

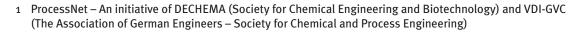




Kommission Reinhaltung der Luft im VDI und DIN – Normenausschuss KRdL

# **Fine Particles**

A Strategy Paper Prepared by the Joint Committee "Fine Particles" of ProcessNet<sup>1</sup>, GDCh<sup>2</sup> and KRdL<sup>3</sup>



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# Table of Contents

1.	Objectives of the Committee	4
2.	Source-Specific Aspects of Fine Particles	5
	2.1 Overview of the Relevant Sources of Fine Particles	5
	2.2 Agriculture	6
	2.3 Firing systems	7
	2.4 Road transport	7
	2.5 Industrial Sources	9
	2.6 Biological Particles	9
	2.7 SOCs and VOCs	10
3.	Further Aspects	12
	3.1 Effect-related metrics of fine particles	12
	3.2 Climate, weather and the transport of fine particles	13
4.	Members of the Joint Committee	14

# 1. Objectives of the Committee

The joint committee "Fine Particles" (JC-FP) represents a body of experts for chemical, chemical-technological and health aspects of fine particles in the environment. The term `fine particles` includes the subset of air-borne aerosols which pass the nose-bronchial filter due to their fineness and enter the pulmonary alveoli. The emergence and prevention of particulate emissions from natural and anthropogenic sources, the measurement and atmospheric transformation of fine particles, the dispersion and the fate of fine particles, as well as the effect on human beings and on the environment are also included. In the context of scientific discussion and environmental political as well as regulatory aspects, the committee pursues the following objectives:

- » Discussion and evaluation of the state of R+D of selected subjects.
- » Identification of knowledge gaps and as the circumstances require demands for action.
- » Initiation of R+D-initiatives in order to remove shortcomings.
- » Release of statements on scientific results and regulatory actions on the topics of the committee.
- » Holistic evaluation of fine particles including costeffectiveness considerations.

The committee usually meets twice a year for one day. Special task forces which create their own working plans may be established for particular projects (e.g. the *writing of statements and publications, or the preparation of special colloquia*). Extraordinary meetings may be held at any time. Exemplary for the results of the preceeding work is the statement of the JC-FP concerning the low-emission zones ("Feinstäube und Umweltzonen"; R. Zellner et al.; Chemie, Ingenieur, Technik, 2009) as well as the status paper "Feinstaub" of September 2010 (K.G. Schmidt, R. Zellner (Ed.), 2010 (download available from the ProcessNet homepage www.dechema.de/processnet or www.processnet.org).

With the intension of an improved protection of humans and the environment, the committee may contribute to push selected topics of fine particles forth by:

The support of an interdisciplinary scientific process with the objective of an improved understanding of the system, e.g. by initiating interdisciplinary and coordinated research projects.

The increase of the dialogue between science, industry and administrative bodies, e.g. for the development of avoidance strategies on already existing health and environmental hazards.

# 2. Source-Specific Aspects of Fine Particles

# 2.1 Overview of the Relevant Sources of Fine Particles

Fine particles are only partially directly emitted as primary fine particles. Approximately 30 - 50% of the urban background pollution can be assigned to the so called secondary aerosol. These are made up of gaseous precursors in the atmosphere, of which  $SO_2$ ,  $NO_x$  and  $NH_3$  as well as NMVOC are the most important. To obtain an impression of the importance of different sources for the concentration of fine particles, the emissions of different sectors are compiled in Table 1.

What is eye-catching in Table 1 is that the amount of the emitted precursors (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and NMVOC) quantitatively exceeds by far those of the direct particle emissions in the fractions PM10 and PM2.5. However, not all of the precursors are converted into particles. Furthermore does the quantitative distinction of particle and precursor emissions tell little about the effective pollution in certain places, due to the geographically extremely diverse distribution of the sector's sources. Thus, for instance, traffic is the dominating factor in street canyons, whereas the urban background pollution may be essentially determined by emissions from agricultural activity.

