

Report

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Survey of natural attenuation of petroleum hydrocarbon plumes in groundwater in Europe

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Survey of natural attenuation of petroleum hydrocarbon plumes in groundwater in Europe

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ABSTRACT

This report presents a study conducted in Europe to characterize the typical size and plume stability condition for groundwater sites impacted by the petroleum hydrocarbons benzene, toluene, ethyl benzene, xylenes (BTEX), and the fuel oxygenates methyl tert-butyl ether MTBE and ethyl tert-butyl ether (ETBE). This “plumeathon” approach have previously been used in the United States (US) in the 1990s (Newell and Connor, 1998; Kamath et al., 2012; Connor et al., 2014).

The study of impacted sites across Europe consisted of an evaluation of 50 sites. Plume length analysis was conducted for a total of 171 plumes using a primary threshold (10 µg/L for all constituents) and 138 plumes using a secondary threshold (constituent-specific drinking water guideline value). Additionally, plume stability analysis was conducted for benzene and MTBE at 29 sites. Key site characteristics and an evaluation of biodegradation were also tabulated for each site.

This study expands the understanding of petroleum plume behaviour to European sites and evaluates site-specific factors that influence this behaviour.

KEYWORDS

Plume length analysis, Europe, fuel oxygenates, methyl tert-butyl ether (MTBE), ethyl tert-butyl ether (ETBE).

INTERNET

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SUMMARY

This report presents a study conducted in Europe to characterize the typical size and plume stability condition for groundwater sites impacted by the petroleum hydrocarbons benzene, toluene, ethyl benzene, xylenes (BTEX), and the fuel oxygenates methyl tert-butyl ether MTBE and ethyl tert-butyl ether (ETBE). This “plumeathon” approach have previously been used in the United States (US) in the 1990s (Newell and Connor, 1998; Kamath et al., 2012; Connor et al., 2014), and this study expands understanding of petroleum impacted plume behaviour to European sites and evaluates site-specific factors that influence this behaviour.

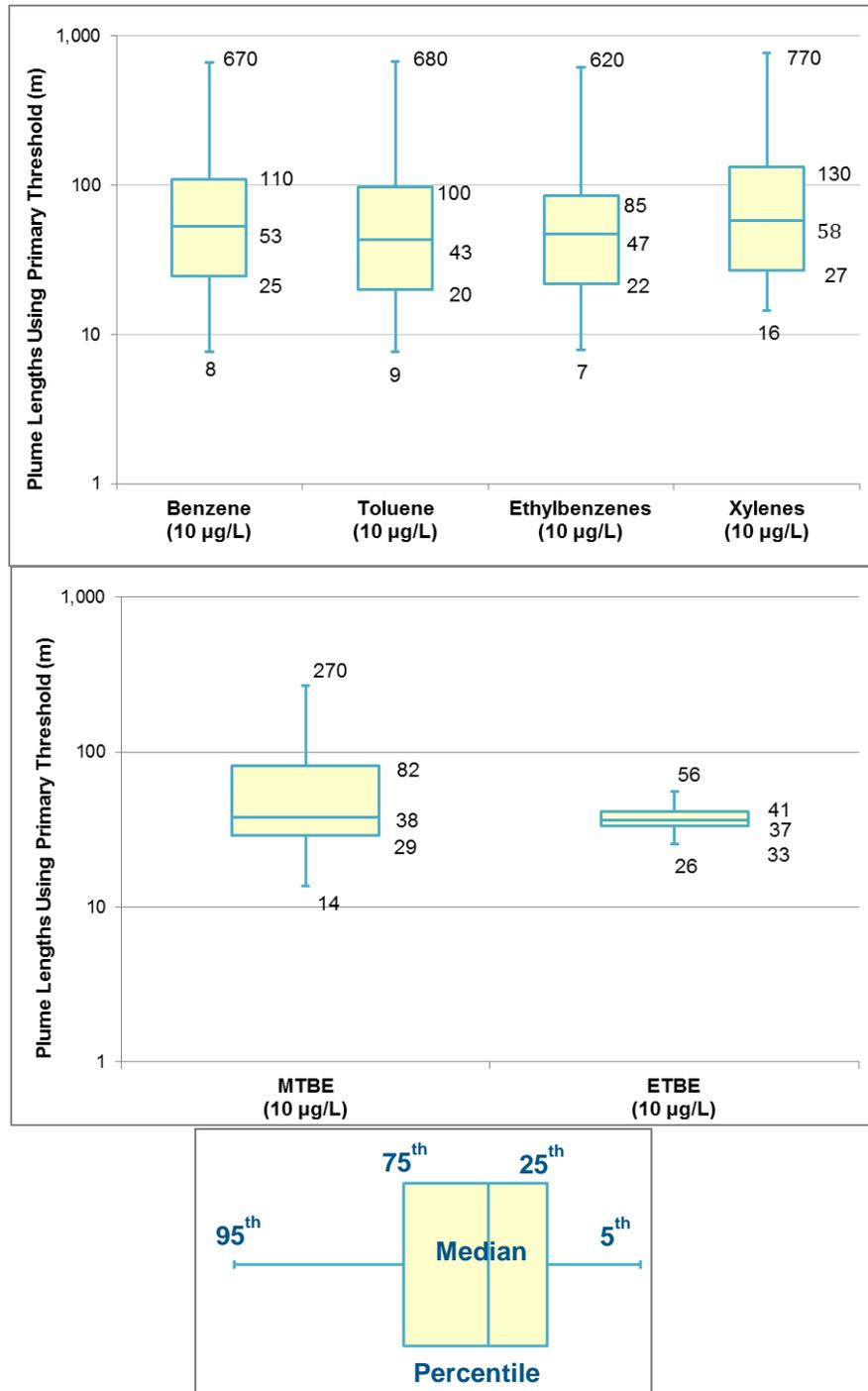
Plume length analyses were conducted using data from Concawe member companies as well as literature sources. Plume stability analysis was conducted for benzene and MTBE using historical datasets. Finally, site specific characteristics, plume lengths, etc. for each site were incorporated into one database and analysed to understand typical plume behaviour.

Key results of the analysis indicate:

A dataset of 50 sites was compiled for analysis of BTEX, MTBE and ETBE plumes. Considering each constituent separately, plume length analysis was conducted for a total of 171 plumes using a primary threshold of 10 µg/L for all constituents. The primary threshold (i.e., 10 µg/L for all constituents) provides a consistent basis for the comparison of plume lengths between constituents and between European and US studies. Within the 171 plumes, 138 plumes were additionally assessed using the constituent-specific World Health Organization (WHO) drinking water guideline value as the threshold. Plume stability analysis was conducted for benzene and MTBE at 29 sites. Key site characteristics and an evaluation of biodegradation were also tabulated for each site.

- Median reported hydraulic conductivity of a sub-set of the sites (n=27) was 3.2 m/day, and the most common soil type in both the vadose and saturated zones are sand and sand/silt.
- Soil and groundwater remediation information was available at 38 and 40 sites respectively. The principal remediation technologies employed were excavation (soil) and pump and treat (groundwater).
- As seen in **Figure 1** below, the median plume lengths for BTEX compounds were similar using the primary threshold, and ranged from 43 to 58 m. Median plume lengths for MTBE and ETBE using the primary threshold were 38 and 37 m, respectively.
- The median plume lengths for BTEX compounds using the secondary threshold ranged from 15 to 53 m. The median plume length for MTBE and ETBE using the secondary threshold were 34 and 37 m, respectively.
- Benzene plumes at high source zone concentration sites were found to be longer than those at low source zone concentration sites ($p = 0.0013$). A similar magnitude difference was found for MTBE, but this was not statistically significant at a 95% confidence level ($p = 0.086$).
- For sites having sufficient data to assess redox conditions in background wells as compared to wells within plumes, a majority of the sites evaluated (13 out of 16) showed evidence of biodegradation.

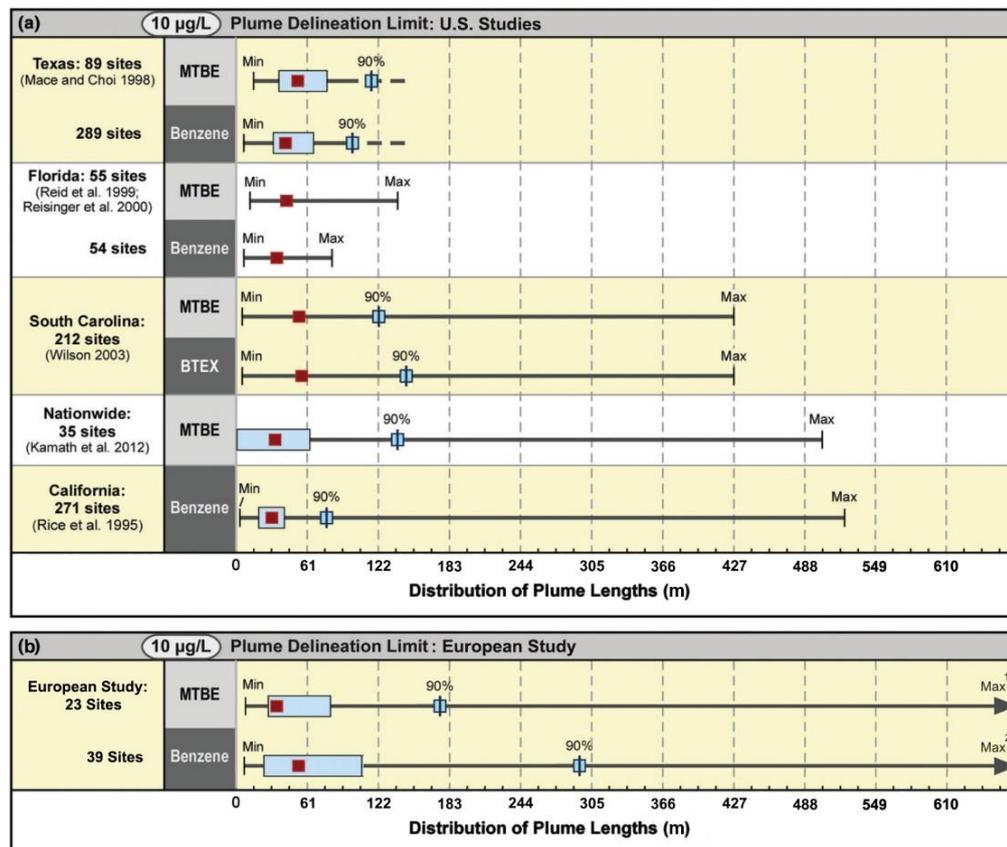
Figure 1: Range of Plume Lengths Groundwater Plumes Affected by the Constituents Analysed Using the Primary Threshold (10 µg/L): BTEX Compounds (top) and Ether Oxygenates (bottom). Box and whisker plots indicate the 5th, 25th, 50th, 75th, and 95th percentiles of the dataset.



Comparing the results from this study to a meta-analysis of US plume studies (Connor et al., 2014) indicates:

- The median plume lengths for benzene and MTBE observed in this study were similar to those reported in the U.S (**Figure 2**).
- At the majority of sites (80% in the US, and 82% in this study), benzene plume are either shrinking or stable. Similarly, for MTBE, the majority of sites (80% in the US, and 86% in this study) are also either shrinking or stable. The level of confidence in benzene and MTBE plume stability analyses was predominantly high or medium.¹
- In this study, less than 10% of all 171 plumes (for BTEX, MTBE and ETBE) using the primary threshold were greater than 500 m. Similarly, in the US, plumes greater than 500 m are rarely observed.

Figure 2: Comparison of Plume Lengths from US Studies (a) and this European Study (b) for a 10 µg/L Delineation Threshold (adapted from Connor et al., 2014). Notes (1) red squares indicate median value; (2) maximum plume length for MTBE in the European Study was 2400 m; (3) maximum plume length for benzene in European Study was 1800m.



¹ High confidence being ≥ 1 historical plume maps with fully delineated plume lengths with > 5 years between first and most current map; Medium confidence being ≥ 1 historical plume maps with either fully delineated plume lengths with 0-5 years between first and most current map, or with estimated plume length with > 5 years between first and most current map).

