Chapter II.3
Ground-Based Observing Systems for Atmospheric Aerosol Chemistry and Composition

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1 Introduction

The most familiar ground-based measurements of atmospheric aerosols are those performed every day in the monitoring stations for air quality in the cities. Adverse health effects of even low levels of atmospheric aerosol concentrations were demonstrated by long-term epidemiological studies, since that of Dockery et al. (1993), which showed a correlation between increased mortality and the concentrations of PM_{10} and, more tightly, of $PM_{2.5}$. "PM_{10}" and "$PM_{2.5}$" refer to the total dry masses of aerosol particles with diameters lower than 10 and 2.5 μm, respectively. These familiar notations contain important concepts of aerosol observations: ambient aerosol particles have a weighable mass; this mass is affected by humidity; the size of aerosol particles is typically in the micron or sub-micron range, and as a natural consequence, aerosol particles can interact with visible light; finally and most importantly, the mass and, in general, *all properties* of the aerosol particles are a function of their size.

Contrary to gases, the aerosol can be filtered out from a sampled air flow, and the filter weighed in a microbalance. Aerosol concentrations are then usually expressed in microgram per cubic metre and can vary from around 1 $\mu g\ m^{-3}$ in very pristine environments and free troposphere air to 5–10 $\mu g\ m^{-3}$ in the clean continental boundary layer, up to 50–100 $\mu g\ m^{-3}$ of dry PM_{10} mass in polluted areas (Van Dingenen et al. 2004). After collection on filters or other sampling substrates, the aerosol samples can also be subjected to a suite of chemical analyses in the same manner of other environmental matrices like soil or water samples, and the resulting concentrations of aerosol chemical components are also expressed in microgram per cubic metre.

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Ambient aerosol particles contain also a significant but variable amount of condensed water, which depends on relative humidity and on the occurrence of hygroscopic materials in the aerosol particles. Condensed water is not included in the PM_{10} or $PM_{2.5}$ mass per definition, and it is believed to be irrelevant for the toxicological properties of aerosol, but it is important to remember that condensed water can make up several tenths of microgram per cubic metre in a humid atmosphere. Condensation of water makes the particles grow to larger sizes, allowing them to scatter more radiation, thus affecting visibility and the transparency of the atmosphere with respect to sunlight. Moreover, humid particles form more efficiently hazes and fogs and modify cloudiness, which has also very important climatic effects (Ramanathan et al., 2001; Kaufman and Koren, 2006).

The interaction of aerosol particles with visible light not only has a great importance for visibility and the atmospheric radiative budget, but also provides tools for determining the aerosol concentration using in situ or remote sensing techniques (e.g. lidars, photometers). A comprehensive and elegant theory of light scattering and absorption by aerosols has been developed by Mie, and it is the subject of several treatises (e.g. Bohren and Huffman, 1983). The aerosol scattering coefficient is measured in situ by common instrumentation, called *nephelometers*, and expressed in mm^{-1}, or km^{-1}. The aerosol scattering coefficient is primarily a function of aerosol size and also of relative humidity, which affects the size of aerosol particles by regulating the amount of condensed water. The scattering coefficient of a heated, dried aerosol is a proxy of total aerosol surface and a broad function of aerosol volume, and measuring it is a simple, cheap and useful way to monitor the total concentration of aerosols with diameter larger than 0.2–0.3 μm. The scattering coefficient has only a weak dependence on the chemical composition. By contrast, the absorption coefficient, which is determined by *aethalometers* and other instruments capable to measure the colour or the darkness of an aerosol sample, is completely dependent on the occurrence of chemical compounds, such as mineral oxides or the so-called black carbon (BC). Other major aerosol compounds, like ammonium sulphates and nitrates and many organic compounds, do not absorb visible light; therefore, the aerosol absorption coefficient is usually weakly correlated with the aerosol mass concentration.

