## CONCAWE Review

Volume 9 • Number 2 • October 2000



CONCAWE is the oil companies' European organization for environment, health and safety. The emphasis of its work lies on technical and economic studies relevant to oil refining, distribution and marketing in Europe. CONCAWE was established in 1963 in The Hague, and in 1990 its Secretariat was moved to Brussels.



Reproduction permitted with due acknowledgement.

Photographs: cover, ©Stock Market Photo Agency Inc.; pages 8 and 9, ©TotalFinaElf/Stolt Nielsen Inland Tanker Service B.V.

This publication was designed and produced by Words and Publications, Oxford, United Kingdom • http://www.words.co.uk

## Foreword

This edition of the *Review* covers a wide range of topics highlighting the diversity of CONCAWE's work in the areas of environment, health and safety.

The majority of the articles in this issue cover developments in the field of environmental protection, dealing with the control of marine vapour emissions, automotive particulate matters, the feasibility and implications of reducing the sulphur content of automotive fuels below 0.005 per cent and the handling of used lubricating oils. They also address the relevant EU legislation currently under consideration. In line with its overall objectives, CONCAWE attempts to provide objective technical facts and figures in order to provide EU decision-makers with a sound basis for preparing this legislation.

An essential part of CONCAWE's work is the assessment of HSE performance trends in the petroleum industry over the past years. One article in this edition highlights the continuing improvement in the safety performance of European refineries.

The last article presents CONCAWE's views on methods for testing the aquatic toxicity of petroleum substances.

We all live in an ever-changing world and CONCAWE cannot escape this trend. In the last *Review* I announced the arrival of Jean-François Larivé. Two further staff changes will now take place. Peter Heinze, Technical Coordinator for Automotive Emissions, will retire at the end of the year. Those of you who know Peter and have worked together with him will appreciate the important role Peter has played in the Secretariat. His dedication to the job and the massive workload he has undertaken on a multitude of issues within his portfolio have contributed effectively to CONCAWE's reputation and achievements. I am pleased to inform you that mean-while Neville Thompson joined the Secretariat in September and will take over Peter's position in December. I am very confident that Neville will continue Peter's successful work. CONCAWE's best wishes go both to the departing and to the new Technical Coordinator.

Finally, I have to announce my own retirement from CONCAWE by the end of the year. I must say that I have enjoyed my second assignment to CONCAWE enormously. The cooperative atmosphere, not only within the Secretariat but also in the relations with colleagues inside and outside the petroleum industry, has helped greatly to deal with a number of difficult issues. I am pleased to inform you that Jean Castelein will take over my functions from December. I am convinced that Jean will be successful in maintaining CONCAWE's mission of generating and analysing data based on technical integrity and objectivity. On behalf of CONCAWE, I wish Jean every success in the future.



Jochen Brandt Secretary-General, CONCAWE

## Contents

#### 5 Emission control at marine terminals *Results of recent CONCAWE studies*

#### Gasoline loading

#### Cost-effectiveness of emission controls for ship loading.

According to IIASA data VOC control on gasoline ship loading would be less cost-effective than many alternative VOC control measures. For the Gothenburg commitment level, these data indicate that vapour recovery at marine loading terminals would not be justified on cost-effectiveness grounds. At the original NEC target proposed by the European Commission, justification would be only marginal even for the very largest terminals.

#### **Ballasting**

#### Emission controls for loading ballast water at gasoline off-loading terminals

Emission control from ballasting operations would be five to fifty times more costly than the most costly alternative VOC control measures in the IIASA database. Installation of vapour recovery systems for ballasting operations is clearly not justified.

Enquiries to: Henk Schipper, CONCAWE

#### 10

#### Regional ozone in Europe Would further VOC control measures be effective?

The influence of anthropogenic VOCs on regional ozone levels is set to decrease dramatically by 2010 as a result of already agreed measures. The actual level of biogenic VOCs is uncertain and could be much higher than originally accounted for, further reducing the impact of anthropogenic sources. New measures to further reduce the latter would therefore not be effective.

Enquiries to: Henk Schipper, CONCAWE

#### 13

#### Automotive particulate matter *The ongoing search for basic knowledge*

With a recent study on particulate emissions from HD diesel engines, CONCAWE has completed a scoping exercise on automotive particles. Whereas the larger, so-called accumulation particles can be adequately characterized and measured, the nucleation mode mechanisms leading to the formation of nano-particles are not well understood and need further research.

Enquiries to: Peter Heinze or Neville Thompson, CONCAWE

### **16** Is a 10 ppm sulphur limit on road fuels desirable?

Although reducing diesel and gasoline sulphur content to less than 10 ppm is technically feasible, the desirability of such a move is far from proven. Beside the significant cost to the refining industry, the truly global effect on both greenhouse gases and air pollutants emissions is neither completely understood nor documented. In this article we highlight CONCAWE's position on the issue in a summary of CONCAWE's response to the EU Commission's consultation on the subject.

Enquiries to: Jean-François Larivé, Peter Heinze or Neville Thompson, CONCAWE

#### 21

#### Disposing of used lubricating oils

As the EU Commission is considering the preparation of a revised Directive for the disposal of used lubricants, this article revisits the conclusions of a previous CONCAWE study and compares them with statistics produced by the EU. Re-refining and burning are the two main disposal routes but, in the opinion of CONCAWE, neither has a clear advantage from an environmental point of view. The key action is to encourage collection; the optimum disposal route will then depend on specific circumstances.

Enquiries to: Eric Martin, CONCAWE

#### 24

#### Downstream oil industry safety statistics Industry safety performance continues to improve

CONCAWE has been collecting downstream oil industry safety statistics since 1993. 1999 shows an overall improvement and particularly a decrease in fatalities.

Enquiries to: Eric Martin, CONCAWE

#### 25

#### Aquatic toxicity testing for petroleum substances

Testing the aquatic toxicity of low-solubility substances such as petroleum fractions is no trivial matter. Amongst the different methods that have been proposed, CONCAWE strongly recommends the Water Accommodated Fraction (WAF) method and supports its universal adoption within the EU.

Enquiries to: Bo Dmytrasz, CONCAWE

27 CONCAWE publications Dear Review Reader,

In this issue of the *Review*, we have introduced a few changes. In the contents section we have included a short outline of each article to help you quickly find out the main issues discussed therein. For each article we also indicate a focal point, usually one of CONCAWE's Secretariat permanent staff, to whom comments or enquiries concerning the specific article can be addressed. Finally we have updated the Secretariat staff information and extended it to include individual e-mail addresses. We welcome your opinion on these changes or on any other issues related to the *Review*.

Wishing you good reading, as well as a happy Christmas and a prosperous New Year.