1. INTRODUCTION

1.1. OBJECTIVES

Understanding the fate and transport of dissolved petroleum hydrocarbons at petroleum release sites is an important component of designing appropriate site remediation and management policies. Key questions include:

1. How long are dissolved petroleum plumes?
2. Are plume lengths expanding, stable, or decreasing?
3. Are there important differences in plume lengths for different dissolved petroleum constituents?
4. Do specific site characteristics influence plume lengths?

These questions have been addressed through “plumeathon” studies conducted in the United States. However, similar studies have not previously been conducted for Europe.

To improve our understanding of dissolved petroleum plumes in Europe, GSI Environmental (GSI) compiled a plume database using the site data provided in site reports provided by Concawe Members and studies of plume behaviour in the published literature. The objectives of this study were to:

- i) better understand the behaviour of dissolved plumes at petroleum release sites (including retail sites, manufacturing sites, etc.) in Europe;
- ii) evaluate plume length, plume stability, differences between constituents, predictors of plume length; and
- iii) compare findings with plume studies previously conducted at sites in the US

1.2. BACKGROUND

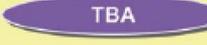
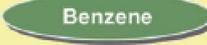
1.2.1. RESULTS OF US STUDIES

Beginning in the 1990s, a number of “plumeathon” studies were conducted in the US to characterize the typical size and plume stability condition for petroleum contaminated groundwater sites (Newell and Connor, 1998). Together, these studies found that most benzene, toluene, ethyl benzene, xylene (BTEX) plumes were less than 70 m long and stable or shrinking in size. These plumeathon studies greatly improved the acceptance of natural attenuation as a remedy for petroleum contaminated groundwater sites in the US by demonstrating that natural attenuation processes provide reliable plume containment for a large proportion of petroleum release sites.

Since the 1990s, the plumeathon approach has been expanded to cover fuel oxygenates such as methyl tert-butyl ether (MTBE) and tertiary butyl alcohol (TBA) (Kamath et al., 2012; Connor et al., 2014). These subsequent studies have found that oxygenate plumes are typically no longer than BTEX plumes (i.e., <70 m; **Figure 3**), although longer oxygenate plumes (> 500 m) have been observed at a very small percentage of sites (i.e., <1%). However, even the unusually long MTBE plumes have been found to persist for only a limited duration. A recent review of sites previously documented to have exceptionally long MTBE plumes found dramatic decreases in plume length at most of these sites in the years following initial plume delineation (McDade et al., 2015).

MTBE was largely eliminated from gasoline in the United States by 2006 (or earlier in some states), and, since that time, MTBE concentrations in groundwater have been observed to be decreasing more rapidly than benzene concentrations for the same populations of sites (Kamath et al., 2012; McHugh et al., 2014). The difference in attenuation rates between benzene and MTBE are largely independent of light non-aqueous phase liquid (LNAPL) recovery efforts (Kulkarni et al., 2015) or application of active plume remediation technologies (McHugh et al., 2014) suggesting that the decreasing concentrations are primarily attributable to natural attenuation processes. MTBE impacts to public water supply wells have been much less frequent than was feared in the 1990s and much less frequent than has been observed for other widely used chemicals such as tetrachloroethene (McHugh et al., 2015) and nitrate from agricultural sources. In Europe, MTBE has been steadily replaced by ETBE and ethanol since the mid-2000's, and therefore trends in MTBE concentrations at European sites may be similar.

Figure 3: Summary of Typical Plume Lengths at Underground Storage Tank (UST) Sites in the US (from Connor et al., 2014). From top to bottom, results are in meters are as follows: 50 m (165 ft); 43 m (140 ft); 58 m (190 ft); 122 m (400 ft); 105 m (345 ft); 128 m (420 ft); 84 m (275 ft); 55 m (180 ft); 162 m (530 ft); 130 m (425 ft).

(A) Results for MTBE, Benzene, and TBA Plumes at UST Sites		
10 µg/L	MEDIAN PLUME LENGTHS	391 sites  MTBE 165 feet
		826 sites  Benzene 140 feet
		108 sites  TBA 190 feet
90th-PERCENTILE PLUME LENGTHS		336 sites  MTBE 400 feet
		772 sites  Benzene 345 feet
		108 sites  TBA 420 feet
(B) Results for MTBE and Benzene Plumes at UST Sites		
5 µg/L	MEDIAN PLUME LENGTHS	96 sites  MTBE 275 feet
		165 sites  Benzene 180 feet
90th-PERCENTILE PLUME LENGTHS		96 sites  MTBE 530 feet
		165 sites  Benzene 425 feet

1.2.2. MOTIVATION FOR A EUROPEAN PLUMEATHON STUDY

The regulatory community in Europe has a good technical understanding of natural attenuation processes and the rationale for applying natural attenuation as a site remedy. However, technical questions remain that limit the application of natural attenuation as a site remedy in some countries. Specifically, most of the large plumeathon studies have been completed using sites in the US, raising questions within the regulatory community as to whether the typical plume lengths and attenuation rates documented in these studies are applicable to sites in Europe. In comparison to sites in the US plumeathon studies, which are dominated by sites in California, Florida, Texas and North Carolina, northern and eastern European sites can exhibit colder groundwater temperatures and impacts to bedrock aquifers may be more common. The purpose of this study was to apply the plumeathon approach to a set of sites in Europe in order to characterize plume lengths and attenuation rates at petroleum sites under these hydrogeological conditions, and to determine the site-specific factors that influence these characteristics

2. METHODOLOGY

2.1. INITIAL SITE SCREENING

For each site, the available site reports were reviewed to determine if sufficient information was available regarding groundwater plume conditions. For a site to have been included in the plume length estimation portion of the study, the available site reports must provide, at a minimum, enough information to allow estimation of the plume length in groundwater for one or more of the constituents of interest. Sites that met the following data requirements were selected for analysis:

- Groundwater concentration data for one or more of the following constituents: benzene, toluene, ethylbenzene, xylenes, MTBE and ETBE, and
- A plume map (or a well location map and tabular concentration data) for most recent groundwater monitoring results that is sufficient to document full or partial plume delineation.

Sites with sufficient information to determine plume length for at least one constituent were included in the database even if other desired information was not available. Missing information concerning plume stability or site characteristics did not result in rejection of the site. Instead, the information fields in the Data Template were recorded as “not available”.

Conversely, the following are examples that caused sites to be rejected from the analysis, as insufficient information was available to determine plume lengths with a minimum level of confidence:

- No well location map available;
- No plume map and no tabulated concentration data per well available
- No concentrations above the study’s plume delineation thresholds; or
- Only data in source area and not enough data in downgradient direction to support estimation of a plume attenuation rate.

2.2. DATA ACQUISITION AND SITE CHARACTERISTICS

All site specific characteristics, plume lengths, etc. for each site were tabulated in the Data Template that served as a form for consistent data acquisition for this study (**Appendix A**). The data template enabled records of key information for each site and included:

1. Estimated release volume and basis for estimate
2. Presence or absence of LNAPL at site including number of wells with LNAPL and current/historical LNAPL thickness
3. The remediation status for the source area (ongoing, excavated, remediated, unknown)
4. Attenuation parameters including:
 - a. site location climate statistics (from literature)
 - b. subsurface geology & hydrogeology
 - c. historic remediation activities (e.g. source removal, pump and treat, application of chemical oxidation)

- d. subsurface redox conditions (e.g. redox probe data or concentrations of redox sensitive species in groundwater, e.g. oxygen, nitrate, sulphate, methane)
 - e. microbiology (e.g. colony counts; microbial assemblages; DNA/RNA)
 - f. site-specific partitioning coefficient (Kd) or retardation coefficient (R) values,
 - g. site-specific half- life values or biodegradation rates from field or microcosm data
5. Plume length information for each key petroleum constituent and oxygenate (e.g., benzene, ethylbenzene, toluene, xylenes, MTBE, and ETBE) including:
 - a. Plume delineation status (e.g., fully delineated, partly delineated, source area only)
 - b. Plume length
 - c. Plume length estimation method (e.g., isocontour map, interpolation, extrapolation)
 - d. GSI's confidence in the plume length (low, medium, high).
 6. The plume status (expanding, stable, shrinking, exhausted), the evidence on which these estimates are based and GSI's confidence in this evidence (low, medium, high)

These files were compiled into an MS Access database for further analysis of typical plume characteristics.

2.3. ESTIMATION OF PLUME LENGTHS

Plume maps or text from contractor reports were used to determine plume length information from each site. The constituents of concern that were analysed include: benzene, toluene, ethylbenzenes, xylenes, methyl tert-butyl ether (MTBE) and ethyl tert-butyl ether (ETBE).

The length of the affected groundwater plumes were determined using two plume delineation thresholds for each constituent (see **Table 1**), the primary threshold (i.e., 10 µg/L for all constituents) provides a consistent basis for the comparison of plume lengths between constituents and between European and US studies and a secondary threshold (i.e., the recommended World Health Organization drinking water standards for each constituent) provides a basis for evaluating the potential impacts to drinking water resources. Each plume length determination was also followed by a qualitative determination in the confidence of that plume length (i.e., low, medium, high).

For each plume map acquired, the following were the order of preference in which the plume lengths were calculated, as well as the level of confidence in the calculation:

1. fully delineated plume (high or medium confidence)
2. report text indicating plume length (medium confidence)
3. extrapolated iso-contour of plume delineation threshold (low confidence)

Table 1: Primary and Secondary Plume Delineation Thresholds for Constituents

Constituent	Primary Plume Delineation Threshold	Secondary Plume Delineation Threshold (using drinking water standards)	Source of Drinking Water Standard
Benzene	10 µg/L	10 µg/L	EU Drinking Water Directive
Toluene	10 µg/L	700 µg/L	World Health Organization
Ethylbenzenes	10 µg/L	300 µg/L	World Health Organization
Xylenes	10 µg/L	500 µg/L	World Health Organization
MTBE	10 µg/L	15 µg/L	World Health Organization
ETBE	10 µg/L	10 µg/L	Based on Health Effects Institute, 1996*

* No drinking water standard applied in US or EU

2.3.1. Method 1: Fully Delineated Plume

In this study, a fully delineated plume was defined as one in which the existing monitoring well network contained at least one clean downgradient well (i.e., with a concentration less than the delineation threshold for a given chemical). For each site with a plume map showing a fully delineated plume, the plume length was determined from the available plume map as the distance in the downgradient direction from upgradient of the source (release area or highest concentration well) to the location where the concentration is less than the delineation threshold.

The level of confidence in this plume length was defined as “high” if the plume length can be determined using minimal interpolation (i.e., the available information supports a plume length estimate within approximately +/- 20%). If a higher degree of interpolation is required, then the level of confidence was defined as medium.

2.3.2. Method 2: Report Text with Plume Length

For sites without a map showing a fully delineated plume, the plume length reported in the text was used, if available. The level of confidence was defined as medium.

2.3.3. Method 3: Estimated (Extrapolated) Plume

An incompletely delineated plume is one in which the existing plume monitoring network does not cover the full extent of the plume. For incompletely delineated plumes, the plume length was estimated based on the observed bulk attenuation rate (i.e., the rate at which the contaminant concentration decreases downgradient of the source area due to the combined effects of dispersion, biodegradation and other attenuation processes (Newell et al., 2002).

First, the bulk attenuation rate was estimated from available concentration vs. distance data from at least three wells as described in Newell et al., 2002. Second, the plume length was estimated using this bulk attenuation rate by calculating the distance required for the constituent concentration to decrease from the concentration in the source area to the delineation standard. Estimated plume lengths were assigned a low confidence.

2.4. PLUME STABILITY OVER TIME

Plume stability classification was assessed for benzene and MTBE by evaluating historical plume lengths using the primary delineation thresholds for each constituent.