The light scattering properties of aerosol particles allow to *count* them too. *Optical particle counters* (OPCs) are widespread instruments where an air stream is sampled at a reduced flow (≤ 1 L min^{-1}) and individual particles are counted by making them scatter a focused light beam. These systems are unsuitable for particles with diameters smaller than 0.1–0.3 μm. More sophisticated instruments, called *condensation particle counters* (CPCs), are able to count also the small particles with diameters down to a few nanometres, using a pre-treatment of the particles in a chamber filled with a supersaturated vapour (usually butanol), where they undergo a condensational growth to larger sizes until they can be counted using optical methods. Aerosol number concentrations span from few tenths per cubic centimetre in very remote environments to some hundreds in relatively pristine environments, to 10^3 cm^{-3} in polluted areas, up to 10^4 cm^{-3} at kerbside sites (Van Dingenen et al., 2004). Despite this general trend towards higher number concentrations with increasing pollution levels, very high number concentrations have been observed sporadically also in the clean atmosphere and related to bursts of freshly formed ultrafine particles (Kulmala et al., 2004). Since the total number of aerosol particles is often dominated by ultrafine particles which, on the other hand, contribute to a very small extent to total particle mass, it is important to keep in mind that aerosol *number* concentrations are not necessarily correlated with *mass* (PM_{10} or $PM_{2.5}$) concentrations. Bursts of ultrafine particles recorded by a CPC will leave only nanogram levels of material on a filter for daily PM_{10} determination. Therefore, the phenomenology of aerosol, with respect to spatial and time variability, can be very different by looking at number concentrations rather than to mass concentrations. The same applies to the phenomenology of aerosol chemical compounds. For instance, mineral dust can make up a substantial fraction of PM_{10}, but it usually accounts for only a minor fraction of the number of particles which, on the contrary, is mainly apportioned into particle types having a smaller modal diameter (like carbonaceous and ammonium sulphate particles). In summary, number-related and mass-related aerosol properties may follow very different dynamics, and this is a consequence of the large breadth and multimodality of the size spectrum of atmospheric aerosol particles. Atmospheric aerosol is an intrinsically heterogeneous system characterized by a continuum size spectrum from 10^{-9} to 10^{-5} m and by varying particle chemical compositions, *mixing state* (i.e. the degree of mixing of the chemical species in the same particles rather than belonging to distinct particle populations), physical properties (density, shape, optical properties) and *internal* structures of the individual particles for an infinite number of degrees of freedom which are simply irreducible by the existing instrumentation. Therefore, the observations of atmospheric aerosols can be much more complex than those of atmospheric gaseous compounds such as CO_2, CO or O_3: the aerosol measurements provide an inherently partial information, and the most appropriate instrumentation must be chosen according to the goals of the specific experiment.

Ground-based observing systems for atmospheric aerosols can be classified into systems measuring the properties and concentration of the aerosol in situ and those looking at columnar properties and vertical profiles using *remote sensing* techniques. Instruments for in situ measurements exploit all principles of aerosol detection, while remote sensing techniques rely on the optical methods alone; therefore, the characterization that can be achieved in situ is much more sophisticated than that retrieved by remote sensing. In particular, remote sensing methods suffer from two major limitations, which are inherent to all optical measurements of the aerosols: (a) they are not sensitive to ultrafine particles and (b) they provide only an indirect information about chemical composition.

A treatment of lidar systems for aerosol observations is given elsewhere in this issue (Chap. I.3). Here, we deal with measurements in situ, with a focus on the techniques of determination of the aerosol chemical composition.

2 Measurements of Aerosol Concentration and Chemical Composition

The determination of the aerosol concentration, size distribution, composition and optical properties can be performed in situ by a large set of instruments that we summarize schematically and in a very simplified form in Figs. II.3.1, II.3.2, and II.3.3. For a more detailed treatment, the reader is referred to the books by Finlayson-Pitts and Pitts (2000) and Heard (2006).

