

Comparison of trace elements in size-fractionated particles in two communities with contrasting socioeconomic status in Houston, TX

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Abstract Levels of ambient air pollutants, including particulate matter (PM), are often higher in low-socioeconomic status (SES) communities than in high-SES communities. Houston is the fourth largest city in the USA and is home to a large petrochemical industry, an active port, and congested roadways, which represent significant emission sources of air pollution in the region. To compare levels of air pollution between a low-SES and a high-SES community, we simultaneously collected a 7-day integrated size-fractionated PM between June 2013 and November 2013. We analyzed PM mass and elements for three particle size modes: quasi-ultrafine particles (quasi-UFP) (aerodynamic diameter $<0.25\ \mu\text{m}$), accumulation mode particles ($0.25\text{--}2.5\ \mu\text{m}$), and coarse mode particles ($>2.5\ \mu\text{m}$). Concentrations of vanadium, nickel, manganese, and iron in the quasi-UFP mode were significantly higher in the low-SES community than in the high-SES community. In the accumulation and coarse modes, concentrations of crustal elements and barium were also significantly higher in the low-SES community compared to the high-SES community. These findings suggest that people living in the low-SES community

may experience higher exposures to some toxic elements as compared to people in the high-SES community.

Keywords Socioeconomic status · Quasi-ultrafine particles · Accumulation particle · Coarse particle · Trace elements

Introduction

The issue of environmental justice in the context of air pollution has been widely studied in the USA and elsewhere (Wing et al. 2008; Kohlhuber et al. 2006; Perlin et al. 2001). Previous studies have examined the impact of nearby industrial facilities, hazardous waste sites, and busy roadways on local air quality. Due to the proximity to these local pollution sources, people living in these communities are more likely to be exposed to high levels of environmental pollution and may face risks for the development of adverse health outcomes. The modification factor may be associated with different socioeconomic status (SES) such as poverty level, education, demographics, or others. In recent studies that have utilized data from ambient air monitoring sites at fixed locations, findings indicate that individuals living in low-SES communities in urban areas are exposed to higher air pollution levels than individuals living in high-SES communities (Fecht et al. 2015; Hajat et al. 2013; Jones et al. 2014; Miranda et al. 2011). For instance, Fecht et al. (2015) reported that daily ambient air levels of particulate matter less than $10\ \mu\text{m}$ in aerodynamic diameter (particulate matter (PM)₁₀) in low-SES communities in England

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and the Netherlands with more than 20% of the population belonging to non-white groups (>20%) were 1.1 to 3.0 $\mu\text{g}/\text{m}^3$ higher as compared to communities with smaller proportions of non-whites (<20%). Jones et al. (2014) also found that individuals from the Multi-Ethnic Study of Arteriosclerosis (MESA) in the USA living in neighborhoods with a predominantly (>60%) Hispanic population were exposed to higher levels of $\text{PM}_{2.5}$ (aerodynamic particle size less than or equal to 2.5 μm) than persons from communities with a lower proportion (<25%) of Hispanics. Thus, these findings of higher levels of air pollution among communities with lower SES suggest heightened health risks in these populations.

It is well documented that ambient $\text{PM}_{2.5}$ is a major risk factor for cardiovascular and respiratory outcomes (Bell et al. 2009; Hoek et al. 2013; Peng et al. 2008). However, the relationships between the impact of ambient $\text{PM}_{2.5}$ and health outcomes in vulnerable communities are widely variable. This may result from the heterogeneity of the chemical constituents in ambient air levels of $\text{PM}_{2.5}$ that vary depending on the mix of sources nearby (e.g., industrial or automotive emissions). Further, studies have found that the toxicity of $\text{PM}_{2.5}$ is primarily dependent on which chemical constituents are present (Peng et al. 2009; Zhou et al. 2011). Yet, few studies have examined variability in levels of chemical constituents in PM in communities on the basis of poverty level. One study utilized data from ambient air quality monitoring sites across the USA to examine differences in constituents of $\text{PM}_{2.5}$ between communities with predominantly non-Hispanic black and Hispanic populations as compared to non-Hispanic whites (Bell and Ebisu 2012). Another study measured personal exposures to $\text{PM}_{2.5}$ and air toxics and reported higher exposures to $\text{PM}_{2.5}$ and benzo[a]pyrene among persons living near local sources in a low-SES community as compared to people living in a community without local pollution sources (Zhu et al. 2011).

Another important consideration is that the levels of chemical constituents in PM may vary according to particle size fractions because different sources emit different size ranges of particles (e.g., combustion-related anthropogenic sources mostly emit particles that are in the fine particle size fraction (e.g., 1 μm in diameter or smaller)). People living near combustion-related sources are likely to inhale a greater dose of more toxic compounds in smaller sizes that can penetrate into deeper regions of the human lung (Sioutas et al. 2005). While studies have examined the spatial and temporal variation of particle-bound polycyclic aromatic hydrocarbons and elemental constituents

of PM in heavily trafficked urban areas or near industrial parks (Kleeman et al. 2008; Kleeman et al. 2009; Arhami et al. 2009; Daher et al. 2013; Saffari et al. 2013), less is known about differences in levels of chemical constituents in different size modes of PM in socioeconomically disadvantaged and advantaged neighborhoods. For a better understanding of local air quality potentially affected by multiple anthropogenic emission sources as well as the distribution of toxic chemical components of PM in different size modes, we performed this study in Houston, TX, between June 2013 and November 2013.

The primary objective of this study was to compare trace elements in ambient air between a low-SES community and a high-SES community in Houston, TX, a city characterized by multiple air pollution sources. Further, a secondary objective was to examine the spatial heterogeneity of size-fractionated airborne trace elements in a low-SES Houston community impacted by nearby industrial and traffic sources compared to a high-SES Houston community without major industrial emission sources.

