

# **catalogue of concauwe special interest reports**

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## INTRODUCTION

The reports listed in this catalogue are CONCAWE special interest reports, also sometimes referred to as "white cover" reports, which have a limited initial circulation. There are a variety of reasons for limiting circulation: the two most common reasons are that the report is expected to have a restricted audience because of its specialist nature, or that the sheer volume of the report makes it uneconomic to print and distribute on a large scale. In the latter case a general circulation ("yellow cover") summary report is often also issued.

Stocks of these special interest reports are small, and readers are therefore asked to assess their requirements carefully before ordering, and to keep the numbers requested to the absolute minimum.

Copies of all CONCAWE reports are held in the Secretariat, including earlier reports, which are not necessarily listed in this catalogue. CONCAWE reports are published only in English. They may be ordered from the Secretariat by completing and returning the Report Order Form.

In addition, a number of recent reports are available on the CONCAWE Website <http://www.concaawe.org> and may be downloaded free of charge.

<b>CONTENTS</b>		<b>Page</b>
<b>INTRODUCTION</b>		<b>II</b>
<b>1.</b>	<b>INDEX TO TITLES OF CURRENT REPORTS</b>	<b>1</b>
1.1.	AIR QUALITY	1
1.2.	FUELS QUALITY AND EMISSIONS	2
1.3.	WATER AND SOIL PROTECTION	4
1.4.	OIL SPILL CLEAN-UP TECHNOLOGY	4
1.5.	PETROLEUM PRODUCTS	4
1.6.	HEALTH	6
1.7.	SAFETY	8
1.8.	REFINERY TECHNOLOGY	8
1.9.	GENERAL	8
<b>2.</b>	<b>SUMMARIES OF CURRENT REPORTS (in chronological order)</b>	<b>9</b>

## 1. INDEX TO TITLES OF CURRENT REPORTS

### 1.1. AIR QUALITY

Proceedings of the CONCAWE seminar on atmospheric emissions and their effects on the environment in Europe with particular reference to the role of hydrocarbons	86/61
US experience of hydrocarbon and NOx emissions control	86/70
Cost-effectiveness of hydrocarbon emission controls in refineries from crude oil receipt to product dispatch	87/52
Sulphur emissions from small stationary oil combustion plant and the availability of low sulphur fuel oil in the EEC	87/61
Capital and operating cost estimating aspects of environmental control technology - residue desulphurization as a case example	88/51
VOC emissions from gasoline distribution and service stations in Western Europe - control technology and cost-effectiveness	90/52
An EC-12/World inventory of greenhouse gas emissions from fossil fuel use.	91/54
VOC emissions from external floating roof tanks: comparison of remote measurements by laser with calculation methods	95/52
Best available techniques to reduce emissions from refineries	99/01
Environmental levels of benzene at the boundaries of three European refineries	99/57

**1.2. FUELS QUALITY AND EMISSIONS**

The relationship between automotive diesel fuel characteristics and engine performance	86/65
Future diesel fuel quality	86/71
An investigation into evaporative hydrocarbon emissions from European vehicles	87/60
Opportunities and costs to upgrade the quality of automotive diesel fuel	88/52
The control of vehicle evaporative and refuelling emissions - the "on-board" system	88/62
Economic consequences of limiting benzene/aromatics in gasoline	89/57
The effects of temperature and fuel volatility on vehicle evaporative emissions	90/51
The sulphur content of diesel fuel and its relationship with particulate emissions from diesel engines	90/54
The chemical composition of diesel particulate emissions	92/51
VOC running losses from canister-equipped vehicles	92/53
Diesel fuel aromatic content and its relationship with emissions from diesel engines	92/54
The effect of gasoline volatility on vehicle exhaust emissions at low ambient temperatures	93/51
Diesel fuel emissions performance with oxidation catalyst equipped diesel passenger vehicles - part 1	94/55
The effect of diesel fuel properties on the exhaust emissions from oxidation catalyst equipped diesel passenger vehicles - part 2	94/56

A review of analytical methods for the quantification of aromatics in diesel fuels	94/58
The influence of heavy gasoline components on the exhaust emissions of European vehicles - Part I - regulated emissions	94/59
The influence of heavy gasoline components on the exhaust emissions of European vehicles - Part II - unregulated emissions	95/53
Interim report on the European refining implications of severe reformulation of gasoline and diesel fuel	95/54
The influence of gasoline mid-range to back-end volatility on exhaust emissions	95/61
The influence of gasoline benzene and aromatics content on benzene exhaust emissions from non-catalyst and catalyst equipped cars - a study of European data	96/51
The measurement of the size range and number distribution of airborne particles related to automotive sources - a literature study	96/56
Diesel fuel/engine interaction and effects on exhaust emissions - Part 1: diesel fuel density Part 2: heavy duty diesel engine technology	96/60
Proposed EU year 2000 gasoline volatility specifications	97/53
A study of the number, size & mass of exhaust particles from European diesel and gasoline vehicles under steady-state and European driving cycle conditions	98/51
Polycyclic aromatic hydrocarbons in automotive exhaust emissions and fuels	98/55
Proposal for revision of volatility classes in EN 228 specification in light of EU fuels directive	99/51
Fuel quality, vehicle technology and their interactions	99/55
EU oil refining industry costs of changing gasoline and diesel fuel characteristics	99/56

Potential of exhaust after treatment and engine technologies to meet future emissions limits 99/62

Impact of a 10 ppm sulphur specification for transport fuels on the EU refining industry 00/54

Measurement of the number and mass weighted size distributions of exhaust particles emitted from European heavy duty engines 01/51

### **1.3. WATER AND SOIL PROTECTION**

An assessment of the application of acute toxicity testing for the monitoring and control of oil refinery effluents 86/55

Best available techniques to reduce emissions from refineries 99/01

### **1.4. OIL SPILL CLEAN-UP TECHNOLOGY**

Oil spill dispersant efficiency testing: review and practical experience 86/52

### **1.5. PETROLEUM PRODUCTS**

CONCAWE statement on the provision of confidential data on petroleum products and chemical additives 84/61

CONCAWE statement on the EEC carcinogenicity classification of straight-run middle distillates 86/57

CONCAWE statement on the classification of petroleum products against EEC skin irritation criteria 86/59

Implications of the EEC directive on product liability 86/63

Guidelines for safe handling of marine fuels 87/55

Guidelines on the classification of lubricant base oils according to EEC carcinogenicity criteria	87/62
Listing of petroleum products and refinery streams in the European inventory of existing commercial chemical substances (EINECS)	88/53
The use of short-term tests to predict the carcinogenicity of mineral oils	88/57
Implications of IARC review of diesel fuels and fuel oils, March 1988	89/51
EC and US environmental regulatory data requirements	89/54
Ecotoxicological testing of petroleum products: a tier testing approach	91/56
Ecotoxicological testing of petroleum products: test methodology	92/56
7th amendment to EC dangerous substances directive - implications for the petroleum industry	93/54
White oils and waxes - summary of 90-day feeding studies	93/56
Existing substances regulation: CONCAWE guidance on completion of Chapter 1 of HEDSET diskette	93/57
The use of the dimethyl sulphoxide (DMSO) extract by the IP 346 method as an indicator of the carcinogenicity of lubricant base oils and distillate aromatic extracts	94/51
Environmental risk assessment of petroleum substances: the hydrocarbon block method	96/52
Acute aquatic toxicity of kerosines - report on CONCAWE test programme	96/55
Acute aquatic toxicity of gasolines - report on CONCAWE test programme	96/57
Exposure profile: gasoline	97/52



Exposure profile: crude oil	98/52
Exposure profile: kerosines/jet fuels	99/52
A test method to assess the 'inherent' biodegradability of oil products	99/59
Revised dangerous preparations directive (1999/45/EC) -- implications for petroleum products	00/56
Environmental classification of petroleum substances - summary data and rationale	01/54

## 1.6. HEALTH

Effects of petroleum hydrocarbons on the nervous system	86/51
Health aspects of worker exposures to oil mists	86/69
Effects of skin contact with gasoline containing methanol	87/54
A management guide to occupational health programmes in the oil industry	89/52
Review of the toxicity of catalytically cracked clarified oil	89/56
An assessment of occupational exposure to noise in Western European oil refineries	90/53
Factors affecting the skin penetration and carcinogenic potency of petroleum products containing polycyclic aromatic compounds	90/55
Review of chemical and UV light induced melanomas in experimental animals in relation to human melanoma incidence	91/52
A preliminary study of ambient air concentrations of benzene around service stations and distribution terminals in Europe	94/53
Catalyst handling procedures to minimize exposures	95/57

A year long study of ambient air concentrations of benzene around a service station	95/63
Review of WHO regional office for Europe proposed short-term SO <sub>2</sub> air quality guideline	96/58
Nitrogen dioxide: evaluation of human health risks in chamber studies	96/59
Overview of the CONCAWE middle distillate programme	96/62
Scientific basis for an air quality standard for carbon monoxide	97/51
The health hazards and exposures associated with gasoline containing MTBE	97/54
Pilot study to investigate airborne benzene levels in service station kiosks	98/53
Scientific basis for an air quality standard for nickel	99/53
Overview and critique of the air pollution and health: a European approach (APHEA) project	99/54
The health effects of PM <sub>2.5</sub> (including ultrafine particles)	99/60
The occurrence of selected hydrocarbons in food on sale at petrol station shops and comparison with food from other shops - a literature survey	00/51
Management of occupational health risks during refinery turnarounds	00/52
An assessment of the reproductive toxicity of gasoline vapour	00/53
A review of trends in hearing thresholds of european oil refinery workers	00/55
A noise exposure threshold value for hearing conservation	01/52
An assessment of occupational exposure to noise in the european oil industry (1989-1999)	01/56

**1.7. SAFETY**

A joint petroleum / petrochemical barge inspection questionnaire 97/55

**1.8. REFINERY TECHNOLOGY**

Interim report on the European refining implications of severe reformulation of gasoline and diesel fuel 95/54

Best available techniques to reduce emissions from refineries 99/01

EU oil refining industry costs of changing gasoline and diesel fuel characteristics 99/56

Impact of a 10 ppm sulphur specification for transport fuels on the EU refining industry 00/54

**1.9. GENERAL**

The European Union legislative process 99/61

## **2. SUMMARIES OF CURRENT REPORTS (in chronological order)**

### **84/61 CONCAWE STATEMENT ON THE PROVISION OF CONFIDENTIAL DATA ON PETROLEUM PRODUCTS AND CHEMICAL ADDITIVES**

Within the petroleum industry very large sums of money are invested in research and development to produce the wide range of oil products used in an industrial society. However, the increasing requirements by Governmental bodies to build up lists of chemical substances, products and preparations have resulted in companies within the petroleum and chemical industries being asked to disclose information on such products which could threaten their confidentiality and the investment they represent. This report gives recommendations on how such requests for confidential data should be handled and indicates what non-confidential data may reasonably be provided.

### **86/51 EFFECTS OF PETROLEUM HYDROCARBONS ON THE NERVOUS SYSTEM**

Recently it has been claimed that long term exposure to low levels of petroleum hydrocarbons may impair behaviour and memory. CONCAWE has therefore reviewed relevant published literature available up to 1984 to appraise the effect of petroleum hydrocarbons on the nervous system. It is concluded that whilst there is good evidence that reversible central nervous system depression can result from exposure to moderately high concentrations of organic solvents and that peripheral nerves can be damaged by exposures to n-hexane and methyl-n-butylketone at levels above currently established exposure standards, there is at present no conclusive evidence that brain damage is caused by long-term exposure to organic solvent vapours.

### **86/52 OIL SPILL DISPERSANT EFFICIENCY TESTING: REVIEW AND PRACTICAL EXPERIENCE**

Laboratory testing of oil spill dispersant efficiency has increased in importance as the range of available dispersants has grown and authorities have become involved in performance specifications. Efficiency is the prime requirement of a dispersant: if a product is ineffective its physical or toxicological properties are irrelevant. In the past, some authorities have considered low toxicity as the prime requirement and this has caused many low efficiency products to be approved. Many different laboratory test methods have been devised to determine the effectiveness of dispersants. This report describes these methods for reference by practitioners in this field, and correlates results given by the tests on a range of dispersants and oils. It is concluded that most of the currently used test methods will rank the performance of a range of dispersants in a similar way. Although sea trials information is limited, visual observations have indicated a correlation between the performance ranking order observed and that obtained in the laboratory test.

### **86/55 AN ASSESSMENT OF THE APPLICATION OF ACUTE TOXICITY TESTING FOR THE MONITORING AND CONTROL OF OIL REFINERY EFFLUENTS**

This report is to provide oil company personnel responsible for relations with authorities, with a sound technical foundation to respond to proposals they may receive to introduce toxicity testing in the controlling scheme of their plant effluent water.

Historically industrial effluents have been controlled and monitored on the basis of chemical analysis, and this has proved effective for refineries. There is extensive evidence that present day refinery effluents are not acutely toxic after effective treatment. In some European countries where acute toxicity testing is a legal requirement, the refining industry as a whole has been exempted. However, there is some evidence of European regulatory authorities being influenced towards acceptance of bioassays as part of effluent monitoring and controlling schemes. The disadvantages though are; poor correlation between laboratory test results and field effects, variability of biological systems makes the interpretation of results difficult, results of tests using different organisms or test systems may not be comparable, slow response time of toxicity tests preclude their use for direct control of effluent treatment processes and bioassays rarely indicate which effluent components are responsible for toxic effects. Also specialised manpower and equipment would be required for such tests which can be costly and would require careful administration and control which may be beyond the capacity of many regulatory authorities.

