



## Review

# Sampling and single particle analysis for the chemical characterisation of fine atmospheric particulates: A review



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## ABSTRACT

To better understand the potential environmental and human health impacts of fine airborne particulate matter (APM), detailed physical and chemical characterisation is required. The only means to accurately distinguish between the multiple compositions in APM is by single particle analysis. A variety of methods and instruments are available, which range from filter-based sample collection for off-line laboratory analysis to on-line instruments that detect the airborne particles and generate size distribution and chemical data in real time.

There are many reasons for sampling particulates in the ambient atmosphere and as a consequence, different measurement strategies and sampling devices are used depending on the scientific objectives and subsequent analytical techniques. This review is designed as a guide to some of the techniques available for the sampling and subsequent chemical analysis of individual inorganic particles.

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## Contents

1. Introduction .....	137
2. Air sampling techniques .....	139
2.1. Active samplers .....	139
2.2. Passive samplers .....	141
2.3. Sampling substrates .....	141
3. Off-line analytical techniques .....	144
3.1. Electron microscopy .....	144
3.2. Atomic spectroscopy .....	145
4. On-line analytical techniques .....	146
5. Conclusion .....	147
Acknowledgements .....	147
Abbreviations .....	147
References .....	147

## 1. Introduction

Compared to trace gases, airborne particulate matter (APM) is a complex mixture of solid and liquid particles of organic, inorganic

and biological substances. Some particulates occur naturally but human activities, such as traffic and industrial emissions, also contribute significant amounts of particulates. The assessment of the potential environmental and human health risks associated with fine APM (>1 μm) requires detailed physical and chemical characterisation. In the past, analysis has concentrated on determination of size, mass concentration and bulk chemistry. Individual particles however, vary in properties such as toxicity, light

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attenuation and hygroscopic behaviour which are functions of their three-dimensional chemical composition. The impact and toxicity of APM is related not only to total elemental composition and size distribution but also to their chemical heterogeneity. Information regarding chemical heterogeneity at the individual particle level (the mixing state) is essential for understanding and predicting the reactivity and environmental and human health impacts of APM. The only means to accurately distinguish between the multiple compositions in APM is by single particle analysis.

Once in the atmosphere, and under favourable weather conditions, particulates can be transported over long distances by prevailing winds and can act as a vector for pollution. As pollutants are commonly taken up on the particle surface, they are typically present in loosely bound forms that are highly mobile and potentially bio-available. The health effects of exposure to APM are well documented (Møller, 2008; Pope et al., 2004; Zanobetti and Schwartz, 2009) and as a result several guidelines have been adopted (Table 1). These guidelines, though not legally binding in most countries, provide a basis for setting standards and limiting airborne particulate pollution. The most frequently used reference guidelines for ambient particulate concentration are the World Health Organisation Air Quality Guidelines (WHO, 2000), the European Union Limit Values for Air Quality (European Union, 2008) and the United States Environmental Protection Agency National Ambient Air Quality Standard (Environmental Protection Agency (EPA), 1997). Most guidelines are measured in  $\mu\text{g}/\text{m}^3$  and averaged over a 24 h time period, however the United Kingdom and European Union standards are averaged over a year. These

**Table 1**

International Air Quality Standards for total suspended particulates (TSP),  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  ( $\mu\text{g}/\text{m}^3$ , 24 h mean unless otherwise stated). (a) (Defra, 2012); (b) (National Environment Protection Council, 1998); (c) (National Environmental Standards for Air Quality, 2004); (d) (SANS, 1929, 2011); (e) (Clean Air Initiative for Asian Cities, 2010); (f) (Clean Air Institute, 2012). WHO AQG – World Health Organisation Air Quality Guidelines; NAAQS – National Ambient Air Quality Standard; EU LVAQ – European Union Limit Values for Air Quality.