Jean-François Larivé, Editor

Website and general e-mail:	http://www.concawe.be	info@concawe.be
Secretary-General	Jochen Brandt Jean Castelein (from 1.12.00)	jochen.brandt@concawe.be jean.castelein@concawe.be
Technical Coordinators:		
Petroleum products	Bo Dmytrasz	bo.dmytrasz@concawe.be
Automotive emissions	Peter Heinze Neville Thompson	peter heinze@concawe.be neville.thompson@concawe.be
Safety management, water and soil protection and waste management, oil pipelines	Eric Martin	eric.martin@concawe.be
Air quality	Henk Schipper	henk.schipper@concawe.be
Health	Jan Urbanus	jan.urbanus@concawe .be
Refinery planning and publications	Jean-François Larivé	jeanfrancois.larive@concawe.be
Administration	Martien Sijbrandij	martien.sijbrandij@concawe.be
Documentation and Library	Annemie Hermans	annemie.hermans@concawe.be
Secretariat	Laurence Evrard Sandrine Faucq Elfriede Geuns Barbara Salter	laurence.evrard@concawe.be sandrine.faucq@concawe.be elfriede.geuns@concawe.be barbara.salter@concawe.be

#### CONCAWE CONTACTS AND SECRETARIAT STAFF



From left to right: Eric Martin, Jan Urbanus, Barbara Salter, Martien Sijbrandij, Elfriede Geuns, Jochen Brandt, Sandrine Faucq, Henk Schipper, Laurence Evrard, Bo Dmytrasz, Annemie Hermans, Jean-François Larivé, Peter Heinze

## Emission control at marine terminals

Results of recent CONCAWE studies

#### BACKGROUND

The European 'Stage 1' Directive 94/63/EC on the control of volatile organic compound (VOC) emissions mandates the installation of vapour emission controls for automotive gasoline storage and loading of road and rail tankers and inland waterway barges. Ship loading emission controls were not included as the EU Commission was awaiting the revision of the International Maritime Organization (IMO) MARPOL Convention to include a new Annex VI on the prevention of air pollution from ships.

Although IMO approved the new Annex in September 1997, it will not come into force until 12 months after ratification by 15 Port States with a combined merchant fleet of not less than 50 per cent of the worldwide gross tonnage.

As there is likely to be a lengthy period before ratification of the new Annex and, due to the non-prescriptive nature of the IMO regulation on VOCs, it is likely that the EU will proceed to develop new legislation on emission controls for ship loading of volatile cargoes. A study sponsored by the Commission is already assessing the economic and technical feasibility of possible measures to reduce emissions of VOCs during the loading and unloading of sea-going ships in Community ports.

CONCAWE Report 92/52 reviewed the costs and cost-effectiveness of installing vapour emission controls for the loading of gasoline onto ships and barges. Due to the small number of project studies at that time, CONCAWE has subsequently undertaken an update of this study. Initial findings were reported in the CONCAWE *Review* in October 1998.

Since then two further studies have been undertaken: gasoline loading in sea-going ships and emissions from ship ballasting at gasoline off-loading terminals. Both studies are discussed on the following pages of this article.

Safety concerns highlighted in CONCAWE Report 92/52 have been addressed in the new MARPOL Annex VI which references IMO publication MSC circular 585 'Standards for vapour emission control systems'.

Continued ...

### Gasoline loading

Cost-effectiveness of emission controls for ship loading

#### **INVESTMENT COSTS**

The initial CONCAWE study published in 1998<sup>1</sup> used the cost data from 20 vapour emission control systems, which had either been installed for gasoline loading in the USA or for benzene loading in Europe, or from project studies. It established that:

- The costs of installing a vapour emissions control system for loading gasoline onto sea-going vessels vary significantly at sites with similar loading rates because of site-specific issues—reported costs for sites with loading rates typical of a large refinery ranged from 4 to 20 million Euros.
- Vapour collection piping will have to be installed on board about 600 sea-going vessels of less than 40 000 dead weight tonnes (DWT) to permit trading at terminals fitted with shore-side vapour emission control systems—the total retrofit costs for these vessels are estimated at 151 million Euros.

The second phase of the CONCAWE study obtained data on 64 terminals in the EU-15 identified as loading gasoline into sea-going ships. These terminals loaded a total of 47.2 million tonnes of gasoline in 1998, representing 32 per cent of the EU-15 gasoline production. The terminals include both refineries and depots where gasoline is imported, stored and onward distributed by ship.

The size of these terminals, the estimated investment cost for installing vapour emission controls and the cost effectiveness of the VOC emissions reduction are shown in Table 1.

Throughput gasoline (kt/a)	Number of gasoline terminals (cumulative)	% of total gasoline loaded (cumulative)	Cumulative cost, million EUR (shoreside facilities only)	Cost-effectiveness kEUR/t abated * (shore and ship)
>3000	I	10.2	8	3.3
>2000	6	37.9	38	3.7
>1000	13	56.5	75	5.0
>750	23	75.1	123	5.9
>500	33	87.7	168	7.0
>250	44	96.6	217	9.2
>100	52	99.3	251	14.5
>0	64	100.0	300	32.2

\* The cost-effectiveness relates to the throughput range only, e.g. 32.2 kEUR/t relates to the range 0–100 kt/a. All other columns are cumulative.

The total cost of vapour collection and emissions control during ship loading of gasoline at the 64 terminals identified would be 451 million Euros, being the sum of the on-board ship costs of 151 million Euros and the shore-side costs of 300 million Euros.

Table 1

<sup>&</sup>lt;sup>1</sup> CONCAWE Review October 1998

#### UNCONTROLLED EMISSIONS

The uncontrolled emissions during gasoline loading can be calculated using an emission factor published by the US EPA of 0.034 per cent by volume loaded and a density of 0.6 kg/l for condensed gasoline vapour. Thus for a total of 47.2 million tonnes per year of gasoline loaded, the annual uncontrolled emissions are equal to 13 190 tonnes. This can be compared to the total annual man-made VOC emissions in the EU-15 of 9.7 million tonnes in 2000<sup>2</sup>.

The emissions from ship loading with gasoline are approximately 0.14 per cent of the total annual man-made VOC emissions in the EU-15 in 2000.

#### **COST-EFFECTIVENESS**

The cost-effectiveness of both on-board and shore-side investments can be calculated from the achievable emission reduction, the annualized cost of the capital investment and the operating and maintenance costs. Using an annual capital charge of 15 per cent, and operating and maintenance costs for on-shore systems of 5 per cent and 2 per cent of capital respectively:

## the overall cost-effectiveness of vapour emission controls on ship loading of gasoline would range from 3300 to more than 32 000 EUR/t, with effectiveness reducing as terminal throughput decreases.

These costs need to be seen in the perspective of other available control measures. Using the example of France, Figure 1 was developed using the IIASA-VOC<sup>3</sup> cost curve from their RAINS<sup>4</sup> model. Here, some eighty control measures are ranked from lowest to highest cost per tonne of VOC abated. Each individual measure is shown as an open blue bar. The width of each bar corresponds to the emission reduction achieved by that measure. Although France is shown here, the IIASA cost curves for other EU countries are similar.

Figure 1 France cost curve for VOC controls from 'No Controls Post 1990' emission levels (Source: IIASA)

To provide for ready comparison, the costs of marine vapour recovery for gasoline loading are shown in Figure 1 in three ranges for terminals with a throughput greater than 1 Mt/a, between 0.25 and 1 Mt/a and less than 0.25 Mt/a respectively.

The emission ceiling target proposed for France in the National Emission Ceilings Directive (NEC) is shown along with France's commitment under the UN-ECE Gothenburg Protocol signed in 1999 (The original EU base case or 'Reference' scenario is also indicated in the graph).



It is clear from Figure 1 that, at the Gothenburg Protocol target level, marine vapour recovery would not be justified, at least on cost-effectiveness grounds. At the original NEC target level proposed by the European Commission, justification would be marginal even for the largest terminals. Control at smaller terminals is clearly much less cost-effective than many other VOC control measures still to be implemented. CONCAWE's analysis also included IIASA data for Italy, Spain and the UK. Conclusions were similar to those for France given above.

<sup>2</sup> Senco 1999; Commission's Second Auto/Oil Programme <sup>3</sup> International Institute for Applied Systems Analysis

<sup>&</sup>lt;sup>4</sup> Regional Air Pollution Information and Simulation model

### Ballasting

#### *Emission controls for loading ballast water at gasoline off-loading terminals*

As mentioned on page 5, the Commission is sponsoring a study that aims to assess measures to reduce emissions both during the loading and the unloading of sea-going ships. The emissions that arise from shore-side gasoline tankage during a ship unloading operation are already covered in the Stage 1 Directive.