Primary fine particles up to 2.5 µm diameters (PM2.5) are mainly caused by emissions from small-scale furnaces and from traffic. Coarse fine particles between 2.5 and 10 µm diameters (the difference between PM10 and PM2.5), on the other hand, are due to abrasion processes of surfaces (brake abrasion, tyre wear particles and road abrasion) in traffic, as well as industrial processes, followed by the cultivation of soil and the keeping of farm animals. The formation of  $SO_2$  still traces back to the combustion of coal in power plants and to process firing plants. For the emissions of NO<sub>x</sub>, especially Diesel engines are responsible, followed up by large power plants and process firing installations. Emissions of NH<sub>3</sub> are almost entirely caused by farm production; NMVOC-emissions trace back to the use of solvents. The short-term threshold for PM10 is frequently still exceeded in Germany. Hence, a further reduction of PM10 emissions is necessary.

In public perception, traffic is usually mentioned as the dominating or even the only cause of fine particle pollution. Therefore, reduction measures in the area of traffic (see environmental zones) are discussed above all. In doing so it is often overlooked that agricultural activities – and more lately the domestic heatings – take a much larger part in the fine particle pollution than commonly assumed.

# Table 1: Emissions of fine particles and precursors in Germany 2010 in Gg/a (Source: PAREST (www.umweltbundesamt.de/publikationen/strategien-zur-verminderung-feinstaubbelastung))

Sector	Unit <sup>1)</sup>	PM10	PM2.5	<b>SO</b> <sub>2</sub>	NO <sub>x</sub>	NH <sub>3</sub>	NMVOC
Energy conversion	Gg/a	10	9	237	258	3	8
Other firings (households, industry, crafts, services)	Gg/a	29	27	48	105	3	85
Industrial firing systems	Gg/a	18	8	62	69	1	4
Production processes	Gg/a	44	13	101	73	10	80
Production and distribution of fossil energy sources	Gg/a	4	1	6	-	_	17
Road traffic	Gg/a	83	28	1	457	9	109
Other mobile sources	Gg/a	16	16	1	201	1	71
Agriculture	Gg/a	27	6	_	83	576	255
Other sources <sup>2)</sup>	Gg/a	9	9	_	_	2	685
Total	Gg/a	240	116	456	1246	605	1314

 $^{1)} 1 \text{ Gg} = 1 \text{ kt}$ ;  $^{2)}$  other sources of fine particles include cigarette smoke and fireworks and solvents (in case of NMVOC)

### 2.2 Agriculture

As already mentioned above, some 30-50 % of the urban background particulate pollution can be attributed to secondary aerosols – which are those that are made up of gaseous precursors. The development of the secondary inorganic aerosols such as ammonium nitrate ( $NH_4NO_3$ ) and ammonium sulfate ( $(NH_4)_2SO_4$ ) is, for instance, regulated by the concentration of  $NH_3$ , whose emission is caused to an extent of roughly 95% by agricultural activities (farming of animals, fertilization). According to latest estimates, the emissions of  $NH_3$  from agriculture in Germany will further slightly increase until at least 2020. A recently detected source of  $NH_3$  is from the catalysts of vehicles (see 2.4).

Due to biochemically induced nitrification processes in animal farming and in plant growth, agricultural activities are also a source of NO. The decomposition of undigested proteins in the dung of farm animals as well as the biogenic emissions from crops are an important source of NMVOC. It is estimated that the relative percentage of NO and NMVOC emissions will further increase in the future.

Agricultural activities like cultivation of soil and farming of animals are also sources for primary fine particles. In 2005, their share in PM10 and PM2.5 emissions in Germany was approximately 10 and 4 %, respectively. This share is expected to further increase in the future, if biomass is increasingly used for energy supply as part of the German turn-around in energy policy.

Moreover, due to the implementation of the EU animal protection directive, it may be expected that the emissions from animal farming will increase significantly in the future, because the intended floor husbandry and the keeping in larger cages causes more fine particles than the keeping in small narrow cages, insofar as additional reduction measures will not be taken. In this context, the emission of multi-resistant germs is also of some concern (see chapter 2.6).

### Scientific Challenges and Call for Action

Agricultural sources are responsible for the largest contribution to the background concentrations of fine particles and thus probably make also a considerable contribution to the health damages associated with such particles. However, the available emission factors are fraught with uncertainty both for primary fine particles as well as for ammonia and NMVOC. It should be added that even the emission processes themselves are often not yet completely understood. What also matters for the estimation of the effect of reduction measures of emissions is the temporal and especially seasonal variability of the emissions, which is only fragmentarily known.

