volatile organic compound emissions: an inventory for western europe

Prepared by the CONCAWE Air Quality Management Group's
Special Task Force on Hydrocarbon emissions from oil refining
and products in the context of total VOC emissions (AQ/STF 25)

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ABSTRACT

This report gives details of an inventory compiled by CONCAWE and covering man-made
non-methane volatile organic compound (VOC) emissions in Western Europe. The survey shows
that the major man-made sources are road transport and solvents, each contributing around
40%. Total oil industry operations account for around 8%, with refining operations
contribution about 2%. The major oil industry contribution comes from the distribution of
gasoline, including vehicle refuelling (5%).

Dit rapport bevat de gegevens van een door CONCAWE samengestelde inventarisatie
betreffende de door menselijke aktiviteiten veroorzaakte emissies in West-Europa van
vluchtige organische verbindingen (VOC). Het overzicht toont aan dat het wegtransport en
oplosmiddelen de belangrijkste emissiebronnen zijn, ieder ongeveer 40% bijdragend. De
totale aardolie-industrie maakt ongeveer 8% van het totaal uit met een bijdrage van
ongeveer 2% van de raffinaderij-operaties. De belangrijkste bijdrage van de
aardolie-industrie komt van de distributie (tussentijdse opslag) van benzine, inklusief
het benzine tanken van auto's (5%).

In diesem Bericht sind von CONCAWE Daten zusammengestellt worden über von Menschen
hervorgerufene Emissionen flüchtiger organischer Bestandteile in West Europa. Der Bericht
zeigt, dass der Hauptanteil dieser von Menschen hervorgerufenen Emissionen von
Strassentransport und von Lösungsmitteln zu 40% je Anteil, stammen. Der gesamte
Operationsbereich der Ölindustrie ist mit 8% beteiligt, davon der Raffineriebereich mit
2%. Der Hauptanteil in der Ölindustrie stammt aus dem Benzinverteilerbereich, davon die
Benzintankstationen mit 5%.

Ce rapport donne des détails d'un inventaire dressé par CONCAWE couvrant des émissions de
composants organiques non-methane de provenance humaine en Europe de l'Ouest.
L'aperçu montre que le gros de ces émissions provient du transport routier et de
dissolvants dont chacun contribue de l'ordre de 40%. L'ensemble des opérations de
l'industrie pétrolière compte pour 8% dont les raffineries contribuent avec 2% le gros de
la contribution de l'industrie pétrolière provient de la distribution des essences, dont
5% des stations de service.

CONCAWE ha desarrollado un inventario de fuentes artificiales de compuestos orgánicos
volatiles (COV), exceptuado el metano, para Europa occidental, utilizando los datos
disponibles más actualizados. El transporte por carretera contribuyen con un 40%. Las
actividades de la industria petrolifera, incluyendo la producción de crudo, el transporte
marítimo y el almacenamiento, refino y distribución de productos, corre con cerca de un 8%
del total y el refino, en sí mismo considerado, con cerca de un 2%. La mayor contribución
individualizada, en lo que concierne a la industria del petróleo, es la distribución de
gasolina, incluyendo el repostado de automóviles, que contribuye con un 5%.

Il rapporto fornisce i particolari di un inventario compilato dal CONCAWE relativo alle
emissioni di Idrocarburi, escluso il metano, in Europa Occidentale provocate da attività
umane. L'indagine mostra che le maggiori fonti di emissione sono il trasporto su strada ed
i solventi, ciascuno con un contributo di circa il 40%. Il complesso delle attività
petrolifere contribuisce per circa l'8%, ed in particolare le operazioni di raffinazione
contribuiscono per circa il 2%. Nelle attività petrolifere il contributo maggiore (5%) è
dato dalla distribuzione delle benzine, incluso il rifornimento dei veicoli.

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SUMMARY

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SUMMARY

In view of the current interest in atmospheric emissions of
hydrocarbons, CONCAWE has compiled an inventory of emissions of
man-made volatile organic compounds (VOCs) in Western Europe, using
the most up-to-date information available.

The survey showed that the two biggest contributors to man-made,
non-methane VOC emissions to the atmosphere in Western Europe are
mobile sources, especially road transport, and solvents. Oil
industry operations contribute much smaller amounts, as shown in
the following table:

Mobile sources	38 - 40 wt%
Solvents	38 - 40 wt%
Oil industry operations (including approximately 2 wt% from refining and 5 wt% from gasoline distribution)	8 wt%

Natural sources of non-methane VOCs are approximately equal to the
total anthropogenic production.