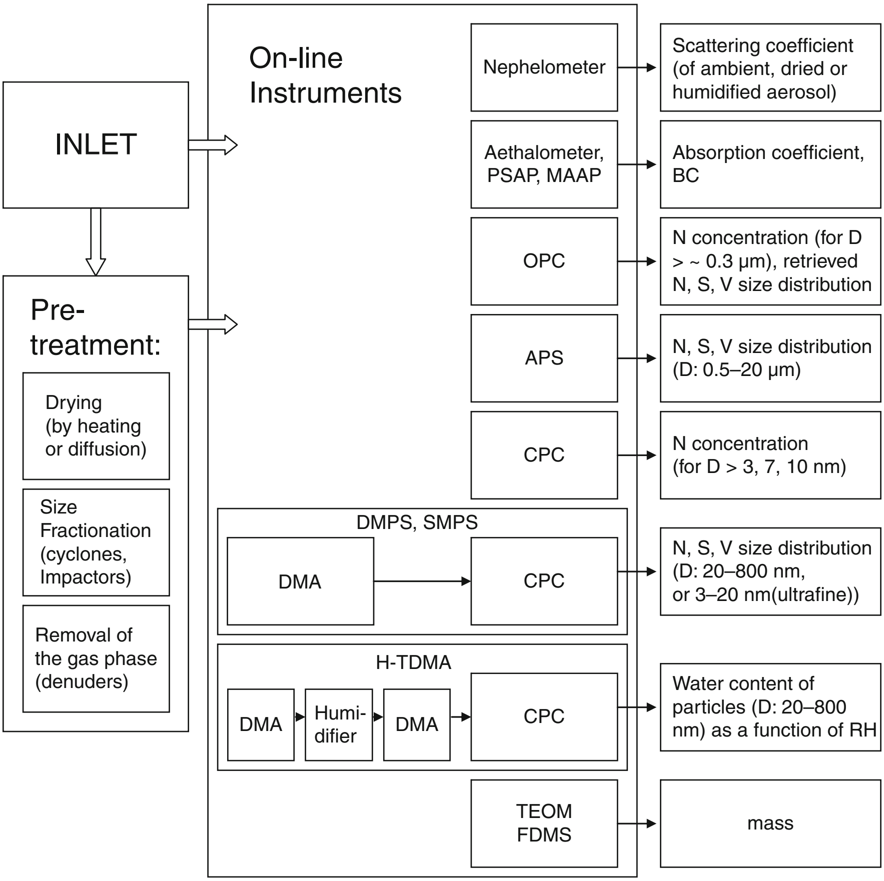

Fig. II.3.1 Online measurements of aerosol concentration and *N* (number), *S* (surface) and *V* (volume) size distributions. APS, aerodynamic particle sizer; CPC, condensation particle counter; DMA, differential mobility analyzer; DMPS, differential mobility particle sizer; FDMS, filter dynamic measurement system; H-TMDA, hygroscopicity tandem differential mobility particle sizer; MAAP, multi-angle absorption photometer; OPC, optical particle counter; PSAP, particle soot absorption photometer; SMPS, scanning mobility particle sizer; TEOM: tapered element oscillating microbalance

