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Alternative metrics for spatially and temporally resolved ambient particle monitoring

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ABSTRACT

Particle active surface area is regarded as an important alternative metric to correlate particle emissions to adverse health effects. We explore the use of a particle counter and a diffusion charger as a relatively simple means to measure spatiotemporally resolved particle concentrations over a wide region using a mobile platform. Although the scanning mobility particle sizer (SMPS) measurements provide particle size distributions (PSDs) that can be converted into many important metrics, this method is impractical to implement for a routine monitoring over a wide region. In this study, an alternative method and new metrics are introduced using the condensation particle counter (CPC) and electrical aerosol detector (EAD). While the CPC and EAD measure particle number (PN) and particle active surface area (PS) concentrations respectively, the ratio of PS/PN provides an additional information of particle size which is important for particle transport. PSDs measured by SMPS during ambient monitoring were used to verify the concept. The study found that alternative metrics (PN, PS, and PS/PN) can be used to monitor spatiotemporally resolved particle concentrations over a wide region.

1. Introduction

Assessment of public exposure to particulate emissions is based on the measurements from stationary monitoring sites, which leads to a discrepancy from actual exposure (Violante et al., 2006). This raises concerns for the real exposure when people are near sources such as being on the road and living near the highways. Numerous studies and monitoring were based upon measurement of PM (Particulate Matter) mass concentrations at stationary monitoring sites. For example, AIRNOW.GOV gives a general trend of where the highest PM concentrations are regionally, but it does not give enough details at finer scale to show the highest concentrations on and near highways. In recognition of this issue, EPA's new air pollution rules require air pollution monitoring near road starting Jan 2014. For example, South Coast Air Quality Management District (SCAQMD, 2013) has implemented four air pollution monitoring stations in the proximity of major highways to monitor nitrogen oxides, fine particulate matter (soot), and carbon monoxide. This implementation is to assess public's real exposure to these air pollutants. There are nearly one million people living within 300 feet from major highways in the South Coast Air Basin (Barboza, 2013).

A common approach to resolve this issue is to monitor PM concentrations using mobile platforms to get temporally and spatially resolved information. Many different researchers had reported particle concentrations and size distributions using particle sizing instruments, such as Scanning Mobility Particle Sizer (SMPS, TSI Inc.) installed on a mobile platform (Durant et al., 2010; Fruin,

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Westerdahl, Sax, Sioutas & Fine 2008; Massoli et al., 2012; Pirjola et al., 2012; Westerdahl, Fruin, Sax, Fine & Sioutas, 2005). They provided detailed size distribution information, which is useful for research. However, particle size distribution measurement is not appropriate for a routine monitoring over a wide region. It is much more costly than other types of measurements for data collection and analysis. It is also difficult to present PSDs to show spatiotemporal evolution and distribution. Ranjan and Dhaniyala (2009) pointed out that a total number concentration is often sufficient to monitor particle exposure level.

Another approach is to monitor particle concentrations using multiple units of low cost particle sensors. Air Quality Egg (AQE) monitors NO_2 and CO gases globally using low cost sensors. Currently there is no true "low cost" PM sensor commercially available. Many of available low cost PM sensors use light scattering to detect particles, which is not appropriate to detect ultrafine particles. A PM mass sensor is being developed by researchers at UC Berkeley (Honicky, Brewer, Paulos & White, 2008). This sensor has the potential to become a true low cost PM sensor, which is also sensitive to ultrafine particles. Unfortunately, this sensor is currently not available in the market.

This study aims to propose and validate alternative metrics to monitor spatiotemporally resolved particle concentrations using relatively simple instruments: a particle counter and a particle surface instrument. There are a few promising commercially available instruments for ambient particle monitoring in real time. A condensation particle counter (CPC) has been widely used to measure number based particle concentrations. A diffusion charger is another promising instrument. The response of a diffusion charger is proportional to the effective surface area for acquiring electrical charge by the diffusion of ions produced in a corona discharge. Particle active surface area is a good metric to correlate with health effects.

