Size-Differentiated Chemical Composition of Re-Suspended Soil Dust from the Desert Southwest United States

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ABSTRACT

As part of the Desert Southwest Coarse Particulate Matter Study which characterized the composition of fine and coarse particulate matter in Pinal County, AZ during 2010-2011, several source samples were collected from several different soil types to assist in source apportionment analysis of the study results. Soil types included native desert soils, agricultural soils (crop farming), dirt-road material adjacent to agricultural areas, paved road dusts, dirt road material from within and adjacent to a cattle feedlot, and material from an active cattle feedlot. Following laboratory resuspension of the soil, size-segregated PM2.5 and PM10 fractions for each source type were collected on filters and characterized for mass, ions, OC, EC, and trace elements. While there are unique chemical composition of soils in this region (e.g., high As and Sb) that reiterate the importance of using local source profiles (e.g., native soils) as compared to Upper Continental Crust or soil profiles from other regions in receptor modeling studies, the study also provides new insights into the impact of land-use modification on source emission profiles. Specifically, high OC and PO_4^{-3} are found in material representative of local cattle feedlot activities while elevated Cu, Sb and Zn are found from sources impacted by motor vehicle traffic. Results of the study indicate that the local native soil composition is only slightly modified by agricultural activities and this study provides the chemical composition of both native soil and agricultural for source apportionment studies in the Desert Southwest.

KEY WORDS: Cattle Feedlots, Dirt Road Dust, Paved Road Dust, Agricultural Soils, Source Characterization

1. INTRODUCTION

Entrainment of crustal material can be a major source of ambient particulate matter (PM) pollution. On a local scale, large fugitive dust sources can contribute to high pollution events of both fine ($PM_{2.5}$, particles with aerodynamic diameter (AD) < 2.5 μm) and coarse (PMc or $PM_{10-2.5}$, particles in the size range between 2.5 and 10 μm AD) particles. Understanding sources of these particles is important as both size ranges have been associated with adverse health effects (Prahalad et al., 1999; Ramanathan et al., 2001; Harrison et al., 2004; Solomon et al., 2011).

Identifying and quantifying the sources of PM is an essential step in developing emissions control strategies designed to reduce levels of air pollutants to below those specified in the US National Ambient Air Quality Standards (NAAQS) (40 CFR 50, 2006) which sets a limit at an annual mean concentration of 12 $\mu g/m^3$ $PM_{2.5}$ averaged over a 3 year time frame (or a 35 $\mu g/m^3$ limit at the 98% percentile averaged over 3 years) and a limit at 150 $\mu g/m^3$ for PM_{10} which is not to be exceeded more than once per year on average over a 3 year time frame. Identifying and quantifying sources is often achieved through observational studies linking emission sources to measured ambient concentrations using receptor models or other statistical tools (Hopke, 1991; Hopke, 2003; Solomon and Hopke, 2008; Watson et al., 2008). In most cases, a detailed knowledge of source composition is needed. Most studies that seek to quantify the contribution of crustal material are limited to a small number of source samples. To improve contribution estimates from different routes of crustal material entrainment to ambient PM concentrations using source attribution approaches, it is essential to understand the chemical composition of different source materials. On a local scale, the extent of crustal material

entrainment strongly depends on soil type, land-use pattern, and wind speed (Holcombe et al., 1997; Macpherson et al., 2008).

The influence of various dust sources can be isolated only by specific chemical markers unique to different source materials, and information on local soil composition is particularly important in areas where crustal material is a substantial contributor to airborne PM. One such region is the arid desert of the southwestern United States (US), including Arizona. A recent study estimated that resuspended dust accounts for as much as 20% of $PM_{2.5}$ and 76% of PMc in the southwestern US (Malm et al., 2007). Pinal County, in central Arizona, is an area that frequently exceeds the PM_{10} NAAQS (U.S. EPA AirData, 2012). Many of these exceedances are attributed to resuspended dust from agricultural activities, unpaved and paved roads, construction, and desert lands. In addition, agricultural activities, like soil preparation, cattle farming, and movement of cattle in feedlots might contribute substantially to local high PM events. However, lack of comprehensive chemical characterization of various soil types in different particle size ranges within the study area limits the understanding of potential sources and the relative impact of each on the PM concentrations on a local scale.

This study presents the detailed chemical composition of local soils in central Arizona with the goal of contrasting chemical composition of material from different land uses to aid in source apportionment. Source profiles are developed for several soil types including soils that are potentially representative of the southwestern US. The specific objectives of the study are to characterize the chemical composition of soils associated with possible sources that contribute to ambient $PM_{2.5}$ and PM_{10} concentrations, and to determine if there are specific chemical markers to differentiate among various soil types. Size-segregated soil samples ($PM_{2.5}$ and PM_{10}) collected from twelve different locations representing 5 different land use types in Pinal County

were analyzed for detailed chemical composition allowing source profiles to be developed and compared to the composition of UCC and to undisturbed native desert soils.

2. EXPERIMENTAL

Soil Sampling

A summary of the sampling locations, local soil types, and soil classifications for this study is provided in Table 1. All soil samples were collected in Pinal County, AZ (USA) in the vicinity of three different air quality monitoring sites, including Pinal County Housing (PCH), Casa Grande (CG), and Cowtown (COW). A detailed description of the monitoring sites and local PM composition can be found elsewhere (Clements et al., 2012), with only a brief summary of the sites given here. PCH is a rural site located in immediate proximity to native (undisturbed) desert land with agricultural fields within a mile in all directions. Soil samples near PCH were collected from native desert soils (denoted native - NAT) from immediate proximity to the air quality monitoring site (less than 40 meters distant), from the edge and within the cropping area of an agricultural field to the north of the monitoring site (denoted agricultural - AGR), and from the edge of a dirt road used to access the agricultural fields in the area (denoted dirt road agricultural – DRA). The CG site is located in the small urban area of Casa Grande, AZ. Potential source samples in the vicinity of CG were collected at the edge of a local roadway representing paved road dust material (denoted paved road dust – PAV). COW is a rural monitoring location with unique local emission sources, which include a grain processing plant to the southwest of the air quality monitoring site and a confined cattle feedlot to the south of the air quality monitoring site. Samples near this location included soils from the native desert in the

immediate vicinity of the monitoring site (less than 40 meters distant) (NAT), within the active and fallowed agricultural fields to the east, north, and west of the site (AGR), the median between the monitoring site and a local two-lane highway (NAT), and the edge of the dirt roads near the cattle feedlot (denoted dirt road feedlot – DRF). Material representative of the soil found within the cattle feedlots was also collected (denoted feedlot – FDL).

Using this set of 11 fixed sampling sites, soil samples were collected during spring, fall, and winter seasons with additional samples collected during unique events (e.g., a cotton field during cotton defoliation). In total, 35 soil samples were collected. All samples were obtained from the top 15 mm of the surface using a trowel, or by a hand broom from paved roadway surface, and placed into pre-baked glass jars for storage and transport (Hagen, 2004). Figure 1 shows the location of the fixed ambient monitoring locations and the nearby locations where soil samples were collected.

Soil Resuspension

All soil samples were resuspended in the laboratory to simulate the process of windblown dust entrainment and to obtain samples representative of the composition of $PM_{2.5}$ and PM_{10} size fractionated aerosols for subsequent chemical analysis. Soil source samples were prepared for resuspension by heating to 110°C for 24 hr (similar to Carvacho et. al. (Carvacho et al., 2004)) and gently compressed to break up large soil aggregates (clumps larger than the size of a dime); care was taken to not grind the material to avoid mechanical abrasion of small particles. Once prepared, samples were introduced into a clean flask and HEPA-filtered air was passed over the sample to resuspend small particles. The resuspended particles were passed through a size-selective cyclone (URG Corporation) and collected on downstream filter media (similar to

Carcacho et. al. (Carvacho et al., 2004)). The operating flow rates were determined based on the cyclone design – 28 L/min for PM_{10} sampling and 42 L/min for $PM_{2.5}$ sampling. These flow rates were well within the range of flow rates used in other soil resuspension experiments (Carvacho et al., 2004; Etyemezian et al., 2007). Three filters, including one Teflon and two quartz-fiber filters were collected in parallel for each reentrainment experiment. A minimum target of ~8 mg of resuspended material was established to provide enough material for the planned detailed chemical analysis. Sampling was periodically stopped so the Teflon filter could be removed to determine the amount of collected material; the filter was reinstalled and sampling continued until the target mass was achieved or exceeded. On average, 20 mg of soil was collected on each filter. Depending on the crustal material studied, a varying period of time was required to reach target mass levels: road dust, for instance, required approximately 20 minutes to collect the minimum mass while some agricultural soils required upwards of 4 hours.

