

**Fine Particles –
Measurements and Exposure
in View of Health Effects**

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1 INTRODUCTION

1.1 Brief History of Particles, Exposure and Health Effects

Airborne particles, also called aerosols, are known for centuries and described already in the early literature. Heavy air pollution caused by incomplete combustion of coal in London in 1273 led to the first abatement measure, the ban of coal combustion. One of the first monographs on air pollution was written by John Evelyn in 1661 with the title “*Fumifugium (or The Inconveniencie of the Aer and Smoak of London Dissipated)*”. He clearly links exposure to health effects and advises King Charles II on how to improve air quality.

„...Inhabitants breathe nothing but an impure and thick Mist accompanied with a fuliginous and filthy vapour, which renders them obnoxious to a thousand inconveniences, corrupting the Lungs, and disordering the entire habits of their Bodies, so that Catharrs, Phthisicks, Coughs and Consumptions rage more in this one City than in the whole Earth besides.“¹

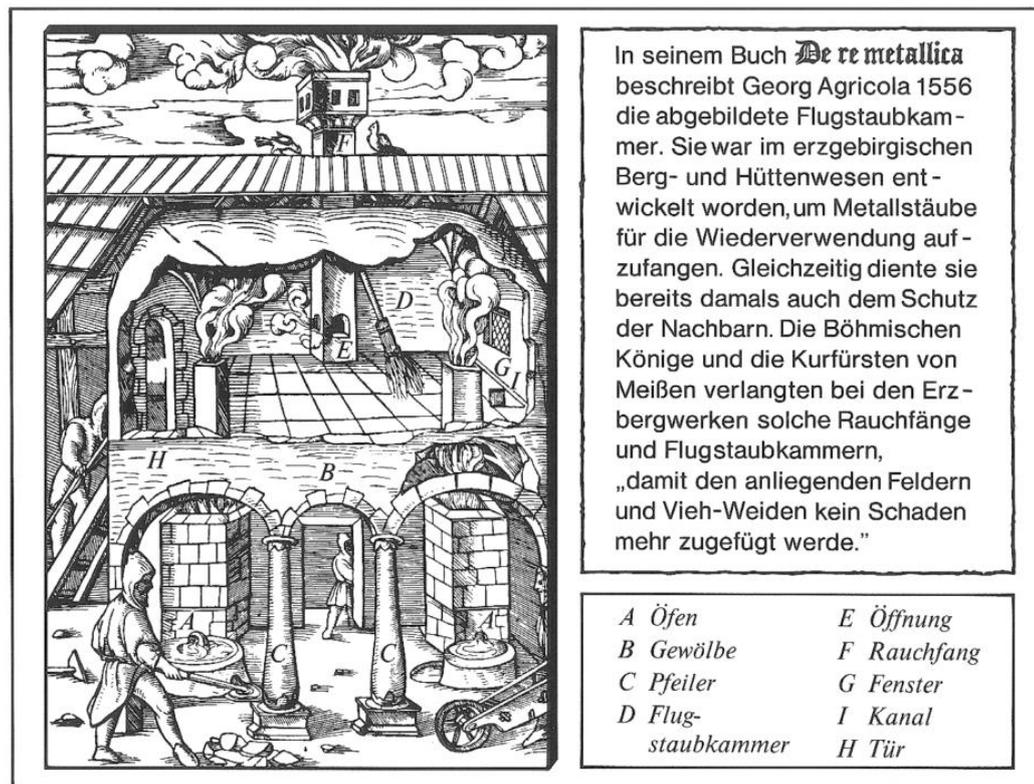


Figure 1: Dust settling chamber for emission cleaning (Fonds der Chemischen Industrie, 1995)

One of the first publications on the health effects of workplace exposure to aerosols was written by Gerg Agricola 1556 in his book “*De Re Metallica*”. He describes the high mortality rate of workers employed in mining and metallurgy in the Erzgebirge. In the same publication Agricola also sketches a dust settling chamber (Figure 1) which was

¹ <http://www.gyford.com/archive/2009/04/28/www.geocities.com/Paris/LeftBank/1914/fumifug.html>
Fumifugium, J. Evelyn, ISBN 0904617068, The Rota, p. 5, 1976.

developed to collect the metal dust for reuse. The bohemian kings and earls made these dust settling chambers mandatory to “avoid further damage of the surrounding agricultural areas and pastures”. Similarly Paracelsus observed in 1530 (Paracelsus, 1567) lung diseases if workers were exposed to high dust levels in mining. The direct linkage of dust from ores and stones with human health effects, especially pneumocinosis and silicosis, was published about 400 years later in 1926 (Collis, 1926).

Another seminal work was published by Sir Percivall Pott² in 1775. He made one of the first epidemiological observations on the effect of particle exposure from incomplete combustion on humans. He noticed the high rate of scrotum cancer of chimneys sweeper. This occupational cancer is most likely caused by the soot and the carcinogenic substances on the soot like benzo(a)pyrene.

Detailed research and investigations in causal chains of air pollution, here more specifically airborne particles, on human health demanded the development of new measurement techniques. Aitken (1888) e.g. developed the first particle number counting apparatus. Also quite a few further measurement techniques used today were developed at the beginning of the 20th century. The “impinger” was developed by e.g. Michaelis (1890) in the Robert Koch laboratory in Germany to allow efficiency testing of filters. Filtration of air to determine airborne particles became common in the 1920’s and Trostel and Egge (1923) for example used this method for the collection of explosive dust. Commercial production of membrane filters began 1927 (Spurny, 1999b), while paper filters were described for the first time by Silverman and Ege in 1943. These and other developments in particle measurement technology enabled health studies and made the adverse effects more known and generally accepted in the 1950’s (Sinclair, 1959, Davies, 1954, Drinker and Hatch, 1954). This and further research and discussions, linking exposure with health effects, led to the regulation of emission and air quality (Figure 2). Table 1 gives a brief overview on the history of air pollution.

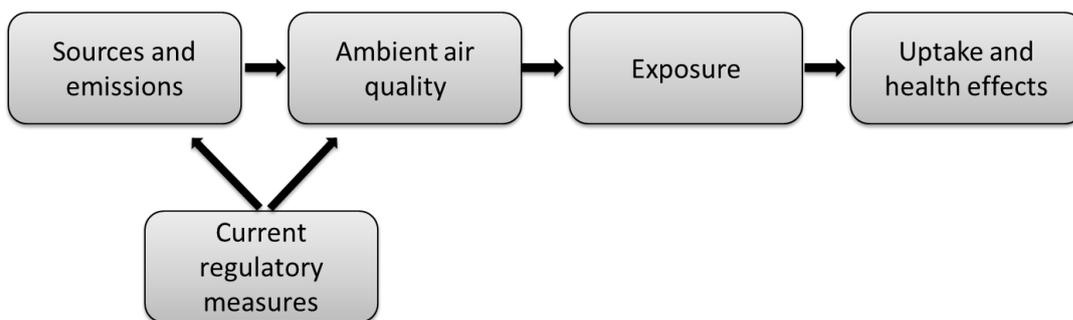


Figure 2: Causal chain of air pollution and health effects leading to the introduction of regulatory measures and legislation

² John R. Brown, John L. Thornton, Percivall Pott (1714-1788) and Chimney Sweepers' Cancer of the Scrotum, *British Journal Industrial Medicine*, 1957 January; 14(1): 68–70.

Table 1: Brief history of events and legal actions with regard to air quality in the UK and EU

<p>1273 UK: Use of coal prohibited in London as being "prejudicial to health".</p> <p>1863 UK- Alkali Works Regulation Act: Required that 95% of the offensive emissions should be arrested.</p> <p>1875 UK- The Public Health Act: Smoke abatement section</p> <p>1946 UK: First smokeless zone and prior approval legislation.</p> <p>1956 UK- Clean Air Act: Introduced Smoke control areas. Prohibited emission of dark smoke from chimneys, with some exceptions.</p> <p>1972 EC Directive 72/306/EEC: Measures against emissions from diesel engines. Limited black smoke emissions from heavy duty vehicles.</p> <p>1980 EC Directive 80/779/EEC: Air quality limit values and guide values for sulphur dioxide and suspended particles.</p> <p>1982 EC Directive 82/884/EEC: Limit value for <i>lead</i> in the air.</p> <p>1989 EC Directive 89/427/EEC: Limit /guide values for sulphur dioxide and suspended particulates. Harmonised measurement methods.</p> <p>1996 EC Directive 96/62/EC: AQ framework directive</p> <p>1999 EU 1st Daughter Directive: AQ limit for SO₂, NO₂, NO_x, PM₁₀ and Lead.</p> <p>2003 EU 4th Daughter Directive: Targets value for PAH, measurement of heavy metals</p>

1.1.1 Ambient Air Quality

The German „Bundestag“ ratified the Bundesimmissionsschutzgesetz (BimSchG, 2000) in 1974 clearly stating the importance of a clean and healthy environment:

§ 1 Zweck des Gesetzes

Zweck dieses Gesetzes ist es, Menschen, Tiere und Pflanzen, den Boden, das Wasser, die Atmosphäre sowie Kultur- und sonstige Sachgüter vor schädlichen Umwelteinwirkungen und, soweit es sich um genehmigungsbedürftige Anlagen handelt, auch vor Gefahren, erheblichen Nachteilen und erheblichen Belästigungen, die auf andere Weise herbeigeführt werden, zu schützen und dem Entstehen schädlicher Umwelteinwirkungen vorzubeugen.

Translation:

§ 1 Aim of the law

The aim of this law is the protection of humans, animals and plants, the soil, water, atmosphere as well as cultural and other material goods against environmental impacts and, as far as it relates to plants requiring permission, also against danger, significant disadvantages and annoyance which occur in a different way, and to avoid adverse effects onto the environment.

Based on this law and other regulations, the mass concentration of total suspended particulate matter (TSP) was used as the particulate related air quality metric for emissions and ambient air quality. TSP refers mainly to the airborne particle size fraction

of $< 30 \mu\text{m}$ in aerodynamic diameter. This was changed with the introduction of the European air quality framework and their daughter directives after 1996. More specifically, with the introduction of the Council Directive 1999/30/EC relating to limit values for sulphur dioxide, nitrogen dioxide and oxides of nitrogen, particulate matter and lead in ambient air (1999/30/EG) PM_{2.5} and PM₁₀ were introduced as new air quality metrics. PM₁₀ here denotes particle penetrating an inlet with a 50% cut-off efficiency for particles with an aerodynamic diameter of $10 \mu\text{m}$. The 50% cut-off for a PM_{2.5} inlet is hence at $2.5 \mu\text{m}$ aerodynamic diameter. Figure 3 shows an idealised relative mass size distribution and the penetration efficiencies for the corresponding PM_{2.5} and PM₁₀ mass size fraction.

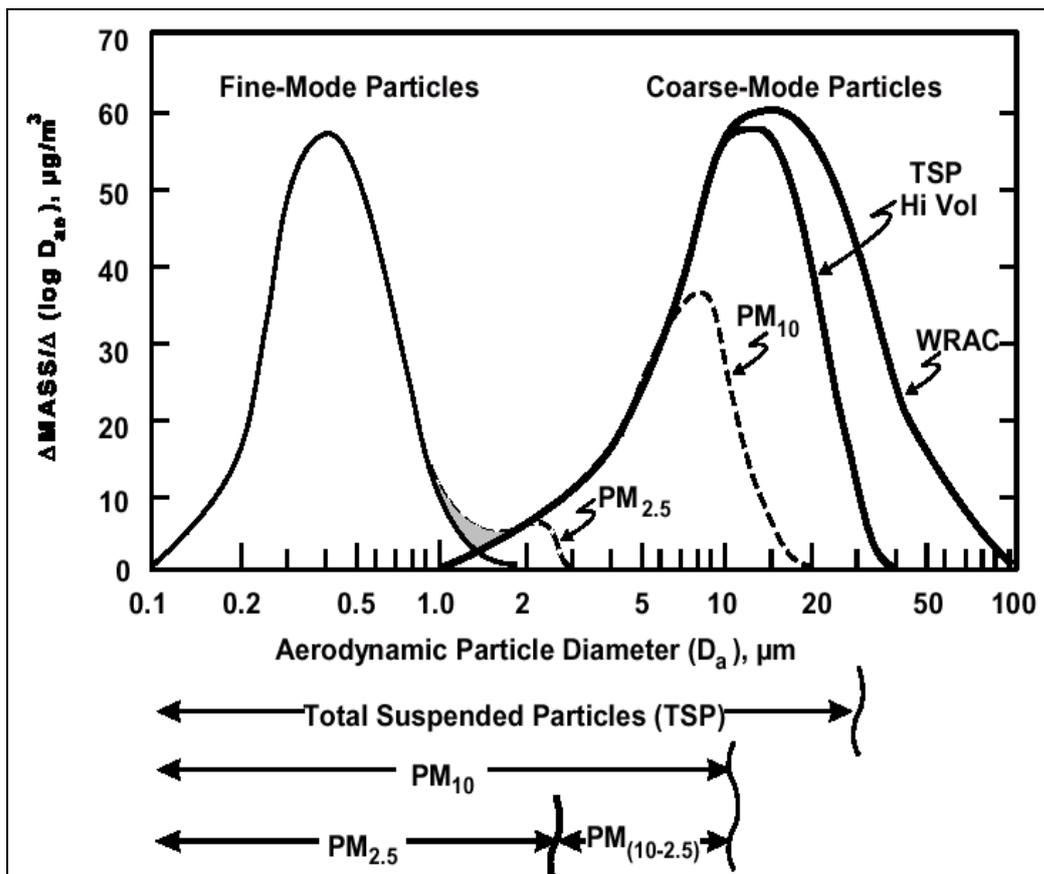


Figure 3: Relative ambient mass size distribution and the PM₁₀ and PM_{2.5} size fractions (Wilson and Suh, 1997)

Figure 4 shows the same particle size distribution as number, surface area and volume (mass) size distribution. It shows how the weighting of the particle size distribution is changed if not mass but surface area or particle number concentration is used for description. Moreover it indicates three main particle modes: nucleation, accumulation and coarse mode. The nucleation mode denotes mainly particles $< \text{ca. } 80 \text{ nm}$ in diameter, is mainly determined by high particle number but low mass concentrations, and is formed by new particles e.g. by homogeneous condensation or particle nucleation. These particles tend to agglomerate with the particles of the same size class (intramodal agglomeration) or the next particle size class, the accumulation mode (ca. 80 nm to $1,000$

nm, intermodal agglomeration). Particles in the accumulation mode grow by either agglomeration or condensation of (semi)volatile substances onto the particle surface. The upper size limit of the accumulation mode is determined by the sedimentation rate, which increases significantly around 1 μm leading to relatively high deposition rates. The coarse mode is mainly of local origin and is formed by the emission of primary particles by e.g. wind erosion. It has to be noted that the size ranges given for the modes are indicative and vary in space and time.

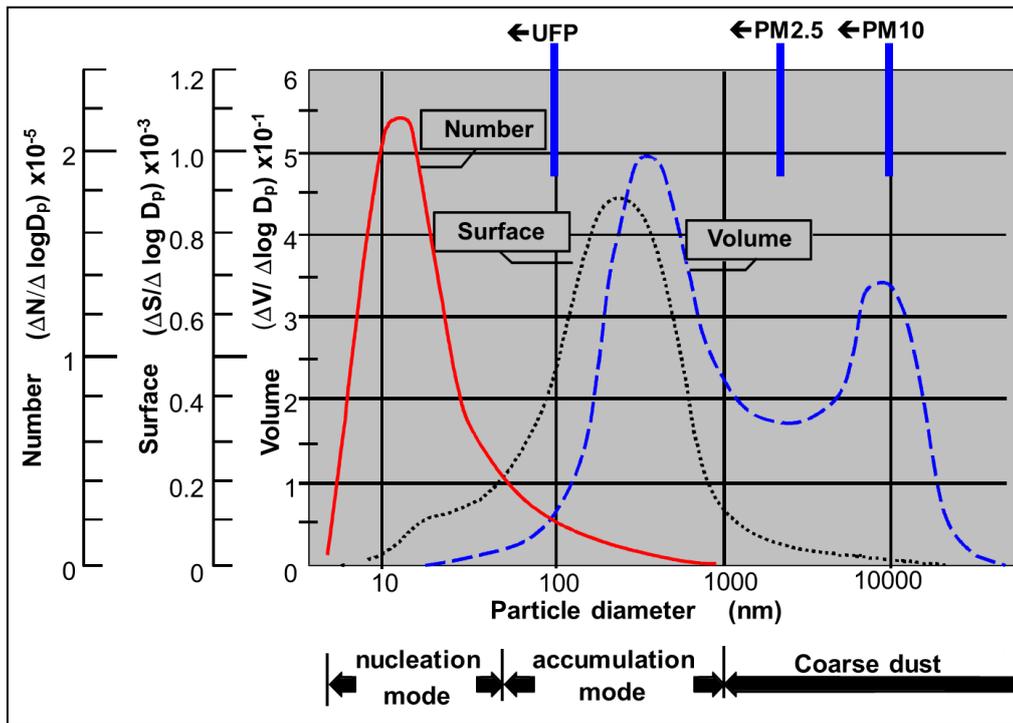


Figure 4: Idealised ambient particle size distribution, modes and weighting to number, surface and volume concentration (based on Whitby, 1978, John, 1993)

The new ambient air quality standards of PM2.5 and PM10 were introduced into legislation at the end of the last century (Directive 96/62/EC).

Table 2: Limit and target values of ambient air quality with regard to airborne particles

<i>Pollutant</i>	<i>Concentration</i>	<i>Avg. period</i>	<i>Legal nature</i>	<i>Permitted exceedences per year</i>
Fine particles (PM2.5)	25 µg/m ³	1 year	Target value entered into force 1.1.2010 Limit value enters into force 1.1.2015	n/a
PM10	50 µg/m ³	24 hours	Limit value entered into force 1.1.2005	35
	40 µg/m ³	1 year	Limit value entered into force 1.1.2005	n/a
Lead (Pb)	0.5 µg/m ³	1 year	Limit value entered into force 1.1.2005	n/a
Arsenic (As)	6 ng/m ³	1 year	Target value enters into force 31.12.2012	n/a
Cadmium (Cd)	5 ng/m ³	1 year	Target value enters into force 31.12.2012	n/a
Nickel (Ni)	20 ng/m ³	1 year	Target value enters into force 31.12.2012	n/a
Polycyclic Aromatic Hydrocarbons	1 ng/m ³ (as Benzo(a)pyrene)	1 year	Target value enters into force 31.12.2012	n/a

The last revision of the European air quality directive was in 2008 when limit values as well as average exposure values were set (Directive 2008/50/EC) (Table 2, Table 3). Revisions of the European Air Quality Directive are foreseen every 5 years also taking possible new and alternative exposure metrics such as particle number concentrations, particle reactivity and particle surface area measurements into account.

Table 3: Average exposure index (AEI)

<i>Title</i>	<i>Concentration</i>	<i>Avg. period</i>	<i>Legal nature</i>	<i>Permitted exceedences per year</i>
PM2.5 Exposure concentration obligation	20 µg/m ³ (AEI)	Based on 3 year average	Legally binding in 2015 (years 2013, 2014, 2015)	n/a
PM2.5 Exposure reduction target	Percentage reduction* + all measures to reach 18 µg/m ³ (AEI)	Based on 3 year average	Reduction to be attained where possible in 2020, determined on the basis of the value of exposure indicator in 2010	n/a

* Depending on the value of AEI in 2010, a percentage reduction requirement (0, 10, 15 or 20%) is set in the Directive. If AEI in 2010 is assessed to be over 22 µg/m³, all appropriate measures need to be taken to achieve 18 µg/m³ by 2020.

1.1.2 Indoor Air and Workplaces

Several environmental compartments can be differentiated beside ambient air to assess particle exposure of humans. One of the environments humans spend most of their time (~90%) indoors. Therefore indoor air quality, whether related to private, public or work environment, has to be carefully considered investigating possible adverse effects. Nevertheless clear differentiations have to be made.

Limit values and regulations can hardly be applied for private indoor environments since those cannot be monitored or observed on a regular and comparable basis. Therefore no specific European wide legislation exists for indoor air quality. Nevertheless, rulings and limit values for gas and particle release from consumer goods such as e.g. paints, carpets and furniture exist to minimize private indoor exposure.

Regulations of air quality in public buildings have been often discussed and especially possible particle concentrations at railway stations, subways and schools are currently seen as possible significant points of exposure (Kim et al., 2011; Kwon et al., 2010; Fromme et al., 2007; Johansson and Johansson, 2003). The focus of discussion to these specific indoor environments is due to the vulnerability of the people exposed, here school kids, or to the high number of people exposed to relatively high particle concentrations. Nevertheless no public ruling has been made. One of the reasons is the difficulty in standardising sampling location and monitoring possibilities. These types of indoor measurements are made on an irregular basis at varying locations mainly to derive information on specific microenvironments for exposure estimates based on personal exposure patterns.

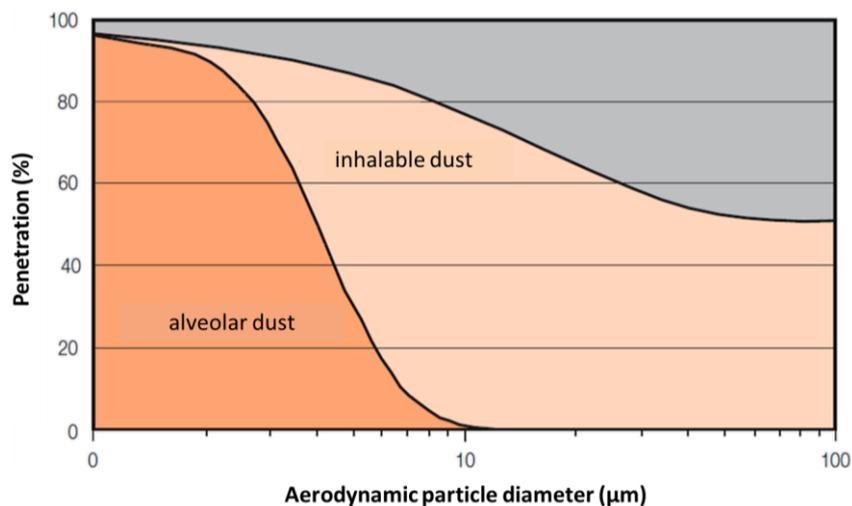


Figure 5: Penetration efficiency of airborne particles according to EN 481 (1993)

The third indoor environments are workplaces. Concentration of air pollutants at workplaces is normally regulated for industrial activities where higher exposure concentrations can be expected, e.g. in chemical industry, steel production, welding workplaces. Exposure limits for workplace environments for biologically inert dust is according to “MAK-Werte” (maximum exposure values), e.g. given in Suva (2011), 3 mg/m³ for the alveolar particle size fraction and 10 mg/m³ for the inhalable size fraction. These exposure values denote an average concentration over a whole work shift

of eight hours. MAK-values are lower if carcinogenic or another relevance is indicated in toxicological tests or epidemiological studies.

1.2 Relevance of fine dust

1.2.1 Health Effects

It can be noted that all limit and target values given in the previous section are based on particle mass concentration or the mass concentration of a chemical compound bound to the particle phase. Basis and the driving factor for the limit values for airborne particulate matter were and will be epidemiological studies. If no epidemiological studies can be performed, as for new materials and compounds toxicological studies are used to identify a possible hazard. Both, epidemiological and toxicological studies for setting limit values must evaluate the dose – effect ratio. Figure 6 shows the simplified causal chain of airborne particles possibly leading to a health effect. Starting from the exposure concentration only a fraction of the particles may cross the barrier into a human body. The main barriers are the skin, the gastro intestinal tract or the lung. Only those particles crossing the barrier lead to an internal dose possibly causing a health effect. On the other hand, not all particles taken up by a human body have the same hazard potential. Hence only if hazardous material is taken up, a possible adverse health effect may occur.

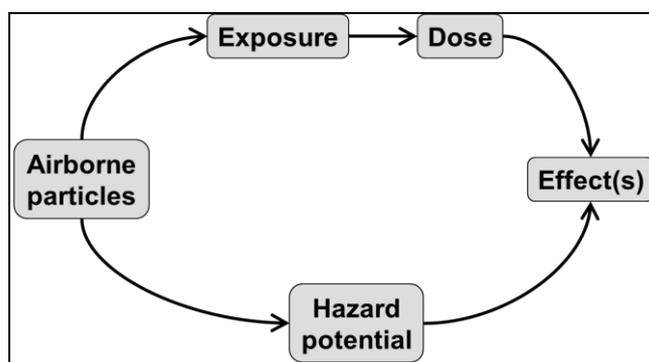


Figure 6: Airborne particles and health effects

Toxicological studies may also have other foci such as (a) biokinetic studies, meaning how and where are particles transported in a body after uptake or (b) what biochemical reactions are triggered by a particle. The latter studies focus more on the mechanisms of reactions caused by particles.

Toxicological studies may be conducted by controlled inhalation exposure or intratracheal instillation of humans and animals (in-vivo), or in-vitro by studies with human or animal cell cultures. No single effect chain or single particle property can currently be linked to all observed adverse health effects. It seems more likely that different particle properties may be linked to different health outcomes. More specifically, it may be that particle number concentrations are more closely linked to cardio-vascular effects while particle mass concentrations may have a closer link to lung functions. Particle properties and aerosol characteristics currently discussed are e.g.

particle number concentrations of ultrafine particles ($< 0.1 \mu\text{m}$, Oberdörster et al., 1995; Peters et al., 1997), pH-value (acid aerosols, Morrow et al., 1994; König et al., 1989), bioaerosols (Matthias-Maser and Jaenicke, 1994; Targonski et al., 1995; O'Hollaren et al., 1991), metals and metal oxides (Müller and Seger, 1985; Kushner et al., 1997; Lay et al., 1998), soot and PAHs' (Gallagher et al., 1994; Heinrich et al., 1995), particle surface area concentrations (Oberdörster et al., 1992; Li et al., 1996) as well as particle reactivity such as its potential to form ROS (Reactive Oxygen Species) (Shi et al., 2003; Lewis et al., 2003; Donaldson et al., 2003).

So far all regulated and discussed particle metrics used for air quality and exposure assessment can only be viewed as indicators to health effects due to the still missing singular linkage between mode of action of particles and health effects. This is also true despite the obvious linkage between particle concentrations and health effects.

Epidemiological studies investigate the correlation of different health effects / health end points (e.g. mortality, morbidity, hospital admission) with varying concentrations of trace gases (SO_2 , NO_x , etc.), airborne particulate matter (e.g. number, surface or mass concentrations) and co-factors, e.g. ambient temperature. Nearly all published epidemiological studies, using multiparameter regression analysis show significant correlations between ambient particle concentration, e.g. $\text{PM}_{2.5}$ and PM_{10} mass as well as ultrafine particle number concentrations and adverse health effects. Correlations exist for short-term effects (lag time of hours to days) as well as for long-term effects (years and decades).

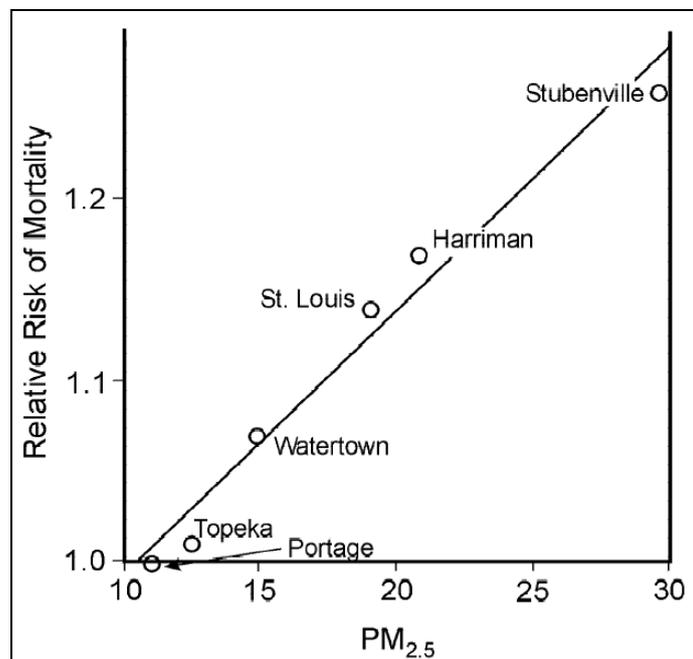


Figure 7: Correlation of the relative risk of mortality with average $\text{PM}_{2.5}$ mass concentration in six cities, the so called Harvard Six City Study (Dockery et al., 1993)

The most influential studies on the effect of ambient air quality on human health were the Harvard Six City Study (Dockery et al., 1993, Figure 7) and the American Cancer Society Study (Pope et al., 1995). Both studies agree well and show highest significant

correlation between PM_{2.5} and the relative risk of mortality, higher than for PM₁₀. The results of these studies were confirmed by an independent expert panel, using the same data as the authors of the original studies (HEI special report, 2000).

It has to be noted that the relative risk of mortality varies significantly between studies from different locations even when the same exposure metrics and statistical tools were employed. This variance is to be expected if the relative mortality risk and PM-exposure are interdependent for one or all of the following reasons: (a) Airborne particle hazard potentials depend on particle size distribution and chemical composition. These characteristics vary with location and time. (b) The investigated population varies in demographic and socioeconomic structures. Even adjustments for this variance cannot take all influences into account. (c) Variance in long-term stressors which vary between regions. (d) Exposure averages used for the statistical analysis may vary significantly between regions e.g. influenced by different distribution of single exposure values. All these parameter influence the results of the analysis and lead to spatial and temporal specific relative mortality risks. A constant relative mortality risk could only be expected if a linear correlation between exposure and health effect over the whole concentration range is given. Furthermore, also a single causal relationship between the chosen exposure metric (here e.g. PM mass concentration) and the health endpoint must exist.

A re-analysis of the Harvard Six City Study by Schwartz et al. (1996) investigated and compared the effects of PM₁₀, PM_{2.5} and PM_{2.5-10}, the latter being the coarse particle size fraction of particles being > 2.5 µm and < 10 µm in aerodynamic diameter. The correlation of daily averages with daily mortality rates showed the highest relative mortality risk as well as the highest statistical significance in all 6 cities for the PM_{2.5} size fraction. Only in one city, Steubenville, a correlation of the coarse particle fraction, PM_{2.5-10}, with mortality was detected. Most of the subsequent studies investigating health effects of PM₁₀ and PM_{2.5} corroborate the above finding of strong correlations between PM_{2.5} ambient concentrations and health effects, especially mortality.

The introduction of the new PM_{2.5} standard in the US in 1997 was mainly based on the results of the above mentioned studies by Dockery et al. (1993), Pope et al. (1995) and Schwartz et al. (1996).

Clancy et al. (2002) analysed the air quality and mortality rates 72 months prior and after the coal ban in Dublin coming into effect 1st September 1990. The decline in black smoke concentrations by 35.6 µg/m³ (70%) after the ban was accompanied by a significant decrease of respiratory (15.5%) and cardiovascular deaths (10.3%). They summarise “About 116 fewer respiratory deaths and 243 fewer cardiovascular deaths were seen per year in Dublin after the ban”.

Pope et al. (2009) published another seminal work, this time showing in their analysis the positive effect of cleaner air on life expectancy. They proofed that a decrease of 10 µg/m³ PM_{2.5} led to an increase in life expectancy of 0.61±0.20 years by comparing air quality and life expectancy of the late 70s’-beginning 80s’ with those of the late 90s’ and early 21st century.

The epidemiological studies overall show a clear link between PM exposure and adverse health effects in the population. Still important and significant uncertainties remain to be investigated to determine the “smoking gun” of PM.

1.2.2 Environmental Effects of Fine Dust

Airborne dust, next to its possible health effects also has further significant implications and environmental effects. Table 4 gives an overview of these effects.

1.2.2.1 Effects on Vegetation and Ecosystems

The human existence depends on the performance of the vegetation and ecosystems. Both, ecosystem structures and -functions are important socioeconomic factors. Society profits from the structural aspects of ecosystems in two ways: (a) the commercial values of products from this system such as fish, vegetables, minerals, wood, pharmaceuticals, gene pool and (b) the use of the ecosystem for recreation.

Ecosystem functions of clean water, air and vegetation are functions needed for these profits. Advantages we have based on the ecosystem functions are e.g. the natural remediation of pollutants, decomposition of natural materials, balance in air composition, regulation of the radiation balance of the earth and the 'storage' of solar energy.

Table 4: List of environmental effects of airborne particulate matter (PM)

Effect on	Keywords
Vegetation	Stress, reduced photosynthesis
Ecosystems	Change in nutrient cycling and pH of soils and waters → reduced productivity, change of the system
Materials	Accelerated corrosion, optical deterioration
Visibility	Reduced visibility by light extinction
Climate	Reduction and changes in the local and global albedo, mainly local to regional implications
Economy	Costs induced by particles through e.g. staining, contamination, corrosion, harvest reduction, changes in ecosystems

Based on the above given examples the importance of the preservation of ecosystem structure is self-evident. But currently only extremely few ecosystems may be viewed as not been hampered with by humans. Airborne particles are chemically diverse and are made of a variety of substances. The airborne transport, especially of the fine particles (< 1 µm in diameter), lead to the ubiquitous distribution worldwide. Hence the deposition and impact on vegetation and ecosystems are of great importance.

Three main deposition pathways of airborne particles exist:

- (1) wet deposition by e.g. rain,
- (2) interception with haze and cloud water,
- (3) dry deposition (slower but continuously).

The deposition of particles onto plants can be influenced chemically as well as physically by the nature and surface of the plant.

Coarse dust

Physical influence: Deposition of inert, bio-persistent particles can lead to changes in leaf temperature, stress and reduced photosynthesis (Guderian, 1986, Eller, 1977).

Chemical influence: Dust from cement factories can lead to reduced photosynthesis and leaf damage when moistened. Closer investigations showed that CaOH was released by the moisture and pH 12 values were reached on the leaf surface. Sea salt even leads to specific ecosystems, the “maritime forests”, which are adapted to the specific condition of salt water and sea salt spray / particles. Sea salt particles in general lead to plant damage by e.g. dry out through osmosis.

Fine dust

Fine dust mainly consist of secondary aerosols, meaning chemical compounds produced from gaseous precursors by either oxidation



or by polymerisation of e. g. isoprene stemming from plants (Figure 8).

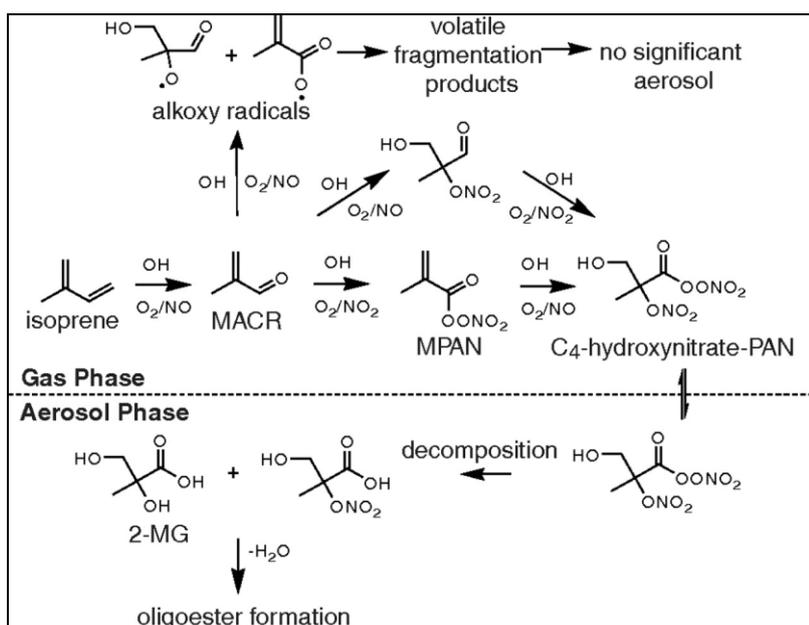


Figure 8: Possible chemical mechanism for the formation of isoprene secondary organic aerosol (SOA) under high-NO_x conditions (from Surratt et al., 2010)

Other important constituents are combustion derived organic and elemental carbon as well as mineral particles. Especially the combustion derived and secondary aerosols are in the particle size fraction < 1 μm aerodynamic diameter.

Nitrogen and sulphur compounds: Puxbaum und Gregori (1998) measured deposition rates for different types of particulate nitrogen and sulphur compounds in oak forests in Austria. A comparison of the determined deposition rates with those independently measured in the US revealed no elevated deposition for the European forests. Studies

investigating directly possible effects by the deposition of nitrogen and sulphur compounds did not show a significant effect for ambient concentrations (Martin et al., 1984)

Acid deposition: Effects of acid deposition onto plants, either gaseous or particle bound, have been studied in detail (Hogan et al., 1998), mainly due to the severe effects observed e.g. in Scandinavia. The most comprehensive overview is given in the NAPAP Biennial Report to Congress: An Integrated Assessment (National Science and Technology Council, 1998). Effects can be differentiated into acute, deposition and exposure to high ion concentrations (several $\mu\text{mol/l}$), and long-term effects of low concentrations (Cape, 1993). Exposure to acid deposition generally can lead to leaf damage, growth rate reduction, reduced buffer capacity, opening of the stomata.

Trace elements are those 80 of the 90 crustal elements which represent less than 0.1 mass-% of the earth's crust. Main sources of these elements in particles (mainly in fine dust $< 10 \mu\text{m}$ dia) stem from anthropogenic activities such as combustion processes, smelters, etc. These trace elements are normally oxidised in the particles (Omrod, 1984). A study of Marchwinska and Kucharski (1987) looking at combined effects of SO_2 and heavy metals in PM_{10} did not reveal any significant effects for peas, carrots and parsley. Nash (1975) showed zinc toxicity in moss close to a zinc smelter. Generally only in vitro studies, using elevated exposure concentrations showed toxicological effects for trace elements (Smith, 1990). Anyhow, those studies were conducted with soluble trace element compounds while particle bound trace elements have low or nearly no solubility in their forms as oxides, phosphates, sulphates etc. (Clevenger et al., 1991). No acute toxicological effects can therefore be expected for trace metals at normal environmental concentrations.

The deposition of particles often leads to indirect effects for plants and the corresponding ecosystem. The indirect effects are often of more importance than the direct effects and are mediated via the soil. Natural soils are a dynamic compartment of the environment. Changes in their structure or in their composition may significantly influence the bacteria and fungi, being important factors for the nutrient cycling. A good overview is given in a report by the US EPA (2000). One major influence of particle to soils is via the deposition of nitrate and sulphate linked to the corresponding acidification of the soils.

Nitrogen/ Nitrate: Some soils of North American forests, e.g. Appalachian Mountains, despite normally being nitrogen limited, show symptoms of saturation by bioavailable nitrogen. This saturation by bioavailable nitrogen is normally only observed in agriculture in cases when significant amounts of nitrogen are artificially added. Reasons discussed for the "saturation" of these soils are mainly the emission of NO_x by combustion processes leading to relatively high deposition rates of nitrate. This leads to increased growth in the corresponding forests, hence increasing the CO_2 sink in the biosphere. Nevertheless increased growth may lead in the long-term to reduced productivity, less root growths and reduced soil fertility (Aber et al., 1989; Aber et al., 1998).

Acid deposition: Acid deposition led in a few regions in Europe, e.g. Sweden and in the East of the USA to significant soil and water acidification (Johnson et al., 1991). Observed changes in the ecosystems of these regions correlated spatially and temporally well with yearly emission rates of NO_2 und SO_2 (Bondiotti and McLaughlin, 1992). The

most important change due to soil acidification is that in the solubility of aluminium (Reuss, 1983) which alters the nutrient availability and uptake via roots (Roelofs et al., 1987). Acidification also changes the uptake of calcium which is essential for the production of wood and viability of the cells. Reduced calcium uptake by plants due to lower pH-values and correspondingly higher aluminium concentrations clearly led to the “Waldsterben“ as well as in reduced productivity and wood quality. Changes in plant structures and ecosystems, like new and increased growth of birch trees in south Sweden, were observed by Falkengren-Gerup (1987).

Nutrient deposition: Airborne transport and deposition of mineral dust is an important factor supplying essential minerals to e.g. the Amazon in Brazil. Measurements and calculation of Swap et al (1992) derived deposition rates of mineral dusts as high as $190 \text{ kg ha}^{-1} \text{ yr}^{-1}$. Ben-Ami (2010) and papers cited therein confirm this finding and identify the Chad region as one of the main areas of mineral dust for the amazon. The importance of this Aeolian transport of mineral dust is nicely expressed by Swap et al. (1992):

Turnover rates for nutrient species deposited with Saharan dust in the Amazon Basin suggest a time scale of 500 to 20,000 years. We believe the dependence of one large ecosystem upon another separated by an ocean and coupled by the atmosphere to be fundamentally important to any view of how the global system functions. Any strategy designed to preserve the Amazonian rainforest or any part thereof should equally concern itself with the inter-relationship between the rain forest, global climate and arid zones well removed from Amazonia.

1.2.2.2 Effects to Materials

Effects of particles, often linked with SO_2 , on materials in the environment may be assessed together. Effects include those only of optical and aesthetic importance as well as those leading to severe structural damages to e.g. churches and bridges.

Corrosion of metals

The additive corrosion effect of air pollutants to natural aging processes depends on concentration and deposition rates of the pollutant as well as the presence of moisture. Different studies have shown that extended exposure to moisture by e.g. fog or rain accelerates corrosion. Moist surfaces along with deposition rates enhanced by fog/rain support corrosive electrochemical surface reaction e.g. on metals (Haynie und Upham, 1974, Sydberg und Ericsson, 1977)

Coatings

It is known that air pollutants such as SO_2 significantly alter the drying time for coatings and the durability of the colours (Holbrow, 1962, Davis et al., 1990). Further studies clearly showed that particles enhance corrosion via some of their substances (Cowling und Roberts, 1954) and damage coatings because of this property (Fochtman und Lagner, 1957, Wolff et al., 1990).

Aging processes of stones and concrete



The property of SO_2 along with moisture, sulphur oxidizing bacteria and fungi to accelerate aging of concrete and stones is known from numerous studies (Young, 1996, Diamkumaku et al., 1995, Sabioni et al., 1992, Schiavon et al., 1995). Further studies by e.g. DelMonte and Vittori (1985) clearly showed that organic and elemental carbon (soot) from deposited particles accelerate even this aging process by catalysing the formation of strong acids based on sulphur.

Figure 9: Soiling by gypsum and soot crusts at Lanyon building, Belfast University (Smith et al., 2011)

Aesthetic aspects

Environmental particles can lead to soiling of surfaces. Soiling is defined here as the deposition of particles, mostly $< 10 \mu\text{m}$ diameter, onto man-made surfaces. Soiling is normally only an optical effect, meaning that light reflection and transmission is altered by the deposition of the particles. Certainly soiling depends on the ambient particle concentration, their size distribution, deposition rate as well as the direction the surface is facing to, e.g. in direction to the main wind direction, and its surface roughness (Haynie, 1986). The chemical composition and particle morphology in combination with the surface characteristic of a building then determine whether soiling becomes visible or not (Nazaroff and Cass, 1991).

1.2.2.3 Effect on Visibility

Visibility is defined as the degree to which visible light penetrates the atmosphere as well as the brightness and colourfastness of the atmosphere (National Research Council, 1993). The visibility range as a metric of air quality was introduced in the USA by the “Clean Air Act” in 1977 to avoid man-made induced visibility impairments in so called “Class I Federal Areas”. Such areas are e.g. nature reserves and national parks.

Light passes air on its way from the object to the observer and is absorbed and scattered. The metric used to describe this is the light extension coefficient which itself is dependent on the (a) light scattering by gases, (b) light absorption by gases, (c) light scattering by particles and (d) light absorption by particles. Light scattering is dependent on particle size, morphology and chemical composition. The efficiency of particles to scatter light significantly decreases for particles $< 0.3 \mu\text{m}$ and $> 1 \mu\text{m}$ (Mie, 1908, Kerker, 1969, McMurry et al., 1996). Hence especially particles in the accumulation mode ($> 0.1 \mu\text{m}$ and $< 0.8 \mu\text{m}$) effectively scatter light and are important for the visibility. Particles of the nucleus range ($< 0.1 \mu\text{m}$) or from the coarse mode ($> 2.5 \mu\text{m}$) are of less importance. Absorption of light is less particle size dependent. Both light absorption and scattering by particles are significantly influenced by humidity and enhanced with increasing water content (Sloane et al., 1991, Omar et al., 1999).

Different indicators are used for the determination of visibility. The potentially best indicators, including those which can be linked to action plans are: (a) PM_{2.5} and its chemical composition, (b) determination of light scattering of the dried ambient aerosol, (c) determination of light scattering using ambient conditions and (d) measurement of the light extinction.

1.2.2.4 Climate Effects of Particles

Airborne particles influence the radiation balance of the earth by scattering and absorption of electromagnetic waves in the range of ultraviolet light (UV), visible light (VIS) and infrared (IR). The actual degree and direction of the effect depend on the optical properties of the particles, like extinction coefficient, single scattering albedo, asymmetric parameter, which all depend on particle parameters such as size, morphology, chemical composition, and internal or external mixture of the chemical compounds. Light extinction is normally largest when particle diameters are in the same size as the corresponding wavelength of the light.

The influence of particles on ultraviolet light is of interest since UV-light (specifically in the UV-B range of 290-315 nm) may cause significant biological damage to humans, animals and plants. Several investigations have proven the correlation between elevated PM concentrations and reduced UV-B radiation (Lorente et al., 1994, Wenny et al., 1998).

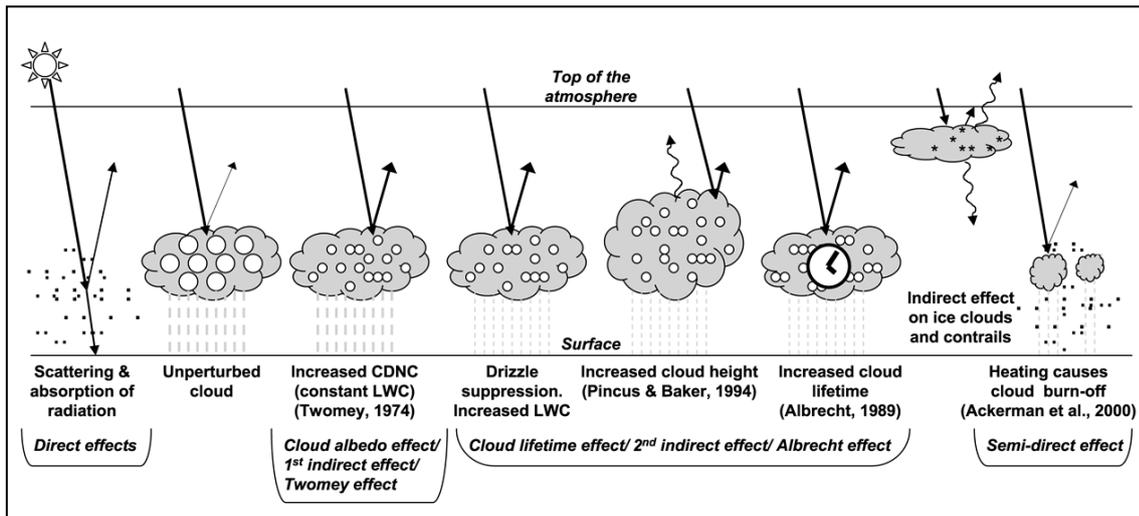


Figure 10: Diagram showing the various radiative forcing mechanisms associated with cloud effects (IPCC, 2007)

“Schematic diagram showing the various radiative mechanisms associated with cloud effects that have been identified as significant in relation to aerosols (modified from Haywood and Boucher, 2000). The small black dots represent aerosol particles, the larger open circles cloud droplets. Straight lines represent the incident and reflected solar radiation, and wavy lines represent terrestrial radiation. The filled white circles indicate cloud droplet number concentration (CDNC). The unperturbed cloud contains larger cloud drops as only natural aerosols are available as cloud condensation nuclei, while the perturbed cloud contains a greater number of smaller cloud drops as both natural and anthropogenic aerosols are available as cloud condensation nuclei (CCN).

The vertical grey dashes represent rainfall, and LWC refers to the liquid water content” (cited from IPCC 2007)

Research into the direct and indirect (via cloud condensation nuclei) effects of particles on the earth’s radiation balance has been significantly increased and some of the most important results were published in Charlson et al. 1987; Twoney, 1977; IPCC, 2004+2007; Tegen et al, 1996; Li-Jones and Prospero, 1998. Figure 10 gives a simplified overview of the different direct and indirect effects particle may have on the earth’s radiation balance. Generally four types of effects can be differentiated: (a) direct effects due to light scattering and absorption by the particles, (b) 1st indirect effect based on higher number concentrations of cloud condensation nuclei in polluted air, hence smaller droplet and therefore increased back scattering of light into space (cooling), (c) 2nd indirect effect based on longer cloud lifetime and formation in higher altitudes due to higher cloud condensation nuclei in polluted air and lower tendency to rain (cooling) and (d) the Ackermann effect based on increased light absorption by soot particles leading to “cloud burn” and heat layers.

The uncertainty in modelling and calculation of the magnitude of particle induced indirect effects is much larger, due to the higher uncertainties of the variables, compared to the direct effects. Calculations of the direct influence of particle bound sulphate on the earth’s radiation budget give values of -0.3 W/m^2 and -1.1 W/m^2 and between -0.4 W/m^2 and -1.6 W/m^2 for the indirect effect. Soot particles, on the other hand are viewed as leading to heating in the order of $0.1\text{-}0.5 \text{ W/m}^2$ along with crustal material (Aeolian dust) of about 0.2 W/m^2 (IPCC, 1994). For comparison, all greenhouse gases emitted between 1870 and 1994 lead to an additional greenhouse effect of about $+2.4 \text{ W/m}^2$. A recent revision of the IPCC data in 2007 confirms the effect of the greenhouse gases and summarises the particle bound overall effect on climate for direct effects to -0.5 (-0.1 to -0.9) W/m^2 and for the indirect effects to -0.8 (-0.4 to -1.8) W/m^2 .

Hence airborne particles, especially fine dust is in global view an important factor counteracting the anthropogenically induced greenhouse gas effects. Nevertheless it has to be noted that particles only remain airborne for a short period (days to weeks) and hence are worldwide not evenly distributed. The cooling effect of airborne particles is therefore more of regional than global relevance. Additionally it also has to be noted, that abatement strategies related to fine dust show much faster effects compared to greenhouse gases. The latter have lifetimes of years to centuries.

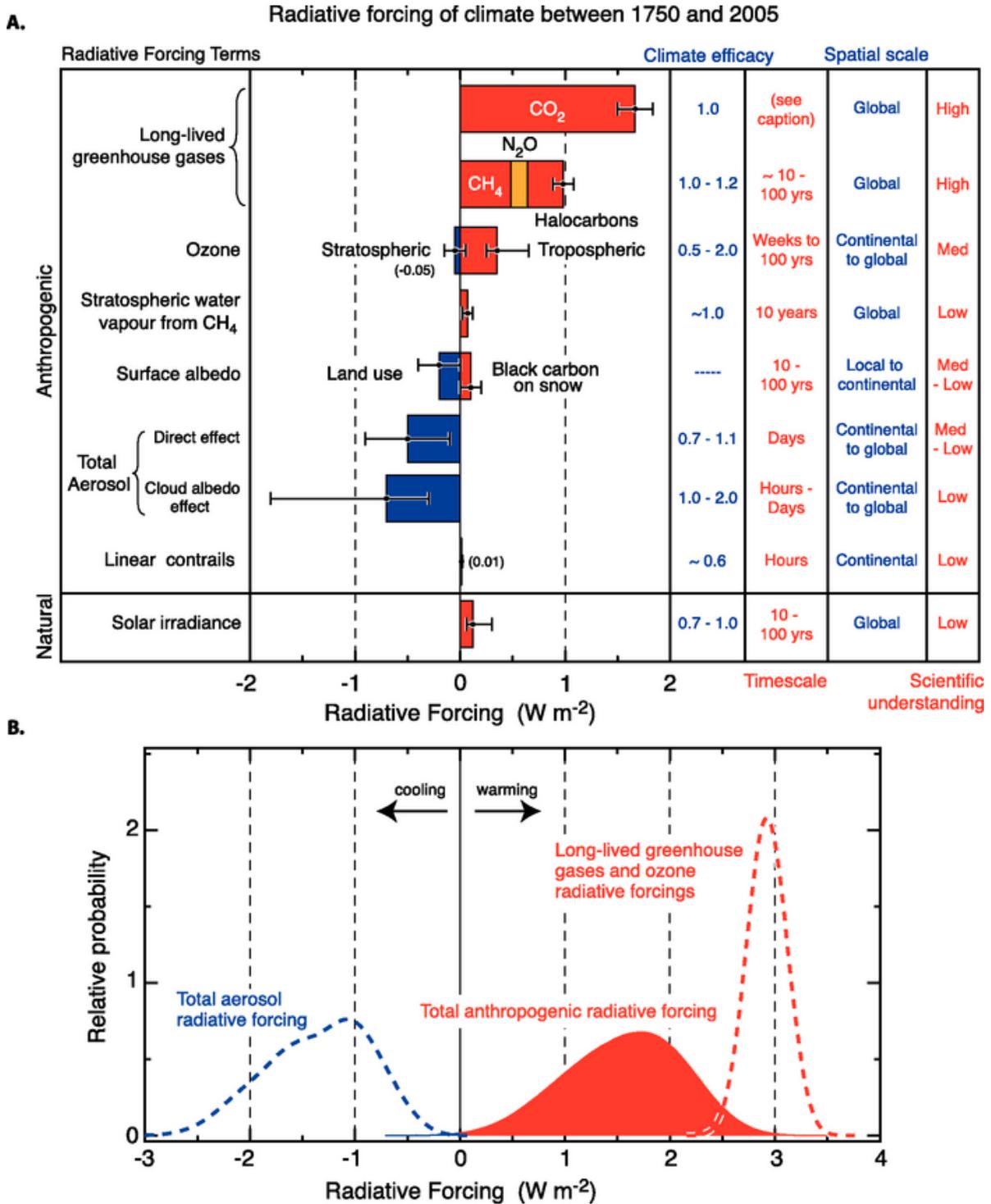


Figure 11: (A) Global mean radiative forcings (RF) and (B) their Probability distribution functions (IPCC, 2007)

“Global mean RFs from the agents and mechanisms discussed in this chapter, grouped by agent type. Anthropogenic RFs and the natural direct solar RF are shown. The plotted RF values correspond to the bold values in Table 2.12. Columns indicate other characteristics of the RF, efficacies are not used to modify the RFs shown. Time scales represent the length of time that a given RF term would persist in the atmosphere after the associated emissions and changes ceased. No CO₂ time scale is given, as its removal from the atmosphere involves a range of processes that can span long time scales, and

thus cannot be expressed accurately with a narrow range of lifetime values. The scientific understanding shown for each term is described in Table 2.11. (B) Probability distribution functions (PDFs) from combining anthropogenic RFs in (A). Three cases are shown: the total of all anthropogenic RF terms (block filled red curve, see also Table 2.12), LLGHGs and ozone RFs only (dashed red curve), and aerosol direct and cloud albedo RFs only (dashed blue curve). Surface albedo, contrails and stratospheric water vapour RFs are included in the total curve but not in the others. For all of the contributing forcing agents, the uncertainty is assumed to be represented by a normal distribution (and 90% confidence intervals) with the following exceptions: contrails, for which a lognormal distribution is assumed to account for the fact that the uncertainty is quoted as a factor of three, and tropospheric ozone, the direct aerosol RF (sulphate, fossil fuel organic and black carbon, biomass burning aerosols) and the cloud albedo RF, for which discrete values based on Figure 2.9, Table 2.6 and Table 2.7 are randomly sampled. Additional normal distributions are included in the direct aerosol effect for nitrate and mineral dust, as these are not explicitly accounted for in Table 2.6. A one-million point Monte Carlo simulation was performed to derive the PDFs (Boucher and Haywood, 2001). Natural RFs (solar and volcanic) are not included in these three PDFs. Climate efficacies are not accounted for in forming the PDFs.“ (cited from IPCC 2007)

1.2.2.5 Effects on the Economy

Assessments of the impact of particle induced damages to buildings, ecosystems and humans onto the economy and therefore onto a regional economy are very complex. A complete assessment of all effects could, due to missing input parameters, not be conducted (US EPA, 2000). The main problems for the assessments of the effect of airborne particles are the quantitative relation between an ambient concentration and an effect as well as the definition of an effect threshold.