Another source is from loading ballast water into the cargo tanks of ships after they have been off-loaded. When a sea-going tanker discharges gasoline at a product off-loading terminal it must take on ballast water to achieve a seaworthy condition for the subsequent voyage. The amount of ballast water taken on board for this purpose varies between 15 per cent and 30 per cent of the total vessel capacity.

Ballast water can be taken into:

- a) segregated ballast tanks (SBTs)—these are tanks dedicated to carry ballast within the cargo spaces of the ship or the double hull spaces (double bottom and wing tanks); or
- b) cargo tanks which have previously held cargo.



Tanks of type 'a' are used solely for carrying ballast, so there are no hydrocarbon emissions when these are being filled. When ballast water is put into a cargo tank of type 'b', however, it will displace any hydrocarbon vapours remaining from a previously held volatile cargo.

IMO regulations state that sea-going vessels of more than 30 000 DWT ordered after 1976 and all others greater than 600 DWT ordered after 1993 shall have dedicated tanks or hull spaces which can carry ballast water. Loading data from seven terminals indicate that less than 20 per cent of volatile products were loaded into non-SBT tankers in 1999. Additionally a study by a major EU refinery showed a gradual downward trend in the use of non-SBT tankers from 45 per cent in 1993 to 13 per cent in 1999.

In any event, emissions due to ballasting will be eliminated in time as older ships that ballast into cargo tanks are either removed from volatile cargo service or decommissioned and replaced with more modern tankers with dedicated ballast water tanks conforming with IMO regulations.

#### UNCONTROLLED EMISSIONS

For a total of 47.2 Mt/a of gasoline loaded into ships in the EU-15, the emissions due to ballasting can be calculated using the following assumptions:

- a) The percentage of total gasoline carried in non-SBT tankers is 20 per cent.
- b) The amount of ballast water loaded averages 25 per cent of ship capacity. Taking the worst

case (i.e. that ballast is always pumped into tanks that previously held gasoline and never into tanks that held nonvolatile products such as automotive diesel fuel) this is equal to 3.2 million  $m^3/a$  of ballast water being loaded into cargo tanks that have previously held gasoline.

The US EPA quote a factor for crude oil ballasting of 0.111 kg of emissions per m<sup>3</sup> of ballast water when the ship had been previously fully loaded. However, there is no



published emission factor for the ballasting of product carriers, although measurements undertaken by a major oil company gave a factor of  $0.1 \text{ kg/m}^3$  ballast. Using this factor and data above:

The total annual emissions due to ballasting are estimated to be 320 tonnes, this is equivalent to 0.003 per cent of the total man-made VOC emissions in the EU-15 in 2000.

#### SHORE-SIDE INVESTMENT

The rate at which ballast water is pumped into cargo tanks depends on the size of the ship and can range from 500 to 1000 m<sup>3</sup>/h. Distribution terminals are served by ships of varying sizes, and thus in the worse case would have to handle vapours emitted at 1000 m<sup>3</sup>/h during ballasting operations, although this occurrence might be rare. For the purposes of this study it has been assumed that all ballasting emission control systems, irrespective of the volume of gasoline off-loaded per year, are designed for a ballasting rate of 750 m<sup>3</sup>/h.

The average cost of installing an emission control system for a vapour flow rate of 750 m<sup>3</sup>/h is about 3 million Euros.

For Marine operations there is currently no requirement to provide vapour recovery facilities at off-loading terminals, as there is no on-board release of hydrocarbon emissions during cargo discharge operations.

#### **COST-EFFECTIVENESS**

Using the same assumptions as for ship loading of gasoline, cost-effectiveness for vapour recovery during ballasting operations would range from 190 000 to 1 900 000 EUR/t of VOC recovered. Compared with the IIASA cost curve given in Figure 1, this indicates that such a requirement would be some five to fifty times more expensive per tonne of VOC controlled than the highest cost measure in the IIASA cost curve, and is therefore not justifiable on economic grounds.

## Regional ozone in Europe

#### Would further VOC control measures be effective?

Over the past three years, both the European Commission and the United Nations Economic Commission for Europe (UN-ECE) have developed response strategies to combat concerns over acidification, eutrophication and regional ozone. In each case 'Integrated Assessment Models' (IAM) were used to support policy development. In the case of the European Commission, this resulted in the National Emission Ceilings (NEC) Directive, which is currently under consideration by the European Institutions. In the case of the UN-ECE, this resulted in the multi-pollutant, multi-effects protocol, known as the Gothenburg Protocol, signed in December 1999.

In both cases the IAM was, in part, designed to identify the required reductions in  $NO_x$  and VOCs in each country to limit exceedances of an 8-hour average ozone target of 120 µg/m<sup>3</sup> (60 ppb)<sup>1</sup>. CONCAWE fully supports the concept of integrated assessment modelling as an important part of a rational process of environmental policy making. However, in this process, it is important to recognize the influence of uncertainties in the input data and key assumptions on model output. In the case of ozone, one such uncertainty, recognized within the scientific community<sup>2</sup>, is the magnitude of biogenic VOC emissions. In this article we explore the implications of these uncertainties on ozone reduction strategies. We shall see that in the 'future 2010 EU', where already agreed measures will have substantially reduced anthropogenic VOCs, further VOC controls offer little potential for improving ozone compliance. This is particularly so if biogenic VOC emissions are higher than currently represented in the EMEP model<sup>3</sup>.

RESULTS OF THE NERI (NATIONAL ENVIRONMENTAL RESEARCH INSITITUTE) DANISH EULEREAN MODEL (DEM) FOR THE 1995 BASE CASE EMISSIONS SCENARIO Number of days with ozone above 120µg/m3 (8-hour values) are shown on the



CONCAWE engaged the National Environmental Research Institute (NERI) of Denmark to study the potential influence of uncertainties in biogenic VOC emissions on predicted ozone levels in Europe. This institute has its own regional ozone modelling capability, the so-called Danish Eulerian Model (DEM). The results of this model were found to be in good agreement with the EMEP model results for the scenarios evaluated under the Commission's NEC work.

Figure 1 shows the results from the DEM for the 1995 Base Case emissions scenario with biogenic emission correlations consistent with those used in the EMEP model. Exceedance days above  $120\mu g/m^3$  (8-hour average) are shown on the EMEP grid scale of 150x150 km.

<sup>1</sup> In the case of the Commission's NEC Directive, this means limiting exceedances to 20 days a year, averaged over three successive years.

<sup>2</sup> Norwegian Meteorological Institute (NMI), Norway, Biogenic VOC Emissions in Europe, Parts I and II

<sup>3</sup> EMEP is the UN-ECE's European Monitoring and Evaluation Programme for Long-Range Transboundary Air Pollution. The EMEP MSC-W Ozone model is a Lagrangian trajectory model and was used to support both the Gothenburg Protocol and NEC Directives.

The exceedance days data for each EMEP grid along the horizontal (H) and vertical (V) transepts, for each biogenic VOC emission scenario, form the basis for the following discussion of the results from the NERI work.

#### IN A '2010 EU' THERE ARE VIRTUALLY NO 'VOC-CONTROLLED' AREAS

Figures 2 and 3 (horizontal and vertical transept respectively) show the exceedance days in 1995 and the predicted exceedance days in 2010 resulting from 'already-agreed measures' for  $NO_x$  and VOC control. The overlaid lines show the 2010 levels if only  $NO_x$  (blue) and only VOC (red) measures were implemented. These data clearly show the areas of Europe that are so-called VOC controlled (e.g. UK) and  $NO_x$  controlled (southern Europe).