Materials and methods

Sampling locations

According to the US Census 2010, Houston is the fourth largest city in the nation with a population that exceeds 2 million (US Census Bureau 2010). With the growth of the population, Houston ranks sixth in the USA for congestion due to traffic (Texas A&M Transportation Institute 2015). Additionally, Houston has about 3700 oil-, gas-, and energy-related companies and more than 540 chemical plants that account for about 40% of the nation's production of chemicals (Greater Houston Partnership 2015). Due to the presence of a large petrochemical complex (i.e., the Houston Ship Channel), an active port and major roadways in Houston, multiple sources likely contribute to ambient levels of PM, particularly in neighborhoods in close proximity to industrial facilities or highways. To capture the impact of these emission sources on ambient air levels, we selected one sampling location that was located approximately 0.5 mi east of a major freeway (I-610) and north of the Port of Houston Authority and shipyards along the Houston Ship Channel. A railroad also runs in an east-west direction, south of the sampling site (Fig. 1). Air sampling equipment was deployed at the fixed ambient air monitoring site on

Clinton Drive that is maintained by the Houston Health Department Bureau of Pollution Control and Prevention (hereafter referred to as the community sampling site (CSS)). For comparison, we selected an urban background location without major nearby emission sources (i.e., the University of Texas Health Science Center at Houston (UTHealth) School of Public Health), which is located in the center of the Texas Medical Center and surrounded by hospitals, universities, and residential areas. Air sampling equipment was deployed on the rooftop of the ten-storey UTHealth School of Public

Health building, hereafter referred to as the UTHealth. Table 1 summarizes the sociodemographic characteristics of the two communities where sampling took place (US Census Bureau 2010). We defined the SES status of communities based on zip codes and 2010 annual median household income (\$49,445) in the USA from US Census Bureau data (US Census Bureau 2010). The CSS sampling site was located in a low-SES community (2010 annual median household income \$31,715), and the UTHealth sampling site was located in a high-SES community (2010 annual median household income \$68,980). Over

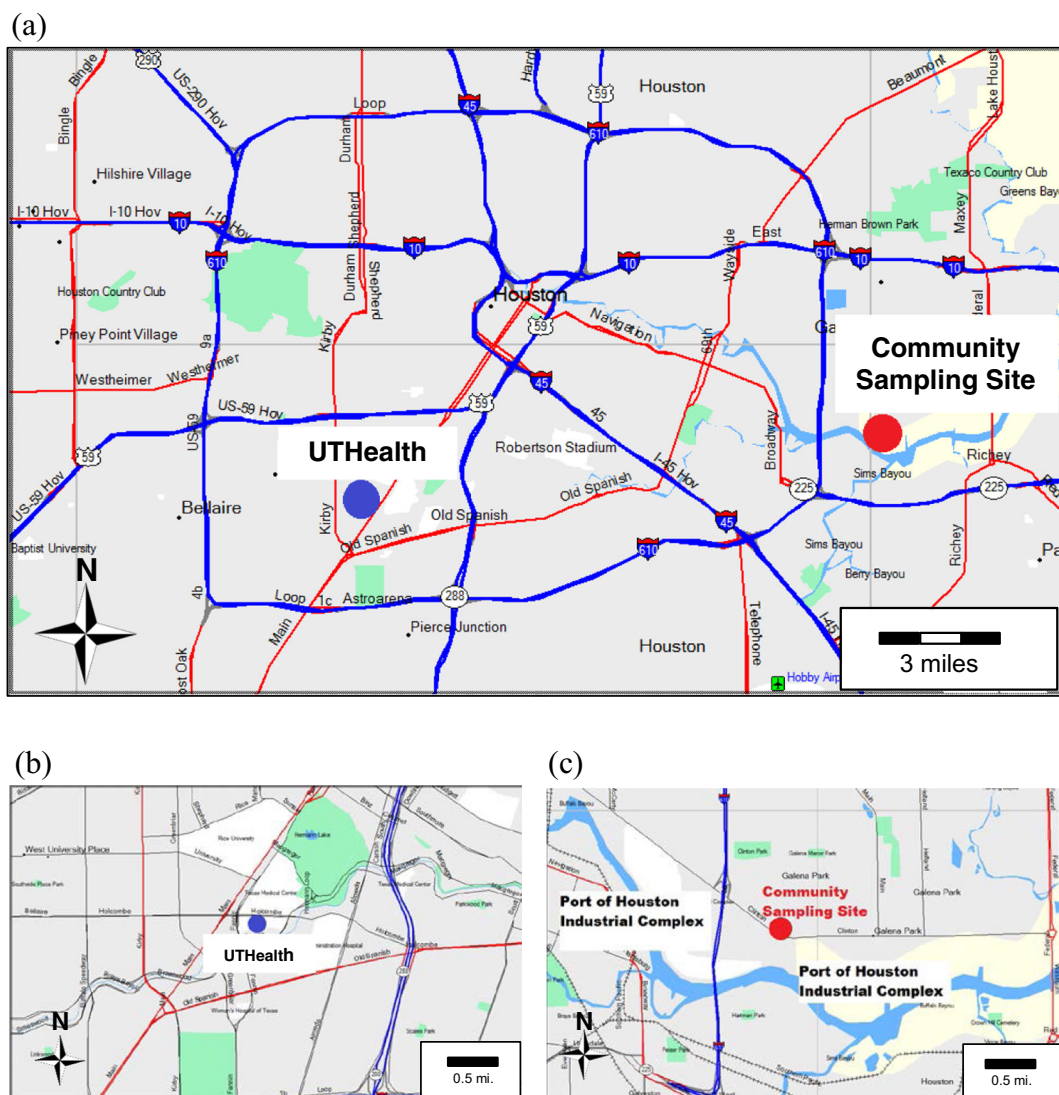


Fig. 1 Map of two sampling locations (a). The University of Texas Health Science Center at Houston (UTHealth) School of Public Health (coordinates 29.7065807, -95.401509) was selected as an urban background sampling site (i.e., high-SES community

site) (b). An ambient air monitoring site (coordinates 29.734154, -95.257583) maintained by the Houston Health Department Bureau of Pollution Control and Prevention was chosen as a low-SES community sampling site (CSS) (c)

Table 1 Comparison of populations, general demographics, and characteristics of housing, income, and socioeconomic status between the two study communities

Category	Low-SES community	High-SES community
Zip code	77,029	77,030
Total population	17,814	10,258
Ethnicity (%)		
Non-Hispanic whites	7.8	58.4
Non-Hispanic blacks	25.3	6.1
Hispanics	66.1	22.4
Non-Hispanic others	0.8	10.2
Median age (years)	31.8	33.6
Total housing units	6340	5759
Occupied housing units	5636 (88.9%)	4902 (85.1%)
Median household income	\$ 31,715	\$ 68,980
Individuals below poverty level	31.2%	12.5%
High school graduation or higher	53.8%	97.7%
Land use	Industrial and residential	Commercial and residential
Nearby sources within 1-mi radius	Refinery, Port of Houston, major freeway (Interstate 610)	Hospitals, universities, local street traffic

Source: US Census Bureau. 2015. Community Facts. Census 2010 Total Population. FactFinder. [Census.gov/faces/nav/jsf/pages/community_facts.xhtml#none](http://factfinder.census.gov/faces/nav/jsf/pages/community_facts.xhtml#none). Accessed on October 20, 2015. (http://factfinder.census.gov/faces/nav/jsf/pages/community_facts.xhtml)

90% of the population was either Hispanic or non-Hispanic black in the low-SES community whereas over 58% of the population was non-Hispanic white in the high-SES community. There were also differences in the percentage of persons living in poverty and educational achievement.