First assessments were based on different approaches and models such as the Contingent Valuation Method (CVM, Hanley und Spash, 1993), the Timber Assessment Market Model (TAMM, Adams und Haynes, 1996), the Willingness to pay model (WTP, Watson und Jaksch, 1982, Harford, 1984), and the Damage Function Approach (DFA, Desvousges et al., 1998).

Table 5 gives an overview of the estimated costs due to particle emission and effects. This overview only comprises a few points induced by the particle burden.

A comprehensive approach assessing economic effects and balancing costs for cleaner air with the benefits was conducted thoroughly within the framework of CAFE (Clean Air For Europe, <http://www.cafe-cba.org/>). The final report on the cost benefit analysis of abatement strategies clearly showed that the benefits outweigh the costs by factors of 14-57 depending on the type of scenario chosen (AEA, 2008). E.g. total costs for abatement actions with regard to national emission ceilings, meaning introducing limitations to the overall allowed emission rate for air pollutants, are estimated to range between 0.9-1.5 billion €/yr. Monetised benefits related to health range from 15-70 billion €/yr and benefits non-health range around 300 million €/yr (AEA, 2008). It has to be noted that the above given results are based on abatement scenarios including gaseous and particulate air pollutants. Still highest correlation between adverse health effects and air pollutants are currently linked with either airborne particulate matter or ozone.

The cost benefit analysis of AEA (2008) clearly shows that further studies and investigations are needed to derive the information on costs induced by changes in the earth's radiation balance and onto ecosystems. Those are currently not adequately addressed in the cost-benefit analysis.

Table 5: Cost estimates induced by airborne particles

Effects	WTP (\$ value of that year)	DFA (\$ value of that year)	Source
House soiling	0.35 US \$ per home and TSP-Unit (1970)	0.32-0.82 \$ per home and year	Manuel et al., 1982 MathTech, 1990
Industry building soiling		0.62-0.98 \$ kg ⁻¹ PM or 45-57 US \$ per home and year (1990)	Tasdemiroglu, 1991
Monuments soiling	72-81 US \$ per home and year(1990)		Grosclaude und Sogue, 1994
Visibility	5-17 US \$ per home and year (1995)		Chestnut, 1997
Ecosystem effects	41 US \$ Albemarle-Pamlico Sounds 27 US \$ Chesapeake Bay 108 US \$ Tampa Bay per 0.5 kg nitrogen deposition (1997)		Avoided-Cost-Approach, EPA, 1998

2 FINE DUST MEASUREMENT TECHNIQUES AND STRATEGIES FOR EXPOSURE

The introduction, summarising the implications of airborne particulate matter on humans, the environment, climate and economy, shows the wide area of interest for fine dust and needed research. One specific and essential research need is that of the characterisation of airborne particles for exposure assessments. Figure 6 in the introduction points to the importance of exposure assessments to assess possible effects of airborne particulate matter on humans, ecosystems or manmade constructions. From the introduction it is also evident that PM mass concentrations do not constitute the sole particle characteristic which can be linked to effects.

2.1 Exposure Measurement Concepts

Exposure assessments can be based on (a) living standards, (b) measurement and modelling of ambient concentrations, (c) personal measurements or assessments based on personal activity pattern and (d) on biological monitoring (Figure 12).

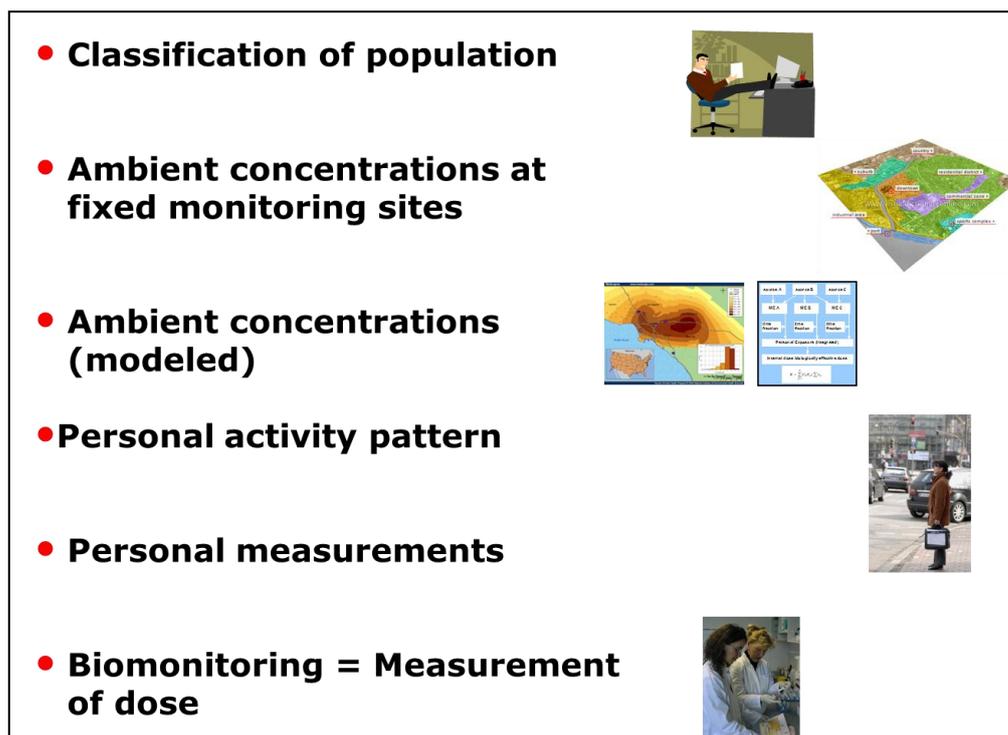


Figure 12: General possibilities of exposure determinations especially in view of epidemiological studies (adapted from J. Cyrus)

The simplest exposure assessment can be done on the classification of the population. This can be based on e.g. income, area of living or educational background. This type of exposure assessment is viewed as poor since it does not really take exposure measurements or scenarios into account but just a classification.

The most common approach for exposure assessment in environmental settings if huge cohort or population based studies are conducted (e.g. Pope III et al., 2009, Hoffmann et al., 2007, Kraemer et al., 2009, Dockery et al., 1993) is based on fixed ambient monitoring sites or modelling approaches. In case of fixed monitoring site approaches normally data from the nearest ambient measurement site are used as exposure values in the health studies. This approach can be refined using a so called land-use-regression model approach (e.g. Ryan and LeMaster, 2007). The basic idea of a land-use-regression model is that ambient concentrations of air pollutants are directly influenced by land use parameters such as population density, distance to major roads, distance to water surfaces, land use such as industry, agriculture, urban in different ranges around a measurement site. By pooling this information along with ambient concentrations of air pollutants from several measurement sites and performing a multi-linear regression analysis dependencies of air quality on land use data are obtained. The dependencies are subsequently used to calculate exposure values for the different members of a cohort study. The degree of information in this case is certainly closer to the “true” value than just data from a fixed monitoring site.

In case of use of population data such as hospital admission, morbidity or mortality data information on fixed monitoring sites can be improved by linking these data with e.g. chemical transport models such as EURAD (Ziegenbein et al, 1994). Chemical transport models base on emission inventories (e.g. Winiwarter et al., 2009) which represent the input parameter to the models beside meteorological data (wind speed, wind direction precipitation, temperature, radiation, etc.). Transport, atmospheric reactions, particle formation, dispersion and deposition of the air pollutants are subsequently modelled to derive three dimensional information on air pollutant concentrations. The latter can be used for exposure estimates if the resolution of the model is fine enough.

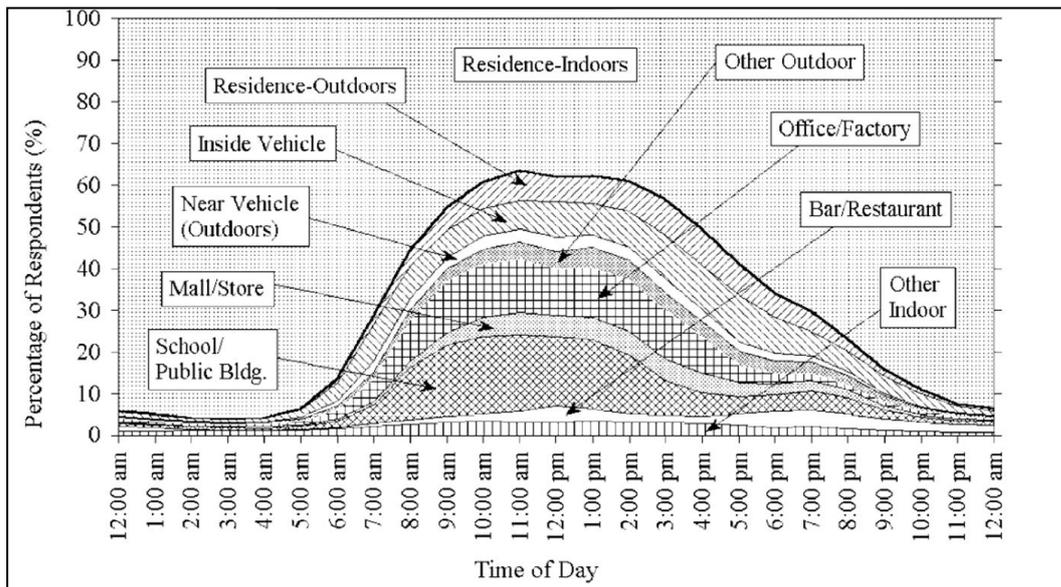


Figure 13: Plot showing the National Human Activity Pattern Survey of California allocating their activities to 10 different locations according time of day (Klepeis et al., 2001)

The personal activity pattern approach and personal measurements are the next class of improved approaches for exposure assessments. The personal activity pattern approach assumes the exposure in specific microenvironments to be comparable and that a common exposure value can be employed (e.g. Duan et al, 1991). Personal exposure can then be calculated for each case if the personal activity pattern was recorded or e.g. a general pattern can be assumed (e.g. Figure 13).

Personal measurements need personal samplers or monitors and subsequent data analysis for each sampled case. This approach is very tedious and labour intensive and hence can only be employed in few cases. Still it gives an excellent possibility of validation of personal exposure assessments based on e.g. activity pattern (Duan et al., 1991).

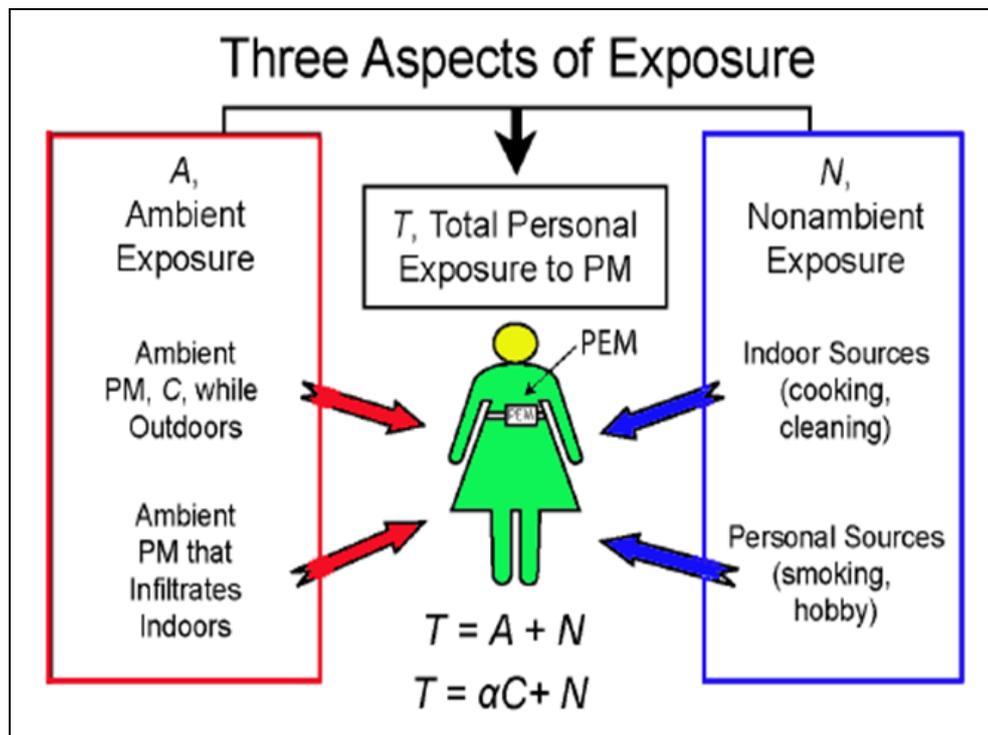


Figure 14: Three aspects of personal exposure (from Wilson and Brauer, 2006)

The different approaches introduced so far determine exposure either using ambient data or personal exposure measurements/calculations. Figure 14 exemplifies the different aspects of personal exposure when differentiating ambient and non-ambient sources of exposure. Exposure to “ambient” particles can be outdoors but also indoors since particles penetrate indoors. Hence the total exposure (T) can be assumed to be

$$T = \alpha C + N$$

with $\alpha > 1$ to account for outdoor particles in indoor micro-environments (Figure 14).

The last and most precise approach is based on biomonitoring. Biomonitoring is the measurement of the body burden of toxic chemical compounds, elements, or their metabolites, in biological substances (NCEH, 2005). Since these measurements are done in the exposed subject, by e.g. analysing blood or urine samples, biomonitoring can be viewed as being closest to the dose (or internal exposure).

It has to be noted that the closeness to the “true” exposure value increases from top to bottom in Figure 12 as well as the work load to achieve the corresponding assessment. The currently mostly used exposure assessments for huge cohort or population based studies are those based on ambient concentrations, either measured or modelled. For smaller cohort or more personal related exposure studies the approaches based on personal activity pattern and personal measurements are more often employed. Here, the approach based on personal activity pattern allows extrapolation while personal exposure measurements are limited to those humans investigated. The most precise but also most limited approach is biomonitoring which is mostly used in in-vivo exposure studies in well-defined settings, such as workplaces or scientific exposure studies.

2.2 Measurement Techniques for Fine and Ultrafine Dust

The selection of measurement and analysis devices and methods depends mainly on the research and monitoring tasks to tackle, their availability and costs. The following sections give an overview on measurement techniques and analytical methods. General overviews on the physics of airborne particles and measurement techniques can be found in Hinds (1999) und Willeke (1993).

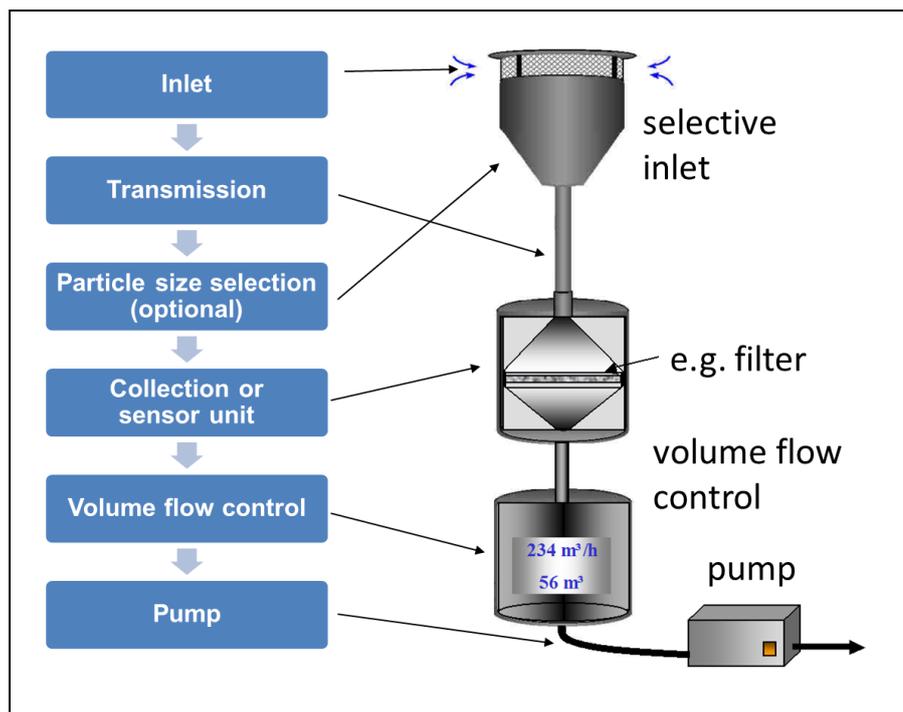


Figure 15: Basic scheme of an airborne particle sampling or sensor unit

Basis of most techniques to characterize ambient particles and aerosol concentrations is the sampling of air (Figure 15). The inlet system determines which particles can be collected or analysed by a sensor. Important is here to note that inlet designs should take care of wind direction and particle size independent sampling to avoid artefacts. Particles may also get lost via deposition in a tube during the transmission from the inlet to the sampling or sensor unit. Deposition in the tube is dependent e.g. on the degree of turbulence (impaction losses), length of tube (diffusional losses, mainly < 100 nm diameter) as well as for larger particles (> 1 μm diameter) on possible bends in the tube

(deposition by impaction). Further artefact may arise by thermodynamic changes taking place in the sampling line due to solar irradiation (heating of the aerosol) leading to evaporation of semivolatile substances. All these artefacts may influence chemical composition, particle size distribution and aerosol concentration. Therefore the sampling line should be kept short and straight. In the next unit, the sampling or sensor unit, particles are either collected on a substrate, filter or in liquids, or are directly analysed e.g. by counting. The volume flow control and pump ensure constant conditions at the inlet to reproducibly sample air and enable the calculation of concentrations by volume. The sum of all the units in a sampling and analysing unit determine the quality of the measurements. Two specific devices are described in the next two subsections while a broader overview on measurement devices is given in section 2.3.

2.2.1 The Cascade Sampling Precipitator

The determination of sources for PM₁₀ and PM_{2.5} and studies investigating factors influencing their ambient mass concentrations gained new importance due to the new challenges introduced by the 1st daughter directive to the European Air Quality Framework Directive in 1999 (EU 99/30/EC). One possibility to determine and quantify sources is based on multi-element analysis of PM and subsequent statistical analysis by multivariate statistics (Paatero and Tapper, 1994) or finger print modelling.

Total x-ray reflectance analysis (TXRF) allows the quantitative measurement of elements with atomic numbers > 13 within one analysis in extremely low concentrations, in ideal cases down to 10^{-8} g. Due to this sensitivity of the TXRF sampling and sample preparation are critical steps for the data quality. For example, some filter materials cannot be used for the detection of trace elements due to their high blank values (Schmeling, 1997).

The high sensitivity of TXRF compared to the normal X-ray fluorescence analysis is due to the total reflection of the x-ray at a “magic angle”, reducing matrix effects and enhancing sensitivity. This also means that the sample carriers have to be extremely flat and no filter can be directly used. If filters are used they have to be digested which is laboursome and leads to artefacts such as particles losses and contamination. Therefore a direct sampling system collecting particles directly on TXRF substrates was developed to avoid these problems (Figure 16, John, 2001; John et al., 2001; John et al, 2000; John et al., 1998). Only internal standard solutions have then to be added to the substrate prior to the multi-elemental analysis via TXRF.

A cascade impactor was specifically designed for particle sizes $< 1 \mu\text{m}$ (PM₁), 1-2.5 μm (PM_{1-2.5}) and 2.5-10 μm (PM_{2.5-10}) to directly determine the chemical composition of the environmentally and legally relevant particle sizes. The PM₁ size fractionation step was introduced in this concept since PM_{2.5} contains a variable fraction of coarse mode particles and a clearer separation of accumulation and coarse mode particles facilitate the identification of particle sources (see also section 1.1.1.).

Figure 16 shows the principal set up of the cascade sampling precipitator. The basic principle used for the collection of the particle size ranges from 2.5-10 μm and from 1-2.5 μm particle diameter, respectively, is impaction. Impaction uses the long stopping distances of larger particles for collection by diverting the air flow in a 90° angle over a substrate. Larger particles tend to “fly” straight ahead, hit the substrate and stick onto the

surface. Glue may be used to avoid bouncing of the particles. Impaction can be used for smaller particles only in low pressure conditions by reducing the interactions between air molecules and particles. In the set-up, an electrostatic precipitator (ESP, Dixkens et al., 1999, Figure 17) was chosen for collection of the submicrometer particles since a) it does not require any low pressure conditions and b) a centralised sampling spot usable for direct subsequent TXRF-analysis could be realised.

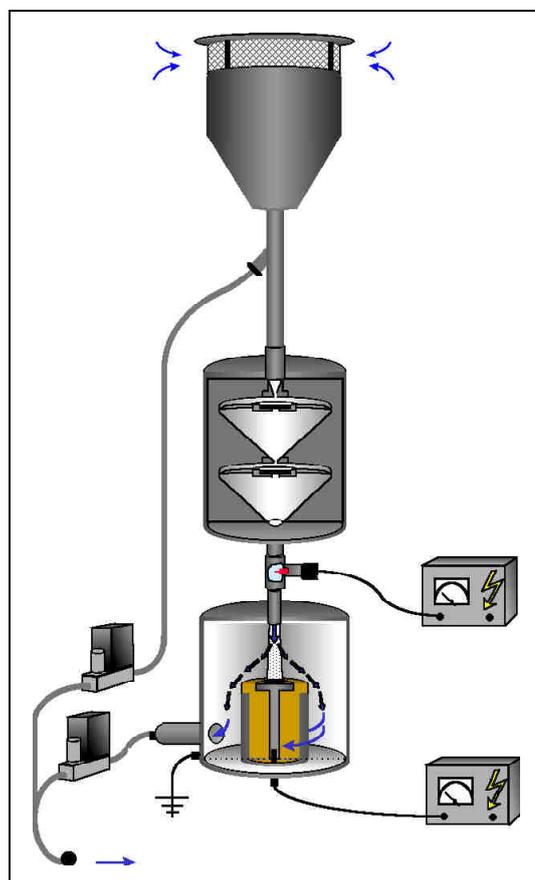


Figure 16: The cascade sampling precipitator for subsequent multi-element analysis by TXRF

The complete sampling system consists of a US-standard PM10 inlet, a cascade impactor for the cut-offs of PM2.5 and PM1, a unipolar charger to charge all particles for subsequent electrostatic sampling in the ESP (Figure 16).

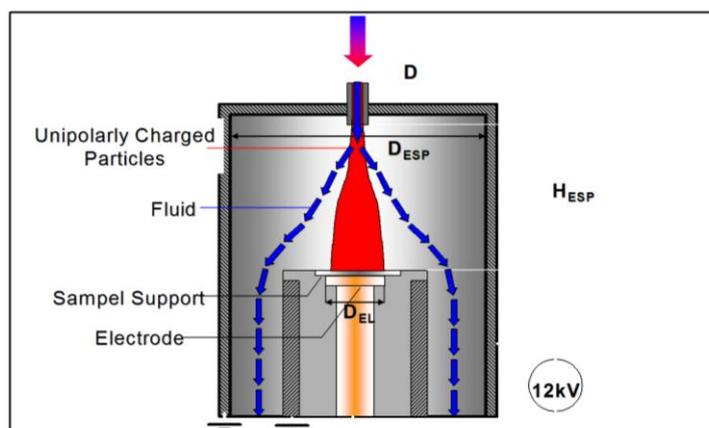


Figure 17: Electrostatic precipitator (Dixkens and Fissan, 1999)

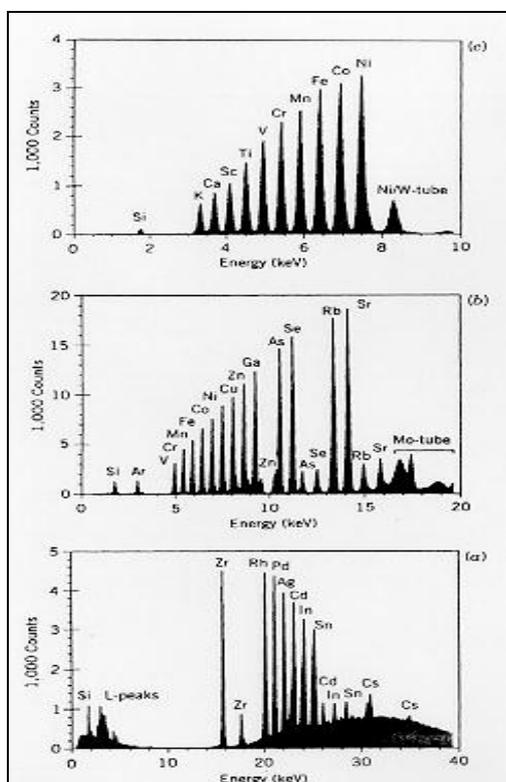


Figure 18: TXRF analysis spectrum of a multi-elemental standard solution as an example

The exemplary TXRF spectrum of a multi-elemental standard solution shown in Figure 18 was determined with X-ray tubes of NiW (a), Mo (b) and W(c).

2.2.2 Online Methods for the Determination of Particle Bound Ions

Several different techniques for the online determination of particle bound sulphate, nitrate and chloride based on a thermo-optical approach have been developed (e. g. Cobourn et al., 1981, Huntzicker et al., 1978, Sturges und Harrison, 1988, Clarke, 1991, Stolzenburg et al. 2003).

One of the devices (Stolzenburg et al., 2003) has been commercialized for sulphate and nitrate (Aerosol Dynamics Inc., USA and Rupprecht & Patashnik, 8400 series, USA) and has been tested in the field (e.g. Chow et al., 2008).

The main principle of this device is the impaction of the humidified PM_{2.5}-fractionated aerosol onto a sample and flash heating cell (d_{p50} ca. 100 nm). The humidifier (98-100 % rH) is used since nitrate and sulphate aerosols are hydrophilic and collection efficiencies by impaction are improved by the increased particle size due to humidification. The sample cell is flashed with clean air after a predetermined sampling period to avoid contamination by gases. The collection substrate is a NiCr-strip which is flash heated by a short burst (0.05 sec.) of high current to 350°C. Ammonium nitrate (NH₄NO₃), potassium nitrate (KNO₃) and sodium nitrate (NaNO₃) release NO_x at this temperature. The flash heating temperature is increased to 600°C for the determination of the particle bound sulphate. At 350°C only sulphuric acid (H₂SO₄) and the sulphate of ammonium sulphate ((NH₄)₂SO₄) are released, but not the corresponding salts of sodium and potassium sulphate. The released sulphur is mainly in the form of sulphur dioxide in the evolved gas. Sulphur dioxide and NO_x analysers are subsequently used to quantitatively determine the sulphate or nitrogen oxides and hence the corresponding mass concentrations.

A second approach currently employed for the detection of particle bound ions is based on in-liquid sampling. Devices for collecting airborne particles in liquid are e.g. the SJAC (Khlystov et al., 1995) or the particle-into-liquid sampler (PILS, Orsini et al., 2003). A commercial available instrument based on the SJAC linked to a high performance liquid chromatography (HPLC) for the detection of soluble ions is the Marga (ten Brink, 2007). Time resolutions of about one hour can be achieved with this method while the thermo-optical methods allow a time resolution of 10-15 minutes.

Overall, both methods allow the determination of diurnal variations. An overview of methods and recent results for online determination of organic and elemental carbon in Europe can e.g. be found in Kuhlbusch et al. (2009b). Similar devices and technologies do not exist currently for the determination of metals or metal oxides.

2.3 Particle Characterization (Kuhlbusch and Asbach, 2011)

The following section is a review which was published in the book: F. R. Cassee, N. L. Mills, D. Newby on “Cardiovascular Effects of Inhaled Ultrafine and nanosized particles” in 2011. This article focuses on the characterization of ultrafine particles since their determination and characterization is very challenging and the high mobility of particles below 100 nm in diameter are currently discussed to be of importance in view of possible health effects. A specific review of measurement techniques for nanoparticles in the environment has been published separately by Kuhlbusch et al. (2008b).

2.3.1 Introduction

Airborne particles are ubiquitous, and their impact on the earth’s ecosystems, climate and human health is generally accepted. The main impact mechanisms to the latter and their dependence on particle metrics such as size dependent number, surface or volume concentrations are still debated or even unknown. One size fraction especially discussed in view of health effects are the ultrafine particles-particles with diameters < 100 nm. Ultrafine particles can be differentiated from nanoparticles by their origin. Nanoparticles, in this chapter, denote (or)refer to intentionally produced engineered particles, while ultrafine particles are naturally produced or the by-products of anthropogenic activities. Both kinds of particles are defined by their size, namely they have diameter of < 100 nm in two or three dimensions. Hence fibres like carbon nanotubes as well as agglomerates in this size range are also viewed as nanoparticles.

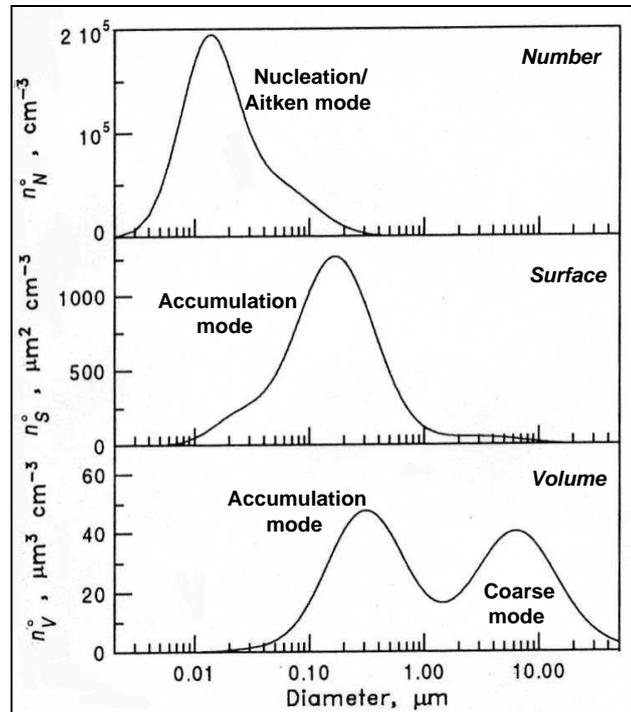


Figure 19: Typical urban aerosol size distribution (adapted from Seinfeld & Pandis, 2006)

Figure 19 shows a typical urban background aerosol size distribution weighted by particle number, surface area or volume (the latter is equivalent to the mass size distribution but with the weighting factor of particle density). These size distributions show how the different modes contribute to the size distribution. The volume size distribution is dominated by coarse and accumulation mode particles, the surface area size distribution by the accumulation mode and the number size distribution by the nucleation and Aitken modes.

2.3.2 Sources of Ultrafine Particles

Different size distribution modes can be identified for airborne particles. Coarse mode particles, particles with diameters mainly $> 1 \mu\text{m}$, are generally produced by mechanical processes like wind erosion and abrasion and are not further discussed in detail. Particle sizes in the submicrometer range can be divided into three modes: nucleation mode, Aitken mode and accumulation mode. The main difference between these modes is their characteristic size range: nucleation mode $< 10 \text{ nm}$, Aitken mode $10\text{-}100 \text{ nm}$, accumulation mode $200\text{-}700 \text{ nm}$. The presence of nucleation mode particles indicates homogeneous processes of aerosol formation by e.g. oxidation or polymerization. The main sink for nucleation particles is their conversion to the Aitken mode due to heterogeneous condensation. Both nucleation and Aitken modes are parts of the ultrafine particle (UFP) size fraction. Particle growth by condensation changes only the size but not their number. The last mode, accumulation mode, is mainly formed by coagulation and agglomeration, possibly coalescing, of Aitken and nucleation mode particles. Therefore these processes are also the main mechanisms of removing nucleation and Aitken mode particles. Particles in the accumulation mode grow until they are removed from the atmosphere by wet (e.g. rain) or dry (e.g. sedimentation) deposition.

The formation of UFP's is, in nearly all cases, by gas-to-particle conversion processes. These processes may be in an engine, in the atmosphere, and in the liquid or in the gas phase. Typical UFP's produced by these processes are

- soot from combustion processes in the exhaust (primary anthropogenic),
- sulphuric and nitrous acid from gaseous SO_2 and NO_x emissions from e.g. coal combustion and subsequent oxidation in the atmosphere (gas or liquid phase). These acids normally combine with ammonia to form the corresponding ammonium salts (secondary anthropogenic),
- soot from vegetation fires (primary natural),
- isoprenes and terpenes from plants and subsequent polymerization leading to a blue haze or iodine oxide and secondary organic oxidation compounds can be related to marine/coastal biota (secondary natural).

In some cases UFP's may be formed from mechanical processes such as bubble bursts (e.g. methane gases) on the surface of water. These bubbles can lead to small droplets which become UFP particles when they dry. A similar process, sprays produced from water pumps may lead to significant concentrations when used indoors. Nevertheless these kinds of source processes are of minor importance.

One of the main sources of UFP's discussed in view of health issues are tailpipe emissions from street traffic. Therefore, particle number measurements will be required for Euro 5 certification of passenger cars with diesel engines (ECE-R83 amendment in 09/2011) and Euro 6 for heavy-duty engines. No particle size distribution of diesel engine emissions are shown since the results very much depend on factors such as dilution ratios, temperature and residence times. Figure 20 shows the particle size distributions determined upwind / downwind of a freeway and the corresponding ratio. It is evident that two particle modes are emitted, one in the nucleation/Aitken size range and another around 100 nm diameter. The former can be attributed to the condensation of unburned / partially burned fuel and possibly also to freshly-formed sulphate particles while the latter very likely stems from soot emission.

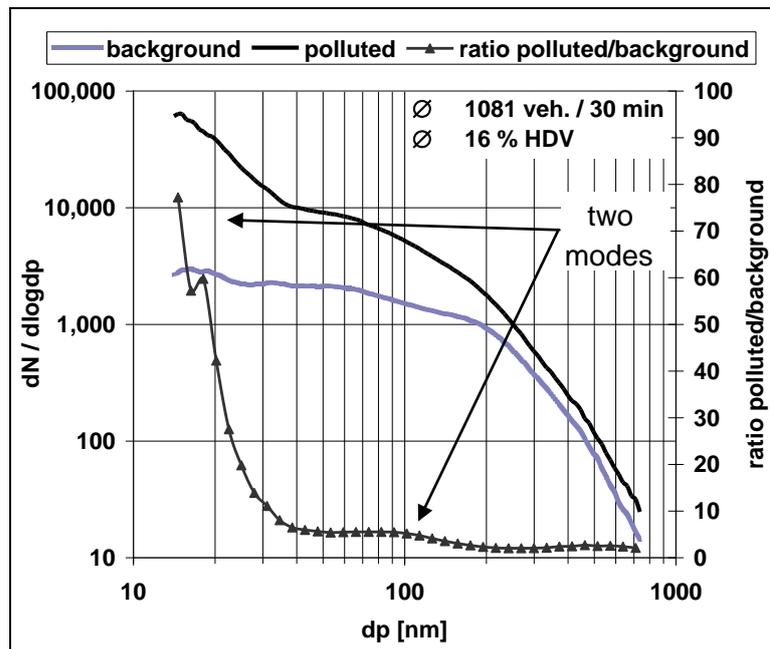


Figure 20: Particle size distributions (based on $\#/cm^3$) upwind and downwind of a freeway (primary y-axis) and the corresponding ratio of the size distributions (secondary y-axis)

The most recent national UFP emission inventory ($PM_{0.1}^3$) is from the UK from 2007. Figure 21 shows the main sectors contributing to $PM_{0.1}$ mass emissions between the years 1970 and 2007. The main anthropogenic sources are industrial processes and traffic related emissions, especially street traffic. The largest decrease from 1970 to 2007 was in industrial emissions while traffic related emissions only slightly decreased. Another source with significant decrease in emissions was domestic combustion, mainly used for heating. The emission inventory here gives reasonable relations between the source groups and can be viewed as a representative for other countries even though only very few emission inventories for UFP's are available. It has to be noted that this inventory does not include natural sources. The high uncertainty of the emission inventory due to the lack of quality input data and difficulties in up scaling emission factors from a few

³ <http://www.airquality.co.uk/reports/empire/naei/annreport/annrep99/index.htm>

investigated sources to all has also to be taken into account. No emission inventories related to particle number or surface area concentrations are known to the authors.

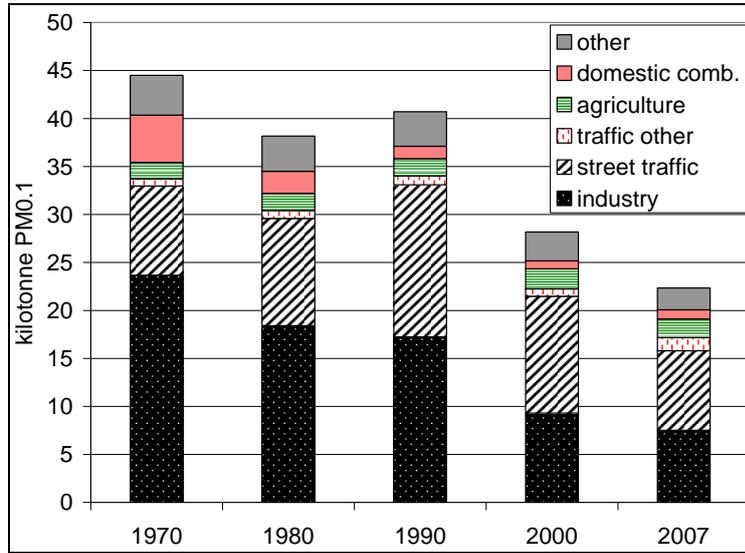


Figure 21: Emission inventory of PM0.1 emission in the UK from 1970 to 2007. Basic data from the National Atmospheric Emission Inventory (NAEI, 2007)

2.3.3 Measurement Techniques

A variety of parameters of airborne particles can be assessed. For regulatory measurements, single particle analysis of their chemical composition, morphology, etc., are usually not feasible. Instead integral parameters such as mass concentrations are measured. The most common aerosol measurement techniques for the different metrics are discussed in this chapter. In general, the measurement techniques can be distinguished or classified into discontinuous methods that determine average values over the measurement period, e.g. 24 h, and continuous methods that determine concentration in (near-) real time. While the discontinuous methods usually operate quite independently and require little maintenance, most continuous samplers require more attention and maintenance, and produce much more extensive data sets that require more time consuming data evaluation.

The determined metrics can be distinguished into size-integrated total concentrations, usually below a certain limiting particle size, and size-resolved distributions. Determined concentration measures comprise particle number, surface area (Fuchs or lung deposited area) and mass concentrations.

2.3.3.1 Discontinuous particle analysis methods

Discontinuous particle analysis methods mainly consist of two steps-the sampling on a substrate and the subsequent analysis of the sample. The sample analysis can be either on the bulk or on single particles. Sampling of airborne UFP's can be done on filters, specific surfaces or in liquids. Techniques for sampling nanoscale particles on surfaces

are generally based on electrostatic forces, thermophoretic forces or the movement due to Brownian motion. Analytical methods and techniques for the measurement of the following will be discussed:

- Mass concentration
- Surface area concentration
- Number concentration
- Number size distribution
- Chemical composition and morphology
- Reactive oxygen species (ROS) potential

Mass Concentration

The most common way of measuring atmospheric airborne particle mass concentration is by deposition of particles onto a filter at a known volumetric flow rate Q_{sample} for a given sampling time (e.g. 24 h). The total mass of particles Δm deposited during the measurement period, Δt , is determined gravimetrically by pre- and post-weighing and divided by the total sample volume to determine the average particle mass concentration. According to DIN EN 12341 (1998), this method is acknowledged as reference method for the determination of particle mass concentrations of particles below 10 μm (PM_{10}).

Due to the dependency of the particle mass on the cube of the particle diameter, this technique is very insensitive to ultrafine particles (<100 nm), because ultrafine particles have very little mass. As an example: one 10 μm particle has the same mass as one million 100 nm particles. Only the use of a pre-selector that removes all particles larger than 100 nm may help. Such a pre-separator, however, would cause a high pressure drop and therefore requires a larger pump than currently used in discontinuous samplers. Furthermore the accumulated mass of ultrafine particles even during a full day sampling may still be too low to be accurately detectable. The discontinuous measurement of ultrafine particle mass concentrations can therefore be considered not feasible.

Cascade Impactors

Impactors sample particles based on particle inertia. The impaction principle is based on the acceleration of the aerosol in a nozzle and the direction of the outflow from the nozzle onto a flat plate, called the impaction plate. The flow is deflected by 90° by the impaction plate and particles above a certain size, the so-called cut-off size, cannot follow the gas flow due to inertia and are deposited on the impaction plate. Besides the physical size, particle density also plays a role in inertial deposition. Hence the deposition efficiency is usually given in terms of the aerodynamic diameter (Hinds, 1999). Cascade impactors, as shown in Figure 22, contain an array of impaction stages with decreasing cut-off diameters, usually backed up by an after filter in the end. Consecutive physical and/or chemical analyses of the impaction plates therefore provide size-resolved information on the sampled particles. Since inertia is directly proportional to the particle mass, it decreases drastically with decreasing particle size. Most cascade impactors are therefore designed to deposit particles in a size range $\geq 1 \mu\text{m}$. In order to deposit ultrafine particles (<100 nm) by inertia, the pressure in the impactor has to be significantly reduced, thus requiring large pumps. The low pressure may also lead to loss of

semivolatiles which has to be taken into account if samples are used subsequently for mass and chemical analysis. The Micro Orifice Uniform Deposit Impactor (MOUDI, MSP Corporation, Marple *et al.*, 1991) deposits particles between 18 μm and 56 nm in 10 stages (model 110), which can be extended down to 10 nm by the addition of three more stages (NanoMOUDI). The last stage of the NanoMOUDI operates at a pressure of only 12.2 kPa. The (Nano-) MOUDI uses up to 2000 micron sized orifices to homogeneously distribute the particles on the sample substrate. To further increase the homogeneity of the deposition, all substrates are rotated during sampling.

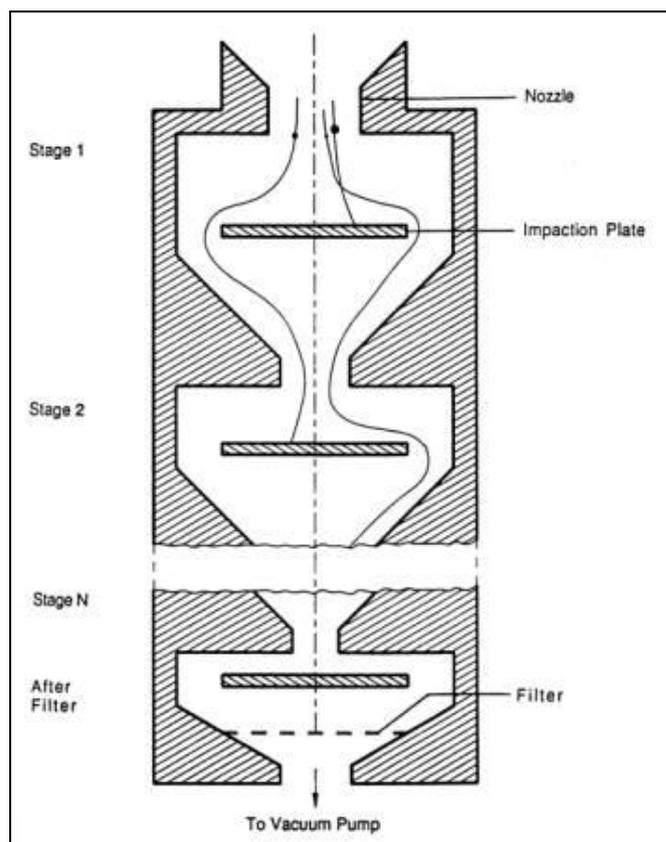


Figure 22: Schematic of a cascade impactor

Another commercial cascade impactor is the Electrical Low Pressure Impactor (ELPI, Dekati, Keskinen *et al.*, 1992). The ELPI uses altogether twelve stages to deposit particles in a size range from 10 μm down to 30 nm on impaction plates. An optional after filter reduces the lower size limit to 7 nm. The last stage operates at a pressure level of only 10 kPa. Prior to deposition, particles get charged by means of a corona charger. Each impaction plate and the after filter are equipped with means for measuring the current induced by the deposition of the particles. With a known charge distribution of the particles, the number concentration of the deposited particles is determined for each stage, thus delivering the particle number size distribution with high time resolution. Marjamäki *et al.* (2000) showed good agreement between ELPI and SMPS size distribution measurements. The impaction stages of the ELPI can be used for consecutive analyses of e.g. the particle mass or the particle chemical composition.

Surface Area

Adsorption of gases is a widely used and accurate technique for total surface area measurements. To determine the surface area, the surface and pore openings of particles, including the irregularities and pore interiors, are completely covered with a condensed gas down to an atomic level. This technique requires a clean surface. Hence the sample is heated to an elevated temperature under vacuum to “outgas”. Nitrogen is often the gas used as its molecular size is well established, it is inert and is available in high purity at a reasonable cost. The “outgassed” sample, under high vacuum in its sample tube, is immersed in a coolant bath of liquid nitrogen at -195°C. At this stage the sample is ready to attract gas molecules onto it when they are admitted to the sample tube.

The amount of gas adsorbed and the resultant sample pressure are determined and recorded. This data is subjected to a number of calculations to obtain surface area. A widely accepted calculation is BET (after Brunauer, Emmett and Teller – 1938). Although the theory is complex, BET measurements are relatively simple to obtain and widely applicable, and the results are highly reproducible. Nevertheless, a minimum particle mass is required for the measurements and hence its applicability to UFPs is very limited.

Number Concentration and Number Size Distribution

There are no real practical offline methods for the determination of number concentrations and number size distributions. Anyhow, nanoscale particles may be deposited onto specific surfaces by thermal precipitation (Azong-Wara et al., 2009, Orr and Martin, 1958), electrostatic precipitation (Dixkens and Fissan, 1999) or in diffusion batteries (Fierz et al., 2009, Sinclair et al., 1976). Particle number concentrations as well as particle number size distributions can be determined by e.g. microscopic analysis (SEM, TEM) and relating the results to sampled volume. Even though that the whole analysis takes some time it allows the combination of size dependent analysis and morphological / chemical analysis of single individual particles.

Chemical Compositions and Morphology

Once the particles are collected on a substrate they can be analysed using modern chemical analytical techniques. These techniques may analyse the bulk material or single particles. Some of the techniques for bulk analysis are high performance liquid chromatography, mass spectrometry MS and atom absorption spectrometry. Techniques for single particle analysis are SEM or TEM (scanning or transmission electron microscopy) coupled with EDX (energy dispersive analysis). A review of analytical methods and their applications in the field can be found in e.g. Chow and Watson (2007), Maynard (2000) and Colbeck (2008).

ROS-Potential

The formation of direct and indirectly particle induced reactive oxidant species (ROS) is believed to lead to oxidative stress, which is nowadays considered to be a major factor for pulmonary toxicity and further adverse health effects (Donaldson et al. 2004, Li et al. 2003). Hence reactive oxygen species can be viewed as a unifying and alternative health indicator (Donaldson et al. 1996, Nel et al. 2006). For the ROS-generation potential and

finally, the possible toxicity of PM, the components of the PM-mixture and the particle size seem to be highly relevant factors. It was shown that especially ultrafine particles have a high reactivity related to PM mass (Li et al., 2003, Ntziachristos et al., 2007b). Still, it has to be noted that the method used for the determination of particle reactivity by Ntziachristos et al. (2007b) has a high sensitivity to ROS coming from aerosol organic hydrocarbons, such as polycyclic aromatic hydrocarbons and quinones (Squadrito et al. 2001), while another method based on electron spin resonance spectrometry (Shi et al., 2003) has a high sensitivity towards metal mediated ROS by e.g. Fenton-like reactions. Both methods can be applied to PM sampled on filters or other appropriate surfaces.

Künzli et al. (2006) used a similar bioassay as Ntziachristos et al. (2007b) and applied it to PM_{2.5} samples from 20 European cities. They concluded, “PM oxidative activity varied significantly among European sampling sites.” Correlations between oxidative activity and all other characteristics of PM were low, both within centers (temporal correlation) and across communities (annual mean). Thus, no single surrogate measure of PM redox activity could be identified. Because these novel measures are supposed to reflect crucial biologic mechanisms of PM, their use may be pertinent in epidemiologic studies. Therefore, it is important to define the appropriate methods to determine oxidative activity of PM.

2.3.3.2 Continuous methods

The following metrics, which can be determined by continuous methods, will be discussed in this section:

- Mass concentration
- Surface area concentration
- Number concentration
- Number size distributions
- Chemical compositions
- ROS-potential

Mass Concentration

Continuous monitors for the determination of ambient particle mass concentrations (also called automatic or online monitors or samplers) are widely used in air quality supervision stations around the world. Compared to manual sampling, automatic sampling offers several advantages. It delivers data with a high time resolution in almost real-time, e.g. airborne particle mass concentration data is available within a matter of minutes (depending on the averaging time span) and can be plotted as diurnal variation. Most commonly used continuous monitors for the determination of ambient particle mass concentrations are the Tapered Element Oscillating Microbalance (TEOM[®]) and different versions of beta-attenuation monitors (also called beta-gauges). Anyhow it has to be noted that the lower detection limits of these online monitors are normally not low enough to detect UFP's reliably.

The Tapered Element Oscillating Microbalance (TEOM®)

The TEOM® is an instrument for continuous measurement of ambient particle mass concentrations. It mainly consists of three components: a size selective inlet, a sensor unit and a control unit.

In the sensor unit, air is continuously drawn through a filter, which is located in an exchangeable filter cartridge on the top end of a tapered element. The tapered element is a hollow tube, clamped on one end and free to vibrate on the other. The oscillation of the tapered element is excited with a constant energy, causing the system containing the tapered element and the filter cartridge to vibrate precisely at its natural frequency, if the total mass remains constant. When an aerosol is drawn through the filter the mass of the filter increases due to the deposition of particles. This causes the frequency f of the oscillator to decrease. The frequency drop is measured and used along with a calibration constant K_0 to determine the corresponding mass Δm of deposited particles (Patashnick et al, 1991).

$$\Delta m = K_0 \left(\frac{1}{f_1^2} - \frac{1}{f_0^2} \right) \quad (\text{E1})$$

Along with the constant volumetric flow rate, the mass concentration is calculated. The time resolution is theoretically only limited by the resolution of the frequency recording (2 sec.). In practice, a noise frequency, which is not related to the mass gain of the filter, is superposed on the oscillation of the tapered element. Since the noise creates a balance of positive and negative artefacts, it can be eliminated by averaging over longer time intervals. For regulatory measurements, the averaging time interval is usually set to 5 minutes or longer.

The TEOM technique is usually used to determine the mass concentration of all particles below 10 μm (PM_{10}), 2.5 μm ($\text{PM}_{2.5}$) and in some cases below 1 μm (PM_1). In the case of ambient PM_1 , the TEOM technique is often operated near its detection limit, because of the low mass of submicron particles. The mass concentration of ultrafine particles, i.e. $\text{PM}_{0.1}$, would even be significantly lower than the PM_1 mass concentration. The use of a TEOM can therefore also not be considered as feasible for ambient UFP measurement.

The Beta Attenuation Monitor

The beta attenuation monitor (also called beta gauge) uses the absorption of beta rays (electrons) by particles collected on a filter (Williams et al., 1993). Inside the monitor, beta rays are emitted by a radioactive source. Commonly used sources are the carbon isotope ^{14}C and Krypton (^{85}Kr).

The aerosol is drawn through a sample inlet, which might be size selective, such as an impactor or a cyclone. From there it is transported to the filter medium, where the particles are deposited on a small spot. As a filter medium, a filter tape (similar to a tape used in an audio tape recorder) is used that is moved stepwise to create a new deposition spot once the current spot reaches the default filter loading threshold. The radioactive source and a Geiger-Müller-Counter are located on two opposite sides of the deposition spot. The Geiger-Müller-Counter counts impulses of incoming beta-electrons. Due to the absorption of beta rays by the particles, the impulse rate decreases as the mass of

deposited particles on the filter increases. Assuming a homogenous distribution of the particles on the filter medium, Lambert-Beer law can be used (e.g. Gebhart, 1993) to determine the mass Δm of the deposited particles:

$$\ln\left(\frac{n_{\beta 0}}{n_{\beta 1}}\right) = \xi \cdot \frac{\Delta m}{A_d} \quad (\text{E2})$$

where ξ is the extinction coefficient and A_d is the surface area of the deposition spot. The mass concentration can then be calculated by dividing the mass change by the total sampled volume during the time period.

For measuring ultrafine particle mass concentrations, the Beta Attenuation Monitor shares the same problem with the TEOM, i.e., the mass of these particles is too low and thus creates too low a signal to be accurately detectable. Therefore the use of a Beta Attenuation Monitor can be considered unfeasible to measure UFP mass concentrations.

Surface Area Concentration

The surface area especially of small particles has been reported to be of particular health relevance (Brown et al., 2001, Oberdörster, 2000). Oberdörster (2000) showed that the health effects observed in rats after instillation of fine (250 nm) and ultrafine (50 nm) TiO₂ particles followed one distinct dose-response relationship, when plotted as a function of the total particle surface area, whereas when plotted versus the particle mass, the relationship was different for the two sizes. As a result, surface area concentration has recently been discussed as being a health relevant metric not only for workplaces in nanotechnology industry, but also for atmospheric measurements.

It should be noted that currently no device exists that directly determines the geometric surface area concentration of airborne particles. All instruments that aim at determining the surface area concentration are based on attachment of ions on the particle surface. This ion attachment rate, however, is not proportional to the geometric surface area of particles, but to the so called Fuchs or active surface area. The Fuchs surface area is a measure for the ion attachment rate and can therefore be a valuable property for atmospheric processes of particles, but not so much for their health relevance. Only one of the commercially available instruments provides a signal that is directly linked to the geometric surface area of airborne particles. The Nanoparticle Surface Area Monitor (NSAM, TSI model 3550) delivers the fraction of the airborne geometric surface area concentration that would deposit in the human lung.

Currently the only way to determine the total geometric surface area concentration is by calculating it from the number size distribution as measured by an SMPS. This, however, requires several assumptions and is only accurate for spherical particles. Maynard (2002) suggested a method to estimate surface area concentrations based on the measurement of number and mass concentration of particles. This method is only suitable for unimodal, lognormal size distributions and therefore not discussed further here. In case of spherical particles, the total surface area can also be determined by deposition of particles and consecutive SEM analysis of the particle cross sectional area. This procedure is very time consuming and thus not discussed further here.

“Surface Area” Monitors

A common way of measuring active surface area is by attachment of ions produced by electrical corona discharge. LQ1-DC (Matter Engineering) uses a corona discharge to charge the particles before they are deposited and the current measured. According to the manufacturer’s specification sheet, LQ1-DC can measure concentrations of 0-2000 $\mu\text{m}^2/\text{cm}^3$ (active surface area) of particles in the size range from a few nanometers to 10 μm with an adjustable time resolution of 3-30 s. Jung and Kittelson (2005) reported that the LQ1-DC results are proportional to $d_p^{1.36}$, thus they agree well with the Fuchs surface area measurements. The instrument has recently been upgraded with a set of diffusion screens with changing mesh sizes to also provide particle size information (Fierz *et al.*, 2009).