As with other studies methane has been excluded from the study as
no realistic data are available. However, it is by no means certain
that this exclusion of methane is technically justified, since it
may, in fact, contribute to the formation of background ozone in
the longer term.

1.   INTRODUCTION

In Europe, the EEC and OECD are both currently involved in the development of emission inventories for a number of pollutants, including sulphur dioxide (SO_2), nitrogen oxides (NO_x), hydrocarbons or volatile organic compounds (VOCs).

The oil industry believes that it has certain information on hydrocarbon emissions available from internal studies which have hitherto not been published. These data reflect developments which have taken place in recent years with regard to the oil industry itself, the size and nature of the motor car population and hence its emissions, and the solvents industry.

This report has therefore been compiled using the industry's information in order to ensure that an accurate data survey on hydrocarbon (VOC) emissions is made available for use by supranational bodies in their current studies.

2. ## VOC EMISSIONS INVENTORY

CONCAWE's best estimates of the main anthropogenic sources of
emissions of volatile organic compounds (VOCs) to the atmosphere
(in tonnes x 10^3) from Western Europe in 1983 are presented in
Table 1. The certainty of the values for individual sources varies
widely. Details of sources used and assumptions made are given in
the later part of this Section. Relatively few of the values are
based on actual measurements made in 1983, but where emission
factors have been used they have been applied to the consumption
figures for 1983 published by the International Energy Agency (1).
In order to facilitate comparison with other official estimates,
figures are also shown for the ten EEC Member States (EEC-10)
during 1983.

The values given exclude methane, which has by convention been
treated as non-reactive in the context of oxidant formation in the
atmosphere. This is an arbitrary decision since it is now believed
that methane may play some role in atmospheric chemistry. However,
the inclusion of methane would be difficult since published data
on, for example, vehicle exhaust emissions would not have been
comparable with emissions from other sources. On the other hand,
organic compounds are considered to consist not only of
hydrocarbons, but also of oxygenated and chlorinated compounds.

In addition, in Table 1 an estimate is given, on the basis of
literature data, of volatile organic compound emissions by natural
sources for comparison with the man-made emissions.

Table 1   Anthropogenic emissions of volatile organic compounds in Western Europe, 1983

Source	Western Europe (OECD)		EEC-10	
	(tonnes x 10^3)	(%)	(tonnes x 10^3)	(%)
MOBILE SOURCES				
Gasoline vehicles	3390	34.4	2725	36.0
Diesel vehicles	300	3.0	225	3.0
Aircraft	40	0.4	30	0.4
Railways	40	0.4	30	0.4
Coastal and inland shipping	10	0.1	10	0.1
Total	3780	38.3	3020	39.9
OIL INDUSTRY				
Production	20	0.2	15	0.2
Marine transport and crude terminals	50	0.5	40	0.5
Refineries	220	2.2	180	2.4
Gasoline distribution	510	5.2	415	5.5
Total	800	8.1	650	8.6
SOLVENTS				
All types	4020	40.8	2850	37.7
MANUFACTURING INDUSTRY				
(not solvents)	405	4.2	300	4.0
NATURAL GAS				
Production	75	0.8	55	0.8
Distribution	570	5.8	550	7.1
Total	645	6.6	605	7.9
SOLID WASTE DISPOSAL	110	1.1	65	0.9
STATIONARY COMBUSTION	85	0.9	58	0.8
TOTAL	9845	100.0	7565	100.0
NATURAL	10000		7500	

Note: All values exclude methane.

2.1 MOBILE SOURCES

In __Table 1__ the figure of 3390 x 10^3 tonnes for gasoline-engined vehicles includes hydrocarbons from both exhaust gases (2428 x 10^3 tonnes) and evaporative losses (962 x 10^3 tonnes), but not re-fuelling losses, which are included under gasoline distribution (__Section 2.2__). This figure is based on recent measurements made during road tests, whereas previous estimates have been based on emission factors and related to total gasoline consumption. In the light of more restrictive legislation on exhaust emissions in recent years, and associated improvements in engine design, the previous emission factors are probably no longer valid. The present study utilises measurements of hydrocarbons in exhaust gas carried out by Warren Spring Laboratory (2) in a range of vehicles in on-the-road tests under urban, rural and motorway driving conditions. The total hydrocarbon emission by gasoline exhausts is derived from the Warren Spring data, OECD (3), together with SMMT data (17) on total car population (115 million cars for OECD Europe), annual mileage (10.5 x 10^3 km) and mileage breakdown. Total evaporative losses from carburettors and fuel tanks resulting from the diurnal cycle and heat soak assume a factor of 0.8 g/km. This is based on the findings of a CONCAWE study currently in progress.

