2007a) estimated a half-life for MTBE in rivers ranging from 30 minutes to 52 days and for lakes from 10 to 193 days.

### Groundwater

The main mechanism for GEO release into groundwater results from point sources. These include leaking storage tanks, accidental spillages during production, storage, transportation and use of gasoline products in retail filling stations, depots and refineries (Rosell et al, 2007a). Once the GEO reach the unsaturated zone and aquifer, a separation from the other gasoline compounds generally occurs – resulting in preferential migration of GEO relative to the aromatic gasoline compounds in most cases. Depending on local conditions, the GEO-plume may even break away from the plume of gasoline compounds (Stupp et al, 2008).

Infiltration of GEO-containing precipitation and volatilisation from the urban atmosphere can act as a diffuse (non-point) source for GEO, resulting in low but measurable background concentrations over large areas. A special feature in this respect can be the infiltration of road runoff and highway runoff, which is partly drained to infiltration systems, such as swales, infiltration trenches and seepage reservoirs. Generally, there is a strong connection between the presence of GEO in groundwater and land use, population density and amount of GEO used in gasoline (Achten et al, 2001). Due to the lower usage of TAME and DIPE in comparison with MTBE and ETBE, their detection in groundwater is significantly lower.

### 1.5. INTRODUCTION TO RISK ANALYSIS FOR GEO RELEASES

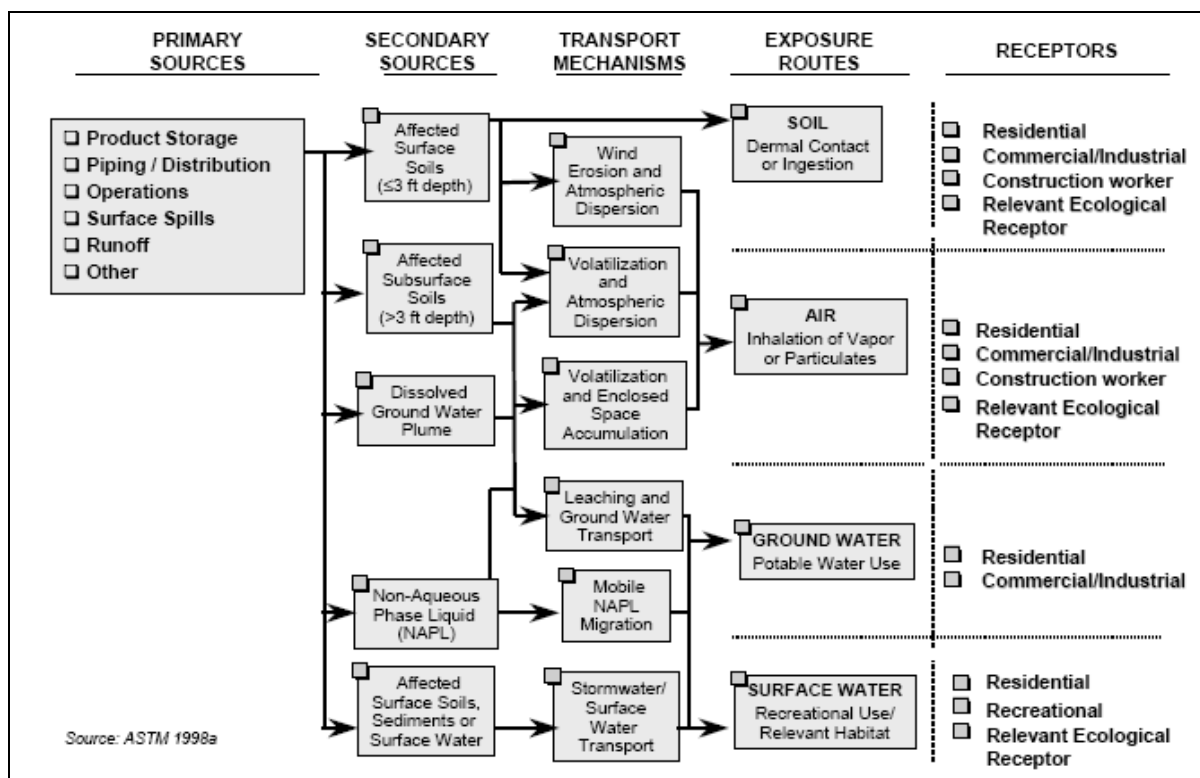
When GEO are identified in or unintentionally introduced into the environment there is a need to establish the potential risks associated with this. This risk analysis applies the well founded Source-Pathway-Receptor approach (e.g. API, 2001) that can establish the presence and severity of any impacts.

On the basis of (API, 2000) sources, pathways and receptors are defined as follows:

- **Sources** include the primary release of the chemical into the environment (e.g. surface spill, or UST system leak). Furthermore, sources also include the environmental compartment (e.g. soil, groundwater) adjacent to the original release (secondary source).
- **Pathways** include the processes by which a chemical may migrate from the source to a point of potential exposure (e.g. ground water migration, vapour migration). Pathways also include the route by which a receptor is exposed to the chemical (e.g. ingestion, inhalation and dermal contact).
- **Receptors** may include any organism (human or other ecological species) or habitat that may be adversely affected by exposure to the chemical.

It is common practice to summarize the actual and potential sources, pathways and receptors related to the chemical release in a conceptual site model (CSM). This can be done in a written summary, a table matrix, a flow chart or a schematic figure. An example is shown in **Figure 1.3**.

**Figure 1.3** Example of an exposure pathway flowchart (after ASTM 1998)



### 1.5.1. Sources of GEO

Contamination of soil and groundwater by GEO can originate from releases of either pure-phase GEO or as a component of gasoline. GEO will initially be transported through the subsurface as a Light Non-Aqueous Phase Liquid (LNAPL), from which both dissolved (aqueous) and vapour-phase impacts may occur. Widespread experience shows that the occurrence of GEO in soil and groundwater is predominantly connected with gasoline releases.

Three different release scenarios can be distinguished. These are GEO release from production plants, refuelling facilities (retail stations and fuel terminals and distribution sites) and other releases including transport accidents. Details of the scenarios are presented in **Table 1.3**.

**Table 1.3** Possible release scenarios of GEO and the resulting type of GEO source

| Release scenario   | Type of possible GEO sources |                       |
|--|------------------------------|-----------------------|
|  | Pure-phase                   | Component of gasoline |
| <b>GEO Production plants:</b>  |                              |                       |
| Leakage from underground storage tanks (LUST) and connecting pipe work | Yes                          | No                    |
| Surface spills and tank over-fills                                     | Yes                          | No                    |
| Losses from pipelines  | Yes                          | No                    |
| Losses from above ground storage tanks                                 | Yes                          | No                    |
| <b>Refuelling Facilities:</b>  |                              |                       |
| Leakage from underground storage tanks (LUST) and connecting pipe work | No                           | Yes                   |
| Surface spills and tank over-fills                                     | No                           | Yes                   |
| Losses from subsurface pipelines                                       | No                           | Yes                   |
| Losses from above ground storage tanks                                 | No                           | Yes                   |
| Losses from leaks in drainage systems and oil-water separators         | No                           | Yes                   |
| <b>Other sources:</b>  |                              |                       |
| Accidental spill during transport (road/rail/ship)                     | Yes                          | Yes                   |
| Release following road traffic accident (car fuel tank rupture)        | No                           | Yes                   |
| Releases to surface water from boats engines                           | No                           | Yes                   |

LUST = Leaking underground storage tanks

### 1.5.2. Pathways for GEO migration

Several GEO migration pathways are possible, including migration in air and soil gas, surface water, groundwater and along utility conduits.

Risk analysis for LNAPL will concentrate on the following pathways:

- Vapour pathway (the exposed medium is the atmosphere or soil air)
- Direct contact pathway (predominantly direct contact with LNAPL-impacted soil; LNAPL)
- LNAPL migration pathway (various exposed media are possible)
- Dissolved phase pathway (dissolved GEO in soil water or groundwater)

#### **Vapour pathway**

The exposed medium by the vapour pathway is either the atmosphere when GEO are spilled directly on sealed ground surfaces or vapour intrusion, defined as migration of volatile chemicals from the subsurface into overlying buildings (USEPA, 2002 OSWER draft guidance).

The potential for vapour intrusion by GEO is discussed in Chapter 4.3.5 (vapour pressure). About 94% of the MTBE spilled on open ground surfaces will partition into air under equilibrium conditions (EU, 2002). The fate of GEO in the atmosphere is dominated by degradation (see Chapter 4.3.7).

#### **Direct contact pathway**

The direct contact pathway describes the potential contact with LNAPL-impacted soil directly to skin or other body parts.

In respect to the relatively controlled sources of GEO, direct contact with GEO in soil or surface water is not likely. Controls, through the use of personal protection equipment (PPE) are standard practice for workers excavating at potentially GEO-contaminated sites, and serves to mitigate the risk of direct exposure to workers. Therefore, the direct contact pathway will not be considered in the following chapters.

#### **LNAPL migration pathway**

It is generally known that once an LNAPL release ceases, subsurface spreading of LNAPL slows and ultimately stops (API Soil and Groundwater Research Bulletin Number 18). For risk analysis, the mobility of LNAPL has to be assessed. LNAPL mobility will not be discussed in the following chapters for detailed information about the mobility of LNAPL see API (2002).

LNAPL in residual saturation (immobile) is an important secondary source of ground water contamination by GEO. This will be discussed in Chapter 4.3.6 (Partitioning from LNAPL).

#### **Dissolved phase pathway**

Dissolved phase groundwater contamination originates primarily from LNAPL in residual saturation. This will be discussed in Chapter 4.3.6 (Partitioning from LNAPL).

### 1.5.3. Management Context

The source – pathway – receptor approach is generally applied in environmental risk assessments. It leads to an assessment of all environmental compartments from the unsaturated zone, aquifer, hyporheic zone (i.e., groundwater – surface water interface) to surface water. Site assessments that do not include the hyporheic zone, if present, may lead to the selection of unnecessarily costly and less sustainable remedial strategies (Landmeyer et al, 2010).

## 1.6. REGULATORY STATUS OF GEO

In Europe, the regulations for GEO are not consistent and are fragmentary with regard to the different environmental compartments. EU legislation does not provide a general requirement. Thus, only a few threshold values and guidelines are in use, referring to water (drinking water and groundwater) and MTBE.

The main reason for the lack of regulatory thresholds and especially drinking water quality standards is related to the fact that the WHO decided not to set up a health-based guideline for MTBE. As indicated by the WHO (2005) and explained by Fawell (2007), the known odour and taste thresholds are 4 to 5 orders of magnitude lower than the concentration for effects on human health, derived from different toxicology studies. Only Denmark has a legally enforced drinking water quality standard for MTBE, with a threshold of 5 µg/l (BEK, 2007).

An evaluation of MTBE in the Netherlands (RIVM, 2004) led to a proposal for a drinking water standard of 1 µg/l, considering the precautionary principle. In parallel this value was compared with the risk limit derived for toxic effects on humans (9,420 µg/l), and odour and taste thresholds of 15 µg/l proposed by WHO (2005) and 20-40 µg/l proposed by the US EPA.

The Dutch Ministry of Housing, Spatial Planning and the Environment recently set a target value for remediation of 1 µg/l within drinking water collection areas for MTBE and ETBE, in accordance with the precautionary principle and the existing drinking water legislation. Outside drinking water collection areas, a value of 15 µg/l was set (VROM, 2008 / van Wezel, et al 2009).

In Germany, the water consortium of the federal states (Länderarbeitsgemeinschaft Wasser = LAWA (Altmayer et al, 2004)) defined a concentration of 15 µg/l that represents the level of insignificance for MTBE in groundwater. This value does not represent a legally binding groundwater quality standard – it is used as an assessment value for groundwater contamination. In addition, Switzerland has a guideline value for MTBE in groundwater of 2 µg/l (BUWAL, 2004b). This is a precaution value and it is used as a tracer for gasoline compounds in groundwater.

The Finnish Government adopted decree 214/2007 (SYKE, 2009), which includes provisions relating to soil contamination and remediation. It indicates a threshold value of 0.1 mg/kg for MTBE. Additionally, lower (5 mg/kg) and higher (50 mg/kg) critical limits are given.

In a study by the Energy Institute and Environment Agency (2009) and the Risk Assessment report for TAME by the EU (2006), it was assumed that the toxicological profiles of ETBE and TAME are quite similar to MTBE. Thus, these components are likely to be detected through odour and taste in concentrations that are below human health effect concentration by orders of magnitude.

The European Water Framework Directive (WFD; 2000/60/EU) sets out goals to ensure that surface water bodies attain good chemical and ecological status and that groundwater bodies achieve good chemical and quantitative status, as well as the prevention and progressive reduction of ground water pollution. Therefore, the interaction between surface water bodies and groundwater bodies is an important aspect which must be understood, especially when -groundwater containing GEO enters surface water. However, the subsequent Groundwater Daughter directive (2006/118/EC) and EQS directive (2008/105/EC) do not provide a value for any GEO.

## 2. USE OF GEO IN EUROPE

### 2.1. GEO MANUFACTURING LOCATIONS

- In 2010 about 55 facilities with GEO (MTBE, ETBE and TAME) production capacity were located in the EU.
- 50% of the total European production capacity is located in Germany, France and the Netherlands.
- There are no records of DIPE and TBA production for blending of gasoline in Europe.

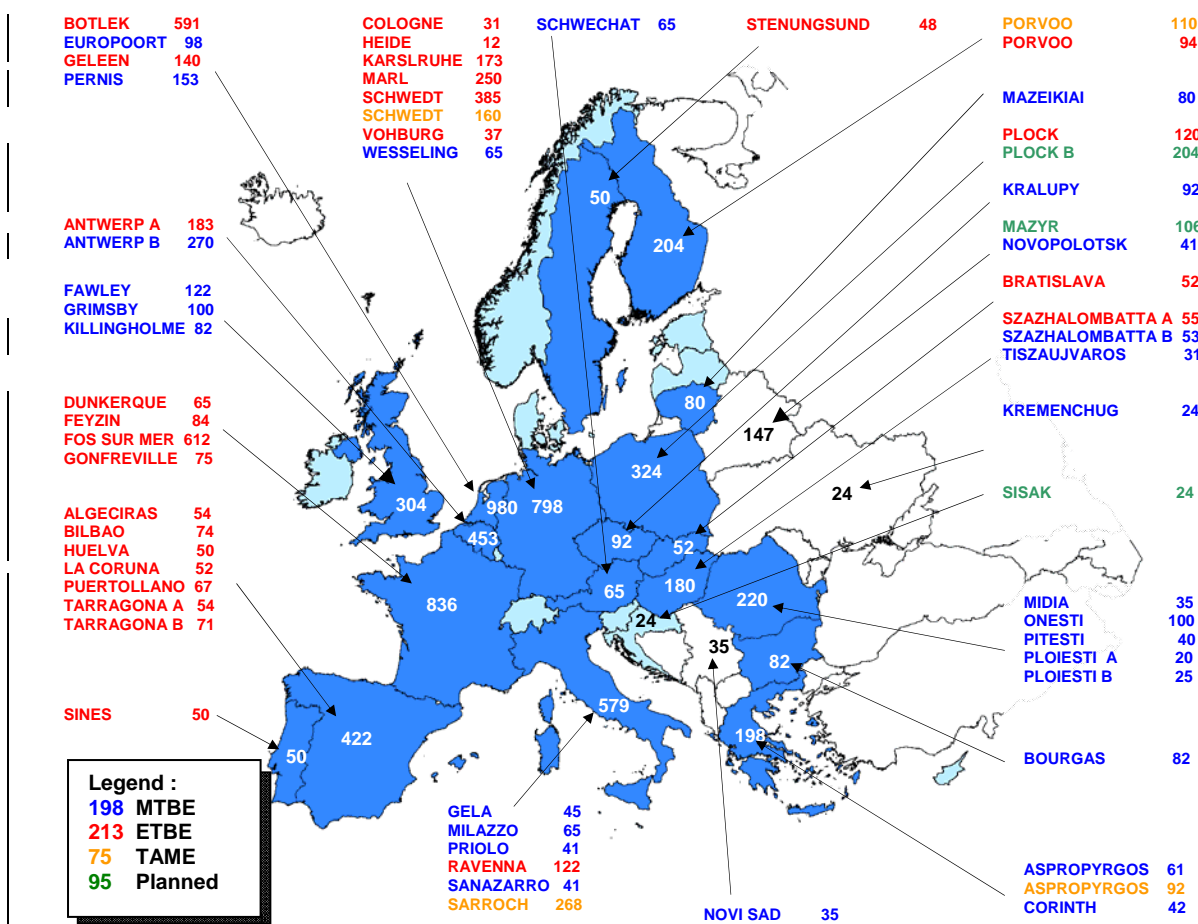
**Figure 2.1** presents the locations of MTBE, ETBE and TAME production sites in European countries in 2010 (EFOA 2010a) with the nameplate capacities. Actual production volumes may be different.

Ether oxygenates are produced by oil companies in refineries and several chemical production companies. The majority of the gasoline oxygenates are produced in the western EU countries with Germany, France and the Netherlands having approximately 50% of the total European production capacity. The two largest production facilities are situated in the Netherlands and France, each with capacities close to 600 ktonne per year. The median capacity of the 55 production facilities is about 75 ktonne per year.

MTBE production sites are distributed over many European countries. Historically the production of ETBE has been mainly located in Germany, France and Spain. Finland is the only producer and consumer of TAAE, the large scale production of TAME started in 1995 and the unit was converted into TAAE production in 2008. Germany, Italy and Greece produce TAME. Germany, Italy and Greece also produce TAAE.

There are no records of DIPE and TBA blending of gasoline in Europe. When produced by the dehydration of TBA, this can be present as an impurity within isobutylene, a base chemical for the production of MTBE and ETBE. TBA is also used as stabiliser for bioethanol.

**Figure 2.1** Countries with GEO production facilities (MTBE, ETBE and TAME) shown in dark blue in 2010. Nameplate capacities are in ktonne per year. Actual production volumes may be different. Countries currently without production facilities are shown in light blue. Countries which are not part of this study are shown in white. No data are available for the current production volume in Norway (EFOA, 2010a)



## 2.2. PRODUCTION CAPACITIES

- The total GEO production capacity (MTBE, ETBE and TAME) has grown from 4,108 ktonne in 2002 to 6,049 ktonne per year in 2010.
- The market share of ETBE has grown from 15% in 2002 to about 60% in 2010, while MTBE production capacity has decreased.
- TAME production capacity was constant from 2005 to 2010, with a 10% market share.
- Actual production volumes are not available.

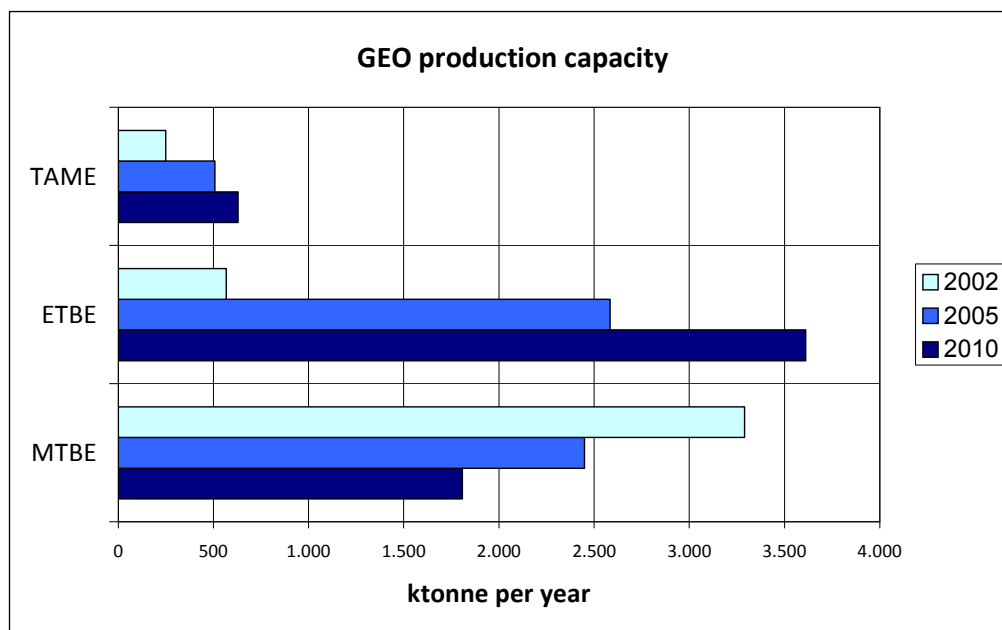
The actual production volumes of GEO in the EU are not communicated openly (commercially sensitive information), although the production capacities of GEO production facilities is publicly available. In general the actual amount of GEO



produced is typically 80 to 90% of the production capacities, based on available data (EU 2002). The production capacities of GEO in 2002, 2005 and 2010 are given in **Figure 2.2**. Country specific production capacities are presented in **Appendix 1**.

**Figure 2.2** presents the market share of MTBE, ETBE and TAME. The total GEO production capacity has grown from 4,108 ktonne in 2002 to 6,049 ktonne per year in 2010. In 2002 the market was dominated by MTBE production, with a production capacity of 3,290 ktonne/yr. In 2002 the market share of ETBE was 15%, when 568 ktonne ETBE were produced in Spain and France. The market share of ETBE has since grown to 47% in 2005 and 60% in 2010, as bio-ethanol based ETBE is accounted as a biofuel. Existing MTBE facilities in Germany and the Netherlands have been converted to ETBE production and a new ETBE facility in Schwedt (Germany) has started production. The major production facilities in Germany, France and the Netherlands now produce ETBE, and many other MTBE plants are evaluating a switch to ETBE.

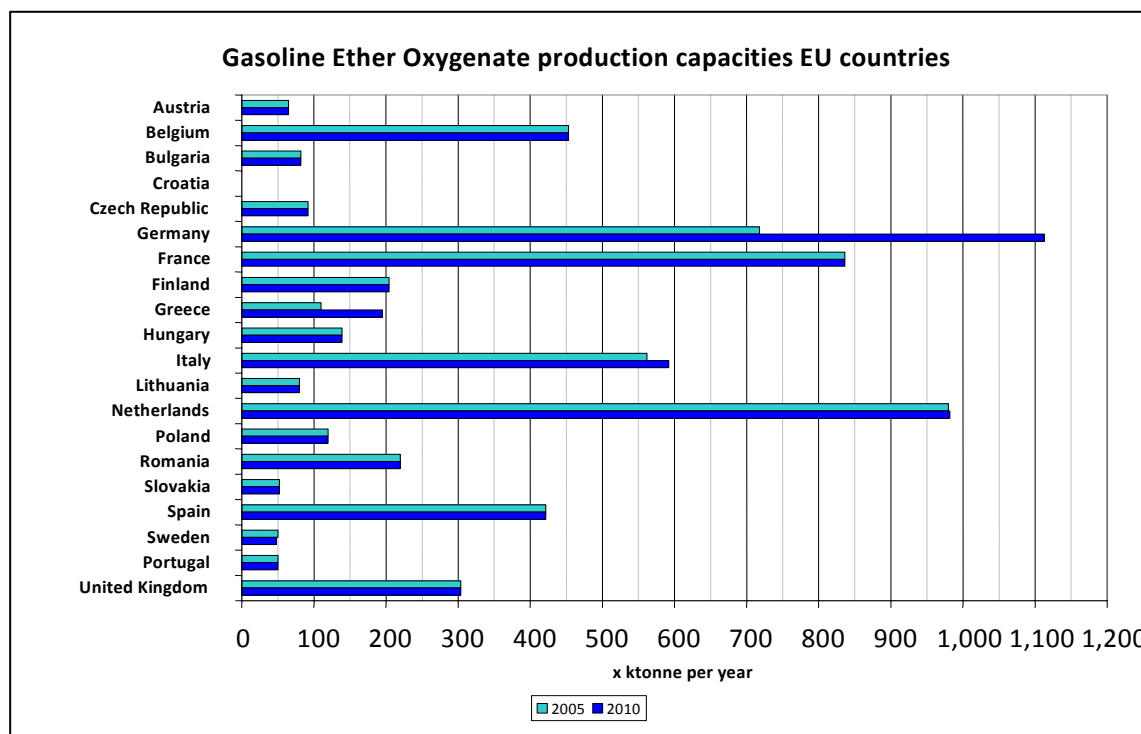
**Figure 2.2** European production capacities of MTBE, ETBE and TAME in 2002, 2005 and 2010 (EFOA, 2010a; EFOA, 2010b; EU, 2002; EU, 2006; Balat, 2007)



TAME and TAEЕ are produced in Germany, Finland, Italy and Greece, with a relatively constant market share of about 10%. Production capacity has increased from 507 kt/yr in 2005 to 630 ktonne/yr in 2010 due to the extension of the existing production in Italy and a new production facility in Greece.

**Figure 2.3** presents the GEO production capacity in individual European countries in 2005 and 2010. Compared with 2005, the production capacity in Germany, Greece and Italy has increased.

**Figure 2.3** GEO production capacities in 2005 and 2010 (EFOA, 2010a)



The Netherlands is one of the few countries that produces far more GEO than it consumes. Therefore, most of the GEO produced in the Netherlands is exported to other countries within the EU.

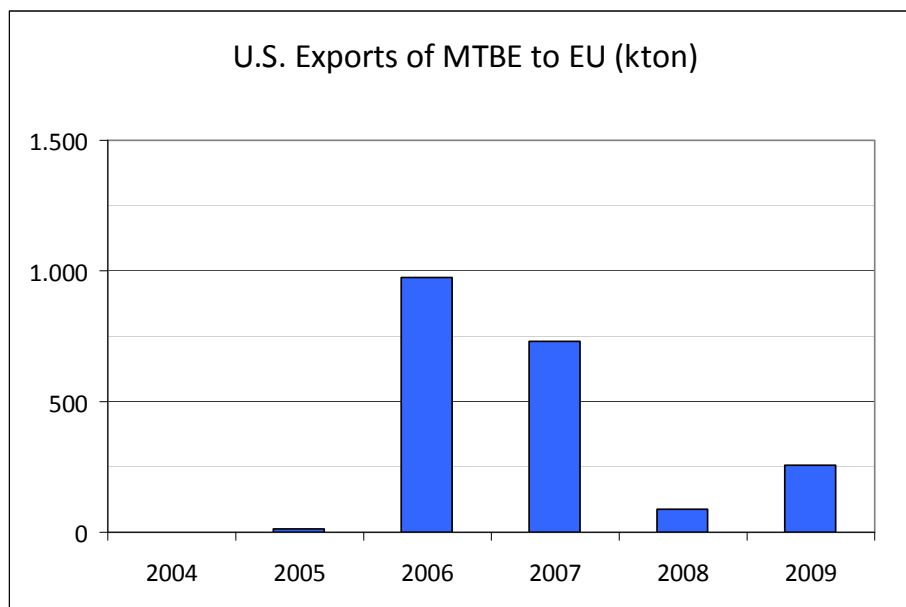
### 2.3. IMPORT AND EXPORT OF GEO

- Detailed import and export numbers of GEO in the EU are currently not known.
- Numbers are most likely constantly changing due to changing regulations, fluctuating production capacities and GEO demand.

In the period 1995 to 1997, 28% of MTBE produced (727 to 904 ktonne MTBE) was exported out of the EU and 8% of the EU's demand was met by imported GEO (EU, 2002). In 2003 2,512 ktonne GEO was produced, of which 539 ktonne (21% of production) was exported. In 2003, a total of 609 ktonne (24% of production) was imported into the EU (EFOA, 2005).

In the period 2005 - 2009 there was a temporary strong increase of MTBE importation into the EU from the United States, with a peak in 2006 of almost 1,000 ktonne/yr (**Figure 2.4**). This equals about 40% of the EU production capacity in 2005. Most of the MTBE was transported to the Netherlands and shipped to other EU countries. This increase was caused by the phasing out of MTBE use in gasoline in the United States, but continued production entering the global market place. Since 2006, US production and exports to the EU have decreased to 250 ktonne/yr in 2009 (EIA, 2010). The exact amount of MTBE imported to, and exported from, countries other than the United States is currently not known.

**Figure 2.4** MTBE export from the United States into the European Union (EIA, 2010)



**2.4. MARKET SHARES OF REGULAR AND PREMIUM GRADE GASOLINE**

- GEO demand is influenced by gasoline demand and GEO concentrations in the different gasoline types.
- GEO are blended with gasoline in different concentrations depending on the gasoline type.
- The total annual gasoline demand in EU27 in the year 2007 was 104,340 ktonne (**Appendix 2**).
- The volume of gasoline sales in EU 15 is decreasing, while the diesel consumption is increasing (2004 – 2008).
- The market share of Premium Unleaded gasoline (RON95) (PUL) has gradually increased from 71% in 2004 to 89% in 2008 at the expense of Regular unleaded gasoline (RON91) (RUL) and Super Premium Unleaded gasoline (PULP).
- The market share of Lead Replacement gasoline (LRG) has decreased to nearly zero in 2008.

GEO are blended with gasoline in different concentrations depending on the gasoline type. To establish the possible presence of GEO in the environment in individual EU countries, the market share of gasoline in different European countries was analysed.

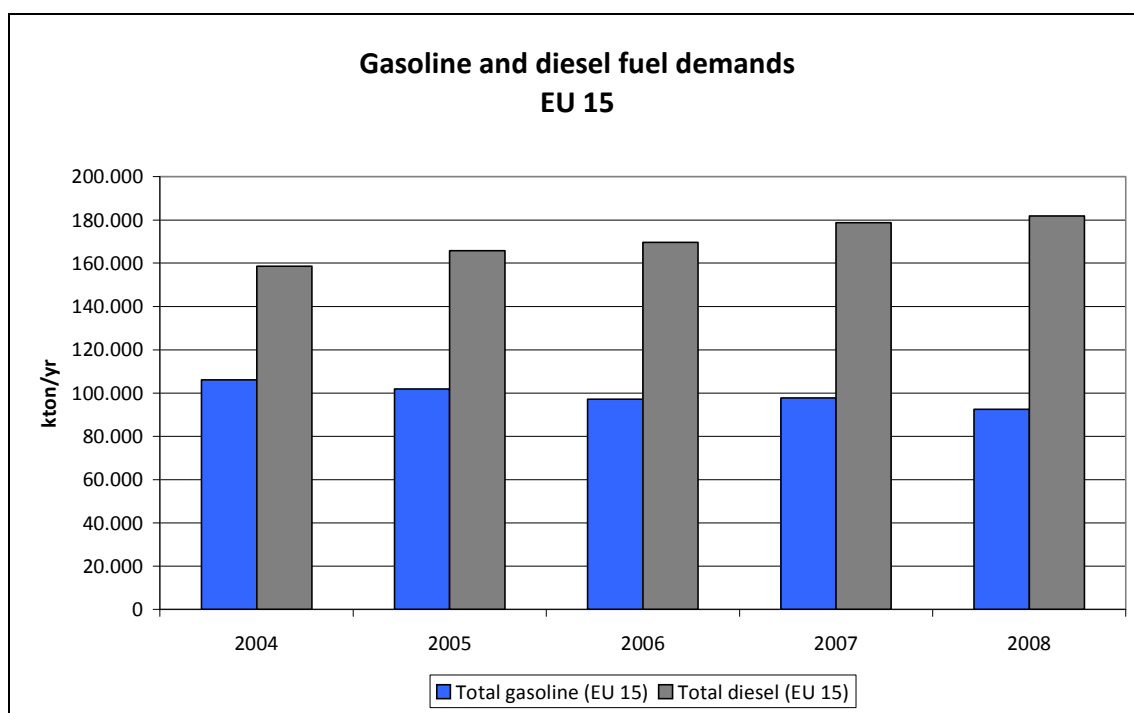
Two databases were consulted to examine the market share of gasoline:

- EU-conducted investigations into the composition of gasoline in the EU, which was performed by AEA Energy and Environment in 2002 to 2007

- EPTC Research undertaken by the Italian Unione Petrolifera in 2004-2008 (EPTC, 2005-2009),

**Figure 2.5** shows the EPTC data of the market demands for gasoline and diesel fuel in 15 mainly western European countries in 2004 - 2008. This number accounts for about 90% of the total fuel demand volume in the 27 EU countries.

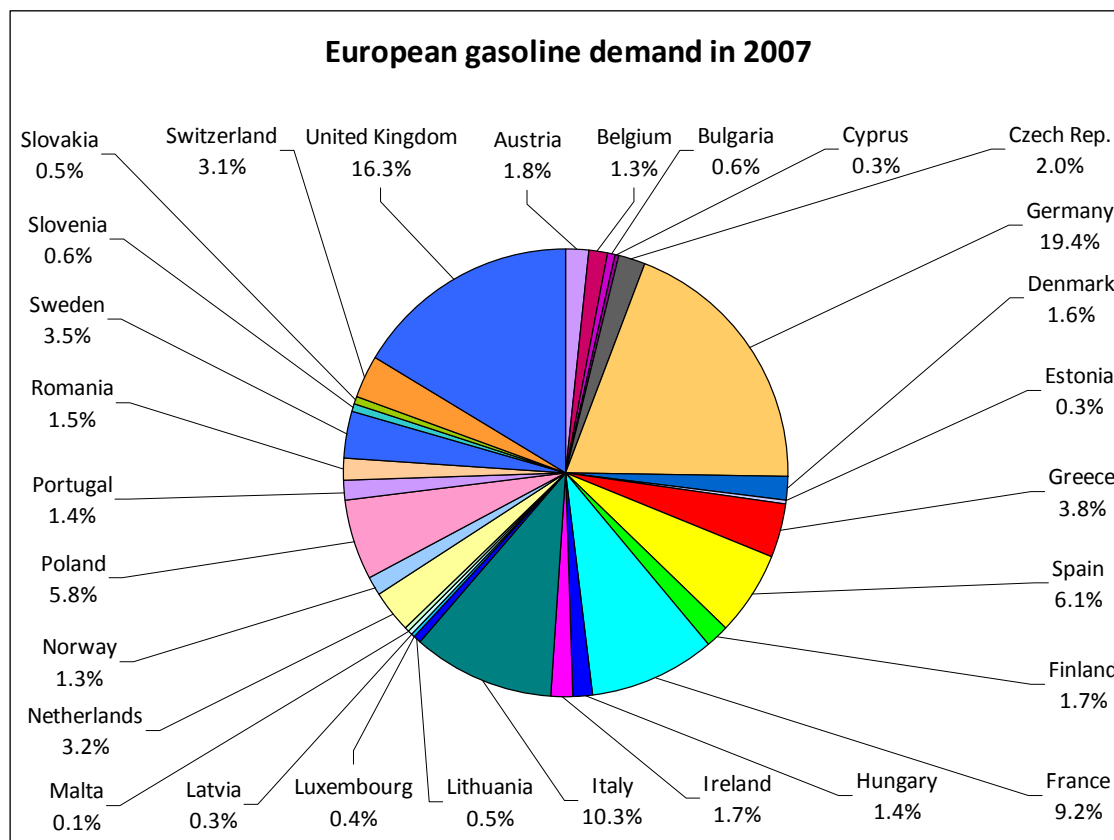
**Figure 2.5** Market demands for gasoline and diesel fuel in 15 countries in Europe (Austria, Belgium, Czech Republic, Denmark, France, Germany, Greece, Italy, Netherlands, Norway, Poland, Spain, Sweden, Switzerland, United Kingdom) in ktonne/yr (EPTC, 2005-2009)



In general the volume of gasoline sales is gradually decreasing in these 15 countries. In contrast diesel consumption is increasing due to the rising share of diesel engine vehicles in the EU (Pock, 2007).

Based on the EU data, the total annual gasoline demand in the EU 27 (without Croatia, Norway and Switzerland) in 2007 was 141,000 million litres or 104,340 ktonne. The proportion of European market share in 2007 by individual countries based on the EU data is presented in **Figure 2.6**. Over 50% of the gasoline is consumed by Germany, United Kingdom, Italy and France.

**Figure 2.6** Gasoline demand of individual countries (EU 27) in 2007, as a percentage of total European demand (EPTC, 2005-2009)

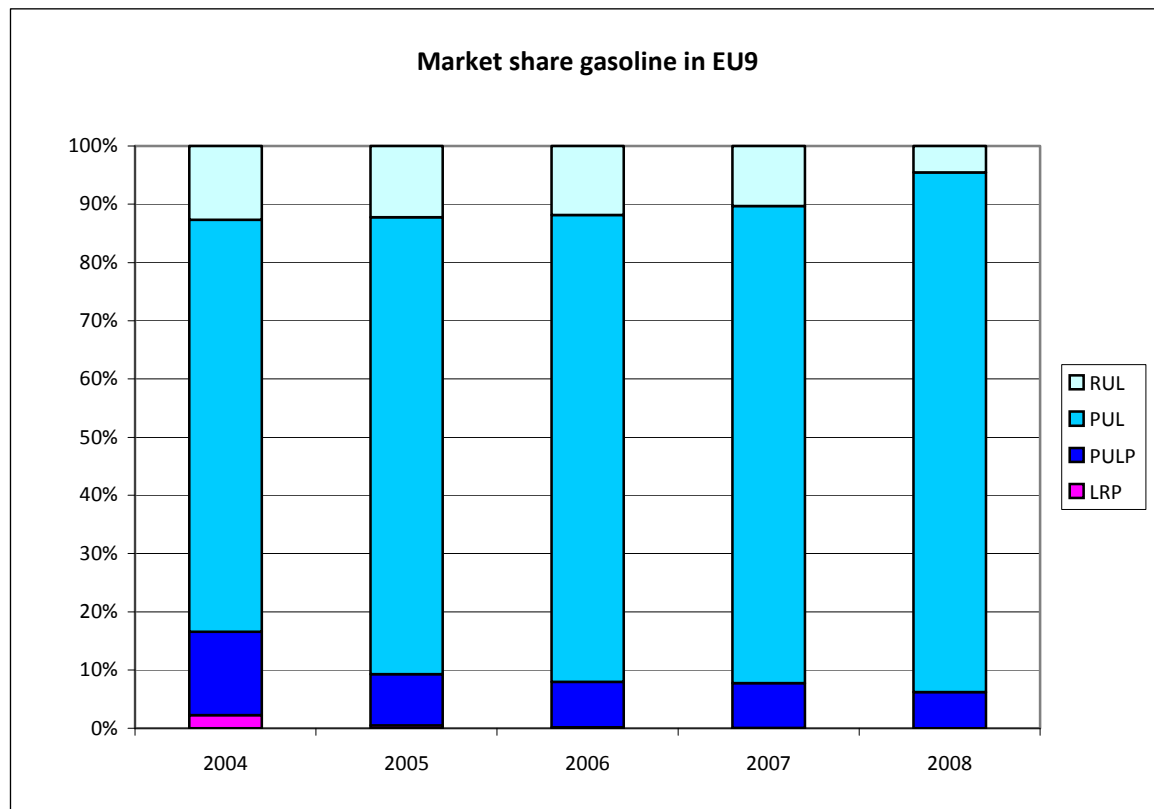


Changes in the demand of individual gasoline types (Lead Replacement Petrol or Gasoline (LRP or LRG), Unleaded RON<sup>2</sup>91 (RUL, Regular UnLeaded gasoline)), Unleaded RON95 (PUL, Premium UnLeaded gasoline) and Unleaded RON98 (PULP, Super Premium UnLeaded gasoline)) in 9 individual countries (Austria, Belgium, Czech republic, Denmark, France, Germany, Greece, Italy, Spain, Switzerland) are presented in **Figure 2.7**.

The market share of PUL (RON95) has gradually increased from 71% in 2004 to about 89% in 2008, at the expense of RUL (RON91). The market share of LRP has decreased to zero in 2008.

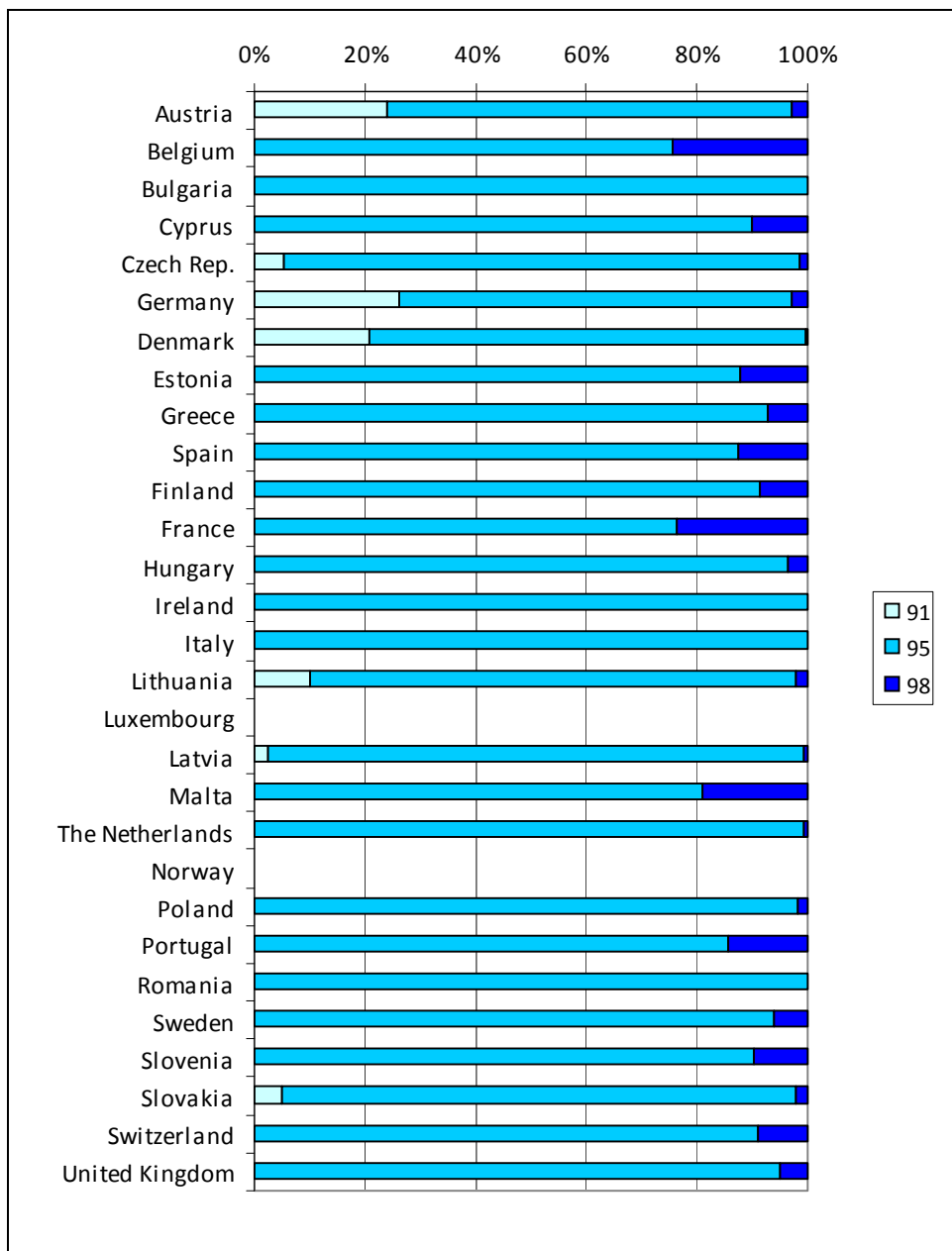
<sup>2</sup> RON : Research Octane Number

**Figure 2.7** Development of individual gasoline type demand (Lead Replacement Gasoline (LRP), Unleaded RON91 (RUL), RON95 (PUL) and RON98 (PULP)) in 2004 – 2008 in 9 European countries (Austria, Belgium, Czech Republic, Denmark, France, Germany, Greece, Italy, Spain, Switzerland) (ETPC 2005-2009)



The distribution of demand in gasoline type in individual EU countries and Switzerland in 2007 is presented in **Figure 2.8**. In most countries PUL (RON95) has the highest market share, varying from 75 to 100%. In Austria, Germany, Denmark and Lithuania RUL (RON91) has a relatively high market share of 10 to 25%. In Belgium, France and Malta PULP (RON98) has a high share in the market of about 20%.

**Figure 2.8** Demand for gasoline type in individual EU countries and Switzerland in 2007 (EPTC, 2005 - 2009)



**2.5. GEO IN REGULAR AND PREMIUM GRADE GASOLINE**

- Data on GEO composition of 6 different gasoline types in EU 27, Norway, Switzerland and Turkey in the period 2000 to 2009 was obtained from SGS and analysed (1,239 sampling events for MTBE, ETBE and TBA and 650 sampling events for TAME and DIPE).
- MTBE was detected in PUL gasoline in all EU 27 in the period 2004 – 2009.

- ETBE use in PUL is less widespread than MTBE in the period 2004 – 2009.
- The highest ETBE concentrations (5 – 12 m/m%) in PUL gasoline were found in France, Spain, Hungary and Austria.
- TAME was detected in PUL and PULP gasoline in concentrations from 1 to 3 m/m% in Cyprus, Finland, Greece, Italy, Malta, Romania and Slovenia.
- DIPE was detected only in 7 out of 650 samples, consistently at concentrations below 1 m/m%.
- TBA traces (0.01 – 0.03 m/m%) were detected in many EU countries.

### 2.5.1. General observations on individual GEO

In 2004-2009 MTBE and ETBE, in general, are present in higher concentrations in gasoline, when compared with TAME, DIPE and TBA. MTBE was detected in PUL gasoline in all European countries. Denmark was the first country to phase out the use of MTBE. France historically has a low MTBE concentration (<1%) in PUL gasoline. Poland and the UK also have a low concentration of MTBE in PUL gasoline.

ETBE use in PUL was less widespread in 2004-2009 compared with MTBE, but its use is growing. The ETBE concentration in PUL was high (5 – 12%) in France, Spain, Hungary and Austria, whereas in Germany the ETBE concentration in PUL was decreasing.

TAME is applied in PUL and PULP gasoline in a few countries. In Cyprus, Finland, Greece, Italy, Malta, Romania and Slovenia TAME is detected in PUL and PULP in a concentration of 1 to 3%. The highest concentration was detected in Finland. This is related to the TAME production facilities in Finland, Greece and Italy.

DIPE was detected in only 7 out of 650 samples in Bulgaria, Greece, Malta, Italy and Sweden in a concentration below 1%. Trace amounts of TBA (0.01 to 0.03%) are detected in many EU countries.

### 2.5.2. General

Data on the GEO composition of 6 gasoline types in 27 EU countries, Norway, Switzerland and Turkey in the period from winter 1999/2000 until summer 2009 was obtained from Société Générale de Surveillance (SGS). In this period, twice yearly, gasoline was sampled and analysed for the presence of GEO in 27 EU countries. Data for Croatia were not available. The database contains information from 1,239 sampling events on MTBE, ETBE and TBA, and 650 sampling events on TAME and DIPE. The data contains minimum, maximum and average concentrations of MTBE, ETBE, TAME, DIPE and TBA in 6 gasoline types in several sampling rounds (country-wide) in summer and winter periods. The average concentrations per country during a period (summer or winter) were calculated and used for analyses in this study.

The complete dataset is held in Excel format at CONCAWE. The observations described in the next sections were extracted from the SGS database. The data available is too extensive to be completely presented in this report. The average concentrations observed in a monitoring round are presented in **Appendix 3** and in figures illustrating the trends are presented in **Appendix 4**.



The dataset is extensive but does not completely cover the period 2000 – 2010 for all countries, gasoline types and GEO. PULP was monitored intensively until 2004, but later the focus in 2006 – 2009 was on PUL. RUL and RL (Regular Leaded) was monitored only in 2000. LRG was monitored only in 2009. In this chapter the focus is on RUL, PUL and PULP. **Appendix 4** presents the GEO concentration in RUL, PUL and PULP for different years in selected EU countries. In addition, monitoring of MTBE, ETBE and TBA in gasoline in the period 1999 to 2002 was made available by CONCAWE, but was only used for reference purposes.

The average (mean), median and maximum concentrations of ether oxygenates found in individual sampling rounds in countries that were observed in the SGS database are presented in **Table 2.1**. Based on the analysis of this data set, 1.5% of the samples contained GEO above 15% MTBE or ETBE.

**Table 2.1** Mean, median and maximum reported concentrations of GEO (m/m%)% in gasoline in 27 European countries in the SGS database. Minimum levels were all 0.00 m/m%.

|  | Ether Oxygenates |                  |                  |                  |                  |
|--|------------------|------------------|------------------|------------------|------------------|
|  | MTBE             | ETBE             | TAME             | DIPE             | TBA              |
| <b>Total number of sampling events (n)</b>   |                  |                  |                  |                  |                  |
|  | 1,239            | 1,239            | 650              | 650              | 1,239            |
| <b>Mean (average) concentration in all European gasoline samples in 2000-2009 (m/m%)</b> |                  |                  |                  |                  |                  |
|  | 5.39             | 0.91             | 0.29             | 0.00             | 0.03             |
| <b>Median concentration in all European gasoline samples in 2000-2009 (m/m%)</b>         |                  |                  |                  |                  |                  |
|  | 4.25             | 0.00             | 0.00             | 0.00             | 0.01             |
| <b>Maximum concentration observed in individual sampling rounds (m/m%)</b>               |                  |                  |                  |                  |                  |
| Concentration (m/m%)   | 20.43            | 15.50            | 11.22            | 0.88             | 1.03             |
| Gasoline type  | PULP             | PUL              | PUL              | PULP             | PULP             |
| Period   | Summer 2001      | Winter 2008/2009 | Winter 2007/2008 | Winter 2002/2003 | Winter 2000/2001 |
| Country  | Romania          | France           | Finland          | Greece           | Switzerland      |

In the following sections general observations from the SGS data presented in **Appendix 3** are presented for the different:

- GEOs
- Fuel types
- Countries

### 2.5.3. GEO Composition of gasoline types

In this chapter the GEO concentration in gasoline will be discussed in relation to the gasoline types. The average concentrations of GEO refer to the data in **Appendix 3**.

### **Lead Replacement Gasoline (LRG or LRP)**

From **Figure 2.7** it can be observed that LRG has a small market share (<1%) from 2005 - 2009. The only data on LRG available is for one sampling round in summer 2009, for Denmark, Greece, Ireland, Malta, Slovakia and Spain. LRG contained an average MTBE concentration of 2.99%, with a maximum of 10.1% in summer 2009 Ireland. ETBE was observed only in Spain, up to 1.53%. TAME was analysed for only in Malta, with an average concentration of 0.92%. DIPE was not detected (investigated only in Malta) and the TBA concentration found were low (maximum 0.05%).

### **Regular and Premium Leaded Gasoline (RL and PL)**

RL and PL had a very small market share. RL gasoline was investigated only in summer 2000 in Bulgaria and Poland. A trace amount of MTBE was found (0.29 and 0.64%, respectively). ETBE and TBA were not detected. TAME and DIPE were not investigated. PL Gasoline was investigated only in summer 2009 in Cyprus, Czech Republic, Greece, Italy, Malta, Poland and Spain. The average MTBE concentration was 1.46% and the highest concentration was found in Spain (8.33%). ETBE was also investigated in these countries but only detected in Spain at 1.24% and in a trace amount in the Czech Republic (0.04%). A trace concentration of TBA (0.01%) was found only in Spain. DIPE and TAME concentrations were not investigated.

### **Regular Unleaded Gasoline (RUL RON 91)**

From **Figure 2.7** it can be observed that RUL had a market share of about 6% from 2005 - 2009. The RUL composition was investigated by SGS only in 2000 in 13 countries. MTBE, ETBE and TBA were analysed. The average MTBE concentration was 1.00%, with a maximum of 16.58% in winter 1999/2000 in Latvia. ETBE was found only in Estonia and Lithuania in very low concentration (0.10% and 0.02%). In 2000 MTBE was the most important GEO used. TBA was found in a trace amount in Estonia (0.09%), Germany (0.46%) and Austria (0.40%). TAME and DIPE concentrations were investigated only in Austria, but were not found above detection limit.

### **Premium Unleaded Gasoline (PUL RON95)**

Premium unleaded gasoline had a market share of about 87% in 2005-2009 (**Figure 2.7**) and was investigated in all countries from summer 2004 to summer 2009. The highest concentration of MTBE (above 10%) was identified in winter 2008 in Bulgaria, Cyprus, Estonia, Latvia and Portugal. Since then, the average MTBE concentration in PUL has dropped. Proportionally, the average ETBE concentration in European PUL has gradually increased, as a replacement for MTBE. The ETBE concentration was highest in Austria, Spain, France and Slovakia. The average TAME concentration in European PUL from 2006 – 2009 was low (< 1%). The maximum concentration of TAME (11.22%) was used in Finland in winter 2007/2008, although the TAME concentration appears to decrease in 2009. DIPE was not detected and TBA was found only in trace amounts of 0.01 to 0.03%. The average TBA concentration tended to rise; however, the reason for this is unclear.

### **Super Premium Unleaded Gasoline (PULP RON98)**

Super premium unleaded gasoline was investigated in all countries from summer 2000 to summer 2004. GEO concentrations of more than 10% have been applied in most countries, besides France, Ireland, Malta, Poland, Spain and the United

Kingdom, which generally had a low concentration of MTBE in PULP. The average MTBE concentration was highest in the winter of 2003. After 2003 the average concentration dropped in 2004 to the lowest value since 2000, caused by the increasing use of ETBE as a replacement for MTBE. The average ETBE concentration in European PULP increased from 0.21% in 2000 to 4.84% in 2004. The ETBE concentration was highest in Austria, France, Poland, Slovakia and Spain, but low in Cyprus, Greece, Norway, Portugal, Sweden and the United Kingdom. The average TAME concentration in European PULP from 2000 – 2004 was <0.20%. The highest concentration of up to 7% was applied in Cyprus in winter 2003/2004. In Greece, Romania and Hungary TAME was also detected at an above average concentration. DIPE was detected at a low concentration in Bulgaria, Greece, Italy and Malta. TBA was also found only at a low concentration of 0.01 to 0.07%. As with PUL the average TBA concentration in PULP tended to rise.

#### **2.5.4. Country by country evaluation**

**Appendix 4** presents the SGS-reported average GEO concentration in RUL, PUL and PULP in different years in all the countries under study. The countries with highest fuel consumption are described in this chapter. Finland is unusual in Europe in so far as TAME is produced and applied widely in gasoline and is, therefore, also described in this chapter.

##### **Finland**

Finland is one of the few producers of TAME and TAAE, where they were applied widely in gasoline. The production capacity in 2007 exceeds the national GEO demand in gasoline, by about 50%.

##### **France**

ETBE dominated the French GEO market and France was one of the first countries to introduce ETBE. MTBE was used only in small amounts. The GEO production capacity in 2007 was below the national GEO demand in gasoline and consequently part of the ETBE will have been imported.

##### **Germany**

In the past MTBE was widely used in PUL and PULP gasoline. Since 2007 the ETBE concentration in PUL has been higher than the MTBE concentration. However, the ETBE concentration in PUL gasoline has dropped to less than 2% since 2008, due to direct blending of ethanol in gasoline. The GEO production capacity exceeded the national market's demands.

##### **Italy**

MTBE was mostly used in the Italian market, but ETBE use is increasing. The production capacity exceeded the national market's demands.

##### **Spain**

ETBE dominated the Spanish GEO market. Together with France, Spain was one of the first countries to introduce ETBE. MTBE was used only in small amounts. The production capacity in 2007 was sufficient to meet the national market's demands.

### United Kingdom

GEO concentrations in UK gasoline were generally low, up to 1 – 2%, and mostly represented by MTBE. TAME was also applied in some instances. The production capacity in 2007 was sufficient to meet the national market's demands.

## 2.6. FUTURE DEVELOPMENTS

- Gasoline consumption is expected to decrease in favour of the use of diesel vehicles and due to enhanced vehicle engine efficiency.
- GEO demand is expected to decrease less than the general gasoline demand due to the biofuel directive.
- Increased use of ETBE derived from bio-ethanol is expected.

The projected gasoline consumption is expected to further decrease towards 2020 in favour of the use of diesel vehicles. Furthermore the enhanced fuel efficiency will also impact the reduction in fuel consumption. With 2005 as a reference year the demand for gasoline in the EU is expected to be 75% in 2010 and only 56% (of the 2005 demand) in 2020.

However the GEO demand is expected to decrease less, as the biofuels directive mandates that the amount of fuels originating from renewable biological resources has to increase over time, up to 10%. The shift from MTBE to ETBE is expected to continue as ETBE derived from bio-ethanol is considered a biofuel. This is also the case for MTBE derived from bio-methanol.

### **3. OCCURRENCE OF GEO IN THE EUROPEAN ENVIRONMENT**

#### **3.1. BASIC INFORMATION ABOUT DATA COLLECTION AND EVALUATION**

A large number of different reports and literature were reviewed to obtain as much information as possible on the occurrence of GEO in different environmental media across selected European countries. The focus was on identification and consideration of already interpreted data and the related conclusions. A general picture for each environmental medium was developed, based on the results obtained within this study.

An additional aspect was to evaluate the current state-of-knowledge on the distribution of GEO in 30 countries in Europe (27 EU countries and Switzerland, Norway and Croatia). This was done by using actual monitoring data obtained by authorities and environmental institutes that have not been published in reports to date. In order to obtain these data, authorities and organisations of the above mentioned countries were contacted by local TAUW network staff. Using a dedicated questionnaire, the authorities and institutes were asked to provide local data on GEO-occurrence in the environment.

Based on this survey, a lot of unevaluated and uncommented data was provided by the responding authorities and organisations. To supplement the information from the different literature resources, these data sets were evaluated in a generalised way leading to the following major observations:

- Number of total measurements per GEO and medium in the provided time scale.
- Number of measurements below the detection limit.
- Number of measurements with values above the detection limit.
- Range of detected concentrations.
- Separation between 'background' values and values from 'contamination spots and plumes' as far as possible.

Having regard to the known chronology of GEO use in fuels in Europe, and to known dates of research publications and environmental survey data on GEO, the study was limited to the period 2000 – 2010.

#### **3.2. OCCURRENCE IN THE ENVIRONMENT**

Within this chapter, a survey of available information on the occurrence of GEO in groundwater, drinking water, surface water, precipitation, air and soil is provided, as far as has been reported in different literature sources and authority information.

In general, most information available refers to the occurrence of GEO in groundwater. Most investigations had been performed in Europe over the last 15 years; the reason for this is the high mobility and the potential risk for contamination of drinking water resulting from the use of groundwater resources.

In contrast, little information and data are available for the research of GEO in soil. With respect to the properties of GEO, there is a relatively low retardation of GEO

within the soil, resulting in a lower frequency of sampling and also lower analytical identification.

A relatively wide range of investigations and data are available for the distribution of GEO in surface water and drinking water. The monitoring systems of Germany, Netherlands, Austria and UK provide the largest contribution to this knowledge.

There is very little information on GEO distribution for runoff water, rain and snow (precipitation) and GEO distribution in air. In this context, only one study and some spot information could be identified.

Due to the variation in amount of information for the different GEO in each country of Europe, the following explanations are structured per compartment. An additional separation of the information obtained per country was performed only for compartments with a wide information background (such as groundwater and surface water).

### **3.2.1. Literature Review and results of enquiry - GEO in groundwater**

- The knowledge on the distribution of GEO in groundwater is relatively good.
- A lot of monitoring and research data and evaluation reports are available for Austria, Denmark, Finland, Germany, the Netherlands, Switzerland, and the UK.
- The reviewed literature showed that MTBE is the most studied GEO, and that it is reported in European groundwater over a wide range (both in terms of location and time).
- Gasoline stations, depots and related production facilities represent the majority of the sites with high concentration levels of GEO.
- Low concentration 'background' MTBE was observed in urban areas – here detection frequencies range from 40 – 60%.
- Low concentration 'background' detection frequencies of MTBE in rural areas are much lower (10 – 20%).
- Diffuse source concentration for MTBE typically range from 0.01 to 1 µg/l (i.e., well below the taste, odour and health-related thresholds).
- Concentrations of MTBE above 1 µg/l are often assumed to be an indication of proximity to a point source.

#### **3.2.1.1. Distribution of GEO in Austrian groundwater**

- The Austrian results (dating from 2002) indicated the presence of MTBE in groundwater at low concentrations.
- Most MTBE detections were found in intergranular porosity aquifers.
- The Federal Environmental Agency of Austria (2004) stated that in cases of MTBE concentrations above 1 µg/l point sources have to be taken into account.
- No groundwater samples in Austria had MTBE at concentrations exceeding the taste and odour thresholds (for potable supply) of 20 µg/l proposed by USEPA.

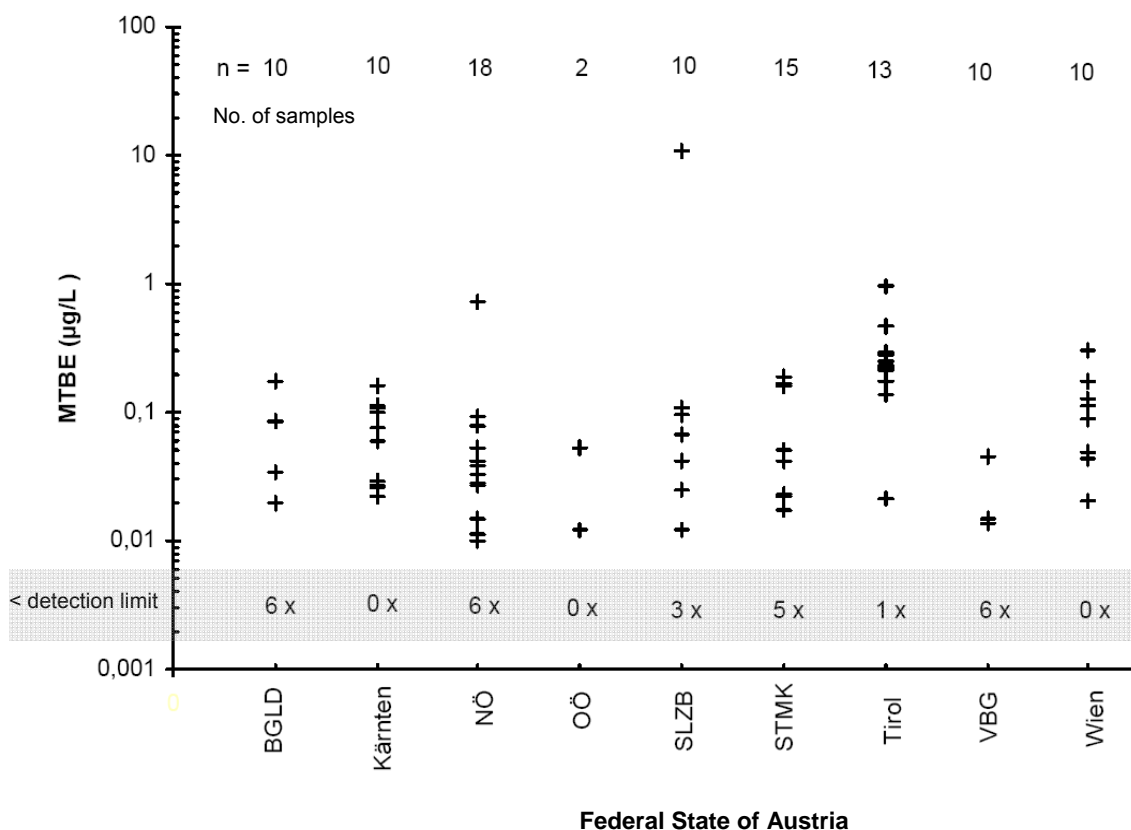
Within the scope of the annual report of water quality by the Federal Environmental Agency of Austria (2002), a number of 101 groundwater samples were evaluated, with 6 samples belonging to known contaminated sites (gasoline stations).

For the 6 samples of the contaminated sites, the MTBE concentration ranged from 0.129 up to 1,594 µg/l, with a median value of 369 µg/l.

**Figure 3.1** provides a graphical evaluation of the remaining 95 groundwater samples per "Bundes Land". Three of these samples belong to a karst/fissure aquifer type, while 92 samples originate from an inter granular porosity aquifer.

The median value for the groundwater samples was 0.036 µg/l, while the highest value was 10.6 µg/l. Only 27 out of 95 samples were below the detection limit (28.4%), indicating a wide concentration distribution of MTBE in the groundwater.

**Figure 3.1** Distribution of MTBE concentration in groundwater from the 2002 annual water quality report of the Federal Environmental Agency of Austria (2002)

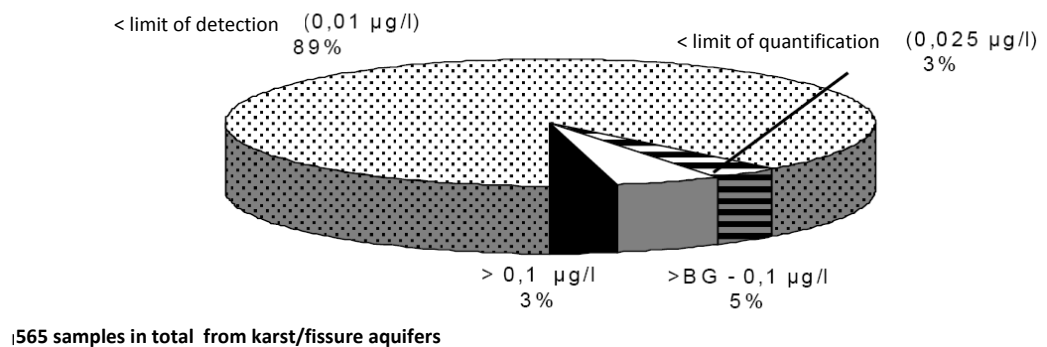


Due to the 2002 results, the Austrian regulators expanded the MTBE monitoring in groundwater, as documented in the annual water quality report of 2004 (Federal Environmental Agency of Austria, 2004).

Within the scope of this monitoring, 217 sampling locations in karst/fissure aquifers were analysed (total number of samples = 565). Only 32 of the 217 locations had samples above the detection limit of 0.01 µg/l (14.7%). The number of positive

samples was 49 from 565 samples (8.7%); the highest concentration noted was 1.2 µg/l. **Figure 3.2** provides an overview on the distribution of the evaluated MTBE concentration. In respect to **Figure 3.2** the detection limit (0.01 µg/l) represents the limit of positive detection, while the limit of quantification (0.025 µg/l) represents the limit for reliable concentration values.

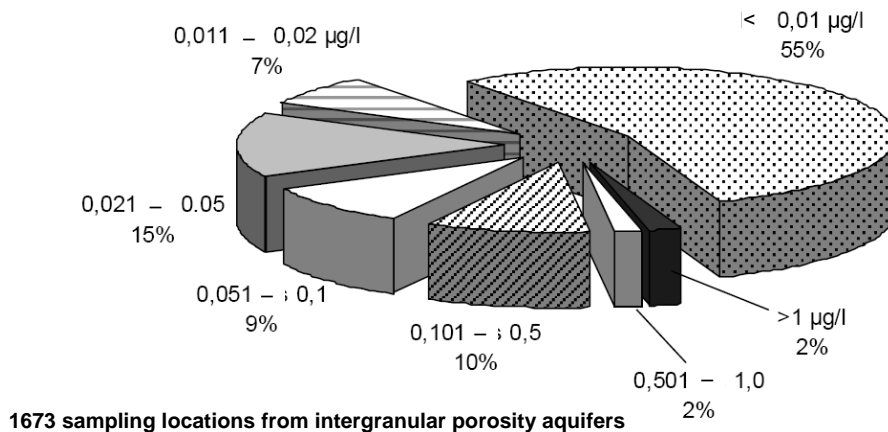
**Figure 3.2** Distribution of MTBE concentration values in groundwater for karst/fissure aquifers, from the Federal Environmental Agency of Austria (2004)



For intergranular porosity aquifers 1,673 sampling locations were analysed (nearly 5,000 samples in total). Around 56.2% of those locations showed results above the detection limit of 0.01 µg/l. The highest value noted was 31 µg/l.

**Figure 3.3** shows the distribution of the observed mean values with regard to the sampling locations.

**Figure 3.3** Distribution of mean values of MTBE concentration in groundwater for intergranular porosity aquifers, from Federal Environmental Agency of Austria (2004)



Based on the large amount of data, the average MTBE concentration (only with regard to samples above the limit of quantification) ranges from 0.04 – 0.06 µg/l



(Upper Austria, Lower Austria, Burgenland, Styria) to 0.08 – 0.12 µg/l (Vienna, Carinthia, Tyrol) and 0.93 µg/l (Vorarlberg).

**3.2.1.2. Distribution of GEO in Swiss groundwater**

- The results from Switzerland show the presence of MTBE in 10 – 19% of the monitored groundwater locations.
- The median MTBE concentration was around 0.1 µg/l.
- MTBE showed the highest detection rate of all volatile organic compounds in Switzerland (BUWAL, 2004).
- The distribution of the other GEO in groundwater is significantly lower, but ETBE and TBA were detected in Canton St. Gallen.
- No groundwater samples in Switzerland had MTBE at concentrations exceeding the taste and odour thresholds (for potable supply) of 20 µg/l proposed by USEPA.

Early observations of MTBE in Swiss groundwater were reported in BUWAL (2002). The report describes no detectable MTBE in groundwater between 1995 to 1997 (limit of detection 0.05 µg/l). For 1998/99, an investigation of 95 locations was reported – here, around 20% of the locations had an MTBE concentration between 0.06 and 0.9 µg/l.

Additional observations were made from 04/2000 to 10/2001 in the scope of the quality observation of groundwater (NAQUA) – here, 9 samples out of 162 in total (39 sampling locations) had an MTBE concentration in the range of 0.12 – 0.34 µg/l.

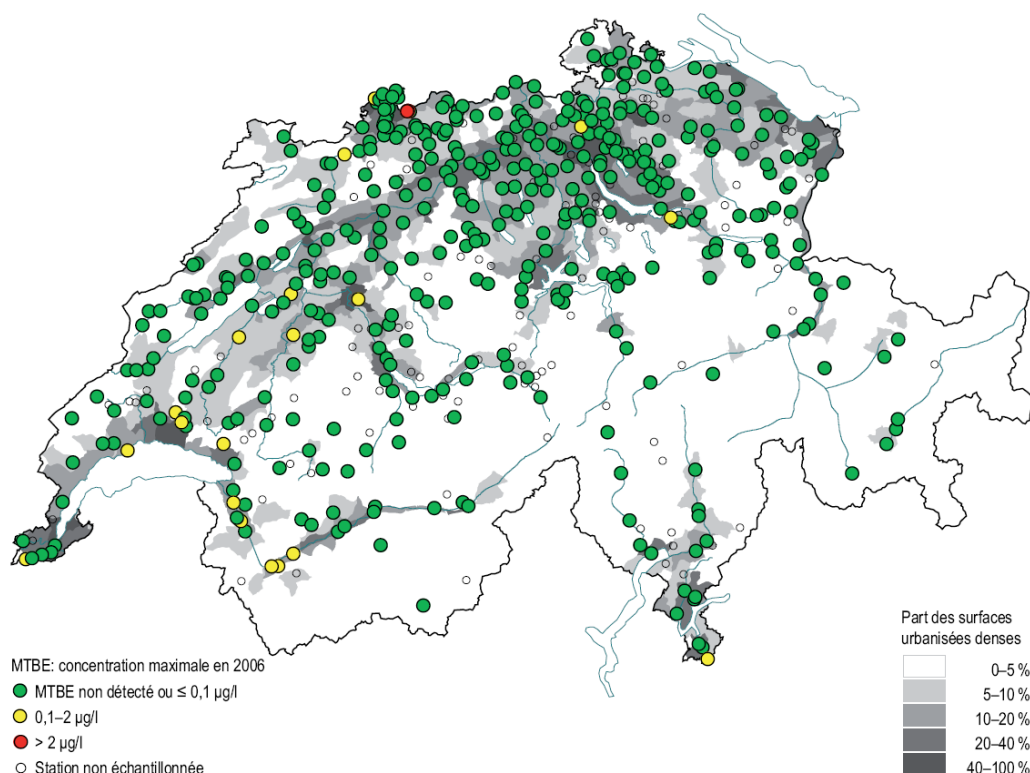
In the scope of the NAQUA programme, BUWAL (2004a) reported 413 sampled locations. In the report of OFEV (Kilchmann et al, 2009) on the groundwater quality, the monitoring results for 2004, 2005 and 2006 are presented. The data from these observations are summarized in the following **Table 3.1**:

**Table 3.1** Results of the MTBE monitoring from 2004 – 2006 in the scope of the NAQUA programme – based on OFEV (Kilchmann et al, 2009) / BUWAL (2004)

| Year   | 2002       | 2003     | 2004      | 2005         | 2006      |
|--|------------|----------|-----------|--------------|-----------|
| Total no. of GW monitoring points (n)                  | 210        | 400      | 421       | 496          | 431       |
| Monitoring points above limit of quantification (n /%) | 21 / 10%   | 72 / 18% | 78 / 19%  | 79 / 16%     | 53 / 12%  |
| Limit of quantification (µg/l)                         | 0.03 – 0.2 | 0.03 - 1 | 0.015 - 1 | 0.015 – 0.25 | 0.024 - 1 |
| Min. value (µg/l)                                      | 0.03       | 0.03     | 0.02      | 0.04         | 0.04      |
| Median value (µg/l)                                    | 0.1        | 0.09     | 0.13      | 0.1          | 0.10      |
| Max value (µg/l)                                       | 0.5        | 2.1      | 4.5       | 0.5          | 6.4       |

The map in **Figure 3.4** provides an overview on the distribution of evaluated MTBE concentration in 2006 in regard to urbanisation, indicating that a higher concentration was more often found in areas with a higher grade of urbanisation.

