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Hygroscopic and Chemical Properties of Aerosols collected near a Copper Smelter: Implications for Public and Environmental Health

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Abstract

Particulate matter emissions near active copper smelters and mine tailings in the southwestern United States pose a potential threat to nearby environments owing to toxic species that can be inhaled and deposited in various regions of the body depending on the composition and size of the particles, which are linked by particle hygroscopic properties. This study reports the first simultaneous measurements of size-resolved chemical and hygroscopic properties of particles next to an active copper smelter and mine tailings by the towns of Hayden and Winkelman in southern Arizona. Size-resolved particulate matter samples collected near an active copper smelter were examined with inductively coupled plasma mass spectrometry, ion chromatography, and a humidified tandem differential mobility analyzer. Aerosol particles collected at the measurement site are enriched in metals and metalloids (e.g. arsenic, lead, and cadmium) and water-uptake measurements of aqueous extracts of collected samples indicate that the particle diameter range of particles most enriched with these species (0.18–0.55 μm) overlaps with the most hygroscopic mode at a relative humidity of 90% (0.10–0.32 μm). These measurements have implications for public health, microphysical effects of aerosols, and regional impacts owing to the transport and deposition of contaminated aerosol particles.

Keywords

Particulate Matter; Soil Dust; Hygroscopicity; Smelter; Arizona; Mine Tailings; Arsenic; Lead; Oxalate; Organic Acid

1. Introduction

The chemical complexity and uncertainties in production mechanisms of atmospheric aerosol species pose a challenge to assess the impact of aerosol particles on public health/

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Supporting Information Available: Additional tables are provided to summarize MOUDI sample collection dates and wind data for the EF analysis. This information is available free of charge via the Internet at <http://pubs.acs.org>.

welfare and for accurate predictions of their interactions with water vapor, radiation, and clouds. These interactions not only depend on composition, but also particle size, which is linked to composition owing to the process of hygroscopic water uptake. The southwestern United States is experiencing rapid population growth, land use change, and drought conditions, which promote natural and anthropogenic emissions leading to aerosol particles with poorly understood chemical and hygroscopic properties. Arid and semi-arid regions such as the US Southwest cover approximately one-third of the global land area resulting in soil dust being the most abundant aerosol type on a mass basis (1). The highest nationwide particulate concentrations of soil dust in the United States are in the Southwest (2). Dust is of special interest in the Southwest owing to a number of factors: (i) effects on the development of clouds and precipitation (3–5); (ii) deposition on snowpacks and expedited snow melt as a result of their light-absorbing properties (6); (iii) rapidly diminishing visibility and exacerbating health and safety risks (e.g. traffic accidents) when in the form of dust storms; and (iv) their ability to carry a suite of allergens, pathogens, toxic metals, and fungi such as *Coccidioides Immitis* (7–9).

An issue related to public and environmental health in the Southwest is the potential enrichment of trace metals and metalloids in the regional aerosol particles, especially soil dust, as a result of anthropogenic activity such as smelting and fossil fuel combustion. Among the highest nationwide atmospheric levels of harmful metals and metalloids such as arsenic (As), copper (Cu), and zinc (Zn) are found in southern Arizona and this has been suggested to be due to the high density of smelting and mining activity in the region (10–11). Mine tailings (i.e. materials left over after the valuable fraction has been extracted from ores) can threaten environmental and public health as a result of transport of toxic compounds into nearby soils, waterways, and the atmosphere (e.g. 12–15). Recent measurements made in the vicinity of a smelter in Arizona suggested that wind-blown dust particles acquire contaminants such as lead (Pb) and As by deposition of sub-micrometer particles from smelting operations (9). Earlier work in Arizona also indicated that airborne particles originating from smelting activities are enriched with high levels of metals and metalloids that can settle and contaminate the local topsoil (10, 16–18).

A topic that has received little attention is the hygroscopicity of such contaminated aerosol particles. Hygroscopicity is a critical aerosol property that governs the ability of a particle to swell or shrink as a result of variations in relative humidity (RH), which influences deposition upon inhalation, light interactions, and the efficacy of aerosols at activating into cloud drops. The role of aerosols as cloud condensation nuclei (CCN) provides a potential explanation for presence of metals and metalloids in wet deposition (19–22). In this work, the hygroscopicity and composition of size-segregated aerosol particles are examined to understand the nature and potential effects of airborne aerosols adjacent to an active smelter and mine tailings site.