**86/57 CONCAWE STATEMENT ON THE EEC CARCINOGENICITY CLASSIFICATION OF STRAIGHT-RUN MIDDLE DISTILLATES**

CONCAWE has reviewed API experimental data from animal tests on exposure effects of various middle distillates boiling in the 49°C-288°C range. CONCAWE's opinion is that the relevance of these tests is limited because the animal studies had an exposure regime which does not reflect human exposure conditions. Therefore it is considered that there is no justification at present for classifying straight run middle distillates (as distinct from middle distillates which distil only partially at temperatures below 288°C and contain higher boiling and cracked components) as EEC Group 2 or 3 carcinogens. Confirmation of this judgement should await further studies at doses not exceeding "maximum tolerated dose" levels and collection of additional information using other tests.

**86/59 CONCAWE STATEMENT ON THE CLASSIFICATION OF PETROLEUM PRODUCTS AGAINST EEC SKIN IRRITATION CRITERIA**

The purpose of this report is to assemble the available skin irritancy data, judge them against the EEC criteria and thus provide the evidence for CONCAWE's previous conclusion in Report No 1/85 that petroleum products are not skin irritants according to EEC criteria.

**86/61 PROCEEDINGS OF THE CONCAWE SEMINAR ON ATMOSPHERIC EMISSIONS AND THEIR EFFECTS ON THE ENVIRONMENT IN EUROPE WITH PARTICULAR REFERENCE TO THE ROLE OF HYDROCARBONS**

The CONCAWE seminar was held on 21 October 1985 in The Hague with speakers, among the foremost experts in their fields, drawn from the EEC, the UN, and from researchers inside and outside the oil industry. Participants were from the oil companies in Europe and from the OECD and TNO (Netherlands). The views expressed are those of the speakers and cover as many aspects of the topic as possible. CONCAWE is convinced that the seminar represents a valuable review of this subject. It will contribute to public awareness of the complexity of the problems involved and the need for further research into the causes, mechanisms and effects of atmospheric pollution.

The proceedings of the seminar (with the exception of Dr. McCabe's paper which is published separately in Report No. 86/70) are fully documented in this report.

**86/63      IMPLICATIONS OF THE EEC DIRECTIVE ON PRODUCT LIABILITY**

This highlights the main provisions of the EEC directive on product liability, 85/374/EEC, as they affect the petroleum industry, and provides a check-list of actions to be considered by CONCAWE participating companies in dealing with the implications. Recommendations are made on assessing and monitoring the safety of products, labelling of packages, supplying health and safety information and keeping appropriate records.

**86/65      THE RELATIONSHIP BETWEEN AUTOMOTIVE DIESEL FUEL CHARACTERISTICS AND ENGINE PERFORMANCE**

This is based on a study of over 40 published papers and data from as yet unpublished oil industry research. Because of the highly complex nature of the interaction between diesel engines and fuel, individual emissions species are discussed in conjunction with the range of fuel properties known to influence those emissions. The relationships show that fuel properties and engine performance necessarily result in conflicting requirements depending on individual engine design. This explains why changes in diesel fuel qualities have only a small influence on overall exhaust emissions, particularly relative to the influence exerted by current engine design and operating conditions.

**86/69      HEALTH ASPECTS OF WORKER EXPOSURE TO OIL MISTS**

This report assesses the broad classes of oils for which the current Threshold Limit Value (TLV) for worker/exposure to oil mist (8-hour time-weighted average (TWA) level of 5 mg/m<sup>3</sup>) is applicable. From a review of published information CONCAWE concludes that a single generic exposure limit is not applicable to all types of oil mists. The ACGIH TLV of 5 mg/m<sup>3</sup> provides an adequate safety margin for a broad range of products recognised as non-carcinogenic: - severely solvent refined oils, severely hydrotreated oils, oils sequentially processed by mild hydrotreating and mild solvent refining, white oils and petrolatums of medicinal or food grade quality. A second broad range of products exist where 5 mg/m<sup>3</sup> may not be applicable but acceptable exposure limits can be based on review of specific formulations: - products containing lighter oils e.g. kerosine, products for which exposures to additives need to be controlled, products used as emulsions. For the third broad range of products containing potentially carcinogenic oils, setting an exposure limit below 5 mg/m<sup>3</sup> would be prudent. Some guidance can be gained from the ACGIH TLV-TWA of 0.2 mg/m<sup>3</sup> for particulate polycyclic aromatic hydrocarbons (as benzene soluble material).

**86/70      US EXPERIENCE OF HYDROCARBON AND NOX EMISSIONS CONTROL**

The report summarizes the presentation given by Dr. L.J. McCabe in October 1985 at the CONCAWE seminar on atmospheric emissions. It is stressed that the original motives for ozone level control (and co-precursors NO<sub>x</sub> and hydrocarbons) were about urban/health issues in the US whereas rural/forest damage considerations predominate in Europe. Hence US regulations may not be appropriate for Europe. However, Europe can benefit from this legislative and regulatory experience. It has to be recognised that appropriate models for estimating rural (as opposed to urban)

ozone levels, are in an early development stage. Also, an effective control strategy should begin with the largest sources e.g. motor vehicles exhaust and evaporative emissions and solvents evaporation. Small items like car refuelling emissions (2% of total man made volatile organic compound emissions) should be lower priority. Also, before specific measures are adopted, adequate evaluation of cost-effectiveness should be carried out for the various technologies available. For example, carbon canisters onboard cars which potentially could save refuelling emissions as well as evaporative emissions, appear more cost effective than service station investments ("Stage 2") which would only recover refuelling emissions.

#### **86/71 FUTURE DIESEL FUEL QUALITY**

This is to publish the results of oil refinery manufacturing planning studies showing the impact of changing refinery processing on diesel fuel quality for the period 1990-2000. The main conclusion is that to provide the necessary scope for the different refinery configurations, crude slates and throughputs and product demand patterns, to keep all the individual refineries viable, will require the specification on cetane index to be reduced to 44 minimum. This results from a clear trend to lower cetane level due to the increased heavy to light product conversion operations required in European refineries. This, together with comments on the specification ranges for density, viscosity and volatility, is being advised to the motor manufacturers and the EEC, with the objective of ensuring that the diesel fuel quality used in developing diesel engine emissions performance and compliance with the emerging legislation, is not set at unrealistic levels.

#### **87/52 COST-EFFECTIVENESS OF HYDROCARBON EMISSION CONTROLS IN REFINERIES FROM CRUDE OIL RECEIPT TO PRODUCT DISPATCH**

This report shows that the cost-effectiveness of oil refinery emission control techniques, measured as the annual cost per ton of additional hydrocarbon retained in the system, rank in the following order:-

- formal programmes of monitoring and maintenance for control of fugitive emissions from refinery process plant have a cost-effectiveness of USD 100 per ton;
- provision of floating covers for waste water separator bays costs about USD 460 per ton of emission reduction;
- fitting rim mounted secondary seals in crude, feedstock and product tanks with external floating roofs has a cost-effectiveness in the range of USD 600 to USD 2840 per ton;
- the use of vapour recovery units at road loading gantries, with or without capture of vapour returned from service station tank filling, has a cost-effectiveness of USD 1000 to USD 1560 per ton of hydrocarbon recovered, respectively.

Vapour recovery units for rail, barge and ship loading cost from USD 3760 to USD 8500 per ton, suggesting a very low priority for implementation. The reason for the low cost-effectiveness relative to road loading is the infrequent use of larger capacity units.

For crude receipt, the changeover to segregated ballast with tanker fleet renewal over time (prescribed in the MARPOL 73/78 Convention) has the side effect of reducing hydrocarbon emissions at crude oil discharge locations.

If all the technology discussed in this report (excluding the crude tanker ballasting case) were applied in all refineries in Western Europe, the net effect would be a reduction of about 140 kt/yr of hydrocarbon emissions or only about 1.4% of the total anthropogenic hydrocarbon emissions in Western Europe; the annual cost would be about USD 170 million.

**87/54 EFFECTS OF SKIN CONTACT WITH GASOLINE CONTAINING METHANOL**

This report documents two studies sponsored by CONCAWE and carried out by Inveresk Research International, Musselburgh, Scotland, in relation to handling and use of gasoline containing methanol. The studies indicated that gasoline containing methanol at levels up to 10 per cent is no more irritant than conventional gasoline, that gasoline is only slightly irritating to the skin under conditions of exposure which are likely to occur during normal handling and use, i.e. occasional splashes, and that only a very small proportion of an applied dose of methanol was likely to be absorbed through the skin. Therefore, it is considered that normal handling and use of gasoline containing methanol does not present any significantly different hazard from conventional gasoline.

**87/55 GUIDELINES FOR SAFE HANDLING OF MARINE FUELS**

When properly used for their intended purpose and with good standards of personal and occupational hygiene, marine fuels are unlikely to present a short or long term health risk, given the closed systems under which they are normally stored, handled and consumed. However, where good standards are not maintained, health hazards can arise. Therefore, to minimize any potential risks, following consultation with health and safety specialists and occupational studies where necessary to assess and control exposures, the preventive measures described in this report should be rigorously applied with responsible adherence to a sensible regime of good personal hygiene and working procedures. This report advises end users of marine fuels of the potential hazards, the recommended precautions for safe handling and use, and on first aid treatment for accidental exposure.

**87/60 AN INVESTIGATION INTO EVAPORATIVE HYDROCARBON EMISSIONS FROM EUROPEAN VEHICLES**

Evaporative hydrocarbon emissions from thirteen cars (three equipped to meet current US emission limits) were measured using a modified SHED (Sealed Housing for Evaporative Determination) test procedure and four were tested using three other procedures to assess the effect of test severity. Using the standard test procedure, emissions varied between 4-16 g/test on a typical European summer fuel and from 9-24 g/test on a more volatile winter grade fuel. The emission controlled cars gave much lower levels, 1-3 g/test on the winter fuel, showing an 85% reduction compared to their equivalent European specification cars. As expected, increasing test severity caused a significant increase in emission levels. For the four cars tested, average emissions increased from 15 g/test to 48 g/test.

The effect of gasoline volatility was tested and RVP was found to be the only significant volatility parameter to affect emissions. Using the standard procedure, a



10 kPa reduction in RVP reduced evaporative emissions by 23%. Oxygenated fuels gave similar or lower emission levels compared to hydrocarbon fuels of equivalent RVP. An MTBE blend in particular produced significantly lower emissions.

A few measurements were made which suggest that diurnal losses are significant for uncontrolled cars, although they are not currently included in the CEC test procedure. Analysis of vapour samples taken from the SHED showed that the vapour consisted essentially of C4 to C6 hydrocarbons. A significantly higher proportion of olefins was found in the vapour than in the base fuel.

Exhaust hydrocarbon emissions levels were shown to increase with reducing gasoline volatility and for normal commercial fuels reducing RVP will tend to increase exhaust emission levels.

An estimate of the total inventory of evaporative hydrocarbon emissions in Europe shows the 1 Mt/yr vehicle evaporative emissions are the third largest source of man-made hydrocarbons in the atmosphere, after solvent evaporation (4 Mt/yr) and vehicle exhaust emissions (2.5 Mt/yr). Refuelling emissions, at only 0.18 Mt, are less than 2% of the total man-made hydrocarbon emissions. The most effective way of reducing evaporative emissions is clearly to fit carbon canister control systems to all vehicles.

#### **87/61      SULPHUR EMISSIONS FROM SMALL STATIONARY OIL COMBUSTION PLANT AND THE AVAILABILITY OF LOW SULPHUR FUEL OIL IN THE EEC**

This report deals with sulphur emissions from combustion plant of capacities less than 50 MWth and the possible effect the implementation of the EEC Large Combustion Plant Directive (LCPD) could have on these emissions. The availability of low sulphur fuel oil in the EEC up to year 2000 is also discussed and the differences between the Northern and Southern member countries highlighted.

Some information is given on the sulphur grades of fuel oil available in the 12 EEC member countries, in order to ascertain whether some standardization would be possible.

Main conclusions are that the introduction of the LCPD should have little effect on the anticipated reductions in sulphur emissions from small plants. However, some adverse effects are possible, due to increased scarcity of low sulphur fuel oil, in terms of a switch from fuel oil to alternative low sulphur fuels with attendant higher consumer costs. By the year 2000, the total supply capability for 1% fuel oil will be inadequate to meet the entire fuel oil consumption in the small plant sector. (Some 23 Mt/yr or 25% of total fuel oil consumption). 1% S fuel oil will anyway be spread unevenly over the countries and additional larger than average demand for low sulphur fuel oil in certain Member States would imply worse-than-average quality fuel oil in other areas. The oil industry itself will have very little flexibility to increase the availability of low sulphur fuel oil but power stations using very high sulphur fuel oil together with Flue Gas Desulphurization could help.