The Nanoparticle Surface Area Monitor (NSAM, TSI model 3550) is an instrument that determines the fraction of the airborne particle surface area concentration that would deposit in either the alveolar or tracheobronchial region of the human lung (Fissan *et al.*, 2007), thus delivering a dose-relevant metric. For the lung deposition, the widely accepted ICRP model (ICRP publication 66) is considered. The lung deposition curves according to this model are shown in Figure 23.

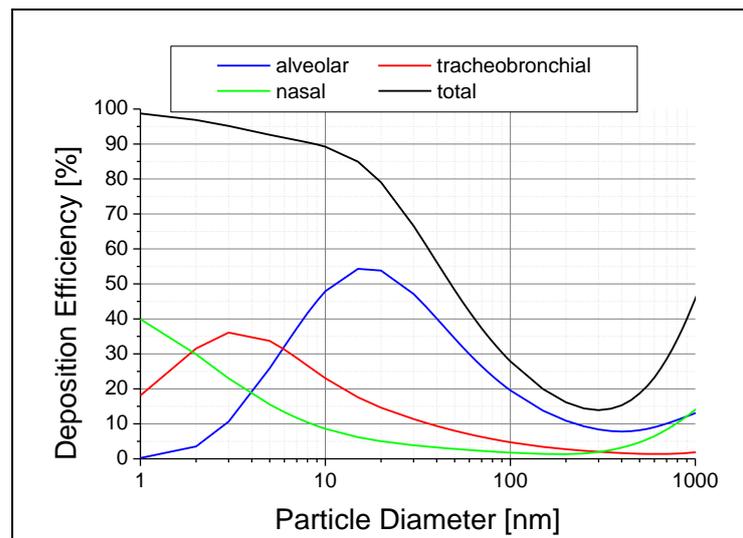


Figure 23: Lung deposition curves, according to ICRP model (ICRP publication 66)

The instrument was originally designed to cover the particle size range up to 1 μm , but Asbach *et al.* (2009b) revealed that it shows highest accuracy between 20 nm and 400 nm. In NSAM, particles are charged in an opposed flow diffusion charger, and excess ions removed in an ion trap. The particles are then deposited on a filter and the resulting current is measured with an electrometer. The voltage in the ion trap can be adjusted to manipulate the penetrating particle size distribution such that the particle induced current is proportional to the geometric surface area concentration of the particles that deposit in either the alveolar or tracheobronchial region of the human lung (Shin *et al.*, 2007). A schematic of the instrument is shown in Figure 24. Recent investigations have shown that nasal as well as total deposition in the respiratory tract can also be determined with this instrument, based on a single measurement without the need for changing the ion trap

voltage (Asbach et al., 2009b). NSAM has a time resolution of 1 s and can thus be used to monitor even quickly changing aerosols. The instrument has been used by the authors in a variety of applications, including nanotechnology workplace monitoring, welding fume measurements and atmospheric measurements. NSAM delivered comparable results with those calculated from number size distribution measurements over a large concentration range, covering several orders of magnitude.

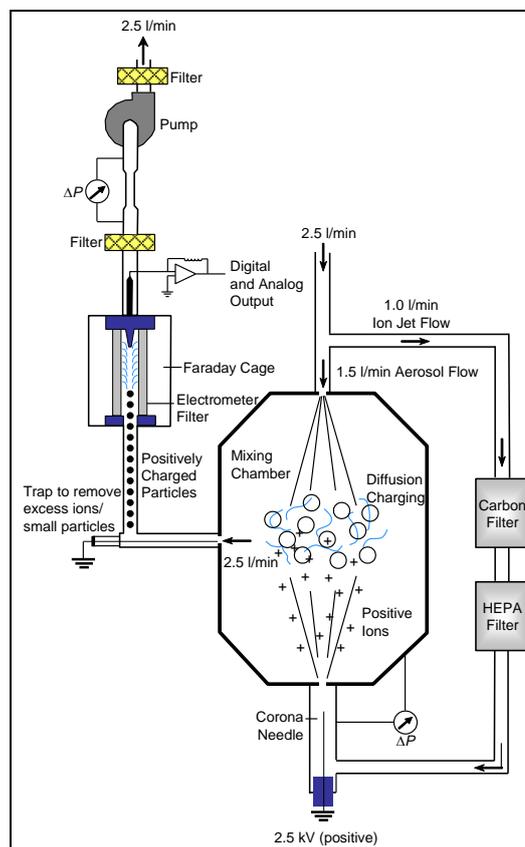


Figure 24: Schematic of Nanoparticle Surface Area Monitor (NSAM, TSI model 3550)

For the sole detection of ultrafine particles, i.e., particles below 100 nm, a pre-separator to remove all particles above 100 nm would be required. This is currently unavailable and the development of such a pre-separator not simple, because a commonly used inertial separator would introduce a significant pressure drop, which may interfere with the charging process and overburden the NSAM pump.

Another instrument to determine particle surface area concentration is the Epiphaniometer. The Epiphaniometer detects the attachment of Lead atoms (^{211}Pb) produced by decay of Actinium (^{227}Ac) on a particle surface as surface area relevant measure (Gäggeler *et al.*, 1989), i.e. it determines the active or Fuchs surface area. Shi *et al.*, 2001 reported good agreement between Epiphaniometer and SMPS/APS measurements. The instrument is not commercialized and thus a pure research instrument. Since it uses a nuclear source and toxic lead, it is not well suited for regulatory

measurements of particle surface area concentrations and therefore not discussed further here.

Determination of Surface Area Concentration From Number Size Distribution

If the number size distribution is available, e.g. from SMPS or FMPS measurements, the surface area size distribution can be easily and accurately calculated if all sampled particles are spherical. To calculate the surface area concentration $\Delta S_i(d_{p,i})$ for each size channel with mean diameter $d_{p,i}$, the number concentration $\Delta N_i(d_{p,i})$ measured in this size channel has to be multiplied with the surface area of a particle with the corresponding diameter :

$$\Delta S_i(d_{p,i}) = \Delta N_i(d_{p,i}) \cdot \pi \cdot d_{p,i}^2 \quad (E3)$$

Figure 25 shows an example for the conversion of a number into a surface area size distribution.

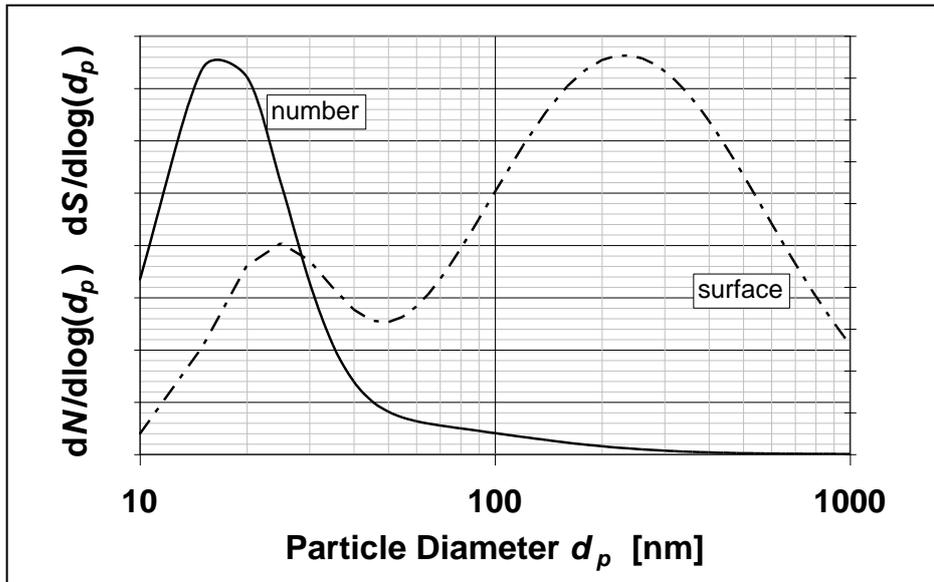


Figure 25: Example for conversion of a number size distribution into surface area size distribution

In order to calculate the total surface area concentration S_{total} within a certain particle size range, the surface area size distribution has to be integrated, i.e. the concentrations in the corresponding size channels added:

$$S_{total} = \sum_i \Delta N_i(d_{p,i}) \cdot \pi \cdot d_{p,i}^2 \quad (E4)$$

Equations E3 and E4 are only valid for spherical particles. The surface area calculation of non-spherical particles is not straightforward and often not possible at all. Instead it is commonly assumed that particles are spherical and the surface area can be calculated based on E3 and E4, using the electrical mobility diameter as particle diameter. Only very few models exist to estimate the surface area of non-spherical particles more accurately. For example, Lall and Friedlander (2006) developed a model that allows calculation of the surface area of chain-like agglomerates as one extreme example. In their model they

consider several assumptions concerning fractal dimension, homogeneity of primary particle sizes and the sintering state of the agglomerate. The difference between the surface area calculated with the Lall and Friedlander model and the simple model assuming spherical particles is usually within $\pm 30\%$.

The lung deposited surface area concentration can be calculated by multiplying the surface area concentration in each size channel with the corresponding lung deposition efficiency $\eta_L(d_{p,i})$:

$$S_{total} = \sum_i \Delta N_i(d_{p,i}) \cdot \eta_L(d_{p,i}) \cdot \pi \cdot d_{p,i}^2 \quad (E5)$$

The authors carried out numerous comparisons between NSAM data and lung deposited surface area, calculated from number size distribution measurements with SMPS or FMPS. The agreement was usually very good, when the particles were assumed to be spherical for the calculation from size distributions.

Number Concentration

The number concentration of particles is independent of particle size and thus weights all particles equally, unlike e.g. the mass concentration which mainly represents large particles because of the d_p^3 dependency. Due to the commonly large number but small surface area and negligible mass of ultrafine particles in ambient air, the number concentration can be considered as the most sensitive integral metric for assessing ultrafine particles in the atmosphere.

Condensation Particle Counter (CPC)

While larger particles are directly detectable by light scattering, ultrafine particles have sizes well below the wavelength of commonly used lasers and can therefore not easily be detected. In a Condensation Particle Counter (CPC), the particles are therefore artificially grown by condensation of a vapour (usually butanol or water) onto the particle surface (Stolzenburg and McMurry, 1991, Hering et al., 2005). The enlarged particles are then optically counted. The principle of the CPC dates back to 1888, when Aitken grew particles by condensation to sizes which he could detect with a magnifying glass (McMurry, 2000). In modern CPCs, particles are singly counted up to a certain concentration, which depends on the specific model and is usually in the range of 10^4 to 10^5 #/cm³. Some models automatically switch to a photometric detection mode above this concentration, which allows the extensions of the upper limit up to 10^7 #/cm³. Condensation particle counters are commercially available in a large variety, with their performance tailored for different application. While some CPCs aim at detecting very small particles (down to 2.5 nm), others can detect very high or very low concentrations, are rugged in design or handheld and battery-operated.

For the detection of solely ultrafine particles, i.e. particles below 100 nm, a pre-separator to remove all particles above 100 nm would be required. This is currently not available and the development of such a pre-separator not simple, because a commonly used inertial separator would introduce a significant pressure drop, which may interfere with the condensation process and overburden the CPC pump.

Number Size Distribution

Number size distributions of airborne particles provide a very versatile data set, because they offer insight into the particle size resolved concentrations which allows for an easy distinction of ultrafine (<100 nm) and coarser particles. A first estimate of potential particle sources may also be possible. Furthermore the number size distribution can be converted into surface area or volume and mass size distributions under several assumptions. The measurement of number size distributions of ultrafine particles, however, is more complex than the measurement of single integral concentration measures such as e.g. particle number concentrations. The most common method to measure number size distributions of submicron particles is by electrical mobility analysis.

Electrical Mobility Particle Sizers

Electrical mobility particle sizers mainly consist of three components:

- a particle charger to charge particles with known particle size dependent efficiency
- a mobility analyzer to classify particles according to their charge to size ratio, i.e. the electrical mobility
- a particle counting device

Commonly used particle chargers include a radioactive source (e.g. ^{85}Kr or ^{241}Am) to produce a bipolar ion atmosphere. Particles exposed to this atmosphere get charged due to collisions of particles with ions. The resulting charge distribution of the particles is nearly at equilibrium (Fuchs, 1963, Wiedensohler, 1988). Another way of charging the particles is by producing unipolar ions with a corona discharge. In order to avoid possible effects of pre-existing charges (Qi et al., 2009), two consecutive corona discharges with opposite polarity can be used. The knowledge about the charge to size ratio, i.e. the electrical mobility of the particles is exploited in the electrical mobility analyzer, where the particles are exposed to an electric field which is perpendicular to the flow direction. Commercial mobility analyzers include two concentric cylindrical electrodes, with the flow in longitudinal direction. Figure 26 shows a schematic of a differential mobility analyzer (DMA) as originally introduced by Pui and Liu (1974). This DMA is a concentric arrangement with an inner and an outer electrode. The aerosol is introduced into the DMA near the outer electrode and separated from the inner electrode by a clean sheath air flow. If a voltage is applied between the two electrodes, particles of one polarity move towards the inner electrode with a radial velocity that depends on their electrical mobility. The location where they hit the inner electrode is thus also a function of the electrical mobility. At the lower end of the inner electrode, a small flow is withdrawn through a slit. Particles in this flow are all within a narrow mobility bandwidth, which can be adjusted by means of the voltage applied to the DMA. The particles in this flow are counted with a counting device, usually a CPC. In some cases the current of the particles is measured with an electrometer to determine the particle concentration. The size of the particles in this flow can be calculated with the known charge of the particles and the following relationship between mobility Z_p and particle diameter d_p and number of elementary charges n :

$$Z_p = \frac{n \cdot e \cdot C_c(d_p)}{3\pi \cdot \eta \cdot d_p} \quad (\text{E6})$$

Where C_c is the Cunningham slip correction factor, which accounts for the fact that at these small particle sizes, the gas can no longer be assumed to be a continuum, e is the elementary charge and η is the viscosity of the gas. As equation E6 shows, the electrical mobility of particles not only depends on the particle size, but also on particle charge, which has to be taken into account for the data evaluation algorithm (e.g. Hoppel, 1978).

In electrical mobility particle sizers, the voltage applied to the DMA is altered to detect different particle sizes. In Differential (DMPS) and Sequential Mobility Particle Sizers (Grimm SMPS), the voltage is changed stepwise (Fissan et al., 1983), whereas in Scanning Mobility Particle Sizers (e.g. TSI SMPS model 3936), the voltage is continuously ramped (Wang and Flagan, 1990). Scanning Mobility Particle Sizers therefore achieve a higher time resolution of about 2 minutes to obtain one full size distribution, compared with around 4 minutes in case of the Sequential and around 10 minutes in case of the Differential Mobility Particle Sizer. The covered size ranges depend among others mainly on the choice of the DMA. The two main manufacturers of mobility particle sizers, TSI and Grimm, both offer two different DMAs. A short DMA (Grimm) or Nano DMA (TSI, Chen et al., 1998) that both cover a size range between approximately 2.5 and 100 nm, and a long DMA for particle sizes between approximately 10 nm and 1 μm , depending on the flow rate settings. The different SMPS systems and some homemade DMPS systems have been subject to a number of intercomparison studies (Dahmann et al., 2001, Helsper et al., 2008, Asbach et al., 2009a). In general an acceptable agreement between the instruments has been reported. Usually the sizing of the instruments was very comparable, but the measured concentrations showed some deviations.

Another instrument for the determination of number size distributions is the Fast Mobility Particle Sizer (FMPS, TSI model 3091) which offers a time resolution of 1 s. The size range covered by an FMPS is between 5.6 nm and 560 nm. The classifier in the FMPS is similar to a DMA, but particles enter the classifier near the inner electrode and are pushed towards the outer electrode, which consists of an array of electrometers. The classifier is operated at a fixed voltage, i.e. the location where a particle hits the outer electrode only depends on its electrical mobility. Each electrometer therefore represents a certain electrical mobility bandwidth. The currents measured with these electrometers are then used to determine the number size distribution. The FMPS offers a lower size resolution of 16 channels per decade than SMPSs (Grimm: 44 channels per decade, TSI: 64 channels per decade) and its accuracy has been reported to be lower than that of SMPSs (Asbach et al., 2009a, Jeong and Evans, 2009).

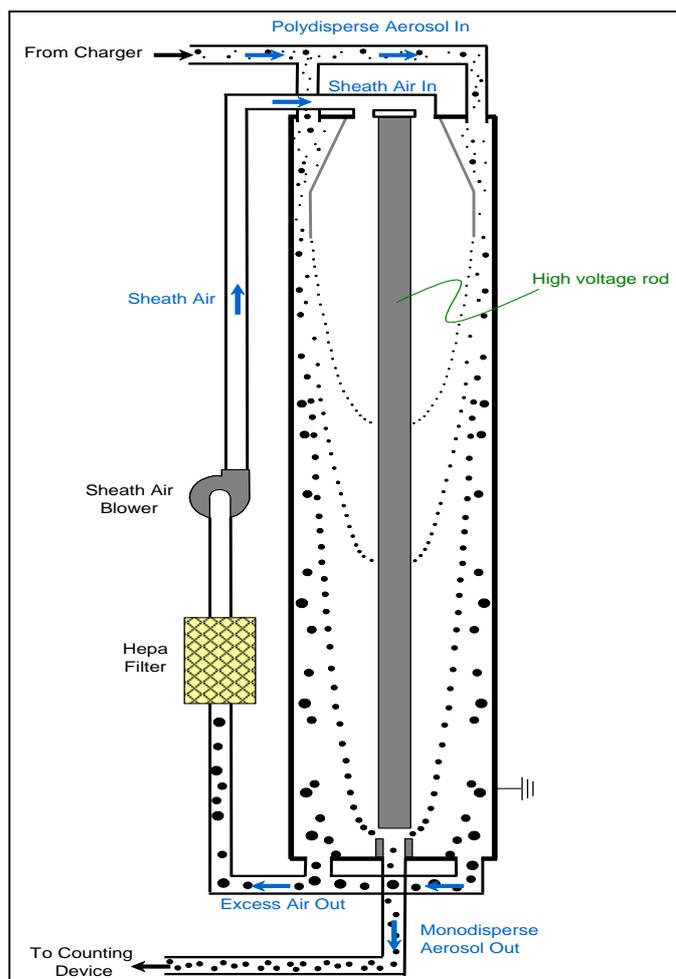


Figure 26: Schematic of a Differential Mobility Analyzer (DMA)

Chemical Compositions

“Quasi”-online chemical analysis of airborne particles can be differentiated into automated systems analysing bulk and single particle composition. The prior and more common analytical techniques are e.g. used in:

- In-liquid samplers with subsequent ion chromatography analysis like the commercially available Marga system (Khlystov et al., 1995), URG 9000 Ambient monitor or Georgia Tech PILS (Weber et al., 2001) or for water soluble organic carbon (Sullivan et al., 2004).
- Samplers on impaction plates like RP 5400 (elemental carbon) or RP 8400 S & N (sulphate and nitrate, Stolzenburg and Hering, 2000), on filters like SunSet Lab (elemental carbon, Bae et al., 2004) or directly in the aerosol phase like Thermo 5020SPA (sulphate). They evaporate the sample and chemically degrade the compounds of interest with e.g. a catalyst so that they can be analysed as gases. Examples of field applications of these instruments are e.g. given in a paper by Lin et al. (2008).
- Thermal desorption chemical ionization mass spectrometry (TDCI-MS) which goes back to Allen and Gould (1981) and Sinha et al. (1982).

In TDCI-MS aerosols are sampled through the aerodynamic lens, focused into a narrow beam, and transmitted into a detection chamber where they are impacted on a heated

surface (Figure 27). Vaporized aerosol species are ionized by electron impact and analyzed via mass spectrometry. Particle time-of-flight from a mechanical beam chopper to the vaporizer is measured to obtain chemically speciated size distributions. This figure shows the simplest version of the AMS. Other existing versions of the AMS which utilize several important instrumental modifications and developments that are not shown in this schematic are discussed in detail in Canagaratna et al. (2007).

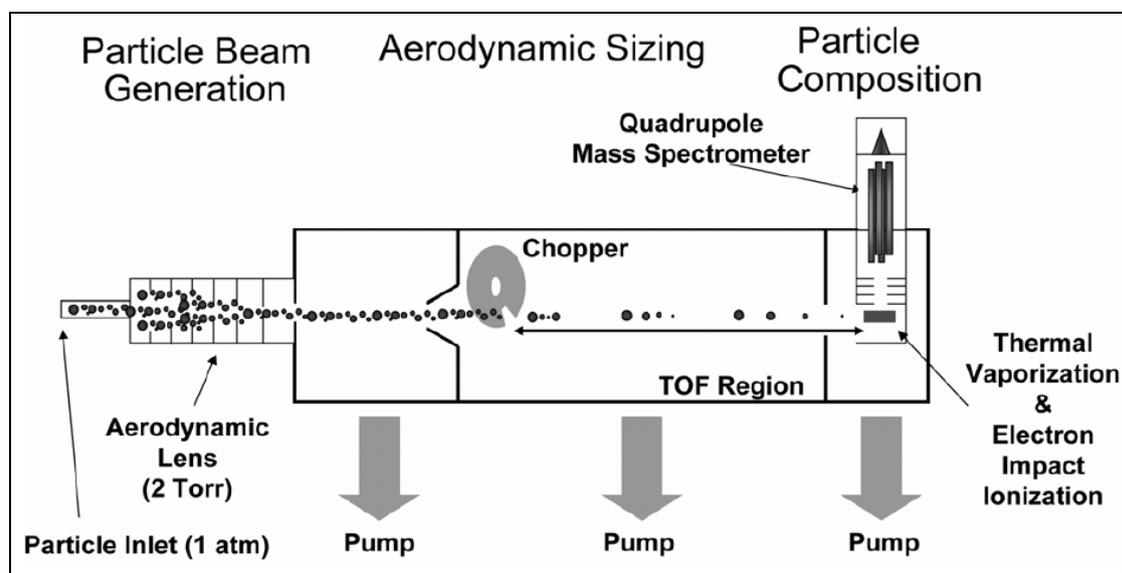


Figure 27: Schematic of an Aerodyne Aerosol Mass Spectrometer (AMS, adapted from Canagaratna et al., 2007)

The only online single particle analysis is the so called real-time single-particle mass spectrometry (RTSP-MS). The principle of this method is the ionization of the particles directly in the mass spectrometer by impacting them on e.g. heated rhenium (Lassiter & Moen, 1974). The resulting ions were then separated in a magnetic sector mass analyzer and detected with an electron multiplier. Recent reviews on this technique can be found in Noble and Prather (2000) and Sullivan and Prather (2005).

Both aerosol mass spectrometer techniques (RTSP-MS and TDCI-MS) have shown that they can analyse particles down to a few 10 nm while the other above mentioned online techniques for chemical analysis besides those for carbon may be limited due to their lower detection limit.

ROS-Potential

For an automated and continuous monitoring of non-specific, particle-bound ROS-formation in ambient aerosols a dichlorofluorescein (DCFH) based fluorescence-method was developed by Venkatachari and Hopke (2008). This analysis system is based on the collection of ambient particles using a particle-into-liquid sampler (PILS) (Weber et al. 2001, Orsini et al. 2003) and analysis of the slurry for organic peroxides mainly using DCFH fluorescence spectrometry.

More in detail the ambient aerosol sample is mixed with a smaller, turbulent flow of steam, in order to initiate a supersaturation of water vapor in which particles grow into

droplets large enough to be collected by impaction. In addition dichlorofluorescein is added and finally a suspension containing the aerosol species and the fluorescence substance is produced. This slurry is passing afterwards the so called membrane reactor, where a radical electron acceptor in form of horseradish peroxidase is catalysing the oxidation of peroxides on the particle surfaces (Hung & Wang 2001, Venkatachari et al. 2005, 2007). Hereby generated peroxide radicals oxidise the deacetylated 2'-7'- dichlorodihydrofluorescein diacetate (DCFH) to its fluorescent product, 2'-7'- dichlorodihydrofluorescein (DCF) which is subsequently detected after an adequate incubation time on excitation and emission wavelengths of 485 and 530 nm, respectively, via fluorescence spectrometry (Venkatachari and Hopke, 2008).

In summary this system is a sophisticated method imaging the ROS-generation potential of atmospheric particle and their on surface-bound organic peroxides, down to nanomolar levels. It has so far not been tested for ultrafine particles.

2.3.4 Typical Physical and Chemical Particle Characteristics

2.3.4.1 Particle Number Concentrations

Measurements of UFP in various environments have shown distinct differences between site types. UFP number concentrations e.g. may range from a few hundred particles per cubic centimeter ($\#/cm^3$) at regional background sites and in remote regions to more than $10^5 \#/cm^3$ at heavy polluted traffic sites. Concentrations in the urban background are generally around $10\text{-}20 * 10^4 \#/cm^3$ while concentrations are even lower indoors $2\text{-}8 * 10^3 \#/cm^3$ when no specific indoor sources are present. Indoor sources besides smoking are e.g. cooking, candles, vacuum cleaning and other electric motors. Figure 10 gives an example of particle number size distributions for different ambient site types.

The size distributions shown in Figure 28 can be assumed to be relatively stable. This is generally true for particle concentrations $< 10^6 \#/cm^3$. Particle number size distributions as e.g. determined in the exhaust of combustion engines of up to $10^{14} \#/cm^3$ are not stable due to very fast coagulation processes. This concentration behaviour with regard to number concentrations leads to the unique effect that no “real” exposure values $> 10^6 \#/cm^3$ exists in ambient environments unless somebody is very close to a source.

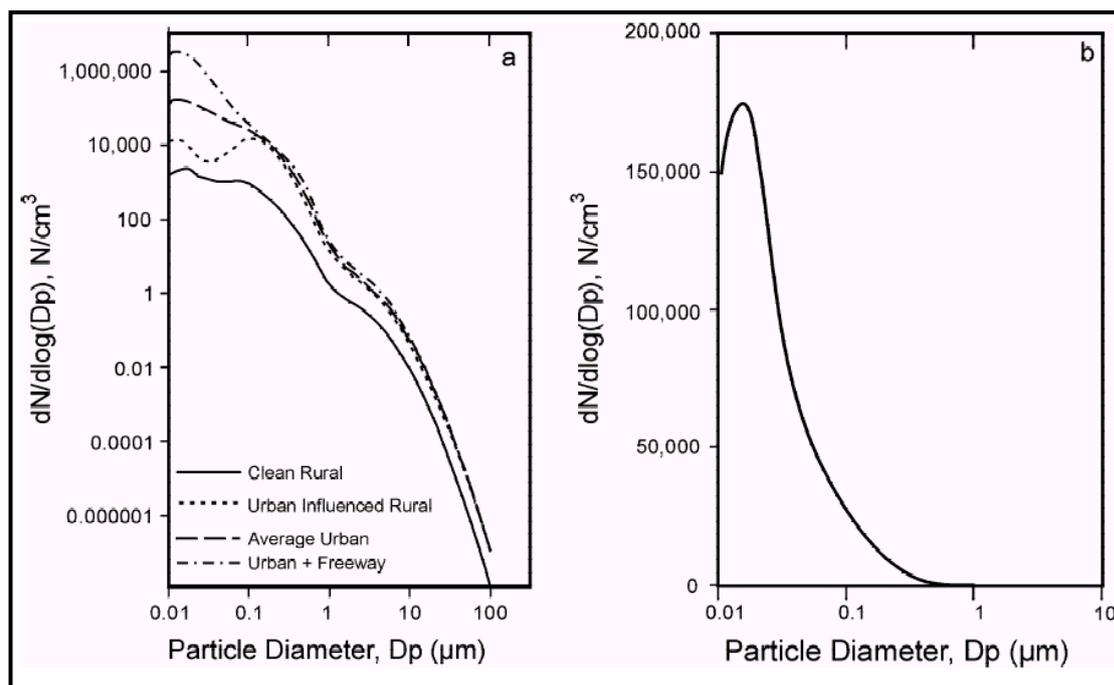


Figure 28: Left: Particle number size distributions for various site types in a log-log plot, right side: average urban of left side with linear concentration axis (Whitby and Sverdrup, 1980)

Further detailed information on UFP particle concentrations and particle size distributions especially in relation to exposure are given in section 7 of this book (Lanki et al., 2006).

2.3.4.2 Particle Surface Area Concentrations

Particle surface area concentrations in ambient air have so far only scarcely been measured. Recently Ntziachristos et al. (2007) reported NSAM (TSI Inc.) measurements at several locations within Los Angeles, e.g. next to a freeway and about 100 m downwind. Mean surface area concentrations determined near the freeway were $\sim 150 \mu\text{m}^2/\text{cm}^3$ while being $\sim 100 \mu\text{m}^2/\text{cm}^3$ for the same period in Feb.-March 2006 100 m downwind. Surface area concentrations determined in a residential area 1 month prior to the freeway period were on average around $50\text{-}70 \mu\text{m}^2/\text{cm}^3$. Measurements conducted in March 2008 by the authors at an urban background site in the Ruhr-Area showed similar results with average values around $30\text{-}60 \mu\text{m}^2/\text{cm}^3$ but quite often peak values of $100 \mu\text{m}^2/\text{cm}^3$ were exceeded during the course of the day.

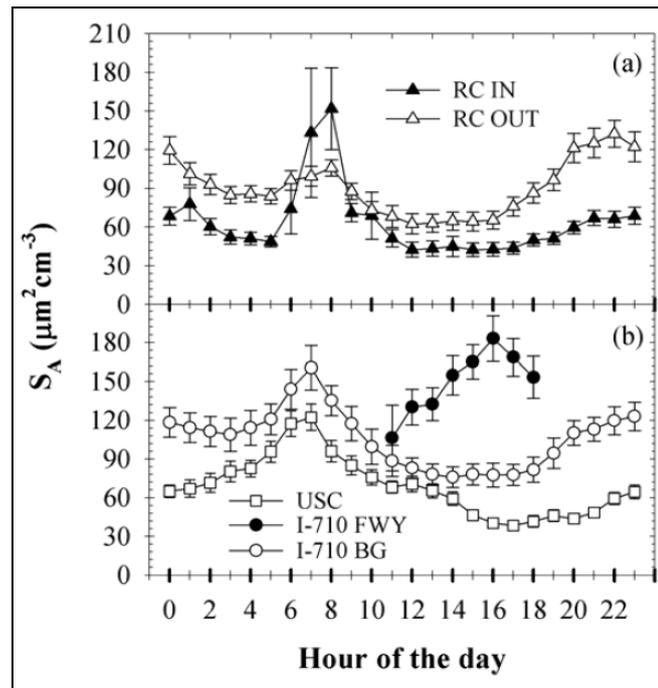


Figure 29: Temporal variation of the particle surface concentration measured with the NSAM at (a) Indoors (IN) and outdoors (OUT) of a retirement community and, (b) an urban background site (USC) and a proximal (FWY) and a distant (BG) site to the I-710 freeway in Los Angeles (from Ntziachristos et al. 2007).

Figure 29 shows the diurnal variations of particle surface area concentrations at different site types in Los Angeles. For all cases, from close to the road to even indoors, a maximum during the morning rush hour can be identified. It also can be noted that the lowest concentrations of the particle surface area concentrations are during the early afternoon hours which may be attributable to sea wind conditions and higher mixing ratios.

In view of exposure it also is important to assess how much of the surface area is attributable to the very fine particles e.g. 16-40 nm as done by Ntziachristos et al. (2007). They found by using particle size distribution measurements (SMPS) and comparing those to the NSAM measurements, that at both sites about 10-20% can be attributed to these particles. A similar analysis of the data at the urban background site in the Ruhr-Area by the authors showed that 12% (8%-17% quartile range) of the NSAM surface area can be attributed to a similar particle size range (14-40 nm). The data above relate to NSAM measurements which determines the fraction of the particle surface area concentration which would be deposited when inhaled.

2.3.4.3 Chemical Composition of Ultrafine Particles

The main chemical constituents of ultrafine particles are carbonaceous materials stemming from combustion processes such as EC and OC and secondary particle components like sulphate, nitrate, ammonia and some organic carbon. The chemical composition of UFP particles (< 100 nm) at a rural background site Melpitz (near

Leipzig, Germany) in November 2003 was $1 \mu\text{g}/\text{m}^3$ EC, $0.2 \mu\text{g}/\text{m}^3$ OC, ca. $0.1 \mu\text{g}/\text{m}^3$ sulphate, $0.3 \mu\text{g}/\text{m}^3$ not explained as well as some minor components such as ammonium and nitrate (Birmili, 2006). The total UFP mass concentration was $2 \mu\text{g}/\text{m}^3$. The average mass concentration at the super site in Los Angeles in June to July 2006 (Ning et al, 2007) was about a factor two higher than that in Melpitz. The highest mass contribution to UFP in this case was organic matter, followed by sulphate and nitrate. EC contributions were nearly negligible all days (Figure 30). Figure 30 also show that the daily variance of the UFP chemical composition was relatively low.

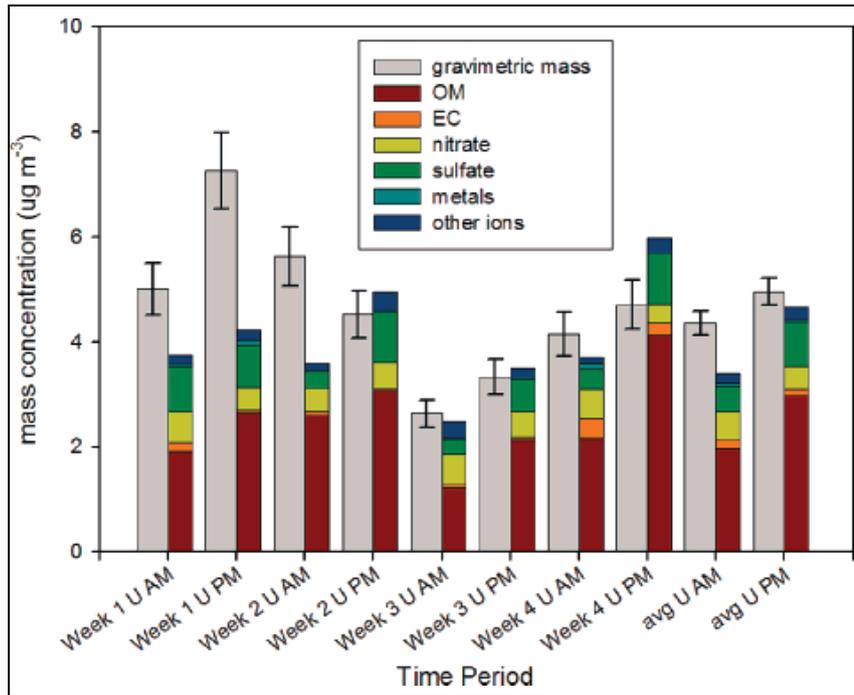


Figure 30: Mass concentration and chemical of the UFP fraction (<math><0.18 \mu\text{m}</math> diameter) in Los Angeles June & July 2006 (Ning et al., 2007)

Other, earlier studies have indicated that about 50-70% of UFP mass consists of carbonaceous material (Berner et al., 1996, Hughes et al., 1998). The above and other measurements indicate that carbonaceous matter, especially organic carbon, is dominating the ultrafine particle size fraction with elemental carbon, sulphate, nitrate and ammonium also representing a significant mass fraction.

Anyhow, so far only few chemical composition measurements for UFP have been made and even less representing a long time series. This is especially due to the low mass concentrations in the UFP size class and hence difficulties in analysis.

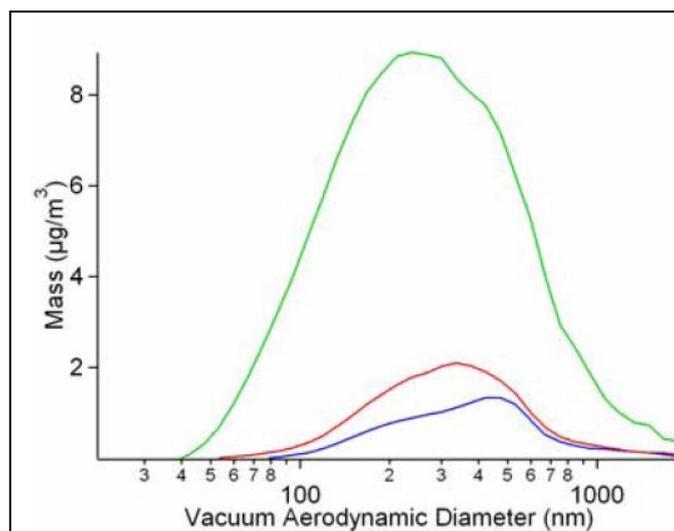


Figure 31: Mass size distribution of organic (green), sulphate (red), and nitrate (blue) components in London Oct. 2006. The C-ToF-AMS (Aerodyne) measures particle size aerodynamically in the free molecular regime, delivering the so-called vacuum aerodynamic diameter (Dall'Osto et al., 2009)

One analytical method promising some improvements for UFP chemical composition is the aerosol mass spectrometer (AMS). This method allows the nearly online size resolved chemical analysis of airborne particles. Figure 31 is giving one example for the capability of AMS to detect size resolved particle compositions (Dall'Osto et al., 2009).

2.3.5 References

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2 Fine Dust Measurement Techniques and Strategies for Exposure

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2.4 Nanoparticles and Exposure: Measurement Technologies and Strategies (Kuhlbusch et al., 2008a)

So far no personal exposure related measurement device to determine nanoparticle and ultrafine particle exposure has been employed in consistent exposure assessment approaches. A personal sampler based on thermophoretic sampling was recently developed and tested by Azong-Wara et al. (2009, 2011). First studies related to ultrafine particle exposure were conducted by using a portable condensation particle counter (TSI 3007) by e.g. Vinzents et al. (2005), Bräuner et al. (2007), Diapouli (2007). Still, the device used in these settings does not (a) differentiate ambient nanoscale particles from engineered nanoparticles and (b) cannot be used as a personal sampler due to the use of a liquid. Therefore specific approaches and measurement strategies are pursued e.g. at workplaces to allow the identification of engineered nanoparticles and their possible sources. The following manuscript by Kuhlbusch et al. (2008a), published in a book edited by Linkov and Steevens "Nanomaterials: Risks and Benefits", gives an example on a measurement strategy to determine possible exposure.

Abstract

Assessments of nanoparticle exposure are needed to enable risk assessments which are needed to achieve a sustainable development of nanotechnology including public perception. Therefore an overview of measurement techniques, needed data quality, comparability, and measurement strategies is given. Additionally some results of exposure related studies are summarized. Overall it is demonstrated that an integrated approach towards nanoparticle exposure assessments in workplaces, but also in the environment is needed, despite the current published results indicating mainly release of nanoparticle agglomerates in the size range larger than 100 nm.

2.4.1 Introduction

Nanoparticles and nanoobjects, intentionally produced particles of nanoscale in three or two dimensions, have specific properties possibly altering the (eco-)toxicological potential when compared to larger particles or the corresponding bulk material. The detailed assessment of this potential risk is a prerequisite to accomplish sustainable nanotechnology since it directly influences the public perception. The risk is generally a function of potential hazard and exposure. The importance of the latter is given by a) that no risk exists if no exposure, b) the dose, leading to possible health effects, is directly linked to the exposure, and c) correct exposure determination is also of importance for e.g. epidemiological studies. To assess exposure it is also necessary to clarify the areas (e.g. workplace, environment), subjects (e.g. humans, animals, ecosystems) and exposure media (air, liquid, solid) of interest.

This paper focuses on the human exposure mainly in workplace environments since highest exposure can be expected in these areas. Also, measurement technologies and strategies can be evaluated and tested in workplace environments since possible sources and hence particle material is known and can be differentiated from ambient nanoscale particles.

The currently discussed main exposure route of nanoparticles is via the airborne state by inhalation. Other discussed routes of exposure such as via the skin or gastrointestinal

tract possibly leading to an uptake are currently seen as of minor importance (Butz et al., 2007) but still have to be investigated.

Two major pieces of information are necessary for the assessment of exposure and possible nanoparticle implications:

- the exposure leading to a dose,
- the hazard, influenced by the particle properties.

Hence, assessments of exposure to nanoparticles have a twofold task. One task is the general determination of an exposure and to quantify the ‘relevant’ aerosol property. The second task is the characterization of the nanoparticle properties since these may have been influenced or changed during the transport period after release. Any changes in these particle properties may have a significant influence on the possible hazard of nanoparticles. Spatial and time resolution of the measurements can therefore play a crucial role in the exposure determination and its evaluation.

This background puts certain demands onto the measurement techniques for airborne nanoparticles as well as the measurement strategies.

2.4.2 Measurement Techniques

Basically, various physical and/or chemical properties of nanoparticles and aerosols can be determined with especially particle size and concentration being physical properties of importance in the case of nanoparticles (<100 nm in 3 dimensions) and nanoobjects (<100 nm in 2 dimensions). For a decision on the instrumentation to be used for exposure measurement the scheme shown in Figure 32 was introduced in Borm et al. (2006):

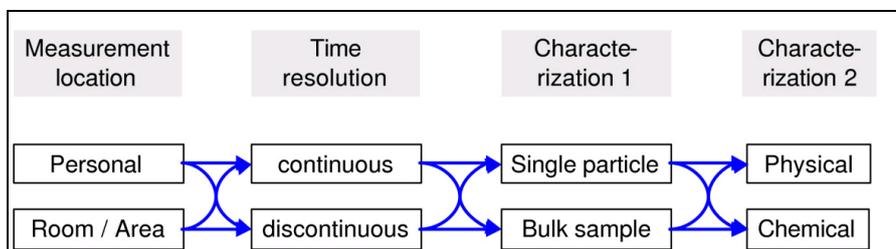


Figure 32: Scheme of particle characterization for exposure measurements (adapted from Borm et al., 2006)

This scheme indicates the different steps of possibilities when choosing the instrumentation for exposure assessment. Certainly, the ideal sampling for exposure assessment would be a personal sampler measuring and reporting continuously the physical and chemical characteristics of all single particles (agglomerates or primary particles) as well as their concentrations entering the measurement system. Since this is not possible, decisions have to be made based on the task to perform and the corresponding availability of adequate measurement technology. If e.g. the question is limited to whether primary particles of a specific nanoparticle product are present in the work area it may be sufficient to use a device determining particle number concentration or particle number size distributions. If, on the other hand, exposure should be determined in the framework of epidemiological studies, possibly certain particle properties such as lung deposited particle surface area become important. Hence, currently an array of

stationary instruments and measurement technologies are employed to study exposure and possible effects of nanoparticles.

To assess particle properties such as morphology or chemical composition, mainly offline methods, namely single particle analysis following particle deposition, commonly by electrostatic precipitation (Dixkens et al., 1999), are usually required. The analytical techniques may comprise electron microscopic (SEM, TEM), atomic force microscopic analysis for physical properties and energy-dispersive x-ray spectroscopy (EDX) for single particle or total reflection x-ray fluorescence spectrometry (TXRF) (John et al., 2000) for total chemical analysis. While the determination of particle concentrations and size distributions from the sample deposits is generally possible by electron microscopy, this method is very limited in its concentration range and also very time consuming.

Technically mature online instrumentation for the determination of various aerosol properties is commercially available. The aerosol properties of interest include particle number and surface area concentration and their size distributions. Mass concentration determination is of minor importance for nanoparticle detection due to the low sensitivity and low mass involved. Total particle number concentrations are commonly determined with a Condensation Particle Counter (CPC), sometimes also called Condensation Nucleus Counter (CNC). In a CPC, particles get exposed to an atmosphere supersaturated with vapour of its working fluid, commonly butanol, isopropyl alcohol or water. The vapour condenses on the particle surface and causes the particles to grow to a size that can scatter a laser beam downstream of the condensation chamber. The number of impulses from the scattering of the laser beam is counted in order to determine the particle number concentration. (McMurry, 2000). The lower detection limit for these devices reaches down to 3 nm (Hermann et al., 2007) and below.

There are no instruments available that can determine airborne concentrations of geometric particle surface area. In electrical diffusion chargers such as the LQ1DC (Matter Engineering) particles are exposed to a unipolar ionic atmosphere and ions are attached to the particle surface due to Brownian diffusion. The current, measured upon deposition of the charged particles, is proportional to the so-called Fuchs surface area (H. Jung and D. Kittelson, 2005), i.e. the surface area of particles available for ion attachment. This “surface area” is proportional to $dp^{1.39}$ and therefore smaller than the geometric surface area, being proportional to dp^2 . Another instrument, called Nanoparticle Surface Area Monitor, NSAM (TSI) also uses an electrical diffusion charger but manipulates the size distribution such that the measured current is proportional to the geometric particle surface area concentration that gets deposited in either the alveolar or tracheobronchial region of the lung (Fissan et al., 2007, Shin et al., 2007).

Number size distributions of airborne particles with diameters down to a few nanometers can be determined using electrical mobility analysis. Particles are initially brought to a known charge distribution (equilibrium) in a neutralizer which can be based on radioactive decay or corona discharge. The particles then enter a Differential Mobility Analyzer (DMA), a concentric, cylindrical set up, near the outer electrode. Charged particles are deflected in the electric field between the two electrodes. The electrical mobility is a function of particle size and charge. Particles of high electric mobility, i.e. small and/or highly charged particles move faster towards the inner electrode than particles of low electrical mobility, i.e. the location where they hit the inner electrode is a function of their electrical mobility. At the other end of the inner electrode, particles of

certain mobility are withdrawn through an outlet slit. The withdrawn mobility is determined by the voltage applied to the electrodes. In a Scanning Mobility Particle Sizer, SMPS (Wang and Flagan, 1990), the voltage is continuously ramped and the particle concentration in the sample flow through the slit measured with a CPC. An algorithm is used to relate the measured concentrations to the corresponding electrical mobilities and along with the known charge distribution to determine the number size distribution as a function of electrical mobility diameter. If the relationship between particle mobility diameter and particle surface area is known, the number size distribution can easily be converted into surface area size distribution. Integration over a desired size range can also provide total number or surface area concentration.

Another, for work related exposure interesting measurement technique is also based on the electrical mobility of charged particles. It is implemented in the Fast Mobility Particle Sizer (FMPS, TSI) which is based on work conducted by Mirme et al. (1984). The main difference to the SMPS described above is that the particle detection is based on an array of 22 electrometers, placed along the outer electrode in a DMA. Simultaneous detection of particles within the entire size range covered (5.6- 560 nm) allows for a time of resolution of one second. For comparison, a comparable size scan by an SMPS would take at least two minutes, because the different sizes are measured sequentially. The downside of the FMPS is the lower size resolution, 16 channels per decade, compared to e.g. possible 64 channels per decade for the SMPS. Nevertheless, the FMPS may be preferred if size distribution have to be determined in work areas with quickly varying work processes and therefore size distribution fluctuations shorter than the time resolution of the SMPS. Extended information on measurement techniques may also be found in Kuhlbusch et al. (2008a⁴).

2.4.3 Comparability and Data Quality

Measurements to assess exposure of e.g. humans should be comparable to actually allow for comparisons between different work environments, different work processes, to enable the evaluation of exposure reduction measures etc. Still, the comparability between different instruments, instruments from different manufacturers, or even between two similar instruments but two different users is not always sufficient. Comparison studies as e.g. reported by Dahman et al. (2001) clearly showed the differences between various instruments and the need of 'understanding' the equipment.

⁴ of reference list 2.4.7

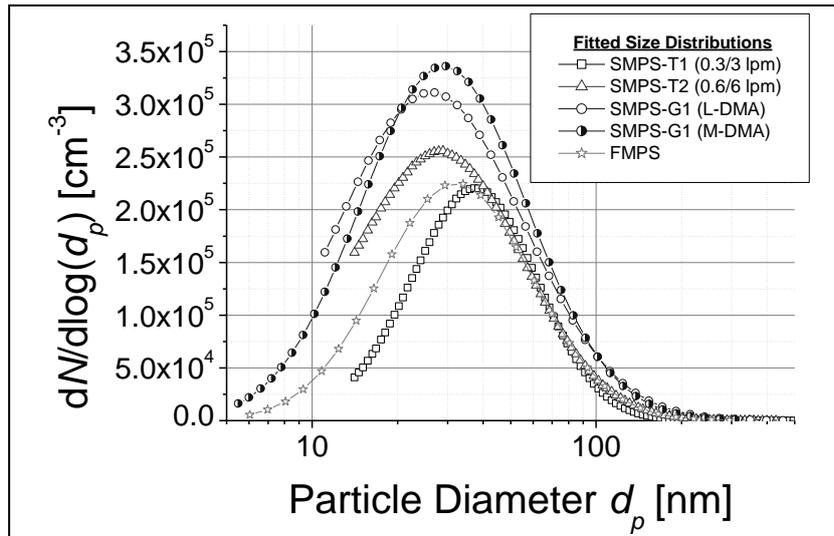


Figure 33: Example of NaCl particle size distribution determined with various SMPS (different providers, volume flow, and type of DMA) and FMPS (adapted from Asbach et al., 2007)

An example of a comparison for sodium chloride test particles is given in Figure 33. This figure clearly shows the comparability of a) number concentration < 100 nm, b) modal diameter, and c) width of the size distribution. This comparability for the ideal test condition is general in the range of around 40% and better. More detailed analysis of the comparability will soon be published.

Nevertheless, it has to be stated that comparability and reproducibility are currently more in an infant state and that standardization as well as good understanding of the measurement devices are still needed.

2.4.4 Measurement Strategies

The difficulties in the determination of nanoparticles in air, water or soils are manifold. In all three matrices the problems of

identification (and differentiation from natural nanoscale particles)

characterization

and quantification

of the specific nanoparticles have to be overcome. In all cases single particle analyses are necessary to enable identification. The problem may be reduced if nanoparticles with very specific and robust properties or specific chemical components, not or only rarely present in the corresponding matrix, are used. Nevertheless, in some cases clear differentiation of the source of the nanoscale particle, natural or manmade, may not be possible as e.g. for iron particles.

Another problem, difficult to be covered with “normal” measurement methods is the alteration of surfaces of nanoparticles. Surfaces of nanoparticles are altered after the production to achieve specific particle properties and to enhance e.g. dispersion of nanoparticles during certain production steps. These surface properties are currently discussed to be of possible importance for toxicological effects. No study, to the

knowledge of the authors, has so far been conducted to investigate whether particle surface properties are stable after e.g. accidental release to the environment.

The above two paragraphs briefly explain the difficulties in the measurements and hence in the setup of measurement strategies for nanoparticles exposure. While the latter case (persistence of particle surface properties) is only of interest if particles are released we may first focus on the task of the general assessment whether nanoparticle exposure exists or not. This is of importance since no risk exists when no exposure is given.

To allow for first tests and evaluations of measurement strategies as well as measurement techniques we first focus on exposure related measurements in work areas since there we know which kind of nanoparticle to determine and we can expect the highest exposure concentrations compared to e.g. in the environment.

Two of the first questions to tackle in the case of exposure measurements are the questions of measurement metric and necessary lower detection limit. Table 6 gives an overview of possible particle metrics and concentration ranges of interest.

Table 6: Particle metrics and concentrations of interest for exposure measurements

Number (N/cm ³)	Surface (µm ² /cm ³)
< 1,000	< 0.01
1,000-100,000	10-1,000
> 100,000	> 1,000

The concentrations given in the first row of Table 6 (< 1,000 N/cm³ or < 0.01 µm²/cm³) are in our view concentration ranges with no or only little effects unless particle specific toxicity as e.g. for asbestos is given. The latter may be assessed in toxicological screening tests. In those cases where such low limits of detection are needed only single particle analysis by microscopes coupled with e.g. EDX can currently be employed as is already used for asbestos. Due to this detection technique it is normally possible to discriminate environmental nanoscale particles from engineered ones. Nevertheless, this method is very time consuming, expensive, and can hardly be employed in routine monitoring.

In cases of particle concentrations exceeding 100,000 N/cm³ (or 1,000 µm²/µm³) a source has to be close to the measurement point and should be identifiable by simple aerial particle number concentration measurements with a CPC.

The most difficult and maybe most important case for exposure assessments is the intermediate concentration range of 1,000-100,000 N/cm³ (or 10-1,000 µm²/cm³). This concentration range covers the usual ambient particle number concentrations which may range from a few hundreds particles per cm³ in clean air conditions, e.g. mountainous regions or sea sides, around 10,000-30,000 N/cm³ in urban background situations, up to possibly 100,000 N/cm³ close to high traffic areas (e. g. Dingenen et al., 2004). Hence, to assess nanoparticle exposure in this concentration range neither microscopic single particle analysis nor simple number or surface area concentration measurements are really an option due to potentially high background contributions and variability. A measurement strategy is therefore needed to overcome this problem but still to allow for a relatively practicable assessment of possible nanoparticle exposure.

The following approach briefly summarized here is described in detail in Kuhlbusch et al. (2008b⁴) and is based on the strategy used in studies at several carbon black production facilities (Kuhlbusch et al., 2004 & 2006).

One of the basic assumptions of this strategy is that the intrusion of ambient particles into a work area can be calculated by a constant, but size dependent ‘penetration factor’. This factor is determined by e.g. measuring with an SMPS concurrently at a comparison site and in the work area when no work activity is taking place. By calculating the ratio of the two measured size distributions a size dependent ‘penetration factor’ is derived.

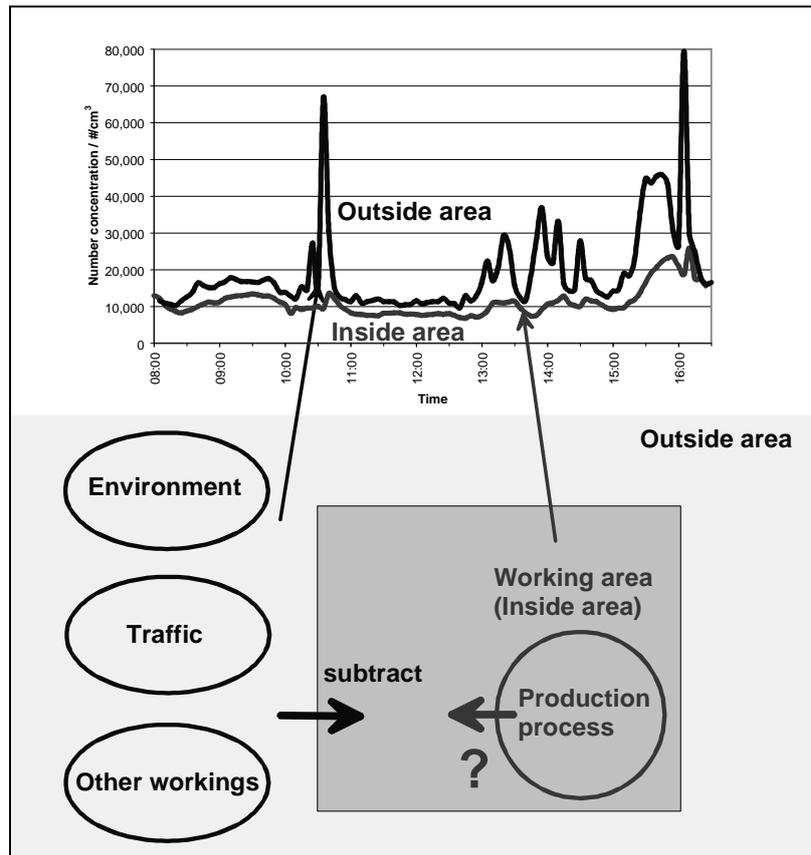


Figure 34: Penetration of particles from outside area into the work area (adapted from Kuhlbusch et al., 2008b⁴)

The same measurements are performed when the work activity of interest is ongoing. Now, the measurements conducted in the work area show the particle size distribution with work activity. The size distribution to be present in the work area when no work activity would have been ongoing can be calculated based on the measurement at the comparison site and the ‘penetration factor’ determined before. The comparison of the two values, in this example particle size distribution, show a) whether nanoscale particles were released by the work activity and b) the size distribution emitted by the process. So far, this value is only indicative, whether nanoparticles may have been released. No nanoparticles have been released by the process if no increased particle concentration is determined. If increased particle concentrations are determined further investigations based on e.g. single particle analysis or time of release (does it correlate with the specific work activity?) are needed. This basic idea is sketched in Figure 34.