2. Methods

Ambient aerosol particles were collected near an active copper smelting site and mine tailings near the towns of Hayden and Winkelman in Arizona (Figure 1). A 10-stage micro-orifice uniform deposit impactor (MOUDI; 23) was used to carry out size-resolved (aerodynamic equivalent $D_{p,50}$ cut sizes of 0.054, 0.10, 0.18, 0.32, 0.55, 1.0, 1.8, 3.1, 6.2, 9.9, 18.0 μm) measurements using teflon filter substrates (PTFE membrane, 2 μm pore, 46.2 mm, Whatman) on the roof of a high school building in Winkelman, approximately 2 km from the mine tailings pile and 1 km from the smelter and smokestack. The hygroscopicity analysis was based on one set of MOUDI samples collected between 9 February 2010 and 25 February 2010, with a sampling program consisting of weekdays between 07:00–15:30 (Local Time). The sampling flow rate was 30 L min^{-1} with a total of 96 h of sampling. The

sample set was fractionated for simultaneous chemical characterization and hygroscopicity analysis. The filters were quantitatively fractionated by gravimetric analysis using an ultra-microbalance (Mettler Toledo XP2U) and Environmental Protection Agency (EPA) Class I equivalent methods. Other MOUDI sample sets were collected for chemical characterization; these samples also experienced a total of 96 h of aerosol collection at a flow rate 30 L min^{-1} (refer to Supplementary Information for sample schedule). For metal and metalloid composition, filters were analyzed using inductively coupled plasma mass spectrometry (ICP-MS; Agilent 7500ce with an Octopole Reaction System). Filters were extracted using the method of Harper et al. (24), which is quantitative for select elements of interest in this study. The filters were extracted using 15 mL of aqua regia (1.03 M HNO_3 /2.23 M HCl trace-metal grade) in sealed glass vials placed in a sonicator at 80°C for 60 min. Extract aliquots of 1.2 mL were diluted to 4 mL with deionized water and then refrigerated prior to ICP-MS analysis. The main sample set of interest was also analyzed with ion chromatography (IC; Thermo Scientific Dionex ICS-5000 anion system with an AS11-HC 2 mm column) and water-uptake measurements. The filter extraction procedure consisted of ultrasonication (15 min) of filter halves with 18.2 mega-ohm Milli-Q water. Syringe filters (Acrodisc filter, $25 \mu\text{m}$) were used to remove any remaining insoluble matter from the extracts after ultrasonication. IC analysis was conducted on triplicate samples using a 38-min multi-step gradient program with sodium hydroxide eluent (1 mM from 0 to 8 min, 1 mM to 30 mM from 8 to 28 min, 30 mM to 60 mM from 28 to 38 min). This work reports relative concentrations of inorganic (chloride (Cl^-), nitrite (NO_2^-), nitrate (NO_3^-), sulfate (SO_4^{2-})) and organic acid (oxalate, formate, acetate, lactate) species. Blank filter samples following the same extraction procedures described above were chemically examined with ICP-MS and IC to confirm the absence of artifacts and contamination that would bias the study results.

Portions of the extracted solutions were atomized into a humidified tandem differential mobility analyzer (HTDMA; Brechtel Manufacturing Inc. (BMI) Model 3002) for size-resolved aerosol hygroscopicity measurements. A constant-rate atomizer with controllable liquid supply flow rate and an in-line desiccant dryer (all of stainless steel construction) (BMI Model 9200) were used for re-aerosolization. The generated polydisperse aerosol sample flow from the atomizer was dried to a RH less than 5% prior to being brought to an equilibrium charge distribution in a charge neutralizer. The particles then entered a differential mobility analyzer (DMA; BMI Model 2000C), which was used to select 40 nm diameter dry particles; this diameter was found to coincide with the peak concentration of re-aerosolized particles from the atomizer. The sample flow containing the monodisperse particles was subsequently humidified to a range of pre-selected RHs, chosen to be 50%, 70%, 80%, and 90%. The humidified particles then entered a second DMA which is coupled to a mixing-type condensation particle counter (BMI Model 1710). The sheath flow in the second DMA was controlled to the same RH set-point as the humidified sample flow. The voltage of the second DMA was varied over time to determine particle concentrations as a function of wet particle diameter and thus the number-size distribution of humidified particles (25). The final parameter quantified is termed the hygroscopic growth factor (GF), which is defined as the ratio of the wet particle diameter ($D_{p,\text{wet}}$) at a defined RH relative to the dry particle diameter ($D_{p,\text{dry}}$): $\text{GF} = D_{p,\text{wet}}/D_{p,\text{dry}}$. Single values of hygroscopic GFs for each MOUDI filter were determined as the modal diameter of the humidified size distribution measured by the second DMA, divided by the corresponding dry diameter. Quality control was performed for HTDMA scans by ensuring that the combined humidifier and sheath air RH variability over the time of a scan did not exceed $\pm 1\%$ and the temperature gradient over the length of the second DMA column did not exceed $\pm 1^\circ\text{C}$. The ambient temperature during HTDMA scans ranged between $19\text{--}24^\circ\text{C}$. It is noted that atomizing the extracted MOUDI samples into the HTDMA effectively re-distributes the components of the collected aerosols on each MOUDI size stage in such a way as to