Maps depicting plumes for at least two points in time were required for the stability analysis. If available, plume maps for three time periods were used to determine the change in plume length as follows:

$$\% \text{ Change} = \frac{(\text{Current Plume Length} - \text{Oldest Historical Plume Length})}{(\text{Oldest Historical Plume Length})} \times 100\%$$

Next, the plume stability classification can be determined based on the following definitions:

- i) Shrinking: More than 20% reduction
- ii) Stable: Reduction or expansion less than 20%
- iii) Expanding: More than 20% increase
- iv) Detached: Visual determination based on highest concentration moving away from source area

In the absence of historical plume maps, we relied upon site reports that characterize the plume stability over time, if available. Finally, the level of confidence in the plume stability (i.e., low, medium, high) was determined using the guidelines in **Table 2** below.

Table 2: Guidelines for Typical Plume Stability Level of Confidence

# of Historical Plume Maps	Time Period between First Historical Plume Map and Current Plume Map	Estimated Plume Lengths?	Fully Delineated Plume Lengths?	Level of Confidence in Plume Stability
≥1	>5 y	No	Yes	High
≥1	>5 y	Yes	No	Medium
≥1	0-5 y	No	Yes	Medium
≥1	0-5 y	Yes	No	Low
None (Report text)	n/a	n/a	n/a	Medium

Note: Table presents typical basis for assigning level of confidence. Level of confidence may be increased or decreased by other available information.

If more than two plume maps were available, the stability determination and confidence were modified based on the consistency in plume trends and conditions across the additional time periods.

3. RESULTS

3.1. OVERALL DATASET

As seen in **Table 3** below, a total of 86 sites were evaluated for this study. Sixty-six (66) of these 86 sites were evaluated using site reports provided by Concawe member companies and 20 sites were evaluated using published literature. Literature studies included those that were evaluated in “Stage 1 Survey of Natural Attenuation of BTEX, MTBE and ETBE Plumes in Groundwater in Europe”, dated 27 May 2015. Of the 86 sites reviewed, 50 were retained for plume length analysis of at least one constituent, and 29 were retained for plume stability analysis.

For each of the 50 retained sites, plume lengths were calculated for one or more constituents using the primary and secondary thresholds. For each constituent, there were between eight and 39 plumes evaluated using the primary and secondary thresholds. For example, 39 of the 50 sites had benzene plumes that could be evaluated while only 8 of the 50 sites had ETBE plumes. Across all constituents, in a total of 171 plumes were evaluated using the primary thresholds, and 138 plumes were evaluated using the secondary thresholds (see **Table 4**). The number of plumes evaluated using the secondary thresholds was smaller because the secondary thresholds were generally higher than the primary thresholds. At some sites, the maximum constituent concentration exceeded the primary threshold of 10 µg/L but was less than the secondary threshold (e.g., 700 µg/L for toluene).

Table 3: Total Number of Sites Evaluated and Retained for Further Analysis

	Site Reports	Literature Studies	Total
Sites Evaluated	66	20	86
Sites Retained for Plume Lengths	48	2	50
Sites Retained for Plume Stability	29	0	29

Table 4: Total Number of Plumes Evaluated for Each Constituent

Plume Length Analysis Using...	Benzene	Toluene	Ethyl-benzene	Total Xylenes	MTBE	ETBE	Total
Primary Threshold	39	33	35	33	23	8	171
Secondary Threshold	39	20	24	27	20	8	138

Key Points

- The dataset included 50 sites that were retained for plume length analysis for at least one constituent and 29 sites for plume stability analysis for either benzene or MTBE.
- Across all constituents, a total of 171 plumes were evaluated using the primary threshold (10 µg/L), and 138 plumes were evaluated using the secondary thresholds (drinking water guidelines).

3.2. GENERAL SITE CHARACTERISTICS

Of the 50 sites retained for plume length analysis, 33 sites are located in either Spain or Hungary, with the other 17 sites located in other countries across Europe. A breakdown of countries and number of sites in each country are listed in **Table 5** below.

Table 5: Sites Analysed, by Country

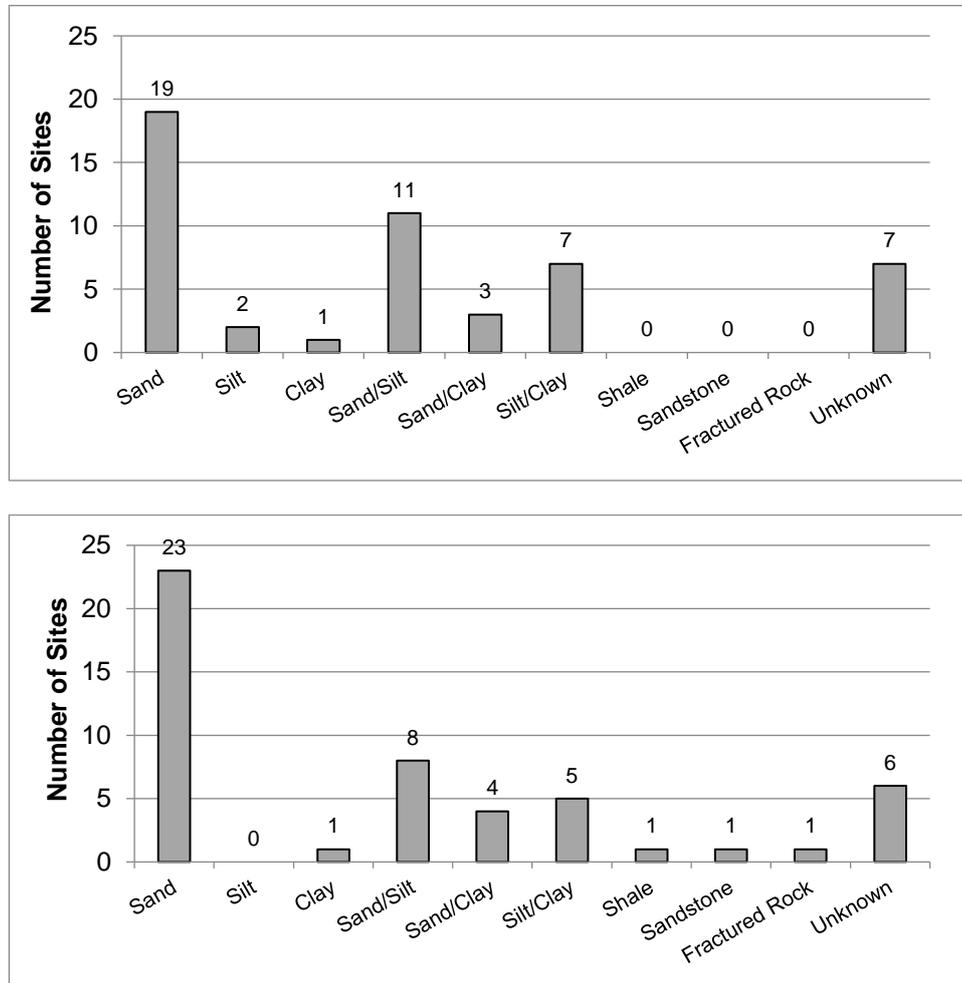
Country	Number of Sites
Belgium	8
Denmark	1
France	1
Germany	2
Hungary	18
Italy	2
Netherlands	1
Spain	15
United Kingdom	1
Unknown*	1
Total	50

*This site was obtained from a published literature study that did not identify the specific country for the site.

Appendix B summarizes the use of MTBE and ETBE in gasoline in European countries. Overall, the average concentration in all European gasoline samples in 2000-2009 was 5.4 m/m% for MTBE and 0.9 m/m% for ETBE (Concawe, 2012).

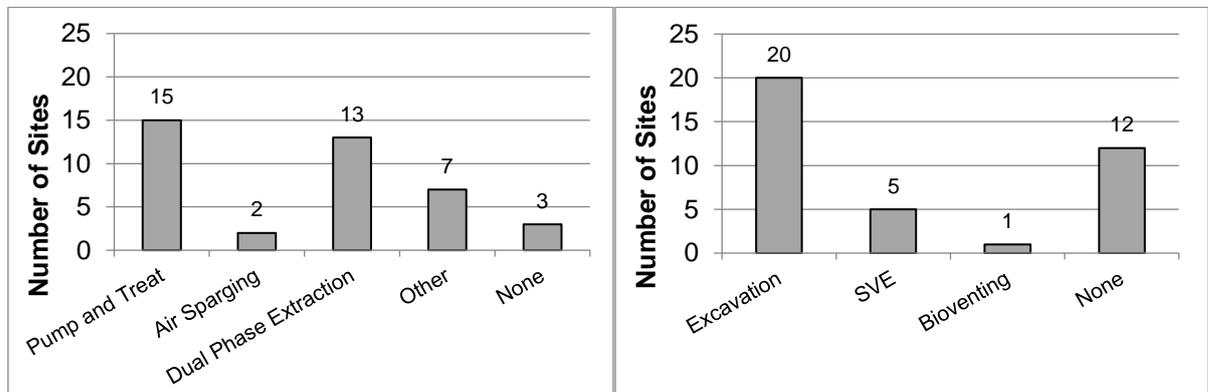
At these sites, the most common soil types, in both the vadose zone and saturated zone, are sand and sand/silt, as seen in **Figure 4** below.

Figure 4: Sites by Predominant Soil Type in the Vadose Zone (top) and Saturated Zone (bottom).



Information was available for soil remediation at 38 sites, and for groundwater remediation at 40 sites. A variety of soil and groundwater remediation technologies have been used at these sites. The principal soil remediation technique is excavation, and the principal groundwater remediation techniques are pump and treat and dual phase extraction (Figure 5).

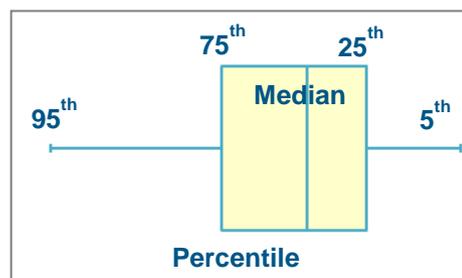
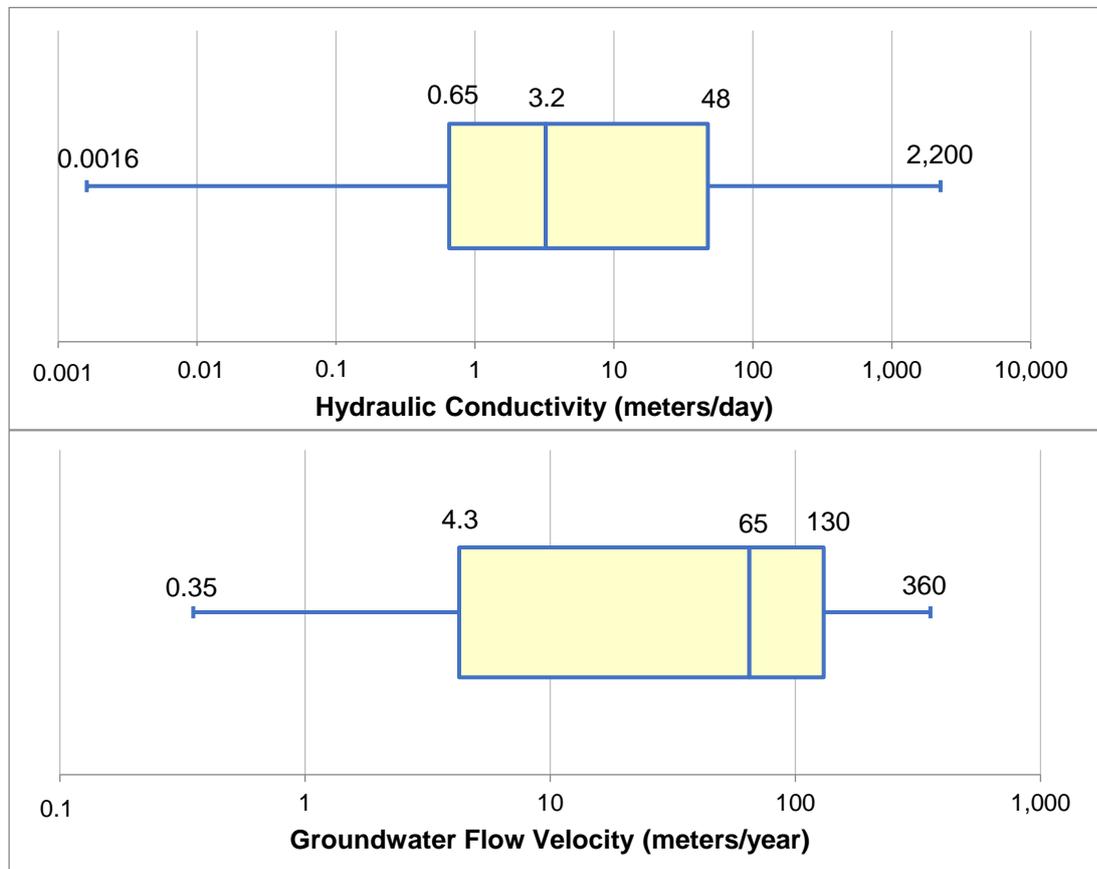
Figure 5: Sites by Soil Remediation Technology (left) and Groundwater Remediation Technology (right). “Other” groundwater remediation techniques include chemical oxidation, bio-stimulation, etc.