Sampling and analytical method

The sampling period was between June 2013 and November 2013. At both sampling sites, we collected one to three weekly (7-day) air samples in each month. To characterize the elemental composition of PM for different size modes at each sampling site, we used Personal Cascade Impactor Samplers (PCISs) (SKC Inc., Eighty Four, PA). The PCIS consisted of four impaction stages that collected particle mass in the following size ranges: (1) aerodynamic diameter 0.25–0.5, (2) 0.5–1.0, (3) 1.0–2.5, and (4) >2.5 μm , as well as one after-filter stage for collecting quasi-ultrafine particles (UFP), i.e., <0.25 μm . The PCIS was assembled with one 37-mm polytetrafluoroethylene (PTFE) filter with a polymethylpentene (PMP) support ring for the after-filter stage (quasi-UFPs) and four 25-mm PTFE filters for the impactor stages. At each sampling location, each PCIS was assembled with a Leland Legacy Pump (SKC Inc., Eighty

Four, PA) and deployed to collect air samples on a weekly (7-day) basis. The sampling train was placed in a customized instrument case (Pelican Products, Inc., Torrance, CA), to protect the sampler from rain or extreme weather conditions. The height of the inlet of the sampling train was approximately between 1.5 and 2.0 m from the ground platform or roof level. All pumps were plugged into power outlets at each site and programmed to start and to end at the same time at both locations. The sampling start and end times were between 10 a.m. and 11 a.m. The pre-flow rate of 9 L/min was calibrated using a flow meter (BIOS Defender 510 MH, Mesa Laboratories, Inc., Lakewood, CO) at each sampling site before sampling commenced. The post-flow rate at the end of sampling was also measured. By taking the average flow rate for each sampler, total volume for each sampler was calculated by multiplying the average flow rate by the elapsed sampling time.

The assembled PCISs with PTFE filters that were collecting five different particle size modes of PM in the air were changed weekly (every 7 days). The total number of filter samples across the 13 weeks of sampling at both sites was 130 (65 at the CSS and 65 at the UTHealth School of Public Health (SPH)). After the completion of each week-long sampling, filters were transferred to the

UTHealth SPH and stored in a filter weighing room that is controlled for temperature and relative humidity. After equilibration of at least 24 h, individual filters were weighed using a CAHN-32 (Thermo Fisher Scientific, Waltham, MA) to measure the concentrations of mass for each particle size range. For mass and elemental analyses, we analyzed three sizes: (1) quasi-UFP ($PM_{0.25}$), (2) accumulation mode PM ($PM_{2.5-0.25}$) by combining three 25-mm filters (sum of 0.25–0.5, 0.5–1.0, and 1.0–2.5 μm), and (3) coarse mode PM ($>2.5 \mu m$).

After measurement of each filter weight, samples were acid digested using a Mars 5 Xpress microwave oven (CEM, Matthew, NC) and analyzed using an Agilent ICP/MS 7500 (Agilent Technologies, Palo Alto, CA). Detailed methods for extraction and analysis were described previously (Han et al. 2012). Briefly, we placed each filter into a 7-mL Teflon digestion vessel (CEM, Matthews NC) and added 60 μL of ultrapure water (Millipore, Billerica, MA) and 225 μL of concentrated optima grade nitric acid (Sigma-Aldrich, St. Louis, MO). We programmed the temperature conditions for the Mars 5 Xpress microwave oven as follows: First, we used a ramp-to-temperature method with a maximum temperature of 165 $^{\circ}C$ and a hold time of 30 min. After the first digestion, we added 100 μL of concentrated optima nitric acid and 40 μL of concentrated Optima grade hydrofluoric acid (Sigma-Aldrich, St. Louis, MO) to each digestion vessel and performed the same ramp-to-temperature method. After the second digestion, we removed the Teflon filters and diluted the sample with ultrapure water. To construct a standard calibration curve, we prepared a standard stock solution by diluting 1 mL of ICP/MS-grade standard solution (10 $\mu g/mL$) for 16 elements (AccuStandard, New Haven, CT) into a 10-mL volumetric flask. We also examined the recovery of the 16 elements with the NIST standard reference material (SRM) 1648a Urban Particulate Matter (National Institutes of Standards and Technologies, Rockville, MD) for quality control. The average recovery rates for all elements were within 15% of the reported values for the SRM (Han et al. 2012).

The instrument detection limit (IDL) was determined with a lowest standard solution of 1 ng/mL for each element in five replicate analyses. The IDL was defined as three times the standard deviations (SD) of the five replicates. The limit of quantification (LOQ) was defined as 3.3 times the IDL. The estimated LOQ ranged from 0.03 to 1.10 ng/m^3 , assuming a 7-day sample at 9 L/min (approximate total sampling volume of

90.7 m^3). For samples with values that were below the LOQ, the values were replaced by one half the LOQ for the data analysis. All 16 elements were detected, and at least 50% of them typically had sample concentrations above the LOQs.

Statistical data analysis

In addition to examining PM mass concentrations by three different size modes (coarse, accumulation, and quasi-UFP modes), we calculated the levels of $PM_{2.5}$ as the sum of mass concentrations in accumulation mode and the quasi-UFP mode; PM_{10} was calculated as the sum of mass concentration in all three particle size modes (coarse, accumulation, and quasi-UFP modes). We performed descriptive statistical analysis by computing means and SD of concentrations of elements in each particle size mode for both the CSS and the UTHealth. We defined spatial heterogeneity of each element as the difference between CSS and UTHealth for each element within the same sampling week. We used the non-parametric Wilcoxon signed-rank test to examine the differences in mean concentrations of each element between the sites and evaluated the results at a significance level of 0.05.

