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AMBIENT PM₁₀ AND METAL CONCENTRATIONS MEASURED IN THE SUNNYSIDE UNIFIED SCHOOL DISTRICT, TUCSON, ARIZONA

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ABSTRACT

The purpose of this study was to measure the airborne concentrations of PM₁₀ and eight metalloids and metals (arsenic, beryllium, cadmium, chromium, cobalt, lead, manganese, and nickel) in the southern Tucson metropolitan area, and to evaluate possible stationary and mobile sources. Over 200 quartz filters from a PM₁₀ network located at six schools in the Sunnyside Unified School District were analyzed. The network was established primarily to detect any airborne beryllium in the vicinity of a ceramics-processing facility, and we were able to analyze the samples for additional metals, at little additional cost. The results show that the study area has good air quality with respect to PM₁₀ and metals, with ambient concentrations meeting US Environmental Protection Agency and World Health Organization standards. Beryllium was detected only once (during a dust storm) and was ascribed to natural-occurring beryllium in the suspended soil. This study was not able to apportion those few metals that were detected to any particular source or sources.

INTRODUCTION

The purpose of this study was to measure the ambient concentrations of PM₁₀ and eight metalloids and metals (arsenic [As], beryllium [Be], cadmium [Cd], chromium [Cr], cobalt [Co], lead [Pb], manganese [Mn], and nickel [Ni]) in southern metropolitan Tucson, and to identify possible natural and anthropogenic sources. The concentration of airborne metals in Tucson has not been previously documented in the peer-reviewed literature.

The work has its origins in an ambient beryllium monitoring network. In cooperation with Brush Ceramic Products (BCP), Pima County Department of Environmental Quality (PDEQ) established a PM₁₀ monitoring network to detect airborne particulate beryllium in the Sunnyside Unified School District where a factory processes beryllium oxide. The network, operating over a 5-year period (2007-2012), is intended to establish beryllium background levels and trends. There is no ambient beryllium standard in the United States, although beryllium and beryllium oxide are toxic, and therefore the network was not designed for source apportionment or compliance verification studies.

Working together, the Sonora Environmental Research Institute, Inc. (SERI), the PDEQ, the University of Arizona, and the Sunnyside Unified School District (SUSD), collected and analyzed the beryllium network quartz filters for seven additional metals. The expanded metal study is the subject of this paper and covers the period July 2007 to December 2009.

There are many potential sources of airborne metals. Beryllium, cadmium, chromium, cobalt, lead, manganese, and nickel are all found naturally

in the soil and so could originate from windblown dust (Bohn et al. 2001). Vehicle traffic is a potential source of cadmium, lead, manganese, and nickel as a result of fuel combustion and the wearing of brakes, tires, and other components (Lough et al. 2005, Johansson et. al. 2009). Emissions from coal burning power plants are a potential source for arsenic, cadmium, chromium, lead, and nickel (Gaffney and Marley 2009) while local ceramic and metal-processing facilities could emit a wide variety of metals including beryllium.

EXPERIMENTAL SECTION

Study Area

A PM₁₀ network was established in 2007 to detect possible beryllium oxide emissions from a local ceramics manufacturing facility, Brush Ceramic Products (PDEQ 2006b). Later, the number of species analyzed in PM₁₀ was temporarily increased to include arsenic, cadmium, chromium, cobalt, lead, manganese, and nickel to better assess air quality in this light industrial sector of Tucson. The network (Fig. 1) is located in the south of the metropolitan area, a few kilometers south from Tucson Electric Power (TEP) which has three electrical-generating units that can burn natural gas or fuel oil and one unit that can burn coal (PDEQ 2007b), which are potentially the largest industrial sources of airborne metals in the study area (US EPA 2010a). Other potential stationary sources are numerous iron works and other light industrial activity, and Tucson International Airport which could be a significant source of airborne lead derived from leaded aviation gasoline (US EPA 2008a). Automobile traffic from the numerous busy