In the absence of recent measurements of on-the-road emissions from diesel vehicles, and the very wide range of performance of such vehicles, an emissions factor of 5 kg/tonne of fuel used has been applied to total European diesel demand in 1983 (1). This emissions factor is based on recent work by Ricardo Consulting Engineers and studies within the oil industry.

No recent data for aircraft, railways or inland/coastal shipping are available. Emissions factors of 2, 10 and 5 kg/tonne of fuel have been applied to 1983 fuel consumption (1) in these sectors. These factors are taken from CRC 1972 (4) for aircraft, EPA 1976 (5) for railways and Norwegian Institute for Air Research 1974 (6) for shipping, respectively. Although developments in engine design may have reduced these factors somewhat during the last ten years, the contribution of these sources to the total atmospheric burden is small, and it will make little difference to the overall picture.

2.2 OIL INDUSTRY

Volatile organic compounds emission from off-shore oil production are estimated by Kort (7) to be 0.01 wt% of production, through fugitive emissions, production water, storage tank losses, etc. This factor has been applied to the Western European 1983 production of crude oil and natural gas liquids of 161 x 10^6 tonnes (1). In common with other sources this figure does not include methane, which may be substantial in this case.

The main source of VOCs from marine transport and storage of crude emissions is hydrocarbon-laden air or inert gas displaced from empty crude tanks by ballast water, a new cargo or inert gassing. Kort (7) estimate a total figure of 60 x 10^3 tonnes for 1979. Pro-rating this to European crude movements for 1983 gives a figure of 50 x 10^3 tonnes.

Based on a limited number of recent estimates in France and Germany the typical level of refinery emissions of VOCs has been calculated to be 0.04 % of the crude oil processed. This factor represents the aggregate of losses from refinery process units, oil/water separators, tankage and product handling. The emission factor would be expected to vary in relation to the conditions of the plants (type, design, age etc.), the complexity of the processes (cracking, deep conversion, lube production etc.), the type of raw materials used (crude oils, light/heavy cut of the barrel), and the shipment pattern (pipeline, tanker, barge, road, rail). Adopting this factor and assuming a throughput of 551.3 x 10^6 tonnes for Western Europe in 1983, refinery losses are estimated at 220 x 10^3 tonnes. Any excursions above/below the level of 0.04 wt% on throughput due for the reasons given would not be expected to result in any significant change in the small contribution of refineries to total VOC emissions.

A CONCAWE study currently in progress found typical losses of gasoline during distribution from the refinery, via the terminal, to the service station and into the vehicle to be 0.49% of the volume of product transferred. Applying this factor to Western European consumption in 1983 of 104.2 x 10^6 tonnes gives a loss to the atmosphere of 510 x 10^3 tonnes. Of this total some 70% is lost at service stations during receipt, storage and dispensing of the gasoline into consumers' vehicles. Refuelling of vehicles is estimated to cause the loss of 0.18% of the volume of the liquid transferred, and this constitutes 188 x 10^3 tonnes of the total 510 x 10^3 tonnes emitted during the entire distribution chain.

2.3 SOLVENTS

Solvents constitute one of the largest sources of VOC emissions although some have low chemical reactivity.

In estimating emissions it has been considered that all materials sold as solvents enter the atmosphere unless they are incinerated or otherwise disposed of. It seems likely that about 15% of solvents used by consumers are disposed of in non-polluting ways (8), leaving some 85% to enter the atmosphere. Total production of solvents in Western Europe is in the region of 4.7 x 10^6 tonnes, which therefore leads 4.0 x 10^6 tonnes being released into the air.

An estimated breakdown of solvent usage in OECD Western Europe is given in Table 2.