To examine the spatial differences of elemental components of PM between the CSS and UTHealth, we calculated the coefficients of divergence (COD) for each element as described elsewhere (Krudysz et al. 2009; Wilson et al. 2005). The COD is defined as the following:

$$COD_{jk} = \sqrt{\frac{1}{n} \sum_{i=1}^n \left(\frac{x_{ij} - x_{ik}}{x_{ij} + x_{ik}} \right)^2}$$

where x_{ij} and x_{ik} are the i th weekly concentrations of a single element at a given particle size mode collected at each site (j is the low-SES CSS and k is the high-SES CSS, respectively), and n is the number of weekly samples ($n = 13$ in our study). CODs greater than 0.2 are indicative of substantial differences in measured concentrations of elements between the sites whereas CODs less than or equal to 0.2 suggest little differences (Wilson et al. 2005).

Furthermore, we examined the enrichment factors and Spearman correlations for 16 elements stratified by two different sample locations. The enrichment factor was calculated based on the following equation:

$$EF_{Al}(X_i) = \frac{\left(\frac{X_i}{Al}\right)_{PM}}{\left(\frac{X_i}{Al}\right)_{Crust}}$$

where $(X_i/Al)_{PM}$ is the normalized ratio of the element concentration of interest (X) to the concentration of aluminum (Al) at a given particle size mode collected at each site and $(X_i/Al)_{Crust}$ is the average ratio of the corresponding elemental crustal soil concentrations of X and Al . Al is considered as the reference element, assuming that ambient air concentrations of Al are not greatly attributed to anthropogenic sources (Gao et al. 2002; Kim and Jo 2006). Enrichment factor (EF) values less than 10 for a measured element in the air are primarily attributed to crustal soil sources while EF values greater than 10 for a measured element in the air indicate emissions largely from anthropogenic sources (Song and Gao 2011). We also performed Spearman correlation analyses to examine whether metals were emitted from similar or mixed sources.

Ratios of weekly concentrations of vanadium (V) to nickel (Ni) were calculated to determine the potential impact of industrial emission sources. The calculated ratios between the two sampling sites were compared for each particle size mode. We report the results using the original data because there were no differences in average ratio values compared with the log-transformed analysis.

Results and discussion

Particle mass concentrations

Mean PM mass concentrations in the low-SES community ($11.6 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $22.9 \mu\text{g}/\text{m}^3$ for PM_{10}) were significantly higher than in the high-SES community ($9.63 \mu\text{g}/\text{m}^3$ for $PM_{2.5}$ and $19.3 \mu\text{g}/\text{m}^3$ for PM_{10}). Seven-day average $PM_{2.5}$ mass concentrations in the low-SES community were 20.5% higher than in the high-SES community. These results are consistent with previous studies, as described in the following. In the Multi-Ethnic Study of Atherosclerosis (MESA) that used data from ambient air monitoring stations operated by US Environmental Protection Agency and additional 27 fixed air monitoring stations in six US cities (Winston-Salem, NC; New York, NY; Baltimore, MD; St. Paul, MN; Chicago, IL; Los Angeles, CA), people living in communities with a significant (>60%) Hispanic population were exposed to 8% higher $PM_{2.5}$ levels than those communities with much smaller (<25%) proportions of Hispanic

residents (Jones et al. 2014). Additionally, Bell and Ebisu (2012) used data from 219 national ambient air monitoring stations in 215 census tracts and estimated that Hispanic and non-Hispanic African-Americans had 3.8 and 9.9% higher $PM_{2.5}$ exposures, respectively, than non-Hispanic whites (Bell and Ebisu 2012).

Among the three different size modes (coarse, accumulation, and quasi-UFP modes) (see Table 2), PM mass concentrations of coarse ($11.3 \pm 2.90 \mu\text{g}/\text{m}^3$) and accumulation modes ($5.84 \pm 1.98 \mu\text{g}/\text{m}^3$) in the low-SES community were 17.5 and 32.4% higher than coarse ($9.62 \pm 2.93 \mu\text{g}/\text{m}^3$) and accumulation ($4.41 \pm 1.97 \mu\text{g}/\text{m}^3$) modes in the high-SES community, respectively. However, there was no significant difference in the quasi-UFP mode between the two communities although the average concentration of quasi-UFP in the low-SES community ($5.76 \pm 1.33 \mu\text{g}/\text{m}^3$) was 10.3% higher than in the high-SES community ($5.22 \pm 1.32 \mu\text{g}/\text{m}^3$).

Partitioning of chemical components across particle size mode

A breakdown of concentrations of trace elements for all three particle size modes for each sampling location is presented in Table 2. Figure 2 presents the percent difference of paired elemental concentrations each week between the CSS in the low-SES community and the UTHealth SPH site in the high-SES community. In the quasi-UFP size mode, 13 elements were higher in the low-SES community than in the high-SES community, although only two elements were significantly higher (82% for vanadium (V) and 78% for manganese (Mn)). The elevated concentrations of V in our study are consistent with a previous air characterization study, which was conducted in 2009 in a location (Deer Park, TX) that is approximately 15 km south from our CSS sampling location (Han et al. 2012). High levels of V may be attributed to fuel oil combustion and refinery activities near the Port of Houston. Studies conducted in Los Angeles confirmed that elevated levels of V may also be related to shipping activities (Saffari et al. 2013; Daher et al. 2013), and in the multiple US city air pollution characterization study, investigators reported that ambient air concentrations of V in cities near the sea coast were significantly higher than in inland cities (Han et al. 2012). The elevated levels of Mn at the CSS in the low-SES community may be due to emissions from multiple anthropogenic sources. Thurston et al. (2011) reported that Mn and Fe in the air were associated with iron and steel industry using the chemical speciation

Table 2 Means \pm standard deviations (SD) of concentrations of trace elements in quasi-UFP, accumulation, and coarse mode PM at both low-SES community and high-SES community sites (June 2013 to November 2013)