normalize the hygroscopic properties of all particles, whether internally or externally mixed, on each stage. It is noted that blank filter and water samples were atomized to confirm the absence of biases associated with the aerosol re-generation process.

To identify transport patterns of air masses originating at the sample site, forward trajectories were computed using the NOAA HYSPLIT model (26). One-day forward trajectories were computed daily between 2005–2010, with the start point being at the MOUDI sampling site (32°59'42.02" N, 110°46'18.04" W). To complement the trajectory analysis, surface-based measurements of precipitation for a representative site in southern Arizona (Tucson; 32°16'49" N, 110°56'45" W) were obtained from the Arizona Meteorological Network (<http://ag.arizona.edu/azmet/>).

3. Results and Discussion

3.1 Composition

Size-resolved composition data are summarized in Figures 2 and 3a. In addition to the two-week period of interest, Figure 2 shows averages of the size-resolved trace species levels for five other MOUDI sample sets collected in the month of February between 2009–2011 to show the consistent nature of the size-resolved elemental composition. There is bimodal concentration profile (cutpoint $D_p \sim 0.18\text{--}0.32\text{ }\mu\text{m}$ and $D_p = 6.2\text{ }\mu\text{m}$) for crustal tracers (indicative of soil dust aerosols) including aluminum (Al), iron (Fe), scandium (Sc), and magnesium (Mg). Aerosol morphology from electron microscopy analysis of filters collected at the same measurement site have shown that the larger size ranges extending above aerodynamic diameters of $1\text{ }\mu\text{m}$ are associated with crushing, grinding, and wind-blown dust while the sub-micrometer particles likely arise from condensation and coagulation of smelting emissions (9). The other trace elements examined, including As, beryllium (Be), cadmium (Cd), Cu, and Pb exhibit the same bimodal distribution, with a number of these species exhibiting higher concentrations in the $0.18\text{--}0.55\text{ }\mu\text{m}$ range including As, Cd, and Pb. The enrichment of trace contaminants in sub-micrometer particles is suggestive of relatively fresh smelter emissions while the presence of these species in super-micrometer particles is likely linked to either deposition of the fine, smelter-derived particles on soil dust particles that are subsequently transported via wind, or of wind-blown particles originating from contaminated soil and tailings.

The water-soluble composition of the collected aerosols (i.e. sum of Cl^- , NO_2^- , NO_3^- , SO_4^{2-} , oxalate, formate, acetate, lactate) indicates that sulfate mass fractions are highest in the sub-micrometer size range, ranging between 55–73%. The main source of sulfate in the sampling area is expected to be associated with smelter emissions and sulfuric acid plant emissions. The highest nitrate and nitrite mass fractions are present in the super-micrometer particles (up to 36% and 9%, respectively, on the $6.2\text{ }\mu\text{m}$ MOUDI stage). Previous work has suggested that owing to reactions of nitric acid (HNO_3) and its precursors with dust, nitrate is associated with super-micrometer particles in the region (27–28). Nitrite can also partition into the aerosol phase as a result of uptake and reactions of nitrogen oxides on the surface of dust particles (29), which could explain its highest mass fractions (8–9%) in the $3.11\text{ }\mu\text{m}$ and $6.2\text{ }\mu\text{m}$ MOUDI stages. Chloride exhibits unexpectedly high mass fractions (up to 16%) since it is typically associated with sea spray particles, which do not influence the measurement site. Furthermore, this species is not typically associated with smelter emissions (30), but rather is emitted by steelworking (31), waste incineration (30), coal burning (32), and is associated with dust due to heterogeneous reactions with HCl(g) (33). It is likely that an anthropogenic source resulted in the high Cl^- levels, with its presence in super-micrometer particles linked to a similar mechanism that likely also explains the abundance of nitrate and nitrite in these larger particles.

