concawe

ENVIRONMENTAL SCIENCE FOR THE EUROPEAN REFINING INDUSTRY

review

Volume 25, Number 1 • July 2016

Environmental science for the European refining industry

Cover photograph reproduced courtesy of Jean Schweitzer/iStockphoto.

Editor: Robin Nelson, Concawe

Design and production: Words and Publications • words@words.co.uk

Foreword

Robin Nelson
Science Director
Concawe

The first three articles in this edition of the Concawe *Review* reflect the origins of the Concawe name, derived from CONservation of Clean Air and Water in Europe. It is clear that the issues that led to the formation of Concawe in 1963 are just as relevant to the petroleum refining industry and the (European) society which it serves today, as in 1960.

In December 2014 the Concawe Oil Pipelines Management Group (OPMG) reported a marked increase in the incidence of product theft from the EU pipeline network. In the fourth *Review* article data from pipeline operators is presented that confirms the scale of this problem, with the annual number of theft-related incidents increasing from 4 in 2010 to more than 150 in 2015.

To conduct our research Concawe draws on recognized experts within our member companies, working together with scientists from leading research institutes and universities. We also are increasingly working to develop staff, who we hope will become future experts with the energy and commitment to continue the work of Concawe for as long as oil products are valued and used by society. As such we thank two of our recent Research Associates, Catarina Caiado and Charlene Lawson for their contribution over the past couple of years and wish them every success upon return to their parent companies.

Contents

For both atmospheric particulates and nitrogen oxides (NOX), the primary focus for emission reductions at both national and local levels is road transport. Against this background, it is vital that the current and future contribution of road transport, and in particular diesel road transport to overall urban air quality in Europe is quantified to provide an appropriate perspective for effective further action at European, national and local levels. The impact of successive improvements in vehicle emission standards which have taken place over the past 15 years, together with the further impact of Euro 6 requirements (commencing September 2015) also needs to be understood. These Euro 6 requirements include the impact of the forthcoming testing regime based on the recently agreed real driving emissions (RDE) conformity factors (CF). Concawe recently commissioned a study to better understand the air quality compliance issues for PM and NO2 in the EU27 countries, with a particular focus on the urban environment. This article discusses some of the results of this study and suggests ways in which compliance can be improved.

Enquiries to: heather.hamje@concawe.org; lucia.gonzalez@concawe.org

Two modern gasoline direct injection (GDI) light-duty vehicles have been tested to investigate the effect of oxygenates (mainly ethanol) on particulates (both mass and number), fuel economy and regulated emissions. The GDI vehicles used in this study met Euro 4 and Euro 5 emissions limits and were tested over the New European Driving Cycle (NEDC) using ethanol-containing gasolines at different oxygen levels and RON values. An ether-containing blend was also tested for comparison. Ethanol and ether blends with matched oxygen content and RON values, and splash blended fuels (in the case of ethanol), were also prepared for testing. Fuels were tested in duplicate using a randomized test order to improve statistical certainty. This article discusses the results of these tests, and the effect of matched and splash-blended oxygenates on both particulate matter (PM) and particulate number (PN).

Enquiries to: heather.hamje@concawe.org

In recent years, increasing quantities of fatty acid methyl esters (FAME) have been blended into automotive diesel fuel to meet the EU's 2020 mandate for renewable energy. One potential consequence of the increased use of FAME is a change in the natural attenuation behaviour of any spilt diesel fuel that reaches the subsurface. To address this uncertainty, Concawe has completed a review of the scientific literature on the physical and chemical properties of FAME, and on its transport and biodegradation behaviour in soil and groundwater. This article presents the main findings from this study, which is described in full in Concawe report 5/16.

Enquiries to: mike.spence@concawe.org

The impact of emission reduction scenarios on air quality limit values

A Concawe study aims to improve understanding of air quality compliance issues for PM and NO_2 in the EU-27 countries.

Despite considerable improvements in European air quality resulting from the progressive implementation of emission reduction measures over the past decade, non-compliance with specific ambient air quality limit values set forth in the Ambient Air Quality Directive (2008/50/EC) persists. The recent revision to the Thematic Strategy on Air Pollution (TSAP) and the accompanying package of measures proposed by the European Commission[1] have taken steps to address this issue, identifying both particulate matter ($PM_{2.5}$ and PM_{10}) and nitrogen dioxide (NO_2) as requiring attention.

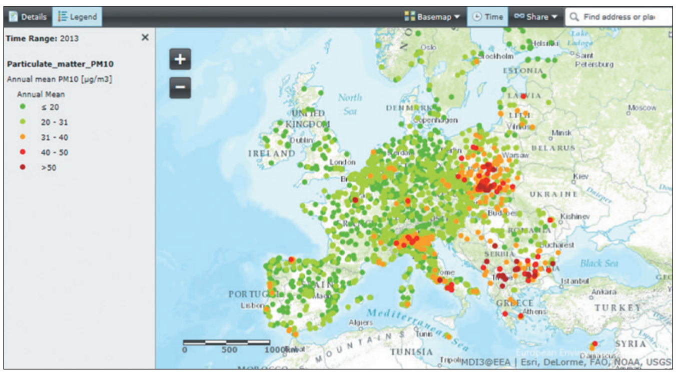
For both atmospheric particulates and nitrogen oxides (NO_X), the primary focus for emission reductions at both national and local levels is road transport. Against this background it is vital that the current and future contribution of road transport, in particular diesel road transport, to overall urban air quality in Europe is quantified to provide an appropriate perspective for effective further action at European, national and local levels. The impact of successive improvements in vehicle emission standards which have taken place over the past 15 years, together with the further impact of Euro 6 requirements (commencing September 2015) also needs to be understood. The Euro 6 requirements include the impact of the forthcoming testing regime based on the recently-agreed real driving emissions (RDE) conformity factors (CF).[2]

Concawe recently commissioned a study, conducted by Aeris Europe, to better understand the air quality compliance issues for PM and NO_2 in the EU-27 countries, with a particular focus on the urban environment. The emissions inventory and projections (IIASA, 2015) considered in the Base Case are the most up-to-date European estimates available at the time of writing but do not take into account the effects of legislation for which the actual impact on future activity levels could not be quantified[3]. As a result, the Base Case should be considered as conservative with respect to anticipated emissions reductions. Road transport emissions have been calculated using the fleet projections included in the TREMOVE 'alternative' scenario and the emission factors of COPERT v4.11, representing a Euro 6 diesel passenger car NO_X emissions conformity factor of approximately 2.8[4].

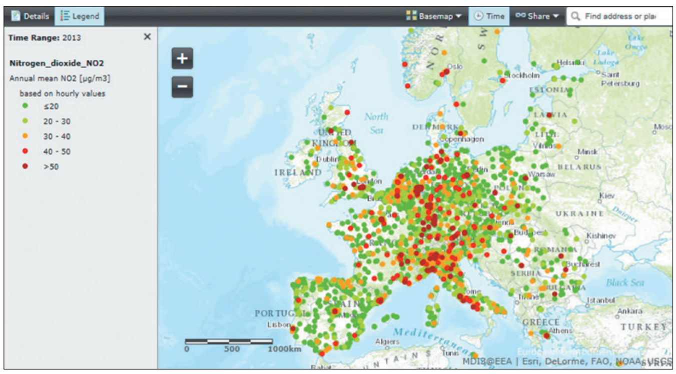
The study was undertaken in two phases. The first phase (Scenarios A to D) was aimed at understanding the maximum possible improvements in $PM_{2.5}$, PM_{10}, and NO_2 compliance from taking action that targets road transport and domestic combustion. This included

Figure 1 Monitoring station data showing PM_{10} and NO_2 compliance, 2013

a) PM_{10} compliance



b) NO_2 compliance



Maps reproduced courtesy of the European Environment Agency

[1] December 2013.

[2] A conformity factor is a multiplication coefficient of the NO_X emissions legislated limited value: (0.08gNO_X/km) for Euro 6 diesel passenger cars (PCDs).

[3] Including the Medium Combustion Plants Directive (MCPD) and the review of the National Emissions Ceilings Directive (NECD).

[4] The COPERT 4 methodology is part of the *EMEP/EEA Air Pollutant Emission Inventory Guidebook* for the calculation of air pollutant emissions. The emission factors generated are vehicle- and country-specific. The PCD NO_X Conformity Factor of 2.8 is therefore an indicative value identified to allow for comparison to the Real Driving Emissions legislation.

exploring some extreme 'beyond the Base Case' scenarios, for example the hypothetical immediate replacement of all diesel-powered road transport with zero exhaust emission vehicles (Scenario B). In the specific context of PM_{10}/$PM_{2.5}$ compliance, given the increasing use of wood burning as a renewable fuel and the continued use of coal in the domestic sector in a number of Eastern European Member States, the impact of a complete removal of solid fuel burning emissions from the domestic sector was also explored (Scenario A).

The second phase (Scenario E) focused on NO_2 compliance and the contribution from diesel passenger cars (PCDs). This included exploring the impact on NO_2 compliance of varying degrees of conformance with legislated Euro 6 emission limits under real driving conditions. An overview of the scenarios explored in this study is included in Table 1.

The study utilised a suite of emission and air quality modelling tools developed and maintained by Aeris Europe which together facilitate the assessment of $PM_{2.5}$, PM_{10} and NO_2 air quality compliance at individual monitoring station level for the whole of the EU. The modelling approach is semi-empirical, drawing on detailed historical data from more than 3,000 monitoring stations in the EEA AirBase database[5] together with other exogenous inputs used to support air policy development in Europe, including national emissions inventories and transboundary source-receptor data.

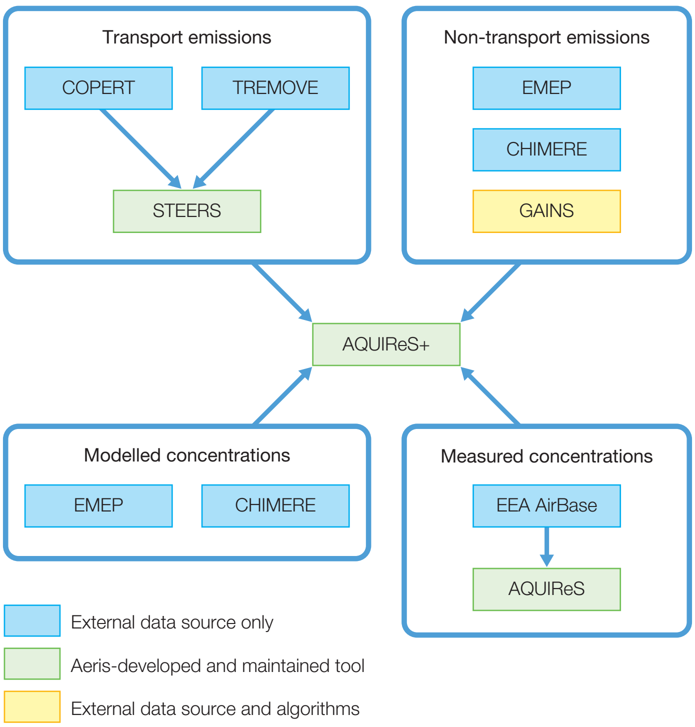
Table 1 Overview of scenarios explored in this study

Scenario		Description	Euro 6 conformity factor[4,6]
Ambient air quality standard, $PM_{2.5}$		Air quality limit value of 20 µg/m³ for $PM_{2.5}$ from 2020	-
A		Removal of solid fuel combustion from the domestic sector by 2020	-
B		Removal of all diesel exhaust emissions from urban areas by 2020	2.8
C	C1	Acceleration of older vehicle replacement: 100% of pre-Euro 5 vehicles replaced with Euro 6 vehicles in each horizon year (2020, 2025, 2030)	2.8
	C2	Acceleration of older vehicle replacement: 25% of pre-Euro 5 vehicles replaced with Euro 6 vehicles in each horizon year (2020, 2025, 2030)	2.8
D	D1	Removal of exhaust emissions from all diesel passenger cars (PCDs) in the urban environment by 2020	2.8
	D2	As scenario D1, additionally removing light-duty diesel vehicles (LDVs)	2.8
	D3	As scenario D2, additionally removing heavy-duty diesel vehicles (HDVs)	2.8
	D4	As scenario D3, additionally removing buses (BUS)	2.8
E	BC0	Scenario E Base Case	2015 onwards: 2.8
	SN1a	These scenarios consider different Euro 6 performance levels and the effect of improving performance by specific dates	2015–2020: 7 2020 onwards: 2.8
	SN1b		2015–2020: 7 2020 onwards: 1.5
	SN1c		2015–2017: 7 2017 onwards: 1.5
	7xLLV		7
	ZEPCD	All diesel passenger cars registered from 1 January 2015 to produce zero NO_X emissions	

[5] AirBase is the air quality information system maintained by the EEA. It contains air quality data from networks and individual stations measuring ambient air pollution within the Member States delivered annually under European Council Directive 97/101/EC.