	Coarse mode			Accumulation mode			Quasi-UFP mode		
	Low SES	High SES	<i>p</i> value	Low SES	High SES	<i>p</i> value	Low SES	High SES	<i>p</i> value
Mass ($\mu\text{g}/\text{m}^3$)	11.3 \pm 2.90	9.62 \pm 2.93	0.014*	5.84 \pm 1.98	4.41 \pm 1.97	0.014*	5.76 \pm 1.33	5.22 \pm 1.32	0.302
Na	235 \pm 134	154 \pm 129	0.625	95.8 \pm 75.1	135 \pm 108	0.152	25.1 \pm 7.26	24.5 \pm 13.5	0.889
Mg	54.1 \pm 24.9	27.0 \pm 17.0	0.018*	27.7 \pm 20.4	28.9 \pm 23.0	0.816	3.72 \pm 2.55	3.22 \pm 2.90	0.650
Al	84.2 \pm 36.6	31.5 \pm 13.0	0.004*	64.9 \pm 57.2	55.7 \pm 54.3	0.380	32.8 \pm 37.0	30.4 \pm 28.3	0.854
K	37.3 \pm 17.4	18.4 \pm 8.65	0.080	37.0 \pm 56.3	30.4 \pm 36.4	0.322	37.9 \pm 36.2	36.1 \pm 32.9	0.896
Ca	117 \pm 43.5	26.1 \pm 12.9	0.009	29.0 \pm 15.9	10.2 \pm 9.57	<0.001*	8.39 \pm 3.50	7.16 \pm 1.15	0.247
V	0.53 \pm 0.18	0.22 \pm 0.11	0.036*	0.75 \pm 0.33	0.45 \pm 0.19	<0.001*	1.72 \pm 0.62	0.73 \pm 0.33	<0.001*
Mn	4.33 \pm 1.65	1.22 \pm 0.38	0.034*	3.46 \pm 1.48	1.91 \pm 0.80	<0.001*	3.51 \pm 1.75	1.45 \pm 0.65	0.001*
Fe	160 \pm 67.0	63.2 \pm 21.8	0.043*	102 \pm 51.4	69.6 \pm 46.0	0.100	50.0 \pm 24.7	32.1 \pm 14.6	0.036*
Co	0.11 \pm 0.06	0.13 \pm 0.08	0.383	0.24 \pm 0.11	0.29 \pm 0.17	0.381	0.17 \pm 0.02	0.18 \pm 0.03	0.545
Ni	0.50 \pm 0.18	0.32 \pm 0.15	0.035*	0.76 \pm 0.31	0.75 \pm 0.34	0.918	1.57 \pm 0.76	1.07 \pm 0.61	0.074
Cu	2.09 \pm 1.09	2.52 \pm 1.04	0.431	8.29 \pm 2.19	7.34 \pm 1.80	0.241	3.19 \pm 1.22	2.84 \pm 0.45	0.352
Zn	6.79 \pm 4.50	6.72 \pm 3.43	0.973	9.10 \pm 4.06	18.2 \pm 11.4	0.016*	7.93 \pm 7.16	19.0 \pm 14.2	0.021*
As	0.19 \pm 0.11	0.23 \pm 0.18	0.629	0.68 \pm 0.36	0.75 \pm 0.42	0.660	0.49 \pm 0.22	0.46 \pm 0.28	0.728
Se	1.59 \pm 1.56	2.16 \pm 1.87	0.539	1.07 \pm 0.58	1.17 \pm 0.56	0.668	0.96 \pm 0.09	0.95 \pm 0.15	0.856
Ba	14.2 \pm 7.52	6.19 \pm 3.74	0.027*	11.1 \pm 6.73	5.01 \pm 3.39	0.009*	1.05 \pm 1.01	0.82 \pm 1.02	0.579
Pb	0.59 \pm 0.30	0.45 \pm 0.10	0.281	0.53 \pm 0.25	0.59 \pm 0.40	0.676	0.85 \pm 0.40	1.04 \pm 0.35	0.206

Unit: ng/m^3 *Comparisons with significant differences ($p < 0.05$)

trend data from 2000 to 2005. Another likely anthropogenic source of Mn and Fe in this community would be vehicular emissions (Amato et al. 2011). Of the remaining 11 elements, the mean differences were 5 to 36% higher in the low-SES community as compared to the high-SES community. Unexpectedly, three elements (cobalt (Co), zinc (Zn), and lead (Pb)) in the quasi-UFP mode at the low-SES community site were lower (2% for Co, 26% for Pb, and 71% for Zn) than at the high-SES community site.

In the accumulation particle size mode, nine elements (magnesium (Mg), aluminum (Al), potassium (K), calcium (Ca), V, Mn, iron (Fe), copper (Cu), and barium (Ba)) in the low-SES community were 11 to 84% higher as compared to the high-SES community whereas the other seven elements (sodium (Na), Co, nickel (Ni), Co, Zn, arsenic (As), selenium (Se), and Pb) in the low-SES community were 11 to 44% lower than in the high-SES community. Among these elements, the differences for Ca, V, Mn, and Ba between the two communities were statistically significant. The elevated levels of Ca in this size mode at the CSS may be attributed to crustal dust or resuspended road dust (Karnae and John 2011). Moreover, measured

concentrations of Ca in the accumulation mode were approximately 25% of those in the coarse size mode, which is expected because these elements are likely emitted from natural sources. Also, studies have found that levels of Ca are approximately 40 to 81% lower in the accumulation size mode than in the coarse particle size (Viana et al. 2015; Krudysz et al. 2009). Our study showed that Ca was 62 and 78% lower in the accumulation size mode than in the coarse size mode in the low-SES community and high-SES community, respectively. Concentrations of Ba were consistently higher in the low-SES community than in the high-SES community. The results may be associated with resuspended road dust or vehicular emissions. Previous studies reported that concentrations of Ba in the accumulation mode were elevated near roadways (Amato et al. 2011; Harrison et al. 2011).

In the coarse particle size mode, 12 of the 16 elements (i.e., not including Co, Cu, As, and Se) were higher in the low-SES community than in the high-SES community. Differences were statistically significant for 7 of these 12 elements: Mg, Al, K, Ca, Mn, Fe, and Ba. They were 39 to 120% higher. While differences did not reach statistical