**Figure 3.4** Distribution of the observed max. MTBE concentration in 2006 with regard to the degree of urbanisation, from OFEV (Kilchmann et al, 2009).



In addition to the above quoted MTBE results, the OFEV (Kilchmann et al, 2009) reports also on ETBE measurements in groundwater samples in 2005 (13 stations) and 2006 (96 stations); none of the locations had a concentration above the limit of quantification ( $0.03 - 0.15 \mu\text{g/l}$ ). Measurements for DIPE at 19 locations in 2006 also did not show any values above the limit of quantification ( $0.05 \mu\text{g/l}$ ).

Monitoring for TBA in 2004 (9 locations), 2005 (13 locations) and 2006 (16 locations) did not show values above the limit of quantification (range  $0.034 - 0.335 \mu\text{g/l}$ ) either.

UFAM (2010) reported measurements for MTBE (498 locations) and ETBE (491 locations). In 2008, MTBE was detected in 71 locations (14.2%), but a concentration  $>1 \mu\text{g/l}$  was reported for two locations only. ETBE was detected in 7 locations (1.4%), but a concentration  $> 1 \mu\text{g/l}$  was not observed in any sample.

Laboratorio cantonale (2008) reported contamination of groundwater with GEO in Morbio Inferiore. The MTBE concentration in 5 groundwater wells at one site was  $83.9$  to  $273 \mu\text{g/l}$ . ETBE was detected in the same wells with a concentration between  $11.8$  and  $13.1 \mu\text{g/l}$ .

Monitoring data from Basel

The city of Basel provided a bigger set of monitoring data for MTBE, ETBE and TAME. The data from these observations are summarized in **Table 3.2**:

**Table 3.2** Results of the GEO monitoring of BASEL

| Substance                          | MTBE        | ETBE        | TAME        |
|------------------------------------|-------------|-------------|-------------|
| Observation period                 | 2002 - 2009 | 2007 - 2010 | 2008 - 2010 |
| No. of sampling locations          | 129         | 58          | 43          |
| No. of samples in total            | 703         | 201         | 116         |
| Locations > limit of detection (%) | 4.65        | 0           | 0           |
| Detection limit (µg/l)             | 0.04 - 1    | 0.05 - 1    | 0.05 - 1    |
| Max. value (µg/l)                  | 0.2         | 0           | 0           |
| Median value of positive samples   | 0.0775      | 0           | 0           |

Monitoring data from Canton St. Gallen

Canton St. Gallen provided a bigger set of monitoring data for MTBE, ETBE and TBA. The data from these observations are summarized in **Table 3.3**:

**Table 3.3** Results of the GEO monitoring of Canton St. Gallen

| Substance                          | MTBE                  | ETBE                  | TBA                   |
|------------------------------------|-----------------------|-----------------------|-----------------------|
| Observation period                 | 2001 - 2009           | 2000 - 2009           | 2000 - 2009           |
| No. of sampling locations          | 65                    | 65                    | 65                    |
| No. of samples in total            | 472                   | 472                   | 472                   |
| Locations > limit of detection (%) | 10.8                  | 1.54                  | 15.4                  |
| Detection limit (µg/l)             | <i>* not provided</i> | <i>* not provided</i> | <i>* not provided</i> |
| Max. value (µg/l)                  | 2.12                  | 1.021                 | 0.11                  |
| Median value of positive samples   | 0.1                   | 0.29                  | 0.078                 |

**3.2.1.3. Distribution of GEO in German groundwater**

- The results from Germany show that MTBE is frequently reported in groundwater, but with the exception of point-source contamination is <20 µg/l.
- For urban areas, MTBE detection frequencies are in the range of 49% to 66% of all sampled locations.
- Non-urban areas have MTBE detection frequencies of 9% to 26%.
- The MTBE detection frequencies at (potentially) contaminated sites varies, but the observed concentrations differ significantly from the “urban background”.
- For ETBE and the other GEO, there is only little information available on groundwater data. Thus, it is not possible to derive a general result in this respect.

The Environmental Agency of the Federal State of Baden-Württemberg reports several groundwater investigations for MTBE at the beginning of the recent decade (LFU, 2002; LFU, 2003). The sampling of 72 potentially contaminated sites between 1999 and 2001 showed a detection of MTBE in 15.8% of all locations. The values obtained ranged from 0.2 to 830 µg/l.

An investigation of 420 observation wells in the Upper Rhine Plain in 2002 showed a detectable MTBE concentration at 65 wells (15.5%). Fifty-nine of these locations belong can be associated to potential emission sources. The values ranged from 0.05 to 18 µg/l, of which only 4 values exceeded 1 µg/l.

The Water Technology Center (TZW) performed an investigation of 170 observations wells in Germany in 1999, within a research programme of the DVGW related to MBTE impact on drinking water (reported in LFU, 2002 and Baus et al, 2008). In the evaluation of the results, the sampled locations were split into urban areas and rural areas. The results are summarised in **Table 3.4**:

**Table 3.4** Results of MTBE monitoring at 170 locations by TZW in 1999

| MTBE µg/l (ranges)                      | < 0.05 | 0.05 – 0.09 | 0.10 – 0.49 | 0.50 – 4.9 | > 5 |
|---|--------|-------------|-------------|------------|-----|
| percentage for rural areas <sup>1</sup> | 91%    | 2%          | 6%          | 1%         | 0%  |
| percentage for urban areas <sup>2</sup> | 51%    | 10%         | 21%         | 14%        | 4%  |

<sup>1</sup> ... 90 locations for rural areas / <sup>2</sup> ... 80 locations for urban areas (170 in total)

The results clearly demonstrate the significant difference in the distribution of MTBE between rural and urban areas.

Baus et al (2008) also quotes the results of MTBE monitoring in Hesse (1999-2002). Here, MTBE was detected in 26% of the rural observation wells (median value <0.01 µg/l, highest value 2.2 µg/l) and in 66% of the urban observation wells (median value 0.26 µg/l, highest value 47.6 µg/l).

More significant contamination of industrial sites and former storage facilities in the territory of the former GDR were reported for Leuna (Saxony – Anhalt,

concentrations up to 185,000 µg/l) and the former WGT site in Münchenbernsdorf (Thuringia, concentrations up to 2,000 µg/l) (Baus et al, 2008).

Effenberger (2001) reported groundwater investigations for 8 gasoline stations and 2 depots. For 5 (50%) of these sites groundwater MTBE concentrations >20 µg/l were reported. The MTBE concentration for the other sites ranged from 0.05 to 2.24 µg/l.

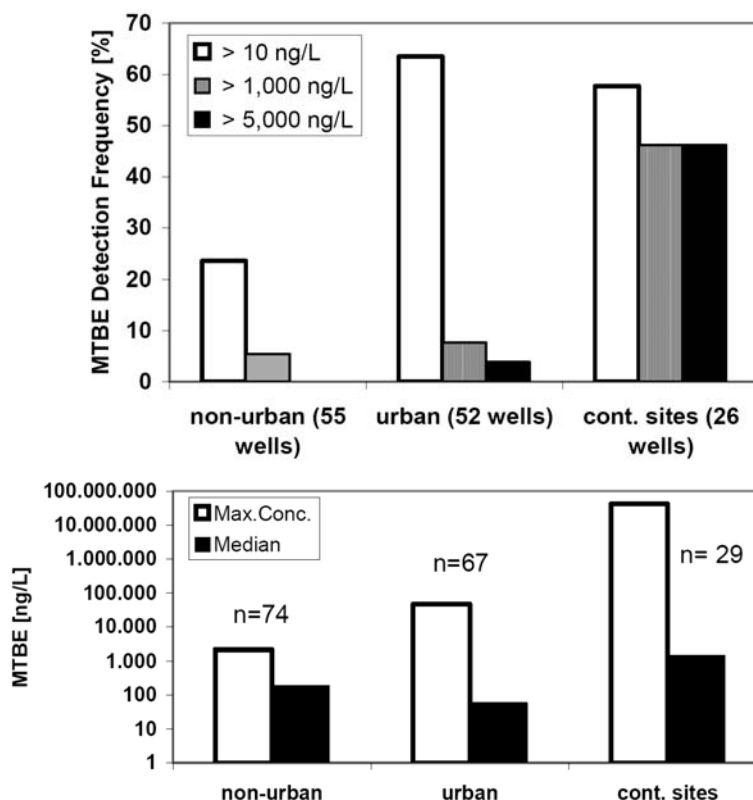
Kolb (2004) reported groundwater investigations from 2000 – 2003, with a focus on Hesse (Rhine-Main area / Main-Kinzig) and some samples from North Rhine-Westphalia and Baden-Württemberg. The samples and locations are summarized in the **Table 3.5**:

**Table 3.5** Summary data for groundwater samples from North Rhine-Westphalia and Baden-Württemberg (Kolb, 2004)

| Total       | contaminated sites | rural area | urban area |
|-------------|--------------------|------------|------------|
| 167 samples | 29 samples         | 74 samples | 64 samples |
| 133 wells   | 26 wells           | 55 wells   | 52 wells   |

The analyses revealed detection frequencies of 58% (contaminated sites), 24% (non-urban sites) and 63% (urban sites) at a detection limit of 0.01 µg/l. A graphical evaluation is provided in **Figure 3.5** below.

**Figure 3.5** Results of MTBE measurements for contaminated sites, rural and urban areas, taken from Kolb (2004)



Kolb (2004) reported that the data from non-urban samples revealed MTBE detection mainly at public supply wells with high pumping rates. In general, these results are comparable with other studies investigating MTBE in groundwater in Germany.

Monitoring data from Regulators in Bavaria, Saxony and Thuringia

The Federal States of Bavaria, Saxony and Thuringia provided a larger set of monitoring data for MTBE and ETBE. This monitoring data is summarized in **Table 3.6**.

**Table 3.6** Results of MTBE and ETBE monitoring in Bavaria, Saxony and Thuringia

| Federal state                      | Bavaria     |             | Saxony      |                           | Thuringia <sup>1</sup> |
|------------------------------------|-------------|-------------|-------------|---------------------------|------------------------|
|                                    | MTBE        | ETBE        | MTBE        | ETBE                      | MTBE                   |
| Observation period                 | 2001 - 2010 | 2006 - 2010 | 2002 - 2008 | 2008 - 2010               | 2003 - 2009            |
| No. of sampling locations          | 626         | 275         | 135         | 275                       | 30                     |
| No. of Samples in total            | 1,408       | 322         | 374         | 610                       | 37                     |
| Locations > limit of detection (%) | 11.98       | 1.45        | 3.7         | 0.32                      | 56.7                   |
| Detection limit (µg/l)             | 0.004 – 4   | 0.01 - 1    | 0.1 – 0.2   | 0.1 – 0.2                 | 0.2 - 1                |
| Max. value (µg/l)                  | 1,600       | 0.331       | 5           | 2.4                       | 31                     |
| Median value of positive samples   | 0.1         | 0.2         | 0.93        | 1.345 (based on 2 values) | 1.3                    |

<sup>1</sup> ... observation in Thuringia mostly on sites with known / assumed incidents

**3.2.1.4. Distribution of GEO in groundwater in the Netherlands**

- The results show a wide distribution of MTBE in Dutch groundwater, albeit generally at very low concentrations.
- Regional groundwater monitoring indicates MTBE detection at >20 µg/l in only 3 out of 207 (1.4%) of abstraction points.
- Detection of ETBE in regional groundwater resources is consistently <1 µg/l, well below the ETBE taste and odour threshold.
- For gasoline stations, MTBE values above 1 µg/l can be found in approx. 45% of the sites investigated.
- Groundwater abstraction wells show detectable MTBE in approx. 25% of all cases.

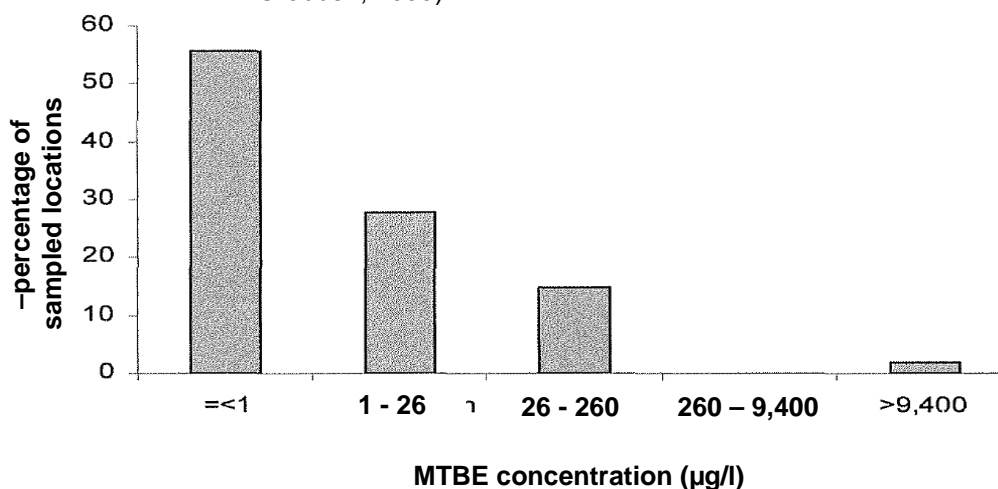
- For ETBE and the other G EO there is no information to support a conclusion, but with regard to their presence in surface water and frequent application of bank filtration, these substances are assumed not to be negligible.

Morgenstein et al (2003) provides some data on groundwater contamination in the Netherlands from the beginning of this decade. The measurements were performed within the framework of drinking water quality measurements.

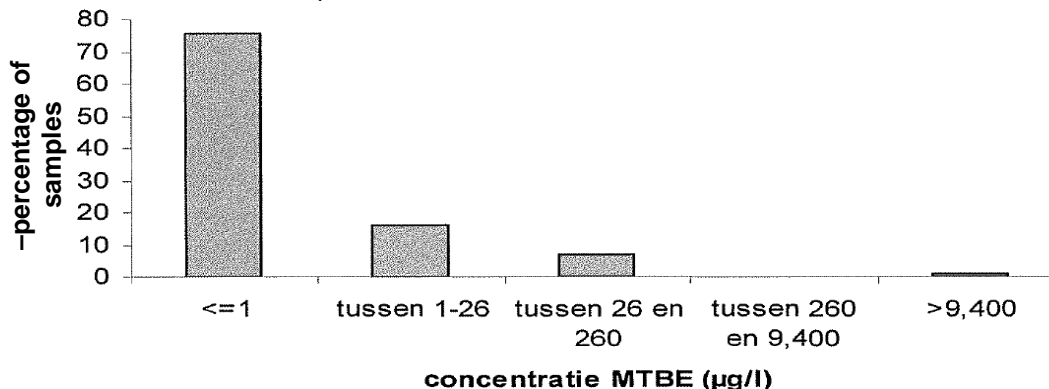
Of 89 samples (taken during two surveying campaigns), 36 samples were below the reporting level of 0.01 µg/l. Thus, MTBE was detected in nearly 60% of all samples. The highest concentration ranged from 0.3 up to 0.41 µg/l, whereas the median concentration was around the detection limit of 0.06 – 0.1 µg/l. The report by RIVM (2002) refers to the same data set and provides a map on the spatial distribution (combined with surface water).

The report by TAUW (Wipfler and Grobber, 2006) shows the results of the investigation of 54 gasoline and 3 depot stations. In total 180 groundwater observation wells were sampled at these stations. **Figure 3.6** and **Figure 3.7** show the results of this investigation.

**Figure 3.6** Distribution of MTBE concentration per location for 54 gasoline stations and 3 depots, monitored by TAUW (Wipfler and Grobber, 2006)



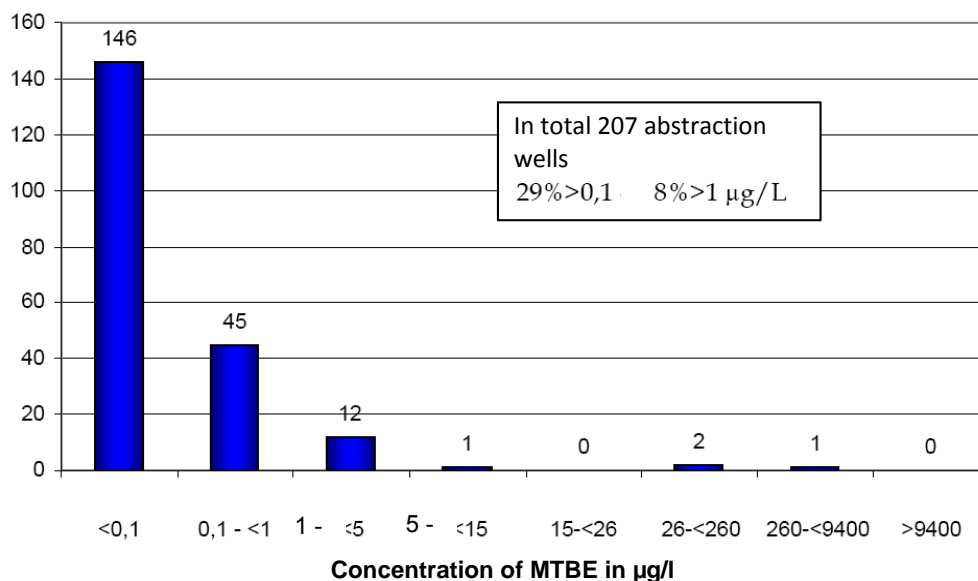
**Figure 3.7** Distribution of MTBE concentration in 180 sampled groundwater observation wells, monitored by TAUW (Wipfler and Grobber, 2006)



The assessment undertaken by TAUW showed that 45% of the gasoline stations investigated had an MTBE concentration in groundwater above 1 µg/l.

The report of Kiwa Water Research (de Voogt et al, 2008) on the presence of MTBE and ETBE in Dutch groundwater refers to groundwater observations at 207 groundwater abstraction wells between May 2002 and September 2007.

**Figure 3.8** Distribution of MTBE concentration in groundwater abstraction wells, taken from de Voogt et al (2008)

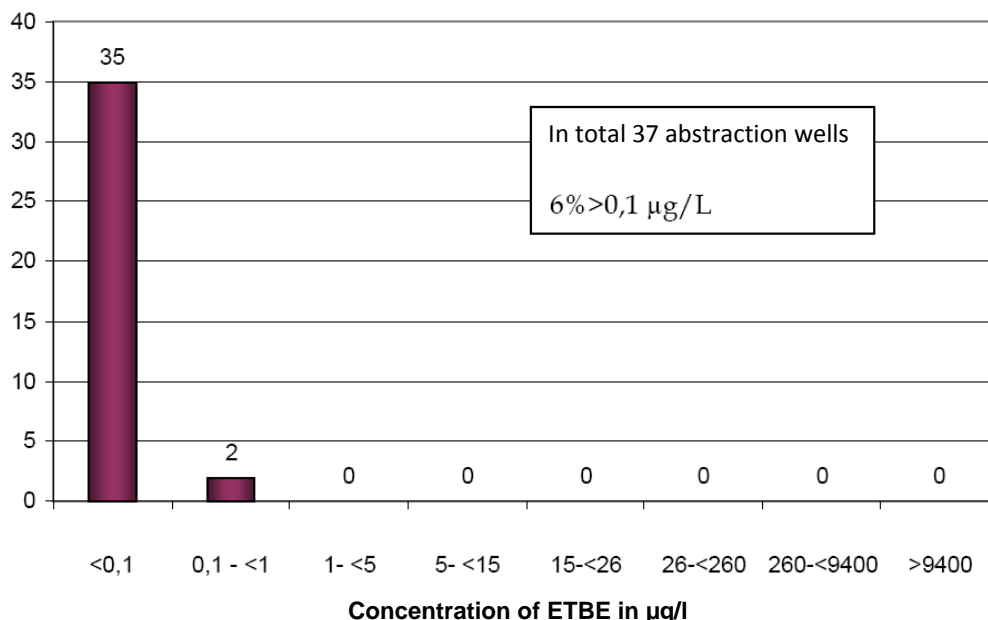


With regard to the different types of aquifers, the results indicate that limestone aquifers and confined aquifers have a much lower percentage of locations with an MTBE concentration above 0.1 µg/l than phreatic aquifers. In comparison, groundwater derived from bank filtrate (in the Netherlands mostly by water from the Rhine and Maas) had a significantly higher MTBE concentration.

For ETBE, the database is smaller. **Figure 3.9** shows the distribution of ETBE originating from 37 abstraction wells.



**Figure 3.9** Distribution of ETBE concentration in groundwater abstraction wells, taken from de Voogt et al (2008)



Regarding the results of de Voogt et al (2008), a clear future trend could not be observed.

**3.2.1.5. Distribution of GEO in groundwater in the UK**

- The majority of the data originates from England and Wales. Except for fuel retail sites, the database for Scotland and Northern Ireland is relatively small. The results from the UK show frequent detection of MTBE at fuel storage facilities, but regional groundwater resources are typically unaffected.
- Recent investigations reveal a significant presence of MTBE at fuel retail sites (65% of all locations sampled with concentrations above 15 µg/l).
- Targeted monitoring at potentially contaminated sites indicates MTBE at 15% of all locations. By contrast routinely monitored (groundwater resource monitoring) locations show an MTBE detection rate of 3.2%.
- Most groundwater samples taken at locations remote from retail filling station sites showed MTBE < detection limit, with a geometric mean of 0.52 µg/l.
- Information on other GEO is locally available.

Results from early investigations

Dottridge et al (2000) reported groundwater monitoring data in the UK from the beginning of the recent decade. In 300 investigations at gasoline retail, storage and distribution sites (99 sites provided by the Institute of Petroleum and 47 sites provided by the Environment Agency), approx. 33% of the sites had MTBE at a detectable concentration. The maximum concentration at different sites was in the range of 1 µg/l up to 800,000 µg/l (i.e., indicative of the presence of pure phase gasoline containing MTBE).

Additionally, regional monitoring data (from the Environment Agency and local water companies) were evaluated with a focus on public water supply wells. Here MTBE was detected at 32 wells (13% of all locations), usually with an MTBE concentration close to the detection limit. Only ten wells (3.9%) had a concentration above 1 µg/l.

Regarding a detailed evaluation of the results, Dottridge et al also report a strong correlation of MTBE detection in water supply boreholes and with areas of high aquifer vulnerability.

Recent investigations by the Energy Institute

The Energy Institute (2009) reported a large collection and evaluation of groundwater quality data for GEO. Based on a large amount of data from Energy Institute member company sites (for details refer to subchapter 3.2.7.1), MTBE and TAME measurements from 524 sites were evaluated. The results are shown in **Table 3.7**. Values for ETBE or DIPE were not reported. Therefore, certain interference is assumed for data used by Dottridge et al (2000) and by the Energy Institute.

**Table 3.7** Distribution of GEO concentration in groundwater at 524 company sites (Energy Institute, 2009)

| Concentration (mg.L <sup>-1</sup> ) | MtBE       |            |            | TAME     |
|-------------------------------------|------------|------------|------------|----------|
|                                     | GC-MS      | GC-FID     | Total      | GC-MS    |
| < detection limit                   | 77 (35%)   | 74 (24%)   | 151 (29%)  | ?        |
| <=0,015                             | 30 (14%)   | 3 (1%)     | 33 (6%)    | 1        |
| 0,015 to 0,05                       | 24 (11%)   | 34 (11%)   | 58 (11%)   | 2        |
| 0,05 to 0,1                         | 9 (4%)     | 23 (8%)    | 32 (6%)    | 1        |
| 0,1 to 1                            | 42 (19%)   | 87 (29%)   | 129 (25%)  | 2        |
| 1 to 10                             | 27 (12%)   | 61 (20%)   | 88 (17%)   | 2        |
| 10 to 100                           | 10 (5%)    | 21 (7%)    | 31 (6%)    | 1        |
| >100                                | 1 (0,5%)   | 1 (0,3%)   | 2 (0,4%)   | 1        |
| <b>Total</b>                        | <b>220</b> | <b>304</b> | <b>524</b> | <b>?</b> |

Notes

1. TAME is not routinely analysed for at oil company sites. The number of sites where TAME was analysed for but not detected is unknown.
2. % refers to % of total for that column.

MTBE was detected at 373 retail filling station sites (71%). 340 sites (65%) had a concentration above 15 µg/l. The issue for samples probably falsely classified as positive (below 50 µg/l for GC-FID samples) is explained in subchapter 3.2.7.1. Regarding the different transport and mobility of GEO in groundwater and soil, it is noteworthy that 43% of the positive detect sites (for groundwater) did not show any detectable MTBE in soil samples.

TAME was detected in 10 groundwater samples, 9 of them with a concentration between 11 to 115,486 µg/l (hot spot). Due to the missing total number of TAME samples, it is not possible to derive a general TAME distribution at the retail filling station sites.

For 25 of the 524 sites, a measurable petrol phase – LNAPL was reported. The geometric mean of the maximum detected concentration at these sites was

3,100 µg/l for MTBE and 12,200 µg/l for TAME – indicating a clear correlation between the presence of petrol - NAPL and high levels of groundwater contamination by GEO.

Additionally, it was reported that the presence of soakaways at the sites does not seem to have a general influence on the contamination level. Contrary to the findings by Dottridge et al (2000), no significant correlation between the contamination level and vulnerability of the aquifer was observed.

Apart from the retail filling station data, the report of the Energy Institute evaluates a large amount of collected data provided by the Environment Agency of England and Wales and the Geological Survey of Northern Ireland. The results are summarised in **Table 3.8**. It has to be noted that early parts of the evaluated data were already used by Dottridge et al (2000).

**Table 3.8** Results of groundwater monitoring for GEO in the UK (regulatory data) as reported by the Energy Institute (2009)

| Substance                                | Scotland   | Northern Ireland | England and Wales                         |                        |   |                    |
|--|------------|------------------|---|------------------------|---|--------------------|
|  | MTBE       | MTBE             | MTBE                                      | ETBE                   | TAME  | DIPE               |
| Observation period                       | since 2005 | 2000 - 2006      | 1990 – 2006                               | 2005 – 2006 (majority) |   | 1997/98 (majority) |
| No. of sampling locations                | 23         | 106              | 2,566 <sup>1</sup> / 771 <sup>2</sup>     | 2,226                  | 2,226   | 14                 |
| No. of Samples in total                  | 52         | 513              | 6,885 <sup>1</sup> / 2,415 <sup>2</sup>   | 3,661                  | 3,661   | 96                 |
| Locations > limit of detection (%)       | 0          | 0.97             | 3.2 <sup>1</sup> / 15 <sup>2</sup>        | 0                      | 0.19<br>0.16 <sup>1</sup> / 0.02 <sup>2</sup> | 0                  |
| Detection limit (µg/l)                   | 1          | 0.2 – 10         | 0.1 – 100<br>majority 0.5 since 2003      | 0.1 - 1                | 0.1 - 1                                       | 50 (majority)      |
| Max. value (µg/l)                        | 0          | detection limit  | 3,900 <sup>1</sup> / 103,000 <sup>2</sup> | 0                      | 0.184 <sup>1</sup> / 117 <sup>2</sup>         | 0                  |
| Geometric mean value of positive samples | ---        | ---              | 0.52 <sup>1</sup> / 5.3 <sup>2</sup>      | ---                    | not reported                                  | ---                |

<sup>1</sup> ... for routine monitoring of groundwater resources

<sup>2</sup> ... for targeted investigations at potentially contaminated sites

Based on the large number of MTBE data, the Energy Institute reported additional evaluations regarding the aquifer types and geographical distribution in the UK.

For the distribution of MTBE in different types of aquifers, no significant difference could be observed. The geographical distribution indicated a higher detection of MTBE in urban areas than rural areas, as expected. Additionally, a correlation between the occurrence of MTBE in routinely monitored wells and proximity to petrol stations was suggested (Energy Institute, 2009).

### 3.2.1.6. Distribution of GEO in groundwater in Denmark

- The available information on GEO in groundwater is relatively small.
- The distribution of MTBE on contaminated / potentially contained sites indicates a range that is comparable with the findings for Germany, the Netherlands and UK.
- The information on background concentrations for MTBE is too small to derive a general estimation.
- Information for GEO other than MTBE could not be identified.

Schmidt et al (2002) reported background MTBE concentrations for Denmark. Results from 1997 reveal 7 samples out of 22 in total with positive detection. Here, the values range from 0.1 to 0.6 µg/l. These samples are not assumed to be representative for the background. At least 10 additional samples with 1 – 5 µg/l were assumed to be under direct influence of a contamination or point source.

Results from 2000 for the County of Funen reveal 7 samples which detected as positive out of 20 in total. Here, the values range from 0.1 to > 30 µg/l. Further background information on this is not reported (Schmidt et al, 2002).

In the report of the Ministry of the Environment (Miljøstyrelsen, 2003), data from 43 contaminated and 7 potentially contaminated sites are collected. For 40 sites, the MTBE concentration in groundwater at the contamination source is provided. Here the values range from 0.22 to 530,000 µg/l. On 8 sites, values >1,000 µg/l were observed. The mean value was calculated as 2,900 µg/l, the median value observed was 44.5 µg/l.

For 9 sites, the concentration at the next downstream observation well was presented. Here, the values range from 0.22 to 290,000 µg/l. On 3 sites, values >1,000 µg/l were observed. The mean value was calculated as 4,590 µg/l, whereas the median value observed was 340.5 µg/l.

#### Key message

The available information on GEO in groundwater is relatively small and dominated by data on suspect sites. The distribution of MTBE at contaminated or potentially contained sites is of a similar magnitude and frequency as the data reported for Germany, the Netherlands and UK.

The information on background concentrations for MTBE is too limited to derive a general estimation. Information for other GEO than MTBE could not be identified.

### 3.2.1.7. Distribution of GEO in groundwater in Finland

- The Finnish results reported by SYRE (2009) showed a wide range in distribution for MTBE and TAME at contaminated sites.
- A comparable high MTBE and TAME concentration was observed at the potentially impacted sites, which are probably connected with the significantly higher MTBE and TAME content in Finnish unleaded petrol.
- The results for the background measurements indicate no general distribution of MTBE and TAME.

- In general, TAME is present at lower contaminations in groundwater than MTBE.
- Data on GEO other than MTBE and TAME could not be obtained.
- Limited baseline groundwater monitoring data indicates 85% of groundwater had no detectable MTBE, and the maximum concentration of 1.4 µg/l is well below the USEPA taste and odour threshold of 20 µg/l.
- TAME was not detected in any baseline groundwater monitoring survey.

The report by the Finnish Environmental Agency (SYRE, 2009) regarding the risk of MTBE and TAME in Finland provides a wide range of data on the distribution of MTBE and TAME.

Based on several investigations on different sites (e.g. gasoline stations) and sites that are assumed to be in the vicinity of a potential source, data of 398 MTBE investigations and 287 TAME investigations were collected and evaluated. The data set is summarized in **Table 3.9**.

**Table 3.9** Monitoring results for MTBE and TAME in Finland (SYRE, 2009)

| Concentration (µg/l) for MTBE / TAME |      |         |      |          |      |             |      |         |      |       |      |  |
|--------------------------------------|------|---------|------|----------|------|-------------|------|---------|------|-------|------|--|
| < 15                                 |      | 15 – 40 |      | 40 – 100 |      | 100 – 1,000 |      | > 1,000 |      | total |      |  |
| MTBE                                 | TAME | MTBE    | TAME | MTBE     | TAME | MTBE        | TAME | MTBE    | TAME | MTBE  | TAME |  |
| <b>Region of Uusimaa</b>             |      |         |      |          |      |             |      |         |      |       |      |  |
| 102                                  | 68   | 5       | 4    | 3        | 3    | 10          | 1    | 10      | 4    | 130   | 80   |  |
| <b>Region of Häme</b>                |      |         |      |          |      |             |      |         |      |       |      |  |
| 121                                  | 135  | 14      | 4    | 8        | 1    | 10          | 4    | 10      | 3    | 163   | 147  |  |
| <b>Region of North Karelia</b>       |      |         |      |          |      |             |      |         |      |       |      |  |
| 41                                   | 18   | 32      | 32   | 6        | 2    | 14          | 6    | 12      | 2    | 105   | 60   |  |
| <b>Total</b>                         |      |         |      |          |      |             |      |         |      |       |      |  |
| 264                                  | 221  | 51      | 40   | 17       | 6    | 34          | 11   | 32      | 9    | 398   | 287  |  |

The data indicates that the majority (66.3%) of the sites had concentrations of these GEO below 15 µg/l. Aside from this; a number of sites with higher MTBE and TAME concentrations were noted, especially in the region of North Karelia. In total, 33.7% (MTBE) and 23% (TAME) of the investigation values were above 15 µg/l. In comparison with other countries, this data reflects a higher contamination level, which might result from the higher percentage of GEO in Finnish petrol. Alternatively, these observations could be a result of the sampling and analysis strategies used.

Apart from the investigation of potentially contaminated sites, background values for groundwater were also reported. Therefore, observations at 10 locations with 19 samples for MTBE and TAME in two campaigns (autumn 2003/spring 2004) were evaluated.

As a result, 16 samples at 8 stations had an MTBE concentration below the limit of detection (range: 0.1 – 0.2 µg/l). Only three samples at the two locations, Kukkila - Hollola / Kunnas kaivo 2 –Lahti, had MTBE concentrations of 0.2 / 0.48 and 1.4 µg/l.

For TAME, none of the 19 samples exceeded the detection limit (range: 0.1 – 0.2 µg/l).

**3.2.1.8. Additional findings for GEO in groundwater of other European Countries**

- With reference to the additional findings for groundwater in Italy, Belgium and Sweden it was noted that these results are within a comparable order of magnitude to the findings of countries like Austria, Switzerland, the UK, The Netherlands and Germany. Thus, certain general parallels might be assumed.
- Investigations at sites that store or distribute gasoline frequently identify MTBE and (less frequently) other GEO.
- Sampling of regional groundwater resources indicates either no detection, or very low concentrations (generally <1 µg/l) at the large majority of sites. This concentration is well below the relevant taste and odour thresholds.

Regulator data of Italy

Two data sets were provided from the region of Bologna for MTBE and ETBE for 2008. These observations are summarized in **Table 3.10**:

**Table 3.10** Results of GEO monitoring in Bologna

| Substance                          | 1 <sup>st</sup> data set       |      | 2 <sup>nd</sup> data set |      |
|------------------------------------|--------------------------------|------|--------------------------|------|
|                                    | MTBE                           | ETBE | MTBE                     | ETBE |
| Observation period                 | 2008                           | 2008 | 2008                     | 2008 |
| No. of sampling locations          | 39                             | 39   | 41                       | 41   |
| No. of samples in total            | 63                             | 63   | 75                       | 75   |
| Locations > limit of detection (%) | 2.65                           | 0    | 4.9                      | 0    |
| Detection limit (µg/l)             | 0.5                            | 0.5  | 0.1                      | 0.1  |
| Max. value (µg/l)                  | 0.9                            | 0    | 0.2                      | 0    |
| Median value of positive samples   | 0.9<br><i>(one value only)</i> | 0    | 0.15                     | 0    |

Regulator data of Ireland

A data set for MTBE measurements carried out from May to November 2009 was provided. In total 504 samples at 263 locations were analysed. None of the samples showed a concentration above the named detection limit (range 0.5 – 2 µg/l).

Data from two gasoline stations in Belgium

No regional groundwater quality monitoring data was available for Belgium. Two data sets for gasoline stations with MTBE groundwater contamination were provided.

The first station is located in the area of Lommel. In total 10 groundwater samples from 6 observation wells at the site were analysed for MTBE. The concentration was between 300 and 25,000 µg/l. The mean value was calculated as 4,835 µg/l, whereas the median value was 2,603 µg/l. TBA was analysed in 4 of the observation wells. Three values did not exceed the limit of detection, but one value was 93 µg/l.

The second station is located in the area of Zammel. A total of 6 groundwater samples from 4 observation wells at the site were analysed for MTBE. The concentration was between 65 and 16,576 µg/l. The mean value was calculated as 5,779 µg/l and the median value was 4,017 µg/l. Groundwater from 2 of the observation wells was analysed for TBA. The well with the highest MTBE concentration had a TBA concentration of 3,013 µg/l. The other well sampled had a TBA concentration of 650 µg/l.

#### Three groundwater measurements in the scope of the national screening in Sweden

The results of 3 selected groundwater measurements are reported in Potter et al (2009) in the scope of the National Screening Programme of Sweden in 2008.

The first sample (Lerum / Skallsjö Ängar – a municipal water source with heavy traffic nearby) had a MTBE concentration of 0.014 µg/l. The value for ETBE was below the detection limit (<0.007 µg/l).

In the second sample (Lerum / natural spring, influenced only by airborne deposition), MTBE was below the detection limit (<0.010 µg/l). The value for ETBE was also below the detection limit (<0.007 µg/l).

The third sample (Stockholm / Ulvsunda, urban site) showed a MTBE concentration of 0.24 µg/l. The value for ETBE was 0.031 µg/l.

### **3.2.2. Literature Review and results of enquiry – GEO in drinking water**

- Drinking water quality in Europe is of a very high standard.
- No MTBE concentrations in drinking water above the USEPA odour threshold of 20 µg/l were reported.
- MTBE has been detected in some European drinking water - mainly in trace concentrations (range 0.01 to 0.2 µg/l).
- Compared with groundwater or surface water, the available knowledge in the published literature of GEO in drinking water is relatively low.
- A good amount of information was available for Germany, the Netherlands, UK and partly for Sweden. Additionally, a smaller amount of information was available for Switzerland and France.
- The detection frequencies range from approx. 30% (Netherlands) to approx. 40% (Germany).
- Typically, the major source for MTBE in drinking water is raw water that originates from surface water.

- Further information on GEO other than MTBE could not be obtained for drinking water.

### 3.2.2.1. Overview of the behaviour of GEO at typical drinking water treatment steps

In Germany the Water Technology Center (Technologiezentrum Wasser, TZW) performed a couple of investigations in the scope of the DVGW research program for the impact of MTBE on drinking water (reported in LFU, 2002 and Baus et al, 2008).

For the behaviour of GEO in typical steps of drinking water treatment, the following aspects were reported (LFU, 2002 and Baus et al, 2008):

- For **river bank filtration**, only a low sorption and a slow microbial degradation is evident, which is probably related to the physical and chemical properties of MTBE, ETBE and TAME. Thus, GEO are only partly reduced in concentration and, if present in river water can persist in the drinking water to a certain extent.
- **Air Stripping** of GEO is generally possible. DIPE, TAME and ETBE show a slightly better removal than MTBE due to their lower Henry's Law coefficients. In practice it is much more challenging to strip GEO than BTEX or light volatile chlorinated hydrocarbons (e.g. tetra- and trichloroethene and their decay products), due to the demand for a much higher water - air ratio and larger columns.
- **Chemical Oxidation** of GEO by ozone up to ozone concentrations of 1 mg/l does not provide sufficient GEO elimination. An advanced oxidation, e.g. a combination of ozone with hydrogen peroxide or UV-radiation leads to a significant increase in the removal rate.
- **Adsorption on activated carbon** is a possible treatment method for GEO reduction and is partly used (mostly due to the parallel adsorption of other compounds). With regard to the relatively low adsorption capacities, GEO removal by activated carbon requires a large amount of activated carbon which may make the technique less sustainable.
- **Membrane filtration** is a possible treatment process that principally achieves good elimination rates for GEO. In practice, a very large membrane surface is necessary.

### 3.2.2.2. Distribution of GEO in drinking water in Germany

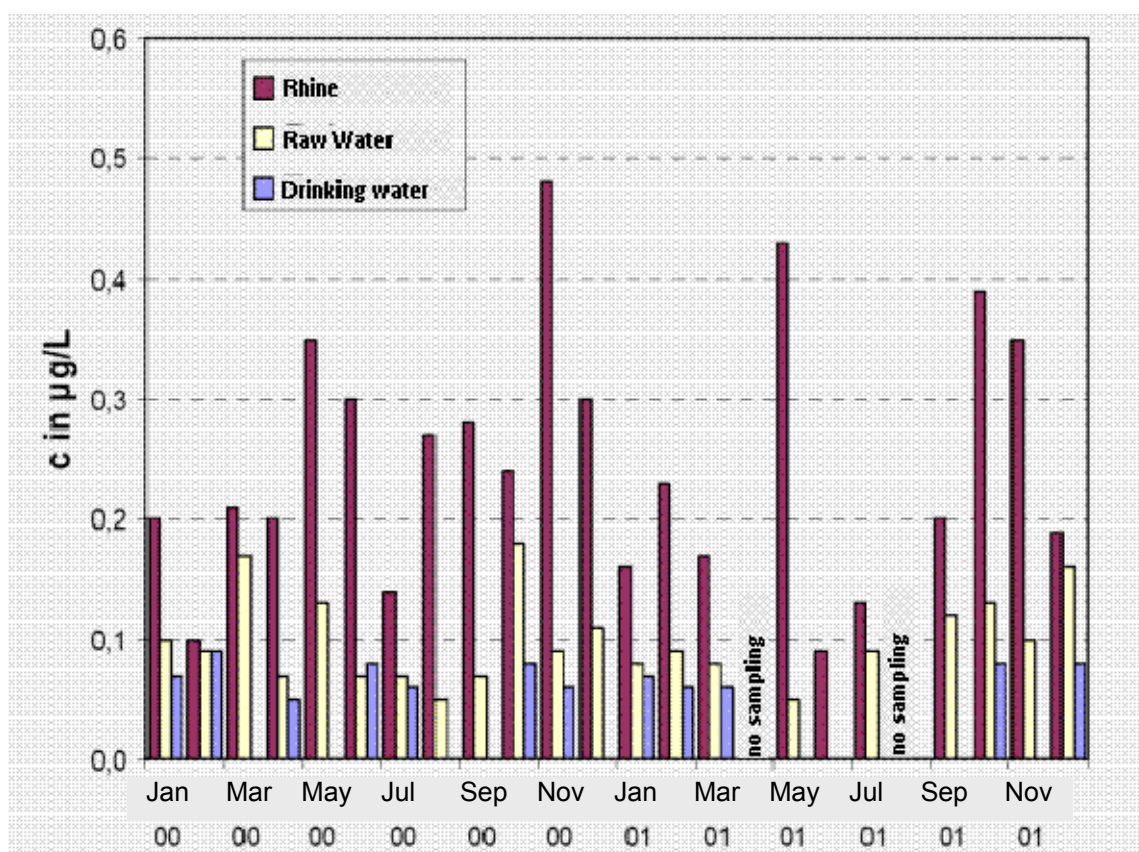
- The German drinking water results indicate a wide distribution and detection frequency of MTBE for Germany: in total 40% (Kolb, 2004).
- When detected, MTBE is usually present in trace concentrations (0.01 to 0.1 µg/l).
- The measured MTBE concentrations in drinking water did not exceed the thresholds for odour and taste.
- The major source of MTBE in drinking water is contaminated groundwater and contaminated surface water that has been used as a raw water source.



- Traditional treatment processes for drinking water sanitation do not completely eliminate MTBE.
- There is insufficient information on the distribution of other GEO in German drinking water to draw any conclusions.

In LFU (2002), the TZW reported an example of a drinking water treatment plant on the River Rhine that uses bank filtrate as a raw water source. The 2000/01 monitoring data for Rhine, raw water and drinking water are shown in **Figure 3.10**.

**Figure 3.10** Comparison of the MTBE concentration in the Rhine, the extracted raw water (river bank filtrate) and produced drinking water for a selected drinking water plant at the Rhine (after LFU, 2002)

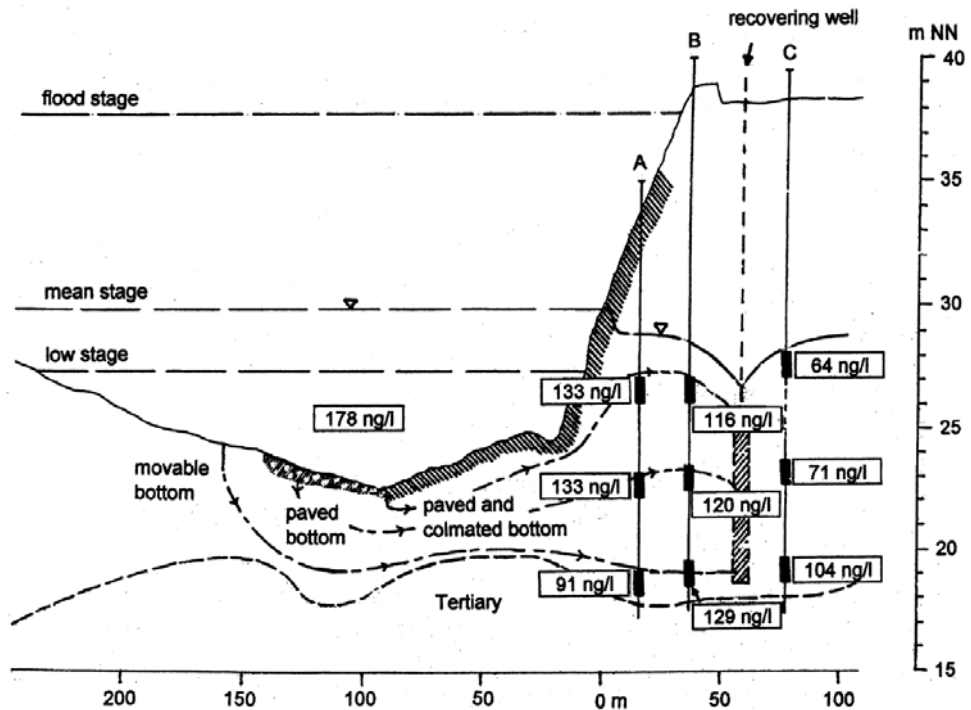


The comparison between the Rhine and the raw water indicates that only part of the MTBE load was attenuated by the bank filtration. Some of the MTBE load also passed the further treatment steps. Thus, the drinking water contained MTBE in the range of 0.04 to 0.09 µg/l.

Detailed investigations on the behaviour of MTBE during transport from the river bank to the Rhine were presented by Achten et al (2002c). Here, the river and three monitoring wells at different distances to the abstraction well were analysed. The results (one example from the three investigated sites) are shown in **Figure 3.11**.

In this study, the MTBE concentration was monitored at two riverbank filtrate water works. The measured concentration ranged from between 0.04 µg/l and 0.06 to 0.11 µg/l.

**Figure 3.11** Hydrogeological cross section through the River Rhine and river bank at the Lower Rhine, including the observed MTBE concentration (ng/l), from Achten et al (2002c)

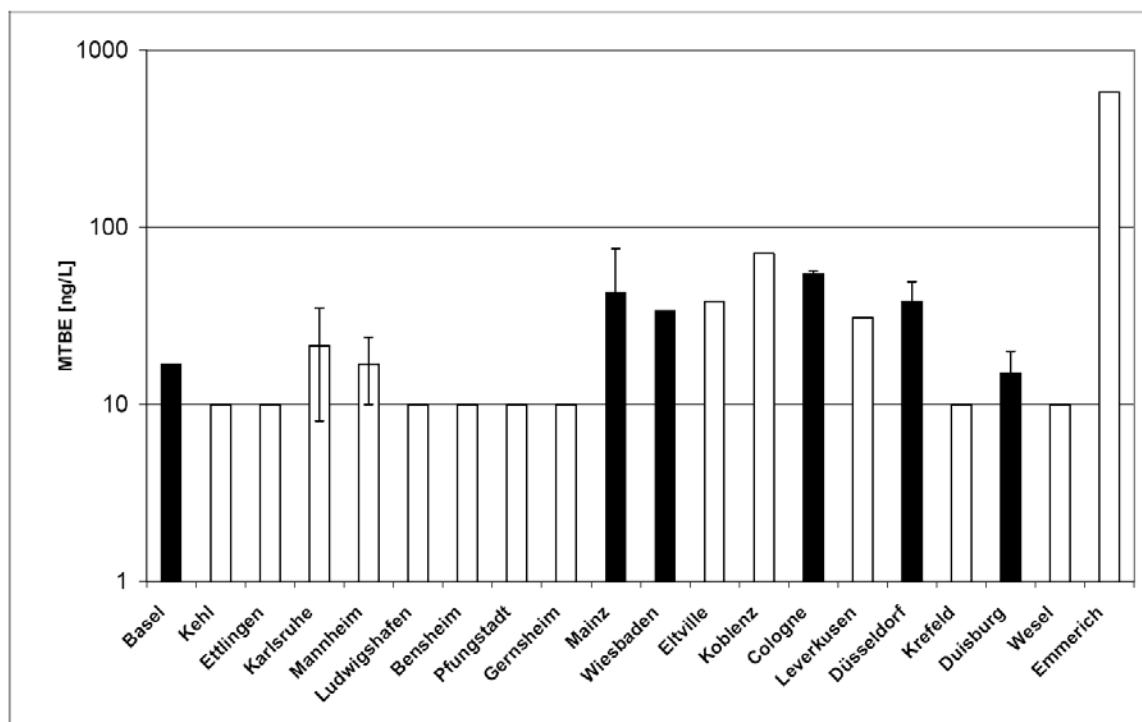


Intensive investigations on German drinking water were published by Kolb (2004) and Kolb & Püttmann (2006b). In these publications, 83 drinking water samples from the public drinking water supply of 50 cities were sampled and analysed for MTBE between November 2000 and September 2003. The detection limit was 0.01 µg/l. Most of the cities are located along the Rhine in Germany. Additionally, bigger cities like Hamburg, Berlin, Munich, Stuttgart, Dresden, Leipzig, Halle, Dortmund and Münster as well as a couple of smaller cities were included. A complete list of all samples is given in Kolb (2004).

As a result, MTBE was detected in 38 samples, with a concentration ranging from 0.017 to 0.712 µg/l. The detection frequency was 40% (20 of the 50 sampled public drinking water supplies). The median of all values above the detection limit was calculated as 0.038 µg/l; the mean value of these samples was 0.089 µg/l.

A special situation was observed for Saxony-Anhalt. The drinking water samples had a detection frequency of 60% and a median concentration of 0.112 µg/l (range of 0.063 - 0.712 µg/l). The highest MTBE concentration was measured in samples from Leuna and Spergau. This high MTBE level is probably connected with a MTBE groundwater contamination at a former production facility in Leuna that produced MTBE over a long time. The groundwater contamination also affects the river Saale that is related to the raw water sources.

**Figure 3.12** Observation of MTBE in public drinking water supplies in cities along the Rhine in (ng/l), taken from Kolb (2004)



The measured MTBE concentration in the public drinking water supply from cities along the Rhine is shown in **0**. The results marked in black represent drinking water that partly originates from bank-filtrated or artificially infiltrated Rhine water. In the geographical region of the upper Rhine valley MTBE could be detected only in samples originating from Basel, Karlsruhe and Mannheim, with a concentration of 0.017, 0.022 and 0.017 µg/l, respectively. In Basel, the groundwater used to provide drinking water contains artificially infiltrated Rhine water. The drinking water of Karlsruhe and Mannheim does not originate from artificially or bank-filtrated Rhine water.

In the geographical region from Mainz to Emmerich, MTBE was detected in all but two (Krefeld and Wesel) drinking water samples. Here, the concentration ranged from 0.015 µg/l in Duisburg to 0.582 µg/l in Emmerich. The sample from Emmerich had the highest MTBE concentration of all samples taken at the Rhine cities.

The remaining drinking water supplies between Mainz and Emmerich had, detectable concentrations below 0.1 µg/l. The drinking water of Mainz, Wiesbaden, Cologne, Düsseldorf and Duisburg is at least partly composed of groundwater affected by Rhine water. The cities of Leverkusen, Krefeld and Wesel do not use raw water that has been influenced by the Rhine.

MTBE was detected in 55% of drinking water supplies along the River Rhine that were sampled for MTBE. The median value was 36 ng/l (0.036 µg/l), the mean value was 69 ng/l (0.069 µg/l).

### 3.2.2.3. Distribution of GEO in drinking water in the Netherlands

- The Dutch results show that MTBE has been detected in drinking water.
- MTBE was detected in raw and treated water used for potable supply with a frequency of about 35%.
- An MTBE concentration above the Dutch “indication value” of 1 µg/l rarely occurs.
- The highest MTBE concentration occurs in raw water and drinking water derived from surface water.
- Groundwater and bank filtrate have lower MTBE concentrations.
- The reported MTBE concentration in drinking water did not exceed the thresholds for odour and taste.
- There is insufficient information on the distribution of other GEO in Dutch drinking water to draw any conclusions.

The publication by RIVM (2002) reports monitoring data for MTBE in Dutch drinking water. Within the first campaign in June/July 2001, 62 locations were sampled; 35% of these locations had an MTBE concentration above the detection limit of 0.01 µg/l. The average concentration was reported as 0.07 µg/l, with the highest observed value as 0.42 µg/l (raw water source: surface water). By splitting the results with regard to the raw water source, the following average results were reported:

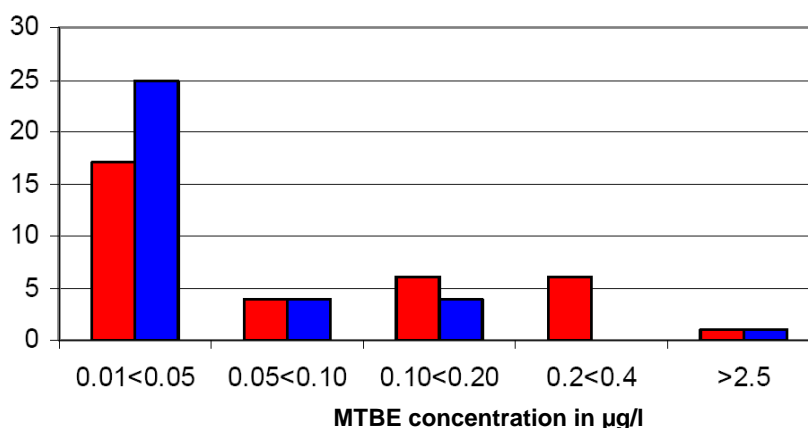
- Surface water: 0.14 µg/l
- Bank filtrate: 0.03 µg/l
- Groundwater: 0.06 µg/l

In the second campaign in September/October 2001, 45 locations were sampled for raw water and drinking water. The average concentration for raw water was 0.13 µg/l, with a highest observed value of 3.2 µg/l (raw water source: surface water, Lekkanaal by Nieuwegein). By splitting the raw water results with regard to the water source, the following average results were reported:

- Surface water: 0.47 µg/l
- Bank filtrate: 0.08 µg/l
- Groundwater: 0.05 µg/l

For drinking water, the average MTBE concentration was 0.09 µg/l (highest value of 2.9 µg/l). **Figure 3.13** provides an overview of the statistical distribution of the single values for raw water and drinking water:

**Figure 3.13** Results of the second observation campaign for raw and drinking water observations (RVIM, 2002)



Monitoring data for MTBE in drinking water was evaluated in the publication of VEWIN (2005). The drinking water from 11 locations investigated originated from surface water. In total 356 water samples were analysed. For 5 of the 11 locations, the median value for drinking water was below the limit of detection (< 0.02 to < 0.05 µg/l). The other locations showed a median concentration of 0.04 – to 0.14 µg/l. The maximum observed concentration ranged from 0.53 to 0.59 µg/l.

The publication of Morgenstern et al (2003) referred to the same investigations as RVIM (2002). Morgenstern et al (2003) also evaluated the correlation of MTBE concentration between raw water and the corresponding drinking water for the second investigation campaign. They estimated a coefficient of  $R^2 = 0.903$ .

With regard to the above mentioned second observation campaign Morgenstern et al (2003) also reported examples for MTBE removal in different treatment steps. These results are presented in **Table 3.11**.

**Table 3.11** Results for MTBE reduction in different treatments step for drinking water formation (Morgenstern et al, 2003)

| Water source  | MTBE concentration in µg/l |             |              |                   |                |
|---------------|----------------------------|-------------|--------------|-------------------|----------------|
|               | Raw water                  | Ozonization | Chlorination | Carbon filtration | Drinking water |
| Surface water | 0.027                      | 0.013       | ---          | 0.010             | 0.010          |
| Surface water | 0.019                      | ---         | ---          | 0.010             | 0.014          |
| Surface water | 0.025                      | ---         | 0.020        | 0.030             | 0.028          |
| Surface water | 0.030                      | 0.020       | ---          | 0.030             | 0.025          |
| Bank filtrate | 0.050                      | 0.030       | ---          | 0.010             | 0.010          |
| Bank filtrate | 0.033                      | 0.030       | ---          | 0.030             | 0.025          |

These results show that conventional drinking water treatment measures are able to reduce the MTBE concentrations to a certain extent, if MTBE is present in trace concentration within the raw water.

Results of drinking water quality monitoring

The report on the Dutch drinking water quality by VROM (2006) refers to drinking water observations between 2001 and 2005. In the scope of this report, 149 locations for raw water and 148 locations for drinking water were investigated, with 1,742 MTBE samples in total. Consequently, a total of 66 public drinking water distribution areas in the Netherlands were observed.

An MTBE concentration above 1 µg/l (Dutch indication value – compare to RVIM, 2004) was detected in 5 sampled locations; 4 belong to surface water as raw water source and one location refers to groundwater. The highest observed value was 7.3 µg/l.

A special situation was reported for the Heel location. The raw water of this water works originates from the Lateraalkanaal (river Maas). Here, the water abstraction was stopped 19 times in 2005 (in all for 144 days) due to spills by a pipeline in an industrial area nearby. The criterion for stopping drinking water abstraction was set at an MTBE concentration of 5 µg/l.

**3.2.2.4. Distribution of GEO in drinking water in Sweden**

In the scope of the Swedish National Screening Programme, 3 samples of municipal drinking water and 3 samples from domestic wells were analysed for MTBE and ETBE (Potter et al, 2009). The results of these investigations are summarized in **Table°3.12**.

**Table 3.12** Results of the Swedish drinking water observations – from Potter et al (2009)

| Type          | Site                  | Unit | MTBE | ETBE | Benzene | Toluene | Ethylbenzene | m+p-Xylene | o-Xylene |
|---------------|-----------------------|------|------|------|---------|---------|--------------|------------|----------|
| Municipal     | Göteborg, Södra vägen | ng/l | 34   | <7.0 | <6.0    | 26      | 1.2          | 4.1        | 2.9      |
| Municipal     | Göteborg, Kärra       | ng/l | <10  | <7.0 | <6.0    | 8.7     | 0.21         | 3.0        |          |
| Municipal     | Kungshamn             | ng/l | <10  | <7.0 | 8.3     | 20      | 1.6          | 7.2        | 1.6      |
| Domestic well | Lerkil 1              | ng/l | 37   | 7.9  | 13      | 18      | 2.2          | 6.4        | 6.2      |
| Domestic well | Lerkil 2              | ng/l | 140  | <7.0 | 7.5     | 7.0     | 0.54         | 2.3        | 1.5      |
| Domestic well | Stångenäs             | ng/l | 34   | <7.0 | <6.0    | 9.4     | 1.5          | 2.0        |          |

The sample in Göteborg Södra Vägen had an MTBE concentration of 0.034 µg/l. As the results for the BTEX compounds analysed at the same time indicate a potential petrol influence is probable for all 6 drinking water samples; MTBE was detected in two samples, but ETBE in one sample only.

**3.2.2.5. Distribution of GEO in drinking water in the UK**

The Energy Institute (2009) reports regular drinking water observations for MTBE by 4 water companies (out of 26 water companies that replied to the query). These companies sampled water mainly from groundwater and partly from surface water sources.

The reported observation frequencies differ depending on the companies and the observed type of water (raw water or treated water) – weekly, monthly, 3-monthly

and annual bases are mentioned. The reported level of detection ranges from 0.06 to 0.5 µg/l.

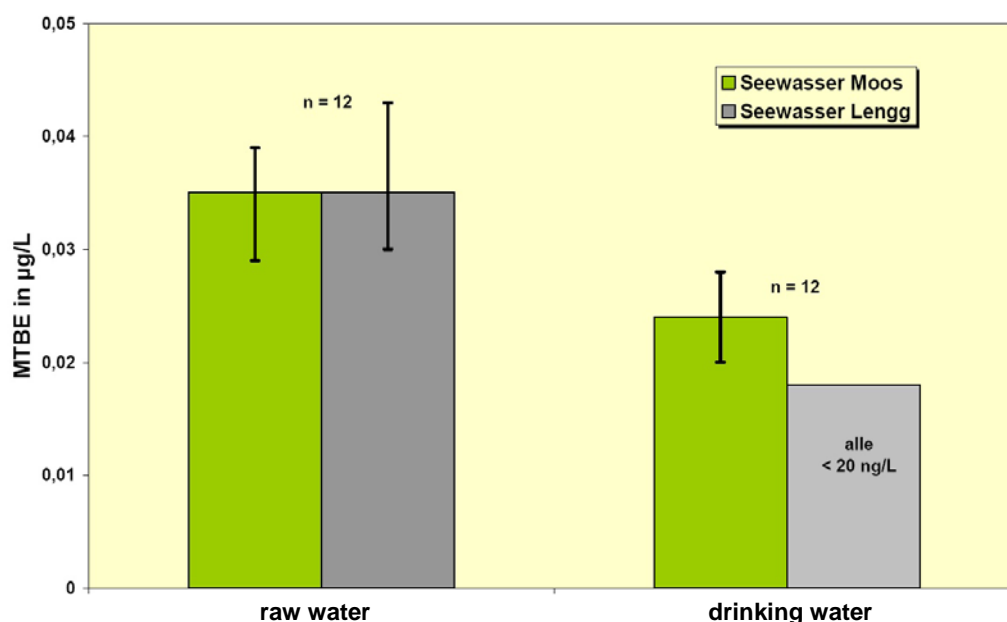
One of the water companies reports raw water values with higher concentrations in three wells (12.29 µg/l / 3.36 µg/l / 0.9 µg/l) for a short period (ranging from one to four months), probably caused by local incidents. Furthermore, the data of 3 water companies regarding 52 groundwater supply wells (51 well fields) indicated concentrations above 0.5 µg/l for 6% of all well fields.

**3.2.2.6. Additional findings for GEO in drinking water of other European Countries**

MTBE in drinking water originating from Lake Zurich (Switzerland)

Baus et al (2008) reported on the drinking water produced from Lake Zurich for the water works at Moos and Lengg in the year of 2005. The results are shown in **Figure 3.14**.

**Figure 3.14** MTBE in the raw water and the drinking water that originates from the lake Zurich - from Baus et al (2008)



MTBE in drinking water at one site in France

The risk assessment undertaken by the European Commission (2002) reported a detection of MTBE in drinking water in France at one site. Here, the average concentration is reported as 0.83 µg/l.

**3.2.3. Literature Review and results of enquiry – GEO in surface water**

- The evaluation of the literature and local enquiries show that most of the data are from larger rivers and their main tributaries. For lakes, however, the amount of published data is much lower.

- There is relatively good data coverage for the Netherlands, Austria, Switzerland, Germany and UK.
- The information on the distribution of GEO in surface waters in other European countries is limited. The documented results from these countries show that GEO have been reported in surface water.
- Intense investigation on large rivers and lakes showed a background GEO concentration from below the detection limit up to ~0.5 µg/l.
- A significant difference between rural and urban catchment areas was evident.
- For lakes and rivers seasonal effects (e. g. due to boating or high and low water levels) were apparent.
- The observed levels for MTBE are mostly higher than those for ETBE, but a slight tendency for an increase of ETBE was also observed (especially in the Netherlands).
- Higher GEO concentrations are usually related to spill events from tankships and sporadic releases at industrial facilities on land. This is especially important for the River Rhine.
- A dense observation network along the Rhine (including Switzerland, Germany and the Netherlands) is operated, leading to a comprehensive characterisation of release events for MTBE and ETBE.

The following chapter provides an overview of the distribution of GEO in European surface water bodies. The evaluation of the literature and local enquiries show that most of the data are available for larger rivers and their main tributaries. For lakes, however, the amount of published data is much lower.

#### 3.2.3.1. Distribution of GEO in Austrian rivers

- Existing results show the presence of MTBE in a number of Austrian rivers.
- MTBE is mainly found in the larger rivers, rather than in smaller rivers in rural areas.

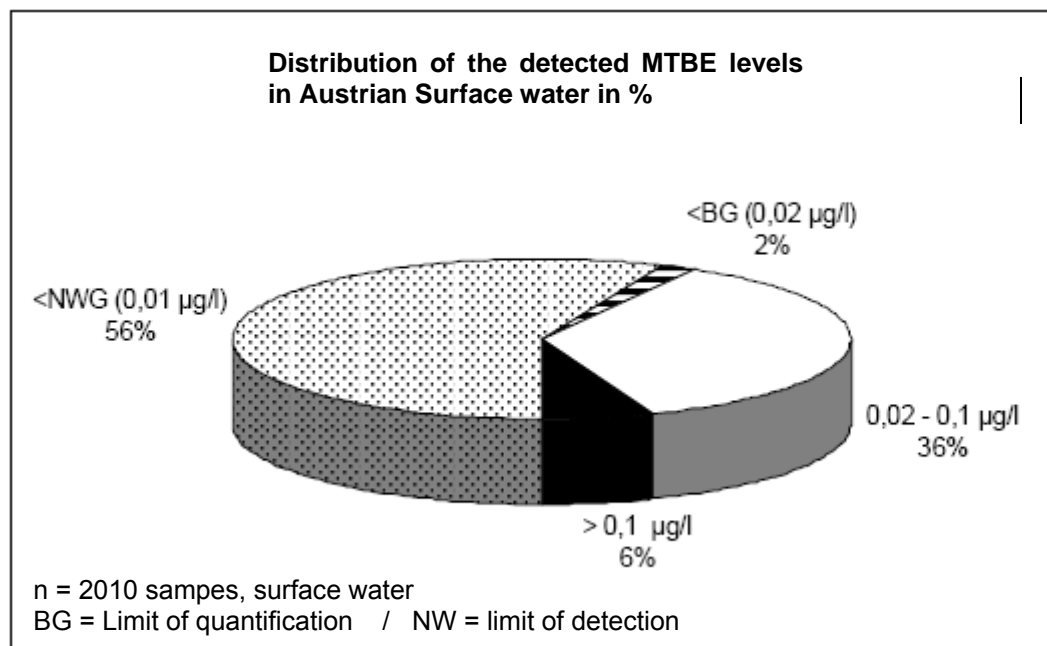
Based on a special monitoring programme, Austria started monitoring for GEO in surface water. The Federal Environmental Agency of Austria (2002) published the first results within a report on the investigation of water quality, reporting on 5 samples taken from the rivers Danube, Inn and Salzach. All these samples had a MTBE concentration above the analytical detection limit in the range 0.019 to 0.11 µg/l (median 0.038 µg/l).

In the following years, monitoring was intensified and the Federal Environmental Agency of Austria (2004) published another report on the water quality, including surface water monitoring based on 2,010 water samples collected at 204 locations.

**Figure 3.15** provides an overview of the MTBE concentration detected in these samples.



**Figure 3.15** Summarized results for the presence of MTBE in Austrian surface water based on information provided by the Federal Environmental Agency of Austria (2004)



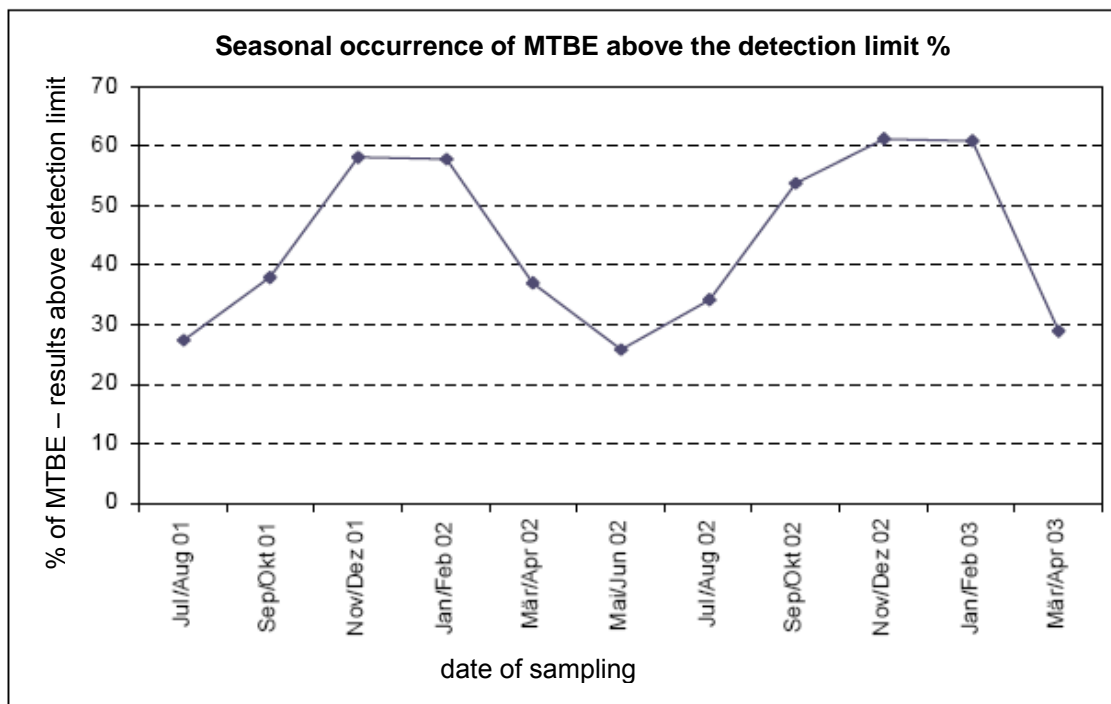
With respect to the sampling locations within this monitoring campaign, 19 of the 204 locations do not show any detectable MTBE. These locations mainly belong to smaller rivers in rural areas.

In contrast, 90% of the sampled locations show at least one sample above the limit of quantification of 0.02 µg/l. This reflects a wide spread of MTBE in Austrian surface water, albeit at a very low concentration.

At 8 sample locations across the Danube, the Drau, the Glan, the Fische and the Wienfluss Rivers, all samples exceeded the analytical limit of quantification (Federal Environmental Agency of Austria 2004). The mean concentration of MTBE in these rivers ranges from 0.04 µg/l (Drau) to 0.12 µg/l (Danube).

An additional evaluation of data from samples exceeding the detection limit shows a seasonal relationship of the occurrence of a measurable MTBE concentration in the surface water. The results are presented in **Figure 3.16**. These show a significant increase in detection of MTBE in the winter season.

**Figure 3.16** Observed seasonal relationship of the occurrence of MTBE in surface water (Federal Environmental Agency of Austria, 2004)

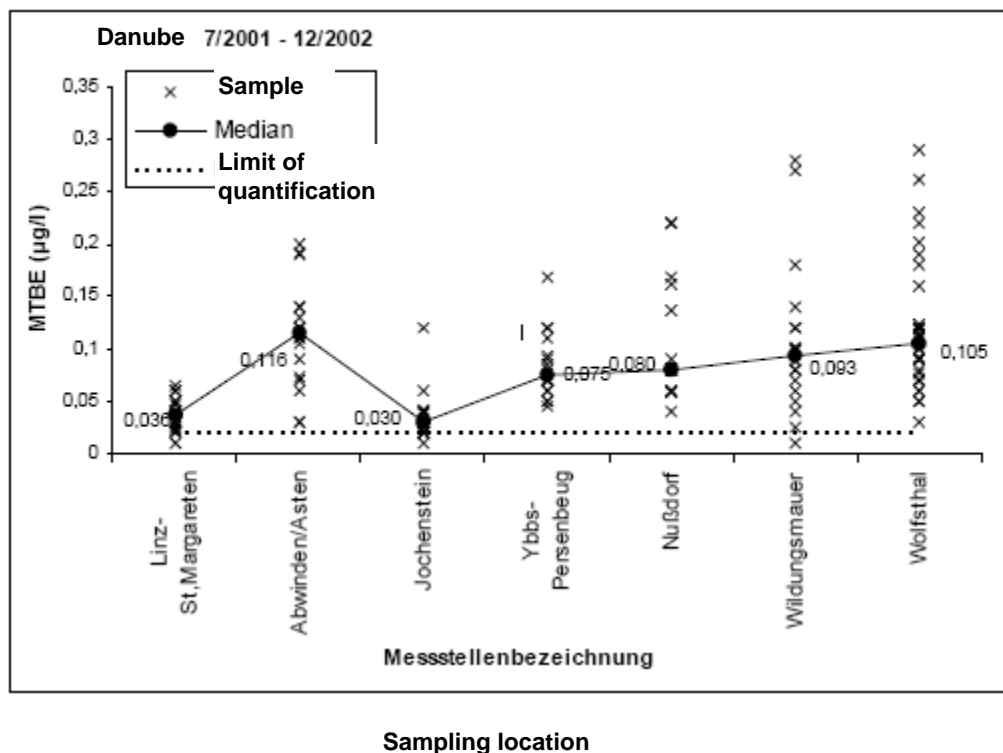


Danube River

The River Danube plays a key role within the surface water quality monitoring in Austria, as it represents the main natural drainage of the country. With respect to this major role, the water quality report by the Federal Environmental Agency of Austria (2004) evaluated the monitoring data of 7 sampling locations along the Danube in Austria. Six of these locations provide data for 18 measuring campaigns; the station Wolfsthal provided data for 36 measuring campaigns.