[6] ICCT (2014).

Figure 2 Schematic of the tools used by Aeris Europe for this study



The robustness of the modelling approach was verified by hind-casting and comparing the predicted concentration levels with historical measurement data from the EEA AirBase database. The tools used in this study are shown in Figure 2.

Air quality management zones (AQMZs) are designated under the ambient air quality directive (2008/50/EC) and oblige Member States to divide their entire territory into zones. Zones can be regarded as the primary territorial units for assessment and management of air quality under the air quality directive. The compliance of individual stations within each zone is used to determine overall zone compliance; specifically, the single least compliant station is chosen for $PM_{2.5}$ and NO_2. This means that zone compliance is reflective of the 'worst' compliance situation within that zone. While AQMZs are intended to be representative of the air quality over the entire area covered it is likely that a single station modelled as non-compliant will result in the entire population of a zone being interpreted as exposed to levels of PM or NO_2 above the limit value. Given that a zone may have a population of 500,000 or more and a traffic station may be measuring an area as small as $200m^2$, exceedance at the traffic station level clearly cannot be taken to be indicative of population exposure within a whole zone. No attempt has been made to allow for this circumstance and detailed analysis of population exposure needs to be undertaken with care.

Some zones are excluded from the modelled results; this is due either to the zone containing no measuring stations or any measuring stations present lacking the required prerequisites for inclusion in the model.

Key findings: particulates ($PM_{2.5}$ and PM_{10})

Primary $PM_{2.5}$ and PM_{10} emissions from road transport

This study highlights the diminishingly small contribution from the exhaust of road transport to overall PM concentrations between now and 2030. By 2020 non-exhaust[7] emissions emerge as the dominant emission from road transport (albeit small in terms of its contribution to the total concentration) and, by 2030, primary $PM_{2.5}$ emissions from road transport are essentially independent of

Table 2 Contribution from road transport to total ppm emissions in EU-27 countries: kilotonnes (% of total)[8]

		2015	2020	2025	2030
PM_{10}	Road-transport exhaust emissions	77 (4%)	38 (2%)	21 (1%)	15 (1%)
	Road-transport non-exhaust emissions	149 (7%)	186 (9%)	199 (11%)	208 (11%)
$PM_{2.5}$	Road-transport exhaust emissions	77 (5%)	38 (3%)	21 (2%)	15 (1%)
	Road-transport non-exhaust emissions	50 (4%)	53 (4%)	54 (5%)	56 (5%)

[7] Brake, road and tyre wear: these sources are present in all road transport including 100% battery-powered vehicles.

[8] All road transport exhaust emissions are $PM_{2.5}$—this fraction is included in the PM_{10} emissions total.

the powertrain, meaning that all vehicles, regardless of motive force, would produce equivalent ppm emissions.

$PM_{2.5}$ compliance with air quality limit values

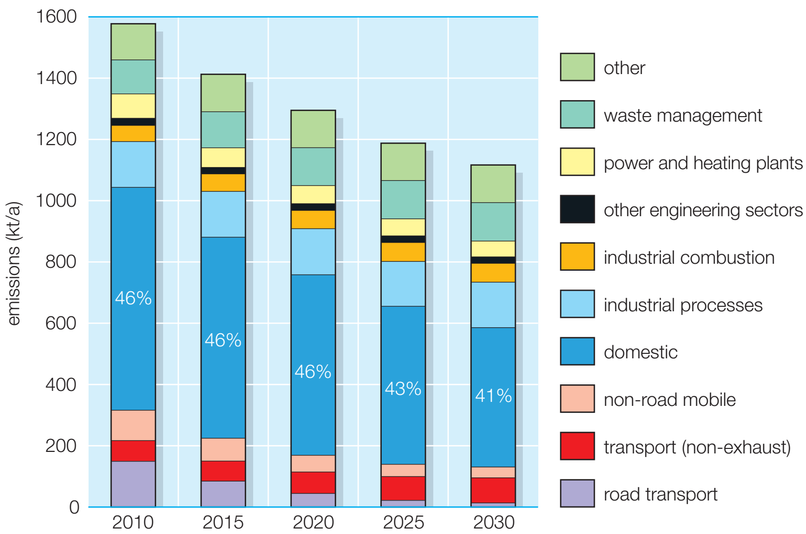
The Base Case results indicate that, in 2015, the percentage of the EU population living in 'likely non-compliant' zones is only 4%; with 68% of the population in 'likely compliant' zones and 28% of the population living in zones that are close to the AQLV (within zones of 'uncertain compliance'). The EU population living in zones of 'uncertain compliance' continues to decline between 2015 and 2030 as already-legislated measures take effect so that the population living in 'likely compliant' zones increases to 77% by 2020 and to 81% by 2030. At the same time, the population living in zones of uncertain compliance reduces to 19% by 2020 and to 15% by 2030. The percentage of population living in 'likely non-compliant' zones remains unchanged at 4% from 2015.

Most of the residual $PM_{2.5}$ non-compliance is seen in Eastern Europe and is attributable to the ppm emissions from domestic combustion of solid fuels that continues to take place in this region of the EU. The results from Scenario A, where solid fuel (e.g. coal, wood) burning in the domestic sector is replaced by 'low ppm' generating fuel (e.g. gas or heating oil), indicates that this measure would improve the compliance picture for both $PM_{2.5}$ and PM_{10}.

Diesel exhaust PM (Scenario B)

Given the small contribution of exhaust emissions from road transportation to total PPM emissions the elimination of diesel vehicles neither improves the overall air quality compliance picture in the future nor does it accelerate the achievement of improved compliance; this is shown in Table 3.

Figure 3 $PM_{2.5}$ emissions aggregated by key sector for EU-27 countries



Domestic solid fuel combustion (Scenario A)

In contrast to the negligible effect that diesel exhaust emissions have on PM concentrations, the removal of solid fuel combustion and its replacement with gas or heating oil shows a marked difference in the proportion of the EU population that are in zones which are borderline compliant for both $PM_{2.5}$ and PM_{10}, resulting in a significant overall improvement in air quality by 2020 and beyond (Table 3). The difference is particularly evident in the case of PM_{10} where approximately 92% of the EU population would live in 'likely compliant' areas and less than 1% in 'likely non-compliant' areas by 2025.

The greatest improvement is observed in countries with high levels of solid fuel burning, particularly Eastern Europe and suggests that those countries experiencing PM_{10} compliance issues could significantly reduce this

Table 3 Percentage of EU-27 population living in zones achieving compliance with ambient air quality standards for $PM_{2.5}$

Base Case v. Scenario A (reducing PPM emissions from solid fuel combustion) v. Scenario B (removing all diesel exhaust emissions)

	EU population living in 'likely compliant' zones			EU population living in 'likely non-compliant' zones		
	Base case	Scenario A	Scenario B	Base case	Scenario A	Scenario B
2015	68%	68%	68%	4%	4%	4%
2020	77%	85%	77%	4%	3%	4%
2025	80%	88%	80%	4%	3%	4%
2030	81%	89%	81%	4%	3%	4%

problem by reducing or eliminating solid fuel combustion in the domestic sector.

Key findings: nitrogen dioxide (NO_2)

NO_2 compliance with air quality limit values in the Base Case

The Base Case modelling results indicate that in 2015 the percentage of the EU population living in 'non-compliant' zones (modelled concentration above 45 g/m^3) is approximately 18%, while 69% of the population live in 'likely compliant' zones (modelled concentration below 35 g/m^3) and 13% of the population live in zones that are close to the AQLV and hence within zones of 'uncertain compliance' (modelled concentration between 35 and 45µg/m^3).

The population living in zones of 'uncertain compliance' continues to decline between 2015 and 2030 as already legislated measures take effect, and by 2030 the population living in 'likely compliant' zones increases to 93%. Importantly, in the period from 2015 to 2030, the pattern of residual non-compliance moves from large contiguous areas to discrete islands of non-compliance. This has important implications for the design of efficient mitigation strategies.

Diesel exhaust NO_X

Compliance with the NO_2 air quality limit value can be improved in the short term by removing all diesel exhaust emissions (HGV, LGV, PCD and Buses) from the urban environment (Scenario B). However, against a Base Case which sees significant improvements in compliance by 2025, the incremental benefit in compliance terms is reduced from 2025. As an alternative, targeted measures to remove old diesel vehicles (pre-Euro V) from the urban environment would be easier to implement than the complete removal of all diesel vehicles, and would accelerate the achievement of improved compliance.

Another solution would be to accelerate turnover of the vehicle fleet, effectively replacing[9] all pre-Euro 5 passenger cars with Euro 6 technology faster than the natural rate of replacement (Scenario C1). This option does offer some improvement by 2020 with the percentage population living in compliant zones increasing from 83% to 88% and the percentage population living in 'likely non-compliant' zones decreasing from 10% to 6%. The benefits of the early replacement of pre-Euro 5 vehicles reduce with time as Euro 6 vehicles naturally achieve prevalence in the fleet, so that the impact from 2025 is negligible. Table 4 presents an overview of these two scenarios and their effect on compliance.

Diesel exhaust NO_x—Euro 6 performance scenarios

In Scenario E, several sensitivity cases regarding the performance of Euro 6 vehicles have been explored (see Table 1 for more details).

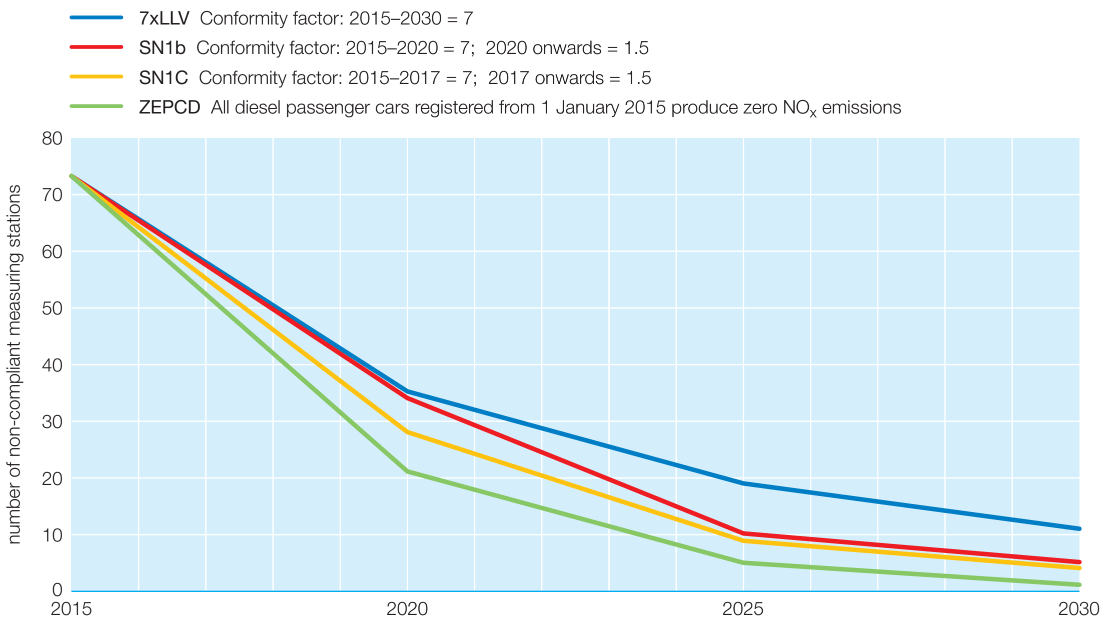
Scenario SN1b reflects[10] the recently proposed conformity factors agreed by the Member States Representatives at the 'Technical Committee—Motor Vehicles' on 28 October 2015 (EC, 2015), however a

Table 4 Percentage of EU 27 population living in zones achieving compliance with ambient air quality standards for NO_2
Base Case v. Scenario B (removing all diesel exhaust emissions) v. Scenario C1 (accelerating fleet turnover)

	EU population living in 'likely compliant' zones			EU population living in 'likely non-compliant' zones		
	Base case	Scenario B	Scenario C1	Base case	Scenario B	Scenario C1
2015	69%	69%	69%	18%	18%	18%
2020	83%	94%	88%	10%	0%	6%
2025	90%	95%	91%	5%	0%	5%
2030	93%	95%	93%	5%	0%	5%

[9] Unlike HDV vehicles, for passenger cars the retrofitting of NO_X control technologies is not a viable option.
[10] Further details can be found in the 'Scenario E' section of the main 'Urban Air Quality Study' report.