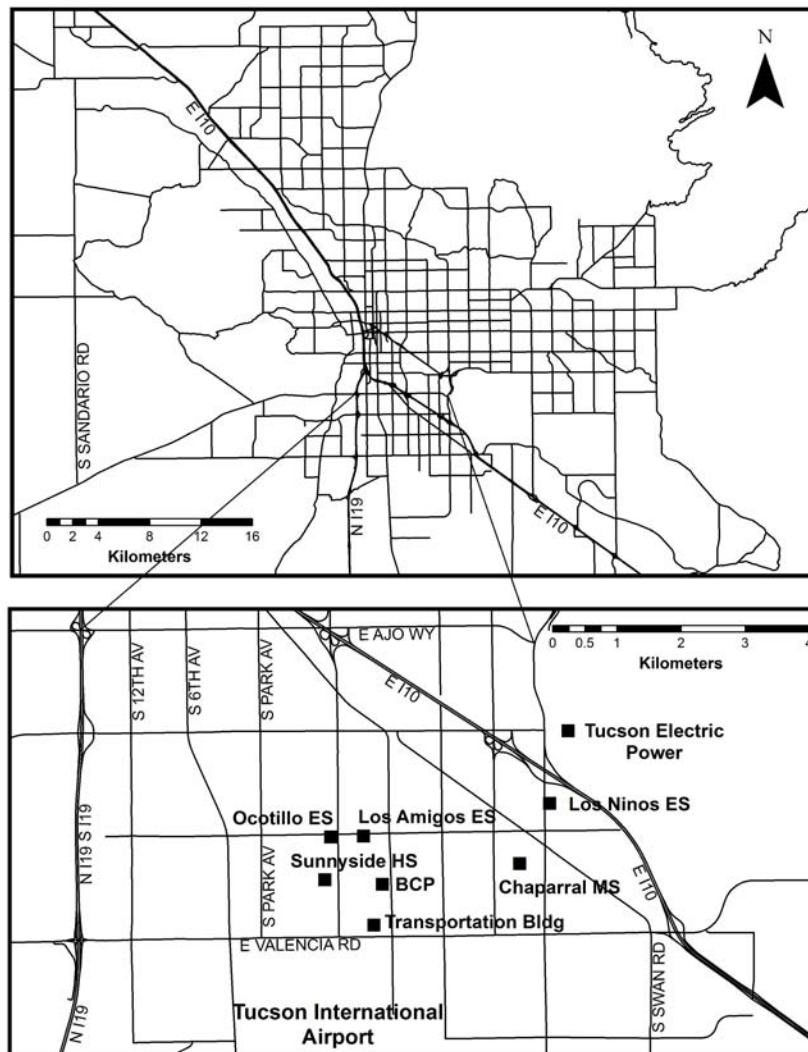


Figure 1. Map of the study area showing the location of the five schools and Transportation Building within the Sunnyside Unified School District where the PM_{10} monitors are located (ES = elementary school, MS = middle school and HS = high school). Also shown are Brush Ceramic Products and Tucson Electric Power. GIS data courtesy of Pima County, Arizona.

streets in the study area, including Interstate 10 and Interstate 19 highways, could also contribute (Johansson et al. 2009).

Sampling

Aerosol collection began on July 11, 2007 using six Anderson PM_{10} monitors equipped with filters that capture airborne particles with an aerodynamic diameter of ≤ 10 micrometers (PDEQ 2007a). Each monitor runs for 24-hours (midnight to midnight) every six days so that one monitor is running all the time. The monitors are located on the roofs of the following buildings in the SUSD: Ocotillo Elementary School, Los Amigos Elementary School, Los Niños Elementary School, Sunnyside High School, Chaparral Middle School and the Transportation Building. A duplicate monitor is located at the Ocotillo Elementary School for quality control pur-

poses. Operational procedures, quality control, calibration and chain of custody protocols are described in the PDEQ Beryllium Monitoring Network Plan (PDEQ 2007a). Quarterly reports of the PM_{10} and beryllium data as well as the Beryllium Monitoring Network Plan are available on the PDEQ website (PDEQ 2010a).

Here we report beryllium results for the period June 1, 2008 to December 31, 2009 ($n=569$); other metals for the period June 2, 2008 to September 1, 2009 ($n=218$); and PM_{10} for the period January 1, 2008 to December 31, 2009 ($n=678$). PM_{10} and beryllium monitoring is ongoing.

Laboratory Analysis

National Institute for Occupational Health and Safety (NIOSH) Method 7102, "Beryllium and Compounds, as Be" (including the use of optional

Table 1. Descriptive statistics for the SUSD metals monitoring project. Concentrations are in ng m⁻³ except for PM₁₀, which is in µg m⁻³. The sampling period for beryllium was June 1, 2008 to December 31, 2009 (n=569). The sampling period for the other metals was June 2, 2008 to September 1, 2009 (n=218). The sampling period for PM₁₀ was January 1, 2008 to December 31, 2009 (n=678).

