Final Report

Effects of Brake and Tire Wear on Particulate Matter Composition, Reactive Oxygen Species, Placental Development and Birth Outcomes in Los Angeles

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Executive Summary

Background

Although a large body of evidence links exposure to fine ($PM_{2.5}$) and coarse particulate matter ($PM_{2.5-10}$) to adverse health effects, little is known about what specific components in the heterogeneous mixture of particles contribute to the observed health effects. $PM_{2.5}$ metal components are thought to play an important role in determining overall $PM_{2.5}$ health effects. Non-combustion sources such as brake and tire wear are an important source of PM-metals in urban areas, but few spatially extensive measurement campaigns have been undertaken. As a result, few models are available to estimate spatial variations in $PM_{2.5}$ metal concentrations for use in population-based studies to assess probable health effects.

Objectives

In this context, we pursued four specific research objectives:

1. Collecting $PM_{2.5}$ and $PM_{2.5-10}$ at 46 locations in two seasons across the Los Angeles Metropolitan Area for a total of 50 samples including four seasonal replicates to determine the metal content and reactive oxygen species (ROS) generation by the particles;

2. Estimating spatial variations in ambient concentrations of PM-metals associated with brake and tire wear and their impact on the generation of reactive oxygen species (ROS) in the human respiratory tract using (a) measurements of the generation of ROS by $PM_{2.5}$ particles, and (b) a newly developed kinetic multi-layer model of surface and bulk chemistry in the epithelial lining fluid (KM-SUB-ELF);

3. Utilizing land use regression (LUR) and co-kriging estimation to develop a predictive model for metals and ROS associated with brake and tire wear that can be used in population health studies; and

4. Use the model estimates developed in (3) above to estimate associations between metals or ROS and placental abnormalities and function and birth outcomes in an ongoing NIH-funded study of pregnant women in Los Angeles and to investigate associations between metals or ROS and pre-term births and low birth weight in 285,614 live births in Los Angeles County from 2017-2019.

Data and Methods

We began with a comprehensive review of the literature on brake and tire wear particles, which served to subsequently inform our selection of elements to represent different emission profiles (e.g., brake, tire, tailpipe and crustal). To support our exposure models, we collected $PM_{2.5}$ and $PM_{2.5-10}$ samples at 46 locations over two seasons (warmer early Fall and cooler mid-winter) across the Metropolitan area of Los Angeles. We also collected four replicate samples to assess seasonal effects. Data on particle mass, black carbon, elemental concentrations, and oxidative stress markers were extracted from the collected filters. After quality assurance and control, we developed land use regression models to interpolate surfaces to all populated areas of the LA metro at 30 m resolution. We also developed a novel co-kriging method that employed both data from our samples and 294 crowd-sourced Purple Air sensors for $PM_{2.5}$. Exposures at a woman's residence derived from the interpolated surfaces were assigned to 285,614 pregnant women in

LA county (based on birth address) and to 161 women from an ongoing cohort study (based on pregnancy address) that measured placental health and other exposures. We then assessed whether metal elements related to brake and tire wear were related to adverse birth outcomes.

Results and Interpretation

Using the positive matric factorization (PMF) model with markers of oxidative stress potential revealed exhaust and non-exhaust sources were contributing to aerosol oxidative stress potential (OSP) separately, with both sources being important contributors. Linking the OSP with CalEnviroScreen indicators suggested positive correlations between measures of social disadvantage and the OAP of particles. The OSP analyses also suggested that there are positive correlations with other adverse exposure and several health outcome indicators included in the CalEnviroScreen.

Although we had limited monitoring data support to fit LUR and Co-kriging models, we were able to fit models that performed well overall, with a few exceptions. We were also able to integrate for the first time information from the crowd-sourced PurpleAir network to our models and this improved model fit significantly.

Results from the epidemiological analyses of birth outcomes and our various exposure measures found associations of metals or OSP predictors, both in fine and coarse mode sizes, with term low birth weight (TLBW) and preterm birth (PTB). Effects sizes estimates for particle components were similar to those for $PM_{2.5}$ mass modeled with input from the Purple Air monitors. Importantly, given its widespread use in epidemiological studies, we note that NO_2 seemed to exert little or no confounding bias on the estimates for metal species or markers of OSP. On the other hand, when including $PM_{2.5}$ mass, we observed attenuation in the metal and OSP markers estimated effect sizes. We attribute this attenuation to the relatively high correlation between $PM_{2.5}$ mass and its elements or OSP markers, likely due to the similar model structure and data inputs used to model both undifferentiated $PM_{2.5}$ mass and speciated $PM_{2.5}$.