The monitoring period for these campaigns covered the period July 2001 until December 2002. The results of this special monitoring are summarized in **Figure 3.17**.

**Figure 3.17** Observed seasonal relationship of the occurrence of MTBE in surface water (Federal Environmental Agency of Austria, 2004)



The results show that MTBE was consistently present at a concentration above the limit of quantification, with the exception of 3 out of 144 samples. The observed range of MTBE concentrations covers approximately 1 order of magnitude. The highest concentration of 0.32 µg/l was observed at Abwinden/Asten.

Within the scope of the monitoring data on GEO in surface water from requested local regulators no further data could be obtained.

**3.2.3.2. Distribution of GEO in Swiss surface water**

- The Swiss results show that there is a relatively low presence of GEO in the Swiss section of the River Rhine and its tributaries in the region of Basel.
- For Lake Zurich, the presence of MTBE at a significant concentration is documented and appears to show a strong positive correlation to boating activities on the lake.

Within this study, only a limited amount of information on GEO in Swiss surface water could be obtained.

Monitoring data from Basel

At the request of the local regulators, the city of Basel provided a bigger set of monitoring data for MTBE, ETBE and TAME for several rivers, including the River Rhine.

The data sets refer to 12 regularly monitored locations in the area of Basel. Two of these locations are sampling locations on the Rhine itself, whereas the other 10 represent tributaries within the area of the city.

For MTBE, the data available were determined between 2003 and 2010. In total, 2,993 water samples were analysed. At the sampling location "Rhine Ruess", six MTBE values were found to exceed the analytical detection limit (varying range 0.1 to 1 µg/l). These six samples were taken in 2003 and they ranged between 1.3 to 4.5 µg/l. Furthermore, two values of the tributaries exceed the analytical detection limit, with a concentration of 1.3 to 1.6 µg/l. These values were measured in 2007/2009.

For ETBE, the available data originate from 2007 to 2010. In total 1,007 water samples were analysed. None of the results exceeded the analytical detection limit of 1 µg/l.

For TAME, the available data was from 2008 to 2010. In total 711 water samples were analysed. None exceeded the analytical detection limit of 1 µg/l.

#### Monitoring station Weil on the River Rhine

On behalf of the Federal Environmental Agency of Switzerland (BAFU) and the Environmental Agency of Baden-Württemberg/Germany (LUBW), a continuous monitoring of the Rhine is performed at the Weil station. The monitoring station has been continuously operated since 1993. The station is supervised by the Agency for Environment and Energy of the city of Basel (AUE).

This monitoring station serves for long-term water quality control of the upstream Rhine section of Basel. Monitoring thereof is integrated in the programme of the International Commission for the Protection of the River Rhine (German abbreviation IKSR) (AUE, 2008).

The AUE publishes the results of the monitoring in annual reports on the internet. During the research for this study, the reports from 2001 till 2008 were available. The reports provide results of the monitoring for MTBE from 2003 until 2008 and ETBE for 2008. The results of the annual AUE reports from 2003 through to 2008 can be summarized as follows:

MTBE / ETBE measurements were made daily, whereby a composite sample was collected every day; this results in 365 / 366 samples each year. In 2003, five samples had MTBE concentration above the analytical detection limit of 1 µg/l. The values range from 1.31 to 4.53 µg/l. The mean concentration of the 5 samples containing MTBE was 2.686 µg/l.

Since 2008, MTBE was not identified in any of the collected samples above the detection limit (1 µg/l). With regard to ETBE there were no samples above the detection limit (1 µg/l) in 2008; no data was available post 2008.

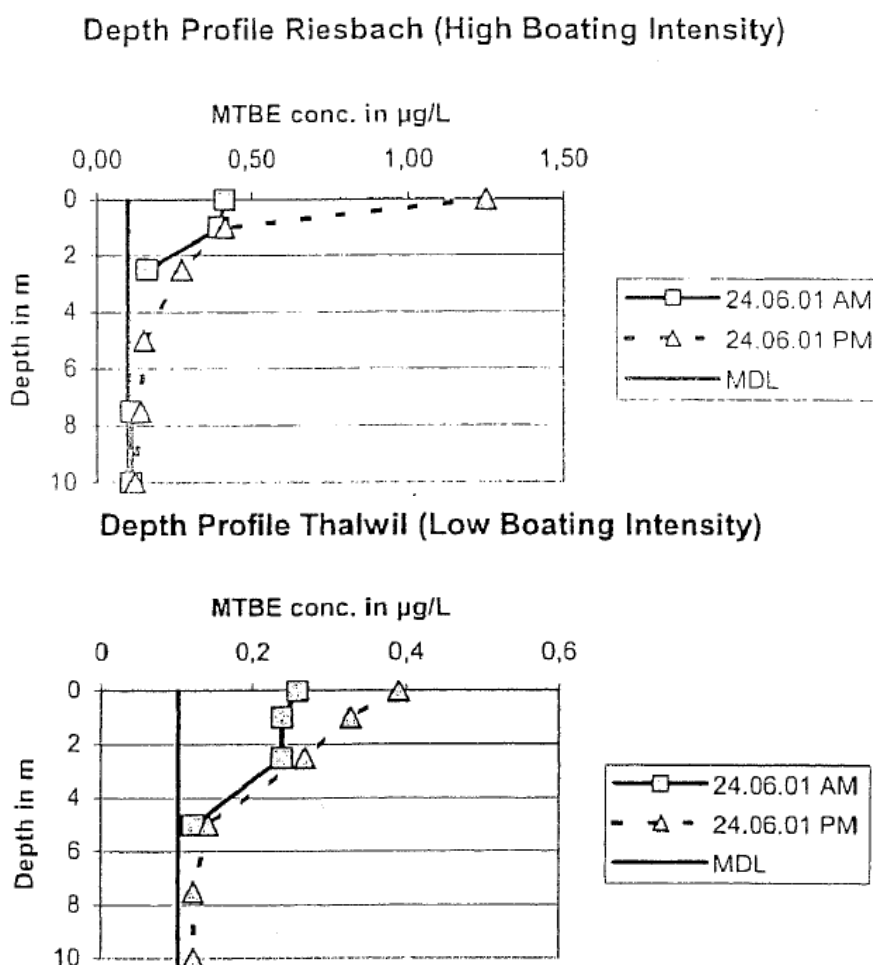
#### Lake Zurich

Schmidt et al (2002) and (2004) reported monitoring results of measurement campaigns for MTBE in Lake Zurich between 2002 and 2004. For the epilimnion (the uppermost layer of a thermally stratified lake), the highest observed MTBE concentration was 1.4 µg/l in the boating season (mean value 0.2 µg/l). For the off-

season, the maximum identified MTBE concentration within the epilimnion was 0.1 µg/l (mean value 0.058 µg/l).

The maximum MTBE concentration of the hypolimnion (deeper, cooler lake water) was 0.048 µg/l (with a mean value 0.037 µg/l). **Figure 3.18** provides two examples of the depth profiles for MTBE recorded by Schmidt et al (2002).

**Figure 3.18** Two examples for a typical MTBE depth profile at Lake Zurich (Schmidt et al, 2002)



The results in **Figure 3.18** also demonstrate the clear effect of a different distribution of MTBE due to boating activity.

Schmidt et al (2004b) concluded that due to the density stratification of holomictic lakes there is hardly any water exchange in summer (boating season). Thus, no significant transport of dissolved GEO from the epilimnion to the hypolimnion occurs. If the GEO are eliminated during the stratification period (e.g. by volatilisation), this will result in a low concentration for the hypolimnion for long periods.

Leemann (2004) reported monitoring data on Lake Zurich in a comparable way to Schmidt et al (2004b). Leemann's findings confirm the major results by Schmidt regarding the distribution of MTBE, and the local and seasonal effect of boating. At this point, Leemann's values will not be quoted because of a discrepancy within the concentration units between Leemann's text and the graphs presented, which could not be clarified.

### 3.2.3.3. Distribution of GEO in German surface water

- The German results show that MTBE and ETBE are present in many surface waters; the detected concentrations of ETBE are, however, much lower than those of MTBE.
- The bigger rivers show that higher concentrations of MTBE are usually related to urban and industrialised areas, whereas measurements in rural areas indicate much lower values.
- As a general trend in recent years, the concentration of MTBE peaks (especially in the Rhine) and the background load are decreasing slightly.
- Due to the low amount of data, a trend for the replacement product ETBE cannot be established for surface water.

#### Overview

Apart from the aspect that a lot of investigations for GEO, especially MTBE and ETBE, had been performed on rivers and large lakes in Germany, this study also indicates that the results of these investigations show variation in spatial and temporal distribution.

There is no institutional and standardised monitoring system for surface water in Germany. The responsibility for monitoring and additional investigations lies with the Environmental Agencies of each federal German state (so called Bundesland). Thus, activities from states with large industrial areas and access to main rivers (like North Rhine – Westphalia, Baden-Württemberg, Hesse) differ a lot compared with activities of other Federal States.

Another reason is to be found in the approach for environmental research work in Germany, i.e. the way this work is conducted and funded. Towards the end of the 1990s, the awareness for MTBE increased significantly, resulting in an increase of research activity by universities, trust- and association-funded research institutes (e.g. TZW Karlsruhe, UFZ Leipzig). With the increasing awareness of the commencement of MTBE replacement by ETBE around 2006, ETBE became an additional focus.

Since approximately 2004/05, research and investigation activities on GEO in Germany decreased significantly. This was due to the knowledge obtained so far and reduced funding on this topic. Most recently, MTBE, ETBE and (to a lesser extent) TBA were integrated into ongoing regular surface water monitoring, differing from state to state. An exception is the special monitoring of the River Rhine, which is described in 3.2.3.4.

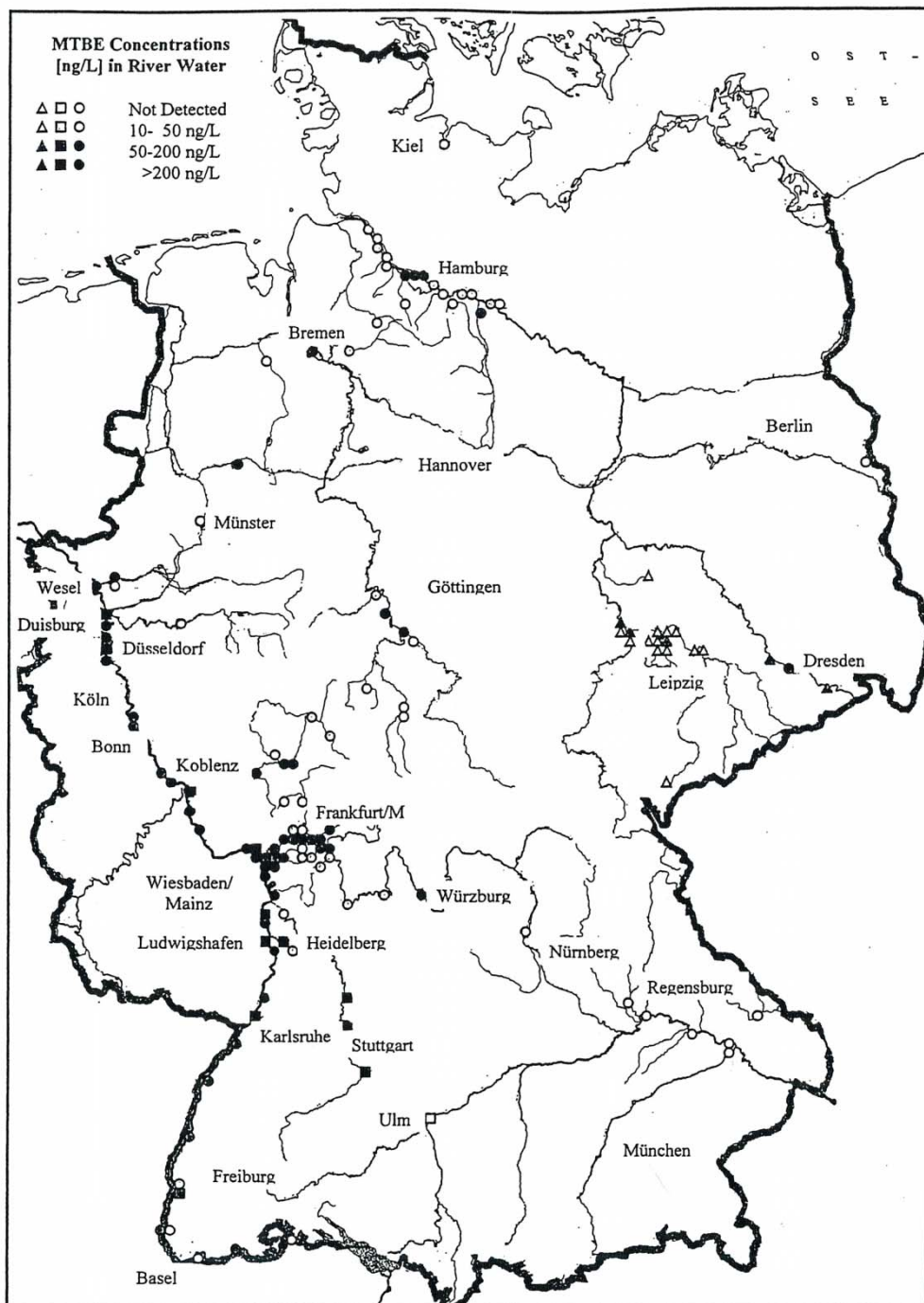
**Table 3.13** from Achten et al (2002b) provides an overview of the MTBE load in the major German rivers during the beginning of the monitoring period in Germany.

**Table 3.13** MTBE concentration in the major German rivers at the beginning of the monitoring period (1999 – 2002), taken from Achten et al (2002b)

| river                         | location                          | sampling date       | no. of samples | MTBE concentration [ng/L] |      |        |         |
|-------------------------------|-----------------------------------|---------------------|----------------|---------------------------|------|--------|---------|
|                               |                                   |                     |                | minimum                   | mean | median | maximum |
| Rhine, upper                  | Schaffhausen – Rastatt            | 11/06/00 – 11/07/00 | 8              | 13                        | 84   | 69     | 195     |
|                               | Vogelgrün – Karlsruhe             | 08/01/99 – 05/01/00 | 8              | 70                        | 115  |        | 160     |
|                               | Karlsruhe – Wesel                 | 05/11/00 – 01/08/01 | 52             | 52                        | 257  | 254    | 579     |
| Rhine, lower                  | Mannheim – Duisburg               | 08/01/99 – 05/01/00 | 52             | 100                       | 233  |        | 990     |
|                               | Wiesbaden/Mainz + Nierstein Mainz | 03/21/99            | 2              | 136                       | 148  | 142    | 160     |
|                               | Mainz                             | 08/01/99 – 05/01/00 | 7              | 120                       | 220  |        | 400     |
| Main, upper                   | Wiesbaden/Mainz                   | 05/11/00 – 01/08/01 | 28             | 107                       | 298  | 301    | 579     |
|                               | Seligenstadt                      | 05/29/00 – 01/11/01 | 4              | nd                        | 26   | 23     | 47      |
|                               | Kleinstheim – Gustavsburg         | 09/19/00 – 01/18/01 | 21             | 25                        | 65   | 87     | 275     |
| Main, lower                   | Offenbach – Gustavsburg           | 05/13/00 – 03/01/01 | 96             | nd                        | 210  | 130    | 2357    |
|                               |                                   | 01/14/99 + 04/21/99 | 2              | nd                        | 12   | 12     | 13      |
|                               | Frankfurt/M                       | 05/26/99 – 04/28/00 | 14             | nd                        | 42   | 32     | 116     |
|                               |                                   | 05/13/00 – 03/01/01 | 24             | 21                        | 161  | 114    | 502     |
|                               |                                   | 08/01/99 – 05/01/00 | 36             | 60                        | 200  |        | 540     |
|                               | Bischofsheim                      | 07/13/00 – 02/21/01 | 32             | 27                        | 205  | 184    | 547     |
| Elbe                          | Boitzenburg – Glücksstadt         | 09/02/00            | 15             | nd                        | 40   | 31     | 87      |
|                               | Hamburg                           | 04/22/99 – 02/14/01 | 9              | nd                        | 61   | 59     | 155     |
|                               |                                   | 08/01/99 – 05/01/00 | 2              | 450                       | 470  | 465    | 480     |
| Donau                         | Dresden                           | 03/28/99            | 1              |                           | 123  |        |         |
|                               | Rathen, Gaudernitz/Dresden        | 03/15/00            | 2              | 83                        | 141  | 141    | 199     |
|                               | Deggendorf – Regensburg           | 10/07/00            | 3              |                           | nd   |        |         |
| Neckar                        | Ulm                               | 08/01/99 – 05/01/00 | 4              |                           | nd   |        |         |
|                               | Mannheim – Deizisau               | 08/01/99 – 05/01/00 | 16             | 50                        | 163  |        | 1200    |
| Weser                         | Neckarhausen                      | 09/06/00            | 1              |                           | 15   |        |         |
|                               | Bremen + Hemeln                   | 09/02/00 + 09/28/00 | 2              | 89                        | 126  | 126    | 162     |
| Oder                          | Frankfurt/O                       | 03/10/99            | 1              |                           | nd   |        |         |
| Schwarzbach                   | Trebur near Frankfurt/M           | 12/07/00 – 02/21/01 | 17             | 38                        | 252  | 155    | 1024    |
| Kinzig                        | Hanau near Frankfurt/M            | 12/07/00 – 02/21/01 | 17             | nd                        | 94   | 32     | 1099    |
| Nidda                         | Frankfurt/M – Nied                | 12/07/00 – 02/21/01 | 17             | nd                        | 34   | 25     | 87      |
| Schwarzbach, Kinzig and Nidda | vicinity of Frankfurt/M           | 12/07/00 – 11/29/00 | 36             | nd                        | 121  | 35     | 1099    |
|                               |                                   | 01/10/01 – 02/21/01 | 15             | 33                        | 141  | 47     | 1024    |
| Landgraben                    | Trebur near Frankfurt/M           | 12/07/00 – 02/21/01 | 4              | 50                        | 1171 | 1183   | 2267    |
| Lahn                          | Coelbe – Limburg                  | 12/07/00 – 02/21/01 | 22             | nd                        | 82   | 43     | 293     |
| Small creeks                  | Germany                           | 06/13/00 – 11/29/00 | 38             | nd                        | 26   | 12     | 198     |
| Channels                      | Germany                           | 06/13/00 – 11/29/00 | 6              | 28                        | 83   | 52     | 204     |

Figure 3.19 provides a spatial overview of MTBE distribution in Germany at the beginning of the last decade (compare with Table 3.13).

**Figure 3.19** Spatial overview of MTBE distribution in Germany at the beginning of the last decade (Achten et al, 2002b)



The above quoted results from Achten et al (2002b) also include the results of earlier work as well as results of Effenberger et al (2001) and measurements by TZW Karlsruhe. Thus, this study is a reliable information source for the surface water contamination by MTBE in Germany at the beginning of the last decade.



Achten et al also report on sampling of the River Rhine along the complete length of the river in Germany, undertaken over the course of two days in November 2000. The results of this sampling campaign indicate the varying MTBE concentration along the river course, with a clear tendency for a lower value in rural catchment areas and a higher value in urban and industrialised catchment areas. Similar results were also reported for bigger rivers including the Main and the Danube.

A separate publication of a measurement campaign by TZW Karlsruhe, reported in LFU (2002), supported the main results of Achten. This publication also mentioned a MTBE sample collected from the Bodensee (Lake Constance), with a value below the detection limit of <0.05 µg/l.

#### Regulator data for the Federal State of Bavaria

The Environmental Agency of Bavaria provided monitoring data for MTBE from 2000 to 2008 and for ETBE in the period from 2006 to 2008.

In total 37 stations were monitored. Six of these locations are along the course of the Danube in Bavaria, while another 4 sampling locations are situated along the course of the River Main in Bavaria. The other sampling locations refer to smaller tributaries.

In total 1,479 samples were analysed for MTBE, in which 878 samples (59.4%) did not exceed the detection limit (range 0.01 to 1 µg/l). The highest concentration observed was 5.5 µg/l, and the average value of all samples above the limit of detection was about 0.03 µg/l. Around 73% of the stations sampled exceeded the detection limit.

In total 344 samples were analysed for ETBE, of which 319 samples (92.7%) did not exceed the detection limit (range 0.01 to 1 µg/l). The highest concentration observed was 0.4 µg/l, with the average value of all samples above the limit of detection being 0.036 µg/l. Around 13.5% of the stations sampled exceeded the detection limit.

#### Regulator data for the Federal State of Saxony

Monitoring data for MTBE and ETBE provided by the Environmental Agency of Saxony were evaluated. The data sets of 2008 and 2009 include both MTBE and ETBE. For 2005 until 2007 only MTBE data was available.

For 2009, a total of 399 stations had been monitored for MTBE and 394 of these were monitored for ETBE. Nine are situated along the course of the Elbe in Saxony. Another 27 sampling locations refer to different lakes in Saxony. The other sampling locations are found on smaller rivers and their tributaries.

In total, 2,077 samples were analysed for MTBE, of which 1,984 samples (95.5%) did not exceed the detection limit (<0.2 µg/l). The highest concentration was 2.1 µg/l, with an average value of all samples above the limit of detection of about 0.3 µg/l. Around 3.5% of the sampled stations exceeded the detection limit.

In total, 2,097 samples were analysed for ETBE, of which 2,094 samples (99.85%) did not exceed the detection limit (range < 0.2 µg/l). The highest concentration was 1.4 µg/l, with an average value of all samples above the limit of detection of about 0.88 µg/l. Around 0.76% of the sampled stations exceeded the detection limit.

For **2008**, all 363 stations were monitored for MTBE and ETBE.

In total, 2,273 samples were analysed for MTBE, of which 1984 samples (93.14%) were at or below the detection limit (<0.2 µg/l). The highest observed concentration was 1.1 µg/l; the average value of all samples above the limit of detection was about 0.33 µg/l. Around 7.2% of the stations sampled exceeded the detection limit.

In total, 2,271 samples were analysed for ETBE, in which 2,263 samples (99.82%) did not exceed the detection limit (range <0.2 µg/l). The highest concentration was 1.2 µg/l; the average value of all samples above the limit of detection was about 0.88 µg/l. Around 1% of the sampled stations exceeded the detection limit.

The MTBE data for the years 2005-2007 can be summarised as follows:

|             |  |                        |
|-------------|--|------------------------|
| <b>2005</b> | no. of sampled stations                            | 152                    |
|             | no. of samples total /below the detection limit    | 1,372 / 1,206 (87,9%)  |
|             | max. concentration /average of positive detection  | 2.7 µg/l / 0.45 µg/l   |
|             | percentage of stations with positive detection     | 4.4%                   |
| <b>2006</b> | no. of stations sampled                            | 381                    |
|             | no. of samples in total /below the detection limit | 2,112 / 1,927 (91,24%) |
|             | max. concentration /average of positive detection  | 2.7 µg/l / 0.52 µg/l   |
|             | percentage of stations with positive detection     | 4.4%                   |
| <b>2007</b> | no. of stations sampled                            | 374                    |
|             | no. of samples in total /below the detection limit | 2,149 / 1,995 (92,83%) |
|             | max. concentration /average of positive detection  | 3.8 µg/l / 0.46 µg/l   |
|             | percentage of stations with positive detection     | 6.4%                   |

#### **3.2.3.4. Special observation at the River Rhine (coordinated with the Netherlands)**

With respect to GEO in large rivers, there is a special situation for the River Rhine. The Rhine is Germany's largest river, a very important source for drinking water for approximately 20 million people, and has been subject to long-term and comprehensive monitoring of organic contaminants, including GEO, over an extended period.

**Figure 3.20** Position of monitoring stations along the River Rhine



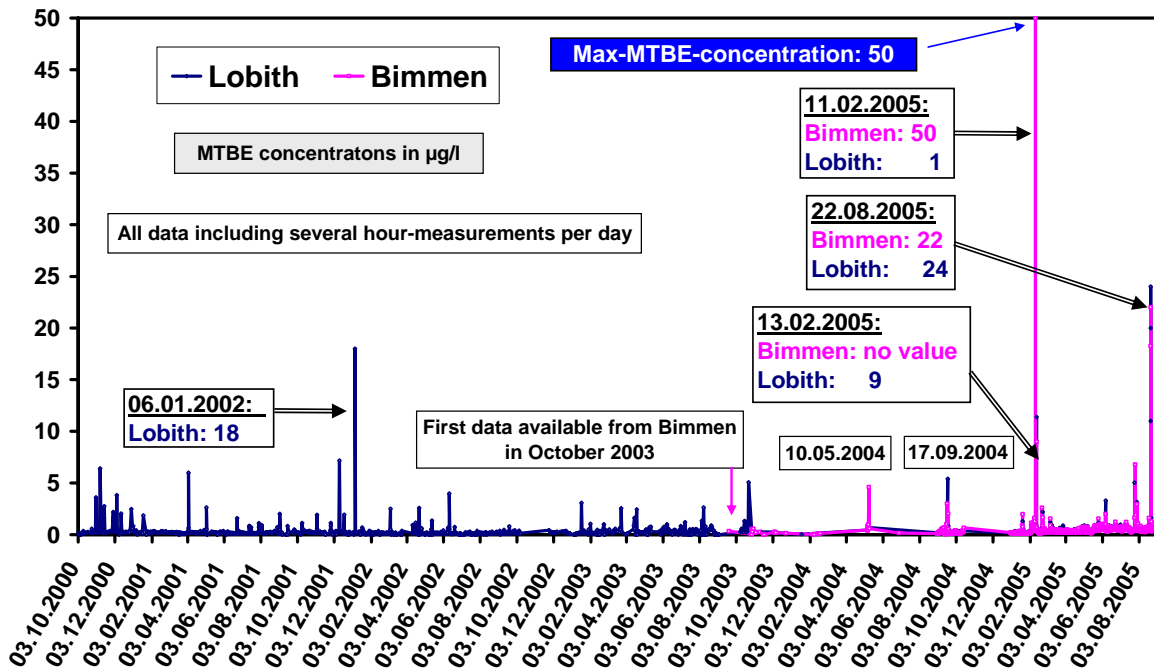
Due to former contamination of the Rhine with inorganic and organic contaminants in the 1960s and 1970s, a monitoring system was established to control the quality of the river water (Figure 3.20). The installation of this warning system started in 1987 and developed into a well established alarm system. At the border between Germany and the Netherlands, the International Monitoring Station Bimmen-Lobith (IMBL) was founded in 2001 in cooperation between RIZA and LANUV. RIZA is the “Dutch National Institute for Inland Water Management and Waste Water

Treatment” and LANUV is the “State Environmental Agency of NORTH RHINE – WESTPHALIA”. The stations Bimmen and Lobith already existed as independent monitoring stations since 1974 and 1970. The data from these stations are available to the public and can be viewed on the internet <http://www.aqualarm.nl>. The MTBE concentration has been measured since 2000 at the Rhine Monitoring Station Lobith and since 2003 at Bimmen.

An elevated (sporadic spike) MTBE concentration has been observed since the beginning of MTBE monitoring at International Measuring Station Bimmen Lobith (IMBL). The historically identified highest spike had an MTBE concentration of 70 µg/l. In 2005, measurements for ETBE were incorporated in the monitoring plan at IMBL and a similar spike of ETBE has have been detected. In addition, from the start of 2006, the MTBE and ETBE sampling frequency at the Rhine Monitoring Stations Bad Honnef and Düsseldorf were intensified (**Figure 3.20**). As a consequence, data from these stations are now used to investigate spikes of both oxygenates in the River Rhine. The main tributaries to the Rhine (e.g. Sieg, Wupper, Ruhr, Emscher and Lippe) are monitored by separate stations.

The average discharge of the River Rhine at the location of IMBL is 2,300 m<sup>3</sup>/s (range from 1,000 – 12,000 m<sup>3</sup>/s). Considering this high river discharge, the MTBE-concentration implies a significant mass of MTBE associated with each recorded spike in concentration. The concentration of MTBE measured in Bimmen and Lobith for the period 2000 – 2005 is presented in **Figure 3.21**.

**Figure 3.21** MTBE concentration (µg/l) in the River Rhine 2000 – 2005, based on data from monitoring stations Bimmen and Lobith



Under contract by EFOA (European Fuel Oxygenates Association), Dr. Stupp Consulting GmbH (DSC) carried out several studies to identify GEO sources in the River Rhine, to deduce the entry points and estimate the amount of GEO released into the river (Dr. Stupp Consulting, 2007 and 2008). From these studies it is evident

that ether transporting vessels (ships) were responsible for the larger releases (spikes). An estimation of the amounts released and the entry points was performed by using the so-called Rhine Alarm Model.

An example of the calculation results for the year 2006 is shown in **Table 3.14**. As listed in the table, seven of the ten peaks originate from the river section between DUISBURG and IMBL, concentrated between Rhine km 790 and 830. The mass of GEO released varied from 230 kg (spike 2) to 1,900 kg (spike 7). The range of MTBE releases varied between 290 kg and 860 kg. For ETBE the amount released ranged from 280 kg to 1,900 kg. The release with the maximum calculated ether amount is spike 7 (1,900 kg ETBE), detected on October 22<sup>nd</sup> at LOBITH.

**Table 3.14** Calibration results of 10 highest spikes of MTBE/ETBE from IMBL in 2006

| Spike No. | Event in 2006 | Monitoring Station with max. conc. | Sub-stance | Spill location Rhine km | Time of spill | Rhine Section   | Spill amount kg of Ether |
|-----------|---------------|------------------------------------|------------|-------------------------|---------------|-----------------|--------------------------|
| 1         | Jan 24        | Bimmen                             | ETBE       | 700                     | 11:00         | Köln-Düsseldorf | 500                      |
| 2         | Feb 11        | Lobith                             | MTBE       | 800                     | 15:00         | Duisburg-IMBL   | 230                      |
| 3         | Mar 11        | Bimmen                             | MTBE       | 810                     | 11:00         | Duisburg-IMBL   | 600                      |
| 4         | Jul 29        | Lobith                             | ETBE       | 790                     | 10:20         | Duisburg-IMBL   | 280                      |
| 5         | Sep 07        | Lobith                             | MTBE       | 830                     | 23:30         | Duisburg-IMBL   | 290                      |
| 6         | Sep 20        | Bimmen                             | MTBE       | 700                     | 00:00         | Köln-Düsseldorf | 860                      |
| 7         | Oct 22        | Lobith                             | ETBE       | 810                     | 16:20         | Duisburg-IMBL   | 1,900                    |
| 8         | Oct 22        | Lobith                             | MTBE       | 825                     | 20:10         | Duisburg-IMBL   | 325                      |
| 9         | Nov 19        | Bimmen                             | MTBE       | 800                     | 22:30         | Duisburg-IMBL   | 300                      |
| 10        | Dec 26        | Bimmen                             | MTBE       | 650                     | 03:20         | Bad Honnef-Bonn | 650                      |

Several actions were initiated. Firstly, a task force under the leadership of EFOA was founded in order to develop concepts for the improvement of the GEO concentrations in the River Rhine. Secondly the report “New code of best practice for handling of ethers” (EFOA, 2008) was published. In addition, further spike calculations were carried out to identify the ships responsible for the GEO releases into the river.

These actions have resulted in an improved water quality in the River Rhine. The spikes have largely disappeared in recent years and are now very seldom observed. For example, in 2010 until the end of October only two spikes of MTBE and ETBE with concentrations >3 µg/l were detected at one of the measuring stations in North Rhine-Westphalia. The most frequently reported MTBE concentration in the river was 0.05 µg/l and ETBE values were in most cases less than the detection limit (<0.05 µg/l). The background concentrations of MTBE, in former years often 0.2 – 0.5 µg/l, were also reduced effectively.

**3.2.3.5. Distribution of GEO in rivers and channels in the Netherlands**

- The Dutch results show a wide distribution of MTBE and ETBE in surface water.
- The highest concentrations of GEO are found in the Rhine, the Maas and the channels.

- A significant background concentration of GEO is present at the majority of the monitored locations.
- Besides the peak values and background level, seasonal effects are found which are assumed to result from increasing boating activities in the summer season.
- For the Maas, there are hints of a specific contamination event with DIPE – for clarification detailed investigations have already started.
- The actual values for ETBE, especially in the Rhine, also reflect the ongoing replacement of MTBE by ETBE.

The Netherlands Waterworks Association (VEWIN, 2005) published a report on the occurrence of MTBE in surface water. This report provides a good overview based on data sets from 2000 to 2004.

In total 22 locations at 18 different water bodies have been considered, including the major Dutch rivers (Rhine, Maas, Schelde), the main channels (Lekkanaal, Noordzeekanaal, Twentekanaal, Lateraalkanaal) and the main lakes (Ketelmeer, IJsselmeer, Haringsvliet).

The evaluation is based on the analysis of 5,685 water samples in total, of which 349 had an MTBE concentration  $>1 \mu\text{g/l}$  (6.14% of all samples).

The highest MTBE concentration was  $62 \mu\text{g/l}$  (observed in the Rhine at Bimmen/Lobith station). Furthermore, a one-time peak at the Maas (near Urmond) was reported with  $194.7 \mu\text{g/l}$ . The average MTBE concentration is between  $<0.1 \mu\text{g/l}$  (IJsselmeer) and  $2.7 \mu\text{g/l}$  (Lateraalkanaal). **Figure 3.22** provides a spatial overview, based on the mean MTBE concentration that has been calculated using the different data sets.

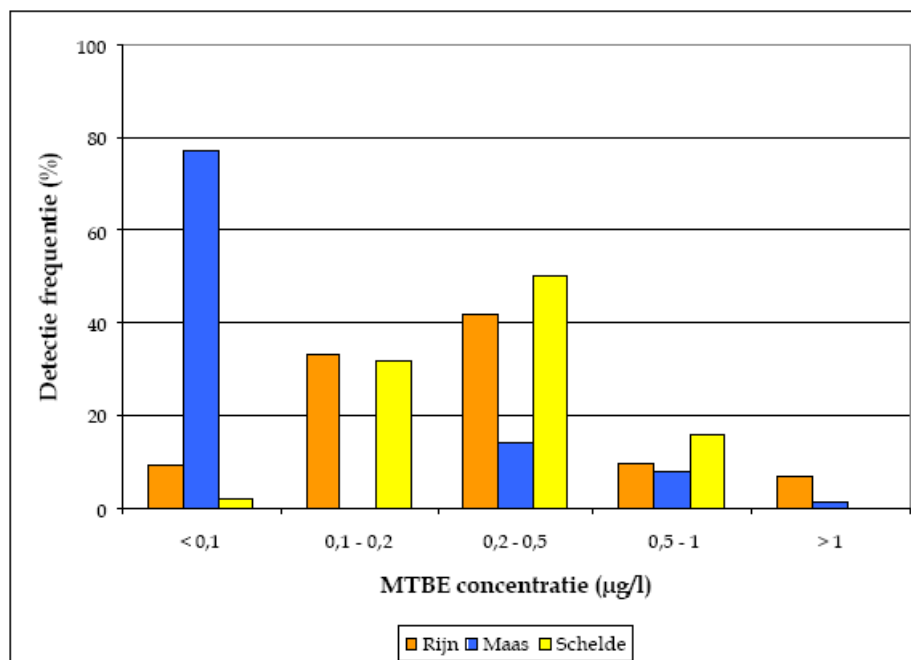
**Figure 3.22** Distribution of MTBE measured in Dutch surface water based on the median values obtained (VEWIN, 2005)



The results, by VEWIN (2005), show that the average MTBE concentration in the Rhine is significantly higher than in the Maas, whereas, the value for the Schelde is

comparable to those of the Rhine. **Figure 3.23** shows the distribution of the measured MTBE concentration in the Rhine, (Rijn) Maas und Schelde.

**Figure 3.23** Distribution of measured MTBE in Dutch surface water based on the median values obtained (VEWIN, 2005)



Rhine

For the Rhine the continuous monitoring data at the Bimmen/Lobith station (refer also to Chapter 3.2.3.3, subsection for Rhine observation) represent a large number of short peaks. Besides, the background load is described in the range of 0.1 to 0.5 µg/l. With regard to the calculated annual mean values for the Rhine from 2000 to 2004, VEWIN identified an increasing trend.

The surface water quality report on the Rhine by RIWA (2006) indicates monthly average values for MTBE and ETBE for 2005 as follows:

Lobith, 363 MTBE / 325 ETBE samples in total

MTBE: 0.186 to 0.761 µg/l, total average 0.405 µg/l  
 ETBE: 0.0697 to 0.998 µg/l, total average 0.231 µg/l

Lekkanaalwater, 26 MTBE / 26 ETBE samples in total

MTBE: 0.11 to 1.65 µg/l, total average 0.533 µg/l  
 ETBE: 0.035 to 0.31 µg/l, total average 0.0685 µg/l

Amsterdams-Rijnkanaalwater, 13 MTBE / 13 ETBE samples in total

MTBE: 0.05 to 0.7 µg/l, total average 0.304 µg/l  
 ETBE: <0.02 to 0.23 µg/l, total average 0.0546 µg/l



IJsselmeerwater, 15 MTBE / 15 ETBE samples in total

MTBE: <0.02 to 0.05 µg/l, total average not determined

ETBE: all <0.02, total average not determined

The surface water quality report on the Rhine by RIWA (2009b) indicates monthly average values for MTBE, ETBE and partly DIPE and TAME as follows:

Lobith, 352 MTBE / 359 ETBE / 13 DIPE samples in total

MTBE: 0.0847 to 0.259 µg/l, total average 0.147 µg/l

ETBE: 0.0624 to 0.233 µg/l, total average 0.139 µg/l

DIPE: <0.01 to 0.09 µg/l, total average 0.0138 µg/l

Lekkanaalwater, 16 MTBE / 16 ETBE / 16 DIPE / 16 TAME samples in total

MTBE: 0.05 to 2.42 µg/l, total average 0.703 µg/l

ETBE: 0.02 to 0.35 µg/l, total average 0.156 µg/l

DIPE: all <0.02, total average not determined

TAME: all <0.02, total average not determined

Amsterdams-Rijnkanaalwater, 13 MTBE / 13 ETBE / 13 DIPE / 13 TAME  
samples in total

MTBE: <0.05 to 0.255 µg/l, total average 0.0962 µg/l

ETBE: <0.02 to 0.162 µg/l, total average 0.10 µg/l

DIPE: all <0.02, total average not determined

TAME: all <0.02, total average not determined

IJsselmeerwater, 13 MTBE / 13 ETBE / 13 DIPE / 13 TAME samples in total

MTBE: all <0.05, total average not determined

ETBE: all <0.02, total average not determined

DIPE: all <0.02, total average not determined

TAME: all <0.02, total average not determined

### Maas

For the Maas, there is a seasonal influence on the MTBE concentration. In winter the range of values is comparable with the background contamination of the Rhine. Another noteworthy finding was that a higher MTBE concentration in the Maas is often connected with low water levels.

Further information on MTBE, ETBE and DIPE concentrations for the Maas is reported in RIWA (2009a). With regard to monitoring data from 2008 at the stations Tailfer, Luik, Eijdsen, Heel, Brakel, Keizersveer and Stellendam, the following maximum GEO concentrations were reported:

MTBE: 0.22 to 1.5 µg/l

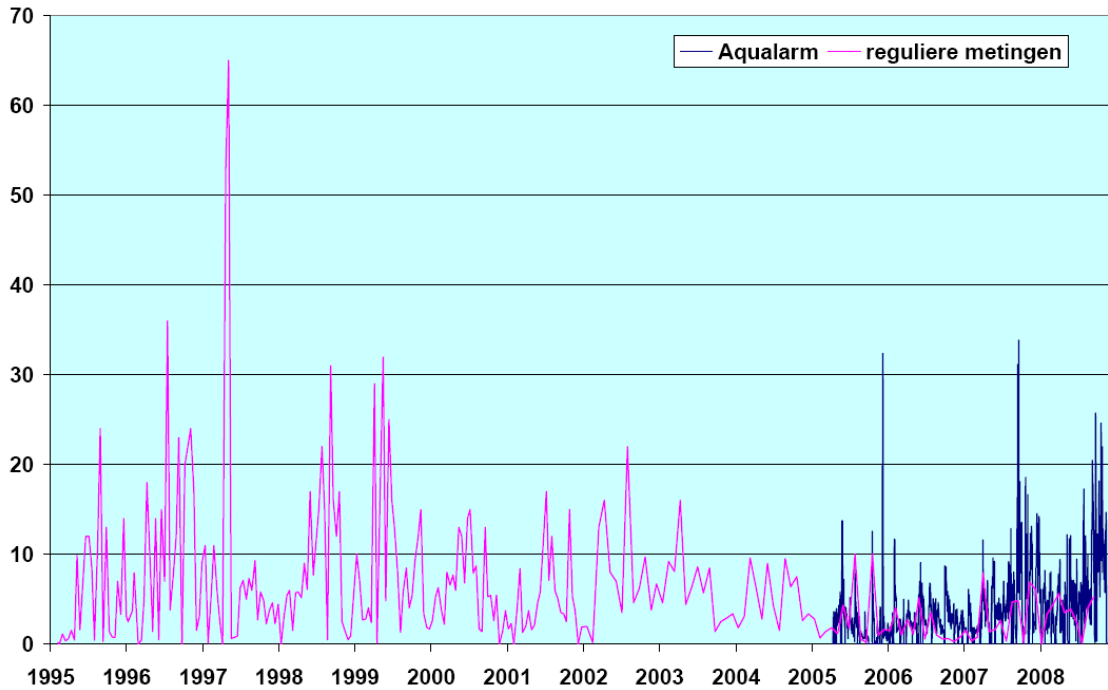
ETBE: <0.05 to 0.7 µg/l

DIPE: 0.07 to 11 µg/l

RIWA (2009a) also reports a seasonal influence on the distribution of MTBE, assumed to reflect the seasonal increase of boating activities in summer. Additionally, a comparison of the average and maximum MTBE concentration at the Keizersveer monitoring station indicates a significant decrease of the MTBE load from 2007 to 2008.

For DIPE, the RIWA report provides a graph with the DIPE concentration in the Maas at the Eijsden station from 1995 to 2008 (**Figure 3.24**).

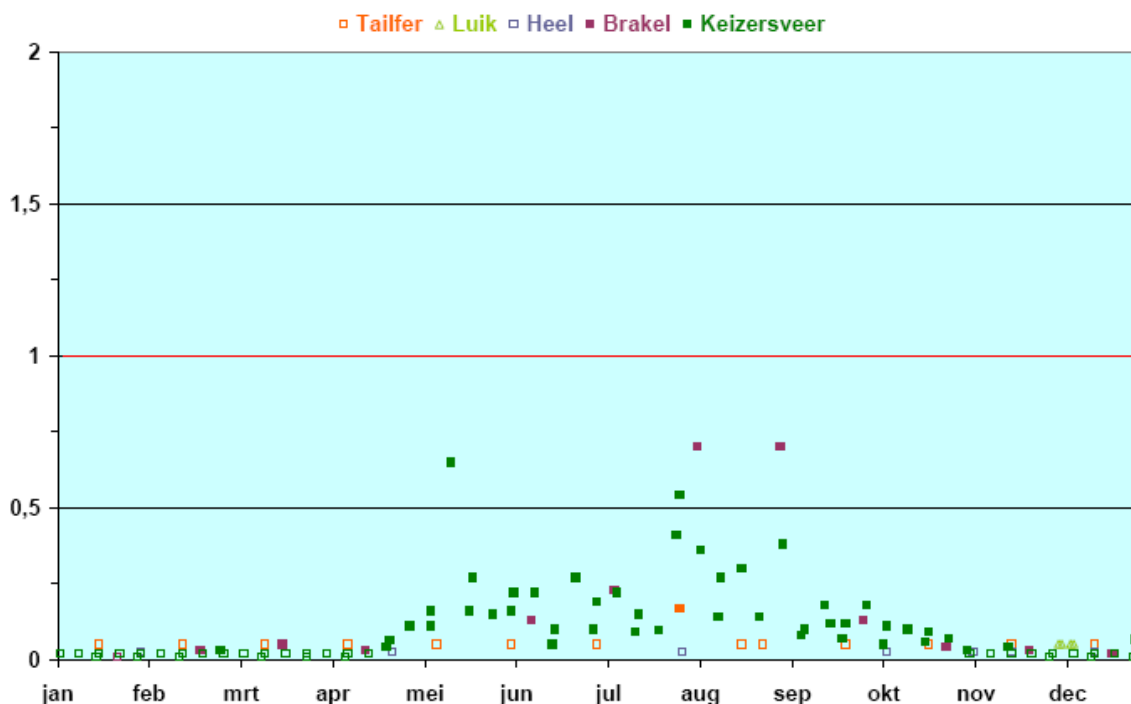
**Figure 3.24** Concentration ( $\mu\text{g/l}$ ) of DIPE in the Maas at the Eijsden station (RIWA, 2009a)



In addition, there has been a significant increase in the cumulative annual DIPE load in the Maas, from 9 tons in 2006 to 19 tons in 2007 and 26 tons in 2008.

For ETBE the 2008 measurements indicate a clear seasonal relationship (see **Figure 3.25**), which is (as with MTBE) assumed to reflect an increase of boating activities and the ongoing replacement of MTBE by ETBE in fuels.

**Figure 3.25** Concentration ( $\mu\text{g/l}$ ) of ETBE in the Maas in (RIWA, 2009a)



The RIWA report also compares the average concentration (2008:  $0.12 \mu\text{g/l}$  / 2007:  $0.1 \mu\text{g/l}$ ) and maximum concentration (2008:  $0.65 \mu\text{g/l}$  / 2007:  $0.68 \mu\text{g/l}$ ) at the Keizersveer station, indicating a constant ETBE level in the river.

**3.2.3.6. Distribution of GEO in surface water in the UK**

- The results imply that most MTBE in surface water is directly linked to local pollution incidents.
- Available data are not adequate to deduce the background concentration of GEO in surface water in the UK.

The Energy Institute (2009) summarised regulatory data on surface water concentrations of MTBE, ETBE, TAME and DIPE. Most of the data belongs to England and Wales (in total 317 water samples from 90 locations). For Scotland, 34 locations with 161 samples (only MTBE) are mentioned. No data was obtained from Northern Ireland, the Channel Islands or the Isle of Man.

In Scotland, none of the 161 samples from the 34 sampling locations had MTBE above the detection limit. The majority of the MTBE samples in England and Wales (250 out of the 317) refer to investigations in response to pollution incidents (no further details on the incidents or the sampling location are given). Of these investigations, 52 samples had an MTBE concentration above the analytical detection limit (range from  $0.2$  to  $108,000 \mu\text{g/l}$ ). It is noteworthy that 38 of these 52 samples belong to just 3 different geographic locations.

The remaining 67 samples (non-investigated data) were interpreted as more representative for the background contamination of surface water in England and Wales. A total of 12 of these 67 samples at 12 different locations had an MTBE

concentration above the analytical detection limit (range from 0.06 to 0.65 µg/l, geometric mean 0.18 µg/l). The analytical detection limit was 0.05 to 0.5 µg/l. The Energy Institute also reports that there is no routine monitoring by regulatory authorities for MTBE in surface water in England and Wales. However, the purpose for the survey of this data is also unknown.

ETBE, TAME and DIPE were not identified in any of the collected samples, from various sampling locations, at concentrations above the respective detection limit.

### 3.2.3.7. Additional findings for GEO in surface water of other European Countries

#### Regulator data of Italy

A data set of surface water samples for Bologna was provided. The data set refers to MTBE measurements in 2008. In total 8 stations were monitored. Most samples had been analysed for MTBE, but all had an MTBE concentration below the detection limit of <0.5 µg/l.

#### Cross reference for surface water measurements in Spain

Arambarri et al (2004), in Könen & Püttmann (2006),) refer to 18 MTBE measurements of surface waters in Spain, of which none had a value above the detection limit of 0.3 µg/l.

### 3.2.4. Literature Review and results of enquiry – GEO in runoff water

- The results from Switzerland and Germany indicate the presence of MTBE, DIPE and TAME in analysed runoff water. Similar results were found in the USA. ETBE was not detected.
- The results also show that the GEO concentrations strongly depend on the rain intensity and land use in the areas where the run off occurs. The detectable concentrations usually range from 0.05 to 1 µg/l.
- Especially for areas with high traffic, higher discharge from the runoff water is assumed as a relevant source for GEO.

The number of publications regarding the presence of GEO in runoff water is very limited. For Europe, information is available for Switzerland only. To gain more knowledge on this topic, a publication from the USA (North Carolina) was additionally taken into account.

#### 3.2.4.1. Results from Switzerland

Baudirektion Kanton Zürich (2009) published a report dealing with the potential contamination of Lake Zurich. In Canton Zurich, the runoff water from 12 km of highway road and 130 km of regular road is drained (partly directly, partly via waste water treatment and partly via soil-groundwater passage) to Lake Zurich.

The annual MTBE mass transfer from roads to the lake is reported as 0.105 kg. **Table 3.15** provides a detailed overview on the different pathways from the road towards the lake.

**Table 3.15** MTBE discharge from road runoff water to lake Zurich, based on local data and balance calculation – (Baudirektion Kanon Zürich, 2009)

|   | MTBE emission [kg/a] | MTBE imission [kg/a] |
|---|----------------------|----------------------|
| <b>Regular roads</b>                        |                      |                      |
| Direct discharge                            | 0.042                | 0.039                |
| Discharge via ponds and creek               | 0.016                | 0.009                |
| Discharge from waste water treatment plants | 0.024                | 0.013                |
| Discharge by flood / high water             | 0.008                | 0.007                |
| Sum regular roads                           | 0.090                | 0.069                |
| <b>Highway A3</b>                           |                      |                      |
| Discharge via ponds and creek               | 0.059                | 0.036                |
| <b>Sum Regular Roads + Highway</b>          | <b>0.149</b>         | <b>0.105</b>         |

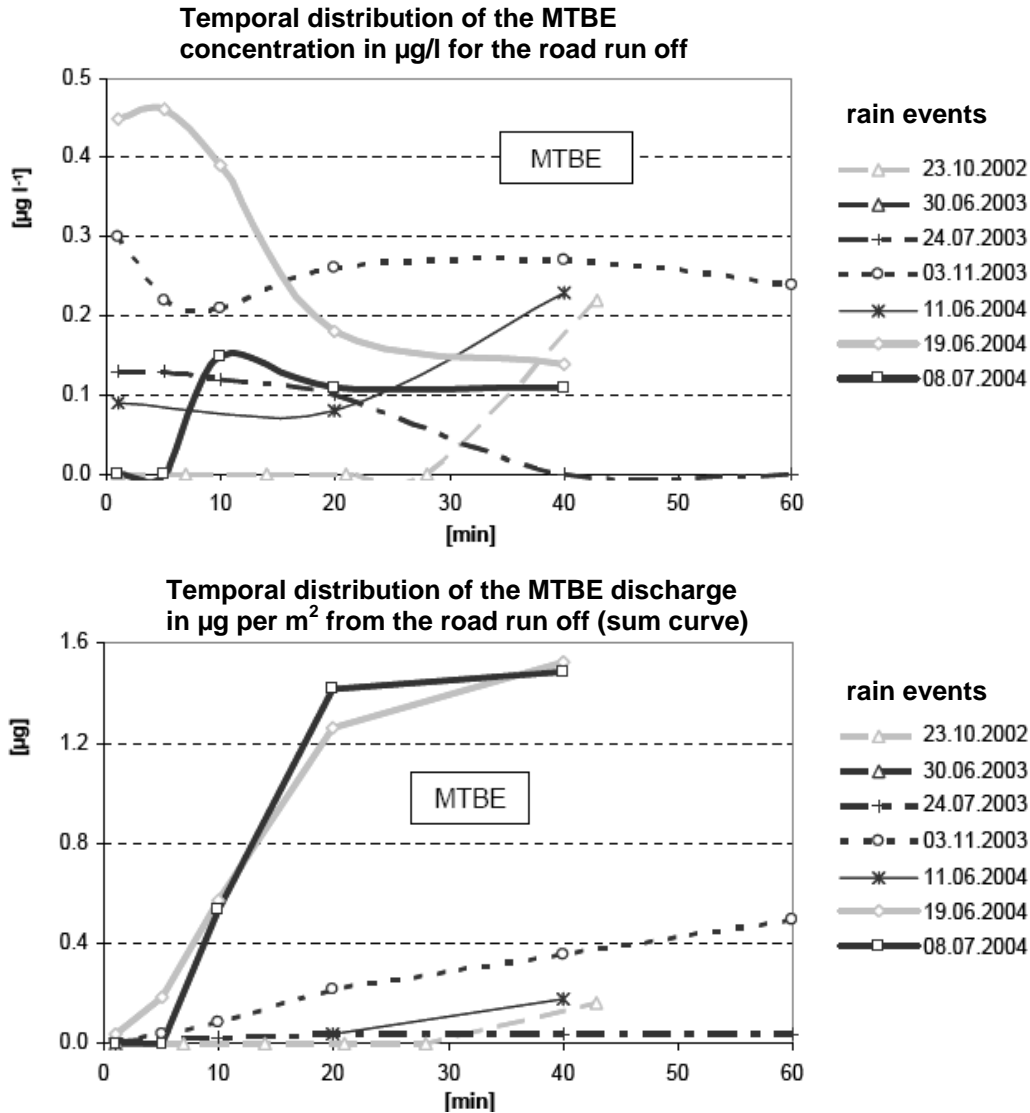
EAWAG (2005) published the results from a research project regarding the presence of hazardous substances in road runoff water from heavy traffic road and their possible retardation by special geotextiles.

The investigations with the pilot plant were conducted close to Burgdorf an der Emme (approx. 20 kms north-east of Bern) on a feeder road to highway A1, a heavy traffic road with approx. 17,000 vehicles per day. Within the investigations, the road runoff from a road area of 1,500 m<sup>2</sup> was collected and sampled.

Within this research, the dynamic distribution of seven precipitation events (between October 2002 and July 2004) was investigated. The average intensity of rain events ranged from 0.07 mm water column per 5 min to 0.98 mm water column per 5 min.

**Figure 3.26** shows the temporal distribution of MTBE concentrations and discharge during the different rain events. The analytical limit of quantification in these measurements was 0.1 µg/l.

**Figure 3.26** Temporal distribution of the MTBE concentration and discharge during the observed rain events – from EAWAG (2005)



The highest observed MTBE concentration during the rain events ranged from 0.13 to 0.46 µg/l. The MTBE discharge per m<sup>2</sup> road varied from 0.04 µg to 1.5 µg. The results also show the important influence of rainfall intensity.

**3.2.4.2. Results from USA used for a comparison**

To compare the results shown by EAWAG (2005), the values reported by Borden et al (2002) for storm water runoff from North Carolina (US) were reviewed.

In Borden et al (2002), 246 storm water samples from 46 different locations were analysed for MTBE, ETBE, DIPE and TAME (among other substances). The locations varied according to the type of land use. The results of the measurements are summarized in **Table 3.16**.

**Table 3.16** Summarized results for MTBE, ETBE, DIPE and TAME measured in storm water, North Carolina, USA (Borden et al, 2002)

|         | Mean (µg/l)     | Median (µg/l) | S.D. (µg/l) | Maximum (µg/l) | Detection frequency (%) |
|---------|-----------------|---------------|-------------|----------------|-------------------------|
| MTBE    | 0.50            | 0.10          | 1.37        | 13.47          | 47.1                    |
| DIPE    | 0.22            | 0.10          | 0.24        | 0.64           | 3.1                     |
| ETBE    | ND <sup>a</sup> | ND            | ND          | ND             | 0                       |
| Benzene | 0.09            | 0.09          | 0.03        | 0.15           | 6.5                     |
| TAME    | 0.09            | 0.07          | 0.06        | 0.23           | 3.1                     |

The median / maximum MTBE concentration found at sites having institutional land use (0.17 µg/l / 0.77 µg/l), commercial land use (0.07 µg/l / 0.24 µg/l), reported by Borden et al (2002), were of the same order of magnitude as the Swiss road results by EAWAG.

**3.2.4.3. Additional findings for MTBE in city runoff from Frankfurt**

Achten et al (2001) report 12 samples of runoff water from the city of Frankfurt/Main that were investigated in the scope of a research on precipitation. The samples had a wide range of MTBE concentration, from 30 to 1,174 ng/l (0.03 – 1.174 µg/l) with a median concentration of 114 ng/l (0.114 µg/l). It was also concluded that about 20% of the detected MTBE concentration in urban runoff is already transported into the atmosphere prior to precipitation, whereas about 80% may be attributed to direct uptake of vehicle emissions and gasoline from the road.

**3.2.5. Literature Review and results of enquiry - GEO in rain and snow**

- MTBE can be present in precipitation, typically at trace concentrations (ng/l).
- The investigations on MTBE concentration in rain samples indicate a much higher MTBE detection rate (86%) for urban areas than for rural areas (18%). The reported MTBE concentration for rain were consistently below 0.1 µg/l.
- Snow samples did not indicate a general difference between MTBE detection in urban and rural areas.
- Generally, the concentration for snow were higher, indicating that snow probably acts as a more efficient scavenger for MTBE from air than rain.
- The detection rate for MTBE in snow, in the limited number of studies, was 65%
- Information on other GEO in precipitation was not available for Europe.

The literature regarding the presence of GEO in precipitation (rain and snow) is very limited. For Europe, comprehensive information is mainly concentrated on Germany.

Precipitation samples, winter 2000/2001

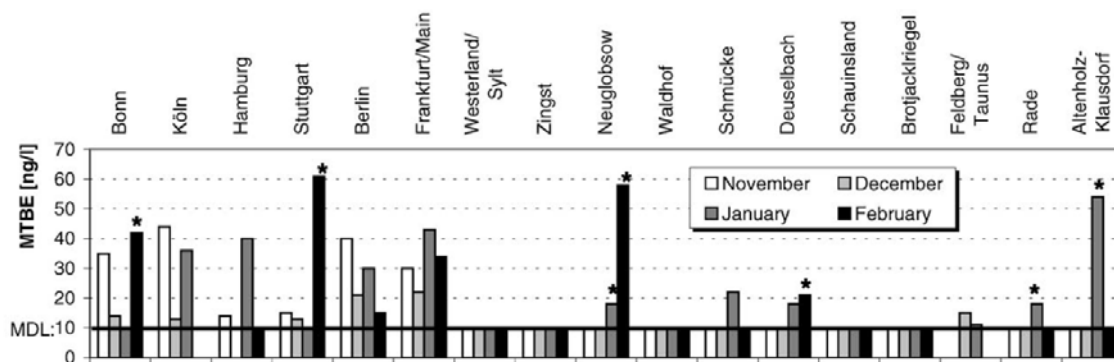
The first extensive investigations on MTBE in rain and snow samples were performed at the beginning of the recent decade. Achten et al (2001) (corresponding to LFU, 2002) reported a sampling campaign with 120 precipitation samples at 17 locations in Germany. The sampling was performed mainly from November 2000 to February 2001. Apart from the usual 4 samples per location, a series of

24 samples in Frankfurt/Main and a series of 8 samples in Berlin were conducted for time series evaluation. The detection limit in this investigation was 0.010 µg/l.

The maximum MTBE concentration determined was 0.085 µg/l. The time series data obtained in Frankfurt (September 2001 – March 2001) had an MTBE concentration from 0.030 to 0.085 µg/l for 49% of the samples (17 in total).

Regarding the complete campaign, the results indicate that MTBE is more often detected in urban precipitation (86%, 78 samples) than in rural precipitation (18%, 42 samples). The results of the campaign are summarised in **Figure 3.27**.

**Figure 3.27** Results for monitoring of precipitation in winter (Achten et al, 2001)



By comparing the results with corresponding temperatures and amounts of precipitation, it was concluded that the detection of MTBE in urban precipitation occurs at ambient temperatures lower than 10 – 15°C.

Snow samples, winter 2001/2002 and 2002/2003

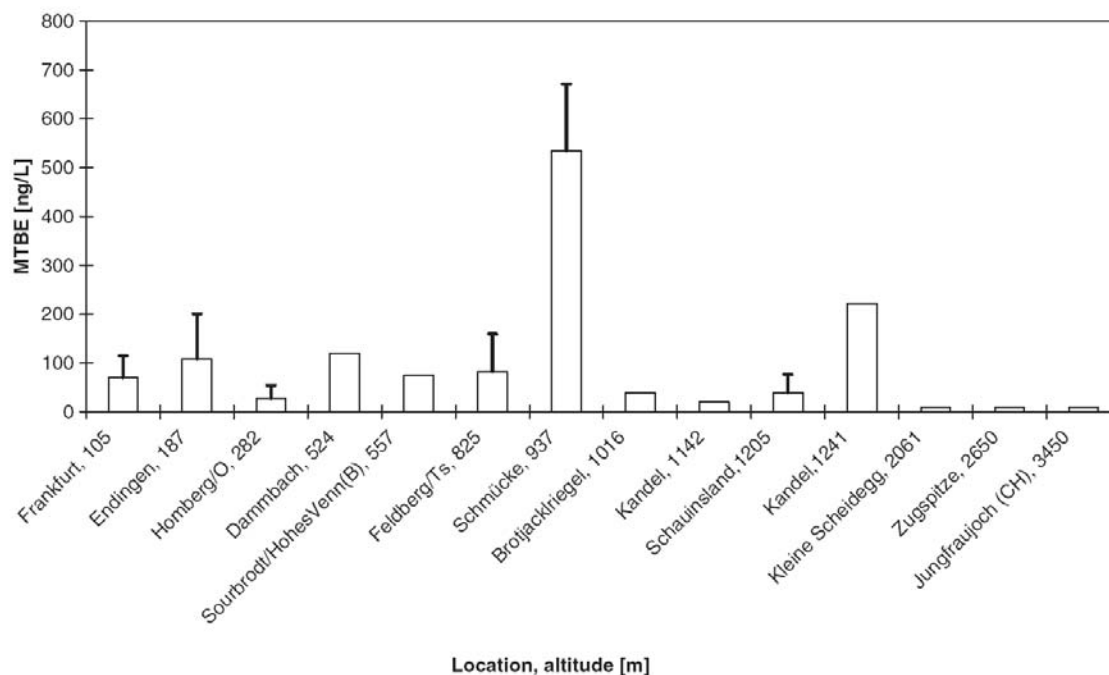
An intensive investigation of snow samples is reported in Kolb (2004) and Kolb et al (2006). In this study, 43 snow samples from 13 locations in Germany, 2 locations in Switzerland and one location in Belgium were analysed for MTBE. The sampling of fresh snow was performed in winter 2001/2002 and 2002/2003. As an example of an urban area, the city of Frankfurt/Main was chosen, the other 12 German locations represent 12 rural areas / remote locations at different altitudes (from 100 up to 3,450 m above sea level) and different geographical regions.

MTBE was detected in 65% of the snow samples. The observed concentration ranged from 0.011 µg/l to 0.631 µg/l.

The observed MTBE concentration was often found at higher values than rain water samples from earlier investigations, confirming that snow acts as a more effective scavenger for MTBE than rain. The results of this investigation are shown in **Figure 3.28**.



**Figure 3.28** Results for monitoring of snow samples in winter (Kolb et al, 2006a)



The results indicate that samples collected from rural and urban locations did not show significant differences in mean MTBE concentration. Thus, it was assumed that the MTBE concentration that could be found even in the snow samples from rural and remote areas may be explained by moving air masses, the lower photochemical degradation of MTBE in winter and ability of snow to scavenge gas phase impurities from the atmosphere. A temperature dependency of the MTBE snow concentration and a wash-out effect were observed in samples from the Schauinsland and Taunus-Observatorium (Kleiner Feldberg) monitoring stations.

**3.2.6. Literature Review and results of enquiry – GEO in air**

- The information regarding the presence of GEO in air for Europe is not adequate enough to draw a general conclusion on the distribution.
- Based on the literature on MTBE in air for Europe it can be assumed that MTBE is present in the European air.
- Results from Finland indicate a clear difference in MTBE concentration between air from urban areas (1.1 – 2.8 µg/m<sup>3</sup>) and rural areas (0.08 to 0.15 µg/m<sup>3</sup>), for a study in 2000-01.
- Results from urban areas in Helsinki are comparable with average values from urban areas in Germany. In addition the results from Zurich (Switzerland) and schools in Belgium also show the same order of magnitude.
- Information on other GEO in air could not be obtained for Europe.

The information regarding the presence of GEO in air for Europe is not adequate enough to draw a general conclusion on the distribution. The following points represent the results from limited findings in Europe.

Results from German publications

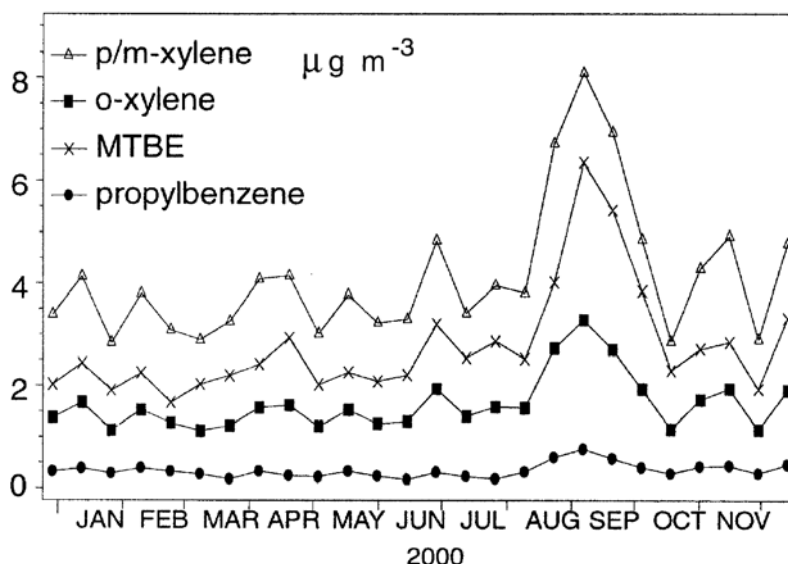
Creutzmacher (in LFU, 2002) reported results on the monitoring for MTBE in air for the federal state Baden-Württemberg. The average concentration was in the range of 1 to 2  $\mu\text{g}/\text{m}^3$  in urban areas. The value for rural areas was reported to be much lower. Some higher concentrations, up to 22  $\mu\text{g}/\text{m}^3$ , were observed, but these values represent only short term peaks.

Achten et al (2001) calculated equilibrium air concentrations for the results of the MTBE measurements in rain and snow. The mean values were estimated to be 0,157  $\mu\text{g}/\text{m}^3$  for urban areas and 0.039  $\mu\text{g}/\text{m}^3$  for rural areas (The original data from Achten et al (2001) was named in ppb. In order to operate with consistent units, the values were transferred into  $\mu\text{g}/\text{m}^3$  by using an air-density of 1,293  $\text{kg}/\text{m}^3$  at a temperature at 0°C.)

Results from Finnish publications

The results for monitoring of volatile organic compounds (including MTBE) in air in Helsinki were reported in Hellén et al (2002). The measurements were mainly from 2000 to 2001 at three air quality monitoring sites in Helsinki (measurement height of 3 m above ground level), a measuring station of the Finnish Meteorological Institute (measurement height of 12m above ground level) and a station in Hyytiälä as a representative for rural areas. The results of an annual time series for the first observation station in Helsinki are provided in **Figure 3.29**.

**Figure 3.29** Annual time series data for the concentration of BTEX compounds and MTBE in air in Helsinki (Hellén et al, 2002)



The range of observed values for the 4 stations in Helsinki are summarised in **Table 3.17**.

**Table 3.17** MTBE concentration in air from 4 observation stations in Helsinki (Hellén et al, 2002)

| Site | Description  | mean value ( $\mu\text{g}/\text{m}^3$ ) | range ( $\mu\text{g}/\text{m}^3$ ) |
|------|--|---|------------------------------------|
| 1    | Crossing with heavy traffic in the city centre (measuring height 3 m above ground level) | 2.8                                     | 1.7 – 6.3                          |
| 2    | Sports field in the city centre (measuring height 3 m above ground level)                | 1.5                                     | 0.9 – 4.3                          |
| 3    | Suburban traffic station (measuring height 3 m above ground level)                       | 2.4                                     | 1.5 – 5.1                          |
| 4    | Industrial area in Helsinki (measuring height 12 m above ground level)                   | 1.1                                     | 0.57 – 2.7                         |

For the rural station in Hyytiälä an MTBE concentration was reported in the range of  $0.146 \mu\text{g}/\text{m}^3$  (winter time) to  $0.078 \mu\text{g}/\text{m}^3$  (summer time).

Vainiotalo et al (1998) reported results of MTBE in ambient air in the vicinity of service stations. The measurements were performed in 1995 at two different service stations and ranged from  $0.5$  to  $121 \mu\text{g}/\text{m}^3$ , with a mean value of  $4.1$  to  $14.1 \mu\text{g}/\text{m}^3$ . The corresponding MTBE concentration measured at the pump islands ranged from  $274$  to  $1,347 \mu\text{g}/\text{m}^3$ .

Furthermore, Vainiotalo et al (2006) reported the air MTBE concentration in the breathing zone for workers that perform tasks like gauge calibration, hose replacements and other repairs / maintenance. Here, the mean concentration for all samples was  $11,000 \mu\text{g}/\text{m}^3$  for MTBE and  $4,700 \mu\text{g}/\text{m}^3$  for TAME. Peak values were up to  $46,000 \mu\text{g}/\text{m}^3$  for MTBE and  $29,300 \mu\text{g}/\text{m}^3$  for TAME.

Results of air measurements for 30 schools in Belgium

In VITO (2010) the results of an air quality investigation in Belgium schools (classroom as well as outside) for volatile organic compounds (including MTBE) are reported. In the study a 5 day average value was sampled for 3 classrooms at each school. Furthermore, 5 days average values were obtained for the outside road and the playground (**Table 3.18**).

**Table 3.18** Results for the air monitoring for MTBE at 30 Belgian schools – from VITO (2010)

| Place              | Number of samples (total) | Median value MTBE ( $\mu\text{g}/\text{m}^3$ ) | Max. value MTBE ( $\mu\text{g}/\text{m}^3$ ) |
|--------------------|---------------------------|--|--|
| Classroom          | 90                        | 0.23   | 3.2  |
| Road outside       | 30                        | 0.20   | 1.14   |
| Playground outside | 30                        | 0.17   | 1.2  |

### 3.2.7. Literature review - GEO in soil

- The knowledge based on the distribution of GEO within soils is relatively low.
- The available information suggests that GEO do not lead to a general background contamination in European soil.
- In urban areas, however, MTBE might be found at a concentration above analytical limits without a local point source existing (see Sweden).

#### 3.2.7.1. Distribution of GEO in soil in the UK

##### Report of the Energy Institute 2009

This study, carried out jointly by the Energy Institute and Environment Agency (Noble and Morgan, 2009), represents the only reliable comprehensive research and literature resource on the distribution of GEO in soil in Europe.

The data evaluated in this report were obtained by consultation with oil companies in the UK and results from the investigation of 632 sites were obtained. One site was a former distribution centre and the remaining 631 were petrol retail sites. The data mainly originate from investigations undertaken from 2003 onwards and includes soil data on 631 of the 632 sites.

Most retail sites were still operational, except for those which have been closed, pending redevelopment. In addition, at 169 sites remediation measures had been implemented and completed, or were still in operation. The majority of these sites were located in England and Wales (569 locations), with 62 sites in Scotland. No data was available from Northern Ireland, the Isle of Man or the Channel Islands. With respect to the sites owned by the interviewed oil companies in 2006, the data reflect 19% of the company-owned sites (in total 3,340 retail stations and seven distribution depots in the UK).

Soil samples were analysed for MTBE and partly TAME at the 631 sites. For TAME only the number of positive results is known, but not the total number of samples analysed for TAME. Furthermore, the data provided does not indicate whether the soil samples analysed belong to the unsaturated zone or the saturated zone.