A wide range of values were reported for the hydraulic conductivity of the affected aquifer, and for the groundwater flow velocity (**Figure 6**). The median reported hydraulic conductivity¹ was 3.2 m/day from a dataset of 27 sites and the median reported groundwater flow velocity was 65 m/y (0.18 m/day) from a dataset of 8 sites.

¹ The median was considered as the better representation of the central tendency since the value of the arithmetic average hydraulic conductivity was heavily influenced by a small number of sand/gravel sites with high hydraulic conductivities, and therefore don't considered to be very meaningful.

Figure 6: Range of Values for Hydraulic Conductivity (top) and Groundwater Flow Velocity (bottom).



Key Points

- Of the 50 sites retained for plume length analysis, the majority (i.e., 33 sites) are located in either Spain or Hungary, with the other 17 sites located across Europe (Belgium, Denmark, France, Germany, Italy, Netherlands, UK).
- The most common soil type in both the vadose and saturated zones are sand and sand/silt.
- Soil and groundwater remediation information was available at 38 and 40 sites respectively. The principal remediation technologies employed are excavation (soil) and pump and treat (groundwater).
- Median reported hydraulic conductivity was 3.2 m/day (n=27 sites) and the median reported groundwater flow velocity was 65 m/y (n=8 sites).

3.3. PLUME LENGTHS OF ALL CONSTITUENTS

Evaluation of typical plume lengths at petroleum release sites helps in understanding the potential for impacts to near-by water wells or other receptors. Differences in plume lengths between constituents also help determine which constituents are the best indicator parameters.

Using the primary delineation threshold of 10 µg/L for all constituents, plume lengths of all sites and constituents are presented in **Figure 7** below. The median plume lengths for BTEX compounds were similar, and ranged from 43 to 58 m. Median plume lengths for MTBE and ETBE were 38 and 37 m, respectively. **Figure 8** presents plume length statistics for all compounds using their individual secondary thresholds.

Summary tables of all primary and secondary plume lengths by confidence interval are provided in **Appendix C**. The percentage of primary plume lengths for constituents with the following confidence levels were as follows: i) high: 19-33%; ii) medium: 54-66%; iii) low: 13-38%. The percentage of secondary plume lengths for constituents with the following confidence levels were as follows: i) high: 22-45%; ii) medium: 33-63%; iii) low: 13-38%.

A separate statistical analysis of the differences in primary plume lengths by confidence interval was also conducted for each compound in which a statistical two-sided t-test was performed between the following subsets: i) high vs. medium confidence; ii) medium vs. low confidence; and iii) high vs. low confidence. Here, the plume lengths between the different confidence interval buckets showed no statistical difference in means at the 95% confidence level ($p < 0.05$ indicates statistically significant difference in means of two subsets) for all constituents and buckets, except between “high” and “medium” confidence plume lengths for xylene.

Figure 7: Range of Plume Lengths of Groundwater Plumes Affected by the Constituents Analysed Using the Primary Threshold (10 µg/L): BTEX Compounds (top) and Ether Oxygenates (bottom). Box and whisker plots indicate the 5th, 25th, 50th, 75th, and 95th percentiles of the dataset.

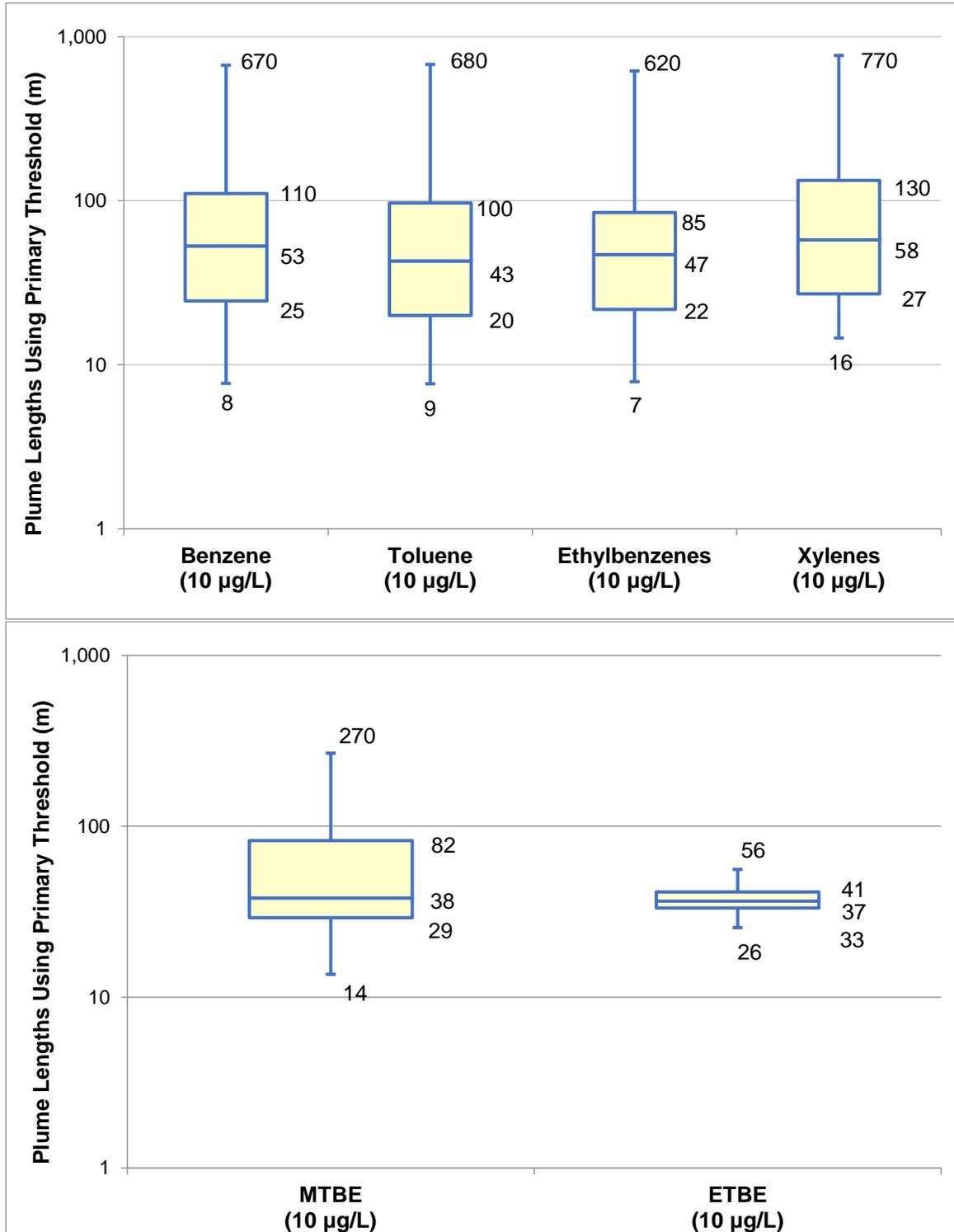
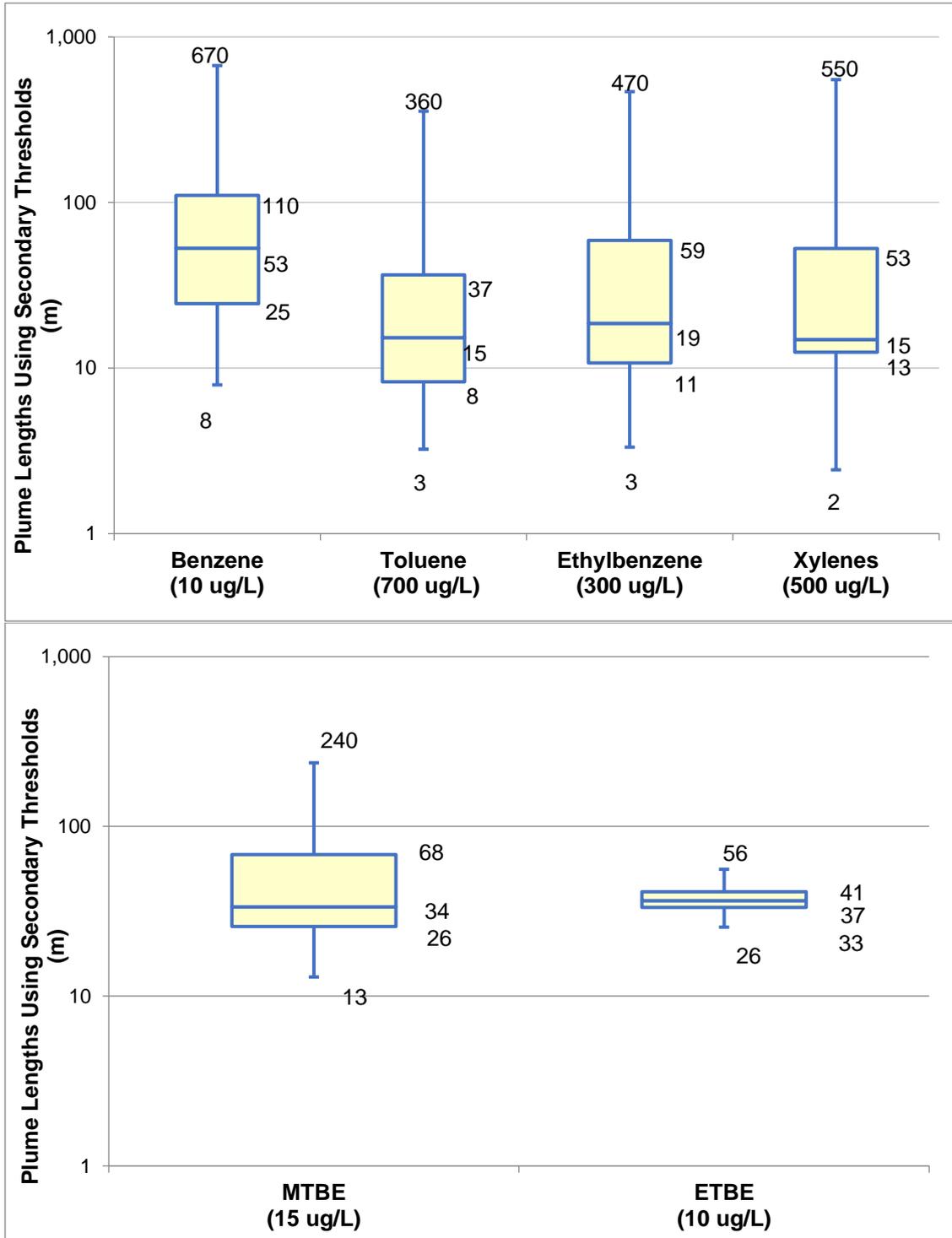


Figure 8: Range of Plume Lengths of Groundwater Plumes Affected by the Constituents Analysed Using the Secondary Threshold (i.e., Drinking Water Guidelines): BTEX Compounds (top) and Ether Oxygenates (bottom). Box and whisker plots indicate the 5th, 25th, 50th, 75th, and 95th percentiles of the dataset.