In Figures 4 and 5, similar plots are given for emission reductions beyond already agreed measures (down to the so-called IIASA<sup>4</sup> 'Maximum Feasible Reductions' (MFR)). Here the picture changes rather dramatically with further  $NO_x$  measures offering a much more significant reduction potential than further VOC measures. This characteristic shift is evident for the whole EU area.



The very large reductions in VOC emissions that are anticipated to occur as a consequence of already agreed measures means that in a '2010 EU' a much higher proportion of residual VOC will be from biogenic sources. It is therefore not surprising to see a move away from the VOC and  $NO_x$ -controlled areas that characterized the 'mid-90's EU' to one that is largely  $NO_x$ -controlled in 2010. We shall now examine the impact of higher biogenic emissions on such a shift.

11

<sup>&</sup>lt;sup>4</sup> International Institute of Applied Systems Analysis, Luxembourg, Austria: this institute developed and maintains the Integrated Assessment Model 'RAINS' which underpins the Gothenburg Protocol and the NEC Directive.

#### WITH HIGH BIOGENIC VOC EMISSIONS, OZONE ABATEMENT POLICIES SHOULD FOCUS ON NO, CONTROL

The DEM model was used to evaluate the impact of a threefold increase in biogenic VOC emissions, well within the range of reported uncertainties. The results are given in Figures 6 through 9 below.

Figure 6 and 7 indicate that if biogenic VOC emissions are three times higher than currently represented in the EMEP model, then even the design of 'already agreed measures' may have over emphasized VOC control measures. The figures also clearly show that already agreed  $NO_x$  measures alone achieve essentially the same improvement as the combined effect of  $NO_x$  and VOC measures. This points to the potential for significant regret investments should the case for high biogenic emissions be confirmed.



Figures 8 and 9 clearly show that further measures aimed at further reducing ozone exceedances should be focussed on  $NO_x$  and that further VOC controls offer essentially no significant improvement.



These findings should be taken into account for setting future priorities within programmes like the Commission's soon-to-be launched CAFE initiative. They also bring a cautionary message to those involved in the finalization of the NEC Directive.

## Automotive particulate matter

#### The ongoing search for basic knowledge

Particulate matter (PM) in the air continues to be the focus of increased attention due to the concern of potential health effects. Accordingly, under the EU Air Quality Framework Directive an air quality standard has been defined with respect to  $PM_{10}$  (particulate with an aerodynamic diameter less than or equal to 10 nm) with a review planned in 2003.

There is concern that automotive tailpipe emissions, as one of the many emitting sources, may make a substantial contribution to ambient particulate concentrations especially within urban areas. Legislation is in place to measure and control the mass of automotive particulate emitted at the tailpipe and tighter limits have been progressively introduced over the years. While there is evidence that adverse health effects are associated with current ambient PM concentrations, it is, as yet, uncertain which feature of the particulate matter, be it chemical or physical, has the most relevance for health studies. Thus, attention has concentrated on the number-based size distribution of the ambient particles.

#### **GUIDANCE STILL REQUIRED ESPECIALLY FROM HEALTH SCIENCE**

At present there is no proven mechanism whereby low-level ambient PM could cause either early death or morbidity and, in terms of plausibility for increased mortality, there are no toxicology data to allow any conclusion.

Guidance from the health and atmospheric scientific community is still awaited on the particulate characteristics relevant for further abatement. Nevertheless it is important that different sectors of industry making significant contributions to ambient particle concentrations have a good understanding of the characterization of the particulate emitted. Automotive particulate emissions appear to have been studied most extensively and a wide range of information is now available.

#### SCOPING EXERCISE ON AUTOMOTIVE PARTICLES COMPLETED

CONCAWE embarked very early on the study of the automotive particulate emissions by mass, number and size. CONCAWE report 96/56 presented a thorough literature study to identify suitable analytical methodology for the measurement techniques of both mass and number distributions of the particles. This work was followed by a scoping exercise to develop an understanding in the area of automotive particulate emissions focusing on a range of light duty diesel and gasoline vehicle technologies and using wide range of marketed fuel specifications (CONCAWE report 98/51, SAE 982600). With a test programme on heavy-duty (HD) engines of two technology levels (Euro 2 and 3) and a fuel matrix matching that used for the light-duty (LD) diesel vehicle study as closely as possible, CONCAWE has now completed its scoping exercise in this field. Results from the heavy-duty study were published recently (SAE 2000–01–2000) and the full CONCAWE report is in its editorial stage. The major findings are reviewed below and put into the context of CONCAWE's current understanding on particle aspects.

#### THE HEAVY-DUTY ENGINE STUDY SHOWED THAT SAMPLING CONDITIONS STRONGLY INFLUENCE THE MEASURED DISTRIBUTION OF NUCLEATION MODE PARTICLES

The investigations of HD engine particulate sizing were based on experience and understanding gained from the earlier work as well as information obtained by contact with the scientific community researching in this area. Thus particle measurements were extended down to 3 nm and sampling effects were addressed as well. The wider range of particles (3–1000 nm) was covered using a Dual Differential Mobility Particle Spectrometer consisting of two analysers applied in parallel to sample simultaneously during the short scanning times available during the steady-state modes of the European HD engine test cycles.

As done in the LD work (and in the absence of any standardized sampling methodology for particle measurements) legislated test and sampling procedures for particulate mass were used for this HD engine study.

The study showed that sampling conditions, such as dilution ratio in the tunnel, residence time of the aerosol from the engine manifold to the inlet of the measurement device and stabilization time for sampling, strongly affect the particle size distribution and integrated total number of particles. The number distribution is bimodal with an accumulation mode (>30 nm) and a nucleation mode (<30 nm). While the accumulation mode particles gave consistent data, nucleation mode particles showed considerable variation with the change of sampling conditions (example given in Figure 1).

In this study only small differences between the tested fuels were observed within the accumulation mode, while larger differences between the fuels were observed in the nucleation mode (nanoparticles) (Figure 2). But such differences were not confirmed by the mass distribution data using a low-pressure cascade impactor.

Figure 1 Sampling conditions, such as dilution ratio and temperature, influence the particle distribution strongly in the nucleation mode (<30 nm), while accumulation mode particles (>30 nm) are only slightly affected. (Results from a Euro 2 engine at one mode condition.)



Figure 2 Differences in fuels were observed for nucleation mode particles which are strongly influenced by the sampling conditions (Figure 1). (ESC weighted averages with Euro 3 engine and individual test runs.)

From the foregoing it is concluded that there is no clear indication how to accurately measure nucleation mode particles. Particle measurements conducted in an atmospheric environment in other current research programmes will help to establish relationships between 'real world' particles and those measured under laboratory conditions. However, early 'real world' measurements also show that accumulation mode particles are quite insensitive to sampling and ambient conditions, while nucleation mode particle measurements are very sensitive, e.g. to ambient temperature.

#### NUCLEATION MODE PARTICLES NEED MORE RESEARCH

For accumulation mode particles the focus of further work should be on precision of size and number and/or mass measurement.

More research is required to better understand the basic generation mechanisms of nucleation mode particles. They are formed by condensation and therefore are observed to be very sensitive to sampling and ambient conditions. Reliable measurements do not seem to be achievable as yet. Further research will have to include engine design and operating conditions including specific fuel properties. Clarification is most urgently required on the contribution of nucleation mode particles to the ambient particulate matter and their relevance to human health.