Reduction potentials of fine particles and their precursors in agriculture do exist sufficiently. As has been shown in the research project PAREST (www.PAREST.de), some of the measures in agriculture even belong into the category of the so-called efficient measures; i.e. those for which the greatest reduction of concentrations and health damages can be expected per invested Euro.

The correlation between the reduction of fine particle precursors like  $NH_3$  and the reduction of the fine particle concentration itself, however, is complex and not linear. Thus, for instance, a reduction of the  $NH_3$  emissions by 10 % from today's levels causes less reduction of the fine particle concentration than an increase of the  $NH_3$  reduction from 10 % to 20%.

The methods for the evaluation of emissions from agriculture and their spatial and temporal distribution should be urgently improved. Once the emissions are better known, the opportunities, potentials and costs for a reduction of the emissions of fine particle precursors and of fine particles should be analysed. Moreover, the obstacles for the realization of measures should be identified.

As mentioned above, an increase in the direct emissions of fine particles from agriculture is expected due to stronger protection measures for farm animals in the EU. If and in how far the reduction of  $NH_3$  will lead to a reduction of the fine particle concentration depends on the chemical and meteorological processes in the atmosphere. Therefore, time and place of the realization of a reduction measure are crucial for its effectiveness. This complex correlation has to be further investigated until effective reduction measures can be identified and taken.

## 2.3 Firing systems

As shown in Table 1, larger shares of the precursors of fine particles are caused by the emissions of  $NO_x$  and  $SO_2$  from larger firing systems. In contrast, smaller firing installations contribute directly to the emissions of primary fine particles.

The measures for reducing NO<sub>x</sub>- und SO<sub>2</sub>- emissions from large combustion plants are long known and have been realized successfully in Germany and other countries. But this does not hold true for emissions of primary fine particles, e.g. from domestic heating installations. A more detailed analysis shows that some 90 % of the fine particle emissions arise from the burning of firewood and wood waste. For this reason, especially the emission reduction measures for wood combustion should be better analysed and implemented. In addition, as opposed to large combustion plants, the emissions from small combustion devices are normally emitted near the ground. For low exchange weather conditions therefore they are responsible for a relatively high share of the near-ground air pollution including the breathing zone. Studies in Switzerland indicate that temporarily up to 80% of the fine particle emissions are due to the combustion of wood in heat generating installations.

### Scientific Challenges and Call for Action

In order to reduce  $CO_2$  emissions, renewable energies should be introduced in all areas where energy is used; also for the heating of rooms. Here one has to differentiate between decentralized heat-generating installations (biomass heating plants) and domestic stoves within the house.

The reason for particle emissions with these installations is on the one hand in the amount of minerals in the biomass, which can be emitted by thermal or mechanical processes. This happens with automatically as well as manually loaded firing systems. By incomplete combustion, on the other hand, emissions of soot particles and low volatile hydrocarbons appear. This problem exists mainly with manual firings with irregular supply of combustibles. The emitted particles, inorganic as well as the organic, are very small (approx. 90 % <1µm or with a maximum in number density distribution at approx. 60 nm) due to the phase transitions such as condensation and resublimation. Potential reduction measures relate primarily to an improvement of combustion technology. On the other hand, particle scavengers are to be developed or are already developed, by whom the exhaust gases may be purified, even from single heating installations.

Particles from the combustion of biomass are notorious with respect to their content of a large variety of organic compounds that potentially have an adverse effect on human health. What is more, during the combustion of biomass polycyclic aromatic hydrocarbons (PCAs) are emitted that either have toxic properties themselves or aquire such properties after functionalization in atmospheric-chemical processes. The role of further organic substances such as phenols and methoxy-phenols for the formation of secondary aerosols and in relation to health aspects is presently part of intensive research.

It is foreseeable that the potential of the development of secondary organic aerosol particles (SOA, see paragraph 2.7) will turn into an important future topic, especially in relation to the combustion of biomass.