2.4.5 Results From Previous Studies

Several studies have now been conducted to assess possible exposure to particles below 100 nm. Most of these studies however deal with unintentionally produced particles such as welding and soldering fumes (e.g. Brouwer et al., 2004). Only a limited number of measurements have so far been conducted determining possible exposure to nanoparticles or nanoobjects in real nanotechnology work areas (Kuhlbusch et al. 2004, 2006, 2008a).

The results of the measurements conducted at three different industrial plants are summarized in Table 7.

Table 7: Summary of particle release below 100 nm at 3 different carbon black plants (adapted from Kuhlbusch and Fissan, 2006)

	Observed increases	Likely origin
Plant 1 Reactor Pelletizer Bagging	No No No	
Plant 2 Reactor Pelletizer Bagging	Yes Yes Yes	Nearby traffic Nearby traffic Propane fork lifts
Plant 3 Reactor Pelletizer Bagging	Yes Yes Yes	condensed oil vapors Flue gas – leak Diesel fork lifts, gas heater

Measurements to determine possible nanoparticle exposure were conducted at overall nine sites. Six of these sites showed elevated concentrations of nanoscale particles. In two cases this could be attributed to nearby traffic by e.g. analysing the wind direction dependency, in further two cases other sources than the nanoparticle production were the reason for the elevated concentrations, namely forklifts and gas heaters. Finally in two cases the reason for the higher concentrations was found to be related with the production facility. One time it was condensed oil vapours coming from maintenance work. These particles were nanoscale particles but no nanoparticles. In the last case a leak in the production facility was the reason for the very high concentrations. In this case true nanoparticles were present.

Another study which investigated the possible exposure to carbon nano tubes was conducted by Maynard et al. (2004). This study showed that there was no evidence of an increase in particle number concentration during the handling of carbon nanotube material. Rather, in all cases, the field number concentration decreased during the periods when the material was handled. But they also state that laboratory investigation showed that nanoparticles, or better here nanoobjects can become airborne when vigorously agitated.

Yeganeh et al. (2008) on the other hand reported significant increases of sub-100 nm particle number concentrations during the handling of carbonaceous nanomaterials including fullerenes. Anyhow, no quantification of release was done due to the highly variable background and hence the actual contribution of engineered nanoparticles to the determined number concentration is not known.

Wake et al. (2002) investigated nanoscale particles at different workplaces in various industries, including workplaces with nanoparticle production. They conclude that there was no evidence of significant ultrafine particle release in unagglomerated form when working with nanoscale powders. In contrast, processes involving heat, such as welding, released high levels of nanoscale particles.

The examples above show that nearly no or little exposure was determined in the field. This may well be due to the materials being produced and the safe work conditions at the investigated workplaces. It now becomes necessary to get comparable measurement technologies and measurement strategies in place to investigate the various workplaces in nanoparticle production facilities. Exposure to nanoparticles and nanoobjects has to be carefully screened to ensure a safe and sustainable development of this technology.

2.4.6 Summary

An array of measurement techniques for the determination of airborne nanoscale particles exists. Still, the demand on these techniques, namely measurements of all possibly health relevant particle parameters online as personal dose (personal sampler) is extremely high and currently not fulfilled. Only few devices exist for personal sampling (only offline and use of imaging techniques) and measurements of the dose. Especially the latter may vary significantly since particle uptake is size dependent (even within the size range below 100 nm) but also dependent of physiochemical characteristics such as solubility and hygroscopicity.

The main techniques currently used are stationary systems such as the SMPS. First assessments of comparability and reproducibility of size distribution measurement devices show discrepancies in e.g. number concentrations, modal size in the range of 30%-40%. Uncertainties can be expected to be larger since no protocols on data correction (e.g. diffusion losses, multiple charge correction), and only limited information on influence of state of agglomeration on the measurement results exist.

Measurement strategies are currently needed beside reproducible and comparable measurement techniques to allow for a first assessment if product nanoparticles / nanoobjects were released. The measurement strategy is crucial to allow for an assessment of uncertainty and of lower detection limits.

Despite all the above problems, which still have to be dealt with, first measurement results indicate that

- a) measurements are currently feasible,
- b) monitoring on a routine base can also be done, and
- c) no or only limited release of nanoscale particles or objects has been detected.

Nevertheless, it has to be kept in mind that the information base for exposure assessment is currently built on an uncertain and limited data base which has to be improved in size, comparability, reproducibility. This can be achieved by working on the feasibility for routine assessments, development of reliable measurement techniques, standardization of measurement techniques, measurement strategies, and implementation of screening / monitoring of nanoscale particles in sensitive work areas.

Future challenges are currently especially seen in the detection of product nanoparticles in the environment, which we believe to be currently nearly impossible for airborne particles.

2.4.7 References

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3 MEASUREMENTS RELATED TO THE IDENTIFICATION OF PARTICLE SOURCES

In view of the legal background and for the safety of population and workers it is important to derive information on the sources of fine dust and nanoparticles. This information can be used to avoid emission and to plan cost effective abatement strategies as e.g. necessary when limit values are exceeded. The need of source apportionment was further extended due to a new EU regulation in 2008 (Directive, 2008). This directive states in § 15:

(15) Contributions from natural sources can be assessed but cannot be controlled. Therefore, where natural contributions to pollutants in ambient air can be determined with sufficient certainty, and where exceedances are due in whole or in part to these natural contributions, these may, under the conditions laid down in this Directive, be subtracted when assessing compliance with air quality limit values. Contributions to exceedances of particulate matter PM₁₀ limit values attributable to winter-sanding or -salting of roads may also be subtracted when assessing compliance with air quality limit values provided that reasonable measures have been taken to lower concentrations. (cited Directive, 2008)

Basis of source apportionment investigations are mainly measurements at sources and at the location of interest. The prior measurements can be used to develop emission inventories while the latter can be employed for statistical analysis in view of source identification and quantification.

Two main source apportionment approaches can be differentiated.

1. Forward directed methods which base on emission inventories and the modelling of the dispersion and reactions during atmospheric transport to derive information on air quality (dispersion modelling).
2. Backward directed methods using measurement data from a specific location (e.g. in workplaces or ambient measurement sites) along with auxiliary information on e.g. workplace activities or meteorological data to determine the source and source strength or rather the contribution of a source to a specific exposure situation.

Examples for the first group are e.g. workplace dispersion models developed and employed by Asbach et al. (2007) or Kuhlbusch et al. (2008c, 2009c) for workplaces or dispersion models as e.g. used in the legal framework of approval procedures for new industrial plants in Germany. Extension to the dimple dispersion models are so called chemical transport models such as EURAD, LOTOS-EUROS or CHIMERE. These models normally run in nested modes to reduce the need of computational power but still allow the modelling in high resolutions. The EURAD model (www.eurad.uni-koeln.de/)

e. g. models first on the European scale with a grid resolution of 125x125 km² subsequently nesting in grid sizes of 25x25 km², 5x5 km² down to 1x1 km².

Sources and their contributions to a local exposure setting can then be studied by altering the source strength or even switching off certain source types and activities.

The main analytical tools used in the second group are the source-receptor models. Those models assume covariance of different pollutants and chemical constituents of particles which in sum represent the contribution of the different sources and source

3 Measurements related to the identification of particle sources

activities. Several sources can be differentiated and their contribution to the ambient burden quantified if enough data are available, e.g. a one year time series of dust and chemical composition data.

The factor and principal component analyses are the most frequently used statistical tools in the identification and quantification of sources of ambient fine particles. This tool was further improved in recent years to the so called positive matrix factor analysis (PMF, Paatero 1994). Figure 35 gives an overview of current receptor model tools. It is interesting and important to note that the degree of a priori information needed increases from left to right meaning, no information of sources and source characteristics are needed, e. g for the PMF while definite chemical source profiles are needed for the chemical mass balance (CMB). A very good overview on methods and source apportionment results can be found in Viana et al. (2008).

Aim of this improved mathematical tool is the grouping of different variables (e.g. chemical composition of PM) into so called factors. The ratios of the different chemical compounds in a factor are one indication of sources. If e.g. sodium, chloride and magnesia are main variables in factor it can be clearly assumed that sea salt emission is the main source. With a run of a multivariate statistic tool, such as the PMF each factor gets a certain weighting for each sample day which subsequent allows a daily attribution of source contributions to the daily air quality (Figure 36).

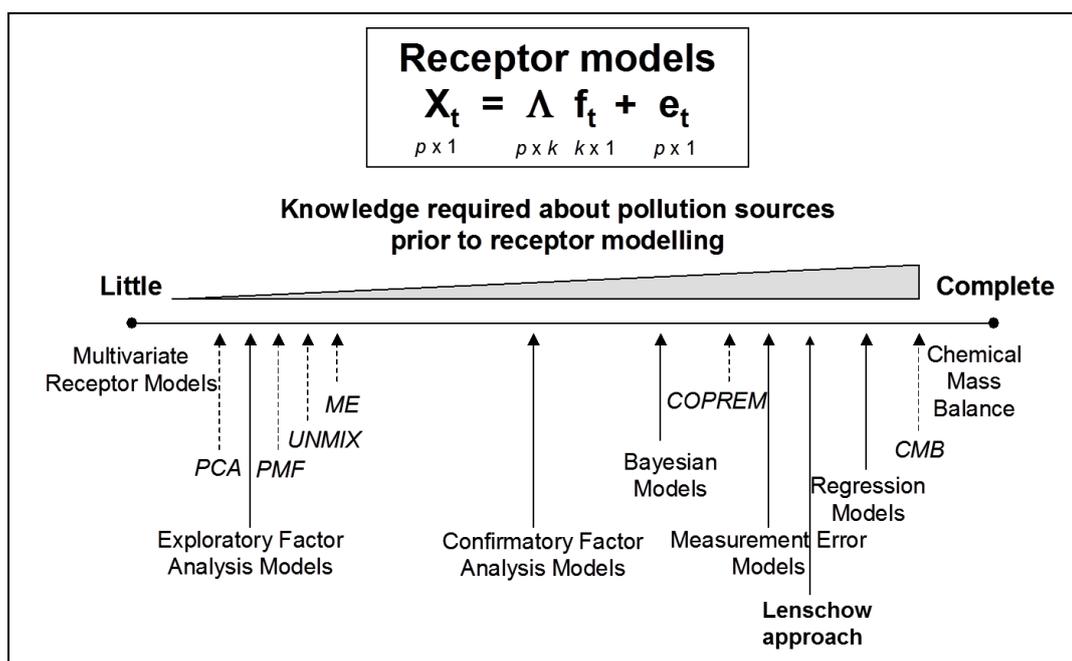


Figure 35: Receptor models and their need of a priori knowledge (based on Viana et al., 2008)

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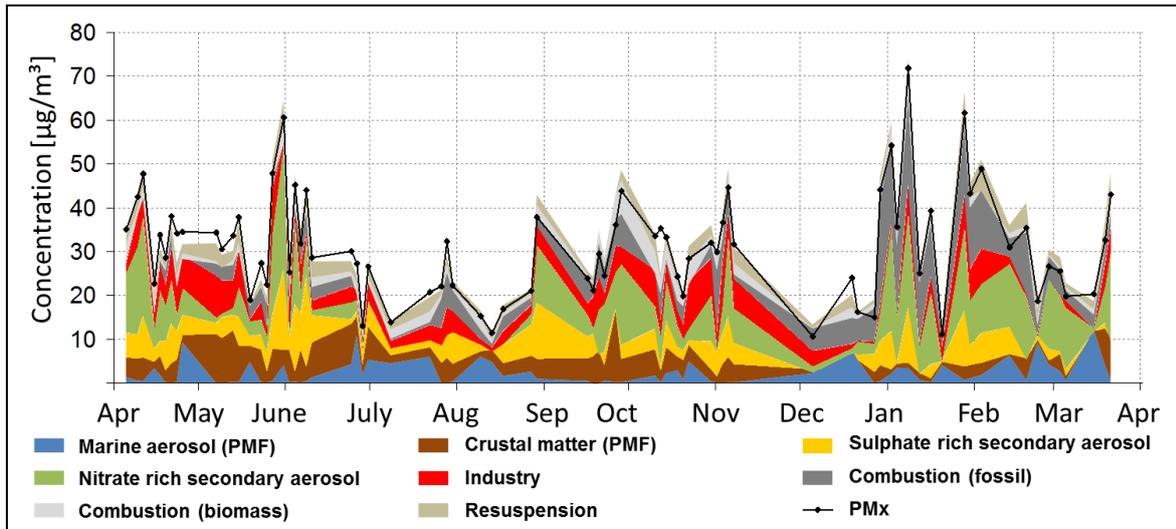


Figure 36: Time series of a PMF study related to an urban background site (Beuck, 2010)

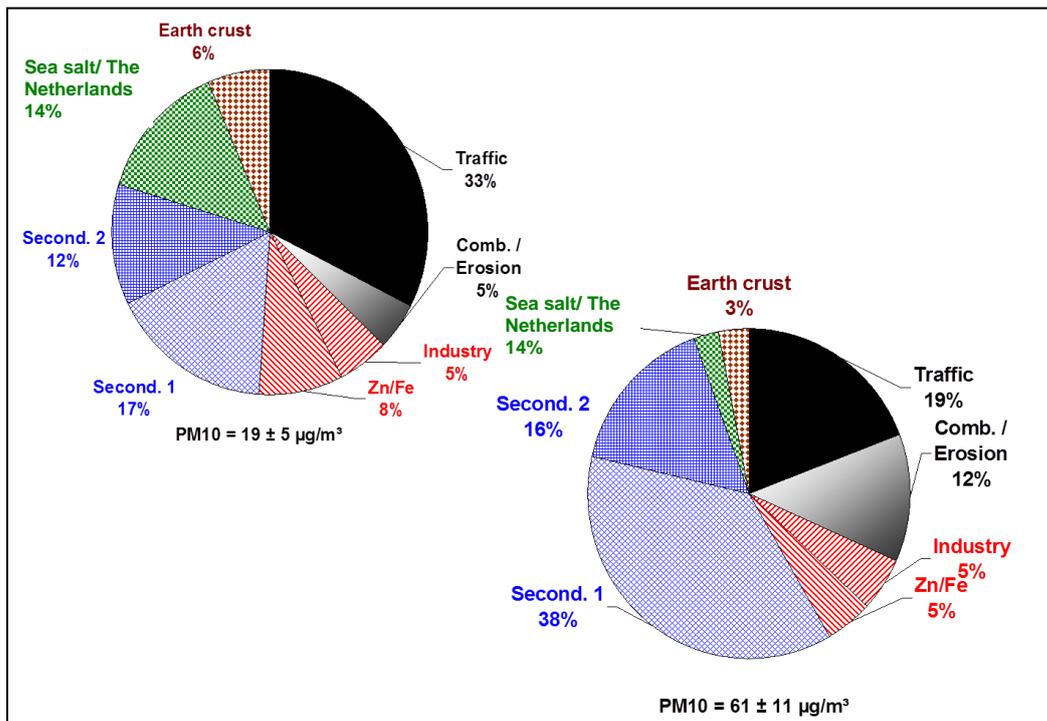


Figure 37: Comparison of factor contributions to PM10 in dependence of ambient mass concentrations (Quass et al., 2004)

Further information on source contribution can be derived when selected subgroups of a time series are studied. This is shown using the example of a factor analysis made for the PM10 fraction in Duisburg (Quass et al., 2004). For days with limit value exceedances (right side Figure 37) contributions of the secondary aerosol represent more than 50% of the atmospheric mass load, while contributions are only about 30% for days with PM10 mass concentration < 30 µg/m³.

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Receptor models allow for the identification and quantification of unknown or not well understood sources and their contributions, since some of these models do not require any a priori information. The downside of this method is demand for long time series measurements of multi-components making this approach time and cost consuming. Another downside is that receptor models have difficulties in separating sources. The models built on covariance and e.g. sea salt reaching Duisburg always travels over land. Hence the sea salt factor derived with PMF analysis always contains some organic and elemental carbon from the combustion processes, e.g. traffic over land. Combination of PMF with further auxiliary data can be used here to derive a) clearer source identification, e.g. by coupling with wind direction analysis and b) differentiation of source contribution from background concentrations by e.g. using diurnal variations. Examples for the identification of natural sources by modelling and PMF can be found in Manders et al. (2010) and Beuck et al. (2011).

A source apportionment method frequently employed in Germany is the so called Lenschow approach (Lenschow et al., 2001). This approach uses simple land used differentiation (hot-spot [industrial or traffic], urban, suburban, rural and regional background) and attributes the concentration gradients to the corresponding general “source group”, e.g. urban. This approach is depicted in Figure 38.

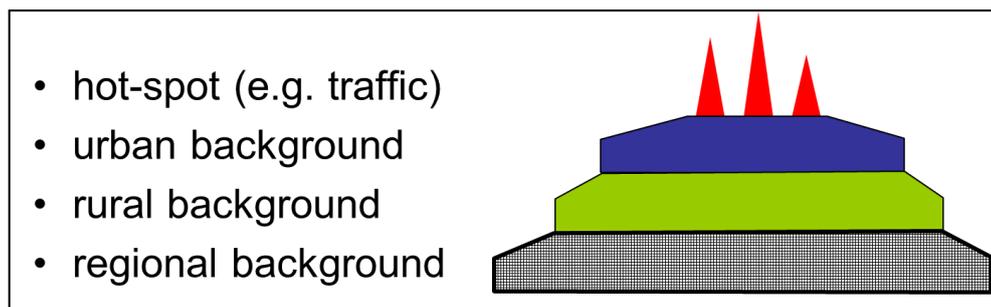


Figure 38: Lenschow approach (2001)

By using the incremental changes in mass concentrations for PM as well as for its chemical composition and linking the chemical composition data along with emission inventories to sources and source groups further detailed source attribution can be made.

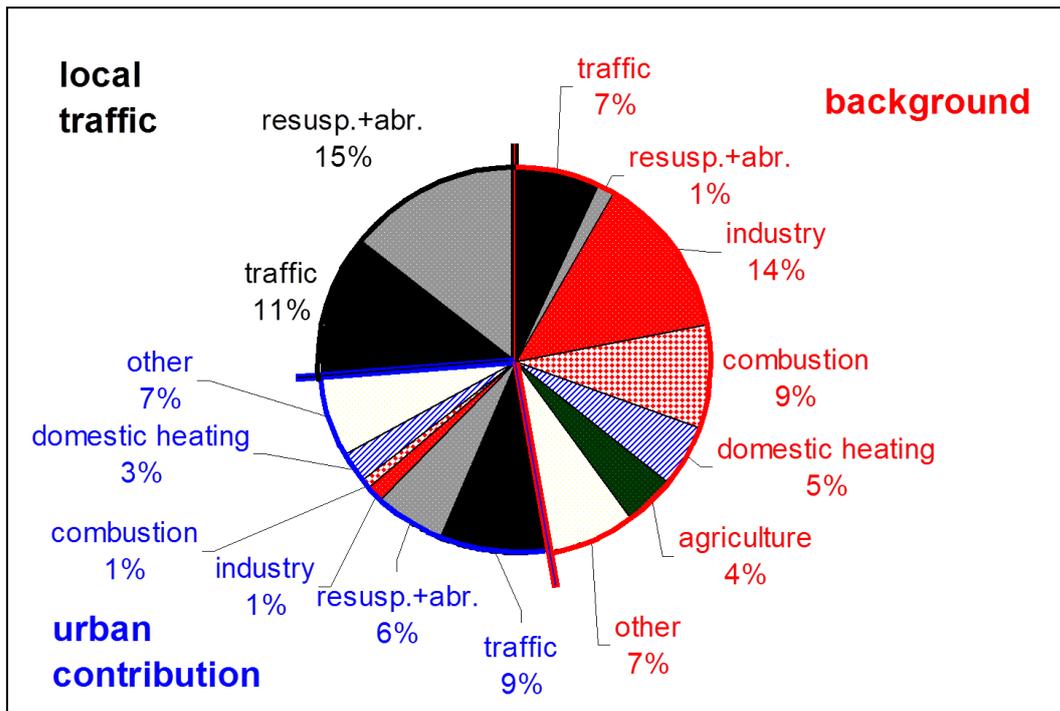


Figure 39: Lenschow approach for Berlin (John and Kuhlbusch, 2004)

Figure 39 gives an example of a source attribution for a Berlin traffic site, Frankfurter Allee. This figure nicely shows that specific traffic contributions decrease significantly with distance to the traffic site. Traffic contributions summed over all contribution areas contribute 49% of the average PM₁₀ mass concentration at this site. Traffic emissions encompass direct tail pipe emission (28%) and diffusive emission such as resuspension, tire wear and break abrasion (21%). The importance of the latter source can be seen by looking at iron oxide concentrations in PM₁₀ at traffic sites (Puteaud et al., 2010, Gietl et al., 2009). Other important contributors to the PM₁₀ load, e.g. power plants, home heating, stem mainly from the background. The linkage of the Lenschow approach (2001) to emission inventories has the advantage of direct source and contributor identification, allow for the planning of straight forward abatement strategies. Nevertheless, sources not present in the emission inventories will be overlooked.

3.1 Emission Measurements And Emission Inventories

Emission measurements and emission inventories are important prerequisites to enable e.g. source apportionment via the Lenschow approach (2001) or to run dispersion models such as Austal 2000 (www.austal2000.de) and chemical transport models such as EURAD, LOTOS-EUROS or CHIMERE. A recent overview on emission inventories, sources and modelling of fine dust in Germany can be found in Kuhlbusch et al. (2010) and Ebel et al. (2010).

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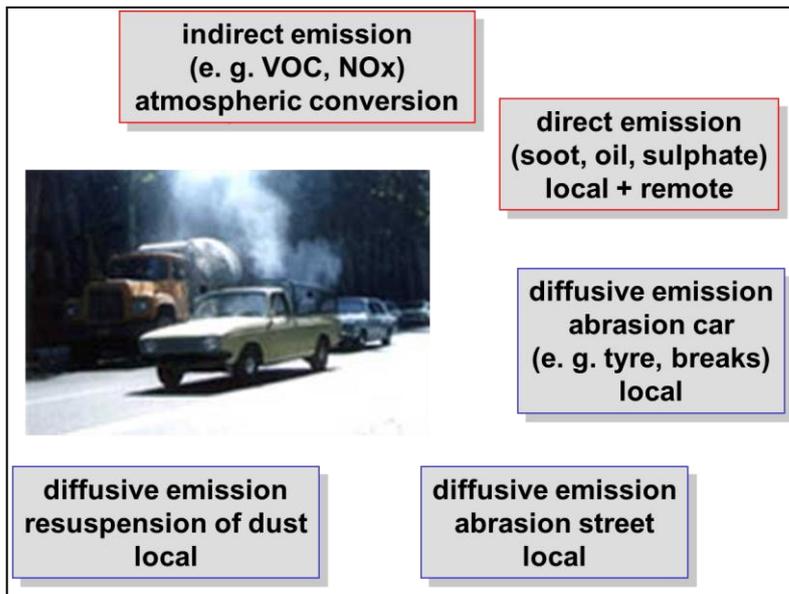


Figure 40: Different particle sources from traffic

With the introduction of the new air quality standards in Europe (1999/30/EG) no device for direct in stack sampling of PM₁₀ and PM_{2.5} existed. John et al. (1999, 2002) developed a specific cascade impactor for this purpose and to enable the determination of corresponding emission rates for different stack sources. Other main sources contributing to urban air quality, as was shown by the study of John and Kuhlbusch (2004) are traffic related emission. Tail pipe emission of particle mass and number concentrations are regularly tested and compiled in e.g. the HBEFA (2010).

While point sources like tail pipe or stack emission can be measured with defined equipment and procedures the determination of emission rates of diffusive sources is much more difficult. Diffusive emissions from traffic are one example (Figure 40). One comprehensive study was conducted by Quass et al. (2008). In this study up and down wind measurements of PM₁ and PM₁₀ as well as its chemical composition was conducted to enable a quantitative differentiation of the different diffusive sources from traffic. The study by Quass et al. (2008) showed that 57% of the total traffic emission at an Autobahn was related to tailpipe emission while 43% could be related to the diffusive sources. The 43% can be divided into 14% attributable to break abrasion, 3% to tyre abrasion, 7% to resuspension of road dust, on average 13% when salt was distributed in winter (an average of 3% otherwise) and 6% which could not be clearly attributed to any of these sources. The emission factors summarised and compared to literature data are shown in Table 8.

Another important diffusive source is the suspension of ores and coal at power plants or steel factories. First studies related to these types of emission were for example conducted within the frame work of setting up a European standard (EN 15445). Hugo et al. (2006) tested different dispersion models for use in the reverse dispersion modelling approach as described in EN 15455. He could show that no significant differences occurred if a Gaussian or Eulerian model was used. Recent detailed studies at coal fired power plants (brown and bituminous coal) showed relative low emission rates for various activities as well as during open storage (Richter et al., 2011). Emission factors range between 0.2-0.8 g/1000 kg coal for PM₁₀ for activities like ship discharge, scooping to move the coal

and scraping. The emission factors for PM2.5 for the same activities only range between 0.02-0.06 g/1000 kg coal (Richter et al., 2011).

Table 8: PM10 emission factors for diffusive traffic emission (based on data from Quass et al., 2008)

Source/process	Emission factor [mg/ km]	Uncertainty [mg/ km]	Comparison values
Salt-resuspension (winter service period)	3 (9)	(5-18)	
Break wear (direct + resus.)	9	5-18	1-7.5 (PKW) [1] 3.5-50 (LKW) [1]
Tyre abrasion	2	1-3	5-13 (PKW) [1] 7.5-200 (LKW) [1]
Street abrasion, resuspension crustal matter	5	3-10	4-40 [1], 11-24 [2]
Tail pipe emission	38	20-70	41 [3], 43 [4]
Not clearly assigned	~12	6-24	

[1] Luhana ,Lakhu, Ranjeet Sokhl, Lynne Warner, Hongjun Mao, Paul Boulter, Ian McCrae, Julian Wright and Daniel Osborn, Characterisation of Exhaust Particulate Emissions form Road Vehicles (PARTICULATES), October 2004.

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[4] Handbuch Emissionsfaktoren des Straßenverkehrs, HBEFA 2.1, Dokumentation zur Version Deutschland erarbeitet durch INFRAS AG Bern/Schweiz in Zusammenarbeit mit IFEU Heidelberg. Hrsg: UBA, 2004.

A summary of a recent compilation of new emission inventories for different air pollutants and the discussion can be found in Jörß et al. (2010) and Table 9. Table 10 gives an overview on global emission rates for different primary and secondary particles. Total primary anthropogenic annual PM10 emission in Germany are given in Table 9 to be 0.26 Mt/a. Worldwide emission of the comparable sources are calculated to be in sum ca. 200 Mt/a according to Table 10, for comparison.

A recent review on the current quality and future needs of emission inventories is given in Winiwarter et al. (2009). They identify major lacks in the inventories for diffusive sources, independent of anthropogenic or natural origin but also information of emission from agricultural activities show high uncertainties.

3 Measurements related to the identification of particle sources

Table 9: Emission rates of primary particles and precursor gases of Germany 2005 (Jörß et al., 2010)

Source groups	Pollutant					
	PM2.5	PM10	NO _x	SO ₂	NH ₃	NMVOC
	kt/a					
Power generation	10	11	283	290	3	8
Non-industrial combustion	27	29	102	78	3	85
Process combustion	9	20	71	65	1	4
Production processes	16	52	91	118	10	86
Production and distribution of fuels	1	4	0	8	0	25
Solvent application , fireworks, tobacco smoke and other consumer goods	9	9	0	0	2	743
Street traffic-benzine	0	0	112	0,5	10	92
Street traffic-diesel	21	21	571	0,5	1	38
Street traffic (evaporation-processes)	0	0	0	0	0	23
Street traffic (tire and break wear)	11	20	0	0	0	0
Street traffic (resuspension)	5	49	0	0	0	0
Other mobile sources and engines	27	21	233	3	1	78
Agriculture	6	26	81	0	577	255
Sum	142	262	1544	563	608	1437

3 Measurements related to the identification of particle sources

Table 10: Global natural and anthropogenic particle emission rates (adopted from Bruckmann et al. (2011) and based on Warneck (1999) and Hainsch (2003))

	Source	Size fraction (dominating)	Main components	global emission rate Mt/a	
Particles from natural sources	primary	Sea Spray	> 1 µm	Sodium, chloride, magnesia, calcium, sulphate	300 - 1300
		Soil erosion	> 1 µm	Silica, aluminium, calcium, Iron, titanium	100-500
		vulcanos	> 1 µm	Silica, aluminium, calcium, Iron, titanium	5 - 30
		Biomass combustion	< 1 µm	Elemental carbon (soot), organic matter, nitrate	5-150
		Biogenic sources	> 1 µm	Organic matter (cell fractions, DNA	30 -80
	<i>SUM</i>				<i>440-2060</i>
	secondary	SO ₂ /H ₂ S/DMS-oxidation	< 1 µm	Sulphate	40 -370
		Nitrate/Ammonium ox.	< 1 µm	Nitrate	60 -200
		VOC-oxidation	< 1 µm	Organic matter (OM)	75 - 200
		<i>SUM</i>			
	<i>Sum of annual global emission rates from natural sources</i>				<i>615-2830</i>
Particles from anthropogenic activities	primary	Industrial processes	> 1 µm	Mineral components and heavy metals	130
		Power plants	< 1 µm	Elemental carbon (soot), organic matter, nitrate	
		Traffic, tail pipe	< 1 µm	Elemental carbon (soot), organic matter, nitrate	
		Traffic, diffusive	> 1 µm	Mineral components and heavy metals	
		Homes	< 1 µm	Elemental carbon (soot), organic matter, nitrate	
		Schüttgutumschlag	> 1 µm	Mineral components and heavy metals	
		Biomasseverbrennung	< 1 µm	Elemental carbon (soot), organic matter, nitrate	
	<i>SUMME</i>				<i>190 - 210</i>
	secondary	SO ₂ /H ₂ S/DMS-oxidation	< 1 µm	Sulphate	110 - 220
		Nitrate/Ammonium-ox.	< 1 µm	Nitrate	20 - 40
		VOC-oxidation	< 1 µm	Organic matter (OM)	10 - 90
<i>SUMME</i>				<i>140 - 350</i>	
<i>Sum of annual global emission rates from anthropogenic activities</i>				<i>330 - 560</i>	
<i>Sum of global emitted or formed particles in the atmosphere</i>				<i>945 - 3390</i>	

The following two sections (3.2 and 3.3) are publications related to ambient and workplace source identification studies in view of possible exposure of humans.

3.2 Sources and Source Contributions to Fine Particles (Kuhlbusch et al., 2009a)

The following manuscript was published in Biomarker.

Abstract

Fine dust is currently seen as one of the major issues in air quality as being responsible for causing adverse health effects. Hence it is important to derive information on particle sources and their contribution to the ambient concentrations. This paper reviews two recent studies related to fine particle sources in Germany. The first study deals with the attribution of source regions to PM₁₀ mass concentrations in nine agglomeration areas of Germany. The second study focuses on the diffusive sources from traffic differentiating and quantifying the various non-exhaust related emissions at an Autobahn.

3.2.1 Introduction

Fine dust, particles of aerodynamic diameters $< 10 \mu\text{m}$, has been associated with different health effects in various toxicological and epidemiological studies. It currently is not definitely known which particle characteristic (e.g. chemical composition, morphology, reactivity) is the one / are the ones actually leading to the determined health effects. Epidemiological studies have shown e.g. significant correlations between fine dust concentrations and mortality (Dockery et al., 1993), as well as cardiovascular effects (Hoffmann et al, 2007) and airway diseases (Krämer et al., 2009).

The “First European Framework Directive for Air Quality“ which was based on these health effect findings came into effect 1996 (Directive 96/62/EC). Still, after 10 years about 67% of agglomeration areas exceeded the daily limit value for PM₁₀ (Figure 41). The revision of this directive and the daughter directives was published 2008, this time also based on results obtained from Integrated Assessment Modelling (Amann et al., 2004). The new air quality directive of the EU (Directive 2008/50/EC, 2008) is extending the focus onto the PM_{2.5}-dust fraction and onto the exposure of the public. The latter is obtained by setting measurement obligations for the determination of an average urban background concentration for agglomeration areas or larger cities.

These mass-related air quality indicators that are linked to limit values and action plans to be set up in cases of exceedances are the background of the studies presented here. Further reports and publications of source apportionment studies in Germany can be found in the following references (Lenschow et al., 2001, Kuhlbusch et al., 2003, Quass et al., 2007a, John and Kuhlbusch, 2004, Werkstatt Feinstaub, Web).

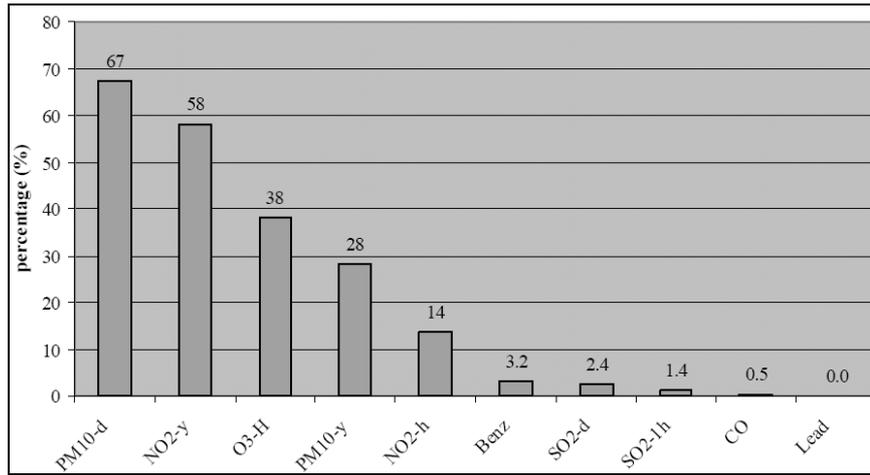


Figure 41: Percentage of zones and agglomeration areas in the EU exceeding the target and limit values of the EU in 2006 (De Leeuw & Vixseboxse, 2007)

The first study deals with the attribution of source regions to PM₁₀ mass concentrations in 9 agglomeration areas of Germany (Quass et al., 2007b). The second study focuses on the diffusive sources from traffic differentiating and quantifying the various non-exhaust related emissions at an Autobahn (Quass et al., 2008).

A very recent overview on source apportionment studies in Europe was conducted within the framework of COST 633 and will soon be published (Viana et al., 2008).

3.2.2 Source Regions of PM

The identification of source regions for air quality parameters and their specific contributions to the local air quality are the basis for assessments of possible abatement strategies and hence the potential success of action plans. Therefore a study based on PM₁₀-data for the years 2003-2005 from the air quality networks in Germany was conducted (Quass et al., 2007b).

3.2.2.1 Agglomeration Areas and Methodology

The nine cities and urban agglomeration areas were Berlin, Hamburg, Munich, Frankfurt and Hannover, Leipzig/Dresden, Ludwigshafen/Mannheim and Bremen as well as the Ruhr Area, represented by the cities of Dortmund and Duisburg at the easterly and westerly boarder. Figure 42 shows that the chosen agglomeration areas cover the whole area of Germany.

Two regional background, two rural background and two urban sites were chosen for each of these areas. This selection of sites was extended by hot-spot sites, two traffic related and where available two industry related sites.

Average diurnal variations (1/2 hourly values) were calculated for each weekday based on the monitoring data, thereby reducing the data set to 336 data points per year, site, and parameter. This data set was further processed by averaging the two corresponding site types per region in case of regional, rural and urban background. This averaging was not done for the hot spots to be able to differentiate possible local differences.



Figure 42: Location of the nine cities and agglomeration areas (stars)

3.2.3 Spatial and Temporal Variability of Contributions

Figure 43 shows on the left side the weekly profile of PM_{10} -mass concentrations averaged for the different site types. A clear increase in concentration from regional background to hot spot location can be seen. Additionally an increase in PM_{10} -mass concentration during the course of the week with a maximum on Fridays is also evident. This trend depicts the build-up of anthropogenic emissions during the week with decreasing concentration on Saturdays and Sundays, during periods of lower emission rates. Interestingly average minimum concentrations at regional background sites and often at rural background sites are determined for Mondays. This can be related to the lower emissions of particle precursors for secondary particles (NO_x , SO_2 etc.) on weekends which on average contribute around 30% to the PM mass and lead to lowest concentrations of e.g. PM-nitrate on Mondays.

Significant differences can sometimes be seen for industrial and traffic related sites. An example is given in Figure 44. PM_{10} mass concentrations at the industrial related site in Ludwigshafen/Mannheim are more similar to the urban background than it is the case for the example taken from the Ruhr Area.

3 Measurements related to the identification of particle sources

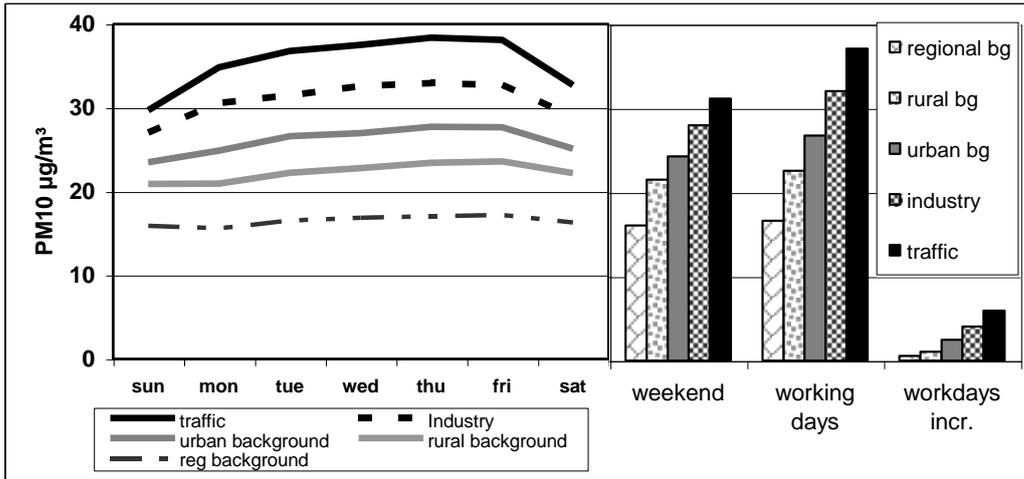


Figure 43: PM₁₀-weekly variation of different site types (average of all sites and years) and comparison for weekend and work days

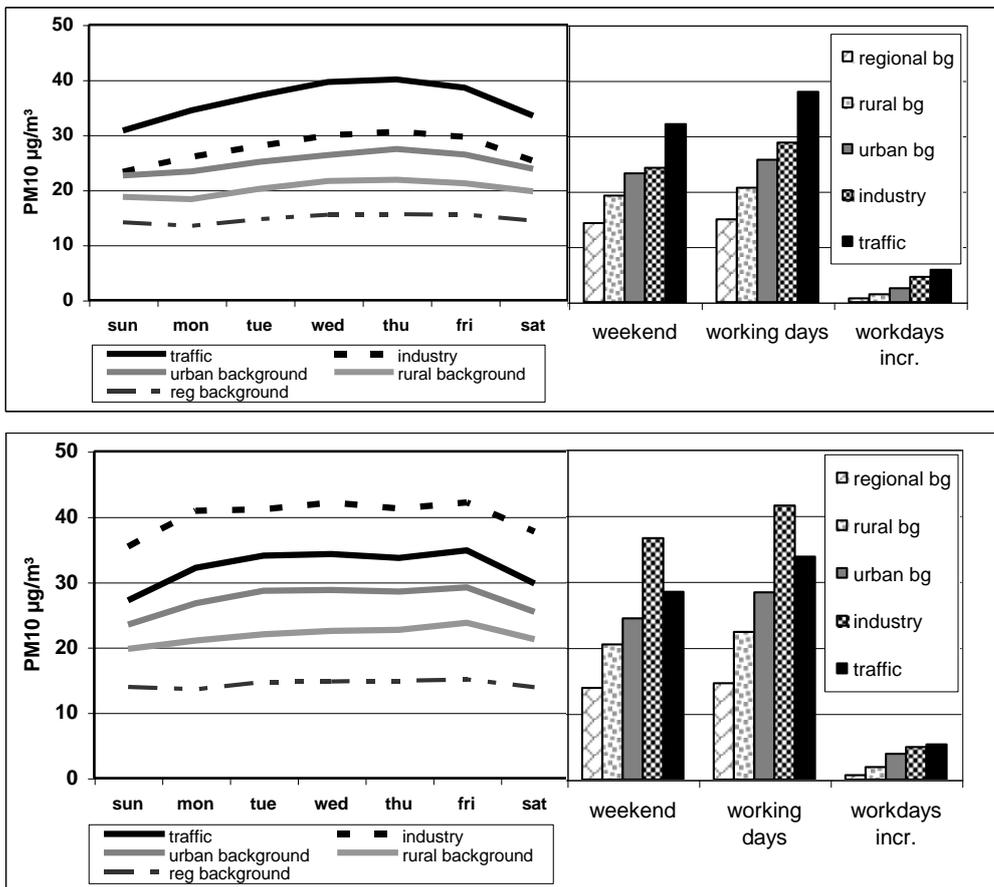


Figure 44: PM₁₀ weekly variation of different site types for the areas of Ludwigshafen/Mannheim (above) and Ruhr Area (below)

3 Measurements related to the identification of particle sources

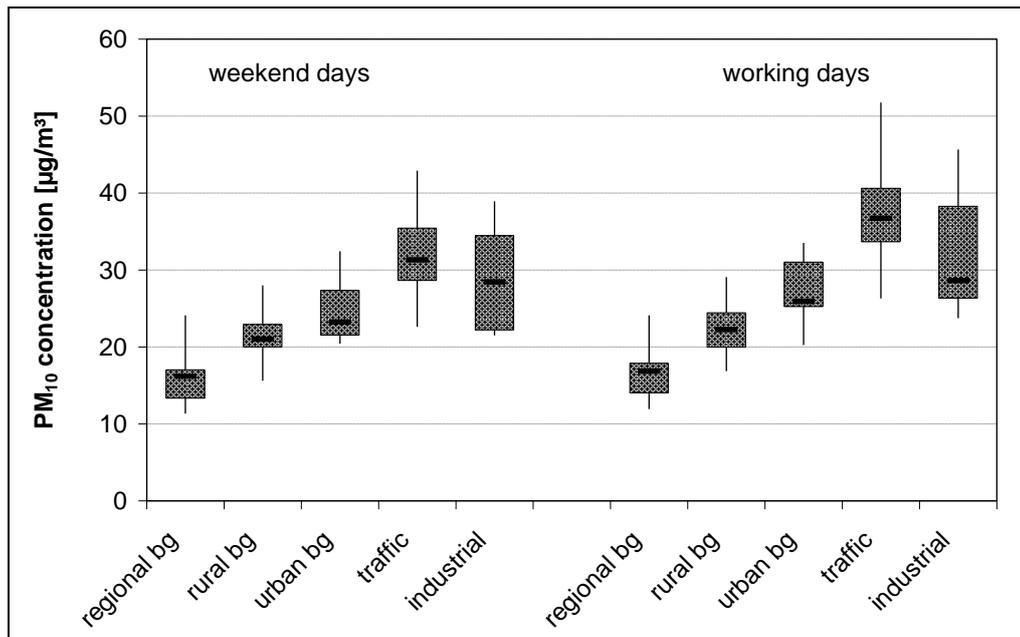


Figure 45: Box-Whisker-Plot with minimum/maximum (vertical line), 25th and 75th percentile (Box), and median (horizontal line) of PM₁₀

Three year average PM concentrations determined for the different site types are summarized in Figure 45 as a Box-Whisker-Plot.

Concentration gradients are evident between the different site types with increasing PM-mass concentrations from the regional background to the urban background and hot-spot sites. This gradient is larger for workdays than for weekend days. It is lowest if only Sundays are averaged (excluding Saturdays).

Table 11: Summary of concentration gradients of neighbouring sites in NRW

Site	difference (µg/m ³)	r ²	Ratio of daily values (av.)
regional/rural background	5-7		
urban/rural background (medium size city)	4 -9	0.2-0.7	1.10-1.60
urban/rural background (agglomeration)	2-6	0.7-0.9	
traffic site/urban background	3-7	0.7-0.9	1.1-1.4
Street canyon/urban background	8-15	0.7-0.8	1.3-1.6
industry/urban background	6-15	0.4-0.6	1.2-1.5

Gradients are in the range of 5-10 µg/m³. This is in the range found in an analysis of the Landesamt für Natur, Umwelt und Verbraucherschutz NRW (LANUV, P. Bruckmann personal communication). Table 11 summarizes results obtained by the LANUV study

for 2005 with the concentration increase, regression coefficient between the site types and ratio range of daily averages.

On average about 50% of the concentrations determined at traffic hot-spots are measured at a corresponding regional background site. This value did not vary significantly between the years 2003-2005 despite quite significantly different meteorological conditions.

3.2.4 Diffuse Traffic Related Emission

Traffic contributions to ambient PM concentration can be related mainly to three emission sources:

1. direct primary particle tail pipe related emission (mainly soot with various amounts of (SV)OC, particle diameter $< 1 \mu\text{m d}_{\text{ae}}$ (Gehrig et al., 2003))
2. diffuse emitted particles (tyre abrasion, break abrasion, street abrasion, resuspension of street dust, particles from corrosive processes, particle diameter mainly $> 1 \mu\text{m d}_{\text{ae}}$)
3. secondary particle generated from gaseous precursors (mainly nitrate from NO_x-emission, and VOC, SO₂, particle diameter $< 1 \mu\text{m d}_{\text{ae}}$)

Secondary particles, according to the last processes (3. above), are of minor importance for local traffic related PM contributions (nevertheless a contribution of 1-2 $\mu\text{g}/\text{m}^3$ in some cases is discussed). On the other hand, a huge fraction of NO_x stems from traffic related combustion processes and therefore a significant amount of ambient background PM-nitrate is due to traffic emission. NO_x-emissions are quite well described due to regulatory obligatory emission testing and measurement of emissions.

Particulate tail pipe emissions are also determined during the regular emission test and emission factors are known for various types of vehicles and driving situations. The emission factors applicable to the German vehicular fleet are summarized in the handbook of emission factors HBEFA (UBA, 2004).

Huge uncertainties on the other hand exist for diffuse traffic related particle emissions. Some studies have been conducted especially in urban areas. These show that between 20-80% of the local traffic related PM₁₀ emission can be attributed to diffuse emissions, depending on traffic flow, fleet composition, road conditions etc.. General emission factors for diffuse emission are given for passenger cars and HDV (Heavy-Duty Vehicle) for example by Düring and Lohmeyer (2004).

So far the contributions of the different diffuse sources (abrasion, resuspension etc.) are not known with sufficient accuracy and therefore effect assessment of activities such as street sweeping cannot be modelled. Studies published so far focussed on heavy trafficked tunnels and urban street canyons which lead to relatively high concentrations of PM, therefore facilitating studies in this direction (Rauterberg-Wulff, 2000, Luhana et al., 2004). Results from these studies however cannot simply be used for high speed rural roads and the Autobahn (motorways). To fill this gap a special project was conducted

3 Measurements related to the identification of particle sources

3.2.4.1 Data and Methodology

Measurements were conducted from 01.09.2005 to 07.01.2007 at the A 61 between AK Meckenheim and AD Bad Neuenahr. Average daily traffic was about 72.000 vehicles/day, with an HDV fraction of 20% (hourly averages 28%, min 2%, max 82%).

PM₁ and PM₁₀-mass concentrations were determined manually as well as automatically (1/2 hour resolution) at upwind and simultaneously downwind conditions along with NO and NO₂ concentrations. The mass concentration difference of the two sites during upwind-downwind condition could directly be related to the traffic emissions, traffic counts, and NO-NO₂ emissions. The difference in PM₁-concentration was evaluated at first approximation as traffic related tailpipe emission while the difference in PM₁₋₁₀-concentration (calculated from PM₁ and PM₁₀ measurements) was interpreted as diffuse traffic related emissions.

The NO_x tracer method (Gehrig et al., 2003) was used for the determination of the total emission factors for PM₁₀, PM₁ and PM₁₋₁₀.

A collection of manually sampled PM₁₀-filters (24 hour samples of 68 days) were chosen for chemical analysis based on meteorological conditions (wind direction, rain, wind speed). Filters of the PM₁-fraction were analysed for the EC and OC content for the verification of tailpipe emissions. EC concentration difference in PM₁ and PM₁₀ served as indicator for tyre abrasion. These data were subsequently used in a Positive-Matrix-Factorisation (PMF) analysis to obtain emission factors for tyre, break, and street abrasion as well as resuspension of dust.

3.2.4.2 Results

Following emission factors were determined by the NO_x tracer method (Table 12):

Table 12: Emission factors for PM_x

	<i>VEH*</i> [g/km*veh]	<i>LDV*</i> [g/km*LDV]	<i>HDV*</i> [g/km*HDV]	<i>VEH**</i> [g/km*veh]
<i>EF PM₁₀</i>	0,067	0,033	0,187	0,066
<i>EF PM₁</i>	0,039	0,017	0,119	0,030
<i>EF PM₁₋₁₀</i>	0,027	0,016	0,068	0,035

Veh: vehicle, LDV: passenger cars, light duty vehicle, HDV: Heavy-Duty Vehicle

** based on 30 min values (TEOM)*

*** based on daily averages (of days chosen for chemical analysis, N=68)*

The values obtained for PM₁₀ compare well to other studies (Gehrig et al., 2003, Düring and Lohmeyer, 2004) and for PM₁ with values calculated based on the information given in HBEFA (UBA, 2004). Significant differences were determined for emission factors of the PM₁₋₁₀ fraction in comparison to values published by Gehrig et al. (2003). Nevertheless, a difference in the range of a factor of 2 can still be viewed as comparable taking the uncertainties in the determination of these factors into account.

Figure 46 shows the chemical composition of the selected PM₁₀ samples. The typical secondary particle components sulphate, nitrate, and ammonium contribute significantly

to the total PM concentrations but are negligible for the local traffic related contributions (difference downwind-upwind). Significant contributions were detected especially for carbonaceous compounds, NaCl, and metals.

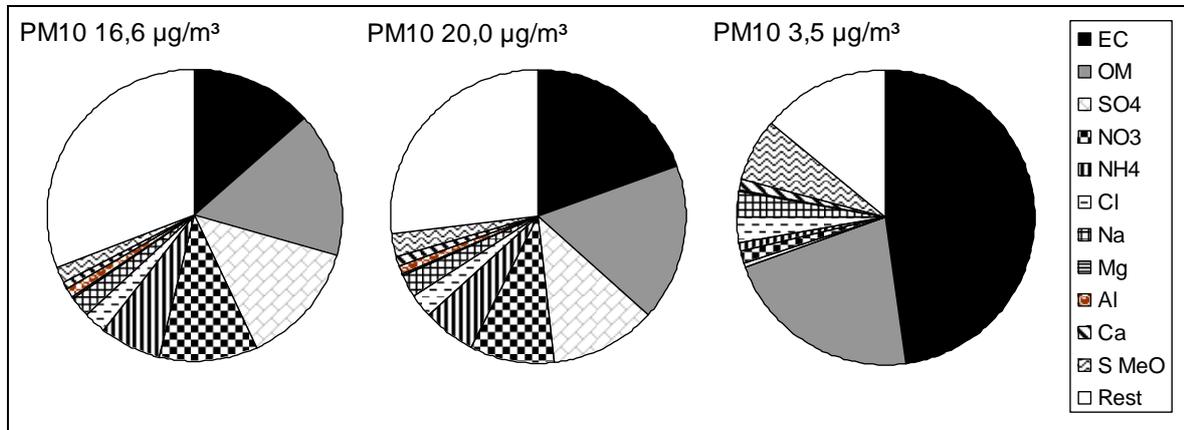


Figure 46: Average chemical composition of the selected PM₁₀-samples (left: upwind samples, middle: downwind samples, right: traffic contribution)

Table 13 summarises the results obtained with PMF-modelling based on the chemical composition data.

Table 13: PMF results (estimated uncertainty range in brackets)

Source process	Fraction of PM ₁₀ -emission [%]	Emission factor [mg/km*veh]
Salt resuspension (only winter period)	13	9 (5-18)
Break abrasion	14	9 (5-18)
Tyre abrasion	3	2 (1-3)
Resuspension, street abrasion	7	5 (3-10)
Tailpipe emission	57	38 (20-70)
Not identified, mainly resuspension	6-19	~12 (6-24)

3.2.5 Summary

The study related to source regions for PM₁₀ showed that about 50% of the concentrations determined at hot-spots are also determined at regional background sites. Nevertheless, significant weekly variations in PM concentrations even at regional background sites show the anthropogenic influence on PM concentrations.

Rural and urban contribution of about 15% add to the regional background concentration leaving on average about 20% of the hot-spot PM₁₀ mass concentration to local activities, here traffic. Variance analysis of regional background concentrations as a fraction of hot-spot concentration did not reveal any significant differences for the years 2003, 2004, and 2005, this despite the highly different meteorological conditions between 2003 and 2005.

3 Measurements related to the identification of particle sources

Diffuse traffic related emissions which are currently not regulated can significantly contribute to local traffic related emission. Only very few studies differentiating the various source processes have been conducted so far, and no study related to rural or autobahn driving conditions to our knowledge.

A corresponding study revealed that about 55-60% of the local traffic related PM₁₀ emission can be attributed to tailpipe emissions. The remaining 40-45% is attributed to break abrasion (ca. 14%), earth crust resuspension (ca. 7%), resuspension of street salt (ca. 13%) and tyre abrasion (ca. 3%). HDV generally showed the highest emission factors, for total PM-emission as well as for the diffuse source processes.

Acknowledgement

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3.3 Particle Characteristics in the Reactor and Pelletizing Areas of Carbon Black Production (Kuhlbusch and Fissan, 2006)

The following manuscript was published in Journal of Occupational and Environmental Hygiene.

3.3.1 Abstract

Physical and chemical characteristics of airborne particles (ultrafine, PM₁, PM_{2.5}, and PM₁₀) in reactor and pelletizing areas during carbon black production were measured to assess process related sources of particles in the work areas. Results from the bagging areas within the same three facilities have been previously published. Particle number and mass concentration measurements were conducted in these work areas and at ambient comparison sites at each of the three carbon black plants. No elevated ultrafine particle number concentrations (UFP, < 100 nm) with respect to ambient were determined in the work areas of Plant 1, intermittently elevated concentrations at Plant 2, and permanently elevated concentrations at Plant 3. The intermittently elevated UFP concentrations in the pelletizer and reactor area of Plant 2 could be related to nearby traffic emissions. The ultrafine particle number concentrations at Plant 2 are comparable to those determined at urban traffic sites. Both work areas of Plant 3 showed elevated UFP concentrations in the reactor and pelletizer area. In the case of the reactor, which was the only enclosed reactor area investigated among the three facilities, the source of the elevated UFP number concentration was most likely attributable to grease and oil fumes from maintenance activities, a conclusion supported by carbon fractionation analysis. The elevated UFP number concentrations in the pelletizing area in this same plant are related to leaks in the production line. These leaks allowed particulate matter to escape to the surrounding areas. Absolute PM₁₀ mass concentrations were all within normal ambient concentrations except for the pelletizing area in Plant 3 that showed continuous levels above ambient. One additional source contributing to peak level PM₁₀ mass concentrations at Plant 2 was due to wind dispersion from a carbon black spill incident the day prior to measurements. It is concluded from these measurements that no carbon black is released in the reactor and pelletizing areas (as UFP or PM₁₀) from the closed production lines under normal operating conditions.

