UFIREG
Ultrafine Particles - an evidence based contribution to the development of regional and European environmental and health policy

Importance of UFP measurements

Report

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The UFIREG project aims to investigate the impact of ultrafine particles (UFPs, less than 100 nm in diameter) and other air pollutants on human health. Whereas its main focus lies on the implementation and harmonization of UFP measurements in the project cities as a basis for the epidemiological studies, it also aims to develop long-term strategies for regular measurements of UFPs. In this report, we discuss the need for regular measurements of UFPs in air quality monitoring networks.

Epidemiological studies have shown that particulate matter (PM) is associated with adverse health effects (Brunekreef and Holgate, 2002; Dockery, 2009; Brook et al., 2010; Rückerl et al., 2011). The vast majority of published epidemiological studies reported the use of PM mass concentrations as an indicator of exposure to PM, because it can be measured relatively simply and accurately on a continuous basis. However, atmospheric particles originate from a variety of sources and possess a wide range of morphological, chemical, physical, and thermodynamic properties. Thus, mass concentration is a relatively broad indicator for the particle mixture contained in ambient air. In general, more specific indicators are often used to characterize ambient PM. Classification of PM by size is quite common because size governs the transport and removal of particles from the air and their deposition within the respiratory system and is at least partly associated with the chemical composition and sources of particles (WHO, 2004).

The diameters of ambient particles range from a few nanometers to 100 µm, spanning 5 orders of magnitude. Typically, the mass-based size distribution of ambient PM shows a bimodal shape, with a "minimum" concentration in the size range of 1–3 µm distinguishing coarse and fine modes: by convention, the coarse mode consists of particles between 2.5 and 10 µm ($PM_{2.5-10}$) in aerodynamic diameter whereas the fine (or accumulation) mode is comprised of particles ≤ 2.5 µm in aerodynamic diameter ($PM_{2.5}$). Both, coarse and fine PM fractions seem capable for inducing toxicity and hence adverse health effects. Coarse particles may reach the upper part of the airways and lung and tend to deposit higher up in the airways whereas smaller particles (in particular $PM_{2.5}$) penetrate more deeply into the lung and may reach the alveolar region. So, epidemiological studies have demonstrated larger health effects for PM_{10} (particles ≤ 10 µm in aerodynamic diameter) and $PM_{2.5}$ than for total suspended particulates (TSP). As a direct consequence of this knowledge, air quality standards for PM_{10} and $PM_{2.5}$ – replacing limits for TSP – were introduced in 1996 and 2008 (EC, 1996; EC, 2008), leading to a continuous evolution of focus from TSP to PM_{10} and further to $PM_{2.5}$. In contrast to $PM_{2.5}$ and PM_{10} particles, all particles < 100 nm in diameter (UFPs) contribute only marginally to particle mass concentration, but this size fraction contains the majority (in numbers) of the ambient particles and an appreciable portion of total surface area.

According to Hinds (1999), UFPs represent more than 85% of the total $PM_{2.5}$ particle number. The UFP size fraction consists of two particle modes: nucleation mode (< 30 nm) and Aitken mode (within 20-100 nm) particles (Hussein et al., 2004). It should be noted, however, that there are also some other studies using different size ranges for nucleation mode and Aitken mode particles. Both particle modes (nucleation and Aitken) are not considered by the mass-based size distribution. The accumulation particles are mostly formed by coagulation and agglomeration of the nucleation and Aitken mode particles.

As the mass concentration (total mass per unit volume of air) of UFPs is very low, the direct measurement of UFPs is challenging. Due to the low mass concentration (typically less than 1 µg/m^3) long collection times on filters are required to obtain sufficient mass that could be detected by a commercial balance. The long collection time can influence in turn another factor (for example gas-to-particle artefact) that could significantly affect the mass measurement method (HEI Review Panel on Ultrafine Particles, 2013). Other methods to determine the UFP concentration in the ambient air are measurements of *quasi-ultrafines* (particles < 0.180 µm ($PM_{0.180}$) or < 0.250 ($PM_{0.250}$) in diameter) by means of cascade impactors, measurements of individual chemical compounds in the $PM_{0.1}$ size range (lower detection limits for individual compounds as for UFP mass concentration), and measurement of the particle surface area concentration. However, the most common measurement of UFP concentration is the determination of particle number concentration (PNC). Because of the reliability of PNC measurements, number concentration data are far more common than particle mass ($PM_{0.1}$), UFP composition or surface area data.

Currently, only mass-based limit values for particulate matter ($PM_{2.5}$, PM_{10}) are established. Scientists criticize that there are no limit values for PNC in ambient air. However, why is the smallest particle size fraction a matter of concern, as those small particles contribute so little to the PM mass concentrations in the ambient air?