Key Points

- The median plume lengths for BTEX compounds were similar using the primary threshold, and ranged from 43 to 58 m. Median plume lengths for MTBE and ETBE using the primary threshold were 38 and 37 m, respectively.
- The median plume lengths for BTEX compounds using the secondary threshold ranged from 15 to 53 m. The median plume length for MTBE and ETBE using the secondary threshold were 34 and 37 m, respectively.

3.4. RELATIONSHIP BETWEEN SOURCE ZONE CONCENTRATIONS AND PLUME LENGTH

The mean benzene and MTBE plume lengths for high and low source concentration sites were compared to determine whether source concentration is a factor in current plume length. For each plume, the source zone concentration for each constituent was defined as the maximum concentration within the delineated plume. First, Shapiro-Wilk tests for normalcy were conducted and determined that the data for plume lengths and maximum source zone concentrations of both benzene and MTBE are not normally distributed. Therefore, non-parametric Wilcoxon t-tests were used to compare the plume lengths at high concentration (source zone concentration greater than median) and low concentration (source zone concentration lesser than median) sites. Benzene plumes at high concentration sites were found to be longer than those at low concentration sites ($p = 0.0013$). A similar magnitude difference was found for MTBE, but this was not statistically significant at a 95% confidence level ($p = 0.086$) (**Table 6**).

Aside from source zone concentration, plume length was not correlated with any other site characteristics, either due to lack of sufficient data or no relationship with site factors. The site characteristics evaluated as potential predictors of plume length were: site type, release volumes, groundwater velocity, and geochemical conditions. This lack of correlation is consistent with prior plume studies conducted in the United States, with the exception of one previous study on exceptionally long MTBE plumes (McDade et al., 2015). Here, exceptionally long MTBE plumes were impacted by the following factors: i) larger release volumes; ii) multiple releases; and iii) high groundwater velocity.

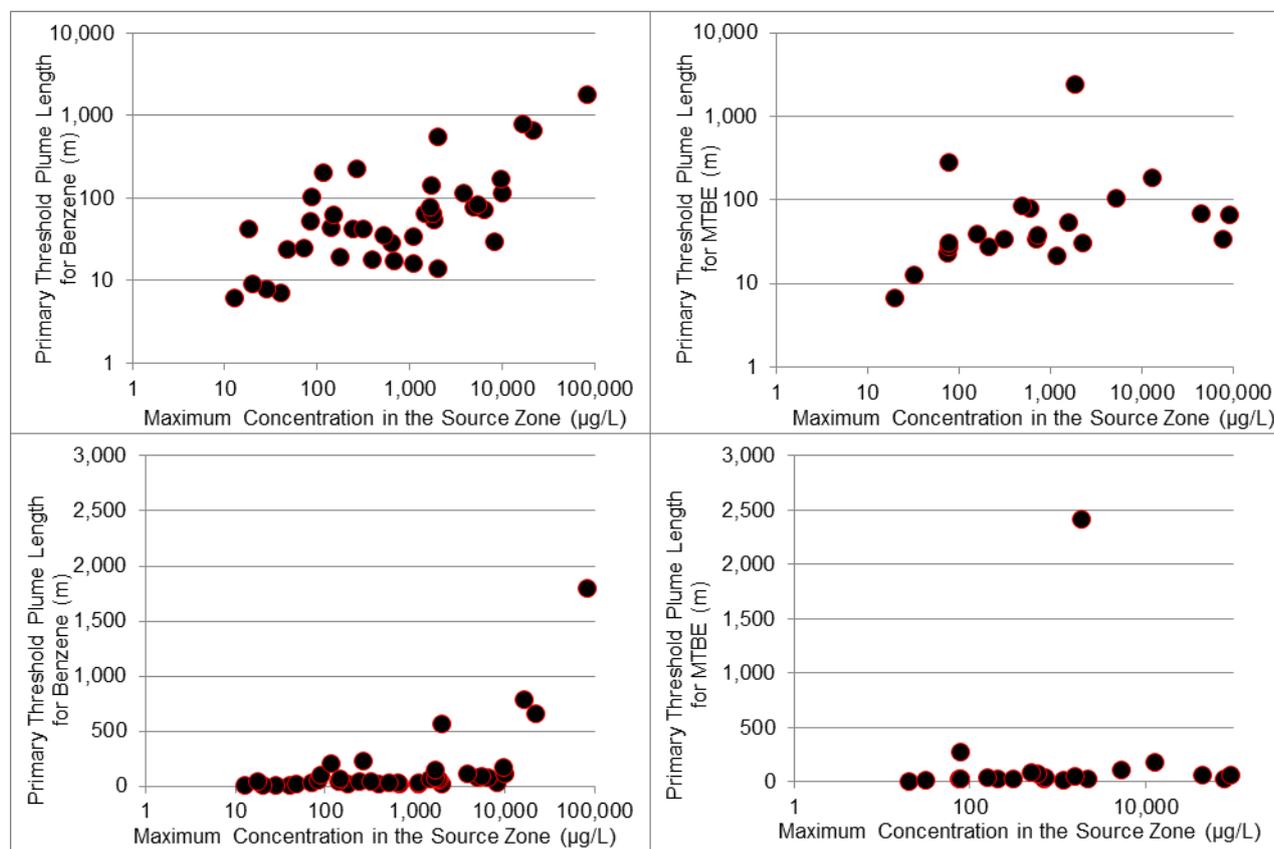
Figure 9 below also shows plume lengths vs. source concentrations for the two compounds.

Table 6: Results of Wilcoxon Test Comparing Plume Lengths at High- and Low-Concentration Sites

Constituent	Mean Plume Length, High Source Concentrations	Mean Plume Length, Low Source Concentrations	p-value	Statistically Significant?
Benzene	260	51	0.0013	Yes
MTBE	277	58	0.086	No

Note: High Concentration defined as $> 680 \mu\text{g/L}$ for benzene, and $> 600 \mu\text{g/L}$ for MTBE

Figure 9: Relationship between Plume Lengths and Source Concentrations for Benzene (left panel) and MTBE (right panel). On the top row, the y-axis is on a logarithmic scale, on the bottom row, a linear scale.



Key Points

- Source concentration (i.e., highest concentration within plume boundary) was evaluated and compared against plume lengths.
- Benzene plumes at high concentration sites were found to be longer than those at low concentration sites, with statistical significance ($p=0.0013$). A similar difference was found for MTBE, but was not statistically significant at 95% confidence ($p=0.086$).

3.5. PLUME STABILITY OF BENZENE AND MTBE

Plume stability conditions for benzene and MTBE were determined using historical data for each site in order to determine whether the plumes are shrinking, stable, expanding or detached. As seen in **Table 7** and **Table 8** below, at the majority of sites, plumes are either stable or decreasing in length.

For expanding and shrinking plumes, the change in plume length over the available monitoring record (ranging from 1 to 10 years with a median of 4 years) was evaluated. For the nine shrinking benzene plumes, the median decrease in plume length was 36 m. For the four expanding benzene plumes, the median increase in plume length was 25 m. For the six shrinking MTBE plumes, the median decrease was 46 m and for the one expanding MTBE plume, the increase was 15 m. For the

benzene plume showing the largest increase in length (from 20 m to 55 m), the current length of 55 m is approximately equal to the median plume length for the full set of plumes (53 m). Similarly, for the one expanding MTBE plume, the current plume length of 66 m is within the middle 50% of plume lengths (29 to 82 m). This suggests that the observation of expanding plumes at these sites was part of the normal plume lifecycle rather than site conditions leading to unusually long plumes.

The level of confidence in benzene and MTBE plume stability analyses was predominantly high or medium (22 out of 23 sites for benzene; 8 out of 14 for MTBE).

Table 7: Summary of Plume Stability Analyses for Benzene

Constituent: Benzene	Shrinking	Stable	Expanding	Detached	Total Sites
No. of Sites	9	10	4	0	23
Median Plume Length Change (m)	-36	2	25	--	--

Table 8: Summary of Plume Stability Analyses for MTBE

Constituent: MTBE	Shrinking	Stable	Expanding	Detached	Total Sites
No. of Sites	6	6	1	1	14
Median Plume Length Change (m)	-46	-1	15	--	--

Key Points

- Plume stability analyses were conducted for benzene and MTBE plumes by evaluating historic data. At 19 out of 23 sites, benzene plumes are either shrinking or stable. At 12 out of 13 sites, MTBE plumes are either shrinking or stable.
- Plume length changes from historical to most recent data were quantified, and indicated that shrinking benzene plumes decreased by a median value of approximately 36 m, and expanding benzene plumes expanded by a median value of approximately 25 m. For MTBE, the shrinking plumes decreased by a median value of 46 m, and the one expanding plume expanded by 15 m.
- Level of confidence in plume stability analyses for both benzene and MTBE was predominantly classified as either medium or high.

3.6. EVIDENCE OF BIODEGRADATION

Natural attenuation is the natural reduction in contaminant concentrations in groundwater without active remediation. Natural attenuation includes destructive processes such as biodegradation and abiotic transformation and non-destructive processes such as dispersion and volatilization. For petroleum hydrocarbons, biodegradation is the most significant natural attenuation process. Following the initial release, natural attenuation processes result in a predictable sequence of events at most petroleum sites consisting of i) initial plume growth followed by, ii) stabilization, iii) shrinking, and then iv) disappearance of the plume.

At most or all petroleum release sites, biodegradation is an important attenuation mechanism. Even at sites with active remediation systems, the majority of mass removal can be attributed to biodegradation processes. In fact, monitored natural attenuation (MNA) is used at many sites as the sole remediation technology (Adamson and Newell, 2014). Additionally, some contaminants such as MTBE are more amenable to biodegradation than previously thought (Adamson and Newell, 2014).

As such, evidence of biodegradation was evaluated at all sites with sufficient data. Here, redox conditions were compared in one well within the plume to an upgradient background well. Biodegradation was considered to be active if the following trends are true:

- Decrease of electron acceptors within plume as compared to background: dissolved oxygen, nitrate and sulphate
- Increase of methane within plume
- Increase of metabolic by-products within plume: Manganese and dissolved iron.

A total of 16 sites had sufficient data to assess redox conditions of both a background well and a well within the plume. Of these, a total of 13 sites indicated one or more of the trends listed above, as a result of biodegradation. As seen in **Table 9**, median concentrations of oxygen, nitrate, sulphate, methane, iron and manganese are presented in a background wells and plume wells.

Table 9: Median Concentrations of Redox Conditions in Background and Plume Wells with Evidence of Biodegradation

Compound	Median Concentration in Background Well (mg/L); (n=number of sites)	Median Concentration in Plume Well (mg/L); (n= number of sites)	Concentration Trend from Background to Plume Wells	Indicative of Biodegradation?
Oxygen	2.2 (n=7)	1.3 (n=6)	Decrease	Yes
Nitrate	39 (n=11)	0.8 (n=10)	Decrease	Yes
Sulphate	63 (n=11)	23 (n=10)	Decrease	Yes
Methane	0.2 (n=6)	2.2 (n=5)	Increase	Yes
Iron	0.4 (n=11)	12.9 (n=10)	Increase	Yes
Manganese	0.3 (n= 7)	2.4 (n=7)	Increase	Yes

Over the last 5-10 years in the US, due to new understanding about LNAPL biodegradation along with new ways to measure degradation rates, practitioners have found that Natural Source Zone Depletion (NSZD) rates are much greater than previously thought via MNA. Additionally, LNAPL biodegradation can target more compounds than just BTEX compounds, such as alkenes. NSZD is the natural loss of LNAPL due to volatilization, dissolution, and most importantly, biodegradation (ITRC, 2009). Because of new understanding about LNAPL biodegradation and new ways to measure NSZD, there has been a movement by the remediation industry to rely on NSZD at many sites to manage LNAPL.

