



Review

Selective Collection of Airborne Particulate Matter[†]

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Airborne particulate matter (PM) or aerosol particles or simply aerosol are ubiquitous in the environment. They originate from natural processes such as wind erosion, road dust, forest fire, ocean spray and volcanic eruption, and man-made sources consuming fossil fuels resulting from utility power generation and transportation, and numerous industrial processes. Aerosols affect our daily life in many ways; PM reduces visibility in many polluted metropolitan areas, adversely impact human health and local air quality around the world (Tang *et al.*, 1981; Mage *et al.*, 1996; Molina and Molina, 2004; Davidson *et al.*, 2005; Baklanov *et al.*, 2016). Aerosol alters cloud cycles and change atmospheric radiation balance (Seinfeld and Pandis, 2000; Lohmann and Feichter, 2005; Flossmann and Wobrock, 2010; Rosenfeld *et al.*, 2014; Seinfeld *et al.*, 2016). Changes in daily mortality associated with particulate air pollution were typically estimated at approximately 0.5–1.5% per 10 $\mu\text{g m}^{-3}$ increase in PM₁₀ concentrations (Pope, 2000). Laden *et al.* (2006) found “an increase in overall mortality associated with each 10 $\mu\text{g m}^{-3}$ increase in PM_{2.5}

concentration either as the overall mean (rate ratio [RR], 1.16; 95% confidence interval [CI], 1.07–1.26) or as exposure in the year of death (RR, 1.14; 95% CI, 1.06–1.22). PM_{2.5} exposure was associated with lung cancer (RR, 1.27; 95% CI, 0.96–1.69) and cardiovascular deaths (RR, 1.28; 95% CI, 1.13–1.44). Improved overall mortality was associated with decreased mean PM_{2.5} (10 $\mu\text{g m}^{-3}$) between periods (RR, 0.73; 95% CI, 0.57–0.95)”. Aerosol particles also play an important role in source identification and apportionment (e.g., Hopke, 2016; Varga *et al.*, 2017). Since the PM problem is associated with many facets of societal issues such as energy production and economic development, making progress on reducing the effects of PM will require integrated strategies that bring together scientists, engineers and decision makers from different disciplines to consider tradeoffs.

PM measurement and monitoring plays the central role in the efforts of developing and designing the integrated strategies. Conventional PM monitoring devices discussed in depth in Kulkarni *et al.* (2011) include various designs based on optical, electrical, or aerodynamic principles to detect and or harvest aerosol particles. Optical sensor (e.g., optical counter) and electrical instrument (e.g., aerosol electrometer) can provide real-time readings of particle concentration as a total count or a distribution of particle size. To determine the chemical and radiological makeup of aerosol particles, the direct and possibly most cost-effective way is to collect or harvest the particles by a single-stage filter or a cascade impactor, and the sample substrate then chemically and or radiologically analyzed using advanced instruments in a laboratory.

Recently, there have been significant advances of aerosol chemical measurement such as the Aerosol Mass Spectrometer (AMS) developed and commercialized by Aerodyne, Inc. (Takegawa *et al.*, 2005), Single Particle Aerosol Mass Spectrometer (SPAMS) developed by Livermore Instrument, Inc. (<http://www.livermoreinstruments.com/spams-overview.html>), and various research-grade single particle mass spectrometers (e.g., Sodeman *et al.*, 2005; Murphy, 2007; Brands *et al.*, 2011; Zelenyuk *et al.*, 2015) around the world. All of these instruments have advantages and disadvantages in either the range of chemical species or the size of particles an instrument can detect. There is no single instrument that can be applied to meet all aerosol measurement needs, and one should not expect such, either.

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The most popular practice of aerosol collection is by filtration with a designed particle size inlet. For example, a PM_{2.5}/PM₁₀ sampler is equipped with a virtual dichotomous inlet to divide particles into two aerodynamic diameter size fractions (those smaller than 2.5 μm and those between 2.5 and 10 μm). A high-volume total suspended particulate (TSP) sampler commonly used by the EPA RadNet monitoring program (<https://www.epa.gov/radnet>) collects every particle suspended in the air. A multiple-stage cascade impactor is also commonly used to harvest airborne PM for determination of aerosol mass and chemical/radiological composition associated with the PM.

These particulate collectors are efficient in terms of accumulating large quantity of particulate material for a given sampling time, but they are not smart. There is no mechanism built in these collectors for identification or confirmation what the particulate material might have been collected at the point of collection. This may be acceptable for many applications that require only measurement of particulate mass, for instance, but insufficient for others that need to further determine trace amount of the chemical and or physical makeup of the aerosol particles. As a result, extensive efforts are devoted to preparing (or “cleaning”) the sample after it is collected such that the analytes can be successfully analyzed. Enormous amount of resources is wasted, if the search for analyte signal this way turns out to be negative. Erroneous information collected this way provides meaningless or useless data to decision making.