3.3.2 Introduction

Ultrafine particles (i.e., <100 nm, aerodynamic diameter) have received growing attention in recent years because of the apparent toxicity associated with their high surface area (Oberdörster, 2000, Oberdörster et al., 2004, Wehmann et al., 2000). The purpose of this study was to characterize carbon black manufacturing work environments with respect to airborne ultrafine particles.

Particle characteristics in the bagging areas of three carbon black plants were reported in a previous paper (Kuhlbusch et al., 2004). This paper reports the measurements and results obtained in the reactor and pelletizing areas of the same three carbon black plants.

3.3.3 Descriptions of the Process and Sites

The carbon black manufacturing process is described in more detail in a companion paper (Kuhlbusch et al., 2004) and is only briefly summarized here. The carbon black production line has three process steps: Reaction-Collection, Pelletizing-Drying, and Packing. In the first process phase, chemical reactions take place in the reactor to produce the primary carbon black particles during partial combustion or thermal decomposition of hydrocarbons by gas-to-particle conversion. Primary particle sizes can range from 1-500 nm with most produced in the 10 nm to 100 nm range. A few to many tens of primary particles immediately form highly branched chains of carbon black called aggregates. During the process of collection, these aggregates are agglomerated in cyclones and bag houses to much larger entities. Carbon black is packed for shipment either as pellets or as “fluffy” agglomerates. In the production of pellets, agglomerates are pelletized into pearl like pellets, a few tenths of a millimetre in diameter. If water is used in this pelletizing process the pellets are dried in an indirect contact dryer. They are then conveyed into storage bins for subsequent shipment in trucks or rail cars for bulk shipment, in large bags (1000 kg) for semi-bulk shipment, or small bags (25 kg). Measurements are reported here for the Reaction-Collection and Pelletizing-Drying process steps in the same three carbon black plants whose packing areas were reported in the previous paper (Kuhlbusch et al., 2004).

Ambient sites

Local measurements in the vicinity of the process equipment are presented and contrasted to ambient measurements taken at outdoor sites within the same production plant properties. Care was taken to ensure that ambient measurement locations were not downwind of ultrafine particle sources. These sites were 200 to 800 meters from the process sites and were chosen to be low traffic areas.

Reactor Area in Plant 1, 2, and 3

At Plant 1, measurements at two different reactor areas, each with 2 reactor lines, were conducted. The measurement devices were situated between two reactor lines in both cases. The measurement devices at Plant 2 were also placed between the two reactor lines. The reactor areas at these two plants were outdoors. At Plant 3, 4 reactors were located inside a building. The measurement devices were placed inside that building about 2-8 m away from the reactors.

Pelletizing Area in Plant 1, 2, and 3

In Plant 1, two pelletizing areas were investigated which were located about 8 m above ground in semi enclosed housings open on two sides. Measurements were conducted in one pelletizing area each, at Plant 2 and Plant 3. Both pelletizing areas were closed.

The measurements conducted in the work areas represent samples of areas where people are likely to periodically perform work but do not represent personal breathing zone samples.

3.3.4 Measurement Methods

Measurements were conducted for PM₁₀ mass concentrations and for submicrometer number size distributions simultaneously (not Plant 1 due to a breakdown of one of the instruments) within the work areas and outside at the comparison site. Prior to and after each sampling campaign all instruments were co-located at the ambient comparison site to check for comparability and to derive information on ambient air particle characteristics. The measurement devices used in the work areas are termed “inside” (even when co-located at the ambient site) whereas the devices placed at the ambient site are termed “outside”.

The instrumentation used is only briefly described. More detailed information is given in Kuhlbusch, et al. (2004).

Particle size distributions were measured with a Scanning Mobility Particle Sizer ([SMPS] Platform 3080 series, Differential Mobility Analyser 3081, Condensation Particle Counter 3025, and model 3077 Aerosol Neutralizer, TSI Inc., Shoreview, Minn.) which determined the particle number size distributions in the size range of 15-734 nm (d_{st} , Stokes diameter). The determined particle size distributions were converted to the aerodynamic diameter (d_{ae}) by assuming a particle density of 1.75 g/cm³. A particle size distribution was determined every five minutes. As noted above, only one SMPS was used at Plant 1 with consecutive sampling at work areas and at the ambient comparison site due to a breakdown of the second SMPS. Two SMPS were employed at Plant 2 and Plant 3 to simultaneously determine the ambient and work environment particle size distributions. The two SMPS systems were compared to each other prior to each measurement campaign and were found to be equivalent. Taking a safety margin into account, due to changing set up locations, differences of a factor of two for particle size classes can be seen as significant when data are compared.

An Aerodynamic Particle Sizer ([APS], model 3310, TSI Inc., Shoreview, Minn.) was used to measure particle size distributions in the range from 0.5-15 μ m (d_{ae} , aerodynamic diameter)(Wilson and Liu, 1980). Average particle size distributions were recorded every 5 minutes.

Two automatic instruments (Tapered Element Oscillating Microbalance, TEOM[®] 1400ab, Rupprecht & Patashnik, Albany, NY) were used as online PM₁₀ mass concentration monitors, one measuring in the work areas and the other at the ambient comparison site. PM₁₀ mass concentrations were averaged and recorded every 5 minutes. PM₁₀ mass concentration ratios between the two samplers (Inside, Outside) during the outdoor comparison measurements ranged between 0.83 and 1.19. Considering the results of the above comparison and a safety margin due to the frequent change of sampling location for TEOM *Inside*, determined mass concentrations can be viewed as significantly different when concentrations are different by a factor greater than 1.5.

Three manual filtration samplers, two LVS 3 (Derenda, Berlin, Germany) and one DHA 80 (DIGITEL, Zurich, Switzerland) equipped with PM₁₀, PM_{2.5}, and PM₁ inlets, respectively, were used in this study to determine mass concentration. The filters were subsequently used for chemical analysis of the different size fractions. The PM₁₀ and PM_{2.5} inlets are in accordance with ambient air quality standards in the US and Europe (40 CFR Part 50 1988 and 1997, EN12341, 1998). No standard for PM₁ is known to exist at the time of this writing. The volume flow of the filtration samplers, as well as of the

online instruments, was checked and inlets cleaned regularly. Preheated Munktell quartz fibre filters (Munktell, Hamburg, Germany) were used throughout the campaign to enable the determination of elemental carbon. Sampled filters were stored cool (2-10°C) in dishes and aluminium foil during transport. The weighing procedure was in accordance to EN 12341 (1998).

Filters were analyzed for elemental and organic carbon to assess and quantify the magnitude and proportion of the carbon fractions of the total particulate matter in the work areas. Elemental carbon (EC) and organic carbon (OC) were analyzed from the preheated quartz fibre filters by a method developed by Cachier et al. (1989) and Kuhlbusch (1995).

Additionally, an Aethalometer (GIV, Breuberg, Germany) was used as a real-time technique for the determination of black carbon (Hansen et al., 1982 and 1984). The term Black Carbon (BC) is used for data obtained with the Aethalometer and refers to the optically absorbing carbon fraction. The term elemental carbon refers to the inert carbon fraction determined by thermal analysis. Since the Aethalometer determines the light-absorbing fraction of carbon it does not clearly differentiate between elemental carbon (EC) and organic carbon (OC). Thus, values given by the Aethalometer are to be looked at as indicative and are used to identify short time changes due to any discontinuous source processes.

All instruments were checked and calibrated in the laboratory prior to each measurement campaign. All reported mass concentration data have been standardised to 0°C and 101.3 kPa.

3.3.5 Results and Discussion

Number size distributions and concentrations are presented separately from mass concentrations and carbon fractionation since the number concentrations are mainly determined by particles < 200 nm whereas the mass concentrations are related to either the accumulations mode (ca. 200-800 nm) or coarse mode (> 1 µm).

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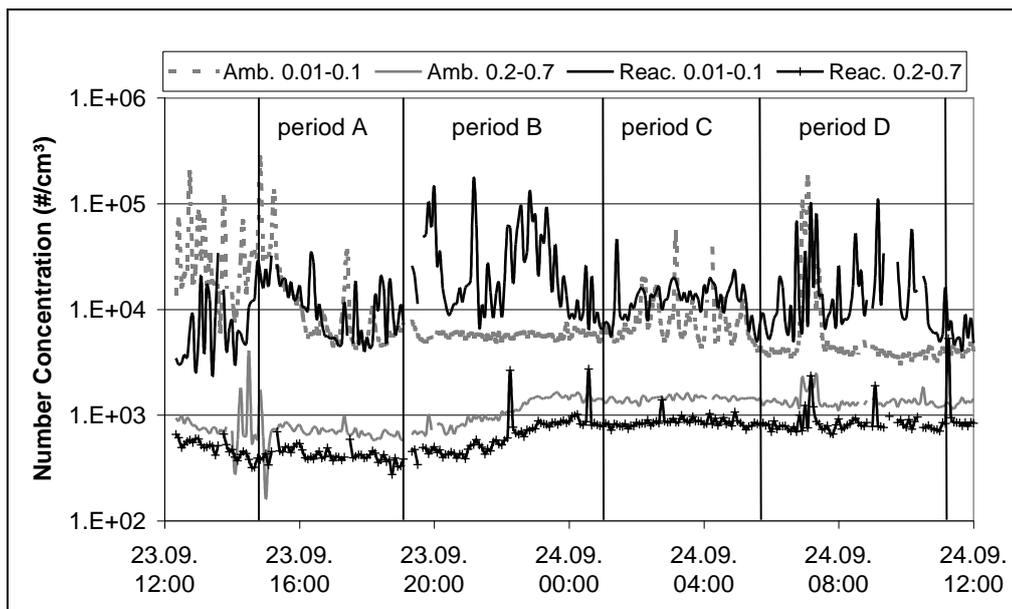


Figure 47: Particle number concentrations in the reactor area of Plant 2, number concentrations for size classes 15-100 nm and 200-700 nm aerodynamic diameter

Reactors and pelletizers are normally operated continuously in the carbon black manufacturing process. Therefore, comparisons between the ambient site and the work area measurements have to be conducted to identify particle sources. This comparison for particle size distributions and mass concentrations are done by comparing time series (e.g. Figure 47 for number concentrations) and separately by dividing the number size distribution determined in the work area by the one determined at the comparison site.

3.3.5.1 Number Concentrations and Size Distributions

Figure 47 shows the time series of the particle number concentrations in the reactor area of Plant 2 as an example. Particle number concentrations are plotted for two size classes (sum of particle number concentrations in the bins between 0.015-0.1 μm aerodynamic diameter d_{ae} , ultrafine particles, 0.2-0.7 μm d_{ae} , accumulation mode particles). Data on the ambient submicrometer particle concentrations, obtained with the second SMPS simultaneously at the ambient site are also plotted.

The results of the comparisons between the average ambient and work area number concentrations are plotted in Figure 48 for all plants. The ambient number concentrations for Plant 1 are plotted separately since those were measured prior and after the work area measurements whereas those of Plant 2 and 3 were measured at the same time.

From this plot it can be seen that the ultrafine and accumulation mode particle number concentrations in Plant 1 and 2 were in the range of the ambient concentrations. This was not the case for the reactor and pelletizer areas of Plant 3. The ultrafine and accumulation mode were always elevated in the pelletizer and reactor areas at Plant 3 indicating a continuous emission source from the processes.

All ultrafine particle number concentrations range between 8,000-44,000 $\#/\text{cm}^3$ (Figure 48a, excluding reactor and pelletizer Plant 3). Number concentrations below 30,000 $\#/\text{cm}^3$ are normal for urban ambient air (Putaud et al., 2004). Number concentrations may reach

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50,000-100,000 #/cm³ at heavily trafficked highway sites (Putaud et al., 2004). Only those of the reactor and pelletizer area of Plant 3 exceed ambient concentrations.

Figure 49 to 52 show the particle number size distributions determined in the different work areas of Plant 1-3 to identify differences and discuss their possible sources. The ratios of work area to ambient air particle size distributions are also plotted in these figures to a) describe the size distribution of the possible sources and b) to show the significance of the difference.

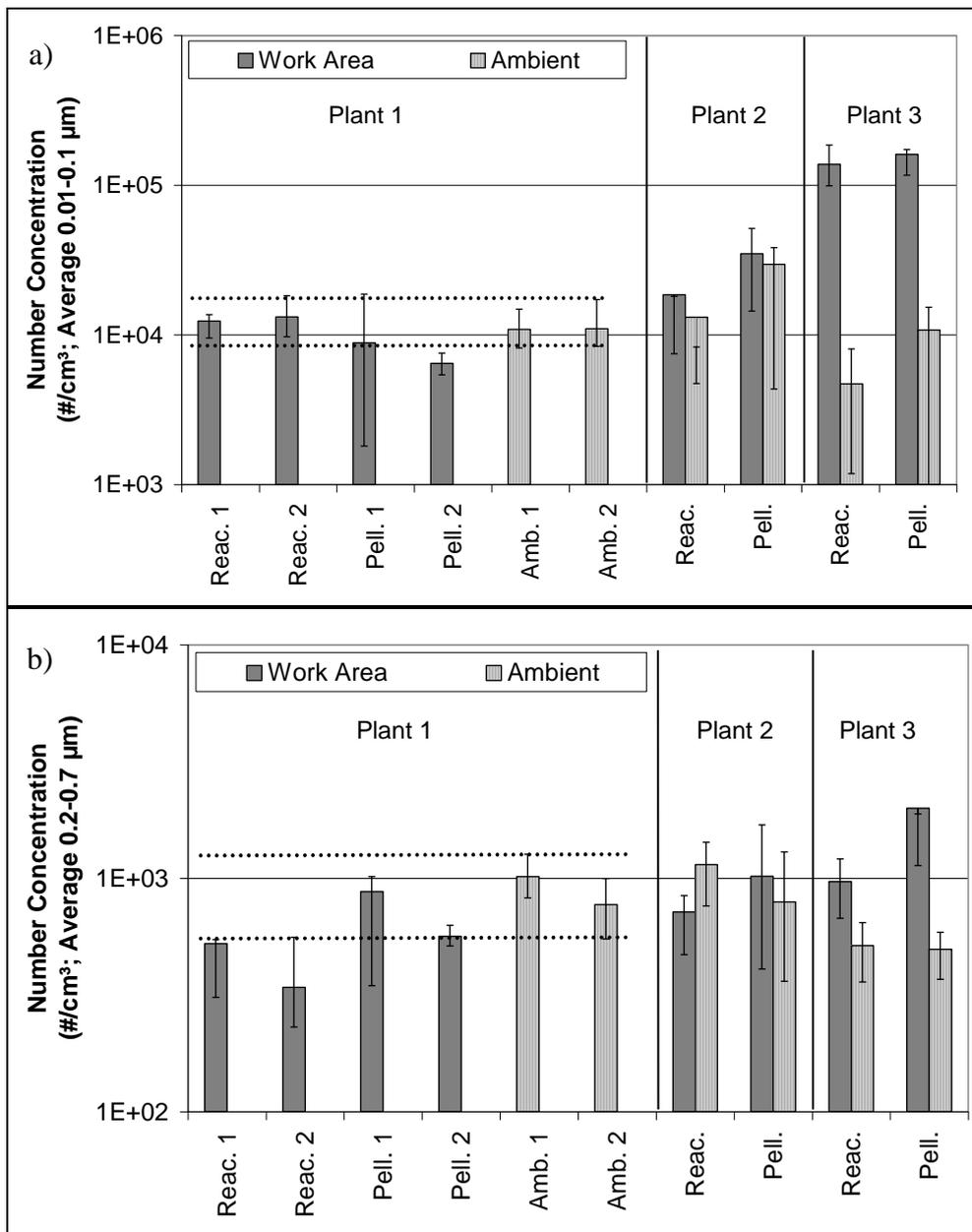


Figure 48: Particle number concentrations in the work areas and at the ambient site for a) ultrafine particles (0.015-0.1 μm) and b) accumulation mode particles (0.2-0.7 μm). Note that the ambient values for Plant 1 were determined prior to and after the work area measurements. The error bars denote the 25 and 75 percentile.

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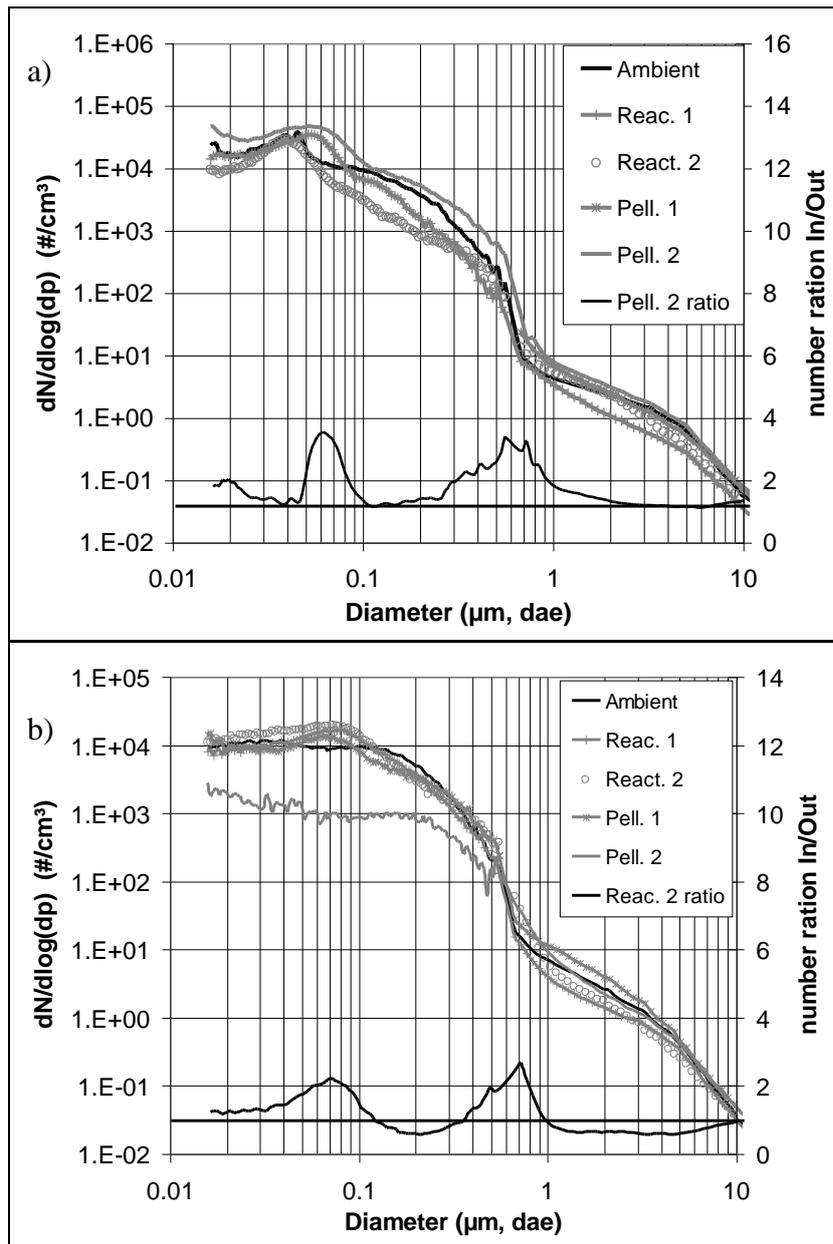


Figure 49: Number size distributions in the pelletizing and reactor areas of Plant 1, a) average from 1 pm to 5 pm, b) average from 10 pm to 4 am

3 Measurements related to the identification of particle sources

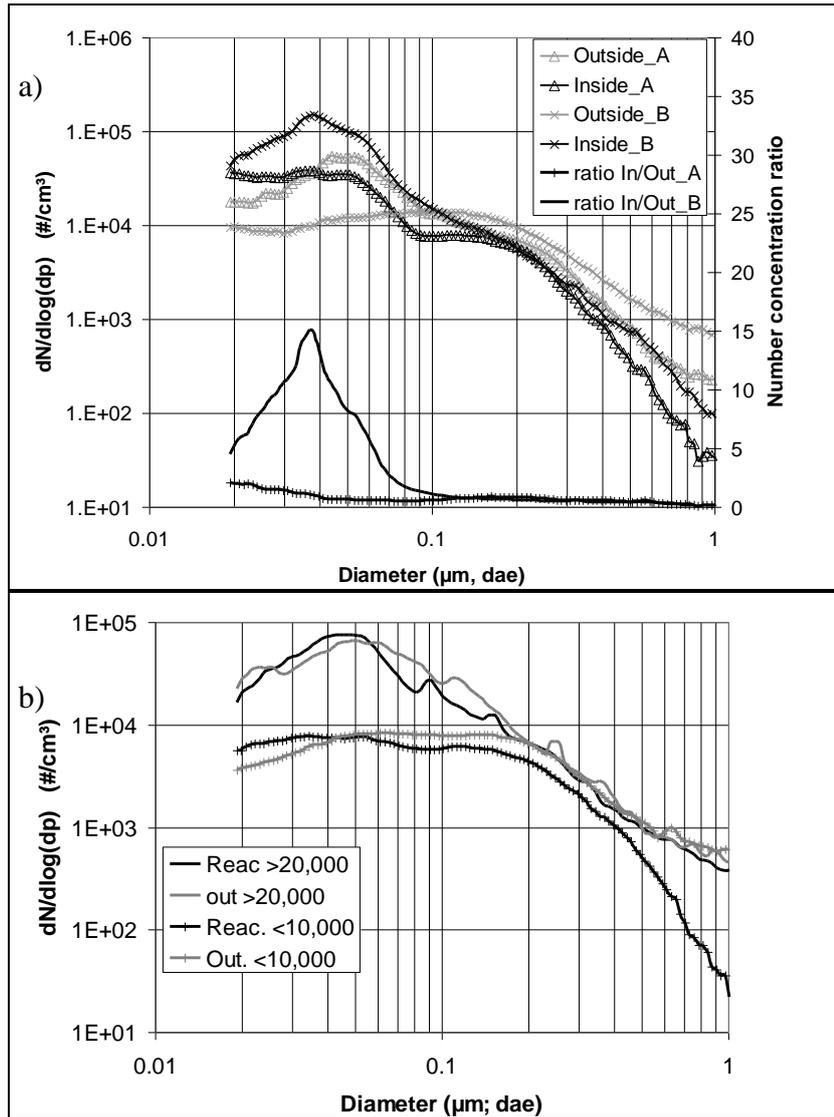


Figure 50: Number size distribution in the reactor area of Plant 2 a) for the time periods A and B (see Figure 47) and b) average submicrometer number size distributions for all periods with number concentrations $< 10,000 \#/cm^3$ and $> 20,000 \#/cm^3$

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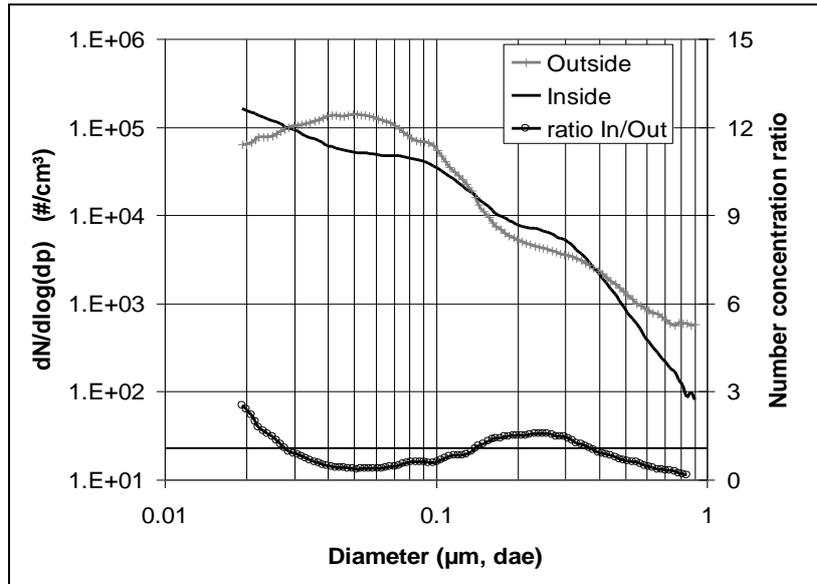


Figure 51: Number size distribution in the pelletizer in Plant 2 period E

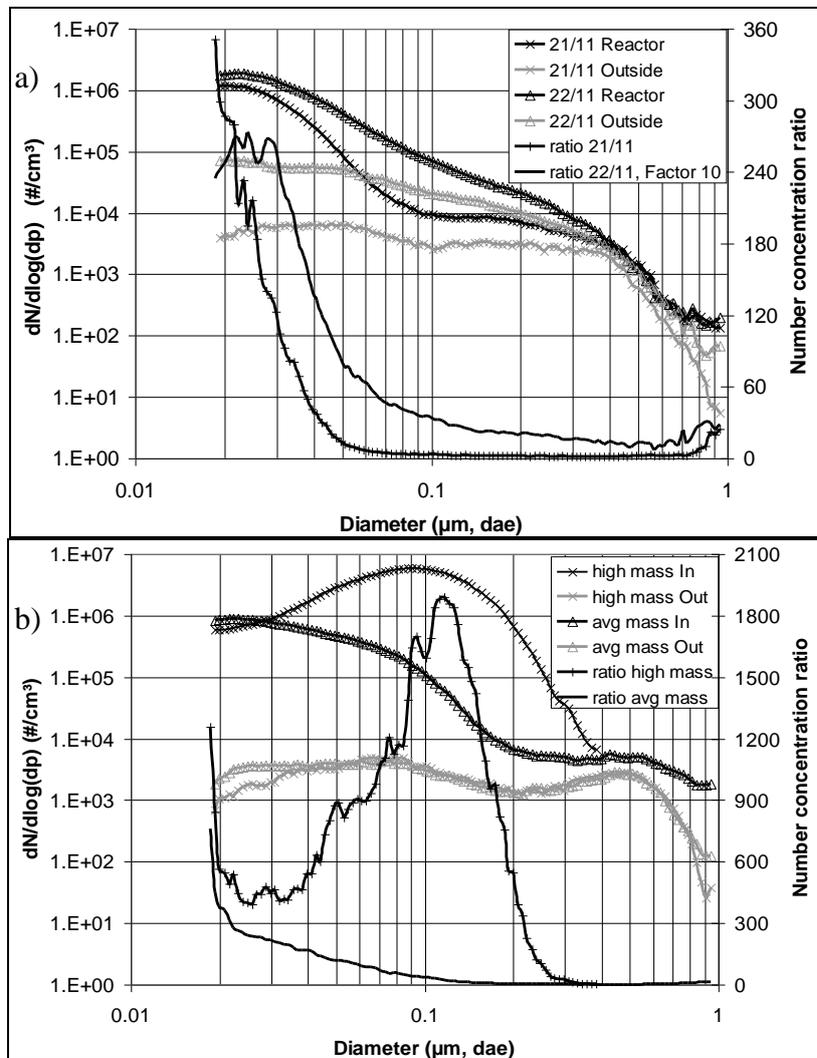


Figure 52: Number size distributions of Plant 3 a) reactor (note: ratio for 22nd Nov. multiplied with 10) and b) pelletizer

Plant 1

Figure 49 a+b show the number size distributions determined in the two reactor and two pelletizing areas of Plant 1 during the production of two different grades of carbon black. Ambient air particle size distributions were determined directly the day after the measurements at the reactors and pelletizers of Plant 1. The weather condition during this period was sunny with low windspeeds and very stable. Hence, we assume and believe that the ambient air particle size distribution can be viewed as representative for the previous days when sampling in the reactor and pelletizing area took place. To check the validity of this assumption two periods of a day were chosen for comparison, one afternoon period (1-5 pm) and one night period (10 pm-4 am). The corresponding size distributions are shown in Figure 49a+b. All size distributions are comparable and lay within a narrow range. The lowest line shown in the figures are the highest ratios of work area to ambient and hence represent the worst case. In both cases two modes, one around 50-60 nm and a second around 700 nm can be seen. These modes are present in 2 out of the 4 possible cases and have a maximum ratio less than 4. These two modes are inconsequential due to the higher uncertainty for the comparison to another day at Plant 1. If the modes are real they can be looked at as of minor relevance in view of exposure by comparison with the general variability of ambient air particle size distributions. From this comparison it is concluded that the reactor and pelletizer at Plant 1 are not a source of ultrafine, accumulation mode or coarse mode particles.

Plant 2

Figure 50a shows the number size distribution and concentration ratio of reactor to ambient for Plant 2 for two periods A+B (see Figure 47). Period A is representative for period C and period B is representative for period D. The ratios (reactor/outside) for the periods A+C range between 0.2-2.7 and for the periods B+D 0.2-15. The number concentration ratio in the cases B and D show a maximum ratio of 15 at 35-50 nm. Due to the different periods and the observation that there were periods with ambient air number concentrations exceeding those determined in the reactor area, average size distribution for all measurements with total particle number concentrations $> 20,000 \text{ \#/cm}^3$ and $< 10,000 \text{ \#/cm}^3$ were calculated and plotted in Figure 50b. This comparison shows very similar size distributions for ambient air and the reactor area. With the knowledge that there were no discontinuous processes in the reactor area, that number concentrations were partially higher in the reactor area and partially higher in ambient air and that the size distributions were very similar in ambient air and in the reactor area when comparing periods with similar number concentrations it is concluded that a source outside of the plant is causing these observations. One source could be a nearby highway coupled with changing wind directions leading to these observations. No simple direct linkage to the wind direction could be established due to the local geographical distribution of the highway and influences in micrometeorological conditions due to buildings and a tower.

The pelletizing area in Plant 2 was relatively more enclosed than those of Plants 1 and 3. The size dependent ratio between pelletizing and ambient particle size distributions range from 0.1 to 2.6 for this time interval (example Figure 51). This size dependent ratio is representative for about 50% of the time measured. Intermittently elevated ultrafine number concentration ratios (up to a ratio of 16) were determined during the remaining

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time. The time periods with elevated number concentrations were all observed during the night with absolute number size distributions not significantly different to the ones measured during daytime outside. A detailed analysis did not give any clear indications of the possible source. Since a) the pelletizing process is continuous, b) no significant increases were determined for day time periods, and c) the absolute concentrations during the night time inside compare well with ambient size distribution determined about 3 hours later, we conclude that observed changes in the number size distribution are most likely not due to the pelletizing process but may be related to nearby traffic emissions outside of the plant.

Plant 3

Figure 52a shows particle size distributions determined in the reactor area of Plant 3 and at the ambient comparison site. Two different time periods were chosen and for both time periods the size distribution ratio of reactor divided by ambient is given. The concentration ratios in both cases range between 1 and 3 over the size range from 0.085 μm to 0.55 μm d_{ae} . The ratios increase significantly below 85 nm to a ratio of about 25 at the 22nd Nov. and even up to 360 at Nov. 21st. The increase above 550 nm is only significant for the 21st (ratio of 20).

Table 14: PM_x carbon fractionation

	Total PM ₁₀ $\mu\text{g}/\text{m}^3$	PM ₁₀ carbon $\mu\text{g}/\text{m}^3$	TC/ PM ₁₀ %	STD (abs) %	EC/ PM ₁₀ %	PM _{2.5} $\mu\text{g}/\text{m}^3$	TC/ PM _{2.5} %	STD (abs) %	EC/ PM _{2.5} %	PM ₁ $\mu\text{g}/\text{m}^3$	TC/ PM ₁ %	STD (abs) %	EC/ PM ₁ %
Plant 1													
Ambient	45	17	38	2	12	25	57	4	14	Sampler defect			
Reactor 1	21	13	61	10	20	16	64		21	Sampler defect			
Reactor 2	34	17	49	16	12	23	59	8	12	Sampler defect			
Pelletizer. 1	47	22	46	2	20	33	56	2	20	33	45	4	16
Pelletizer. 2	45	19	43	1	18	25	57	2	23	30	49	9	11
Plant 2													
Ambient	25	3	15		9	18	34		21	14	22		9
Reactor	27	13	44		30	22	37		13	18	62		18
Pelletizer	47	40	86		60	34	54		41	16	91		28
Plant 3													
Ambient	15	4	30		16	14	28		21	12	23		11
Reactor	24	19	81		30	24	60		17	22	31		17
Pelletizer	1308	1262	92		74	781	74		66	483	88		74

STD: standard deviation from duplicate samples only plant 1

The sharp increase of the size distributions below 85 nm and the maxima below 30 nm indicate relatively recent emissions. Simultaneous Black Carbon (BC) measurements with the Aethalometer do not show any significant higher BC mass concentrations (see also EC values in Table 14) indicating a source other than carbon black for these ultrafine particles.

Figure 52b shows number size distributions determined in the pelletizer area and at the ambient comparison site at Plant 3. Two different time periods were chosen according to significant features in the size distribution. During a time period with high mass concentration (PM₁₀, TEOM[®]) the number size distribution shows a maximum (mode) around 90 nm whereas no such mode was determined during a time period with average mass concentrations. These two different size distributions occur and partially mix during the whole measurement time.

For both time periods the size distribution ratios of pelletizer divided by ambient are also provided. It has to be noted that the ratios determined in the pelletizer were the highest measured during all measurement campaigns. The cause of the high number concentrations and the high mass concentrations is due to emissions stemming from one or more leaks in the production line, specifically, seals on dryers immediate adjacent to the pelletizing area, as observed by one of the authors (TK).

3.3.5.2 PM_x Mass Concentrations

Column 3 in Table 15 gives the average PM₁₀ mass concentration determined in the work areas for the whole measurement time at that site as determined with the TEOM. All PM₁₀ mass concentrations, except the levels measured in the pelletizer in Plant 3, are in the range of 10-50 µg/m³. These mass concentrations are within the normal variability of ambient air mass concentrations at urban and rural sites in Europe and within the daily average limit values for PM₁₀ of the EU (Putaud et al., 2004).

Table 15: Average mass concentration ratios work area to outside and average mass size ratios

		Automatic sampler		Manual sampler	
		PM ₁₀ In µg/m ³	TEOM In/TEOM Out	PM _{2.5} /PM ₁₀	PM ₁ /PM ₁₀
Plant 1	Ambient	43±20	97%	56%	n.d.
	Reactor 1	18±20	119%	66%	n.d.
	Reactor 2	29±25	99%	70%	n.d.
	Pelletizer 1	38±35	90%	71%	70%
	Pelletizer 2	47±39	106%	57%	69%
Plant 2	Ambient	19±10	119%	75%	70%
	Reactor	24±7	125%	84%	76%
	Pelletizer ⁽¹⁾	50±50	474%	72%	38%
Plant 3	Ambient	10±3	94%	85%	67%
	Reactor	21±12	214%	92%	93%
	Pelletizer ⁽²⁾	2080±1613	8156%	71%	34%

⁽¹⁾ difference In/Out mainly due to some peaks during the sampling period

⁽²⁾ values uncertain due to possible overload of samplers/inlets

n.d. not determined

The PM₁₀ mass concentrations in the pelletizing area of Plant 3 showed mass concentration maxima about every 2-3 hours of up to 4000 µg/m³ with concentrations of 200-300 µg/m³ during the intermediate time intervals. These high mass concentrations in

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this work area were due to leaks in the production line as observed by one of the authors (TK).

The PM_x -mass concentration ratio of 'TEOM In / TEOM out' is given in column 4 of Table 15 as a more sensitive indicator of particle emissions influencing the PM_{10} mass concentrations. Co-location of the two TEOMs prior and after the measurement campaign showed good agreement of 94% to 119% (ambient values, column 4, Table 15) between the two samplers. Based on this agreement, PM_{10} mass concentration ratios (column 4, Table 15) were significantly elevated in the pelletizing area of Plant 2 and the reactor and pelletizing area of Plant 3. Absolute PM_{10} mass concentration differences were $40 \mu\text{g}/\text{m}^3$ for the pelletizing area Plant 2, $11 \mu\text{g}/\text{m}^3$ for the reactor area Plant 3, and $2055 \mu\text{g}/\text{m}^3$ for the pelletizing area Plant 3. The mass concentrations in the other work areas investigated were comparable to the simultaneously determined ambient concentrations. The higher PM_{10} mass concentration in the pelletizing area of Plant 2 is due to three peak time intervals while the concentrations in the reactor and pelletizing area of Plant 3 were consistently above the ambient concentrations. The mass concentration peaks in the pelletizer of Plant 2 do not correlate with the changes in UFP concentrations discussed above. The occurrence of peaks makes the pelletizer an unlikely source since it runs continuously. One reasonable explanation could be the loose carbon black adjacent to the pelletizer from a storage container which broke shortly before the measurement started. Clean up work and wind gusts seem likely processes leading to the determined higher mass concentrations.

The determined increased PM_{10} mass concentration in the reactor of Plant 3 is in agreement to the determined increased particle number concentrations in the particle size range of up to $0.9 \mu\text{m}$ (Figure 51). The continuous source of these particles cannot be explained by the particle number size distribution or mass concentration measurements shown so far. The source of the particle in the pelletizing area of Plant 3 was given above.

The $PM_{2.5}/PM_{10}$ and PM_1/PM_{10} mass concentration ratios determined in the work areas and at the ambient comparison site with the manual filtration samplers are given in the columns 5 and 6 of Table 15. From these ratios it can be deduced that the main particle size range contributing to the elevated PM_{10} mass concentrations in the pelletizing area of Plant 2 were particles $> 1 \mu\text{m}$ (low PM_1/PM_{10} ratio). This result is in agreement with the determined particle size distributions for this work area.

The elevated PM_{10} mass concentrations in the reactor area of Plant 3 can be attributed to submicrometer particles (high PM_1/PM_{10} ratio) also in agreement with the determined number size distributions (Figure 52a). The relatively low PM_1/PM_{10} ratio determined in the pelletizing area of Plant 3 may have been influenced by an overload of the manual filtration samplers. Assuming that the determined concentrations are correct, this and the $PM_{2.5}/PM_{10}$ ratio indicate that mainly particles $> 1 \mu\text{m } d_{ae}$ led to the increased PM_{10} mass concentrations. Nonetheless, significant amounts of submicrometer particles were released as can be seen in Figure 52b.

3.3.5.3 Carbon Fractionation

Table 14 gives an overview of the determined carbon fractions TC (total carbon) and OC (organic carbon) from the samples collected with the manual filtration samplers in the work areas and at the ambient comparison site. Carbon fractions were determined for the

PM₁₀, PM_{2.5}, and PM₁ size fractions. The average PM_X mass concentrations are given along with the percentages of the carbon fractions.

Particle size distributions and PM_X mass concentrations in the reactor and pelletizing areas of Plant 1 as well as in the reactor area of Plant 2 did not differ from those of the ambient comparison site. A similar result was obtained for the carbon fractionations in most of these work areas (Table 14). Some difference may be observed for the PM₁₀ TC concentration at reactor area one of Plant 1 (TC/PM₁₀ 61%). This increased ratio does not correspond with increased mass or particle number concentrations, and does not represent a significant mass concentration (4.5 µg/m³ TC) at workplace environments.

The PM₁₀ and PM₁ TC ratios in the reactor area of Plant 2 (TC/PM₁₀ 44%, TC/PM₁ 62%) were higher than at the ambient comparison site some days before. EC/PM_X ratios, an indicator for carbon black or soot from diesel engines, were also elevated in the PM₁₀ and PM₁ fraction. In the section on number size distribution it was shown that this site was influenced by some outside sources of submicrometer particles some part of the time (Figure 47 and Figure 50). This outside source, previously related to outside traffic, may be the cause for the elevated TC/PM_X and EC/PM_X ratios in the PM₁₀ and PM₁ fraction.

Influences on particle number and/or mass concentrations were determined for the following work areas, pelletizing Plant 2, reactor Plant 3 and pelletizing Plant 3. These areas also show some changes in the carbon fractionation. All size fractions in the pelletizing area of Plant 2 show elevated TC and EC to PM_X ratios. The measurements of the size distributions at this site did show some slight differences to the ambient site, likely related to outside traffic emissions, and PM₁₀ mass concentration measurements showed 2-3 peaks most likely related to wind gusts eroding loose carbon black particles from the broken storage container previously noted. Hence the determined elevated TC and EC mass concentrations can be related to those sources.

The reactor area of Plant 3 was the only enclosed reactor area (four reactor lines) investigated. Significant increased UFP number concentrations and slightly elevated mass concentrations were determined in this work area. The results of the carbon measurements (Table 14) show changes for EC but even higher increases in the TC concentrations. We conclude that about 10 µg/m³ additional mass concentration (seen for all measured size fractions PM₁, PM_{2.5}, PM₁₀) in the enclosed reactor is mainly organic carbon. Since the reactors are enclosed the most likely source of the elevated number and mass concentrations in this reactor area is grease and oil used in the maintenance of the reactor. This increase in the enclosed reactor area could not be determined in the open reactor areas as a result of the minor source strength and the dilution effects of ambient conditions.

The highest of all particle concentrations were determined in the pelletizing area of Plant 3. The results of the carbon measurement are as expected for combusted and uncombusted tail gas with high EC and only slightly higher TC concentrations.

3.3.6 Summary and Conclusions

Measurements of particle size distributions in the pelletizing and reactor areas of three carbon black plants showed intermittently elevated ultrafine particle (UFP) concentrations in Plant 2 and continuously elevated concentrations in Plant 3.

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Elevated UFP concentrations in Plant 2, reactor and pelletizer, could be attributed to nearby traffic outside of the plant. UFP concentrations were in the normal concentration range for ambient sites. Elevated UFP were observed in both work areas of Plant 3. The most likely composition of the elevated UFP in the reactor of Plant 3 (the only enclosed reactor area including four reactor lines) was determined to be mainly organic carbon with some EC (indicator of carbon black or soot). The highest UFP concentrations were measured in the pelletizing area of Plant 3. The source of these particles was from leaks in the production line with high amounts of EC.

Table 16: Ultrafine particle number concentrations at carbon black work areas

		Observed increases	Likely origin
Plant 1	Reactor	No	
	Pelletizer	No	
	Bagging	No	
Plant 2	Reactor	Yes	Near-by traffic
	Pelletizer	Yes	Near-by traffic
	Bagging	Yes	Propane fork lifts
Plant 3	Reactor	Yes	condensed oil vapors
	Pelletizer	Yes	Flue gas-leak
	Bagging	Yes	Diesel fork lifts, gas heater

The results obtained with the number size distribution measurements are in close agreement with the PM₁₀ mass concentration measurements conducted simultaneously at the ambient comparison site and inside of the work areas. PM₁₀ mass concentrations in the work areas were within normal ambient range excluding the results from the pelletizer of Plant 3. The source for the high mass concentration was the same as for the high UFP concentrations, leaks in the production line. Elevated PM₁₀ mass concentrations, a factor of 4 higher than ambient, were determined in the pelletizing area of Plant 2. The additional mass concentration of about 40 µg/m³ was related to three concentration peaks within the measurement interval. One possible cause identified is fugitive dust re-entrained by wind gusts from the previously noted spill. The source of the slightly elevated PM₁₀ mass concentration in the reactor area of Plant 3 (ca. 10 µg/m³) was most likely the same as for the UFP, grease and oil used for the maintenance of the reactor lines.

Summarizing the findings of this investigation it can be concluded that particle number and mass concentrations in the reactor and pelletizing areas of carbon black production plants appear to fall in the range of ambient air values when normal operating and preventive maintenance conditions exist.

Table 16 summarizes the results obtained from this study and the previously published study on the bagging operations at these three plants (Kuhlbusch et al., 2004).

Acknowledgement

This work was sponsored by the International Carbon Black Association (ICBA). We also would like to thank all the companies and workers involved in the measurement campaigns for their welcome help and understanding.

3.3.7 References

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4 RELEASE AND EXPOSURE TO NANOPARTICLES

Release and diffusive emissions at ambient conditions were summarised in the previous section, especially 3.1. Chapter 4 solely deals with the release and exposure related measurements at industrial workplaces or work activities which are somehow related to nanomaterials. The first paper by Stahlmecke et al. (2009) describes a specific method investigating agglomerate stability of nanoparticles. This is of importance in view of health effects since deagglomeration of powders during release may lead to finer particles which possibly have a higher mobility after human uptake. Deagglomeration may also be of importance after uptake in biological fluids e.g. lung lining as investigated in the NanoCare project (Schulze et al., 2008, Kuhlbusch et al., 2009c).

The second manuscript, Kuhlbusch et al. (2011) is a review of workplace exposure related studies as well as laboratory simulations of possible release from nanomaterials. This review summarises investigations including those of Kuhlbusch et al. (2004), Kuhlbusch and Fissan (2009) and Stahlmecke et al. (2009) and reviews the current state of the art with indicating limitations and research needs.

4.1 Investigation of Airborne Nanopowder Agglomerate Stability in an Orifice Under Various Differential Pressure Conditions (Stahlmecke, Kuhlbusch et al., 2009)

The following manuscript was published in Journal of Nanoparticle Research.

4.1.1 Abstract

The stability of agglomerates is not only an important material parameter of powders but also of interest for estimating the particle size upon accidental release into the atmosphere. This is especially important when the size of primary particles is well below the agglomerate size, which is usually the case when the size of primary particles is below 100 nm. During production or airborne transportation in pipes, high particle concentrations lead to particle coagulation and the formation of agglomerates in a size range of up to some micrometers. Binding between the primary particles in the agglomerates is usually due to *van der Waals* forces. In case of a leak in a pressurized vessel (e.g. reactor, transport pipe, etc.) these agglomerates can be emitted and shear forces within the leak can cause agglomerates to break up. In order to simulate such shear forces and study their effect on agglomerate stability, a method was developed where agglomerate powders can be aerosolized and passed through an orifice under various differential pressure conditions. First results show that a higher differential pressure across the orifice causes a stronger fragmentation of the agglomerates, which furthermore seems to be material dependent.

4.1.2 Introduction

Nanoagglomerate powders are used in a variety of applications such as color pigments in paints (e.g. Liz-Marzán, 2004, Quinten, 2001), as carriers for instance during drug delivery (e.g. Panyam and Labhasetwar, 2003, Soppimath et al., 2001) or sun screens (Schulz et al., 2002). Recently, the use of powders containing nanoscale particles is becoming more and

more common, because they can provide new functionalities and applications (Maynard, 2007), such as in scratch resistant paint or self-cleaning surfaces. Nanoscale TiO₂ particles are e.g. used in cosmetics (Kaur and Agrawal, 2007), because they become transparent as opposed to microscale particles but maintain their other functionality. Other materials promise to yield the same effects as powders consisting of primary particles in the micrometer size range by using much less material, i.e. during use in an automotive catalytic reactor. Furthermore, due to their unique properties, they show the perspective of new functionalities and therefore new applications (Anselmann, 2001, Siddiquey et al., 2008). The production of nanoagglomerate powders has been significantly increased over the past few years. Hence, the likelihood of accidental release of these particles is also increased and possible implications need to be investigated.

Primary particles are produced e.g. by pyrolysis inside a closed vessel. In the dry state and under the high concentrations usually dominant during production these particles tend to form agglomerates (consisting of primary particles and / or tightly bound aggregates) of a size of up to some micrometers (Hinds, 1999). Chemical bonds may also be formed by chemical or physical reactions (e.g. sintering during the passage of a high temperature zone inside the vessel), which lead to the formation of tightly bound aggregates consisting of several primary particles. These aggregates are usually stable in a sense that they cannot be broken down into primary particles once they are formed. Agglomerates, found in nanopowders are commonly agglomerations of aggregates and/or primary particles. The most prominent mechanism for uncharged particles to form agglomerates is coagulation induced by Brownian motion of the particles. Binding between the particles takes place due to *van der Waals* forces. The effect of *van der Waals* force on bonding of spherical particles was described in detail by Hamaker (1937). Furthermore, electrostatic or-in the case of magnetic particles-magnetostatic interactions may lead to bigger agglomerates. The binding energy in an agglomerate is greatly enhanced in the presence of water within the powder. The effect of a meniscus building up between two primary particles may lead-depending on the distance between the particles and the relative humidity-to an immense increase of the binding energy (Eber, 2004).

A variety of methods exists to study the binding energy between primary particles or aggregates. The most basic method is to investigate the energy necessary to split up a couple of two primary particles. This can be achieved by using atomic force microscopy (AFM, Blum, 2006), when one of the particles is attached to the tip and the other is attached to a substrate. By increasing the distance between the tip and substrate, the necessary energy to split up the agglomerates can directly be measured. The drawback of this method is the limited number of experiments which can be conducted in an adequate time and the simplification of studying the bonding between only two particles or agglomerates/aggregates.

Another method to study the binding energy of airborne agglomerates is the production of primary particles of the desired size and the subsequent controlled agglomeration (Froeschke et al., 2003, Seipenbusch et al., 2007). The agglomerates are formed in the airborne state and are subsequently deposited by impaction under various low pressure conditions on a grid for transmission electron microscope (TEM) investigations. The differential pressure leads to an acceleration of the particles und thus to a high impact velocity. This corresponds to a high impact energy which can lead to a deagglomeration of the agglomerates. This effect can subsequently be studied by high resolution electron microscopy, in which a gap between primary particles indicates the break-up of the bond between these particles. By comparing

the number of broken bonds of the impacted agglomerates with the number of bonds of “softly“ deposited agglomerates the binding energy can be calculated for a known deposition velocity and an estimated agglomerate mass. In contrast to the AFM-method, statistical information of the binding energy can be achieved, but the number of experiments is still limited. The effect of impact fragmentation on compact agglomerates is also studied numerically (e.g. Thornton et al., 1996, Sator et al. 2008) which helps to understand the microscopic factors influencing the fragmentation behaviour.

In both aforementioned cases only a limited number of agglomerates can be investigated due to the tedious analysis of the experimental results. Furthermore, these experiments were designed to reflect “perfect agglomerates” in the sense, that they are intentionally produced for these studies in order to determine material properties, whereas the intention of the study presented here was to investigate the stability of industrially produced nanopowders.

In this paper, we present a novel method applicable to aerosolized nanopowders, which is based on the deagglomeration in an orifice under various differential pressure conditions. The fact that agglomerates do get destructed during the passage of an orifice is common knowledge and has been applied before for the deagglomeration of micron-sized agglomerates (Fonda et al., 1999). Similar experiments were also conducted by Kurkela et al. (2008) on particles in the same size range. They studied the deagglomeration process within a turbulent stream and found out that this method can effectively be used to separate compact particles from each other. The study presented in this paper allows the comparison of the stability of different nanoparticle agglomerates with each other. The deagglomeration of some typical nanopowders mainly based on metal oxides was studied and the results are presented.

4.1.3 Experimental Setup

In order to mimic the flow and shear forces and the resulting deagglomeration in a leak in a pressurized vessel, nanoagglomerate powders were aerosolized and passed through an orifice under various differential pressure conditions. The number size distribution of the aerosol after passing the orifice was measured and compared to the size distribution of an “unstressed” aerosol.

The experimental setup is shown in Figure 53. Experiments consisted of three steps: In the first step the powder, as delivered from the manufacturer, was aerosolized. This is done in a pressurized beaker by magnetically stirring the powder under a constant carrier gas flow. In the study presented here, particle free, dry nitrogen with a relative humidity well below 10% was used as carrier gas in order to eliminate possible unknown enhancement of the binding energy caused by water vapour.

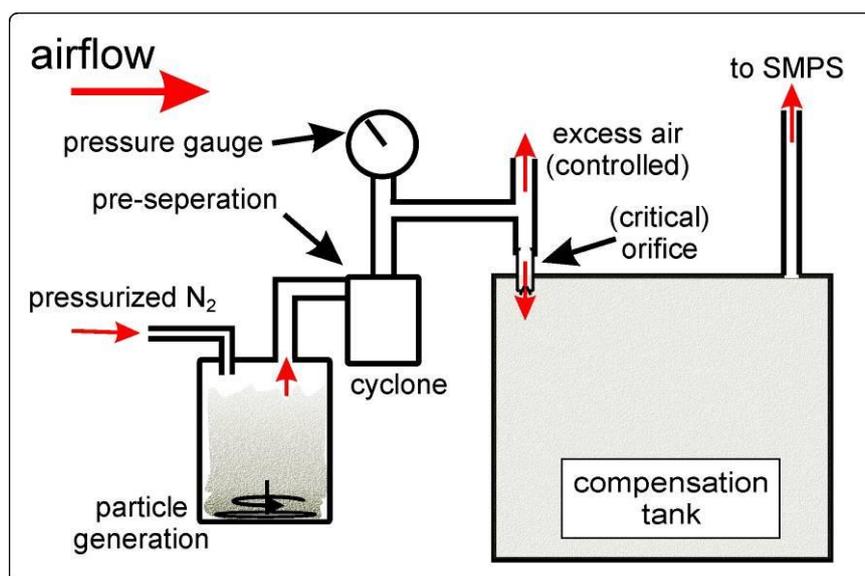


Figure 53: Schematic measurement setup. After generation within a pressurized beaker the aerosol containing nanoparticle agglomerates passes a (critical) orifice under different overpressure conditions. The resulting number size distribution of the aerosol is measured after a compensation tank using an SMPS at ambient pressure.

After generation of the aerosol, airborne agglomerates with an aerodynamic diameter above approximately $1.3 \mu\text{m}$ are removed by two consecutive cyclones (in Figure 53 shown as one cyclone). After this pre-separation of bigger agglomerates the aerosol passes the orifice with an aperture of $508 \mu\text{m}$ diameter and sharp edges (O’Keefe controls, type: K4-20) into a compensation tank with a volume of 10 litres which is used to damp possible influences of concentration spikes. The whole setup (besides the beaker made of pressure resistant glass) is electrically grounded to minimise particle losses. Due to the shear forces within the orifice a deagglomeration of the agglomerates into smaller fragments or even primary particles occurs. The magnitude of the shear forces is defined by the pressure gradient within the (critical) orifice, i.e. with increasing overpressure the flow Reynolds number and thus the turbulence inside the orifice is increasing.

The effect of shear forces was studied by applying overpressures starting from 10 kPa and then 20 kPa up to 140 kPa in 20 kPa steps. Above an overpressure of about 100 kPa the orifice becomes critical. The pressure downstream of the orifice was maintained at ambient level. To maintain a constant flow rate of 2 litres per minute (at operating pressure) through the cyclones a second orifice is used to release excess air (the size of this orifice is depending on the applied overpressure).

To investigate the effect of the different shear forces a measurement of the “as prepared” aerosol is also conducted for reference. In this case the aerosol enters the compensation tank with a flow rate of 2.0 litres per minute without passing the orifice. Only the pre-separation process in the cyclones takes place. The resulting size distribution of these measurements will be referred to as reference number size distribution or reference measurement within this paper. Altogether measurements with nine different pressure levels including the reference measurement were conducted for each nanopowder under investigation.

The number size distribution of the resulting aerosol in the compensation tank was measured using a conventional Scanning Mobility Particle Sizer (SMPS) from TSI (Model

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3080, long-DMA model 3081 in connection with a water based UCPC, model 3786) with a covered size range from 14.1 up to 736.5 nm electrical mobility diameter with a maximum resolution of 64 size channels per decade. The aerosol flow rate was set to 0.3 liters per minute with a sheath flow rate of 3.0 liters per minute. The scan time to obtain one number size distribution was set to 240 seconds to minimize possible scan time effects (Russel et al., 1995). The next scan started after 20 seconds retrace and additional 40 seconds wait time.

In addition to the pre-separation of the cyclones, an impactor with an orifice size of 710 μm was used throughout the measurements as a pre-separator at the SMPS inlet. This impactor was carefully chosen, considering the largest electrical mobility diameter and the particle density. This was crucial since the particle generation system produced fairly large amounts of particles larger than the upper size limit of the SMPS. If sampled by the SMPS, these would bias the results upon multiple charge correction.