A series of LNAPL research projects determined that anaerobic biodegradation processes are more important than previously thought, and these processes act in both the vadose and saturated zones to generate methane. In fact, the amount of degradation that was expressed by the vertical gas transport exchange was almost 100 times the biodegradation from the electron acceptors measured in the saturated zone (Suthersan et al., 2015).

Currently, four key measurement methods are available to quantify NSZD rates at LNAPL sites:

- Carbon Traps (E-Flux, 2016);
- Dynamic Closed Chamber Method (Sihota et al., 2011; LI-COR, 2015),
- Gradient Method (Lundegard and Johnson, 2006; Sihota, et al., 2011); and
- Thermal NSZD (Thermal NSZD, 2016).

These technologies have been employed at a number of sites, and the NSZD rates have been measured to be on the order of 1000 L/ha/y to 100,000 L/ha/y of petroleum (McCoy et al., 2012; Sihota et al., 2011; LA LNAPL Workgroup, 2015).

Key Points

- Majority of sites evaluated (13 out of 16) indicated evidence of dissolved plume biodegradation through the analysis of redox conditions in background wells as compared to wells within plumes.
- In addition, recent research has indicated that most or all petroleum sites exhibit significant source zone biodegradation that is not accounted for by the evaluation of dissolved plume indicators. This research has documented that typical source area biodegradation results in source attenuation on the order of 1000 L/ha/y to 100,000 L/ha/y of petroleum (McCoy et al., 2014; E-Flux, 2016; Sihota et al., 2011; Thermal NSZD, 2016).

3.7. COMPARISON TO RESULTS FROM US STUDIES

Figure 10 and **Table 10** below summarize the comparison of plume lengths and plume stability for benzene and MTBE in the US with those observed in this European study.

The median plume lengths for benzene and MTBE observed in this study were similar to those reported in the US. In this study, less than 10% of all 171 plumes (for BTEX, MTBE and ETBE) using the primary threshold were greater than 500 m. Similarly, in the US, plumes greater than 500 m are rarely observed. The results from this study included one or two outliers with unusually long plumes, though a determination could not be made whether this was a random difference or reflects a true difference between conditions in the US vs. Europe.

In comparison to the US studies, the European study is based on data for a relatively small number of plumes (23 MTBE plumes; 39 benzene plume). This increases the potential for random bias, for example, if reported MTBE and benzene plumes are from sites with different geology. The potential significance of such effects was assessed by comparing benzene and MTBE plume lengths at sites where both were present (and therefore migrating under similar ground conditions). In this subgroup of 16 sites the median benzene plume length (32.5 m) was just shorter than the MTBE plume length (36.2 m), whereas for the full population of plumes the median length was 53 m for benzene and 38 m for MTBE (according to the primary delineation

threshold). These differences suggest that geological bias could be a factor and that median plume lengths in the US and Europe may be not be significantly different. Additional data from European sites would be needed to confirm this.

Additionally, at the majority of sites in the database (80% in the US, and 82% in Europe), benzene plume are either shrinking or stable. Similarly, for MTBE, the majority of sites in the database (80% in the US, and 86% in Europe) are also either shrinking or stable.

Appendix B summarizes the use of MTBE and ETBE in gasoline in the US as well as European countries. Overall, the average concentration in all European gasoline samples in 2000-2009 was 5.4 m/m% for MTBE and 0.9 m/m% for ETBE (Concawe, 2012).

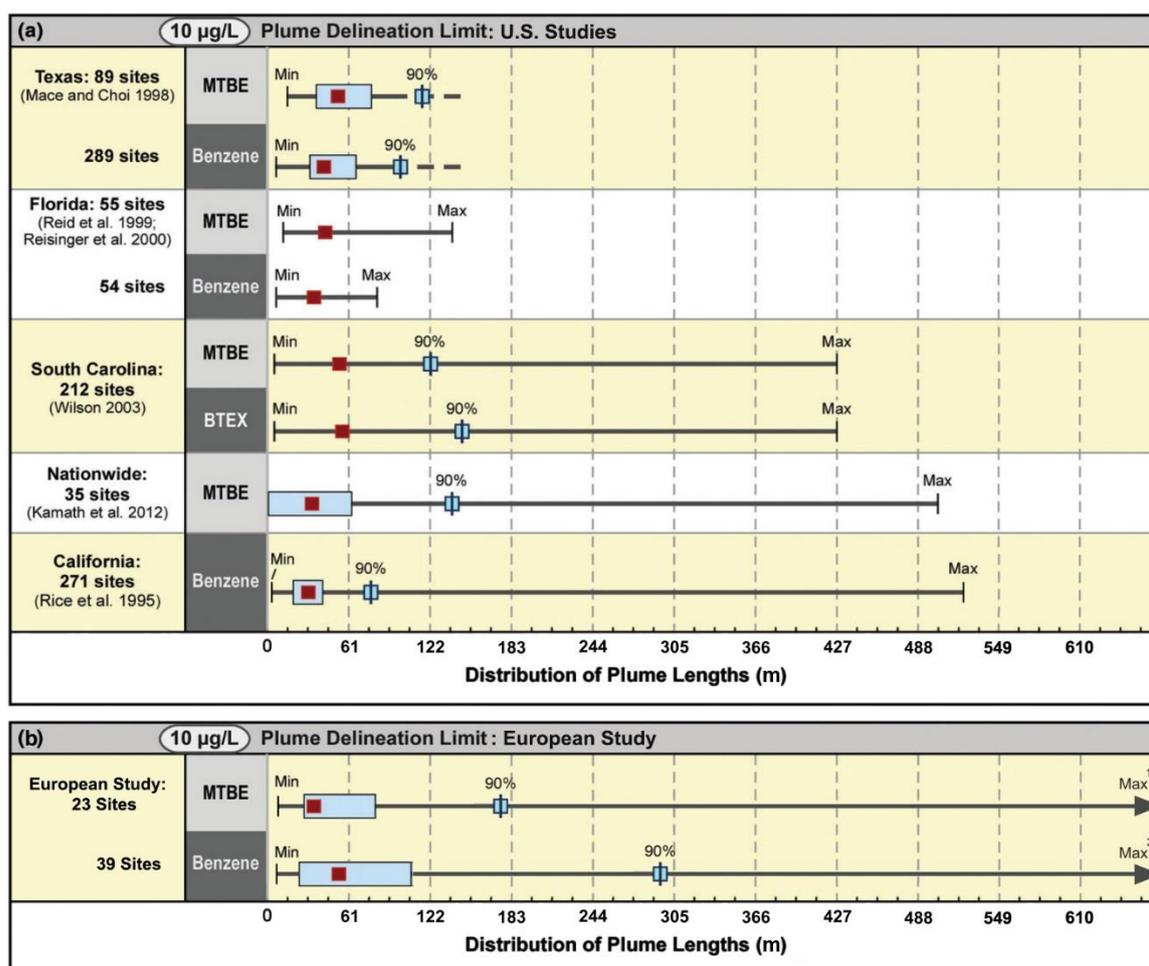
In the US, typical concentrations ranged from 5% to 10% by volume, although they can be up to 17% (ACGIH, 2002). Use of MTBE in the US was phased out in the early 2000s and largely ended by 2006.

Table 10: Comparison of Methodology and Results of US and European Studies

	US Studies	This Study of European Sites
Methodology	<ul style="list-style-type: none"> Plume lengths evaluated for Benzene and MTBE Length defined per hand-drawn contours to downgradient clean well No level of confidence analysis Extrapolated bulk attenuation rate for some plume lengths with insufficient info. Plume stability analysis using plume lengths vs. time as well as concentration vs. time analysis in individual downgradient well (Connor et al., 2014) 	<ul style="list-style-type: none"> Plume lengths evaluated for BTEX, MTBE and ETBE Length defined per hand-drawn contours to plume downgradient boundary with a clean well Length analysis categorized as: high, medium, low confidence Extrapolated bulk attenuation rate to define some plumes with insufficient info. Plume stability analysis using 1 to 2 historical datasets for benzene and MTBE and comparison of plume lengths vs. time.
Median Plume Length Using 10 µg/L Threshold (m)		
Benzene (n=no. of sites)	43 m (140 ft) (n=826) (Connor et al., 2014)	53 m (n=39)
Toluene (n=no. of sites)	--	43 m (n=33)
Ethylbenzene (n=no. of sites)	--	47 m (n=35)
Xylenes (n=no. of sites)	--	58 m (n=33)
MTBE (n=no. of sites)	50 m (165 ft) (n=391) (Connor et al., 2014)	38 m (n=23)
ETBE (n=no. of sites)	--	37 m (n=8)
Plume Stability		
Benzene	Increasing – 6% Stable – 48% Decreasing – 32% Detached – 0% (Connor et al., 2014)	Increasing – 17% Stable – 43% Decreasing – 39% Detached – 0%
MTBE	Increasing - 6% Stable - 20% Decreasing - 60% Detached – 2% (Connor et. al., 2014)	Increasing – 7% Stable – 43% Decreasing – 43% Detached – 7%

Note: (--) indicates data not available.

Figure 10: Comparison of Plume Lengths from US Studies (a) and this European Study (b) for a 10 µg/L Delineation Threshold (adapted from Connor et al., 2014). Notes (1) red squares indicate median value; (2) maximum plume length for MTBE in the European Study was 2400 m; (3) maximum plume length for benzene in European Study was 1800m.



Key Points

- The median and upper end plume lengths for benzene and MTBE, as well as plume stability analysis results indicate that hydrocarbon plumes in the US and Europe both exhibit similar characteristics.
- At the majority of sites in the database (80% in the US, and 82% in Europe), benzene plume are either shrinking or stable. Similarly, for MTBE, the majority of sites in the database (80% in the US, and 86% in Europe) are also either shrinking or stable.

4. CONCLUSIONS

A plumeathon study of sites impacted by the petroleum hydrocarbons BTEX, and fuel oxygenates MTBE and ETBE across Europe was done and consisted of an evaluation of 50 sites and analysis of BTEX, MTBE and ETBE plumes, respectively. Plume length analysis was conducted for a total of 171 plumes using the primary threshold and 138 plumes using the secondary threshold. Additionally, plume stability analysis was conducted for benzene and MTBE at 29 sites. Key site characteristics and an evaluation of biodegradation were also tabulated for each site.

Key results of the analysis indicated:

- Median reported hydraulic conductivity of a sub-set of the sites (n=27) was 3.2 m/day, and the most common soil type in both the vadose and saturated zones are sand and sand/silt.
- Soil and groundwater remediation information was available at 38 and 40 sites respectively. The principal remediation technologies employed were excavation (soil) and pump and treat (groundwater).
- The median plume lengths for BTEX compounds were similar using the primary threshold, and ranged from 43 to 58 m. Median plume lengths for MTBE and ETBE using the primary threshold were 38 and 37 m, respectively.
- The median plume lengths for BTEX compounds using the secondary threshold ranged from 15 to 53 m. The median plume length for MTBE and ETBE using the secondary threshold were 34 and 37 m, respectively.
- Benzene plumes at high concentration sites were found to be longer than those at low concentration sites ($p = 0.0013$). A similar magnitude difference was found for MTBE, but this was not statistically significant at a 95% confidence level ($p = 0.086$).
- Aside from source concentration, plume length was not found to be correlated with any other site characteristics (e.g., soil type, any soil and/or groundwater remediation techniques applied, and hydraulic conductivity).
- For sites having sufficient data to assess redox conditions in background wells as compared to wells within plumes, a majority of the sites evaluated (13 out of 16) showed evidence of biodegradation.
- Comparing the results from this study to a meta-analysis of US plume studies (Connor et al., 2014) indicates:
 - The median plume lengths for benzene and MTBE observed in this study were similar to those reported in the US
 - At the majority of sites (80% in the US, 82% in this study), benzene plume are either shrinking or stable. Similarly, for MTBE, the majority of sites (80% in the US, 86% in this study) are either shrinking or stable. At the majority of sites (80% in the US, and 82% in this study), benzene plume are either shrinking or stable. Similarly, for MTBE, the majority of sites (80% in the US, and 86% in this study) are also either shrinking or stable. The level of confidence in benzene and MTBE plume stability analyses was predominantly high or medium.²
 - This study's relatively small dataset could increase random bias, explaining some discrepancy between this study and US studies.