Our analysis of ischemic placental disease (IPD), while underpowered, revealed consistent patterns of association with all estimated coefficients having the expected sign. We interpreted this result as suggestive of an effect of metals and oxidative stress potential on IPD.

Policy Implications

Our findings have several policy implications. With the climate imperative of reducing CO_2 emissions, one strategy is to simultaneously increase the renewable component of the energy mix while also shifting driving toward electric and hydrogen vehicles. If fleet composition continues to shift toward vehicles with no tailpipe emissions, it is likely that transportation-related particle emissions will decline, but the remaining particles may have higher oxidative stress potential due to the relatively larger contribution from brake and tire wear. In such an instance, health effects from traffic-related air pollution may not decline as much as would be expected if based solely on particle mass reductions. Other measures to reduce vehicle miles travelled through changes to urban design that promote walking and biking as well as improvements to public transit may also be needed to reduce emissions from brake and tire wear. In addition, technology-forcing regulations may also be required to minimize emissions from brake and tire wear. Measures such as particle traps around braking systems may be needed. Regulations could also be put in place to change the matrix of materials in brakes and tires so the emitted particles become less toxic. On monitoring, given the high cost of assessing elemental compositions with gravimetric filters, other modeling approaches that increase exposure assessment precision, such as the co-kriging

methods developed here will be needed. This approach requires widespread data support from low costs sensors such as the Purple Air monitors. Although we did not quantitatively assess the spatial patterns of Purple Air monitors, our visual inspection revealed that many disadvantaged areas have either no or much less dense monitoring than those areas with more economic resources. State and local air agencies may have to augment these low-cost sensor networks with monitors to increase the density of sensors. This will be essential for developing exposure models that predict accurately in all areas, regardless of socioeconomic position.

1 Vehicular non-exhaust emissions: a review

1.1 Introduction

Traffic is one of the most important sources of air pollution in urban environments. Traffic-related air pollution has been associated with a wide range of adverse health effects such as asthma and respiratory symptoms (Jerrett et al. 2008), cardiovascular diseases (Le Tertre et al. 2002), birth defects (Brauer et al. 2008), and increased mortality (Beelen et al. 2008). The large body of literature on health effects linked to TRAP exposures has led to the implantation of various regulations and policies to mitigate vehicle exhaust emissions. On the other hand, the significance of non-exhaust emission sources have remained underestimated, and they have not been directly targeted by the previous emission control polices.

Non-exhaust emissions include both non-vehicular and vehicular sources. The vehicular non-exhaust emissions generate particles from the abrasion of auto parts, mainly from brake and tire wear, while non-vehicular emission sources include mineral dust and resuspended road dust (Pant and Harrison 2013a). Non-exhaust particulate matter (PM) emissions are among the major anthropogenic sources of metals and trace elements (Grieshop et al. 2006a; Lawrence et al. 2013). PM metals can increase PM toxicological effects by facilitating the generation of free radicals and contributing to oxidative stress (Bates et al. 2019; Gao et al. 2020b; Padoan and Amato 2018; Rönkkö et al. 2018).

Source apportionment studies have shown that non-exhaust emissions contribute to both $PM_{2.5}$ (aerodynamic diameter < 2.5 μm) (Habre et al. 2020a; Mousavi et al. 2018) and PM_{10} (aerodynamic diameter < 10 μm) (Amato et al. 2016a; Lawrence et al. 2013). Environmental organizations in Europe and United States have reported a gradual increase in the relative contribution of non-exhaust emission sources to overall PM_{10} traffic emissions in recent years, reporting a relative contribution in the range 75-95% in 2015 (Farrow and Oueslati 2020). In addition, it has been shown that the contribution of non-exhaust emission sources to $PM_{2.5}$ in metropolitan areas has increased by 21%-27% per year between 2011 to 2016 (Jeong et al. 2020a).

Despite the growing body of research that emphasizes the important role PM-metals in contributing to overall PM health effects, the knowledge on vehicular non-exhaust emission sources as one of the major contributors to PM-metals is limited. Moreover, with the rapid growth in the hybrid and electric vehicles (EVs) market in upcoming years, more emerging sources of non-exhaust emissions such as Li-ion batteries are introduced, whose contributions to non-exhaust PM emissions have not been studied in the past. The knowledge gap in this area prevents an organized effort for regulating vehicular non-exhaust emissions in the future. The goal of this work is to provide insight into the vehicular non-exhaust emission sources and identify the most influential factors affecting them by reviewing the current state of knowledge in this area to facilitate future regulatory efforts for mitigating these emerging traffic sources.