Soil Chemical Analysis

Re-suspended soil samples were analyzed chemically in a manner similar to ambient PM samples; details including procedures, analytical precision, and quality control measures are reported elsewhere (Clements et al., 2012). Gravimetric mass was determined by the difference between the pre- and post-collection weights of the Teflon filter. Sample weights were measured under controlled temperature (22 ºC < T < 24 ºC) and humidity conditions (45% < RH < 55%) to minimize water-uptake (U.S. EPA, 2006).

Following gravimetric analysis, the Teflon filter was microwave-digested using an acid mixture according to the method of Upadhyay, et al. (Upadhyay et al., 2009; Clements et al., 2012). The extract was then analyzed for 63 elements by high-resolution inductively coupled

plasma mass spectrometry (ICP-MS) (ThermoFinnigan ELEMENT 2) using an internal indium standard. Elements quantified included, but were not limited to, Al, As, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, Sb, Se, Sn, Ti, U, V, Zn. All elements reported were measured well above method detection limits (MDLs). The elements in $PM_{2.5}$ and PM_{10} soil samples are one order (As, Cd, Cr, Na, Ni, and Sb) to five orders (Pb) of magnitudes higher than the MDLs. The instrument was calibrated using a multi-element standard (SPEX Certiprep Inc.) and two National Institute of Standards and Technology standard reference materials (San Joaquin Soil SRM 2709 and Urban Dust SRM 1649) were also measured for quality control and element concnetrations were within 100 ± 20% of the certified values.

Water-soluble ions were measured from a set of two 1.5-cm^2 punches that were removed from one of the two quartz-fiber filters and extracted in 7.5 mL ultrapure water for 15 minutes in an ultrasonic bath at room temperature. Extracts were filtered using a syringe filter (Millex GP 0.22 μm pore size PES membrane filter) and then analyzed for cations (sodium, potassium, ammonium, calcium, and magnesium) and anions (chloride, nitrate, phosphate, and sulfate) by ion chromatograph (Dionex IC20, Dionex Corporation) equipped with CG12A and AS12A analytical columns.

Bulk organic carbon (OC), elemental carbon (EC), and total carbon (TC) concentrations were analyzed by thermal-optical transmittance (TOT) (Birch and Cary, 1996) using a slightly modified thermal protocol as described in Clements et al. (Clements et al., 2012).

3. RESULTS AND DISCUSSION

Chemical composition

Bulk chemical concentrations, including organic carbon and major mineral elements such as Al, Fe, K, etc., showed consistency within each sample type across each season, so samples were aggregated into groups according to land use type. Tables 2 and 3 present the average chemically speciated source profiles for $PM_{2.5}$ and PM_{10} for each land use type, respectively. Standard deviations (SD) represent the pooled average of the land use samples for each category. Method detection limits (MDLs) for chemical species were calculated based on 3 x SD of filter blanks and the notation BDL refers to concentrations below the MDL. Analytical uncertainties are given in Tables S1 (for $PM_{2.5}$) and S2 (for PM_{10}) for reference. The chemical profiles for each category were similar for both $PM_{2.5}$ and PM_{10} with the contribution for most chemical species within the range of the variability as measured by the SD. For all categories, Al, Ca, Fe, K, and Mg are the most abundant elemental species observed (weight percent ≥ 1%), which is consistent with the known bulk composition of crustal material. Organic carbon (OC) is also a substantial component (> 1% of PM mass) for all categories in both size fractions.

The relative abundance of OC is between 2 and 17 times higher for feedlot (FDL) samples compared to the other soil types. Sulfate and phosphate are both found in the greatest abundance in FDL samples. The elevated abundance of these particular species is consistent with nature of the cattle feeding operations and possibly is influenced by the chemical composition of the feed used in the feeding operation.

The relative abundance of OC in paved road (PAV) samples is about 2-6 times higher than in the other source categories excluding FDL. The relative abundance of elemental carbon (EC) is more than an order of magnitude higher in PAV than in samples from other categories, including native soils (NAT), dirt roads near agricultural site (DRA), and agricultural soils (AGR), where the EC values are below detection limits. Several anthropogenic elements

203 including copper, lead, and zinc are higher by factors of 3 -10 in PAV relative to other source
204 categories; the presence and abundance of these elements suggest origin from vehicular sources.
205 These results are consistent with high concentrations of these elements in ambient $PM_{2.5}$
206 collected from a parking garage in Tempe, Arizona (Majestic et al., 2009). Lough et al (Lough et
207 al., 2005) suggest that the dominance of Ba, Cu, Pb, and Zn are associated with both tail-pipe
208 emissions and mechanical abrasion of vehicle brakes and lead weights used to balance tires.
209 The relative abundance of the chemical components measured in NAT, AGR, and DRA
210 samples are, in general, similar to each other even though samples were collected from different
211 land use regions. Exceptions to this observation include sulfate and sodium ion, which are about
212 4 – 10 times higher in DRA compared to NAT and AGR samples. Although there is variability in
213 composition among the source categories, ammonium, nitrate, and chloride ions also show a
214 pattern of being an order of magnitude higher in DRA compared to NAT and AGR samples. All
215 5 of these species are components of agricultural wastes and fertilizers and have been reported as
216 markers for active agricultural farms (Cao et al., 2008). Elevated relative abundances of these
217 ions found in DRA samples, rather than AGR samples, are likely due to overspray of fertilizers,
218 the active mixing of soil by moving vehicles, deposition of windblown dust onto the road from
219 the adjacent agricultural fields, runoff from fields, and/or the lack of regular irrigation that occurs
220 on AGR during growing season. Depressed abundances of these ions in the NAT and AGR
221 samples may be the result of uptake by native or agricultural plants.
222 The relative abundances of calcium, nitrate, and sulfate were higher in samples from dirt
223 roads near the feedlot (DRF) than the other source categories; both $PM_{2.5}$ and PM_{10} size fractions
224 showed this enrichment. Calcium and sulfate species may be a result of the application of
225 calcium lignosulfonate, a by-product of wood processes, which has been used as both a dust

suppressant (Ouyang et al., 2006) and as an animal feed binder (Kaliyan and Vance Morey, 2009). Concentrations of these species, in excess of that found in the natural soils, may be distinct fingerprints for the use calcium lignosulfonate as a soil stabilizer or animal feed binder.

As previously noted, the relative abundances of Al, Ca, Fe, K, and Mg are high in all source types in both the $PM_{2.5}$ and PM_{10} size fractions. However, the lowest relative abundance of Al and Fe are found in feedlot samples due to the higher fraction of organic material in this source category (Tables 2 and 3). Of particular interest in the FDL samples, the concentrations of PO_4^{3-} and P are considerably higher than observed in other source categories. PO_4^{3-} and P abundances were 2.8% and 1.4% in $PM_{2.5}$ and 3.9% and 1.3% in PM_{10}, respectively. Higher concentrations of nutrient-related species (e.g., K, K^+, Na^+, Mg^{2+}, Cl^-, Ca^{2+}, Ca, and SO_4^{2-}) are also found in FDL samples. These results indicate that PO_4^{3-} and K^+ may be distinct markers for entrainment of material from FDL sources, although soluble K (K^+) also is a marker for wood combustion (Calloway et al., 1989; Khalil and Rasmussen, 2003). These results are consistent with feedlot profiles reported in a previous study (Chow et al., 2003). The correlation of cations to anions can be used to corroborate possible ion pairing: Cl^- in FDL is fairly well-correlated with Na^+, K^+, and NH_4^+ ($R^2 \geq 0.82$), while Mg^{2+} and PO_4^{3-} as well as K^+ and SO_4^{2-} are also well-correlated ($R^2 \geq 0.90$) indicating that these correlated components may be associated as salts.

Total versus soluble fractions of elements

Samples were extracted in water to determine the water-soluble ionic concentration which was compared to the total element concentration determined by acid digestion to determine a water-soluble fraction. The water-soluble fraction of selected elements in $PM_{2.5}$ and PM_{10} is shown in Figure 2 for each source type. The mole fraction of P in PO_4^{3-} (soluble P) was used to calculate the water-soluble fraction of phosphorus. Results show enhanced solubilityfor

Na, K, and P in FDL samples and Ca in DRF samples relative to the other source types. Greater than half of the Na is in the form of soluble salts in both size fractions for DRA, DRF, and FDL and for AGR in PM_{10}. The other components were fairly insoluble showing greater amounts of insoluble material compared to soluble material. All components were measured to be more soluble in PM_{10} than $PM_{2.5}$. The higher solubility of the selected elements in PM_{10} rather than in $PM_{2.5}$ suggests that the soluble ions associated with soil fertility are predominantly present in the coarse particle size range.