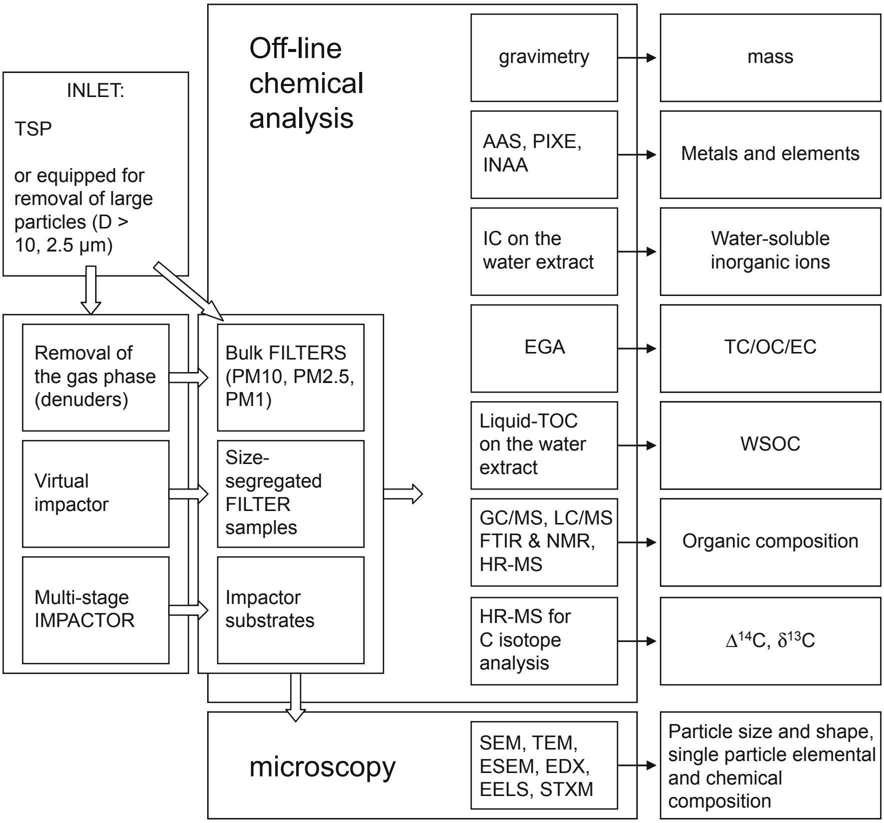

Fig. II.3.2 Off-line chemical, gravimetric determination and microscopic characterization of total suspended particulate (TSP) matter samples and size-segregated aerosol samples. TC, OC, EC and WSOC stand for total, organic, elemental and water-soluble organic carbon. AAS, atomic absorption spectroscopy; EDX, energy-dispersive X-ray spectrometry, EGA, evolved gas analysis; EELS, electron energy loss spectroscopy; ESEM, environmental scanning electron microscopy; FTIR, Fourier transform infrared spectroscopy; GC/MS, gas chromatography/mass spectrometry; HR-MS, high-resolution mass spectrometry; IC: ion chromatography; INAA, instrumental neutron activation analysis; LC/MS: liquid chromatography/mass spectrometry; NMR, nuclear magnetic resonance spectroscopy; PIXE, particle-induced X-ray emission; SEM, scanning electron microscopy, STXM, scanning transmission X-ray microscopy; TEM, transmission electron microscopy; TOC: total organic carbon

All instruments have *inlets* and sampling lines to bring the air sample to the detector/collector. Since aerosol particles have an inertia and tend to deviate from the streamlines, the sampling efficiency can be reduced for particles with a large aerodynamic diameter. Therefore, the inlets can be suitable for sampling the total suspended particulate (TSP) matter or, alternatively, only the aerosol particles smaller than a given size depending on the flow rate and on the geometry of the inlet and of the sampling line. Commercially available inlets with a calibrated geometry

