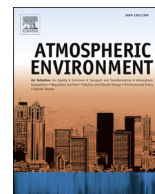




Contents lists available at ScienceDirect

## Atmospheric Environment

journal homepage: [www.elsevier.com/locate/atmosenv](http://www.elsevier.com/locate/atmosenv)

# Passive sampling to capture the spatial variability of coarse particles by composition in Cleveland, OH



Eric J. Sawvel<sup>a</sup>, Robert Willis<sup>b</sup>, Roger R. West<sup>c</sup>, Gary S. Casuccio<sup>c</sup>, Gary Norris<sup>b</sup>, Naresh Kumar<sup>d</sup>, Davyda Hammond<sup>e</sup>, Thomas M. Peters<sup>a,\*</sup>

<sup>a</sup> The University of Iowa, Iowa City, IA, USA

<sup>b</sup> US EPA, Office of Research and Development, National Exposure Research Laboratory, Research Triangle Park, NC, USA

<sup>c</sup> RJ Lee Group, Inc., Monroeville, PA, USA

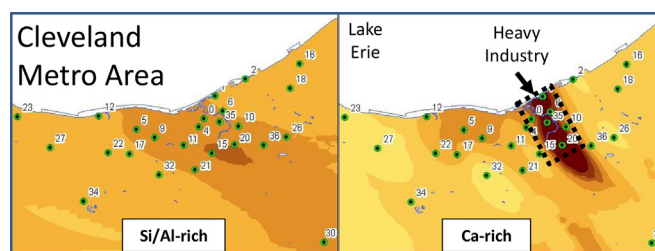
<sup>d</sup> University of Miami, Miami, FL, USA

<sup>e</sup> Oak Ridge Associated Universities, Oak Ridge, TN, USA

## HIGHLIGHTS

- Examined spatial variability of PM<sub>10-2.5</sub> and components in Cleveland, OH.
- Used passive samplers with automated microscopy to classify particles.
- Components associated with steel and cement production highest in industrial area.
- Components associated with crustal material more uniformly distributed.
- Method may be useful to reduce exposure misclassification in epidemiological studies.

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Article history:

Received 2 November 2014

Received in revised form

12 January 2015

Accepted 14 January 2015

Available online 15 January 2015

### Keywords:

PM<sub>10-2.5</sub>, particulate matter  
Computer controlled scanning electron microscopy  
Single particle analysis

## ABSTRACT

Passive samplers deployed at 25 sites for three, week-long intervals were used to characterize spatial variability in the mass and composition of coarse particulate matter (PM<sub>10-2.5</sub>) in Cleveland, OH in summer 2008. The size and composition of individual particles determined using computer-controlled scanning electron microscopy with energy-dispersive X-ray spectroscopy (CCSEM-EDS) was then used to estimate PM<sub>10-2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) and its components in 13 particle classes. The highest PM<sub>10-2.5</sub> mean mass concentrations were observed at three central industrial sites ( $35 \mu\text{g m}^{-3}$ ,  $43 \mu\text{g m}^{-3}$ , and  $48 \mu\text{g m}^{-3}$ ), whereas substantially lower mean concentrations were observed to the west and east of this area at suburban background sites ( $13 \mu\text{g m}^{-3}$  and  $15 \mu\text{g m}^{-3}$ ). PM<sub>10-2.5</sub> mass and components associated with steel and cement production (Fe-oxide and Ca-rich) exhibited substantial heterogeneity with elevated concentrations observed in the river valley, stretching from Lake Erie south through the central industrial area and in the case of Fe-oxide to a suburban valley site. Other components (e.g., Si/Al-rich typical of crustal material) were considerably less heterogeneous. This work shows that some species of coarse particles are considerably more spatially heterogeneous than others in an urban area with a strong industrial core. It also demonstrates that passive sampling coupled with analysis by CCSEM-EDS is a useful tool to assess the spatial variability of particulate pollutants by composition.

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\* Corresponding author. 145 N Riverside Drive, S331 CPHB, University of Iowa, Iowa City, IA, USA.

E-mail address: [thomas-m-peters@uiowa.edu](mailto:thomas-m-peters@uiowa.edu) (T.M. Peters).

## 1. Introduction

Exposure to fine atmospheric particulate matter (PM<sub>2.5</sub>) has been associated with increased adverse cardiopulmonary health effects (Brook et al., 2010; Hoek et al., 2013), although effect estimates vary significantly among studies with heterogeneity in fine particle composition suspected as a factor of considerable uncertainty (Hoek et al., 2013). The evidence is less clear for adverse health effects associated with exposure to coarse particulate matter (PM<sub>10-2.5</sub>), with recent meta-analyses reporting a lack of evidence for mortality (Hoek et al., 2013), but ‘suggestive evidence’ for increased morbidity and mortality not explained by simultaneous co-exposure to PM<sub>2.5</sub> (Adar et al., 2014). These meta-analyses stress the need to better characterize the heterogeneity of particulate matter exposures by composition to reduce uncertainty in effect estimates, especially for PM<sub>10-2.5</sub>.

The combination of varying sources and short atmospheric lifetimes often leads to substantial heterogeneity in the concentration and chemical makeup of PM<sub>10-2.5</sub>. Coarse atmospheric particles are emitted primarily by widely varying mechanical and resuspension sources, leading to a complex mixture of material from roads, soil, wear of automotive parts (e.g., tires and brakes), and biological material from vegetation (Kelly and Fussell, 2012). Particle settling velocity scales with diameter squared causing coarse particles to settle substantially faster than fine particles (Seinfeld and Pandis, 2012; Zhang and He, 2014). Consequently, the use of data from spatially sparse networks of regulatory samplers can result in substantial exposure misclassification for PM<sub>10-2.5</sub> that can attenuate the power of epidemiological studies (Chang et al., 2011).