Figure 2 shows DRF samples are distinct in terms of soluble Ca with about 70% and 80% solubility in $PM_{2.5}$ and PM_{10}, respectively. This further supports the application of calcium lignosulfonate as a soil stabilizer (Ouyang et al., 2006) or animal feed binder (Kaliyan and Vance Morey, 2009) as it readily disassociates into ionic species in water. In other soil types, soluble Ca ranged between 10% to 20% in both size fractions. Higher solubility of Ca in conjunction with high SO_4^{2-} appears to be a good source signature for DRF samples. Samples from the feedlot showed that all (ratio ≈ 1) of the Na, K, and P found in these samples were soluble. Overall, results suggest that the soils from cattle feedlots are chemically different and that there are markers species, especially K^+ and PO_4^{3-}, that can be used to discriminate this soil type from the natural soil sources, although care must be taken as soluble K (K^+) is also a marker for wood combustion (Calloway et al., 1989).

Reconstructed Mass Balance

A mass balance relating gravimetrically determined mass to measured species can verify that major components have been accounted for by the targeted analytes. The mass balance summary and major components are shown in Figure 3. The *organic matter* (OM) component

was determined as 1.4 x OC (Turpin and Lim, 2001). The *crustal* component was based on the reconstructed soil mass determined by the IMPROVE (Interagency Monitoring of Protected Visual Environment) approach (Eldred, 2003; DeBell, 2006).

$$\text{Crustal} = 2.20*\text{Al} + 2.49*\text{Si} + 1.63*\text{Ca} + 2.42*\text{Fe} + 1.94*\text{Ti} \qquad (1)$$

The carbonate and water components that are sometimes included in the crustal calculation were not directly measured in the study and are excluded from the calculation of crustal material. Since Si was not analyzed by ICP-MS, its mass in the sample was estimated based on the average ratio of Si/Al = 3.5. This value was estimated based on the average Si/Al ratios of 3.8 observed in the average composition of upper continental crust (UCC) (Taylor and McLennan, 1995); 3.0 observed road dust or agricultural soil in the Imperial Valley, California, USA (Watson and Chow, 2001); and 3.5 observed in road dust in an urban area of Texas, USA (Chow et al., 2004). The *phosphate* and *sulfate* components are pure components containing just the ionic species themselves. The *non-crustal K* component was determined by difference between the measured K and the calculated crustal K where crustal K was determined as 0.6 x Fe (Malm et al., 2004). The *trace element* component was determined as the sum of all other trace metals not included in the crustal equation with no correction factors applied and ionic species not accounted for as pure components. The reconstructed mass balance showed that these components, on average, account for 102 ± 8% of resuspended crustal mass across all sites, with crustal as the single dominant component of $PM_{2.5}$ and PM_{10}.

Crustal material is the dominant contributor to all source types in both size fractions (Figure 3). It accounts for 63% and 81% of FDL and DRF, respectively and from 94-100%

across the other source types in the $PM_{2.5}$ size range. For PM_{10}, crustal material accounted for 44% of FDL and between 78-91% of the other source types. These results are consistent with findings that report soil elements are the major components of fine and coarse PM in Phoenix (Katrinak et al., 1995; Tolocka et al., 2001; Lewis et al., 2003). OM in $PM_{2.5}$ ranged from less than 5% in NAT, AGR, and DRA to 10% in PAV, to a maximum of 37% in FDL. SO_4^{2-} was a major contributor to the composition of DRF accounting for approximately 21% of the PM_{10} mass. The smaller crustal fraction in FDL samples is offset by the higher fraction of OM, SO_4^{2-}, and non-soil K, with these three components accounting for 49% PM_{10} mass. This is consistent with cattle feeding and farm operations where manure adds a significantly amount of organic material to native soils. Overall, the mass closure agrees to within 20%. These results indicate that human activities, such as motor vehicle traffic and cattle farming activities, can greatly modify the chemical characteristics of entrained crustal material from sources impacted by these activities.

Enrichment Factors

Enrichment factors (EFs) compare the abundance of a given component in a sample (e.g., soil or ambient sample) relative to that same component in a reference material to isolate unique features to potentially use as tracer species. In this case, the average trace element composition of each source type is compared to an average reference crustal material and EFs greater than 1 show enrichment of the given element, likely due to a local source. The EF of an element (X) is most commonly calculated relative to the published average composition of UCC (Taylor and McLennan, 1995) using Al or Fe as the reference element (R) (Dodd et al., 1991) where EF = $[X/R]_{sample}$ / $[X/R]_{UCC}$. The EFs of selected elements relative to UCC using Al as the reference

element are reported in Figure 4. While similar trends are observed for both $PM_{2.5}$ and PM_{10}, several unique features are observed. Numerous elements including As, Ca, Cu, Ni, P, Pb, Sb, and Zn are enriched (EF>5) in most source types with As, Cu, P, Sb, and Zn enriched by an order of magnitude or more compared to UCC. All samples, including NAT and AGR soils are enriched in As and Sbsuggesting regional influence from localemission sources and/or pollutants from long-range transport. Vehicular emissions are important sources of Cu, Pb, and Zn (Seinfeld and Pandis, 1997; Maykut et al., 2003; Utsunomiya et al., 2004; Solomon and Hopke, 2008) and could account for enrichment in PAV samples and others potentially influenced by vehicle and train traffic. Consistent with the mass abundance in the soil profiles, P has the highest EF of 75 ± 50 in $PM_{2.5}$ and 76 ± 52 in PM_{10} in the FDL samples, indicating the significant alteration of the local soil caused by the cattle feeding operations.

To more realistically account for local geochemical conditions to isolate if sources are enriched by local sources, EFs were determined based on a comparison to the composition of unaltered desert soils (NAT) obtained during this study (Figure 5). Al was again used as the reference element. The native land in the sampling area is not routinely cultivated or fertilized and was not part of a roadway, so this material is less impacted by agricultural chemicals and vehicular emissions. For most elements, there were very minimal alterations compared to EFs based on UCC composition. However, the enrichment of As and Sb in all soil types when EFs are determined relative to UCC is not observed when the EFs are determined relative to local native desert soil. This indicated the ubiquitous nature of these elements in crustal material common to this region. This observation is supported by reports of elevated As levels in soil and ground water in the southwestern US (Focazio et al., 2000). No elements are enriched in AGR relative to NAT, suggesting little anthropogenic influence of trace elements through farming

practices. Progressively, a larger number of elements are enriched in DRA, PAV, DRF, and FDL samples. Vehicular and industrial emissions appear to impact the elemental components (such as Pb, Cu, Sb, Sn, and Zn) of paved road dust samples and P enrichment is significant in FDL samples.

4. CONCLUSIONS

Samples collected from crustal material representing different land uses in Pinal County, AZ were resuspended in the laboratory, including material representing native desert (NAT), agricultural crop farming (AGR), dirt roads adjacent to agricultural areas (DRA), paved roads (PAV), dirt road within and adjacent to a cattle feedlot (DRF), and a cattle feedlot (FDL). Following resuspension of the material in the laboratory, size-segregated $PM_{2.5}$ and PM_{10} fractions for each source type were collected on filters and characterized for mass, ions, OC, EC, and trace elements. Results showed that the chemical abundances for the majority of species are similar between source category between $PM_{2.5}$ and PM_{10} samples. Elements common to crustal sources (Al, Ca, Fe, and Mg,), K, and OC are abundant in all soil types (mass percent by weight ≥1%), Ca and Ca^{+2}, and SO_4^{2-} are most abundant in DRF, and OC and PO_4^{3-} are most abundant in FDL. Data suggest that soluble phosphate is a possible unique marker for entrainment of crustal material from cattle feedlots. Calcium and sulfate present in some dirt road samples may be related to calcium lignosulfonate used as a dust suppressant or animal feed binder.

Vehicular movement and wind likely help mix agricultural soil with dirt roads within the vicinity of agricultural fields (DRA). This mixing and overspray of fertilizers likely results in an increase in the relative abundance of NH_4^+, SO_4^{2-}, NO_3^-, Cl^-, and Na, which are an order of

magnitude more abundant in DRA when compared to NAT and AGR samples. Decreased abundances of these elements in the fallow and crop lands are suggestive of removal from the soils by plant uptake. Fugitive dust emissions from the undisturbed agricultural soil may therefore contain lower concentrations of these species than the dust released during the land preparation. The abundance of Cu, Pb, and Zn are an order of magnitude higher in PAV compared to other source types, consistent with motor vehicle sources. Mass balance analysis showed that the crustal component comprises most of the particle mass in both $PM_{2.5}$ and PM_{10}.