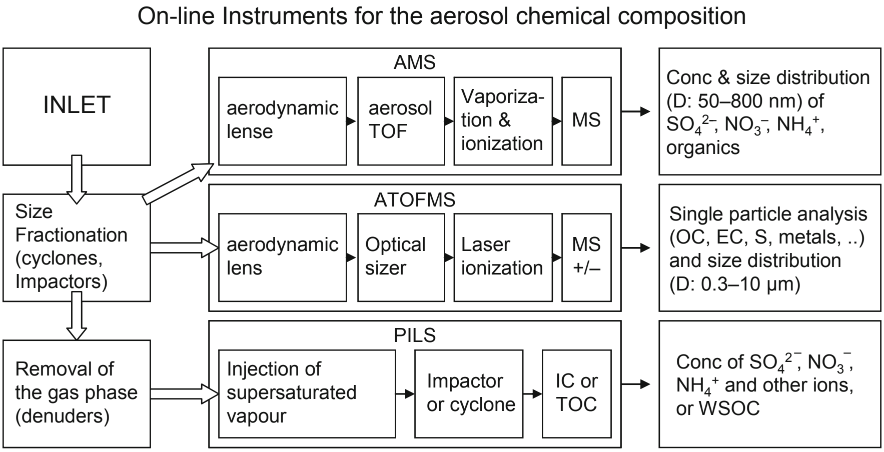

Fig. II.3.3 Online instruments for the aerosol chemical composition. AMS, aerosol mass spectrometer; ATOFMS, aerosol time-of-flight mass spectrometer; IC, ion chromatography; PILS, particle-into-liquid sampler; TOC, total organic carbon analysis; TOF, time-of-flight; WSOC, water-soluble organic carbon

(sampling heads) allow selective preseparation of PM_{10} and $PM_{2.5}$ for standard flow conditions. Inertial fractionation of the particles can be achieved also using cyclones and impactors, which allow the isolation of particles with aerodynamic diameter smaller than 1 μm, down to 0.1 μm in the case of impactors. Once drawn into the instrument, the air sample may undergo further treatments to eliminate possible interference with aerosol detection/collection. For instance, humidity can be removed by heating the sample or by employing diffusion driers, and gas-phase compounds can be eliminated using absorbing plates or cylinders (denuders).

The detection, observation and characterization of aerosol particles exploit several measurement principles already mentioned in Sect. 1 and that can be summarized as follows:

(1) aerosol have a weighable mass
(2) they scatter and absorb visible light
(3) they absorb water vapour (and organic vapours)
(4) after sampling they can be subjected to chemical, biological and mineralogical analysis

Moreover:

(5) they can be ionized in a mass spectrometer
(6) they can be observed using an optical or electronic microscope

There are other principles of detection, like attenuation of β rays, but these will not be treated in this review.

Detection can be carried out *online* with the sampling or *off-line* after the end of the sampling.

Online instruments employ more sensitive techniques allowing continuous or quasi-continuous measurements of the aerosol particles, which, in most of the cases, are analysed directly in their gaseous medium. Conversely, in the off-line methods, time-integrated samplings are performed using filters and other substrates to separate the aerosol particles from the air stream, and the sampled substrates are then transferred to a laboratory for subsequent analysis.

Online systems have been first developed for measuring the aerosol scattering and absorption coefficients, the aerosol number concentrations and the aerosol number size distribution (Fig. II.3.1). All these instruments exploit the sensitivity of the optical detection. OPC can count particles and estimate the size distributions of accumulation mode and coarse particles. More accurate sizing can be achieved exploiting aerodynamic forces (using APSs, aerodynamic particle sizers) or, for smaller particles, their electrical mobility in a magnetic field (with DMAs, differential mobility analyzers, Flagan 1998). The differential mobility particle sizer (DMPS) is DMA coupled to a CPC, and it is by far the most common and accurate instrument for the determination of the number size distributions of ultrafine, Aitken and accumulation mode aerosol particles. DMA and CPC were revolutionary instruments in the aerosol science and they have been used in smart arrangements to characterize several properties of aerosol particles. For instance, a tandem DMA assemble called H-TDMA (hygroscopicity tandem DMA) is used to measure the particle growth from a dry to a humidified atmosphere, hence allowing determination of the aerosol water content as a function of relative humidity.

Measuring particle mass online is a challenging task, because of the very small mass that can be accumulated with a high-frequency sampling. The microbalances equipped with an oscillating tapered element on which the aerosol is continuously deposited are the only available technical solutions for measuring the aerosol mass with a resolution higher than 1 min^{-1} (Patashnick and Rupprecht, 1991). The original products called TEOMs are not widespread because they need to dry the sample heating it at 50°C causing substantial mass losses by evaporation. However, the new version equipped with a FDMS (filter dynamic measurement system) can account for the semivolatile material and it is a promising solution.