The deficiency of using a single metric is a lack of particle size information. Particle transport such as particle lung deposition is a strong function of particle size. None of single metrics such as mass, surface area, and number gives such information. There are previous studies to use combination of a particle surface instrument and a particle counter for ambient air monitoring in an attempt to extract information regarding particle size. Ntziachristos, Polidori, Phuleria, Geller and Sioutas (2007) reported the mean diameter obtained from the ratio of Nanoparticle Surface Area Monitor (NSAM, TSI) and a CPC measurements were in good agreement with the SMPS arithmetic mean diameter from particle size distributions. The NSAM and Electrical Aerosol Detector (EAD, TSI) are similar instruments, because their fundamental measurement techniques. On the other hand, Frank, Saltiel, Hogrefe, Grygas and Garland Lala (2008) conducted calibration tests using a CAST (Combustion Aerosol STandard) system and reported that EAD/CPC and SMPS have a poor agreement in terms of the mean particle diameter and total aerosol length. They attributed the difference in the mean particle size determined by the SMPS against the EAD/CPC method to the difference in concentrations measured by the CPC and SMPS rather than any difference in the characteristics of the three instruments (SMPS, EAD and CPC). They reported that the EAD and SMPS measurements were consistent with each other because it was assumed that both instruments were affected by the same diffusion losses. Although Frank et al. (2008) was mainly focused on comparing the particle mean diameter, they did not look into the relationship between particle size distribution and EAD/CPC ratio. Frank et al. (2008) overlooked characteristics of the EAD such as power law dependence and could not properly extract information contained in EAD/CPC ratio.

This study aims to investigate particle active surface area concentration, particle number concentration, and their ratio as alternative metrics for spatially and temporally resolved ambient particle monitoring. A CPC and an EAD were used to monitor PN (particle number), PS (particle active surface area), and its ratio for this study. The study reports theoretical background and physical meaning of the EAD/CPC ratio. Data obtained from ambient air was analyzed to validate the concept. This method can be applied to mobile monitoring for a wide region using multiple platforms simultaneously considering relatively inexpensive instrumentation and convenience of data processing and presentation.

2. Theory

2.1. Background

Particles are charged by diffusion of ions and the charges carried by particles are detected by an electrometer in a unipolar diffusion charger equipped with an electrometer. The response of a diffusion charger is well predicted by diffusion charging theories, but some discrepancies exist for small particles. Pui, Fruin, and McMurry (1988) showed experimental data agree well with Fuchs charging theory above 10 nm. Biskos, Reavell, and Collings (2005) compared Fuchs charging theory with experimental data and calculation results by the birth-and-death theory. They found the experimental results agree well with Fuchs charging theory while the calculation fits well with the experimental results between 10 and 300 nm. On the other hand, Filippov (1993) compared Fuchs charging theory to Monte Carlo simulation and found out the agreement is down to 30 nm due to difference in treating ionic velocity distribution. Keller, Fierz, Siegmann, Siegmann and Filippov (2001) defined active surface area as the part of surface area that is responsible for exchanging energy and momentum in contrast to passive surface area which does not interact with carrier gas and diffusing species. This active surface area is a similar concept to Fuchs surface area, which was first defined by Pandis, Baltensperger, Wolfenbarger, and Seinfeld (1991). Keller et al. (2001) proposed scaling laws which relate mass transfer coefficient to mobility and diffusion constant. They concluded the influence of image force is negligible above 20 nm for diffusion charging.

Pui et al. (1988) determined the mean free path of the dominant ion species for diffusion charging as 14.5 nm. This makes the range of continuum and transition regime wider but makes free molecular regimes narrower for the size range of interest between 10 nm to 1 μm. Fuchs or active surface area is proportional to d_p^x where the exponent x is 1 for Kn≪1 and 2 for Kn≫1 for the mass transfer of diffusing species. In the transition regime, the exponent x varies as a function of Kn. Response of a diffusion charger can be approximated as a power of particle diameter for the range of measurement interest. Jung and Kittelson (2005) characterized