	TSP	$\text{PM}_{10}$	$\text{PM}_{2.5}$
WHO AQG	–	50	25
NAAQS (United States)	–	50	35
EU LVAQ (Europe)	–	50	25 (annual mean)
United Kingdom (a)	–	50	25 (annual mean)
Australia (b)	–	50	25
New Zealand (c)	–	50	–
South Africa (d)	–	120	65
China (e)	300	150	–
Hong Kong (e)	–	100	75
India (e)	–	100	60
Japan (e)	–	100	35
Bangladesh (e)	–	150	65
Bhutan (e)	200	100	–
Indonesia (e)	230	150	–
Malaysia (e)	260	150	–
South Korea (e)	–	100	50
Mongolia (e)	150	150	50
Nepal (e)	230	120	–
Singapore (e)	–	150	35
Pakistan (e)	500	150	35
Philippines (e)	230	150	–
Sri Lanka (e)	–	150	50
Thailand (e)	330	120	–
Vietnam (e)	200	150	–
Bolivia (f)	–	150	–
Brazil (f)	–	150	–
Colombia (f)	–	100	50
Chile (f)	–	150	50
Ecuador (f)	–	150	65
Mexico(f)	–	120	65
Peru (f)	–	150	50
Puerto Rico(f)	–	150	35

guidelines are based on clinical, toxicological, and epidemiological evidence and were established by determining the concentrations with the lowest observed adverse effect, however, to date there is no evidence to support a threshold level below which no adverse health effects occur (Kim et al., 2015). Standards have also been implemented for other air toxins such as lead, cadmium, arsenic and mercury, however these are outside the scope of this review.

Atmospheric residence time, deposition rates, and inhalation processes are predominantly influenced by the size of the particles. Even though many particles are not spherical, they are typically classified to size by their aerodynamic diameter which is defined as the diameter of a spherical particle of density  $1 \text{ g}/\text{cm}^3$  having a settling velocity equal to that of the particle in question (John, 2011). The aerodynamic diameter is useful for particles larger than  $0.5 \mu\text{m}$  and is considered to be the most appropriate measure to describe particle motion in the atmosphere (Sullivan and Prather, 2005) and the ability of the particle to penetrate and deposit at different sites within the respiratory tract (Pinkerton, 2000). The aerodynamic properties of particles also depends on density and shape.

The health risks associated with APM arises from the deposition of particles in the human respiratory system (Fig. 1). After inhalation, particles in the  $2.5\text{--}10 \mu\text{m}$  size fraction (thoracic particles) are primarily deposited in the tracheal and bronchial region, from where they are transported by mucociliary processes and typically swallowed, thus reaching the gastrointestinal tract. Finer particles can travel deeper into the alveolar region (respirable particles) where they interact with lung fluids (Asgharian et al., 2001). Ultrafine particles ( $<0.1 \mu\text{m}$ ) can not only deposit in the respiratory tract, they can traverse the alveolar epithelium to be absorbed directly into the bloodstream. Their large specific surface area, with its increased surface reactivity, has the potential to result in greater toxicity (Oberdörster et al., 2005). These ultrafine particles not only have an enhanced inflammatory potential, they also have a higher deposition efficiency within the pulmonary system. There has been an increasing awareness of the impacts of these ultrafine particles, however methods for characterizing these particles is outside the scope of this review.

The site of particle deposition within the respiratory system strongly influences the health effects of exposure to these particles and as a result, regulation and monitoring of APM has evolved over time from total concentrations (total suspended particulates, TSP) to a focus on smaller inhalable particles that can be deposited into the respiratory system, namely fine ( $\text{PM}_{2.5}$ ) and coarse ( $\text{PM}_{10}$ ) particles, which are defined as particles with an aerodynamic diameter of less than  $2.5 \mu\text{m}$  and  $10 \mu\text{m}$  respectively.

While methods for measuring particle concentrations and size distribution are well established, the compositional analysis of single particles remains problematic. A variety of methods and instruments are available, which range from filter-based sample collection for off-line laboratory analysis to on-line instruments that detect the airborne particles and generate size distribution and chemical data in real time, however, a single practical technique does not exist for obtaining all the required information, specifically the size, morphology, composition and molecular structure of fine particulate matter (Pratt and Prather, 2012a). The ultimate goal of analytical techniques developed for APM is to quantitatively identify all species within each individual particle but as single particles are complex mixtures containing in the order of  $\sim 10^2\text{--}10^{15}$  molecules per particle, which translates to masses in the order of  $\sim 10^{-20}$  to  $10^{-6} \text{ g}/\text{particle}$ , measurement can be challenging (Pratt and Prather, 2012a).

Off-line techniques generally allow for greater molecular and structural speciation than on-line techniques however, on-line techniques are able to examine the chemical changes in APM on

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