Another noteworthy aspect is that 332 of the 631 samples were analyzed using GC-FID-based methods that might have led to false positive detection in the range below 100 µg/kg. The results of the other samples that were analyzed by GC-MS, with a detection limit of 10 µg/kg, are more reliable. The results were summarized by the Energy Institute (2009) in the following **Table 3.19**.

**Table 3.19** Reported GEO concentrations at sites of the major 4 oil companies in the UK (Energy Institute, 2009)

| Concentration (mg.kg <sup>-1</sup> ) | MtBE       |            |            | TAME     |
|--------------------------------------|------------|------------|------------|----------|
|                                      | GC-MS      | GC-FID     | Total      | GC-MS    |
| < detection limit                    | 224 (75%)  | 145 (44%)  | 369 (58%)  | ?        |
| < 0.1                                | 21 (7%)    | 45 (14%)   | 66 (10%)   | 9        |
| 0.1 to 1                             | 26 (9%)    | 54 (16%)   | 80 (13%)   | 6        |
| 1 to 10                              | 14 (5%)    | 51 (15%)   | 65 (10%)   | 1        |
| 10 to 100                            | 11 (4%)    | 31 (9%)    | 42 (7%)    | 3        |
| >100                                 | 3 (1%)     | 6 (2%)     | 9 (1%)     | 0        |
| <b>Total</b>                         | <b>299</b> | <b>332</b> | <b>631</b> | <b>?</b> |

Notes

1. TAME is not routinely analysed for at oil company sites. The number of sites where TAME was analysed for but not detected is unknown.
2. % refers to % of total for that column.

Apart from the uncertainty of the GC-FID samples below 0.1 mg/kg, a general outcome is that 58% of the samples had no detectable MTBE concentration in soil. It is also clear that the proportion of MTBE concentrations below the detection limit is significantly higher for GC-MS analysis (75%) compared with the GC-FID-analysis (45%). This may be an effect of the uncertainty associated with GEO detection using the GC-FID methods.

In total 31% of the soil samples have an MTBE concentration above the analytical uncertainty concentration of 0.1 mg/kg. Less than 10% of the total soil samples have contamination above 10 mg/kg. The geometric mean of all values above the detection limit was 0.82 mg/kg, and the maximum reported concentration was about 73,500 mg/kg, indicating a NAPL-contamination.

TAME was detected in 19 samples, in a range of 0.011 mg/kg up to 49 mg/kg, whereas in 15 samples the value was below 1 mg/kg. This results in a total geometric mean of 0.3 mg/kg.

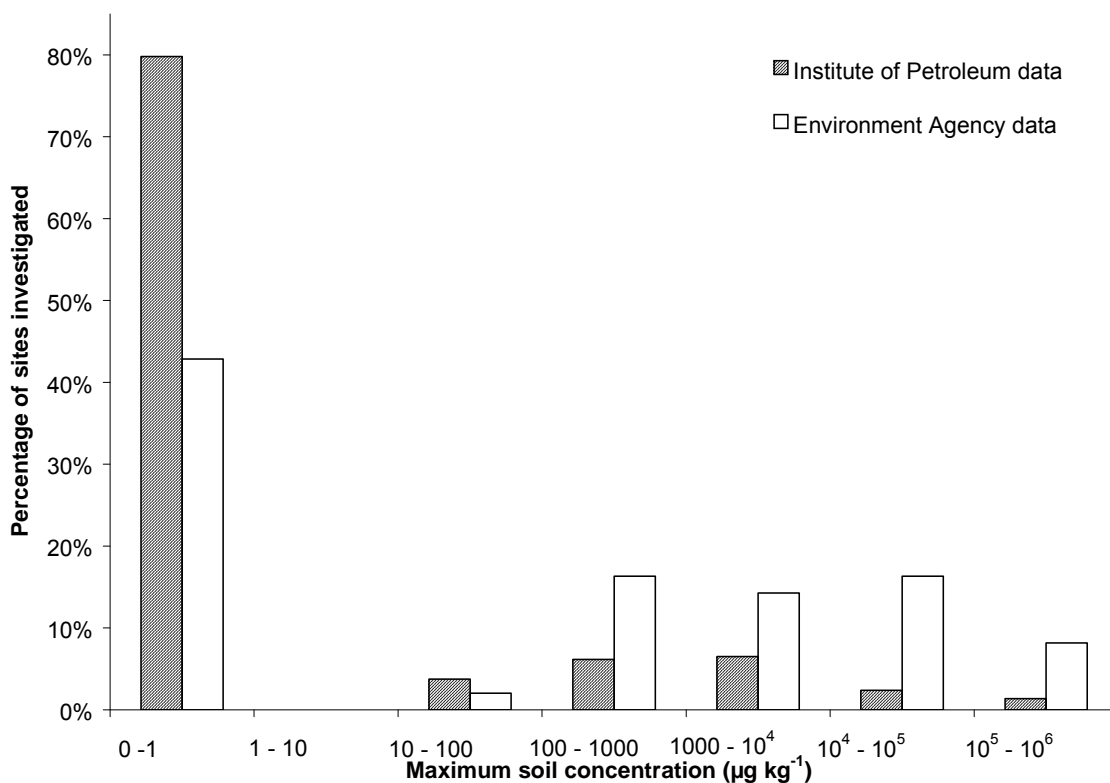
The same sites with contaminated soil were also investigated for GEO contamination of groundwater. A comparison of the groundwater results with the soil results shows that of 151 sites with no detectable groundwater contamination, 39 sites (26%) have an MTBE concentration in soil from 0.005 to 76.5 mg/kg.

Earlier Investigations in the UK by the Institute of Petroleum and the Environment Agency

Dottridge et al (2000) reported MTBE concentrations based on existing data (before 2000) by the Institute of Petroleum and the Environment Agency. It is believed that a significant part of this data was also used in the study by the Energy Institute in 2009.

**Figure 3.30** gives an overview on the distribution of the reported MTBE concentration in soil, collected in 2000. The MTBE concentration is plotted only for sites where MTBE was detected in groundwater: 99 sites (total 292) by the Institute of Petroleum and 47 sites (total 59) by the Environment Agency.

**Figure 3.30** Distribution of maximum MTBE concentration in soil from site investigation data from the Institute of Petroleum and the Environment Agency (Dottridge et al, 2000)



**3.2.7.2. Monitoring data for GEO in soil in Sweden**

The Swedish Environmental Research Institute (Potter et al, 2009) reported monitoring data for soil within the scope of results from the Swedish National Screening Programme 2008.

Within this programme, 5 sites were sampled for MTBE and ETBE in soil. The Kritineberg site was sampled as a representative for a background contamination. A traffic-related site in Göteborg was sampled to represent soil in urban areas. Furthermore, a gasoline station in Göteborg, a sample location 1 km north of the Preemraff Lysekil AB refinery, and a site of an ETBE Manufacturer (Borealis AB in Stenungsund), were sampled as representative for point sources.

The sample from Kristineberg had an MTBE concentration of 22 µg/kg. The concentration for ETBE was reported as <0.81 µg/kg. The sample in Göteborg had an MTBE concentration of 37 µg/kg, while ETBE was <0.69 µg/kg. The gasoline station had an MTBE concentration of 18 µg/kg, whereas ETBE was <0.57 µg/kg. The site near the refinery had 11 µg/kg MTBE and <0.65 µg/kg ETBE. The highest soil concentration was found in Stenungsund, with 55 µg/kg for MTBE and <0.89 µg/kg for ETBE.

Due to the relatively high concentration for MTBE in Kristineberg, the IVL assumed an urban influence at this site. In contrast, even close to the ETBE manufacturing site, the ETBE soil concentrations were below the limit of quantification.

**3.2.7.3. Soil concentrations for MTBE in Finland**

The Report of the European Commission (EU, 2002) contains results from 1998/99 relating to soil contamination of various sites in Finland that are known for petrol/oil contamination, or sites that had been used as service stations.

The detected MTBE concentration ranged from 0.1 to 1,000 mg/kg for contaminated gas and service stations. Furthermore, the soil gas concentration was in the range 100 to 130 mg/m<sup>3</sup> for one case, which probably reflects the presence of free-product in the subsurface.

The Finnish Environment Institute (SYKE, 2009) reported measurement of MTBE in 25 soils. The highest observed concentration was 1,400 mg/kg. The distribution of the concentration is summarized in (Table 3.20).

**Table 3.20** Reported measurement of MTBE in 25 soils Finnish Environment Institute (SYKE, 2009)

| <b>MTBE &lt; 0.1</b> | <b>0.1 &gt; MTBE &lt; 5</b> | <b>5 &gt; MTBE &lt;50</b> | <b>MTBE &gt; 50</b> |
|----------------------|-----------------------------|---------------------------|---------------------|
| <b>mg/kg</b>         | <b>mg/kg</b>                | <b>mg/kg</b>              | <b>mg/kg</b>        |
| <b>more than 10%</b> | <b>more than 20%</b>        | <b>nearly 40%</b>         | <b>nearly 30%</b>   |

## 4. FATE AND TRANSPORT OF GEO IN SOIL AND GROUNDWATER

The transport of GEO in the subsurface is influenced by the physical-chemical properties of the GEO as well as the specific hydrogeological and geochemical conditions of individual sites and the site-specific release conditions.

In the following chapters 4.1 to 4.3 the different environmental media, the conceptual model and the fate and transport processes in the subsurface are presented. In chapters 4.4 to 4.6 a literature review of the fate and transport characteristics of GEO in the unsaturated zone, groundwater and at the groundwater / surface water interface is provided.

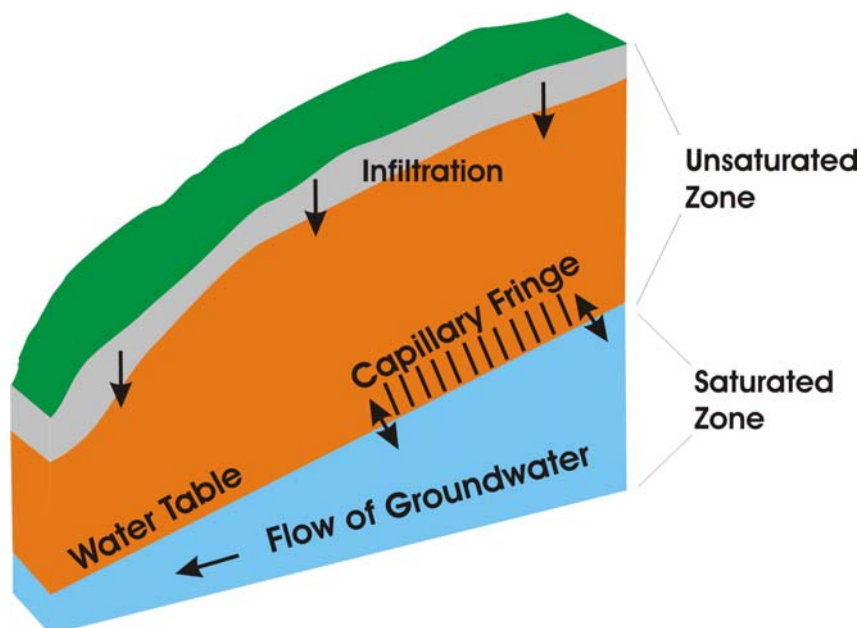
### 4.1. ENVIRONMENTAL MEDIA

The environmental media considered in the chapter fate and transport of GEO in soil and groundwater are:

- Unsaturated zone
- Saturated zone
- Hyporheic zone

From a hydrological perspective the subsurface below the soil zone can be classified into an unsaturated zone (also termed vadose zone) and a saturated zone (**Figure 4.1**). In terms of risk assessment the transition zone between groundwater and surface water must be considered too.

**Figure 4.1** Conceptual model of unsaturated and saturated zone



The unsaturated zone is the portion of earth between the land surface (soil) and the water table. Voids in the unsaturated zone are partially filled with water and the remainder of the pore space is taken up by air. Water in the unsaturated zone has a



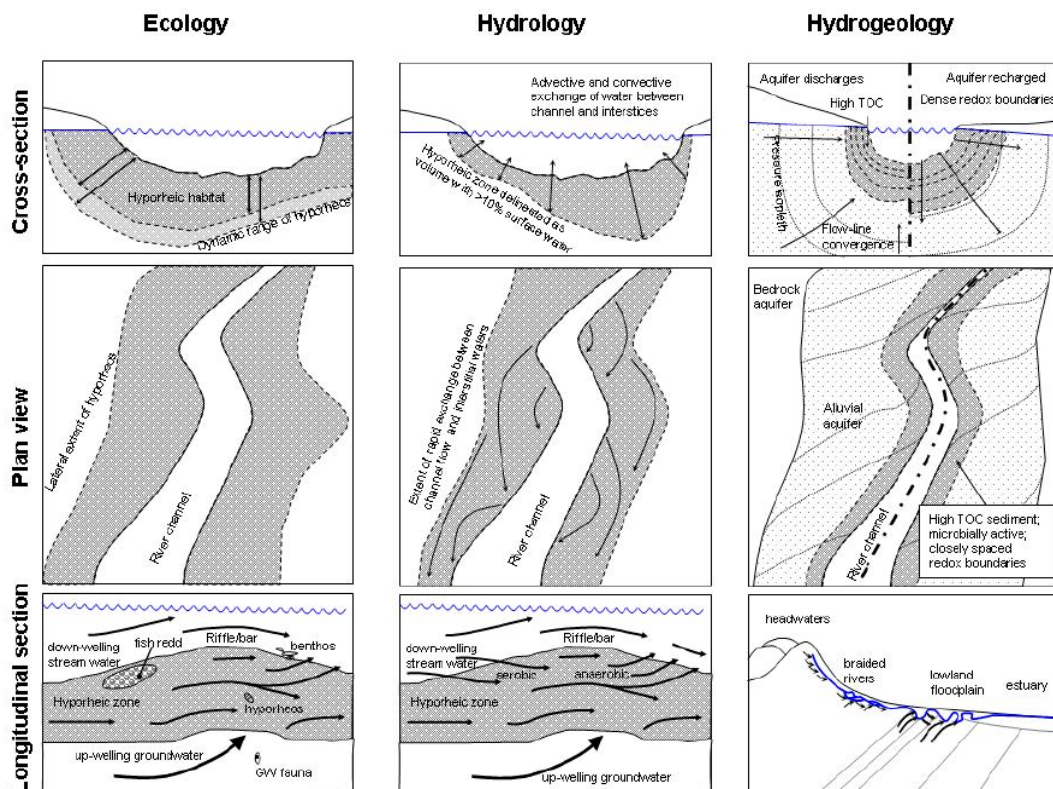
pressure head less than atmospheric pressure, and is retained by a combination of adhesion (funicular groundwater) and capillary action (capillary groundwater). The saturated zone refers to that part of the subsurface in which the whole pore-space is filled ('saturated') with water.

The top part of the unsaturated zone is the soil, a natural body consisting of layers (e.g. soil horizons but also bedrock) of mineral constituents of variable thickness with particular organic matter, which differ from the parent materials in their morphological, physical, chemical, and mineralogical characteristics. Soil will not be considered separately in this report, because releases of GEO do not usually affect natural soils. Consequently there is only limited literature dealing specifically with GEO and natural soils.

Another environmental compartment is the transition zone between groundwater and surface water, called the hyporheic zone. Several definitions and conceptual models of the hyporheic zone have been proposed by researchers in different scientific disciplines (ecology, hydrology, hydrogeology). This is visualized in **Figure 4.2**. Concentrating on the attenuation of GEO at the groundwater / surface water interface, this report defines the hyporheic zone as:

The water-saturated transitional zone (horizontally and vertically) between surface water and groundwater.

**Figure 4.2** Conceptual models of the hyporheic zone reflecting the aims of different researchers (Smith, 2005)



The hyporheic zone can be further characterised by:

- Increased organic matter content and influx of dissolved oxygen, primarily from surface water (but also groundwater where this is aerobic).
- Sharp chemical and redox gradients (Bencala, 2000) that combine to act as a sink for many inorganic (Fuller and Havey, 2000; Moser et al, 2003; Gandy et al, 2007) and organic contaminants (Lendvay et al, 1998a, 1998b; Lendvay and Adrianes, 1999; Smith and Lerner, 2008).
- Mixing between surface water and groundwater (Winter et al, 1998; Woessner, 2000).

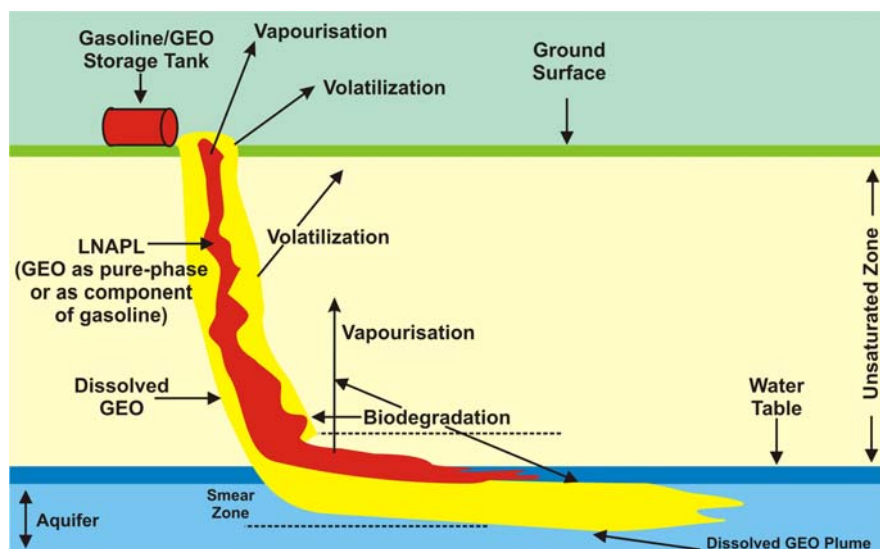
Compared with the groundwater environment, the hyporheic zone has higher variability in physical properties, chemical composition and microbial community.

#### 4.2. CONCEPTUAL MODEL OF LNAPL FATE AND TRANSPORT IN THE SUBSURFACE

- Risk assessment is systematically done on the basis of a conceptual model.
- The conceptual model of a GEO release on the ground surface shows the relevant fate and transport processes involved during GEO migration in the subsurface (**Figure 4.3**).
- Additionally the LNAPL migration is systematically described in a written summary.

The conceptual model including the release of GEO (as pure-phase or as a component of gasoline) at the ground surface and transport of Light Non-Aqueous Phase Liquid (LNAPL) and dissolved phase components through the unsaturated zone to the water table is shown in **Figure 4.3**.

**Figure 4.3** Conceptual model: Release of GEO at the ground surface and LNAPL/dissolved phase transport through the unsaturated zone to the water table



The idealized geology of the conceptual model consists of two principal components:

1. Unsaturated zone, where GEO migration in permeable material is primarily downward, but also laterally in response to variations in geological features (e.g. low permeability layers or fractures, etc) which induce preferential migration in this direction under the effects of gravity, with limited spreading. Contamination with GEO in the unsaturated zone consist of the four different phases air, water, soil/sediment and LNAPL.
2. Saturated zone where GEO migration is principally horizontal as pure and dissolved phase under the effect of the piezometric regime in an aquifer. Limited vertical movement may result from displacement of pore water and water table fluctuations. Contamination with GEO in the saturated zone consist of the three different phases water, sediment, and LNAPL.

A detailed discussion about different geological and hydrogeological environments throughout Europe is not part of this report. The conceptual model will focus on porous aquifers (intergranular porosity aquifers, unconsolidated and consolidated) which is the widest described European aquifer type in the literature. For detailed information about MTBE contamination, fate and transport in a dual porosity aquifer we refer to Thornton et al (2006).

The conceptual model of LNAPL migration in the porous subsurface can be described using the framework presented in API (2002). It should be noted that the “pancake” on water table model is highly idealised to provide a general conceptual model for the actual LNAPL transport and distribution. Site-specific factors like LNAPL distribution and source zone geometry can vary widely depending on geological and hydrogeological factors and site setting. This will be influenced by, for example, variation in matrix porosity, fracture orientation and connectivity (if present), water table fluctuations, groundwater flow direction and rate.

1. LNAPL release on the ground surface is influenced by the actual meteorological conditions (temperature, wind, rain) and the soil characteristics of the release site. LNAPL which does not migrate into the soil will vapourise directly into the atmosphere or dissolve in rainwater.
2. LNAPL migration in the subsurface begins with vertical drainage of LNAPL under gravity and capillary forces gradients. The drainage is strongly influenced by soil characteristics and occurs most rapidly in dry, high permeability soils, and more slowly in wet or low permeability soils. As the LNAPL moves downward through the unsaturated zone it will be subject to physical and chemical processes that include volatilisation, vapourisation, entrapment of part or all of the LNAPL as residual phase (immobile), and dissolution of LNAPL components in soil water (**Figure 4.3**).
3. If the release of LNAPL is sufficiently large to exceed the residual retention capacity of the unsaturated zone soils, the LNAPL will eventually encounter the groundwater table. As LNAPL encounters pore space completely saturated with water, the weight of the LNAPL causes it to displace pore water until hydraulic equilibrium is achieved. At the same time the large vertical gradient through the unsaturated zone dissipates into a lateral gradient in the water table zone.

4. Once the release of the free product stops, the LNAPL in the water table zone will eventually cease to move as the resistive forces in the water-wet sediments balance the driving forces in the LNAPL pool. An absolute endpoint of this movement is when the LNAPL reaches field residual saturation, a condition where the effective hydraulic conductivity of the host matrix to LNAPL movement is zero. This leaves a mass of LNAPL for secondary dissolved and vapour phase transport.
5. During the evolution of the LNAPL body external hydraulic factors may act to redistribute all or portions of it. For example, water table fluctuations will tend to smear LNAPL vertically throughout the range of the hydraulic variation, and often below the normally observed oil/water interface in a monitoring well. This occurs particularly in fractured rock aquifers, where the head of LNAPL created by the connected height of product above the water table can exceed the entry pressure into the fractures (Wealthall et al, 2002).
6. As soon as the LNAPL encounters groundwater dissolution of soluble compounds of the LNAPL by groundwater moving below and through the LNAPL-impacted zone begins. Thus a dissolved phase plume starts to develop and, with time, grows in the down gradient direction.
7. The size of a dissolved GEO plume depends primarily on the:
  - rate (flux) of release (flux) to the aquifer from the source (both in the unsaturated and saturated zones);
  - rate of transport in the aquifer (controlled by the advective groundwater flux);
  - rate of dispersion, vapourisation and volatilisation that lead to concentration reduction
  - the rate of biodegradation, which removes mass and reduces GEO concentrations (controlled in part by the flux of dissolved electron acceptors into the plume relative to the kinetics of the respective biodegradation processes).

Depending on the relationship between these four factors a plume may grow, stabilise or shrink.

On the basis of the physical-chemical properties and available experience, three different stages of MTBE (as an example GEO) plume development over time can be differentiated (Stupp, 2007):

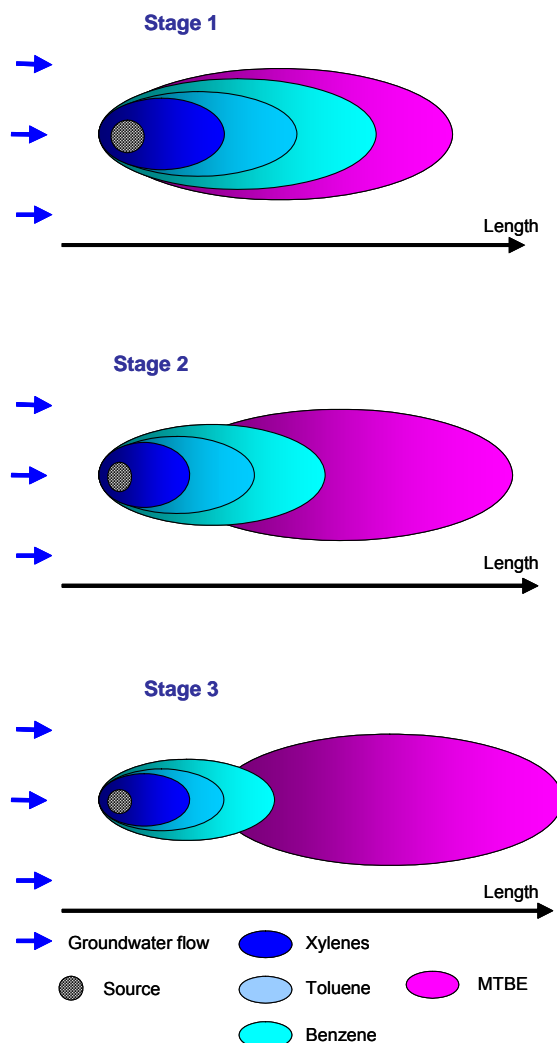
- Stage 1: The MTBE plume development corresponds almost to that of the BTEX plume.
- Stage 2: The MTBE plume development is further advanced along the flow path than the BTEX plume.
- Stage 3: The highest MTBE concentration has already left the source area while the BTEX plume core is still present in the source area or very close to it (a 'detached plume').

The three different spreading scenarios can be explained by different source strengths in connection with different relative time scales of development, illustrated in **Figure 4.4**. In groundwater MTBE has the potential for the

formation of relatively large plumes. These plumes are generally longer than BTEX plumes, but shorter than plumes formed by light volatile chlorinated hydrocarbons (e.g. tetra- and trichloroethene and their decay products), when present under aerobic conditions (Stupp et al, 2007).

8. As dissolution and volatilisation of more soluble and volatile LNAPL compounds continues, the LNAPL becomes increasingly depleted, resulting in decreasing concentrations of these compounds in the source area and eventual contraction of the dissolved plume.
9. If the dissolved plume migrates in a direction to surface water the water-saturated transition zone between groundwater (hyporheic zone) will influence fate and transport. This zone has sharp biogeochemical gradients, increased organic matter content and higher dissolved oxygen content relative to most aquifers. This leads to an increased potential for sorption, biodegradation and dilution due to mixing with surface waters.

**Figure 4.4** Schematic illustration of stages in MTBE plume development in groundwater over time for an idealised release of unleaded gasoline (Stupp, 2007)



### 4.3. FATE AND TRANSPORT PROCESSES IN THE SUBSURFACE

In a first step of risk assessment all processes that affect the fate and transport of GEO in the subsurface are identified.

- Dispersion and diffusion will dilute dissolved GEO concentrations in soil water and groundwater.
- Sorption and volatilisation will transfer GEO mass from dissolved GEO into the soil phase and gas phase.
- Vapourisation and dissolution will transfer LNAPL into the gas phase and water phase.
- Destructive processes that degrade GEO to CO<sub>2</sub> or other degradation products.

In a second step of risk assessment the most important processes for fate and transport of GEO in the subsurface are identified.

- The most important processes are vapourisation and biodegradation.

Processes affecting the fate and transport of GEO in the subsurface can be classified into non-destructive and destructive processes (**Table 4.1**). Non-destructive processes which do not affect the total GEO mass will dilute dissolved GEO concentrations in groundwater, transfer dissolved GEO to the gas phase and soil or transfer LNAPL to the water and gas phase. Destructive processes will reduce the GEO mass by abiotic degradation or biodegradation. All the processes listed in the **Table 4.1** are discussed in detail in the following chapters.

**Table 4.1** Overview of processes affecting the fate and transport of GEO in the subsurface

| Type of Process           | Effect on GEO in groundwater      | Process  | Controlling factors in the subsurface  |
|---------------------------|-----------------------------------|--|--|
| Non-destructive processes | Dilution                          | Dispersion   | Heterogeneity in groundwater flow and aquifer matrix physical properties                             |
|                           |                                   | Diffusion  | Heterogeneity in chemical concentration gradients and tortuosity of the porous media                 |
|                           | Phase transfer from dissolved GEO | Sorption<br>(Partitioning from GEO dissolved in groundwater into soil)           | Aquifer matrix and organic carbon content<br>Organic-carbon water partition coefficient ( $K_{oc}$ ) |
|                           |                                   | Volatilisation<br>(Partitioning from GEO dissolved in groundwater into soil air) | Henry's law constant   |

| Type of Process       | Effect on GEO in groundwater | Process  | Controlling factors in the subsurface  |
|-----------------------|------------------------------|--|--|
|                       | Phase transfer from LNAPL    | Vapourisation<br>(Partitioning from LNAPL into soil air or atmosphere) | Vapour pressure  |
|                       |                              | Partitioning from LNAPL into groundwater                               | Groundwater flux and effective solubility                                    |
| Destructive processes | Transformation               | Abiotic degradation  | Hydrochemical environment  |
|                       |                              | Biodegradation   | Hydrochemical environment and microbiological potential (aerobic, anaerobic) |

#### 4.3.1. Dispersion

Dispersion is the mixing of water due to groundwater movement (advection) and aquifer physical heterogeneity. It depends only on aquifer properties and the scale of observation. On the microscopic scale dispersion is caused by three mechanisms:

1. Different velocities in a pore channel due to the drag exerted to the fluid by the roughness of the pore surface.
2. Different velocities due to differences in pore size along the flow path of the water molecules.
3. Tortuosity, branching and inter-fingering of pore channels that creates differences in the path length.

Dispersion has three components, represented in terms of a transverse vector in the y- and z-direction, and a longitudinal vector in the x-direction. In most cases the magnitude of longitudinal dispersion is several orders of magnitude greater than that of the transverse components. However in all cases and for a given velocity, the extent of dispersion is determined by the dispersivity (units of cm).

Dispersion leads to a reduction of dissolved GEO concentrations by simple dilution of the compounds with water that does not contain these, during transport along the flow path, but mass is conserved.

#### 4.3.2. Diffusion

Diffusion is the movement of particles due to molecular/atomic motion. It depends on contaminant properties and concentration gradients (described by Fick's law). In groundwater diffusion is generally unimportant relative to dispersion at most groundwater flow velocities (Wiedemeier et al, 1999), but may be important in very low permeability environments (e.g. clay strata) where advection is very limited or in dual porosity systems like chalk aquifers (Thornton et al, 2006). Diffusion acts to retard the transport of GEO in the subsurface.

For vapour phase transport diffusion is the most important process under natural conditions (ITRC, 2009). In the unsaturated zone transport of GEO will be influenced by diffusion of these chemicals within the internal structure of soil and aggregates.

#### 4.3.3. Sorption

Sorption is the physical adherence of a soluble compound to a solid surface, such as the soil/aquifer matrix. Sorption depends on properties of the sorbed compound (compound-specific parameters: solubility, hydrophobicity, octanol-water partitioning coefficient) as well as on the subsurface characteristics (subsurface-specific parameters: organic carbon content, clay mineral content, specific surface area, total porosity). Compound-specific sorption can be estimated from the  $K_{oc}$  (organic carbon partition coefficient) value for the organic chemical.

The log  $K_{oc}$  values for GEO range from 0.4 for TBA ( $K_{oc} = 2.5$  l/kg) to 1.6 for TAME ( $K_{oc} = 39$  l/kg) (**Table 1.2**).

MTBE, DIPE, TAEE, THxME and TBA show significantly weaker sorption than benzene to typical geological materials. The sorption characteristics of THxEE and TAME are similar to those of benzene (the log  $K_{oc}$  for benzene is approx. 1.5 – 2.1). The log  $K_{oc}$  value of ETBE ranges from 0.94 to 1.57.

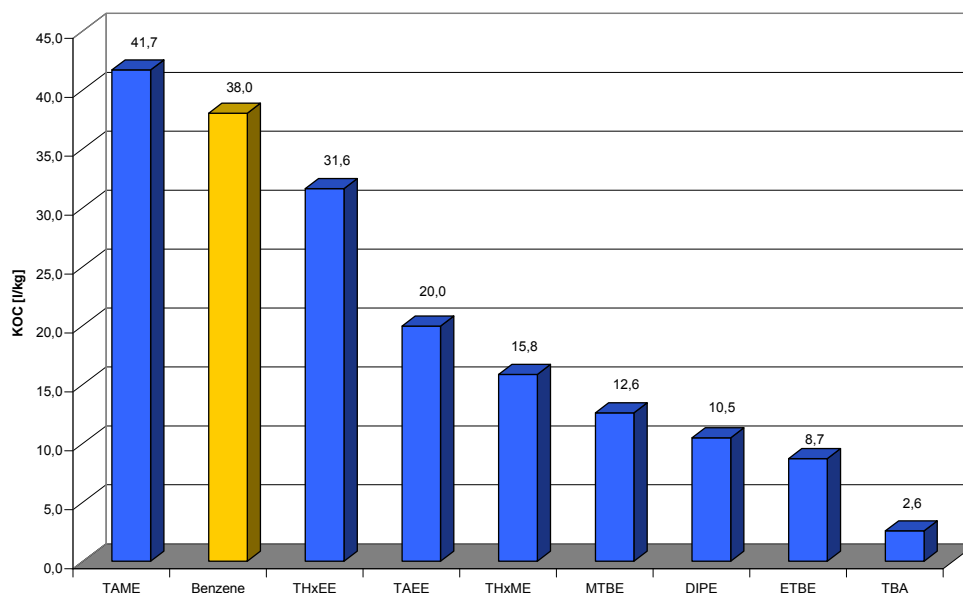
Compound-specific sorption has the following order:

TBA < MTBE < DIPE < THxME < TAEE < ETBE (at log  $K_{oc}$  1.57) < THxEE < TAME.

The influence of sorption processes on GEO transport in the subsurface is generally low, and GEO are typically transported in groundwater without significant retardation by sorption to subsurface media. All GEO except TAME sorb less to the soil matrix than benzene and are, therefore, more mobile in groundwater (**Figure 4.5**).



**Figure 4.5** Koc values for different GEO and benzene



The effect of sorption on GEO transport can be expressed by the retardation factor, which is the rate of movement of groundwater divided by the rate of movement of the GEO. The retardation factor (R) can be calculated:

$$R = 1 + (\rho/n) \times f_{oc} \times K_{oc}$$

R = Retardation factor (dimensionless)

$K_{oc}$  = organic carbon partition coefficient (l/kg)

$\rho$  = bulk density of aquifer sediment (assumed with 1.72 kg/l)

n = porosity (assumed with 0.35)

$f_{oc}$  = fraction of organic carbon in sediment (mg/mg) (range for aquifer sands is from 0.0001 (low) to 0.001 (median) to 0.01 (high) (Wiedemeier et al, 1999)

Calculated retardation factors due to sorption for different GEO are summarized in **Table 4.2**.

**Table 4.2** Retardation factors for GEO and benzene for different fractions of organic carbon in aquifer sediments

| Compound | Log KOC | $f_{oc} = 0.0001$<br>Low for aquifers | $f_{oc} = 0.001$<br>Median for aquifers | $f_{oc} = 0.01$<br>High for aquifers<br>Typical for soil |
|----------|---------|---------------------------------------|---|--|
| TBA      | 0,41    | 1,00                                  | 1,01                                    | 1,13   |
| ETBE     | 0,94    | 1,00                                  | 1,04                                    | 1,43   |
| MTBE     | 1,10    | 1,01                                  | 1,05                                    | 1,51   |
| DIPE     | 1,02    | 1,01                                  | 1,06                                    | 1,62   |
| THxME    | 1,20    | 1,01                                  | 1,08                                    | 1,78   |
| TAEe     | 1,30    | 1,01                                  | 1,10                                    | 1,98   |

| Compound | Log KOC | foc = 0.0001<br>Low for aquifers | foc =0.001<br>Median for aquifers | foc = 0.01<br>High for aquifers<br>Typical for soil |
|----------|---------|----------------------------------|-----------------------------------|---|
| THxEE    | 1,50    | 1,02                             | 1,16                              | 2,55  |
| Benzene  | 1,58    | 1,02                             | 1,19                              | 2,87  |
| TAME     | 1,62    | 1,02                             | 1,20                              | 3,05  |

#### 4.3.4. Volatilisation

The evaporation (volatilisation) of an organic compound dissolved in water is described by its temperature-dependent (dimensionless) Henry’s law constant (H). According to Henry’s Law the concentration of a compound in the gaseous phase is directly proportional to the concentration of the compound in the aqueous phase. The Henry’s law constant increases with increasing temperature. Compounds with a Henry’s law constant >0.05 readily evaporate from water (ITRC, 2005).

$$H = C_a/C_w$$

H = Henry’s law constant (dimensionless)

C<sub>a</sub> = Concentration in air (mg/l)

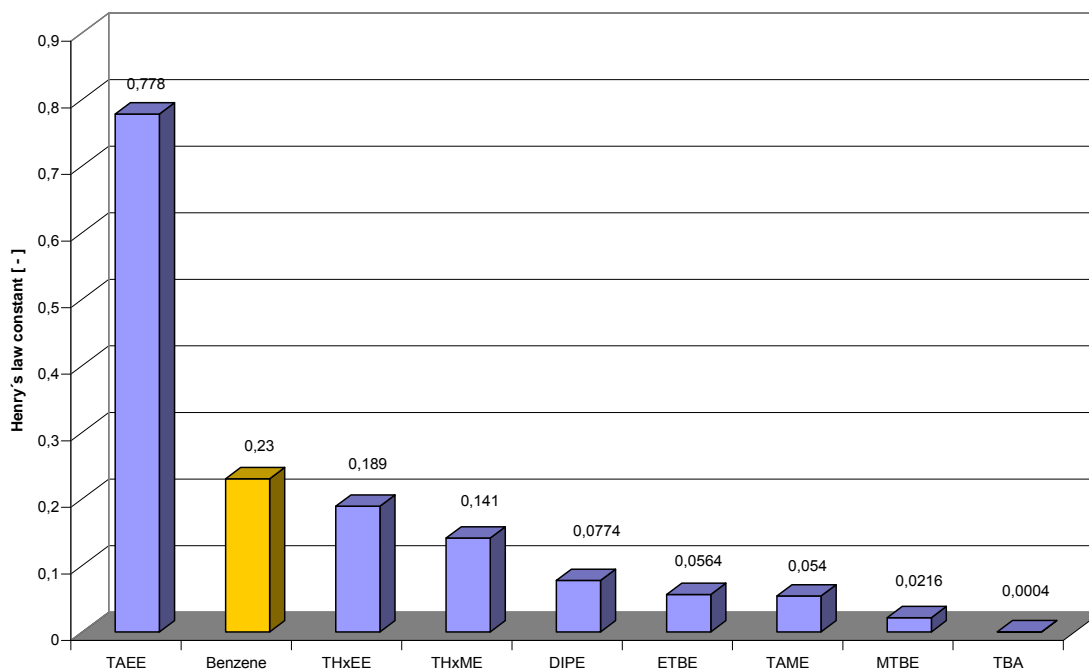
C<sub>w</sub> = Concentration in water (mg/l)

Henry’s law constants for GEO range from 0.778 for TAAE to 0.0004 for TBA, spanning almost 3 orders of magnitude (**Figure 4.6**). Of the GEO, only TAAE passes more easily from the aqueous phase to the gaseous phase than benzene (Henry’s law constant of approx. 0.23). The influence of Henry’s law constant on the GEO transport in the subsurface is generally low, and GEO do not readily partition from dissolved aqueous phase (groundwater) into the gas phase.

Henry’s law constant has the following order:

TBA < MTBE < TAME ≤ ETBE < DIPE < ThxME < ThxEE < TAAE.

**Figure 4.6** Henry's Law constant for different GEO and benzene



For risk analysis concerning vapour intrusion the differences in Henry's Law constants for GEO must be considered.

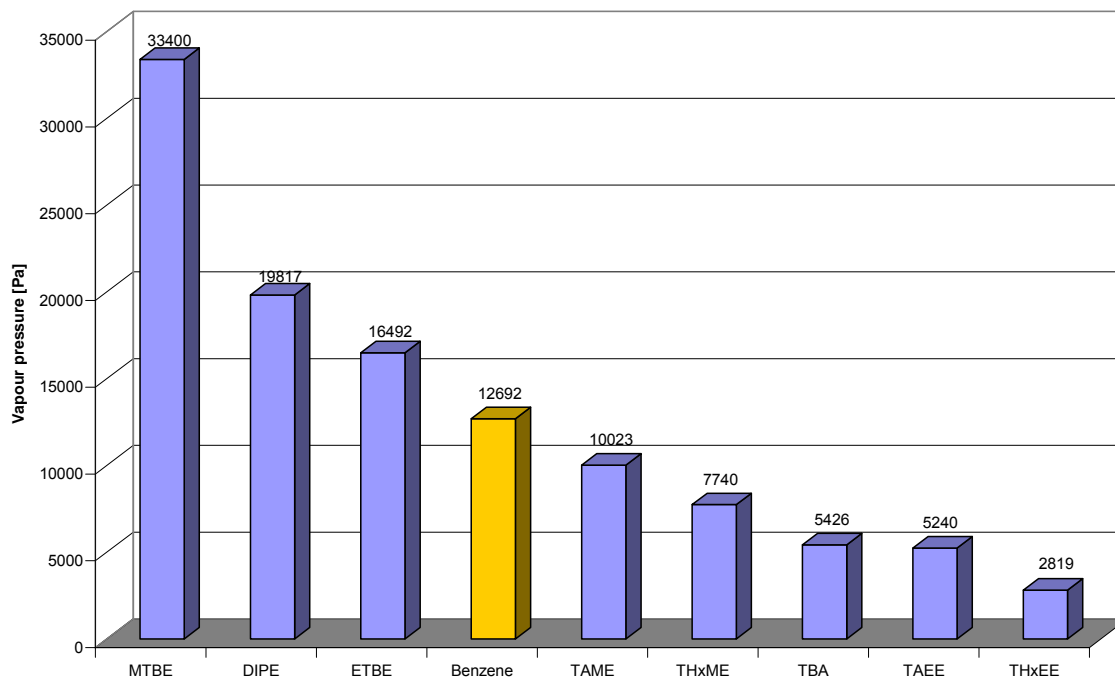
#### 4.3.5. Vapourisation

Vapourisation is influenced by the vapour pressure of each compound. Vapour pressure describes the tendency of a compound to evaporate and increases with temperature. In a mixture, the partial pressure of a volatile compound in equilibrium with gasoline corresponds to the pure phase vapour pressure multiplied by its mole fraction in gasoline (Schwarzenbach et al, 1993).

GEO are VOC (volatile organic compounds (boiling point <250 C°)) and have vapour pressure values ranging from approx. 2,800 Pa for THxEE up to approx. 17,600 Pa (at 10°C) for MTBE (**Figure 4.7**). MTBE has the highest vapour pressure and easily transfers into the gas phase.

The vapour pressures of MTBE, DIPE and ETBE are higher than the vapour pressure of benzene (vapour pressure approx. 12,000 Pa), and GEO generally partition readily from an organic liquid (e.g. gasoline) into the vapour phase. Vapour pressure reduces in the order of MTBE > DIPE > ETBE > TAME > THxME > TBA > TAE > THxEE.

**Figure 4.7** Vapour pressure for different GEO and benzene



For risk analysis concerning vapour intrusion the differences in vapour pressure for GEO must be considered.

**4.3.6. Partitioning from LNAPL into water**

When LNAPL is in contact with groundwater (and sometimes recharge water) dissolution into the aqueous phase occurs. Dissolution of GEO from LNAPL is the primary source of dissolved GEO in groundwater. Partitioning from LNAPL into water depends on the groundwater flux in contact with LNAPL and the GEO properties (solubility, effective solubility, age). In groundwater MTBE has the potential for the formation of relatively large plumes. These plumes are generally longer than BTEX plumes but shorter than plumes of light volatile chlorinated hydrocarbons (e.g. tetra- and trichloroethene and their decay products) formed under aerobic conditions (Stupp et al, 2007).

**Groundwater flux**

The primary factors controlling groundwater movement in the LNAPL zone (smear zone) are the LNAPL distribution, LNAPL/water saturation, the corresponding water relative permeability, the hydraulic conductivity and hydraulic gradient.

Under the LNAPL zone, where LNAPL saturation is zero, groundwater flux is simply governed by the hydraulic gradient and water-saturated hydraulic conductivity of the aquifer material. Above the groundwater potentiometric surface, in the unsaturated zone, the water pressure is less than atmospheric and water movement is primarily vertical.

Between the base of the LNAPL zone and the effective groundwater potentiometric surface groundwater movement is restricted to varying degrees by the presence of LNAPL that occupies a fraction of the pore space. Zones of higher LNAPL saturation imply lower water saturation and therefore lower relative permeability with respect to water.

For more detailed description about multiphase flow and partitioning from LNAPL we refer to API (2002) and ITRC (2009).

### **GEO properties**

The most important GEO property affecting the partitioning from LNAPL into water is the aqueous solubility. It is defined as the property of a compound to dissolve in water to reach an equilibrium concentration under standard conditions. Solubility is temperature dependant and increases with increasing temperature.

The aqueous solubility of pure-phase GEO ranges from 2,450 mg/l for DIPE up to approximately 50,000 mg/l for MTBE (**Figure 4.8**). Alcohols, such as TBA, are miscible with water in any ratio. The pure-phase solubility of all GEO is thus higher than the pure-phase solubility of benzene (approx. 1,800 mg/l).

For GEO present in a mixture with other organic compounds (e.g. gasoline), the effective solubility ( $S_i$ ) of each component compound is approximately described by Raoult's Law:

$$S_i = S * X_i$$

$S_i$  = effective solubility of chemical i in a mixture (mg/l)

S = pure-phase aqueous solubility of chemical (mg/l)

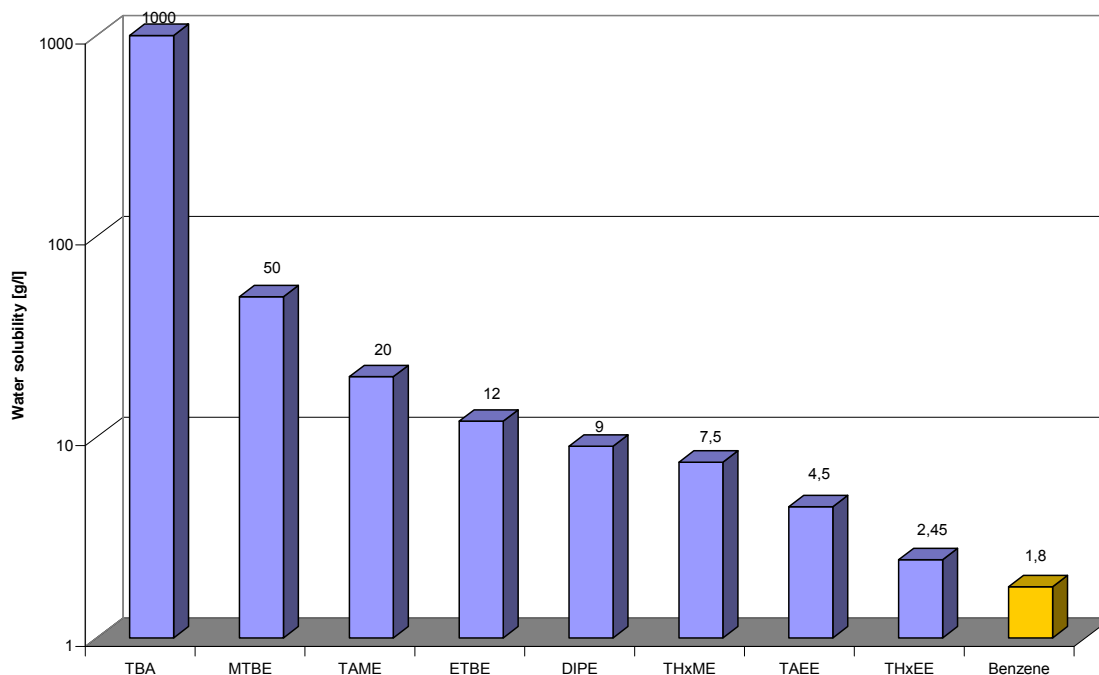
$X_i$  = mole fraction of chemical i in a mixture (dimensionless)

The pure-phase solubility of GEO decreases in the order: TBA > MTBE > TAME > ETBE > DIPE > THxME > TAEE > THxEE.

Example: In a gasoline with a MTBE content of approx. 1% per moles, or mole fraction of 0.01 (which in case of gasoline is roughly equivalent to 1% by volume or 1% by mass), the MTBE would have an effective equilibrium solubility of approx. 500 mg/l, while the pure MTBE would have a solubility of approx. 50,000 mg/l (Moyer, 2003).

The higher relative solubility of GEO explains the general observation that GEO are released quickly from an LNAPL source and form larger plumes with higher concentration than other components (e.g. BTEX) in gasoline.

**Figure 4.8** Aqueous solubility of different GEO and benzene



**4.3.7. Abiotic degradation**

Abiotic degradation is the transformation of a compound into a simpler compound without the influence of biological agents.

In the atmosphere hydroxyl radicals (free OH<sup>-</sup>) quickly react with MTBE (API, 2007). Smith et al (1991) determined a half-life of 3.9 days for MTBE in the atmosphere. Therefore, the transformation of MTBE by OH<sup>-</sup> radicals in the atmosphere is an important process that can remove substantial amounts of MTBE from the environment.

In the subsurface there are no significant abiotic degradation processes for MTBE in groundwater (Environment Agency UK, 2002). Therefore abiotic degradation of GEO will not be further considered for the unsaturated zone, groundwater and hyporheic zone.

**4.3.8. Biodegradation**

Under aerobic conditions, micro-organisms can use MTBE, ETBE, TBA, DIPE and TAME as the exclusive carbon and energy source for metabolism (Debor & Bastiaens, 2007; Müller et al, 2008; Shah et al, 2009). Aerobic biodegradation for MTBE, TBA and TAME has been demonstrated at many sites (**Table 4.3**).

Anaerobic biodegradation of MTBE to CO<sub>2</sub> was proven in microcosms with surface water sediments under sulphate-reducing, iron-reducing, manganese-reducing and nitrate-reducing conditions (Bradley et al, 2001). Furthermore, the biodegradation of MTBE to TBA under methanogenic conditions was documented (Bradley et al,

2001). Anaerobic biodegradation of TBA to CO<sub>2</sub> was also proven with surface water sediments for nitrate-reducing, iron-reducing, manganese-reducing and sulphate-reducing conditions (Bradley et al, 2002).

ETBE biodegradation under anaerobic conditions in microcosms was shown by Yeh & Novak (1994). TAME biodegradation under methanogenic conditions in surface water sediments was shown by Somsamak et al, 2005. **Table 4.3** gives a summary of sites where biodegradation of MTBE, TBA, TAME and ETBE was demonstrated. No data are available on the anaerobic biodegradation of other GEO.

**Table 4.3** Number of sites with observed biodegradation at field scale and in microcosms (predominantly intergranular porosity aquifer material)

| Redox conditions  | MTBE | TBA  | TAME | ETBE |
|-------------------|------|------|------|------|
| Aerobic           | 35   | 11   | 4    | n.d. |
| Nitrate reducing  | 10   | 4    | n.d. | n.d. |
| Fe-reducing       | 5    | n.d. | n.d. | n.d. |
| Mn-reducing       | 3    | 2    | n.d. | n.d. |
| Sulphate reducing | 4    | 5    | n.d. | n.d. |
| Methanogenic      | 21   | 8    | 1    | n.d. |
| Anaerobic*        | 19   | 14   | 1    | 1    |

n.d. = no data

\* not specified anaerobic conditions (not included in sum of above mentioned redox conditions)

Biodegradation is the biologically-mediated transformation of a compound. Generally biodegradation is often described by a first-order process, although other kinetic descriptions also exist. First-order biodegradation rates are indicated in the present report. First-order rate constants and half-lives are related by the following expression.

$$\lambda = T_{1/2} / \ln 2$$

Where  $\lambda$  is the first-order rate constant (units of 1/time) and  $T_{1/2}$  is the half life (units of time). The half-life is the time period in which the contaminant concentration is reduced by 50%. Typical first-order biodegradation rates are converted into a corresponding half-life in **Table 4.4**.

**Table 4.4** Comparison of first-order degradation rates to half-life

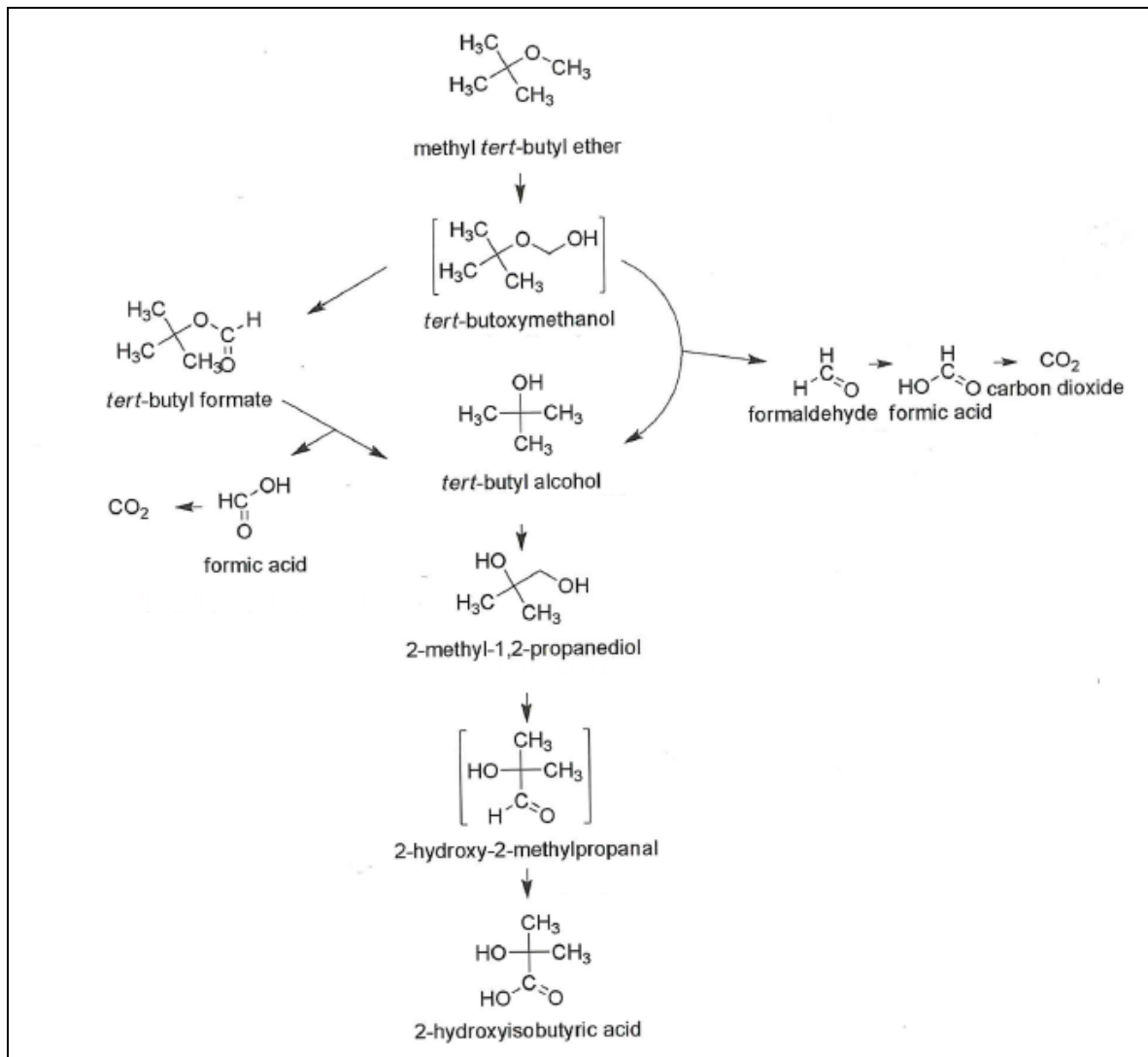
| First-order biodegradation rates | Half-life                            |
|----------------------------------|--------------------------------------|
| 0.0001 d <sup>-1</sup>           | 6931 days; approx. 19 years          |
| 0.001 d <sup>-1</sup>            | 693 days; approx. 1.9 years          |
| 0.01 d <sup>-1</sup>             | 69 days; approx. 0.19 years 10 weeks |
| 0.1 d <sup>-1</sup>              | 6,9 days; approx. 0.019 years 1 week |

A distinction is made between aerobic and anaerobic biodegradation. During aerobic biodegradation, organic compounds (electron donors) are transformed using oxygen as an electron acceptor. Aerobic processes are thermodynamically the most favourable (i.e., release the greatest energy for the microbe) and dominate as long as there is sufficient oxygen *in situ* (generally taken as dissolved oxygen above 1 to 2.0 mg/l) (Environment Agency, 2000; Finneran and Lovley, 2003). There is no further information available on other GEO.

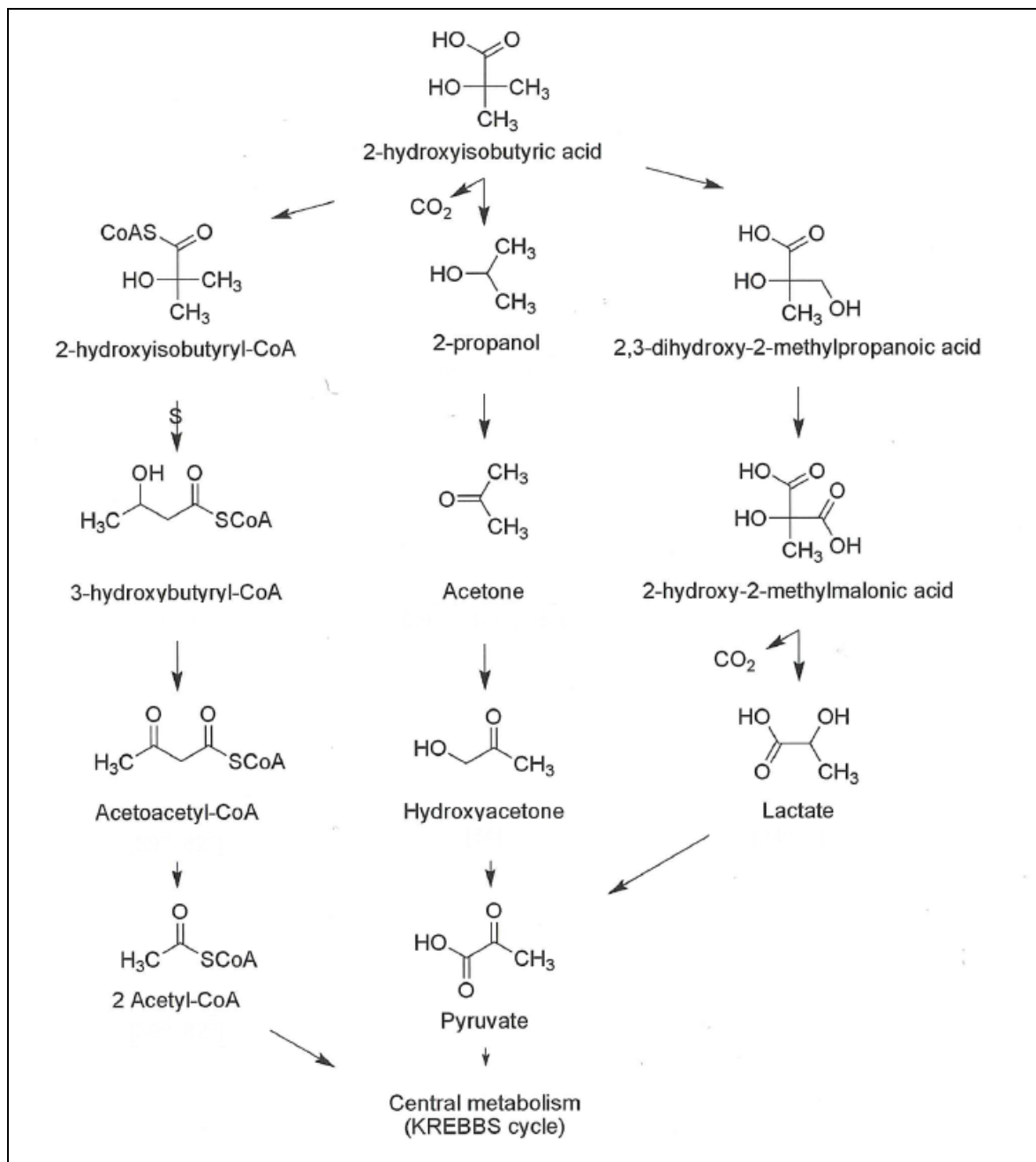
The biodegradation pathway of MTBE has been analysed under aerobic conditions and with various bacterial cultures. Current findings are presented in **Figure 4.9** and **4.10**. A distinction is made between an upper pathway of MTBE transformation to 2-hydroxyisobutyric acid (HIBA) (**Figure 4.9**) and a lower pathway of HIBA transformation to CO<sub>2</sub> (**Figure 4.10**). MTBE is biodegraded to HIBA through TBA, with the production of formaldehyde. For further biodegradation of HIBA to CO<sub>2</sub> and bacterial biomass, three different pathways have been described by Steffan et al (1997). For more details of the aerobic biodegradation of MTBE we refer to Debor & Bastiaens (2007). Recently, the enzymes and genes that are responsible for MTBE-biodegradation have been reviewed by Müller et al (2007) and Ferreira et al (2006), respectively.



**Figure 4.9** Reported aerobic biodegradation pathways to transform MTBE/TBA into HIBA (Debor & Bastiaens, 2007)

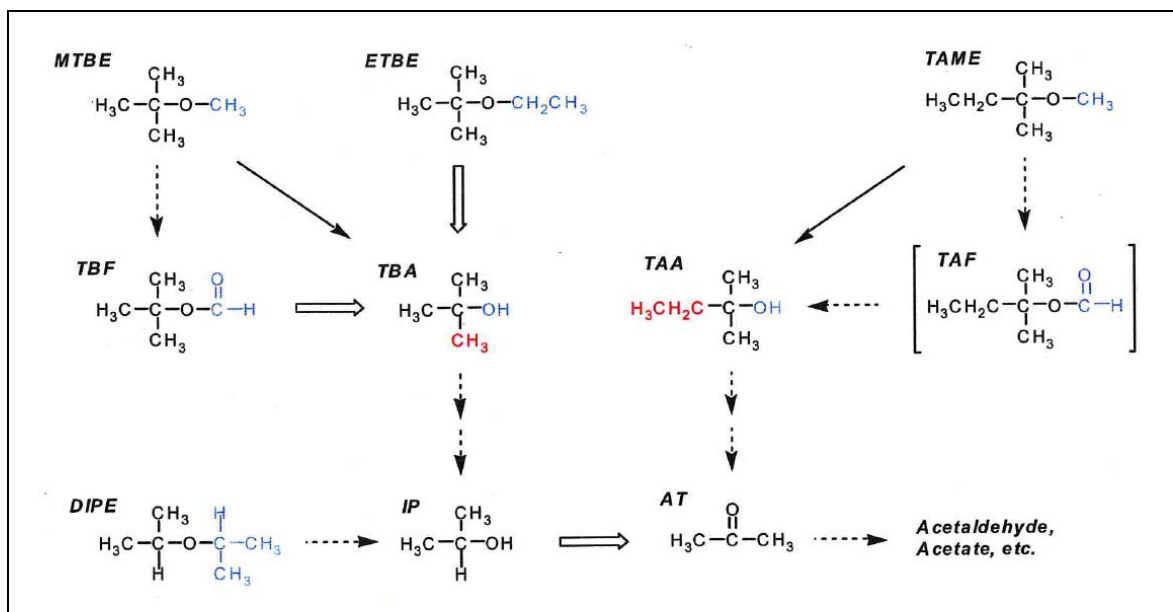


**Figure 4.10** Reported aerobic biodegradation pathways to convert HIBA to CO<sub>2</sub>; s: suggested, but not detected (Debor & Bastiaens, 2007)



Church et al (2000) examined the biodegradation pathways of MTBE, ETBE, TAME and DIPE with the bacteria strain PM1 (**Figure 4.11**). The research concluded from the similarity in biodegradation rates of TAME, ETBE, DIPE, TBA, TAA and MTBE and the formation of metabolites that similar enzyme systems were involved for these reactions. There is no information available on the biodegradation pathways of the other GEO.

**Figure 4.11** Proposed biodegradation pathway of fuel oxygenates by PM1. Open arrows: transformations that were directly observed (Church et al, 2000). Solid arrows: observed, but may occur via intermediates. Dashed arrows: hypothesized steps.



Anaerobic biodegradation occurs using dissolved and mineral-based electron acceptors such as nitrate, ferric iron ( $\text{Fe}^{3+}$ ), manganic manganese ( $\text{Mn}^{4+}$ ), sulphate, or under methanogenic conditions. According to the thermodynamic calculations of Schmidt et al (2004), biodegradation of MTBE and TBA is feasible under aerobic, nitrate-reducing, sulphate-reducing, iron-reducing and methanogenic conditions. However, in general anaerobic biodegradation of GEO appears to occur at rates which are much slower than those for corresponding aerobic conditions (Somsamak, et al, 2005; Shah et al, 2009).

Little is known about the biodegradation pathways of MTBE or TBA under anaerobic conditions, as anaerobic bacteria strains have so far not been cultivated. Stable isotope data from methanogenic (Kuder et al, 2005) and sulfidogenic (Somsamak et al, 2005, 2006) microcosms suggest anaerobic biodegradation is related to cleavage of the oxygen-methyl carbon bond, speculatively attributed to acetogenic bacteria (Somsamak et al, 2006).

Biodegradation at field-scale under aerobic, nitrate-reducing, iron-reducing, manganese-reducing, sulphate-reducing and methanogenic conditions is discussed in Chapters 4.4, 4.5 and 4.6. In addition, Chapter 4.5.5.3 considers special issues regarding the influence of other contaminants on GEO biodegradation. The occurrence of lag times before onset of biodegradation is discussed in Chapter 4.5.5.4 and isotopic investigation methods for evaluation of biodegradation are presented in Chapter 4.5.5.5.

#### 4.4. LITERATURE REVIEW – UNSATURATED ZONE

The following issues are important for risk assessment:

- The vapour-to-indoor exposure pathway should be assessed near a GEO release, where there is potential for exposure of a receptor.
- Existing guidelines for the assessment of the vapour-to-indoor pathway are available for petroleum compounds and chlorinated VOC. Specific guidance for GEO does not exist, but existing protocols can be adapted to allow evaluation of GEO risks.
- GEO in contact with water will quickly transfer into the water phase.
- GEO biodegradation is hard to investigate and might be too slow to prevent GEO transfer to groundwater or into the vapour phase.

##### 4.4.1. Introduction

Specific literature on the unsaturated zone in relation to GEO fate and transport is sparse. Several publications describe MTBE transport from the soil surface through the unsaturated zone into groundwater (Stupp et al, 2008; API, 2007; ITRC, 2005; Wilson, 2003).

The unsaturated zone contaminated with GEO consists of the four different phases gas, water, soil and LNAPL. In the following chapters the most relevant processes for fate and transport of GEO in the gas, soil and water phase are described.

##### 4.4.2. GEO transfer into the gas phase

Most GEO have relatively high vapour pressures (Chapter 4.3.5). This indicates that in the unsaturated zone near a GEO release potential vapour intrusion exposure pathways should be assessed.

Due to relatively low Henry's Law constants for all GEO, the transfer of dissolved GEO in water into the gas phase is of minor importance.

Detailed guidance concerning the assessment of the vapour exposure pathways for GEO, taking into account the high vapour pressures, low Henry's Law constants and the high solubility in water of the GEO, does not exist, but existing guidance on petroleum compounds and chlorinated VOCs may be adapted.

A site-specific approach for assessing vapour exposure pathways was first addressed in API (1998). USEPA published a tool to conservatively evaluate the risk profile associated with site specific vapour pathways (USEPA, 2002). However, the tool may not be an appropriate mechanism for screening the vapour pathway at underground storage tank (UST) sites, due to the associated level of conservatism. Specifically for petroleum hydrocarbon release sites, API (2005) provides guidance of collecting soil gas samples to assess the vapour exposure pathways, and more recent research and model development such as BioVapour (API, 2010) provide a toolkit for more representative risk-assessment of petroleum compounds.

#### **4.4.3. GEO transfer into the water phase**

The transfer of GEO is described in Chapter 4.3.6 and in the conceptual model (Chapter 4.2).

#### **4.4.4. GEO transfer to the soil phase**

The particulate organic matter content, represented by the fraction of organic carbon ( $f_{oc}$ ), is generally higher in the soil/subsoil of the unsaturated zone than in the saturated zone (aquifer), although exceptions are possible, such as in the Netherlands, where peat is often found in the saturated zone. Therefore, retardation in the unsaturated zone due to sorption onto particulate organic matter is generally higher than in the saturated zone. GEO retardation by sorption to natural organic matter in the unsaturated zone is, nevertheless, a weak process that results in only slightly increased travel times through the unsaturated zone when compared with water transport.

#### **4.4.5. GEO biodegradation**

When GEO are transported as dissolved components of gasoline, they are usually present at the leading edge of the contaminant front (if in the unsaturated zone) or contaminant plume (if in the saturated zone) due to their high solubility in water, low retardation, and high vapour pressure. Therefore, GEO transported down through the unsaturated zone may have the most contact with oxygen in the soil vapour and air. Under these conditions, aerobic biodegradation of MTBE is feasible (ITRC, 2005). Transverse to the direction of flow there is limited dispersion and chromatographic separation of BTEX and GEO so that oxygen is available for biodegradation of both contaminants. Biodegradation in the unsaturated zone can also occur, particularly if atmospheric exchange allows the continued introduction of oxygen (in air) to promote aerobic respiration (Baehr et al, 2001).

Lawrence and Erickson (2004) analysed MTBE biodegradation in the unsaturated zone and determined biodegradation rates of 0.5 – 1.0 mg/(kg\*day) in the soil. They concluded that the unsaturated zone may provide a highly efficient sink for MTBE. In contrast, Pasteris et al (2002) concluded from a lysimeter experiment with 13 gasoline fuel components and MTBE that biodegradation of MTBE in the unsaturated zone is too slow to prevent MTBE vapour migration to groundwater. It can be concluded that biodegradation in the unsaturated zone is difficult to investigate and, while it is likely to occur wherever oxygen is present, a reliable estimate of the biodegradation rate has to be determined on a site-specific basis.

### **4.5. LITERATURE REVIEW – GROUNDWATER**

The following issues are important for risk assessment:

- Vapourisation only takes place in the unsaturated zone.
- Vapour exposure pathways should be checked although vapourisation is of minor importance.
- A conceptual model in respect to LNAPL dissolution and plume development should be developed

- Biodegradation is widely observed under aerobic conditions in aquifers but less so under anaerobic conditions for MTBE.
- Biodegradation of ETBE, TBA, TAME and DIPE occurs.
- Compound-specific isotope analysis (CSIA) is a valuable investigation method for assessment of GEO biodegradation.

#### **4.5.1. Introduction**

The fate and transport in particular of MTBE and TBA in groundwater has been studied quite extensively. Most of the literature focuses on MTBE and TBA biodegradation in different hydrogeological (aquifer settings) and hydrochemical environments (redox-conditions). Another focus of the literature is directed towards the microbiology of MTBE and TBA biodegradation.

The saturated zone contaminated with GEO consists of the three different phases: water, soil and LNAPL. In the following chapters the most relevant processes for fate and transport of GEO are described.

#### **4.5.2. GEO transfer into the gas phase**

Due to the relatively low Henry's Law constants for all GEO, the transfer of dissolved GEO in water into the gas phase is of minor importance in respect to the vapour-to indoor-air exposure pathway. However, for risk assessment the vapour-to indoor-air exposure pathway should be checked.

The transfer from dissolved GEO into the vapour phase is discussed for the unsaturated zone in chapter 4.4.2.

#### **4.5.3. GEO transfer to groundwater**

Dissolution of GEO from LNAPL is the primary source of dissolved GEO in groundwater. The transfer of GEO into groundwater is described in Chapter 4.3.6 and in the conceptual model (Chapter 4.2).

#### **4.5.4. GEO transfer to the soil phase**

Sorption of GEO in groundwater to particulate organic matter depends mainly on the organic carbon content of the aquifer minerals in the saturated zone. This is represented in models by the fraction of organic carbon ( $f_{oc}$ ), and typically ranges from 0.0001 to 0.01 (mean 0.001) (Wilson, 2003). The  $f_{oc}$  is obtained by dividing the measured weight% of particulate organic carbon per unit mass of soil or aquifer material by 100, and is controlled by the geological strata, its depositional environment and post-depositional diagenesis.

The influence of sorption processes on GEO transport in the subsurface is generally low, and GEO are typically transported in groundwater without significant retardation (Chapter 4.4.4).

#### 4.5.5. GEO Biodegradation

In 1988 a field experiment was carried out at CFB Borden in Canada to investigate the biodegradation of MTBE in groundwater. The first analysis of the field data (Hubbard et al, 1994) suggested that MTBE was recalcitrant in groundwater. Based on these and other results MTBE was thought for a number of years to be persistent in groundwater. However, further sampling in 1995 at the same site showed that after 3,000 days only 3% of the original MTBE mass was present (Schirmer and Barker, 1998; Schirmer et al, 1999). Using microcosms (laboratory investigations) to evaluate aerobic biodegradation of MTBE it could be verified that this mass reduction was primarily the result of biodegradation.