The unique chemical composition of soils in this region (e.g., high As and Sb) shows the importance of using local soil profiles (e.g., native soils) as compared to UCC or soil profiles from other regions in receptor modeling studies. Failure to use a region specific source soil profile in an apportionment study may lead to inappropriate apportionment of crustal particles to other sources. In this case, elevated arsenic concentrations in the native soil could have been mischaracterized as anthropogenic rather than natural which might have lead to apportionment of the aerosol to a smelting source not present or contributing to PM concentrations in this airshed. Source material composition also indicates the impact of local sources modifying the soil composition from NAT. For example, high OC and PO_4^{3-} were associated with the cattle feedlot and Cu, Sb, and Zn were associated with paved roads.

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List of Table Titles

Table 1. Source Sampling Details

Site Number	Season	Closest Monitor	Sampling Location	Soil type	Sample Category	Classification
1	Spring	PCH	20 Meters SW of Monitoring Site	Fine Sandy Loam	Native	NAT
1	Fall	PCH	20 Meters SW of Monitoring Site	Fine Sandy Loam	Native	NAT
1	Winter	PCH	20 Meters SW of Monitoring Site	Fine Sandy Loam	Native	NAT
2	Spring	PCH	40 Meters SW of Monitoring Site	Fine Sandy Loam	Native	NAT
2	Fall	PCH	40 Meters SW of Monitoring Site	Fine Sandy Loam	Native	NAT
2	Winter	PCH	40 Meters SW of Monitoring Site	Fine Sandy Loam	Native	NAT
3	Spring	COW	20 Meters SW of Monitoring Site	Clay Loam	Native	NAT
3	Fall	COW	20 Meters SW of Monitoring Site	Clay Loam	Native	NAT
3	Winter	COW	20 Meters SW of Monitoring Site	Clay Loam	Native	NAT
4	Winter	COW	Median between Site and Highway	Clay Loam	Native	NAT
5	Spring	COW	East Alfalfa Field	Clay Loam	Agricultural	AGR
5	Fall	COW	East Alfalfa Field	Clay Loam	Agricultural	AGR
5	Winter	COW	East Alfalfa Field	Clay Loam	Agricultural	AGR
6	Spring	COW	West Alfalfa Field	Clay Loam	Agricultural	AGR
6	Fall	COW	West Alfalfa Field	Clay Loam	Agricultural	AGR
7	Spring	PCH	Winter Wheat Field - Edge	Fine Sandy Loam	Agricultural	AGR
7	Spring	PCH	Winter Wheat Field - Center	Fine Sandy Loam	Agricultural	AGR
7	Fall	PCH	Winter Wheat Field - Center	Fine Sandy Loam	Agricultural	AGR
7	Winter	PCH	Winter Wheat Field - Center - Cut	Fine Sandy Loam	Agricultural	AGR
8	Fall	PCH	Defoliated Cotton Field	Fine Sandy Loam	Agricultural	AGR
8	Winter	PCH	Fallow Cotton Field	Fine Sandy Loam	Agricultural	AGR
9	Spring	PCH	Dirt Road Dust - South Edge	Fine Sandy Loam	Dirt Road - Ag	DRA
9	Fall	PCH	Dirt Road Dust - South Edge	Fine Sandy Loam	Dirt Road - Ag	DRA
9	Winter	PCH	Dirt Road Dust - South Edge	Fine Sandy Loam	Dirt Road - Ag	DRA
10	Spring	PCH	Dirt Road Dust - North Edge	Fine Sandy Loam	Dirt Road - Ag	DRA
10	Fall	PCH	Dirt Road Dust - North Edge	Fine Sandy Loam	Dirt Road - Ag	DRA
11	Winter	CG	Paved Road - Edge Composite	Fine Sandy Loam	Paved Road	PAV
12	Spring	COW	Dirt Road Dust - Near Feedlot	Clay Loam	Dirt Road - Feed	DRF
12	Fall	COW	Dirt Road Dust - Near Feedlot	Clay Loam	Dirt Road - Feed	DRF
13	Spring	COW	Feedlot Material	Clay Loam	Feedlot	FDL
13	Fall	COW	Feedlot Material	Clay Loam	Feedlot	FDL
13	Fall	COW	Feedlot Material	Clay Loam	Feedlot	FDL
13	Winter	COW	Feedlot Material	Clay Loam	Feedlot	FDL
14	Winter	COW	Empty Feedlot Material	Clay Loam	Empty feedlot	FDL
15	Winter	COW	Old Feedlot Surface Material	Clay Loam	Empty feedlot	FDL

PCH - Pinal County Housing; COW - Cowtown; CG - Casa Grande

NAT - Native Soil; AGR - Agricultural Soil; PAV - Paved Road Dust; DRA - Dirt (Unpaved) Road Dust from an Agricultural Area; DRF - Dirt (Unpaved) Road Dust from a Cattle Feedlot Area; FDL - Soil from a Cattle Feedlot

Table 2. Average (±SD) of source profile chemical compositions (weight percent by mass) of resuspended $PM_{2.5}$ material[a]

Species	Soil Classification				
	NAT	AGR	DRA	PAV[b]	DRF
OC	1.2 ± 0.7	3 ± 2	1.7 ± 0.5	7	2.8 ±
EC	BDL	BDL	BDL	0.2	0.002 ±
TC	1.2 ± 0.7	3 ± 2	1.7 ± 0.5	7	2.8 ±
Cl^-	0.02 ± 0.03	0.04 ± 0.03	0.2 ± 0.2	0.08	0.5 ±
NO_3^-	0.05 ± 0.09	0.03 ± 0.02	0.2 ± 0.2	0.05	0.8 ±
PO_4^{3-}	0.06 ± 0.03	0.09 ± 0.06	0.02 ± 0.02	0.15	0.10 ±

SO_4^{2-}	0.03	±	0.04	0.1	±	0.2	0.6	±	0.2	0.2	17	±
Na^+	0.09	±	0.05	0.20	±	0.08	0.8	±	0.2	0.2	0.46	±
NH_4^+	0.06	±	0.05	0.12	±	0.10	0.5	±	0.5	0.02	0.2	±
K^+	0.11	±	0.03	0.14	±	0.07	0.07	±	0.02	0.07	0.369	±
Mg^{2+}	0.02	±	0.01	0.04	±	0.02	0.02	±	0.01	0.02	0.09	±
Ca^{2+}	0.4	±	0.2	0.7	±	0.5	0.5	±	0.2	0.6	6	±
Al	8	±	1	7	±	3	7	±	1	7	5	±
As	0.0012	±	0.0003	0.0014	±	0.0003	0.0014	±	0.0002	0.002	0.001	±
Ba	0.054	±	0.004	0.05	±	0.02	0.05	±	0.01	0.06	0.04	±
Ca	2.2	±	0.3	4	±	2	6	±	1	3	10	±
Cd	0.0001	±	0	0.0001	±	0	0	±	0	0.0002	0.00003	±
Co	0.002	±	0	0.002	±	0.001	0.0016	±	0.0002	0.002	0.0010	±
Cr	0.005	±	0.001	0.004	±	0.001	0.005	±	0.000	0.007	0.003	±
Cs	0.0016	±	0.0003	0.002	±	0.001	0.0011	±	0.0002	0.001	0.0011	±
Cu	0.008	±	0.002	0.010	±	0.005	0.0049	±	0.0004	0.03	0.007	±
Fe	4.0	±	0.2	3.7	±	1.0	3.7	±	0.4	4	2.5	±
Ga	0.0022	±	0.0002	0.002	±	0.001	0.0017	±	0.0002	0.002	0.0015	±
K	3.6	±	0.7	3.0	±	0.5	3.2	±	0.9	2	2.8	±
Mg	1.9	±	0.4	1.6	±	0.5	2.1	±	0.5	2	1.2	±
Mn	0.15	±	0.01	0.13	±	0.04	0.12	±	0.02	0.09	0.08	±
Na	0.31	±	0.03	0.4	±	0.1	1.2	±	0.2	0.6	0.56	±
Ni	0.004	±	0.001	0.004	±	0.001	0.003	±	0	0.005	0.002	±
P	0.12	±	0.01	0.13	±	0.04	0.11	±	0.01	0.2	0.26	±
Pb	0.007	±	0.002	0.006	±	0.002	0.003	±	0	0.02	0.004	±
Rb	0.013	±	0.002	0.011	±	0.004	0.012	±	0.002	0.01	0.008	±
Sb	0.0002	±	0.0001	0.0002	±	0.0001	0.00015	±	0.00001	0.001	0.0002	±
Sr	0.019	±	0.002	0.03	±	0.01	0.045	±	0.005	0.02	0.041	±
Th	0.0017	±	0.0003	0.002	±	0.001	0.0015	±	0.0003	0.002	0.0014	±
Ti	0.30	±	0.04	0.27	±	0.09	0.27	±	0.05	0.3	0.17	±
V	0.0081	±	0.0003	0.008	±	0.002	0.007	±	0.001	0.008	0.006	±
Zn	0.020	±	0.002	0.020	±	0.007	0.014	±	0.001	0.1	0.018	±