Organic acids are ubiquitous in ambient aerosols and are of especial importance owing to their influence on the hygroscopicity of aerosols (e.g. 34). A series of low molecular weight organic acids (lactate, acetate, formate, oxalate) accounted for 10 to 36% of the water-soluble mass for individual MOUDI stages, with a higher average mass fraction in the super-micrometer stages ($23\% \pm 10\%$) as compared to sub-micrometer stages ($16\% \pm 9\%$). Among just the organic acids, acetate exhibited the highest total mass fraction ($11\% \pm 8\%$), with a greater average contribution to the total organic acid mass for super-micrometer sizes ($52\% \pm 7\%$) as compared to the sub-micrometer sizes ($30\% \pm 22\%$). Formate and lactate exhibited nearly identical average organic acid mass fractions in the sub- and super-micrometer size ranges (formate $\sim 20\%$; lactate $\sim 25\%$), but oxalate exhibited its highest organic acid mass fractions in sub-micrometer sizes ($26\% \pm 13\%$) as compared to super-micrometer sizes ($14\% \pm 21\%$). Comparable mass fractions for acetate, formate, and oxalate relative to sub-micrometer water-soluble aerosol mass have been observed in urban and remote regions, in addition to smog chamber organic photooxidation experiments (35). Previous work suggests that the presence of such carboxylic acids in coarse particles may be associated with their condensation and adsorption onto pre-existing coarse particle surfaces (36), with potential heterogeneous processing (37). Gas-to-particle conversion is mostly responsible for presence of these low-volatility organic acids in the fine mode, specifically for oxalate, which has been documented to be an important secondary organic aerosol tracer, especially in the aqueous phase for fine particles (38).

3.2 Enrichment Factors at Sample Site

A common way to quantify the enhancement of specific species in aerosols when comparing different areas or sources is the enrichment factor (EF). This is the ratio of a given element to a reference element at a site of interest divided by the same ratio at a baseline site. The reference element chosen here is Sc, which has insignificant anthropogenic sources and is a commonly-used crustal tracer species in EF analysis (39–40). Enrichment factor is therefore calculated as follows:

$$EF = [C_{n(\text{sample})}/C_{\text{ref}(\text{sample})}]/[B_{n(\text{baseline})}/B_{\text{ref}(\text{baseline})}] \quad (1)$$

where $n = \text{As, Cd, or Pb}$ and $\text{ref} = \text{Sc}$. Enrichment factors larger than unity would indicate that an element is enriched in the sample aerosols relative to the baseline site chosen. Three baseline sites are used (shown in Figure 1): (i) same location of the normal measurements except that the smelter was shut off with similar average frequencies of wind speeds and directions based on data from two weather stations co-located with the MOUDI sampler (refer to Supporting Information for details); (ii) the city of Green Valley, which is 150 km south of the measurement site and within 1.5 km of a metal-free mine tailings pile; (iii) on the roof of the Physics and Atmospheric Sciences building of The University of Arizona, in Tucson, which represents an urban setting (metropolitan population ~ 1 million; US Census Bureau 2009) and is 110 km south of the measurement site. The EF results are based on between four and six MOUDI sample sets collected at the baseline sites and 59 MOUDI sample sets collected at the measurement site when the smelter was on. The reader is referred to the Supporting Information for sample collection dates for sample sets included in the EF analysis.