Because different conditions pertain across the Community, country specific studies would be required to obtain a better insight into their individual situations. Also, fuel oil grade structures vary widely as a result of tailoring to meet market requirements and it would be difficult to formalise standard community-wide low, medium and high

sulphur grades. The benefits of doing so are in any case not likely to be significant in terms of emission control.

**87/62      GUIDELINES ON THE CLASSIFICATION OF LUBRICANT BASE OILS ACCORDING TO EEC CARCINOGENICITY CRITERIA**

CONCAWE Report No. 1/85 ("Precautionary labelling of petroleum products in packages - a labelling system") concluded that severely refined oils were clearly not classifiable as EEC carcinogens and that vacuum distillates, mildly acid refined oils and distillate aromatic extracts should be classifiable as EEC Category 2 carcinogens and it did not distinguish between Categories 2 or 3 for mildly solvent refined or mildly hydrotreated oils. In addition, no guidance was provided on the classification of mixtures containing oils in different EEC categories, nor on what constitutes "severely" and "mildly" refined. The encouragement being given by EEC to recycling of waste oils rather than burning has also emphasised the need to address the carcinogenicity classification of reclaimed or re-refined oils. It is apparent that the need for expert interpretation of relevant information against EEC Category definitions, plus some differences in national views and regulations, make the harmonization of carcinogenicity classifications amongst oil suppliers in different European countries difficult to achieve. However, this is clearly a desirable objective and it has therefore been the subject of extensive discussion amongst CONCAWE member company experts. This document addresses problem areas and suggests some approaches which CONCAWE believes could lead to the desirable greater harmonization amongst oil suppliers and regulatory authorities.

**88/51      CAPITAL AND OPERATING COST ESTIMATING ASPECTS OF ENVIRONMENTAL CONTROL TECHNOLOGY - RESIDUE DESULPHURIZATION AS A CASE EXAMPLE**

Capital and operating cost estimates require a defined data framework to allow comparative scrutiny and meaningful correlation to be made with other estimates for the same and alternative technologies. The various elements of control technology cost estimating are described under the headings of:

- Capital cost
- Capital charge
- Fixed and variable operating costs
- Loss
- Indirect costs/benefits

Special emphasis has been put on capital cost estimating and the concept of capital charge. An accuracy of  $\pm 30\%$  is considered sufficient for capital cost estimates to allow ranking of different control technologies. This means putting emphasis on the identification and inclusion of the main cost items, rather than going into fine detail on equipment, and on clear statements of the assumptions made. A worked example is shown of a particular residue desulphurization case. This shows that capital charge makes the largest single contribution to annual costs followed by energy and loss.

**88/52 OPPORTUNITIES AND COSTS TO UPGRADE THE QUALITY OF AUTOMOTIVE DIESEL FUEL**

Future automotive diesel fuels are expected to have lower cetane numbers and higher densities than in the past. A survey has been made of possible routes and their costs to increase the quality of diesel fuels.

Three options have been identified:

- Selective blending of distillate pool components.
- Processing causing significant yield change, e.g. hydrocracking, solvent extraction.
- Hydrogenation with little or no yield change.

The hydrogenation process, although not in commercial operation and highly capital intensive, would be the only processing option to produce a significant cetane number increase across the EEC-12 diesel fuel pool. A two cetane number pool improvement would require a capital investment of USD 3.5-4.5 billion, a yearly total cost (including capital charge) of USD 1.3-1.7 billion and an additional hydrocarbon consumption of 3 Mt/yr.

A non-processing alternative viz. the use of an additive to improve cetane number would require some 30 kt/yr of additive for a two cetane number pool improvement and cost USD 42-50 million/year including dosing facilities and testing costs. This latter option is clearly less expensive than the processing route and is more flexible cost-wise in coping with fluctuating situations, but other parameters such as density and aromaticity are not changed.

**88/53 LISTING OF PETROLEUM PRODUCTS AND REFINERY STREAMS IN THE EUROPEAN INVENTORY OF EXISTING COMMERCIAL CHEMICAL SUBSTANCES (EINECS)**

The European Inventory of Existing Commercial Chemical Substances (EINECS) has been published as an advance edition version in the English language. It will come into full legal effect after translation into all the official EEC languages and publication in the Official Journal of the European Communities. EINECS identifies "existing substances placed on the market in the EEC for commercial purposes between 1st January 1971 and 18th September 1981". Future marketing of a substance not covered by an entry in EINECS will be subject to pre-marketing notification involving provision of a wide range of technical and toxicological data.

This report provides lists of petroleum product and refinery stream entries in EINECS number and alphabetical orders. These lists will enable individual companies to identify their own products and streams in EINECS.

**88/57 THE USE OF SHORT-TERM TESTS TO PREDICT THE CARCINOGENICITY OF MINERAL OILS**

A CONCAWE Workshop held in September 1986 reviewed the possibilities for shorter tests and reduced animal usage in test methods for assessing potential carcinogenicity of mineral oils. The tests examined included various in vitro assays, two short-term in vivo assays, and shortened versions of long-term skin painting assays. In combination with other data, such tests may provide some indication of

the likely carcinogenicity of oils. However, further development and validation are necessary, the need remains for some long-term skin painting studies, and account must be taken of the inherent uncertainties in extrapolating animal data to humans.

**88/62 THE CONTROL OF VEHICLE EVAPORATIVE AND REFUELLING EMISSIONS - THE "ON-BOARD" SYSTEM**

In this research programme to further investigate "on-board" control of evaporative Volatile Organic Compound (VOC) emissions from gasoline fuelled vehicles, an Opel Ascona and a Honda Civic were tested with specially installed enlarged carbon canisters to control refuelling emissions. Both installations achieved refuelling emissions below 0.05 g/litre of fuel dispensed representing control efficiencies of about 97% or more. The total evaporative emissions as normally measured, being the sum of diurnal, hot soak and refuelling losses, were below 2 g/test with all the fuels used which ranged between 61 kPa and 103 kPa RVP. No significant changes in exhaust emissions or in hot or cold weather drivability were seen between the standard and converted cars, and the tests demonstrated that regeneration of the canisters was achieved much more quickly than required to cope with the vapour from subsequent refuellings.

**89/51 IMPLICATION OF IARC REVIEW OF DIESEL FUEL AND FUEL OILS, MARCH 1988**

The IARC Monograph, Volume 45, concerning occupational exposures in petroleum refining: crude oil and major petroleum fuels was drafted in the March 1988 IARC review to evaluate carcinogenic risks to humans. CONCAWE has prepared this present Report No. 89/51 to relate IARC conclusions to the compositions of commercially available diesel fuels and fuel oils (heating oils).

The implications with respect to classification against EC Dangerous Substances Directive criteria are also reviewed.

**89/52 A MANAGEMENT GUIDE TO OCCUPATIONAL HEALTH PROGRAMMES IN THE OIL INDUSTRY**

Within CONCAWE's Health Management Group and its Industrial Hygiene, Medical and Toxicology Subgroups, experience has shown that broadly-based occupational health programmes produce positive business as well as human benefits. A pro-active and preventive team approach by experienced professionals in the competent disciplines of occupational health can ensure early identification of potential problems for management, employees, customers and the general public and define the actions required to prevent their occurrence.

Increasing awareness of potential occupational and environmental health risks associated with the manufacture, distribution, use and disposal of petroleum products and the development of additional legislation, all emphasise the need to consider the impact of these issues on oil refining and marketing operations.

This briefing paper has therefore been prepared on behalf of CONCAWE's Health Management Group to outline the broad nature and benefits of successful multi-disciplinary occupational health programmes.

The programmes identified in this paper should be seen as guidelines. The appropriate elements should be selected by CONCAWE member companies and will depend on the nature and scope of their operations.

**89/54 EC AND US ENVIRONMENTAL REGULATORY DATA REQUIREMENTS**

Current and proposed environmental legislation in the EC and US is reviewed. The report provides a summary of the intent of the legislation, the developments foreseen and an assessment of the types of petroleum products which might be effected. Where possible, the product ecotoxicological data that are required under the various items of legislation are defined.

**89/56 REVIEW OF THE TOXICITY OF CATALYTICALLY CRACKED CLARIFIED OIL**

Catalytic cracking is used in petroleum refineries to break down higher boiling feedstocks into lower boiling components which are fractionated into various distillate streams. The bottoms fraction which is known as slurry oil, is passed through a slurry settler to remove catalyst. The resulting brownish-black liquid is known as catalytically cracked clarified oil (CCCO), or alternatively, clarified (or catalytic) slurry oil, decant oil or heavy clarified oil. According to the American Petroleum Institute (API), CCCO (CAS No. 64741-62-4) contains "hydrocarbons having carbon numbers predominantly greater than C<sub>20</sub> and boiling above approximately 350°C (622°F)". The primary uses of CCCO are as feedstocks to coker units and as a component in heavy fuels (i.e. Bunker C and No. 6 fuel oil). It may also be used in cut-back and emulsified bitumens; feedstocks for petroleum cokes, petroleum pitch and carbon black; dust suppressant road oils; enhanced oil field recovery oil; and rubber extender oils, process oils and ink oils. Some of these uses have declined or have been discontinued in recent years. It is estimated that CCCO production in Europe (OECD countries) is approximately 8.3 million tons per annum.

The principal route of exposure to CCCO is dermal. The petroleum industry has long recognized the hazardous nature of CCCO and has instituted stringent industrial hygiene practices in its plants. Therefore CCCO tends to be processed in closed systems and where workers wear protective clothing and are instructed on safe handling procedures. Customers are provided with hazard communications (i.e. Material Safety Data Sheets (MSDS) and/or notification letters) which warn of the dangers and provide advice on safe working practice with CCCO. The number of people potentially exposed and the extent of their exposure to CCCO are believed to be small.

**89/57 ECONOMIC CONSEQUENCES OF LIMITING BENZENE/AROMATICS IN GASOLINE**

This report records the economic consequences to four different types of refineries in Europe if the benzene content of gasoline is required to be limited to 3% vol or 1% vol. The consequences of also setting limits on aromatics content are also investigated. The study utilized refining planning computer models optimized by linear programming techniques.

European gasoline currently contains on average 2.6% vol benzene and 34% vol aromatics. These levels would increase to 3.2% vol and 43% vol, respectively, if all gasoline were to be supplied as 95 octane unleaded grade; depending on individual

refinery configuration, the production would range from 2.3 to 5% vol benzene and 35 to 56% vol aromatics, with the highest levels resulting from simple refineries (hydroskimming/thermal cracking) processing Brent-type crude oils. The levels also depend on the amount of oxygenates and isomerization capacity available.

A restriction of benzene in gasoline to 3% vol would mainly affect the simple refineries (still representing 40% of the number of refineries and 20% of the capacity in EC), which would need benzene extraction facilities, and isomerization capacity if not already installed. The investment for the refining sector in EC would be USD 1100 million. The manufacturing cost increase would range from a minor increase for complex refineries (catcracking/hydro-cracking/coking) up to USD 10-12/ton for simple refineries.

Further reduction of benzene below 3% vol would need benzene extraction facilities also in complex refineries. A 1% vol benzene limit would require an investment of USD 1750 million in EC. The manufacturing cost increase would go up to USD 8-12/ton for complex refineries and to USD 16-20/ton gasoline for simple refineries.

About 2 million t/yr of benzene would have to be extracted and disposed of in a European market of 5 million t/yr as result of a 1% benzene limit. The aromatic content of gasoline from simple refineries could only be reduced by some 5 percentage points through the additional use of oxygenates and isomerization, resulting in average aromatics levels still exceeding 40% vol. Further aromatics reduction in simple refineries would result in yield losses of up to half or more of the gasoline production. Complex refineries could achieve aromatics levels generally in the range of 30 to 35% vol through the wide use of oxygenates as well as additional isomerization.

## **90/51 THE EFFECTS OF TEMPERATURE AND FUEL VOLATILITY ON VEHICLE EVAPORATIVE EMISSIONS**

Evaporative emissions of gasoline vapours have been measured from five matched pairs of European vehicles, with and without carbon canister evaporative emission control systems. Measurements of hot soak and running losses were made at various ambient temperatures using different fuel volatilities and driving patterns. Measurements of benzene emissions were also made.

Hot-soak losses and running losses from uncontrolled vehicles increased progressively with ambient temperature and fuel volatility (RVP) and have been summarized by simple three-term mathematical models. The response of the emissions to ambient temperature and volatility was found to be similar for all cars and in good agreement with previous studies. A 1°C change in ambient temperature was found to have the same effect on evaporative emissions as a 3.8 kPa change in fuel RVP. Fuel tank temperatures and the consequent hot-soak emissions increased with more severe, i.e. higher speed and longer duration warm-up of the vehicles, whereas the running losses (per km) were approximately independent of the driving pattern.

Carbon canisters were found to be effective at controlling evaporative emissions to very low levels at all except unrealistically high combinations of ambient temperature and fuel volatility, and total daily emissions were reduced by around 95%.

Running losses could not be detected from canister equipped cars with the procedure used in this programme. Due to the inadequacies of the procedure used to measure running losses, an improved measuring technique is needed to be certain they do not occur.

Reducing fuel RVP had no significant effect on emissions from vehicles equipped with carbon canisters.

Benzene emissions from uncontrolled vehicles varied significantly between vehicles, and the benzene content of the vapour could be more or less than that of the fuel. The carbon canister control system effectively reduced benzene emissions in line with total hydrocarbons.