[a]SD was calculated from the average of the aggregated samples from each site
[b]No SD is included for PAV because only one sample was collected
BDL - below detection limit as defined in the text

Table 3. Average (±SD) of source profile chemical compositions (weight percent by mass) of resuspended PM_{10} material[a]

	Soil Classification				
Species	NAT	AGR	DRA	PAV[b]	DRF
OC	1.3 ± 0.4	2 ± 1	2.0 ± 0.5	8	3.9 ± 0.
EC	BDL	BDL	BDL	0.2	0.02 ± 0.
TC	1.3 ± 0.4	2 ± 1	2.0 ± 0.5	8	3.9 ± 0.
Cl^-	0.02 ± 0.02	0.01 ± 0.01	0.2 ± 0.2	0.11	0.3 ± 0.
NO_3^-	0.07 ± 0.12	0.03 ± 0.02	0.2 ± 0.2	0.04	1 ± 2
PO_4^{3-}	0.07 ± 0.03	0.1 ± 0.1	0.03 ± 0.03	0.18	0.12 ± 0.
SO_4^{2-}	0.04 ± 0.05	0.1 ± 0.1	0.8 ± 0.2	0.2	24 ± 2
Na^+	0.09 ± 0.04	0.2 ± 0.2	0.8 ± 0.2	0.2	0.61 ± 0.
NH_4^+	0.05 ± 0.04	0.1 ± 0.1	0.5 ± 0.4	0.02	0.3 ± 0.
K^+	0.10 ± 0.02	0.12 ± 0.06	0.06 ± 0.00	0.07	0.51 ± 0.
Mg^{2+}	0.02 ± 0.01	0.03 ± 0.02	0.011 ± 0.004	0.02	0.09 ± 0.
Ca^{2+}	0.4 ± 0.2	0.6 ± 0.4	0.4 ± 0.3	0.7	8 ± 8
Al	7 ± 1	7 ± 1	5 ± 2	7	5 ± 2
As	0.0011 ± 0.0004	0.0014 ± 0.0003	0.0013 ± 0.0002	0.002	0.0013 ± 0.
Ba	0.049 ± 0.007	0.044 ± 0.007	0.046 ± 0.003	0.06	0.04 ± 0.
Ca	2.2 ± 0.5	4 ± 3	5.8 ± 1.0	3	9 ± 3
Cd	0.00006 ± 0.00002	0.00006 ± 0.00001	0.00004 ± 0.00001	0.0002	0.00004 ± 0.
Co	0.0016 ± 0.0001	0.0015 ± 0.0002	0.0014 ± 0.0001	0.001	0.0010 ± 0.
Cr	0.004 ± 0.001	0.0035 ± 0.0005	0.0044 ± 0.0001	0.007	0.003 ± 0.
Cs	0.0014 ± 0.0003	0.0015 ± 0.0003	0.0010 ± 0.0002	0.001	0.0012 ± 0.
Cu	0.007 ± 0.002	0.008 ± 0.001	0.0045 ± 0.0004	0.03	0.006 ± 0.
Fe	3.7 ± 0.4	3.3 ± 0.5	3.4 ± 0.2	4	2.4 ± 0.
Ga	0.0019 ± 0.0002	0.0018 ± 0.0003	0.0016 ± 0.0001	0.002	0.0014 ± 0.
K	3.2 ± 0.8	2.9 ± 0.6	3.0 ± 0.6	2	2.8 ± 0.
Mg	1.7 ± 0.5	1.5 ± 0.3	1.8 ± 0.6	1	1.1 ± 0.
Mn	0.13 ± 0.02	0.11 ± 0.02	0.11 ± 0.01	0.09	0.07 ± 0.
Na	0.31 ± 0.03	0.36 ± 0.08	1.2 ± 0.3	0.7	0.62 ± 0.
Ni	0.004 ± 0.003	0.0029 ± 0.0003	0.003 ± 0	0.005	0.0022 ± 0.
P	0.11 ± 0.02	0.11 ± 0.02	0.10 ± 0.01	0.2	0.26 ± 0.
Pb	0.006 ± 0.002	0.005 ± 0.001	0.0032 ± 0.0003	0.01	0.004 ± 0.
Rb	0.012 ± 0.003	0.010 ± 0.003	0.010 ± 0.001	0.01	0.009 ± 0.
Sb	0.0002 ± 0	0.0002 ± 0.0001	0.0001 ± 0	0.001	0.0002 ± 0

Sr	0.018	±	0.003	0.03	±	0.01	0.046	±	0.006	0.03	0.039	±	0
Th	0.002	±	0	0.0015	±	0.0004	0.0012	±	0.0004	0.002	0.001	±	0.
Ti	0.28	±	0.05	0.24	±	0.04	0.26	±	0.02	0.3	0.19	±	0.
V	0.0075	±	0.0005	0.007	±	0.001	0.0070	±	0.0004	0.007	0.005	±	0.
Zn	0.018	±	0.003	0.016	±	0.003	0.014	±	0.003	0.1	0.018	±	0.

[a]SD was calculated from the average of the aggregated samples from each site
[b]No SD is included for PAV because only one sample was collected
BDL - below detection limit as defined in the text

Table S1. Average analytical uncertainty of source profile chemical components (weight percent by mass of resuspended $PM_{2.5}$ material)

	Soil Classification					
Species	**NAT**	**AGR**	**DRA**	**PAV**	**DRF**	**FDL**
OC	0.1	0.2	0.1	0.4	0.2	1.1
EC	0.0	0.03	0.02	0.03	0.02	0.08
TC	0.1	0.2	0.1	0.4	0.2	1.1
Cl^-	0.007	0.007	0.007	0.007	0.007	0.007
NO_3^-	0.007	0.007	0.007	0.007	0.007	0.007
PO_4^{2-}	0.006	0.006	0.006	0.006	0.006	0.1
SO_4^{2-}	0.006	0.006	0.006	0.006	6	0.1
Na^+	0.001	0.001	0.001	0.001	0.001	0.001
NH_4^+	0.004	0.004	0.004	0.004	0.004	0.004
K^+	0.005	0.005	0.005	0.005	0.005	0.005
Mg^{2+}	0.002	0.002	0.002	0.002	0.002	0.002
Ca^{2+}	0.003	0.003	0.003	0.003	0.003	0.003
Al	0.3	0.4	0.4	0.5	0.2	0.2
As	0.0003	0.0004	0.0003	0.0003	0.0002	0.0004
Ba	0.002	0.002	0.002	0.003	0.001	0.002
Ca	0.2	0.3	0.3	0.1	0.7	0.4
Cd	0.00001	0.00001	0.00001	0.00002	0.00001	0.00001
Co	0.0001	0.0002	0.0001	0.0002	0.0001	0.0002
Cr	0.0002	0.0003	0.0003	0.0003	0.0002	0.0003
Cs	0.00003	0.00005	0.00004	0.00011	0.00004	0.00005
Cu	0.0003	0.0006	0.0004	0.0005	0.0004	0.0007
Fe	0.1	0.1	0.1	0.01	0.1	0.04
Ga	0.0001	0.0002	0.0001	0.0003	0.0001	0.0002
K	0.1	0.1	0.1	0.2	0.1	0.3
Mg	0.1	0.1	0.1	0.1	0.1	0.1
Mn	0.003	0.005	0.004	0.002	0.002	0.002
Na	0.02	0.03	0.05	0.03	0.05	0.07
Ni	0.0003	0.0006	0.0004	0.0001	0.0004	0.0006
P	0.003	0.005	0.008	0.003	0.006	0.053
Pb	0.0002	0.0003	0.0002	0.0011	0.0002	0.0002
Rb	0.001	0.001	0.001	0.001	0.001	0.001
Sb	0.00001	0.00002	0.00001	0.00005	0.00001	0.00002
Sr	0.0004	0.0005	0.0012	0.0003	0.0008	0.0015
Th	0.00005	0.00006	0.00006	0.00003	0.00005	0.00003
Ti	0.006	0.012	0.007	0.008	0.005	0.008
V	0.0003	0.0005	0.0003	0.0008	0.0003	0.0003
Zn	0.001	0.001	0.001	0.008	0.001	0.006