In the off-line methods, aerosol particles are collected on a filter for subsequent gravimetry, chemical analysis or for observation using an electron microscope (Fig. II.3.2). For instance, for air quality monitoring purposes, filters are collected typically on a daily basis for gravimetric determination of PM_{10} or $PM_{2.5}$. Routine measurements employ fully automated sampling equipments, called sequential samplers, which can be left unattended for days. The chemical methods that have been used to characterize atmospheric aerosol samples involve a large diversity of techniques that cannot be reviewed comprehensively in this chapter. However, the techniques that allowed the higher recovery, in terms of aerosol mass speciated, are, for $PM_{2.5}$, ion chromatography (IC) and evolved gas analysis (EGA), which determine the water-soluble ionic composition and the carbon concentration, respectively. Atomic absorption spectroscopy (AAS), particle-induced X-ray emission (PIXE) and other techniques for the quantification of aluminium, silica and metals can account for a non-negligible fraction of $PM_{2.5}$ and a major fraction of PM_{10}. In principle, the mass apportioned into chemical species should approach the aerosol mass obtained by gravimetric determination. However, the closure of the aerosol mass budget is a challenging exercise, because of the uncertainty associated with the amount of oxygen and water contained in the organic matter and in the mineral fraction (Putaud et al., 2000). This problem is more severe for the organic matter which can exhibit a very variable stoichiometry and a very complex molecular composition. Scholars speculate that an aerosol sample can contain 10,000 to more than 100,000 individual organic compounds, each occurring in very low concentrations. On the other hand, state-of-the-art high-resolution mass spectrometric techniques are able to determine the molecular formulas of up to 3,000 compounds (Wozniak et al. 2008). Such techniques are not designed for routine analyses and for samples collected in remote areas where the sampled mass is low and sensitivity of the analytical techniques is critical. Overall, our understanding of the chemical composition of the aerosol organic carbon is still unsatisfactory for most environments, although a lot of new information has been gained in the last 10 years (Fuzzi et al., 2006).

The determination of the aerosol mass size distribution and the size-resolved chemical composition has been attempted using *cascade impactors* (Hering et al., 1978), which allow the collection of aerosol particles in different size intervals on different collection plates, which are then weighed and analysed at the same manner of PM_{10} and $PM_{2.5}$ filters. The number of the stages of cascade impactors varies between 5 and 15 for the size range 0.05–20 μm, hence the size resolution is coarse if compared to that of volume size distributions obtained by DMPS. The number of impactor stages is limited not only by the efficiency of the inertial separation (which is lower compared to that based on the electrical mobility) but also by the need of ensuring enough aerosol mass to be deposited on each impactor plate for subsequent weighing and analysis. Another limiting factor is the chemical analysis, which can be very labour intensive and time expensive. As a result, multistage impactors are not used for monitoring purposes and a very few climatologies of the aerosol composition based on impactor measurements are available in the literature (Cabada et al., 2004). On the contrary, cascade impactors have been widely used in field campaigns (e.g. Cavalli et al., 2006; Herner et al., 2006). An example of a time series of impactor measurements of the aerosol mass distribution taken in parallel with online measurements of PM_{10} and $PM_{2.5}$ from TEOM is given in Fig. II.3.4.

