Pollutants From Combustion: Formation & Impact on Atmospheric Chemistry

Pollutants from Combustion

Formation and Impact on Atmospheric Chemistry

edited by

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Kluwer Academic Publishers

Dordrecht / Boston / London

Published in cooperation with NATO Scientific Affairs Division

Proceedings of the NATO Advanced Study Institute on
Pollutants from Combustion Formation and Impact on Atmospheric Chemistry
Maratea, Italy
13-26 September 1998

A C.I.P. Catalogue record for this book is available from the Library of Congress.

ISBN 0-7923-6134-2 (HB)

Published by Kluwer Academic Publishers,
P.O. Box 17, 3300 AA Dordrecht, The Netherlands.

Sold and distributed in North, Central and South America
by Kluwer Academic Publishers,
101 Philip Drive, Norwell, MA 02061, U.S.A.

In all other countries, sold and distributed
by Kluwer Academic Publishers,
P.O. Box 322, 3300 AH Dordrecht, The Netherlands.

Printed on acid-free paper

Printed in the Netherlands.

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Series C: Mathematical and Physical Sciences – Vol. 547

PREFACE

This volume is based on the lectures presented at the NATO Advanced Study Institute: (ASI) « Pollutants Formation from Combustion. Formation Mechanisms and Impact on Atmospheric Chemistry » held in Maratea, Italy, from 13th to 26th september 1998.

Preservation of the environment is of increasing concern in individual countries but also at continental or world scales. The structure of a NATO ASI which involve lecturers and participants of different nationalities was thought as especially well suited to address environmental issues.

As combustion is known to substantially contribute to the damaging of the atmosphere, it was natural to concentrate the ASI program on reviewing the currently available knowledge of the formation mechanisms of the main pollutants liberated by combustion systems. In most situations, pollutants are present as trace components and their formation and removal is strongly conditioned by the chemical reactions initiated by fuel consumption. Therefore specific lectures were aimed at defining precisely the general properties of combustion chemistry for gaseous, liquid and solid fuels. Physical factors can strongly affect the combustion chemistry and their influence was also considered.

An interesting peculiarity of this specific ASI was to complement the program with a substantial part concerned with the impact of the main combustion pollutants: NOx, aromatics, soot, VOCs, sulphur and chlorinated compounds, on atmospheric chemistry. That bringing together scientists from two different domains: combustion and atmospheric chemistry would be very fruitful to derive an overview of pollution linked to combustion was first suggested by my colleague Georges Le Bras, and I am pleased to thank Georges for this very positive suggestion.

I am deeply grateful to Richard Wayne who accepted to take in charge the organization of the section on atmospheric chemistry. Richard deserves a special thank for having done all the work without the pleasure of being present in Maratea since health problems forced him to stay in Oxford. My colleague Gilles Poulet carried on Richard's work and I value his contribution.

Selim Senkan, Karel Svoboda and Jürgen Warnatz have been involved as co-organizers in the preparation of the combustion section and their contribution was very valuable.

I would like to express a special thanks to all lecturers for the care taken to prepare lectures understandable by participants coming from the two different areas considered in this ASI. As high-level specialists in their domains they are involved in many tasks and their efforts to be present and available for free discussions with the participants deserve to be mentioned.

Of course the success of an ASI depends strongly on the active involvement of the participants. I am very pleased to thanks again all of them for their interest and the friendly relationships that prevailed during the whole duration of the ASI.

Finally, I thank the NATO Scientific and Environmental Affairs Division for their sponsorship.

Christian Vovelle

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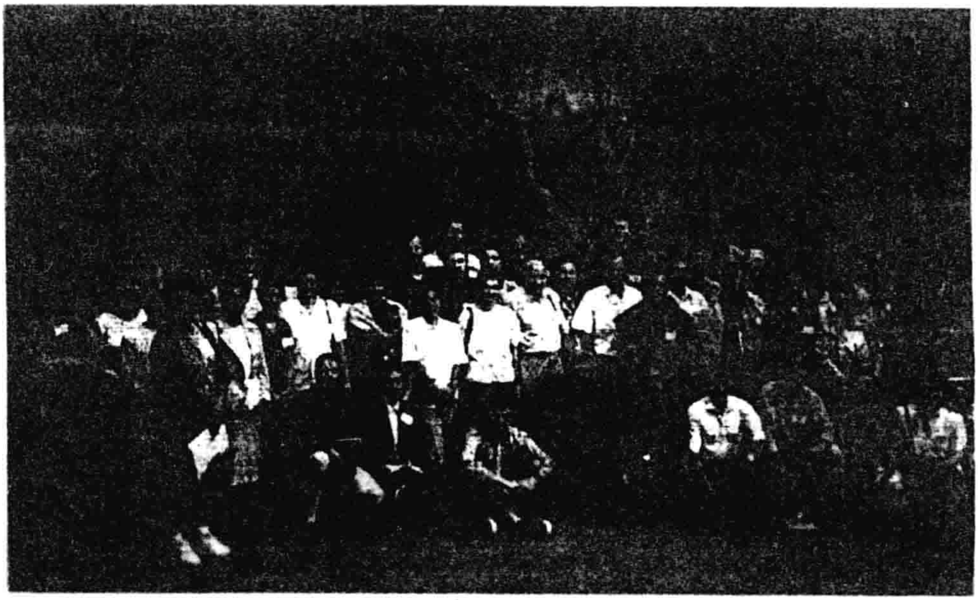

