New Methods and Standards for Fine Dust

Jürgen Spielvogel, Stefan Hartstock and Hans Grimm
Grimm Aerosol Technik GmbH, Dorfstrasse 9, 83404 Ainring, Germany
ECOMESURE, 3, rue du Grand Cèdre, 91640 JANVRY, France
E-mail: jsp@grimm-aerosol.

Abstract. There seems to be common agreement that PM10 is a suboptimal quantity for the quantification of potential dangers from fine dust due to a number of reasons, notably because the chemical composition of the particles is not considered, because the size distribution is disregarded, and because of sampling artefacts. In a first step for improving the particle measurements, the European Community has published new directives for ambient air in June 2008 (EU 2008), which as a main part included new regulations for PM2.5 measurements, in addition to the further on valid regulations for PM10. The comparison of PM2.5 and PM10 may allow a source apportionment and a better assessment of the influence of fine dust on human health. The source apportionment may allow more effective fine dust reduction strategies.

1. The coming PM2.5 regulations in Europe
In order to prepare the coming limits for PM2.5, the European directive 2008/50/EC requires new PM2.5 measurement sites in addition to the existing PM10 measurement sites. The measurements must be started at January 1st 2009 and the new directive and regulations must be part of governmental laws and process before the 11th of June 2010. The regulations state that a minimum number of PM2.5 measuring points must be installed, namely one PM2.5 site for every million people in metropolitan areas and at least one measuring point in cities with more than 100.00 people. As an additional general rule, one PM2.5 site must be installed for two PM10 sites. The regulations required also a minimum total number of measuring points (PM2.5 and PM10), which depends on PM10 concentrations and which is higher for polluted areas.

As for the data quality, the regulations state that the uncertainty must be smaller than 25 % and that the minimum data capture must not be less than 90%. The data must be reported to the European Union.

| Year | Concentration [µg/m³] | Limit type |
|------|----------------------|------------|
| 2010 | 25                   | Target     |
| 2015 | 25                   | Limit      |
| 2020 | 20                   | Limit      |
The new regulations include future limits for PM2.5 concentrations as listed in Table 1. As a rule of the thumb, PM2.5 concentrations are typically ~60% of the PM10 concentrations, and considering the present limits for PM10, annual mean < 40 μg/m³ and not more than 35 days > 50 μg/m³, these new limits for PM2.5 are hardly stronger than the old limits for PM10.

There will, however, also be exceptions for the coming PM2.5 limits. Such exceptions are permitted by the EC for a country, region, or city, if the authorities present scientific reasons based on long time measurement for the impossibility of compliance with the limits. Potential reasons could be high background values due to long-range transport or special local orographic or weather conditions.

Each candidate Method (CM), which will be used to measure PM2.5 concentrations needs an approval according to EN14907, similar to the approval for PM10 according to EN12341. For such an approval, daily values in μg/m³ measured by the CM are compared against a Reference Methods (RM).

2. Principle of optical aerosol spectrometers

Unlike nephelometers, aerosol spectrometers measure scattering intensity of each individual particle as a proxy for particle size. The light scattering process has three size regions according to the value of the optical size parameter α, \( \alpha = \pi \frac{D_p}{\lambda} \), with the aerosol particles diameter \( D_p \) and the wavelength of the incident light \( \lambda \) (Mie 1908). For \( \alpha \ll 1 \), Rayleigh-scattering occurs, where the intensity of the scattered light is proportional to the sixth power of particle diameter (\( I \sim D_p^6 \)). The range of \( \alpha \approx 1 \) is addressed as Mie-scattering, where a strong interaction between the aerosol particle and the incident beam takes place. For Mie scattering, the intensity of the scattered light is a complex function of particle diameter and refractive index. The GRIMM optical spectrometers use however a special procedure to retrieve the contained size information to a large extent. The case \( \alpha \gg 1 \), Geometric Optics, comprises reflection, refraction diffraction and absorption, and the intensity of the scattered light is usually proportional to the particle cross-sectional area (\( I \sim D_p^2 \)).

![Figure 1. Principle of the GRIMM optical spectrometer.](image)

Principle of the GRIMM aerosol spectrometers is illustrated in Figure 1. A laser-diode is used as light source (\( \lambda = 655 \text{ nm} \)), and the beam is focused into a well defined, nearly homogeneously illuminated optical measuring volume. The scattered light from a scattering angle of 90° with an aperture angle of 30° is collected by on a photodiode. This geometry is part of the measures used to minimize the influence of refractive index on scattering intensity and to compensate the ambiguities of Mie-scattering. The detected signal is then amplified and used by a pulse height analyzer. Thus the GRIMM instruments reports particle number concentrations for 31 channels covering the size range of 0.25 – 32 μm with a time resolution of 6 s. Higher time resolution of 1 s only is possible for a reduced size range and allows the use of the instrument for flux measurements with eddy-correlation methods.

The measured number size distributions are converted into the required PM10, PM2.5 or, if desired, PM1 concentrations. The conversion is a challenge because it requires detailed investigation of aerosol properties.
These GRIMM aerosol spectrometers are very small, rugged, and require only a minimum of maintenance essentially without consumables. They can be equipped with a variety of accessories such as Nafion dryers to reduce humidity in sampling air, heated inlets to investigate the content of semivolatiles, outdoor housings, integrated filters for the collection of detected particles, or integrated batteries for mobile use. Here we describe three versions of the optical spectrometer, namely the Environmental Dust Monitor (EDM) 180 for official PM10 and PM2.5 measurements, the Wide Range Aerosol Spectrometer for measuring size distributions also of particles smaller than the lower size limit of the aerosol spectrometer (0.25 µm), and The EDM 265 for quantifying the semivolatile mass fraction.