The issue with using current samplers for trace analysis is primarily in the signal-to-noise ratio embedded in the PM. In other words, how much analytically observable material (e.g., mass or analyte concentration) actually exists in the filter to enable a quantitatively meaningful identification and measurement of a specific atom, molecule, or compound. Mostly likely, the background information will overwhelm the actual signal in a sample that had accumulated large number or mass of particles. Furthermore, a short sampling time maybe encountered in many applications, which would leave no alternative for harvesting species in the particulate material of interest by using current sampling methods described, previously.

Smart sampling is to collect relevant or essential materials during sampling either at the right time or at the right place or both. Thereby, smart sampling could potentially enhance signal collection by orders of magnitude. Only when a sampler can collect the target analytes, selectively (in time, space, and or by analyte), there is a hope that the downstream effort to prepare for environmental monitoring or analysis can be meaningfully minimized and the signal quality can be substantially improved.

Recently, there have been several interesting developments in smart environmental sampling. These developments represent the current trends and concepts in the design and research for the next-generation environmental samplers. This FOCUS paper will highlight these developments and use them to entice future development of smart environmental samplers.

SELECTIVE COLLECTION BY MAGNETIC PROPERTY OF AEROSOL

Separation and purification is a rational approach that has been employed for many decades since the beginning of modern analytical chemistry to directly enhance analyte signal. Aerosol particles have unique material properties that can be exploited for selective collection; for instance, thermal property (as in volatility), magnetic susceptibility, electrical conductivity, ionic property, and so on. Here we use the magnetic property as an example to illustrate our idea of selective collection. Environmental particulate matter consists of many magnetic materials. Materials like Fe, Ni, and Co and their oxide forms are ferromagnetic, while others like a large number of elements in the periodic table are paramagnetic meaning that they can become magnetically separable given sufficient magnetic field is present (Coe, 2010). If elements of interest are magnetically amenable to collection, use of magnetic separation upfront to select the aerosol particles before collection is a warranted approach for conditioning signal and actually a smart sampling approach.

Thus, use of a magnetically assisted collector for particulate sampling represents a conscious effort in the collection of application-relevant materials (Hunt, 1986). The magnetic collector would collect only particles with magnetic signature and leave out those that do not; thus, its function of simultaneous collect signal and screen or separate the noise from an aerosol mixture represents the basic requirement a next-generation collector needs to have. Again, this selective collection could save significant amount of resources devoted to otherwise required subsequent cleaning of samples. The concept of high-gradient permanent magnetic sampling (HGPMS) developed in 2014 and the following three years represents an example of such a particle collection technique (Cheng *et al.*, 2014, 2016a, b, 2017).

The HGPMS is a versatile technology that can be customized for a wide range of applications. The HGPMS technology consists of a core configuration with embedded multi-stage NdFeB permanent magnets that form reversing-pole assembly (Cheng *et al.*, 2014; 2017). The maximum magnetic field strength produced by the HGPMS technology at its current configuration is about 16 T/m. An aerosol collector built on the HGPMS principle does not require power to operate, if it rides on the auxiliary flow of other systems and can be operated for a long time such as months or years depending on operational requirement. A HGPMS sampler has a high collection efficiency for collecting aerosol particles similar to that of a High Efficiency Particulate Air (HEPA) filter without the pressure drop, but HGPMS preferentially collects magnetic aerosol particles while HEPA collects all PM (Cheng *et al.*, 2018b).

SELECTIVE COLLECTION USING AERIAL SAMPLING VEHICLE

It is commonly known that a location fixed ground-based collector or even a network of ground-based collectors can be deficient in fully characterize air contaminants in an

airshed and for source identification, for instance. If the ground-based collector is not in the plume, the collector does not have the chance to harvest any useful materials from the plume for assessment. Also, one would collect large amount of background or noisy particulate matter to dilute the signal that one has intended to collect. Thus, it is necessary that an environmental sampler has the ability to home-in the plume and collect the sample on-demand. That is smart sampling. Use of a mobile sampling platform like unmanned aerial systems (UAS) for air sampling represents a significant step toward smart sampling.