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SOURCES OF POLLUTANTS

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1. Introduction

Sources of pollutants to the atmosphere are manifold. A rough division into the following source categories can be made:

a) combustion processes,
b) industrial production processes,
c) storage and distribution of goods, including fossil fuels,
d) application of volatile organic solvents and solvent containing products,
e) biological and natural processes.

Combustion processes comprise, for example, internal combustion vehicles and combustion plants and furnaces. Industrial processes with relatively high emissions of pollutants are production of coke, cement, glass, pig iron, steel, castings, mineral oil products, wood pulp and sugar. The distribution of oil and gas causes VOC emissions, handling of loose materials leads to dust emissions. Solvent containing products include paints and varnishes, printing inks, consumer goods for private use (e. g. products for cleaning and personal hygiene, washing agents), metal degreasing agents, refrigerants or adhesives.

A large variety of biological and natural processes emit trace substances, for example the activities of methagene bacteria in wetlands (moor, floodplains), rice fields, waste disposal sites, excreta (liquid manure, dung), digestive tracts of animals (in particular ruminants and termites) and sediments (oceans and freshwater). Other examples are processes of biosynthesis (VOC emissions of deciduous trees and conifers), erosion and sea salt (particulate emissions), biological processes in aquatic ecosystems and soils, lightning (NO emissions), and NH_3 emissions form livestock and use of inorganic fertilisers.

Corresponding to the subject of this book, this chapter focuses on combustion processes. These are responsible for more than 95% of the CO_2 and NO_x emissions, about 92% of CO emissions, 85% of SO_2 emissions and 40% of NMVOC emissions in the countries of the European Union (1994) and about 60% of PM10 emissions in Germany (1996).

C. Vovelle (ed.), Pollutants from Combustion. 1–15.

2. Combustion processes

Combustion processes can be split into mobile and stationary sources. Mobile sources include internal combustion engines of road and track vehicles, ships and aircraft. Stationary sources contain many different sizes and techniques with as well large power plants with several thousand of megawatts of thermal power as small firings for heating or warm water production with a power of a few kilowatts.

A large number of different pollutants is emitted from combustion processes, which can be classified into

- products of complete combustion (CO_2, H_2O),
- by-products of complete combustion (NO_x),
- products of incomplete combustion (CO, soot, volatil organic compounds as e.g. alkanes, alkenes, alkynes, aromatics and aldehydes),
- products stemming from additional substances contained in the fuel (SO_2, NO_x, particulates, heavy metals, HCl, HF and others).

2.1. PRODUCTS OF COMPLETE COMBUSTION

Products of a complete combustion of fossil fuels are carbon dioxide and water, following the overall reaction scheme

$$C_nH_m + (n+m/4)\,O_2 \rightarrow n\,CO_2 + m/2\,H_2O \qquad (1)$$

where C_nH_m represents fossil fuels or other organic materials with a certain ratio of C/H and $(n+m/4)\,O_2$ indicates the stoichiometric amount of oxygen, which is theoretically required for a complete combustion. The CO_2 emission factor decreases with decreasing C/H ratio of the fuel (Table 1). CO_2 emissions can be reduced by switching to a fuel with lower C-content, by using renewable or nuclear energy, by using energy in more efficient way or by saving energy.

TABLE 1. CO_2 emission factors for complete combustion of fossil fuels

fuel	Emission factor kg CO_2/G J
Lignite	112
Hard coal	93
Heavy fuel oil	78
light fuel oil / diesel	73
gasoline	71
natural gas	55

2.2. NITROGEN OXIDES

NO is formed during combustion by processes involving as well nitrogen from the air (thermal NO and prompt NO) as nitrogen contained in the fuel (fuel NO). The detailed mechanisms of NO formation are explained in detail in the contribution of Tom Bowman in this book.