Figure 4 NO_x compliance scenarios using different conformity factor assumptions



CF of 7 is used to 2020 (ICCT, 2014) rather than the 2.8 agreed by the technical committee as this was the best available data at the time this work was undertaken. The 7xLLV scenario assumes the worst case that CF of 7 continues to 2030. The ZEPCD scenario represents the immediate substitution of new diesel passenger car sales with zero NO_X emissions alternatives. Comparing the SN1b 'real-world' scenario with a potential 'best-case' helps to highlight how much improvement might be achieved in practical terms (Table 5).

In the SN1b scenario, by 2020 the percentage of the EU population living in 'non-compliant' zones is 12% reducing to 6% by 2025 and 5% by 2030. This compares to 9% by 2020, 5% by 2025 and 0% by 2030 in the ZEPCD scenario. The plateauing of compliance from 2025 in SN1b is due to a very small number (0.5%) of non-compliant roadside air quality measuring stations. This number does reduce to 0.2% by 2030; however they are located in large urban conurbations with high population density. By expediting the achievement of a CF of 1.5 by

Table 5 Percentage of EU 27 population living in zones achieving compliance with ambient air quality standards for NO_2

Base Case v. Scenario SN1b (Euro 6 'central conformity' scenario) v. ZEPCD (removing diesel vehicles from sale)

	EU population living in 'likely compliant' zones			EU population living in 'likely non-compliant' zones		
	Base case	Scenario SN1b[11]	ZEPCD	Base case	Scenario SN1b	ZEPCD
2015	69%	69%	69%	18%	18%	18%
2020	83%	80%	85%	10%	12%	9%
2025	90%	89%	92%	5%	6%	5%
2030	93%	93%	94%	5%	5%	0%

[11] SN1b uses a 'worse than base case' CF of 7 from 2015 to 2020 for Euro 6 PCD; this is responsible for the initial decrease in compliance.

three years as in scenario SN1c it is possible to reduce the number of non-compliant stations by 2020.

The high level of compliance observed in the SN1b scenario is consistent with the recent assessment work undertaken in the UK by the Department for Environment, Food and Rural Affairs (DEFRA, 2015). Based on modelling of around 2000 individual road links in Greater London, this work indicates that by 2025 any residual NO_2 compliance issues will be confined to very small areas within a largely compliant urban agglomeration. Such small islands of non-compliance lend themselves to local, tailored strategies rather than significantly more costly and potentially disruptive city or country-wide responses.

While today NO_2 air quality limit value compliance varies widely in the urban environment, future non-compliance is anticipated to be limited to small, discrete areas. The distribution of this non-compliance strongly supports the implementation of targeted, specific solutions rather than sweeping or wide-ranging measures. Primary PM_{10} and $PM_{2.5}$ emissions from engine/exhaust road transport is a decreasingly small contributor to the total emissions.

Conclusions

The conclusions of the study are as follows:

- For PM, elimination of diesel exhaust emissions in the studied time frame will not help to achieve earlier compliance with air quality standards.
  - Replacement of solid fuels with cleaner burning alternatives in selected areas of non-compliance is expected to help achieve future compliance.
- NO_2 ambient air quality compliance is reliant on:
  - The short term (2020 horizon): targeted local measures.
  - The medium-long term (2025–2030 horizon): Euro 6/VI achieving compliance with conformity factors.
- Local options for NO_2 could include:
  - Accelerating the replacement of pre-Euro 5 vehicles by newer vehicles; to have real impact this could be very expensive.
  - Targeting captive fleets (e.g. buses) with replacement and/or retrofit schemes.
  - Introducing 'Ultra Low Emission Zones' (ULEZ) in selected locations.

References

DEFRA (2015). *Improving air quality in the UK—Tackling nitrogen dioxide in our towns and cities*. UK Government, Department for Environment, Food & Rural Affairs.

EC (2015). *Commission welcomes Member States' agreement on robust testing of air pollution emissions by cars*. European Commission, 28 October 2015. http://europa.eu/rapid/press-release_IP-15-5945_en.htm.

ICCT (2014). *Real-world exhaust emissions from modern diesel cars: A meta-analysis of PEMS emissions data from EU (Euro 6) and US (Tier 2 BIN 5/ULEV II) diesel passenger cars*. International Council on Clean Transportation (ICCT) white paper, October 2014.

IIASA (2015). WPE2014-CLE scenario for 2030 using the PRIMES 2013 Reference Activity Projection and COPERT v4.11 emission factors. In *Adjusted historic emission data, projections, and optimized emission reduction targets for 2030 – A comparison with COM data 2013*. Thematic Strategy on Air Pollution (TSAP) Report No.16, International Institute for Applied Systems Analysis.

Effect of fuel composition on particulate emissions from two GDI cars

A new study of the effects of fuel composition on particulate emissions extends previous work to include two modern GDI vehicles and a wider range of fuels.

Gasoline direct injection (GDI) engines typically emit higher particulate number (PN) emissions than conventional port fuel injected (PFI) engines due to the reduced time required for fuel atomization in the combustion chamber and the greater possibility of fuel impingement on the cylinder surface. For this reason, PN emissions limits were added for GDI vehicles in Europe, starting with the Euro 5 emissions regulations that are expected to be in full force by 2017.

A number of technology options are likely to be available to help meet these new regulations. This article describes recent work by Concawe which investigated the effects of fuel properties, in particular the use of two different fuel oxygenates representative of current and future fuels, on particulate and other regulated emissions from two modern European GDI cars.

Introduction

Emissions reduction has been the focus of worldwide vehicle legislation for more than 25 years. Although regulations initially focused on gaseous emissions of carbon monoxide (CO), hydrocarbons (HC) and nitrogen oxides (NO_x), particles emitted from vehicles and from other sources are now recognised as having an impact on air quality and on human health (Concawe, 1999). European emissions legislation has set limits for particulate emissions from diesel vehicles since the early 1990s. The introduction of clean fuels and advanced engine and after-treatment technologies has resulted in a substantial reduction in automotive particulate matter (PM) emissions with a corresponding improvement in air quality.

Conventional PFI gasoline vehicles generally emit very low levels of particulates, because the fuel is well mixed with the intake air before combustion. As a result, the focus of much research and legislation has remained on diesel vehicles which inherently have much higher particulate emissions. More recently, GDI vehicles have been introduced into the market which share some similarities with diesel vehicles in that the fuel is injected directly into the cylinder. The injected fuel has much less time to evaporate and mix before combustion starts and this can lead to greater particulate formation. There are a growing number of research studies on particle emissions from gasoline engines, and the Euro 5 and 6 particulate emission limits now apply to GDI vehicles from 2017 as well as diesel vehicles. Without the use of a gasoline particulate filter (GPF), more particles may be emitted from GDI engines than from diesel vehicles equipped with a diesel particulate filter (DPF).

From 2014, GDI cars must meet an interim PN limit of $6x10^{12}$ particles/km going down to $6x10^{11}$ particles/km in 2017. A recent article published by SAE (SAE, 2015) suggests that a range of solutions are likely to be available including combined port fuelled and direct injection systems and highly controlled direct injection systems as well as gasoline particulate filters. In addition, Bosch recently presented data on a 350-bar gasoline direct injection system demonstrating reduced particulate number compared to a 200-bar system (Bulander, 2015) and meeting the 2017 limits.

In parallel to the developments on vehicle technology and emissions regulation, the European Renewable Energy Directive (RED, 2009/28/EC) will require 10% renewable energy in transport fuels by 2020 while the Fuel Quality Directive (FQD, 2009/30/EC) will also require reductions in GHG emissions intensities from transport fuels by 6%.

Most of the available literature on particulate emissions is based on diesel vehicle studies, and although this provides valuable insights there are fewer studies in the literature on particulates from direct injection engines and the effect of oxygenates on gasoline particulates.

Concawe study objective

In a previous study (Concawe, 2005), Concawe investigated PM and PN emissions from two GDI vehicles. These were 2003 and 2004 vehicles meeting Euro 3 and Euro 4 emissions levels, respectively. Two different gasolines were used in these tests but neither contained oxygenates. Oxygenated fuels were used in another study which included a Euro 4 GDI vehicle along with two port fuel injected vehicles (Martini, *et al.,* 2013). In this study, although fuel properties including octane and oxygenates were widely varied, fuel effects were found to be small compared to vehicle-to-vehicle and drive-cycle differences. The study did demonstrate the difference in particulates between PFI and GDI vehicles.

The current study extends the previous work to include two more modern (Euro 4 and Euro 5) GDI vehicles using the European New European Driving Cycle (NEDC), the current test cycle used for homologation of vehicles. A wider range of fuels has also been investigated including ethanol and ETBE with matched oxygen content or research octane number (RON) and splash blended fuels (in the case of ethanol) to better understand the effect of these components, if any, on PM and PN. Ethanol and ETBE produced from renewable ethanol are already widely used in Europe and their use will increase to meet regulatory demands.

Test vehicles and fuels

Two GDI vehicles were selected for testing that are representative of the current road vehicle population. Both were stoichiometric GDI vehicles equipped with three-way catalysts and demonstrated to meet either Euro 4 or Euro 5 emission standards. The larger six-cylinder vehicle was naturally aspirated while the medium sized four-cylinder vehicle was turbocharged (Table 1).

The seven test fuels specially blended for this study had three levels of Research Octane Number (RON) (around 95, 98 and over 100) and three levels of oxygen content (0%, around 3.7 wt% and 7 wt% or higher), corresponding to ethanol levels of E0, E10 (or 22% ETBE) and E20. The fuels were blended to achieve a matrix with RON and oxygen content as the primary variables.

Figure 1 Test fuel matrix

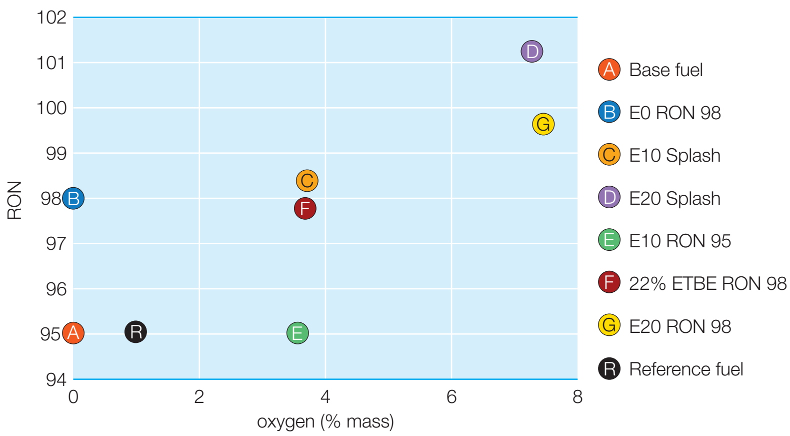


Table 1 Test vehicles

	Vehicle 1	Vehicle 2
Vehicle class	Upper Medium	Medium
Emission standard (homologation)	Euro 4	Euro 5
Engine displacement (litres)	2.5	1.8
Max. power (kW)	140	118
Inertia class (kg)	1590	1470
Cylinders	6	4
Valves	24	16
Aspiration	Natural	Turbo
Combustion type	Homogeneous stoichiometric	Homogeneous stoichiometric
Injection system	DI	DI
After-treatment device	TWC	TWC
Drive	RWD	FWD
Transmission	Manual 6-speed	Manual 6-speed
E10 compatible	Yes	Yes
Registration date	15 June 2007	4 June 2009
Mileage at start of test (miles)	23,354	8,890

The fuel matrix is shown in Figure 1. A CEC Euro 5 reference fuel of 95 RON/85 MON was also run interspersed with the test fuels in a randomized order as a repeatability check.