Many other investigations have demonstrated that biodegradation of MTBE and TBA varies from one gasoline release site to another. Kane et al (2003) constructed microcosms with material from seven MTBE release sites in California. MTBE was biodegraded in sediment from only three of the sites. In sediment from the other four sites, MTBE was not biodegraded within the period of incubation (170 days to 350 days depending on the site). When Schirmer et al (1999) used sediment from the Borden aquifer to construct laboratory microcosms, acclimation to biodegrade MTBE was a rare event; only 3 of 40 microcosms were acclimated after 20 months of incubation. However, once acclimation occurred in laboratory microcosms, biodegradation was rapid and extensive.

The prospects for *in situ* biodegradation of MTBE by native micro-organisms (bacteria) appears to be related to the age of the release, the time available for acclimation of the native micro-organisms to MTBE and to the seepage velocity of the ground water (USEPA, 2005).

In the following chapters biodegradation will be discussed primarily in the context of field investigations of aquifers. Investigations of aquifers and groundwater can be done on a field-scale basis primarily with groundwater sampling or at laboratory-scale using experiments (microcosms and columns) with site-specific sediment and groundwater samples.

##### 4.5.5.1. Biodegradation in aquifers under aerobic conditions

MTBE has been shown to biodegrade in aquifer sediments under aerobic conditions (Salanitro et al, 2000; Wilson et al, 2002; Hunkeler et al, 2001; Kane et al, 2001; Schirmer et al, 2003; DeVuall et al, 2004; Shah et al, 2009). TBA, which can be an intermediate metabolite of MTBE, has also been shown to biodegrade in aquifer sediments under aerobic conditions (Hunkeler et al, 2001; Kane et al, 2001; Wilson et al, 2002; Schirmer et al, 2003).

When GEO are transported together with gasoline, oxygen is depleted by bacteria, (bacteria very quickly consume oxygen for the biodegradation of BTEX) so that anaerobic redox-conditions generally prevail. Consequently, aquifers that are contaminated with GEO and which have aerobic redox-conditions in the plume are rare.

Aerobic biodegradation of MTBE has been investigated at the field-scale on two North American sites very intensively. First-order biodegradation rates from studies on the sites at Port Hueneme, USA and CFB Borden, Canada are listed in **0**. For the field-scale investigation biodegradation rates of  $0.016 \text{ d}^{-1}$  for Port Hueneme and  $0.0012 \text{ d}^{-1}$  for Borden were estimated. First-order biodegradation rates determined in the laboratory for both sites are higher than the ones obtained at field-scale (Port

Hueneme 0.11 d<sup>-1</sup>; Borden 0.055 d<sup>-1</sup>). The biodegradation rate estimated using laboratory column experiments for Borden is a little lower than the one from the field (0.00071 d<sup>-1</sup>). At Port Hueneme compound-specific isotope analysis (CSIA) was also carried out (Hunkeler et al, 2001). The results are presented in chapter (4.5.5.5).

Biodegradation of MTBE in a dual porosity aerobic aquifer (UK Chalk) was investigated by Thornton et al (2006). Aerobic biodegradation rates estimated using microcosms ranged from 0.0025 d<sup>-1</sup> to 0.0075 d<sup>-1</sup>. Later laboratory microcosm studies using groundwater and aquifer material from this site showed that aerobic MTBE biodegradation occurred at rates which varied as a function of MTBE concentration, up to 5 mg/l (Shah et al, 2009).

**Table 4.5** Results of MTBE biodegradation in aquifer sediments under aerobic conditions

| Site                | Investigation Scale | Redox-Conditions | Biodegradation rate d <sup>-1</sup> | Reference                              |
|---------------------|---------------------|------------------|-------------------------------------|--|
| Port Hueneme, USA   | Field scale         | Aerobic          | 0,016*                              | Salanitro et al, 2000                  |
| Port Hueneme, USA   | Microcosms          | Aerobic          | 0.11*                               | Salanitro et al ,2000                  |
|                     |                     |                  |                                     |  |
| Borden, Canada      | Field scale         | Aerobic          | 0.0012                              | Schirmer et al, 1999                   |
| Borden, Canada      | Microcosms          | Aerobic          | 0.04 – 0.07<br>(mean 0.055)         | Schirmer et al, 2003                   |
| Borden, Canada      | Microcosms          | Aerobic          | (1)                                 | Hunkler et al, 2001                    |
| Borden, Canada      | Column              | Aerobic          | 0.00071                             | Schirmer et al, 2003                   |
|                     |                     |                  |                                     |  |
| St. Albans, England | Microcosms          | Aerobic          | 0.0025 – 0.0075<br>(mean 0.005)     | Thornton et al, 2006                   |
|                     |                     |                  |                                     |  |
| Fairfax Conty, Va   | Microcosms          | Aerobic          | 0.063                               | Zoeckler et al, 2003                   |
| Fairfax Conty, Va   | Microcosms          | Aerobic          | 0.037                               | Zoeckler et al, 2003                   |
|                     |                     |                  |                                     |  |
| Vandenberg AFB, CA  | Microcosms          | Aerobic          | 0.03 – 0.15<br>(mean 0.09)          | Wilson et al, 2002                     |
|                     |                     |                  |                                     |  |
|                     | Microcosms          | Aerobic          | Mean 0.06                           | Calculated from above mentioned values |

\* rate in Wilson 2003

(1) CSIA investigations Hunkeler et al 2001, -1.52 +- 0.06 per mille, -1.97+-0.05 ‰

Kolhatkar et al, 2000 determined field-scale biodegradation rates of 0.011 d<sup>-1</sup> and laboratory values of 0.014 d<sup>-1</sup> for aerobic to nitrate reducing conditions. The results of MTBE biodegradation in microcosms with aquifer sediments are shown in **Table 4.5**. The mean first-order biodegradation rate in microcosms is 0.06 d<sup>-1</sup>.



The results of TBA biodegradation in microcosms with aquifer sediments are summarized in **Table 4.6**. The mean first-order biodegradation rate is 0.1175 d<sup>-1</sup>.

**Table 4.6** Results of laboratory investigations of TBA biodegradation in aquifer sediments under aerobic conditions

| Site                | Investigation Scale | Redox-Conditions | Biodegradation rate d <sup>-1</sup> | Reference            |
|---------------------|---------------------|------------------|-------------------------------------|----------------------|
| Borden site, Canada | Column              | Aerobic          | 0.00073                             | Schirmer et al, 2003 |
| Borden site, Canada | Microcosms          | Aerobic          | 0.12                                | Schirmer et al, 2003 |
| Borden site, Canada | Microcosms          | Aerobic          | (1)                                 | Hunkler et al, 2001  |
| Palo Alto, CA       | Microcosms          | Aerobic          | 0.1 – 0.13*<br>(mean 0.115)         | Kane et al, 2001     |
|                     |                     |                  | Mean 0.1175                         |                      |

\* rate in Wilson 2003

(1) CSIA investigations Hunkeler et al 2001

The results of **Table 4.5** and **Table 4.6** indicate that the first-order biodegradation rate of TBA is typically greater than the biodegradation rate of MTBE in aquifer sediments under aerobic conditions. A general conclusion concerning the accumulation of TBA during MTBE biodegradation cannot be drawn. However the relative biodegradation rates imply that TBA will be biodegraded faster than MTBE in groundwater. Transient accumulation of TBA was found in several studies under strongly anaerobic conditions (e.g., Schirmer et al, 2003; Hunkler et al, 2001 and Kane et al, 2001), but biodegradation occurred following a return to aerobic conditions. TBA was never reported to be recalcitrant in aquifer or surface water sediments under aerobic conditions (Bradley et al, 2002; Schmidt et al, 2004).

Aerobic biodegradation of ETBE by the bacteria strains L108 and IFP2001 has been described (Rosell et al, 2007). Furthermore, ETBE biodegradation has been demonstrated with the pure culture PM1 (Church et al, 2000). Purswani et al (2008) and Steffan et al (1997) showed ETBE biodegradation for a number of bacteria strains.

Aerobic TAME biodegradation by bacterial strains was demonstrated by Purswani et al (2008), Steffan et al (1997) and Church et al (2000). Aerobic biodegradation of TAME has also been demonstrated to occur simultaneously with MTBE in laboratory microcosms, for representative groundwater concentrations of both GEO (Shah et al, 2009).

Aerobic DIPE biodegradation was demonstrated by Hernandez-Perez et al (2001) with the bacteria strain *Gordonia terrae*. In addition, Church et al (2000) observed DIPE biodegradation with the pure culture PM1.

Information about biodegradation of other GEO in aerobic aquifers is currently not available.

**4.5.5.2. Biodegradation in aquifers under anaerobic conditions**

When GEO are transported with gasoline in groundwater, dissolved, oxygen is depleted by bacteria, (bacteria very quickly consume the available oxygen for biodegradation of BTEX) so that anaerobic redox conditions prevail. Therefore,

knowledge about anaerobic biodegradation of GEO is very important. Biodegradation of MTBE and TBA in anaerobic aquifers or under anaerobic conditions in plumes does not occur at all sites, as observed in numerous laboratory investigations (Mormille et al, 1994; Somsamak et al, 2001; Bradley et al, 1999; Bradley et al, 2002; USEPA, 2007; Shah et al, 2009; Thornton et al, 2011).

Furthermore in field-scale investigations several redox conditions may develop simultaneously. Usually redox-conditions vary spatially and temporally in an aquifer.

Many reports demonstrate MTBE biodegradation in aquifer sediments under nitrate-reducing conditions (Bradley et al, 2001a; Bradley et al, 2001b), sulphate-reducing conditions (Bradley et al, 2001a; Somsamak et al, 2001), iron-reducing conditions (Landmeyer et al, 1998; Bradley et al, 2001b; Finneran and Lovley, 2003) and methanogenic conditions (Mormille et al, 1994; Wilson et al, 2000; Bradley et al, 2001b; Kolhatkar et al, 2002; Somsamak et al, 2005; Wilson et al, 2005a).

TBA has been shown to biodegrade under nitrate-reducing, sulphate-reducing, manganese-reducing and methanogenic conditions (Bradley et al, 2002; Yeh and Novak, 1994; DeVaul et al, 2003; Wilson, et al, 2005a; USEPA, 2007).

ETBE biodegradation is reported to occur in anaerobic aquifer sediments (Yeh and Novak, 1994). In contrast, Somsamak et al (2001) found no indication of ETBE biodegradation in surface water sediments under nitrate-, iron-, sulphate-reducing conditions or methanogenic conditions.

TAME biodegradation has been documented in surface water sediments under sulphate-reducing conditions (Somsamak et al, 2001) and under general anaerobic conditions (Landmeyer et al, 2010). Experiments reported by Somsamak et al (2001) indicated no TAME biodegradation under nitrate, iron-reducing and methanogenic conditions in surface water sediments.

Biodegradation rates for anaerobic conditions in aquifer sediments were evaluated from the literature. First-order laboratory-scale and field-scale biodegradation rates for MTBE and TBA are summarized in **Table 4.7** to **Table 4.9**, respectively.

The results of laboratory-scale investigations of MTBE biodegradation rates in aquifer sediments under aerobic to nitrate-reducing, iron-reducing and methanogenic conditions are presented in **Table 4.7** Furthermore mean values are calculated for each redox-condition with at least three values. A mean value for aerobic to nitrate-reducing conditions is  $0.0041 \text{ d}^{-1}$  and for methanogenic conditions is  $0.0199 \text{ d}^{-1}$ .

**Table 4.7** Results of laboratory investigations of MTBE biodegradation in aquifer sediments under anaerobic conditions

| Site                | Investigation Scale | Redox-Conditions   | Biodegradation rate d <sup>-1</sup> | Reference                      |
|---------------------|---------------------|--------------------|-------------------------------------|--------------------------------|
| Borden, Ontario     | Microcosms          | Aerobic or Nitrate | 0,00071*                            | Church et al, 2000             |
| Farmington Hill, Mi | Microcosms          | Aerobic or Nitrate | 0,00110*                            | Church et al, 2000             |
| Turnersville, NJ    | Microcosms          | Aerobic or Nitrate | 0,00099*                            | Church et al, 2000             |
| Amoco Site, MI      | Microcosms          | Aerobic or Nitrate | 0,01370                             | Javanmardian and Glasser, 1997 |
|                     |                     |                    | Mean 0,0041                         |                                |
| Beaufort, SC        | Microcosms          | Iron- reducing     | 0,00016                             | Landmeyer et al, 1998          |
|                     |                     |                    |                                     |                                |
| Empire, MI          | Microcosms          | Sulphate-reducing  | Yes, no rate                        | Mormille et al, 1994           |
|                     |                     |                    |                                     |                                |
| Elizabeth City, NC  | Microcosms          | Methanogenic       | 0,00822                             | Wilson et al, 2000             |
| Empire, MI          | Microcosms          | Methanogenic       |                                     | Mormille et al, 1994           |
| Pasippany, NJ       | Microcosms          | Methanogenic       | 0,03151                             | Kolhatkar et al, 2002          |
|                     |                     |                    | Mean 0,0199                         |                                |

\* rate in Wilson 2003

The evaluation of the literature for field-scale investigation of MTBE biodegradation rates shows a broad range only for methanogenic conditions, summarized in **Table 4.8**. The mean field-scale biodegradation rate for MTBE under methanogenic conditions is 0.013 d<sup>-1</sup>. Borden et al (1997) report a biodegradation rate for aerobic to nitrate reducing conditions of 0.001 d<sup>-1</sup>. Wilson (2003) reported a mean value of 0.00274 d<sup>-1</sup> for 10 measured rates for anaerobic conditions.

**Table 4.8** Results of field-scale investigations of MTBE biodegradation in aquifer sediments under anaerobic conditions

| Site               | Investigation Scale | Redox-Conditions | Biodegradation rate d <sup>-1</sup> | Reference             |
|--------------------|---------------------|------------------|-------------------------------------|-----------------------|
| Elizabeth City, NC | Field scale         | Methanogenic     | 0,00822                             | Wilson et al, 2000    |
| New York           | Field scale         | Methanogenic     | 0,02986                             | Kolhatkar et al, 2000 |
| Florida            | Field scale         | Methanogenic     | 0,01973                             | Kolhatkar et al, 2000 |
| Pennsylvania       | Field scale         | Methanogenic     | 0,02685                             | Kolhatkar et al, 2000 |
| Long Island, NY    | Field scale         | Methanogenic     | 0,01425*                            | Kolhatkar et al, 2000 |
| Pasippany, NJ      | Field scale         | Methanogenic     | 0,00115*                            | Kolhatkar et al, 2000 |
| Washington, DC     | Field scale         | Methanogenic     | 0,00118*                            | Kolhatkar et al, 2000 |
| NJ                 | Field scale         | Methanogenic     | 0,00156                             | Kolhatkar et al, 2000 |
| PA-2               | Field scale         | Methanogenic     | 0,00115                             | Kolhatkar et al, 2000 |

| Site               | Investigation Scale | Redox-Conditions | Biodegradation rate d <sup>-1</sup> | Reference             |
|--------------------|---------------------|------------------|-------------------------------------|-----------------------|
| FL-2               | Field scale         | Methanogenic     | 0,00082                             | Kolhatkar et al, 2000 |
| Vandenberg AFB, CA | Field scale         | Methanogenic     | 0,04600**                           | Mackay et al, 2007    |
|                    |                     |                  | Mean 0,01371                        |                       |

\* rate in Wilson 2003

\*\* Addition of ethanol

At the laboratory-scale a TBA biodegradation rate of 0.0259 d<sup>-1</sup> was determined by White et al (1986) for general (unspecified) anaerobic conditions, whereas Kolhatkar et al (2002) estimated a rate of 0.030 d<sup>-1</sup> for methanogenic conditions. The TBA biodegradation rate range at the field-scale level much wider (see **Table 4.9**). Investigations at a chemical plant by Day and Guliver (2003) showed that under iron- to sulphate-reducing conditions the TBA biodegradation rate varied along the flow path. The mean biodegradation rate was 0.0021 d<sup>-1</sup>. For methanogenic conditions the mean TBA biodegradation rate was 0.01978 d<sup>-1</sup>.

**Table 4.9** Results of field-scale investigations of TBA biodegradation in aquifer sediments under anaerobic conditions

| Site   | Investigation Scale | Redox-Conditions | Biodegradation rate d <sup>-1</sup> | Reference              |
|--|---------------------|------------------|-------------------------------------|------------------------|
| Passadena, Texas, flowpath from location 150, Chemical plant | Field scale         | Fe - SO4         | 0,00301                             | Day and Guliver, 2003  |
| Passadena, Texas, flowpath from location 57, Chemical plant  | Field scale         | Fe - SO4         | 0,00266                             | Day and Guliver , 2003 |
| Passadena, Texas, flowpath from location 165, Chemical plant | Field scale         | Fe - SO4         | 0,00071                             | Day and Guliver, 2003  |
|  |                     |                  | Mean 0,0021                         |                        |
| New York   | Field scale         | Methanogenic     | 0,02411                             | Kolhatkar et al, 2000  |
| Florida  | Field scale         | Methanogenic     | 0,02000                             | Kolhatkar et al, 2000  |
| Pennsylvania   | Field scale         | Methanogenic     | 0,01973                             | Kolhatkar et al, 2000  |
| Maryland   | Field scale         | Methanogenic     | 0,00167*                            | Kolhatkar et al, 2000  |
| Long Island, NY  | Field scale         | Methanogenic     | 0,03808*                            | Kolhatkar et al, 2000  |
| District of Columbia   | Field scale         | Methanogenic     | 0,01507*                            | Kolhatkar et al, 2000  |
|  |                     |                  | Mean 0,01978                        |                        |

\* rate in Wilson (2003)

The summary of the literature review concerning biodegradation rates for MTBE and TBA under various redox-conditions is shown in **Table 4.10**. Although the database on some issues is not very broad, the conclusions are:

- The comparison between laboratory-scale and field-scale investigation of biodegradation rate shows that the laboratory values are slightly

higher than biodegradation rates measured in the field. This can be explained by different conditions between the laboratory and the field.

- The biodegradation rate for methanogenic conditions seems to be higher than biodegradation rates for other anaerobic redox conditions.
- Field-scale biodegradation rates of MTBE under methanogenic conditions are in the range of reported biodegradation rates for natural aerobic conditions.
- Under similar redox conditions TBA biodegradation rates are higher than biodegradation rates for MTBE.

**Table 4.10** Mean first-order biodegradation rates ( $d^{-1}$ ) of MTBE and TBA from field-scale and laboratory investigations

| Scale      | MTBE                    | TBA     |
|------------|-------------------------|---------|
|            | <b>Oxygen / Nitrate</b> |         |
| Field      | 0.016 – 0.0012          | -       |
| Laboratory | 0.0041                  | 0.1175  |
|            | <b>Fe – SO4</b>         |         |
| Field      | -                       | 0.0021  |
| Laboratory | 0.0002*                 | -       |
|            | <b>Methanogenic</b>     |         |
| Field      | 0.01371                 | 0.01978 |
| Laboratory | 0.0199                  | 0.03**  |

\* single value Landmeyer et al (1998)

\*\* single value Kolhatkar et al (2002)

The Environment Agency UK (2002) tabulated MTBE biodegradation rates by reviewing and validating published papers, conference proceedings and unpublished information. Anaerobic biodegradation rates range from 0.0035 to 0.00035  $d^{-1}$ , which is typical for most petroleum hydrocarbon-contaminated sites.

As a general conclusion MTBE biodegradation is highly site-specific and numerous reports fail to demonstrate significant biodegradation. ETBE biodegradation under anaerobic conditions in microcosms was shown by Yeh & Novak (1994).

TAME biodegradation under methanogenic conditions in surface water sediments was shown by Somsamak et al (2005). There have been few studies on the biodegradation of TAME, ETBE and DIPE. Information about biodegradation in anaerobic aquifers for the other GEO (except MTBE) is not available at present.

#### 4.5.5.3. Influence of other organic compounds on GEO biodegradation in aquifers

GEO are often found together with BTEX and other petroleum hydrocarbons at sites with gasoline releases. Furthermore, other organic compounds like sugar, alcohol or organic acids may also be present in contaminated groundwater, often as partial breakdown products of the biodegradation of non-GEO components in the mixture. The biodegradation of GEO can be influenced by these compounds in different ways. This can be demonstrated with the findings for MTBE:

- 
- Results of Salanitro et al (1995) indicated that labile organic carbon (sugars, alcohols and organic acids) can inhibit MTBE and BTEX biodegradation in aerobic and anaerobic environments.
  - Deeb et al (2001) found for the pure culture PM1 that aerobic biodegradation of MTBE was especially inhibited by benzene and toluene. In addition, xylene and ethylbenzene slowed MTBE biodegradation.
  - Finneran & Loveley (2001) reported that humic compounds stimulated MTBE biodegradation under iron-reducing conditions.
  - Debor & Bastiaens (2007) reported that alkanes, alcohols and co-contaminants like benzene can be used as a growth substrate for co-metabolic aerobic MTBE biodegradation.
  - Field experiments by Mackay et al (2007) showed that in an existing MTBE plume MTBE was transformed to TBA under methanogenic conditions when benzene, toluene, o-xylene and ethanol were added. Another experiment at the same site with the addition of benzene, toluene and o-xylene without ethanol showed no MTBE biodegradation.
  - USEPA (2000) found in methanogenic microcosms that MTBE removal did not begin before the removal of the BTEX compounds was completed.
  - A laboratory study by Shah et al (2009) showed that MTBE and TAME can be biodegraded simultaneously under aerobic conditions and that the presence of TAME as a co-contaminant did not affect the rate of MTBE biodegradation.

Information about the influence of other organic compounds on the biodegradation of other GEO is not available at present.

#### **4.5.5.4. Observed lag time during GEO biodegradation in aquifers**

In field and laboratory studies MTBE and TBA biodegradation under aerobic and anaerobic conditions does not often begin immediately, but after a lag time. This lag time can be 40 – 80 days as demonstrated in many aerobic and anaerobic microcosms. Up to now no correlation between the length of a lag time and any hydrochemical, microbiological or hydrogeological parameter has been found. However, Shah et al (2009) observed different lag periods for aerobic biodegradation of MTBE, according to the presence of pre-existing contamination. In microcosms containing inocula from an MTBE plume, there was immediate biodegradation of MTBE, or this occurred after a short lag phase of 15-33 days. In contrast, using uncontaminated inocula from the same site, the lag period was 120 days. This illustrates the importance of pre-exposure to GEO, as a mechanism that may influence MTBE biodegradation.

The most common explanation for a lag time is that in aerobic MTBE biodegradation studies isolated microbial organisms have been observed to grow at relatively slow rates (Deeb et al, 2000). This slow rate of growth means that an initial population of organisms capable of biodegrading MTBE may require a relatively long lag time (or acclimation time) before the exposed biomass has grown to a population sufficient to produce a measurable rate of MTBE biodegradation (API, 2007; Shah et al, 2009).

#### 4.5.5.5. Compound-specific stable isotope analysis (CSIA)

In recent years compound-specific stable isotope analysis (CSIA) has become a versatile tool for characterising and assessing *in situ* biodegradation of organic pollutants in contaminated aquifers (Meckenstock et al, 2004; Schmidt et al, 2004). CSIA makes use of kinetic isotope fractionation processes that occur during biodegradation and uses the enrichment of heavy isotopes ( $^{13}\text{C}$  and  $^2\text{H}$ ) in the residual fraction as an indicator for *in situ* biodegradation (Rosell et al, 2007b). Detailed information on the use of CSIA for assessing biodegradation is given in USEPA (2008).

Smallwood et al (2001) was one of the first to use stable isotopes to identify different MTBE sources in a field study. Surprisingly the MTBE  $\delta^{13}\text{C}$  values for several gasolines were in a relatively narrow range (-28.3 to 31.7‰). O'Sullivan et al (2003) measured  $\delta^{13}\text{C}$  values of commercial MTBE from the USA (10 samples) and worldwide (25 samples from Europe, Asia, South America and Africa). They found carbon isotope ratios for MTBE between -31.7‰ to -28.3‰ for the USA samples and -32‰ to -27.4‰ for the worldwide set. For any MTBE release with an unknown original  $\delta^{13}\text{C}$  composition, a value more positive than -27‰ ( $\pm$ instrumental precision) would suggest *in situ* biodegradation (Kuder et al, 2005).

#### MTBE biodegradation under aerobic conditions

Carbon isotope enrichments due to aerobic biodegradation of MTBE have been observed in microcosms (Hunkler et al, 2001; Gray et al, 2002; Thornton et al, 2008). Carbon isotope effects in aerobic microcosms for MTBE show enrichment factors ( $\epsilon$ ) around -2‰ or lower. Very low isotope effects have been observed for aerobic biodegradation of MTBE by two aerobic strains (L108, IFP2001) by Rosell et al (2007b), indicating that CSIA may not be sensitive enough for certain types of aerobic bacteria. The carbon isotope enrichment factor for MTBE has also been shown to vary according to the availability of dissolved oxygen for aerobic biodegradation (Thornton et al, 2008). In a recent study, Thornton et al (2011) used carbon isotope analysis of field and laboratory microcosm samples to show that the aerobic fringe of GEO plumes is an important but under-researched zone of enhanced microbiological activity for MTBE biodegradation in aquifers.

Hydrogen isotope enrichment factors obtained by Gray et al (2002) were -29‰ to -66‰ (PM1 and sediment microcosms). Values from Rosell et al (2007b) were lower relative to the lower carbon enrichment factors. To date no CSIA case studies documenting carbon isotope fractionation for aerobic biodegradation in the field have been published (Kuder and Philp, 2008). The recent study by Thornton et al (2011) measured the carbon isotope composition of MTBE along the flow path of an unleaded fuel plume in a fractured chalk aquifer. However, biodegradation of the MTBE could not be resolved from the isotope composition of MTBE in the plume source area, using the carbon isotope data in isolation, even though this was proven in laboratory microcosms using material from the same aquifer and plume. The study concluded that the use of dual isotopes (e.g. C and H) may provide improvements in this respect.

**Table 4.11** Results of aerobic laboratory CSIA investigations for MTBE, TBA and ETBE

| GEO  | Culture                                      | <sup>13</sup> C enrichment factor [‰] | <sup>2</sup> H enrichment factor [‰] | Reference           |
|------|--|---------------------------------------|--------------------------------------|---------------------|
| MTBE | Enrichment culture (Borden aquifer)          | -1.52 to -1.97<br>± 0.06              | n.a.                                 | Hunkler et al, 2001 |
| MTBE | VAFB mixed consortium                        | -1.5 to -1,8<br>± 0.1                 | -29 to -66<br>± 3 – 4                | Gray et .al, 2002   |
| MTBE | PM1  | -2.0 to -2,4<br>± 0.1 – 0.3           | -33 to -37<br>± 4 - 5                | Gray et .al, 2002   |
| MTBE | L108   | -0.48<br>± 0.05                       | n.d. (-0.2)<br>± 8                   | Rosell et al, 2007b |
| MTBE | IFP2001 (resting cells)                      | -0.28<br>± 0.06                       | n.d. (+5)<br>± 17                    | Rosell et al, 2007b |
| MTBE | R8   | -2.4<br>± 0.1                         | -42<br>± 4                           | Rosell et al, 2007b |
| MTBE | Chalk aquifer microcosm with no oxygen limit | -1.53<br>± 0.38                       | n.a.                                 | Thonton et al, 2008 |
| TBA  | Enrichment culture (Borden aquifer)          | -4.21<br>± 0.07                       | n.a.                                 | Hunkler et al, 2001 |
| ETBE | L108   | -0.68<br>± 0.06                       | -14<br>± 2                           | Rosell et al, 2007b |
| ETBE | L108 (resting cells)                         | -0.8<br>± 0.01                        | -11<br>± 3                           | Rosell et al, 2007b |
| ETBE | IFP2001                                      | -0.8<br>± 0.1                         | -11<br>± 4                           | Rosell et al, 2007b |

n.a. not analysed  
n.d. not detected

**MTBE biodegradation under anaerobic conditions**

Carbon isotope enrichment factors are significantly higher for anaerobic MTBE biodegradation, ranging from  $-8.2 \pm 3.1\text{‰}$  to  $-15.6 \pm 4.1\text{‰}$  in several publications on methanogenic and sulphate reducing conditions in microcosms. Hydrogen isotope enrichment factors were determined for one methanogenic sample set, with  $-16 \pm 5\text{‰}$  (Kuder et al, 2005).

Field-scale applications of CSIA for the assessment of MTBE biodegradation *in situ* have been published by Kolhatkar et al (2002) and Wilson et al (2005b). Kuder et al (2005) and Zwank et al (2005) used carbon and hydrogen CSIA for further investigation of biodegradation pathways for MTBE.

**Conclusion MTBE**

Combined carbon and hydrogen CSIA (2-D CSIA) for MTBE can readily discriminate between the aerobic and anaerobic processes. In summary, aerobic biodegradation of MTBE results in low carbon isotope fractionation and proportionally higher hydrogen isotope fractionation. Anaerobic biodegradation of MTBE results in strong carbon isotope fractionation and proportionally lower hydrogen isotope fractionation. Kuder and Philp (2008) state that carbon CSIA is sufficient to detect strong isotope



ratio enrichments to interpret anaerobic MTBE biodegradation, whereas combined 2-D CSIA is recommended to interpret aerobic biodegradation.

**Table 4.12** Results of anaerobic laboratory CSIA investigations for MTBE and TAME

| GEO  | Redox condition                    | Culture                          | C enrichment factor [‰] | H enrichment factor [‰] | Reference             |
|------|------------------------------------|----------------------------------|-------------------------|-------------------------|-----------------------|
| MTBE | Methanogenic                       | Sediment enrichment culture (NJ) | -9<br>± 5               | n.a.                    | Kolhatkar et al, 2002 |
| MTBE | Methanogenic                       | Field                            | -8.1<br>± 0.85          | n.a.                    | Kolhatkar et al, 2002 |
| MTBE | Methanogenic                       | Enrichment culture (Arthur Kill) | -15 to -16<br>± 4 – 5   | n.a.                    | Somsamak et al, 2005  |
| MTBE | Methanogenic                       | Enrichment culture (NJ)          | -13<br>± 1              | -16<br>± 5              | Kruder et al, 2005    |
| MTBE | Sulphate reducing and methanogenic | Serveral enrichments cultures    | -14.4<br>± 0.7          | n.a.                    | Somsamak et al, 2006  |
| TAME | Methanogenic                       | Enrichment culture (Arthur Kill) | -11 - -14<br>± 3 - 5    | n.a.                    | Somsamak et al, 2005  |

n.a. not analysed

### TBA

In aerobic microcosms cometabolism of TBA produced carbon isotope enrichment factors of  $4.21 \pm 0.07\text{‰}$  (Hunkeler et al, 2001). The published CSIA data for TBA at several anaerobic gasoline station sites show very little variability in  $\delta^{13}\text{C}$  values (Kolhatkar et al, 2002; Kuder et al, 2005; Zwank et al, 2005). This indicates either negligible extent of *in situ* TBA biodegradation or a biodegradation process that results in only small isotope fractionation. Bush-Harris et al (2008) reported high  $^{13}\text{C}$  enrichment of microbial fatty acids from Bio-Sep probes using  $^{13}\text{C}$ -labelled TBA at a gasoline site, with potentially anaerobic biodegradation of TBA.

### ETBE

Aerobic ETBE biodegradation by the bacteria strains L108 and IFP2001 was found to be associated with a low carbon isotope fractionation (-0.68 to -0.8‰) and a slightly higher hydrogen isotope fractionation (-11 to -14 ‰) (Rosell et al, 2007b).

### TAME

Biodegradation of TAME in anaerobic microcosms under methanogenic conditions produced carbon isotope enrichment factors of -11 to -14‰ (Somsamak et al, 2005).

No information was found on the use of CSIA for the other GEO.

#### **4.6. LITERATURE REVIEW – GROUNDWATER / SURFACE WATER INTERFACE**

The following issues are important for risk assessment:

- Very little is known about the groundwater / surface water interface (hyporheic zone) in respect to solute fluxes and biodegradation processes.
- The hyporheic zone should be taken into account in risk assessment.
- The hyporheic zone has a high potential for microbial activity and therefore for biodegradation.
- Sorption in the hyporheic zone is more important than in the aquifer, due to a higher content of particulate organic carbon in the former.
- Identification of single processes by investigations is not easily achieved.

##### **4.6.1. Introduction**

In the literature there are only few references dealing with the impact of GEO-contaminated groundwater on surface water. Two papers refer to GEO-contaminated groundwater reaching surface water. However many publications describe research on river bed sediments that were used since 1999 to investigate the biodegradation of GEO because of their known high potential for microbial activity. A comprehensive report about the current state of knowledge, nature of and processes at the groundwater / surface water interface is available (Smith, 2005).

##### **4.6.2. GEO transfer into the gas phase**

Volatilisation of GEO in the hyporheic zone is not different to volatilisation in groundwater (chapter 4.5.2) and can generally be assumed to be negligible.

##### **4.6.3. GEO transfer to groundwater**

Dissolution of GEO from LNAPL in the hyporheic zone is not different to the processes in groundwater (chapter 4.5.3).

##### **4.6.4. GEO transfer to the soil phase**

Greenwood et al (2007) determined the sediment-water distribution coefficient ( $K_d$ ) for MTBE and TBA at a site in Ronan, Montana, in seven sediment samples from the hyporheic zone. A linear isotherm was found to most accurately represent the sorption of MTBE. The Freundlich model provided a more accurate representation of TBA sorption, indicating a non-linear relationship between sorption and aqueous concentration of TBA.

The fraction of organic carbon ( $f_{oc}$ ) at the reported site ranged from 0.012 to 0.068 and is higher than that for typical aquifer sediments (see Chapter 4.5.4).

The soil characteristics were (Greenwood et al, 2007):

Sand content 28 - 55%, Silt content 38 – 50% and Clay content 7 – 22%.

A comparison of  $K_{oc}$  values estimated by Greenwood et al (2007) with values in literature is given in **Table 4.13**.

**Table 4.13** Comparison between log  $K_{oc}$  values from the literature and estimated for hyporheic zone sediments by Greenwood et al (2007)

| Compound | log $K_{oc}$ (literature) | log $K_{oc}$ (Greenwood et al 2007) |
|----------|---------------------------|-------------------------------------|
| MTBE     | 1.1                       | 2.13                                |
| TBA      | 0.41                      | 0.7                                 |

The  $K_{oc}$  value for MTBE from Greenwood et al (2007) is ten times higher than the literature value, indicating that sorption is ten times higher than predicted by the literature. The sorption of TBA is about two times higher than predicted by the literature value. Other researchers (e.g. Smith & Lerner, 2008) have shown that the organic carbon content of the hyporheic zone is greater than that of underlying aquifers and leads to a significantly increased potential for retardation of organic contaminants.

The extent of sorption of other GEO can be estimated, according to their individual  $K_{oc}$  values (see Chapter 1.3). Sorption in the hyporheic zone is more important than in the aquifer, due to the relatively higher content of particulate organic carbon in this environmental compartment.

#### 4.6.5. GEO Biodegradation

Surface water sediments from the hyporheic zone have been used in many microcosm studies to investigate the biodegradation of GEO under various redox-conditions. Biodegradation was observed for MTBE, TBA, TAME under aerobic conditions (**Table 4.14**).

**Table 4.14** Results from microcosm investigations of GEO biodegradation in surface water sediments under aerobic conditions

| Site                              | Investigation scale | Compound        | Biodegradation confirmed? | Redox condition | Reference             |
|-----------------------------------|---------------------|-----------------|---------------------------|-----------------|-----------------------|
| Cecil Field, Jachson Ville, FL    | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Cranbery Lake, NJ                 | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Laurens, SC                       | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Lidy Hot Spring, ID               | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| NWIRP Dallas, TX                  | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Oasis 1, Charleston, SC           | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Oasis 2, Charleston, SC           | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Pensacola Bay, FL                 | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Penascola Swamp, FL               | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Picatinny Arsenal, NJ             | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Ronan, MO                         | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001c  |
| Laurens, SC                       | Microcosms          | TBA             | Yes                       | aerobic         | Bradley et al, 1999   |
| Charleston, SC                    | Microcosms          | TBA             | Yes                       | aerobic         | Bradley et al, 1999   |
| Laurens, SC                       | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 1999   |
| Charleston, SC                    | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 1999   |
| Charleston, SC                    | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001a  |
| Cecil Field, Jachson Ville, FL    | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001a  |
| Picatinny Arsenal, NJ             | Microcosms          | MTBE            | Yes                       | aerobic         | Bradley et al, 2001a  |
| Carmans River, fresh water, NY    | Microcosms          | MTBE, TBA, TAME | Yes                       | aerobic         | Landmeyer et al, 2010 |
| Ford Pond, fresh water, NY        | Microcosms          | MTBE, TBA, TAME | Yes                       | aerobic         | Landmeyer et al, 2010 |
| Tiana Bay, brackisch water, NY    | Microcosms          | MTBE, TBA, TAME | Yes                       | aerobic         | Landmeyer et al, 2010 |
| Great South bay, saline water, NY | Microcosms          | MTBE, TBA, TAME | Yes                       | aerobic         | Landmeyer et al, 2010 |

Biodegradation has been observed for MTBE and TBA under nitrate-reducing conditions in surface water sediments (Table 4.15).

**Table 4.15** Results from microcosm investigations of GEO biodegradation in surface water sediments under nitrate-reducing conditions

| Site                           | Investigation scale | Compound     | Biodegradation confirmed? | Redox condition | Reference            |
|--------------------------------|---------------------|--------------|---------------------------|-----------------|----------------------|
| Charleston, SC                 | Microcosms          | MTBE and TBA | Yes                       | Nitrate         | Bradley et al, 2001b |
| Pensacola, FL                  | Microcosms          | MTBE and TBA | Yes                       | Nitrate         | Bradley et al, 2001b |
| Athur Kill estuarien inlet, NY | Microcosms          | MTBE         | No                        | Nitrate         | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | ETBE         | No                        | Nitrate         | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | TAME         | No                        | Nitrate         | Somsamak et al, 2001 |
| Charleston, SC                 | Microcosms          | MTBE         | Yes                       | Nitrate         | Bradley et al, 2001a |
| Cecil Field, Jachson Ville, FL | Microcosms          | MTBE         | Yes                       | Nitrate         | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE         | Yes                       | Nitrate         | Bradley et al, 2001a |

Biodegradation has been observed for MTBE under iron- and manganese-reducing conditions in surface water sediments (**Table 4.16**).

**Table 4.16** Results from microcosm investigations of GEO biodegradation in surface water sediments under iron- and manganese-reducing conditions

| Site                           | Investigation scale | Compound | Biodegradation confirmed? | Redox condition | Reference            |
|--------------------------------|---------------------|----------|---------------------------|-----------------|----------------------|
| Athur Kill estuarien inlet, NY | Microcosms          | MTBE     | No                        | Fe(III)         | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | ETBE     | No                        | Fe(III)         | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | TAME     | No                        | Fe(III)         | Somsamak et al, 2001 |
| Charleston, SC                 | Microcosms          | MTBE     | Yes                       | Fe(III)         | Bradley et al, 2001a |
| Cecil Field, Jachson Ville, FL | Microcosms          | MTBE     | Yes                       | Fe(III)         | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE     | Yes                       | Fe(III)         | Bradley et al, 2001a |
| Charleston, SC                 | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Cecil Field, Jachson Ville, FL | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Charleston, SC                 | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Cecil Field, Jachson Ville, FL | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Charleston, SC                 | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Cecil Field, Jachson Ville, FL | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE     | Yes                       | Mn(IV)          | Bradley et al, 2001a |

Biodegradation has been observed for MTBE and TAME under sulphate-reducing conditions in surface water sediments (**Table 4.17**).

**Table 4.17** Results from microcosm investigations of GEO biodegradation in surface water sediments under sulphate-reducing conditions

| Site                           | Investigation scale | Compound | Biodegradation confirmed? | Redox condition | Reference            |
|--------------------------------|---------------------|----------|---------------------------|-----------------|----------------------|
| Athur Kill estuarien inlet, NY | Microcosms          | MTBE     | Yes                       | Sulphate        | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | TAME     | Yes                       | Sulphate        | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | ETBE     | No                        | Sulphate        | Somsamak et al, 2001 |
| Charleston, SC                 | Microcosms          | MTBE     | Yes                       | Sulphate        | Bradley et al, 2001a |
| Cecil Field, Jackson Ville, FL | Microcosms          | MTBE     | Yes                       | Sulphate        | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE     | Yes                       | Sulphate        | Bradley et al, 2001a |

Biodegradation has been observed for MTBE under methanogenic conditions in surface water sediments (**Table 4.18**).

**Table 4.18** Results from microcosm investigations of GEO biodegradation in surface water sediments under methanogenic conditions

| Site                           | Investigation scale | Compound | Biodegradation confirmed? | Redox condition | Reference            |
|--------------------------------|---------------------|----------|---------------------------|-----------------|----------------------|
| Laurens, SC                    | Microcosms          | TBA      | No                        | Methanogenic    | Bradley et al, 1999  |
| Charleston, SC                 | Microcosms          | TBA      | No                        | Methanogenic    | Bradley et al, 1999  |
| Laurens, SC                    | Microcosms          | MTBE     | No                        | Methanogenic    | Bradley et al, 1999  |
| Charleston, SC                 | Microcosms          | MTBE     | No                        | Methanogenic    | Bradley et al 1999   |
| Athur Kill estuarien inlet, NY | Microcosms          | MTBE     | No                        | Methanogenic    | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | ETBE     | No                        | Methanogenic    | Somsamak et al, 2001 |
| Athur Kill estuarien inlet, NY | Microcosms          | TAME     | No                        | Methanogenic    | Somsamak et al, 2001 |
| Charleston, SC                 | Microcosms          | MTBE     | Yes                       | Methanogenic    | Bradley et al, 2001a |
| Cecil Field, Jackson Ville, FL | Microcosms          | MTBE     | Yes                       | Methanogenic    | Bradley et al, 2001a |
| Picatinny Arsenal, NJ          | Microcosms          | MTBE     | Yes                       | Methanogenic    | Bradley et al, 2001a |

The results of the microcosm studies with surface water sediments indicate that biodegradation of MTBE has been observed under all redox-conditions (**Table 4.19**). Biodegradation of TBA has been observed for aerobic and nitrate-reducing conditions, whereas biodegradation of TAME has been shown for aerobic and sulphate-reducing conditions.

**Table 4.19** Biodegradation of GEO in surface water sediments under various redox-conditions

| Redox condition    | Biodegradation observed |
|--------------------|-------------------------|
| Aerobic            | MTBE, TBA, TAME         |
| Nitrate reducing   | MTBE, TBA               |
| Iron reducing      | MTBE                    |
| Manganese reducing | MTBE                    |
| Sulphate reducing  | MTBE, TAME              |
| Methanogenic       | MTBE                    |

Specific investigations into the biodegradation of MTBE, TBA and TAME in the hyporheic zone were conducted by Landmeyer et al (2001, 2010). Landmeyer et al (2001) demonstrated for a concrete-lined ditch that MTBE concentrations were reduced by 96% over a 6.5 m distance in the subsurface. The MTBE mass loss was attributed to biodegradation and not dilution or dispersion. In microcosm studies complete biodegradation of MTBE to CO<sub>2</sub> was observed within 80 days.

Landmeyer et al (2010) investigated natural attenuation processes such as dispersion and biodegradation relative to the rate of groundwater discharge in the hyporheic zone at four sites impacted by MTBE, TBA and TAME. The results showed that about 3% of the observed attenuation for MTBE, TBA and TAME was caused by biodegradation. The primary natural attenuation mechanisms observed in the hyporheic zone in these studies were dilution (by mixing of groundwater and surface water) and dispersion. The contrary results of Landmeyer et al (2001, 2010) demonstrated that biodegradation is site-specific and that knowledge of the interaction of groundwater and surface water is needed on a site-specific basis.

## **5. IDENTIFICATION OF KNOWLEDGE GAPS AND RESEARCH NEEDS**

### **5.1. USE OF GEO IN EUROPE**

The available data that have been used to evaluate the production and use of GEO in EU does not cover all countries, nor the entire period 2000-2010. Despite the incomplete data set and partially fragmented information, this report comprises a representative overview of the production and use of GEO in the EU (and selected European countries) between 2000-2010. In 50% of the countries the gasoline demand market has been analysed in detail. Those countries investigated in detail represent more than 90% of the total EU market.

The following important limitations should be considered when the results of the analysis are evaluated:

- Name plate GEO capacities of existing plants were used to estimate the production volumes. The actual production volumes are not known, but will generally be lower, most likely 80-90% of the name plate capacity. Individual production facilities may not be operational on GEO.
- Detailed GEO import and export data are not available and can have a considerable effect on the GEO volumes used in Europe.
- GEO levels in gasoline were investigated by SGS in narrow timeframes (PULP in 2000-2004 and PUL only in 2006-2009). A complete overview is not available.
- There is one unit producing TAEE and THxEE in Europe.

### **5.2. OCCURRENCE OF GEO IN THE EUROPEAN ENVIRONMENT**

#### **5.2.1. Gaps on GEO in groundwater**

The results of this study indicate that a relatively large amount of information is available, most of it referring to MTBE. The following points were identified as general gaps in this context:

- For 18 countries no publications or any data were available for a reliable evaluation.
- Most of the information refers to MTBE. With the exception of a small number of countries, data on the other GEO are not adequate to produce a representative picture of their presence in European aquifers.

#### **5.2.2. Gaps on surface water**

For a number of countries (e.g. Germany, the Netherlands, Switzerland and Austria) a good record of observations is available to understand the behaviour of MTBE in surface water systems. For other GEO minor information was available.

The results in this study indicate that GEO are present to a certain extent in surface water bodies. Regarding the importance of release events and background loads in large rivers, data is available only for the Rhine (including some tributaries).



For other surface water bodies with high risk of GEO releases better monitoring would help to ascertain the distribution and occurrence of release events. Thereby the other GEO should also be taken into account more intensively.

### **5.2.3. Gaps on drinking water**

The results for drinking water indicated that information on MTBE was mainly available for Germany, Netherlands, UK and Sweden.

Monitoring data in the other countries is necessary in regard to the strong link between drinking water and surface water as a raw water source, and the aspect that regular treatment methods do not lead to complete removal.

Due to the ongoing replacement of MTBE, monitoring of drinking water for the other GEO is currently a data gap.

### **5.2.4. Gaps on run-off water**

With regard to the findings for MTBE in road run-off water in Switzerland, for urban run-off water in Frankfurt/Main (Germany) and the comparison of results from the United States, it is clear that run-off water is an important diffuse source for GEO in surface water and groundwater.

The existing information / data on this is insufficient to provide a general conclusion on the presence, behaviour and discharge to other environmental compartments for GEO in Europe. Therefore, focussed research on this topic, also in regard to the other GEO beside MTBE, may be appropriate by the relevant authorities.

### **5.2.5. Gaps on precipitation**

Knowledge on GEO in precipitation is mainly limited to Germany and MTBE. In regard to the knowledge required on precipitation as a diffuse source of GEO for groundwater and surface water, a more extensive monitoring programme would be required. Therefore the other GEO as well as the climate background need to be taken into account.

### **5.2.6. Gaps on air**

The identified results on MTBE in air for Germany, Finland, Belgium and Switzerland indicated a certain presence, especial in urban areas. Regarding the missing information on other GEO and missing information for other countries a more intensive observation for GEO in air is necessary. Larger cities with heavy traffic should be especially considered, due to the potential large contribution of GEO as a diffuse source.

### **5.2.7. Gaps on soil**

Beside the common opinion of low retardation for MTBE and other GEO in soil, the results from the UK, Sweden and Finland show a certain presence of MTBE and partly TAME in soil, especially at contaminated sites. The results also indicated that MTBE was found in soil where no groundwater contamination was detected.

As a result of this study, a general background load of GEO in European soil is not expected (beside known contaminated sites). Thus, a general GEO monitoring (comparable to surface or groundwater) is not considered necessary.

### **5.3. FATE AND TRANSPORT OF GEO IN THE SUBSURFACE**

Knowledge gaps and the need for further research are listed according to the environmental media: unsaturated zone (Chapter 4.4), groundwater (Chapter 4.5) and groundwater / surface water interface (Chapter 4.6).

#### **Unsaturated zone**

Field-scale information about the fate and transport of GEO in the unsaturated zone is sparse. Most of the conclusions are derived from the general understanding of multi-phase transport in the unsaturated zone and the physical-chemical properties of the GEO. This gap probably originates from the field experience of MTBE releases that are usually investigated only in respect to the areal extent of a release in the unsaturated zone. After the extent of the release in the unsaturated zone is known and risks to human health and the environment established, appropriate risk-management actions are implemented. Therefore, no detailed investigations into the unsaturated zone are performed.

Up to now no guidelines about the GEO vapour-to-indoor-air pathway exist. Therefore research is recommended into the fate and transport behaviour of GEO in the unsaturated zone, especially on vapour phase transport and attenuation.

#### **Groundwater**

As soon as GEO are detected in a groundwater body detailed understanding about transport behaviour is needed to determine the extent of the impact. Later on during the risk assessment stage knowledge about the site-specific biodegradation of the individual GEO is required.

The transport behaviour of MTBE and TBA at the field-scale is well documented in shallow (primarily unconsolidated) intergranular porosity aquifers, predominantly from studies in the USA. Biodegradation of MTBE and TBA is known to be very site-specific and most information on this is available from the USA, whereas, information from European hydrogeological environments is much more limited.

Therefore research / data collation is recommended into the transport and biodegradation behaviour of the other GEO in groundwater, especially at the field-scale, for the different aquifer types present in Europe.

Furthermore, research into the development of technologies to measure biodegradation quantitatively at the field-scale (e.g. BACTRAPs<sup>®</sup>, Bio-Trap<sup>®</sup> Sampler) is advised.

#### **Groundwater / surface water interface**

The groundwater / surface water interface is an environmental compartment that has come to public attention in terms of risk assessment quite recently. Therefore, relatively little is known about the transport and fate processes affecting GEO transfer from groundwater into surface water, as these are site-specific. In this

respect, GEO does not differ from other substances. Two major questions have to be answered for the hyporheic zone. These are:

- How important is biodegradation and sorption of GEO in the hyporheic zone considering the different milieus, or is physical mixing the predominant process for attenuation of GEO here?
- To what extent do attenuation processes in the hyporheic zone reduce the transfer of GEO mass from groundwater into surface water?

The mass loss of GEO in the hyporheic zone may be taken into account for the assessment of GEO plumes close to, or entering, a surface water body. For each surface water body the tolerable maximum GEO concentration has to be determined to ensure risks to ecology and other legitimate water users are appropriately managed.

## 6. CONCLUSIONS

### 6.1. USE OF GEO IN EUROPE

Data was collated on the production and use of MTBE, ETBE, TAME, DIPE and TBA in 30 countries (27 EU countries and Croatia, Norway and Switzerland) to inform the debate on the environmental management issues relating to GEO. The report comprises data on gasoline production and use in Europe that were provided by CONCAWE and obtained from the European Commission.

Conclusions from the investigations into the production of GEO in Europe can be summarised as follows:

- In 2010, about 55 facilities with MTBE, ETBE and TAME production capacity are located in the EU.
- 50% of the total European production capacity is located in Germany, France and the Netherlands.
- There are no records of DIPE and TBA production for blending of gasoline in Europe.
- The total GEO production capacity (MTBE, ETBE and TAME) has grown from 4,108 Ktonnes in 2002 to 6,049 Ktonnes per year in 2010.
- The market share of ETBE has grown from 15% in 2002 to about 60% in 2010 while MTBE production capacity has decreased.
- Combined TAME and TAAE production capacity was constant from 2005 to 2012 with a 10% market share.
- Actual production volumes at present are not publicly available.

GEO are blended with gasoline in different concentrations depending on the gasoline type and country specific practices. Consequently GEO demand is influenced by gasoline demand and GEO concentrations in the different gasoline types. Conclusions from the demand and use of gasoline in Europe can be summarised as follows:

- The total annual gasoline demand in EU27 in the year 2007 was 104,340 Ktonnes.
- The volume of gasoline sales in EU 15 during 2004-2008 decreased while the diesel consumption increased.
- The market share of Premium Unleaded gasoline (RON95) (PUL) has gradually increased from 71% in 2004 to 89% in 2008 at the expense of Regular unleaded gasoline (RON91) (RUL) and Super Premium Unleaded gasoline (PULP).
- The market share of Lead Replacement gasoline (LRG) has decreased to nearly zero in 2008.

Data on GEO composition of 6 different gasoline types in EU 27, Norway, Switzerland and Turkey in the period 2000 to 2009 was obtained from SGS (Société Générale de Surveillance) and analysed (1,239 sampling events for MTBE, ETBE

and TBA and 650 sampling events for TAME and DIPE). The main conclusions can be summarized as follows:

- MTBE was detected in PUL gasoline in all EU 27 in the period 2004 – 2009.
- ETBE use in PUL was less widespread than MTBE in the period 2004 – 2009.
- The highest ETBE concentrations (5 – 12 m/m%) in PUL gasoline were found in France, Spain, Hungary and Austria.
- TAME was detected in PUL and PULP gasoline in concentrations from 1 to 3 m/m% in Cyprus, Finland, Greece, Italy, Malta, Romania and Slovenia.
- DIPE was detected only in 7 of 650 samples at a concentration consistently below 1 m/m%.
- TBA traces (0.01 – 0.03 m/m%) were detected in many EU countries.

Future gasoline consumption is expected to decrease in favour of the use of diesel and due to increasingly fuel efficient vehicles. The report shows how the use of GEO has changed from MTBE to ETBE in certain countries and onwards towards a lesser use of GEO (presumably in favour of direct ethanol blending) in certain markets.

## 6.2. OCCURRENCE OF GEO IN THE EUROPEAN ENVIRONMENT

A major aspect was the investigation of GEO distribution in groundwater, drinking water, surface water, run-off water, precipitation (rain/snow) and air. Apart from the general sources of literature for the study, local environmental authorities and institutes in 30 countries (27 EU countries and Croatia, Norway and Switzerland) have been contacted for additional information. As a result, information from 11 countries was obtained for different environmental compartments, while for 19 countries no specific data on GEO in the environment were available.

It can be concluded that:

- GEO in air mainly originate from industrial emissions, fugitive emissions at petrol stations and the exhaust emissions from gasoline powered vehicles, due to incomplete combustion or reduced efficiency of the catalytic converter during the cold run period of vehicles. Atmospheric MTBE degradation rates are reported to be rapid, with an MTBE half-life in the order of 3 - 7 days. Consequently significant build-up of GEO in the atmosphere is not anticipated. The information regarding the presence of GEO in air for Europe is not sufficient to draw a general picture on the distribution and is likely to be very variable as climatic conditions control air movement.
- GEO in soil originate predominantly from release events. MTBE may be found in urban areas near roads with heavy traffic at levels above the analytical detection limits without an existing local point source. The knowledge based on the distribution of GEO within soils in Europe is relatively low.
- Analysis of highway run-off water from Switzerland, Germany and the USA demonstrate the presence of MTBE, DIPE and TAME in some samples. ETBE was not detected. The results show that the GEO concentrations strongly depend on the rain intensity and land use in the areas where the road run-off occurs. The detectable MTBE concentration usually ranged from 0.05 to 1 µg/l.

- GEO in precipitation originates mainly from air emissions. The amount of GEO in precipitation is generally higher in urban and industrial areas than in rural areas. Elimination of GEO is predominantly done by volatilisation at the air-water interface. Biodegradation in precipitation is considered to be negligible. For rain, urban areas have a much higher MTBE detection rate (86%) than samples from rural areas (18%) (data from Germany). The reported MTBE concentration for rain was consistently below 0.1 µg/l.
- In drinking water, an MTBE concentration above the USEPA recommended guide for MTBE odour threshold of 20 µg/l was not reported. MTBE is observed in some European drinking water mainly in trace concentration (range 0.01 to 0.2 µg/l). The major source for MTBE in drinking water is raw water originating from surface water. However blending, water treatment and other water quality management measures ensure that European drinking water is not significantly impacted by GEO.
- In surface water, low background concentrations of GEO originate from precipitation, run-off from urban areas and effluent of waste water treatment plants. Higher GEO concentrations are sometimes found in rivers or lakes from industrial releases or illegal dumping of tank washings from tank ships. In recreational areas, the use of watercrafts leads to seasonal elevated GEO concentrations in lakes. GEO are eliminated from surface water mainly by volatilisation over the air-water interface. The reported half-life of MTBE in rivers is from 30 minutes to 52 days. In lakes, the half-life ranges from 10 to 193 days.
- In groundwater background concentrations of GEO are typically non-detectable. Regional GEO impacts typically originate from precipitation and run-off. The main reasons for GEO occurrence in groundwater are direct and very localised (point-source) releases from production plants, refuelling facilities (retail filling stations, fuel terminals and distribution sites) and other releases such as transport accidents. GEO elimination by natural attenuation processes in groundwater is often slower than in the other environmental media, but due to generally slow groundwater velocities this may still be sufficient to mitigate the risk to near-by receptors.

In general GEOs can be observed in the environment, however, not in concentrations that warrant this to be classified as a generic environmental issue that requires active management, except for situations that are the result of specific and local release events that are normally already identified and subject to remedial control measures.

### 6.3. FATE AND TRANSPORT OF GEO IN SOIL AND GROUNDWATER

When GEO are accidentally released into the subsurface, they have the potential to cause impact due to their high solubility and low taste and odour thresholds. Furthermore, GEO are weakly adsorbed to aquifer and soil minerals and are less easily biodegraded than other constituents in gasoline; GEO plumes in groundwater, for example, tend to be larger and more persistent than BTEX plumes. However, it is now generally accepted that, after an initial acclimation period, GEO will biodegrade in the subsurface environment, albeit often at a slower rate than the BTEX compounds.

A review of the international literature was undertaken with a focus on international reports and scientific publications to give an overview on the known fate, transport

and degradation mechanisms of GEO, to inform risk-management strategies that may rely on natural attenuation processes.

The conclusions of the international literature review can be summarised as follows:

- To evaluate the risk of a GEO release systematically, a conceptual model for the subsurface media: soil, unsaturated zone, saturated zone and transition zone between groundwater and surface water (hyporheic zone) should be established.
- In a first step of risk assessment, all processes that affect the fate and transport of GEO in the subsurface have to be identified. This should take into account that dispersion and diffusion will decrease dissolved GEO concentrations in groundwater, sorption and volatilisation will transfer GEO mass from dissolved GEO into the soil phase and gas phase, vapourisation and dissolution will transfer LNAPL into the gas phase and water phase. The only process that will reduce the mass of GEO in the subsurface is biodegradation.
- In the unsaturated zone vapourisation is an important process due to the relatively high vapour pressures of GEO. Consequently, the vapour exposure pathways may need to be considered in a risk assessment, where potential receptors are present.
- In groundwater MTBE may form plumes which are generally longer than BTEX plumes and shorter than plumes by light volatile chlorinated hydrocarbons (e.g. tetra- and trichloroethene and their decay products).
- Biodegradation in aquifers under aerobic conditions has frequently been demonstrated for MTBE, TBA, ETBE, TAME and DIPE. Aerobic first-order biodegradation rates for MTBE and TBA were collated for many sites. No degradation rate data were available on the aerobic biodegradation of other GEO.
- Biodegradation in aquifers under anaerobic conditions was often reported for MTBE and TBA, and less frequently for TAME and ETBE. For many sites, first-order biodegradation rates for MTBE and TBA in aquifers under anaerobic conditions were compiled. No data on the anaerobic biodegradation of other GEO were available.
- In recent years, an investigation method called compound-specific stable isotope analysis (CSIA) has become a useful tool for characterising and assessing *in situ* biodegradation of organic pollutants in contaminated aquifers. CSIA was first used in 2001 to identify different MTBE sources in a field study. Up to now CSIA has been successfully applied to assess biodegradation of MTBE, TBA, ETBE and TAME in aquifers.
- The hyporheic zone has a relatively high potential for microbial activity. Furthermore sorption in the hyporheic zone is more important than in the aquifer, due to a higher content of particulate organic carbon in the former. Therefore the hyporheic zone should be taken into account in risk assessment. However in detail very little is known about the groundwater / surface water interface (hyporheic zone) in respect to solute fluxes and biodegradation.

Finally, it can be concluded that at sites without nearby receptors, monitored natural attenuation (MNA) of GEO contamination may be a viable risk management strategy, either in isolation, or as part of a remediation treatment train approach.

## 7. GLOSSARY OF TERMS

|       |   |
|-------|---|
| AUE   | Agency for Environment and Energy of the city of Basel                                    |
| BAFU  | Federal Environmental Agency of Switzerland   |
| BTEX  | Benzene, toluene, ethyl-benzene and xylene compounds                                      |
| CSIA  | Compound specific isotope analysis  |
| DIPE  | Di-isopropyl ether  |
| EFOA  | European Fuel Oxygenate Association   |
| EPTC  | European Petroleum Technical Cooperation  |
| ETBE  | Ethyl tertiary butyl ether  |
| EU    | European Union  |
| EU15  | The 15 Member States of the European Union (at 2005)                                      |
| EU27  | The 27 Member States of the European Union (at 2011)                                      |
| GEO   | Gasoline ether oxygenates   |
| IKSR  | International Commission for the Protection of the River Rhine (German abbreviation IKSR) |
| IMBL  | International Measuring Station Bimmen Lobith   |
| LAWA  | In Germany, the water consortium of the federal states (Länderarbeitsgemeinschaft Wasser) |
| LNAPL | Light non-aqueous phase liquid  |
| LRG   | Lead replacement gasoline   |
| LUBW  | Environmental Agency of Baden-Württemberg/Germany (LUBW),                                 |
| LUST  | Leaking underground storage tanks   |
| MNA   | Monitoring natural attenuation  |
| MTBE  | Methyl tertiary butyl ether   |
| PUL   | Premium unleaded gasoline (see RON95)   |
| PULP  | Super premium unleaded gasoline (see RON98)   |
| RON91 | Research Octane Number 91   |
| RON95 | Research Octane Number 95   |



|       |   |
|-------|---|
| RON98 | Research Octane Number 98                     |
| RUL   | Regular unleaded gasoline                     |
| SGS   | Société Générale de Surveillance              |
| TAAE  | Tertiary amyl ethyl ether                     |
| TAME  | Tertiary amyl methyl ether                    |
| TBA   | Tert-butyl alcohol                            |
| TEL   | Anti-knock additives tetra-ethyl lead         |
| THxEE | Tertiary hexyl ethyl ether                    |
| THxME | Tertiary hexyl methyl there                   |
| USEPA | United States Environmental Protection Agency |
| VOC   | Volatile Organic Compounds                    |
| WFD   | EU Water Framework Directive                  |
| WHO   | World Health Organization                     |

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## APPENDIX 1 GASOLINE ETHER OXYGENATE PRODUCTION CAPACITY IN EUROPE

| Fuel Ether production capacity (kton/yr) |      |              |              |
|--|------|--------------|--------------|
|  | 2002 | 2005         | 2010         |
| Austria                                  |      | 65           | 65           |
| Belgium                                  |      | 453          | 453          |
| Bulgaria                                 |      | 82           | 82           |
| Croatia                                  |      | 0            | 0            |
| Czech Republic                           |      | 92           | 92           |
| Germany                                  |      | 718          | 1,113        |
| France                                   |      | 836          | 836          |
| Finland                                  |      | 204          | 204          |
| Greece                                   |      | 110          | 195          |
| Hungary                                  |      | 139          | 139          |
| Italy                                    |      | 562          | 592          |
| Lithuania                                |      | 80           | 80           |
| Netherlands                              |      | 980          | 982          |
| Poland                                   |      | 120          | 120          |
| Romania                                  |      | 220          | 220          |
| Slovakia                                 |      | 52           | 52           |
| Spain                                    |      | 422          | 422          |
| Sweden                                   |      | 50           | 48           |
| Portugal                                 |      | 50           | 50           |
| United Kingdom                           |      | 304          | 304          |
| <b>Total</b>                             |      | <b>5,539</b> | <b>6,049</b> |

|              | 2002         | 2005         | 2010         |
|--------------|--------------|--------------|--------------|
| MTBE         | 3,290        | 2,449        | 1,808        |
| ETBE         | 568          | 2,583        | 3,611        |
| TAME         | 250          | 507          | 630          |
| <b>Total</b> | <b>4,108</b> | <b>5,539</b> | <b>6,049</b> |

Source (EFOA 2010a), (EFOA 2010b), (EU 2002), (EU 2006), (Balat 2007).

**APPENDIX 2 GASOLINE DEMAND IN 2007 (EU 2007)**

|                        | 2007 (million liters) |                |              |                |            |
|------------------------|-----------------------|----------------|--------------|----------------|------------|
|                        | RON                   |                |              | Total          | %          |
|                        | 91                    | 95             | 98           |                |            |
| <b>Austria</b>         | 642                   | 1,944          | 76           | 2,662          | 1.8        |
| <b>Belgium</b>         | 0                     | 1,429          | 458          | 1,887          | 1.3        |
| <b>Bulgaria</b>        | 0                     | 822            | 0            | 822            | 0.6        |
| <b>Cyprus</b>          | 0                     | 427            | 48           | 475            | 0.3        |
| <b>Czech Rep.</b>      | 161                   | 2,732          | 40           | 2,933          | 2.0        |
| <b>Germany</b>         | 7,548                 | 20,458         | 824          | 28,830         | 19.4       |
| <b>Danmark</b>         | 507                   | 1,917          | 11           | 2,435          | 1.6        |
| <b>Estonia</b>         | 0                     | 404            | 56           | 460            | 0.3        |
| <b>Greece</b>          | 0                     | 5,197          | 401          | 5,598          | 3.8        |
| <b>Spain</b>           | 0                     | 7,917          | 1,137        | 9,054          | 6.1        |
| <b>Finland</b>         | 0                     | 2,272          | 209          | 2,481          | 1.7        |
| <b>France</b>          | 0                     | 10,414         | 3,210        | 13,624         | 9.2        |
| <b>Hungary</b>         | 0                     | 1,970          | 74           | 2,044          | 1.4        |
| <b>Ireland</b>         | 0                     | 2,472          | 0            | 2,472          | 1.7        |
| <b>Italy</b>           | 0                     | 15,239         | 0            | 15,239         | 10.3       |
| <b>Lithuania</b>       | 80                    | 702            | 18           | 800            | 0.5        |
| <b>Luxembourg</b>      | no data               | no data        | no data      | 610            | 0.4        |
| <b>Latvia</b>          | 12                    | 453            | 3            | 468            | 0.3        |
| <b>Malta</b>           | 0                     | 77             | 18           | 95             | 0.1        |
| <b>The Netherlands</b> | 0                     | 4,757          | 30           | 4,787          | 3.2        |
| <b>Norway</b>          |                       |                |              | 1,950          | 1.3        |
| <b>Poland</b>          | 0                     | 8,418          | 166          | 8,584          | 5.8        |
| <b>Portugal</b>        | 0                     | 1,845          | 305          | 2,150          | 1.4        |
| <b>Romania</b>         | 0                     | 2,297          | 0            | 2,297          | 1.5        |
| <b>Sweden</b>          | 0                     | 4,914          | 322          | 5,236          | 3.5        |
| <b>Slovenia</b>        | 0                     | 741            | 80           | 821            | 0.6        |
| <b>Slovakia</b>        | 42                    | 751            | 17           | 810            | 0.5        |
| <b>Switzerland</b>     | 0                     | 3,605          | 348          | 4,630          | 3.1        |
| <b>United Kingdom</b>  | 0                     | 22,890         | 1,221        | 24,111         | 16.3       |
| <b>Total</b>           | <b>8,992</b>          | <b>127,064</b> | <b>9,072</b> | <b>148,365</b> | <b>100</b> |

Source : EU Fuel monitoring 2003-2006  
EPTC (2005-2009)

### APPENDIX 3 GASOLINE COMPOSITION 2000 – 2009 (SGS) COUNTRY WIDE AVERAGE LEVEL IN HALF YEAR PERIODS

#### INTRODUCTION

For many years SGS (<http://www.sgs.com/en.aspx>) has been gathering information on gasoline by obtaining gasoline samples from randomly selected retail stations in the EU-27 Member States, Norway, Turkey and Switzerland.

For the purpose of this report CONCAWE has acquired the results of these analyses on GEO in these samples that were taken between winter 1999/2000 and summer 2009. The total number of samples analysed for GEOs and TBA are 1239. These are a subset of the 5089 samples taken in that period, because the blending of GEOs was not done in all these Countries in the period covered. In Table A3-1 the distribution of these over the Countries included in the di-annual SGS-sampling together with the number of samples per gasoline type that were tested.