The results in Figure 4 point to major enhancements in As, Cd, and Pb at the sample site during periods when the smelter is active, with the largest EFs observed for As in the smallest size range examined ($D_p < 1 \mu\text{m}$). When the baseline chosen was Hayden without active smelting operations, there was a reduction in all three EFs as a function of size in the super-micrometer range as compared to the other two baseline sites. This suggests that the larger aerosol particles near the sample site by Hayden and Winkelmann are contaminated to a larger extent with these three toxic species relative to the other sites and that active

smelting does not significantly alter the background levels of these species for $D_p > 9.9 \mu\text{m}$. These results indicate that the transport of aerosols near the sample site to other nearby locations has the potential to impact public health owing to the enrichment of As, Cd, and Pb. To put the results of Figures 2 and 4 in perspective, Pb is currently one of six criteria pollutants that the Clean Air Act requires the EPA to set National Ambient Air Quality Standards (NAAQS) for with a current primary standard of $0.15 \mu\text{g m}^{-3}$ on a rolling three-month average time basis (<http://www.epa.gov/air/lead/standards.html>). The integrated Pb concentrations in Figures 2a and 2b are $0.018 \mu\text{g m}^{-3}$ and $0.028 \mu\text{g m}^{-3}$, respectively.

3.3 Aerosol Hygroscopicity

Figure 3b summarizes the size-resolved sub-saturated hygroscopic GFs of the study aerosol particles. For comparison to the experimental data, GF predictions for a representative inorganic species (ammonium bisulfate) are provided. These are based on a modified Kohler model that includes a parameterization for the solution osmotic coefficient that approaches the correct thermodynamic limit as the solution becomes infinitely dilute (41–42). Representative values for an organic acid salt (oxalic acid, $\text{C}_2\text{H}_2\text{O}_4$) are also included based on laboratory experiments for this pure test aerosol (43).

At a RH of 50%, the hygroscopic GFs of the sample aerosols were within the narrow range of 1.04–1.08, with an increasing range of values at the higher RHs (1.14–1.25 at 70%; 1.25–1.39 at 80%; 1.43–1.88 at 90% RH). At RHs exceeding 50%, GFs were systematically higher for sub-micrometer particles as compared to larger sizes and this is most likely due to higher relative amounts of sulfate (55–73%) in the five smallest MOUDI stages. The reduction in GFs at high RH for super-micrometer particles is most likely associated with a greater contribution of less soluble material such as organics that are less hygroscopic than the more sulfate-rich sub-micrometer particles. The measurements almost always lie in between the range of GFs for the two model salts at each of the four RHs examined. The exceptions are for the super-micrometer stages where the measured GFs are below that of pure oxalic acid, suggestive of the influence of less hygroscopic species.

The results in Figures 2 and 3 are important from a health effects perspective since particles can be exposed to very humid conditions ($> 90\%$) in the human respiratory tract (e.g. 44). While the metals and metalloids (Figure 2) do not contribute as much as the water-soluble species (Figure 3) to the hygroscopic properties of the aerosol, the overall hygroscopicity of particles is crucial information to predict where they deposit upon inhalation. The maximum concentrations of As, Cd, and Pb in the $0.18\text{--}0.55 \mu\text{m}$ range, which overlaps with the most hygroscopic mode at a relative humidity of 90% ($0.10\text{--}0.32 \mu\text{m}$), are of especial concern from a health perspective as this size is within the range ($\sim 0.1\text{--}1 \mu\text{m}$) most effective at penetrating the extrathoracic, conducting, and pulmonary airways of the human body upon inhalation (45–46). In terms of deposition efficiency in the respiratory tract, it is known that an absolute minimum of single-breath deposition efficiency of aerosols occurs at an aerodynamic diameter of $0.3 \mu\text{m}$ (47). This minimum arises from the competition between particle residence time and rates of settling/diffusion of particles onto exposed surfaces. The enhanced GFs that occur at high RH (Figure 3b) suggest that deposition efficiency of the most contaminant-laden particles would be higher than expected when just considering dry size alone. We note that the behavior of the growing particles in the human respiratory system might be quite complex. The residence times in various regions of the lung vary from a few milliseconds (bronchi) to a few hundred milliseconds (alveolar sac), which is similar to the characteristic time for growth of hygroscopic particles at 100% RH (48–49). It could result in the deposition of sub-micrometer particles deep within the terminal bronchi or the alveoli where they subsequently grow and deposit more efficiently than expected. Their fate would depend on the aerosol hygroscopicity and initial size distribution, and would need to be modeled in order to gain a more insightful understanding.