² High confidence being ≥ 1 historical plume maps with fully delineated plume lengths with > 5 years between first and most current map; Medium confidence being ≥ 1 historical plume maps with either fully delineated plume lengths with 0-5 years between first and most current map, or with estimated plume length with > 5 years between first and most current map).

5. GLOSSARY

BTEX	Benzene, toluene, ethyl benzene, xylenes
GSI	GSI Environmental Inc.
ETBE	Ethyl tert-butyl ether
Kd	Site-specific partitioning coefficient
LNAPL	Light non-aqueous phase liquid
MNA	Monitored natural attenuation
MTBE	Methyl tert-butyl ether
NSZD	Natural Source Zone Depletion
R	Site-specific retardation coefficient
TBA	Tertiary butyl alcohol
US	United States

6. ACKNOWLEDGEMENTS

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APPENDIX A: DATA TEMPLATE

Table 1A Data Template used in this study in order to get consistent data acquisition

Issued: 1 August 2016				
A. SITE INFORMATION	DATA NEEDED	RESPONSE TYPE	NOTES	DATA SOURCE (Report, Page, Table/Figure)
1.	Database Site ID	Text (e.g., UK-001)		
2.	Facility Name	Text		
3.	Address, City	Text		
4.	Country	Text		
5.	Language of Site Reports	Text		
6.	Is site an active petroleum facility?	Dropdown (Yes / No)		
7.	If No. 5 is 'No', date taken out of service?	Date (DD/MM/YYYY)		

B. SITE ACTIVITY RECORD					
<i>Release Event</i>					
8.	Date of most recent release occurrence, or date before which release occurred (i.e., "Before 1986 if exact date unknown)		Date (DD/MM/YYYY)		
9.	Source material: (gasoline, diesel/mixed product, other (describe))		Dropdown (gasoline, diesel, mixed product, other(describe), n/a)		
<i>Affected Soils (i.e. Vadose Zone) Remediation</i>					
10.	Has soil remediation strategy been implemented at site?		Dropdown (Yes / No)		
11.	Soil treatment method: (i.e., Excavation, SVE, Other(describe))		Dropdown (Excavation, SVE, Other (describe), n/a)		
12.	If No. 10 is 'Yes', START date of treatment		Date (DD/MM/YYYY)		
13.	If No. 10 is 'Yes', END date of treatment		Date (DD/MM/YYYY)		

Affected Groundwater (i.e., Saturated Zone) Remediation					
14.	Has groundwater remediation system been installed?				
15.	If No. 11 is 'Yes', Groundwater remediation method (i.e. Pump and Treat, SVE, Air Sparging, Air Sparging w/SVE, Other (describe))		Dropdown (Yes / No) Dropdown (Pump and Treat, SVE, Air Sparging, Air Sparging w/SVE, Other (describe))		
16.	If No. 11 is 'Yes', START date of treatment		Date (DD/MM/YYYY)		
17.	If No. 11 is 'Yes', END date of treatment		Date (DD/MM/YYYY)		
18.	Has NAPL recovery effort been implemented?		Dropdown (Yes / No) Dropdown (skimming, bailing, dual-phase extraction, other (describe))		
19.	NAPL recovery method		Date (DD/MM/YYYY)		
20.	If No. 15 is 'Yes', START date of recovery		Date (DD/MM/YYYY)		
21.	If No. 15 is 'Yes', END date of recovery		Date (DD/MM/YYYY)		

C. SITE-SPECIFIC CONDITIONS	Presence of Affected Groundwater											
22.	Depth to top of affected water bearing unit (i.e., to top of aquifer, not water table) (m BGS):				Number (meters bgs)							
23.	Predominant vadose zone soil type: (e.g., clay, silt, sandstone)				Dropdown							
24.	Predominant saturated zone soil type: (e.g., clay, silt, sandstone)				Dropdown							
25.	If measured, organic fraction of predominant soil in vadose zone				Number							
26.	Principal groundwater flow direction in affected unit (N, S, SE, NW, etc.):				Dropdown (N,S,SE,NW, etc.)							
27.	Number of wells used in groundwater flow direction analysis				Dropdown							
28.	How many monitoring events used in groundwater flow direction analysis?				Number							
29.	Confidence (professional judgement) in groundwater flow direction				Dropdown							
30.	Aquifer porosity				Number							
31.	Source of reported aquifer porosity				Dropdown							
32.	Aquifer hydraulic conductivity (m/d)				Number (m/d)							
33.	Source of reported aquifer hydraulic conductivity				Dropdown							
34.	Groundwater flow velocity (m/y)				Number (m/y)							
35.	Source of reported groundwater flow velocity				Dropdown							

Presence of NAPL									
36.	Has the presence of free-phase hydrocarbons been noted?					Dropdown (Yes / No)			
37.	If free-phase hydrocarbons noted, in how many wells?					Number			
38.	Last date free-phase hydrocarbons noted:					Date (DD/MM/YYYY)			
39.	Approximate average LNAPL thicknesses observed in wells					Number (mm)			
	Attenuation Parameters								
40.	Mean annual air temperature at site location (°C, typically available from online weather resource)					Number (°C) from online weather resource			
41.	Microbiology information available?					Dropdown (Yes / No)			
42.	If No. 35 is 'Yes', Microbiology: colony counts					Number (cfu/mL)			
43.	If No. 35 is 'Yes', Microbiology: microbial assemblages					Text			
44.	If No. 35 is 'Yes', Microbiology: DNA/RNA					Text			
45.	Site-specific partitioning coefficient (Kd)					Number			
46.	Site-specific retardation coefficient (R value)					Number			
47.	Site-specific half-life value from field or microcosm data					Number (days)			
48.	Site-specific dispersivity value								
49.	Redox conditions in background: average oxygen concentration (mg/L)					Number (mg/L) from 1 upgradient well			

50.	Redox conditions in background: average nitrate concentration (mg/L)			Number (mg/L) from 1 upgradient well		
51.	Redox conditions in background: average methane concentration (mg/L)			Number (mg/L) from 1 upgradient well		
52.	Redox conditions in background: average sulfate concentration (mg/L)			Number (mg/L) from 1 upgradient well		
53.	Redox conditions in background: average iron concentration (mg/L)			Number (mg/L) from 1 upgradient well		
54.	Redox conditions in background: average manganese concentration (mg/L)			Number (mg/L) from 1 upgradient well		
55.	Redox conditions in source: average oxygen concentration (mg/L)			Number (mg/L) from 1 well on downgradient edge of source area		
56.	Redox conditions in source: average nitrate concentration (mg/L)			Number (mg/L) from 1 well on downgradient edge of source area		
57.	Redox conditions in source: average methane concentration (mg/L)			Number (mg/L) from 1 well on downgradient edge of source area		
58.	Redox conditions in source: average sulfate concentration (mg/L)			Number (mg/L) from 1 well on downgradient edge of source area		
59.	Redox conditions in source: average iron concentration (mg/L)			Number (mg/L) from 1 well on downgradient edge of source area		
60.	Redox conditions in source: average manganese concentration (mg/L)			Number (mg/L) from 1 well on downgradient edge of source area		
61.	Is there evidence of degradation?			Dropdown		

<i>Preferential Pathways</i>							
62.	Does man-made preferential pathway affect plume migration?				Dropdown (Yes / No)		
63.	If No, is 'Yes', then what type? (e.g., gravel-filled service trenches, subsurface conduits, other (describe))				Text (gravel-filled service trenches, subsurface conduits, etc.)		
D. CALCULATED PLUME LENGTH INFORMATION (PRIMARY DELINEATION THRESHOLD)							
<i>Benzene (10 ppb threshold)</i>							
64.	Date of most recent plume map				Date (DD/MM/YYYY)		
65.	Plume delineation status (e.g., fully delineated, estimated)				Dropdown (fully delineated, estimated)		
66.	Benzene plume length (m):				Number (m)		
67.	Plume length estimation method (e.g., isocontour map, interpolation, extrapolation, report text)				Dropdown (Isocontour map, interpolation, extrapolation, report text)		
68.	Confidence in plume length (low, medium, high)				Text (low, medium, high)		
<i>Toluene (10 ppb threshold)</i>							
69.	Date of most recent plume map				Date (DD/MM/YYYY)		
70.	Plume delineation status (e.g., fully delineated, estimated)				Dropdown (fully delineated, estimated)		

71.	Toluene plume length (m):			Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
72.	Plume length estimation method (e.g., isocontour map, interpolation, extrapolation, report text)			Text (low, medium, high)		
73.	Confidence in plume length (low, medium, high)					
Ethylbenzene (10 ppb threshold)						
74.	Date of most recent plume map			Date (DD/MM/YYYY)		
75.	Plume delineation status (e.g., fully delineated, estimated)			Dropdown (fully delineated, estimated)		
76.	Ethylbenzene plume length (m):			Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
77.	Plume length estimation method (e.g., isocontour map, interpolation, extrapolation, report text)			Text (low, medium, high)		
78.	Confidence in plume length (low, medium, high)					
Xylenes (100 ppb threshold)						
79.	Date of most recent plume map			Date (DD/MM/YYYY)		
80.	Plume delineation status (e.g., fully delineated, estimated)			Dropdown (fully delineated, estimated)		
81.	Xylenes plume length (m):			Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
82.	Plume length estimation method (e.g., isocontour map, interpolation, extrapolation, report text)					

83.		Confidence in plume length (low, medium, high)		Text (low, medium, high)		
MTBE (10 ppb threshold)						
84.		Date of most recent plume map		Date (DD/MM/YYYY)		
85.		Plume delineation status (e.g., fully delineated, estimated)		Dropdown (fully delineated, estimated)		
86.		MTBE plume length (m):		Number (m)		
87.		Plume length estimation method (e.g., isocountour map, interpolation, extrapolation, report text)		Dropdown (isocountour map, interpolation, extrapolation, report text)		
88.		Confidence in plume length (low, medium, high)		Text (low, medium, high)		
ETBE (10 ppb threshold)						
89.		Date of most recent plume map		Date (DD/MM/YYYY)		
90.		Plume delineation status (e.g., fully delineated, estimated)		Dropdown (fully delineated, estimated)		
91.		ETBE plume length (m):		Number (m)		
92.		Plume length estimation method (e.g., isocountour map, interpolation, extrapolation, report text)		Dropdown (isocountour map, interpolation, extrapolation, report text)		
93.		Confidence in plume length (low, medium, high)		Text (low, medium, high)		

E. CALCULATED PLUME LENGTH INFORMATION (SECONDARY DELINEATION THRESHOLD)										
Benzene (10 ppb threshold)										
94.	Date of most recent plume map							Date (DD/MM/YYYY)		
95.	Plume delineation status (e.g., fully delineated, estimated)							Dropdown (fully delineated, estimated)		
96.	Benzene plume length (m):							Number (m)		
97.	Plume length estimation method (e.g., isocontour map, interpolation, extrapolation, report text)							Dropdown (Isocontour map, interpolation, extrapolation, report text)		
98.	Confidence in plume length (low, medium, high)							Text (low, medium, high)		
Toluene (700 ppb threshold)										
99.	Date of most recent plume map							Date (DD/MM/YYYY)		
100.	Plume delineation status (e.g., fully delineated, estimated)							Dropdown (fully delineated, estimated)		
101.	Toluene plume length (m):							Number (m)		
102.	Plume length estimation method (e.g., isocontour map, interpolation, extrapolation, report text)							Dropdown (Isocontour map, interpolation, extrapolation, report text)		
103.	Confidence in plume length (low, medium, high)							Text (low, medium, high)		