Experimental set-up and test design

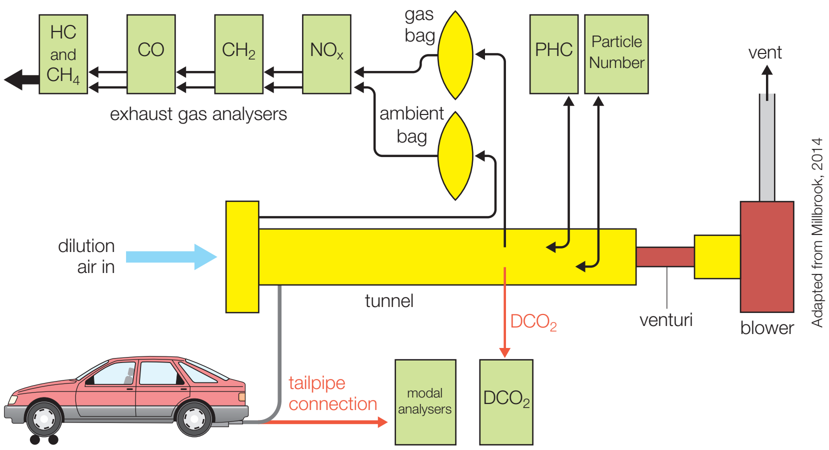
To improve repeatability, each vehicle was driven by its designated driver throughout the duration of the programme. The vehicle drive speed traces and dynamometer load were recorded second by second for each test and found to be closely repeatable. Vehicle 1 proved to be more variable than Vehicle 2 in spite of careful preparation and confirmation that the vehicle was on specification.

PM emissions were determined by sampling the vehicle tailpipe emissions using industry standard constant volume sampling (CVS) technology. Integrated bag sample emissions were collected for each phase of the test and corrected for ambient contaminants using a bag sample taken from the intake air to the CVS. In addition, undiluted exhaust gas from the tailpipe was connected directly to modal analysers, with the dilute measurements corrected for the fraction of the raw exhaust that was drawn from the tailpipe. Gaseous emissions of HC, CH_4, CO, CO_2, and NO_x were measured. Particulate emissions measurements were made using a line from the CVS dilution tunnel which was also taken to a dilute CO_2 analyser. PM was collected on a single-phase particulate filter, following the standard test procedure specified in the European vehicle emission test procedure. Fuel consumption was calculated by the regulated carbon balance method using individual carbon.

The PN measurements were made from the same sampling point as the PM emissions using the procedure developed for European regulatory testing. A gas sample from the dilution tunnel was taken to a cyclonic separator along with two thermodiluters in series which remove volatile components from the particulates. The separated sample then passed to a condensation particle counter (CPC) which gives a count of total PN. Although there can be considerable variations in repeat PN tests, the method allows discrimination down to much lower emission levels than the PM test. In these tests the CPC was complemented by an electrical low pressure impactor (ELPI). This simultaneously measures emissions in 12 different size ranges with mid points from 0.063 microns up to 9.99 microns, thus providing information on size distributions.

In all tests, second-by-second measurements were taken to allow analysis of vehicle operation in greater detail at various points in the test.

Figure 2 Experimental set-up

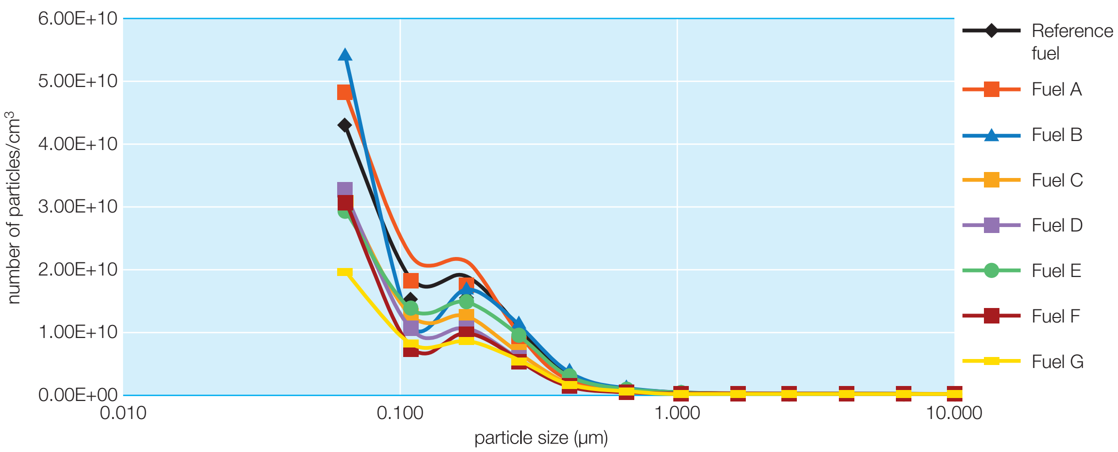


The test design specified two separate tests to be performed on each fuel in each vehicle to allow statistical evaluation of fuel effects. The fuels were tested in a randomized order with the reference fuel interspersed at regular intervals. To ensure that the individual test results were comparable, care was taken that the vehicle condition and test experience was the same for each test.

Particulate size distributions

Particle size showed a bimodal distribution with peaks around 300 nm (0.3 μm) and below the smallest 63 nm (0.063 μm) size category measurable by the ELPI. This pattern is similar to that seen in other studies including those on diesel vehicles. Although PM is dominated by

Figure 3 Particle size distribution, Vehicle 2



the larger particles, the highest number of particles was recorded in the lowest size range which suggests that there may be smaller particles that are not recorded by the ELPI.

Fuel effects on PM/PN emissions and fuel consumption

The fuel matrix was designed to cover a range of current and future fuel formulations including ethanol and ETBE blends. The presence of oxygen in the fuel has been shown in previous studies to affect PM and PN emissions in diesel vehicles so it may reasonably be expected to influence GDI combustion as well. The RON of all the fuels was adequate for the test vehicles' needs, so performance effects due to RON were not expected. However, since the oxygenate components have high octane, RON was also included as a variable in the fuel matrix. The variability in the data, particularly for the PM and PN emissions which were very low, meant that paired comparisons need to be treated with care, and that fuel-effect trends are best examined using statistical analysis of the whole fuel matrix.

For the oxygen-free fuels there were no statistically significant trends with octane number although there were directional trends for higher particulates, CO_2 and NO_x (on one vehicle) on the 98 RON fuel compared with the 95 RON fuel. The two fuels were very similar in terms of their carbon and aromatic contents. For the oxygenate-containing fuels there was evidence of a reduction in PN with the higher-octane E20 fuel compared to the base fuel, but this was only significant in vehicle 1.

There is evidence for a reduction in particulate emissions as oxygen content increases (Figures 4 and 5).

Figure 4 PM emissions versus oxygen content (arithmetic means)

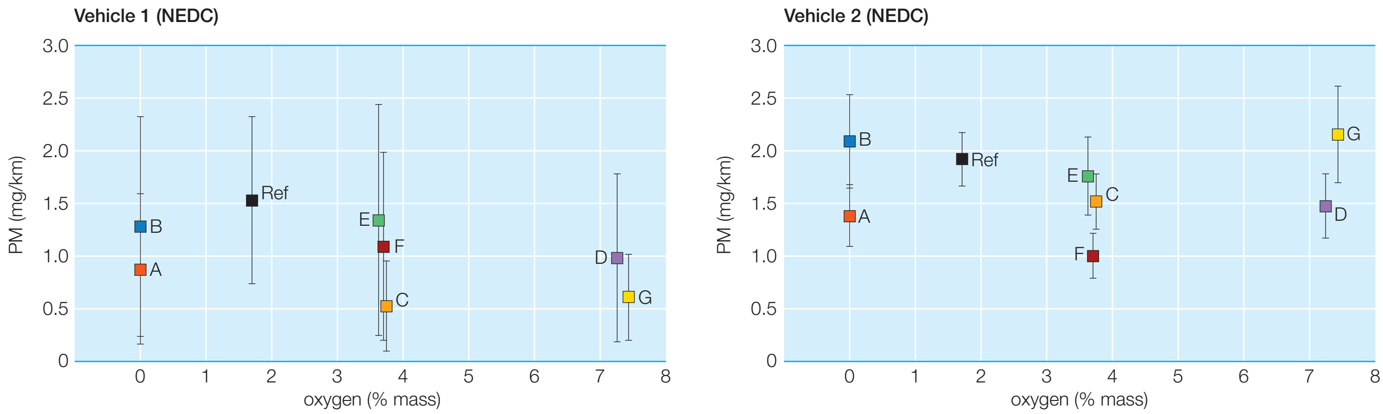
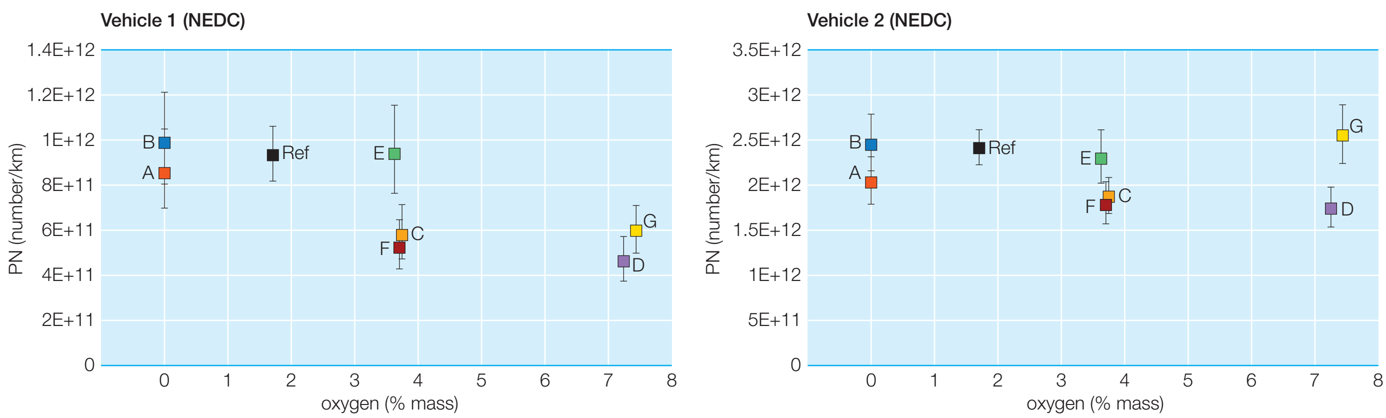


Figure 5 PN emissions versus oxygen content (geometric means)



The effect is most evident for Vehicle 1 where the trend was statistically significant for PN but not for PM, perhaps reflecting the better discrimination of the PN measurement. The correlation for Vehicle 2 was not statistically significant.

In these plots the two test runs on each fuel were averaged and the confidence band shown on the graph based on ±1.4 times the standard error of the mean. The arithmetic mean was used for the PM results, but because there is greater numerical variation in the PN results, a geometric mean was considered more appropriate.

The PM emissions on both vehicles were very low, with Vehicle 1 having lower, but more variable, emissions than Vehicle 2. No clear trend with oxygen content can be seen and the vehicles performed equally well on all of the fuels.

The better discrimination of the PN test method can be seen in the smaller error bars, although Vehicle 1 still showed more variability than Vehicle 2. There is a trend in Vehicle 1 for lower PN emissions at higher oxygen concentrations, but this is not clearly seen for Vehicle 2. For the fuels at 3–4 wt% oxygen there is a trend for the higher octane fuels (C and F) to give lower PN emissions, but the opposite trend is seen for the hydrocarbon-only fuels (A and B).

As expected, volumetric fuel consumption increased as the oxygen content of the fuel increased. Similar effects were seen on both vehicles and the results are shown in Figure 6. The higher variability for Vehicle 1 can be seen from the larger error bars.