Parameter	Average conc.	Minimum conc.	Maximum conc.	Standard deviation	# Samples < PQL	Total # samples
Beryllium	0.00	0.00	0.35	0.015	520	569
Arsenic	0.27*	0.00	6.20	0.756	173	218
Cadmium	0.10*	0.00	4.84	0.486	204	218
Cobalt	0.09*	0.00	1.16	0.212	177	218
Chromium	1.89	0.00	35.61	3.684	112	218
Manganese	12.78	1.36	58.53	8.295	0	218
Nickel	0.74	0.00	23.38	1.941	74	217
Lead	2.75	0.00	12.09	1.618	3	218
PM ₁₀	25.50	1.50	152.90	15.555	0	678

*The average concentration is less than the Practical Quantitation Level (PQL) of 0.582 ng m⁻³ for arsenic, 0.503 ng m⁻³ for cadmium and 0.265 ng m⁻³ for cobalt.

hydrofluoric acid) was used for the extraction of PM₁₀ samples. The mixture of hot concentrated nitric acid, sulfuric acid, and hydrofluoric acid results in the total digestion of any anthropogenic beryllium oxide and naturally occurring beryllium silicates. The Pima County Regional Wastewater Reclamation Department (PCRWRD) Compliance & Regulatory Affairs Office (CRAO) Laboratory analyzed the quartz filters for metals according to the 1999 US Environmental Protection Agency (EPA) methods for inductively coupled plasma spectroscopy (Compendium Method IO 3.4) or ICP-mass spectroscopy (Compendium Method IO 3.5). The State of Arizona has certified the PCRWRD CRAO Laboratory for these methods and many others.

During the study period, 521 valid beryllium samples were collected, and 218 of the filters (collected from June 2, 2008 to September 1, 2009)

were also analyzed for arsenic, cadmium, total chromium (i.e., Cr (III) + Cr (VI)), cobalt, lead, manganese, and nickel.

RESULTS

The most abundant airborne metals in southern metropolitan Tucson are lead and manganese (see Table 1 and Fig. 2). Chromium and nickel were detected in about half of the PM₁₀ filters, while the average cadmium, arsenic, and cobalt concentrations were below the Practical Quantitation Limit (PQL).

Beryllium was detected in only one of the 521 samples. A dust storm which originated in the Casa Grande area (approximately 100 km north of Tucson) occurred on July 22, 2009. On this day, the beryllium concentration at the Transportation Building was 0.35 ng Be m⁻³ and the PM₁₀ concen-

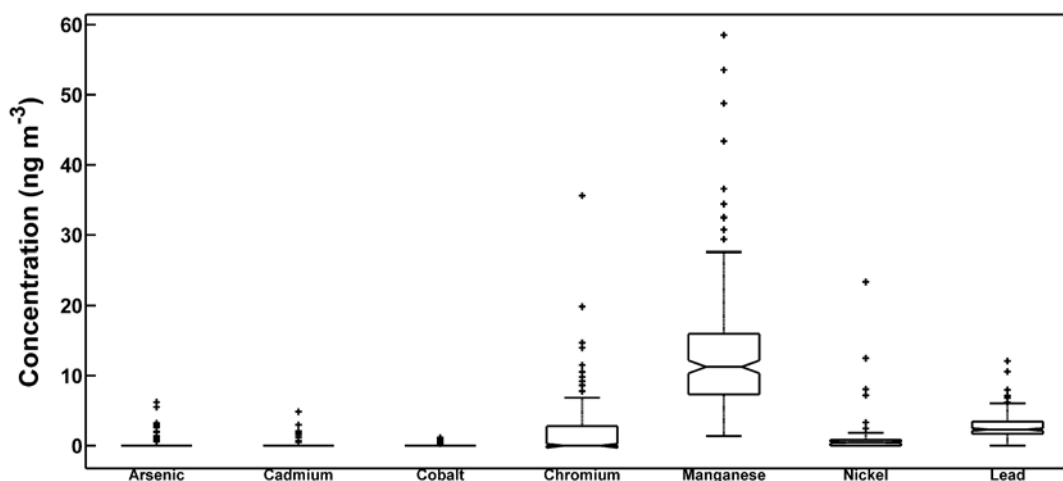


Figure 2. Box plot of metals found in PM₁₀ collected in SUSD. The tops and bottoms of each "box" are the 25th and 75th percentiles and the "whiskers" are the ends of the inter-quartile ranges. The line in the middle of each box is the sample median. Outliers are displayed with a + sign. The sampling period was from June 2, 2008 to September 1, 2009, and n=218.