**90/52 VOC EMISSIONS FROM GASOLINE DISTRIBUTION AND SERVICE STATIONS IN WESTERN EUROPE - CONTROL TECHNOLOGY AND COST-EFFECTIVENESS**

This report firstly identifies the sizes of the emissions from gasoline distribution and retail networks both within the EC-12 and 17 OECD European countries. Secondly it estimates the emission reductions achievable with various control measures. The control techniques are described and their cost and cost-effectiveness in reducing emissions reviewed.

Within the EC-12 countries there are 860 terminals (at refineries and intermediate storage facilities) at which gasoline is loaded into road tankers. The majority of terminals (64%) have throughputs less than 100 kt/yr but these handle only about 20% of the total gasoline. Some 54 terminals, mainly at refineries, additionally have rail loading facilities. A fleet of about 17 500 road tankers delivers the gasoline to 137 000 service stations.

Installing single-stage vapour recovery units (VRUs) at all road tanker loading terminals and Stage 1 vapour return equipment at all service stations could recover 75-81% of the gasoline vapours emitted. Only about another 2% could be recovered by using double-stage VRUs. Taking into account that some vapour control measures are already installed, it is estimated that installing Stage 1 at all terminals and service stations would require between 800 M\$ to 1225 M\$ investment. The average cost-effectiveness of Stage 1 on road deliveries is USD 2630/t. The range around this average is large depending upon the throughput of the terminal.

Stage 1 value for money decreases sharply for road delivery networks handling less than 120 kt/yr.

Equipping rail delivery terminals with vapour recovery equipment would cost another \$54-73 million investment and at 82% recovery would recover another 2.8% of total uncontrolled Stage 1 emissions.

Stage 2 equipment to reduce car refuelling emissions would be less than 60% efficient with the current European car population. At this low efficiency, the cost-effectiveness of Stage 2 would be \$5800/t. Stage 2 efficiency and hence cost-effectiveness could only be improved by modifications being made to car fill pipe and tank vent designs, and therefore would depend upon the replacement rate of the car population.

**90/53 AN ASSESSMENT OF OCCUPATIONAL EXPOSURE TO NOISE IN WESTERN EUROPEAN OIL REFINERIES**

CONCAWE has summarized and analysed results of over 800 nominal fullshift personal noise exposure measurements made available by its member companies. The data, collected in five Western European countries in the period 1982-1988 relate to work in areas such as refinery plant, utilities and workshops, and were obtained using measurement procedures consistent with the CONCAWE guidelines for conducting personal noise dosimetry.

The objectives of the work were to investigate the trends in noise exposure and to compare the levels with occupational noise exposure limits set down in the EC Noise Directive. For these purposes, the 1982-84 data were separated from the 1985-88 data and both sets assessed to ascertain the percentage of measurements less than 80, 85, 90 and 95 dB(A).

It is concluded that there has been a reduction in noise exposure levels for the period 1985-1988 compared with those in the period 1982-1984. However, in spite of this improvement, many noise exposures still exceed one or both of the  $L_{EP,d}$  (8h) action levels specified in the EC noise directive at which the implementation of effective hearing conservation programmes is required.

**90/54 THE SULPHUR CONTENT OF DIESEL FUEL AND ITS RELATIONSHIP WITH PARTICULATE EMISSIONS FROM DIESEL ENGINES**

This report gives the results of a research programme to investigate the influence of diesel fuel sulphur content on diesel engine exhaust particulate emissions. A representative range of current light-duty vehicles (nine) and heavy-duty engines (four) were tested using European test procedures with four fuels of 0.31, 0.22, 0.12 and 0.055% wt sulphur. The test fuels were produced by progressively desulphurizing the base fuel, avoiding changes in other fuel quality variables which may have influenced the emissions results.

The study found a consistent, but small influence of diesel fuel sulphur on the mass of particulate emissions from the light-duty vehicles: exhaust particulates declined about 7% as fuel sulphur content reduced from 0.3 to 0.05% wt.

For the heavy-duty engines, there was no consistent trend linking reducing particulate levels with reducing fuel sulphur content. This finding differs from the trends found in some US studies. However, those studies used sulphur doped fuels, an artificial situation considered not representative of refining practice in the production of low sulphur fuels.

**90/55 FACTORS AFFECTING THE SKIN PENETRATION AND CARCINOGENIC POTENCY OF PETROLEUM PRODUCTS CONTAINING POLYCYCLIC AROMATIC COMPOUNDS**

The potential of oils to produce skin cancer depends on their polycyclic aromatic compound (PAC) content and on the bioavailability of the PAC, i.e. penetration of the PAC into the cells of the viable epidermis. Residual aromatic extracts are less carcinogenic than their PAC content would imply. It has been suggested this is due to their lack of bioavailability. Factors influencing bioavailability include the barrier properties of the skin, the partition coefficient between the oil and the skin,



concentration of the specific PAC in the oil, duration of contact, the size and shape of the specific PAC, and the metabolism of PAC to a form that can interact with the DNA of epidermal cells. The interaction of all these factors is not fully understood.

Results from chemical analysis, dermal penetration, and dermal carcinogenicity studies of oils with viscosities between 6 and 415 cSt show that the carcinogenic activity is highly correlated with the concentration of 3 to 7 ring PAC in the oil, and suggest that viscosity has a minimal effect on carcinogenic activity. Over the viscosity range of 6 to 415 cSt, the dermal penetration rate of benzo-a-pyrene from oils was inversely proportional to the log of the viscosity. No data are available which relates oil composition to carcinogenic activity of oils with viscosities above 415 cSt; although the viscosity of these high boiling oils may have no greater effect on dermal penetration, further work is needed to substantiate this. Therefore, the viscosity of the mixture is not expected to be a major factor in determining carcinogenic potency.

Further work on high viscosity oils is needed to investigate to what extent the viscosity of the oil affects its carcinogenic activity and to what extent the carcinogenic activity is governed by composition.

**91/52 REVIEW OF CHEMICAL AND UV LIGHT-INDUCED MELANOMAS IN EXPERIMENTAL ANIMALS IN RELATION TO HUMAN MELANOMA INCIDENCE**

Following the IARC conclusion that there is an increased incidence of melanoma among refinery workers, a literature review was conducted to determine the strength of evidence relating exposure to polycyclic aromatic hydrocarbons (PAH) with occurrence of melanoma.

Of the PAHs, only 7,12-dimethylbenz(a)anthracene (DMBA) had been reported to induce melanoma, and then only in a few animal species under experimental conditions involving high dose levels.

In view of the low level of exposure to DMBA in refineries and the apparent inability of other PAHs or complex hydrocarbon mixtures to produce melanomas, together with the absence of any observed increase in other skin cancers in refinery workers, it was considered unlikely that any increase in melanoma incidence in such workers was the result of PAH exposure.

**91/54 AN EC-12/WORLD INVENTORY OF GREENHOUSE GAS EMISSIONS FROM FOSSIL FUEL USE**

An inventory of emissions of carbon dioxide from fossil fuels for the European Community (EC-12) from 1987 to 2010 is developed. The predictions are based on published data and set in the context of world emissions. Although information for methane is both limited and uncertain, some data are provided to illustrate its potential significance relative to carbon dioxide; nitrous oxide emissions are assumed to be negligible.

Two scenarios predict increasing emissions from the EC-12 although their percentage contribution to World totals decreases as emissions from non-EC-12 countries increase further. A third scenario predicts a significant reduction in EC-12 emissions but within the timeframe considered this can only be indicative of the possible effects of energy conservation and fuel switching.

The contribution of oil to EC-12 emissions falls from 50% to 45% in all three scenarios.

The data clearly illustrate that concerted global action would be required to limit significantly the predicted growth in emissions.

**91/56 ECOTOXICOLOGICAL TESTING OF PETROLEUM PRODUCTS: A TIER TESTING APPROACH**

There is considerable knowledge of the effects of oil on the environment from studies of oil spills. However, relatively few data on oil products have been generated for regulatory purposes.

The deficiencies of the current ecotoxicological database for petroleum products have been recognized by the industry and the present report provides a framework within which relevant data requirements can be identified and addressed. The report describes a structured tier-testing scheme. It also recognises the principle of "read across" whereby data for one product can be used to predict the toxicity of structurally related products. In combination, these two approaches will allow data to be obtained in a manner which ensures efficient utilization of resources, which minimizes the use of test organisms and shortens the overall time taken for testing and evaluation.

**92/51 THE CHEMICAL COMPOSITION OF DIESEL PARTICULATE EMISSIONS**

CONCAWE Report No. 90/54 described a programme which measured the particulate emissions from a wide range of diesel vehicles and heavy duty diesel engines on fuels of 0.31, 0.22, 0.12 and 0.055% weight sulphur content. The work also included chemical analysis of the particulates. The procedures used in that programme for the chemical analysis of particulates are described and the chemical composition data obtained are discussed.

Six light duty diesel vehicles and four heavy duty diesel engines, representative of the current European parc were tested over EC legislated cycles. Particulate samples were analysed for total carbon, sulphate, and hydrocarbons in the fuel and lubricating oil boiling ranges. In addition to the analyses given above, selected samples were analysed for total and individual polycyclic aromatic hydrocarbons (PAH).

The analysis failed to establish any trend of chemical composition with fuel sulphur content, apart from the expected decrease in particulate sulphate level with reduced fuel sulphur content. Wide differences in particulate mass and composition were determined within engine and vehicle groups, over legislated test cycles. These differences were, however, broadly in line with published data.

When expressed on a direct mass/mass basis, PAH levels on particulate obtained under ECE-15+EUDC conditions were approximately an order of magnitude greater than those obtained under ECE-R49 conditions. Wide differences in PAH emission levels were determined within vehicle and engine groups.

Considerable problems were encountered in this programme in terms of methodology for the chemical analysis of diesel particulate. The problems led

directly to the formation of an Institute of Petroleum working group whose purpose is to publish standard methods in this area.

**92/53 VOC RUNNING LOSSES FROM CANISTER-EQUIPPED VEHICLES**

Six European vehicles fitted with carbon canisters have been tested under severe conditions to establish if evaporative losses of volatile organic compounds occur under European driving conditions - so called "running losses". The programme entailed the development of a point source measurement technique which has a number of advantages over other methods currently in use.

Following the development and validation of the measurement technique, the six vehicles were tested at 28°C over a range of driving cycles on a gasoline with a Reid vapour pressure of 90 kPa. None of the vehicles exhibited classical running losses, i.e. losses during higher-speed driving. This was due to the effectiveness of canister purging in these conditions. However, significant volatile organic compound (VOC) losses were observed for several vehicles during idle after a period of driving had heated the fuel. Substantial car-to-car variation was observed in the losses obtained. The losses were always more severe over longer idling periods, and more severe than hot soak over comparable periods. This may have important implications for urban pollution.

Critical factors affecting running losses are fuel temperature and purging strategy. Higher fuel temperatures increase vapour generation and hence the canister charging rate. Purging rates must be sufficient to overcome the charging rate. Large carbon canisters (LCC) were found to be more effective than small carbon canisters (SCC) in reducing running/idling losses because of the extra adsorbent capacity available. Mitigation of refuelling losses is an added benefit. Systems that combine the canister with a pressurized fuel tank, in order to limit VOC charging of the canister, were shown to run the risk of VOC losses from sources other than the canister vent.

**92/54 DIESEL FUEL AROMATIC CONTENT AND ITS RELATIONSHIP WITH EMISSIONS FROM DIESEL ENGINES**

This report provides the results of a research programme designed to investigate the influence of diesel fuel aromatic content and cetane number on diesel engine exhaust emissions.

A representative range of seven current light-duty vehicles, together with two heavy-duty engines, was tested using European test procedures with six fuels having aromatics contents in the range 15 to 37% volume. The test fuels were produced by deep hydrogenation of the base fuel. This process influences other fuel quality parameters, including density, sulphur content and cetane number. To balance these changes the matrix included sulphur and ignition improver additive-doped fuels. A hydrocracked fuel was also included in order to study the influence of aromatic type.

The study found a significant influence of fuel properties on carbon monoxide and particulate emissions from light-duty vehicles. The strongest correlations were obtained with cetane number. Inclusion of aromatics terms in correlation with cetane number gave no improvement over correlations incorporating only cetane number.

**92/56 ECOTOXICOLOGICAL TESTING OF PETROLEUM PRODUCTS: TEST METHODOLOGY**

This is the second of two CONCAWE reports considering ecotoxicological testing of petroleum products. An assessment is made of the relevance and validity of the standard ecotoxicological test protocols for the assessment of the environmental hazards presented by oil products. The report concludes that in many cases it is technically feasible to adapt standard methods for aquatic toxicity, bioaccumulation and ready biodegradability, but that further work is needed, particularly to develop appropriate "*inherent biodegradability*" tests for oil products. A number of specific recommendations are made with the intention of promoting a uniform approach to the generation of ecotoxicological test data on petroleum products.