3. The EDM 180 for official PM measurements
This 19” rack version of the aerosol spectrometer is designed for the long term measurement in official networks, it is certified for PM10 according to EN12341 and the certification for PM2.5 according to EN14907 will soon be accomplished. The instrument includes a dehumidification system based on a NAFION tube, which is activated at an ambient relative humidity above 70%. It has a GESYTEC MODE for online data transfer.

A considerable advantage for the coming PM2.5 regulations is that the system can simultaneously measure both, PM2.5 and PM10 measurements. Thus a single instrument constitutes a PM10 and a PM2.5 site, and existing users of the EDM 180 will not need any new equipment measuring PM2.5. The system is also able to measure PM1 concentrations or total number concentrations for which, in the long run, new regulations might be established.

4. The Wide Range Aerosol Spectrometer (WRAS) to measure the full size range
The optical spectrometers can be combined with a GRIMM Scanning Mobility Particle Sizer with Condensation Particle Counter as detection system (SMPS+C). Such a SMPS+C, when operated with a differential mobility analyser (DMA) of the type M-DMA, covers the size range of 5 – 350 nm. Therefore the WRAS, i.e. optical spectrometer with additional SMPS+C, covers the size range from 5 nm to 32 µm with 72 size channels. This constitutes a comprehensive information on the particle size distribution essentially over the complete range of airborne particles. A special software accomplishes the synchronisation of optical spectrometer and SMPS+C, the time resolution is determined by the scan time of the SMPS+C and equals 4 min. If desired, the EDM data in better time resolution are still available on the internal memory of the device.

The WRAS is also available as stand-alone outdoor system, with stainless steel housing, temperature and humidity sensor, optional rain and wind sensor, suitable for long term measurement.

Figure 2. Example for particle size distribution measured with a WRAS system on top of the Fronau tower in Berlin at 11th of June, 2007. The high concentrations of ultrafine particles in the evening are caused by photochemistry.
Figure 2 shows an example of size distributions measured with WRAS in a comprehensive study on the sources of fine dust in Berlin (Pesch 2007). In this study the excellent size resolution of the WRAS was used, together with chemical speciation and gas measurements, for source apportionment. The result was that local sources are only a small contribution to the total PM concentrations. Thus the fine dust concentrations in Berlin can only be reduced effectively if the contribution from long range transport is reduced; important sources for the long range transport are power plants in Eastern Europe. The study highlighted also a poor correlation between PM2.5 and particle number concentrations, a case for considering particle number concentrations in future fine dust regulations.

5. The EDM 265 for measurements of the semi-volatile fraction

Existing epidemiological studies require a sophisticated statistics to detect significant but small health effects from fine dust. Part of the reason for the only small effects is probably that the used PM10 or PM 2.5 data are not a good measure for adverse health effects from ambient dust since the composition of the aerosol is disregarded. It can be suspected that the health effects of ambient particulate matter are rather caused by the solid non-volatile constituents than by the semivolatile fraction, and we speculate that an epidemiological study on the connection of non-volatile particulate matter and health effects would show much stronger effects than previous results based on total PM concentrations.

Numerous studies have shown that the contribution of semivolatile compound to the PM2.5 mass concentration is typically 20 – 50% and that these semivolatile compounds are mostly nitrates, ammonium, and organic material.

The EDM 265 is devoted to the measurement of the volatile fraction, it is a stand-alone outdoor system with a heated sample pipe to evaporate semivolatile material. The sample pipe is however not continuously heated as in other instruments that just loose the semivolatile fraction, but only periodically so that the semivolatile fraction can be assessed from the concentration difference between heated and non-heated periods. The temperature of the sampling pipe can be selected between ambient and 300°C, the use of different temperatures allow to distinguish between different classes of semivolatile material.

![Figure 3. Data from the GRIMM model 265, shown are 7 from 31 size channels. The strong regular concentration changes are induced by heating – and non-heating periods of the sample pipe and allow a quantification of the semivolatile fraction.](image)

6. Conclusions and Outlook

We expect that the PM values will be reduced and that future fine dust regulations will include limits for PM-1 or even for number concentrations of nano particles, similar to new EURO5 regulations for emissions from car engines. GRIMM spectrometers can already today measure PM1 values and they can be adapted to additional nano-particle measurements with accessories that fit in existing outdoor shelters.
References

[1] EN 12341 (1998). Air quality - Determination of the PM10 fraction of suspended particulate matter - Reference method and field test procedure to demonstrate reference equivalence of measurement methods; Beuth Verlag, Berlin.

[2] EU directive 2008/50/EC.

[3] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2008:152:0001:0044:EN:PDF

[4] Mie G 1908 Ann. Phys. 25 377

[5] Pesch M 2007 2. Zwischenbericht zur Ursachenanalyse der PM 2,5-Immissionen in Berlin. Technical report, TU-Berlin Fachgebiet Umweltverfahrenstechnik