Thermal NO starts to be formed at about 1300 ° C, the production rises rapidly with rising temperature. In addition the NO formation is increased with increasing oxygen surplus and increasing residence time of the fuels in the zone with high temperature. Prompt NO normally causes only a limited share of the total NO formation.

Fossil fuels, especially coal and oil products, contain organic nitrogen compounds, the nitrogen content amounts to about 0,8-1,5 % for coal and 0,1-0,6 % for fuel oil. The combustion of the fuel leads to a partly oxidation of the fuel nitrogen. With average conditions, about 20-40 % of the nitrogen in coal, 60-70 % of the nitrogen in heavy fuel oil and 80-90 % of the nitrogen in light fuel oil is oxidised. The formation of fuel NO depends on the oxygen surplus, whereas the temperature only has a minor influence.

Table 2 shows NO_x emission factors for typical combustion processes without special primary and secondary measures for NO_x reduction. The highest emission factors are allocated to passenger cars with Otto engine - without three way catalyst - driving with high speed followed by diesel cars using high engine power. With decreasing speed NO_x emissions decrease, so the main source of NO_x is traffic on highways. As new passenger cars possess catalysts, the share of the NO emissions caused by heavy duty vehicles is increasing. For stationary combustion, plants with high thermal power have higher temperatures and so higher NO emissions than smaller plants.

TABLE 2. NO_x-emissions of selected technical combustion processes without specific impairment measures (derived from [1])

Combustion process	NO_x-concentration^a (exhaust) in mg/m^3	NO_x-emission in mg/kWh
Energy production		
Hard coal dust firing		
wet bottom	1.200 - 3.000	1.400 - 3.600
dry bottom	700 - 1.800	900 - 2.300
Soft coal firing	600 - 1.000	850 - 1.400
Industry		
Roasting furnace	150 - 650	200 - 950
Fluidised bed furnace		
steady state	100 - 1.000	140 - 1.400
circulatory	80 - 300	100 - 410
Oil combustion	300 - 1.100	300 - 1.200
Gas combustion	100 - 800	85 - 700
Residential sectors		
Oil combustion	80 - 250	80 - 260
Gas combustion		
blowpipe	60 - 170	50 - 150
atmospheric burner	100 - 200	85 - 170
Machinery		
Passenger car with Otto engine	1.000 - 8.000	900 - 7.000
Passenger car with Otto engine (idling)	20 - 50	18 - 50
Passenger car/lorry with Diesel engine	400 - 3.000	1.000 - 3.500
Passenger car/lorry with Diesel engine (idling)	20 - 50	70 - 180

^{a}partly related to different excess air values

2.3. PRODUCTS OF INCOMPLETE COMBUSTION

In practice technical combustion processes are more or less incomplete. One reason for this could be a lack of oxygen. In general, the amount of oxygen needed for the combustion process is higher than the stoichiometric amount, due to unavoidable inhomogenities of fuel/oxygen mixtures. Other reasons of incomplete combustion might be low combustion temperatures or a residence time of the fuel in the burner zone being too short.

Incomplete combustion of fossil fuel leads to emissions of CO (an intermediate product of the oxidation of carbon to CO_2), soot and VOCs. Hydrocarbons can enter the flue gas in a partly oxidised form (e. g. aldehydes) or even in an unoxidised form (e. g. alkanes, aromatics). Also the formation of new hydrocarbons (alkenes, alkines, aromatics) by radical reactions can be observed.

TABLE 3. Total VOC emission factors of the combustion of fuels in different sectors in Germany; values given in kg VOC/TJ_{fuel} (derived from [2] and [3])

source type	further description	gasoline	diesel
passenger car	gasoline, without catalyst	473	
passenger car	gasoline, with 3-way-catalyst	17	
passenger car	diesel, before 1990		38
passenger car	diesel, after 1996		33
light duty vehicle	gasoline, without catalyst	454	
light duty vehicle	gasoline, with 3-way-catalyst	17	
light duty vehicle	diesel		49
heavy duty vehicle	diesel, before 1990		170
heavy duty vehicle	diesel, after 1996		135
moped	2-stroke	4859	
moped	2-stroke, with uncontrolled catalyst	2168	

		natural gas	light fuel oil	heavy fuel oil	coal	wood/ solids
households	space heating	5	5		400	350
households	warm water	8	5			1600
households	cooking	25				2000
other consumers		5	5		30	210
industry		5		8	30	210
power plants		0.6		7	3	65*