**93/51 THE EFFECT OF GASOLINE VOLATILITY ON VEHICLE EXHAUST EMISSIONS AT LOW AMBIENT TEMPERATURES**

Eight European vehicles, four of which were equipped with 3-way catalysts, have been tested on two gasolines with significantly different front-end/mid-range volatilities. The investigation was conducted over the new ECE+EUDC test cycle at various ambient temperatures. It was found that emission levels varied widely between individual vehicles and that the effect of fuel volatility on emissions was much less than the effect of temperature. Carbon Monoxide (CO) and Hydrocarbon (HC) emissions increased dramatically as test temperature was reduced. For catalyst cars, CO emissions increased by over 500 per cent and HC emissions by around 300 per cent as the temperature was reduced from 25 to -5°C. NO<sub>x</sub> emissions were much less affected by test temperature.

**93/54 7TH AMENDMENT TO EC DANGEROUS SUBSTANCES DIRECTIVE - IMPLICATIONS FOR THE PÉTROLEUM INDUSTRY**

The European Community (EC) rules relating to the classification, packaging and labelling of dangerous substances have been amended/updated with the publication of the 7th Amendment (92/32/EEC) of the Dangerous Substances Directive (67/548/EEC). This report discusses the 7th Amendment highlighting those changes which are considered relevant to the oil industry.

**93/56 WHITE OIL AND WAXES - SUMMARY OF 90-DAY FEEDING STUDIES**

CONCAWE has conducted a study on white mineral oils and waxes which are representative of those used for food applications. The aim was to help clarify the mixed results found in other toxicity studies with laboratory animals and the implications from a human health viewpoint.

It was found that microcrystalline waxes and 100 cSt white oil elicited no adverse effects in the 90-day feeding studies carried out. Biological effects were inversely related to molecular weight, viscosity and melting point, but oil type and processing did not appear to be determinants.

The absorption of mineral hydrocarbons and the nature of the occurrences of the biological responses were confirmed and a new effect for paraffin wax has been identified. Further studies are required to elucidate more fully the mechanism for the responses observed.

**93/57      EXISTING SUBSTANCES REGULATION: CONCAWE GUIDANCE ON COMPLETION OF CHAPTER 1 OF HEDSET DISKETTE**

Advice is given in a step by step manner on the completion of Chapter 1 of the HEDSET diskette for the reporting of petroleum substances under the Existing Substances Regulation. Chapter 1 deals with the manufacturer-related data that oil companies in the EC are required to submit to the European Commission.

**93/58      USING THE HEDSET PROGRAMME TO CREATE DATA SETS FOR THE EXISTING SUBSTANCES REGULATION 793/OF 23 MARCH 1993**

The EC Existing Substances Regulation 793/93 requires manufacturers and/or importers to submit data sets on the risks of those substances to man and the environment. The European Commission has developed a computer program, HEDSET, with which to compile such data sets, and this report gives recommendations and practical advice on how to use the software and organize the data storage.

**94/51      THE USE OF THE DIMETHYL SULPHOXIDE (DMSO) EXTRACT BY THE IP 346 METHOD AS AN INDICATOR OF THE CARCINOGENICITY OF LUBRICANT BASE OILS AND DISTILLATE AROMATIC EXTRACTS**

Untreated lubricant base oils have been associated in the past with the development of human skin cancer. To give a better understanding of these health effects, industry has conducted an extensive range of long-term dermal carcinogenicity studies with the objective of identifying the influence of different types of refinery processing and to establish the important base oil compositional factors. The studies have led to improved refining techniques and to the development of simple markers for control purposes based on a standard analytical test.

However, with the increasing emphasis on the regulatory classification and labelling of petroleum products, it is proposed that the same markers can be effectively used for the classification of base oils.

The report describes the development of markers for the prediction of base oil carcinogenicity and examines the relative merits of two particular candidates, one based on dimethyl sulphoxide extraction by method IP 346 and the other based on benzo(a)pyrene (BaP) concentration.

**94/53      A PRELIMINARY STUDY OF AMBIENT AIR CONCENTRATIONS OF BENZENE AROUND SERVICE STATIONS AND DISTRIBUTION TERMINALS IN EUROPE**

CONCAWE initiated a programme to measure the ambient air concentrations of benzene in the vicinity of a number of gasoline stations and distribution terminals in Europe. The steps in the programme were: review previous studies/measurement methodologies; establish sampling and analytical procedures; perform a pilot study to establish a protocol and validate techniques; conduct the main study. The results showed benzene-in-air levels at the service station boundaries ranging from 1.6 to 119  $\mu\text{g}/\text{m}^3$ , with the highest results generally recorded downwind of the service station. In general, the results obtained during the study were similar to previously reported levels for urban air. For distribution terminals the results indicated benzene-in-air levels of a similar magnitude.

**94/55 DIESEL FUEL EMISSIONS PERFORMANCE WITH OXIDATION CATALYST EQUIPPED DIESEL PASSENGER VEHICLES - PART I**

This study confirms the very effective control of diesel exhaust particulate, carbon monoxide and hydrocarbon emissions with oxidation catalyst technology. In addition, the investigation documents the ability of an exhaust oxidation catalyst to effectively reduce the variation in emission levels with fuels meeting the EN 590 specifications. However, a balanced design of catalyst, engine and vehicle seems to be important if optimum performance is to be achieved.

**94/56 THE EFFECT OF DIESEL FUEL PROPERTIES ON EXHAUST EMISSIONS FROM OXIDATION CATALYST EQUIPPED DIESEL PASSENGER VEHICLES - PART II**

This report further evaluates the study reported in CONCAWE Report No. 94/55 and documents the ability of oxidation catalyst technology to reduce the influence of fuel properties on exhaust emissions performance. The relationship between fuel properties and exhaust emissions was very vehicle dependent. Particulate emissions were found to correlate best with fuel density but no correlation was identified with total aromatics. Oxidation catalysts reduce particulate emissions by oxidizing hydrocarbons and are generally tolerant to fuel sulphur.

**94/58 A REVIEW OF ANALYTICAL METHODS FOR THE QUANTIFICATION OF AROMATICS IN DIESEL FUELS**

The definition of "aromatics" in diesel fuel has caused difficulties, both within the oil industry and amongst other interested parties. By virtue of the boiling range of the fuel, the majority of its hydrocarbons will have between ten and twenty carbon atoms. This means that any molecule considered aromatic may also have a "non-aromatic" component that is greater than the aromatic portion. The level of aromatics that is measured is therefore dependent on the technique used. This report summarizes the various analytical procedures currently available for the determination of diesel fuel "aromaticity".

**94/59 THE INFLUENCE OF HEAVY GASOLINE COMPONENTS ON THE EXHAUST EMISSIONS OF EUROPEAN VEHICLES - PART I - REGULATED EMISSIONS**

Ten European vehicles, meeting the requirements of the EU 'Consolidated Emissions Directive', have been tested on a matrix of seven gasolines over the current ECE+EUDC test cycle. The programme was designed to investigate the effects of heavy gasoline components in terms of both distillation and composition on the emissions performance of a fleet of modern fuel injected catalyst cars.

Gasoline back end volatility and composition both had some effect on regulated emissions performance. For HC and CO emissions, back end volatility overall had a larger effect than composition. However, the back end effects were discontinuous, with no measurable effect between the 160°C and 180°C T90 fuels. The fuel effects on NO<sub>x</sub> emissions were in the opposite direction to those for HC and CO, and compositional influences in this instance were greater than those due to back end volatility.

The back end volatilities of all the test fuels differed to an increasing extent from mid range (T50) to final boiling point (FBP). It was not possible to ascribe the fuel effects to any one distillation point within this range; neither to distillation temperatures at

percent volumes recovered (T values), nor to percent evaporated volumes at certain temperatures (E values). Throughout all the tests, it was evident that emissions performance differences between the cars were substantially higher than differences observed across the fuel matrix.

**95/52 VOC EMISSIONS FROM EXTERNAL FLOATING ROOF TANKS: COMPARISON OF REMOTE MEASUREMENTS BY LASER WITH CALCULATION METHODS.**

Hydrocarbon emissions from storage tanks are normally calculated using procedures published by the American Petroleum Institute.

A laser-based technique has been used to measure remotely the emissions of hydrocarbons from floating roof tanks. The measurements obtained have confirmed the accuracy of the recently updated API estimation method for external floating roof tanks.

The ability of the remote measurement technique was demonstrated by comparison with direct measurements of emissions during the loading of a barge.

**95/53 THE INFLUENCE OF HEAVY GASOLINE COMPONENTS ON THE EXHAUST EMISSIONS OF CATALYST EQUIPPED EUROPEAN VEHICLES - PART 2 - UNREGULATED EMISSIONS**

This report extends the investigation published as CONCAWE Report No. 94/59, which studied the influence of gasoline back-end volatility and composition on regulated emissions. This Part 2 report describes the full hydrocarbon and aldehyde/ketone speciation measurements conducted as an integral part of the programme. The results demonstrated that cars have a much greater effect on speciated emissions than fuel parameters. No consistent trends were observed in speciated emissions with the less volatile fuels, and the minor fuel effects were more related to fuel composition than back-end volatility. Ozone forming potential was also evaluated.

**95/54 INTERIM REPORT ON THE EUROPEAN REFINING IMPLICATIONS OF SEVERE REFORMULATION OF GASOLINE AND DIESEL FUEL**

This report records the results of a study to assess the EU-12 refining industry implications of severe universal reformulation of gasoline and diesel fuel individually and together, in terms of investment and operating costs, energy consumption and carbon emissions. It concludes that the edge of the envelope within which a practical set of reformulation options might lie would require investment of up to US \$40-60 billion, would increase refining own use and loss from about 8% to 11% of throughput and would increase overall CO<sub>2</sub> emissions by up to 55 Mt/yr.

This is an interim report and the study will go on to assess fuels packages lying within this envelope which are options needing cost/benefit assessment under the European Auto/Oil Programme.

**95/57 CATALYST HANDLING PROCEDURES TO MINIMIZE EXPOSURES**

This report outlines a generalized approach to reducing worker exposure to solid and liquid catalysts during refinery catalyst handling activities such as reactor loading, unloading and screening.

Many catalyst handling operations have the potential for generating airborne dust, mist or vapour, depending on the nature of the catalyst involved, and due care and attention is necessary to prevent adverse health effects. The report stresses that engineering controls should be adopted as the primary means of minimizing exposures, although it acknowledges that the use of personal protective equipment, such as respirators, goggles and gloves, is also necessary in many catalyst handling activities.

**95/61 THE INFLUENCE OF GASOLINE MID-RANGE TO BACK-END VOLATILITY ON EXHAUST EMISSIONS**

This report is a literature review of recent published studies evaluating the effects of mid-range to back-end volatility on regulated emissions from gasoline powered vehicles. It is concluded that there are no wholly definitive data defining exactly which distillation parameters are the true causative factors in influencing tail-pipe emissions, partly due to the necessary physical constraint of a certain degree of intercorrelation between adjacent distillation parameters, e.g. T50 and T60 or E100 and E110. There is, however, a balance of evidence suggesting that the effect is best described by parameters in the mid-range region, typically between T50 and T70, or E100 and E120.

Back-end chemical compositional effects on emissions have been studied by CONCAWE and AQIRP. CONCAWE found that changing back-end composition from aromatics to paraffins and then to olefins gave relatively small but progressive reductions in HC and CO and increases in NO<sub>x</sub> emissions from catalyst cars. Fuel Volumetric Air Demand (FVAD) i.e. kg of air per litre of fuel, was thought to describe the magnitude of transient air fuel ratio excursions and hence emission effects observed, although intercorrelation between FVAD and other fuel parameters prevented firm conclusions. Distillation effects were found to be somewhat more important than back-end compositional effects for HC and CO emissions, but for NO<sub>x</sub> the opposite was true. The AQIRP Phase 2 Heavy Hydrocarbon study reported no strong effects of back-end chemical composition on emissions. The reason for the rather different findings of the two programmes is not clear.

**95/63 A YEAR LONG STUDY OF AMBIENT AIR CONCENTRATIONS OF BENZENE AROUND A SERVICE STATION**

This report describes the results obtained from measurements of ambient benzene levels around a single service station in the UK over a one year period. Continuous sampling was achieved using diffusion tubes and results compared with those from intermittent active sampling using pumps and adsorption tubes. Results obtained around the service station were compared with those from a corresponding "green field site" in the same local area.



**96/51 THE INFLUENCE OF GASOLINE BENZENE AND AROMATICS CONTENT ON BENZENE EXHAUST EMISSIONS FROM NON-CATALYST AND CATALYST EQUIPPED CARS - A STUDY OF EUROPEAN DATA**

An analysis of data on the effect of gasoline benzene and aromatics contents on exhaust benzene emissions has been conducted. It was based on data from CONCAWE member companies and an Italian industry programme, and included the results of emission tests on 21 conventional non-catalyst and 34 catalyst cars. Although none of these programmes was specifically aimed at investigating the combined effects of gasoline benzene and aromatics content on benzene exhaust emissions, the combination of data from the individual programmes allowed some insight into these relationships.

Earlier programmes conducted with non-catalyst cars - using the ECE-15 test cycle - demonstrated that the main effect on benzene exhaust emissions derived from the benzene content of the gasoline employed. However, higher aromatics also influenced benzene exhaust emissions, albeit to a lesser extent. The effect of benzene in those earlier programmes was about twelve times higher than that of higher aromatics. Analysis of new emissions data over the combined ECE15+EUDC test cycle indicated that similar relationships existed for both non-catalyst and catalyst cars.

