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Spatiotemporal distribution of airborne particulate metals and metalloids in a populated arid region

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HIGHLIGHTS

- Spatiotemporal analysis of $PM_{2.5}$ elemental composition in southern Arizona.
- Concentrations of crustal elements exhibit a distinct day-of-week pattern.
- There have been significant reductions in toxic species concentrations since 1988.
- Toxic element concentrations are enhanced during 'dust events' in urban areas.
- Phoenix has the highest concentrations of airborne Cu, Zn, and crustal elements.

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ABSTRACT

A statistical analysis of data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network of aerosol samplers has been used to study the spatial and temporal concentration trends in airborne particulate metals and metalloids for southern Arizona. The study region is a rapidly growing area in southwestern North America characterized by high fine soil concentrations (among the highest in the United States), anthropogenic emissions from an area within the fastest growing region in the United States, and a high density of active and abandoned mining sites. Crustal tracers in the region are most abundant in the summer (April–June) followed by fall (October–November) as a result of dry meteorological conditions which favor dust emissions from natural and anthropogenic activity. A distinct day-of-week cycle is evident for crustal tracer mass concentrations, with the greatest amplitude evident in urban areas. There have been significant reductions since 1988 in the concentrations of toxic species that are typically associated with smelting and mining. Periods with high fine soil concentrations coincide with higher concentrations of metals and metalloids in the atmosphere, with the enhancement being higher at urban sites.

1. Introduction

Atmospheric aerosol particles impact the planet's radiation balance, the hydrologic cycle, public health and welfare, and biogeochemical cycling of nutrients. Aerosol particles act as carriers for potentially harmful contaminants and beneficial nutrients to ecosystems. Potential health effects of aerosol particles are linked to their physicochemical properties that govern their transport patterns, method of deposition, and ultimate health impact upon deposition. Of most concern for health effects are fine particles (mean aerodynamic diameter <2.5 μm), as they are most effective at penetrating the extrathoracic, conducting, and pulmonary airways of the human body upon inhalation (Park and Wexler, 2008). While the size and hygroscopicity of particles will largely govern where particles deposit upon inhalation, knowledge of their composition is critical for predicting their ultimate health effect upon deposition.

Metals and metalloids, of which many are considered by the Environmental Protection Agency (EPA) to be air toxins (e.g., arsenic (As), lead (Pb), nickel (Ni), chromium (Cr), manganese (Mn)), are commonly found in ambient particles. They are natural

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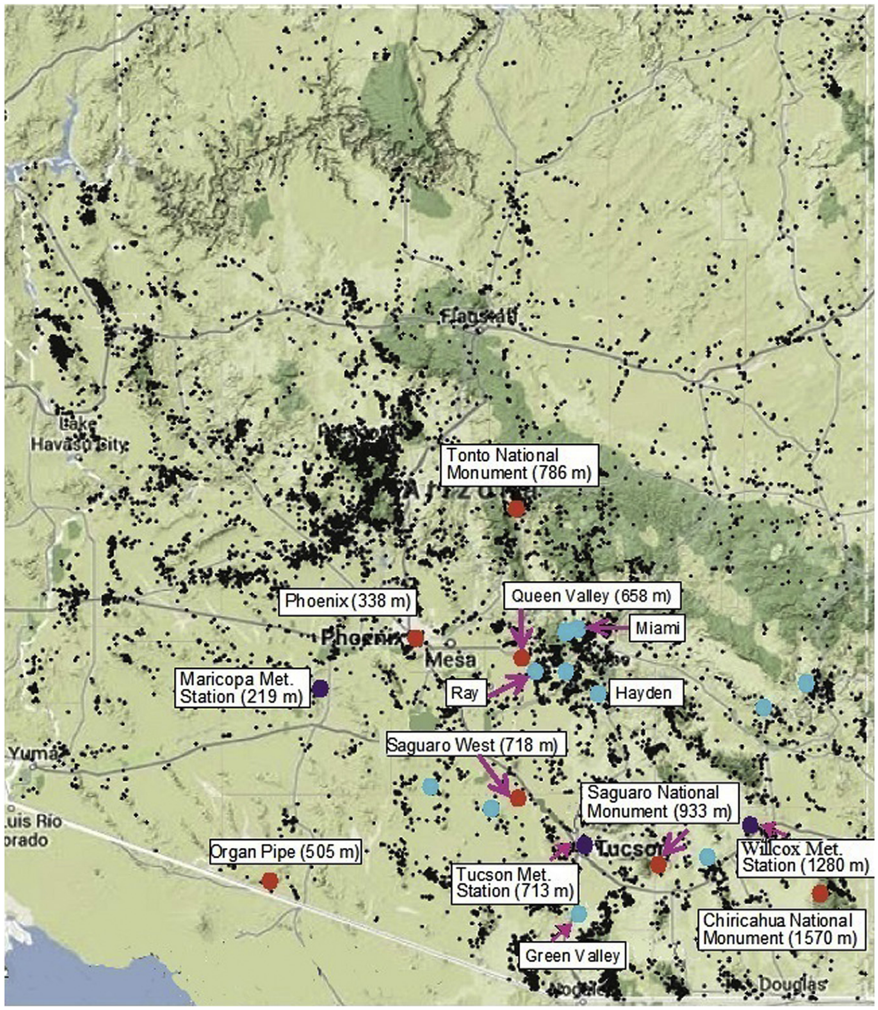