The compensation tank was flushed with particle free nitrogen prior to particle generation, assuring a background particle concentration level below 1000 particles per cm^3 . Concentrations were at least between one and three orders of magnitude higher during actual measurements, which were conducted after the number size distribution within the compensation tank, had reached stable conditions. The resulting number size distributions were then repeated five times for each pressure step to obtain a reliable data base and to calculate an average number size distribution.

Table 17: Overview of nanoparticle materials under investigation in this study. Overall five metal oxides and one carbonate were investigated during the measurements. The primary particle size and BET-surface area were provided by the manufacturer, the primary particle shape was investigated by high resolution SEM.

Material	Abbreviation	Primary particle size	Primary Particle shape	BET surface area
Cerium Dioxide	CeO_2 -1	40-50 nm	Spherical (partly irregular)	63 m^2/g
Cerium Dioxide	CeO_2 -2	40 nm	Spherical (partly irregular)	33 m^2/g
Titanium Dioxide	TiO_2 -1	10-20 nm	Spherical	Not available
Titanium Dioxide	TiO_2 -2	Not available	Spherical	40 m^2/g
Titanium Zirconium Aluminium Oxide	TiZrAlO	Not available	Spherical	Not available
Strontium Carbonate (hydrophobized)	SrCO_3	15-30 nm	Elongated	8.9 m^2/g

The nanopowder agglomerates under investigation are summarized in Table 17. Five metal oxides and one carbonate with different properties were investigated. The size range of the primary particles is on the order of 10 nm to 50 nm and quite similar for the different nanopowders (in the case of TiO_2 -2 and TiZrAlO the primary particle size is unknown, but it is most likely within the same size range as the TiO_2 -1). The shape of the primary particles as

investigated by high resolution scanning electron microscopy (SEM) ranges from nearly perfect spheres (the different TiO_2 -modifications) over sphere-like particles (the different CeO_2 -modifications show some edges on the high resolution scanning electron micrographs) to elongated particles with an estimated aspect ratio of about 2-5 in case of the SrCO_3 .

4.1.4 Data Evaluation

For data evaluation a resolution of 16 channels per decade (this corresponds to a size range from 16.5 nm up to 697.8 nm) was used throughout our study. In Figure 54 an example of the average number size distributions as a function of the electrical mobility diameter of CeO_2 -1 is shown for the reference measurement and for four different overpressure values (for the sake of clarity only the reference measurement and measurements at 20, 60, 100 and 140 kPa are displayed within this graph). Figure 54 shows that the reference size distribution has a mode diameter of about 500 nm, which shifts towards smaller diameters with increasing overpressure. Furthermore, the number concentration within each channel and thus the total concentration increases with increasing overpressure.

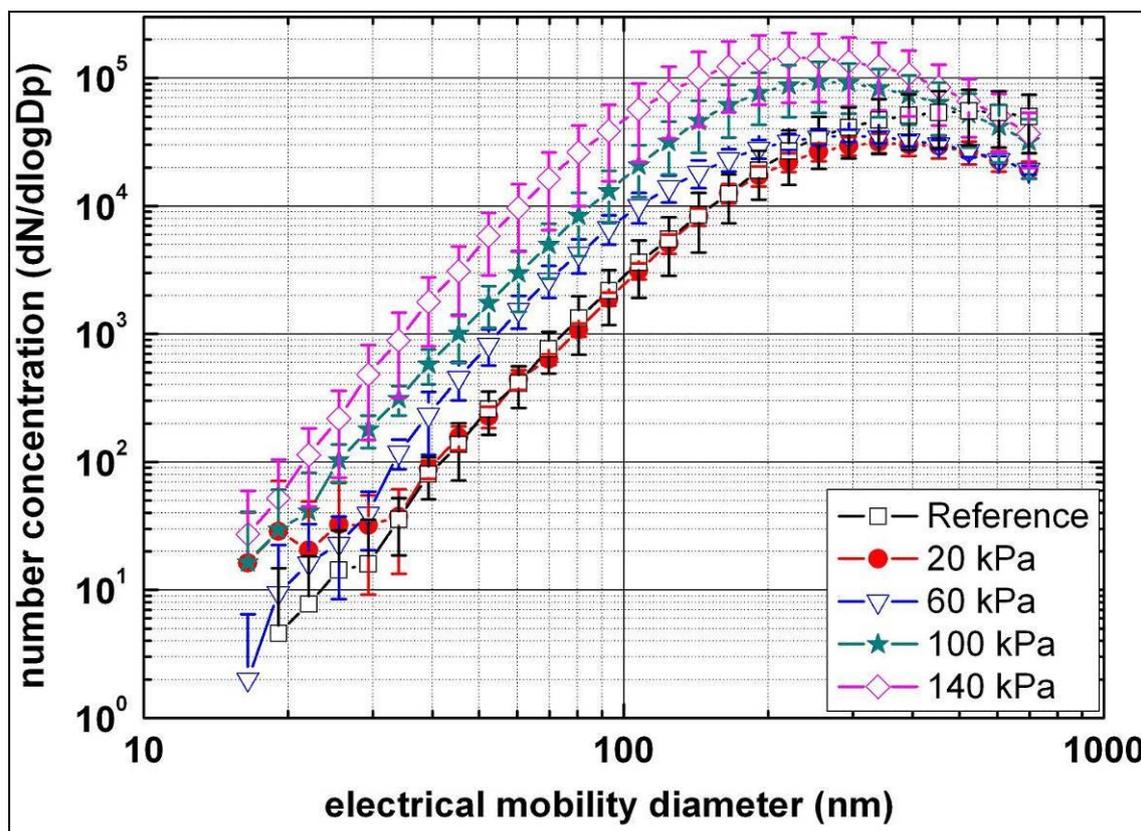


Figure 54: Number size distribution of CeO_2 -1 for the reference measurement and four different overpressure levels.

Due to differences in the total concentration for each substance and also for each pressure step (which results from the aerosol generation process and the passage through the orifice) the obtained number size distributions were further normalized. This allowed for direct

comparison of the different pressure steps for one substance and comparable results for the different nanopowders.

The relative number size distribution was therefore calculated in a first evaluation step by dividing the concentration in each size class by the total number concentration of all size classes. The resulting relative size distributions are shown in Figure 55. In this diagram the shift of the modal diameter with increasing overpressure becomes more visible. Furthermore, the graph shows that the relative number concentration for the higher size classes decreases with increasing overpressure, whereas the relative number concentration in the lower size classes increases with increasing overpressure. This gives rise to the assumption that larger agglomerates break up in the orifice, thus producing smaller agglomerates, aggregates or primary particles.

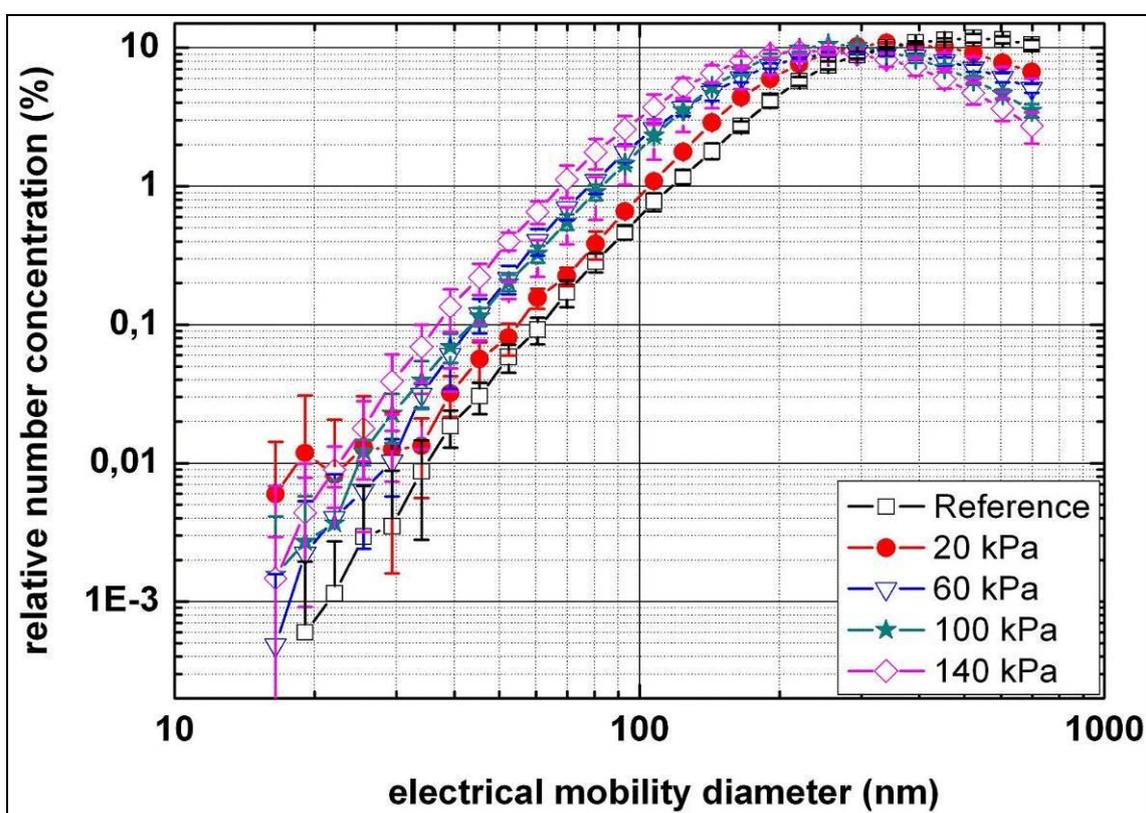


Figure 55: Relative number size distribution of CeO₂-1 calculated from values in Figure 54. In this diagram the shift of the modal diameter towards smaller particles with increasing overpressure is better visible.

In order to better display this process, the relative change of particle concentration within one size class was calculated in a second step for each given overpressure. The relative number concentration in each size channel of one pressure step was therefore divided by the corresponding relative number concentration of the reference size distribution. The results for CeO₂-1 are shown in Figure 56.

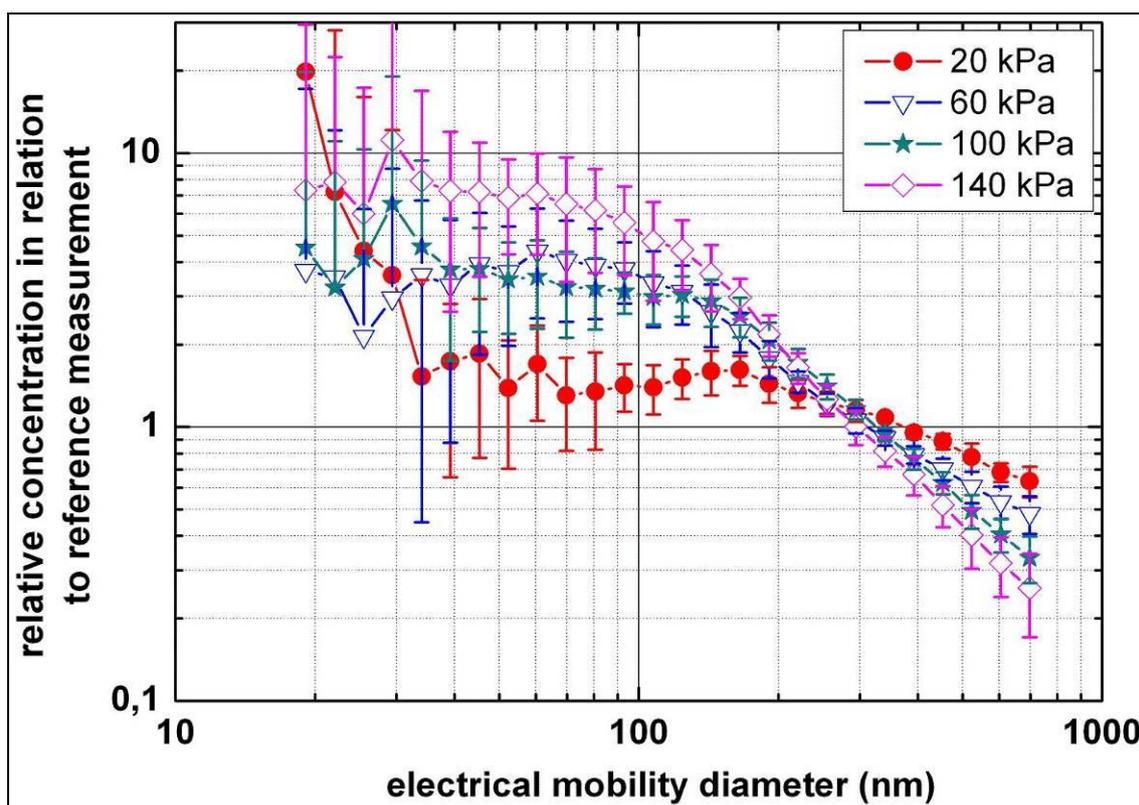


Figure 56: Ratio of the relative number size distribution of CeO₂-1at four different overpressure levels with regard to the relative number size distribution of the reference measurement. An increase in the relative particle concentration for mobility diameters below about 300 nm takes place.

Figure 56 shows the ratio of the four different overpressure measurements with regard to the reference measurement. A ratio above one indicates that the relative number concentration within the according size class is increased and a value below one indicates that it is decreased. It becomes apparent, that the fraction of particles with an electrical mobility diameter below approximately 300 nm is increasing with increasing overpressure, whereas the fraction of particles above approximately 300 nm is decreasing with increasing overpressure. This shows that agglomerates are fragmented in the orifice and that this effect is pressure dependent. For exposure assessment following an accidental release in a pressurized nanopowder production or transport vessel, it may therefore not be sufficient to evaluate only particle sizes as known from the size distribution inside the vessel as these may change when the particles pass a leak.

One complication during the experiments outlined here are possible particle losses due to impaction or diffusion in the sampling lines which were ignored during our study. These particle losses might occur within the orifice (Chen et al., 2007), within the compensation tank or the measurement unit. Also, resuspension most likely in the vicinity of the critical orifice by the turbulent flow might occur (Reeks et al., 1988). Since the whole setup was electrically grounded, it can be assumed that electrophoretic losses are negligible. Possible particle losses, however, only affect number size distributions shown in Figure 54 whereas the size dependent losses cancel each other out during the calculation of the relative size distributions.

4.1.5 Results and Discussion

By applying the method described above the change in the number size distribution of a variety of nanopowders was investigated. The results for an overpressure of 140 kPa are summarized for six different powders in Figure 57. In case of the lower size channels the error bars get larger and there are some data points missing (see for instance the data of TiO_2 -1 or SrCO_3). This is due to the low number concentration within these size channels for the reference measurement which leads to a higher uncertainty for these size classes. If no data for the reference measurement are available at all, the data points for the ratio of the overpressure measurement are subsequently also unavailable.

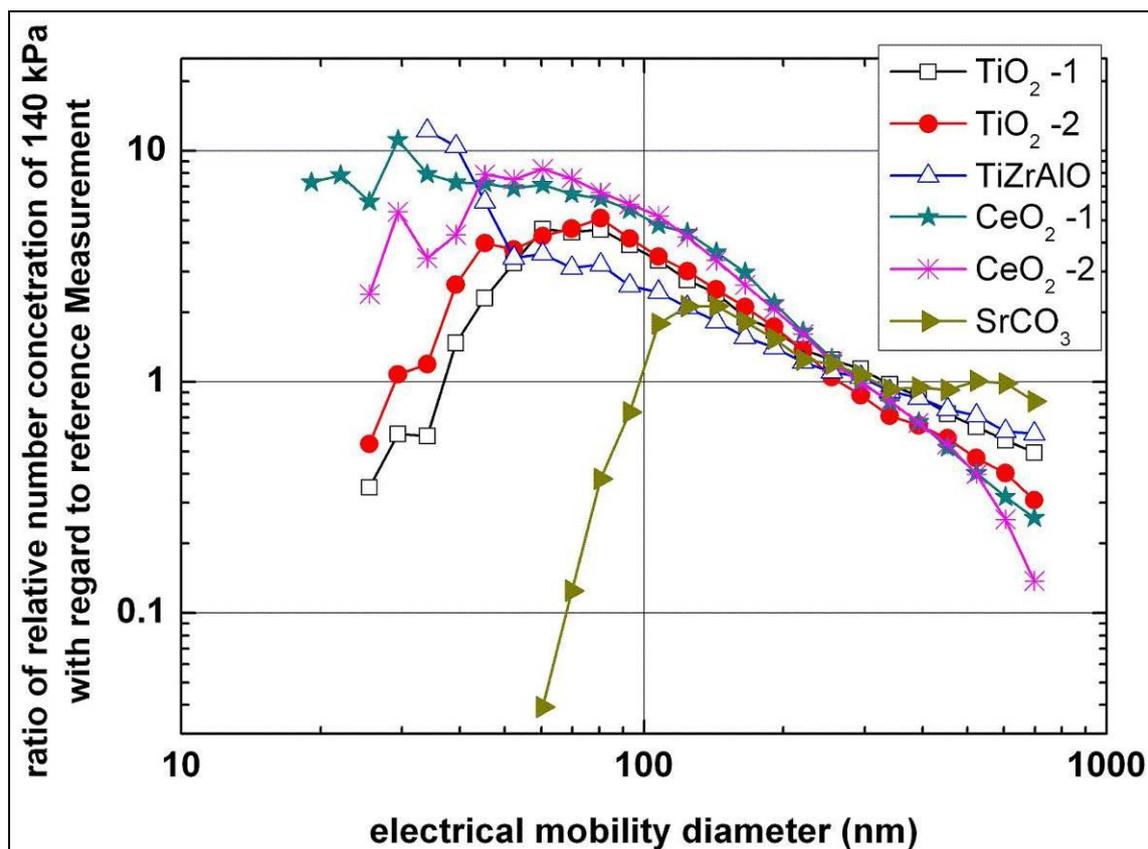


Figure 57: Comparison of the ratio for the 140 kPa overpressure measurements with respect to the reference measurements for six different nanopowders.

As one can see in Figure 57 the different substances show a generally similar behaviour, except for SrCO_3 . All other substances show a clear decrease in the relative number concentration for a size above roughly 300 nm electrical mobility diameter. This is caused by deagglomeration of the agglomerates within this size range. Also, all other substances show a pronounced increase of the relative number concentration below about 300 nm due to the fact that smaller fragments are introduced by the deagglomeration process. For SrCO_3 , the number concentration is increased only in a size range of approximately 100-300 nm, but it steeply decreases below 100 nm. In the size range above approximately 300 nm, the relative concentration is close to unity. It is assumed that the bonding energy between primary particles is higher (as compared to spherical primary particles) for these agglomerates, because of their elongated morphology, allowing for larger contact area. Therefore

agglomerates do not break up as easily, causing the concentration of larger particles to remain almost constant. Still it is surprising that the concentration of small particles decreases so drastically. This may be caused by different losses during the measurement of the reference size distribution and those during overpressure. Furthermore it is noteworthy that concentrations of the SrCO_3 in the sub-100 nm size range were generally quite low leading to an increased uncertainty of the measured number size distributions.

In any case, Figure 57 indicates that the deagglomeration behaviour in the orifice seems to be material dependent. The two different CeO_2 -modifications show the highest degree of deagglomeration with a factor of about seven to eight for the relative increase of particle number in a size range of about 50 nm to 80 nm. The number concentrations for the modifications of TiO_2 showed in the same size range only a five to six fold increase. The TiZrAlO shows only an increase by a factor of about three. For the case of SrCO_3 the maximal increase by a factor of two in the relative number concentration is reached for a size of 130 nm electrical mobility.

To illustrate the relative particle concentration change with pressure for the six substances, the mean ratios of the concentration change for two different sizes are shown as a function of overpressure in Figure 58. The mean value at 100 nm is calculated as the geometric mean value of the four size classes around 100 nm (80.6, 93.1, 107.5 and 124.1 nm) and the mean value at 600 nm is calculated as the geometric mean value of the three size classes around 600 nm (523.3, 604.3 and 697.8 nm). As can be expected from Figure 57 the same trends regarding the behaviour of the different substances are also visible in Figure 58. This is indicated by the fitting of the data points. It should be noted that linear fitting was used only as a first approximation to describe the deagglomeration behaviour as a function of the pressure difference. With more data points available in the future, the relationship may become non-linear. The horizontal black lines correspond to a ratio of one, thus indicating no change in the relative particle concentration. The magnitude of the increase of the relative number concentration for the smaller fragments (top) and the deagglomeration of bigger agglomerates (bottom) are illustrated in this figure. Values below one for the case of the smaller fragments as well as values above one for the case of the bigger agglomerates are due to measurement uncertainties.

As described above, the strontium carbonate shows only a little increase of the concentration in the size range of 100 nm at higher overpressure values and almost no change in the 600 nm size range, indicating that there may not be any deagglomeration. For the case of the different titanium dioxide modifications a more or less continuous increase (with some deviations for single overpressure steps) in relative particle number concentration occurs in the 100 nm size range. The highest increase in this size range was observed for the two different CeO_2 modifications, indicating that these substances are more easily fragmented than the other substances examined here. This can also be seen at the data for the 600 nm size range where the CeO_2 shows the highest degree of deagglomeration.

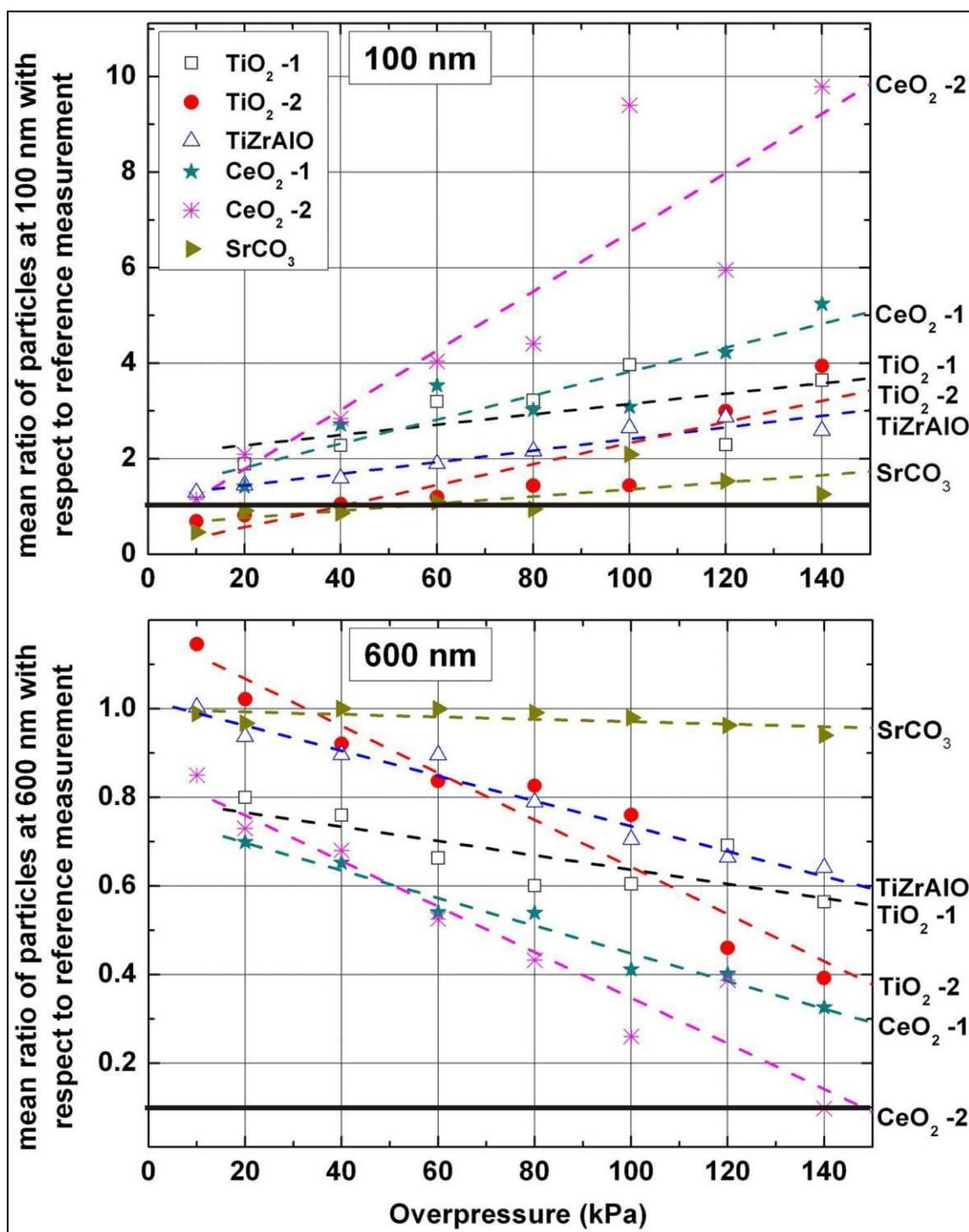


Figure 58: Mean values of the ratio of relative particle concentration for the different overpressure measurements with regard to the reference measurements for a particle size of 100 nm (top) and 600 nm (bottom) respectively.

The differences in the deagglomeration behaviour of the substances evaluated within this study might be due to the shape of the primary particles. Figure 59 shows scanning electron micrographs of three different substances (CeO₂-1, TiO₂-1 and SrCO₃). In case of the CeO₂-1 it is revealed that the primary particles have a sphere-like morphology with some irregularities. In principal the same morphology is found for the CeO₂-2. The TiO₂-1 consists of agglomerates with primary particles having a spherical morphology, which is also true for

the second modification of TiO_2 and the TiZrAlO . The primary particles of the SrCO_3 have, as revealed by the SEM-images, an elongated shape with an estimated aspect ratio of about 2-5.

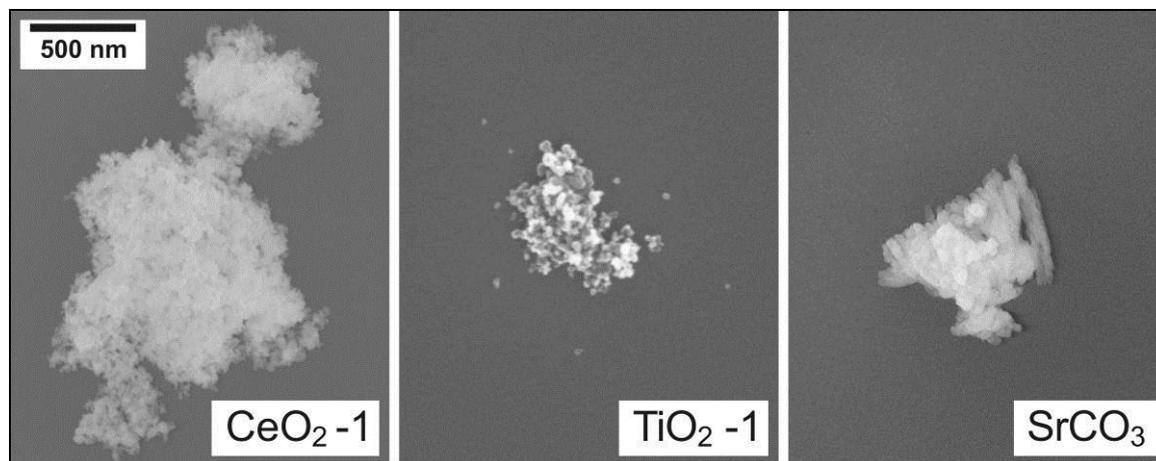


Figure 59: Scanning electron micrographs for three different substances. The morphology of the primary particles consists of spherical particles which are partly irregular (CeO_2 -1, left), more perfect spheres (TiO_2 -1, middle) to elongated particles (SrCO_3 , right). The scale bar is valid for all three images.

In case of the SrCO_3 we find only a low tendency of deagglomeration. The elongated shape of the primary particles might lead to tightly bound agglomerates due to an enhanced contact area and therefore an enhanced binding energy. Once dispersed by the magnetic stirrer, these agglomerates remain intact during the higher mechanical stress occurring within the orifice. In the case of the different TiO_2 modifications the primary particles have a spherical shape and presumably only one contact point between each other. In this case the binding energy is supposed to be lower which is also reflected by the higher deagglomeration as measured within this study. The highest degree of fragmentation into smaller particles was measured for the two CeO_2 modifications. This might also be explained by the shape of the primary particles which is nearly spherical. But in contrast to the TiO_2 some rough edges are also visible in the scanning electron micrographs. Since the *van der Waals* force depends strongly on the distance between the particles, an increase of the distance by some edges with a lower contact area might lead-even in the case of nanoparticles-to a higher fragmentation when shear forces are applied. For the case of bigger particles in the micrometer size range this effect is used by the introduction of nanoparticles to enhance the flowability of such powders (Eber and Zimmermann, 2004, Zimmermann et al., 2004).

Figure 60 shows the fraction of particles below 100 nm (with respect to the total number concentration of the SMPS measurement) as a function of overpressure. The characteristics of the different substances are nearly the same as it is obvious by a comparison of Figure 58 and Figure 60. In case of the reference measurement Figure 60 shows that all substances have a fraction around 1 % of particles with an electrical mobility diameter below 100 nm. In the case of strontium carbonate this fraction is mainly below this value for all overpressure conditions, indicating no significant fragmentation. For the different titanium dioxide modifications the fraction of particles below 100 nm slightly increases to about 3 % and in

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one case to about 5 % of the overall number concentration at an overpressure of 140 kPa. The CeO₂ shows a higher fraction of particles below 100 nm even at moderate overpressure conditions and a value of up to 12 % at 140 kPa overpressure (Table 18).

Table 18: Comparison of the linear fit data based on the data within Figure 59 of the increase of particles below 100 nm for the different nanopowder materials.

Material	Slope of linear fit (%/kPa)	Standard deviation of linear fit (%/kPa)	Correlation coefficient R
CeO ₂ -1	0.0326	0.0068	0.8894
CeO ₂ -2	0.0783	0.0130	0.9160
TiO ₂ -1	0.0125	0.0074	0.5648
TiO ₂ -2	0.0315	0.0076	0.8446
TiZrAlO	0.0213	0.0032	0.9296
SrCO ₃ (hydrophobized)	0.0010	0.0025	0.1531

To establish a more reliable ranking of the different materials under investigation a linear fit of the data in Figure 60 was made as a first approximation of the increase of particles below 100 nm as a function of increasing pressure difference. A higher slope indicates a stronger deagglomeration of the material. As can be expected from the results discussed above, the two CeO₂ materials have the highest slope with modification 2 (slope: 0.0783 %/kPa) showing about a doubled deagglomeration behaviour when compared with modification 1 (slope: 0.0326 %/kPa). These values might be linked to the BET surface where the modification 2 has a value which is about 50 % of the value of modification 1, i.e. the tendency of deagglomeration seems to be inversely proportional to the BET surface area for these materials. For a comparison of different substances the BET surface area is not indicative to predict the dispersability. This is obvious for the case of TiO₂-2 (slope: 0,0315 %/kPa) with a BET surface in the same range as the CeO₂-2 but a slope in the same range as the CeO₂-1. In case of SrCO₃ with a slope of 0.001 %/kPa there seems to be only a vague dependency of the deagglomeration on increasing pressure difference. This is also confirmed by the correlation coefficient R: In case of the SrCO₃ there is nearly no linear relationship between the data points (R = 0.1531). For the TiO₂-1 the relationship is also rather bad with a value of R = 0.5648. The other substances show a good correlation coefficient with values between R = 0.8446 up to R = 0.9296, indicating that a linear fit might be used as a first approximation for the increase of particles below 100 nm with increasing pressure difference.

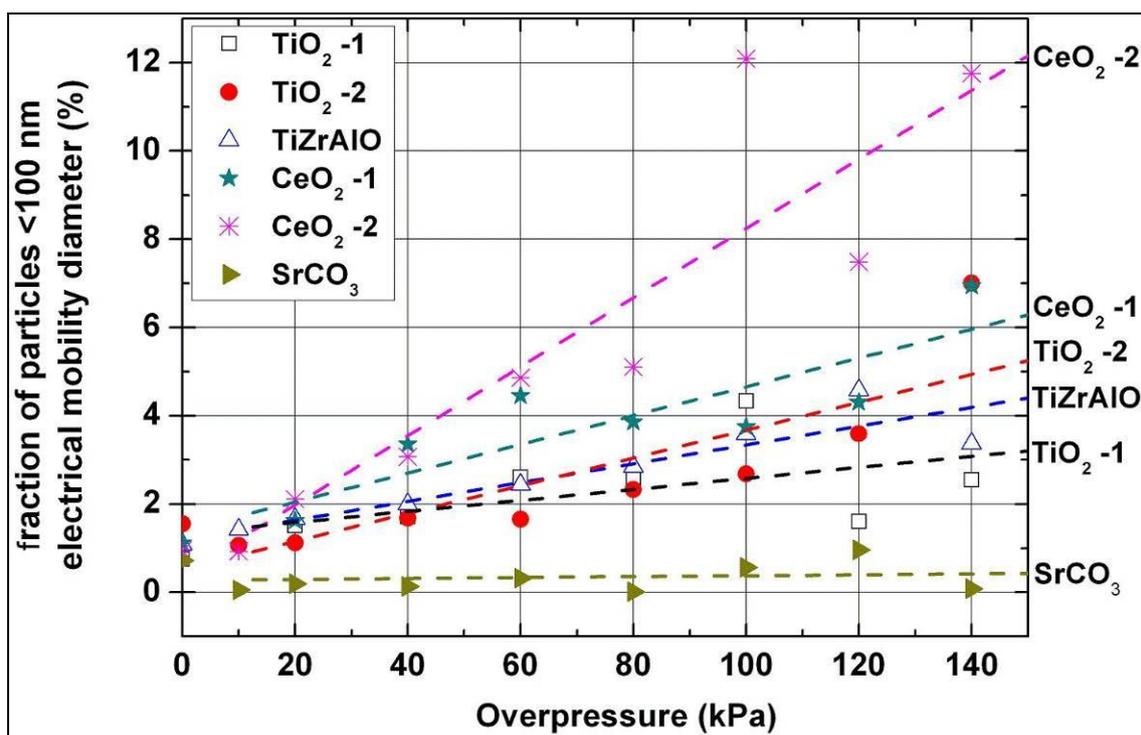


Figure 60: Fraction of particles with a size below 100 nm (with regard to the total concentration).

These results provide evidence for a connection between the shear forces occurring within a (critical) orifice and the number size distributions of the aerosolized nanopowders. Due to the influence of shear forces, agglomerates can get separated into smaller fragments, depending on their material. Therefore, the relative number of particles in the higher size classes is decreased, and the relative number of particles in the lower size classes increases by a factor of up to ten. Furthermore, the influence of the shear forces is increasing with increasing overpressure, as it can be expected due to the higher energy within the orifice.

From a microscopic point of view an agglomerate of a size of some hundred nanometers consists of thousands of primary particles. In the simplest case of spherical (equally sized) particles, one particle can have up to six bonds with the surrounding particles. The number of bonds and thus the binding energy strongly depends on the unknown local morphology of the agglomerate. A detachment of (smaller) fragments, i.e. primary particles, aggregates or smaller agglomerates, most likely takes place at the weakest link between the fragments. In case of agglomerates with a diameter of some hundred nanometers this might be between two more or less equally sized fragments. Small fragments are more likely detached from the outer parts of an agglomerate. Due to the vast number of primary particles and aggregates forming an agglomerate the process of deagglomeration is stochastic. A downsizing of the agglomerates for a given shear force is therefore obvious but no preferential size class for the fragmented parts can be expected. This behaviour is observed within these experiments: with increasing overpressure the size distribution is continuously shifted towards smaller particles.

The differences in the deagglomeration between the substances presented here can presently not be traced back to intrinsic material parameters like the Hamaker constant of the materials, because this is presently not well determined for most nanomaterials. The binding energy between primary particles of the agglomerates remains also unknown and cannot be

calculated due to the complex shape of the agglomerates and the unknown number of primary particles.

4.1.6 Conclusions

We have presented a method to compare the stability of airborne nanoparticle agglomerates. After the powder, containing agglomerates and aggregates of primary particles within the nanometer range, is dispersed under different overpressure conditions it passes a (critical) orifice. This method can be considered to simulate a leak in a pressurized nanoparticle production or transport line. During the passage through the orifice the agglomerates are fragmented due to the shear forces present within the orifice. The magnitude of the shear forces and thus the degree of fragmentation depends on the overpressure at the inlet of the orifice. A clear trend to a higher degree of fragmentation is found with increasing overpressure. The degree of deagglomeration furthermore seems to be material dependent. The highest degree of fragmentation is found for CeO₂ powders with nearly spherical primary particles containing some edges. In contrast, SrCO₃ with elongated primary particles shows no clear fragmentation when passing the orifice. This method is sensitive to the measurement of small particles and is used for a differentiation between nanopowder materials. For exposure assessment, especially important following an accidental release from a pressurized vessel, these results show that it is not sufficient to know the particle size distribution inside the vessel, but also to evaluate the resulting size distribution after passing the leak.

Further studies on the deagglomeration taking place during agglomerate flow through an orifice are currently under way. Within these studies other materials consisting of primary particles within the nanometer size range are being tested to provide a more detailed “ranking” of these materials with regard to their morphological features (i.e. primary particle size and shape, BET-surface). Also, these measurements will provide a more quantitative analysis of the degree of fragmentation as a function of more general quantities like the flow Reynolds number and the intrinsic material properties of the different nanopowders.

Acknowledgement

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4.2 Nanoparticle Exposure at Nanotechnology Workplaces: A Review (Kuhlbusch et al., 2011)

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4.2.1 Abstract

Risk, associated with nanomaterial use, is determined by exposure and hazard potential of these materials. Both topics cannot be evaluated independently. Realistic dose concentrations should be tested based on stringent exposure assessments for the corresponding nanomaterial taking into account also the environmental and product matrix. This review focuses on current available information from peer reviewed publications related to airborne nanomaterial exposure. Two approaches to derive realistic exposure values are differentiated and independently presented, those based on workplace measurements and those based on simulations in laboratories. An assessment of the current available workplace measurement data using a matrix, which is related to nanomaterials and work processes, shows, that data are available on the likelihood of release and possible exposure. Laboratory studies are seen as an important complementary source of information on particle release processes and hence for possible exposure. In both cases, whether workplace measurements or laboratory studies, the issue of background particles is a major problem. From this review, major areas for future activities and focal points are identified.

4.2.1.1 Introduction

Research and product developments in the area of nanotechnology have steadily increased especially due to new, beneficial properties of nanomaterials. Nanotechnology as a cross-cutting technology, nowadays used in electrical devices, in construction and composite materials, as catalysts and as antibacterial coatings, is more and more present in workplaces as well as consumer products. This steady increase is accompanied with larger production, handling and processing facilities for nanostructured materials and higher tonnage of nanomaterials.

New nanomaterials, an inherent part of nanotechnological developments, allow on the one hand new products and solutions to e.g. societal problems related to natural resources, drinking water, energy generation and storage, but also raise concerns due to their new specific properties. The major concern is, that the new properties and the high mobility of some nanomaterials may lead to health or environmental effects. This concern has been identified early and was taken seriously by public bodies and the industry. First specific research investigations in toxicology related to particles at the nanoscale were already conducted in the late 1980's [1]. During the last two decades the amount of toxicological research on nanomaterials has increased from less than 10 publications before 1998 to more than 200 in 2010 (ISI Web of Knowledge, 02/2011). A risk, however, may only arise if both a hazard potential of the nanomaterial and exposure exist. Therefore first studies of workplace related exposure were initiated by the International Carbon Black Association (ICBA) in 1998 [2, 3]. In parallel Maynard et al. [4] conducted first studies related to nanomaterial exposure of carbon nanotubes (CNT). The number of workplace studies and published results has

increased significantly since then and a first ISO-guideline on inhalation exposure characterization and assessment has been set-up [5].

Different approaches can be pursued to derive exposure relevant information in workplaces: (a) Studies based on real workplaces and (b) process based studies in simulated workplaces and of simulated work processes. The major advantage of the prior approach is that data from real work conditions are obtained. Still, due to various background aerosols originating from the general work environment or the process itself, extensive and time consuming measurement campaigns have to be conducted. The latter approach, based on simulations in laboratories allows the clear differentiation of a release from the investigated process from background aerosols coming from other sources than the process. It has to be noted that background aerosols may stem from areas outside of the work area and from the process itself. The latter can again be divided into background aerosols released from e.g. electrical motors used in the process [6, 7, 8] or unintentionally produced particles from the production process as a side product. This clear differentiation of background aerosols is also necessary to understand what type of background aerosol can be differentiated by which measurement approach.

Process based studies in simulated workplaces and of simulated work processes also enable investigations on how variances in handling, process conditions influence release rates. Release rates are important pieces of information for workplace modelling approaches as investigated and described in detail by Schneider et. al. [9]. Still, laboratory simulations do not represent the real work conditions but represent an important complementary way to derive information on possible nanomaterial exposure at workplaces.

The purpose of this manuscript is to present an overview of publications related to the exposure of engineered nanomaterials in the workplace and processes which may lead to such exposure. Workplace investigations and laboratory simulations are presented and discussed separately since strategies and methodologies employed differ significantly for both approaches. In some cases references to investigations are given in both parts. Those are valuable studies linking process studies with real work conditions, even though this quite often only refers to small bench workplaces.

4.2.2 Methods, Devices and Measurement Strategies for Airborne Nanoobjects and Nanomaterials

Methods and Devices

Measurement and sampling devices for airborne nanoscale particles used in workplace exposure studies can generally be divided into four types, i.e. any combination of size resolved and size integrated with time resolved and time integrated as shown in Table 19.

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Table 19: Selected measurement and sampling devices for airborne particles

Measurement/sampling device	Size range / time resolution / metric + equivalent diameter	References
Size resolved <i>Time resolved</i>		
Scanning mobility particle sizer (SMPS)	2.5 nm-1000 nm > 30 s number PSD, based on electrical mobility diameter	[10, 11]
Electrometer based mobility particle sizer: fast mobility particle sizer (FMPS) / engine exhaust particle sizer (EEPS)	5.6 nm-560 nm 1 s / 0.1 s number PSD, based on electrical mobility diameter	[12, 13]
Electrical low pressure impactor (ELPI)	6 nm-10 µm 0.1 s number size distribution, based on aerodynamic dia.	[14]
Optical particle sizer (OPS): - laser aerosol spectrometer (LAS)	(> 60 nm) >300 nm-20 µm 1 s number PSD, based on light scattering equiv. dia.	[15]
Inertial spectrometer / time of flight instruments: - aerodynamic particle sizer (APS)	500nm-20µm 1 s number PSD of the aerodynamic dia.	[16]
Size resolved <i>Time integrated</i>		
Low pressure cascade impactor	> 20 nm n.a. mass size distribution, chemical analysis, morphology	[17, 18]
Micro orifice uniform deposit impactor (Moudi)	10nm -20µm n.a. mass size distribution, chemical analysis, morphology	[18]
Wide range aerosol system (WRAS)	5.5 nm-32 µm 5 min number size distribution	[19]
Thermal precipitator (TP)	20-ca. 300 nm n.a. size distribution	[20]
Size integrated <i>Time resolved</i>		
Condensation particle counter (CPC)	5.5 nm-9 µm 1 s particle number concentration (NC)	[21, 22]
Surface area monitors (e.g. electrical aerosol detector (EAD), nanoparticle surface area monitor (NSAM), LQ1-DC)	10 nm->1 µm 1 s aerosol length (EAD), active surface area (LQ1-DC), lung deposited surface area (NSAM)	[23 - 25]
Aerosol photometer	250 nm-20 µm 1 s Mass concentration	
Size integrated <i>Time integrated</i>		
Electrostatic precipitator (ESP)	> 20nm n.a. chemical analysis, morphology	[26]
Thermal precipitator (TP)	> 20 nm n.a. chemical analysis, morphology	[20]
Filtration (e.g. PM10, PM2.5)	mass concentration, chemical composition	

Size resolved, time resolved: These devices usually use detection principles based on the particles' optical properties or electrical mobility. Hence the results are based on optically or electrically equivalent spheres. In electrical mobility analysis, the equivalence is only valid for the given particle orientation in the classifier, which in case of non-spherical particles may be unknown and hence bias the measurement accuracy. Handling and calibration of mobility particle sizers are standardised in ISO 15900:2009 [27]. Problems of equivalency for different particles also exist for optical detection methods if optical properties vary significantly, e.g. carbon black versus TiO₂. It also is important to take the time resolution into account. Particle size distributions may vary in time scales of seconds which is not resolved by the SMPS (3-5 minutes time resolution) but often used for such measurements. Hence fast particle sizers like FMPS or ELPI should be employed for example to analyse fast changes in the particle size distributions.

Size integrated, time resolved: The particle size weighting is dependent on the metric. Number based detectors give the same signal weight to each particle, while geometric surface area based devices increase the signal weight according to the particle diameter squared. Condensation particle counters (CPC) detect particles from a few nanometres to a few micrometres [22], depending on the model used. Contribution of particles to the surface area is often below detection limit at particle sizes below approximately 20 nm due to the metric immanent weighting of particle size distributions towards larger particles. Diffusion charger based surface area monitors are most accurate in a size range from approximately 20 nm to 400 nm [28]. The lower detection limit is usually not critical due to the low surface area contribution of such small particles in most real particle size distributions. The upper limit, however, may cause more significant errors, because even a small number of these large particles can have a significant contribution to the total surface area.

Size resolved, time integrated: Stahlmecke et al. [29] showed that particles tend to further deagglomerate with increasing pressure difference across an orifice. Hence devices like the low pressure impactor may lead to significantly artificial change of the particle size distribution in the measuring device. A second major error using time integration is the change in physical and chemical characteristics during sampling on the substrate due to interaction with other particles or the gas. One example is the adsorption of volatile organic gases onto collected particles and the substrate.

Size integrated, time integrated: The ESP or TP as shown in Table 19 can efficiently sample down to 20 nm and below. While both types of samplers can generally also sample micrometre sized particles, they can easily be lost in the sampling lines or inlet system. Thermophoretic deposition is more or less independent of particle size up to approximately 400 nm. Hence TPs can provide a homogenous deposit of particles which can also be evaluated for the particle size distribution [20]. The sampling efficiency for fibrous particles in TPs has to be investigated since inefficiencies in collection may exist.

Common measurement methods of time integrated samples applied after collection are the bulk chemical analysis of a filtration or cascade impactor sample, morphological studies using electron microscopy (SEM, TEM) and single particle chemical analysis using Energy Dispersive X-ray analysis (EDX).

More detailed information on measurement techniques for airborne nanoscale particles are given in [15, 30-32].

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Despite all the progress made during the recent years with regard to measurement techniques and strategies for exposure assessments towards airborne nanomaterial, there is still the lack of clear performance criteria to achieve comparability and hence nanomaterial specific workplace safety criteria. The development of easy to use devices for the measurements by hygienists is urgently needed even though no agreement on the best metric exists. First personal samplers for time resolved nanoparticle measurements have been recently developed. Such devices can currently not be employed in the breathing zones of workers. In summary, we currently lack information on personal exposure and rely on estimates based on static measurements and first measurements with recently developed devices.

Measurement Strategies

Measurement techniques and measurement strategies have to be optimally combined to allow sensitive and cost effective determination of airborne engineered nanomaterials at workplaces. Measurement strategies can mean tiered approaches as has been suggested by [33] and [34]. Tiered approaches may facilitate cost effective screening of many workplaces if sensitive sensors exist. The NEAT strategy, as developed by NIOSH [33], applies a handheld device which determines particle number concentrations. Particle number concentrations are quite sensitive for nanoscale particles and thus a real-time screening allows identification of possible “hot spots”. If the initial screening reveals that the workplace is “clean”, i.e. measured concentrations are below a certain threshold value, no further investigations may be necessary. If a nanomaterial release is suspected due to increased concentration levels, the next step of investigations of the tiered approach will be more detailed. This can be personal exposure related, process related or approaches closely linked to toxicological and epidemiological questions.

Directly after the screening detailed measurements related to the above mentioned aims and approaches may be pursued in the areas, which are possibly affected by airborne nanomaterial. Personal exposure approaches will either be based on personal devices and samplers or areal measurements combined with the recording of personal activity patterns to allow the calculation of personal exposure. So far no legal binding framework concerning nanoparticle specific limit values exists. Personal exposure studies related to nanomaterial exposure are therefore dominantly linked to research studies. A good discussion can be found in Maynard and Aitken [35].

Process related approaches will mainly be based on real measurements, allowing for the concurrent use of several and more sensitive devices. Measurement locations will be often close to the process, handling or work activity of interest.

Approaches with the aim of deriving detailed information for toxicological and/or epidemiological studies will use a variety of sampling and measurement devices with possibly health relevant metrics. The variety of devices used for this purpose is high as the metric which best fits to nanomaterial related health effects is still under discussion.

One considerable problem is the distinction of nanomaterial from the background aerosol. Four basically different approaches for background distinction can be differentiated as follows:

- time series approach,

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- spatial approach,
- approach based on comparative studies with and without nanomaterial,
- (size resolved) chemical and/or morphological analysis.

The different background distinction approaches are part of the measurement strategies presented above. The task to be tackled determines the combination of needed measurement strategy and devices. Time series analysis is generally coupled with online detection methods. This analysis basically assumes that the concentration determined during no work activity is the background concentration and any increased concentrations during the work activity can be attributed to the process, the nanomaterial or both.

The spatial analysis assumes that a background measurement location is representative for the background at the workplace of interest. Any difference between the determined background and workplace concentrations can be linked to the work activity and the nanomaterial investigated.

Most of the studies published so far use a combined approach of time series and spatial analysis, or link the spatial analysis with morphological or chemical analysis (see Table 20). Morphological analyses are often included when single particle chemical analyses are conducted (SEM-EDX) or distinct features of the nanomaterial can be used for its identification like a fibre structure.

Table 20: Measurement Strategies for background distinction (only articles with original measurements)

	With/without activity	With/without nanomaterial
Time series analysis	[36], [37], [38], [39], [40], [41]	[42] ^{2,3}
Time series and, or only spatial analysis	[36] ¹ , [38] ¹ , [43], [44], [33] ^{2,3} , [45], [46], [47], [2] ² , [3] ² , [48], [50], [49], [51], [52] ² , [53] ^{2,3} , [54] ^{2,3} , [55] ^{2,3}	

¹: only in some cases

²: Additional background distinction by chemical analysis (filtration sample or single particle analysis)

³: Additional TEM or SEM analysis

⁴: Nearly clean room conditions: [4], not in table

Two approaches are represented by only one published study each (see Table 20). The approach based on comparative studies with and without nanomaterial was published only once [35] but according to the authors' personal communications is often used in industry. This approach assumes any handling and processing of the composite matrix without nanomaterial to be the point of comparison and any changes during handling or processing of the composite with nanomaterial can be attributed to the nanomaterial.

The "clean room approach" was not set up for the assessment of possible worker exposure but needed to avoid product material contamination by ambient dust. Nevertheless, the case in a facility producing carbon nanofibres which flushed a tent with nearly particle free air is an

excellent example to avoid background particles from the outside. Still by-product particles either from the production process itself or from other sources such as electrical devices may influence the measurements.

Overall 25 studies related directly to workplace exposure were identified in the literature. More than 50 % of these pursued the combined approach based on time series and spatial analysis. Quite often this approach was backed up by chemical and/or morphological analyses since none of the online devices for the detection of airborne nanomaterials is sufficiently selective to unambiguously determine product nanoobjects or their agglomerates. The combined use of the measurement devices significantly enhances possibilities for the distinction of nanoobjects from the background.

4.2.3 Exposure Related Workplace Measurements

For this review, we have taken into account exposure related nanoparticle measurements at workplaces, where engineered nanoobjects and their agglomerates or products using nanomaterials are processed or used. These include industrial production facilities, processing plants, pilot plant investigations, crafting of nanomaterials (drilling, sawing etc.) as well as research related work area settings. Table 21 and 22 summarize the reviewed articles. The work activities investigated can be structured along the production pathway: production, handling and refinement of the raw material, bagging and shipping of the nanomaterial, processing of the nanomaterial and work processes with the nanomaterial product. No strict separation according to types of nanomaterials (suspension or powders) is pursued but are indicated by the process investigated, e.g. production via the liquid phase.

Production: Nanoobjects are either produced top-down by milling and grinding of bulk material or bottom-up starting from nucleation with subsequent particle growth by condensation and/or coagulation. The bottom-up process is the most common industrial synthesis route for nanoobject production. Two major process parameters significantly influence the possible release of nanomaterial: production via the gas [e.g. 51] or liquid phase [41] and production in an open [45] or closed process [2, 3]. Gas phase production process can further be differentiated into nucleation-condensation (metals), vapour deposition (CNTs) and gas-to-particle conversion via oxidation (metal oxides). The latter differentiation itself does not influence the possibility of release but may determine whether the process is open or closed (references given after the brief description of the process indicate corresponding exposure related studies).

Handling and refinement: Once the nanomaterial is produced, it is removed from the process by filtration or opening of a reactor. Relevant, sometimes manual work steps are filtration [3], pelletizing [2], cleaning of raw nanomaterial [56], drying [41], grinding and milling [56].

Bagging and shipping: The next working steps are bagging and shipment of the nanomaterial. The nanomaterial may either be handled as liquid suspension or dry powders. The latter is often discussed and investigated as a source for airborne exposure [2, 4] while the prior significantly decreases the likeliness of airborne release. No investigations related to bagging/filling and shipping of suspensions have been published so far.

Processing: The area of activities related to processing of nanomaterials covers a wide range such as mixing nanomaterial powders in liquids, cutting, drilling etc. of composites, drying

and spraying. Therefore one single study can rarely cover all processing activities of one nanomaterial. E.g. the handling and mixing of CNF was investigated by Mazzuckelli et al. [52] but similar processes for TiO₂ or other nanomaterials are not published. We may differentiate processing steps with the nanomaterial itself as a powder or in a suspension and those processes, e.g. drilling or cutting, when the nanomaterial is embedded in a matrix.

Work process with nanomaterial products: the use of nanomaterial coated products at medical workplaces, workplaces using nanomaterial products, such as dry cleaners, parquet sanding are also nanomaterial related workplaces. Exposure scenarios towards the nanomaterial for these workplaces have so far not been published to our knowledge and are not further covered in this review.

An overview on processes and investigated materials is given in Table 21. It can be seen that most of the materials and processes have been investigated. Still quite a few processes were only part in overview studies, e.g. by Methner et al. [33] or Möhlmann et al. [43], which summarize results but do not go into detail and give no clear results whether the actual nanomaterial was released or not.

Table 21: List of workplace processes and nanomaterials being investigated for possible exposure

Nanomaterial	Production	Handling & refinement	Bagging & shipping	Processing	
				Powder or suspension	In a fixed matrix
Carbon Black	[3]	[3], [37]	[2], [37]	[38]	
CNT, CNF, fullerenes	[4], [33], [46], [48], [49], [50], [51], [54]	[38], [33], [4], [46], [45], [50], [49] [54]	[4], [45]	[38], [45], [50], [51], [52]	[42], [52]
Ag	[41], [55]	[41]		[33], [53]	
TiO ₂	[39], [56], [55]	[39], [33], [56]			
SiO ₂	[39], [43]	[39], [43]	[36]	[43]	
Al ₂ O ₃	[39]	[39]		[44], [53], [54]	[44], [54]
Metals	[39], [33], [51]	[39], [33]		[33]	[33]
Metal oxides	[39], [43], [33], [45]	[39], [43], [33], [45]	[45]	[43], [33], [45], [47]	
Others	[39]	[39]		[40]	[40]

The lack of a harmonized approach concerning measurement strategies and techniques, metrics and size ranges as well as data analysis procedures complicates the summary of the studies listed in Table 21 and 22. The different measurement strategies and measurement devices were introduced in the prior section. From Table 22 it is evident that all studies used particle number concentrations (NC), either directly determined by a CPC or derived from particle size distribution (PSD) measurements, in their analysis for possible exposure. This is not surprising since number concentration measurements are relatively easy, cheap and very sensitive towards nanoparticles. The main problem for a combined assessment of health effect studies related to NC and PSD is that no defined lower detection limits for particle size are used. This leads to difficulties in the comparison of absolute values, especially in case of number concentrations, where contributions of sub-10 nm particles can be significant.