First of all, it is unlikely that all particles, irrespective of size/chemical composition/source, will have the same health effect. Compared to the larger particle size fractions, UFPs have some different properties and represent an additional independent characteristic of the urban aerosol not fully characterized by $PM_{2.5}$ and PM_{10}. The correlation between the two particle fractions (PM mass concentration – UFPs) is rather low. Furthermore, UFPs are often suggested as a marker of locally emitted primary particles. They are produced in large numbers in urban areas especially by combustion processes such as traffic (tailpipe emissions from motor vehicles), coal-fired power plants or domestic heating (Morawska et al., 2008; Zhu et al., 2002).

Another source of UFPs is the secondary particle nucleation from gaseous precursors (Brock et al., 2002; Holmes, 2007; Kulmala and Kerminen, 2008). Also, the chemical composition of UFPs differs from that of larger particles. The main chemical constituents of UFPs are carbonaceous material stemming from combustion processes such as elemental carbon (EC) and organic carbon (OC) and to lesser extent secondary particle components like sulfate, nitrate, and ammonia.

As stated before, UFPs contribute only slightly to PM_{10} or $PM_{2.5}$ mass but have large surface-to mass ratio and high number concentration. The high number concentration along with a large surface-to mass ratio results in a large bio-available surface, which leads to a greater bio-availability of the adsorbed or condensed toxic air pollutants (oxidant gases, organic compounds, and transition metals) on the particle surface (Oberdörster, 2001). The different properties of UFPs when compared with larger particles make this particle fraction of great concern because of their potential adverse human health effects. The most important properties were summarized by Peters et al. (2011):

- They deposit with higher efficiency in the alveolar region and to a lesser extent in the larger airways.
- Their motion is defined by diffusion rather than their aerodynamic properties.
- They have little mass but high number and surface area concentration.
- They are not well recognized and cleared by macrophages in the alveolar space.
- They potentially translocate into cells (and consequently into extra-pulmonary organs) through diffusion mechanism.

The first evidence of the health effects related to exposure to UFPs came from animal and in vitro toxicological studies. It was postulated that the number of UFPs is a more relevant exposure metric than their mass, because of their larger surface area. The first epidemiological studies on UFPs have been panel studies, which generally showed associations between short-term exposure to UFPs and occurrence of acute respiratory symptoms and lung function (Peters et al., 1997; Penttinen et al., 2001). However, few epidemiological studies have assessed more severe end points such as daily mortality and hospital admissions (e.g. Wichmann et al., 2001; Forastiere et al., 2005; Stolzel et al. 2007; Breitner et al., 2009). In these studies, UFPs have been found to have health effects of similar magnitude as larger particles, but the effects are suggested to be independent of the effects of $PM_{2.5}$ or PM_{10} (Pekkanen et al., 2002; Stolzel et al., 2007). However, although there is a growing body of scientific literature that addresses the health effects related to UFPs, it is not enough to draw definitive conclusions about the specific consequences of exposure to UFPs.

As concluded in a recently published review on the health effects of UFPs (HEI Review Panel on Ultrafine Particles, 2013), there are still limitations and inconsistencies in the findings from short-term studies on health effects of UFPs, and until now there are no long-term studies on health effects of UFPs. Furthermore, only relatively few studies have directly compared UFPs with other particle size fractions and no quantitative summary of the effects of UFPs could be made because of the paucity of data. Moreover, the large majority of the short-term effect studies on UFPs were conducted primarily in Western European countries and almost no studies were conducted in other parts of the world. In conclusion, the rare data about health effects of UFPs do not allow defining target or limit values for this particle fraction yet (HEI Review Panel on Ultrafine Particles, 2013).

Similar conclusions were drawn by the WHO project REVIHAAP, which was designed to inform revisions of European Union policies on air quality in 2013 (WHO, 2013). It was concluded that although there is considerable evidence that UFPs can contribute to the health effects of PM, the scientific base is too small to work on a guideline for the number of UFPs and to propose a guideline value. Moreover, as there are no epidemiological studies on long-term exposure to UFPs the data on the concentration–response function are too scarce to evaluate and recommend an air quality guideline.

In general the REVIHAAP project identified three critical data gaps regarding UFPs: (a) lack of epidemiological evidence on the effect of UFPs on health, with only a handful of studies published on this topic; (b) insufficient understanding of whether the human health effects of UFPs are independent of those of $PM_{2.5}$ and PM_{10}; and (c) evidence of which ultrafine particle physical or chemical characteristics are most significant to health.

One reason for the limited number of epidemiological studies on UFPs is that in most locations measurement of ambient UFPs is not conducted routinely at the monitoring stations operated by the local air quality network. It seems that it is a chicken–egg problem:

a) UFPs are not routinely monitored by the air quality monitoring network as there are no limit values for the smallest particle fraction.
b) The limit values could not be proposed due to a limited number of studies published on this topic.
c) The insufficient amount of data about health effects of UFPs is largely because these are not routinely monitored by the air quality monitoring network (see a).