The modelling of emissions from small firing installations is currently not possible because of the high variability in combustibles and firing conditions and the corresponding lack of emission factors. Moreover, it is presently not even clear who is going to provide such factors. As a consequence precautionary measures to reduce health effects from small firing installations are currently not possible. A WHO publication on the health effects due to emissions from the combustion of firewood and wood wastes is in preparation.

### 2.4 Road transport

As can be seen from Table 1, traffic still belongs to the largest sources of environmental particles. This is true for both size fractions, PM10 and PM2.5. Not for nothing the locations of the highest fine particle pollutions and of the highest threshold exceedance frequencies for PM10 are located in traffic-related hotspots. Also the PM2.5 values show the highest values at such hotspots and even the

concentrations of ultrafine particles (UFPs) at these spots often exceed those observed for the urban background by at least an order of magnitude. Due to the spatial proximity of an assessment spot in the area around the polluted streets, mainly primary source processes by local car traffic are relevant. These include the fine particle emissions from the exhaust pipes as well as traffic related swirl up and abrasion emissions.

Soot particles that are primarily emitted by Diesel engines are of special importance concerning the health effects of traffic emissions. A broad consent exists that such particles are health-damaging. In a recent evaluation by the International Agency for Research on Cancer (IARC) of the WHO, fine particles from Diesel and Otto engines are even assigned to have cancerogenous effects. Regarding the health effect of traffic emissions, the emitted aerosol as a whole (gas and particle phase) is possibly of importance. Hence, synergistic effects of gas and particle phase might be considered at the same time, where  $NO_x$  and semi-volatile hydrocarbons (including PCAs and nitroaromates) do play a role.

#### Scientific Challenges and Call for Action

The evaluation of measures for the reduction of immissions of fine particles from road transport requires a complete picture of the sources and source strenghts, including information on important chemical contents (e.g. EC, organic markers like source-specific hopanes) or alternative metrics (e.g. ultra-fine particles). It also demands the ability to model the formation and transport process of such particles.

A need for research concerning the traffic emissions exists in the further clarification of the emission strength of Diesel powered vehicles in dependence on the driving dynamics and the efficiency of the after-treatment of exhaust gases for different operation conditions. Currently, also the type and amount of particles that are emitted even from modern Otto engines is vague.

Recent observations from field measurements in street canyons have demonstrated evidence for the emission of  $NH_3$  from vehicles with catalysts. As discussed above, ammonia is responsible for the formation of secondary aer-

osols such as ammonium nitrate and ammonium sulfate. Whether this source is competitive in rural areas with the primary source of ammonia from agriculture remains to be investigated.

An important field of further clarification concerns the so called source metric of exhaust emissions. In the presentday guidelines of environmental protection the focus of evaluation is exclusively laid upon the mass of particles, differentiated by size fractions such as PM10 or PM2.5, and in the number of particles. But it is left unclear which components of such emissions (soot, elementary carbon (EC) or organic carbon (OC)) correlate best with health effects and if the number of particles is more important than the mass of particles. The important question how these emissions will change with increasing substitution of fossil Diesel and gasoline fuels by fuels from renewable sources such as biodiesel or ethanol should be added. In addition to that, the high ratio of short-distance travel in the cities also requires an improved analysis of the effects of cold start conditions (i.e. with restricted effective retention systems) on particle emissions.

An important point regarding the emissions from road transport with combustion engines are also the metal emissions from engine wear and tear as well as from additives in lubrication oils. Especially for older Otto engines, metal oxide particles can make up to 30% of the total particulate emission with size ranges between 15-25 nm. Moreover, zinc oxide emissions from the metal additive content of lubrication oils are also considered to cause adverse health effects, which need to be restricted.

After all, a significant lack of knowledge exists about road dust resuspension and abrasion from surfaces (including brakes and tyres) as well as their dependence on the driving dynamics and/or the frequency of street cleaning. Particles from such processes contain silicates and carbonates, but also toxic metals such as zinc, copper and antimony. These source types are active even with the cleanest vehicles (and in an extreme case also with electric vehicles), so that their consideration and quantification is by no means negligible. It should be noted that measures such as more frequent or more intense street cleaning has not lead to a measurable or significant improvement. Possible reduction measures of emissions from traffic are either of technological nature (type of drive, emission control, road surfaces, etc.) or they imply a change in mobility (modal split, logistics, management, speed limits, travelling distances). Measures with limitations of traffic like environmental zones represent a restriction of collective and individual needs for mobility.