Oxygen affects fuel consumption primarily through its effect on the volumetric energy content of the fuel. This

Figure 6 Effect of oxygen content on volumetric fuel consumption (arithmetic means)

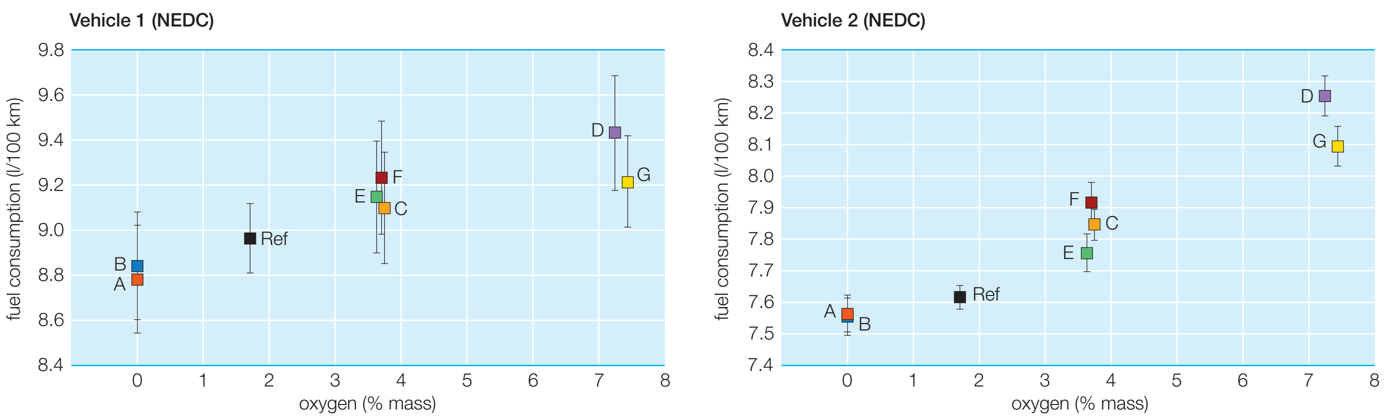
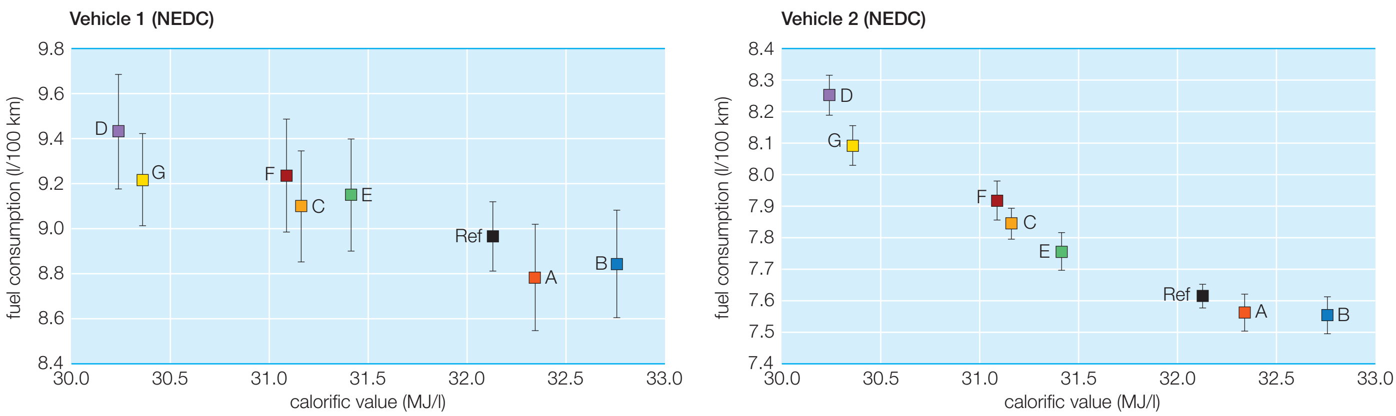


Figure 7 Fuel Consumption versus fuel calorific value (arithmetic means)



is illustrated in Figure 7 and the trends were statistically significant for both vehicles. There was also a correlation with the volatility parameter E100°C, but this is considered to be an effect of the way in which E100°C correlates with oxygenate content in this fuel matrix. These results are in line with previous studies on the effect of oxygenates on volumetric fuel consumption (Martini *et al.,* 2013). Comparison of fuels containing ethanol or ETBE at the same oxygen content showed no statistically significant differences.

Conclusions

It can be concluded from the results that no fuel effect or test variability (apart from one HC data point) was sufficiently large to cause either vehicle to exceed its relevant emissions limits. Although only two GDI vehicles were tested, in each case the vehicle had a greater impact on particle and gaseous emissions than the fuel and driving cycle. In particular, PM measured gravimetrically was difficult to interpret for fuel effects because the PM emission levels were very low from these modern GDI vehicles.

Oxygen content had no measurable effects on PM or gaseous emissions over the NEDC cycle. However, a step-change down in PN emissions for Vehicle 1 was observed for fuels containing >3.7% mass oxygen. At the same oxygen content, ETBE showed no different effect on volumetric fuel consumption compared to ethanol.

Volumetric fuel consumption increased with increasing fuel oxygen content and was due to the influence of oxygen on fuel calorific value. Although not the focus of the study, it was seen that varying RON between 95 and 98 RON without the presence of oxygenate had no consistent effect on emissions or volumetric fuel consumption in these vehicles.

Acknowledgments

Concawe would like to thank Coryton Advanced Fuels Ltd, Essex, UK for fuel blending and Millbrook Proving Ground, Bedford, UK for carrying out the vehicle testing. Thanks also go to members of the Concawe FE/STF-20 and FE/STF-25 groups and their member companies for their contributions to this work.

References

Concawe (1999). *The health effects of $PM_{2.5}$ (including ultrafine particles)*. Concawe Report No. 99/60. Brussels.

Concawe (2005). *Fuel effects on the characteristics of particle emissions from advanced engines and vehicles*. Concawe Report No. 1/05. Brussels.

SAE (2015). *Automotive Engineering*. SAE International. June 2015 issue.

Bulander, R. (2015). Powertrain optimization using a comprehensive systems approach. In *Proceedings of the 36th International Vienna Motor Symposium*, 2015.

Martini, G. *et al.* (2013). *Effect of oxygenates in gasoline on fuel consumption and emissions in three Euro 4 passenger cars*. European Commission Joint Research Centre Institute for Energy and Transport, 2013.

The natural attenuation of fatty acid methyl esters in soil and groundwater

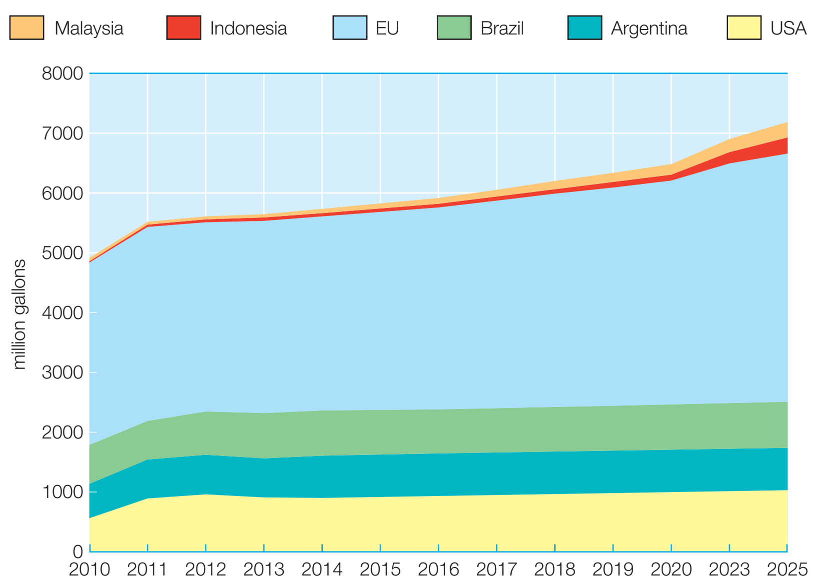
Current evidence suggests that FAME is readily biodegradable—but is there a need for further study to enhance understanding of the impacts of petroleum biodiesel released into the environment?

Introduction

Fatty acid methyl esters (FAME) are a group of organic compounds that can be synthesised through the process of esterification of fatty acids with methanol (Energy Institute, 2008; Ginn *et al.,* 2009). FAME are of considerable environmental and economic importance as they are a key constituent of commercial 'biodiesel' fuel, which may comprise neat FAME but more typically is a FAME/petroleum diesel blend (Ginn *et al.,* 2009). With the increasing use of FAME in fuel, the potential for releases of FAME and biodiesel to the environment exists. Effective management of such releases will require an understanding of the fate and transport of FAME and the impact of FAME on the fate and transport of other diesel constituents.

FAME may be sourced from a variety of feedstocks, including vegetable oils (rapeseed, soy, palm, sunflower and maize), animal fats (tallow, lard, poultry and fish oils) and waste oils and fats (used cooking oils) (Concawe, 2009; Ginn *et al.,* 2009). A number of methods are in use for the production of FAME and have been described in detail elsewhere (Ginn, *et al.,* 2009; ITRC, 2011; Concawe, 2009; Moser, 2009). In the most common process, FAME are produced via the transesterification of the feedstock material through reaction with methanol in the presence of a catalyst. The resulting mixture contains FAME and glycerine (glycerol). The latter is separated from the FAME prior to use.

Figure 1 World biodiesel production projections (FAPRI, 2011)



Fuels containing FAME are recognised by the percentage of FAME in the mixture using the letter B followed by the percentage in the fuel. B100 is pure FAME and, while B100 can be used directly as a fuel, it is usually blended with petroleum-derived diesel to produce an amended biofuel (Prince *et al.,* 2008). Typically blends include 5 and 7% (V/V) biodiesel and are designated B5 and B7. Biodiesel is internationally recognised as an alternative to conventional fuel, and a number of standards have been developed that specify the key properties of biodiesel (e.g. ASTM D6751, BS EN 14214). Globally, current mandates for blends of FAME with conventional diesel vary from B2 to B10 with the State of Minnesota planning a move to B20 (Smith, 2014; Lane, 2013). In Europe, the maximum FAME content in EN590 diesel fuel is 7% (V/V) (B7), and a new standard is being developed (FprEN 16734) that allows blending of up to 10% (V/V) FAME (B10) in accordance with the EN14214 specification.

Climate change and fuel security issues have resulted in an increasing interest in the manufacture and sale of renewable fuels globally (Fuller *et al.,* 2013). For biodiesel, this trend is driven primarily by legislation (e.g. The European Union Renewable Energy Directive (RED, 2009/28/EC) mandates the use of 10% renewable energy in road transportation and non-road mobile machinery by 2020) to reduce greenhouse gas emissions and enhance energy security.

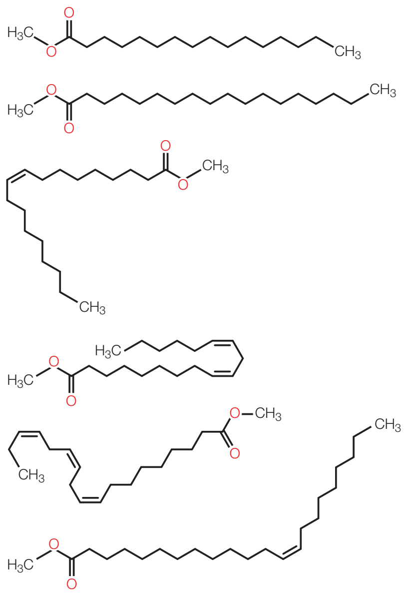
Industrial scale production of biodiesel fuel has been undertaken in Europe since 1991 (EBTP 2011). Biodiesel production is predicted to increase globally in the coming decades, as illustrated in Figure 1.