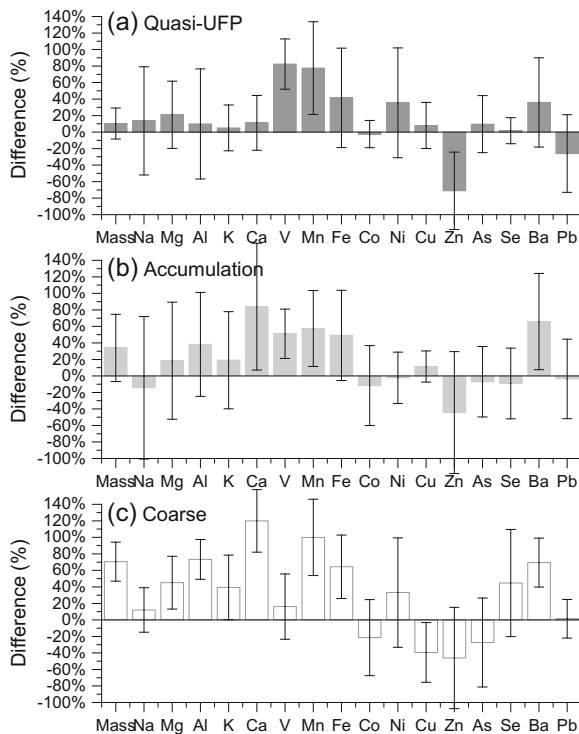


Fig. 2 Mean differences of weekly paired elements between the low-SES community and the high-SES community in **a** quasi-UFP, **b** accumulation, and **c** coarse modes between June and November 2013. % Difference = $[(\text{low-SES conc.} - \text{high-SES conc.}) / \{(\text{low-SES conc.} + \text{high-SES conc.}) / 2\}] \times 100$

significance, Na, V, Ni, Se, and Pb were also elevated (1 to 45%) in the low-SES community as compared to the high-SES community. The relatively high levels of Mg, Al, K, Ca, Mn, Fe, Na, and V in the low-SES community may be attributed to mineral and sea salt natural sources (Daher et al. 2013; Krudysz et al. 2009; Minguillon et al. 2008). The sampling site at the low-SES community is near the Port of Houston; thus, the elevated levels of these elements were expected.

Unlike mineral and sea salt elements, the major sources of Ba in the coarse mode in the low-SES community may be associated with traffic sources such as brake pad emissions (Thorpe and Harrison 2008; van der Gon et al. 2013). Studies have reported high concentrations of Ba in coarse particles in samples collected near major highways; further, concentrations of Ba were lower in locations with less trafficked roadways (Birmili et al. 2006; Gietl et al. 2010). It is likely that the elevated Ba concentrations in the low-SES community may be related to a large volume of traffic, especially trucks that transport commercial goods from the Port of Houston to elsewhere.

Spatial heterogeneity

We plotted box plots of the CODs of concentrations of particle mass and speciated components for the low-SES and the high-SES communities (Fig. 3). In quasi-UFP mode, eight elements were larger than 0.2 (suggesting spatial differences) and eight elements as

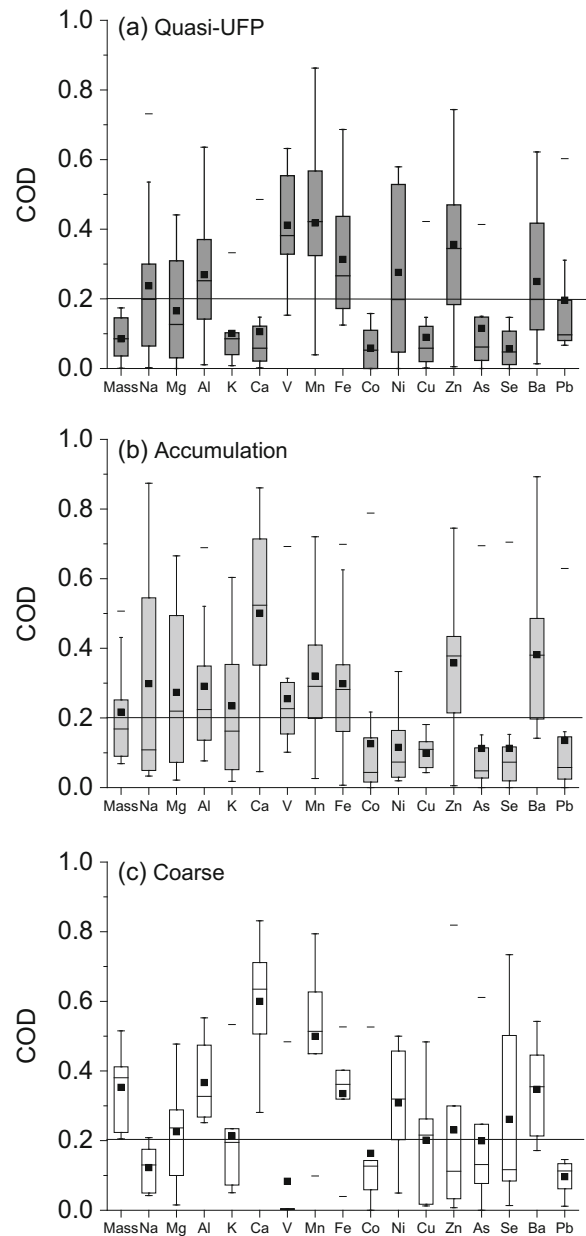


Fig. 3 Box plots of coefficients of divergence (COD) of PM mass and elements for **a** quasi-UFP, **b** accumulation, and **c** coarse size modes. Plots show minimum, 25th percentile, median, mean (filled square), 75th percentile, and maximum

well as PM mass were smaller than 0.2 (suggesting little heterogeneity between the two sampling locations). The highest weekly mean for the CODs was found for Mn (0.42) followed by V (0.41), Zn (0.36), Fe (0.31), Ni (0.28), Al (0.27), Ba (0.25), and Na (0.24). The weekly averaged CODs for the remaining eight elements and PM mass ranged from 0.06 to 0.19.

In the accumulation particle size mode, the results show that ten elements plus PM mass had mean COD values above 0.2 whereas six elements had mean COD values that were smaller than 0.2. The highest COD was found for Ca (0.50) followed by Ba (0.38), Zn (0.36), Mn (0.32), Na (0.30), Fe (0.30), Al (0.29), Mg (0.27), V (0.26), K (0.23), and PM mass (0.22). The weekly mean CODs for the remaining six elements ranged from 0.10 to 0.14.

In the coarse size mode, we found that mean CODs of 12 elements (Mg, K, Al, Ca, Mn, Fe, Ni, Cu, Zn, As, Se, and Ba) and PM mass were at or above 0.2 while only a few elements (Na, V, Co, and Pb) were less than 0.2. The average COD for Ca (0.60) was the highest while that for Pb (0.10) was the lowest in the coarse size mode.