The off-line (i.e. filter-based and impactor-based) techniques allow a more sophisticated chemical characterization than online techniques, but sensitivity, and consequently time resolution, is much lower and it is further reduced by the interference of the substrate matrix with the analysis (the "blank"). Another problem of the off-line techniques is that the time-integrated sampling over a filter or another collection substrates can cause modifications of the aerosol mass and composition, which are usually referred to as *sampling artefacts*. Examples of sampling artefacts are the collection of vapours by the matrice of the filter or the evaporation of semivolatile constituents of the aerosol. There are several technical solutions for

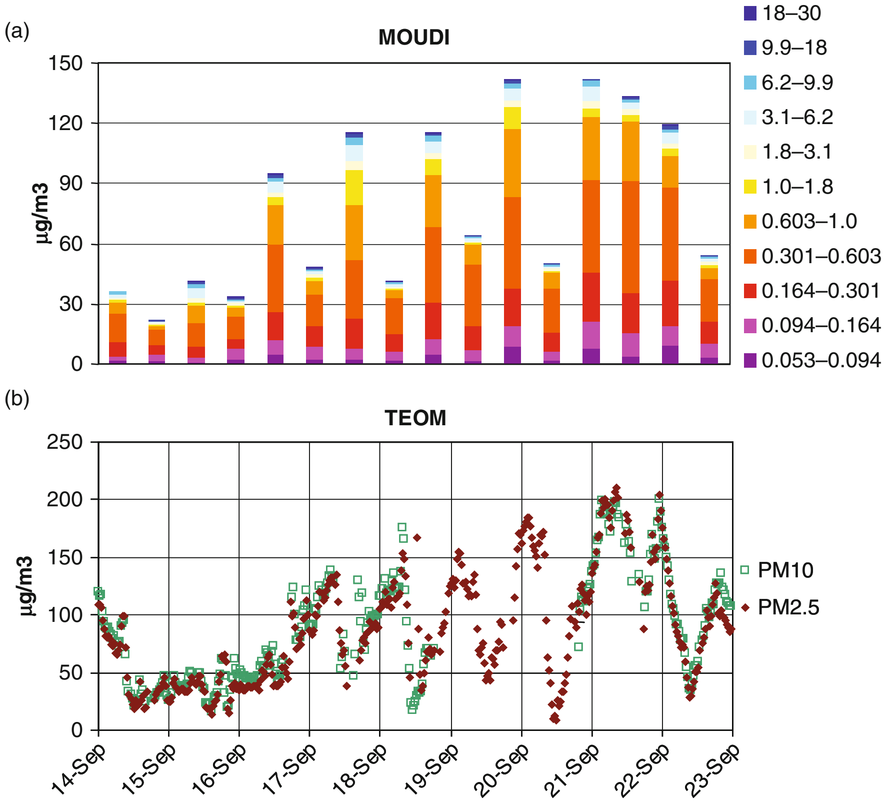

Fig. II.3.4 Measurement of aerosol mass **a** off-line with a cascade impactor (MOUDI: micro-orifice uniform deposition impactor) and **b** online with a tapered element oscillating microbalance (TEOM). MOUDI provides the size-segregated mass in 11 size ranges with 12-h time resolution. The upper and lower size limits (diameters in micrometers) are given in the legend on the right. Two TEOM systems were used equipped with PM_{10} and $PM_{2.5}$ inlets. Time resolution of TEOM measurements is here 30 min

limiting the sampling artefacts but they are not universal and must be evaluated specifically for each experiment.

In order to overcome these major limitations of the off-line chemical analyses, new instruments have been designed without any filter or other sampling substrates and the chemical characterization is performed online (Fig. II.3.3). The particle-into-liquid sampler (PILS) allows the measurement of water-soluble ions and carbon with a time resolution of 5–10 min (Orsini et al., 2003). The time resolution is even higher for the techniques employing mass spectrometry for detection (Heard 2006). The aerosol mass spectrometer (AMS) from Aerodyne is able to measure the concentration of major non-refractory aerosol chemical compounds and provide information on the organic chemical composition and it is also able to measure the size distribution of selected compounds (Canagaratna et al., 2007).