#### NUCLEATION PROBLEM EXISTS WITH TRAPS AS WELL

Particulate traps can substantially reduce carbonaceous particulate (i.e. accumulation particles). However, drastically reducing the carbonaceous material will also remove the possibility of condensation of volatile particles. This would further increase the difficulties of

controlled sampling and measurement since both particle modes are at a different level of understanding. As observed in the CONCAWE study, the reduction of fuel sulphur would not necessarily solve the problem of nucleation of particles. Even with very low levels of sulphur (< 10 mg/kg) large numbers of particles can be produced if the engine is equipped with a highly effective oxidation catalyst in the exhaust. Such effects were observed when a Euro 3 HD engine was equipped with a CRT (Continuous Regenerating Trap) to meet Euro 4 emission limits (Figure 3).



PRODUCTION OF NUCLEATION MODE PARTICLES,

Figure 3 Particulate filter trap equipped Euro 3 engine adjusted to meet Euro 4 shows strong increase in nucleation mode particles even with a very low sulphur (<10ppm) containing fuel. Accumulation mode particles are completely trapped versus the Euro 3 setup without the trap. Data given for medium operating condition (ESC mode 4). (Sampling effect, see Figure 1.)

#### A PRAGMATIC APPROACH

The CONCAWE study provides further evidence of the complexity of the sampling and measurement of the full range of particles. In view of these results reported also by other workers and also observed under 'real world' conditions, it might be appropriate to consider the work on these issues in segments. There is currently a greater understanding of the practical handling of accumulation mode particles, while more research is needed to understand the complex nucleation processes and the resulting nucleation mode particles. Of course it has to be kept in mind that both areas of the automotive particle phenomenon have to be put into perspective with regard to other sources and their relevance to human health.

CONCAWE continues to be involved in the work on particle aspects as e.g. shown in its participation in the DETR/SMMT/CONCAWE PM Research Programme and in the PM Consortium managed by the European Commission's Directorate for Transport and Energy.

## Is a 10-ppm sulphur limit on road fuels desirable?

The German government proposal for the introduction of road fuels with less than 10 ppm sulphur issued at the end of last year re-launched the debate on the sulphur issue. In the meantime, the auto industry has been vocal in demanding 'ultra low sulphur' (ULS) fuels which it deems essential to meet its commitment on vehicle  $CO_2$  emissions. In May of this year, the EU Commission submitted a 'call for evidence' to all stakeholders to gather facts and opinions on the various implications of a move to sulphur levels of 30 and 10 ppm in EU road fuels. This article gives an overview of CONCAWE's response and reiterates the key messages.

#### **IS THERE A NEED FOR ULTRA LOW SULPHUR FUELS?**

Although some after-treatment technologies are reported to require ULS fuels, other available options have to be considered as well. In addition, technologies are evolving fast. Detailed cooperative investigations should be conducted to evaluate the optimum level of sulphur in the fuels on an integrated basis.

In order to support its commitment to a reduction of  $CO_2$  emissions from vehicles, the EU motor industry is looking at new engine technologies. As all such technologies must also meet the increasingly stringent exhaust emissions limits, they must be linked to an appropriate after-treatment system. Much research has been and is being carried out in these fields and a number of options have emerged.

Currently, the most promising route to improved gasoline engine efficiency is the lean-burn G-DI engine technology. Such engines need effective reduction of  $NO_x$ . Although G-DI technology as such is not sulphur-sensitive, the  $NO_x$  storage catalyst systems required to reduce the  $NO_x$  emissions to the desired (Euro 4) level are currently affected by sulphur.

Although the effect is mostly reversible, higher levels of sulphur in the fuel result in faster catalyst deactivation and more frequent regeneration and desulphation cycles. Such regeneration and desulphation cycles involve a temporary change to a rich mixture with an associated increase in fuel consumption and therefore  $CO_2$  emissions. Very little data are publicly available to quantify the fuel efficiency penalty as a function of the fuel sulphur content. According to the limited information published so far, such gasoline  $NO_x$  storage catalysts would satisfactorily operate with fuels up to 50 ppm sulphur.

 $NO_x$  storage catalysts are a viable option for direct injection gasoline engines with a lean burn concept to optimize engine efficiency. Similar technologies are still in the research and development phase for light-duty diesel vehicles and may become a valid option for heavyduty engines. For the latter, after-treatment technologies such as the Selective Catalytic Reduction (SCR) systems are options that can operate satisfactorily with a maximum of 50 ppm sulphur level in the fuel. Among the particulate trap systems the Continuous Regenerating Trap (CRT) is reported to require a lower sulphur level. Detailed evidence is, however, not yet available to determine to what extent ULS fuels would allow this technology concept to deliver its full potential. CONCAWE's submission to the Commission includes a detailed analysis of the vehicle technologies available to the auto industry to contribute to both its  $CO_2$  commitment and the mandated exhaust emission levels.

#### OIL INDUSTRY INVESTMENTS AND EMISSIONS

Although technologies to produce ULS fuels exist, the cost of implementation is high and the additional emissions of greenhouse gases from refineries would be significant. The extra financial pressure on refiners could result in under-investment and tightening of the supply.

#### Refinery technology

A reduction of sulphur from 50 to 10 ppm in road fuels may seem small in absolute terms but would be far from trivial for the refiners. Taking into account the margins required to ensure the specification is met at the pump, refineries would have to produce fuels at a sulphur level of 6–7 ppm. This is indeed nearly one order of magnitude less, a very significant change in terms of, for example, the required desulphurization catalyst activity.

The bulk of the sulphur in gasolines originates from the FCC (Fluid Catalytic Cracker) streams that would therefore need to be almost completely desulphurized. This leads to some olefin saturation resulting in turn in a loss of octane. Technology is moving fast in this field and it is now possible, by a suitable combination of splitting and treating, to remove most of the sulphur while still keeping a fair proportion of the olefins. Some octane loss is still unavoidable and needs to be counterbalanced by increased use of high-octane components such as oxygenates or reformate, use of the latter being limited by the aromatics specification. Many gasoline components hitherto considered as 'sulphur-free' do in fact contain a few ppm of sulphur (e.g. alkylates, oxygenates, butane). While, with higher sulphur limits, they serve as a sulphur diluent, this is no longer the case for ULS scenarios where they have to be considered at best as sulphur neutral. In some cases additional treatment would also be required.

Virtually all diesel components would require desulphurization. Recent advances in hydrodesulphurization (HDS) catalysts make it possible to extend the range of this process to the very high levels of desulphurization compatible with the production of ULS diesel. This can be achieved in some of the existing plants, albeit at the cost of a capacity reduction, and/or in new plants similar to existing HDS plants but with comparatively larger amounts of catalyst and generally higher pressure levels. Deep hydrogenation (involving a second treatment stage on noble-metal catalysts) would not be required. Extra hydrogen and energy consumption would consequently be relatively small in absolute terms. Other properties of the product (such as density, cetane, aromatics) would only be marginally changed. ULS diesel would, however, have very low lubricity and conductivity and extra additives would have to be used to maintain quality, at a significant extra cost.

Although most of the processes required for both gasoline and diesel are based on proven technology some would use novel catalysts and/or processing schemes. In terms of practical day-today operation, reducing gasoil sulphur by three to four orders of magnitude is largely uncharted territory. This raises concerns with regard to the reliability of the HDS process with the potential for relatively frequent disturbances in production. Generally a learning curve would undoubtedly apply to the new processing schemes and might result in decreased reliability and localized short-term supply disturbances.

#### Logistics and distribution

Refinery oil movements and shipping systems as well as distribution networks, all of which are shared to some extent, would need to be carefully reviewed. Hardware as well as operating procedures would have to be adapted to minimize the risk of contamination (as little as 0.1 per cent of jet fuel could be enough to make a batch of diesel off-spec for sulphur whereas it would still be perfectly suitable with regards to other specifications). This would lead to additional costs as well as a reduction in general efficiency and therefore some increase in energy consumption (as an example, introduction of 50 ppm diesel in the UK has led to trucking back to refineries cross-contaminated material from multi-product pipelines). It also has the potential for creating short-term supply disruptions.