**Table A3-1. SGS-Gasoline sampling and GEO analyses between 1999 and 2009**

| Country         | Total | LRG | PL | PUL | PULP | RL | RUL | No info |
|-----------------|-------|-----|----|-----|------|----|-----|---------|
| Austria         | 50    | -   | -  | 20  | 20   | -  | 10  | -       |
| Belgium         | 40    | -   | -  | 20  | 20   | -  | -   | -       |
| Bulgaria        | 44    | -   | -  | 20  | 17   | 5  | 2   | -       |
| Cyprus          | 27    | -   | 2  | 13  | 11   | -  | 1   | -       |
| Czech Republic  | 49    | -   | 3  | 20  | 20   | -  | 6   | -       |
| Denmark         | 49    | 1   | -  | 20  | 19   | -  | 9   | -       |
| Estonia         | 43    | -   | -  | 20  | 14   | -  | 9   | -       |
| Finland         | 40    | -   | -  | 20  | 20   | -  | -   | -       |
| France          | 40    | -   | -  | 20  | 20   | -  | -   | -       |
| Germany         | 49    | -   | -  | 20  | 20   | -  | 9   | -       |
| Greece          | 47    | 2   | 4  | 20  | 20   | -  | -   | 1       |
| Hungary         | 49    | -   | -  | 20  | 20   | -  | 9   | -       |
| Ireland         | 37    | 4   | -  | 20  | 12   | -  | 1   | -       |
| Italy           | 39    | -   | 4  | 20  | 15   | -  | -   | -       |
| Latvia          | 44    | -   | -  | 20  | 15   | -  | 9   | -       |
| Lithuania       | 49    | -   | -  | 20  | 20   | -  | 9   | -       |
| Luxemburg       | 40    | -   | -  | 20  | 18   | -  | 2   | -       |
| Malta           | 30    | 12  | 2  | 15  | 1    | -  | -   | -       |
| The Netherlands | 40    | -   | -  | 20  | 20   | -  | -   | -       |
| Norway          | 40    | -   | -  | 20  | 20   | -  | -   | -       |
| Poland          | 44    | -   | 1  | 20  | 20   | 3  | -   | -       |
| Portugal        | 40    | -   | -  | 20  | 20   | -  | -   | -       |
| Romania         | 35    | -   | -  | 20  | 15   | -  | -   | -       |



| Country        | Total       | LRG       | PL        | PUL        | PULP       | RL        | RUL       | No info  |
|----------------|-------------|-----------|-----------|------------|------------|-----------|-----------|----------|
| Slovakia       | 42          | 2         | -         | 20         | 20         | -         | -         | -        |
| Slovenia       | 36          | -         | -         | 17         | 17         | -         | 2         | -        |
| Spain          | 47          | 3         | 4         | 20         | 20         | -         | -         | -        |
| Sweden         | 40          | -         | -         | 20         | 20         | -         | -         | -        |
| Switzerland    | 40          | -         | -         | 20         | 20         | -         | -         | -        |
| Turkey         | 29          | -         | 3         | 14         | 10         | 2         | -         | -        |
| United Kingdom | 40          | -         | -         | 20         | 20         | -         | -         | -        |
| <b>Total</b>   | <b>1239</b> | <b>24</b> | <b>23</b> | <b>579</b> | <b>524</b> | <b>10</b> | <b>78</b> | <b>1</b> |

GEO in Lead Replacement Gasoline (LRG or LRP) in % m/m

| Oxygenate<br>Fuel Type | Country        | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |             |
|------------------------|----------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|-------------|
|                        |                | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |             |
|                        |                | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer      |
|                        | Austria        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Belgium        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Bulgaria       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Croatia        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Cyprus         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Czech Republic | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,00        |
|                        | Denmark        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Estonia        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Finland        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | France         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Germany        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Greece         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 5,93        |
|                        | Hungary        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Ireland        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 4,29        |
|                        | Italy          | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Latvia         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Lithuania      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Luxembourg     | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 3,26        |
|                        | Malta          | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Netherlands    | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Norway         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Poland         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Portugal       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Romania        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Slovakia       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,96        |
|                        | Slovenia       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Spain          | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 3,48        |
|                        | Sweden         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | Switzerland    | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | United Kingdom | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
|                        | <b>Average</b> |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        | <b>2,99</b> |

| Oxygenate<br>Fuel Type | ETBE   |        | LRG    |        | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |             |
|------------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|-------------|
|                        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |             |
|                        | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer      |
| Austria                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Belgium                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Bulgaria               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Croatia                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Cyprus                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Czech Republic         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Denmark                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,00        |
| Estonia                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Finland                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| France                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Germany                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Greece                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,00        |
| Hungary                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Ireland                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,00        |
| Italy                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Latvia                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Lithuania              | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Luxembourg             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Malta                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,03        |
| Netherlands            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Norway                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Poland                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Portugal               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Romania                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Slovakia               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Slovenia               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0,00        |
| Spain                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Sweden                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Switzerland            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 1,53        |
| United Kingdom         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| <b>Average</b>         |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        | <b>0,26</b> |

| Oxygenate<br>Fuel Type | TAME<br>LRG |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |             |  |
|------------------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|-------------|--|
|                        | Year        |        | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |             |  |
|                        | Winter      | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |             |  |
| Austria                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Belgium                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Bulgaria               | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Croatia                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Cyprus                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Czech Republic         | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Denmark                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Estonia                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Finland                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| France                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Germany                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Greece                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Hungary                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Ireland                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Italy                  | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Latvia                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Lithuania              | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Luxembourg             | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Malta                  | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Netherlands            | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Nonway                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Poland                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Portugal               | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Romania                | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Slovakia               | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Slovenia               | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Spain                  | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Sweden                 | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| Switzerland            | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| United Kingdom         | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |  |
| <b>Average</b>         |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        | <b>0.92</b> |  |

| Oxygenate<br>Fuel Type | DIPE<br>LRG | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |             |  |  |  |
|------------------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|---|-------------|--|--|--|
|                        |             | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |   |             |  |  |  |
|                        |             | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |   |             |  |  |  |
| Austria                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Belgium                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Bulgaria               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Croatia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Cyprus                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Czech Republic         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Denmark                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Estonia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Finland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| France                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Germany                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Greece                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Hungary                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Ireland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Italy                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Latvia                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Lithuania              |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Luxembourg             |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Malta                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | 0.00        |  |  |  |
| Netherlands            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Norway                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Poland                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Portugal               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Romania                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Slovakia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Slovenia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Spain                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Sweden                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| Switzerland            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| United Kingdom         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | -           |  |  |  |
| <b>Average</b>         |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   | <b>0.00</b> |  |  |  |

| Oxygenate<br>Fuel Type | TBA<br>LRG | Year   |  | 2000   |  | 2001   |  | 2002   |  | 2003   |  | 2004   |  | 2005   |  | 2006   |  | 2007   |  | 2008   |  | 2009   |  |        |  |
|------------------------|------------|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|--------|--|
|                        |            | Winter |  | Summer |  | Winter |  | Summer |  | Winter |  | Summer |  | Winter |  | Summer |  | Winter |  | Summer |  | Winter |  | Summer |  |
|                        |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Austria                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Belgium                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Bulgaria               |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Croatia                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Cyprus                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Czech Republic         |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Denmark                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Estonia                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Finland                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| France                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Germany                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Greece                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Hungary                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Ireland                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Italy                  |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Latvia                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Lithuania              |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Luxembourg             |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Malta                  |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Netherlands            |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Norway                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Poland                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Portugal               |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Romania                |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Slovakia               |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Slovenia               |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Spain                  |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Sweden                 |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| Switzerland            |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| United Kingdom         |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |
| <b>Average</b>         |            |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |        |  |

GEO in Regular Leaded Gasoline (RL) in % m/m



| Oxygenate<br>Fuel Type | MTBE<br>RL | Year   |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |  |  |  |
|------------------------|------------|--------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---|--|--|--|
|                        |            | 2000   |             | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |   |  |  |  |
|                        |            | Winter | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |   |  |  |  |
| Austria                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Belgium                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Bulgaria               |            | -      | 0,29        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Croatia                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Cyprus                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Czech Republic         |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Denmark                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Estonia                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Finland                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| France                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Germany                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Greece                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Hungary                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Ireland                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Italy                  |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Latvia                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Lithuania              |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Luxembourg             |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Malta                  |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Netherlands            |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Norway                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Poland                 |            | -      | 0,46        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Portugal               |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Romania                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovakia               |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovenia               |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Spain                  |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Sweden                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Switzerland            |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| United Kingdom         |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| <b>Average</b>         |            |        | <b>0,37</b> |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |  |  |  |

| Oxygenate<br>Fuel Type | ETBE<br>RL | Year   |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |
|------------------------|------------|--------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|---|---|--|--|--|
|                        |            | 2000   |             | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |   |   |  |  |  |
|                        |            | Winter | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |   |   |  |  |  |
| Austria                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Belgium                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Bulgaria               |            | -      | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Croatia                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Cyprus                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Czech Republic         |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Denmark                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Estonia                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Finland                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| France                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Germany                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Greece                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Hungary                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Ireland                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Italy                  |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Latvia                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Lithuania              |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Luxembourg             |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Malta                  |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Netherlands            |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Norway                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Poland                 |            | -      | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Portugal               |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Romania                |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovakia               |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovenia               |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Spain                  |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Sweden                 |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Switzerland            |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| United Kingdom         |            | -      | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| <b>Average</b>         |            |        | <b>0,00</b> |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |

| Oxygenate<br>Fuel Type | TAME<br>RL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
|------------------------|------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|--|--|--|--|--|
|                        |            | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |  |  |  |  |  |
|                        |            | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |  |  |  |  |  |
| Austria                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Belgium                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Bulgaria               |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Croatia                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Cyprus                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Czech Republic         |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Denmark                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Estonia                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Finland                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| France                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Germany                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Greece                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Hungary                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Ireland                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Italy                  |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Latvia                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Lithuania              |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Luxembourg             |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Malta                  |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Netherlands            |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Norway                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Poland                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Portugal               |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Romania                |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Slovakia               |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Slovenia               |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Spain                  |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Sweden                 |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| Switzerland            |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| United Kingdom         |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |
| <b>Average</b>         |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |  |  |  |  |  |



| Oxygenate<br>Fuel Type | TBA<br>RL | Year | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |  |
|------------------------|-----------|------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--|
|                        |           |      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |  |
|                        |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Austria                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Belgium                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Bulgaria               |           |      |        | 0.00   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Croatia                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Cyprus                 |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Czech Republic         |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Denmark                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Estonia                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Finland                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| France                 |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Germany                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Greece                 |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Hungary                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Ireland                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Italy                  |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Latvia                 |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Lithuania              |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Luxembourg             |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Malta                  |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Netherlands            |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Norway                 |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Poland                 |           |      |        | 0.00   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Portugal               |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Romania                |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Slovakia               |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Slovenia               |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Spain                  |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Sweden                 |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| Switzerland            |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| United Kingdom         |           |      |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |
| <b>Average</b>         |           |      |        | 0.00   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |  |

GEO in Premium Leaded Gasoline (PL) in % m/m

| Oxygenate<br>Fuel Type | MTBE<br>PL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |      |  |  |
|------------------------|------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---|------|--|--|
|                        |            | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |   |      |  |  |
|                        |            | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |   |      |  |  |
| Austria                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Belgium                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Bulgaria               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Croatia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Cyprus                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Czech Republic         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Denmark                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Estonia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Finland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| France                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Germany                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Greece                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Hungary                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Ireland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Italy                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Latvia                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Lithuania              |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Luxembourg             |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Malta                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Netherlands            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Norway                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Poland                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Portugal               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Romania                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Slovakia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Slovenia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Spain                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Sweden                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| Switzerland            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| United Kingdom         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |      |  |  |
| <b>Average</b>         |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   | 1,46 |  |  |

| Oxygenate<br>Fuel Type | ETBE<br>PL | Year   |   | 2000   |   | 2001   |   | 2002   |   | 2003   |   | 2004   |   | 2005   |   | 2006   |   | 2007   |   | 2008   |   | 2009   |   |        |   |             |
|------------------------|------------|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|-------------|
|                        |            | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   |             |
|                        |            |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |             |
| Austria                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Belgium                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Bulgaria               |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Croatia                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Cyprus                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Czech Republic         |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Denmark                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Estonia                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Finland                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| France                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Germany                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Greece                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Hungary                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Ireland                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Italy                  |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Latvia                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Lithuania              |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Luxembourg             |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Malta                  |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Netherlands            |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Norway                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Poland                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Portugal               |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Romania                |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Slovakia               |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Slovenia               |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Spain                  |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Sweden                 |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| Switzerland            |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| United Kingdom         |            | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |             |
| <b>Average</b>         |            |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   | <b>0,18</b> |



| Oxygenate<br>Fuel Type | TAME<br>PL | Year   |        | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |
|------------------------|------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
|                        |            | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |
| Austria                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Belgium                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Bulgaria               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Croatia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Cyprus                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Czech Republic         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Denmark                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Estonia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Finland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| France                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Germany                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Greece                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Hungary                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Ireland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Italy                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Latvia                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Lithuania              |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Luxembourg             |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Malta                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Netherlands            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Norway                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Poland                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Portugal               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Romania                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Slovakia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Slovenia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Spain                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Sweden                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Switzerland            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| United Kingdom         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| <b>Average</b>         |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |

| Oxygenate<br>Fuel Type | DIPE<br>PL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |   |  |  |
|------------------------|------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|---|---|---|--|--|
|                        |            | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |   |   |   |  |  |
|                        |            | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |   |   |   |  |  |
| Austria                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Belgium                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Bulgaria               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Croatia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Cyprus                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Czech Republic         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Denmark                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Estonia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Finland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| France                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Germany                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Greece                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Hungary                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Ireland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Italy                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Latvia                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Lithuania              |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Luxembourg             |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Malta                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Netherlands            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Norway                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Poland                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Portugal               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Romania                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Slovakia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Slovenia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Spain                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Sweden                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| Switzerland            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| United Kingdom         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |
| <b>Average</b>         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - | - |  |  |

| Oxygenate<br>Fuel Type | TBA<br>PL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        | Average     |
|------------------------|-----------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|-------------|
|                        |           | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |             |
|                        |           | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |             |
| Austria                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Belgium                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Bulgaria               |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Croatia                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Cyprus                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0.00   | 0.00        |
| Czech Republic         |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Denmark                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Estonia                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Finland                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| France                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Germany                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Greece                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0.00   | 0.00        |
| Hungary                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Ireland                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Italy                  |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Latvia                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0.00   | 0.00        |
| Lithuania              |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Luxembourg             |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Malta                  |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0.00   | 0.00        |
| Netherlands            |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Norway                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Poland                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Portugal               |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0.00   | 0.00        |
| Romania                |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Slovakia               |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Slovenia               |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Spain                  |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| Sweden                 |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | 0.01   | 0.01        |
| Switzerland            |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| United Kingdom         |           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -           |
| <b>Average</b>         |           |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        | <b>0.00</b> |

GEO in Regular Unleaded Gasoline (RUL) in % m/m

| Oxygenate<br>Fuel Type | 2000        |             | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |
|------------------------|-------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
|                        | Year        |             | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        |
|                        | Winter      | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |
| Austria                | 0.52        | 0.73        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Belgium                | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Bulgaria               | 0.31        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Croatia                | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Cyprus                 | 3.81        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Czech Republic         | 1.61        | 2.01        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Denmark                | 0.47        | 0.06        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Estonia                | 2.30        | 2.01        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Finland                | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| France                 | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Germany                | 0.37        | 0.23        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Greece                 | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Hungary                | 0.26        | 0.56        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Ireland                | 2.62        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Italy                  | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Latvia                 | 1.88        | 1.07        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Lithuania              | 0.74        | 0.27        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Luxembourg             | 1.94        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Malta                  | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Netherlands            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Norway                 | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Poland                 | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Portugal               | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Romania                | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Slovakia               | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Slovenia               | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Spain                  | 0.31        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Sweden                 | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Switzerland            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| United Kingdom         | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| <b>Average</b>         | <b>1.32</b> | <b>0.87</b> |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |

| Oxygenate<br>Fuel Type | ETBE<br>RUL | Year        |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |
|------------------------|-------------|-------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|---|---|--|--|--|
|                        |             | 2000        |             | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |   |   |  |  |  |
|                        |             | Winter      | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |   |   |  |  |  |
| Austria                |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Belgium                |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Bulgaria               |             | 0,00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Croatia                |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Cyprus                 |             | 0,00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Czech Republic         |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Denmark                |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Estonia                |             | 0,00        | 0,02        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Finland                |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| France                 |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Germany                |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Greece                 |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Hungary                |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Ireland                |             | 0,00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Italy                  |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Latvia                 |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Lithuania              |             | 0,00        | 0,00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Luxembourg             |             | 0,00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Malta                  |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Netherlands            |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Norway                 |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Poland                 |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Portugal               |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Romania                |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovakia               |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovenia               |             | 0,00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Spain                  |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Sweden                 |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Switzerland            |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| United Kingdom         |             | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| <b>Average</b>         |             | <b>0,00</b> | <b>0,00</b> |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |

| Oxygenate<br>Fuel Type | TAME<br>RUL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |  |  |  |
|------------------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---|--|--|--|
|                        |             | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |   |  |  |  |
|                        |             | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |   |  |  |  |
| Austria                |             | -      | 0,00   | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Belgium                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Bulgaria               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Croatia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Cyprus                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Czech Republic         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Denmark                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Estonia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Finland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| France                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Germany                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Greece                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Hungary                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Ireland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Italy                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Latvia                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Lithuania              |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Luxembourg             |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Malta                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Netherlands            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Norway                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Poland                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Portugal               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Romania                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovakia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovenia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Spain                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Sweden                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Switzerland            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| United Kingdom         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Average                |             | 0,00   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |  |  |  |

| Oxygenate<br>Fuel Type | DIPE<br>RUL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        | Average |
|------------------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---------|
|                        |             | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |         |
| Country                |             | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |         |
| Austria                |             | -      | 0,00   | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Belgium                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Bulgaria               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Croatia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Cyprus                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Czech Republic         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Denmark                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Estonia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Finland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| France                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Germany                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Greece                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Hungary                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Ireland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Italy                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Latvia                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Lithuania              |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Luxembourg             |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Malta                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Netherlands            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Norway                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Poland                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Portugal               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Romania                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Slovakia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Slovenia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Spain                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Sweden                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Switzerland            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| United Kingdom         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |
| Average                |             | -      | 0,00   | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |         |



| Oxygenate<br>Fuel Type | TBA<br>RUL | Year        |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |
|------------------------|------------|-------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|---|---|--|--|--|
|                        |            | 2000        |             | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |   |   |  |  |  |
|                        |            | Winter      | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |   |   |  |  |  |
| Austria                |            | 0.01        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Belgium                |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Bulgaria               |            | 0.00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Croatia                |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Cyprus                 |            | 0.00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Czech Republic         |            | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Denmark                |            | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Estonia                |            | 0.01        | 0.01        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Finland                |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| France                 |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Germany                |            | 0.04        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Greece                 |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Hungary                |            | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Ireland                |            | 0.00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Italy                  |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Latvia                 |            | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Lithuania              |            | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Luxembourg             |            | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Malta                  |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Netherlands            |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Norway                 |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Poland                 |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Portugal               |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Romania                |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovakia               |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovenia               |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Spain                  |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Sweden                 |            | 0.00        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Switzerland            |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| United Kingdom         |            | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| <b>Average</b>         |            | <b>0.00</b> | <b>0.00</b> |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |

GEO in Premium Unleaded Gasoline (PUL) in % m/m

| Oxygenate<br>Fuel Type | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |   |
|------------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---|
|                        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        | Year   |        |   |
|                        | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |   |
| Austria                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Belgium                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Bulgaria               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Croatia                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Cyprus                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Czech Republic         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Denmark                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Estonia                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Finland                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| France                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Germany                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Greece                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Hungary                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Ireland                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Italy                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Latvia                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Lithuania              | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Luxembourg             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Malta                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Netherlands            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Norway                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Poland                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Portugal               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Romania                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Slovakia               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Slovenia               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Spain                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Sweden                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| Switzerland            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| United Kingdom         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |
| <b>Average</b>         |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |

| Oxygenate<br>Fuel Type | ETBE   |        | PUL    |        | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |      |
|------------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|
|                        | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |
|                        | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year   | Year |
| Austria                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Belgium                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Bulgaria               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Croatia                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Cyprus                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Czech Republic         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Denmark                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Estonia                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Finland                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| France                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Germany                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Greece                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Hungary                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Ireland                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Italy                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Latvia                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Lithuania              | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Luxembourg             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Malta                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Netherlands            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Norway                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Poland                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Portugal               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Romania                | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Slovakia               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Slovenia               | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Spain                  | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Sweden                 | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| Switzerland            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| United Kingdom         | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    |
| <b>Average</b>         |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |

| Oxygenate<br>Fuel Type | TAME<br>PUL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |
|------------------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|------|---|---|--|--|--|
|                        |             | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009 |   |   |  |  |  |
|                        |             | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |      |   |   |  |  |  |
| Austria                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Belgium                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Bulgaria               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Croatia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Cyprus                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Czech Republic         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Denmark                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Estonia                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Finland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| France                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Germany                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Greece                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Hungary                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Ireland                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Italy                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Latvia                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Lithuania              |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Luxembourg             |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Malta                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Netherlands            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Norway                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Poland                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Portugal               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Romania                |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovakia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Slovenia               |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Spain                  |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Sweden                 |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| Switzerland            |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| United Kingdom         |             | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -    | - | - |  |  |  |
| <b>Average</b>         |             |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |      |   |   |  |  |  |

| Oxygenate<br>Fuel Type | DIPE |   | PUL |   | Year |   | 2000   |   | 2001   |   | 2002   |   | 2003   |   | 2004   |   | 2005   |   | 2006   |   | 2007   |   | 2008   |   | 2009   |   |        |   |        |   |
|------------------------|------|---|-----|---|------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|--------|---|
|                        |      |   |     |   |      |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   | Winter |   | Summer |   |
|                        |      |   |     |   |      |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |
| Austria                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      |   |
| Belgium                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Bulgaria               | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Croatia                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Cyprus                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Czech Republic         | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Denmark                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Estonia                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Finland                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| France                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Germany                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Greece                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Hungary                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Ireland                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Italy                  | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Latvia                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Lithuania              | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Luxembourg             | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Malta                  | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Netherlands            | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Norway                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Poland                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Portugal               | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Romania                | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Slovakia               | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Slovenia               | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Spain                  | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Sweden                 | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| Switzerland            | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| United Kingdom         | -    | - | -   | - | -    | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - | -      | - |
| <b>Average</b>         |      |   |     |   |      |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |        |   |

| Oxygenate<br>Fuel Type | TBA<br>PUL | Year   |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |  |  |  |
|------------------------|------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---|--|--|--|
|                        |            | 2000   |        | 2001   |        | 2002   |        | 2003   |        | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |   |  |  |  |
|                        |            | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |   |  |  |  |
| Austria                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Belgium                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Bulgaria               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Croatia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Cyprus                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Czech Republic         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Denmark                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Estonia                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Finland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| France                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Germany                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Greece                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Hungary                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Ireland                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Italy                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Latvia                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Lithuania              |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Luxembourg             |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Malta                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Netherlands            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Norway                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Poland                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Portugal               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Romania                |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovakia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovenia               |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Spain                  |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Sweden                 |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Switzerland            |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| United Kingdom         |            | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| <b>Average</b>         |            |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |        |   |  |  |  |

GEO in Super Premium Unleaded Gasoline (PULP) in % m/m



| Oxygenate<br>Fuel Type | MTBE<br>PULP | Year        | 2000        |             | 2001        |             | 2002        |             | 2003        |             | 2004   |        | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |
|------------------------|--------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
|                        |              |             | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |
|                        |              |             |             |             |             |             |             |             |             |             |        |        |        |        |        |        |        |        |        |        |        |        |
| Austria                | -            | 11.70       | 13.02       | 14.15       | 12.91       | 14.11       | 13.53       | 10.79       | 1.67        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Belgium                | -            | 8.84        | 9.03        | 7.67        | 10.30       | 12.23       | 11.32       | 8.11        | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Bulgaria               | -            | -           | 9.83        | 11.34       | 11.24       | 11.04       | 11.44       | 11.10       | 9.46        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Croatia                | -            | -           | -           | -           | -           | -           | -           | -           | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Cyprus                 | -            | -           | -           | 14.31       | 9.43        | 9.02        | 11.28       | 12.51       | 12.50       | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Czech Republic         | -            | 9.12        | 11.48       | 13.40       | 14.03       | 12.36       | 12.23       | 9.08        | 3.09        | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Denmark                | -            | 8.01        | 7.29        | 8.31        | 2.69        | 4.77        | 9.54        | 11.16       | 6.81        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Estonia                | -            | -           | -           | 8.77        | 8.44        | 10.31       | 11.62       | 14.45       | 13.82       | 9.47        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Finland                | -            | 9.92        | 10.09       | 10.55       | 8.61        | 11.46       | 10.71       | 11.80       | 0.37        | 0.54        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| France                 | -            | 2.61        | 4.32        | 4.31        | 1.64        | 4.83        | 2.28        | 0.96        | 0.38        | 0.06        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Germany                | -            | 12.35       | 11.13       | 11.02       | 10.67       | 9.66        | 6.49        | 4.15        | 4.29        | 5.62        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Greece                 | -            | 7.21        | 8.46        | 10.76       | 12.50       | 13.46       | 11.74       | 12.81       | 12.08       | 11.44       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Hungary                | -            | 10.19       | 11.07       | 14.47       | 11.28       | 13.19       | 12.49       | 5.05        | 4.60        | 1.84        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Ireland                | -            | 4.45        | 3.99        | 0.45        | 1.12        | 1.67        | -           | -           | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Italy                  | -            | -           | -           | 10.73       | 8.61        | 13.69       | 9.97        | 13.32       | 11.06       | 5.86        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Latvia                 | -            | -           | -           | 10.36       | 6.62        | 8.66        | 11.54       | 13.17       | 14.01       | 10.87       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Lithuania              | -            | 5.50        | 6.64        | 10.33       | 11.82       | 9.75        | 11.16       | 13.32       | 13.54       | 10.46       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Luxembourg             | -            | -           | 9.11        | 9.38        | 9.72        | 10.36       | 12.15       | 12.57       | 7.12        | 13.45       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Malta                  | -            | -           | -           | -           | -           | -           | -           | 0.43        | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Netherlands            | -            | 8.29        | 9.50        | 11.93       | 10.41       | 9.59        | 10.77       | 6.58        | 4.44        | 11.06       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Norway                 | -            | 0.00        | 0.06        | 6.28        | 2.22        | 6.09        | 7.84        | 11.18       | 10.28       | 10.28       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Poland                 | -            | 6.78        | 5.15        | 2.53        | 5.00        | 9.54        | 6.54        | 7.68        | 0.14        | 0.03        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Portugal               | -            | 8.49        | 7.15        | 10.99       | 8.69        | 12.87       | 11.74       | 12.05       | 13.15       | 13.42       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Romania                | -            | -           | -           | 13.08       | 10.47       | 12.46       | 12.60       | 10.49       | 10.10       | 10.48       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Slovakia               | -            | 9.34        | 9.98        | 10.92       | 12.98       | 12.80       | 11.77       | 7.10        | 2.19        | 2.57        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Slovenia               | -            | -           | 11.53       | 11.87       | 12.79       | 13.91       | 12.17       | 12.10       | 10.15       | 0.83        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Spain                  | -            | 8.54        | 6.00        | 7.98        | 1.58        | 2.19        | 1.53        | 1.93        | 1.90        | 0.61        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Sweden                 | -            | 10.07       | 6.96        | 7.83        | 5.74        | 7.42        | 12.16       | 13.45       | 11.80       | 10.51       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| Switzerland            | -            | 10.26       | 7.39        | 9.27        | 9.32        | 9.99        | 8.68        | 6.85        | 10.37       | 11.53       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| United Kingdom         | -            | 1.04        | 0.46        | 1.99        | 1.86        | 1.77        | 1.86        | 2.86        | 2.10        | 2.26        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      |
| <b>Average</b>         |              | <b>7.63</b> | <b>7.81</b> | <b>9.46</b> | <b>8.25</b> | <b>9.55</b> | <b>9.83</b> | <b>9.28</b> | <b>7.39</b> | <b>6.51</b> |        |        |        |        |        |        |        |        |        |        |        |        |

| Oxygenate<br>Fuel Type | ETBE<br>PULP | Year   |             |             |             |             |             |             |             |             |             |        |        |        |        |        |        |        |        |        |        |   |  |  |  |
|------------------------|--------------|--------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|---|--|--|--|
|                        |              | 2000   |             | 2001        |             | 2002        |             | 2003        |             | 2004        |             | 2005   |        | 2006   |        | 2007   |        | 2008   |        | 2009   |        |   |  |  |  |
|                        |              | Winter | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer | Winter | Summer |   |  |  |  |
| Austria                |              | -      | 0.00        | 0.00        | 0.04        | 0.00        | 0.05        | 0.21        | 0.28        | 3.82        | 11.24       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Belgium                |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.49        | 0.28        | 0.84        | 2.93        | 2.93        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Bulgaria               |              | -      | 0.00        | 0.00        | 0.01        | 0.00        | 0.02        | 0.11        | 1.86        | 1.76        | 1.76        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Croatia                |              | -      | -           | -           | -           | -           | -           | -           | -           | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Cyprus                 |              | -      | -           | -           | 0.00        | 0.00        | 0.00        | 0.03        | 0.02        | 0.01        | 0.01        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Czech Republic         |              | -      | 0.32        | 0.00        | 0.02        | 0.00        | 0.93        | 0.57        | 4.72        | 10.85       | 10.85       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Denmark                |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Estonia                |              | -      | 0.00        | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.03        | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Finland                |              | -      | 0.00        | 0.00        | 0.00        | 0.14        | 0.74        | 0.25        | 1.02        | 11.70       | 11.90       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| France                 |              | -      | 3.62        | 1.71        | 2.34        | 3.97        | 2.96        | 6.68        | 11.46       | 13.51       | 13.05       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Germany                |              | -      | 0.00        | 0.00        | 0.12        | 0.16        | 2.71        | 4.99        | 6.30        | 8.00        | 5.31        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Greece                 |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.16        | 0.20        | 0.28        | 1.06        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Hungary                |              | -      | 0.00        | 0.00        | 0.00        | 0.02        | 0.04        | 0.29        | 6.12        | 8.04        | 12.02       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Ireland                |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | -           | -           | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Italy                  |              | -      | -           | -           | 0.00        | 0.00        | 0.20        | 0.61        | 0.28        | 2.64        | 5.44        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Latvia                 |              | -      | -           | -           | 0.00        | 0.00        | 0.00        | 0.03        | 0.08        | 0.00        | 2.05        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Lithuania              |              | -      | 0.00        | 0.00        | 0.00        | 0.03        | 0.00        | 0.01        | 0.37        | 0.00        | 2.34        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Luxembourg             |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.09        | 0.64        | 0.70        | 3.84        | 0.48        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Malta                  |              | -      | -           | -           | -           | -           | -           | -           | 0.53        | -           | -           | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Netherlands            |              | -      | 0.00        | 0.00        | 0.00        | 0.02        | 0.01        | 0.03        | 3.97        | 4.19        | 0.43        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Norway                 |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.01        | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Poland                 |              | -      | 0.00        | 0.66        | 3.37        | 2.95        | 1.42        | 4.14        | 3.83        | 9.79        | 11.07       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Portugal               |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.09        | 0.13        | 0.00        | 0.00        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Romania                |              | -      | -           | -           | 0.00        | 0.00        | 0.00        | 0.11        | 0.64        | 0.19        | 0.76        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovakia               |              | -      | 0.02        | 0.17        | 0.12        | 0.18        | 0.00        | 0.03        | 4.40        | 10.88       | 11.65       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Slovenia               |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 1.82        | 1.78        | 2.86        | 11.66       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Spain                  |              | -      | 0.00        | 4.82        | 2.11        | 6.87        | 7.19        | 9.19        | 11.43       | 11.35       | 12.01       | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Sweden                 |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.02        | 0.24        | 0.00        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| Switzerland            |              | -      | 0.16        | 0.04        | 0.03        | 0.08        | 1.44        | 4.31        | 5.59        | 2.29        | 2.28        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| United Kingdom         |              | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.01        | 0.04        | 0.04        | 0.02        | -      | -      | -      | -      | -      | -      | -      | -      | -      | -      | - |  |  |  |
| <b>Average</b>         |              |        | <b>0.21</b> | <b>0.32</b> | <b>0.29</b> | <b>0.52</b> | <b>0.66</b> | <b>1.28</b> | <b>2.49</b> | <b>4.30</b> | <b>4.84</b> |        |        |        |        |        |        |        |        |        |        |   |  |  |  |

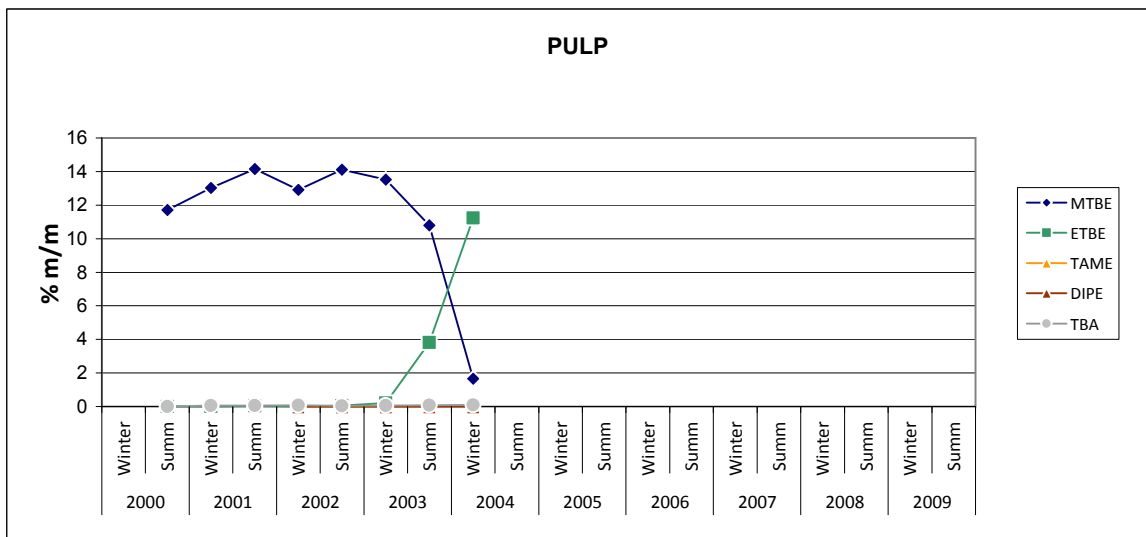
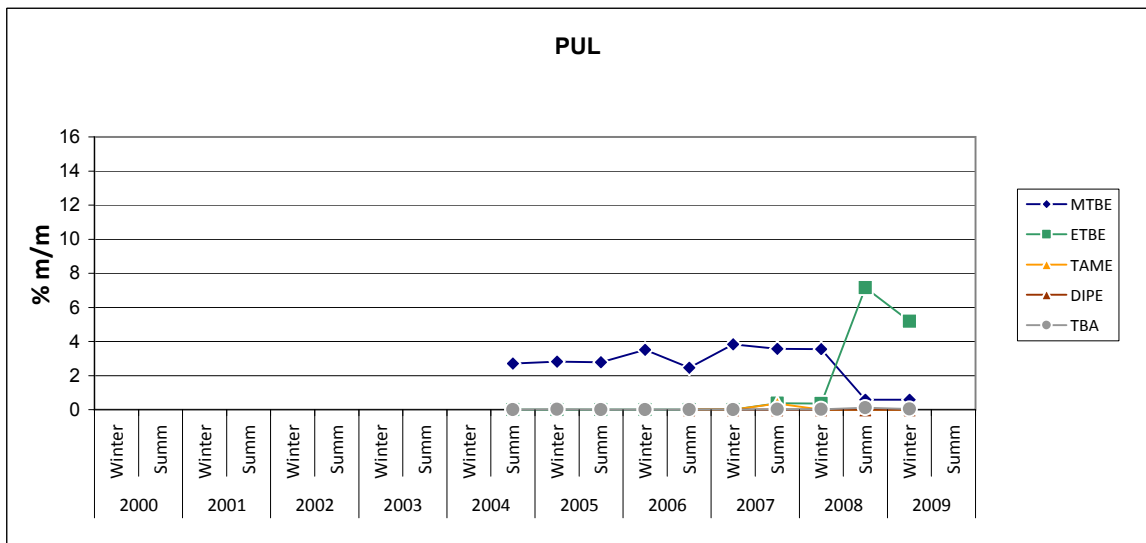
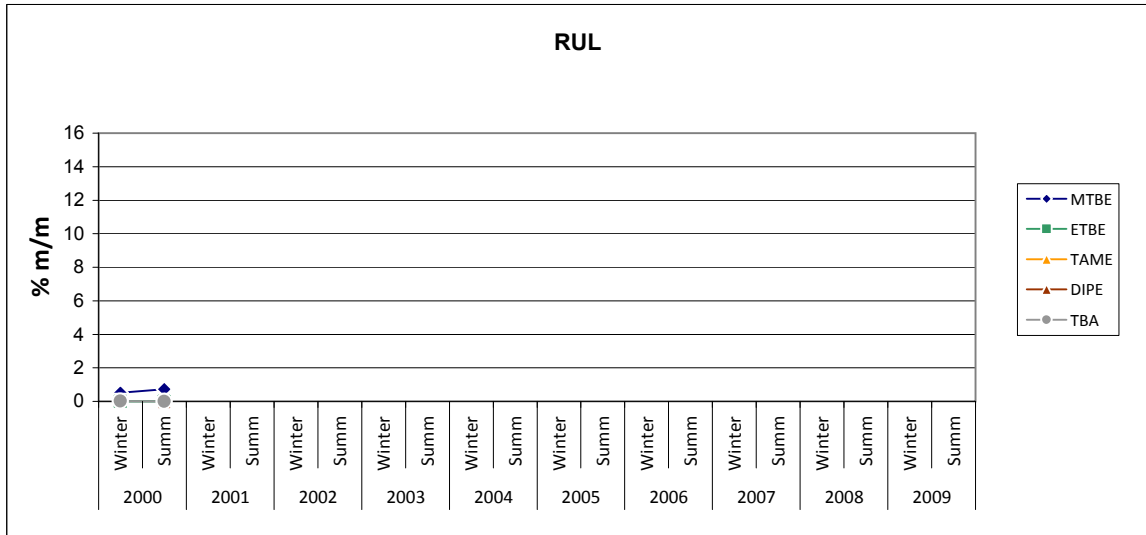
| Oxygenate<br>Fuel Type | TAME   |        | PULP   |        | Year   |        | 2000        |             | 2001        |             | 2002        |             | 2003        |             | 2004        |             | 2005        |             | 2006        |             | 2007        |             | 2008        |             | 2009        |             |             |
|------------------------|--------|--------|--------|--------|--------|--------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
|                        | Winter | Summer | Winter | Summer | Winter | Summer | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      |             |
|                        |        |        |        |        |        |        |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |
| Austria                | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.08        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Belgium                | -      | -      | -      | -      | -      | -      | 0.05        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Bulgaria               | -      | -      | -      | -      | -      | -      | 0.44        | 0.10        | 0.11        | 0.05        | 0.65        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Croatia                | -      | -      | -      | -      | -      | -      | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Cyprus                 | -      | -      | -      | -      | -      | -      | 0.59        | 2.46        | 1.71        | 0.97        | 1.62        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Czech Republic         | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.01        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Denmark                | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Estonia                | -      | -      | -      | -      | -      | -      | 0.20        | 0.29        | 0.01        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Finland                | -      | -      | -      | -      | -      | -      | 1.36        | 0.60        | 0.70        | 0.37        | 0.09        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| France                 | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Germany                | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Greece                 | -      | -      | -      | -      | -      | -      | 0.39        | 0.59        | 0.58        | 0.50        | 1.65        | 2.86        | -           | -           | 1.65        | 2.86        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Hungary                | -      | -      | -      | -      | -      | -      | 0.02        | 0.02        | 0.63        | 0.50        | 0.34        | 0.14        | -           | -           | 0.34        | 0.14        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Ireland                | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Italy                  | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.12        | 0.03        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Latvia                 | -      | -      | -      | -      | -      | -      | 0.27        | 0.49        | 0.01        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Lithuania              | -      | -      | -      | -      | -      | -      | 0.17        | 0.60        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Luxembourg             | -      | -      | -      | -      | -      | -      | 0.06        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Malta                  | -      | -      | -      | -      | -      | -      | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Netherlands            | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Norway                 | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Poland                 | -      | -      | -      | -      | -      | -      | 0.02        | 0.00        | 0.00        | 0.00        | 0.10        | 0.00        | 0.00        | 0.00        | 0.10        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Portugal               | -      | -      | -      | -      | -      | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Romania                | -      | -      | -      | -      | -      | -      | -           | 0.09        | 0.25        | 1.31        | 0.58        | 1.39        | -           | -           | 0.58        | 1.39        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Slovakia               | -      | -      | -      | -      | -      | -      | 0.00        | 0.00        | 0.13        | 0.42        | 0.00        | 0.01        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Slovenia               | -      | -      | -      | -      | -      | -      | 0.01        | 0.14        | 0.29        | 0.14        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Spain                  | -      | -      | -      | -      | -      | -      | -           | 0.00        | 0.00        | 0.03        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Sweden                 | -      | -      | -      | -      | -      | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Switzerland            | -      | -      | -      | -      | -      | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| United Kingdom         | -      | -      | -      | -      | -      | -      | -           | 0.00        | 0.00        | 0.00        | 0.34        | 0.00        | 0.00        | 0.00        | 0.34        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| <b>Average</b>         |        |        |        |        |        |        | <b>0.18</b> | <b>0.19</b> | <b>0.17</b> | <b>0.15</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> | <b>0.20</b> |

| Oxygenate<br>Fuel Type | DIPE<br>PULP |        |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |  |
|------------------------|--------------|--------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|--|
|                        | Year         |        | 2000        |             | 2001        |             | 2002        |             | 2003        |             | 2004        |             | 2005        |             | 2006        |             | 2007        |             | 2008        |             | 2009        |             |             |  |
|                        | Winter       | Summer | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      |             |  |
| Austria                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Belgium                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Bulgaria               | -            | -      | 0.00        | 0.04        | 0.05        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Croatia                | -            | -      | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |  |
| Cyprus                 | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Czech Republic         | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Denmark                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Estonia                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Finland                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| France                 | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Germany                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Greece                 | -            | -      | 0.00        | 0.00        | 0.06        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Hungary                | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Ireland                | -            | -      | 0.00        | 0.00        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |  |
| Italy                  | -            | -      | 0.00        | 0.00        | 0.03        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Latvia                 | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Lithuania              | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Luxembourg             | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Malta                  | -            | -      | -           | -           | -           | 0.05        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |  |
| Netherlands            | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Norway                 | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Poland                 | -            | -      | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Portugal               | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Romania                | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Slovakia               | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Slovenia               | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Spain                  | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Sweden                 | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| Switzerland            | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| United Kingdom         | -            | -      | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |  |
| <b>Average</b>         |              |        | <b>0.00</b> | <b>0.00</b> | <b>0.01</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> | <b>0.00</b> |  |

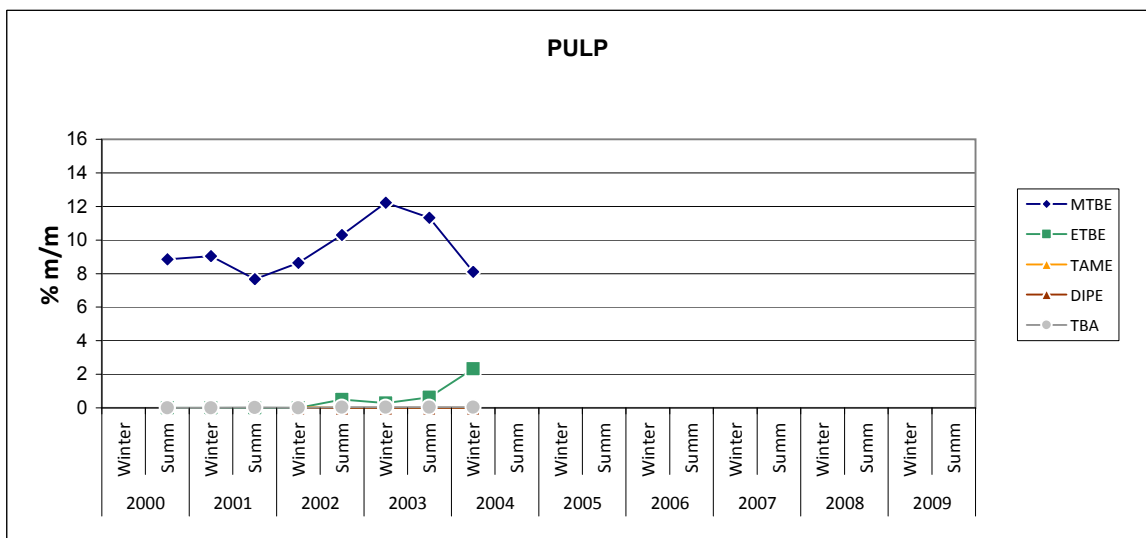
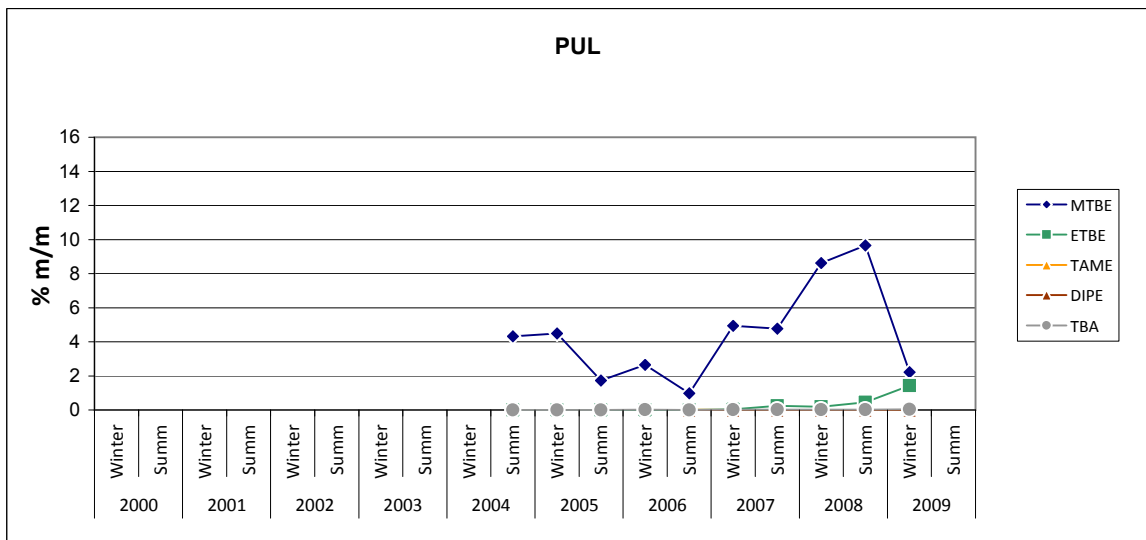
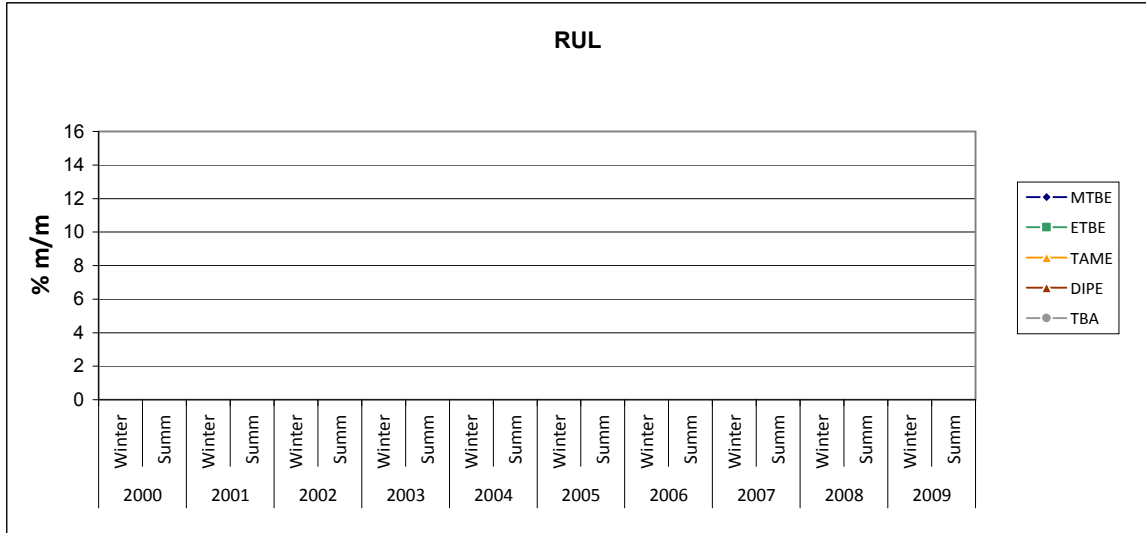
| Oxygenate<br>Fuel Type | 2000        |             | 2001        |             | 2002        |             | 2003        |             | 2004        |             | 2005        |             | 2006        |             | 2007        |             | 2008        |             | 2009        |             |             |
|------------------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
|                        | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      | Winter      | Summer      |             |
|                        | Year        |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |             |
| Austria                | -           | 0.00        | 0.06        | 0.05        | 0.04        | 0.04        | 0.05        | 0.07        | 0.09        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Belgium                | -           | 0.01        | 0.02        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.03        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.03        | 0.03        |
| Bulgaria               | -           | -           | 0.00        | 0.03        | 0.11        | 0.05        | 0.05        | 0.08        | 0.04        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Croatia                | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Cyprus                 | -           | -           | -           | 0.02        | 0.00        | 0.00        | 0.03        | 0.01        | 0.02        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Czech Republic         | -           | 0.00        | 0.06        | 0.04        | 0.11        | 0.16        | 0.05        | 0.10        | 0.12        | 0.05        | 0.05        | 0.10        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Denmark                | -           | 0.02        | 0.00        | 0.02        | 0.00        | 0.00        | 0.01        | 0.01        | 0.00        | 0.00        | 0.01        | 0.01        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        |
| Estonia                | -           | 0.01        | 0.02        | 0.04        | 0.09        | 0.12        | 0.03        | 0.04        | 0.03        | 0.02        | 0.03        | 0.04        | 0.03        | 0.02        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        |
| Finland                | -           | 0.01        | 0.02        | 0.01        | 0.01        | 0.03        | 0.05        | 0.09        | 0.21        | 0.19        | 0.09        | 0.09        | 0.06        | 0.13        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        |
| France                 | -           | 0.05        | 0.03        | 0.03        | 0.09        | 0.06        | 0.08        | 0.09        | 0.06        | 0.13        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        |
| Germany                | -           | 0.02        | 0.03        | 0.04        | 0.09        | 0.16        | 0.14        | 0.13        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        | 0.07        | 0.09        |
| Greece                 | -           | 0.00        | 0.00        | 0.02        | 0.02        | 0.09        | 0.03        | 0.01        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        |
| Hungary                | -           | 0.02        | 0.04        | 0.04        | 0.11        | 0.04        | 0.05        | 0.17        | 0.04        | 0.17        | 0.04        | 0.17        | 0.04        | 0.17        | 0.04        | 0.17        | 0.04        | 0.17        | 0.04        | 0.17        | 0.04        |
| Ireland                | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Italy                  | -           | -           | -           | 0.01        | 0.03        | 0.06        | 0.03        | 0.02        | 0.01        | 0.05        | 0.01        | 0.05        | 0.01        | 0.05        | 0.01        | 0.05        | 0.01        | 0.05        | 0.01        | 0.05        | 0.01        |
| Latvia                 | -           | 0.00        | 0.01        | 0.01        | 0.06        | 0.07        | 0.04        | 0.02        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        |
| Lithuania              | -           | 0.03        | 0.07        | 0.05        | 0.08        | 0.08        | 0.03        | 0.04        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        | 0.03        | 0.01        |
| Luxembourg             | -           | -           | 0.01        | 0.01        | 0.02        | 0.04        | 0.05        | 0.04        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        | 0.04        | 0.03        |
| Malta                  | -           | -           | -           | -           | -           | -           | -           | 0.00        | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           | -           |
| Netherlands            | -           | 0.00        | 0.00        | 0.02        | 0.06        | 0.02        | 0.03        | 0.05        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        | 0.03        |
| Norway                 | -           | 0.00        | 0.00        | 0.01        | 0.00        | 0.01        | 0.02        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        |
| Poland                 | -           | 0.01        | 0.03        | 0.02        | 0.06        | 0.01        | 0.04        | 0.05        | 0.08        | 0.03        | 0.04        | 0.05        | 0.08        | 0.03        | 0.04        | 0.05        | 0.08        | 0.03        | 0.04        | 0.05        | 0.08        |
| Portugal               | -           | 0.02        | 0.00        | 0.01        | 0.00        | 0.02        | 0.04        | 0.02        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        | 0.00        | 0.02        |
| Romania                | -           | -           | -           | 0.01        | 0.04        | 0.07        | 0.04        | 0.03        | 0.01        | 0.04        | 0.03        | 0.01        | 0.04        | 0.03        | 0.01        | 0.04        | 0.03        | 0.01        | 0.04        | 0.03        | 0.01        |
| Slovakia               | -           | 0.01        | 0.06        | 0.01        | 0.04        | 0.07        | 0.04        | 0.11        | 0.04        | 0.19        | 0.04        | 0.19        | 0.04        | 0.19        | 0.04        | 0.19        | 0.04        | 0.19        | 0.04        | 0.19        | 0.04        |
| Slovenia               | -           | 0.08        | 0.03        | 0.08        | 0.06        | 0.06        | 0.06        | 0.06        | 0.02        | 0.15        | 0.02        | 0.15        | 0.02        | 0.15        | 0.02        | 0.15        | 0.02        | 0.15        | 0.02        | 0.15        | 0.02        |
| Spain                  | -           | 0.02        | 0.04        | 0.08        | 0.07        | 0.06        | 0.10        | 0.14        | 0.14        | 0.17        | 0.14        | 0.17        | 0.14        | 0.17        | 0.14        | 0.17        | 0.14        | 0.17        | 0.14        | 0.17        | 0.14        |
| Sweden                 | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.01        | 0.03        | 0.03        | 0.02        | 0.04        | 0.02        | 0.04        | 0.02        | 0.04        | 0.02        | 0.04        | 0.02        | 0.04        | 0.02        | 0.04        | 0.02        |
| Switzerland            | -           | 0.07        | 0.21        | 0.05        | 0.05        | 0.11        | 0.16        | 0.19        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        | 0.08        |
| United Kingdom         | -           | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.00        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        | 0.01        |
| <b>Average</b>         | <b>0.01</b> | <b>0.03</b> | <b>0.02</b> | <b>0.05</b> | <b>0.05</b> | <b>0.06</b> | <b>0.05</b> | <b>0.07</b> | <b>0.05</b> | <b>0.06</b> | <b>0.05</b> | <b>0.07</b> | <b>0.05</b> | <b>0.06</b> | <b>0.05</b> | <b>0.07</b> | <b>0.05</b> | <b>0.06</b> | <b>0.05</b> | <b>0.07</b> | <b>0.05</b> |

**APPENDIX 4 GASOLINE ETHER OXYGENATES (GEO) IN 2000 – 2009  
(IN % MM) ON A COUNTRY BY COUNTRY BASIS**

# Austria

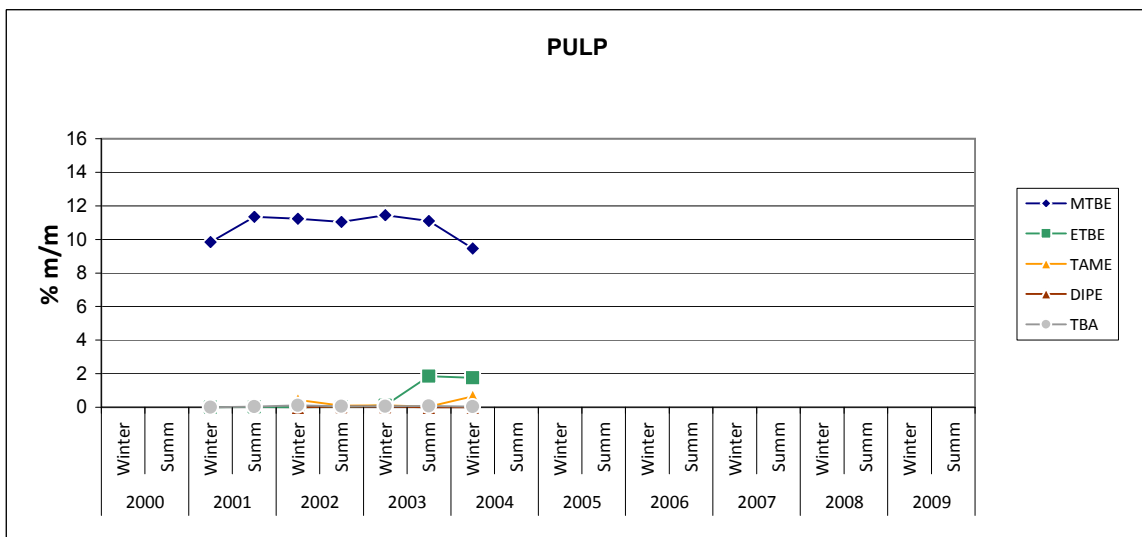
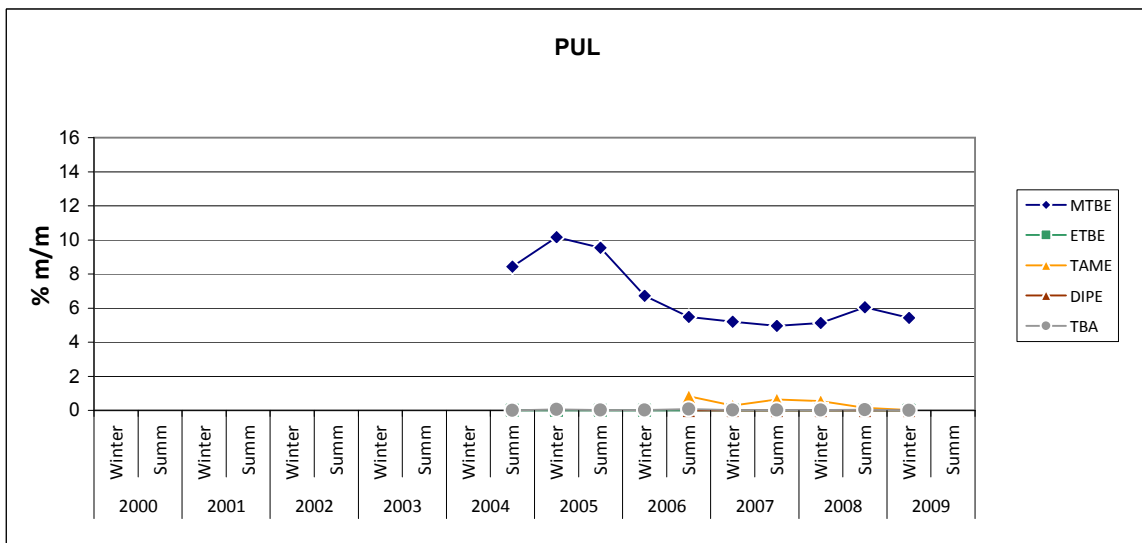
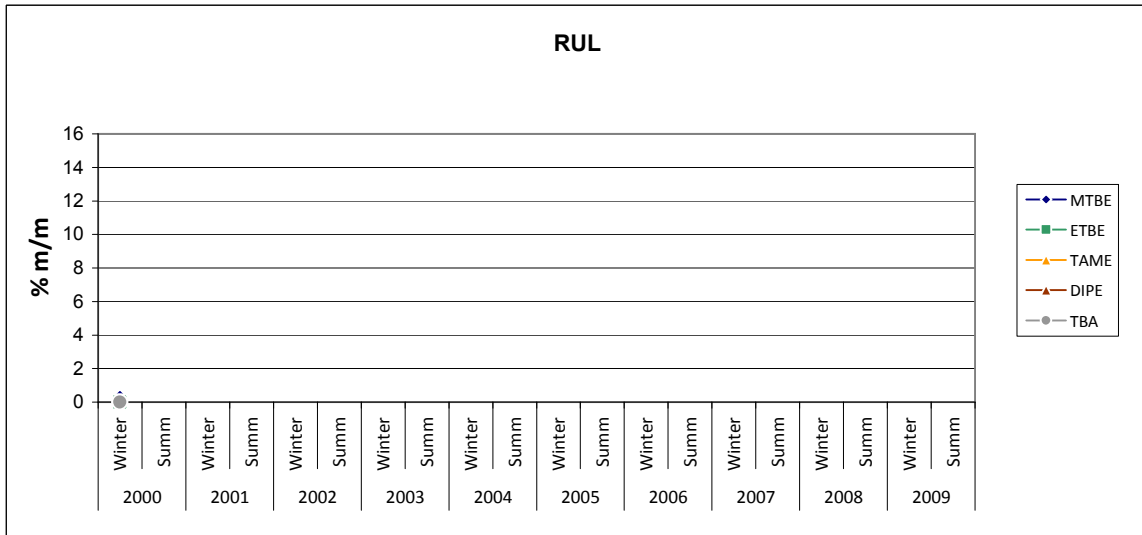


# Belgium

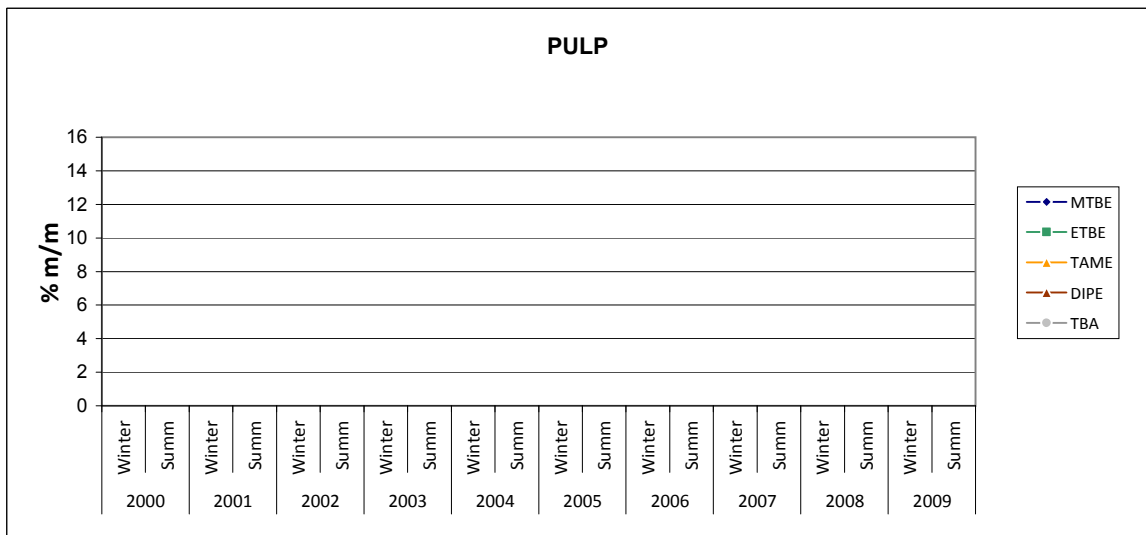
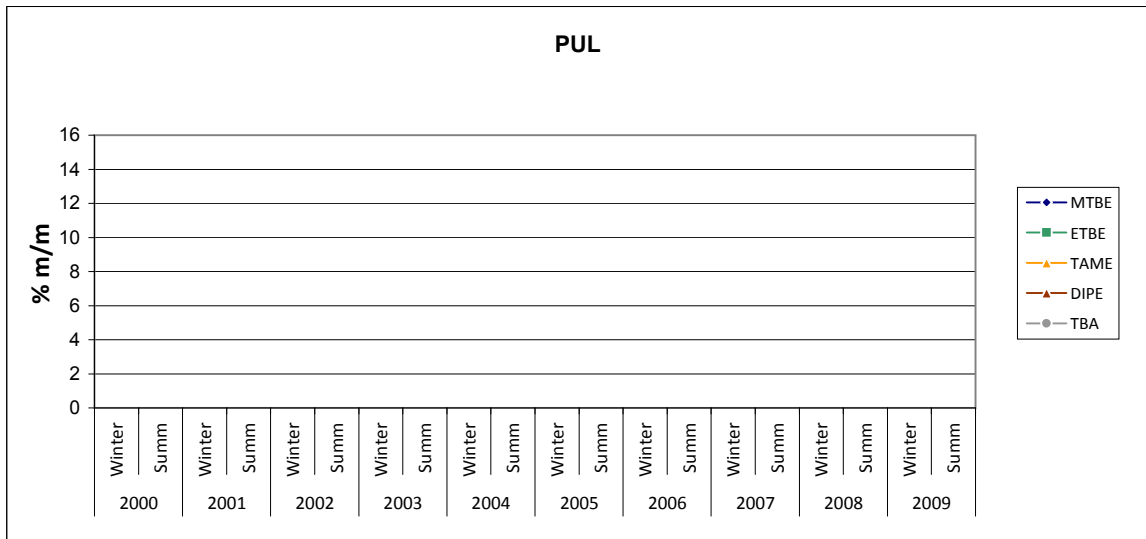
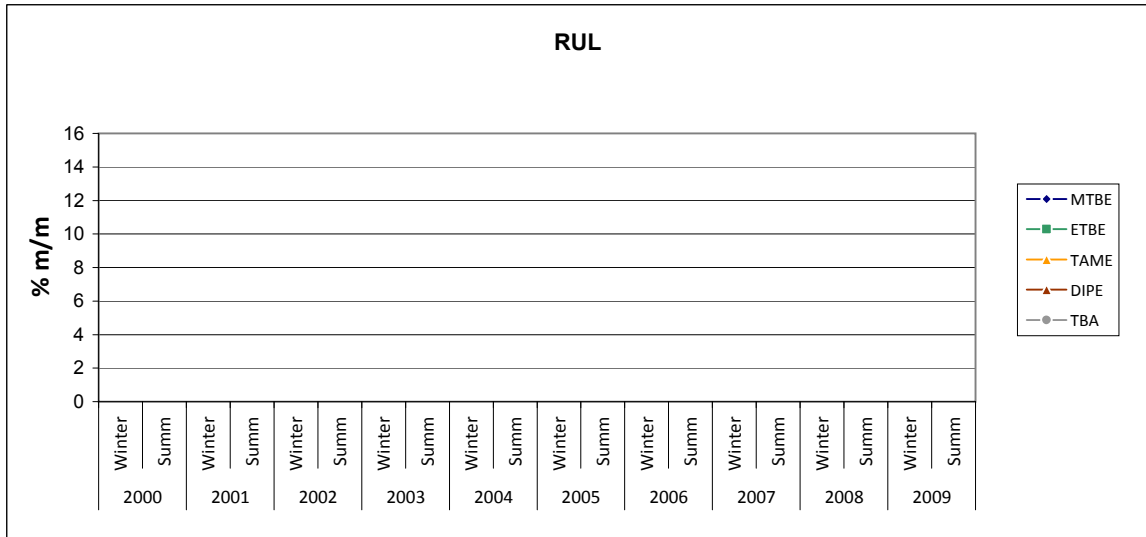




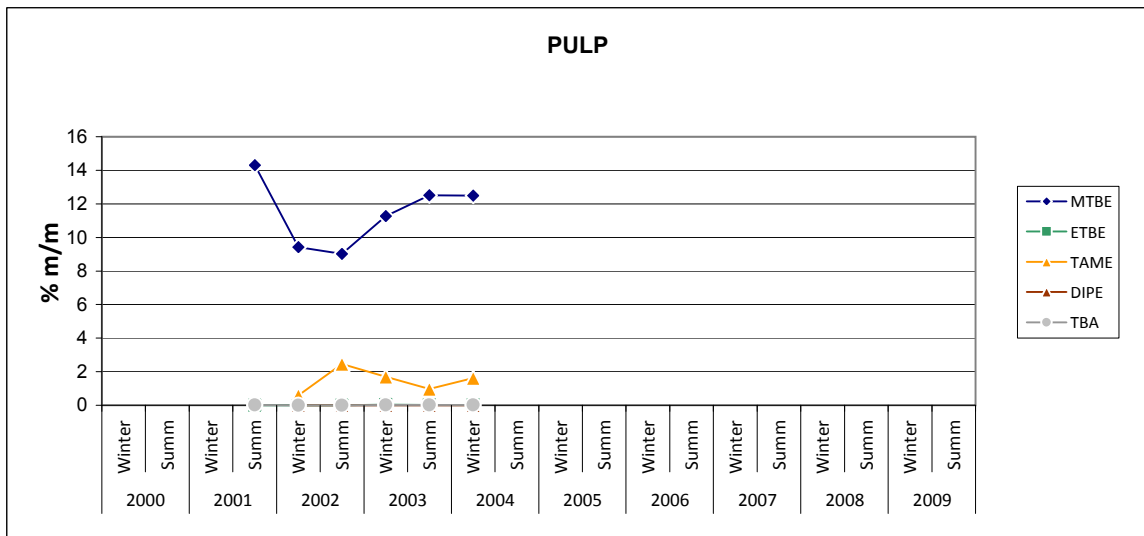
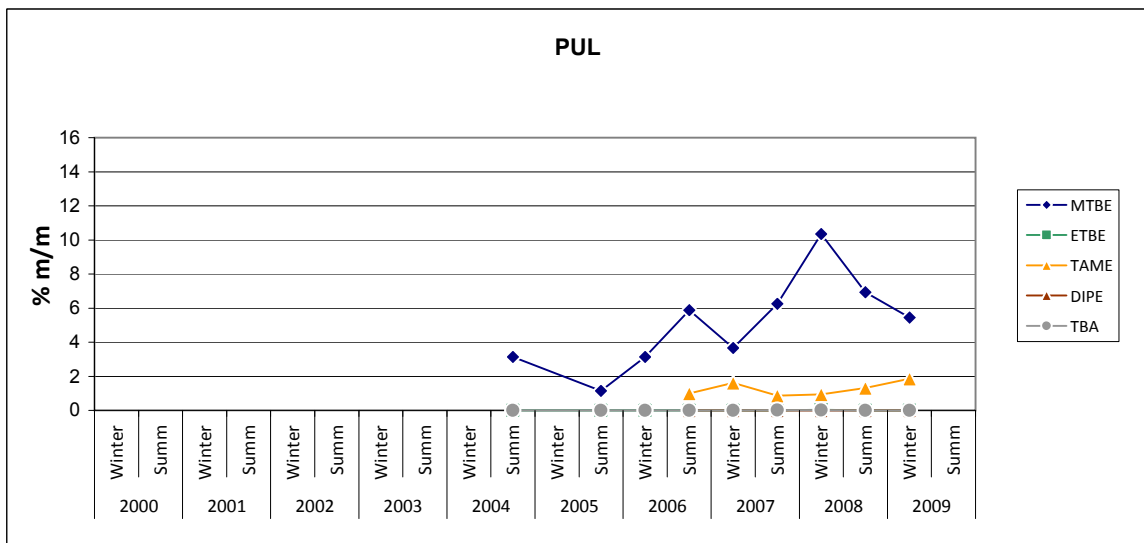
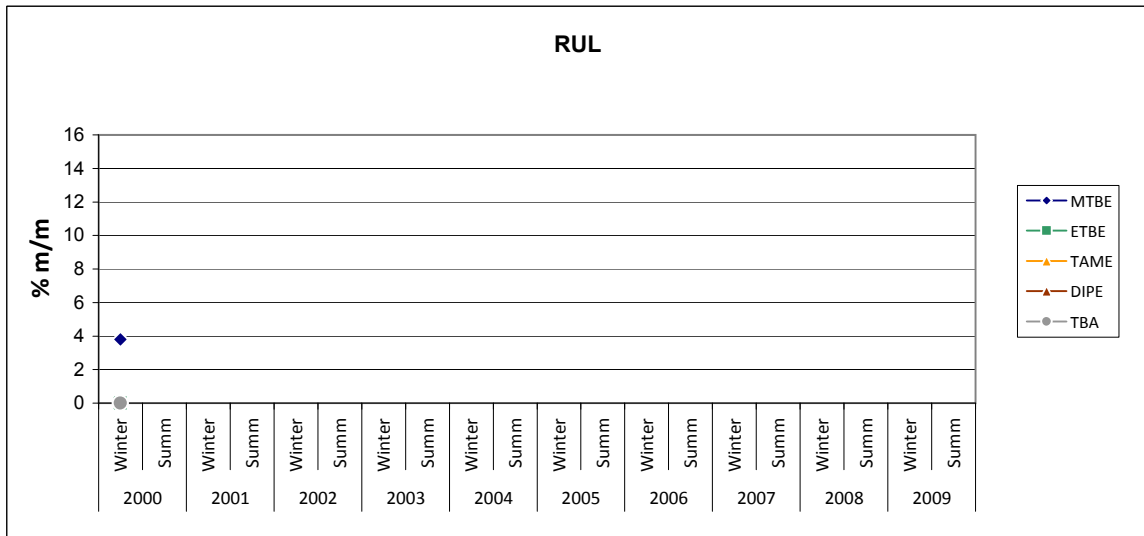
# Bulgaria



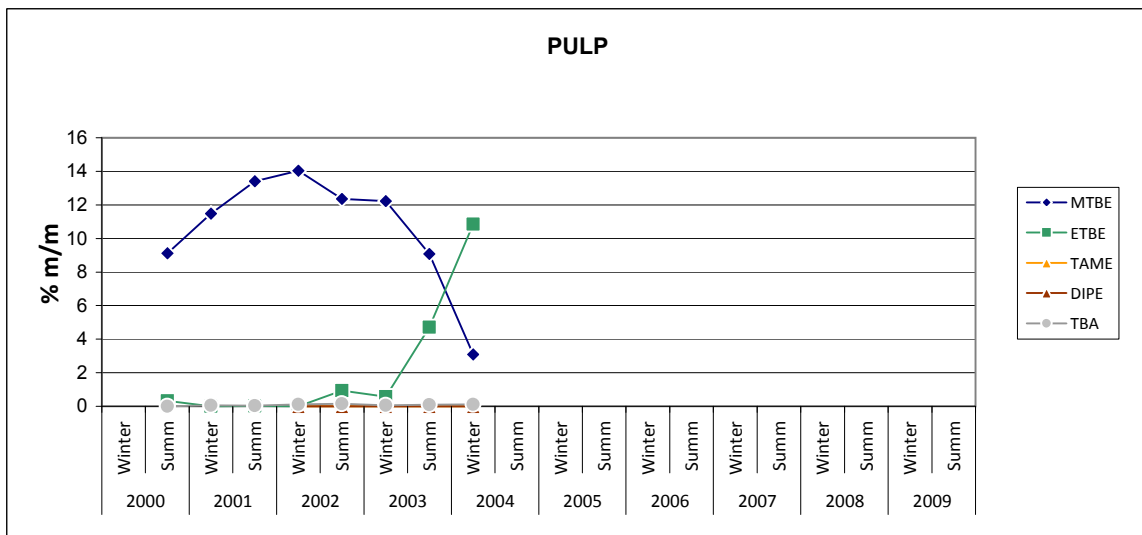
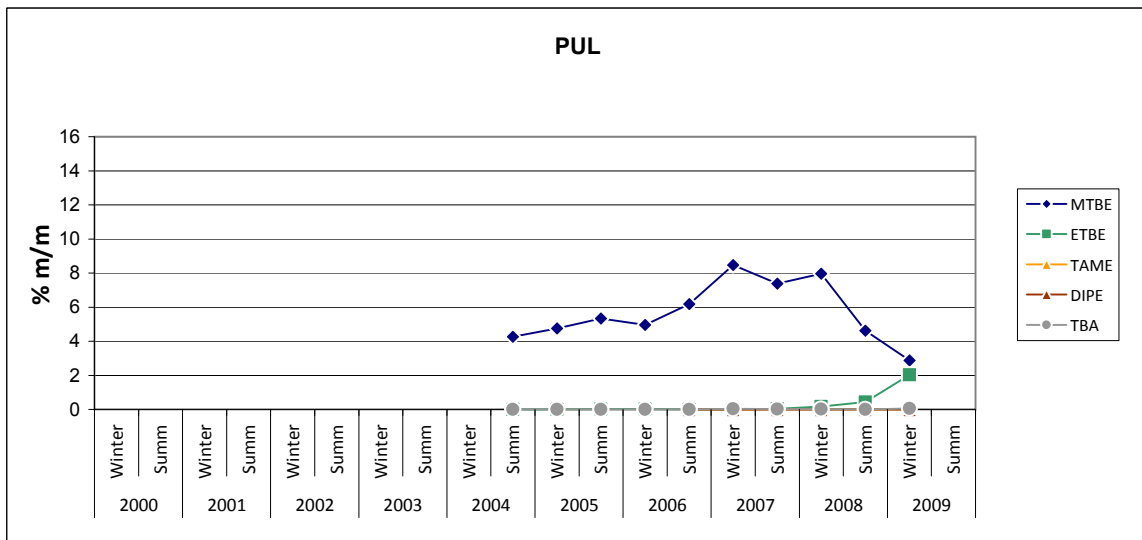
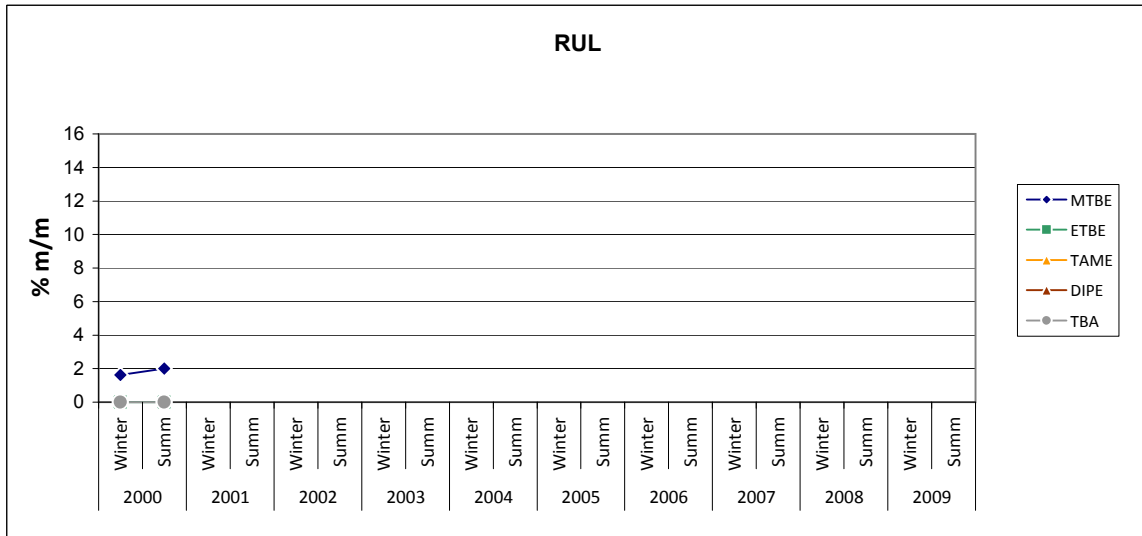
# Croatia