Table 2 Breakdown of solvent usage in OECD Western Europe

Solvent (tonnes x 10^3)	Sector					
	Paint	Printing	Adhesives	Pharmaceutical	Other	Total
Aliphatic hydrocarbons	485	35	100	–	500	1120
Aromatic hydrocarbons	560	90	30	–	270	950
Alcohols	255	50	25	230	430	990
Other oxygenates	510	70	80	120	260	1040
Chlorinated hydrocarbons	–	–	60	–	570	630
Totals	1810	245	295	350	2030	4730

Comparative data show an overall trend towards reduction in solvent usage due to advances in technology of paints and coatings.

2.4   MANUFACTURING INDUSTRY

Process and product storage emissions for the chemical industry are assumed to be related to its petroleum feedstock by the same ratio as emissions from an oil refinery to its gasoline production. Taking the consumption of petroleum feedstocks to be 40 x 10^6 tonnes in 1983 (1), the emissions are 85 x 10^3 tonnes.

Other industries which are significant emitters of volatile organics, other than solvents, include brickmaking, coking and fermentation. Total emissions derived using EPA emission factors (5) are estimated at 320 x 10^3 tonnes per year.

2.5   NATURAL GAS

The EPA emission factor (including methane) for natural gas production is 3.1 x 10^{-3} kg/m^3. If the average content of non-methane hydrocarbons is taken as 11% (see Section 3.5), then the VOC loss arising from a production rate of 2.2 x 10^{11} m^3 is 75 x 10^3 tonnes of hydrocarbons, excluding methane.

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Substantial leakage of natural gas takes place through joints in distribution pipe designed for town gas. Assuming a 3% loss, then the non-methane hydrocarbons from this source amount to 570×10^3 tonnes per year. If methane were included the figure would be an order of magnitude higher.

2.6 SOLID WASTE DISPOSAL

Brice and Derwent (8) have estimated total emissions of VOCs from this source in the UK. In the absence of better data, these estimates have been pro-rated upwards sevenfold, this being the ratio of UK population to that for Western Europe in total. The same authors estimate that 50% of the emissions are methane, resulting in an emission of 110×10^3 tonnes per year of non-methane VOC's.

2.7 STATIONARY COMBUSTION

Estimates of VOC emissions resulting from steam-raising/heating/ power generation are based on EPA emission factors (5) and estimated 1983 consumption of oil, coal and gas in these sectors (1). Data used are given in Table 3.

Table 3 1983 fuel consumption and emissions estimates

Fuel	Emissions factor	Consumption	Total emissions (tonnes x 10^3)
Oil	0.15 kg/tonne	52.4×10^6 tonnes	8
Gas	50 kg/10^6 m^3	2×10^{11} m^3	10
Coal			
Electricity generation	0.15 kg/tonne	246×10^6 tonnes	37
Industrial	0.7 kg/tonne	34×10^6 tonnes	24
Domestic	2.0 kg/tonne	31×10^6 tonnes	62
Total			141

If it is assumed that approximately 40% of the emissions are methane (16) then the total figure for non-methane VOC emissions in 1983 is 85×10^3 tonnes.

2.8   NATURAL SOURCES

Three major natural sources of VOC emissions are recognised:
production of methane by bacterial decay under anaerobic conditions
in swamps; emissions of terpenes, etc. by trees; and release of
esters, ketones and other oxygenates from grassland, especially
after cutting. Taken together they greatly exceed anthropogenic
emissions. Methane from vegetable decay is probably responsible for
the high (1.5 ppm) concentration of this material in the global
atmosphere (9). Recent work (10) has suggested that it is
increasing at a rate of 1.2 - 1.9% per year. As in the case of
anthropogenic emissions, methane is not included in the inventory
of natural emissions, in view of its relative non-reactivity (see
Section 4).

Based on measurements of terpene flux above actual forests (11) and
in the laboratory (12), it is possible to conclude an average
emission factor of 30 $\mu g/m^2$ x min for typical European weather
conditions. Based on a forest area of 52 x 10^6 hectare in Western
Europe, the total annual emission from forests is 8 x 10^6 tonnes.

A further quantifiable source of natural emissions, from leaf
litter and pasture, is given (13) as 2.7 $\mu g/m^2$ x min in summer.
Taking the appropriate area to be 60% of the total Western Europe
land area (250 x 10^6 hectare), the emissions rate is 2 x 10^6 tonnes
per year.