#### Refinery investments and extra CO<sub>2</sub> emissions

The CONCAWE methodology for evaluating the cost of a certain measure, based on linear programming (LP), has been described in detail in CONCAWE report 99/56. It is assumed that EU refineries would invest in order to continue to meet the forecast demand while having access to the same crudes (only one Middle Eastern crude is allowed as marginal feedstock). In this way possible trading options which would change the EU-15 global import/export balance are factored out. In reality a mixture of investment and trading options would be used but market forces would then ensure that the global cost remains more or less the same. The results of the CONCAWE study for the production of 10 ppm sulphur fuels are summarized in the box below.

The extra costs and  $CO_2$  emissions to move from 50 ppm sulphur to ULS fuels are of the same order of magnitude as the figures previously published for the Auto/Oil I measures and some Auto/Oil II scenarios.

Results of the CONCAWE	study for the production of 10 p	opm sulphur fuels		
		Gasoline	Diesel	Total
Net Present Value*	GEUR	4.8	6.7	11.5
Extra CO <sub>2</sub> emissions**	Mt/a	3.1	1.5	4.6
	% of total for road fuels	0.3	0.2	0.5

\*As per Auto/Oil I methodology:

\*\*Including changes in fuel heating value

For gasoline, investments would mainly concentrate on the generally larger and more complex FCC refineries and aim at both removing sulphur and re-establishing the octane balance. With only a small extra hydrogen requirement, the additional energy consumption would mainly be due to the energy use inherent to the additional processing plants.

For diesel, most refineries would have to invest in larger, higher-pressure HDS plants or at least in major revamps of existing plants. Generally the new plants would not consume much more energy than the existing ones while the extra hydrogen consumption would be small. For that reason additional  $CO_2$  emissions are relatively limited. Investments as well as extra operating costs (e.g. for extra additivation) are high.

In reality, some refiners may decide not to invest and to produce limited volumes through a combination of throughput reduction, appropriate crude selection and components trading. This could potentially cause serious tensions on the markets and lead to volatility and localized

supply shortage. In this respect gradual introduction, following the market demand for such fuels and possibly linked to tax incentives, would allow phasing of investments as supported by market conditions. Logistics would, however, be much more complicated (e.g. to ensure that the new fuels are used by the cars that need them).

#### IMPACT ON AIR QUALITY

For all the pollutants of concern the introduction of ULS fuels would have a negligible impact on either emissions or regional, as well as urban, air quality. Ammonia emissions would potentially increase leading to higher levels of secondary particulates.

At the 50 ppm level mandated for 2005, the contribution of road transport to the total  $SO_2$  emissions is already extremely small. For other air pollutants, the maximum effect is less than 0.5 per cent. The Auto/Oil II emission trends based on 50 ppm sulphur fuels from 2005 are not visibly affected by a move to ULS fuels from 2008. It is evident that such a minute change in emissions from transport would have a negligible impact on air quality in an urban environment. Given the negligible impact on NO<sub>x</sub> and VOC emissions, a move to ULS sulphur fuels offers essentially no contribution to improving the level of ozone attainment in the EU.

Gasoline engine catalysts are known to emit small amounts of ammonia. Once emitted in the atmosphere ammonia will neutralize acidic sulphate or nitrate aerosols to form the ammonium salt, thus adding to the total mass of secondary particulates. There is some evidence to suggest that lower sulphur in the gasoline will result in higher ammonia production, thus contributing adversely to air quality. Although the true magnitude of the increase is uncertain the growing concerns about the health effects of particulates warrants more study of this phenomenon.

#### **GLOBAL EFFECT ON GREENHOUSE GASES EMISSIONS**

In terms of  $CO_2$  emissions, the benefits from ULS fuels would not necessarily surpass the  $CO_2$  debit due to extra refinery processing.  $N_2O$  emissions could increase, further contributing to the overall greenhouse gases load.



Although it is reported that ULS fuels would allow an increase in the overall fuel efficiency of

lean burn G-DI cars equipped with  $NO_x$  storage catalysts, there is little published data to indicate the magnitude of the effect. On the basis of what limited information is available we have considered two scenarios assuming that the fuel efficiency would be improved by 1 to 2 per cent when reducing gasoline sulphur level from 50 to 10 ppm. The  $CO_2$  benefit over the years depends very much on the rate of introduction of such sulphur-sensitive technologies. We have derived figures from projections recently published by ACEA/EUCAR and have assumed that ULS gasoline would be introduced from 2008.

Unless the vehicle efficiency gains are well above 1 per cent, the  $CO_2$  increase in the refineries is not adequately compensated, so that the cumulative  $CO_2$  load only becomes negative over a very long time horizon. For diesel engines the development of  $NO_x$  storage catalysts is, in our understanding, less advanced and scenarios of overall  $CO_2$  balances are therefore even less clear.

Vehicles equipped with a three-way catalyst produce  $N_2O$  before the catalyst has reached full operating temperature. There is a strong possibility that the increased catalyst activity in a sulphur-free environment would lead to increased  $N_2O$  formation. Although warm-up time will be significantly reduced with the introduction of more advanced catalyst systems to meet the new exhaust emission standards taking effect from 2000 and 2005,  $N_2O$  emissions from the total vehicle fleet will continue to be of importance for a number of years to come. As a greenhouse gas,  $N_2O$  may be as much as three hundred times more potent than  $CO_2$  so that even a modest increase in emissions would markedly affect the global greenhouse gases load. Based on available COPERT<sup>1</sup> data, we have estimated that a 20 per cent increase in the  $N_2O$  emissions is plausible. This would result in an increased greenhouse gases load of some 2.3 Mt/a  $CO_2$  equivalent.

#### CONCLUSIONS

Although ULS fuels might bring benefits to certain vehicle technologies, there are a number of identified counterbalancing effects in terms of cost and  $CO_2$  emissions. There is also some evidence of potentially negative consequences in terms of air quality and greenhouse gases that require further studies. It is CONCAWE's opinion that the desirability of ULS fuels should be studied in a comprehensive joint programme that would uphold the principles of cost-effectiveness, sound science and transparency as well as be consistent with the Precautionary Principle.

<sup>&</sup>lt;sup>1</sup> Computer Programme to Calculate Emissions for Road Transport

## Disposing of used lubricating oils

The disposal of used lubricating oils is regulated in the EU by the Waste Oil Directive<sup>1</sup> (as well as other waste directives). This Directive requires the Commission to obtain statistical data on the collection and disposal of used oil in the Member States. The Commission has recently published a report on these statistics<sup>2</sup> and is planning to issue a revised Waste Oil Directive.

Under current EU legislation, used engine oil is considered a hazardous waste and Member States must ensure safe collection and disposal. They can apply subsidies for this. The Waste Oil Directive gives a hierarchy of disposal options. The first priority is for regeneration, i.e. rerefining. After this, used oil may be used as fuel as long as this is under environmentally acceptable conditions. Finally, if these options cannot be used, it must be subject to safe destruction or disposal. In the following paragraphs we revisit the main conclusions from a previous CONCAWE study and compare some of the results with the EU data.

CONCAWE carried out a study in 1994/5 based on 1993 data. This study covered all lubricating oils in the whole of Western Europe, not just motor oils. It did not include other types of waste oils such as those recovered from effluent treatment systems. One of the problems with such studies is to determine which types of oil are included in the data, particularly when there is the potential for different types of oil being mixed at the collection facilities. The study looked at the market situation, used oil collection, disposal routes, re-refining technology, and re-refining economics. The full results are published in CONCAWE report no. 5/96.