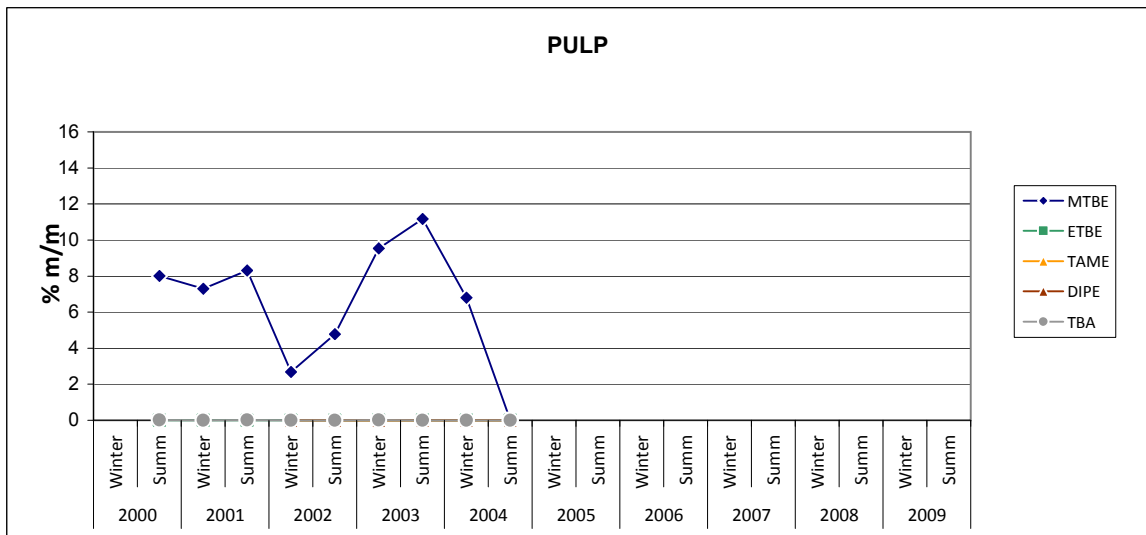
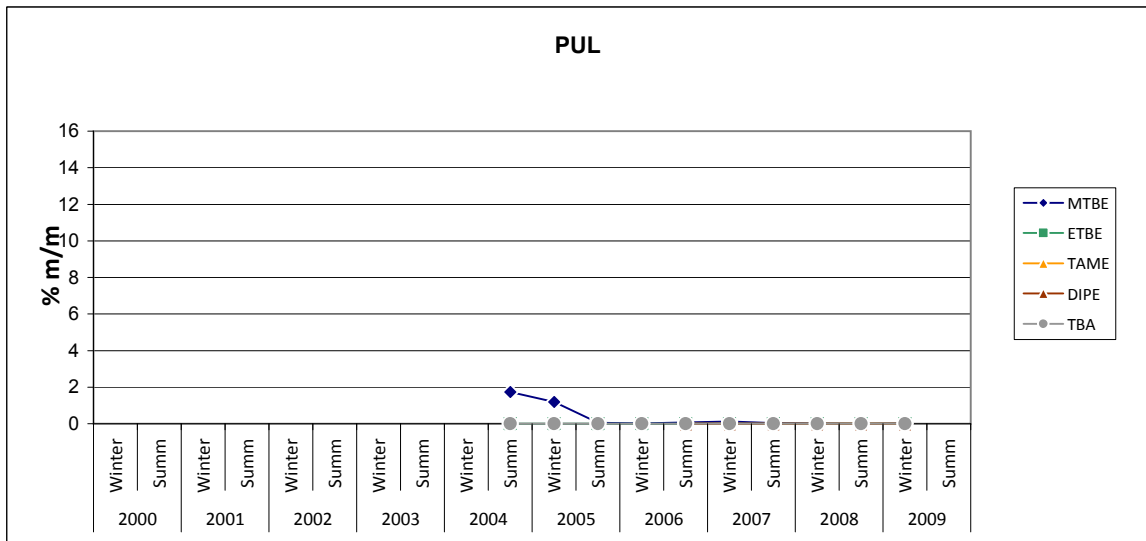
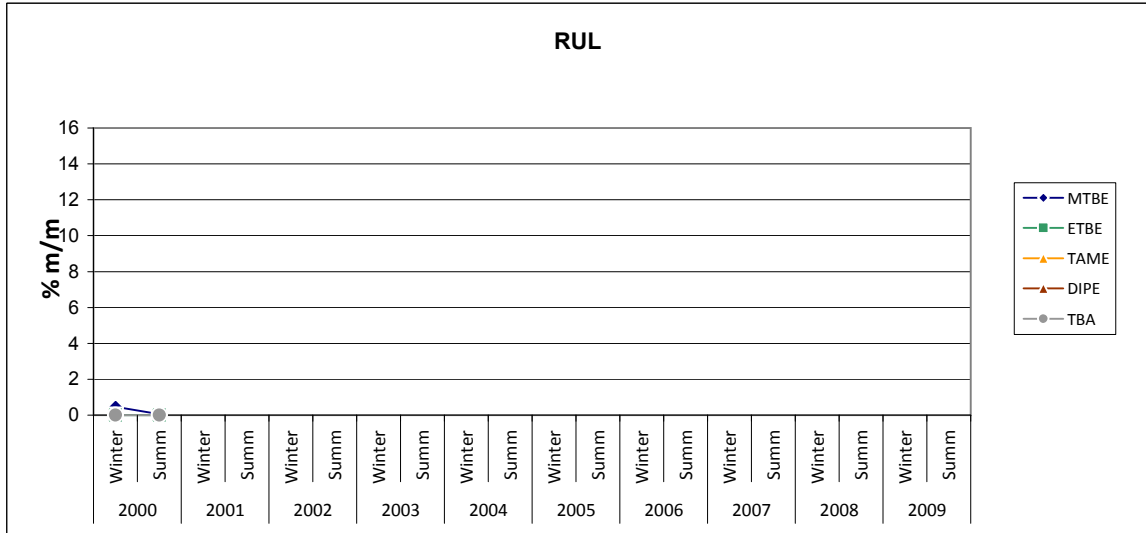
# Cyprus



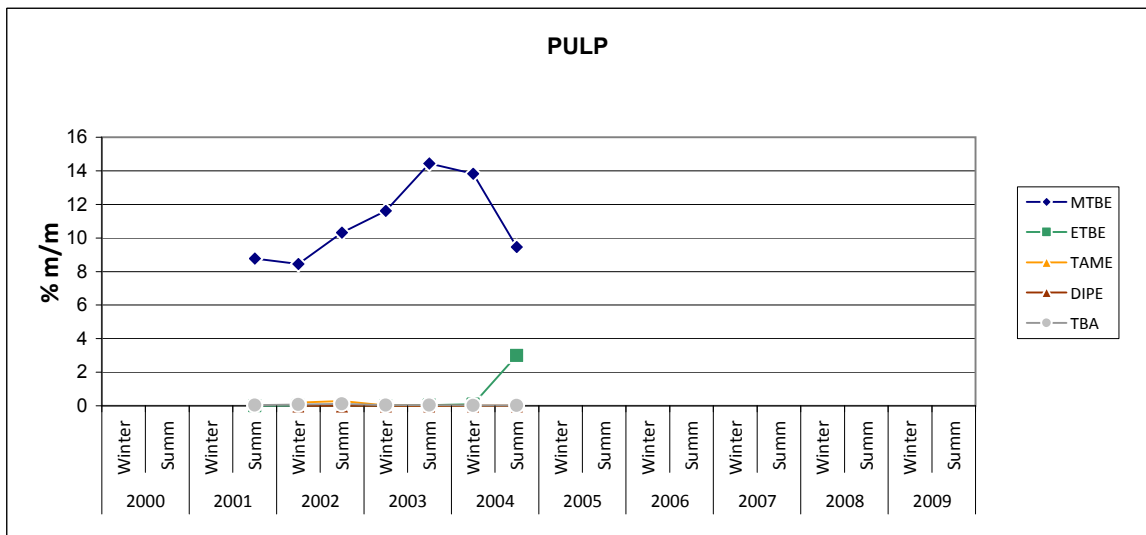
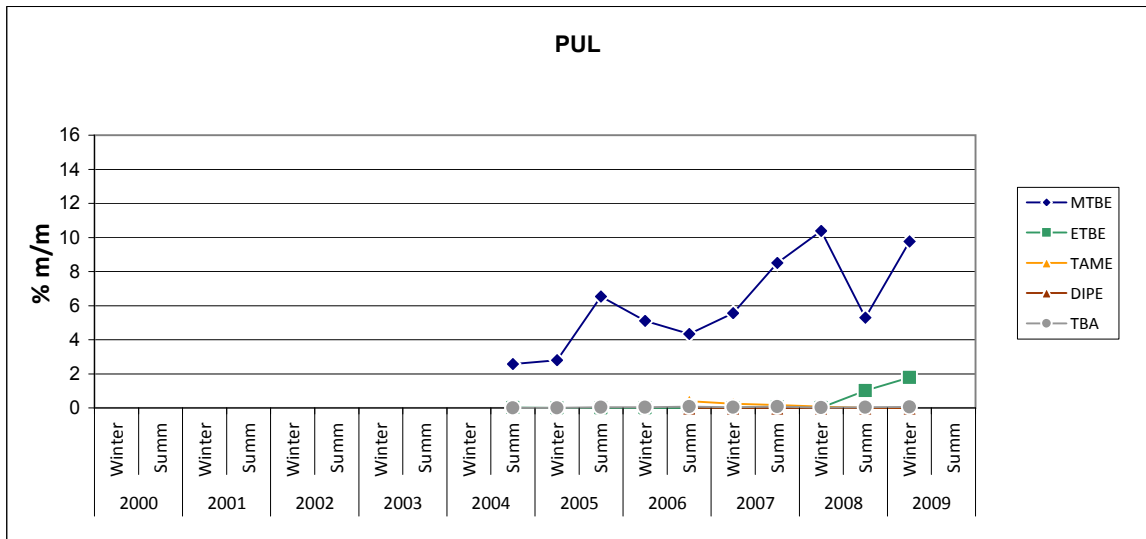
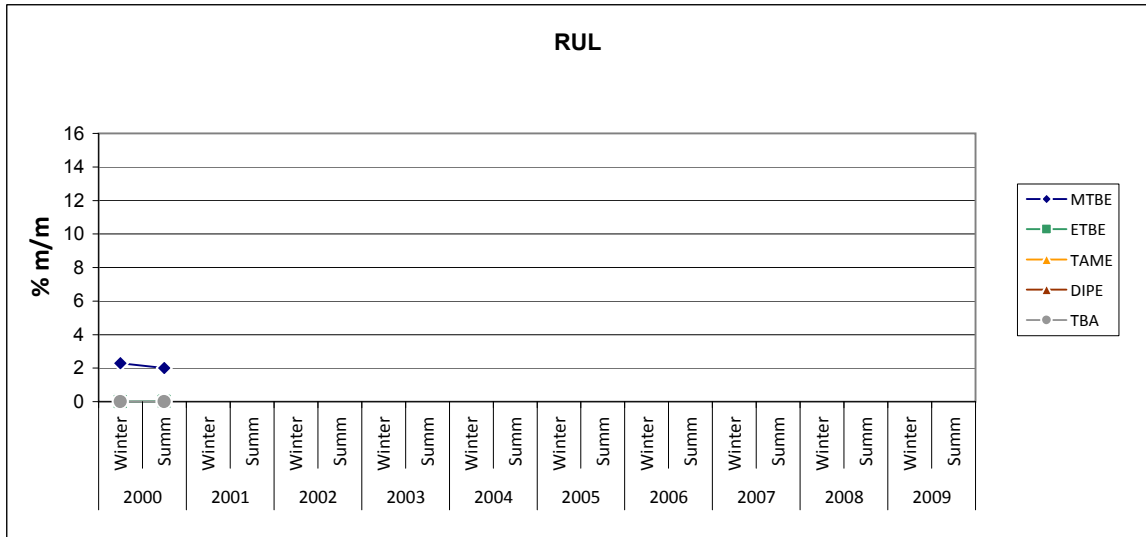
# Czech Republic



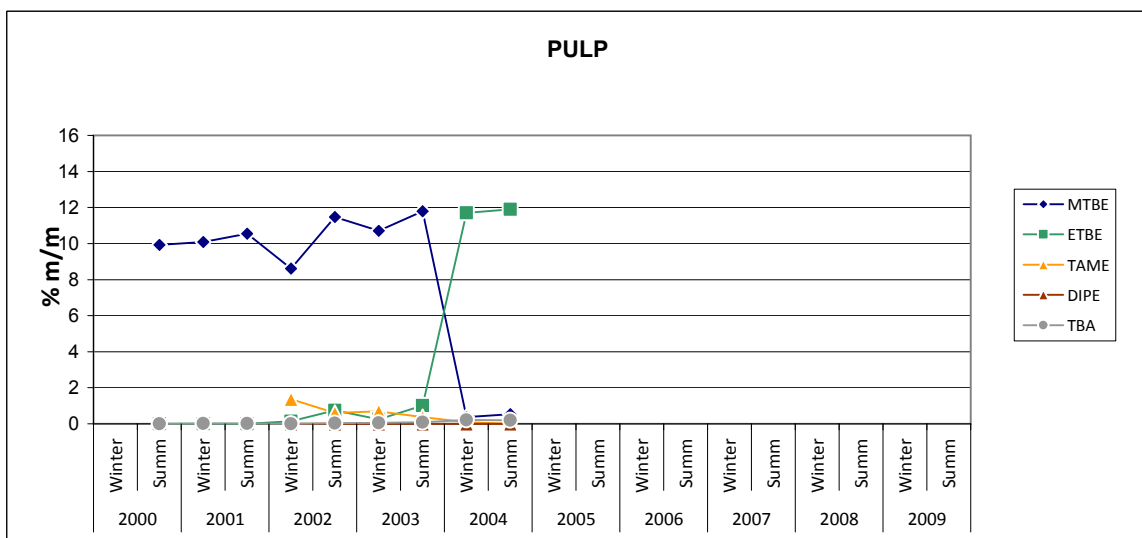
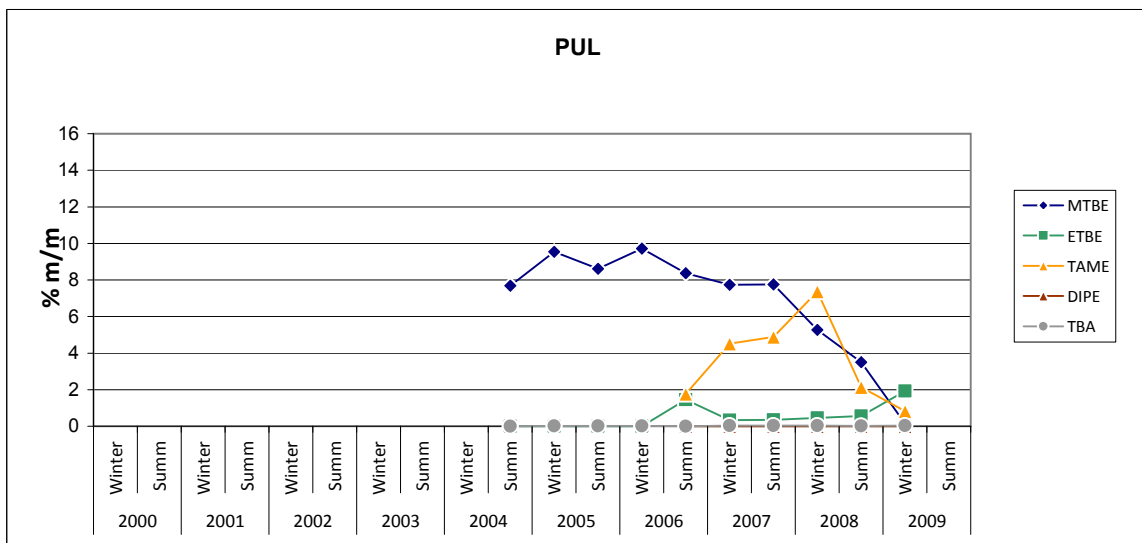
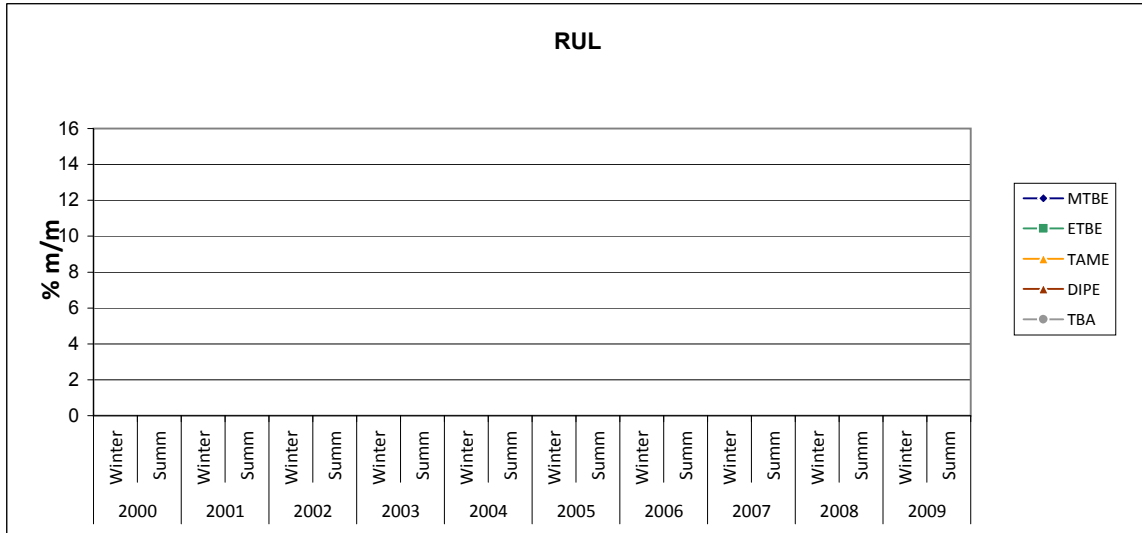
# Denmark



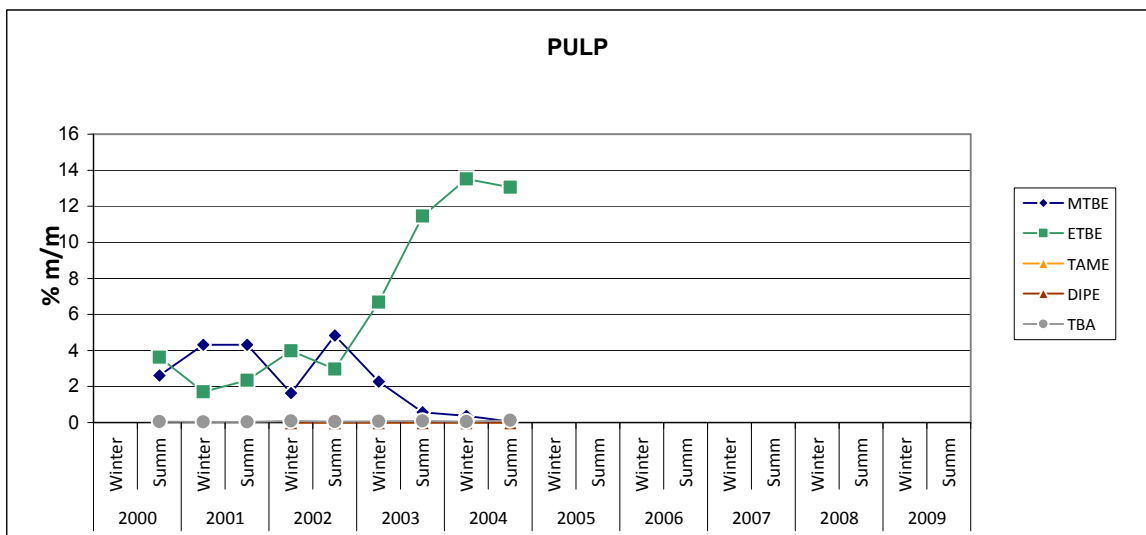
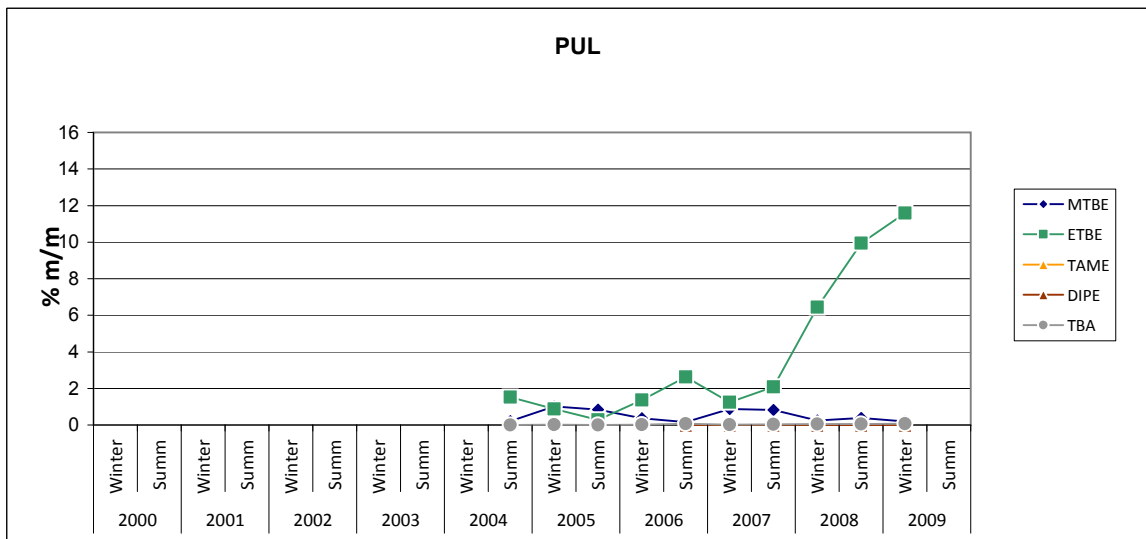
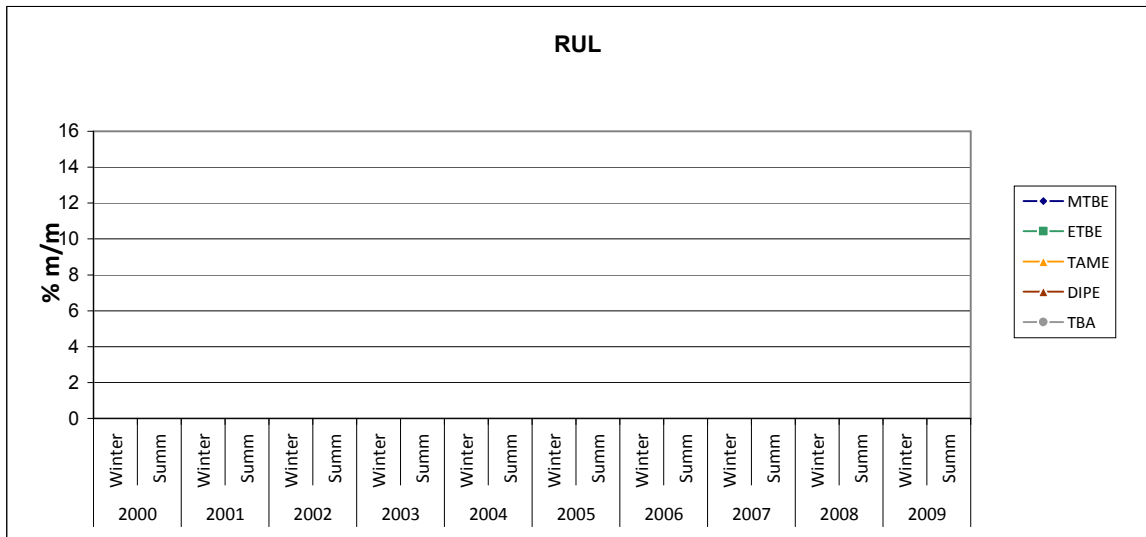
# Estonia



# Finland

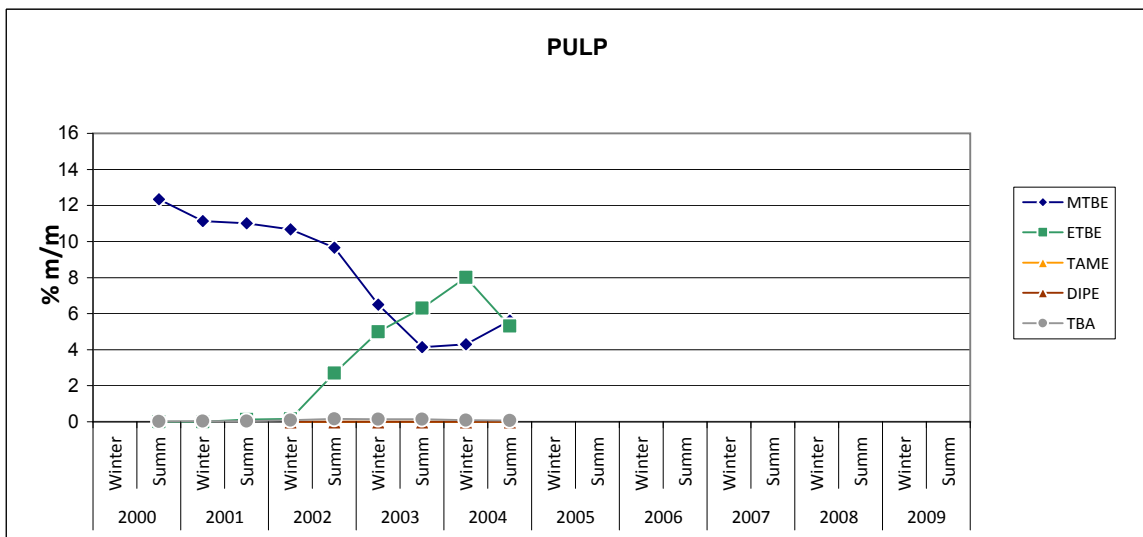
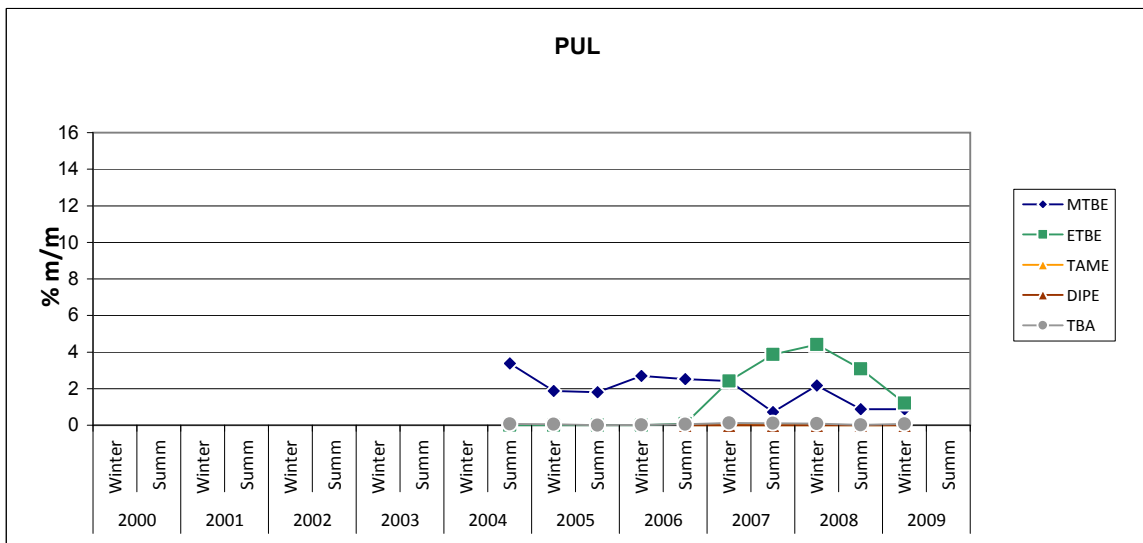
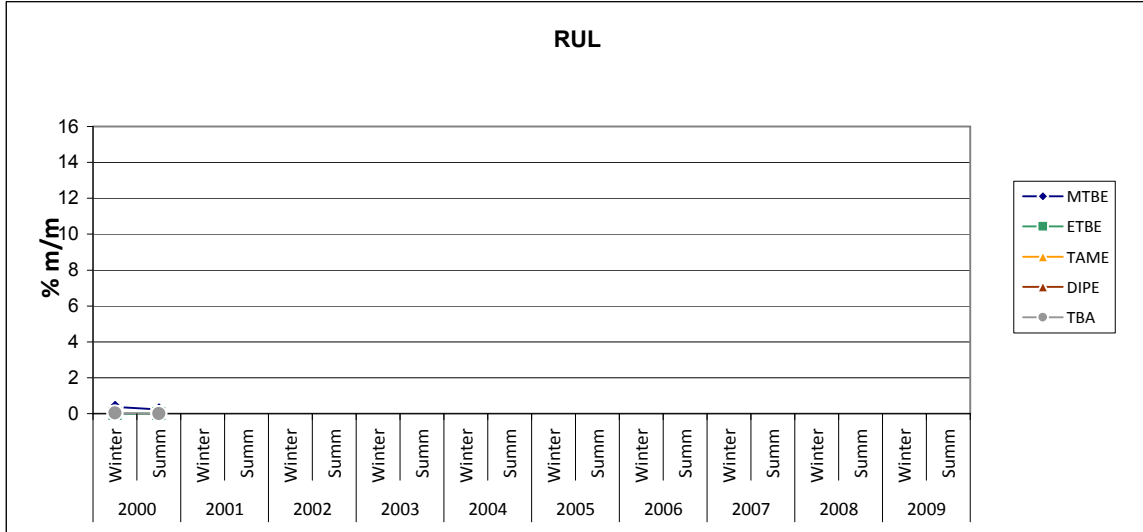


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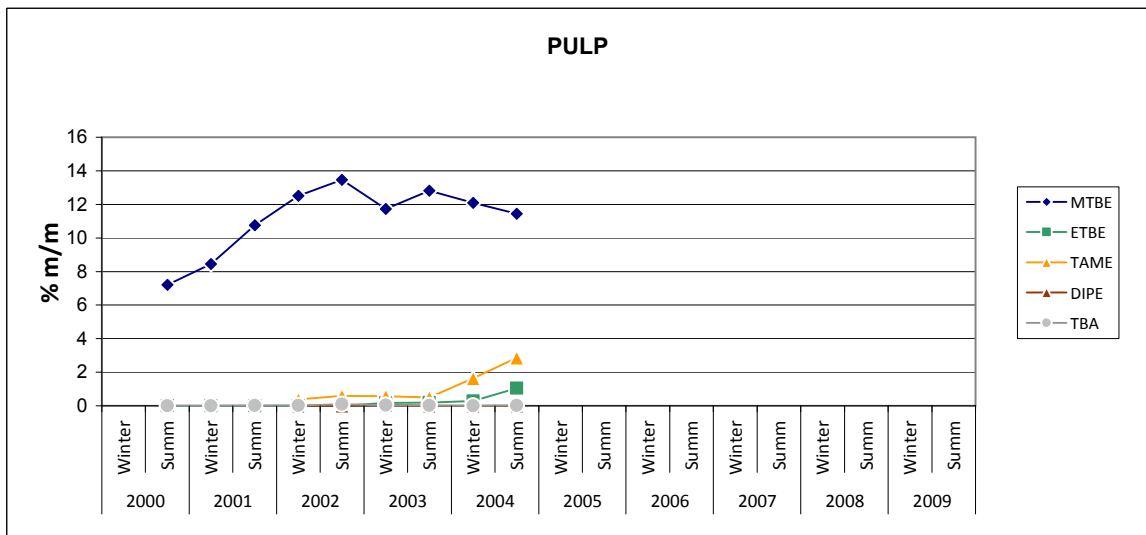
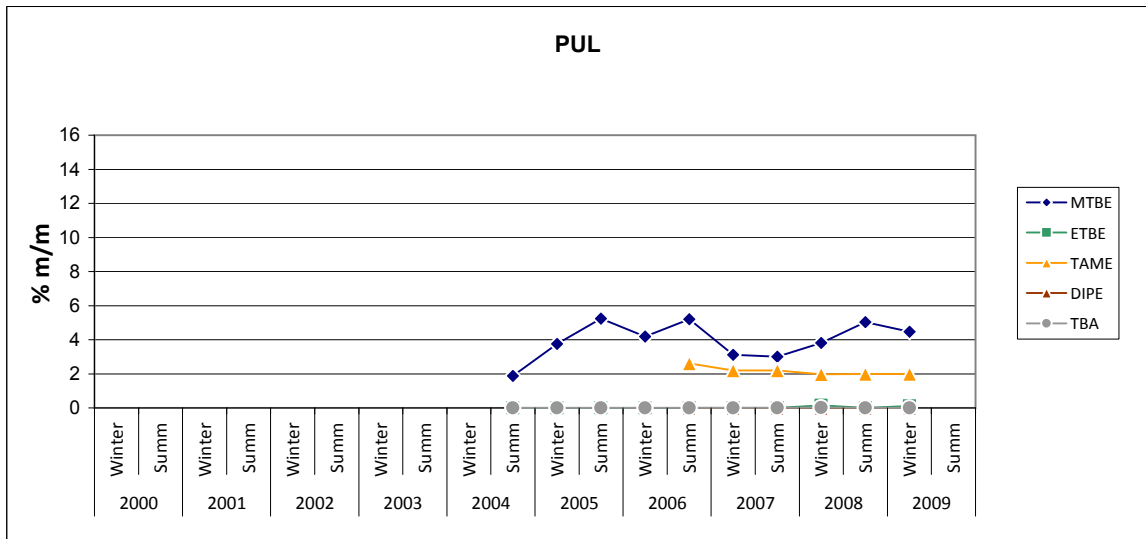
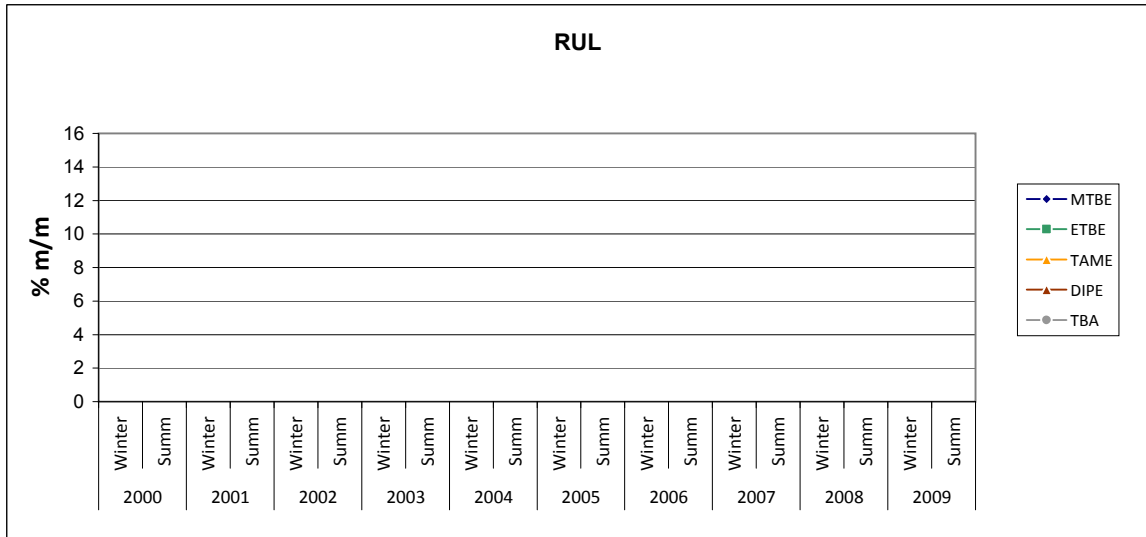




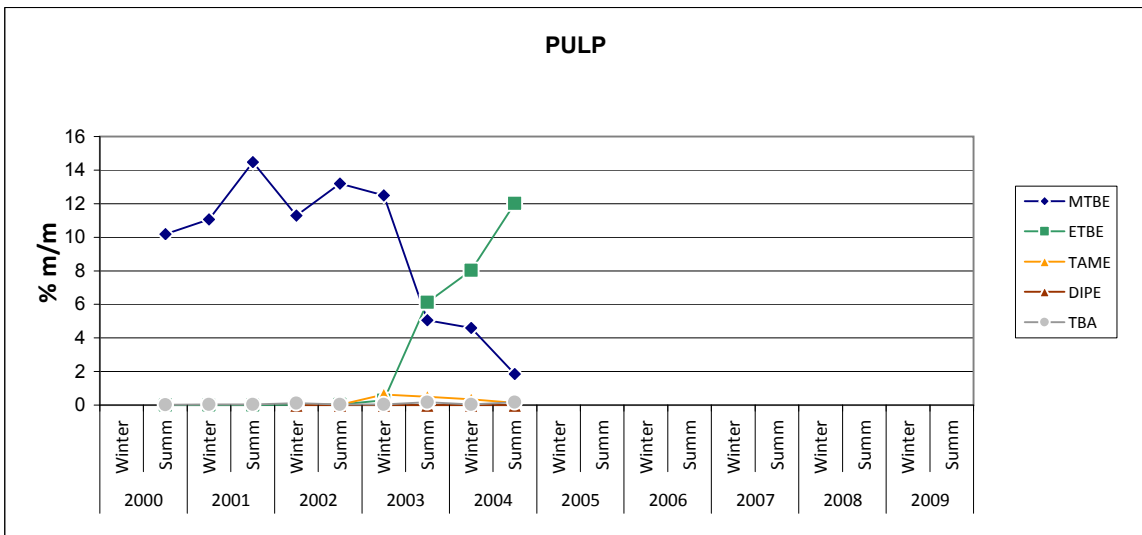
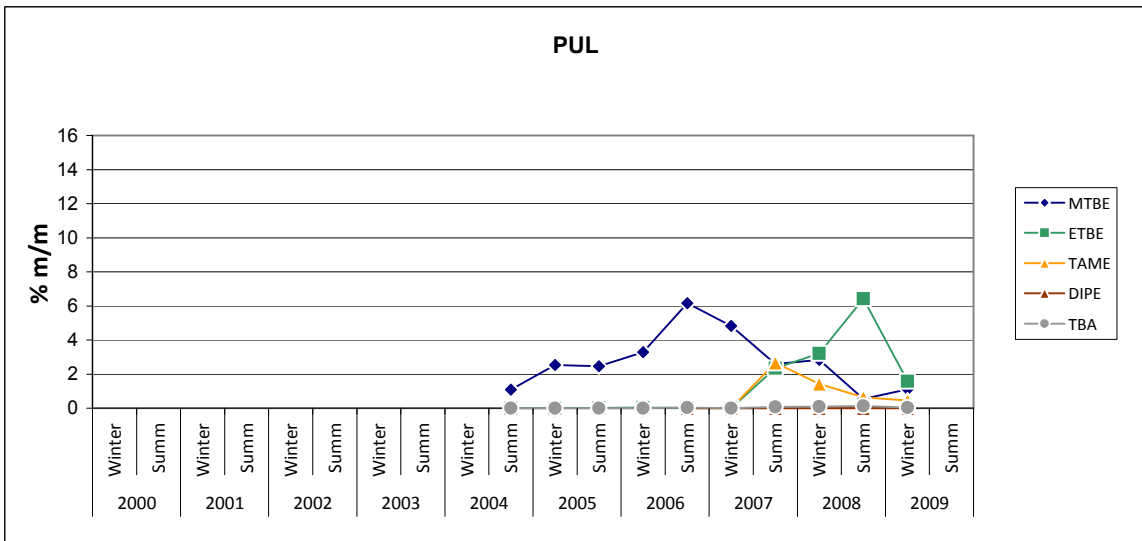
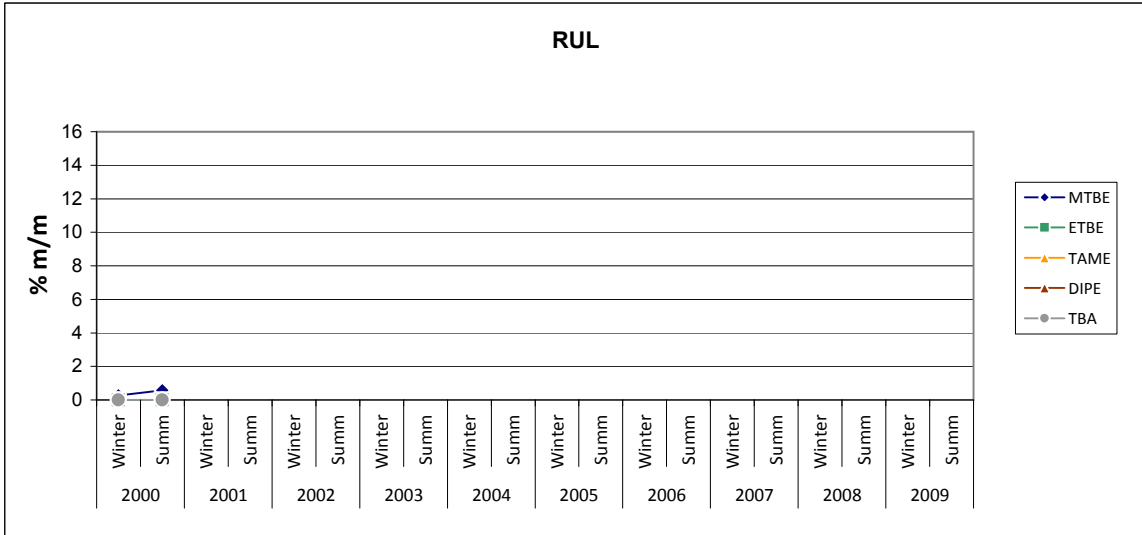
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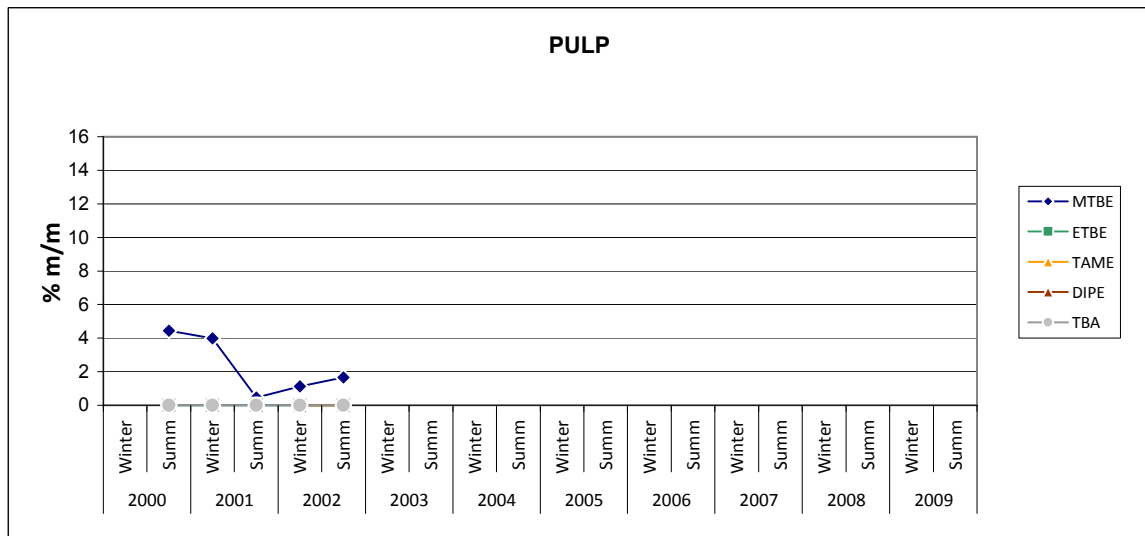
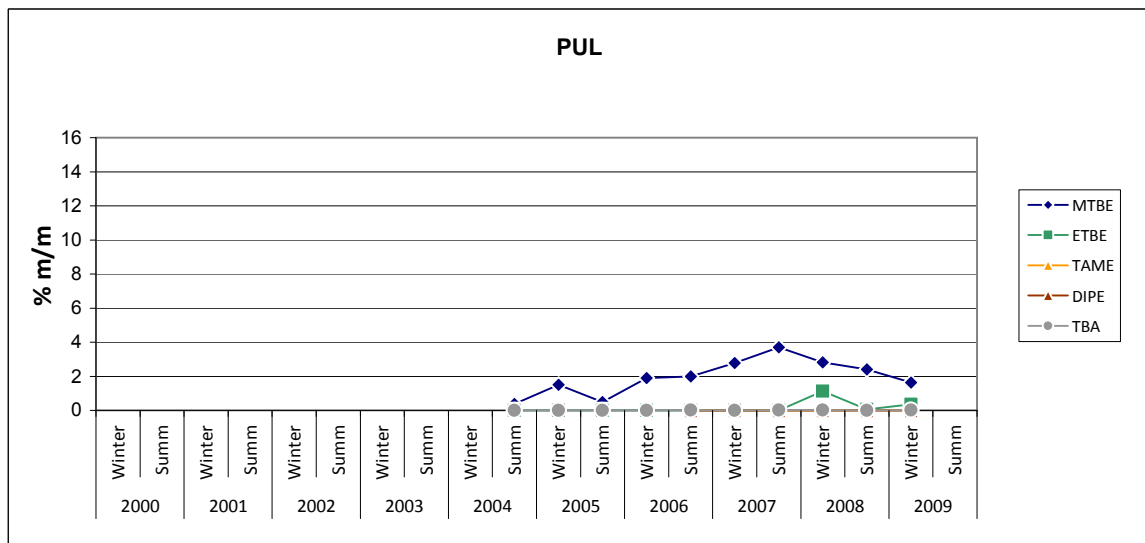
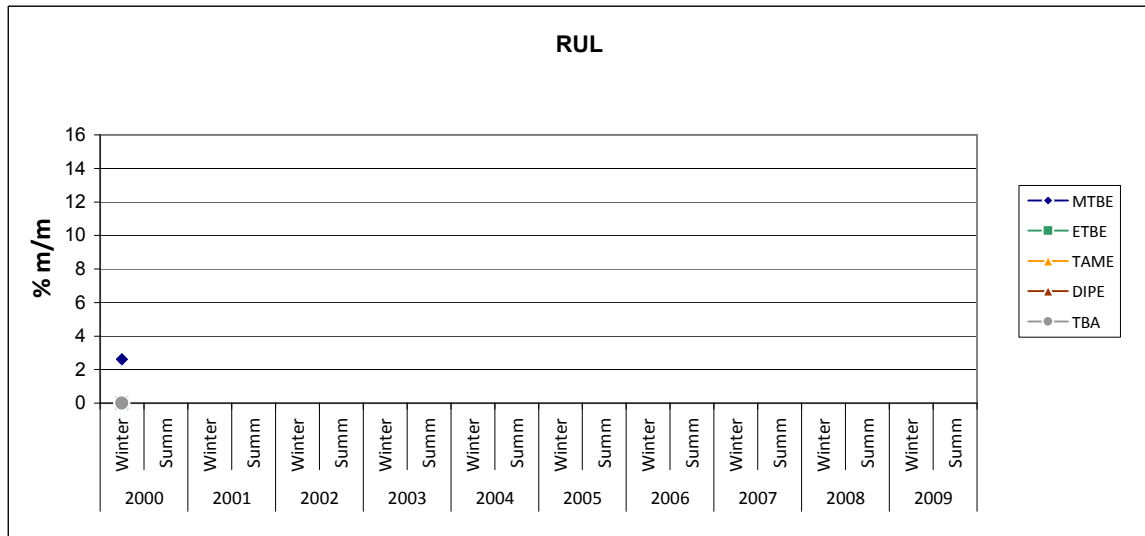
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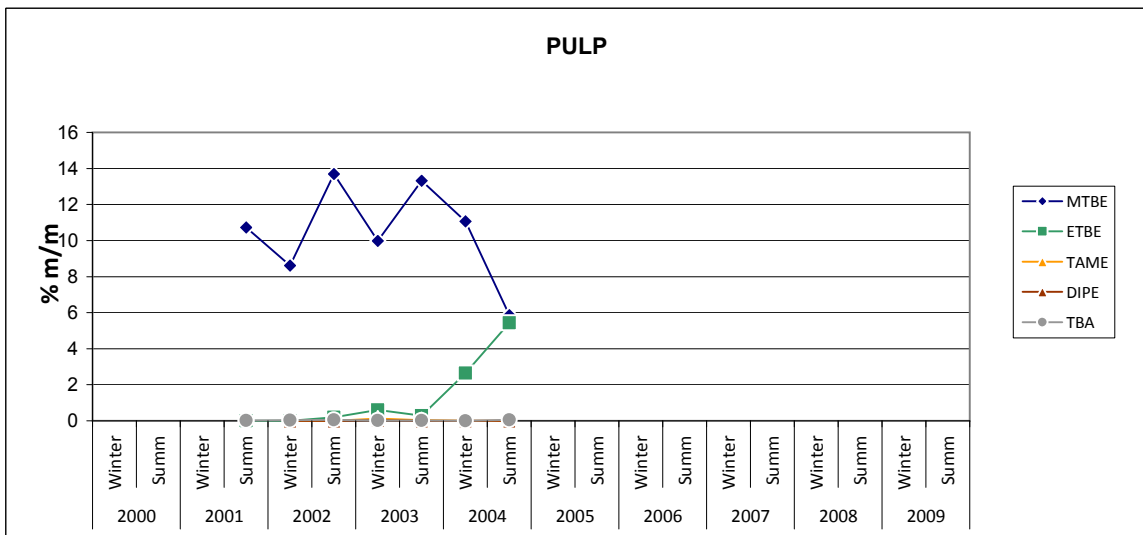
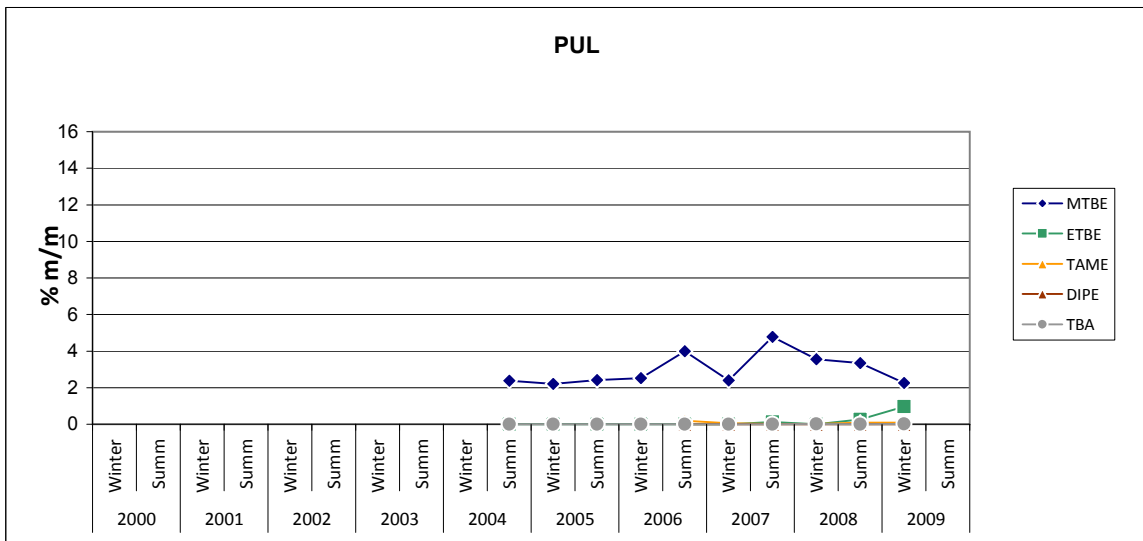
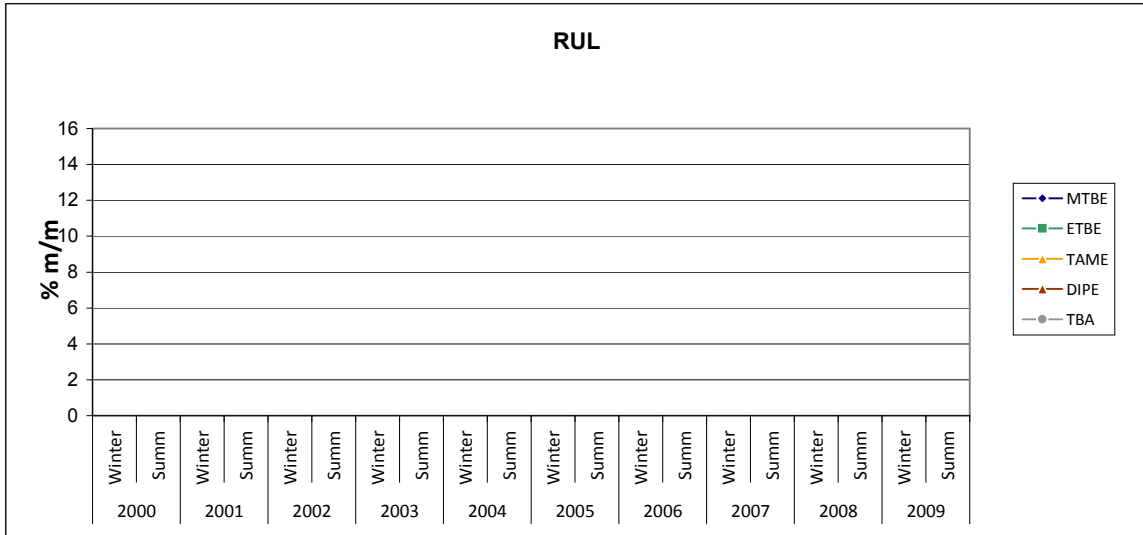
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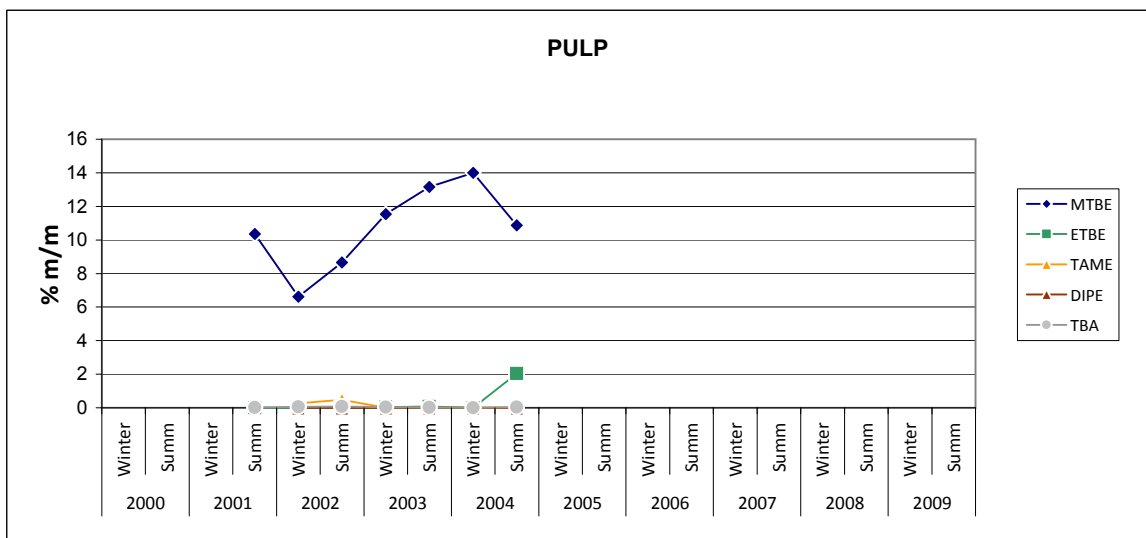
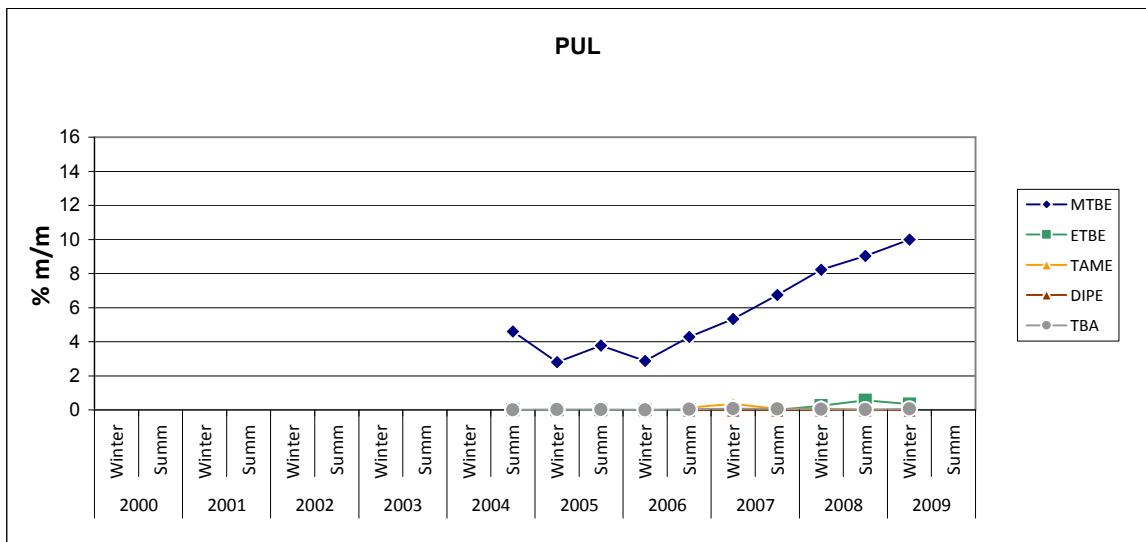
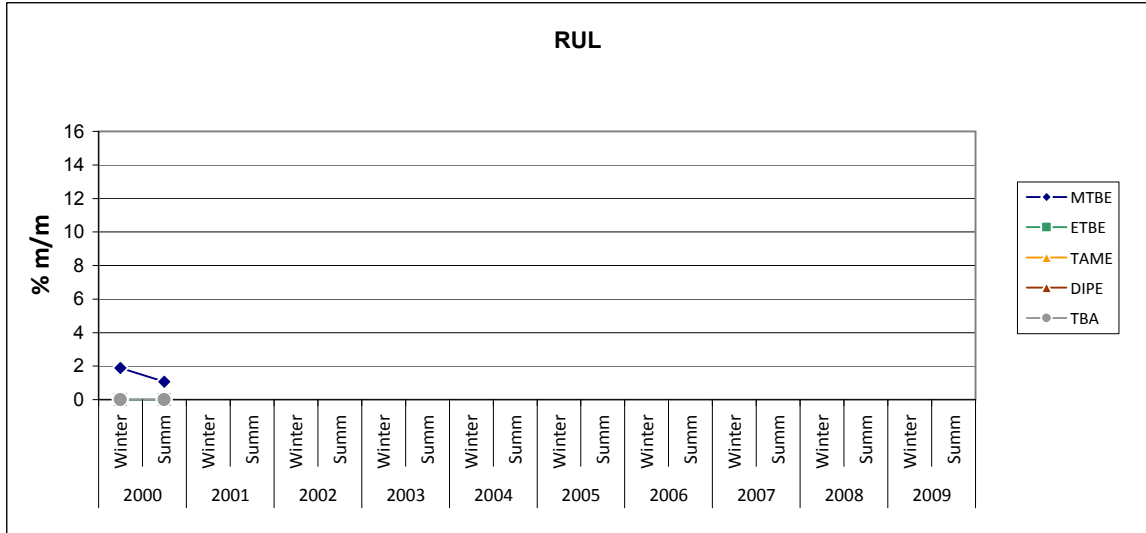
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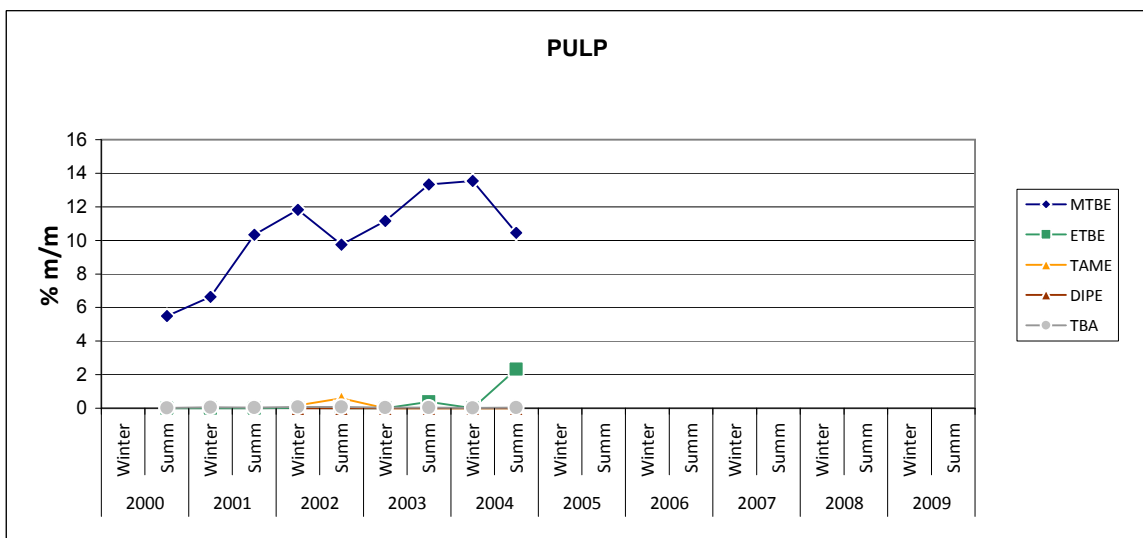
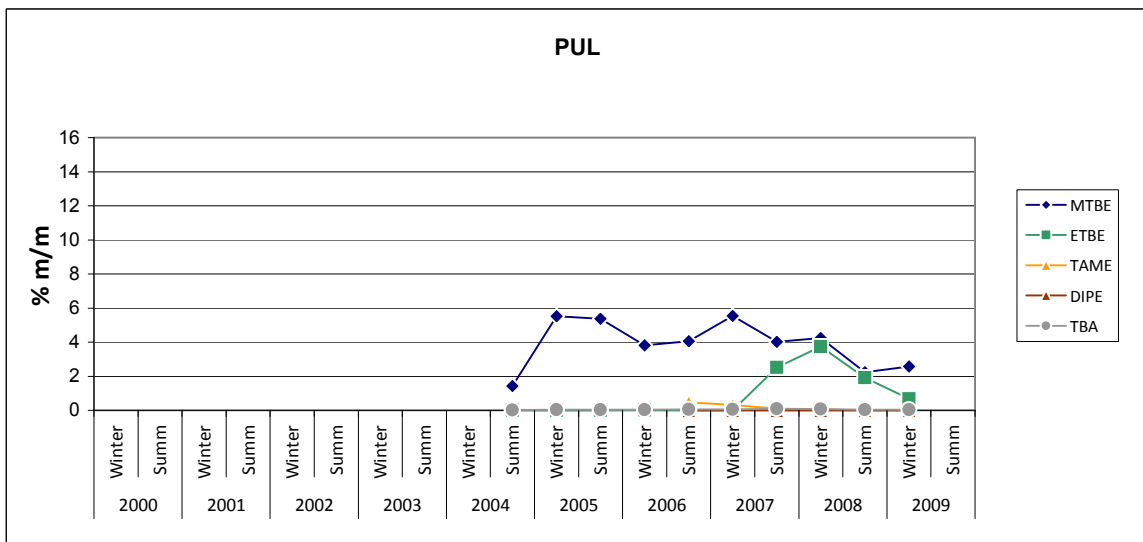
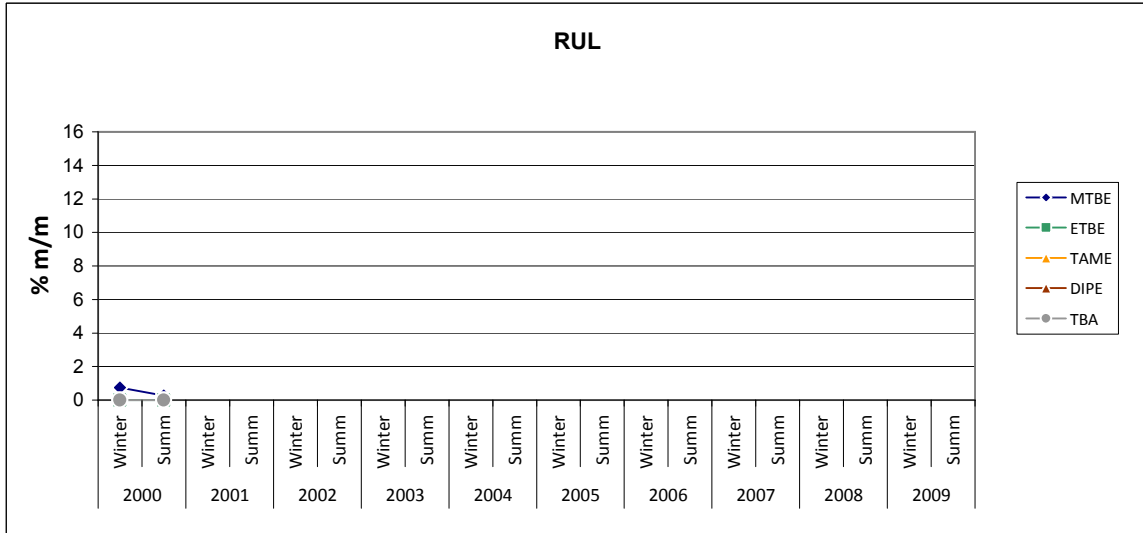
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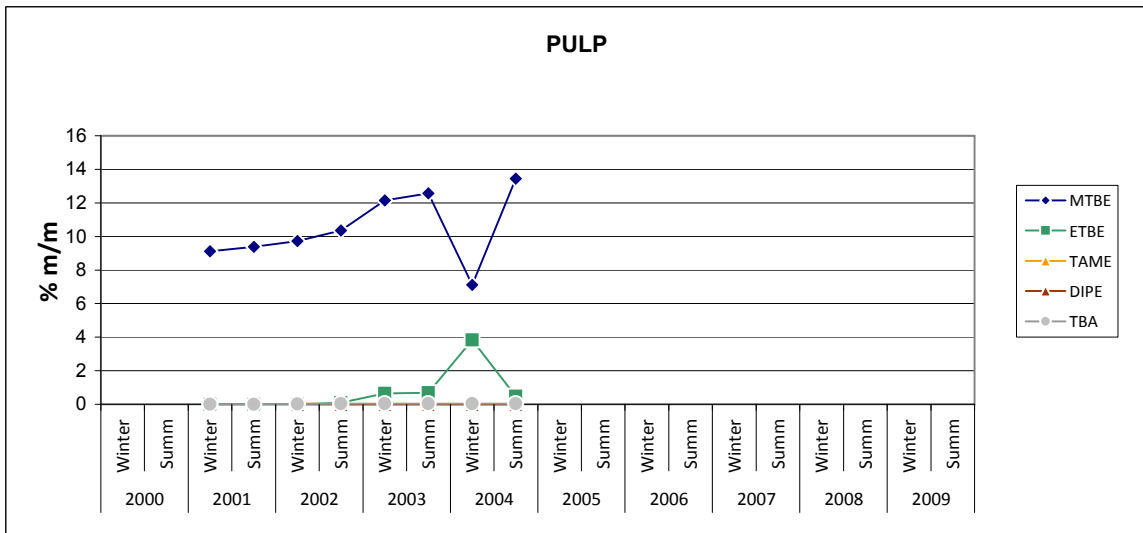
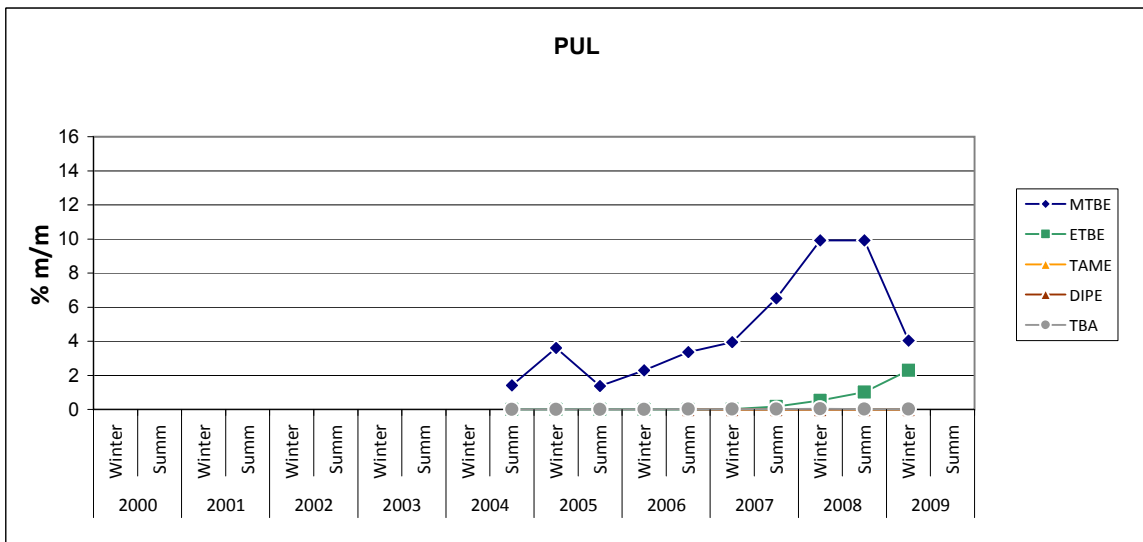
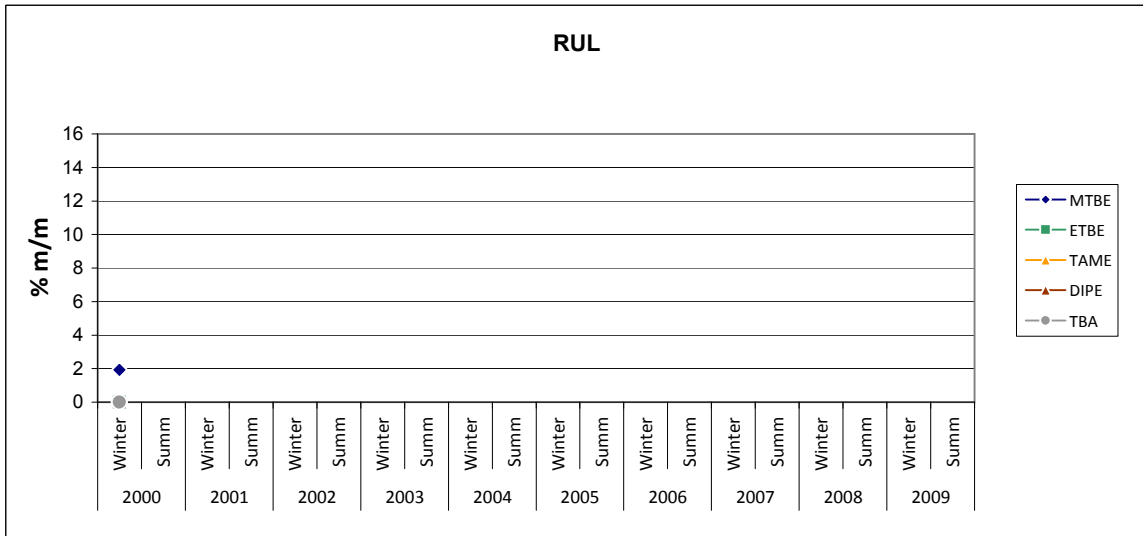
# Latvia