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21 out of the 25 studies summarized here also determined the particle size distribution down to below 100 nm particle size. This is seen to be essential to actually allow for the differentiation of larger agglomerates from the more mobile particle size fraction below 100 nm. While the time resolution of NC measurements is normally relatively high (1 Hz), time resolution of PSD devices may become crucial when studying processes with quickly changing aerosols. It is also important to note that determined size distributions, either by electrical mobility or optical properties are based on diameters of electrically or optically equivalent spheres, which may not be very meaningful for non-spherical particles.

Slightly more than half of the studies determined also the mass concentration (MC). Various devices, e.g. optical particle sizer based measurements, stationary and personal filtration samplers, and PM1, PM2.5, PM10 samplers were employed. The use of filtration samplers and MC determination is seen to be important to enable the linking with conventional workplace exposure assessment and legislative limits. Nevertheless, this metric is dependent on the particle volume, i.e. particle diameter to the third power and is hence very insensitive to nanomaterials in the lower submicrometer size range.

The importance of the use of electron microscopy (EM) for nanomaterial exposure related studies is stressed by the fact that 12 studies included this time consuming analysis. Even though the use of EM may not be seen as quantitative it is still the only method, which allows for a definitive identification of product nanomaterial, especially when linked with single particle chemical analysis. Good examples are here the detection of CNTs and CNFs.

Eight studies also used consecutive chemical analysis to derive more information on whether the airborne material may have been the nanomaterial in use. Bulk chemical analysis can be used for the different size fractions as indicators, but only single particle analysis can provide definitive differentiation from the background.

The possible importance of the particle surface as exposure metric was presented by Oberdörster et al. [57] and stressed by several other researchers [58, 59]. Easy to use, portable devices became available only in recent years and 4 studies already used these devices. None of the currently available measurement devices measures the geometric surface area of particles, but the lung deposited surface area or “active” surface area. Although the delivered metric is not the geometric surface area of the particles, the devices output can directly be linked to health relevant toxicological reaction mechanisms. The practicability is a further advantage of this measurement technique. On the other hand, this metric is increasingly insensitive for decreasing particle sizes.

The lack of any uncertainty and detection limit discussion in all of the studies is another problem in the assessment of the published exposure related studies at workplaces. Kuhlbusch et al. [56] have recently published a first uncertainty estimate for the method they employed. They showed that no single uncertainty value can be given due to the high dependency on local conditions, like variance in background concentrations. If two or more devices are used simultaneously to obtain a spatial distribution, the uncertainty is furthermore influenced by the measurement accuracy and intercomparability of the measurement devices. Even two devices of the same type can show deviations up to approximately 30 % [60, 44].

Some general conclusions on the focal point, possible airborne exposure to nanomaterials, can be drawn from the studies. 22 of the 25 studies indicate release of nanomaterial particles > 100 nm. The remaining three studies reported increased values in the work area but stated that the elevated concentration may not be attributed to the nanomaterial. Two of those were

laboratory scale production studies related to CNTs. This ambiguity in the identification is much more common for the reported values for particles below 100 nm particle size. 13 of the 25 summarized workplace studies in Table 4 indicate release of nanomaterial below 100 nm particle size. But, only few studies were able to clearly identify the nanomaterial, Kuhlbusch et al. [3], Fujitani et al. [48], Tsai et al. [49]. In other cases, results were much less clear as in Yeganeh et al. [46] and Evans et al. [50]. This listing shows that the identification of nanomaterial exposure is not straight forward and that well described and harmonized methodologies are needed a) to unambiguously link elevated particle concentrations to the nanomaterial under investigation and b) come to quantitative results. The latter is extremely difficult and possibly needs process studies on the laboratory scale.

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Table 22: Summary of the workplace related exposure studies

	Workplace	Type of activity	Nanomaterial	Metric	Results-remarks
[36]	Toner and printing inks industry	Bag emptying of powders	Fumed silica	NC (< 1 µm), PSD (< 1 µm), ASA (< 1 µm), morph., CC	Significantly increased ¹ NC (>100 nm) and ASA detected during bag emptying, confirmed by TEM analysis
[37]	Industrial manufacturing plant	Manual packaging, warehouse, pelletizing	carbon black	PSD (< 1 µm), LDSA (< 1 µm)	Higher NC and LDSA concentrations during activity than during non-activity
[46]	Small commercial nanotechnology production facility	Production of fullerenes (arc reaction), sweeping, vacuum cleaning	Fullerenes	PSD (14 nm-673 nm) , PM2.5 MC, PAH MC	Slightly elevated NC in work area compared to background at one day out of 4 possibly related to cleaning of fume hood, Very good containment of the nanomaterial in the fume hood (production and handling area)
[45]	Industrial production	Metalloxiide production (gas burner) and embedding into a porous oxide matrix, bagging, handling, cleaning and maintenance	MeO (no further information)	NC (10-1000 nm) PSD (14-760 nm), MC PM1 (0.1-1000 nm).	Long term study on possible release of nanomaterial, Significant release of nanomaterial by 'open' production line, handling and cleaning < 1000 nm, Increased NC < 100 nm concurrent with production activity.
[47]	Industrial production	Wet mill	Lithium titanate metal oxides	NC (10-1000 nm), PSD (300 nm-10 µm)MC (respirable fraction), CC, morph.	Only large agglomerates have been detected
[2]	Industrial production	Bagging areas of three plants	Carbon black	PSD(15-675 nm), MC, NC, CC	No significant release of nanoparticles detected, release of agglomerates (> 400 nm) of nanoparticles in all cases of bagging detected if open systems were used, Other sources also significantly influence nanoscale particle concentrations

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	Workplace	Type of activity	Nanomaterial	Metric	Results-remarks
[3]	Industrial production	Production and pelletizer areas of three plants	Carbon black	PSD(15-675nm),MC,NC, CC	Significant release of nanoparticles ($> 10^6$ #/cm ³) and their agglomerates detected in case of a leak in the pelletizing area, in case of good maintenance no significant release of NP from closed production and pelletizing processes, other sources significantly influenced particle number concentrations
[48]	Industrial production	Bagging and agitation including use of vacuum cleaner during these work steps	Fullerenes	PDS (15nm-10 μ m), morph	Release of particles < 100 nm were observed during bagging and vacuum cleaning, also release of particles > 2 μ m was observed during all work steps, including agitation.
[50]	Industrial production	Production and processing (bagging, handling CNF in dryer, thermal treatment, removal from dryer)	Carbon nanofibers	NC, MC respirable , ASA, photoelectric response, CO and CO2	Elevated NC and MC indicate release of significant amounts of nanoscale particles and their agglomerates, no definite indication on release of single and agglomerated carbon nanofibres.
[51]	Industrial production pilot plant, Industry processing	Production and maintenance (silicon), extrusion of CNT nanocomposites	Silicon, CNT	PSD (5-600 nm), NC, ASA	No changes in PSD and NC was observed during production, but spikes during cleaning of mostly agglomerated silicon (> 200 nm), High NC concentration observed in the extrusion area, but no specific CNT detection method was employed,
[41]	Industrial manufacturing facility	Liquid phase process, drying, grinding, handling,	Silver	PSD (15nm-675nm), morph	Significant release of particles < 100 nm as well as of agglomerates was observed during all processing steps as soon as the reactor, dryer and grinder were opened, leading to possible exposure even for wet production processes
[56]	Industrial manufacturing	Production, filtration, bagging	TiO ₂ , Al ₂ O ₃	PSD (5-600 nm), MC PM1, CC, morph	Wet and combustion production processes were compared and no significant release of particles < 100 nm observed, in one case a bag was overfilled and release of agglomerates > 400 nm observed
[55, 61]	Simulated industry workplace	Compounding of nanocomposites with nanoscale alumina	Al ₂ O ₃	PSD (5.6-560 nm), morph.	Significant release, confirmed by STEM analysis

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	Workplace	Type of activity	Nanomaterial	Metric	Results-remarks
[38]	Research Laboratory for use of carbon based ENMs	Transfer of CNMs, sonication in environmentally relevant matrices	Fullerenes, MWCNT, carbon black	PSD (300-10,000 nm), NC (10-1,000 nm)	Each activity resulted in increased particle number concentrations, TEM images clearly show CNM
[39]	Research laboratories	Scalable flame spray pyrolysis	NaCl, BiPO ₄ , CaSO ₄ , Bi ₂ O ₃ , TiO ₂ , SiO ₂ , WO ₃ , Cu/ZN, Cu/SiO ₂ , Cu/ZrO ₂ , Ta ₂ O ₅ /SiO ₂ , Pt/Ba/Al ₂ O ₃	PSD (15-675 nm), NC (> 7 nm, > 10 nm), MC (< 1 μm, < 10 μm)	Concentration in near field and far field higher than in background in 40% of measured cases
[40]	Research laboratories	Plasma enhanced CVD, PVD, compounding of polymers with nanofillers	Nanofillers (not further specified)	PSD (5 nm-20 μm), NC (< 370 nm)	Increased concentrations detected, but likely not caused by ENP release
[43]	Various ²	Mixing of powder and liquid, filling/emptying oven, suspension spraying, flame spraying	TnO, ZnO, InZnO, SiO ₂	PSD (14nm-20 μm), NC (< 1 μm), MC (respirable and inhalable)	No evidence of release of ZnO and InZnO during handling, very high concentrations during spraying of silane and flame spraying of SiO ₂ suspension
[33]	Laboratory to industrial workplace	Synthesis of nanoobjects, handling and production of composite materials	CNT, CNF, Carbon Nanopearls, fullerenes, TiO ₂ , Ag, Mn, Co-oxide, Fe-oxide, Al, SiFe, QDs	NC (15-1000 nm) for screening, PSD (300-1000 nm), MC, CC (not size selective)	Increased NC in all three investigated size classes (10-1000 nm, 300-500 nm, 500-1000 nm) indicate Release of nanomaterial during various of the investigated sites, no systematic analysis of the results is presented

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	Workplace	Type of activity	Nanomaterial	Metric	Results-remarks
[4]	Laboratory and industrial production facility	Normal activities during batchwise production of SWCNT: collection, removal, cleaning, opening container, vacuum cleaning	SWCNT	NC (10-1000 nm), MC (size fraction not indicated), morph, CC	Likelihood of CNT exposure during production given, period of exposure relatively short (ca. 1h) but concentration are sometimes high the exposure nearly pure nanomaterial.
[49]	Laboratory scale production	Production by chemical vapour deposition (CVD)	SWCNT, MWCNT	PSD (5 nm-20 µm), morph	SWCNT and MWCNT release was determined in the production area in the fume hood, depending on process conditions, No significant amounts of CNT were detected in the breathing zone of a worker and the background.
[42]	Laboratory scale production	Machining / cutting	CNT hybrid composites	NC, PSD (5 nm-20 µm), morph, PM10 MC	Small increases in NC during wet cutting, significant increases (ca. 300,000 #/cm ³) during dry cutting, fibres detected in concentrations of 1-4 fibres/cm ³ during dry cutting.
[52]	Laboratory scale production and handling	Weighing, mixing with solvent, cutting	raw CNF and CNF composite	NC (10-1000 nm), PSD (10 nm-10 µm), ASA, morph	Slight increases in NC for weighing, mixing of CNF and wet cutting. TEM picture reveal the release of CNF during these processes. Minor airborne CNF concentration during normal handling, Main increase in PSD for sizes < 400 nm.
[53]	Laboratory handling	Handling in fume hoods of nanomaterial powders, pouring, transferring	Al ₂ O ₃ , Silver	PSD (5-600 nm), morph	increased NC in the breathing zone of a worker mainly in size range > 100 nm but also partially < 100 nm during handling activity,
[54]	Laboratory scale production and handling	Growth, removal, shaving and transfer of CVD derived CNT	CNT	PSD (5-600 nm), NC (10-1000 nm), TP, ESP, MC	Neither TEM nor NC analysis reveal a release of CNT during these processes
[62]	Laboratory and industrial production	Four production facilities (2 x TiO ₂ by combustion, Ag by plasma and in liquid via citrate), collection of powders in fume hood and in liquid	TiO ₂ , Silver	PSD (15-710 nm), MC, CC, morph	Lowest number concentration detection for in liquid production, higher particle number concentrations during combustion but also release from electro engines and other side activities

4.2.4 Nanoparticle Release Studies Under Laboratory Conditions

Systematic and quantitative analysis of processes and materials can be performed under defined boundary conditions in the laboratory. Therefore, potential sources must be previously identified and selected. Relevant process-related parameters have to be chosen for the transferability to real exposure situations. To meet these requirements, some studies used standardized procedures for the experimental process simulation [53, 63-66]. Due to the multitude of processes which lead to the release of particles, the number of studies on new characterization methods is steadily increasing [29, 54, 66, 67].

Process control (e.g. avoidance of background concentration and particle contamination, sampling), conditioning of the generated aerosol (e.g. dilution, neutralization, size selective particle deposition), and the combination with suitable measurement technology are crucial for the quality of investigations on particle release in the laboratory. Particle number based aerosol measurement devices, originating from clean room monitoring, enable the highest sensitivity for the quantification of low particle amounts. Therefore the nanoparticle release is expressed as a particle number released from a sample of defined size [68].

Nearly all of the reviewed laboratory studies are based on reducing background concentrations. The easiest way to ensure a stable and well known background concentration is to isolate the test room atmosphere from outside influences as performed by Bello et al. [42, 54, 69]. Lower background concentrations require encapsulation and purging with filtered air or gas as realized by the most studies. The lowest background concentrations were achieved by using laminar flow benches which also provide larger working space as employed by Vorbau et al. [64] and Göhler et al. [67].

To ensure the comparability of different tests or to estimate the relevance for a certain workplace situation the volumetric flow over the treated sample, the analysed volumetric flow of the measurement device and the dilution ratio must be measured to calculate the number of emitted nanoparticles (defined as nanoparticle release) from the measured particle number concentration and particle size distribution. Furthermore, the comparability requires the reporting of treated sample size, for instance sample mass. Relating the released nanoparticle number to this sample mass delivers a kind of emission factor of a certain material in a certain treatment process.

Generally, nanomaterial release studies can be structured in following steps:

- reducing background particle concentration
- sample treatment and/or process simulation
- nanomaterial release and aerosolisation
- aerosol sampling and conditioning
- aerosol/particle characterization

Nanomaterials are subjected to mechanical, thermal and environmental stress situations during production, processing and use. Published studies, based on the characterization of the particle release into air due to individual treatment processes, can be roughly classified by the investigated nanomaterial group (powders, suspensions, coatings, composites) as shown in Table 23. According to these groups the experimentally simulated treatment processes (shown in column two) are different like fluidized bed processes for powders or sanding processes for

coatings. Comparable processes are separated additionally. Moreover, the processes are ordered downwards with increasing energy input.

Powders

Based on the various application processes (e.g. transport, dosing, filling) of powders and their characteristic parameters (e.g. energy input, flow velocity, sample amount), different methods have been developed in the past for the investigation of material dependent particle release, the so called dustiness. Additional material dependent parameters, like powder flow properties also affect the results of such dustiness tests and limit their comparability. Nevertheless different types of test setups are described in national standards and two test procedures based on different drop conditions (rotating drum test, continuous drop test) were introduced as international standard (EN 15051:2006). These methods, developed for MC assessments, were taken, modified and combined with aerosol measurement devices by Schneider & Jensen [65], Jensen et al. [66] and Tsai et al. [53] for the characterization of the particle release. Schneider & Jensen [65] and Jensen et al. [66] investigated the particle size distribution and particle number concentration of different metal oxides powders (TiO_2 , SiO_2 , $\text{FeO}(\text{OH})$, $\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$, Al_2O_3), bentonite and organoclay powders. Typical results show a number based submicrometer fraction of about 60 % and < 10 % for particles < 100 nm. Similar results were obtained by Tsai et al. [53] for TiO_2 and ZnO metal oxide powders with slightly modified measurement devices (SMPS, APS, and MOUDI). Isbaseta & Biscans [70] investigated TiO_2 and SiO_2 in free drop conditions. Despite large deviations in the PSD determined by an ELPI, both powders show a release of nanoparticles of $10 \text{ mg}\cdot\text{m}^{-3}$ and $40 \text{ mg}\cdot\text{m}^{-3}$ for TiO_2 and SiO_2 , respectively, for the same initial sample mass.

Studies based on fluidized bed processes represent a second test type for dustiness. Maynard [71] aerosolized TiO_2 powder in a two component fluidized bed to determine the agglomerate size distribution between 5 nm and 20 μm . Furthermore, airborne particles were electrostatically-precipitated for subsequent TEM analysis. Results showed about 10 % of the particle number concentration to be < 100 nm diameter and 70 % in the submicrometer range. Baron et al. [72] and Maynard et al. [4] introduced a new method for agitating CNTs and metal oxide powders to achieve higher particle concentrations. Powder samples or mixtures of powders and beads were filled in a HEPA-filtered air purged shaker-agitated centrifuge tube. The PSD measurements (3 nm-20 μm) showed that fumed alumina powder released more particles < 100 nm than the investigated SWCNT powder. The agitating principle of Baron et al. [72] and Maynard et al. [4] was also used by Ogura et al. [73] to characterize the amount, PSD and morphology from different nanostructured powders (TiO_2 , ZnO) and carbon materials (SWCNT, MWCNT, fullerenes). In addition to the PSD, Ogura et al. [73] relate the measured number concentrations to the applied sample volume or sample mass. Lee et al. [74] used a similar shaker-principle for the PSD characterization of MWCNTs. They also employed TEM-analyses. Results of atomized, dried and neutralized suspensions of the same MWCNT and ultra-pure water show similar peak diameters at approximately 200 nm up to 300 nm in comparison to the shaker-generated aerosol. In each case fractions of particles in the nanometer size range were observed. An extension of the fluidized bed to aerosolize CNTs was presented by Plitzko et al. [75]. To improve the aerosolisation process, the sieve plate of the fluidized bed was agitated by a shaker. First results based on SMPS measurements and TEM investigations on TP-precipitated particles show a considerable release of CNTs with material specific fractions of particles/agglomerates in the nanometer

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size range. The amount of particles with electrical mobility diameters < 100 nm was determined to be between 10 % and 60 % of the determined number based particle size distribution < 1000 nm.

The procedures and results presented up to here are based on a low energy input to the powder. High energy processes, on purpose or by accident, can result in significantly higher energy input to the nanomaterial. Therefore, worst-case scenarios have to be investigated. The particle release due to a leak in a pressurized vessel was experimentally simulated by Stahlmecke et al. [29]. Different metal-oxide powders (TiO_2 , CeO_2 , SrCO_3 , TiZrAlO) were aerosolized using a pressurized beaker in combination with a magnetic stirrer and passed through an orifice. Results are given as PSD and relative concentrations based on the comparison with reference measurements of powders dispersed without orifice, mimicking the leak. In the measured size range < 736 nm, the amount of particles < 100 nm of the number weighted particle size distribution increases from 1 % up to max. 12 % with increasing overpressure from 0 kPa to 140 kPa, depending on the material. Another worst case scenario, preferred by NIOSH and introduced in ISO DIS 12025:2010 [68], is the method described by Boundy et al. [76], where a small amount of powder-around 4 g-is completely sucked and dispersed in an evacuated bottle.

Nearly all powder studies suffer from incomplete determination of the energy input during sample treatment. Some powder drop tests were affected by the material properties, e.g. flowability, themselves. Furthermore repeated treatment of powders in rotating drum or fluidized bed can generate secondary agglomerates. In this case the number of released particles rapidly decreases during the test [72, 75]. Fast measuring devices may help to avoid this problem [65, 67]. Furthermore, treatment processes stressing a well-defined sample only once allow a straight correlation of the measurement data to the treatment process. Repeated energy input leads to permanent changes in the experimental conditions.

Suspensions

Johnson et al. [38] investigated the particle release of fullerenes, MWCNT and carbon black into air due to the sonication of suspensions in comparison to different laboratory handling processes, especially the weighing and transferring of powder material. Size specific particle number concentrations (300 nm-10 μm) and total particle number concentration (10 nm-1 μm) were analysed. In addition, the generated aerosols were filtrated for subsequent TEM analysis. Johnson et al. (2010) [38] found that sonication leads to the release of water droplets with embedded nanomaterials. CPC measurements showed an increase in concentration by sonication in comparison to the dry powder bulk handling, while OPC measurements showed a decrease in the concentration of coarse particles. It remains unclear whether all droplets contained nanomaterial.

Spray simulation with nano-silver and nanoparticle-free suspensions were carried out by Hagendorfer et al. [77] inside a particle free glove box. Two different types of commercial spray dispensers (pump spray, propellant gas spray) were investigated. The generated aerosols were dried by a thermodesorber before PSD measurement and sampling by an ESP for subsequent TEM and EDX analysis (0.8 m away from the generation zone). The pump spray experiments showed no significant increase in the number concentration (10 nm to 500 nm) in comparison to the background. This can possibly be explained by the formation of only large droplets which deposited inside the glove box before sampling. Propellant gas spray, on the other hand showed a broad PSD. The silver particle sizes, determined with SMPS and TEM,

depended on the droplet size generated by the dispenser. The measured number based PSDs of the propellant gas spray generated aerosol show a fraction of around 80 % < 100 nm.

Coatings

Engineered nanoparticles are often intentionally embedded in the matrix material of a coating, which can be understood as a thin layered composite on a substrate. Several studies were performed to determine potential exposure of consumers by typical application processes of such coatings like abrasion, scraping or sanding. Besides the determination of the particle concentration and the PSD, additional appropriate chemical and morphological analyses (e.g. SEM, TEM, EDX, ICP-MS) are necessary for the distinction of unintentionally produced particles in the nanoscale and the engineered nanoobjects in the airborne emissions.

Vorbau et al. [63] employed a commonly used and standardized Taber Abraser (DIN 68861-2:1981 [78], DIN EN 13523-16:2005 [79], DIN EN ISO 7784:2006 [80]) for determining the particle release from different coatings with and without ZnO nanoparticle additives. This device consists of two abrasion wheels, which act on the sample surface. Investigations were performed by operating the Taber Abraser in a particle free environment inside a laminar flow bench. Due to the weak abrasion process, the nanoparticle concentration was too low for determining PSDs by SMPS. The average total particle concentration based on the CPC measurements ranged between 1 cm^{-3} to 20 cm^{-3} . No significant differences were observed between coatings with and without added nanomaterial. TEM images of electrostatically collected wear particles showed the nanomaterial only embedded in the matrix. In another study Guiot et al. [63] used the Taber Abraser test method for investigations on polyvinylchloride layers with and without nanoclays on a PET substrate. In contrast to Vorbau et al. [64] the generated and aerosolized particles were locally sampled near the abrasion zone. The abrasion zone was encapsulated and purged by HEPA-filtered air. Measurement data show an increase in the particle concentration for nanoclay-doped samples. The authors subtracted the measured PSD of the PVC layer without nanoclays from that with nanoclays. This calculation assumes same particle release rates in both experiments for balancing particle amounts, which was not shown. Furthermore, the difference in the released number of nanoparticles may not represent the nanoclay release due to changed mechanical matrix properties introduced by the addition of nanoclay.

Nanoparticle release due to light at different wavelengths, air flow and mechanical scraping for surface coatings containing nanomaterials were investigated by Hsu and Chein [81]. Coatings with different TiO_2 additives on wood, polymer and tile were tested in a closed, purged chamber and PSDs were measured (15 nm-616 nm). The highest particle concentrations of about 630 cm^{-3} were found for the coating on tile and wood during parallel use of UV-light, airflow and scraping. No comparative measurements of coatings without added nanomaterial were performed, so a clear interpretation of the measured data is not possible. Bello et al. [54] investigated the particle release during the removal of CNT forests, which were artificially grown by CVD on a silicon substrate, by cutting with a razor blade. No significant difference between background concentration and material handling was observed using an FMPS and a CPC. SEM and TEM-images of electrostatically and thermophoretically collected and filtrated aerosol particles showed typical background particles but no CNTs, whether individual or bundles.

A commercial hand-held orbital sander with an internal dust removal fan was operated by Koponen et al. [7, 8] for the characterization of the sanding dust. The exhaust air of the sander

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was connected to an exposure chamber for determining the PSD (5 nm-20 μm). Additionally, particles were electrostatically sampled for subsequent physicochemical and toxicological analysis. Coatings containing and not containing engineered nanoparticles were investigated. Some coatings showed higher others lower release rates when engineered nanoparticles were added to the coating. Sanding of surface coatings with and without ZnO and Fe₂O₃ nanoparticle additives was investigated by Göhler et al. [67]. The sanding zone was encapsulated and a particle free environment was provided by a laminar flow bench. PSD (5 nm-20 μm) and number concentrations (CPC) were measured and ESP samples analysed by SEM, TEM and EDX. Despite a considerable generation of nanoscale particles due to the sanding process, neither matrix-free engineered nanoparticles nor significant differences in the particle size distribution between coatings with and without engineered nanoobjects were observed in both studies.

Composites

Particle release due to Taber Abraser tests, customized sanding and UV-light weathering was compared by Wohlleben et al. [82] for different composites containing SiO₂, CNT, CSH nanomaterial. In each case the morphology, the size distribution and surface chemistry of the wear powders were analysed. Aerosol particles were measured by SMPS for the first two processes. Only the polymer-composite with CNTs behaved significantly differently to the same material without nanoobjects, where increased UV-light adsorption resulted in faster matrix degradation.

The ability for the release of CNTs from composite materials was also investigated by Bello et al. [42, 69] during wet and dry cutting and solid core drilling. In both studies the same types of CNT-composites were analysed. Bello et al. [42] used a band saw for the simulation of a dry cutting process, while a rotating cutting wheel was operated for the wet cutting process. Samples for the analyses were taken at two different locations-near the source for the dry cutting process and at the breathing zone. The PSD (5 nm-20 μm), NC (10-1000 nm) and MC (PM_{2.5}, optically determined) were measured. Analysis for particle chemistry and morphology were carried out by SEM, TEM coupled with EDX on ESP and TP substrates. A considerable generation of nanoscaled particles during the dry cutting process was observed. Results show no significant difference between samples with and without CNTs with regard to the particle number concentration. SEM and TEM analysis showed no free CNTs and no CNTs which are bundled or attached to coarser particles.

A commercial drill press was employed by Bello et al. [69] to simulate dry and wet solid core drilling. The latter one was simulated by continuous spraying of deionized water to the drilling zone. In comparison to Bello et al. [42] a WRASS was added to the measurement devices in [69] for size selective particle deposition and subsequent ICP-MS analysis. During the drilling processes smoke generation was observed. Also clusters of CNT were determined with TEM. Whether these were released directly from the drilling process or originate from contaminations remains unclear.

Table 23: Studies focused on the particle release of ENMs by laboratory testing

Study	Process	Materials	Instrumentation	Metric	Results
<i>Powders</i>					
[53]	Rotating drum test (free fall, stirring, ...)	TiO ₂ (Aeroxide P25), ZnO	SMPS, APS, MOUDI	PSD (15 nm-20 µm), NC, DI	< 10 % particles < 100 nm 60% particles < 1 µm
[65]	Rotating drum test (free fall, stirring, ...)	TiO ₂ , SiO ₂ , FeO(OH), Mg ₃ Si ₄ O ₁₀ (OH) ₂ , Al ₂ O ₃	FMPS, APS, Filtration	PSD (5.6 nm-20 µm), NRP (0.5 µm-20 µm), NRP (5.6 nm-560 nm), DI	Undefined fraction of particles < 100 nm
[66]	Rotating drum test (free fall, stirring, ...)	Organoclay, Bentonite	FMPS, APS, Filtration	PSD (5.6 nm-20 µm), NRP (0.5 µm-20 µm), NRP (5.6 nm-560 nm), DI	Undefined fraction of particles < 100 nm
[70]	Free fall	TiO ₂ (G5), SiO ₂ (Aerosil 200)	ELPI, SEM (ELPI)	PSD(30 nm-10 µm), MC, MC/M	Fraction of particles < 100 nm
[71]	Fluidized bed	TiO ₂ (Aeroxide P25)	SMPS (LDMA, NDMA), APS, TEM (ESP)	PSD (4 nm-20µm)/NC _{max}	10 % particles < 100 nm 70 % particles < 1 µm
[4, 72]	Vortex shaker (fluidized bed, agitation)	SWCNT, alumina powder	SMPS (LDMA, NDMA), APS	PSD (4 nm-20 µm), NC	Fraction of particles < 100 nm, alumina powder released more NP than SWCNT
[73]	Vortex shaker (fluidized bed)	SWCNT, MWCNT, TiO ₂ , ZnO	SMPS, HHCPC, APS, OPC	PSD (10 nm-20 µm), NC (10 nm-> 1 µm)/V, NC (10 nm-> 1 µm)/M	Fraction of particles < 100 nm
[74]	Shaker method	MWCNT	SMPS, APS, TEM(ESP, CI)	PSD (14 nm-20 µm), NC(dt)	Fraction of particles < 100 nm, peak at 200-300nm
[75]	Fluidized bed with oscillating sieve plate	MWCNT	SMPS, SEM (TP)	PSD (< 1 µm), NC(dt)	Fraction of particles < 100 nm
[29]	Stirring and dispersing in orifice (leak in pressurized vessel)	TiO ₂ , CeO ₂ , SrCO ₃ , TiZrAlO	SMPS	PSD (14 nm-736 nm), fractions, relative values	Increase of the fraction of ENPs by increase of the overpressure (up to 12 %)
<i>Suspensions</i>					
[38]	Weighing/transferring of powders and sonication of suspensions	fullerenes, MWCNT, CB	HHCPC, HHPC, TEM-EDX (filtration)	PSD (0,3 µm-10 µm), NC (10 nm-1 µm)	Suspension sonication leads to droplets with embedded ENM

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Study	Process	Materials	Instrumentation	Metric	Results
[77]	Spraying	suspensions with and without Ag	SMPS, TEM(ESP)	PSD (10 nm-500 nm), NC (< 100 nm), NC(< 500 nm)	High fraction of particles < 100 nm
<i>Coatings</i>					
[63]	Weak abrasion process (Taber Abraser)	PVC layer with/without ENPs (nanoclay)	SMPS, CPC	PSD (5 nm-1 µm)	
[64]	Weak abrasion process (Taber Abraser)	coatings with/without ENPs (ZnO)	SMPS, CPC, SEM/TEM-EDX (ESP)	PSD (16 nm-626 nm), NC (> 6 nm), wear mass	Very low concentrations, ENPs still embedded
[81]	UV light, wind erosion, scrubbing	coatings with TiO ₂	SMPS	PSD (15 nm-661 nm), NC (15 nm-661 nm)	Comparison with non-doped samples is missing
[54]	Shaving (razor blade)	CNT	FMPS, HHCPC, SEM & TEM-EDX (TP)	NC (10 nm -1 µm), PSD (5.6 nm-560 nm)	No significant change in concentration, no free CNTs were observed
[7, 8]	Sanding process (orbital sander)	coatings with/without ENPs (TiO ₂ , CB, SiO ₂ , CaCO ₃)	FMPS, APS (ESP)	PSD (5.6 nm-20 µm)	General release of NP, spark particles contamination
[67]	Sanding process (Dremel)	coatings with/without ENPs (ZnO, Fe ₂ O ₃)	FMPS, CPC, OPC SEM/TEM-EDX (ESP)	PSD (5.6 nm-20 µm), NC (< 100 nm), NRP (<100nm), NRP (< 10 µm), swarf mass, material, morphology	General release of NP but ENPs still embedded in the matrix
<i>Composites</i>					
[82]	UV-light , weak abrasion process (Taber Abraser), customized sanding	POM with/without CNT, PA with/without SiO ₂ , cement with/ without CNT, cement with/without CSH	SMPS, UNPA, SEM, AUC, XPS, SIMS	PSD (14 nm-820 nm), Morphologie, material	No free SiO ₂ -particles or CNTs detected
[42]	Dry and wet cutting (band-saw, rotatory cutting wheel)	composites with and without CNT	FMPS, APS, HHCPC, DT, SEM/TEM-EDX (ESP, TP)	PSD (5.6 nm-20 µm), NC (5.6 nm-560 nm), NC (0.5 µm-20 µm) MC, material, morphology	No free CNTs observed
[69]	Dry and wet solid core drilling	Composites with and without CNT	FMPS, APS, HHCPC, DT, DC, SEM/TEM-EDX (ESP, TP) WRASS+ICP-MS	PSD (5.6 nm-20 µm), NC, MC (< 35 µm), SA, material, morphology	Smoke generation, free CNT clusters observed

4.2.5 Discussion and Conclusions

Two different basic approaches to derive information on possible workplace exposure were summarised and the state of the art presented in this review. It is evident that both approaches, workplace measurements and laboratory studies, are needed for a concise assessment to derive e.g. model based predictions on possible release of nanomaterial from a given process.

All studies can be ordered in a systematic scheme of different workplaces, work processes and nanomaterials. This systematic evaluation shows that all basic parameters have been pursued and published in the peer-reviewed literature. Thus, the amount of data becoming now available will allow first meta-analysis investigating material structure and release relationships in detail. Despite the major progress made in recent years, open issues still exist hindering the set-up of a coherent and concise exposure assessment for nanomaterials at workplaces. A coherent approach for the likeliness of release and workplace exposure assessment for all relevant nanomaterials and work processes as e.g. outlined in Table 21 is recommended.

Measurement metrics and corresponding measurement technology remain to be an extremely important issue. The metric best related to possible health effect is still not identified. The lack of comparability always became evident, when trying to compare results from the various studies in this review, because a harmonized approach for data evaluation concerning metric, size range etc. is still missing. This lack hinders the development of general conclusions.

Particle number concentrations and particle number size distributions are the most commonly used metrics within the reviewed workplace and laboratory studies. This approach is currently seen as the one to further develop due to the high sensitivity of the metric to airborne nanoobjects as well as due to the availability of the measurement devices. The latter allow the use in tiered approaches starting with a screening e.g. only measuring a size-integrated concentration quantity such as the number concentration. Intensified measurements can be conducted in a second tier in identified areas of increased concentration.

A major drawback of current state of the art measurement devices is their lack of differentiation of background from nanomaterial related particles. Aerosol mass spectrometer is currently the only instrument sizing and chemically analysing nanoscale particles online. Such instrumentation, once it is capable of also analysing metals and metal oxides, which is currently in development, would be an ideal analytical instrument to be used for areal measurements at workplaces if costs and size of the instrument are neglected. On the other hand particle number concentration and particle size distribution measurements at workplaces can at best only be viewed as indicative measurements for the presence or absence of airborne nanomaterials but without a definitive proof. The latter can only be provided by particle sampling with subsequent electron microscopy, if possible linked with a single particle chemical analysis.

A second drawback may be the sensitivity and comparability. While CPCs can reliably measure concentrations down to 0.001 cm^{-3} , size resolved measurements for nanoscaled particles need significantly higher concentrations. In addition, the commonly noisy background makes it impossible to detect a release which only amounts to a small

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fraction of the total concentration. In case of the need for detection of concentration increases of a few particles per cm^3 other devices and methods have to be employed. An obstacle related to this topic is the current lack of information of lower and upper detection limits related to measurement devices and especially when combined with a measurement strategy. Further uncertainty arises from the calibration of the devices which sometimes may differ by up to 30 % [60]. Work on the uncertainties and detection limits is certainly needed to achieve data qualities good enough for comparison of results from different studies.

The method mostly used for the identification of nanomaterials is electron microscopy (SEM or TEM) coupled with single particle chemical analysis such as EDX. This method was regularly employed in different studies but is not standard in routine workplace assessments due to the high demand of person hours. A difficulty for this approach is the limitation of the measurements. Nevertheless the use of single particle analysis, combining morphological and chemical information, is currently the only approach proving the presence of engineered nanomaterials. A definitive proof of their absence may not be possible due to the limitation of the subsequent quantification to a few thousand particles, even if automated particle identification software is used.

There is still an urgent need for a systematic approach of harmonization and standardization. The needed areas to be covered are test procedures simulating workplace activities and processes as well as coherent workplace exposure assessments. First steps in simulating workplace activities and work processes are investigations on sensitivities of nanomaterial emissions to specific work parameters such as in sanding on the type of sanding paper, the acting force onto the surface, rotational speed. Further steps in exposure assessments is the European, better worldwide, agreement on a harmonized measurement metric, strategy, and data treatment and analysis including statistics.

From the discussion above and the outcomes of the studies presented in this review, we conclude that a tiered approach is viewed as most practical for workplace and laboratory measurements, because complete measurement campaigns are very time and hence cost intensive and may only be necessary if there is evidence of an increased particle concentration. The proposal brought forward by NIOSH [33] and the consent report [34] are scientific reasonable and pragmatic starting points for further refinements. Laboratory studies are the consequence of the tiered approach.

Some studies reported a release of particles. In fewer studies engineered nanoscale particles were observed. The latter was mainly caused by maintenance problems, open gas phase production processes, open handling of nanopowders or smoke generation during processing.

A release of agglomerated nanoobjects, mainly > 300 nm in the number weighted diameter, was regularly observed, especially during open handling of dry nanomaterials [2, 3]. Release of nanomaterials < 100 nm was only observed in the few special cases mentioned above. The use of fume hoods and appropriate ventilation systems seemed to significantly reduce potential exposure concentrations.

The review of the laboratory test procedures shows that many workplace related processes are currently simulated in the laboratory. While some approaches are quite advanced we still lack a coherent, systematic approach over all work related processes as well as studies on single simulations. Questions like: How shall abrasion tests be

pursued? How does the normal force of the abraser influence the release? Shall heating of the sample be kept at a minimum? Shall the test simulate worst case scenarios? How to avoid background particles? have to be answered before a simulation method can be performed. This area, important for modelling of work processes and subsequent possible exposure, has to be further developed on the basic research level as well as in view of a standardized method.

Generally all reviewed studies underline that the amount of released nanoparticle is the result of the combination of the treatment process and the employed material. All powder handling processes released some nanoobjects. Release of free engineered nanoparticles was not observed in laboratory studies of workplace related processes based for treatment processes on coatings and composites because the nanomaterial was still embedded in the matrix material.

Still one of the major issues to be tackled in the near future is the question of how to detect and define level of detection needed for workplace safety assessment. In the case of a specific health hazard of a nanomaterial, lower detection limits may be down to single particles are necessary. Generally, particle number concentration based exposure values may be sufficient in most cases. When assessing the metric to be used for limit values, possible other parameters, such as particle surface area and particle reactivity [e.g. 83], have to be evaluated. The use of mass concentration measurements is currently viewed as being too insensitive to assess toxicological effects related to airborne nanomaterials.

A final point revealed by this review is that a certain set of minimum information is needed for all workplace related studies, either real workplaces or laboratory simulations. Release assessments and comparability between different studies require sample size specific nanoparticle release data, which can only be obtained by simultaneous measurement of several more parameters than simply particle size distributions and number concentrations. Nevertheless, in each case, whether workplace studies or laboratory investigations, comparisons with reference activities, reference materials or different treatment processes are of fundamental importance for the discussion on possible nanomaterial exposure. The nanomaterials and processes studied have to be described carefully and in sufficient detail, including contextual information, to be able to compare the results with those of other studies. All methods employed to identify the corresponding nanomaterial should be described and a clear conclusion given if a release or exposure was determined. A last but very important piece of information is the reporting of particle sizes and size resolved concentrations. Integrated particle number and surface area concentrations alone may be used as indicators in a tiered approach, but do not describe exposure in sufficient detail.

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Abbreviations

APS: Aerodynamic Particle Sizer, AUC: analytical ultracentrifugation, CNF: carbon nanofibers, CNT: carbon nanotubes, CPC: Condensation Particle Counter, CSH: calcium silicate hydrate, DI: Dustiness Index, DT: Dust Track, EDX: energy dispersive X-ray spectroscopy, EEPS: Engine Exhaust Particle Sizer, ELPI: electrical low-pressure impactor, EM: electron microscopy, ENM: engineered nanomaterial, ENP: engineered nanoparticle, ESP: electrostatic precipitator, FMPS: Fast Mobility Particle Sizer, HHCPC: hand-held condensation particle counter, HHPC: hand-held particle counter, ICBA: International Carbon Black Association, ICP-MS: Inductively Coupled Plasma-Mass Spectrometry, M: mass, MC: mass concentration, MOUDI: micro-orifice uniform deposit impactor, NC: number concentration, NRP: number of released particles, NSAM: Nanoparticle Surface Area Monitor, OPC: Optical Particle Counter, PM: particulate matter, PSD: particle size distribution, SA: Surface Area, SEM: scanning electron microscope, SIMS: secondary-ion mass spectroscopy, SMPS: Scanning Mobility Particle Sizer, TEM: transmission electron microscope, TP: thermophoretic precipitator, UNPA: universal nanoparticle analyzer, V: volume, WRASS: Wide Range Aerosol Sampling System, XPS: X-ray photoelectron spectroscopy

4.2.6 References

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5 PARTICLE EXPOSURE METRICS AND HEALTH EFFECT STUDIES

The previous chapters introduced the background of particle exposure and their possible effects (chapter 1), overviews on the current knowledge on exposure assessment strategies, exposure related measurement strategies and measurement technologies (chapter 2). Examples on how these strategies were employed, how sources of particles in workplaces and in ambient air were identified as well as investigations on agglomerate stability were given in the chapters 3 and 4. The latter chapter also presents a comprehensive review on nanoparticle related workplace measurements and laboratory studies of release processes. This chapter concludes by giving examples on studies of new possibly health relevant metrics.

5.1 Exposure Metric Ultrafine Particles-Spatial and Temporal Variability

One of the main sources of ultrafine and submicrometer particles in urban areas is street traffic. This can nicely be seen in Figure 61 where Weijers et al. (2008) present results of average particle size distributions in three different distances to the road. It can be seen that the number concentrations of particles below 300 nm diameter decrease rapidly with the distance.

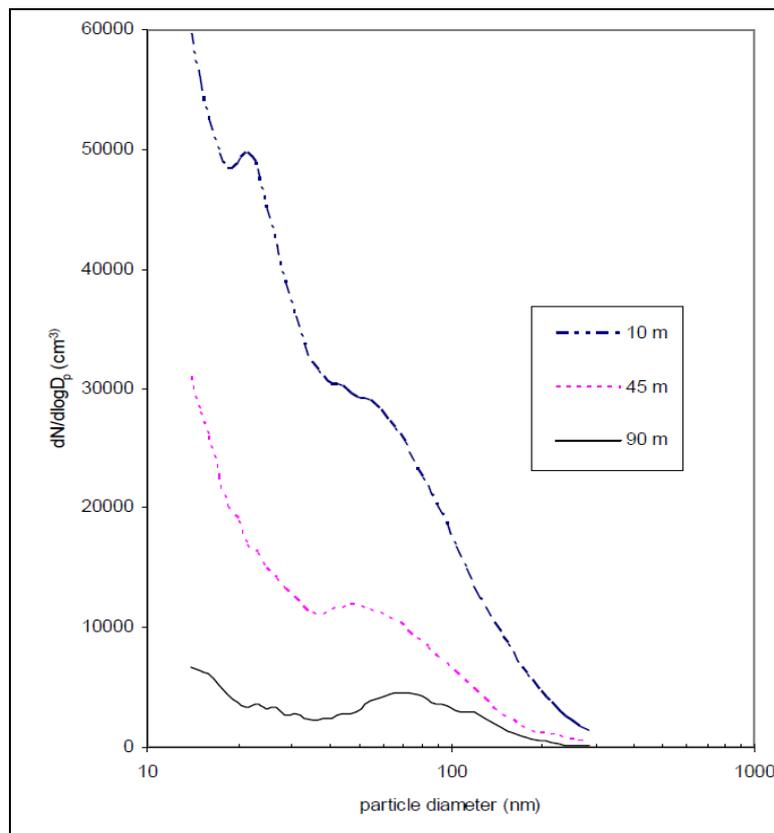


Figure 61: Particle size distributions in dependence on distance to a motorway (Weijers et al., 2008)

5 Particle exposure metrics and health effect studies

Some further detailed studies by Zhu et al. (2002) and Hinds et al. (2006) show beside the dependency for the particle number concentration (6-230 nm diameter) also clearly decreasing concentrations for carbon monoxide, black carbon and particle mass concentrations (Figure 62). The prior two parameters show a similar strong dependency like the ultrafine particles downwind of the road. PM mass concentrations show the lowest dependency. This finding can be explained by the different contributions to the background concentrations. Concentrations of CO, black carbon and ultrafine particle numbers are dominated by traffic emissions near the road while PM mass concentrations are comparably little influenced by the road traffic.

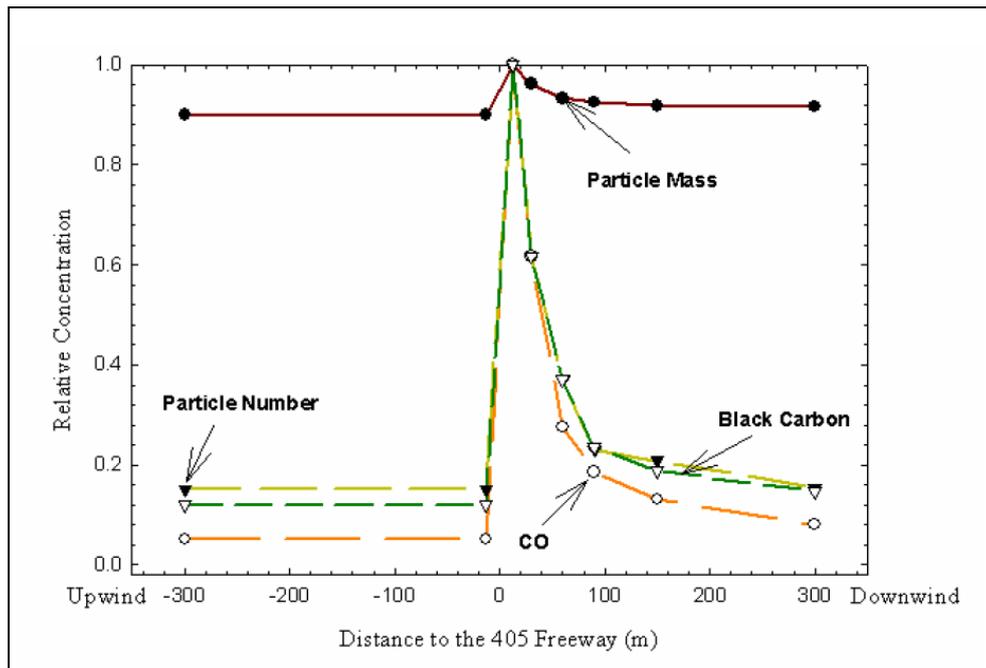


Figure 62: Changes in particle number, mass and black carbon concentration with distance to a motorway (Zhu et al., 2002, Hinds et al., 2006)

These results are important in view of exposure assessments since it demonstrates different spatial patterns for PM mass concentrations compared to black carbon and particle number / ultrafine particle concentrations.

This dependency with the distance to the road is not only valid for freeways and autobahns as shown in Figure 63. The presented measurements were conducted around a traffic site in a street canyon (Gladbecker Strasse, Essen, Germany). A handheld CPC (TSI 3007) was used to record particle number concentrations at 59 sites along and in transects of 90 degree angle to the main road. The measurements were repeated 17 times at comparable meteorological conditions. The number concentrations shown for the transects in Figure 63 indicate in all cases a decrease with the distance as could be expected from the motorway studies. The initial number concentration decrease of 7-9% per 10 m distance to the main road agrees well with results of a field study by Hagler et al. (2009). It is interesting to note that the transect 4 shows an increase of the particle number concentration again when another main street is approached.

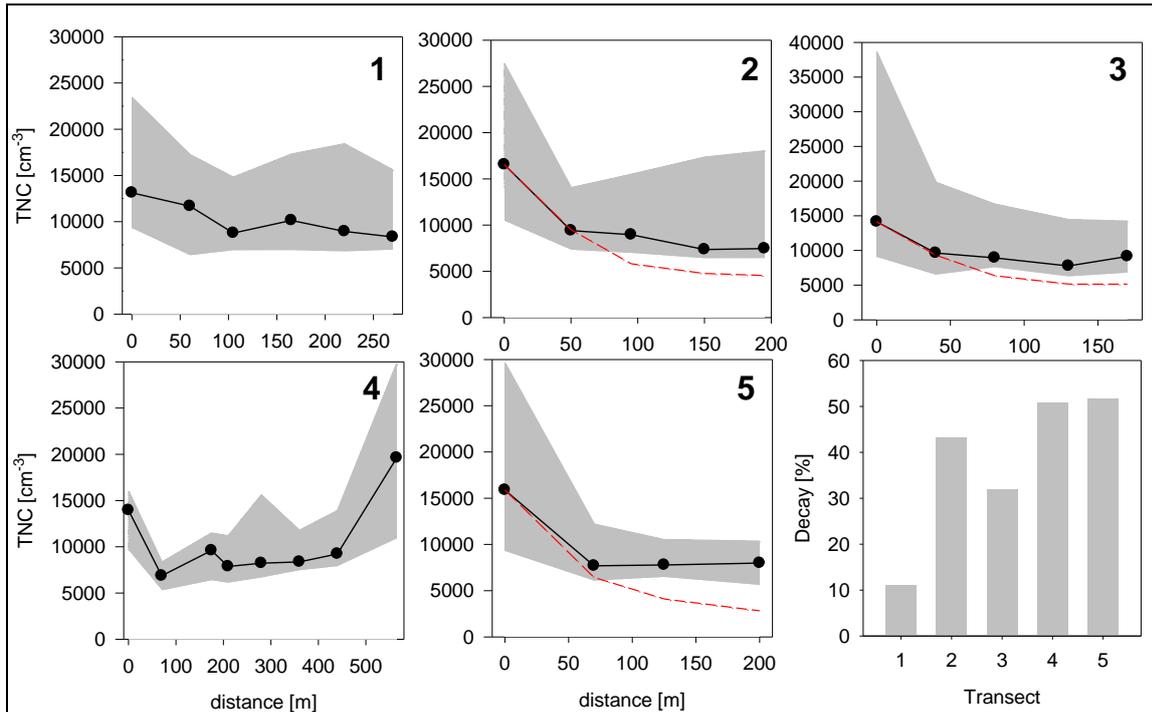


Figure 63: Total particle number concentrations at five transects in the study area Gladbecker Strasse, Essen, Germany. Particle number decay (red dashed line) according to Hagler et al. (2009), solid line median and grey shading inter-quartile range (Nickel et al., 2009).

Overall consistent results are observed for the spatial variability of ultrafine particle number concentrations with the distance from roads.

Investigations of temporal and spatial variability by Cyrus et al. (2008) showed for Augsburg a high degree of correlation between different urban background sites. Correlation coefficients (R) ranged between 0.91 for the two urban background sites, 0.84 for the two traffic sites and from 0.77 to 0.92 between the site types. The average particle number concentration was only slightly elevated at the traffic influenced sites with 20,000-24,000 #/cm³ compared to 15,000-20,000 #/cm³ for the urban background sites (Cyrus et al., 2008).

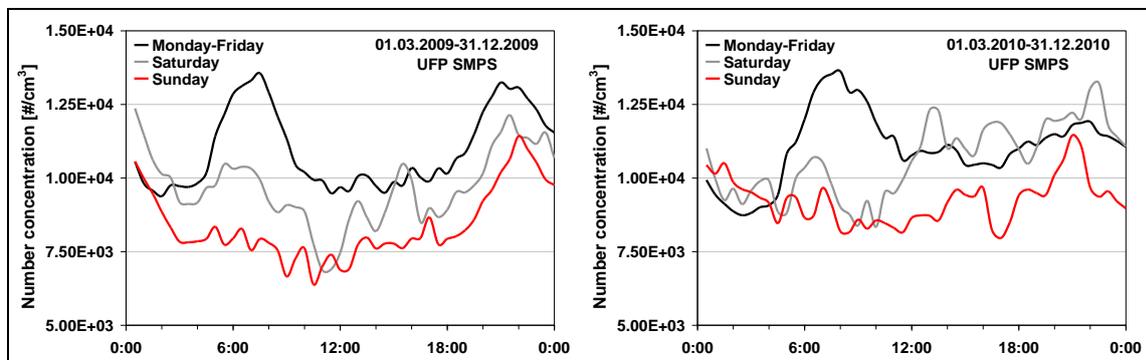


Figure 64: Average diurnal variation of ultrafine particles at an urban background station in Mülheim Styrum (< 100 nm) in 2009 (left) and 2010 (right) (Kaminski et al. 2009, Kaminski and Kuhlbusch et al. 2011)

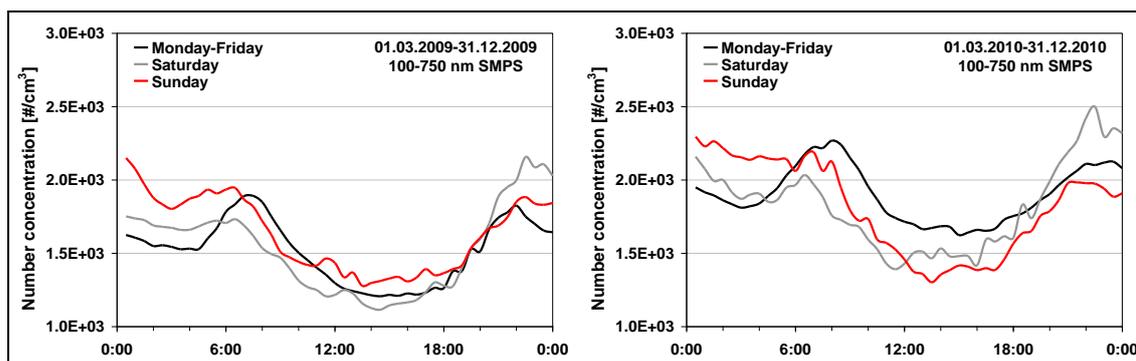


Figure 65: Average diurnal variation of submicrometer particles at an urban background station in Mühlheim Styrum (100-750 nm) in 2009 (left) and 2010 (right) (Kaminski et al. 2009; Kaminski and Kuhlbusch et al. 2011)

Figure 64 and Figure 65 show the diurnal patterns of particle number concentrations for the ultrafine particles and the particle size fraction of 100-750 nm. The measurements were conducted at an urban background site in Mühlheim-Styrum, Germany (Kaminski et al., 2009, Kaminski and Kuhlbusch, 2011). The diurnal variations were separately calculated for weekdays and weekend days since anthropogenic activity patterns vary significantly. Several conclusions can be drawn from the figures:

- normally two minima can be observed: one around 4 am and the second in the early afternoon,
- the absolute minimum concentration for the ultrafine particles is normally observed for the early morning hours while that of the coarser size fraction (100-750 nm) for the early afternoon,
- intraday variations are more pronounced and variable for the ultrafine fraction compared to the coarser fraction,
- interday variations (weekday to weekend) are also more pronounced for the ultrafine particle size fraction.

The above findings agree with those derived in a study by Kuhlbusch et al. (2001) and Birmili et al. (2006). Explanations for the diurnal variations are the meteorological variations along with emission pattern and atmospheric physical-chemical processes. E.g. the morning rush hour leads to significantly higher concentrations due to a) emission and b) low atmospheric mixing heights. Mixing heights increase during the course of the day leading to a maximum mixing height along with high wind speeds in the early afternoon. Hence high mixing ratios of emissions and therefore a minimum in ambient particle concentrations can be observed. Increased particle concentrations towards the evening are due to increased traffic and decreasing mixing heights. Depending on the season this may be coupled leading to increased maxima or decoupled. A more detailed discussion can be found in Kuhlbusch et al. (2001).