Unmanned aerial vehicle (UAV) or simply known as drone is an aircraft without a human being onboard. It is a part or a component of UAS, which consists of a drone or drones, a remote controller and a communication system. A drone however can be man-piloted or autonomous. Most of the civilian UAS these days are used for recreational purpose, land survey, aerial photography, law enforcement surveillance, agriculture guidance, Earth science and so on (Calo, 2011; Zhang and Kovacs, 2012; Anderson and Gaston, 2013; Colomina and Molina, 2014; Brouwer *et al.*, 2015; Madden *et al.*, 2015; Marinelli *et al.*, 2015; Wegener *et al.*, 2015). Interestingly, there have been applications recently where a drone carrying sensors and collectors is used for air sampling and meteorological measurements (Corrigan *et al.*, 2008; Lopez-Yglesias *et al.*, 2014; Brosy *et al.*, 2017), and monitoring methane leak (Patel, 2017). The US EPA has recently begun research investigating the use of unmanned aerial systems/sensors for emission monitoring (Gullette *et al.*, 2015) for various sources like open burning/open detonation and prescribed forest fires for which that the EPA does not have a standard sampling protocol to characterize source emissions.

SELECTIVE COLLECTION BY MODEL PROGNOSTICS

The last example of developing a smart sampler can be found in the area of collection of fallout particles (Cheng *et al.*, 2018a), particularly the volatile fraction of fallout particles. Fallout particles are a unique set of entities that only have a short lifetime compared with many environmental particles. Fallout particles are man-made and produced only through the use of atomic weapons. The majority of the particulate mass of fallout particles is comprised of background materials that are affected by the violent action of detonation. These particles would also contain unique chemical and isotopic species specific to the weapons used. The process of gas-particle phase partitioning of the elements leads to an elemental distribution on the fallout particles (Freiling, 1961; Thompkins, 1968). The refractive fraction of the fallout particles consists of those elements that rapidly transfer into the particulate phase after detonation. Elements that condense above 3,000 K include Al, Ca, Ti, Y, Zr, Ce, Pm, Sm, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, Ir, Np, Am, and Cm. Elements that condense under 1,000 K include H, P, Cl, K, As, Se, Br, Kr, Rb, Mo, Ru, I, Xe, Cs, and Po. Because of the condensation temperature of the matrix/carrier material

(~2,000 K), species in the first group will be found throughout the volume of the particles, whereas those in the second group will only be found on the surface of particles. The volatile fallout particles are generally smaller in size than those refractive ones; therefore, the lower settling velocity contributes to the long-range transport (on the order of hundreds of miles) for the volatile fractions. So, in theory, the volatile fraction of the fallout particles can be reasonably collected at farther distances downwind if a sufficient number of equivalent fissions can be collected at the distance.

The EPA RadNet has employed TSP high-volume sampler for the past several decades, which we know is ineffective in precision particle sizing (Watson *et al.*, 1983 and references therein). Thereby, such a sampler would encounter the issue discussed previously that is accumulating large number of unwanted particles and could potentially “dilute” the radiological fractionation content. This is not desirable for collect fallout particles. To design a smart sampler collecting the target volatile fraction of fallout particles, a precise estimate of the particle cut size is needed. This information can be retrieved from other physics-based model like the Department of Defense Fallout Prediction System (DELFI, 1979). The model was used to simulate the elemental fractionation in the atmosphere following a nuclear event (Cheng *et al.*, 2018a). The model calculations show that individual particles with a diameter of 64 μm or less should be volatile. Similarly, any sample of multiple particles that has an activity-weighted average particle size of 64 μm or less should also be volatile. Thus, applying the smart sampling concept a particulate collector designed to optimize collection of only particles of size smaller than or equal to 64 μm would be effective in harvesting volatile fallout particles. Again, a sampler without a properly defined inlet particle size would actually be misleading by diluting the volatile fraction in the sample and running at a risk of losing/washing the signal. Selective particle collection or smart sampling was accomplished by the aid of the DELFI model prognostics for the optimal particle size.

In summary, with the increased complexity in air pollution around the world and convolution of sampling airborne particulate matter for making correct decision, we need advanced samplers to provide useful data for solving contemporary particulate problems. Digital manufacturing technology, materials, unmanned mobile platforms, and aerosol science have advanced significantly in the past decade. It is time to take advantages of these technology advances for development of next-generation PM samplers that have the ability to be selective in collecting/harvesting target analytes in aerosol particles at the right time and right place.

CURRENT CHALLENGES IN SELECTIVE COLLECTION OF AEROSOL PARTICLES

Selective collection or material-property based collection in aerosol sampling is for direct signal enhancement that is critical for many decision-making processes. There are many challenges in the execution and or implementation of

selective environmental collection. For instances, one does not know the exact nature of the signal and its origin (as in the example of using model prognostics). The magnitude of the aerosol signal to be detected is embedded in a mixture of background material (as in the example of magnetic separation). The availability of suitable sampling platform (as in the example of aerial sampling platform) is lacking. Selective aerosol collection depends highly on the specific requirements and conditions of a sampling campaign. However, the key to a successful selective collection is to understand the nature of the aerosol material signal to be collected before a sampling platform is deployed or a new sampler is designed.

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