# 2.5 Industrial Sources

The contribution of industrial sources to fine particle emissions amounts to approximately 20%. Taking into account the precursors for secondary particles this fraction may be even much higher. On the one hand, industrial sources contribute considerably to the large scale background pollution. This applies especially to confined sources such as chimneys with a high plume emission height. On the other hand, the local contribution of industrial sources shows a large variability with relative amounts between a few and more than 50%.

Besides the emission strength, especially the plume height of the emissions is crucial for their contribution to the fine particle pollution in ambient air. In close proximity (approximately 1 km distance) of industrial locations with considerable mostly diffuse emissions from low sources (e.g. metallurgy, opencast mines, quarries, handling of bulk materials, coal bunkers, cement manufacturing), the permitted daily mean of  $50 \ \mu g/m^3$  for PM10 might often be exceeded, partly even more than the maximum permissible 35 days per year. An important contribution to fine particles from diffuse emission sources is from building sites and building machineries, including off-road vehicles.

Since the particle emissions from industry are mostly high only on small-scales, i.e. short distance to the source, and the number of measuring stations is limited, it may be assumed that currently only part of the exceedances in both space and time in the vicinity of industrial sources is documented.

### Scientific Challenges and Call for Action

The contributions of industrial sources to the background particulate pollution and to local exceedances of limits

have to be better quantified in order to reduce the fine particle pollution on the whole by appropriate measures at the source. In doing so, the acquisition and reduction of diffuse particle emissions still causes substantial difficulties. A need for research exists especially in developing methods for the determination of source strengths of diffuse sources. This not only applies to industrial sources like e.g. opencast mines and the handling of bulk materials, but also for diffuse sources of particles in agriculture. Besides its quantification there is a need for the development of economical methods to reduce diffuse particle emissions. Furthermore, the characterization of industrial sources of ultrafine particles in respect of source strengths and composition is an important topic of future research work.

Some industrial sources are due to their special chemical contents of considerable health relevance. Thus, the need exists for clarification of the contribution of important components in industrial emissions (e.g. metal compounds, soot) to health effects. Data on the emissions of ultrafine particles by industrial sources do hardly exist yet.

## 2.6 Biological Particles

Biological aerosol particles are often referred to as primary biological aerosol particles (PBAP) in specialized literature and consist of biological materials which are emitted directly from the biosphere to the atmosphere. These include a variety of different particles types like for example complete microorganisms (bacteria, algae), reproductive units (pollen, fungal, fern and moss spores), abrasions of plants, parts of insects and biopolymers (DNA, chitin, cellulose and other polysaccharides). The diameters of PBAP range from a few nanometers (protein molecules and viruses) to hundreds of micrometers (pollen, plant spores and abrasion of plants). PBAP are ubiquitous in the global atmosphere and in some cases - especially in clean air above vegetation covered areas – have shares up to 50% and more of the total airborne particle mass. Microscopic studies show that PBAP in the air of rural and forested areas amount to 30% of the mass of particles in fine particle or in the submicron size range (<1µm) and to 70% in coarse dust or supermicron range (>1µm). Estimates of the global PBAP emissions range from approximately

60 Tg per year in fine particles to approximately 1000 Tg per year in total suspended matter. Fungal spores alone account for typical mass concentrations of circa 1  $\mu$ g m<sup>-3</sup> in the continental boundary layer and for global emissions in the dimension of approximately 50 Tg per year. The atmospheric transport of PBAP is an important pathway for the distribution of organisms within and the translocation between different eco systems.

Pathogenic PBAP can cause infectious diseases and allergies in humans, animals and plants, and play an important role in the areas of hygiene, health and agriculture.

Scientific studies also show that in the proximity to sites for biological waste and water treatment installations as well as for industrial farming the concentrations of biological particles may be enhanced. Currently, endotoxins and microorganisms in an increased concentration have been detected at a distance of 250 m in a Dutch study. However, resistances or multi-resistances of bacteria from biological aerosols against antibiotics does not seem to be of larger relevance in view of the ubiquitous occurrence of resistant microorganisms in the environment and outside of medical facilities with patients with risks of infection.