* waste incineration

The more incomplete a reaction is, the greater are the emissions. Relative to energy consumption, internal combustion engines are of major importance. As it is shown in Table 3, two-stroke engines of mopeds are characterised by the highest VOC emission factor, here expressed in kg of emitted VOCs per TJ fuel used. The exhaust gas of two-stroke engines contains not only uncombusted or partly combusted gasoline but also uncombusted or partly combusted lubricants in large amounts. VOC emission factors for 4-stroke gasoline engines (without catalyst) are considerably lower, followed by diesel engines of heavy duty vehicles, light duty vehicles and, finally, diesel engines of passenger cars with the lowest emission factors for engines without emission control. However, the lowest emission factors for mobile sources are allocated to gasoline engines equipped with 3-way catalysts.

Decisive emission parameters controlling emission rates from the combustion of fossil fuels in stationary plants and furnaces are fuel type and combustion technique, giving an indication on the completeness of combustion processes. It is obvious that the combustion of gaseous and liquid fuels in general leads to lower VOC emissions than the combustion of coal, wood or other solids. The only exceptions are coal dust combustion installations, to which lower emission factors are allocated than to heavy fuel oil power plants.

Especially for solid fuels, the VOC emission factor for large combustion plants is by far lower than the emission factor for small furnaces used in households. The reason for this is that fossil fuel power plants are equipped with complex and costly combustion technologies, ensuring more or less complete combustion of specially prepared coal, whereas the construction of small furnaces in the household sector is relatively simple and more designed for easy operation than for really optimised combustion. Medium size industrial and commercial combustion installations have emission factors between the extreme values.

It is not only the total amount of VOCs emitted that is an important parameter in air quality evaluation procedures, but also the specific composition of the VOC.

Examples of the composition of VOC tailpipe emissions are summarised in Table 4 for conventional passenger cars without catalyst, passenger cars with 3-way-catalysts and for diesel cars and duty vehicles. The values represent the average composition for different driving conditions. Detailed information on the dependency of the composition of VOC tailpipe emissions on driving modes are subject of ongoing research.

Some of the more important aspects of the VOC profiles discussed in Patyk et al. [4] are:

- 50 to 75 % of the VOC emissions from passenger cars with Otto engines consist of fuel compounds which are not combusted,
- the efficiency of 3-way-catalysts in passenger cars with Otto engines is around 90 %, but differs from species to species,
- some hydrocarbons are produced in significant amounts by the initially combustion process or by subsequent catalytic reactions (e. g. high amounts of ethene and ethyne in conventional vehicles with Otto engines and high amounts of methane in the exhaust gas of cars with 3-way-catalysts).

Especially for diesel engines, the 'other VOCs' indicated in Table 4 are normally high-molecular paraffins [5]. However, Patyk and Höpfner [4] emphasize the large uncertainty in VOC compound profiles for diesel engines.

TABLE 4. Composition of VOC emissions from internal combustion engines; values given in % mass , based on [4].

	otto engine 4-stroke without catalyst	otto engine 4-stroke 3-way-catalyst	otto engine 2-stroke without catalyst	diesel engine
paraffins				
methane	4.0	14.0	7.0	2.4
ethane	0.8	2.0	1.0	
propane	0.3	0.5		
butane	3,0	6.0	2.0	
iso-butane	2.0	3.0	0.3	
pentane	2.0	2.5	3.0	
iso-pentane	5.0	7.5	4.0	
olefines				
ethene	7.0	4.5	5.0	12.2
propene	4.0	2.5	2.0	4.7
1-butene	0.4	0.1		
iso-butene	2.5	1.5		0.8
cis-2-butene	0.2	0.2	0.4	0.8
trans-2-butene	0.8	0.6	0.2	0.7
1,3-butadiene	0.6	0.4		1.1
pentene	1.1	0.7		
alkynes				
ethyne	5.5	3.0	4.0	2.8
aromatic hydrocarbons				
benzene	5.0	6.0	5.0	1.9
toluene	11.5	10.0	12.1	0.8
xylene	10.0	9.0	11.0	0.8
ethylbenzene	2.5	2.0	2.8	0.3
C_9-aromatics	7.5	6.0	8.3	
aldehydes				
formaldehyde	1.5	1.0	0.6	8.1
acetaldehyde	0.7	0.6	0.2	4.2
acrolein	0.4	0.2	0.0	2.1
benzaldehyde	0.3	0.2	0.2	1.8
tolualdehyde	0.5	0.4	0.2	1.0
ketones				
acetone	0.6	0.5	0.1	1.5
other VOCs	20.3	15.1	30.6	52.0