The CONCAWE data is compared with those collected by the EU Commission as well as those from another study by Coopers & Lybrand (C&L) in Table 1. While there are differences between the three sets of data, and they cover different years, the results are all basically similar. The most important feature is that only about half of the potentially collectable waste oil is recorded as collected and, of this, only some 36 per cent is in fact re-refined.

	<b>CONCAWE</b> (1993)	<b>EU</b> (1997)	<b>C&amp;L</b> (1995)	Average	Percer	nt of a	/erage
Sold	5319	4892	5109	5107	100		
otential waste oil	2624	2601	2448	2558	50	100	
Collected	1500	1915	1827	1747	34	68	100
egenerated	645	607	658	637	12	25	38
Burnt	732	1274	1169	1058	21	41	62
lissing	1124	686	621	810	16	32	

#### Table 1

Results of the different studies compared (kt/a). Note that, for each study, only about half of the potentially collectable waste oil is recorded as collected and, of this, only some 36 per cent is in fact re-refined.

<sup>&</sup>lt;sup>1</sup> Council Directive 75/439/EEC of 16 June 1975 as amended by Council Directive 87/101/EEC of 22 December 1986

<sup>&</sup>lt;sup>2</sup> Report from the Commission to the Council and the European Parliament on the Implementation of Community Waste Legislation for the period 1995–97 (COM (1999) 752 final, 10 January 2000, p.43)

It is important to realize that 'used oil' contains only about 70–80 per cent of actual oil. The type of oil is variable and probably a mixture of various grades. It also contains water (up to 10 per cent), unburned fuel (up to 10 per cent), metals (ca. 0.5 per cent), heavy ends, additives, and sulphur (up to 1 per cent). An important point is that used oil from gasoline engines is carcinogenic (because of PAH formation in the engines).

In the CONCAWE study a number of disposal routes were identified and the quantities of oil going to each route estimated. The results are given in Table 2. Burning in cement kilns uses the fuel value, organic contaminants are destroyed and the metals are locked in the cement. Burning in space heaters also uses the fuel value but is likely to cause local air pollution. Burning in hazardous waste incinerators should certainly control emissions and residues but may appear to use the energy content inefficiently. However, such incinerators usually require support fuel, and

Disposal route	kt/a	% of total
eclaiming industrial oils	165	П
urning (cement kilns, space heaters, incinerators)	480	32
-processing to fuel (mild by removal of water d sediments; and severe by dewatering, flashing		
d vacuum distillation)	375	25
-refining (to new lubricating oil)	480	32
otal	1500	100

if the waste oil replaces this, then the energy content is used effectively. Burning after mild or severe re-processing can also have advantages but the degree of processing and the environmental effects depend very much on the type of combustion installation being considered.

The CONCAWE study looked at various re-refining methods. Some of these gave good product quality but it was considered that severe hydrotreatment is probably required to achieve adequate removal of carcinogenic components. The main problems identified with re-refining were that it is capital intensive, and probably not economic without subsidies. It is difficult to confirm a lack of carcinogenicity in the product and it is probably not possible to produce premium quality grades of lubricating oil, particularly low friction oils, from the typical mixed feed. There are also emissions from re-refining processes and residues have to be disposed of.

A number of Life Cycle Analyses (LCA) have been carried out by others. These are difficult to do, as the uncertainties are large in comparison with the differences between the options. Most have concluded that re-refining is 'better' than burning, usually on grounds of energy efficiency. The CONCAWE study did not claim to be a full Life Cycle Analysis but it did consider the environmental aspects of the various options. It attempted to consider the impact on the whole refining industry using the CONCAWE refinery model. This is a complicated question as, amongst other things, it depends on the sulphur balance in the future European oil market. Also, the differences between the re-refining and burning options were small compared with the uncertainties and there was no convincing way to model the replacement of coal as a fuel for cement kilns by used lubricating oil. CONCAWE (which represents both lubricating oil manufacturers and re-refining plant operators) concluded that none of the possible disposal options had a clear advantage and that the results might well differ from place to place depending on local circumstances and the quantities of used oil available for disposal.

Table 2 CONCAWE identified a number of disposal routes for used oil, and the quantities of oil going to each route were estimated. Such an LCA should really also consider effects on the fuel economy of the vehicle using the oil. The difference in fuel consumption between a top-quality low friction oil and an 'ordinary' oil can be more than 5 per cent. Over 10 000 km a typical modern car uses ca. 1000 litres of gasoline and one fill of ca. 5 litres of engine oil of which perhaps 3 litres is recoverable oil. The possible difference in fuel use is 50 litres, which is much greater in energy conservation terms than any differences arising from different used oil disposal methods. CONCAWE's view is therefore that any incentives used to encourage re-refining should not include a compulsion to use re-refined oil in all new oils. There is no shortage of virgin base stocks for the manufacture of lubricants so that re-refining does not have an advantage from that point of view.

CONCAWE could not come to a clear choice on the 'best environmental option' for disposal of used oils and the optimum solution may vary with both place and time. The differences in energy requirements between the various disposal options are much less than the energy content of the used oil. The most important action is therefore to encourage the collection of used oil and select the disposal route that is most beneficial and environmentally acceptable in each specific case.

## Downstream oil industry safety statistics

Industry safety performance continues to improve

CONCAWE has now been collecting data on the safety performance of the downstream oil industry in Europe for seven years. This year, 21 CONCAWE members provided data, down from 27 last year. The decrease is partly due to the merging of companies. However, the statistics still cover a large majority of the companies refining and marketing oil in Western Europe.

Of the various statistics collected, the most reliable are those for the Lost Workday Injury Frequency (LWIF). This is the number of injuries sustained at work, which lead to an employee not being fit to work for one day or more per 1 million hours worked. The figure of 4.3 reported this year is slightly lower than that recorded for the last four years and the long-term average of 4.5. However, larger improvements have been achieved in other areas. The All Injury Frequency (AIF) at 9.4 is lower than the average for the previous five years, which was 10.3. The Lost Workday Injury Severity, the average number of days lost per incident, has been steadily decreasing during the course of the survey from 25.7 in 1993 to 19.3 in 1999. The Road Accident Rate (RAR) has also consistently improved over the years, reducing every year from 3.8 in 1993 to 0.9 in 1999.

Perhaps the most notable feature of the 1999 statistics was the number of fatalities, which at eight was the lowest number recorded in the seven years of the survey and resulted in the lowest recorded Fatal Accident Rate (FAR) of 1.8, down from 5.0 in 1993. Six of these fatalities resulted from road accidents, the other two being from construction and maintenance works. This indicates that the oil industry is well aware of the flammability and explosion hazards of the materials it handles and has developed procedures to reduce the risks to a low level. In spite of the improvement in the RAR, road accidents are still by far the main cause of fatalities in the industry.



One point that is obscured by the averages quoted above is the large differences between companies. The LWIF reported by the best company was only 0.34, whereas the highest was 14.6. Some of this variation can probably be explained by differences in categorization of incidents and reporting standards in different countries but there do seem to be real differences. The best performers tend to be those companies who recognize that safety is part of good business management, and in particular, makes good economic sense. In such companies, 'safety first' is not just a catch phrase but a positive commitment for management, employees and contractors.

# Aquatic toxicity testing for petroleum substances

The EU Dangerous Substances Directive (DSD) provides a systematic approach for the classification of substances based on knowledge of their intrinsic properties and in accordance with defined hazard criteria. The 18th Adaptation to Technical Progress (ATP) of the DSD provides the specific criteria for the classification of substances for environmental hazards.