Fig. 1. Geographic location of southern Arizona IMPROVE stations (orange markers), with altitudes above sea level reported in parentheses. Locations of selected mines, prospects, quarries, and processing mills and plants (including abandoned mines) are shown as black dots (State of Arizona Department of Mines and Mineral Resources, http://mines.az.gov/Publications/AzmilsMap.pdf); the emissions strength of metals and metalloids from these sites is uncertain. Also shown are the location of major active copper mining and smelting operations (blue markers) (http://www.admmr.state.az.us/), and meteorological stations close to the IMPROVE sites (purple markers). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

components of crustal materials (e.g., wind-blown dust) and are also generated from high-temperature processes (smelting and fossil fuel combustion) (Thornton, 1991; Alloway, 1995; Reheis et al., 2009; Schauer et al., 2006; Navarro et al., 2008; Ettler et al., 2014). The transport of contaminants via particles is spatially more extensive than by other means such as waterways (e.g., surface and ground water) (Csavina et al., 2012). Metal and metalloid contaminants can also be found in rain and snow (e.g., Barbaris and Betterton, 1996; Cheng et al., 2011), indicating that sources of particles enriched with metals and metalloids can influence distant regions via the role of particles in serving as cloud condensation nuclei (CCN) and ice nuclei (IN) and depositing via precipitation. Concentrations of crustal-derived species (calcium, magnesium, potassium, sodium) peak between March and June in both $PM_{2.5}$ and precipitation in the southwestern United States (Southwest), indicative of the role of dust as both CCN and IN (Sorooshian et al., 2013). If toxic species, or even fungi, allergens, and pathogens, are associated with such dust particles that can deposit as precipitation, they have the potential to adversely impact downwind ecosystems and biota inhabiting those areas.

The potential link between atmospheric aerosol particles, especially soil dust, and harmful metals and metalloids is a concern for arid and semi-arid regions that cover approximately one-third of the global land area. The long lifetime of metals in soils (Alloway, 1995; Kelly et al., 1996) provides a significant residence time frame for these contaminants to be transported to downwind sites via soil erosion and atmospheric transport, followed by subsequent deposition (Galloway et al., 1982; Johnson et al., 1994). In North America, the region most impacted by soil dust is the Southwest (Malm et al., 2004), which includes the focus region of this study (Fig. 1). Naturally unvegetated or anthropogenically disturbed soil surfaces, such as dry lakes ("playas"), dry washes, gravel pits, large construction sites and fields (after harvest) can be major sources of wind-blown dust. Mine tailings in particular can provide large uncovered surfaces for dust emissions and contain higher concentrations of metals and metalloids than other soil surfaces (Boulet and Larocque, 1998). While regulations and initiatives for dust emission suppression (e.g., revegetation, surface moistening) are in place for active mines, mine tailings in abandoned mining sites are allowed to dry and can stay unvegetated for decades (Mendez and Maier, 2008), thus becoming sources of contaminated dust (Moreno et al., 2007).

Southern Arizona is an ideal setting to study the pervasiveness of metals and metalloids in the atmosphere, especially in relation to