From the above studies and publication like Kwasny et al. (2010) it becomes evident that ultrafine particle number concentrations do not correlate well with PM mass concentrations leading to possible differences for exposure assessments. Johansson et al. (2007) nicely summarise the difference in exposures by comparing PM10 mass and particle number concentrations in urban environments. They conclude:

”Annual mean urban background PM₁₀ levels are relatively uniformly distributed over the city, due to the importance of long range transport. For PNC (particle number concentrations, added by TK) local sources often dominate the concentrations resulting in large temporal and spatial gradients in the concentrations. Despite these differences in the origin of PM₁₀ and PNC, the spatial gradients of annual mean concentrations due to local sources are of equal magnitude due to the common source, namely traffic. Thus, people in different areas experiencing a factor of 2 different annual PM₁₀ exposures due to local sources will also experience a factor of 2 different exposures in terms of PNC. This implies that health impact studies based solely on spatial differences in annual exposure to PM₁₀ may not separate differences in health effects due to ultrafine and coarse particles. On the other hand, health effect assessments based on time series exposure analysis of PM₁₀ and PNC, should be able to observe differences in health effects of ultrafine particles versus coarse particles.”

5.2 Exposure Metric Particle Surface Area – Correlation to Particle Number Concentration

A second possibly health relevant metric is the particle surface area. The particle surface area deposited in a lung, is e.g. the area interacting with the biological lung fluid. Particle surface areas can be either directly determined (see p. 47) or calculated from particle number size distributions as shown in Figure 66. Figure 66 shows the ambient particle surface area distribution as well as the fraction which is deposited in a ‘standard lung’ of a male worker. It has to be noted that the surface area concentration is calculated based on the assumption of unit density (1 g/cm³) and spherical particles. The latter assumption is certainly not true for agglomerates e.g. from traffic exhaust particles.

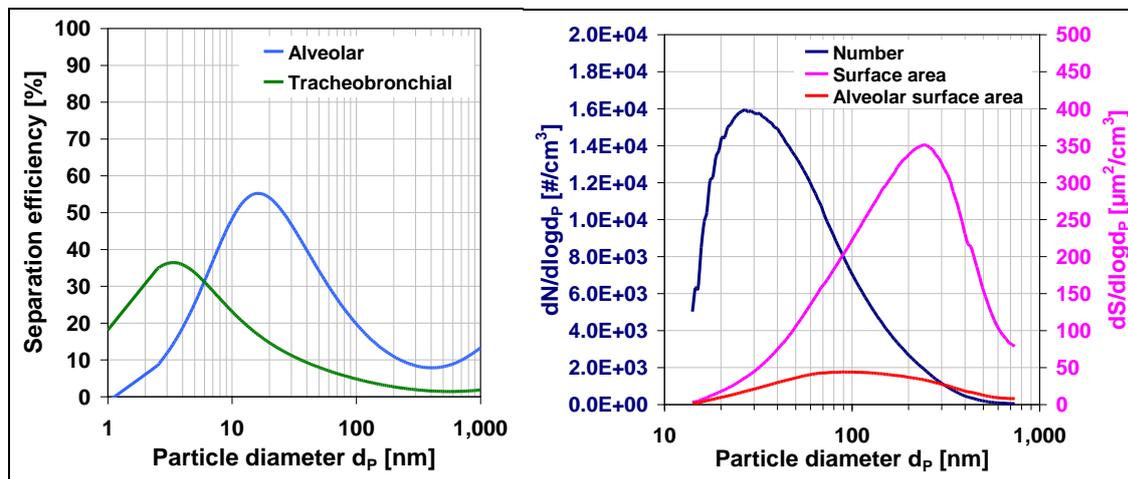


Figure 66: Left: Deposition probability of inhaled particles in the alveolar and tracheobronchial lung region of reference worker according to ICRP (1994), right: Exemplary ambient particle size distribution (blue), calculated corresponding particle surface area (pink) and the alveolar deposited fraction (red, see chapter 5.3 for lung deposition)

A comparison of measured lung deposited particle surface area concentrations with those calculated according to the approach of Figure 66 is shown in Figure 67. The 30

minutes averages used for this comparison show very good agreement for both years investigated. Regression coefficients (R^2) of 0.94 and 0.98 for more than 12,000 paired values and slopes of 1.01 and 0.97 show a high degree of equivalence.

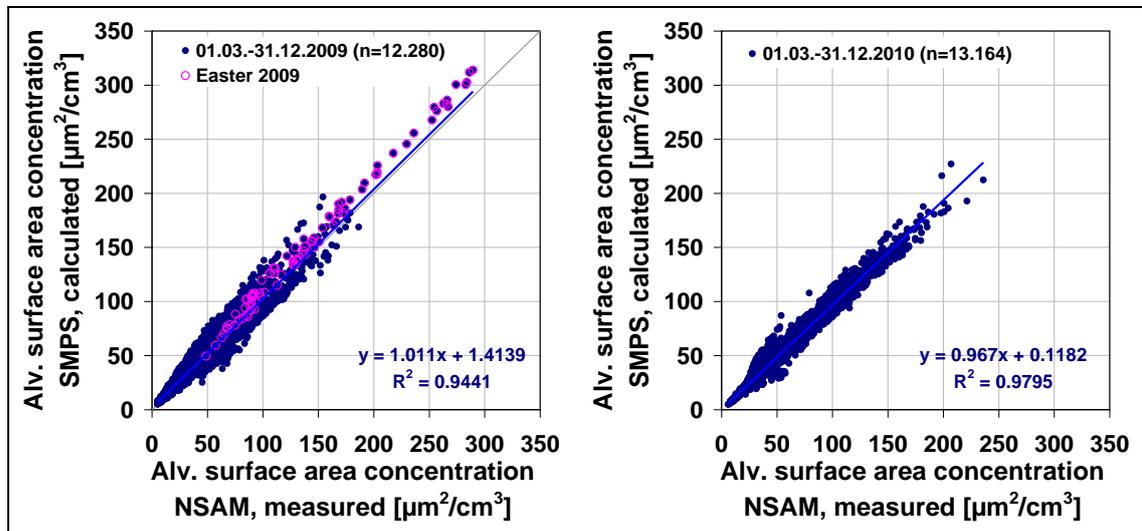


Figure 67: Correlation analysis of the alveolar deposited surface area concentration measured with an NSAM and calculated from particle size distribution measurements (30 min averages) 2009 (left) and 2010 (right) (Kaminski et al., 2010)

This equivalence is somewhat surprising knowing that a) the comparison bases on assumptions and b) that the aerosol was dried for the SMPS but not for the NSAM measurements. Detailed investigations and comparisons of two NSAMS with and without a dryer showed that the particle surface area determined by the NSAM was not influenced by humidity. The electrometer used in the NSAM for detection has to be heated and stabilized at 40°C to achieve the needed sensitivity and reproducibility. This heating also heats and hence dries the aerosol which makes the NSAM reading independent from ambient humidity. From the comparison it is also deduced that the effect of ambient particle morphologies being different from spheres on the particle surface area concentrations is within the errors of measurements.

One important issue when discussing a metric and its possible relation to induced health effects is its independence from other metrics, especially those already employed in air quality monitoring. Figure 68 shows the correlation of more than 12,000 paired values of particle surface area concentration measurements with the NSAM and particle number concentrations for particles < 100 nm (UFP) and 100-750 nm. Only a very low correlation can be seen in Figure 68 for UFP number and surface area concentrations while this correlation significantly increases for the correlation with particles between 100-750 nm diameters. In Figure 4 the influence of the particle size weighting on an exemplary ambient particle size distribution is shown. From this it is evident that the mode of the particle surface area is in the particle size range of 100-1000 nm and hence the correlation between particle number and surface area concentration for this size range can be explained.

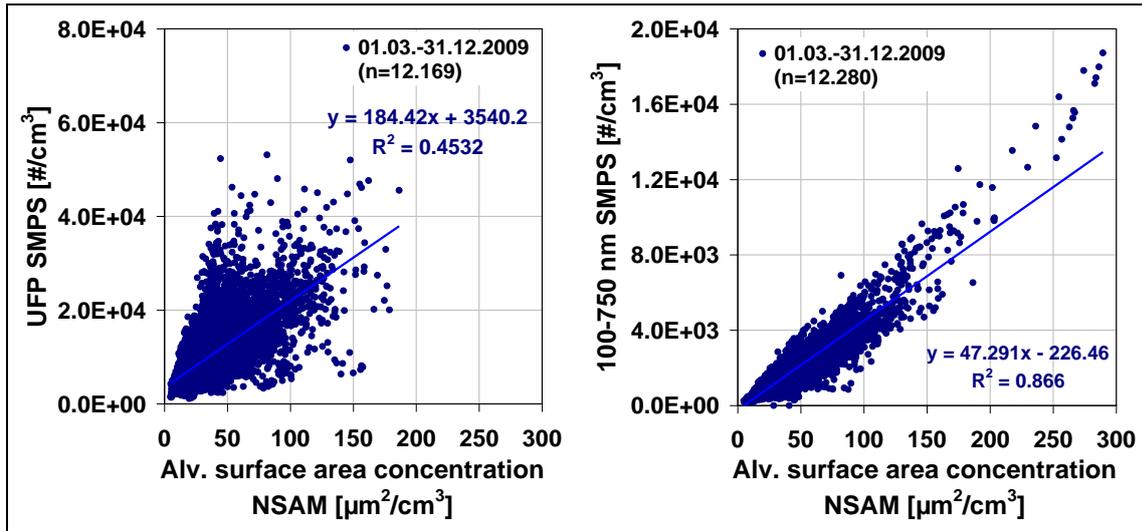


Figure 68: Correlation of particle surface area with number concentrations for the ultrafine size fraction and particles in the size range of 100-750 nm

The studies and results shown for particle number, surface area and mass concentrations in this chapter agree nicely with what is shown in Figure 4. The status for the discussed exposure metrics is that

- they are independent from each other,
- methods for their measurement exist,
- the measurements can be used to investigate temporal and spatial variations,
- they can be used for exposure assessments and epidemiological studies.

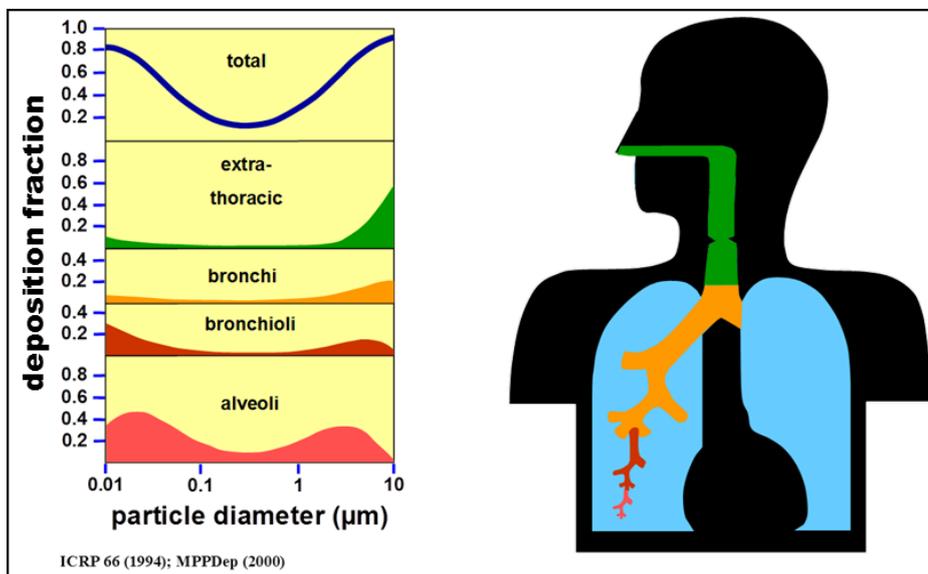


Figure 69: Particle size dependent deposition probability for different lung compartments (from W. Kreyling, Helmholtz-München)

5.3 Exposure to Dose

Already in the section “Surface Area” monitors (page 41) the concept of lung deposited surface area measurements was described. Figure 69 illustrates where particles dependent on their size are deposited in a lung.

This size dependent deposition in human lungs makes particle exposure and the dose calculation unique. Deposition of gaseous pollutants can be calculated from exposure by using one uptake or deposition factor. From Figure 69 it becomes evident that this deposition factor may vary by a factor of 5 dependent on particle size. To demonstrate the effect of size dependent lung deposition, exposure and deposition fractions of particle number and particle surface area concentrations were calculated for three different exposure situations for particles in the size range from 14-750 nm (Figure 70).

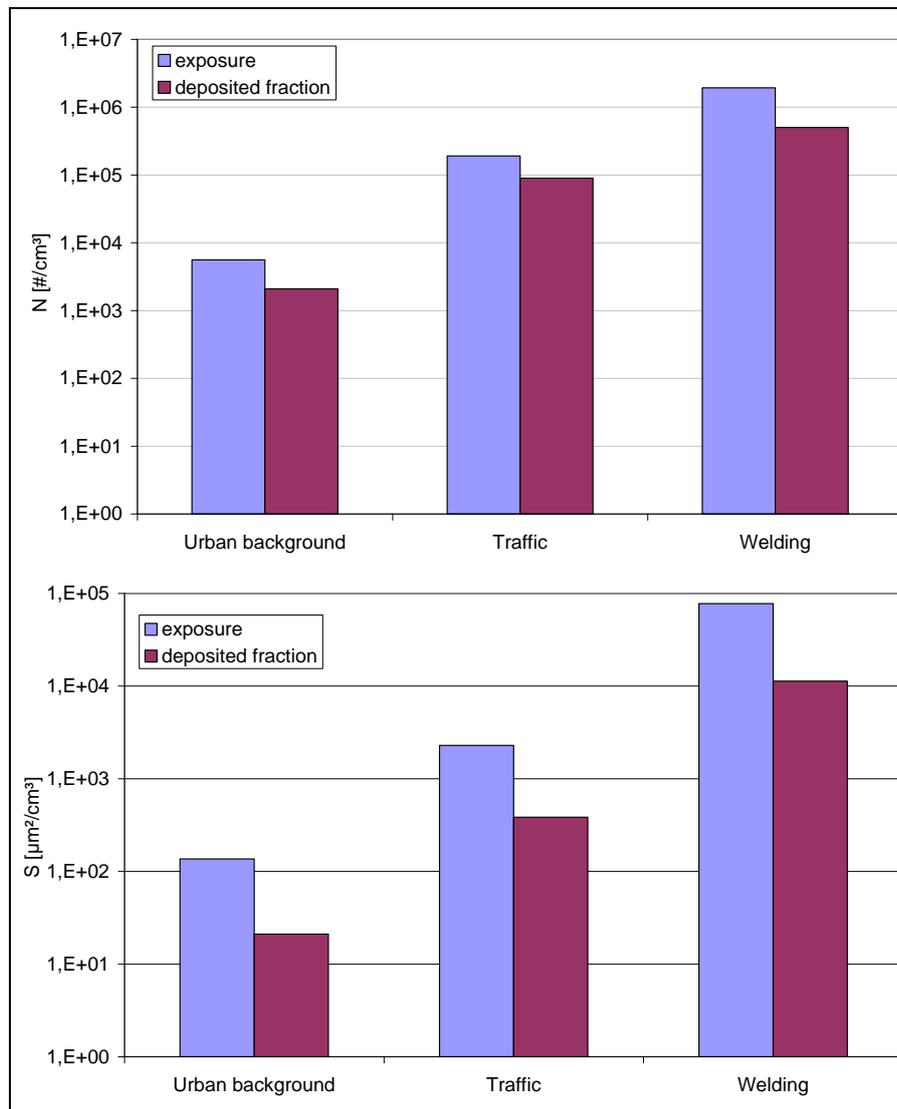


Figure 70: Exposure and alveolar deposited particle size fraction for different exposure scenarios, top: number concentration, bottom: surface area concentration

Figure 70 shows that the deposited fraction is smaller for surface area compared to number weighting of the size distribution in all cases. Table 24 summarises the results shown in Figure 70. From this it becomes evident that surface area is less dependent on

modal values and absolute concentration of exposure than particle number concentrations. Deposition probabilities for the latter vary between 26% and 47% while the prior is around 16% for all three exposure scenarios.

Table 24: Exposure concentrations and deposited particle fraction in dependence of exposure scenario and particle size weighting

Exposure scenario	Particle number [#/cm ³]		Percent of the exposure deposited in the alveolar region of the lung
	exposed concentration	deposited concentration	
Urban background	5.58E+03	2.09E+03	37%
Traffic	1.90E+05	8.95E+04	47%
Welding	1.93E+06	5.01E+05	26%

Exposure scenario	Particle surface area [μm ² /cm ³]		Percent of the exposure deposited in the alveolar region of the lung
	exposed concentration	deposited concentration	
Urban background	1.36E+02	2.11E+01	15%
Traffic	2.29E+03	3.84E+02	17%
Welding	7.75E+04	1.13E+04	15%

This finding can be explained by the weighting of the size distribution metric and deposition likeliness. For surface area concentration the variance in deposition probability is relative small in the interesting size range between 100-1000 nm. This is certainly different looking at the size range between 10-100 nm which is mainly of interest for number concentrations. The modal value for the traffic exposure scenario was < 15 nm which also shows the highest fraction of deposition (47%). On the other extreme the modal value for welding fumes was > 100 nm which led to the lowest fraction of deposition of 26%.

5.4 Exemplary Health Studies Investigating Different Exposure Metrics

Some alternative exposure metrics were introduced in the chapters 5.1-5.2. To identify the significance of correlations between alternative metrics and possible health effects, specific studies have to be designed comparing health outcomes with exposure concentrations. E.g. Lanki et al. (2006) conducted such a study based on the voluntary exposure of test people in three European cities, Amsterdam, Erfurt and Helsinki. They measured PM_{2.5} mass, ultrafine number concentrations and absorbance as exposure metrics. They also analysed the filters for chemical composition and used these data for a PMF source apportionment. This allowed them to study a health outcome with sources as an additional exposure metric. The results for ST segment depression as health outcome are shown in Figure 71. ST segment depression is an indicator for ischemia, restriction in blood supply.

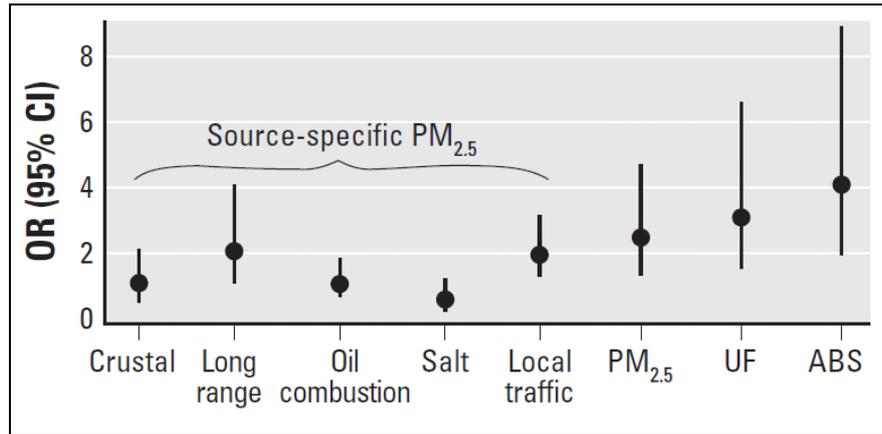


Figure 71: Odds ratio (OR) and 95% confidence interval for 2-day lag for association of different exposure metrics with ST segment depressions (Lanki et al., 2006)

From Figure 71 odds ratios above 1 indicate a positive correlation between exposure metric and the health outcome. Here the highest effects are seen between absorbance, indicator for soot, and ultrafine particle number concentrations and ST segment depression is observed. PM_{2.5} showed higher ratios than any of the identified sources. Interestingly long range transported aerosol seems to have a similar odd ratio as local traffic. While traffic can be expected to be of relevance the outcome for long range transport is somewhat surprising and indicates that possible oxidation products of volatile organic carbons (VOCs) may be health relevant (Jiminez et al., 2009).

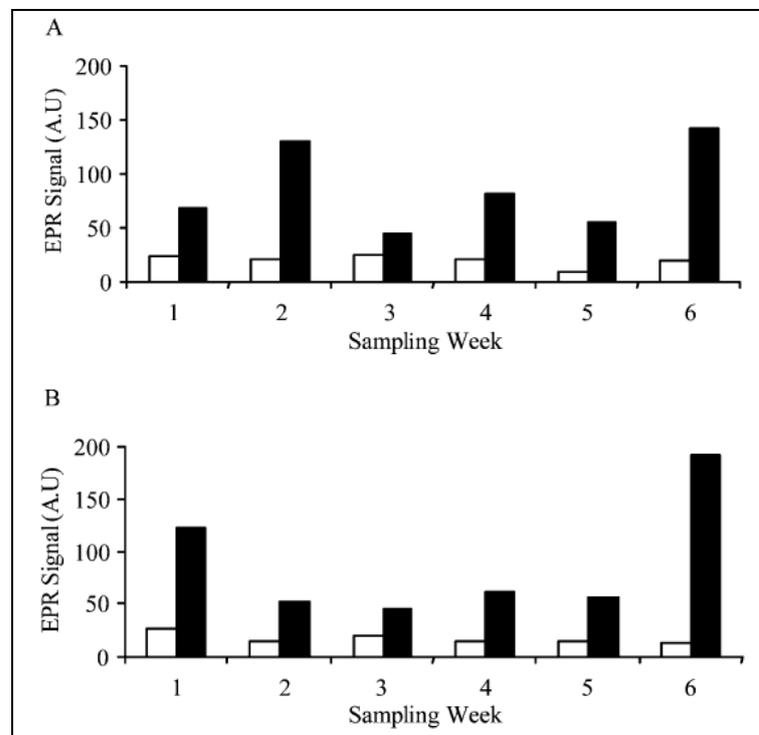


Figure 72: ROS formation by fine (A) and coarse (B) PM samples in Hettstedt (■) and Zerbst (□) (Shi et al., 2003)

Another alternative, possibly health relevant particle characteristic is the particles' ability to form reactive oxygen species (ROS). Shi et al. (2003) studied the formation potential in non cellular media using the electron paramagnetic resonance spectrometry (EPR). After testing the analytical method they investigated the formation potential from PM samples of Zerbst, a less industrialised and Hettstedt, a highly industrialised (smelter) town and demonstrated that ambient particulate matter has varying ability to generate hydroxyl radicals / ROS in dependency of the chemical compounds, particle size and source. Overall the samples from Hettstedt showed a higher ROS formation potential than the Zerbst samples at equal mass.

These two towns were earlier investigated by several epidemiological studies consistently indicating higher morbidity due to respiratory diseases and allergies for Hettstedt despite of similar PM mass concentrations (Heinrich et al., 1999). Gavett et al. 2003 confirmed these results showing a significant increase in airway responsiveness and lung inflammation caused by the Hettstedt but not by the Zerbst dust in a mouse model study. Finally Schaumann et al. 2004 demonstrated distinct airway inflammation in healthy humans living in Hettstedt compared to people living in Zerbst. Both latter studies assume that the detected higher concentration of metals might be responsible for the increased inflammation caused by the Hettstedt dust.

In summary, these results obtained with the EPR analysis can be used to explain the increases in inflammatory effects in Hettstedt. Thereby the ROS generation potential itself seems to play a key role in inducing an inflammatory response and can be taken as an environmental monitoring parameter linked to PM induced inflammatory effects.

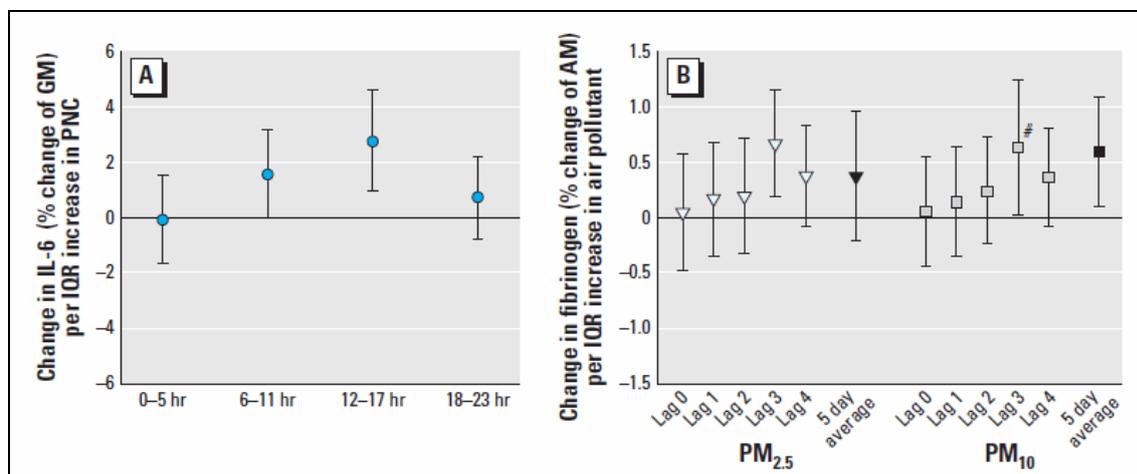


Figure 73: Effects at different lag intervals of particle number concentrations on IL-6 (A) and of PM_{2.5} and PM₁₀ (B) on fibrinogen (Rückerl et al., 2007)

When comparing effect ratios for different metrics a further complication has to be taken into account, the lag time. The effect of different lag times can nicely be seen in the study by Rückerl et al. (2007). They studied the development of biomarkers in blood in dependence on co-determined exposure metrics in this multicity study. IL-6 is an inflammatory blood marker while fibrinogen is a marker for possible clot formation possibly also influenced by inflammation.

5 Particle exposure metrics and health effect studies

Figure 73 nicely shows that the blood marker IL-6 is linked with UFP exposure and that the highest response was found at a lag time of 12-17 hours after elevated exposure. Lag times for PM mass exposure, both PM10 and PM2.5, were significantly longer with the highest response after 4 days with regard to fibrinogen concentrations in blood. Hence, comparing short-term effects and health outcomes of different exposure metrics have to take different lag times into account.

The above discussed health studies were focussed on short term health effects by air pollutants. Long-term health effects are a main issue since (a) they include the sum of all short-term effects and (b) also take into account effects due to chronic exposure. The latter effects include e.g. lung cancer and myocardial infarction. One approach for an exposure assessment for long-term average concentrations to traffic-related air pollution was developed by Hochadel et al. (2006) for the Ruhr-area, Germany. This GIS-based approach was subsequently used by Kraemer et al. (2009) to study possible effects of traffic-related air pollution (TAP) on children. The birth cohorts GINIplus and LISApplus of the Ruhr-area were used for this study of possible correlations between TAP and respiratory or eczema effects. No correlations between TAP and respiratory impairments for children at the age of 6 were observed. In contrast, significant associations between TAP exposure and eczema, prevalence and longer lasting, were observed. This was even observed for small town areas with generally low exposure levels close to the Dutch-German border (Kraemer et al., 2009).

Exposure to air pollution, here specific airborne particulate matter, and the study of possible effects to humans is a complex task which has already been investigated for more than a century. Despite all knowledge gained, major tasks still remain to be tackled. Of which the question of the “best” exposure metric and that of a threshold level, also called level of no effect, remain to be resolved.

6 SUMMARY AND OUTLOOK

Airborne particles have numerous effects on humans, the environment, climate and anthropogenic constructions. Examples are

- short-term and long-term health effects for humans leading to higher morbidity and mortality rates,
- ecosystem effects through acid deposition and nitrification causing ‘waldsterben’,
- climate effects via direct and indirect radiative forcing leading mainly to cooling and
- corrosive effects by enhanced degradation of anthropogenic constructions.

Hence, these effects also lead to significant economic losses. Both, our wish to protect and preserve our lives and the environment, and to save money and be more competitive worldwide drive the research into airborne particles.

In recent years, as described in the introduction, it became obvious that especially fine particles, particles less than 2.5 µm in diameter are linked to most of the abovementioned effects which enhanced corresponding research and technological developments.

Section 2 describes and summarises both, the existing devices to determine and characterise airborne particles as well as the current shortcomings leading to further needs in developments. The focus in this area will be online measurement methods for particle sizes from the nanometer up to 10s’ of micrometre giving multiparameter information. This may be size and chemical composition or size and morphology.

Another focus in this area will be the miniaturisation and development of low cost devices to facilitate the determination and characterisation indoors, on personal level as well as multi-location monitoring in any environment of interest.

These developments will enable improved exposure assessments for humans as well as other living and dead matter in the environment. Exposure assessments in general also include the determination of sources leading to harmful exposure levels. Source identification and quantification is still a major issue in ambient air quality since no simple straight forward methods exist. Section 3 gives examples on how source apportionment can be conducted and summarises the current knowledge. The latter has certainly to be improved to enable source related and hence cost effective abatement measures. Besides improved data availability by the new devices, validation of current source apportionment methods including comprehensive evaluation of source receptor models and chemical transport models (dispersion models) results are needed and should be pursued.

Human exposure at workplaces, especially towards product nanoparticles and nanomaterials, is another focus in exposure research. This is dealt with in section 4 where both, measurement strategies to determine workplace exposure as well as results of exposure related field measurements, are presented and discussed. One major problem in assessing exposure and ensuring workplace safety at ‘nano’ related workplaces is that not enough data for a generalisation is available. Hence two approaches are currently pursued. One building on new measurement devices and exposure related measurements and the other using detailed studies of handling and processing of nanomaterials to assess possible release by the process and hence possible exposure. Another major issue for workplace exposure assessment is the discrimination of background particles, composed

6 Summary and outlook

of other particles than those produced in the plant. This is still an issue and future work has to be put into the harmonisation of approaches for different types of exposure settings and processes.

The release of fine dust into the environment and its detection and characterisation has to be assessed in view of possible health effects. This is the topic of section 5 which also is the topic with the highest uncertainties. It is known and was shown in the introduction that airborne particles, especially fine dust lead to health effects. Current investigations mainly base on mass concentrations as an exposure metric. Other, alternative metrics such as ultrafine particle number concentration, surface area concentrations, black carbon or the potential to form reactive oxygen species are currently also discussed to be of relevance and some investigations are ongoing, studying the correlation of these metrics to health effects. This certainly will be one focus in the future. It may well be that one metric will correlate better to some health endpoints than to others and that another metric is more closely linked to the other health endpoints.

The major aim of the research area “airborne particles and health” is to advance our knowledge on release, exposure, dose and health effect and the interactions inbetween to reduce the adverse health impacts and improve our quality of life.

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1. Bruckmann, P., Kuhlbusch, T.A.J., John, A., Quass, U., Kasper, M. (2011): Staub ist überall- Auch in der Luft, Feinstaub-Klein, fein und gemein, In Chemie über den Wolken, (Eds.) R. Zellner und GDCh, Wiley-VCH, ISBN 978-3-527-32651-8, Weinheim, Germany, 105-119.
2. Kuhlbusch, T.A.J., Asbach, C. (2011): Particle Characterization, In Cardiovascular Effects of Inhaled Ultrafine and nanosized particles, (Eds.) F. R. Cassee, N. L. Mills, D. Newby, John Wiley & Sons, ISBN: 9780470433539, New York, USA, 59-87.
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4. Richter, C.-J., Braun, F.J., Hermsdörfer, K., Höfl, H.-C., Scherer, P., Beyer, M., Hugo, A., Jarzyna, D., Kuhlbusch, T.A.J. (2011): Bestimmung von Emissionsfaktoren für den Kohleumschlag, VDI-Berichte Nr. 2113, ISSN 0083-5560, ISBN 978-3-18-092113-6, Beuth Verlag, Berlin, Germany, 107-130.
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8. Gartiser, S., Nickel, C., Stintz, M., Damme, S., Schaeffer, A., Erdinger, L., Kuhlbusch, T.A.J., Behaviour of nanoscale titanium dioxide in laboratory wastewater treatment plants according to OECD 303, Setac Europe 2011, Mailand, Italy, 15.-19.05.2011.
9. Kuhlbusch, T.A.J., NanoGEM Nanomaterialien – Gesundheit, Exposition und Materialeigenschaften, Sitzung der DECHEMA/VCI Working Group „Responsible Production and Use of Nanomaterials“, Frankfurt, Germany, 17.03.2011.
10. Kuhlbusch, T.A.J., NanoGEM Nanomaterialien – Gesundheit, Exposition und Materialeigenschaften, Bridging NanoEHS Research Efforts-A Joint US-EU Workshop, Washington, USA, 09.03.2011.
11. Asbach, C., Kaminski, H., Fissan, H., Azong-Wara, N., Stahlmecke, B., Kuhlbusch, T.A.J., Broßell, D., Horn, H.G., Portable system for detailed aerosol analysis in the workplace, Annual Forum for NanoSafety, Berlin, Germany, 17.03.2011.
12. Kuhlbusch, T.A.J., Beurteilung der Exposition-Messen alleine reicht nicht, Kompaktkurs, Bochum, Germany, 08.02.2011.
13. Brock, T.H., Berges, M., Pelzer, T., Bachmann, V., Plitzko, S., Wolf, T., Engel, S., Götz, U., ...Asbach, C., Kuhlbusch, T., Tiered-Type Approach to an Exposure Assessment of Nanoscale Aerosols Released from Engineered Nanomaterials in Workplace Operations, Nano 2011 INRS Conference, Nancy, France, 05.-07.04.2011.
14. Nickel, C., Hellack, B., Gartiser, S., Gabsch, S., Stintz, M., Maes, H., Schäffer, A., Erdinger, L., Kuhlbusch, T.A.J., Fate and behaviour of TiO₂ Nanomaterials in the environment influenced by their shape, size and surface area, NIN Conference, Lausanne, Switzerland, 14.-18.02.2011.
15. Hellack, B., Albrecht, C., Schins, R.P.F., Kuhlbusch, T.A.J., Gesundheitsschädigende Effekte von (ultrafeinen bzw. nanoskaligen) Partikeln, Geoökologisches Kolloquium, Braunschweig, Germany, 13.01.2011.
16. Kuhlbusch, T.A.J., Nanomaterialien-Chancen, Risiken, Perspektiven, Dialog-Forum "Nanomaterialien am Arbeitsplatz", Dortmund, Germany, 17.01.2011.

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17. Azong-Wara, N., Asbach, C., Stahlmecke, B., Kaminski, H., Fissan, H., Plitzko, S., Kuhlbusch, T.A.J., Ein neuer personengetragener Thermalpräzipitator (TP) für die Messung nanoskaliger Partikel an Arbeitsplätzen, Dialog-Forum "Nanomaterialien am Arbeitsplatz", Dortmund, Germany, 17.01.2011.
18. Stahlmecke, B., Ravi, L., Azong-Wara, N., Asbach, C., Fissan, H., Kuhlbusch, T.A.J., CarboSafe Airborne CNT-Towards an Online-Measurement Tool, Inno.CNT Jahreskongress 2011, Karlsruhe, Germany, 25.-27.01.2011.
19. CarboSafe Konsortium (auch Stahlmecke, B., Kiesling, Fissan, H., Kuhlbusch, T.A.J., Voetz, M., CarboSafe-Measurement of nanoobjects in the air you breath, Inno.CNT Jahreskongress 2011, Karlsruhe, Germany, 25.-27.01.2011.
20. Stanley, N., Pui, D.Y.H., Kuehn, T.H., Asbach, C., Kuhlbusch, T.A.J., Fissan, H., Nanoparticle fate from a simulated leak during nanoparticle manufacturing, Annual AAAR Conference, Portland, OR, USA, 25.-29.10.2010.
21. Asbach, C., Fissan, H., Stahlmecke, B., Kaminski, H., Pui, D.Y.H., Kuhlbusch, T.A.J., 450 nm cut-off pre-separator with low pressure drop for diffusion charger based nanoparticle measurement, Annual AAAR Conference, Portland, OR, USA, 25.-29.10.2010.
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23. Kuhlbusch, T.A.J., Asbach, C., Fissan, H., Kaminski, H., Atmospheric particle surface area concentration and number size distribution measurements, AAAR Conference, Portland, USA, 26.10.2010.
24. Brand, P., Gube, M., Gerards, K., Bertram, J., Kaminski, H., John, A., Kuhlbusch, T.A.J., Wiemann, M., Eisenbeis, C., Winkler, R., Kraus, T., Innere Exposition, biochemische Effekte und Lungenfunktion bei Schweißern nach Kurzzeit-Exposition mit Schweißrauch von verschiedenen Schweißverfahren und Verfahrensvarianten, Deutsche Gesellschaft für Arbeitsmedizin und Umweltmedizin e.V., 50. Wissenschaftliche Jahrestagung, Dortmund, Germany, 16.-19.06.2010.
25. Asbach, C., Kuhlbusch, T.A.J., Stäube/Feinstäube – Expositionsbestimmung, -erfassung und -beurteilung, 51. Kongress der Deutschen Gesellschaft für Pneumologie, Hannover, 19.03.2010.
26. Asbach, C., Nickel, C., Kaminski, H., Fissan, H., Kuhlbusch, T.A.J., Bestimmung der persönlichen internen Dosis inhalierter Nanopartikel, ProcessNet Fachausschuss Partikelmesstechnik, Karlsruhe, Germany, 09.03.2010.
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29. Kuhlbusch, T.A.J., ENP Characterization in View of Health Studies, Co-Nanomet, Nürnberg, Germany, 28.-29.04.2010.
30. Stahlmecke, B., Ravi, L., Azong-Wara, N., Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Characterization of Dimensions of Carbon Nanotubes using Online Techniques, AAAR, Portland, USA, 29.10.2010.
31. Kuhlbusch, T.A.J., Asbach, C., Kaminski, H., Particle Surface Area Measurements-Are Current Techniques Applicable?, AAMG-RSC and AirMonTech Conference, London, UK, 14.-15.12.2010.
32. Kuhlbusch, T.A.J., AirMonTech 2010 – 2013, AAMG-RSC and AirMonTech Conference, London, UK, 14.12.2010.

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33. Jacobi, S., Kaminski, H., Bruns, K., Kuhlbusch, T.A.J., Neue normkonforme PM10-/PM2,5-Messtechnik aktuelle Hinweise aus der Praxis und Weiterentwicklung, KRdL-Expertenforum, Bonn, Germany, 02.12.2010.
34. Kuhlbusch, T.A.J., John, A., EC/OC/Black-Smoke-Messungen in Europa-Neue europäische Standardisierung, KRdL-Expertenforum, Bonn, Germany, 02.12.2010.
35. Kuhlbusch, T.A.J., Release of and Exposure to Nanoobjects, CeNIDE Science Talk, Essen, Germany, 18.11.2010.
36. Hellack, B., Stahlmecke, B., Albrecht, C., Schins, R.P.F., Kuhlbusch, T.A.J., Intrinsisches ROS Bildungs- und Scavengingpotential verschiedener Kohlenstoffnanoröhren (CNTs), Doktorandenkolloquium, Universitätsklinikum Essen, Germany, 19.11.2010.
37. Nickel, C., Klemm, O., Kuhlbusch, T.A.J., Verhalten und Verbleib von nanoskaligen Partikeln in der Umwelt, IUTA Seminarreihe "Spezielle Fragestellungen der Verfahrens-, Energie- und Umwelttechnik", Duisburg, Germany, 16.11.2010.
38. Kuhlbusch, T.A.J., Asbach, C., Fissan, H., Kaminski, H., Atmospheric Particle Surface Area Concentration and Number Size Distribution Measurements, AAAR Conference, Portland, USA, 28.10.2010.
39. Stahlmecke, B., Kuhlbusch, T.A.J., Fissan, H., CarboLifeCycle Kick-Off-Treffen BAYER Vorhabensbeschreibung IUTA, CarboLifeCycle Kick Off, Leverkusen, Germany, 11.11.2010.
40. Azong-Wara, N., Asbach, C., Stahlmecke, B., Kaminski, H., Fissan, H., Plitzko, S., Kuhlbusch, T.A.J., Experimentelle Evaluierung eines personengebundenen Thermalpräzipitators (TP) für Nanopartikel, CeNIDE Vollversammlung, Duisburg, Germany, 10.11.2010.
41. Hellack, B., Sugiri, D., Quass, U., Albrecht, C. Schins, R.P.F., Krämer, U., Kuhlbusch, T.A.J., PM2.5 induzierte Entstehung von ROS und Diabetes im Raum Duisburg-Wesel, IUTA Seminarreihe "Spezielle Fragestellungen der Verfahrens-, Energie- und Umwelttechnik", Duisburg, Germany, 11.11.2010.
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43. Kuhlbusch, T.A.J., Gesundheit, Umwelt, Sicherheit und Nachhaltigkeit beim Einsatz von Nanomaterialien, BMBF Strategie-Treffen, Themen des 8. FRP, Frankfurt a.M., Germany, 09.11.2010.
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45. Kuhlbusch, T.A.J., Wer oder was verursacht die Feinstäube in unserer Umwelt?, 670. DECHEMA-Kolloquium "Feinstäube: Erkenntnisse, Maßnahmen und Bewertungen", Frankfurt a.M., Germany, 27.10.2010.
46. Nickel, C., Kuhlbusch, T.A.J., Hellack, B., Stintz, M., Gabsch, S., Schiwy, A., Maes, H., Erdinger, L., Gartiser, S., Fate and behavior of TiO₂ nanomaterials in the environment, influenced by their shape, size and surface area, Workshop Maurach "Nanoparticles and Nanomaterials in Aquatic Systems", Maurach, Germany, 29.9.-01.10.2010.
47. Kuhlbusch, T.A.J., Weber, St., Nickel, C., Asbach, C., Kaminski, H., Ultrafeine Partikel im Ruhrgebiet, Zeitliche und räumliche Varianz, Nanosymposium Nanopartikel in der Atmosphäre, Berlin, Germany, 15.09.2010.
48. Kuhlbusch, T.A.J., Test procedures and models related to nanomaterial exposure, 2nd Nanosafety Autumn School, Venedig, Italy, 04.-08.10.2010.
49. Nickel, C., Kuhlbusch, T.A.J., Suspensionsherstellung von Nanomaterialien, Workshop Suspensionserstellung, UBA, Dessau, Germany, 09.09.2010.

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50. Kaminski, H., Asbach, C., Nickel, C., Weber, St., Kuhlbusch, T.A.J., Evaluation of Particle Size Distribution Measurement Techniques for Urban Air Quality Monitoring, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
51. Azong-Wara, N., Asbach, C., Stahlmecke, B., Kaminski, H., Fissan, H., Plitzko, S., Kuhlbusch, T.A.J., Experimental evaluation of a thermophoretic personal sampler for nanoparticles, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
52. Stahlmecke, B., Ravi, L., Azong-Wara, N., Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Development of Online Techniques for the Measurement of Carbon Nanotubes, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
53. Kuhlbusch, T.A.J., Weber, St., Nickel, C., Kaminski, H., Particle Size Dependent Variability and Spatial Representativity of Traffic Site Measurements for Exposure Assessments, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
54. Asbach, C., Fissan, H., Stahlmecke, B., Kaminski, H., Pui, D.Y.H., Kuhlbusch, T.A.J., Development and Evaluation of a pre-separatr for nanoparticle measurement devices, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
55. Wang, J., Asbach, C., Huelser, T., Fissan, H., Kaminski, H., Kuhlbusch, T.A.J., Pui, D.Y.H., Exposure Measurement during the Production of Silicon Nanoparticles in a Pilot Scale Facility, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
56. Fissan, H., Stahlmecke, B., Asbach, C., Kuhlbusch, T.A.J., Wegner, K., Possible Nanoparticle Release from Liquid and Solid Materials into Environment, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
57. Kuhlbusch, T.A.J., Asbach, C., Fissan, H., Kaminski, H., Particle Surface Area Concentrations and Number Size Distributions in View of Exposure Assessment, IAC 2010, Helsinki, Finland, 29.8.-03.9.2010.
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60. Kuhlbusch, T.A.J., Partikel in urbanen Gebieten, Seminarvortrag KIT-IMK, Karlsruhe, Germany, 06.06.2010.
61. Azong-Wara, N., Asbach, C., Stahlmecke, B., Kaminski, H., Fissan, H., Plitzko, S., Kuhlbusch, T.A.J., Development and experimental validation of a thermophoretic personal sampler (TP) for nanoparticle exposure studies, GAeF Workshop Workplace Aerosols, Karlsruhe, Germany, 28.06.-02.07.2010.
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63. Huelser, T.P., Asbach, C., Fissan, H., Kaminski, H., Kuhlbusch, T.A.J., Pui, D.Y., Wang, J., Assessment of Exposure to Engineered Silicon Nanoparticles in a Pilot Plant, Materials Research Society Fall Meeting 2010, Boston, USA, 29.11.-03.12.2010.
64. Kuhlbusch, T.A.J., Hellack, B., Nanomaterial Exposure Measurements and Monitoring for Risk Assessment, Nanotoxicology 2010, Edinburgh, Scotland, 03.06.2010.
65. Kuhlbusch, T.A.J., Nickel, C., Kaminski, H., Weber, S., Räumliche und zeitliche Variabilität von ultrafeinen Partikeln im Ruhrgebiet, Messtechnisches Kolloquium 2010, Eisenach, Germany, 10.-12.05.2010.

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66. Kuhlbusch, T.A.J., Exposure scenarios and exposure monitoring, Workshop "Innovation by nanotechnology and nanomaterials-current aspects of safety assessment and regulation", Dresden, Germany, 22.04.2010.
67. Kuhlbusch, T.A.J., Airborne Nanoparticles: Measurement techniques, characterization and exposure, EMPA, St. Gallen, Switzerland, 14.04.2010.
68. Kuhlbusch, T.A.J., Sustainable Nanotechnology-Nanoparticle exposure, Professional Training on Nanoparticle Synthesis, CeNIDE, IUTA, Duisburg, Germany, 01.03.2010.
69. Nickel, C., Kaminski, H., Weber, St., Kuhlbusch, T.A.J., Räumliche und zeitliche Variabilität von ultrafeinen Partikeln im Ruhrgebiet, LANUV NRW, Essen, Germany, 10.02.2010.
70. Stahlmecke, B., Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Dziurawitz, N., Plitzko, S., Gierke, E., CarboSafe – Erzeugung und Charakterisierung von luftgetragenen CNT, Inno.CNT Jahrestreffen 2010, Marl, Germany, 20.-21.01.2010.
71. Fissan, H., Asbach, C., Stanley, N., Kuehn, T., Pui, D.Y.H., Kuhlbusch, T.A.J., New Techniques to Quantify Nanoparticle Exposure and Internal Dose, DFG-NFS Research Conference "Sustainable Use of Nanomaterials for Novel Engineering Solutions", New York, USA, 14.-17.10.2009.
72. Stahlmecke, B., Kuhlbusch, T.A.J., Asbach, C., Fissan, H., Ravi, L., CarboSafe Projekttagung Bayer, Stand der Arbeiten – IUTA, CarboSafe Projekttagung Bayer, Leverkusen, Germany, 18.11.2009.
73. Stanley, N., Pui, D.Y.H., Kuehn, T., Asbach, C., Kuhlbusch, T.A.J., Fissan, H., The fate of Accidentally released nanoparticles into a simulated workplace environment, AAAR 28th Annual Conference, Minneapolis, USA, 26.-30.10.2009.
74. Asbach, C., Rating, U., van der Zwaag, T., Fissan, H., Kuhlbusch, T.A.J., CFD modeling of nanoparticle dispersion in a workplace upon accidental release, AAAR 28th Annual Conference, Minneapolis, USA, 26.-30.10.2009.
75. Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Pui, D.Y.H., Wang, J., Analytical-statistical modeling of nanoparticle deposition probability on inverted surfaces at low pressure, AAAR 28th Annual Conference, Minneapolis, USA, 26.-30.10.2009.
76. Stahlmecke, B., Wagener, S., Asbach, C., Kaminski, H., Fissan, H., Kuhlbusch, T.A.J., Stability of Nanopowder Agglomerates in Orifice Flow, AAAR 28th Annual Conference, Minneapolis, USA, 26.-30.10.2009.
77. Stanley, N., Kuehn, T.H., Pui, D.Y.H., Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Experimental and Numerical Simulation of the Fate of Airborne Nanoparticles from a Leak in an Manufacturing Process to Assess Worker Exposure, NSF/EPA-Meeting, Las Vegas, USA, 9.-10.11.2009.
78. Kaminski, H., John, A., Asbach, C., Wiemann, M., Bruch, J., Eisenbeis, C., Winkler, R., Gube, M., Brand, P., Kraus, T., Kuhlbusch, T.A.J., Investigation of physico-chemical and toxicological properties of particles emitted from different welding processes, AAAR 28th Annual Conference, Minneapolis, USA, 30.10.2009.
79. Fissan, H., Kuhlbusch, T.A.J., Auf dem Wege zur ökologischen Nachhaltigkeit von Nanoprodukten, GHUP Jahrestagung, Stuttgart, Germany, 08.10.2009.
80. Quass, U., Beuck, H., Pfeffer, U., Bruckmann, P., Kuhlbusch, T.A.J., PM10 und natürliche Feinstaubquellen-Bestimmung in der Routine, KRdL-Expertenforum, Bonn, Germany, 07.10.2009.
81. Nickel, C., Kaminski, H., Quass, U., John, A., Kuhlbusch, T.A.J., Calculation particle number emission factors of motorway traffic in Germany, AAAR 28th Annual Conference, Minneapolis, Minnesota, USA, 26.-30.10.2009.
82. Fissan, H., Asbach, C., Stahlmecke, B., Kuhlbusch, T.A.J., Determination of CNT-Emission and Exposure in Air During Nanocompound Manufacturing and Handling, CNTComp 2009, Hamburg, Germany, 20.-23.09.2009.

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83. Kuhlbusch, T.A.J., Die neue EU-Luftqualitätsrichtlinie und ihre Anforderungen an die Standardisierung, KRdL-Förderkreissitzung, Karlsruhe, Germany, 30.09.2009.
84. Richter, C.-J., Braun, F.J., Höfl, H.-C., Enderle, K.-H., Wiegele, A., Beyer, M., Hugo, A., Jarzyna, D., Kuhlbusch, T.A.J., Ableitung von Emissionsfaktoren für Staub aus offenen Kohlelagern von Steinkohle und Braunkohlekraftwerken, VDI-Tagung "Diffuse Emissionen", Düsseldorf, Germany, 15.-16.09.2009.
85. Hugo, A., Beyer, M., Jarzyna, D., Kuhlbusch, T.A.J., Richter, C.-J., Anwendung des Reverse Dispersion Modelling (RDM) bei Gasen, Stäuben und Partikeln, VDI-Tagung "Diffuse Emissionen", Düsseldorf, Germany, 15.-16.09.2009.
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87. Fissan, H., Asbach, C., Kaminski, H., Kuhlbusch, T.A.J., Messtechnik für Produktnanopartikel in der Umwelt Anforderungen und Stand der Technik, PTB-Seminar Nanopartikel-Charakterisierung, Braunschweig, Germany, 18.-19.05.2009.
88. Fissan, H., Asbach, C., Kuhlbusch, T.A.J., Exposure to Nanoparticles, GAeF-Workshop, Karlsruhe, Germany, 05.09.2009.
89. John, A.C., Kaminski, H., Wiemann, M., Bruch, J., Eisenbeis, C., Winkler, R., Gube, M., Brand, P., Kraus, T., Kuhlbusch, T.A.J., Physico-chemical and toxicological characterization of welding fumes from different processes, European Aerosol Conference 2009, Karlsruhe, Germany, 06.-11.09.2009.
90. Hugo, A., Beyer, M., Kuhlbusch, T.A.J., Braun, F., Wiegele, A., Richter, C.J., Fugitive PM10/PM2.5 emission factors for coal fired power plants, European Aerosol Conference 2009, Karlsruhe, Germany, 06.-11.09.2009.
91. Asbach, C., Rating, U., van der Zwaag, T., Fissan, H., Kuhlbusch, T.A.J., Modelling of accidental nanoparticle release into a workplace, European Aerosol Conference 2009, Karlsruhe, Germany, 06.-11.09.2009.
92. Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Pui, D.Y.H., Wang, J., Deposition probability of nanoparticles on inverse surfaces at low pressure, European Aerosol Conference 2009, Karlsruhe, Germany, 06.-11.09.2009.
93. Stahlmecke, B., Wagener, S., Asbach, C., Kaminski, H., Fissan, H., Kuhlbusch, T.A.J., Comparison of Agglomerate Stability of Different Nanopowders, European Aerosol Conference 2009, Karlsruhe, Germany, 06.-11.09.2009.
94. Nickel, C., Kaminski, H., Quass, U., John, A., Kuhlbusch, T.A.J., Field measurements of particle number emission factors for freeways, European Aerosol Conference 2009, Karlsruhe, Germany, 06.-11.09.2009.
95. Kuhlbusch, T.A.J., Aerosol exposure and health effects: Do we know the basis?, EAC 2009, Karlsruhe, Germany, 06.-11.09.09.
96. Asbach, C., Qi, C., Shin, W.G., Pui, D.Y.H., Fissan, H., The effect of particle pre-existing charge on unipolar charging and its implication on electrical aerosol measurements, EAC 2009, Karlsruhe, Germany, 6.-11.09.09.
97. Azong-Wara, N., Asbach, C., Stahlmecke, B., Fissan, H., Kaminski, H., Plitzko, S., Kuhlbusch, T.A.J., A new personal thermophoretic sampler with simplified analysis routines for nanoparticle exposure studies, EAC 2009, Karlsruhe, Germany, 06.-11.09.09.
98. Nickel, C., Weber, St., Kaminski, H., Pfeffer, U., Bruckmann, P., Kuhlbusch, T.A.J., Spatial and temporal variability of ultrafine particles within an urban agglomeration, the Ruhr-Area, Germany, EAC 2009, Karlsruhe, Germany, 06.-11.09.09.
99. Hellack, B., Quass, U., Wick, G., Albrecht, C., Schins, R.P.F., Kuhlbusch, T.A.J., Analysis of ROS generated by PM10 sampled at a rural and urban location in North Rhine Westphalia, EAC 2009, Karlsruhe, Germany, 06.-11.09.09.

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100. Quass, U., Sperber, O., Romazanowa, O., Luther, F., Caspari, A., Beyer, M., Pfeffer, U., Zang, T., Bruckmann, P., Kuhlbusch, T.A.J., Approaches to determine the contribution of natural sources to PM10 concentrations in North-West Germany, EAC 2009, Karlsruhe, Germany, 06.-11.09.09.
101. Pisani, F., Kuhlbusch, T.A.J., Kaminski, H., Hänninen, O., Jöckel, K.-H., Hoffmann, B., Personal exposure to UFP in distinct microenvironments, 54. Jahrestagung der Deutschen Gesellschaft für Medizinische Informatik, Biometrie und Epidemiologie e.V. (GMDS), Essen, Germany, 07.-10.09.2009.
102. Kuhlbusch, T.A.J., Charakterisierung von Feinstaub zur Beurteilung von Quellen und Exposition, GDCh-Wissenschaftsforum Symposium "Feinstaub-Prozesse und Wirkungen", Frankfurt a.M., Germany, 31.08.2009.
103. Nickel, C., Kuhlbusch, T.A.J., Studie zur Emission von Nanopartikeln aus Produkten in ihrem Lebenszyklus und ihre ökologische Bewertung-Messtechnik-, UBA Fachgespräch-Projektvorstellung -, Berlin, Germany, 26.08.2009.
104. Nickel, C., Kuhlbusch, T.A.J., Studie zur Emission von Nanopartikeln aus Produkten in ihrem Lebenszyklus und ihre ökologische Bewertung, UBA Fachgespräch, Berlin, Germany, 26.08.2009.
105. Asbach, C., Fissan, H., Kuhlbusch, T.A.J., Mögliche Projektthemen, Processnet-Partikelmesstechnik, Frankfurt a.M., Germany, 03.07.2009.
106. Asbach, C., Kuhlbusch, T.A.J., Exposure Measurements – Examples, Catholic University of the Sacred Heart, Rom, Italy, 30.06.2009.
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