1.2 Research approaches for studying vehicular non-exhaust emissions

Based on the literature review, it has been found that vehicular non-exhaust emissions have been studied through various study designs, including ambient measurements, laboratory modeling,

and on-road sampling. Each of these study approaches can investigate different aspects of non-exhaust emissions. In ambient measurement studies, the overall contribution of vehicular non-exhaust emission sources, including brake and tire wear particles, to traffic emissions are quantified by source apportionment techniques. However, studies based on ambient measurements and source apportionment cannot address the effects of individual vehicle properties on brake and tire wear particles. Therefore, laboratory studies which can provide better control on experimental variables are necessary for modeling a vehicle's brake and tire wear emissions.

Laboratory studies can investigate the behavior of brake and tire wear particles while parts are detached from the vehicle and studied separately. The brake wear particles have been studied using different brake dynamometers (Alemani et al. 2018; Farwick zum Hagen et al. 2019b; Garg et al. 2000; Hagino et al. 2015; Kukutschová et al. 2011; Sanders et al. 2003) and pin-on-disc tribometers (Nosko and Olofsson 2017; Wahlström et al. 2010b; Wahlström 2015). In addition, tire wear particles have been studied in laboratory settings using road and tire simulators (Dahl et al. 2006; Gustafsson and Eriksson 2015; Kim and Lee 2018).

Laboratory studies on the brake and tire wear particles have also been performed while driving vehicles under different emission test cycles using chassis dynamometers (Kwak et al. 2013; Mathissen et al. 2019). While studies based on sampling on chassis dynamometers are limited, these studies can provide valuable information about the effect of driving conditions on brake and tire wear particles and their chemical composition. On-road sampling studies are required for understanding the behavior of brake and tire wear particles under more realistic driving conditions, which cannot be modelled in the laboratory studies (Beji et al. 2020; Farwick zum Hagen et al. 2019a; Kwak et al. 2013; Mathissen et al. 2012, 2018, 2019; Oroumiyeh and Zhu 2021). The on-road studies have investigated the effects of various factors, including brake temperature (Farwick zum Hagen et al. 2019a; Mathissen et al. 2018), vehicle velocity (Beji et al. 2020; Mathissen et al. 2012), road condition (Mathissen et al. 2012), driving condition, and lateral acceleration (Kwak et al. 2013), as well as vehicle mass and braking intensity (Oroumiyeh and Zhu 2021) on the generated brake and tire wear particles. In the following sections, vehicular non-exhaust emission sources and their physical and chemical characteristics are discussed in detail.

1.3 Characterization of vehicular non-exhaust emission sources

1.3.1 Brake wear emissions

Light-duty vehicle (LDV) brake systems are categorized into two general groups of drum and disc brakes. The disc brake technology has been introduced more recently compared to drum brakes that have been used in the auto industry for a longer period. While some of the LDVs are still equipped with rear-wheel drum brakes, front-wheel brakes, responsible for more than 70% of total vehicle braking force, are increasingly equipped with disc brakes due to their superior performance (Grigoratos and Martini 2015a).

Disc brakes generally consist of a rotor, a brake caliper assembly, and brake pads. During a braking event, the applied hydraulic force generated by brake pedals moves the piston inside the caliper assembly and presses the brake pads to the rotor, which is connected to the wheel hub. As a result of this procedure, the required friction force for stopping the vehicle is supplied. The mechanical interaction between the brake pad and brake rotor can produce brake wear particles in different sizes (Wahlström et al. 2009).

While cast iron has been reported as the most common brake rotor material, steel, ceramic, carbon-carbon, and Al, Si, and Ti have also been used as the base material for brake rotors (Kukutschová and Filip 2018; Qu et al. 2009; Sadagopan et al. 2018). Other elements including,

Mn, P as well as minor amounts of other alloying elements, including Cr have been used for improving the wear properties of brake rotors (Filip 2013). In contrast, the chemical composition of brake pads is largely variable, and it has improved over time. The brake lining friction components, their function, and their material are summarized in Table 1.1 (Eriksson et al. 2002; Grigoratos and Martini 2015a; Thorpe and Harrison 2008):