tration was 149.7 $\mu\text{g m}^{-3}$. Soil in the Tucson area is known to contain natural beryllium, leading us to hypothesize that the ambient beryllium was of soil origin. To test this hypothesis, PM₁₀ filters from the PDEQ Orange Grove site (approximately 25 km upwind from the study area) were analyzed for beryllium. If there was high beryllium at Orange Grove on July 22, but not on other, non-windy days, then one could reasonably conclude that the beryllium at both sites was due to wind-blown soil and not from BCP emissions. The Orange Grove beryllium concentration was 0.66 ng Be m⁻³ on July 22, compared to a non-dusty day average of just 0.09 ng Be m⁻³ on July 10 and July 16. This suggests that the beryllium originated naturally from the dust because there is no other known beryllium source near the Orange Grove site. Similarly, the ratio of beryllium to PM₁₀ (i.e., to “dust”) is approximately equal at both Orange Grove (2.4 ppm) and Sunnyside (2.1 ppm), as one would expect if the beryllium was due to airborne dust. These ratios are reasonably close to the known concentration of beryllium in local soils (not PM₁₀), which ranges from about 0.2 to 1.2 ppm (PDEQ 2010b).

The Be/Mn ratio should be approximately equal at both Orange Grove and Sunnyside if the same type of dust was sampled. Manganese is commonly found in soils so one would expect that it would also be high at both sites on the dusty day. The Orange Grove Be/Mn ratio on July 22 was 0.26%. This is close to the Sunnyside Be/Mn ratio of 0.30% on the same day. The two sites appear to have sampled the same wind-blown soil because both beryllium and manganese were higher in approximately the same proportion (within natural variability).

US EPA and World Health Organization (WHO) Standards

Table 2 summarizes how the Tucson monitoring results compare with US EPA standards (US EPA 2010b). A major source of PM₁₀ is dust that originates from soil (Seinfeld and Pandis 2006) and the highest PM₁₀ concentration (152.90 $\mu\text{g m}^{-3}$) in the study area occurred on a very windy day. WHO standards for metals (van Leeuwen 2002) are not enforceable in Arizona, but are included in Table 2 as guidelines where US EPA standards do not exist. Two nickel measurements exceeded the lower limit of the WHO standard but are well within the 50 ng m⁻³ upper limit. The WHO nickel standard has a range so that regulators can adapt it to local conditions. There are no WHO or US EPA limits for ambient beryllium, cobalt or total chromium.

Comparison with Other Cities

Table 3 compares Tucson PM₁₀ and metals concentrations with results from other border cities and Los Angeles. The methods used in each study are compared in Table 4. Three studies used x-ray fluorescence (XRF) rather than acid digestion and ICP analysis, which offers the advantage of lower detection limits (Rasmussen et al. 2006).

The study area in southern metropolitan Tucson has good air quality compared to the other cities listed in Table 3, with only Tampico, Mexico, having lower average PM₁₀ values. Unlike the El Paso and Los Angeles metropolitan areas, Tucson is in compliance with the US EPA, PM₁₀ standard of 150 $\times 10^3$ ng m⁻³ (24-hour mean). The study area also has lower ambient metal concentrations than the other border cities listed in Table 3.

Table 2. Comparison of Tucson ambient metal and PM₁₀ levels with US EPA (2010b) and World Health Organization (WHO) standards.

Parameter	Standard	Units	Organization	# Times over standard
PM ₁₀	150 (24-hour mean)	$\mu\text{g m}^{-3}$	US EPA	1
Lead	150 (rolling 3-month mean)	ng m^{-3}	US EPA	0
Beryllium*	None	ng m^{-3}	None	Not applicable
Manganese	150	ng m^{-3}	WHO	0
Arsenic ⁺	4-13 (annual mean)	ng m^{-3}	WHO	0
Nickel	10-50	ng m^{-3}	WHO	2
Cadmium	5	ng m^{-3}	WHO	0
Chromium	None	ng m^{-3}	None	Not applicable
Cobalt	None	ng m^{-3}	None	Not applicable

*A representative average concentration for US cities is 0.2 ng m⁻³ (ATSDR 2010).
⁺Exceeds lower limit only

Table 3. PM_{10} ($\mu g m^{-3}$) and metals concentrations ($ng m^{-3}$) obtained from other studies.