Enrichment factor and Spearman correlation analyses

The EF was calculated for each element to examine the relative contribution of crustal and anthropogenic sources at each location (i.e., the CSS and UTHHealth) (Fig. 4). At both sampling locations, the EFs of Ca, Mg, K, Fe, Na, and Mn for all particle size modes were generally below 10, suggesting that these elements were mostly emitted from natural sources whereas the EFs of the rest of the elements (V, Ni, Co, Ba, Pb, Zn, Cu, and As) for all particle size modes were mostly above 10, indicating that these elements were significantly affected by anthropogenic sources. At CSS in the low-SES community, the EFs for V, Ni, and Pb in the quasi-UFP mode were higher as compared to those in the accumulation and coarse particle size modes. The same pattern was observed for V, Ni, and Pb in the high-SES community. Sources of V and Ni may be associated with shipping and refinery activities, oil combustion, and traffic (Amato et al. 2011; Pandolfi et al. 2011; Pey et al. 2013; Ito et al. 2016). Our findings for Ba are consistent with previous studies that show that airborne concentrations of Ba are generally associated with crustal sources in quasi-UFP mode or with vehicular emissions from brake abrasion in accumulation and coarse modes (Cahill et al. 2016; Harrison et al. 2012; Oakes et al. 2016).

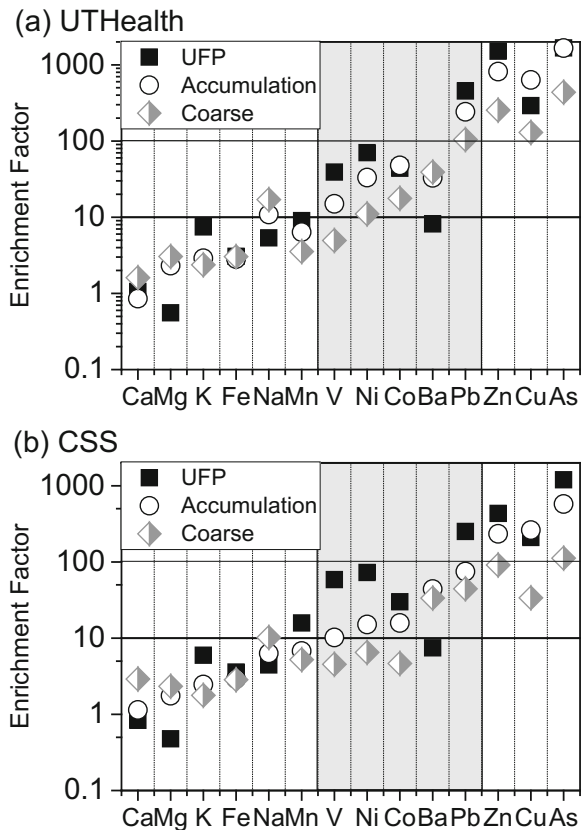
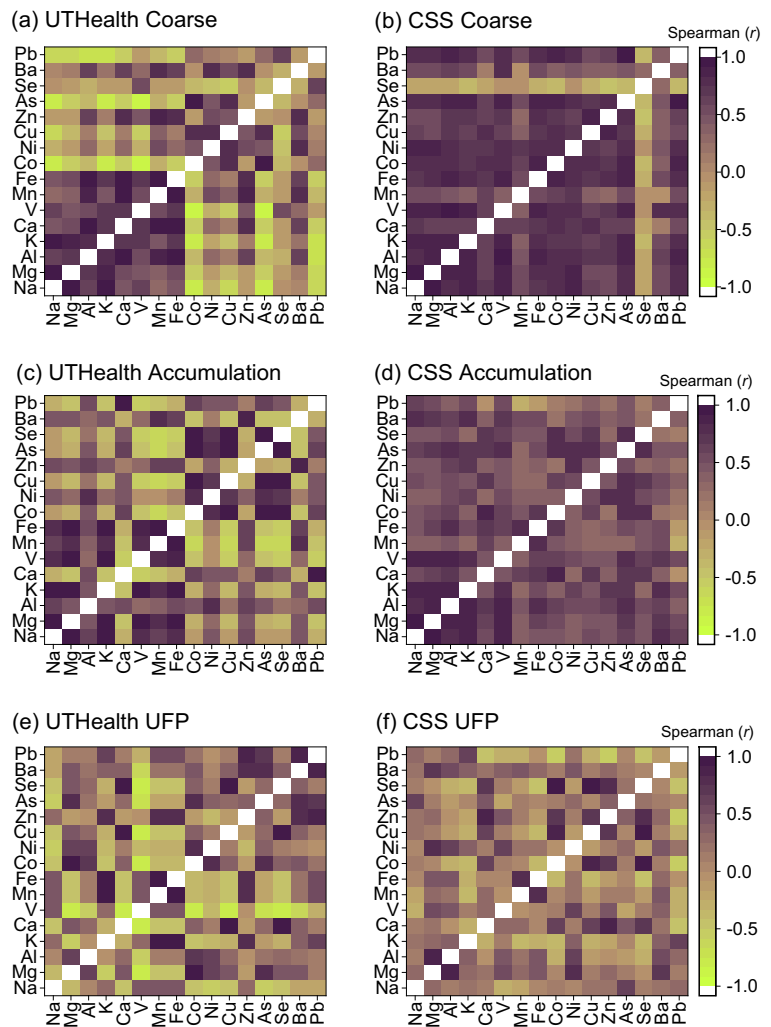


Fig. 4 Enrichment factors for selected elements normalized to Al for quasi-UFP, accumulation, and coarse particle size mode at **a** the UTHHealth and **b** the CSS sampling locations between June and November 2013

Figure 5 shows the Spearman correlation coefficients among 16 elements by sampling location for all particle size modes. Concentrations of crustal elements (Na, Mg, Al, K, Ca, V, Mn, and Fe) and Ni, Cu, Zn, As, and Ba were highly correlated with each other at the UTHHealth sampling location in the coarse particle mode (Fig. 5a). At the CSS location in coarse particle mode (Fig. 5b), most elements were highly correlated with each other, possibly due to various common natural and anthropogenic sources near the sampling location. For accumulation mode particles as compared to the coarse mode particles, the correlations between pairs of all 16 elements were similar but marginally lower at the UTHHealth location (Fig. 5c) and nearly the same at the CSS location (Fig. 5d). For quasi-UFP mode particles at the UTHHealth location (Fig. 5e), the highest correlations were observed among the following groups of metals: (1) Mn, Fe, and K; (2) Co, Ni, Cu, Mg, and Al; and (3) As, Zn, Ba, and Pb. For quasi-UFP mode particles at the

Fig. 5 Spearman correlation among 16 elements for quasi-UFP, accumulation, and coarse particle size mode at the UTHealth and the CSS sampling location between June and November 2013. **a** UTHealth coarse. **b** CSS coarse. **c** UTHealth accumulation. **d** CSS accumulation. **e** UTHealth UFP. **f** CSS UFP



CSS location (Fig. 5f), the highest correlations were for Al and Mg; Mn and Fe; Co, Cu, Zn, and Se; and Ni, Mg, and V. Due to the relatively small number of samples ($n = 13$) at each location, no additional source receptor modeling for source apportionment was performed.