Table 1.31.3: Brake lining components and materials

Brake lining component	Function	Material
Binders	Adhering the brake lining material together	Mostly phenolic resins (high resistance against temperature and strong wear properties)
Structural fibers and particulates	mechanical strength	Metallic, mineral, carbon, and ceramic material
Fillers	Minimizing production costs; improving thermal properties	Inert or active materials, including minerals (i.e., stone powders, mica, silicates, glass, and vermiculite) and antimony sulfate, chromium oxides, and barium sulfate
Frictional additives	Improving frictional properties while reducing the wearability	Includes lubricant (Graphite, organic materials such as nut powders, as well as metal sulfides and rubber) and abrasives (metal oxides and minerals such as silica and zircon)

While the brake lining material has been generally classified into four classes of non-asbestos organic (NAO), low-metallic (10-30% metals), semi-metallic (metals <65%) brakes in older brake lining material classifications (Thorpe and Harrison 2008), newer brake lining material including ceramic brake pads were developed more recently, which should also be considered as a major type of brake lining materials (Kumar and Kumaran 2019).

Ceramic brake pads have been developed more recently compared to other brake lining materials. Despite their substantial price, ceramic brakes have been increasingly used in passenger cars with the expansion of ceramic technology in the auto industry over the past few years (Li et al. 2020). The high frictional properties and reliable thermal resistance at a wide range of operating temperatures up to 1000 ºC are the advantages of the ceramic brake pads (Kumar and Kumaran 2019; Li et al. 2021). Ceramic brakes generate less PM compared to other types (Seo et al., 2021). For instance, carbon ceramic discs have been shown to reduce PM10 emissions by up to 70% (Hesse et al., 2021). While ceramic brake pads have been mostly popular in high-end vehicles due to their high cost (Li et al., 2020), they have become more widespread in recent years due to their durability and superior performance (Li et al., 2021).

1.3.1.1 Chemical composition

The chemical composition of brake components, including brake pads, discs, and rotors, have been changing by brake manufacturers in the past few decades to improve braking capacity while providing optimized frictional, mechanical, and wear properties. The elemental composition of brake components have been analyzed through multiple laboratory measurement studies (Chandra Verma et al. 2015; Hagino et al. 2016; Hulskotte et al. 2014; Iijima et al. 2007; Kukutschová et al. 2011; Liati et al. 2019; Schauer et al. 2006a; Verma et al. 2016; Von Uexküll

et al. 2005; Wahlström et al. 2010a). For instance, the elemental composition of conventional low-metallic and semi-metallic European brake pads and brake rotors have been investigated by X-ray fluorescence (XRF) spectroscopy, and it has been found that Cu and Fe, as the primary brake elements, as well as a few other elements, including Sb, Sn, and Zn contribute to up to 90% of the weight of the brake elements (Hulskotte et al. 2014). Elemental speciation of Low-metallic and NAO brakes has been reported to be dominated by Oxygen and metals such as Ba, Cu, Fe, Mn, Ti, and Zn (Wahlström et al. 2010a), while other elements, including Al, Ca, Cd, Cr, K, Mo, Ni, Pb, Si, and Zr have also been observed in different brake pads (Figi et al. 2010; Hjortenkrans et al. 2008; Kukutschová et al. 2011; Sethupathi et al. 2021; Vontorová et al. 2017). While previously rare-earth elements (REEs) were reported to be predominantly associated with mineral dust (Amato et al. 2016a; Pakbin et al. 2011), a group of RREs, including Gd, Ho, Lu, Pr, and Tb have been recently reported in brake pads (Mleczek et al. 2021). On the other hand, Fe has been reported to be the most dominant element in the brake discs, while a few other elements, including Al, Cd, Cu, Mn, Mo, Ni, P, Sb, Sn, Ti, and Zn had lower mass concentrations in brake discs (Hulskotte et al. 2014).

While the majority of the previous studies have focused on the elemental composition of brake components, fewer studies have investigated the organic constituents of brake pads (Gadd and Kennedy 2000; Hagino et al. 2016; Plachá et al. 2015; Rogge et al. 1993; Zhao et al. 2015). The total carbonaceous fraction of brake wear PM_{10} has been reported to be largely variable (5%-76%) depending on the brake pad material, braking velocity, and brake temperature (Alves et al. 2021; Hussain et al. 2014; Malachova et al. 2016). More recently, approximately 150 organic compounds have been identified in brake wear PM_{10}, including n-alkanes, n-alkenes, n-alkanols, glycerol compounds, phenolic compounds, and polycyclic aromatic hydrocarbons (PAHs) (Alves et al. 2021). The concentrations of organic compounds were shown to be higher during light braking than heavy braking events, indicating the potential degradation of organic compounds during heavy braking conditions, which shows the effect of driving style on the chemical composition of the generated brake wear particles (Alves et al. 2021; Menapace et al. 2019).