The aerosol–TOF–mass spectrometer (ATOFMS) from TSI provides a more qualitative information on the chemical composition but it is sensitive also to refractory compounds, and it provides the composition of individual particles (Murphy 2007). These instruments represented a real breakthrough for the aerosol chemical measurements. Still, the information that can be gained by their use is less sophisticated than that provided by the off-line methods, especially in respect to the organic molecular composition. At present, PILS and AMS are widely used in the aerosol characterization experiments and are able to provide unsurpassed information about the aerosol processes at short timescales, but they have not been designed for monitoring purposes. By contrast, the traditional samplers employing filters are robust, weather resistant and relatively cheap and are still the best solution for monitoring networks and for routine sampling in remote places.

3 Strategies for Ground-Based Aerosol Observations

We can conclude from the above discussion that an ideal instrument or a simple set of instruments for measuring all relevant properties of the atmospheric aerosol do not exist and appropriate solutions must be identified for the specific applications.

So far, two general strategies have been pursued for implementing ground-based observation systems for atmospheric aerosols (Heard et al., 2006): (1) continuous measurements of the aerosol concentrations from networks of stations covering different spatial scales and (2) multiplatform-intensive observation periods at the regional scale (field campaigns).

In the case of monitoring networks, only the more cost-effective, automated and robust aerosol instruments are employed. For instance, for air quality monitoring in the urban environment, normally only the determination of the daily mass concentrations of PM_{10} and, less commonly, $PM_{2.5}$ is performed. In Europe, routine analysis of the aerosol chemical composition is performed by the EMEP (European Monitoring and Evaluation Programme) network, which, however, has a rather patchy spatial coverage. Another network of stations, equipped with instruments for measuring both the aerosol optical properties, the number size distribution and the chemical composition has just become operative in the frame of the project EUSAAR (European Supersites for Atmospheric Aerosol Research, http://www.eusaar.net/). The monitoring networks for the chemical composition are more diffused in the USA, with the IMPROVE (Interagency Monitoring of Protected Visual Environments, http://vista.cira.colostate.edu/improve/) and the STN (EPA Speciation Trends Network).

In remote locations, also the maintenance needs and power consumption can be limiting factors. Fully automated, standalone, weather-resistant instruments, powered by solar cells and transmitting the data via satellite connection, like the photometers of the AERONET network, are definitely ideal instruments to be used for monitoring purposes. Analogous instruments for the in situ measurement of the aerosol concentration and composition have not been developed yet.

Consequently, such measurements rely on the existence of permanent stations, which can guarantee power and maintenance to the aerosol instruments even in remote locations (Bonasoni et al., 2008; Jourdain et al., 2008).

During multiplatform regional experiments, aerosol measurements are taken simultaneously in situ, using mobile platforms (ships, aircrafts), and by satellite remote sensing, over a spatial scale of 500–1,000 km for ca. 1 month. During these experiments, the state-of-the-art instrumentation is deployed, trying to achieve a comprehensive characterization of the aerosol concentration, composition and distribution, attempting to expand our knowledge about aerosol processes and impacts. The regional experiments have been conducted in several areas of the world but always for limited periods of time and collecting more data in clear-sky conditions. Comparison of the results obtained in the different regions (often after many years between one experiment and the other) is affected by the technological development of the instruments and by the changes in the emissions. Observations were more concentrated in polluted marine coastal areas, where the satellite measurements are more accurate (Quinn and Bates, 2005). The regional experiments have taught us a lot about how aerosols form and are transported and processed in the atmosphere and about their impacts on light extinction and on cloud properties (Quinn et al., 2005; Guo et al., 2007). However, such load of data are biased geographically and temporally, therefore, care must be taken when extrapolating these results to unexplored environments and weather regimes.

The regional studies have shown that the intrinsic limitations of the various aerosol instruments can be overcome by collocating measurements from different observing systems, including ground-based in situ and remote sensing systems, research aircrafts and the satellite sensors. Still, some important issues remain unsolved, e.g. assessing the spatial distribution and chemical composition of ultrafine particles, quantifying the biogenic and anthropogenic sources of aerosol organic matter and understanding the effects of aerosol on precipitating clouds at the global scale.

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