TABLE 5: Estimated composition of VOC emissions from combustion installations; values given in % mass (data mainly based on[5])

	Coal	Wood	Fuel oil	Gas
Alkanes				
Methane	75	25	10	75
Ethane	3.75	7.5		12
Propane	2.5	1.13		4
Butane		0.38		4
Alkanes > C_4	1.25	2.25		
Alkanes, not specified			65	
Alkenes				
Ethene	7.5	22.5		1.8
Propene	1.25	4.5		0.6
Butene		0.38		0.6
Pentene		1.5		
Alkenes > C_3	1.25			
Alkenes, not specified			10	
Alkynes				
Ethyne	1.25	7.5		
Propyne		0.38		
Aromatic hydrocarbons				
Benzene	1.25	11.25		
Toluene	0.25	3.75		
o-Xylene	0.09	0.38		
m-Xylene	0.08	0.54		
p-Xylene	0.08	0.55		
Ethylbenzene		0.38		
Aromatics, not specified			5	
Aldehydes				
Formaldehyde		1.5		1.8
Acetaldehyde		0.75		0.12
Propionaldehyde				0.04
Acrolein		1.5		
Aldehydes ~ C_4		2.25		0.04
Aldehydes, not specified			10	
Other VOCs				
Ketones, not specified		1.13		
VOCs, not specified	4.5	3		

At present 3-way-catalysts represent the most effective measure for the control of VOC exhaust gas emissions from vehicles with Otto engines (see Tables 1 and 2, above). Emissions can be reduced by about 90 % (average) with 3-way-catalysts relative to a vehicle without a catalyst. In principal, another reduction measure is to keep internal combustion with an excess of air ($\lambda > 1$). However, current European emission standards for exhaust gas from motor vehicles (EURO-3 and 4) and the planned tightening (EURO-5) cannot be met with this technology, unless it is combined with a NO_x reducing catalyst.

The relative importance of VOC emissions from the combustion of solid fuels has already been mentioned. The values listed for combustion installations in Table 3 represent typical values over longer time periods. However, short-term peeks with very high emission rates can be observed during the start-up and shut-down of combustion installations, during operations under partial load or just after the feeding of solid fuel combustion installations with new material.

The relative composition of VOC emissions from combustion installations varies considerably with different fuel types. The values in Table 5 give only rough clues about the VOC-profiles from smaller furnaces. Due to their limited importance, corresponding data for large combustion installations are not yet available. However, the table shows clearly that methane is the most important component in coal and gas fired installations. With the combustion of wood, significant amounts of alkenes, benzene and other aromatic hydrocarbons are released into the atmosphere. The composition of oil and gas fired installations indicates high amounts of unburned fuel, with significant amounts of higher molecular alkanes and ethane emitted respectively.

Possibilities for the abatement of VOC emissions from small solid fuel furnaces are mainly by ensuring their correct construction and proper operation. Concerning construction options, combustion with a stable and uniform flame has to be ensured even in case of feeding varying amounts of coal or wood. A proper operation mode means that, for example, feeding the combustion installation with wet or damp fuel wood and completely closing ventilation flaps should be avoided.

CO emissions show a similar pattern (Table 6). Combustion of solid fuels causes higher emissions than combustion of liquid or gaseous fuels. The larger the plant, the better the combustion and the lower the CO emissions.

TABLE 6. Energy-related emissions of combustion plants; values in kg CO/TJ fuel used [2],[6].

	Wood	Coal	Light fuel oil	Heavy fuel oil	Gas	Diesel fuel	Petrol
Households							
Space heating	2.120	5.000	45		60		
Hot water	12.000		45		60		
Cooking	15.000				300		
Other consumers	2.460	500	45		45	970	
Industry	2.460	100	10	10	45		
Power stations		17		3	1		

Table 7 shows CO emission factors for road transport. Especially for situations with congestion very high emission factors occur.

TABLE 7. CO emission factors of vehicles for selected driving patterns,in g CO/km (derived from[3])

	motorway	other non-urban transport	urban transport	congestion
passenger car (without catalyst)	3,0	1,5	3,1	12,9
light duty vehicle	5,5	2,1	3,5	11,8
heavy duty vehicle	2,0	2,0	4,7	12,4