Guidance on the safe use and handling of biodiesel has been developed both in Europe and the USA (Concawe, 2009; NREL, 2009). However, there is also interest in understanding the fate and behaviour of biodiesel in the event of a release to the subsurface as a result of accidents, leakages or spills (California EPA, 2015). Considerable work has been undertaken to examine the fate of other biofuels such as ethanol (Morgan *et al.,* 2014) but far less research appears to have been published on biodiesel. Possible reasons for this include the higher prevalence of ethanol-based biofuels (FAPRI, 2011), the low usage of biodiesel in the USA, and the initial understanding that FAME is non-toxic and readily biodegradable (US EPA, 2004; NBB, 2012). While aerobic and anaerobic biodegradation of biodiesel has been documented, uncertainty remains regarding how biodiesel partitions and degrades in the subsurface and its effect on the fate of petroleum hydrocarbons. A review of technical literature has therefore been completed to bring together the available data in this area, and the findings of this review (shortly to be published as a Concawe report) are summarised here.

Use of fatty acid alkyl esters in biodiesel

The predominant fatty acid alkyl ester (FAAE) used in European biodiesel production is FAME, although in future other FAAE may also be used, such as fatty acid ethyl ester (FAEE). Work is under way to extend the test methods and specifications in EN 14214 to include FAEE, and to investigate the performance of FAEE-containing blends in engines and vehicles. The successful completion of this work is a prerequisite for accepting FAEE as a diesel fuel blending component. A further barrier to FAEE use in biodiesel is the higher price of ethanol compared to methanol. If methanol prices were to increase relative to ethanol, and if greenhouse gas emission reduction become a critical factor for legal compliance, FAEE production could become an economically attractive alternative to FAME (Concawe, 2009).

Figure 2 Examples of FAME produced from vegetable oils used in biodiesel production



Note: numbers describing each acid indicate the number of carbon atoms in the chain followed by the number of unsaturated carbon-carbon bonds in the chain.
Adapted from www.Chemspider.com

FAME composition in biodiesel

FAME have the following general molecular structure:
$CH_3(CH_2)nCOOCH_3$ (saturated); and
$CH_3(CH_2)_n(CH)_xCOOCH_3$ (unsaturated).
Examples of FAME produced from the main vegetable oils used in biodiesel production are summarised in Figure 2.

The composition of FAME will depend on a number of factors including the origin of the feedstock used and the manufacturing process (Energy Institute, 2008). In addition, the genotype, growing seasons and growing conditions have all been found to affect oil content and fatty acid profiles (Hollebone, 2009).

Key findings of the review

Individual FAME compounds are of low aqueous solubility, low volatility and low mobility. FAME, based on its low bulk density and aqueous solubility, is expected to exist as a light non-aqueous phase liquid (LNAPL) in the subsurface. A FAME LNAPL source would be relatively immobile, potentially long-lived (dependent on the volume of the release), but have a relatively small region of influence. In this context, a B5 or B20 FAME/petroleum diesel blend may be expected to behave similarly to petroleum diesel in the subsurface. The mixtures of FAME that have been studied in peer review literature do not appear to enhance the solubility of hydrocarbons as a whole or individual components such as PAH or monoaromatic hydrocarbons.

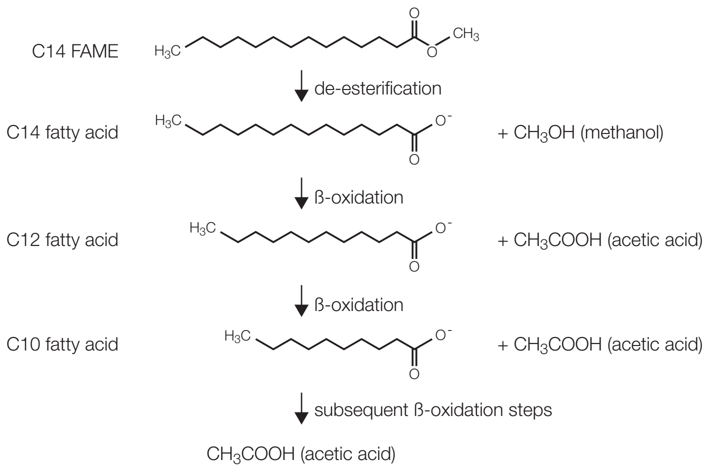
Numerous laboratory studies have been conducted to investigate the biodegradation of FAME (both bulk FAME and individual compounds) from various feedstocks. These studies have utilised different test conditions, microbial inocula, amendments and analytical measurements:

- Multiple laboratory studies have demonstrated that biological mineralization of FAME to carbon dioxide occurs under aerobic conditions. Oxygen depletion in response to FAME has also been observed at the two field sites.
- Multiple laboratory studies have demonstrated biodegradation of FAME under methanogenic conditions. Methane production in response to FAME has also been observed at the two field sites.
- A more limited number of laboratory studies have provided evidence that biodegradation of FAME occurs through nitrate and sulphate reduction. Nitrate and sulphate depletion in response to FAME biodegradation has also been observed at one field site.
- Although no laboratory studies were identified that evaluated biodegradation of FAME through iron or manganese reduction, increases in dissolved iron in response to FAME have been observed at the two field sites. No data have been reported on manganese reduction.

These observations are consistent with FAME biodegradation through all major redox processes involved in natural attenuation.

Biodegradation of FAME under anaerobic conditions has the potential to produce significant quantities of methane. In two field studies biodegradation of FAME was reported to be associated with high concentrations of methane in groundwater (30 mgl^{-1}) and soil gas (up to 67%). The release of significant quantities of methane into the subsurface during biodegradation could lead to a fire or explosion risk in the event that it accumulates in confined areas, such as basements or utility conduits. The potential for methane production should, therefore, be taken into account during the risk assessment of FAME release sites. Off-gassing of methane and carbon dioxide can enhance volatilization of BTEX and other volatile compounds from groundwater and thereby increase the risk of vapour intrusion into buildings. Studies conducted at other biofuel sites, such as ethanol release sites, indicate that the rate of methane formation will likely be dependent on factors

Figure 3 General pathway for metabolism of FAME



such as geology, depth to water, soil moisture content and other factors. Methane production rates may vary dependent on whether the release is pure FAME, or a FAME/petroleum diesel blend. Additional field studies are needed to determine whether this is the case.

FAME appears to enhance the biodegradability of diesel at concentrations of B20 and higher, but this effect has not been demonstrated at the field scale in the context of a subsurface release of a FAME/diesel mixture. At sites with limited electron acceptors and macronutrients (nitrogen and phosphate), microorganisms that degrade FAME have the potential to deplete available electron acceptors and nutrients resulting in an extended time for diesel biodegradation. However, the significance of this in field studies has not been reported.

While studies support the overall conclusion that FAME is readily biodegradable under both aerobic and anaerobic conditions, the specific details—rate, observation of a lag period, extent of degradation, preferential degradation of specific FAME—varied from study to study. Site-specific assessment of natural attenuation processes, in accordance with lines-of-evidence based good practice on monitored natural attenuation (MNA), remain necessary to demonstrate MNA on a site-by-site basis.

FAME has been reported to undergo relatively rapid auto-oxidation and hydrolysis in aqueous solution, with 5–10% conversion to free fatty acids and methanol over a 24-hour period. These more soluble, but equally biodegradable substances could increase the concentration of dissolved organic carbon in groundwater beyond that expected for the parent FAME. Confirmation of complete FAME biodegradation requires more than disappearance indicated by gas chromatography (GC) because intermediates produced through auto-oxidation and hydrolysis are not detectable by standard GC methods for FAME and diesel. With the exception of methanol, these intermediates are not known to be toxic, but could continue to have an impact on water quality. Additional work is needed to explore these effects.

Conclusions and recommendations

Overall it was concluded that natural attenuation processes appear to be significant in controlling the fate, behaviour and potential risks posed by biodiesel. Significant attenuation mechanisms are likely to include sorption, autoxidation and biodegradation via a variety of redox processes: the exact role and contribution of each will depend on the nature of the release, the characteristics of the individual FAME, the FAME-diesel blend and the environmental setting. Such attenuation has not been observed to hinder the degradation of other diesel constituents, but could generate undesirable effects such as excessive methane, which need to be assessed and managed.

To date, only a limited number of studies have been published on the fate and behaviour of FAME at the field scale. Additional studies of either controlled or accidental releases of FAME or FAME-diesel mixtures would enhance understanding of the biodegradation processes discussed above and the behaviour of these processes in different geologies. Of particular interest would be additional information on the impact of FAME on the fate and transport of petroleum diesel and the production of methane.

References

California EPA (2015). *Multimedia Evaluation of Biodiesel*. California Environmental Protection Agency, Staff Report. State of California. http://www.calepa.ca.gov/CEPC/2015/BiodieselRpt.pdf

Concawe (2009). *Guidelines for handling and blending FAME*. Report 9/09. Conservation of Clean Air and Water in Europe. Brussels, Belgium.

Energy Institute (2008). *EI literature review: Biofuels – Potential Risks to UK Water Resources*. Final Report. Energy Institute. London, UK.

EBTP (2011). *Fatty Acid Methyl Esters (FAME)*. Biofuel Fact Sheet. European Biofuels Technology Platform. Brussels.

FAPRI (2011). *World Agricultural Outlook*. Food and Agricultural Policy Research Institute, Iowa State University, Iowa, USA.

Fuller. S., Spikmans, V., Vaughan, G. and Guo, C. (2013). Effects of Weathering on Sterol, Fatty Acid Methyl Ester (FAME), and Hydrocarbon Profiles of Biodiesel and Biodiesel/Diesel Blends. In *Environmental Forensics*, Vol. 14, Issue 1, pp. 42-49.

Ginn, T. R., Hatch, T. J., McKone, T.E. and Rice, D. W. (2009). *California Biodiesel Multimedia Evaluation Tier I Report.* University of California, Davis and University of California, Berkeley. USA.

Hollebone, B. (2009). *Biofuels in the Environment. A Review of Behaviours, Fates and Effects & Remediation Techniques*. Environment Canada. Presentation at Freshwater Spills Symposium, St Louis, Missouri, USA.

ITRC (2011). *Biofuels: Release Prevention, Environmental Behaviour, and Remediation.* Interstate Technology & Regulatory Council. USA.

Lane, J. (2013). Biofuels Mandates around the World: 2014. Biofuels Digest (website article). http://www.biofuelsdigest.com/bdigest/2013/12/31/biofuels-mandates-around-the-world-2014.

Morgan, P., Firth, S. and Hildenbrand, B. (2014). Behaviour and effects of alcohol-blended petrol in the subsurface. In *Quarterly Journal of Engineering Geology and Hydrogeology*, Vol. 47, No. 3, pp. 267-279.

Moser, B. R. (2009). Biodiesel production, properties and feedstocks. In *Vitro Cellular & Developmental Biology—Plant*, Vol. 45, No. 3, Biofuels, pp. 229–266.

NBB (2012). *Biodiesel: Environmental & Safety Information*. Biodiesel Fact Sheet. National Biodiesel Board. USA.

NREL (2009). *Biodiesel Handling and Use Guide. Fourth Edition*. National Renewable Energy Laboratory. NREL/TP-540-43672. USA.

Prince, R. C., Haitmanek, C. and Lee, C. C. (2008). The Primary Aerobic Biodegradation of Biodiesel B20. In *Chemosphere*, Vol. 71, Issue 8, pp.1446–1451.

Smith, D. L. (2014). *Biofuels development in Canada and the US*. European Biofuels Technology Platform, 6th Stakeholder Plenary Meeting, SPM6, Biofuels for Low Carbon Transport and Energy Security. Brussels. October 2014.

US EPA (2004). *U.S. EPA gives $69,000 to University of Nevada to find ways to improve alternative fuel*. Press Release Date: 5 August 2004. United States Environmental Protection Agency, Washington, DC, USA.

Product theft from oil pipelines

The theft of oil from product pipelines is a fast-growing issue for European pipeline operators.