Table S2. Average analytical uncertainty of source profile chemical components (weight percent by mass of resuspended PM_{10} material)

	Soil Classification					
Species	**NAT**	**AGR**	**DRA**	**PAV**	**DRF**	**FDL**
OC	0.1	0.2	0.1	0.4	0.2	1.2
EC	0.0	0.03	0.02	0.04	0.02	0.07
TC	0.1	0.2	0.1	0.4	0.2	1.2
Cl^-	0.007	0.007	0.007	0.007	0.1	0.007
NO_3^-	0.007	0.007	0.007	0.007	0.007	0.007
PO_4^{2-}	0.006	0.006	0.006	0.006	0.006	0.1
SO_4^{2-}	0.006	0.006	0.006	0.006	8	0.1
Na^+	0.001	0.001	0.001	0.001	0.001	0.001
NH_4^+	0.004	0.004	0.004	0.004	0.004	0.004
K^+	0.005	0.005	0.005	0.005	0.005	0.005
Mg^{2+}	0.002	0.002	0.002	0.002	0.002	0.002
Ca^{2+}	0.003	0.003	0.003	0.003	0.1	0.003
Al	0.4	0.4	0.4	0.7	0.5	0.2
As	0.0003	0.0005	0.0002	0.0001	0.0002	0.0003
Ba	0.002	0.002	0.002	0.002	0.002	0.001
Ca	0.1	0.2	0.3	0.0	0.5	0.2
Cd	0.00001	0.00001	0.00001	0.00002	0.00001	0.00001
Co	0.0001	0.0002	0.0001	0.0001	0.0001	0.0002
Cr	0.0002	0.0003	0.0003	0.0005	0.0002	0.0004
Cs	0.00005	0.00006	0.00004	0.00005	0.00005	0.00004
Cu	0.0003	0.0004	0.0003	0.0002	0.0003	0.0005
Fe	0.1	0.1	0.1	0.20	0.2	0.04
Ga	0.0001	0.0002	0.0002	0.0002	0.0001	0.0001
K	0.1	0.1	0.1	0.2	0.2	0.3
Mg	0.1	0.1	0.1	0.2	0.1	0.1
Mn	0.004	0.004	0.005	0.005	0.002	0.002
Na	0.02	0.02	0.06	0.07	0.04	0.04
Ni	0.0005	0.0003	0.0003	0.0011	0.0003	0.0006
P	0.005	0.006	0.006	0.009	0.008	0.072
Pb	0.0003	0.0003	0.0001	0.0009	0.0002	0.0002
Rb	0.001	0.001	0.001	0.000	0.001	0.001
Sb	0.00001	0.00002	0.00001	0.00007	0.00001	0.00001
Sr	0.0004	0.0008	0.0012	0.0011	0.0010	0.0007
Th	0.00005	0.00006	0.00004	0.00007	0.00004	0.00005
Ti	0.010	0.010	0.015	0.016	0.005	0.006
V	0.0003	0.0003	0.0003	0.0005	0.0002	0.0003
Zn	0.001	0.001	0.001	0.002	0.001	0.002

List of Figure Captions

Figure 1. Depiction of the ambient sampling locations and the associated soil sampling locations in close proximity.

Figure 2. Soluble fraction of selected elements in $PM_{2.5}$ and PM_{10} across six sampling sites. The mole fraction of P in PO_4^{3-} (sol P) was used to calculate the water-soluble fraction of phosphorus.

Figure 3. Mass balance for resuspended soil collected as $PM_{2.5}$ and PM_{10}. Error bars represent the total propagated error calculated from the standard deviation of samples within each soil type for each chemical component. The definition of each category can be found under the heading Reconstructed Mass Balance. The category titles are given in italics.

Figure 4. Enrichment factors for elements in $PM_{2.5}$ and PM_{10} resuspended soil samples relative to average composition of upper continental crust. Ba, Ce, Fe, Rb, Th, Ti, V, and Y have EFs between 1 and 2 and are not included in the plot. The horizontal green line denotes an EF of 1 while boxed numbers indicate the EFs for elements that go off-scale.

Figure 5. Enrichment factors for elements in $PM_{2.5}$ and PM_{10} resuspended soil samples relative to native soil collected at PCH and COW. Ba, Ce, Fe, Rb, Th, Ti, V, and Y have EFs between 1 and 2 and are not included in the plot. The horizontal green line denotes an EF of 1 while boxed numbers indicate the EFs for elements that go off-scale.

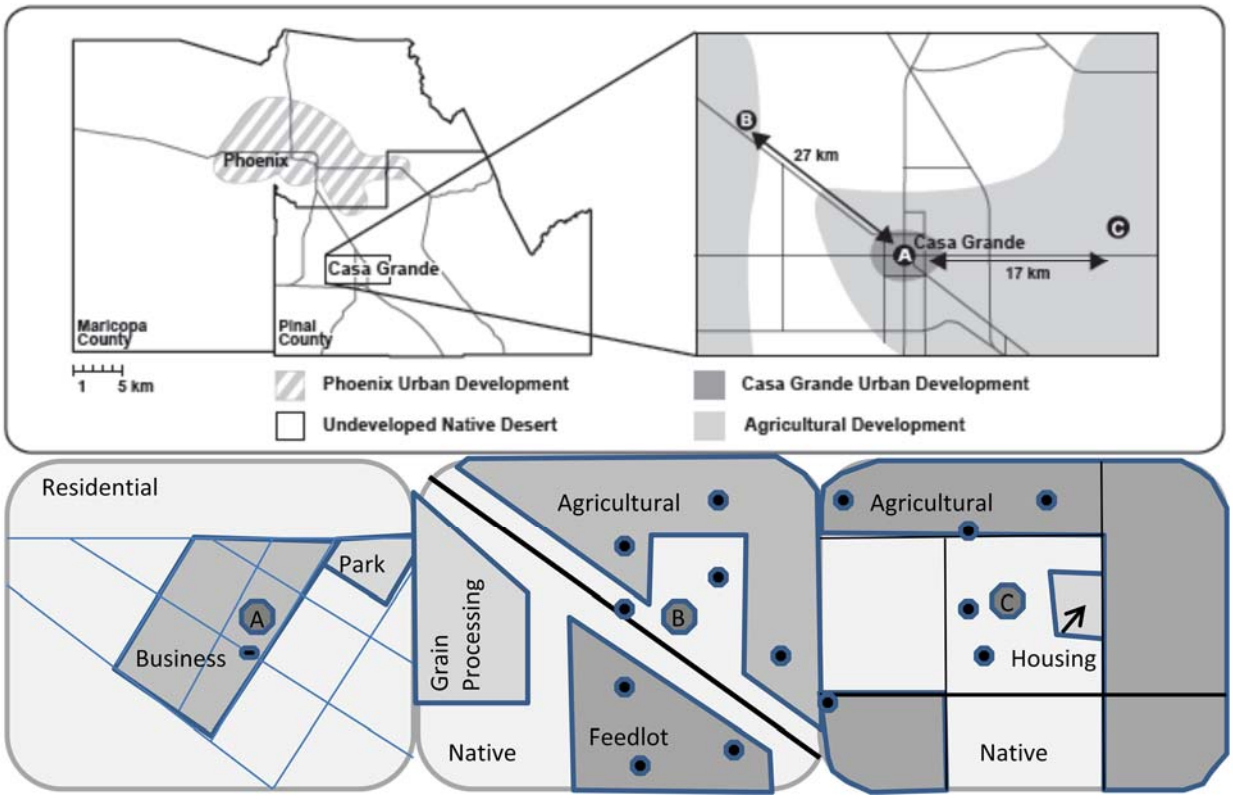
Phoenix
Casa Grande
Maricopa County
Pinal County
1 5 km
Phoenix Urban Development
Undeveloped Native Desert
Casa Grande Urban Development
Agricultural Development
B
27 km
Casa Grande
A
C
17 km
Residential
Park
A
Business
Grain Processing
Agricultural
B
Feedlot
Native
Agricultural
C
Housing
Native