Ethylbenzene (300 ppb threshold)								
104.	Date of most recent plume map				Date (DD/MM/YYYY)			
105.	Plume delineation status (e.g., fully delineated, estimated)				Dropdown (fully delineated, estimated)			
106.	Ethylbenzene plume length (m):				Number (m)			
107.	Plume length estimation method (e.g., isocountour map, interpolation, extrapolation, report text)				Dropdown (isocountour map, interpolation, extrapolation, report text)			
108.	Confidence in plume length (low, medium, high)				Text (low, medium, high)			
Xylenes (500 ppb threshold)								
109.	Date of most recent plume map				Date (DD/MM/YYYY)			
110.	Plume delineation status (e.g., fully delineated, estimated)				Dropdown (fully delineated, estimated)			
111.	Xylenes plume length (m):				Number (m)			
112.	Plume length estimation method (e.g., isocountour map, interpolation, extrapolation, report text)				Dropdown (isocountour map, interpolation, extrapolation, report text)			
113.	Confidence in plume length (low, medium, high)				Text (low, medium, high)			
MTBE (15 ppb threshold)								
114.	Date of most recent plume map				Date (DD/MM/YYYY)			
115.	Plume delineation status (e.g., fully delineated, estimated)				Dropdown (fully delineated, estimated)			

116.	MTBE plume length (m):			Number (m) Dropdown (isocountour map, interpolation, extrapolation, report text)		
117.	Plume length estimation method (e.g., isocountour map, interpolation, extrapolation, report text)			Text (low, medium, high)		
118.	Confidence in plume length (low, medium, high)					
ETBE (10 ppb threshold)						
119.	Date of most recent plume map			Date (DD/MM/YYYY)		
120.	Plume delineation status (e.g., fully delineated, estimated)			Dropdown (fully delineated, estimated)		
121.	ETBE plume length (m):			Number (m) Dropdown		
122.	Plume length estimation method (e.g., isocountour map, interpolation, extrapolation, report text)			(isocountour map, interpolation, extrapolation, report text)		
123.	Confidence in plume length (low, medium, high)			Text (low, medium, high)		
F. PLUME STABILITY INFORMATION						
Benzene (10 ppb threshold)						
124.	Enough information to evaluate plume stability?			Dropdown (Yes / No)		
125.	How many historical plume delineation maps used in analysis?			Number		
126.	Date of Historical Plume 1 map			Date (DD/MM/YYYY)		
127.	Historical Plume 1 length (m)			Number (m)		

128.	Historical Plume 1 estimation method (e.g., isocountour map, interpolation, extrapolation, report text)		Dropdown (Isocountour map, interpolation, extrapolation, report text)		
129.	Date of Historical Plume 2 map		Date (DD/MM/YYYY)		
130.	Historical Plume 2 length (m)		Number (m) Dropdown		
131.	Historical Plume 2 estimation method (e.g., isocountour map, interpolation, extrapolation, report text)		Dropdown (Isocountour map, interpolation, extrapolation, report text)		
132.	Date of Historical Plume 3 map		Date (DD/MM/YYYY)		
133.	Historical Plume 3 length (m)		Number (m) Dropdown		
134.	Historical Plume 3 estimation method (e.g., isocountour map, interpolation, extrapolation, report text)		Dropdown (Isocountour map, interpolation, extrapolation, report text)		
135.	Plume stability (expanding, stable, shrinking, exhausted)		Dropdown (expanding, stable, shrinking, exhausted)		
136.	Confidence in plume status (low, medium, high)		Dropdown (low, medium, high)		
MTBE (10 ppb threshold)					
137.	Enough information to evaluate plume stability?		Dropdown (Yes / No)		
138.	How many historical plume delineation maps used in analysis?		Number		
139.	Date of Historical Plume 1 map		Date (DD/MM/YYYY)		
140.	Historical Plume 1 length (m)		Number (m)		

141.	Historical Plume 1 estimation method (e.g., isocountour map, interpolation, extrapolation, report text)	Dropdown (Isocountour map, interpolation, extrapolation, report text)			
142.	Date of Historical Plume 2 map	Date (DD/MM/YYYY)			
143.	Historical Plume 2 length (m)	Number (m)			
144.	Historical Plume 2 estimation method (e.g., isocountour map, interpolation, extrapolation, report text)	Dropdown (Isocountour map, interpolation, extrapolation, report text)			
145.	Date of Historical Plume 3 map	Date (DD/MM/YYYY)			
146.	Historical Plume 3 length (m)	Number (m)			
147.	Historical Plume 3 estimation method (e.g., isocountour map, interpolation, extrapolation, report text)	Dropdown (Isocountour map, interpolation, extrapolation, report text)			
148.	Plume stability (expanding, stable, shrinking, exhausted)	Dropdown (expanding, stable, shrinking, exhausted)			
149.	Confidence in plume status (low, medium, high)	Dropdown (low, medium, high)			
Other Constituent					
150.	Constituent	Text			
151.	Enough information to evaluate plume stability?	Dropdown (Yes / No)			
152.	How many historical plume delineation maps used in analysis?	Number			
153.	Primary delineation threshold (ppb)	Number (ppb)			
154.	Date of Historical Plume 1 map	Date (DD/MM/YYYY)			

155.	Historical Plume 1 length (m)								
156.	Historical Plume 1 estimation method (e.g., isocontour map, interpolation, extrapolation, report text)						Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
157.	Date of Historical Plume 2 map						Date (DD/MM/YYYY)		
158.	Historical Plume 2 length (m)						Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
159.	Historical Plume 2 estimation method (e.g., isocontour map, interpolation, extrapolation, report text)						Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
160.	Date of Historical Plume 3 map						Date (DD/MM/YYYY)		
161.	Historical Plume 3 length (m)						Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
162.	Historical Plume 3 estimation method (e.g., isocontour map, interpolation, extrapolation, report text)						Number (m) Dropdown (Isocontour map, interpolation, extrapolation, report text)		
163.	Plume stability (expanding, stable, shrinking, exhausted)						Dropdown (expanding, stable, shrinking, exhausted)		
164.	Confidence in plume status (low, medium, high)						Dropdown (low, medium, high)		
H. NOTES									
165.									
166.									
167.									

INSTRUCTIONS					Complete fields 1-164 per available data. If not available: insert "n/a"	
a)					Record Report, page number and table/figure of information found	
b)					For fields with "Other", please describe in Column NOTES	
c)						

APPENDIX B: USE OF MTBE AND ETBE IN GASOLINE

In Europe, ether oxygenates were used primarily to maintain the octane rating in the absence of lead. Italy began using MTBE in gasoline in 1979, and much of Europe followed suit by the mid 1980's (von Krauss and Harremoës 2002). MTBE was added to gasoline blends initially at low concentrations (2-5 wt-%), and later was added at higher concentrations (11-15 vol-%). The average concentration in all European gasoline samples in 2000-2009 was 5.4 m/m% for MTBE and 0.9 m/m% for ETBE (Concawe, 2012). Table 1 below summarizes the minimum to maximum % by mass for both MTBE and ETBE in each of the countries included in this study. Denmark was the first country to phase out the use of MTBE (Concawe, 2012). Other changes in the use of MTBE in Europe have been market driven.

ETBE was first used in France in 1992, and is widely used and manufactured today in most major gasoline markets in Europe. The ETBE concentration in Premium Unleaded Gasoline was high (5-12%) in France, Spain, Hungary and Austria (Concawe, 2012). The EU Biofuel Directive set requirements for the biofuel contribution in gasoline to be 2% in 2005, rising to 5.75% in 2010, and may even rise to 8% by 2020. As such, the production and use of ETBE will increase as compared to MTBE, as it is made from bio-ethanol and is considered a biofuel (Concawe, 2012).

MTBE in the US was used almost exclusively as a blending component or octane-enhancing agent in gasoline, with typical concentrations ranging from 5% to 10% by volume, although they can be up to 17% (ACGIH, 2002). MTBE usage in the United States began in 1979 and became increasingly prominent in the 1990's due to the 1990 amendment to the Clean Air Act. By the late 1990's, concerns about MTBE in drinking water had been raised. Use of MTBE in the US was phased out in the early 2000s and largely ended by 2006.

Table 1B Concentration Ranges of MTBE and ETBE in Gasolines in Various Countries in Europe, 2000-2009 (Concawe 2012)

Country	Additive (wt%) 2000-2009
Range MTBE	
	Range
Belgium	1-12%
Denmark	0.1-11%
France	0.1-5%
Germany	0.2-12%
Hungary	0.3-14%
Italy	2-14%
Netherlands	0.3-12%
Spain	0.3-9%
United Kingdom	0.2-3%
Range ETBE	
Belgium	0-11%
Denmark	0%
France	0.3-14%
Germany	0-8%
Hungary	0-12%
Italy	0-5%
Netherlands	0-4%
Spain	0-12%
United Kingdom	0-0.1%

APPENDIX C: SUMMARY OF ALL PRIMARY AND SECONDARY PLUME LENGTHS BY CONFIDENCE INTERVAL

Table 1C Statistical Summary of Primary Threshold Plume Lengths, by Confidence Levels

	Benzene	Toluene	Ethylbenzene	Xylenes	MTBE	ETBE
High Confidence (plume lengths, meters)						
Minimum	7	6	4	10	27	--
5th Percentile	8	8	8	--	--	--
25th Percentile	16	12	18	--	--	--
Median	25	17	72	39	72	--
75th Percentile	79	56	82	--	--	--
95th Percentile	294	87	92	--	--	--
Maximum	561	97	98	81	277	--
Mean	83	35	56	44	102	--
Number of Sites	13	8	8	6	6	--
% of Sites	33%	24%	23%	19%	26%	--
Medium Confidence (plume lengths, meters)						
Minimum	6	8	7	14	7	23
5th Percentile	18	9	14	16	10	--
25th Percentile	35	22	25	27	23	--
Median	62	49	42	58	34	39
75th Percentile	142	95	87	203	68	--
95th Percentile	786	658	541	581	1074	--
Maximum	1795	1114	865	1384	2411	60
Mean	215	141	151	193	229	41
Number of Sites	21	18	21	21	13	5
% of Sites	54%	55%	60%	66%	57%	63%
Low Confidence (plume lengths, meters)						
Minimum	8	23	6	28	33	31
5th Percentile	--	23	--	--	--	--
25th Percentile	--	27	--	--	--	--
Median	53	83	56	100	60	35
75th Percentile	--	305	--	--	--	--
95th Percentile	--	727	--	--	--	--
Maximum	206	830	800	1052	85	38
Mean	73	228	171	289	59	35
Number of Sites	5	7	6	5	4	3
% of Sites	13%	21%	17%	16%	17%	38%

Table 2C Statistical Summary of Secondary Threshold Plume Lengths, by Confidence Levels

	Benzene	Toluene	Ethylbenzene	Xylenes	MTBE	ETBE
High Confidence (plume lengths, meters)						
Minimum	7	4	2	9	22	--
5th Percentile	8	5	5	--	--	--
25th Percentile	16	6	10	--	--	--
Median	22	17	14	14	51	--
75th Percentile	75	32	47	--	--	--
95th Percentile	298	59	74	--	--	--
Maximum	561	76	79	54	240	--
Mean	80	22	28	25	82	--
Number of Sites	12	9	10	6	6	--
% of Sites	32%	45%	42%	22%	27%	--
Medium Confidence (plume lengths, meters)						
Minimum	6	2	2	1	5	23
5th Percentile	18	2	5	2	9	--
25th Percentile	35	10	15	6	18	--
Median	62	13	48	17	29	39
75th Percentile	142	95	112	52	46	--
95th Percentile	786	524	475	413	1129	--
Maximum	1795	638	638	692	2292	60
Mean	215	127	129	83	230	41
Number of Sites	21	8	8	16	12	5
% of Sites	55%	40%	33%	59%	55%	63%
Low Confidence (plume lengths, meters)						
Minimum	8	12	9	13	33	31
5th Percentile	--	--	--	--	--	--
25th Percentile	--	--	--	--	--	--
Median	53	46	26	50	56	35
75th Percentile	--	--	--	--	--	--
95th Percentile	--	--	--	--	--	--
Maximum	206	341	519	652	69	38
Mean	73	133	106	204	54	35
Number of Sites	5	3	6	5	4	3
% of Sites	13%	15%	25%	19%	18%	38%

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