Taken together, natural sources, excluding methane, contribute
about 10 x 10^6 tonnes of VOCs per year, which is approximately
equal to the annual total anthropogenic emissions.

3.   COMPOSITION OF VOC EMISSIONS

3.1   MOBILE SOURCES

Fairly consistent data are available from a wide variety of sources
on the detailed composition of exhaust from gasoline engines, but
those for diesel exhaust, other than on polynuclear aromatics, are
scant. No data are available for aircraft, rail or ship. Some
typical data are given in Table 4.

Table 4   VOC composition of typical gasoline engine exhaust

Compounds		Quality (wt %)
Saturates % wt	C_2	1.2
	C_5	7.5
	C_6	4.6
	Total	13.3
Unsaturates % wt	C_2	10.0
	C_3	6.0
	C_4	1.5
	Total	17.5
Aromatics % wt	C_6	8.8
	C_7	16.5
	C_8	19.3
	C_9	10.6
	Total	55.2
Others		14.0
Total		100.0

These compositional data do not necessarily correspond with the
hdyrocarbon composition of the in-going gasoline. It is known that
extensive chemical transformation of unburnt hydrocarbons takes
place in the combustion chamber.

Although copious data are available on the breakdown of polynuclear
aromatics associated with particulates in diesel exhaust, there is
little recent information available on the VOC composition of
gaseous diesel exhaust. Hurn and Marshall (14) report the following
typical results from a study of 50 diesel engines.

Table 5   VOC composition of typical diesel exhaust

Compounds	Total hydrocarbons (mol%)	
	Idle	Full power
C_2 – C_5	32	44
C_6 – C_{11}	14	2
C_{12} – C_{18}	35	38
C_{18} – C_{24}	10	15
Other	9	1
Total	100	100

The components in the range C_{18} – C_{24} and a large part of those in the C_{12} – C_{18} range are likely to be aromatic in type. These data are broadly consistent with those published by Springer and Baines (15) and other.

3.2   OIL INDUSTRY

As might be expected, relative concentrations of individual hydrocarbons and types of hydrocarbons vary substantially from one location to another. It is therefore almost impossible to obtain a true "average" refinery emissions composition. Perhaps the best that can be done is to measure composition at a location downwind of the refinery, although it is by no means certain that this is truly representative. Measurements made at two refineries within the process area are shown in Table 6.

Table 6   Hydrocarbon composition of refinery air samples
(process area)

Compounds	Air sample composition (wt%)	
	Refinery A	Refinery B
Alkanes,		
Ethane	14.8	2.4
Propane	13.7	14.5
C_4^+	55.9	68.9
Alkenes		
Ethene	3.3	0.5
C_3^+	8.3	0.4
Aromatics		
Benzene	1.8	2.6
Toluene	2.2	5.7
Xylene	--	5.0
Total	100.0	100.0

Limited data suggest that refinery emissions are essentially
saturated, as has been assumed previously, the saturated
hydrocarbon content lying between 80 - 90 wt%. The balance of 10 -
20 wt% is unsaturated and/or aromatic hydrocarbons, the actual
values depending on the nature of the refinery processes installed.

As illustrated in Table 7, these compositional data correspond
fairly closed with those for gasoline losses. However, where a
refinery is also associated with a chemicals complex the VOC
emissions profile may be significantly different.

Measurements of hydrocarbon composition in the gasoline distribution
system are more consistent reflecting the fact that we are dealing
with a fairly constant, single product. Thus, measurements taken at
tank vents, and downwind at the terminal fence in unrelated locations
give essentially the same result.

Table 7   Hydrocarbon composition of gasoline terminal air samples

	Tank vent (Terminal 1) (wt%)	Downwind fence (Terminal 2) (wt%)
Alkanes		
Ethane	-	-
Propane	-	2.0
C_4+	89.2	89.1
Alkenes		
Ethene	-	-
C_3+	6.9	6.5
Aromatics		
Benzene	1.1	1.5
Toluene	2.0	0.9
Xylene	0.8	-
Total	100.0	100.0

Limited data on emissions from service stations are consistent with the known composition of emissions from terminals handling gasoline.

3.3   SOLVENTS

Since no measured data on the composition of VOC emissions from solvents are available, the logical assumption has been made that, on a global basis, solvent VOC emissions will be proportional to production/consumption of individual solvents. These are estimated in Table 8.