Networks of active samplers—samplers that collect particles from an aspirated volume of air—have been used to measure the spatial variability of PM<sub>10-2.5</sub> in urban settings. Burton et al. (1996) used paired PM<sub>10</sub> and PM<sub>2.5</sub> filter samplers at eight sites to show that coarse particles (calculated by subtraction: PM<sub>10</sub> – PM<sub>2.5</sub>) were heterogeneously distributed across Philadelphia, PA. This subtraction method, however, is subject to multiple measurement error from two filter samplers, introducing measurement uncertainty in gravimetric measurement that is amplified in chemical analysis (Goldman et al., 2011). Using two impactors in series, Thornburg et al. (2009) measured coarse particles separately from particles of other size. They found that PM<sub>10-2.5</sub> measured at five sites in Detroit, MI were temporally correlated and had low spatial heterogeneity. The spatial heterogeneity of PM<sub>10-2.5</sub> and its components has been studied intensively in Los Angeles, CA with networks of cascade impactors (Cheung et al., 2011; Cheung et al., 2012; Fruin et al., 2014). At 10 downtown and suburban sites, PM<sub>10-2.5</sub> mass was observed to be moderately heterogeneous (Pakbin et al., 2010) with greater heterogeneity observed for components of coarse particles (Cheung et al., 2011; Cheung et al., 2012). Fruin et al. (2014) identified substantial within-community heterogeneity in PM<sub>10-2.5</sub>.

Networks of passive samplers have been used to investigate the spatial and temporal variability in PM<sub>10-2.5</sub>. Compared to active sampling, passive samplers are relatively inexpensive, require no electricity to operate, and can be deployed at numerous locations easily and cost-effectively (Wagner and Leith, 2001). Ott et al. (2008b) used passive samplers at 30 sites with analysis by light microscopy to show that coarse PM was highly heterogeneous at a spatial scale of 4.4 km in a medium-sized Midwest city. Lagudu et al. (2011) used a network of 25 passive samplers with analysis by computer-controlled scanning electron microscopy coupled with energy-dispersive X-ray spectrometry (CCSEM-EDS) to show PM<sub>10-2.5</sub> mass and its components were highly heterogeneous at a spatial scale of 2 km across Rochester, NY.

Less work has been done to assess the spatial heterogeneity of PM<sub>10-2.5</sub> in cities with substantial industrial activity, such as Cleveland, OH. Cleveland, OH is a ‘rust belt’ city with substantial steel and cement production in a central river valley. In a previous publication, the spatial heterogeneity of iron-containing particles within the Cleveland metropolitan area was investigated using arrays of passive samplers analyzed by CCSEM-EDS (Ault et al., 2012). Results indicated that anthropogenic iron-containing coarse particles were highly heterogeneous and subject to physicochemical transformation as they moved away from their source.

The goal of the present work was to more broadly investigate the spatial heterogeneity of PM<sub>10-2.5</sub> mass and compositional components in Cleveland, OH using a network of passive samplers coupled with single particle analysis by CCSEM-EDS. Particles were classified into 13 compositional classes based on their X-ray spectra. The spatial variability of PM<sub>10-2.5</sub> and the 13 components were evaluated with visual and quantitative indicators of heterogeneity. We report that anthropogenic particles from the industrial core are more heterogeneous than crustal material. These results may be important for interpreting epidemiological data for cities with strong industrial cores. Moreover, passive sampling with single particle analysis represents an alternative exposure assessment method for the epidemiology of PM<sub>10-2.5</sub>.

## 2. Methods

### 2.1. Study area

Sampling was conducted in the Cleveland, OH metropolitan area (Fig. 1). Included in the study area is Cleveland’s Flats District, a low-lying topography along the banks of the lower Cuyahoga River from the river’s mouth at Lake Erie stretching south approximately 8 km. The elevation of the river surface is approximately 180 m above sea level. The surrounding bluffs start at an elevation of 213 m and extend to a height of approximately 365 m above sea level. The river valley width varies from 0.8 km at its narrowest point and widens to approximately 2.4 km.

Within the Flats, large quantities of steel are produced by integrated and electric arc furnace. Steel production uses large quantities of aluminum and calcium, some of which results in slag, a waste byproduct composed mostly of alumina, lime, and trace metals (van Oss, 2009). Slag is then used as a raw material in the manufacturing of cement, another industry common to the Flats. Other industries in this area include asphalt, gravel, petroleum, and aluminum processing, along with road salt production and storage.

### 2.2. Site selection

A method designed to optimize the capture of spatial variability in PM<sub>10-2.5</sub> was used to select sampling sites. As described by Kumar et al. (2011), a preliminary ‘demand surface’ of PM<sub>10</sub> with a high spatial resolution (90 m) was generated for the study area, using the empirical relationship between satellite-based aerosol optical depth and ground-based PM<sub>10</sub> measurements. PM<sub>10</sub> was used as the best available surrogate for PM<sub>10-2.5</sub>. Sites were identified and ranked that would maximize the variability observed in the preliminary surface. This approach, unlike classical sample site selection, was adopted to minimize redundancy in sites by controlling for spatial autocorrelation. The locations of optimal sites were adjusted by moving them to the closest schools, churches, fire stations, and private homes as practical for logistic and security reasons. One sampler was damaged during the first week, and this site was dropped from the remainder of the study, leaving a total of 25 sites for the study.

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