Aerosol particles in the size range most enriched with Pb and As (0.18–0.55 μm) exhibit the highest GF at RHs exceeding 50%, which is indicative of a highly hygroscopic aerosol with a greater potential to activate into droplets and participate in cloud formation and subsequent wet deposition. While the coarse aerosol particles deposit via dry deposition more effectively than fine aerosols, they still exhibit significant hygroscopicity, especially in the largest MOUDI stage examined ($> 18 \mu\text{m}$). Dust particles have previously been shown to become more hygroscopic with age as a result of coating by soluble species such as sulfate (e.g. 50), which allows dust particles to serve as CCN. The results of this study suggest that water-soluble species are associated with super-micrometer particles in the study region and this may be linked to the coating effect mentioned.

3.4 Regional Transport

It is of interest to examine the extent to which the contaminated aerosols near the sample site may impact distant areas. It is noted that there are other such active smelters and abandoned mine tailings in southern Arizona and these trajectory results are meant to provide a case study example as to how emissions from one such source can influence the greater region. The projected influence of aerosols originating from the study site, as predicted based on 24 h forward trajectories of air masses from the HYSPLIT model, is summarized in Figure 5 as a function of time of year. The cities of Tucson and Phoenix, with populations of over one and four million, respectively (US Census Bureau, 2009), could be influenced substantially by air parcels released from the study site. One difference among the various months is that the areal extent of forward trajectories is reduced in the summer, most likely due to the absence of prevailing westerly winds during the monsoon season (28). Independent of the HYSPLIT results, analysis of surface precipitation measurements in southern Arizona (Figure 5) also suggests that wet scavenging would limit the spatial influence of aerosols transported from the measurement site. The seasonal cycle of precipitation in southern Arizona is characterized by two modes: the first between November and March and the second and most dominant mode being during the summertime monsoon season that typically occurs between July and August. The other months show similar levels of areal influence with the spring season exhibiting slightly more spatial influence on downwind regions. These results have implications for southern Arizona aerosols (especially soil dust) that are impacted by smelting emissions, which have the ability to transport contaminants to downwind regions, where New Mexico is shown to be the most impacted adjacent state from the point source examined in this work.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.
(Left) Map of southern Arizona indicating the location of the measurement site and two other locations (Tucson and Green Valley) that are used as baseline sites for the “Enrichment Factor” analysis. (Right) Close-up of the actual measurement site near the towns of Hayden and Winkelman. The mine tailings are approximately 3 km long.

Figure 2.

(a) Size-resolved aerosol chemical measurements for one MOUDI sample set collected between 9 February 2010 and 25 February 2010, and (b) for the average of five other sets of February measurements between 2009–2011 ($n = 5$) with standard deviation bars provided.

Figure 3.

(a) Size-resolved aerosol water-soluble composition measurements (reported as mass fractions) for one MOUDI sample set collected between 9 February 2010 and 25 February 2010 near Hayden, Arizona. (b) Size-resolved hygroscopic growth factor (GF) measurements after re-aerosolizing a fraction of the same water-extracted samples examined in panel (a). Thermodynamic GF predictions for ammonium bisulfate (41–42) and experimental data for laboratory-generated oxalic acid (C₂H₂O₄) particles (43) are provided at the four RHs examined for reference in panel (b).

Figure 4.

Size-resolved enrichment factors (Eq. 1) of aerosols sampled adjacent to an active copper smelter and mine tailings. Error bars represent standard deviations. Three baseline locations are shown for comparison including the same site when the smelter was shut down, Green Valley, and Tucson (sites shown in Figure 1). The analysis is based on 59 MOUDI sample sets collected at the measurement site when the smelter was active. The data shown in this figure are from between December 2008 and April 2011. Specific sample collection dates are available in the Supporting Information.

Figure 5.

Average forward trajectory frequency plots as a function of time of year using daily data between 2005–2010 from the HYSPLIT model. (The model simulated the release of air parcels from above the surface of the mine tailings near the towns of Hayden and Winkelman, denoted by the star label in top left panel, and tracked for 24 h.) Colors indicate the minimum frequency of air parcels at a given point when originating from the source point. Locations of the two metropolitan centers of Tucson and Phoenix are shown in the top left panel. The bottom right panel shows the annual profile of daily precipitation accumulation (average of daily data for each month, including days with no rainfall) averaged over 2000–2009 at a representative location near the source point (Tucson), which independently suggests that wet scavenging of aerosols limits the transport range of particulate matter between July and August as compared to other months.