Location	PM_{10}	As	Cd	Co	Cr	Mn	Ni	Pb	Reference
Juárez, Mexico	197.0	2.0	2.0	3.0	5.0	65.0	4.0	48.0	Li et al. (2001)
Mexicali, Mexico	130.0	0.8	2.9	0.0	6.8	64.0	4.0	95.4	Chow and Watson (2001)
El Paso, TX	113.0	4.0	4.0	1.0	12.0	58.0	2.0	43.0	Li et al. (2001)
Calexico, CA	61.9	0.8	1.7	0.0	2.7	29.8	1.8	38.2	Chow and Watson (2001)
Tucson, AZ	25.5	<0.582	<0.503	<0.265	1.9	12.8	0.7	2.8	This work
Tampico, Mexico	4.4	n/a	1.0	1.0	3.0	154.0	6.0	18.0	Flores-Rangel et al. (2007)
Los Angeles, CA	n/a	n/a	n/a	n/a	6.09	12.81	4.98	12.5	Singh et al. (2002)

Table 4. Sampling and analysis methods used in other studies. For each study, the average period was the entire study.

Reference	Start sampling	End sampling	Instrument	Method
Li et al. (2001)	8/1/1999	7/31/2000	Dichotomous	XRF
Chow and Watson (2001)	3/13/1992	8/29/1993	Minivol	XRF
This study	6/1/2008	9/2/2009	Hivol	ICP
Flores-Rangel et al. (2007)	10/22/2003	12/27/2003	Hivol	ICP
Singh et al. (2002)	9/2000	1/2001	MOUDI	XRF

Ambient beryllium has been less studied than the other seven metals featured in this analysis. Japanese investigators found an average beryllium concentration of 0.03 to 0.04 $ng m^{-3}$ in the industrial area of Fukuoka, Japan (Nguyen et al. 2010). Our study has a higher beryllium PQL of 0.265 $ng m^{-3}$, making comparison with the Nguyen et al. study difficult.

Thorat et al. (2001) conducted a beryllium monitoring study in the vicinity of a beryllium processing facility near Mumbai, India, which uses the metal to manufacture X-ray tube windows. Within the boundaries of the facility, they obtained an average ambient beryllium concentration of $0.48 \pm 0.43 ng m^{-3}$ ($n=397$). Soil beryllium concentrations near the facility were in the range of 1.42-2.75 ppm by mass. The researchers concluded that the soil was a major source of ambient beryllium.

Correlations Between PM_{10} and Metals

The correlation between an independent variable (PM_{10}) and a dependent variable (constituent metal) is often evaluated using linear least squares analysis, which assumes a normal distribution (Milton 1999). However, histograms of the PM_{10} dataset and each metal dataset suggested gamma distributions rather than normal distributions, which are characterized by a large number of low concentration values and a much smaller number of high values.

The Edwards-McKee algorithm (Edwards and McKee 1997) was used to transform the lead, PM_{10} , and manganese datasets from gamma distributions into normal z-distributions that could then be

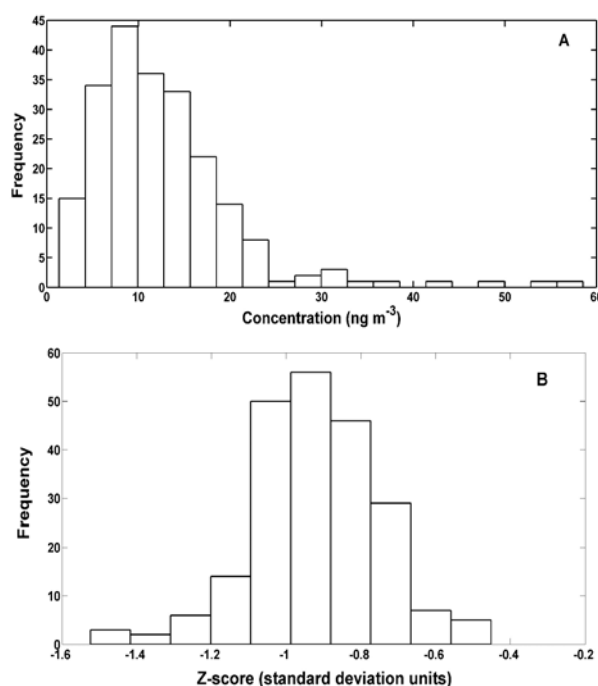


Figure 3. Frequency distribution of manganese mass/volume concentration ($ng m^{-3}$) before (A) and after (B) a Z-distribution transformation. The number of samples is 218.