If benzene emissions are expressed as a percentage of total hydrocarbons emitted, then the effect of gasoline benzene content and other aromatics varies between vehicle type. More specifically, the influence of fuel benzene content was found to be over 18 times greater than that of non-benzene aromatics for non-catalyst cars. For catalyst equipped cars, the effect of benzene content was 10 times greater than that of other aromatics. Moreover, benzene exhaust emissions from catalyst cars were substantially lower. On average, emissions were reduced by around 85%, demonstrating the efficient control provided by the catalysts employed.

It has also been demonstrated that the regression equations developed predict the trends and magnitude of the benzene exhaust emissions observed over the modified ECE(11 s)+EUDC cycle, as used in the EPEFE programme and the 1994 CONCAWE gasoline study. This cycle employs a shorter idle period at the start of the test and collects exhaust emissions immediately from cranking the engine.

**96/52 ENVIRONMENTAL RISK ASSESSMENT OF PETROLEUM SUBSTANCES: THE HYDROCARBON BLOCK METHOD**

This report describes an approach to environmental risk assessment for petroleum substances compatible with the principles described in EU Technical Guidance Documents.

**96/55 ACUTE AQUATIC TOXICITY OF KEROSENES - REPORT ON CONCAWE TEST PROGRAMME**

This report describes the experimental procedures and the results obtained in acute ecotoxicity tests on three kerosine samples. The samples were tested for toxicity to the rainbow trout, *Oncorhynchus mykiss*, the crustacean zooplankter, *Daphnia magna* and the alga, *Selenastrum capricornutum* using water accommodated fractions. These results assist in determining the environmental hazard from kerosines.

**96/56 THE MEASUREMENT OF THE SIZE RANGE AND NUMBER DISTRIBUTION OF AIRBORNE PARTICLES RELATED TO AUTOMOTIVE SOURCES - A LITERATURE STUDY**

This report reviews the literature relating to particulate size measurement and the techniques suggested for different applications. Successful measurements have been made using cascade impactors, which give gravimetric analysis of sized fractions and electrical mobility techniques, which give number distributions of sized fractions. It is recommended that this approach be employed in the assessment of automotive particulate emissions. Because the subsequent fate of these emissions are subject to many complex and confounding factors, their measurement "at the tail-pipe" has only a tenuous relationship with air quality particulate inventories.

**96/57 ACUTE AQUATIC TOXICITY OF GASOLINES - REPORT ON CONCAWE TEST PROGRAMME**

This report describes the experimental procedures used, and the results obtained in conducting acute ecotoxicity tests on eleven gasoline samples to meet OECD guidelines and recognised principles of good laboratory practice. The loading rate approach that was used, gave results for studies involving fish, Daphnia and algae. These results assist in determining the environmental hazard classification of both gasoline components and gasoline itself.

**96/58 REVIEW OF WHO REGIONAL OFFICE FOR EUROPE PROPOSED SHORT-TERM SO<sub>2</sub> AIR QUALITY GUIDELINE**

The World Health Organization (WHO) regional office for Europe recommends an SO<sub>2</sub> guideline of 500 µg/m<sup>3</sup> (0.175 ppm) for a 10 minute sampling time, an uncertainty factor of 2, and a threshold of about 1000 µg/m<sup>3</sup> (0.35 ppm).

Based on SO<sub>2</sub> chamber studies described herein the following conclusions are summarized:

- Population at risk

The WHO-EU recommended guideline is set to protect the most susceptible individuals among the most susceptible population, namely exercising asthmatics not receiving medication.

- 10 minute sampling time

The data suggest an exercising asthmatic's response to SO<sub>2</sub> begins within minutes, reaches a maximum in about 10 minutes, and does not increase with longer exposures (and may actually decrease with longer or repeated exposures). Further, bronchoconstriction may be reduced at rest compared with that experienced while exercising even if exposure continues.

- Healthy subjects, COPD patients, and resting asthmatics are not at risk of an adverse response to SO<sub>2</sub> exposure at concentrations as high as 1 ppm SO<sub>2</sub>.
- The interpretation of the exposure-response relationships of exercising asthmatics, depends on the definition of an adverse effect. The WHO-EU definition is the most conservative one in the literature.

- The basis for inclusion of an uncertainty factor of two is unclear. The chamber studies include sensitive subjects, representative of the asthmatic population.

**96/59 NITROGEN DIOXIDE: EVALUATION OF HUMAN HEALTH RISKS IN CHAMBER STUDIES**

The World Health Organization /European region (WHO/EU) proposes a 0.11 ppm 1-hour guideline for ambient NO<sub>2</sub> concentrations. This guideline is based on reversible 1 hour effects on lung function of greater than 5% and increased airway responsiveness (AR) observed in mild asthmatics at 30 minute exposures to 0.2 to 0.3 ppm.

In summary, the chamber study data indicate little or no change in lung function as a result of short-term exposure NO<sub>2</sub> levels ranging from ppb to ppm concentrations for both healthy subjects and patients with asthma and obstructive airway disease. Concentrations of NO<sub>2</sub> likely to be found in ambient air increase airway responsiveness to irritants such as cold air, SO<sub>2</sub>, and O<sub>3</sub>, in both asthmatics and healthy individuals. However, the actual reduction in lung function associated with the increased responsiveness is small and not an effect that can be distinguished from background variability and the effects of exposure to air alone.

**96/60 DIESEL FUEL/ENGINE INTERACTION AND EFFECTS ON EXHAUST EMISSIONS  
PART 1: DIESEL FUEL DENSITY  
PART 2: HEAVY DUTY DIESEL ENGINE TECHNOLOGY**

CONCAWE has investigated two major aspects of fuel/engine interaction and the resulting effects on exhaust emissions:

- The impact of fuel density on the particulate emissions of a light duty turbocharged passenger car fitted with an advanced engine technology/electronic management system.
- The influence of technology change on two generations of the same model of heavy duty diesel engine.

For both investigations fuels of the EPEFE (European Programme on Emissions, Fuels and Engine Technologies) diesel fuel matrix were used. This matrix represents the optimum concept to study effects of decorrelated fuel properties (density, poly-aromatics, cetane number and T-95). The study concluded that changes in engine technology and engine management systems had a profound effect on emissions performance which far outweighed any benefits accruing from changes in fuel characteristics.

**96/62 OVERVIEW OF THE CONCAWE MIDDLE DISTILLATE PROGRAMME**

This report reviews the results from a three phase programme of work designed to investigate the factors influencing the skin carcinogenicity of middle distillates. In particular, it concentrates on the final phase, which consisted of a 2 year skin painting study in mice with two gas oils and a kerosine, each applied at three different concentrations.

It was concluded that materials containing significant concentrations of PACs probably produce skin tumours by a genotoxic mechanism. Undiluted straight-run gas oil and kerosine, containing low or undetectable levels of PACs may produce tumours but only when moderate to severe skin irritation is also present. Such effects are probably related to the influence of the continuous cycle of cell damage and repair prompted by chronic skin irritation.

The results are an important consideration in terms of the hazard and risk assessments for middle distillate fuels.

**97/51 SCIENTIFIC BASIS FOR AN AIR QUALITY STANDARD FOR CARBON MONOXIDE**

Carbon Monoxide (CO) is emitted into the atmosphere mainly as a product of the incomplete combustion of carbonaceous material. The major sources of CO exposure for the general, non-smoking population are exhaust emissions from combustion engines and the burning of fossil fuels. Smoking provides an additional source of CO for the non-smoking as well as the smoking public. In addition to these exogenous sources, CO is generated endogenously mainly from the breakdown of haem proteins. Healthy individuals can tolerate low level exposures to CO but it can be hazardous at higher concentrations and even at low concentrations for those with unusual susceptibility.

The primary toxic action of carbon monoxide is the inhibition of cell oxidation following exposure by inhalation. The brain, heart, and embryo/foetus have critical needs for oxygen. It follows that the major health effects associated with CO exposure include cardiovascular, central nervous system, and developmental toxicities. Of these, the most critical target of carbon monoxide is the cardiovascular system. Furthermore, the most susceptible populations are those individuals with pre-existing cardiovascular disease.

The absorption and elimination of CO from the body is influenced by concentration, duration of exposure and pulmonary ventilation. Most absorbed CO binds reversibly with haemoglobin (Hb) forming carboxyhaemoglobin (COHb), reducing the oxygen carrying capacity of the blood. The relationship between CO exposure and the formation of COHb has been mathematically described. The extent of COHb saturation may be used as a biological marker of exposure. In addition, specific adverse health effects have been linked to characteristic COHb levels, which have in turn been associated with CO exposure levels, and these relationships can serve as a basis for an Air Quality Standard (AQS).

Chronic angina patients are presently viewed as the most sensitive group at risk after exposure to CO, and the most sensitive health endpoint is earlier onset of angina with exercise. A COHb saturation of 2.5% posed no significant health effect to the non-smoking population including those with angina. This COHb level

corresponds to continuous CO exposures at 15 ppm. A safety factor was applied to the CO exposure conditions to adjust for the uncertainty associated with persons having other pre-existing disease conditions, e.g., anaemia or pulmonary disease. This adjustment resulted in a predicted COHb concentration of 1.6%, corresponding to continuous CO exposure at 10 ppm. [Thus, an 8 hour running average of 10 ppm is a scientifically supportable AQS for carbon monoxide.]

**97/52 EXPOSURE PROFILE: GASOLINE**

This report details the available exposure data for gasoline and gasoline products. It provides information on estimated release rates; measured human exposure data for occupationally exposed groups, consumers and the general public; and environmental exposure data for air, water, soil and sediments. The potential for indirect exposure to gasoline hydrocarbons via the food chain is also addressed. As gasoline is a complex substance, the exposure data is presented in terms of total hydrocarbons and the main hazardous constituents, namely 1,3-butadiene, n-hexane, benzene, toluene, ethylbenzene and xylenes.

The report also gives a summary of the composition, hazards and occupational exposure limits for gasoline and lists supply and consumption figures.

**97/53 PROPOSED EU YEAR 2000 GASOLINE VOLATILITY SPECIFICATIONS**

A CONCAWE study identified the proposed flat maximum RVP limit across Europe to be of greatest concern within the gasoline volatility specification in the year 2000 EU Draft Fuels Directive. The 60 kPa RVP defined for a six month summer period would lead to problems with regard to driveability and exhaust emissions during intermediate seasons and safety requirements in the more extreme Nordic countries. A flat RVP would result in unbalanced evaporative HC emissions across Europe.

The report concludes that a constant RVP limit for Europe is not practical due to the widely varying summer temperatures. It provides examples for adjusted RVP limits which will provide low but balanced evaporative HC profiles across Europe. Geographically and seasonally adjusted volatility specifications are recommended - an approach which has been applied historically to ensure safe operation and good driveability.

**97/54 THE HEALTH HAZARDS AND EXPOSURES ASSOCIATED WITH GASOLINE CONTAINING MTBE**

This report compares the toxicological properties of gasoline with and without MTBE. It also reviews the available occupational and consumer exposure data for MTBE.

**97/55 A JOINT PETROLEUM / PETROCHEMICAL BARGE INSPECTION QUESTIONNAIRE**

This report provides an agreed checklist to be used when carrying out pre-chartering checks of barges intended for the transport of petroleum products and similar petrochemicals within Western Europe. It has been produced by representatives of oil and chemicals companies active in this area to provide a

consistent basis for such checks and allow easy interchange of information between companies.

**98/51      A STUDY OF THE NUMBER, SIZE & MASS OF EXHAUST PARTICLES EMITTED FROM EUROPEAN DIESEL AND GASOLINE VEHICLES UNDER STEADY-STATE AND EUROPEAN DRIVING CYCLE CONDITIONS**

This study investigates the measurement of the mass and the number of light duty automotive (diesel and gasoline) exhaust particles and their related size distributions. Different analytical techniques for particle size determination are assessed and compared and recommendations made for future work. Selected aspects of particle emissions are also investigated across a limited number of vehicles and fuels, but covering a wide range of vehicle technology and marketed fuel quality.

**98/52      EXPOSURE PROFILE: CRUDE OIL**

This report details the available exposure data for crude oil. It provides information on estimated release rates; measured human exposure data for occupationally exposed groups; and environmental exposure data for air, water and soil. The potential for indirect exposure to crude oil hydrocarbons via the food chain is also addressed. As crude oil is a complex substance, the exposure data are presented in terms of total hydrocarbons and the main hazardous constituents, namely n-hexane, benzene, toluene, ethylbenzene, xylenes and polycyclic aromatic hydrocarbons.

The report also gives a summary of the composition, hazards and occupational exposure limits for crude oil and lists supply and consumption figures.

**98/53      PILOT STUDY TO INVESTIGATE AIRBORNE BENZENE LEVELS IN SERVICE STATION KIOSKS**

This report describes the results of a small pilot study which was carried out in two service stations in the UK. The purpose of the study was to compare the concentrations of benzene in the forecourt air with that in the kiosk at each of the service stations and possibly identify reasons for any observed differences.

The results showed that the ambient concentrations of benzene on the forecourts of the two service stations differed significantly. A possible explanation for this observation is that the service station with the lower ambient benzene concentrations had a stage I vapour balancing system installed, thereby reducing releases of hydrocarbons into the air during tanker discharging.