Figure 1

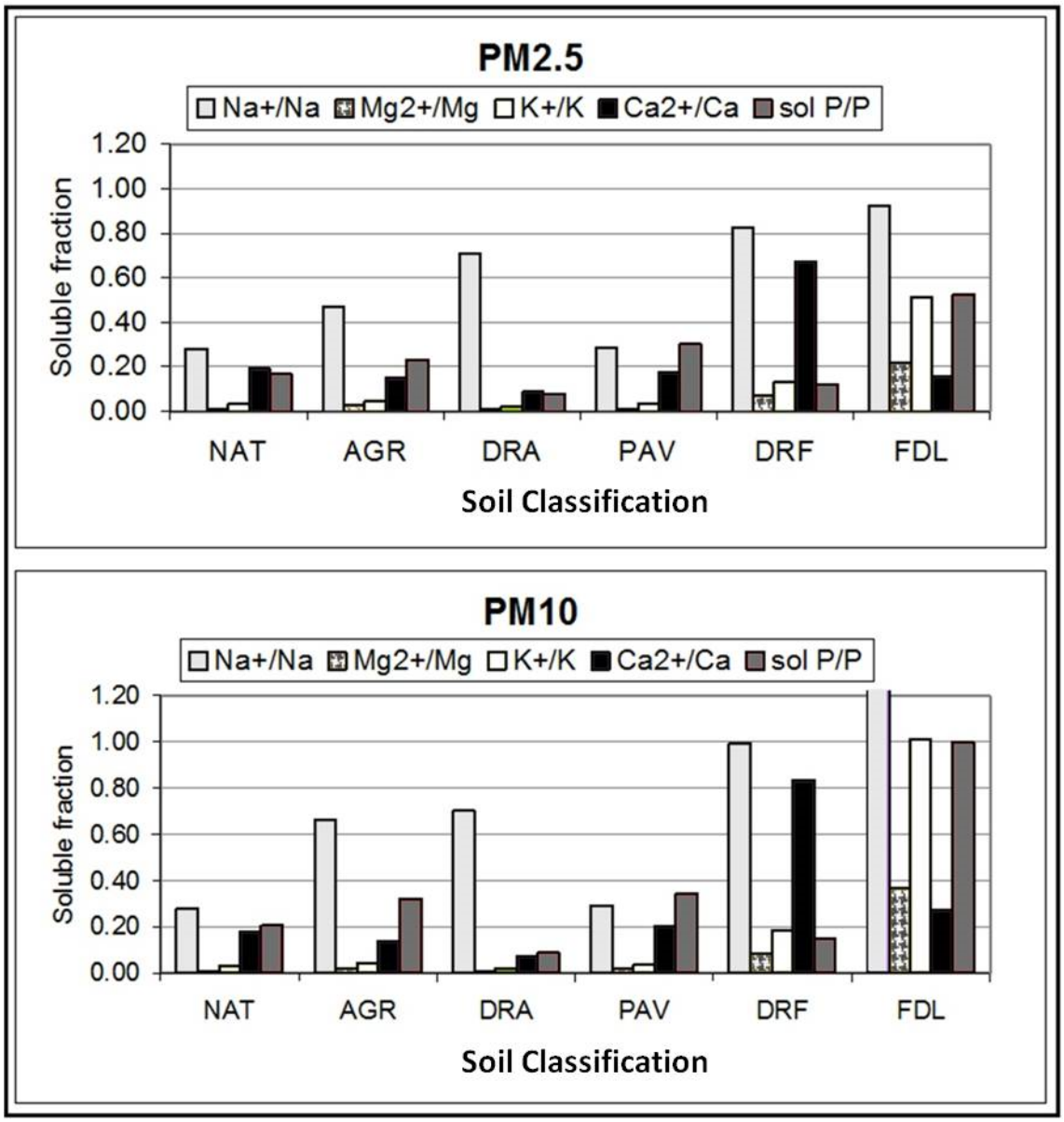
PM2.5
Na+/Na
Mg2+/Mg
K+/K
Ca2+/Ca
sol P/P
Soluble fraction
1.20
1.00
0.80
0.60
0.40
0.20
0.00
NAT
AGR
DRA
PAV
DRF
FDL
Soil Classification
PM10
Na+/Na
Mg2+/Mg
K+/K
Ca2+/Ca
sol P/P
Soluble fraction
1.20
1.00
0.80
0.60
0.40
0.20
0.00
NAT
AGR
DRA
PAV
DRF
FDL
Soil Classification

Figure 2

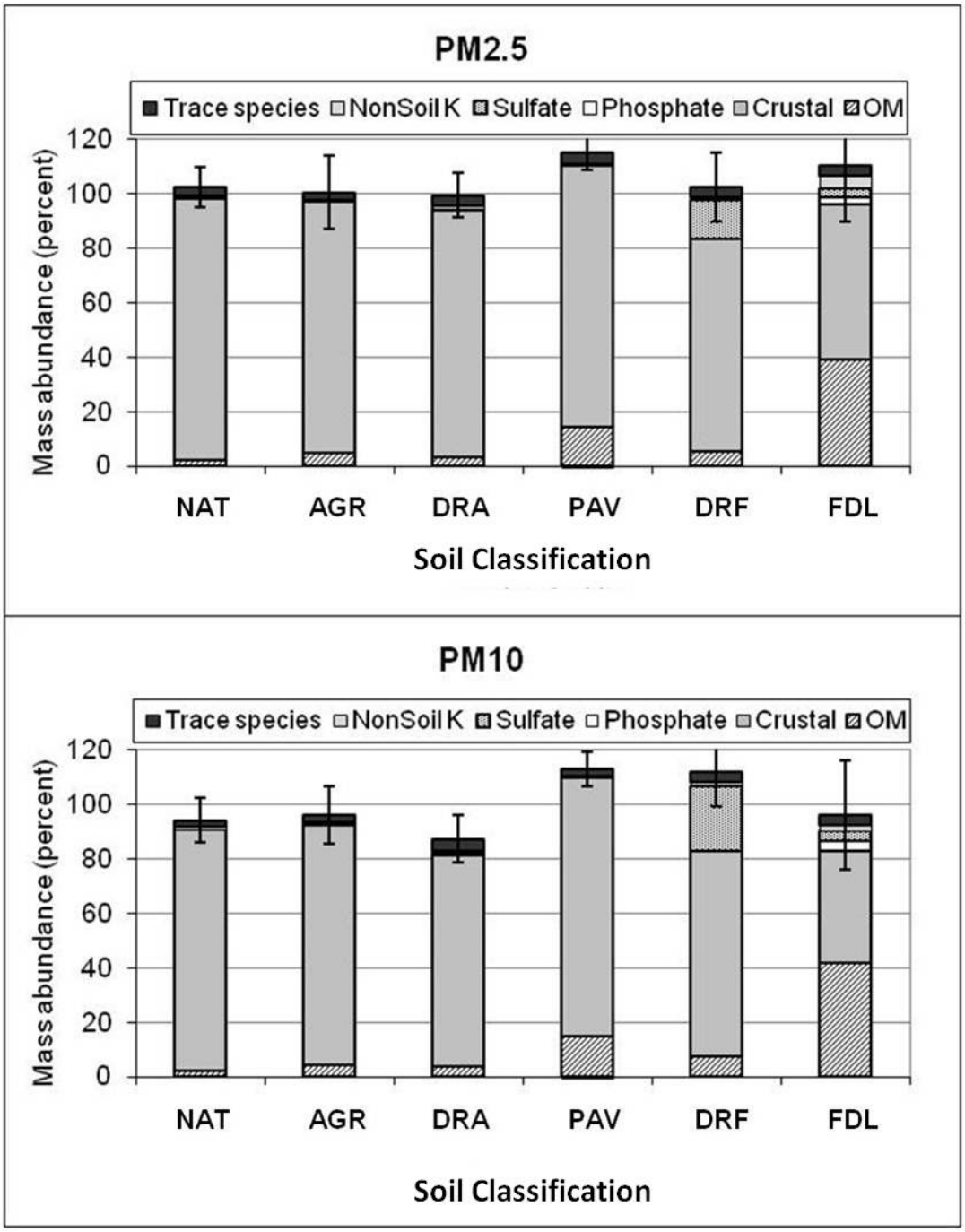
PM2.5
Trace species
NonSoil K
Sulfate
Phosphate
Crustal
OM
Mass abundance (percent)
120
100
80
60
40
20
0
NAT
AGR
DRA
PAV
DRF
FDL
Soil Classification
PM10
Trace species
NonSoil K
Sulfate
Phosphate
Crustal
OM
Mass abundance (percent)
120
100
80
60
40
20
0
NAT
AGR
DRA
PAV
DRF
FDL
Soil Classification