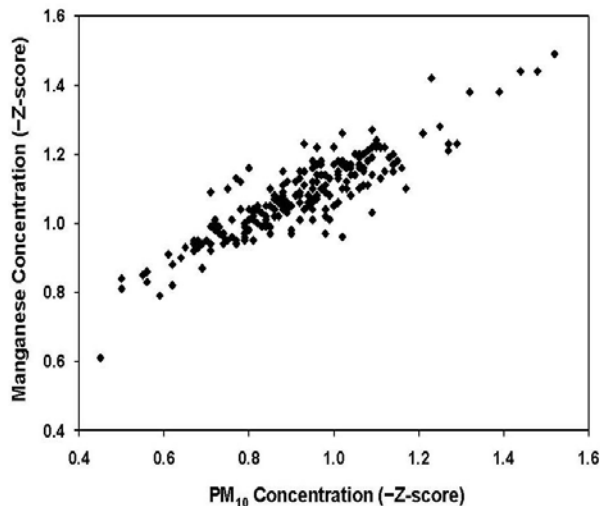


Figure 4. Scatter plot of PM_{10} concentration vs. manganese concentration. The PM_{10} and manganese concentrations were converted to z-scores using the Edwards-McKee algorithm ($y=0.60x+0.52$; $R^2=0.793$). The sampling period is June 2, 2008 to September 2, 2009 and $n=218$.

analyzed by the linear least squares method. The original and transformed manganese distributions are shown in Figure 3. The other metal distributions were not suitable for the Edwards-McKee transformation because they contained too many values below the PQL. For example, cadmium was detected in only 14 of 218 samples while beryllium was detected only once in 521 samples (Table 1). After transformation, a correlation coefficient of 0.891 was obtained between PM_{10} and manganese. Its significance was evaluated by calculating the Student's *t* statistic which was 28.86, i.e., it is highly significant and indicates a strong linear relationship between the two variables (see Fig. 4). The transformed lead distribution did not meet the Lilliefors criteria for normality (Milton 1999) and could not be analyzed further.

The major source of PM_{10} is wind-blown dust (Seinfeld and Pandis 2006). The strong correlation between manganese and PM_{10} implies that soil is a major source of manganese. Manganese, an important plant nutrient, occurs naturally in local soils at concentrations of about 400-800 ppm by mass

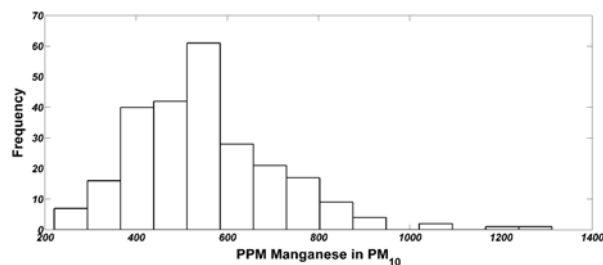


Figure 5. Frequency distribution of mass fraction manganese (ppmw) in PM_{10} . Local soils are known to contain approximately 400-800 ppmw Mn supporting the hypothesis that airborne Mn originates largely from windblown dust. The sampling period is June 2, 2008 to September 2, 2009; $n=218$. In this graph, the original data are used rather than the z-scores.

(Bohn et al. 2001). Converting airborne manganese concentrations from $ng\ m^{-3}$ to mass fractions of PM_{10} shows that manganese in PM_{10} is of the same order of magnitude as in soil (see Fig. 5).