The results also showed that for each service station, the benzene concentration inside the kiosk was similar to that immediately outside. It was also shown that increases in ambient benzene concentrations following forecourt activities, was followed by a corresponding increase inside the respective kiosk.

**98/55 POLYCYCLIC AROMATIC HYDROCARBONS IN AUTOMOTIVE EXHAUST EMISSIONS AND FUELS**

A comprehensive literature review of polycyclic aromatic hydrocarbons (PAH) in automotive exhaust emissions and fuels has been conducted. Sources and mechanisms of their formation in the internal combustion engine are discussed. Analytical techniques for PAH determination are described and experimental designs to elucidate the sources of PAH are proposed. PAH emissions reduction techniques employing either emissions control technology or alternative fuels are reviewed. The limitations of current understanding of polycyclic aromatic hydrocarbon formation are discussed and recommendations are made for further work. While the study reports on PAH related to automotive issues, some general information is included on environmental and health concerns.

**99/01 BEST AVAILABLE TECHNIQUES TO REDUCE EMISSIONS FROM REFINERIES**

BAT Reference documents (BREFs) are to be prepared by the European IPPC Bureau established at JRC/IPTS in Seville, and the Refinery BREF is scheduled for 1999. It is understood that the oil industry will be involved in the preparation of this document. The oil industry wishes to make a positive and informed contribution to the exercise, based on actual data on facilities installed in refineries, their capital and operating costs, and delivered performance capability. CONCAWE therefore established Special Task Forces WQ/STF-28 and AQ/STF-55 to study this subject. This report details their findings.

This CONCAWE report considers a wide range of emissions control techniques for refinery operations, the cost of installing and operating them, and the performance they have been demonstrated to deliver. The document is divided into five sections as follows:

- I. General Introduction
- II. Emissions to Air
- III. Emissions to Water
- IV. Waste
- V. Soil and Groundwater

**99/51 PROPOSAL FOR REVISION OF VOLATILITY CLASSES IN EN 228 SPECIFICATION IN LIGHT OF EU FUELS DIRECTIVE**

CONCAWE has reviewed the current gasoline volatility specifications within EN228 relating to hot weather driveability, i.e. RVP, E70 and Vapour Lock Index (VLI), in anticipation of changes to volatility characteristics after year 2000, due to the impact of the new EU Fuels Directive (98/70/EC).

This study utilises the assessment of the hot weather driveability (or Hot Fuel Handling (HFH) performance of current European vehicles and the trends of current and year 2000 car populations to determine the volatility requirements of individual European markets for year 2000. The generation and interpretation of such data is based upon extensive knowledge in this field accumulated by member companies over many years.

Along with appropriate consideration of future trends in gasoline composition, the study leads to the conclusion that for year 2000 summer volatility classes, a VLI

specification is no longer necessary. Data also shows that for other seasons volatility classes (non-summer time), a VLI specification is also generally no longer necessary. Only during the transition periods between summer and winter for four markets, identified as critical, might a VLI be used as an alternative solution to ensure satisfactory driveability.

This report had been made available to CEN / TC19 / WG21 during their review of EN 228 and contains total customer satisfaction curves as a means of allowing the appropriate selection of volatility classes for individual European markets in accordance with their climatic variation and car populations. Further proposals for changes to year 2000 volatility, in addition to VLI removal, are included in this report.

Due to the proposed increase in minimum levels of E70 and E100, in addition to the introduction of an E150 minimum limit, the new version on EN 228 should improve the cold weather driveability performance of gasoline vehicles.

#### **99/52 EXPOSURE PROFILE: KEROSINES/JET FUELS**

This report details the available exposure data for kerosines and jet fuels. It provides information on estimated release rates; measured human exposure data for occupationally exposed groups; and environmental exposure data for air, water and soil. The potential for indirect exposure via the food chain is also addressed.

The report also summarises the compositions, hazards and occupational exposure limits for kerosines and jet fuels, and gives supply and consumption figures for the European Union over the years 1992 to 1996.

#### **99/53 SCIENTIFIC BASIS FOR AN AIR QUALITY STANDARD FOR NICKEL**

The health effects due to nickel exposure are reviewed in this document for the purposes of understanding the scientific basis for an Air Quality Standard (AQS). The report provides an overview of two approaches for deriving a nickel AQS by presenting both a threshold/safety factor and a linear non-threshold approach. Using a threshold/safety factor approach with a lowest observed-adverse-effect-level (LOAEL) of 1000  $\mu\text{g Ni/m}^3$  for soluble nickel compounds based on the occupational epidemiology data, exposure limits of 0.6  $\mu\text{g Ni/m}^3$  for soluble nickel compounds and 6  $\mu\text{g Ni/m}^3$  for less soluble compounds (e.g., oxidic and sulfidic compounds), as an annual average, are justified on scientific grounds. Use of a single value of 0.6  $\mu\text{g Ni/m}^3$  (annual average) is recommended since it would be protective for both soluble and less soluble nickel compounds. By contrast, exposure limits derived using the animal and human data and a linear non-threshold approach are approximately an order of magnitude lower and range from 0.01-0.03  $\mu\text{g Ni/m}^3$  (annual average) for both soluble and insoluble compounds.

In general, nickel compounds are not acutely toxic. However, nickel is a proven sensitizer; therefore nickel compounds should be regarded as potential sensitizers. The primary hazard associated with exposure to certain nickel compounds is the ability to adversely affect the respiratory system and to produce respiratory cancers. This effect is consistent between animals and humans. Oral exposures do not appear to result in the biological activities exhibited by the inhalation route. Because inhalation is the most pertinent exposure route for an AQS and inhalation is the



most toxic route for nickel compounds, inhalation-related health effects (excluding nickel carbonyl) and the bioavailability of the relevant forms of these compounds to relevant target tissues are considered most appropriate. In particular, the critical effect associated with nickel exposure, for the purposes of setting an ambient air standard, is respiratory cancer.

The choice of human versus animal datasets for deriving a nickel AQS does not strongly influence the final recommended value. However, the choice of extrapolation method (i.e., a threshold/safety factor versus linear non-threshold approach) has an impact on the recommended AQS value. A common weakness of both approaches is that the characteristics of nickel exposures in animal and human studies are qualitatively different from exposures present in ambient air. Specifically, the strongest respiratory cancer associations identified in the human and animal studies are with sulfidic and oxidic nickel; sulfidic nickel is not likely to be detected in ambient air while nickel oxide may be present up to a maximum of 8%.

Overall, the weight of the evidence strongly supports the use of a threshold/safety factor approach for deriving a scientifically justified and defensible AQS for nickel. Factors that support the use of the threshold/safety factor approach include the following:

- the existence of a NOAEL for pulmonary inflammation of 130  $\mu\text{g Ni/m}^3$  (nickel dust) derived from inhalation studies,
- the lack of carcinogenic response in the absence of pulmonary inflammation,
- the presence of good evidence for an empirical threshold for lung and nasal cancer in epidemiology studies of occupationally exposed groups,
- the presence of a NOAEL for pulmonary carcinogenesis of approximately 100  $\mu\text{g Ni/m}^3$  observed in recent rodent studies conducted with three nickel compounds administered by inhalation, the relevant route of exposure,
- no evidence of genotoxicity in *in vivo* studies conducted by the inhalation route of exposure in humans and rodents.

Although the finding of genotoxicity from *in vitro* studies of nickel compounds may support the use of a default linear non-threshold approach, the combined weight of the evidence suggests that this would likely produce an unduly conservative AQS value. Accordingly, a value of 0.6  $\mu\text{g Ni/m}^3$  (annual basis) is recommended as an AQS for nickel to protect the general public against carcinogenic and other potential hazardous effects associated with exposure to both soluble and less soluble nickel compounds. This value is derived from the threshold/safety factor approach.

#### 99/54 **OVERVIEW AND CRITIQUE OF THE AIR POLLUTION AND HEALTH: A EUROPEAN APPROACH (APHEA) PROJECT**

The relationship between air pollution and adverse health outcomes is a subject of current concern in the area of environmental health. The Air Pollution and Health: a European Approach (APHEA) project is a co-ordinated study of the short-term effects of air pollution on mortality and hospital admissions in sixteen cities in eleven European countries. The Institute for Environment and Health have been requested

by CONCAWE to carry out an independent review and critique of the APHEA project. Twenty-three published papers were identified and reviewed.

### **Methodology**

A standardised protocol was developed and adopted by all centres in the APHEA project for the collection of air pollutant data, health outcome data and information on potential confounders, such as temperature, relative humidity and unusual events. Poisson regression, allowing for autocorrelation and overdispersion was used by each centre to analyse their own data and the procedure for the building statistical models was specified in detail. Each city fitted the model of 'best fit' to their data, so that the city-specific models differed in the number of variables they included and the transformations and lag times used for the time-dependant variables. The meta-analyses combined the relative risks obtained from the best fitting models for the individual cities to give overall summary estimates. A fixed effects model was first fitted and a test of heterogeneity was carried out. If heterogeneity was significant, a random effects model was then fitted, with explanatory variables.

### **Results**

- Small increases in the relative risks were consistently found in the individual cities and in the combined analyses for several air pollutants and health outcomes
- The relative risks for total mortality ranged from 0.98 to 1.13, most being between 1.01 and 1.03.
- The strongest effect for total mortality was found for sulphur dioxide (SO<sub>2</sub>), with particulates showing a slightly weaker association.
- Consistently elevated relative risks were found for both nitrogen dioxide (NO<sub>2</sub>) and ozone (O<sub>3</sub>) and the estimates from the combined analyses were statistically significant.
- The results for mortality attributable to respiratory disease and cardiovascular disease were less consistent with a wider variation in risk estimates between cities and few being significantly elevated.
- Similar patterns were found for digestive system diseases, chosen as a control group, although risk estimates tended to be lower.
- Increased risk of hospital admissions for respiratory disease was found for SO<sub>2</sub> in the elderly (aged 65 and over), and for black smoke (BS) in the 15-64 age group.
- Results for NO<sub>2</sub> and respiratory disease admissions were inconsistent.
- A significantly positive association was found for respiratory admissions in London with O<sub>3</sub>, and the combined estimate was significantly raised and was larger in the elderly.
- A combined analysis from four cities for emergency hospital admissions for asthma found associations with NO<sub>2</sub> in both children under the age of 15 years and adults aged from 15-64 years, and for SO<sub>2</sub> in children that were significantly raised.

- Emergency admissions for chronic obstructive pulmonary disease were significantly associated with all pollutants using the combined estimates, with the strongest and most consistent effect being found for O<sub>3</sub>.
- The effects of temperature and season varied between cities and by pollutant and health outcome. There was a tendency for effects to be stronger in the summer.
- Correlations between pollutants within cities and the extent to which synergy between pollutants existed also varied considerably.

### **Critique**

The systematic and co-ordinated approach is one of the strengths of the APHEA project, and the results have contributed to the previous lack of meaningful information in Europe. The results support those found in many other studies of the adverse effects of air pollution. There was a tendency, however, for there to be a stronger association with SO<sub>2</sub>, than studies, for example from the USA, and risk estimates for particles were generally lower than those obtained in the American studies.

However, the project was limited by the quality of some of the data, in particular the exposure information. There were variations in the numbers of monitors per geographical area and per population, the location of the monitors, the measurement and sampling techniques, and the correlations between values from the monitors. Ecological studies such as this are subject to a high degree of misclassification due to the use of ambient air exposure data as a surrogate for personal exposure. There were also problems of completeness of diagnosis for some of the hospital admission data, and it was not possible to separate emergency from planned admissions in some centres.

The published papers do not report the results in a standardized format. Information important for the interpretation and comparison of the results between the cities was not consistently presented. For example, it would have been useful to know, for all the cities, the values of the correlations between pollutants measured in order to assess potential collinearity, the risk estimates for all the covariates included in the models, changes in the values of the coefficients of the pollutants as variables were added to or removed from models, and an indication of the fit of the models. The correct adjustment for potentially strong confounders, such as those relating to climatic conditions, is particularly important when elevation of the relative risk estimates is so small. Variables, such as population size, age structure, migration patterns and proportion of smokers, which might have undergone change over the study periods and influenced either health or pollution or both, were described for some cities but were not considered as variables for inclusion in the models. Other time-varying factors, such as pollen, which might influence respiratory diseases such as asthma, were not included in the analyses.

The lack of homogeneity in some of the data and the use of risk estimates derived from the 'best fit' models, with varying choice of variables, lag times and transformations, necessitates caution when evaluating the summary estimates obtained from the meta-analyses.

### **Recommendations**

Although the results from the APHEA project concur with many other studies in showing an association between a range of airborne pollutants and adverse health outcomes, they have also helped to emphasise the wide variation in both the magnitude and direction of the estimates and the predominant pollutants.

Clarification of the relationship between levels of air pollution measured by outdoor fixed-site monitoring, levels indoor and personal exposures is required. The role of each pollutant, both as an individual risk factor and in its contribution to a synergistic effect, the importance of different sources and mixtures of pollution, the interrelationships of pollution and climate and the influence of microenvironments are all areas which require further research. The development of a European air pollution monitoring network, which uses the same method of sampling and analysis and is more evenly distributed by area and population is also essential.