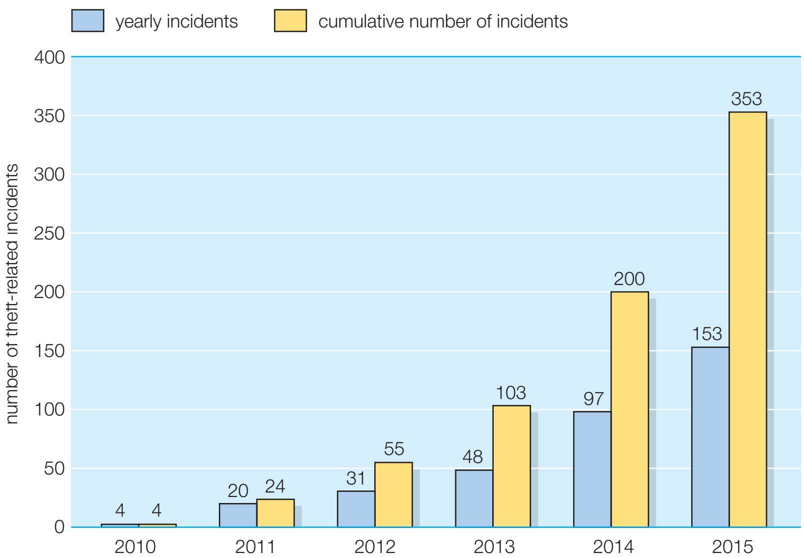
Background

Since 1971, Concawe has carried out an annual survey of spillages from European cross-country oil pipelines and published the results in an annual report, along with a detailed analysis of the primary causes of the spillages. Over the years, figures have consistently shown a steady reduction in the frequency of spillages (spillages per 1000 km of pipeline). Of the total number of spillages reported, those caused by factors within the direct control of operators, including mechanical failures, corrosion and operational errors, have accounted for a decreasing percentage, while the number of spillages related to third-party activities has been on the increase.

Over the years, a small number of third party-related spillages have resulted from successful or attempted product theft. All of these occurred in Southern and Eastern Europe. However, starting in 2011, a new trend began to emerge, with product theft events being reported in several areas of Europe, including in a number of countries that had hitherto not been affected. The number of reported cases has increased year-on-year since then.

It became clear that the reporting system for pipeline spillages was not providing a complete picture of the scale of product theft, since it only captured theft events that resulted in a spill. In response, Concawe carried out a special survey in 2015 to record both successful and attempted product theft incidents in the European oil pipeline network since the beginning of the decade. This article presents an analysis of the 2010–2015 survey results and looks at the impact of this new phenomenon on long-term European pipeline spill statistics.

Figure 1 Annual and cumulative numbers of theft-related incidents, 2010–2015



Product theft attempts: historical development

The theft incident survey performed by Concawe in mid-2015 was updated in early 2016 to capture the total number of events recorded in 2015. It included all 78 operators who regularly contribute to the annual spillage survey, 57 of which responded representing nearly 90% of the total inventory. Eighteen operators representing 60% of the total inventory reported theft attempts in a total of eight countries spread across Europe. It is believed that all operators who suffered theft attempts responded and that Concawe has therefore been able to capture virtually all such events in Europe.

Figure 1 shows the annual and cumulative numbers of theft-related events between 2010 and 2015. A large increase is evident, from 4 to over 150 cases per year over the 6-year period.

Figure 2 Theft-related incidents per country, 2010–2015

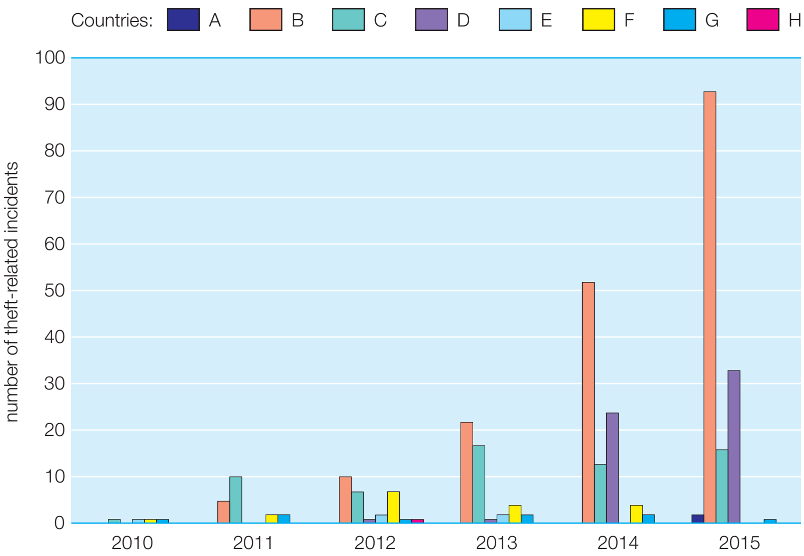
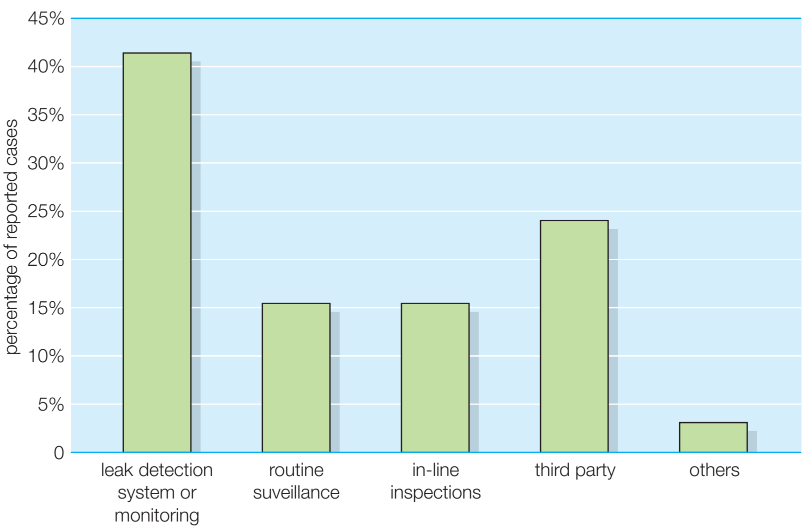


Figure 2 shows the distribution of events among the eight countries that have been affected. Some countries have been particularly targeted; three countries account for 86% of all cases.

As would be expected, white product pipelines (gasoline, diesel, jet fuel) are frequent targets. Indeed, 93% of all events involved theft attempts from these lines. While 25 incidents affecting crude oil lines were reported, there is evidence that in 8 of these cases the crude oil line had been targeted in error. Eighteen of the incidents on crude oil lines were in the same country.

Figure 3 Discovery mode of illegal connections (reported for 42% of cases)



Modus operandi

Although the circumstances of product theft vary, the typical modus operandi is described below.

Thieves generally target buried pipeline sections in relatively isolated areas, mostly rural or semi-rural and with some form of vegetation cover. A reported 78% of illegal tappings were installed in underground sections. Other cases involved above-ground installations, particularly valve stations. The type of land use was reported for about a third of all recorded cases; 84% of cases were in rural areas of either open land or shrub, while the balance of cases involved more 'public' areas such as car parks, lay-bys or even buildings.

Thieves typically excavate around the pipeline, probably under cover of darkness, and install a connection using a variety of means ranging from welded 'hot taps' to clamp devices. The tapping is then connected to a hose running to a nearby collection point which is generally outside in an area accessible to vehicles (e.g. lay-bys, car parks etc.), although some cases have involved buildings with fixed storage vessels. The distance between the tapping and collection point varies considerably (46% under 10 m; 27% between 10 and 100 m; and the balance up to 1 km). The type of hose used also varies a great deal from specialist high pressure hoses to low pressure 'garden' water hoses, which are often unable to withstand the pressure in the pipeline and therefore fail, leading to spillage or injury.

Detection

Abstraction of product at low flow rates, and often intermittently, can be difficult to detect and some tappings may remain unnoticed for long periods of time. Figure 3 shows a breakdown of the ways illegal connections have been discovered. Leak detection systems, together with monitoring by control room operators, are the most prevalent means of discovery. Theft incidents may also be reported by third parties in the event that connections leak, and passers-by or land owners see or smell hydrocarbons.

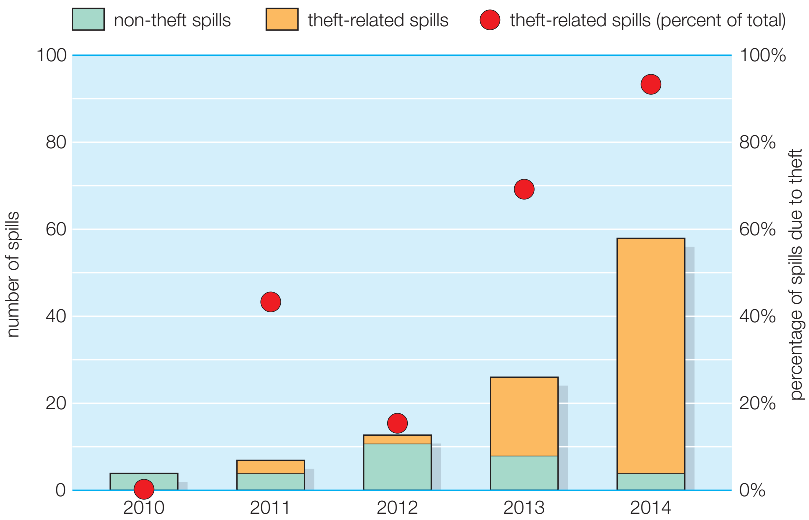
Implications for Concawe's annual spillage survey

Out of the 353 theft-related incidents reported from 2010–2015, 184 (52%) resulted in a reportable spill. Figure 4 shows that theft-related spills, as a percentage of the annual total, increased from close to zero in 2010 to 93% 2014.

Including these spills in the long-term Concawe pipeline performance statistics, which are intended to reflect pipeline integrity in the absence of intentional damage, would create a major distortion. Accordingly the reporting format was adapted from the 2013 reporting year to show long-term trends with, and without, theft-related events, where appropriate.

The 2010–2015 theft incident survey has confirmed the importance of recording the number of attempted product theft incidents, as well as the number of incidents associated with reportable product spills. Accordingly, the annual Concawe survey of pipeline operators will be updated in 2016 to include summary data on attempted theft incidents. This will allow Concawe to track the development of this new phenomenon and monitor the success of measures taken by both operators and authorities to address this issue.

Figure 4 Number and percentage of product spills arising from theft attempts vs other causes, from 2010-2014 (percentages indicated by yellow circles)



Interview with Concawe research associate, Catarina Caiado

Catarina Caiado (Galp Energia) talks about her experience as Research Associate at Concawe.

Q: Before we talk about your work at Concawe, please tell us about yourself (where you come from, your company, your background and your position in your company).

A: *I come from Lisbon, Portugal. My background is in chemical engineering and before moving to Brussels I was working for Galp Energia, the Portuguese oil company, as a process engineer. During this time I worked with refining information systems and advanced process control and was also involved in the commissioning activities of the new hydrocracker unit at one of Galp's refineries.*

Q: How did you learn about the opportunity to join Concawe as a Research Associate?

A: *I was not participating in any of the Concawe working groups before, so I learnt about this opportunity through some of the colleagues that are collaborating with the Association.*

Q: Why were you interested in taking this position?

A: *What interested me the most about this position was the opportunity to gain international experience while learning more about the challenges of the oil refining industry and the EU legislative processes.*

Q: What projects have you been working on during your time at Concawe?

A: *During my assignment at Concawe I was involved in the Oil Refining Fitness Check*[1]*, collecting and analysing data from member companies to provide to the Commission, and also reviewing and commenting on the draft reports.*

Other responsibilities in Concawe were to run the Fleet and Fuels model to conduct an internal analysis of the impact of different options for future transport policies and also as part of the JEC consortium as an input for the Biofuels Study update.

Q: What else have you been working on?

A: *I've also participated in several projects such as the Urban Air Quality study, the ReCap project (Carbon Capture in Oil refineries), the CO_2 Allocation study (defining refining CO_2 intensities for oil products) and lately I did some work as part of the Sub-group on Advanced Biofuels for the Sustainable Transport Forum.*

Q: What did you enjoy the most about your assignment in Brussels?

A: *During this year and a half I had the privilege of working with people from around the world and from different organizations, and this was probably what I enjoyed the most. You learn a lot on work-related subjects, of course, but on the human relations field as well. I'm sure that such learning experience will be very useful throughout life.*

Q: Can you tell us about your experience living in Brussels?

A: *Living in Brussels can be very exciting! It is probably one of the most international cities you will ever know and, above all, it is very easy to travel to anywhere in Europe! The fact that you can go for a coffee in cities like Paris or London, is great!*

Q: How has this experience helped you in your career, and how do you believe it might help in the future?