Identification of Possible Sources

The concentrations of the eight metals and PM_{10} at each monitoring site were mapped to evaluate potential differences between sites. Figure 6 (metals) and Figure 7 (PM_{10}) suggest that the concentrations at each site are similar and one site is not disproportionately affected by a significant metals source. The data were further sorted into two categories based on the proximity of each monitoring site to TEP and BCP, two industrial facilities in the study area (see Fig. 1). Los Niños Elementary School and Chaparral Middle School were grouped as the northeast sites, while Los Amigos Elementary School, Ocotillo Elementary School, Sunnyside

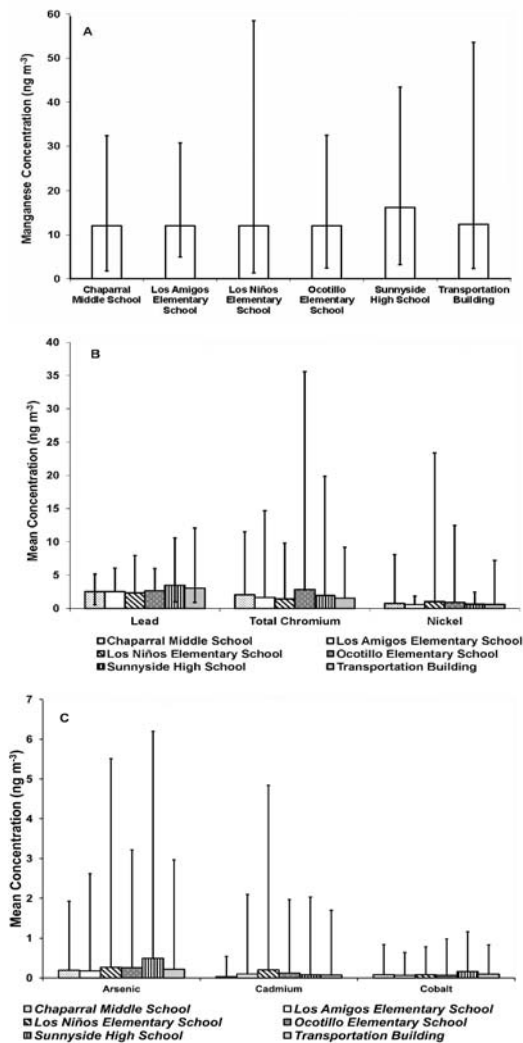


Figure 6. Concentrations of metals at each of the six SUSD sites. For clarity, the metals are grouped into similar concentrations. The range lines for each bar are the highest and lowest values in the data sets. (A) Manganese. (B) Lead, total chromium and nickel. (C) Arsenic, cadmium and cobalt. See Table 2 for the US EPA and WHO standards.

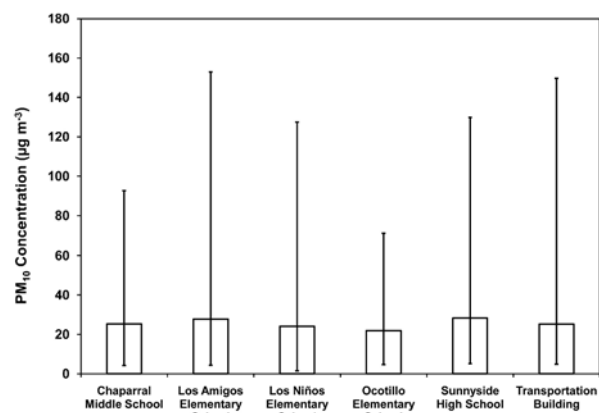


Figure 7. PM₁₀ concentration at each SUSD monitoring site. The sampling period is January 1, 2008 to December 31, 2009; n=512. The US EPA standard for PM10 is 150 µg m⁻³ (24-hour average).

High School and the SUSD Transportation Building were grouped as the southwestern sites.

The results are summarized in Table 5. For PM₁₀, arsenic, cadmium, chromium, cobalt, manganese, and nickel, there were no statistical differences between the northeast and southwest groups. (The beryllium data were not analyzed because it was detected only once.) However, the southwest group has significantly higher lead concentrations than the northeast group. The four southwest monitoring sites are closer to both the Transportation Building and Tucson International Airport which could both be lead sources. SUSD diesel school buses, which are parked and serviced at the Transportation Building, are potential sources of particulate lead from brake linings and engine parts (Lowenthal et al. 1994). While lead is not added to jet fuel used in commercial or military jets, some piston-engine powered airplanes use leaded aviation

gasoline (100 octane low lead (100LL) avgas), and so the airport could also be a source (US EPA 2008a). Comparing lead data from the Transportation site to the other sites in the southwest group showed no significant difference using the Wilcoxon-Mann-Whitney rank sign test, suggesting that school bus emissions are not the sole or dominant lead source within this group of sites.