Figure 3

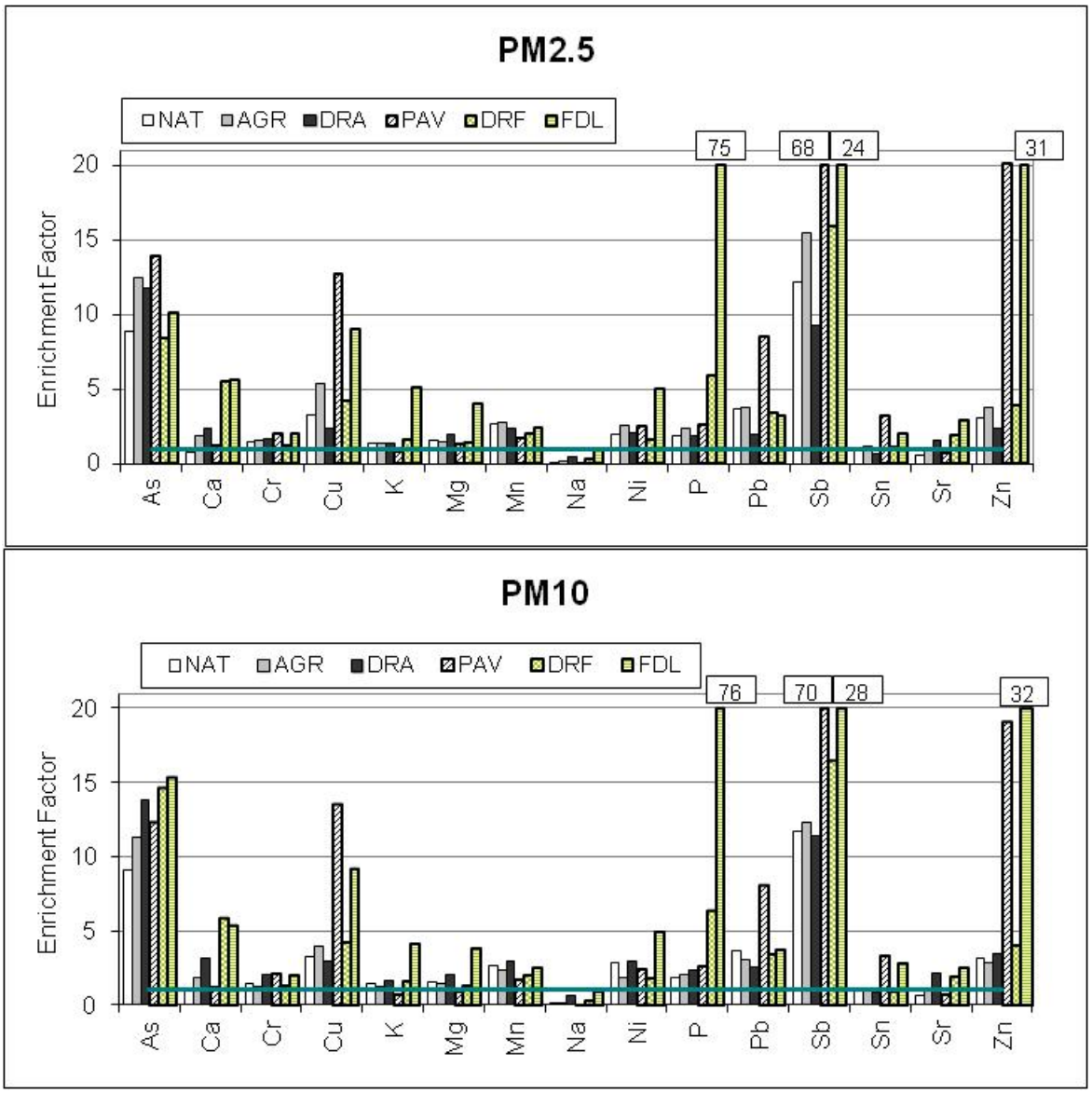
PM2.5
NAT AGR DRA PAV DRF FDL
Enrichment Factor
0
5
10
15
20
75
68
24
31
As Ca Cr Cu K Mg Mn Na Ni P Pb Sb Sn Sr Zn
PM10
NAT AGR DRA PAV DRF FDL
Enrichment Factor
0
5
10
15
20
76
70
28
32
As Ca Cr Cu K Mg Mn Na Ni P Pb Sb Sn Sr Zn

Figure 4

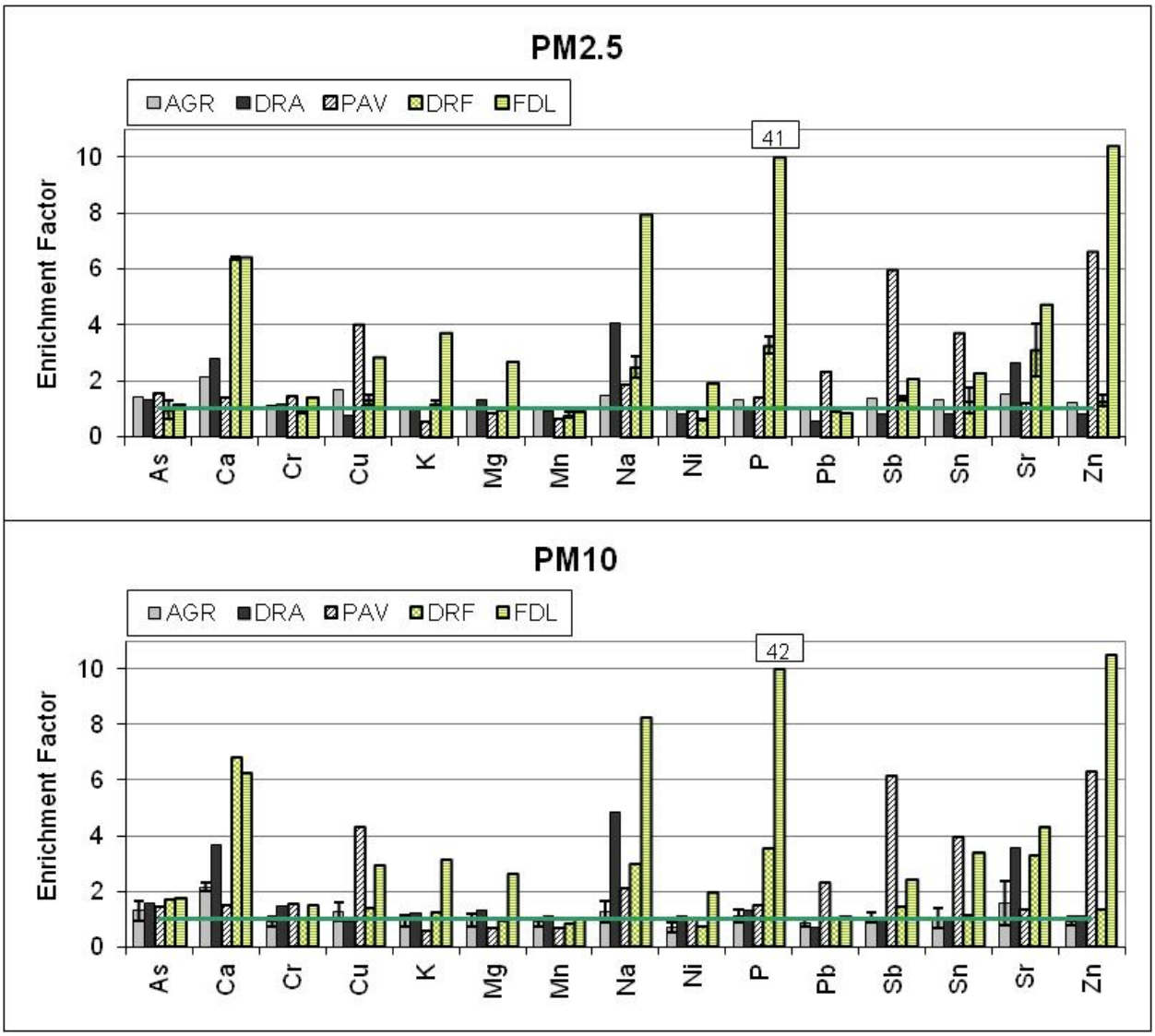
PM2.5
AGR
DRA
PAV
DRF
FDL
41
Enrichment Factor
0
2
4
6
8
10
As
Ca
Cr
Cu
K
Mg
Mn
Na
Ni
P
Pb
Sb
Sn
Sr
Zn
PM10
AGR
DRA
PAV
DRF
FDL
42
Enrichment Factor
0
2
4
6
8
10
As
Ca
Cr
Cu
K
Mg
Mn
Na
Ni
P
Pb
Sb
Sn
Sr
Zn

Figure 5