In view of the lack of consistency of reporting of the results from the APHEA project, a paper summarising these in a standard format, as suggested earlier, for all cities, pollutants and health outcomes, would be useful. The result for diseases of the digestive system should also be given for all cities to assist comparisons. Although there are limitations in the APHEA data, reanalyses of the combined data sets using the raw data from all the relevant centres, would be helpful. Lag times, transformations and weights could be fitted, and explanatory variables and interaction terms characterising the individual centres could also be included.

Ecological study designs are limited in their inherent assumption that individual risk can be estimated using group data. Spurious associations may be identified through an unobserved variable or by incomplete adjustment for confounding. Collinearity between pollutants may make it difficult to identify exactly which pollutant is important and there may be incomplete knowledge about the time-exposure-response relationship. The regression coefficients obtained from the models used cannot be used to quantify the effect of pollution. A Cohort study approach is needed to assess causality and also estimate the potential public health benefit of a reduction in pollution.

**99/55 EU OIL REFINING INDUSTRY COSTS OF CHANGING GASOLINE AND DIESEL FUEL CHARACTERISTICS**

CONCAWE has studied the many aspects important when considering fuel quality. The report reviews the complex interactions between fuels, vehicle technology, test cycles and reference fuels with regard to their relative influences on vehicle emissions, fuel consumption, CO<sub>2</sub>, durability and customer acceptance. Implications for the refining industry and trends in vehicle technology are also discussed as these are fundamental for a cost effective approach to contribute to meeting air quality standards.

The study concludes that effects of fuel changes alone on emissions and performance are relatively small, but benefits arise when they are used to enable new technologies. Therefore, fuels and engines need to be developed together as a common system. Such developments have to be assessed in view of their global impact on a "cradle to grave" basis. To produce sufficient quantities of fuel, flexibility of the refineries has to be ensured by specifying fuel properties only where a clear link to vehicle performance or emissions is proven. Harmonising fuel specifications can only go in parallel with emissions limits, vehicle technology and test cycles.

CONCAWE believes that more joint industry technical programmes, such as EPEFE, AQIRP and JCAP, are required to expand the existing sound scientific database to the rapidly developing new technologies.

**99/56 EU OIL REFINING INDUSTRY COSTS OF CHANGING GASOLINE AND DIESEL FUEL CHARACTERISTICS**

This report presents the results of a study to assess the EU-15 refining industry implications of changing gasoline and diesel fuel characteristics, for year 2005 onwards. The study's starting point is the EU Council Common Position on the AO1 proposed year 2000 road fuels, i.e. gasoline aromatics 42%, sulphur 150 ppm, olefins 18% and diesel cetane number (CN) 51, sulphur 350 ppm.

The costs to oil refiners and the CO<sub>2</sub> emissions effects have been calculated with regards to the changes to gasoline and diesel fuel characteristics given in the Fuels Directive (98/70/EC) agreed in Conciliation. These are for gasoline: sulphur content from 150 ppm to 50 ppm and aromatics content from 42% to 35%; for diesel: sulphur content from 350 ppm to 50 ppm. In addition an increase in CN from 51 to 55 and further towards 58, if achievable, is reported on.

The complex interactive analysis was carried out using a purpose built supply/demand refinery LP model featuring four refinery types and seven regions. This degree of definition is essential to reduce the over optimisation of such models which otherwise seriously underestimate effects. On the other hand, some component transfers between refinery types are allowed. This approach results in some equalisation of qualities (especially aromatics and olefins contents of gasoline) and provides some low cost networking solutions. The results are published in this report and in the associated detailed tabulations available as computer files to assist in the analysis of fuel related vehicle emissions measures.

The single parameter cost for reformulation depends on the sequence applied as a result of synergy or antagonism between the required processing needs.

The CO<sub>2</sub> emissions effects are in the unwanted direction, reflecting additional fuel and hydrogen in processing.

**99/57 ENVIRONMENTAL LEVELS OF BENZENE AT THE BOUNDARIES OF THREE EUROPEAN REFINERIES**

A year-long study of benzene-in-air concentrations at the boundaries of three refineries has been carried out.

The sampling technique employed was passive diffusion tubes. Sampling locations were fixed at 12 or 16 points around the refineries.

Continuous samples were collected for 26 two-week periods to provide the annual averages for each sampling location. Two-weekly wind direction data were also gathered at each refinery.

The results reflect the relative sizes and complexities of the three refineries. Variations between the two-weekly samples reflect changes in wind direction and occasional operational events and incidents.

**99/59 A TEST METHOD TO ASSESS THE 'INHERENT' BIODEGRADABILITY OF OIL PRODUCTS**

This report describes the development of a test method for assessing the potential of oil products to be biodegraded in the environment ('inherent' biodegradability). The method produced was based on International Standard ISO 14593 ('CO<sub>2</sub> Headspace Test') but used a pre-exposed inoculum and an extended test duration. The work of the CONCAWE Biodegradation Task Force is summarised and details are given of an international ring test which demonstrated the practicality of the new method and determined its precision. As a preliminary to submission of the method to the Organisation for Economic Co-operation and Development (OECD) for consideration as a new test guideline for assessing 'inherent' biodegradability, the method has been prepared in 'OECD format' (included as an appendix to this report).

During the course of the Task Force's work, CONCAWE funded a one year project with the University of Hull to evaluate techniques for measuring the incorporation of test substance carbon into new microbial biomass during biodegradation under the conditions of the CONCAWE test method. The report also describes this research and summarises its main findings.

**99/60 THE HEALTH EFFECTS OF PM<sub>2.5</sub> (INCLUDING ULTRAFINE PARTICLES)**

A review has been prepared of the health effects of exposure to PM<sub>2.5</sub> particles (aerodynamic diameter < 2.5 µm), including so-called ultrafine particles (aerodynamic diameter < 0.1 µm). The report covers briefly sources, composition and characteristics of PM<sub>2.5</sub>. It covers in more detail the dosimetry of inhaled particles, and toxicological and epidemiological investigations. A series of conclusions is presented.

**99/61 THE EUROPEAN UNION LEGISLATIVE PROCESS**

This report updates Report 95/64 and describes the legislative process in the European Union, as amended by the Amsterdam Treaty of 1997. The legislative process in the EU is illustrated in the form of a flow chart.

**99/62 POTENTIAL OF EXHAUST AFTER TREATMENT AND ENGINE TECHNOLOGIES TO MEET FUTURE EMISSIONS LIMITS**

In view of the European Auto Oil Process (AO2) a CONCAWE study reviewed literature (up to 9/1998) and in-house information on improved exhaust after treatment and engine technologies to reduce emissions from gasoline (stoichiometric, lean-burn, G-DI) and diesel vehicles (light and heavy duty). Engine features include advances in engine combustion, fuel injection equipment, exhaust gas recirculation systems and management systems/strategies. The various technologies are summarised with regard to their application, advantages, disadvantages, fuel implications, fuel consumption and their likelihood to emerge on the European market.

Based on the knowledge available at the time of the study, CONCAWE's experts assessed the potential of different technologies to contribute to meeting the year 2005 emissions limits. While it is concluded that vehicle technologies already exist to meet such limits even with current fuel quality, for conventional gasoline vehicles,

no current light duty diesel vehicle achieves these limits. For light and heavy duty diesel vehicles, several technologies exist (traps, de-NOx) which will be further developed, and fuel quality will depend on the conversion efficiency and durability needs of such catalyst technology to meet 2005 limits and beyond. The SCR (urea de-NOx) system would be expected to be sulphur tolerant and applicable for heavy duty engines. Continuous regenerative traps (CRT) should operate satisfactorily with the 2005 sulphur level (50 mg/kg). De-NOx catalyst systems required for gasoline direct injection systems (G-DI) are under rapid development and improvements in their sulphur tolerance could be expected. In the Fuels Directive 98/70/EC fuel sulphur has been specified to enable new technologies to meet future emission limits.

**00/51 THE OCCURRENCE OF SELECTED HYDROCARBONS IN FOOD ON SALE AT PETROL STATION SHOPS AND COMPARISON WITH FOOD FROM OTHER SHOPS - A LITERATURE SURVEY**

This study presents the results of a review of reports on the occurrence of some hydrocarbons in food in relation to the sales location, with a particular emphasis on petrol station shops. The principal selected hydrocarbons are volatile components of gasoline. The reported findings are discussed and compared, and put in the context of total daily exposure. The relevance of gasoline sales to the levels of the selected hydrocarbons in food items is assessed and compared with other sources of the hydrocarbons.

**00/52 MANAGEMENT OF OCCUPATIONAL HEALTH RISKS DURING REFINERY TURNAROUNDS**

This report addresses the occupational health issues which are an intrinsic part of refinery turnaround activities. It recognises the underlying importance of an effective management system and is intended to assist planners, managers and specialists to achieve a high standard in controlling health risks. Health is viewed within the framework of a generic management system that can be applied to the various phases of turnarounds, from planning to close-out. The report focuses on health risk management, prevention of risk, health surveillance, the employment of contractors and skill requirements.

Hazards to health in the various phases of a turnaround are identified, the risks listed according to severity and control strategies suggested. General guidance is also given on monitoring, health surveillance and the reporting of results.

While it is appreciated that safety and environmental issues are also encountered in turnarounds, the report focuses primarily on the occupational health risks.

**00/53 AN ASSESSMENT OF THE REPRODUCTIVE TOXICITY OF GASOLINE VAPOUR**

The reproductive toxicity of gasoline vapour in rats was studied in order to generate relevant toxicity information for gasoline in humans via the inhalation route. The two-generation study was designed and conducted according to international guidelines. Exposure levels were 0 and approximately 5000, 10 000 and 20 000 milligram per cubic metre. No treatment-related adverse effects relevant to humans were observed. It was concluded that the No-Observed-Adverse-Effect-Level for reproductive toxicity in rats in this study was 20 000 milligram per cubic metre.

**00/54      IMPACT OF A 10 PPM SULPHUR SPECIFICATION FOR TRANSPORT FUELS ON THE EU REFINING INDUSTRY**

Production of road fuels to a 10 ppm sulphur specification is feasible but costly both in terms of refinery investments and CO<sub>2</sub> emissions. Other fuel properties would not be significantly affected.

**00/55      A REVIEW OF TRENDS IN HEARING THRESHOLDS OF EUROPEAN OIL REFINERY WORKERS**

The change of hearing thresholds was assessed in a study population of over 1000 noise-exposed oil refinery workers subject to noise at work regulations in Member States of the European Union. Audiometric data covering a period of approximately 12 years were retrieved from refinery occupational health departments. The data were screened using established procedures, standardised for age according to international guidelines and subjected to statistical analysis to identify time trends and differences between subgroups of younger and older workers. From the analyses of the data it was concluded that the workers' hearing did not deteriorate more than expected from natural ageing.

**00/56      REVISED DANGEROUS PREPARATIONS DIRECTIVE (1999/45/EC) -- IMPLICATIONS FOR PETROLEUM PRODUCTS**

This report highlights and explains the significance of the changes regarding the classification and labelling of preparations introduced by this revision (1999/45/EEC) to the Dangerous Preparations Directive, including the classification and labelling of preparations for environmental endpoints, revised disclosure requirements, Safety Data Sheets and implications for distance selling.

**01/51      MEASUREMENT OF THE NUMBER AND MASS WEIGHTED SIZE DISTRIBUTIONS OF EXHAUST PARTICLES EMITTED FROM EUROPEAN HEAVY DUTY ENGINES**

This study investigates the measurement of the mass and the number of heavy duty automotive particle emissions and their related size distributions. Limited additional test work has investigated the effect of sampling and measurement conditions on these distributions. Two engines representing Euro 2 and Euro 3 technology were examined, using a selection of diesel fuels representative of European market quality.

**01/52      A NOISE EXPOSURE THRESHOLD VALUE FOR HEARING CONSERVATION**

As part of a proposed EU directive for health and safety at work, a noise exposure threshold level of 75 decibels (A-weighted) over an 8-hour working day has been specified. This is stated to be the value below which continuous and/or repetitive exposure has no adverse effect on the health and safety of workers. This review considers the scientific background to the current international standard used to set the threshold level. Other literature data are reviewed, taking into account hearing loss due to the combination of natural ageing and exposure to noise. It is concluded that a threshold level of 75 dB(A) is overly conservative and that a daily noise exposure threshold level of 80 dB(A) can deliver suitable hearing protection.



**01/54 ENVIRONMENTAL CLASSIFICATION OF PETROLEUM SUBSTANCES - SUMMARY DATA AND RATIONALE**

Environmental data on the fate and effects of petroleum substances are summarised. Technical issues relating to the choice of test methodology for the evaluation of environmental impacts of petroleum substances are discussed. Proposals for self-classification according to EU criteria, as defined in the Dangerous Substances Directive, are presented for individual petroleum substance groups, on the basis of available test data and structure activity relationships (based on composition).

**01/56 AN ASSESSMENT OF OCCUPATIONAL EXPOSURE TO NOISE IN THE EUROPEAN OIL INDUSTRY (1989-1999)**

Daily personal noise exposure data available from member companies for European oil industry work environments (refinery, product distribution and others) were collected and analysed. Comparisons are made with historical data and with current EU exposure limits. The impact of a possible lowering of exposure limits is examined.

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