Effect of Wind Speed on PM₁₀ and Metal Concentrations

Threshold wind velocities for dust production from a variety of Southwestern surface types has been summarized by Brazel (1987). The threshold velocity is a function of soil moisture, soil type, and the type of vegetation cover, but here we use a value of 9.0 m s⁻¹, which lies between that for disturbed desert (8.1 m s⁻¹) and a dry wash (10.0 m s⁻¹). Meteorological data from Tucson International Airport was used to distinguish windy days (2-minute wind gust ≥9.0 m s⁻¹) from non-windy days.

As expected, the PM₁₀ concentration is significantly higher on windy days (Table 6), but by contrast, nickel and cadmium concentrations were significantly higher on non-windy days suggesting that a local source(s) unrelated to natural soils may be significant. However, more data are needed to distinguish potential sources such as vehicles and industry.

Manganese, which is highly correlated with PM₁₀, also has higher concentrations on windy days but the difference between calm and windy days is not statistically significant (Table 6). Vehicle emissions, a potential source of manganese (Lough et al. 2005, Johansson et al., 2009), may confound efforts to correlate wind speed with ambient manganese.

Table 5. Comparison of average concentrations for the northeastern group (Los Niños and Chaparral) and the southwest group of monitoring sites (Los Amigos, Ocotillo, Sunnyside High School and the SUSD Transportation Building) using the Wilcoxon-Mann-Whitney rank sign test at a probability of 0.05.

Parameter	Northeastern group average	Southwestern group average	Statistically significant difference?
PM ₁₀ (µg m ⁻³)	24.71	25.87	No
Arsenic (ng m ⁻³)	0.23*	0.29*	No
Cadmium (ng m ⁻³)	0.12*	0.09*	No
Chromium (ng m ⁻³)	1.71	1.98	No
Cobalt (ng m ⁻³)	0.08*	0.10*	No
Lead (ng m ⁻³)	2.44	2.91	Yes
Manganese (ng m ⁻³)	12.06	13.16	No
Nickel (ng m ⁻³)	0.88	0.75	No

*Less than the PQL.

Table 6. Comparison of average PM₁₀ and metals concentrations on windy (≥ 9 m/s) and non-windy days using the Wilcoxon-Mann-Whitney rank sign test ($p=0.05$).

Parameter	Windy day average	#<PQL	Non-windy day average.	#< PQL	Statistically significant difference?
PM ₁₀ ($\mu\text{g m}^{-3}$)	28.18	0	23.82	0	Yes
Arsenic (ng m^{-3})	0.31	74	0.23	99	No
Cadmium (ng m^{-3})	0.00	93	0.18	111	Yes
Chromium (ng m^{-3})	2.20	43	1.65	69	No
Cobalt (ng m^{-3})	0.11	74	0.08	103	No
Lead (ng m^{-3})	2.62	2	2.85	1	No
Manganese (ng m^{-3})	13.61	0	12.16	0	No
Nickel (ng m^{-3})	0.44	41	0.97	33	Yes

CONCLUSIONS

This nearly 3-year study of hundreds of PM₁₀ samples demonstrates that the southern metropolitan Tucson area has good air quality with respect to metals and particulate matter. The measured PM₁₀ and metal concentrations are below both US EPA and WHO standards. Ambient metal and PM₁₀ concentrations are also low compared to Los Angeles, Mexico City and border cities such as El Paso.

Beryllium, a metal of major concern to the south Tucson community, was detected only once in 521 samples (probably from wind-blown dust) indicating that BCP was not a significant ambient beryllium source in the study area over this time period. This study found that TEP, a potential source of airborne metals, is not disproportionately affecting the schools closest to the facility. Because TEP uses natural gas more often than it uses coal, the potential for the facility to be a significant metals source is probably reduced.

We observed statically higher lead concentrations in the southwestern group of monitoring sites. In light of the new ambient lead standard which has been lowered from 1500 ng m^{-3} to 150 ng m^{-3} (3-month rolling mean) (US EPA 2008b), PDEQ resumed daily lead monitoring at one of their sites in 2011, which may shed more light on the origins of the airborne lead.

This study found a strong correlation between PM₁₀ and manganese, implying that the soil is an important source of ambient manganese. Further study is needed to evaluate whether the soil is also an important source for other airborne metals that were detected less frequently in this study.

As expected, high winds lead to significantly higher PM₁₀ concentrations. On non-windy days, cadmium and nickel concentrations are statistically higher, and further study is needed to understand this phenomenon.

ACKNOWLEDGMENTS

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