Airborne microorganisms from agriculture are subject to a rapid dilution in ambient air and are influenced in their survivability by climatic factors like relative humidity, temperature and radiation. However, it has to be considered that the results of model calculations on the dispersion of PBAP currently differ considerably from the results of measurements.

#### Scientific Challenges and Call for Action

For biological aerosols in ambient air neither dose-effect relationships are known, nor do limiting values exist. This is due to the fact that bio-aerosols may contain a large variety of different types of microorganisms in different concentrations, dependent on production facilities and process specifics, meteorological conditions as well as seasonal and local situations and so forth, and many of these are to be found in nature. In addition, health effects of bio-aerosols (irritative, allergic, toxic, infectious) are dependent on the individual predisposition and on the immune status of the exposed individual. For risk groups including immune restricted persons, allergy sufferers and persons with respiratory diseases, already the existing background concentrations might cause an additional health risk. Therefore, the total pollution and thus the immissions from existing facilities in the neighbourhood should be considered in the evaluation. But the measurements of the concentrations of bio-aerosols that are needed for this evaluation are not available in most cases. In addition, as a surrogate for the natural background pollution that is customary in a place, values taken from literature are usually used.

# 2.7 SOCs and VOCs

Semi-volatile organic compounds (SOC) as well as volatile organic compounds (VOC) can contribute to the so-called secondary organic aerosol (SOA) by atmospheric-chemical processing. A prerequisite is, however, that the processing of SOC or VOC leads to oxidation products with a very low vapour pressure. The formation process might be the direct nucleation of such products from the gaseous phase or it might also be the result of a condensation of semi-volatile or non-volatile transformation products on pre-existing surfaces. Many of the substances involved like polycyclic or functionalized hydrocarbons are toxic and pose a danger for health, on the one hand by direct inhalation, on the other hand by food, because they tend to accumulate in the environment in the marine and terrestrial food chain. Currently, the total amount of secondary organic aerosols is difficult to quantify, just as its distribution in the different PM size fractions.

Semi-volatile organic compounds (SOC) are formed by oxidation of VOC, like e.g. dicarboxylic acids and hydrocarboxylic acids, or emitted from anthropogenic (e.g. pesticides, chlorinated industrial chemicals) and biogenic sources (e.g. paraffines). The chemistry of SOC differs from the chemistry of VOC by two principal features: (1) It occurs in both phases of the atmospheric aerosol (gas and condensed phase) and thus are transported to a significant extent as part of airborne particles. (2) As far as SOC constituents are concerned, which are not readily degradable after their deposition onto surfaces of soil, water and vegetation, these will be typically multicompartment compounds. As far as substances are concerned that do not decompose rapidly after deposition in surface water, in the soil or on surfaces of vegetation, we are concerned with multi-compartment substances. Therefore, by consecutive cycles of (re-)volatilization and deposition, distinct temporal and spatial characteristics of the concentration distributions may appear. For instance, the translocation of pesticides from the tropics and subtropics into higher geographical latitudes or from the lowlands to mountain regions by "multi-hopping" or "grasshopper effect" is wellknown. Dependent on the solubility in water and lipids as well as the persistence of the SOC in terrestrial and aquatic environmental compartments, total residence times in the environment result that exceed the atmospheric residence times by orders of magnitude (up to years and decades). Many SOCs bio-accumulate along aquatic and terrestrial food chains which is relevant for ecosystems and human health.

Volatile organic compounds in ambient air (VOC) arise from anthropogenic and biogenic sources, with the chemical nature, however, being different for both sources. While saturated and aromatic hydrocarbons (solvents, fuels, e.g. benzene) are typical for the anthropogenic sources, predominantly unsaturated hydrocarbons (e.g. isoprene, terpenes, ethene) are emitted from biogenic sources The atmospheric transformations of VOCs leads – just as those of the SOC – to a formation of secondary organic aerosols (SOA) via reactive intermediates and oxidized hydro-carbons.

#### Scientific Challenges and Call for Action

Among all aerosols SOA, as a secondary aerosol from SOC and VOC, shows by far the most complex chemical composition. Up to now and with the availability of most sensitive and specific analytical methods, it has not been successful to identify the variable chemical constitution of SOA aerosols completely. Only some 20-30% of the SOA mass are currently identified on a molecular level. It can be expected that this part will grow in the future with increasing analytical efforts; but the full knowledge is not to be expected for now.