The air quality monitoring strategy proposed by the REVIHAAP project could help to break this vicious circle. One of the main implications was that more monitoring is needed, both regularly by local air quality networks and in the framework of projects with health specialists. The use of "supersites" to perform simultaneous studies using the same monitoring and health evaluation approaches across Europe was strongly suggested.

The air quality monitoring strategy proposed by the REVIHAAP project could help to break this vicious circle. One of the main implications was that more monitoring is needed, both regularly by local air quality networks and in the framework of projects with health specialists. The use of "supersites" to perform simultaneous studies using the same monitoring and health evaluation approaches across Europe was strongly suggested. At such supersites additional air quality parameters, such as size-segregated UFPs, online PM speciation measurements, surface area, oxidative potential and other parameters should be measured. New studies should be conducted with a multi-pollutant approach for establishing concentration-response functions for those additional PM exposure metrics (WHO, 2013).

The necessity of the multi-pollutant approach was also underlined by the HEI panel review (HEI Review Panel on Ultrafine Particles, 2013). Many epidemiological studies on UFP effects on human health did not account or adjust for the potential association with gaseous pollutants (especially of those associated with traffic such as CO or NO_2) or other particle metrics. The authors pointed out that one of the factors that has limited the comparison and interpretation of the epidemiological studies conducted to date on the short-term effects of ambient UFPs is the variability in study designs, both in exposure methods and measurements (including co-pollutants) as well as in the health outcomes across individual studies and cities. This makes any kind of meta-analysis to strengthen interferences from those short term studies (as already done for $PM_{2.5}$ and PM_{10}) difficult and no such analyses have been conducted up to now. An implementation of UFP measurements at "supersites" across Europe might lead to more harmonization of the exposure methods, measurements and study designs (adjustment for co-pollutants) and will make future epidemiological studies more comparable.

The harmonization and quality assurance of UFP measurements is an important point. As recently confirmed by the AirMonTech project (http://www.airmontech.eu/), the measurement techniques for PNC are not as advanced and harmonized as those for PM_{10}, $PM_{2.5}$ or black carbon. Moreover, the quality of the existing data may be variable and not directly comparable. AirMonTech is a European project aiming at the compilation of the knowledge and information needed to harmonize current air pollution measurements and to guide decisions about future monitoring.

One of the key recommendations of AirMonTech was that additional pollutants or characteristics of known pollutants may also be of importance for public health and should thus be included into a comprehensive AQ monitoring strategy. Priority parameters for extended field trials are real-time methods for black carbon, particle surface area concentration, particle number concentration (i.e. UFPs), and some others.

The focus of networks required by the Air Quality Directive should be broad enough at least to include an assessment of compliance with EU standards in background and hotspot sites, and the assessment of population-based exposure appropriate for health effect studies. It was strongly recommended to integrate permanent "research sites" measuring a large range of pollutants in carefully-chosen sites into the air quality monitoring networks. It was made explicit that national monitoring networks have aims beyond compliance monitoring, such as clarification of health effects, source apportionment, and abatement assessment.

Summary

Ultrafine particles (UFPs) contribute only slightly to PM_{10} or $PM_{2.5}$ mass but have large surface-to mass ratio and high number concentration. The different properties of UFPs when compared with larger particles make this particle fraction one of great concern because of their potential adverse human health effects. Epidemiological studies have provided evidence that the adverse health effects of exposure to UFPs differ from those of larger particles. However, as concluded by the REVIHAAP project (WHO, 2013) the scientific base is too small to work on a guideline for PNC in the ambient air and to propose a guideline value. Moreover, there are limitations and inconsistencies in the findings from short-term studies on health effects of UFPs, and there are no epidemiological studies on long-term effects of UFPs on human health (HEI Review Panel on Ultrafine Particles, 2013). One of the major limitations of the current studies is that UFPs have been assessed in different ways by using different measurement techniques in the different studies. This is because ambient monitoring of UFPs is not conducted by the air quality monitoring network in most locations, and the measurements were done by researchers without any harmonization of the measurement techniques.

As concluded by some European projects (REVIHAAP, AirMonTech), more monitoring of UFPs in air quality monitoring networks is needed, both regularly by local air quality networks and in the framework of projects with health specialists. The use of "supersites" to perform simultaneous studies using the same monitoring and health evaluation approaches across Europe was strongly suggested. It was pointed out that standardized measurement techniques for PNC monitoring are needed, and that also additional pollutants or characteristics of known pollutants may be included into a comprehensive AQ monitoring strategy. More data on UFPs will allow to investigate the spatial and temporal variation of UFPs (which can be very large), will provide information on possible trends, and last but not least will provide a good database for future epidemiological studies on health effects of UFPs. More epidemiological findings related to UFPs will allow establishing concentration-response functions and the development of limit values for PNC in the ambient air.

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