Table 8   Estimated relative VOCs from solvents

Solvent type	Relative production/consumption (wt%)
Saturated hydrocarbons	24
Aromatic hydrocarbons	20
Alcohols	21
Ketones esters and ethers	22
Chlorinated hydrocarbons	13
Total	100

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3.4.   MANUFACTURING INDUSTRY

Brice and Derwent (8) quote data on the types and quantities of
emissions from this source in the UK in 1977, as shown in Table 9.

Table 9   VOC emissions from UK manufacturing industry in 1977

Industry	Emission	
	Type	Amount (wt%)
Synthetic rubber	Butadiene, styrene, acrylonitrile	10.0
Fermentation	Alcohols, esters, ketones, aldehydes	45.0
Plastics	Vinyl chloride	0.5
Organic chemicals	Phthalic and maleic anhydrides, quinones	0.5
Bricks	Hydrocarbons from combustion	6.0
Paint	Solvent	37.0
Total		100.0

3.5   NATURAL GAS

The composition of natural gas from individual fields is known with
considerable accuracy. Since the contribution of natural gas to
atmospheric VOCs is exclusively through losses during production and
in transmission, the composition of this contribution is identical
to the composition as found. Table 10 shows typical ranges for
natural gas from North Sea production only.

Table 10   Typical composition of North Sea natural gas production

Compound	Composition (wt%)
Methane	84 – 88
Ethane	6 – 8
Propane	1 – 3
Butane	0.5 – 1.5
C_5-C_9 saturates	0.5 – 1.0
C_6-C_7 aromatics	0.2
CO_2	0.5 – 5.0

3.6   SOLID WASTE DISPOSAL

No information on composition has been found.

3.7   STATIONARY COMBUSTION

Trijonis et al (16) conclude that 60% of total VOC emissions from stationary sources should be included in the non-methane VOC category but do not give typical compositions.

4.   DISCUSSION AND CONCLUSIONS

The two biggest contributors of man-made VOC emissions to the
atmosphere in Western Europe are mobile sources (especially road
transport) and solvents, each contributing by about 40%. Road
vehicle exhaust emissions are already being tackled by the EEC and
other countries by progressively tightening up legislation limiting
total hydrocarbon and other gaseous emissions. Assuming that
proposals for new automotive emission limits currently being
considered by the Council of Environmental Ministers are eventually
accepted in their present form, something like a 30% reduction in
tonnage of hydrocarbon emitted from passenger cars can be expected
by 2000 AD. Proposals for control of emissions from heavy-duty
diesel vehicles are also expected in the near future. In the
absence of significant reductions in other sources of VOCs this
will mean a significant change in the relative contribution from
mobile sources.

The oil industry in its own operations is responsible for about 8%
of man-made VOC emissions. Of this, about 2% is directly from
refineries. There is some degree of uncertainty about emission
factors from refineries, as there is also for other sources, but
even if the factor were doubled, refineries would still contribute
less than 5% either in Western Europe as a whole or in the EEC
Member States.

The main contributor from the oil industry point of view is
gasoline distribution, with about 5% of total anthropogenic
emissions. This fact has already been recognised by the oil
industry which has taken appropriate steps (for example, bottom
loading and vapour recovery) and is instituting further measures to
recover valuable products which otherwise represent an economic
loss.

Estimates for the ten EEC Member States show slightly higher
contributions from mobile sources, natural gas and the oil
industry, than those for OECD Western Europe as a whole, reflecting
higher traffic density in the Community, substantial natural gas
production in UK and the Netherlands and the fact that a high
proportion of Europe's refineries are situated in EEC Member
States.

Estimates show that VOC emissions from natural sources are
approximately equal to those produced by man. In this context the
emissions directly attributable to oil industry operations are only
about 4% of the total, and refineries as such contribute only about
1% of the overall atmospheric burden. However, it is recognised
that in specific urban or industrial areas the contribution from
anthropogenic sources may be higher.

Methane has traditionally been excluded from VOC and hydrocarbon
inventories on the grounds that it plays a very limited role in
short-term photoxidant formation. However, in the long term it may
contribute significantly to the formation of background ozone. For
this reason there is a good case for considering the substantial
emissions of methane from both natural and anthropogenic sources
when quantifying atmospheric VOC emissions.

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