A: *Having an international experience always helps boosting your career and this is because during this experience you leave your comfort zone, to expose yourself to everyday challenges, to learn about what's beyond your 'nutshell'! Companies are very much aware of this so it is a valuable experience that would be of benefit to anybody's future career.*

Q: Would you recommend your colleagues to undertake a similar development path?

A: *I would recommend it to everyone! I believe that you should have an experience like this at least once in your lifetime! Whether your goal is to learn more about this and other related industries or experience what it is like to live and work in an international environment, both as a professional and at the social level, you will always benefit greatly from it!*

[1] As part of its Better Regulation policy the Commission initiated a programme for Regulatory Fitness and Performance (REFIT) in 2010. Fitness Checks provide an evidence-based critical analysis of whether EU actions are proportionate to their objectives and delivering as expected. The oil refining fitness check evaluates how ten pieces of the most relevant EU legislation drawn from the fields of environment, climate action, taxation and energy affect the petroleum refining sector. The analysis covers a wide range of important aspects including five key evaluation criteria (effectiveness, efficiency, coherence, relevance and EU added value). Consideration is also given to the sector's competitiveness position from 2000 to 2012 and issues such as excessive regulatory burden, overlaps, gaps, inconsistencies or obsolete measures.

Interview with Concawe research associate, Charlene Lawson

Charlene Lawson (Shell) talks about her experience as Research Associate at Concawe.

Q: Before we talk about your work at Concawe, please tell us about yourself (where you come from, your company, your background and your position in your company).

A: *I am originally from Warrenton, Virginia. I relocated to Houston, Texas to work for Shell Global Solutions in 2014. My background is in atmospheric chemistry and air quality modelling. I worked as an Air Science Consultant in the Environmental Science team at Shell Technology Center, Houston.*

Q: How did you learn about the opportunity to join Concawe as a Research Associate?

A: *The Research Associate position was presented to me by my supervisor at Shell. He felt that it would be a great developmental role and that I was an ideal candidate for the position.*

Q: Why were you interested in taking this position?

A: *Shell is a global company, so it is very important to be well-rounded and knowledgeable about industry challenges on a global scale. The Research Associate position provided the opportunity to broaden my knowledge of global technical issues and EU environmental legislation. This position also enables you to gain experience in managing and coordinating research activities with a diverse group of member company experts, which is vitally important for professional growth, leadership development and networking.*

Q: What projects have you been working on during your time at Concawe?

A: *My projects have been focused primarily in the area of refinery emissions monitoring and reporting. I have supported the development and implementation of quantitative optical gas imaging (QOGI) field trials, coordinated work on the strategy for the evaluation and assessment of the impact of HCN emissions from fluid catalytic cracking (FCC) units, and developed experimental work to minimise the uncertainties associated with DIAL[1] and SOF[2] measurements by improving the wind characterization.*

Q: What else have you been working on?

A: *I've had the opportunity to write technical papers on some of the projects and also to present that work at international conferences.*

Q: What did you enjoy the most about your assignment in Brussels?

A: *This is a hard question because it's really been a great experience. I would have to say that working with people from all over the world, conducting projects in different countries, and seeing first-hand how important technical research is with regard to advocacy efforts have been the best parts.*

Q: Can you tell us about your experience living in Brussels?

A: *Taking this assignment was my first time in Europe so I was not quite sure what to expect but I absolutely love Brussels! It reminds me of Washington, DC. It's a small city with a very diverse population. The food is great, the people are friendly, and it's centrally-located, making it easy to visit other countries in your free time.*

Q: How has this experience helped you in your career, and how do you believe it might help in the future?

A: *The breadth of knowledge and exposure to the industry that I gained has truly been invaluable, not only for me but for my home company as well. I have grown both personally and professionally, and made lasting relationships. I feel confident that I can return to Shell and make even more impactful contributions to the businesses.*

Q: Would you recommend your colleagues to undertake a similar development path?

A: *I strongly encourage my colleagues to undertake this type of development path. It's very hands-on and the work is challenging yet exciting.*

1 Differential absorption LIDAR (light detection and ranging)

2 Solar occultation flux

Abbreviations and terms

AirBase	The European air quality database maintained by the EEA
AQLV	Air Quality Limit Value
AQMZ	Air Quality Management Zone
BTEX	Benzene, Toluene, Ethylbenzene and Xylenes (some of the volatile organic compounds (VOCs) found in petroleum derivatives such as gasoline)
CEC	Coordinating European Council
CF	Conformity Factor
CH_4	Methane
CO	Carbon Monoxide
CO_2	Carbon Dioxide
COPERT 4	Software tool used to calculate air pollutant and greenhouse gas emissions from road transport
CPC	Condensation Particle Counter (sometimes called Condensation Nucleus Counter, CNC)
CR	Compression Ratio
CVS	Constant Volume Sampling
DEFRA	UK Government, Department for Environment, Food & Rural Affairs
DI	Direct Injection
DIAL	Differential Absorption LIDAR
DPF	Diesel Particulate Filter
Exx	Gasoline blend containing xx% ethanol
Exx°C	% fuel evaporated at xx°C
EC	European Commission
EEA	European Environment Agency
ELPI	Electrical Low Pressure Impactor
ETBE	Ethyl Tertiary Butyl Ether
EU	European Union
EU-27	The 27 Member States that comprise the European Union
EUCAR	European Council for Automotive Research and development
Euro 5, 6	European emission standards for light-duty vehicles
FAAE	Fatty Acid Alkyl Ester
FAEE	Fatty Acid Ethyl Ester
FAME	Fatty Acid Methyl Ester
FCC	Fluid Catalytic Cracking
FWD	Front-Wheel Drive
FTP	Federal Test Procedure
GC	Gas Chromatography
GDI	Gasoline Direct Injection
GPF	Gasoline Particulate Filter
HC	Hydrocarbon
HCN	Hydrogen Cyanide (also Hydrocyanic Acid)
HGV	Heavy Goods Vehicle
ICCT	International Council on Clean Transportation
IIASA	International Institute for Applied Systems Analysis
JEC	JRC-EUCAR-Concawe consortium
JRC	Joint Research Centre of the European Commission
LCV	Lower Calorific Value (same as LHV)
LGV	Light Goods Vehicle
LHV	Lower Heating Value (same as LCV)
LIDAR	Light Detection And Ranging
LNAPL	Light Non-Aqueous Phase Liquid
MJ	Megajoule
NEDC	New European Driving Cycle
MNA	Monitored Natural Attenuation
NMHC	Non-Methane Hydrocarbon
MON	Motor Octane Number
NO_2	Nitrogen Dioxide
NO_x	Nitrogen Oxides
PCD	Passenger Car Diesel
PFI	Port Fuel Injection
PM	Particulate Matter or Mass
$PM_{2.5}/PM_{10}$	Particulate matter with an aerodynamic diameter less than or equal to 2.5/10 µm
PMP	Particulate Measurement Programme
PN	Particulate Number
QOGI	Quantitative Optical Gas Imaging
RDE	Real Driving Emissions
RED	European Union Renewable Energy Directive
RON	Research Octane Number
RWD	Rear-Wheel Drive
SOF	Solar Occultation Flux
TSAP	Thematic Strategy on Air Pollution
TWC	Three-Way Catalyst
ULEZ	Ultra Low Emission Zones
VOC	Volatile Organic Compound

Concawe contacts

Director General

John Cooper
Tel: +32-2 566 91 05
E-mail: john.cooper@concawe.org

Science Director

Robin Nelson
Tel: +32-2 566 91 61 Mobile: +32-496 27 37 23
E-mail: robin.nelson@concawe.org

Science Executives

Air quality
Lucia Gonzalez Bajos
Tel: +32-2 566 91 71 Mobile: +32-490 11 04 71
E-mail: lucia.gonzalez@concawe.org

Fuels quality and emissions
Heather Hamje
Tel: +32-2 566 91 69 Mobile: +32-499 97 53 25
E-mail: heather.hamje@concawe.org

Health
Hans Ketelslegers
Tel: +32-2 566 91 63 Mobile: +32-493 25 51 54
E-mail: hans.ketelslegers@concawe.org

Petroleum products • Safety
Klaas den Haan
Tel: +32-2 566 91 83 Mobile: +32-498 19 97 48
E-mail: klaas.denhaan@concawe.org

REACH and Petroleum products
Hannu Keränen
Tel: +32-2 566 91 66 Mobile: +32-492 72 54 85
E-mail: hannu.keranen@concawe.org

REACH Implementation Manager & Legal Advisor
Sophie Bornstein
Tel: +32-2 566 91 68 Mobile: +32-497 26 08 05
E-mail: sophie.bornstein@concawe.org

Refinery technology
Alan Reid
Tel: +32-2 566 91 67 Mobile: +32-492 72 91 76
E-mail: alan.reid@concawe.org

Water, soil and waste • Oil pipelines
Mike Spence
Tel: +32-2 566 91 80 Mobile: +32-496 16 96 76
E-mail: mike.spence@concawe.org

Office management and support

Office Support
Marleen Eggerickx
Tel: +32-2 566 91 76
E-mail: marleen.eggerickx@concawe.org

Sandrine Faucq
Tel: +32-2 566 91 75
E-mail: sandrine.faucq@concawe.org

Jeannette Henriksen
Tel: +32-2 566 91 05
E-mail: jeannette.henriksen@concawe.org

Anja Mannaerts
Tel: +32-2 566 91 78
E-mail: anja.mannaerts@concawe.org

REACH Support
Julie Tornero
Tel: +32-2 566 91 73
E-mail: julie.tornero@concawe.org

Julien Harquel
Tel: +32-2 566 91 74
E-mail: julien.harquel@concawe.org

Vanessa Kondagba
Tel: +32-2 566 91 65
E-mail: vanessa.kondagba@concawe.org

Finance, Administration & HR Manager
Didier De Vidts
Tel: +32-2 566 91 18 Mobile: +32-474 06 84 66
E-mail: didier.devidts@concawe.org

Finance, Administration & HR Support
Alain Louckx
Tel: +32-2 566 91 14
E-mail: alain.louckx@concawe.org

Madeleine Dasnoy
Tel: +32-2 566 91 37
E-mail: madeleine.dasnoy@concawe.org

Communications Manager
Alain Mathuren
Tel: +32-2 566 91 19
E-mail: alain.mathuren@concawe.org

Communications Support
Lukasz Pasterski
Tel: +32-2 566 91 04
E-mail: lukasz.pasterski@concawe.org

Legal Advisor
Gloria Crichlow
Tel: +32 2 566 91 22
E-mail: gloria.crichlow@concawe.org

Concawe publications

Reports published by Concawe from 2015 to date	
10/16	Gasoline direct injection particulate study
9/16	Emission factors for metals from combustion of refinery fuel gas and residual fuel oil
8/16	Environmental fate and effects of poly- and perfluoroalkyl substances (PFAS)
7/16	Performance of European cross-country oil pipelines—Statistical summary of reported spillages in 2014 and since 1971
6/16	*Critical review of the relationship between IP346 and dermal carcinogenic activity*
5/16	*The natural attenuation of fatty acid methyl esters in soil and groundwater*
4/16	*Review of recent health effect studies with sulphur dioxide*
3/16	*Assessing the aquatic toxicity of petroleum products: comparison of PETROTOX calculations and SPME-GC screening*
2/16	*Analysis of N-, O-, and S- heterocyclics in petroleum products using GCxGC with specific detection*
1/16	*Sulphur dioxide emissions from oil refineries in Europe (2010)*
10/15R	*First Aid Reference Guide—2015*
9/15	*Hazard classification and labelling of petroleum substances in the European Economic Area—2015*
8/15	*Monitoring method for inhalation exposure to gas oil vapour and aerosol*

Adobe PDF files of virtually all current reports, as well as up-to-date catalogues, can be downloaded from Concawe's website at:
https://www.concawe.eu/publications

This page is intentionally blank

Concawe

Boulevard du Souverain 165, B–1160 Brussels, Belgium

Telephone: +32-2 566 91 60 • Telefax: +32-2 566 91 81

info@concawe.org • www.concawe.org