Besides the improved analytics of SOA, a considerable need for research exists also in the further clarification of

processes. This includes for the SOC: (1) the provision of relevant model aerosols and the study of heterogenous photochemical reactions of SOC in laboratory experiments, (2) the characterization of the sorption processes of SOC and their mobility (and reactivity) in particulate matter as well as (3) the determination of physical and chemical immobilization processes of SOC in multiphase systems in the soil. Furthermore, the organic particle phase of urban and continental atmospheric aerosols with regard to their semi-volatile substances, their phase and size distributions, as well as the health relevance of inhaled particles has to be better characterized. Finally, the development of models for predicting atmospheric and multi-compartment chemistry as well as the transport of SOC should definitely be advanced. In the area of VOC, an improved quantification of anthropogenic and biogenic parts as well as their seasonal variability is necessary.

Only by such measures it may be expected that spatial and temporal large-scale balances are compiled and that the total residence time and the persistence of the secondary aerosol may be better understood. As much as toxic or ecotoxic substances are concerned, also an evaluation of the risk to the environment and its management should be possible.

Finally, by the support of the dialogue between science, industry and political decision makers for the compilation and communication of the spatial and temporal use and emission patterns of SOC and VOC, inherent eco-friendly substance properties (substance design) are supported. Further, adjustment and avoidance strategies with already existing risks for the environment are developed.

# 3. Further Aspects

# 3.1 Effect-related metrics of fine particles

As clearly demonstrated by epidemiological observations and scientific studies - beginning with the London Smog in 1952 - environmental particles lead to adverse health effects. However, early research papers concerning the exposition of particles and health effects were always based upon the concentration of the total suspended particle mass in the air as an effective metric. Under the dictum of effect-related dust measurement in industrial hygiene, a focus on the alveolar fraction of mineral particles turned out to be more target-aimed, e.g. for the evaluation of the silicosis threat by particles. Advances in particle measurement technology around the 1980s led to the fact that also in environmental hygiene better associations of health effects with smaller particle fractions < 2.5 µm diameters could be demonstrated in the following two decades. A further aspect is the variety of the observed health effects which can hardly be related to a consistent and common mode of biological action. Examples include the acute and chronic effects on the respiratory tract or systemic effects like cardiovascular diseases or coagulation disorders of the blood.

According to conclusions from recent toxicological studies in relation to the specific relevance of the different components of the environmental aerosols for health, the combustion aerosols from traffic, power plants and industry - commonly summarized under the general term "black carbon" (BC) - are to be assessed extremely critically. However, as has been concluded in recent reviews on BC from EPA (http://www.epa.gov/ncea/isa/pm.htm) and WHO (http://www.euro.who.int/en/health-topics/ environment-and-health/air-quality/publications/2012/ health-effects-of-black-carbon), the soot kernel may be regarded as a universal carrier for a large and complex number of chemicals with different toxicities. According to this picture it is not the soot kernel itself but rather its surface concentrations of toxic chemicals which is regarded as a cause for adverse effects: ".....suggested that BC may not be a major directly toxic component of fine PM, but it may operate as a universal carrier of a wide variety of chemicals of varying toxicity to the lungs, the body's major defence cells and possibly the systemic blood circulation."

Moreover, in a recent clinical epidemiological study, an extremely short exposure of 1 hour could be identified as a relevant exposition of PM2.5 for the triggering of an acute coronary syndrome. Particles in the accumulation mode size or ultra-fine particles, however, did not show a statistically relevant correlation with the clinical effects.

### Scientific Challenges and Call for Action

In the beginning of the 1990s of the last century, it became clear that not only the concentration of the particle masses and their chemical composition influence the health effects of particles, but other particle properties as well. These include the number densities of particles, especially for very small particles with dimensions of < 100 nm, the surface concentrations of particles as well as further particle properties such as the morphology, the reactivity of particles – especially in biological media – and the biopersistence, i.e. how long a particle can exist in a biological environment before it dissolves or is excreted. Here, the size of particles is always an important factor, because it influences the above mentioned properties directly and indirectly.