CONCAWE has based its recommendations for the environmental classification of petroleum substances on results from Water Accommodated Fraction (WAF) studies. Some regulatory bodies (GESAMP<sup>1</sup>, UK Department of the Environment), along with industry associations (CEFIC, CONCAWE) and others, recommend the WAF approach as the most suitable method for testing the aquatic toxicity of 'difficult' substances. Its wider acceptance by EU Member States remains, however, under discussion.

Current OECD guidelines for the acute aquatic toxicity testing of substances are unsatisfactory when the substances contain components which are poorly soluble in water or volatile. However, during 1998, the OECD set up a working group to specifically address the problems of testing these 'difficult' substances with the aim of improving the current advice given in its ecotoxicity testing guidelines. The WAF method is included in a January 2000 OECD draft guidance document on the aquatic toxicity testing of difficult substances.

Test methods are only part of the problem of testing these difficult substances. Expression of the results is also problematic. All these difficulties are experienced in dealing with the aquatic toxicity testing of petroleum substances. Some of the problems encountered, and the solutions that have been found, are dealt with in the following paragraphs.

Because of the low water solubility of the hydrocarbon components, adding petroleum substances to water to produce solutions for the evaluation of aquatic toxicity rapidly results in the production of two-phase systems. A number of approaches have been developed to produce 'media' for testing, and these have previously been reviewed<sup>2,3</sup>. Many early studies described the preparation of water extracts of petroleum substances at very high loading rates (the term 'loading' or 'loading rate' has been frequently used as shorthand for the amount of hydrocarbon added to a specific volume of the test medium). The water extract was then diluted for testing, and the results expressed in terms of the percent dilution of the extract, or alternatively, in terms of the concentration of particular constituent hydrocarbons measured in the water. Data from this type of study, usually referred to as the WSF (Water Soluble Fraction) approach are not useful for classification; the percent dilution result is not in line with the quantitative criteria defined (expressed in terms of mg/l of test substance). Results based upon concentrations of specific dissolved components are again not suitable because the qualitative composition of the water fraction produced at a high loading rate may differ considerably from that of a solution prepared at a lower loading rate.

<sup>&</sup>lt;sup>1</sup> IMO/FAO/UNESCO/WMO/WHO/IAEA/UN/UNEP Joint Group of Experts on the Scientific Aspects of Marine Pollution

<sup>&</sup>lt;sup>2</sup> Connell, D.W. and Hawker, D.W. (1988). Use of polynomial expressions to describe the bioconcentration of hydrophobic

chemicals by fish. Ecotox Environ Safety 16, 242-257.

<sup>&</sup>lt;sup>3</sup> Girling, A.E. (1989). Preparation of aqueous media for aquatic toxicity testing of oils and oil-based products: a review of the published literature. Chemosphere 19, 10–11, 1635–1641.

Test methods based upon varying the loading concentration of the test substance in order to determine the toxicity, produce data more applicable to classification. Often, with petroleum substances composed primarily of low molecular weight hydrocarbons such as naphthas, nearly all of the substance is dissolved within the loading range where acute toxicity is observed. However, heavier petroleum substances contain a major proportion of components that are not completely soluble. In these cases, there are three methodological options available:

- 1. Remove the insoluble hydrocarbon portion;
- 2. Stir the mixture continuously to keep the insoluble material dispersed;
- 3. Allow the two phases to remain without physical mixing.

Approaches 1 and 2 have been most commonly utilized. The disadvantage of the second and third approaches is that the undissolved hydrocarbon may result in physical fouling and/or entrapment of the organisms, particularly *daphnia* and algae; the stirring apparatus used in the second approach may also cause physical harm to such small organisms. The first approach is favoured by CONCAWE, as it precludes physical effects and measures chemical toxicity. The water phase used in this approach is usually known as the Water Accommodated Fraction (WAF), since it is not usually possible to demonstrate that it is a true solution. Toxicity data resulting from these tests are expressed in terms of 'lethal loading' (LL) to distinguish them from the conventional lethal concentration (LC) data developed for single substances. The results of lethal loading studies are usually expressed as  $LL_{50}$  (the loading rate killing 50 per cent of organisms) or  $EL_{50}$  (the loading rate causing 50 per cent effect), rather than the corresponding  $LC_{50}$ , or  $EC_{50}$  values.

The concentration of each individual chemical species dissolved in the water phase at any particular 'loading' must be maximized. The maximum possible water concentration of each component is achieved through prolonged stirring of the water-petroleum substance mixture. In addition, a sealed system approach is necessary to measure the inherent toxicity of the more volatile petroleum substances.

Chemical analyses of the aqueous concentrations of all constituents are not possible due to the complexity of the composition, but equilibration should be confirmed by analysis of selected (representative) hydrocarbons or total dissolved hydrocarbons. Analyses are also typically undertaken to confirm whether the concentrations of selected components (and by inference, all components) decrease due to volatilization, or other processes such as biodegradation, during the period of exposure. Renewal of the WAF may be done daily, or at less frequent intervals, depending on stability. The concentrations of individual hydrocarbons in test media are not needed to calculate the  $LL_{50}$  value, since the results are expressed in terms of the entire substance (the concentration, or loading rate, used to make the initial WAF).

As a general rule, the use of auxiliary solvents or surfactants in the preparation of media for the testing of petroleum substances is not recommended. The presence of solvents during the preparation of test media from complex substances poses particular problems, as a consequence of their potential to influence partitioning between the dissolved and undissolved phases.

In conclusion, determination of the toxicity of petroleum substances is not a trivial matter. CONCAWE reaffirms its view based on sound technical arguments for the WAF method as the most suitable method for the aquatic toxicity testing of petroleum substances and strongly supports its regulatory acceptance by the EU.

#### CONCAWE PUBLICATIONS, 1999 TO DATE

General circulation (yellow cover) reports:					
1/99	European downstream oil industry safety performance—statistical summary of reported incidents—1998				
2/99	Environmental exposure to benzene				
3/99	Performance of cross-country oil pipelines in western Europe—statistical summary of reported spillages 1998				
4/99	Motor vehicle emission regulations and fuel specifications—part I summary and annual 1998/99 update				
Special	interest (white cover) reports				
99/5 I	Proposal for revision of volatility classes in EN 228 specification in light of EU fuels directive				
99/52	Exposure profile: kerosines/jet fuels				
99/53	Scientific basis for an air quality standard for nickel				
99/54	Overview and critique of the air pollution and health: a European approach (APHEA) project				
99/55	Fuel quality, vehicle technology and their interactions				
99/56	EU oil refining industry costs of changing gasoline and diesel fuel characteristics				
99/57	Environmental levels of benzene at the boundaries of three European refineries				
99/58	Occupational health auditing (1): occupational hygiene				
99/59	A test method to assess the 'inherent' biodegradability of oil products				
99/60	The health effects of PM <sub>2.5</sub> (including ultrafine particles)				
99/61	The European Union legislative process				
99/62	Potential of exhaust after treatment and engine technologies to meet future emissions limits				
00/5 I	The occurrence of selected hydrocarbons in food on sale at petrol station shops and comparison with food from other shops—a literature survey				
00/52*	Management of occupational health risks during refinery turnarounds				
Product dossiers					
99/110	Petroleum waxes and related products				
* Available shortly					
Up-to-c	late catalogues of CONCAWE reports are available via the CONCAWE website at www.concawe.be				
New reports are generally also published on the website.					

CONCAWE Madouplein 1 B–1210 Brussel Belgium

Telephone: 32-2-220 3111 Telefax: 32-2-219 4646 E-mail: info@concawe.be World Wide Web: http://www.concawe.be



Printed on cblorine-free paper made from recycled waste, sawmill residues, forest thinnings and fibre from sustainable forests.