Ratios of V to Ni

The concentration ratios of V to Ni can be used to apportion contributions from various combustion sources (Pey et al. 2013; Pandolfi et al. 2011; Minguillon et al. 2008). Ratios higher than 1.5 indicate fuel oil combustion whereas ratios between 1.0 and 1.5 indicate industrial sources and ratios smaller than 1.0 indicate traffic sources (emissions from vehicles powered with gasoline or diesel engines). Minguillon et al. (2008) reported ratios of concentrations of V to Ni near the ports of Los Angeles and Long

Beach that ranged from 2.96 to 4.17 for $PM_{2.5}$. The ratios of concentrations of V to Ni in the low-SES community in our study were 1.14 ± 0.46 , 1.20 ± 0.68 , and 1.25 ± 0.48 for the coarse, accumulation, and quasi-UFP modes, respectively (Fig. 6). Our findings that the ratios of V to Ni increased with smaller particle size suggest that the sampling site in the low-SES community is affected by emissions from marine vessel activities as well as refinery facilities. On the other hand, at the high-SES community site, the ratios of V to Ni were 0.92 ± 0.33 , 0.72 ± 0.34 , and 0.87 ± 0.50 for the coarse, accumulation, and quasi-UFP modes, respectively (Fig. 6). There were no decreasing or increasing patterns of the ratios of V to Ni observed as a function of particle size in the high-SES community, suggesting little contribution of emissions from oil fuel combustion or refinery facilities but rather primarily from traffic sources.

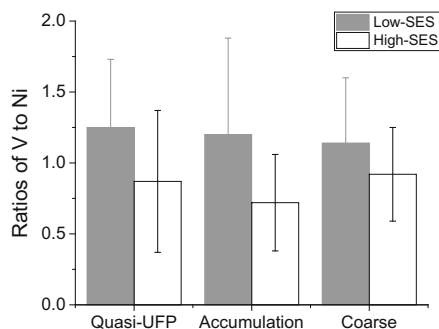


Fig. 6 Comparison of average ratios of V to Ni at both sampling sites for quasi-UFP, accumulation, and coarse particle size modes. Error bars represent standard deviation

Limitations and future work

Although we simultaneously deployed size-fractionated air samplers to evaluate ambient air levels of elemental components of PM in two contrasting communities in Houston, we could not fully evaluate temporal variations in levels of these elements because the sampling period covered only a 6-month period. Additionally, the number of samples (13 weekly measurements for each location) was relatively small. Thus, we could not estimate the contribution of local sources by conducting a formal source apportionment such as positive matrix factorization, which requires far more data than we collected.

Another limitation of our study relates to the differences in heights of the air samplers that were deployed at the CSS site (ground level) and the UTHealth site (ten-storey building height). However, in a pilot study, we simultaneously deployed PM_{2.5} samplers on the rooftop of the UTHealth School of Public Health and on the ground level of the UTHealth School of Public Health for 24 h. The preliminary (unpublished) data ($n = 5$ days) showed that there was no difference in daily PM_{2.5} concentrations between locations ($9.16 \pm 2.87 \mu\text{g}/\text{m}^3$ for the UTHealth School of Public Health roof and $9.14 \pm 1.80 \mu\text{g}/\text{m}^3$ for the UTHealth School of Public Health ground level). Our preliminary results are consistent with previous studies. Jung et al. (2011) reported that outdoor air concentrations of 14-day integrated PM_{2.5} samples that were collected on rooftops of buildings with 0–2 floors, 3–5 floors, and 6–32 floors in New York City, NY, were 10.7 ± 4.1 , 12.0 ± 3.2 , and $10.7 \pm 3.1 \mu\text{g}/\text{m}^3$, respectively. Wu et al. (2014) also reported little (3.6%) differences in PM_{2.5} levels between the ground level and the rooftop of an 11-storey building in Boston, MA. Although we have not fully evaluated the vertical differences of PM and its constituents for all

particle size modes, our initial results suggest that any discrepancy by height may be small.

One important strength of our study was that our air sampling campaign was conducted simultaneously in a low-SES community and a high-SES community that allowed for comparisons of elemental compositions by different particle size modes (coarse, accumulation, and quasi-UFP). Additionally, we demonstrated that concentrations of certain toxic elements (e.g., V and Ni) in the low-SES community were the highest in the quasi-UFP mode compared to the larger particle size modes (accumulation and coarse particles). Because quasi-UFP can be deposited at a much higher efficiency in the alveolar region than fine and coarse PM, this finding provides useful information to examine potential risks associated with exposures to particles in different size ranges, especially in disadvantaged communities. For a future study, it would be worthwhile to analyze organic compounds along with elemental constituents for size-fractionated particles as well as gaseous compounds for a better understanding of local sources in these communities. Furthermore, including more sampling locations within each community and studying additional communities would provide better information concerning intra-urban variations in concentrations of chemical constituents of PM of different particle sizes. Such an enhanced sampling strategy would further our understanding of differences in levels of chemical constituents by particle size mode in communities that vary in terms of the mix of air pollution sources and by SES, and this, in turn, may help to address disparities in environmental health.

Conclusions

In this study, we characterized elemental concentrations in ambient PM fractionated by three size ranges in a low-SES community and a high-SES community in Houston, TX. Concentrations of most elements were higher in the low-SES community than in the high-SES community. In the quasi-UFP size mode, the largest difference in levels of individual constituents of PM between the two communities was for V, possibly attributed to the close proximity of the low-SES community to shipping and refinery activities near the Port of Houston. In the accumulation size mode, concentrations of Ca, Mn, and Ba were higher in the low-SES community than in the high-SES community. This difference may result from suspended road dust and heavy-duty truck emissions. Concentrations of crustal-related elements were mostly

abundant in the coarse size mode in the low-SES community because the low-SES community is located near the Port of Houston. The results of this study suggest that people living in a disadvantaged community near multiple air pollution sources may be exposed to substantially elevated levels of combustion-related toxic elements (e.g., V, Ni, and Mn) compared to people living in a high-SES community without major point sources nearby.

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