An important point for future scientific research is the identification of critical components in complex environmental particles. Not only the mass of aerosols is responsible for their effect, but they are those components from which special health effects emanate or are expected. These components include soot, some metals and metal oxides, but also organic compounds. Also, concerning the behaviour of particles in a biological environment, like the interaction with proteins, other bio-molecules and components of cells, the dynamics of aggregation, deaggregation and dissolution, as well as the transport in the biological system, require further clarification. For instance, the intracellular displacement volume of aggregated nanoparticles with otherwise little biological effects (e.g. AIOOH) turned surprisingly out to be the effect-determining metric.

Toxicological studies of aggregates of nanoparticles from combustion processes have shown acute health effects (within hours), the cause of which is probably indicative for the presence of fast dissolving chemical components. Such types of effects are also apparent for the dynamics of dissolution in the context of the lateral diffusion in the lung (e.g. bio-availability of PCAs). One of the tasks of the following years will be to unravel the correlations between different properties of aerosol particles with the endpoints of health effects, both epidemiological and toxicological, and to develop and validate the necessary (routine) measuring technique. In addition, the scientific basis for the assessment of particle exposition is to be improved and a new metric has to be introduced in order to achieve an improved prevention of particle-induced health effects.

# 3.2 Climate, weather and transport of fine particles

Apart from their importance for air-guality, fine particles are also important for the radiation balance in the atmosphere and thus for the climate on Earth. A complex and only partially resolved problem in climate research is the role of aerosols for the so called radiative forcing. In general, aerosols have scattering properties, principally reduce the intensity of solar radiation by backscattering and thus have a cooling effect on the globe. Intensively absorbing aerosols like soot particles and some mineral aerosols (i.e. iron oxides) are an exception. These also absorb the solar radiation and thus have a warming effect on the layers of air, in which they are contained. Equally relevant as for the radiative forcing are aerosols with regard to cloud formation and consequently precipitation. Thus, they are important factors of influence for the weather. Therefore, it is necessary to improve the treatment of fine particles (or the atmospheric aerosole as a whole) not only in chemistry transport and climate models, but also in weather and air quality forcast models.

Fine particles or aerosols in the atmosphere are transported similar to gases, except that they can also be subject of sedimentation. Thus, the concentration of fine particles is always the result of the combined effect of emission, transport and dynamics. Since the residence time of fine particles in the lower atmosphere is between several days and one week, fine particles can principally be transported over long distances (up to some 1000 km).

For the determination of the atmospheric transport of fine particles from different sources, chemistry transport models are applied. Their accuracy strongly depends on the availability of reliable emission data. This is stressing the need of reliable measurements of emission data and derivation of aerosol source characteristics. They are important not only for anthropogenic, but also for natural emissions, e.g. in the case of volcanic eruptions or dust storms. Such measurements can be carried out in-situ on the ground and by air-born platforms or by remote methods employing airplane and satellite experiments (as in the case of volcanic ash clouds, forest fire aerosols and industrial exhausts).

Tethered balloons turned out to be very suitable instruments for the study of the near-ground air layers and the distribution of fine particles. Their special strength is the clarification of the distribution of fine particles in the inversion layers in urban atmospheres as well as the vertical and horizontal distribution of fine particles from wood log heating in residential areas. Usually, tethered balloons are equipped with measurement techniques for the determination of vertical profiles of meteorological parameters (most important are measurements of wind and temperature) and of concentrations of fine particles and their size distribution. Furthermore, samples of fine particles can be collected in different heights, in order to analyse their structure and chemical composition in the laboratory.

### Scientific Challenges and Call for Action

From the perspective of application, more intensive and improved studies of special aerosol events like dust storms, sudden turbidities or volcanic dust emissions (air traffic), are necessary. Pending questions also exist in the field of modelling with regard to the parameterisation of the transport of fine particles, primarily in the planetary boundary layer. A further scientific issue is the determination of the influence of meteorology on the concentration of fine particles – relating, for instance, to elemental carbon (EC) – and *vice versa*. An important issue is the improvement of chemical mechanisms for the formation of biogenic aerosols.

Environmental policy and management are benefitting from the fact that aerosol-chemistry-transport models can well be applied to the source-receptor analysis of atmospheric pollutants, among which fine particles are highly relevant regarding human health.

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