# Lithuania

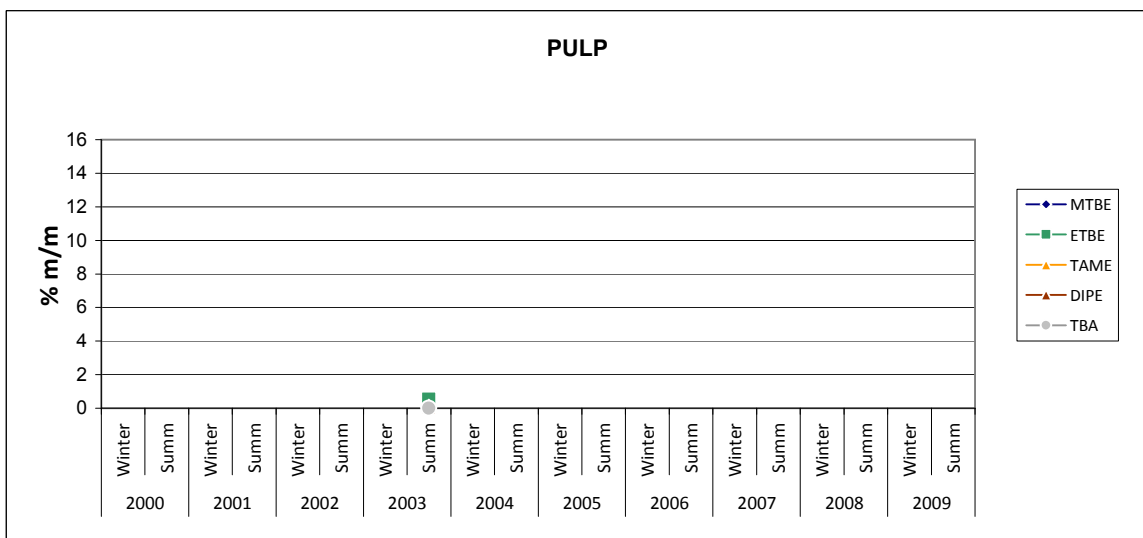
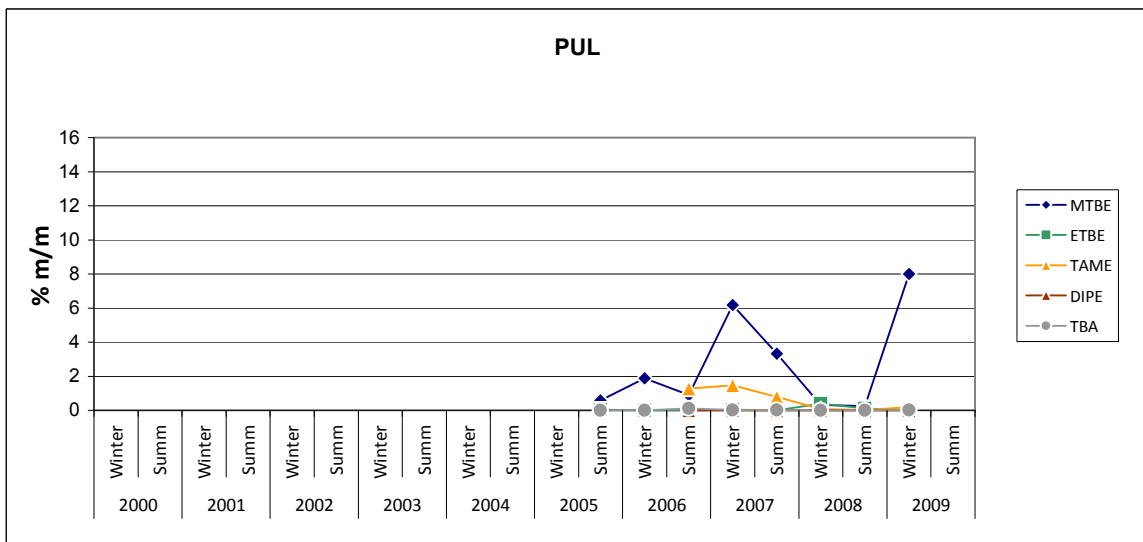
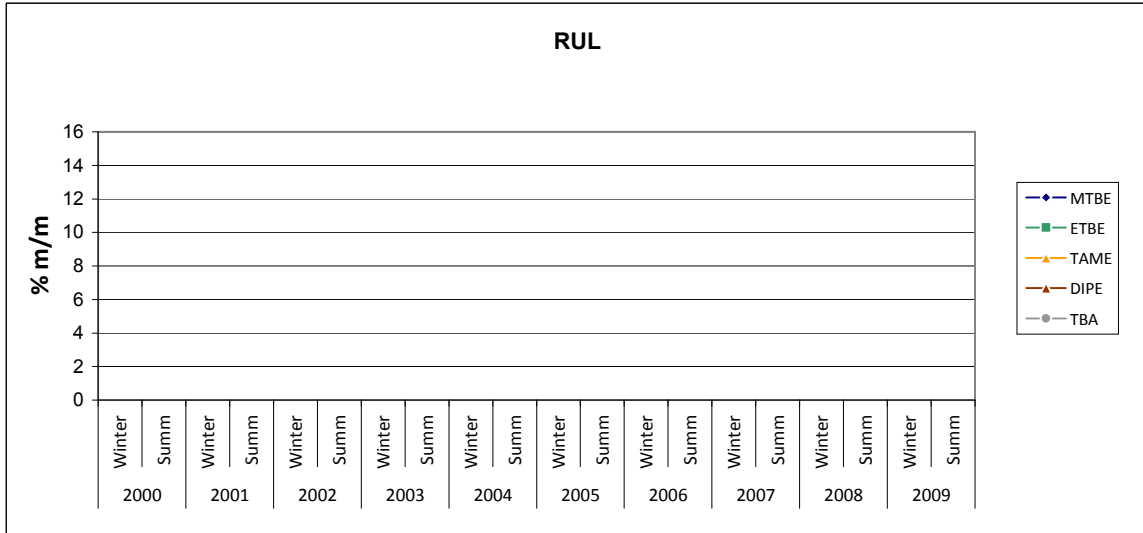


# Luxembourg

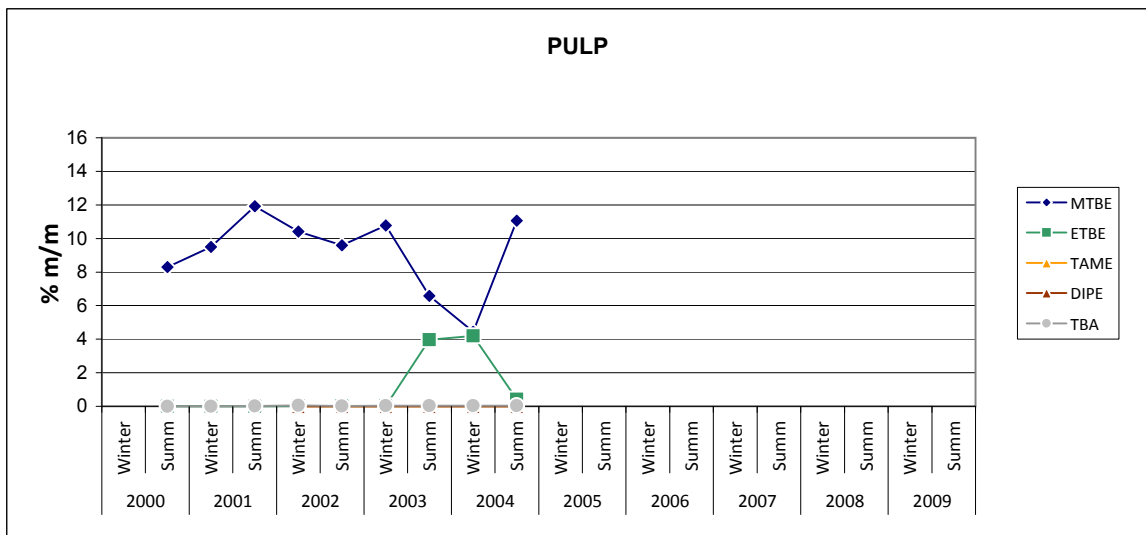
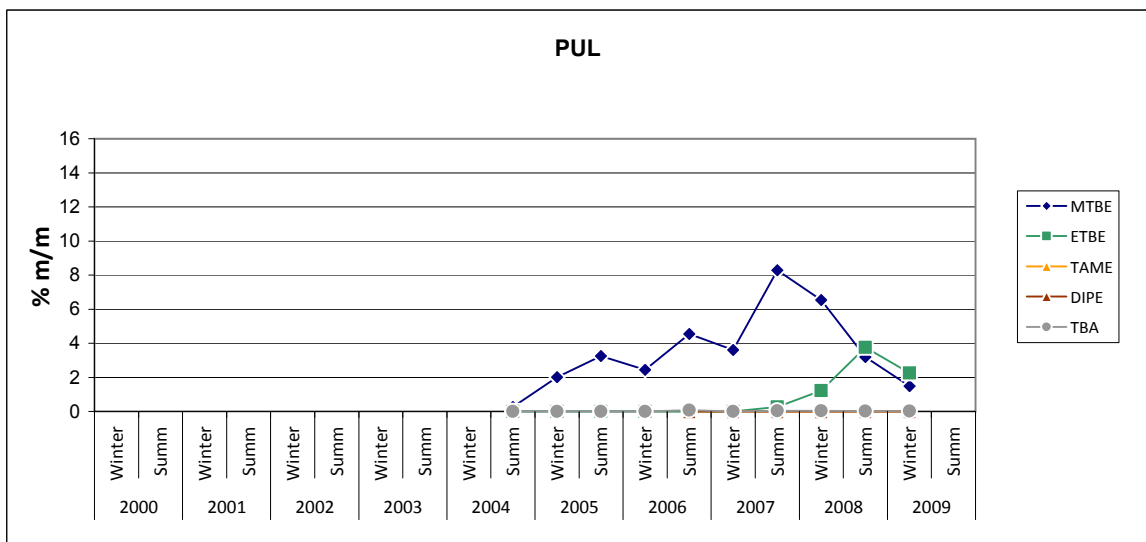
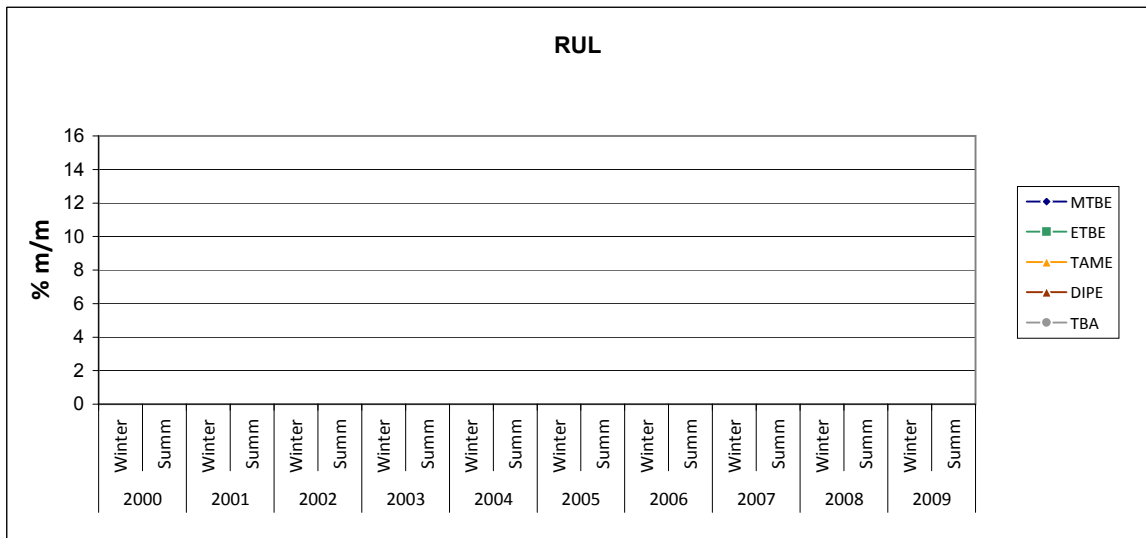




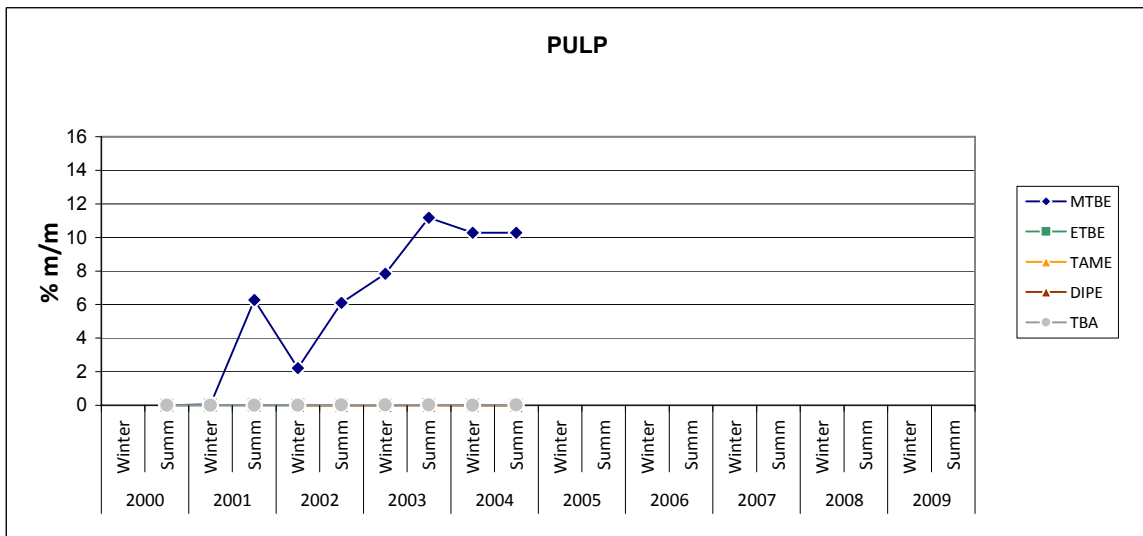
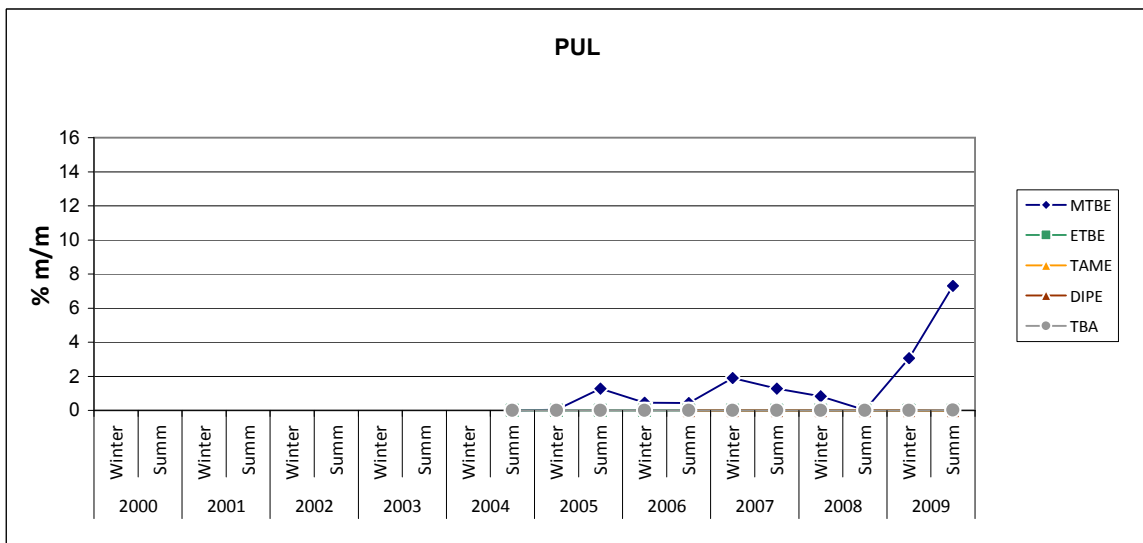
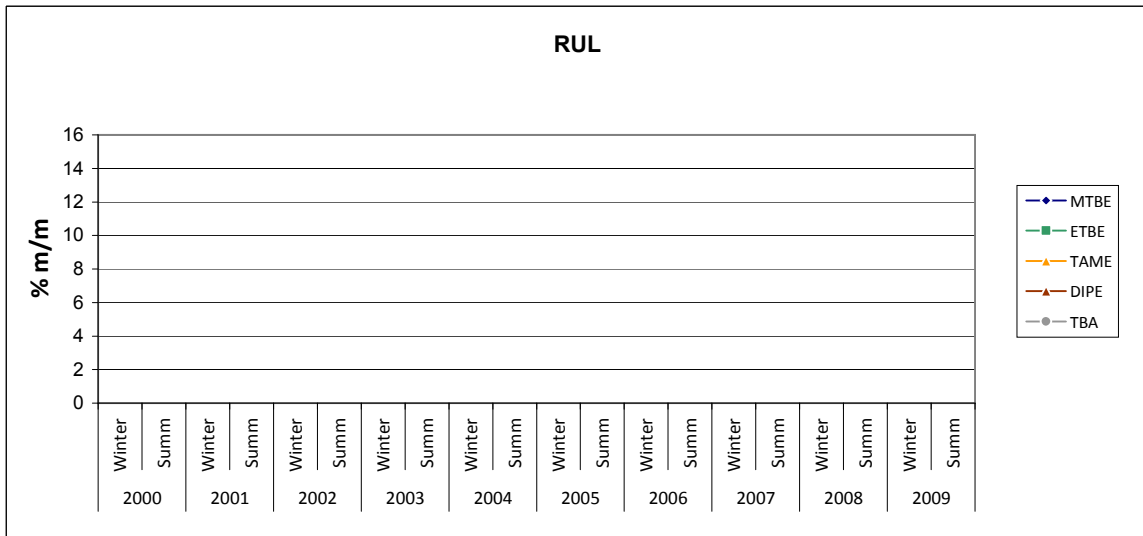
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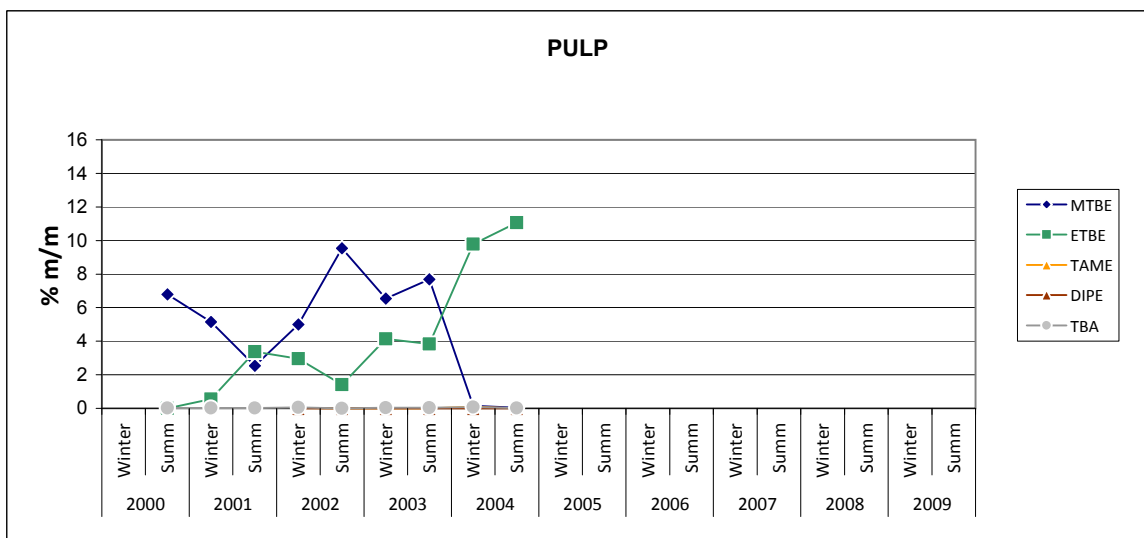
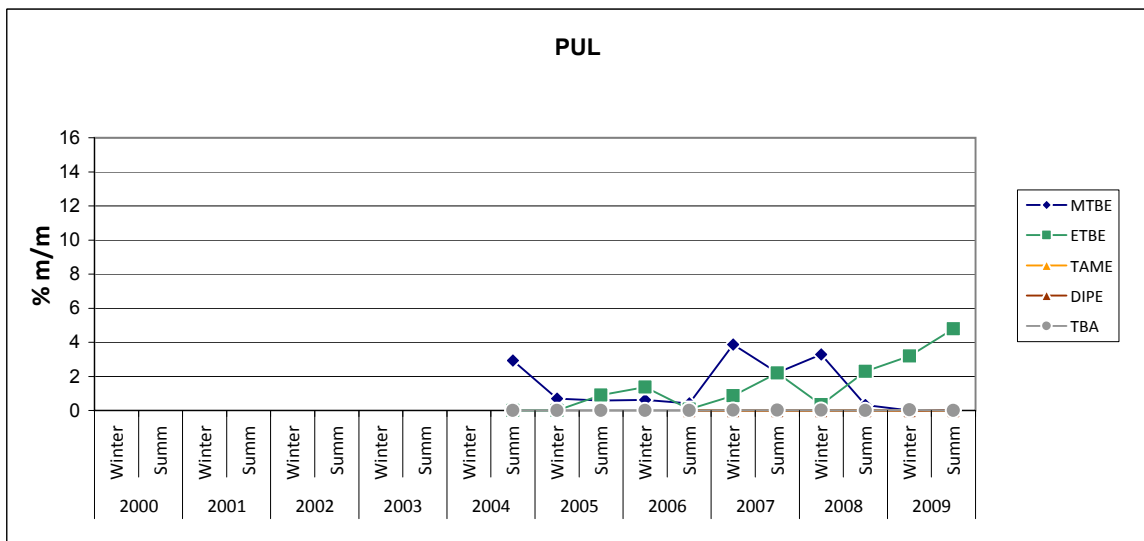
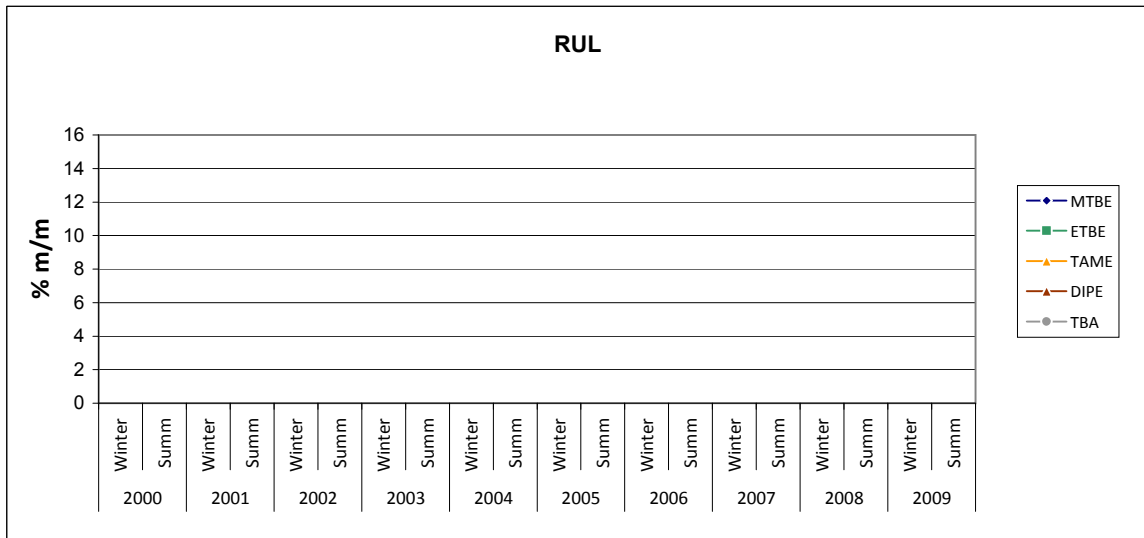
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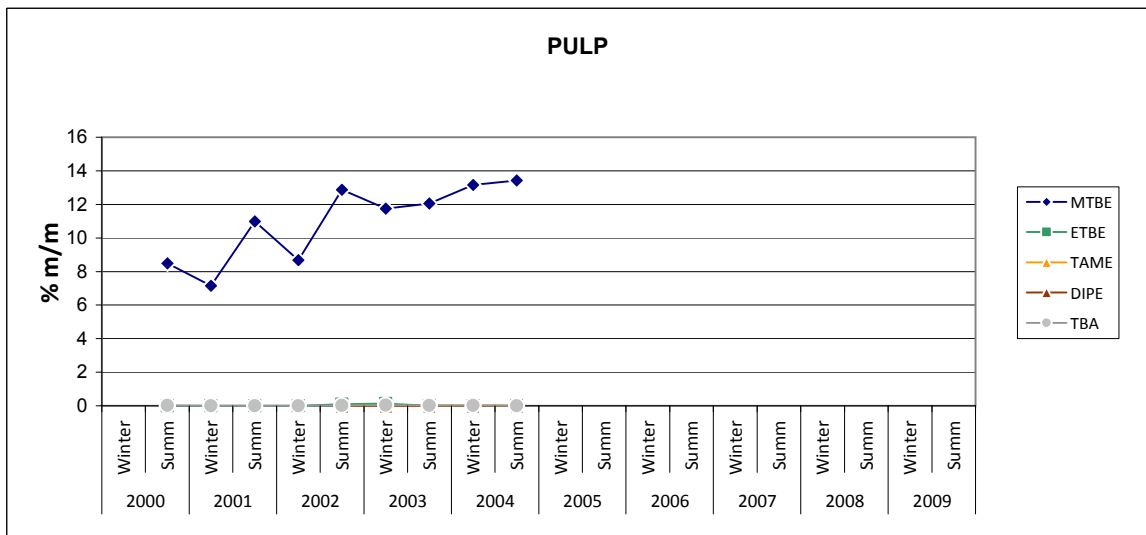
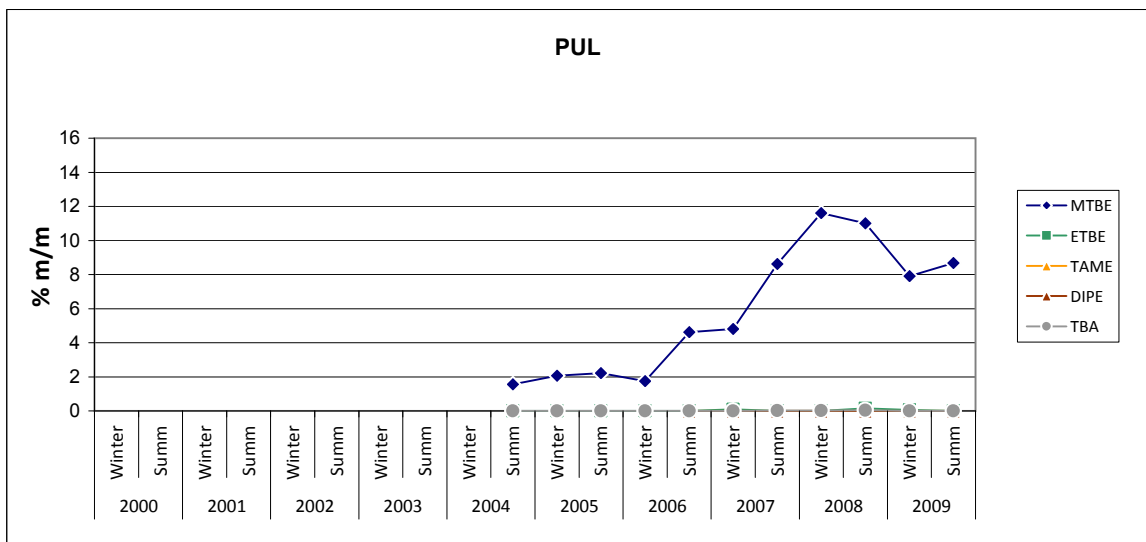
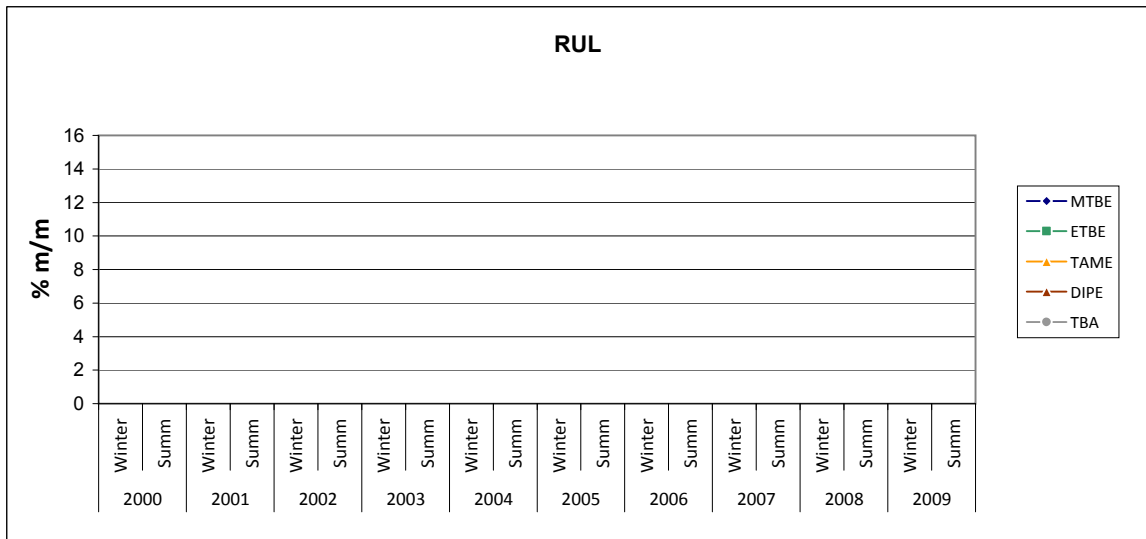
# Norway



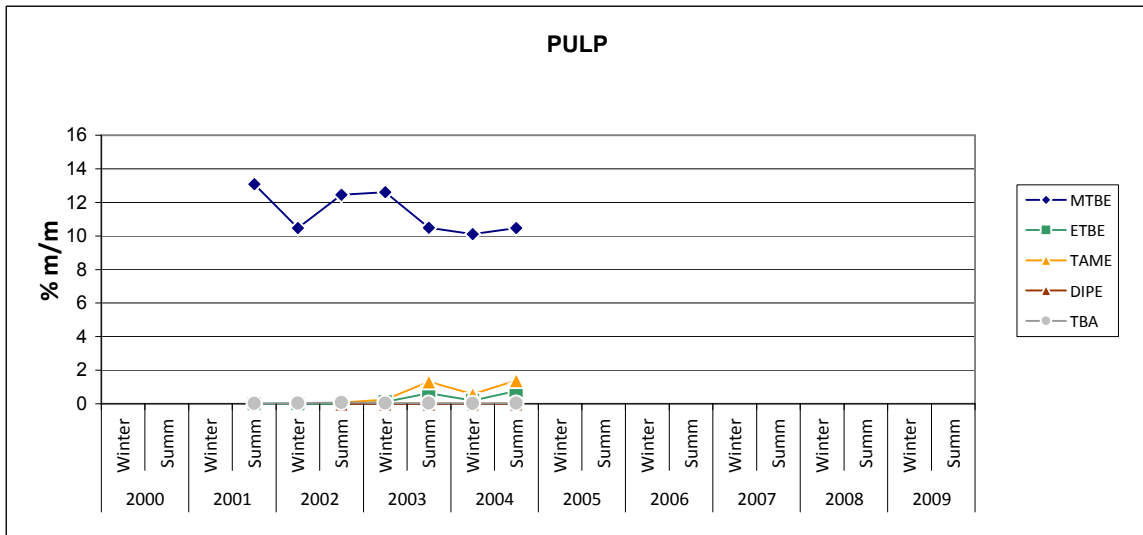
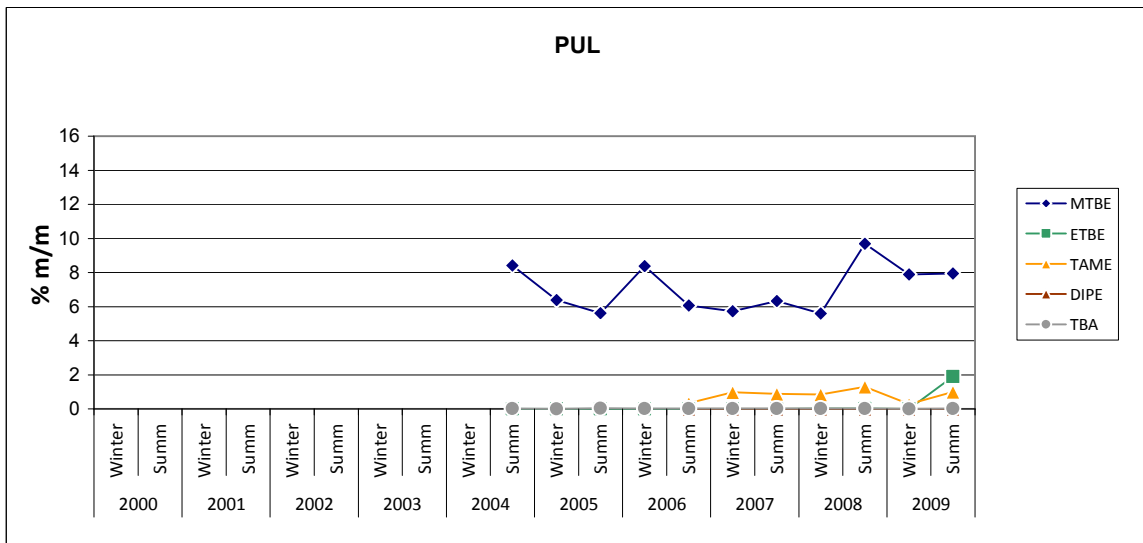
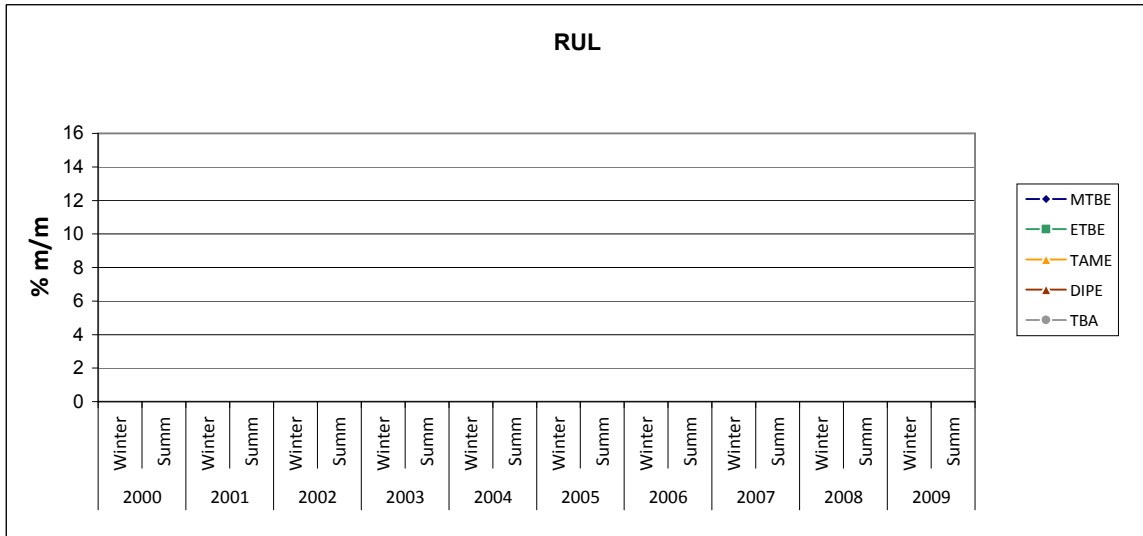
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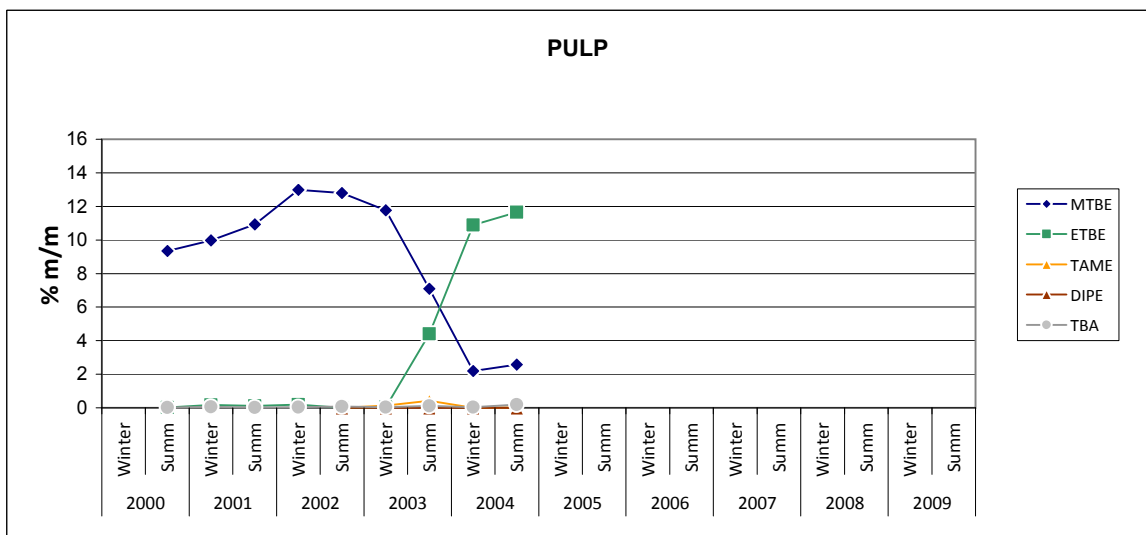
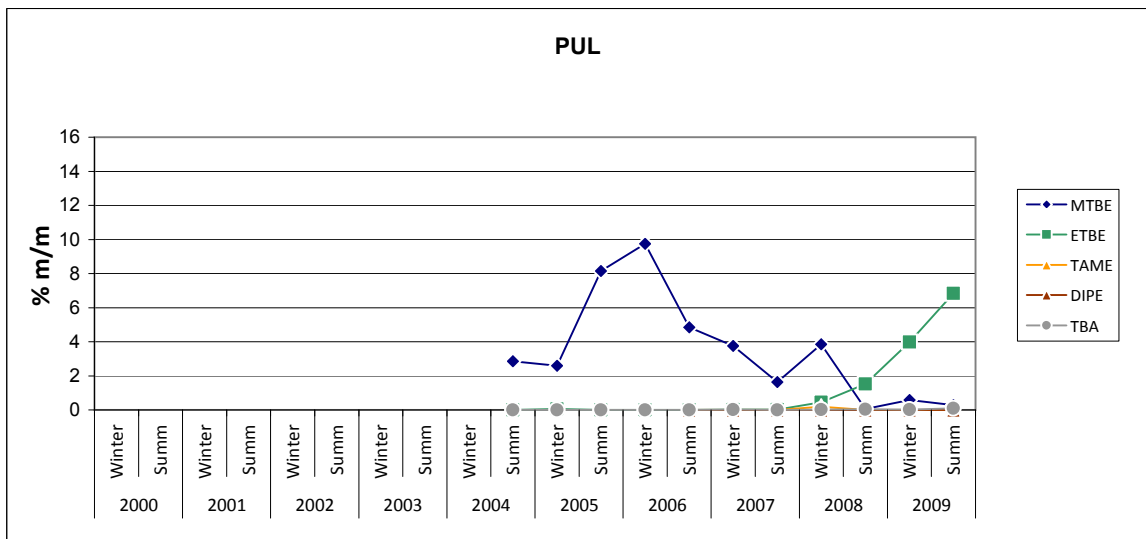
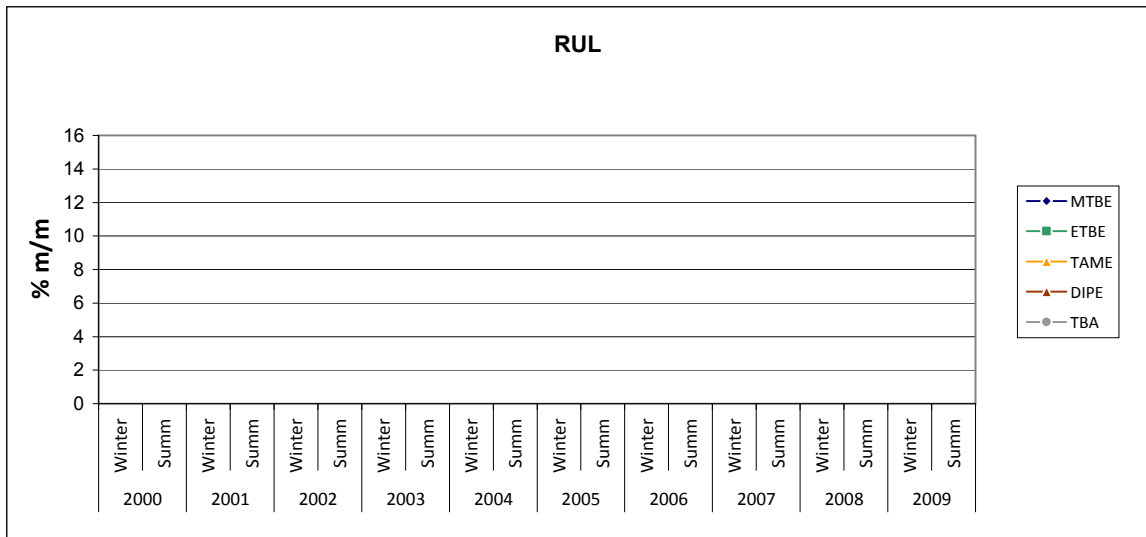
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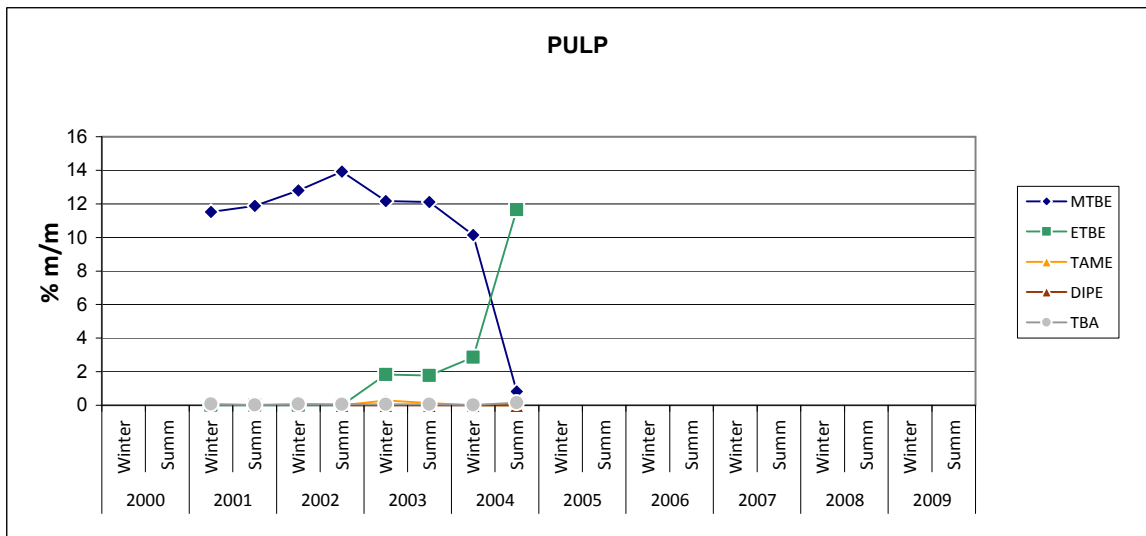
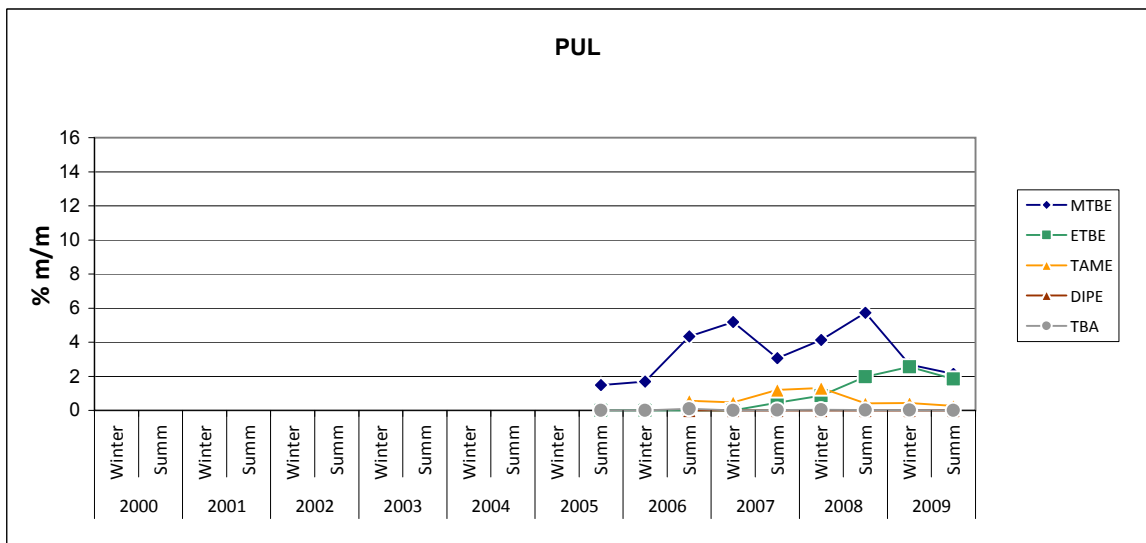
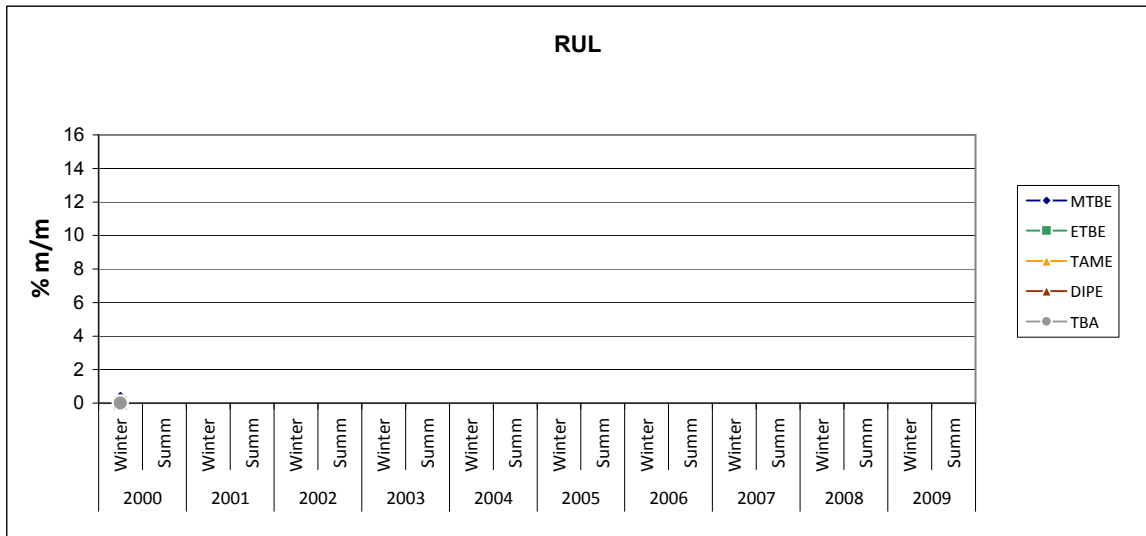
# Romania



## Slovakia

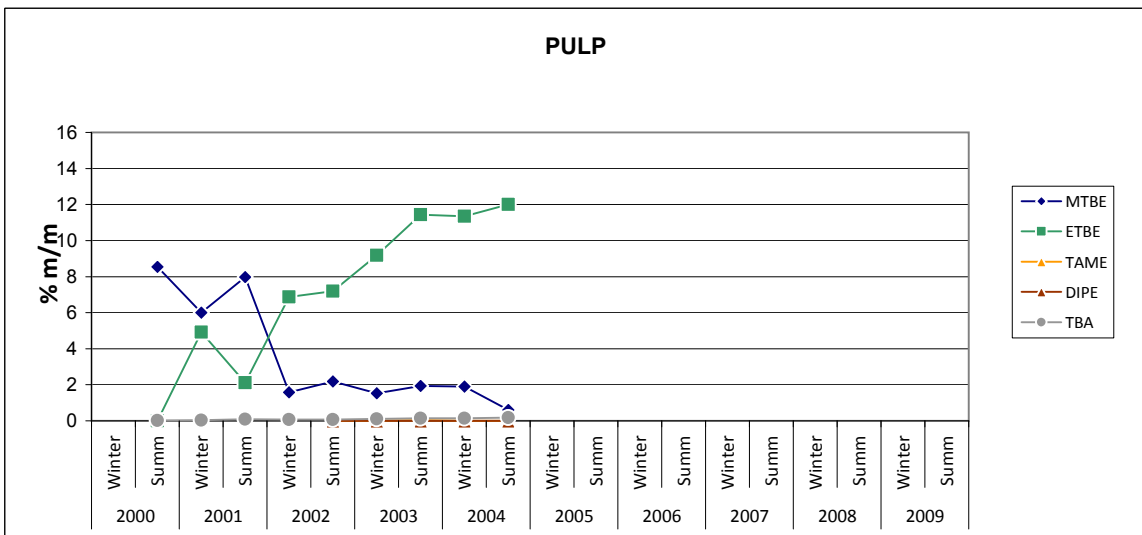
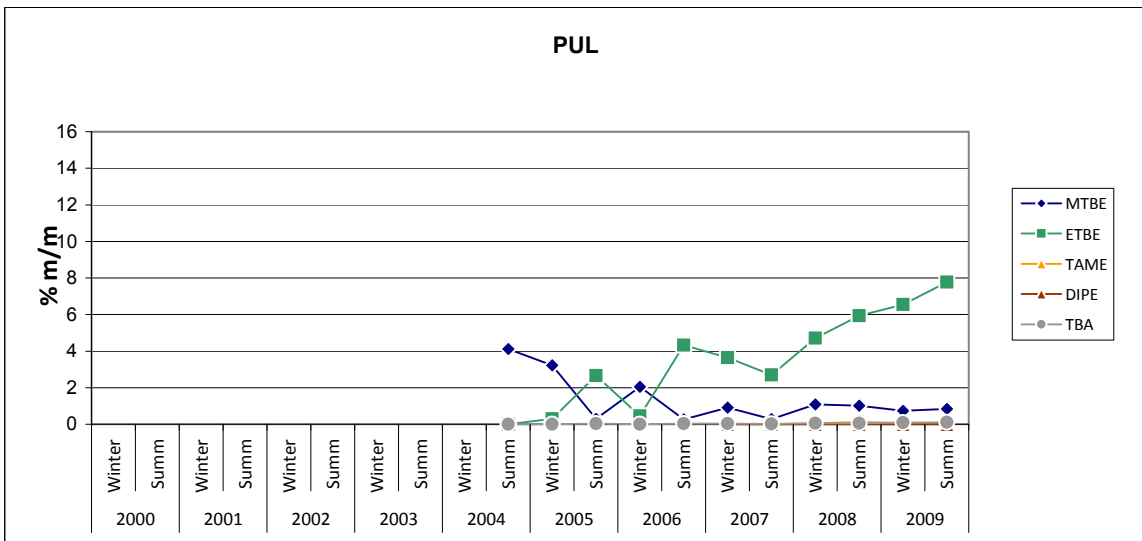
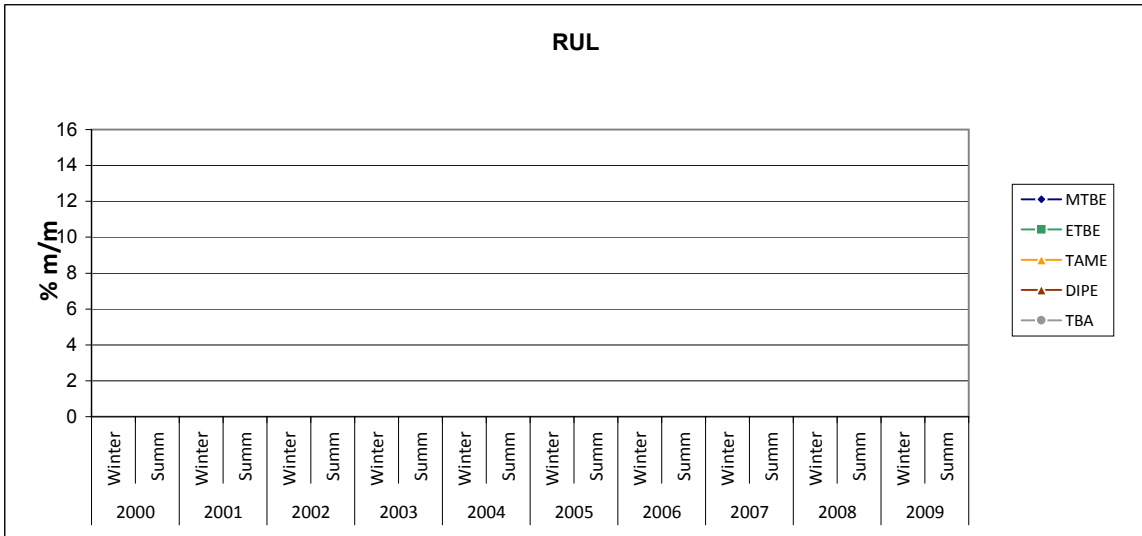


## Slovenia

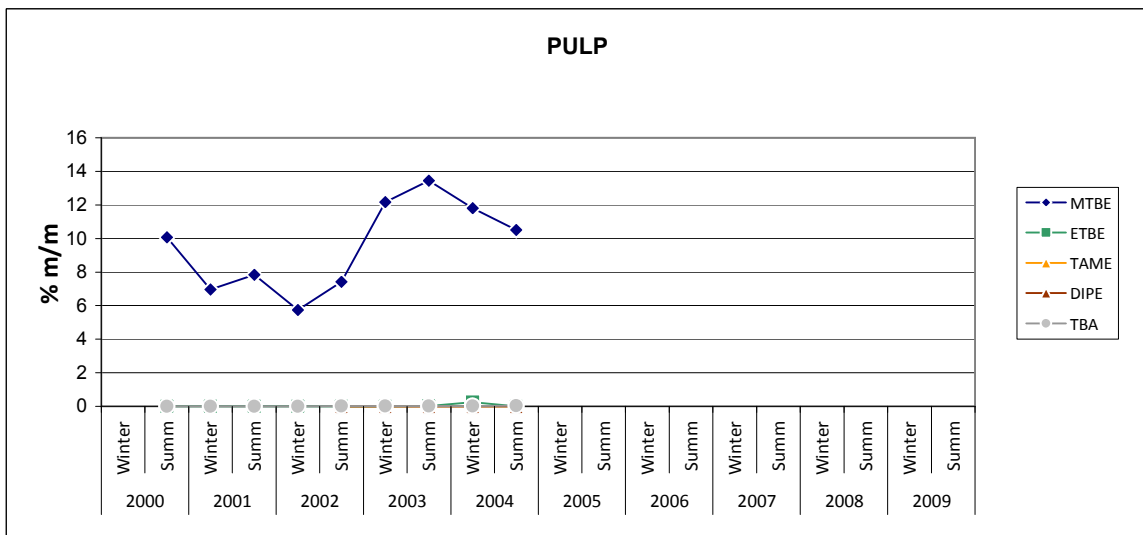
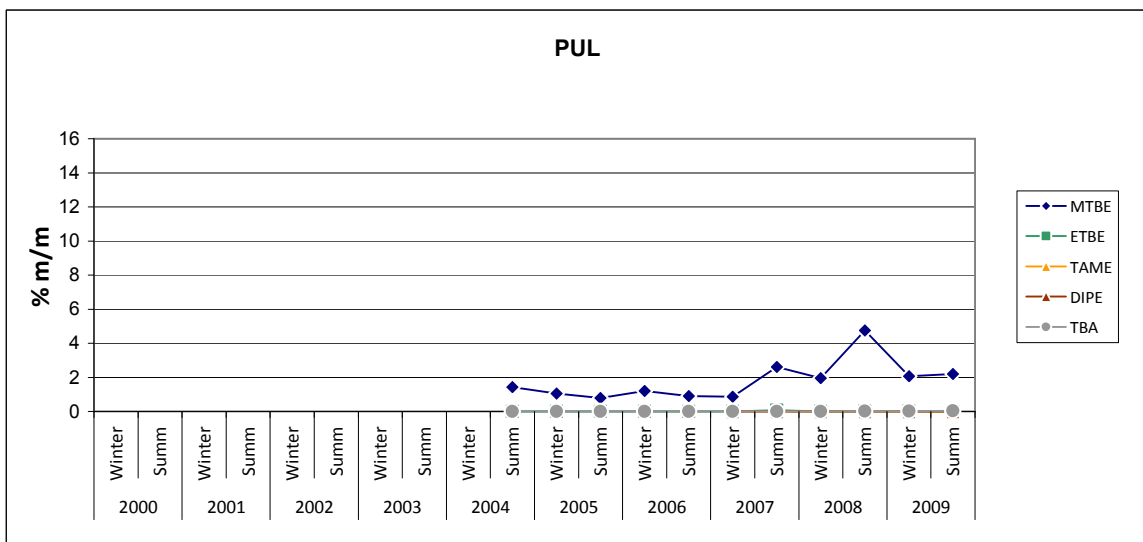
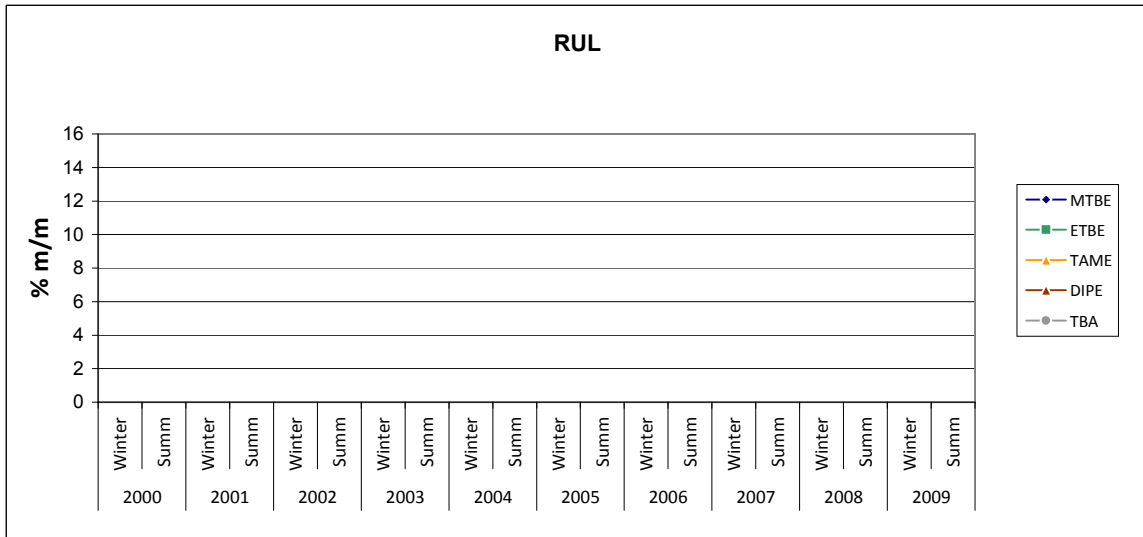




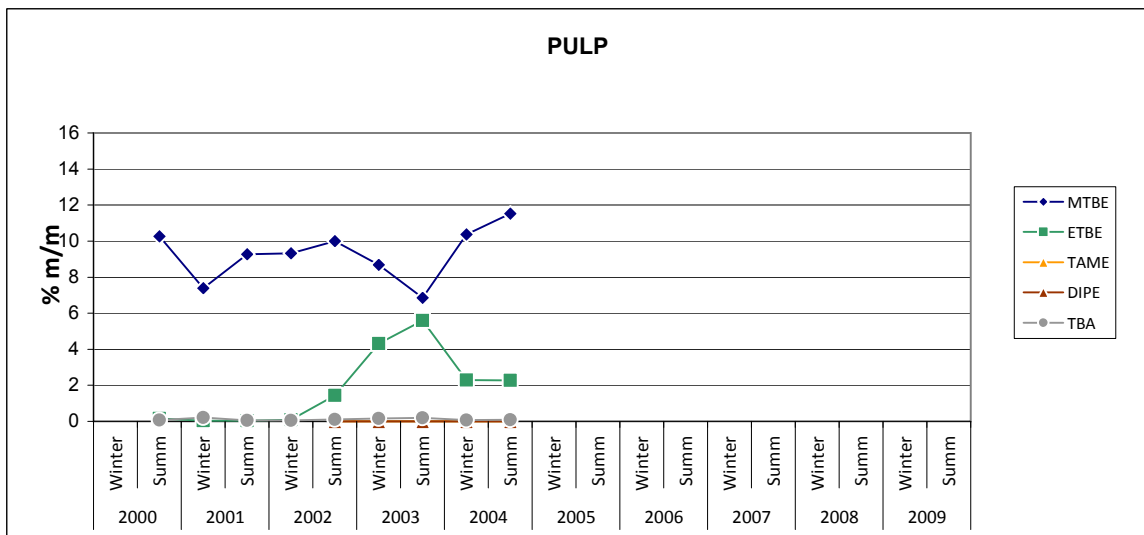
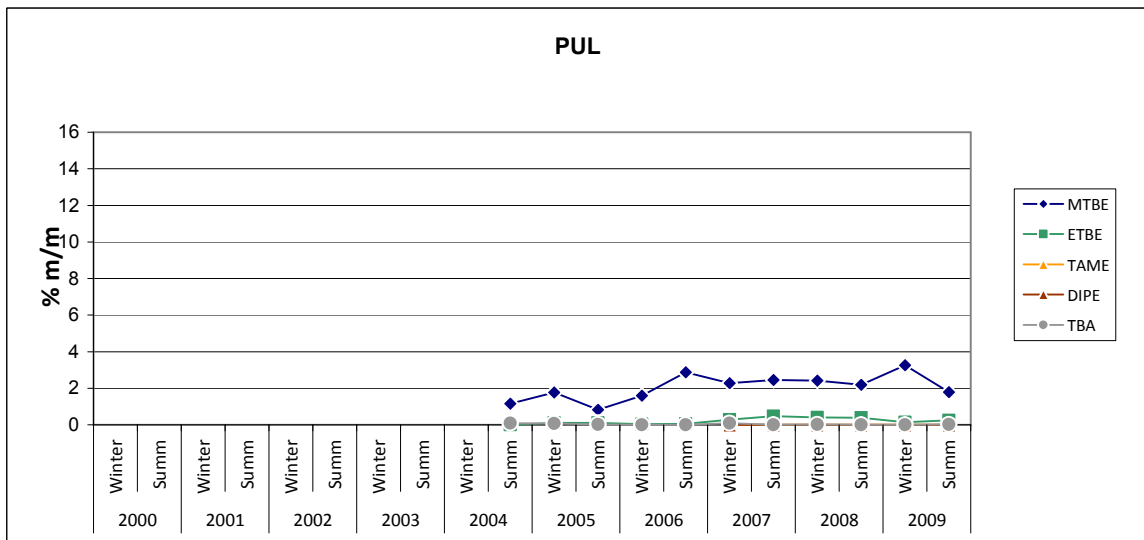
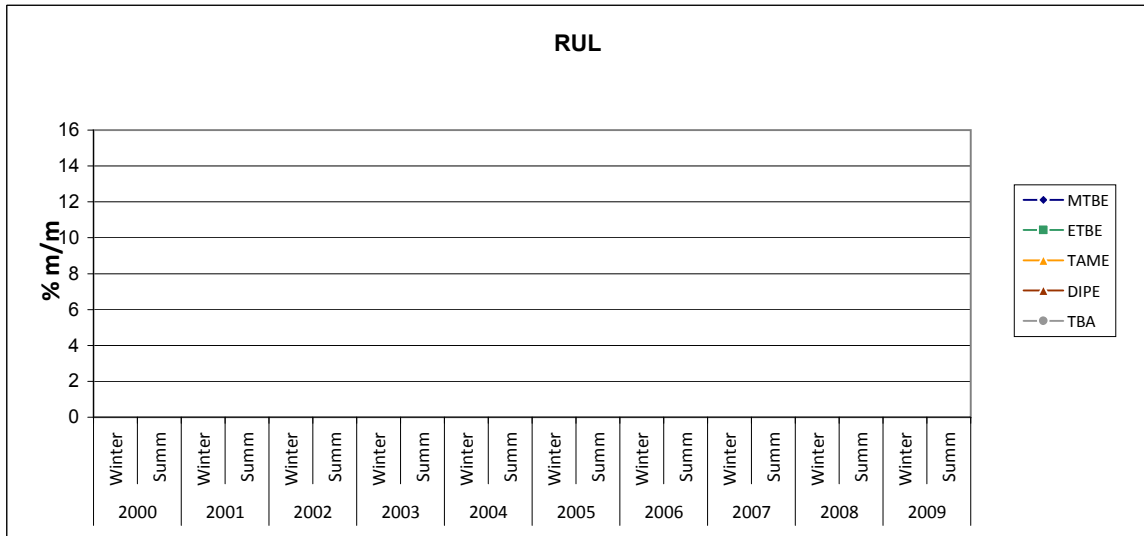
## Spain



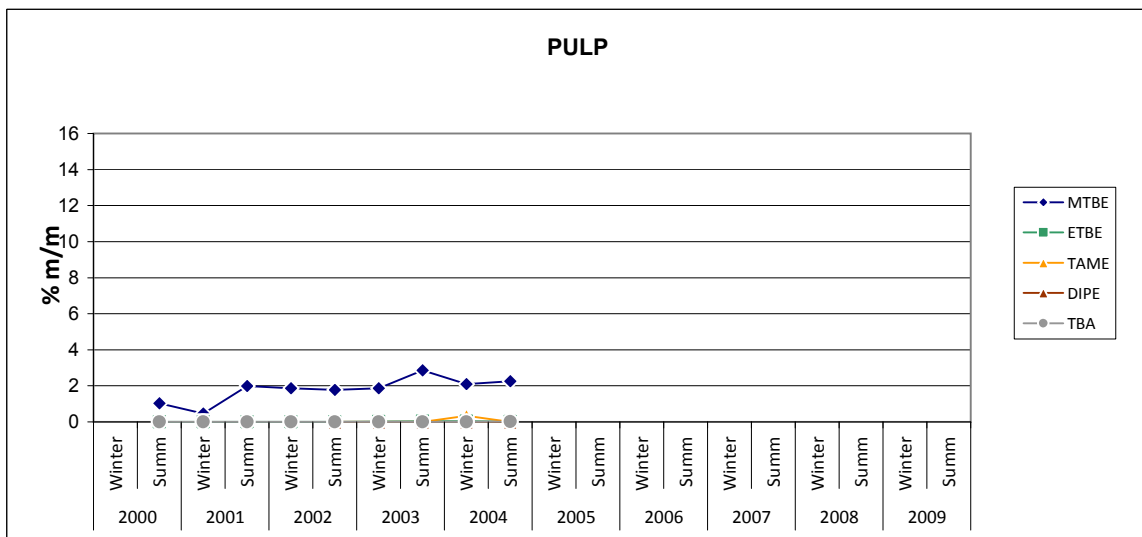
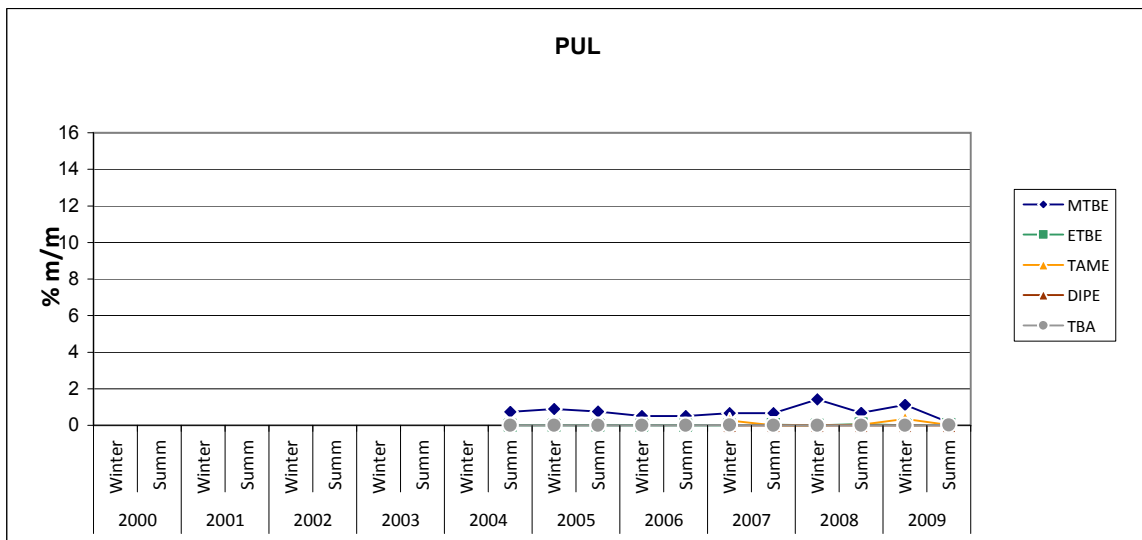
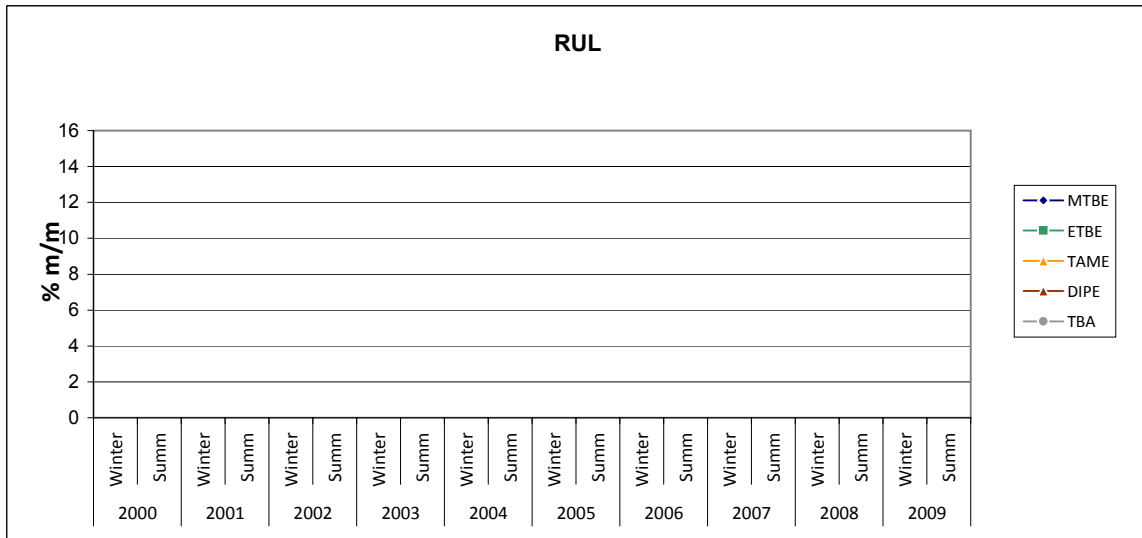
# Sweden



# Switzerland



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