Various elements have been used as brake wear tracers for source apportionment of fine and coarse particles. Ba, Cu, Sb, and Sn have been most frequently used as brake wear tracers (Almeida et al. 2020; Gietl et al. 2010; Hagino et al. 2016; Iijima et al. 2008; Jeong et al. 2020a; Lawrence et al. 2013; Oroumiyeh et al. 2022; Schauer et al. 2006a; Sternbeck et al. 2002). In addition, a few other elements, including As, Cr, Fe, Mn, Mo, Sr, Ti, Zn, and Zr have been recommended as brake wear tracers in some of the previous studies (Bukowiecki et al. 2009; Duong and Lee 2011; Jeong et al. 2019; Wang et al. 2021). However, many of the elements from the second group have been associated with other emission sources. For instance, Zn has been associated with tire wear emissions and industrial emissions (Jeong et al. 2019), and Fe, Ti, Mn, and Sr have been associated with various sources, including road dust and industrial emissions (Cesari et al. 2016; Pakbin et al. 2011; Querol et al. 2007). Since Ba, Cu, Sb, and Sn are more specific to brake wear emissions, they are expected to be the most suitable brake wear tracers.

Due to the potential contribution of brake wear emissions to adverse health effects, brake wear materials have been subject to various environmental regulations in different parts of the world, which have been previously discussed in detail (Grigoratos 2018). In Europe, Pb, Cd, Cr, and Hg have been eliminated from brakes since 2003 under the End of Life Vehicle Directive. In the United States, brake wear emissions have not been directly regulated through federal legislation to this date. However, some of the states, including California, New York, Rhode Island, and Washington have enforced various legislations to gradually reduce toxic species, including Cu from auto brakes by 2025, while monitoring Ni, Sb, and Zn for the potential further regulations (Grigoratos 2018). These states have also specified maximum concentration limits of 0.1% weight for Cr, Pb, Hg, and 0.01% weight for Cd in the manufactured brake pads. With the continuous

changes in brake wear chemical composition by the brake manufacturers, as well as the enforcement of new regulations on brake materials, the chemical composition of brake wear particles is expected to alter in the next decade. Therefore, continuous analysis of the chemical speciation of the brake components is crucial for understanding the physiochemical characteristics of brake wear particles and their associated health effects.

1.3.1.2 Size distributions

The brake wear particle size distribution has been investigated using various sampling approaches, as discussed in section 1.2. As shown in Figure 1.1, while mass-based particle size distribution with mode diameter in fine and ultrafine size range has been reported in a previous brake dynamometer study (Alemani et al. 2016), the majority of the previous studies have reported a unimodal brake wear mass-based particle size distribution with mode diameters in the range of 1-10 µm using various sampling approaches (Hagino et al. 2016; Iijima et al. 2007, 2008; Kukutschová et al. 2011; Oroumiyeh and Zhu 2021; Sanders et al. 2003; Von Uexküll et al. 2005).

The reported brake wear particle number distributions are more variable than brake wear mass-based particle size distributions (Figure 1.1). As shown in Figure 1.1, the mode diameter of brake wear particle number distributions has been reported from nano-scale to coarse size range. While a few studies observed a unimodal brake wear particle number distribution, the distributions were reported to be bimodal and multimodal in the majority of the previous studies.

The brake wear particle number distribution has been shown to be affected by a few factors, including brake lining material (Park et al. 2021; Sanders et al. 2003) and brake pad maintenance history (Farwick zum Hagen et al. 2019a). As shown in Figure 1.1, the measurement instruments and sampling approaches should also be considered while comparing the reported brake wear particle size distributions. For instance, Iijima et al., (2007) and Iijima et al., (2008) measured brake wear particles using Aerodynamic Particle Sizers (APS 3321, TSI Inc., Shoreview, MN) and reported a unimodal brake wear particle number distribution, without reporting a second mode in the ultrafine size range (diameter <0.1 µm), due to the limited detection size range of APS (0.5-20 µm). On the other hand, many studies reported multimodal brake wear particle size distributions with mode diameters in the fine and ultrafine size ranges (Alemani et al. 2016; Farwick zum Hagen et al. 2019a, 2019b; Nosko and Olofsson 2017; Park et al. 2021).

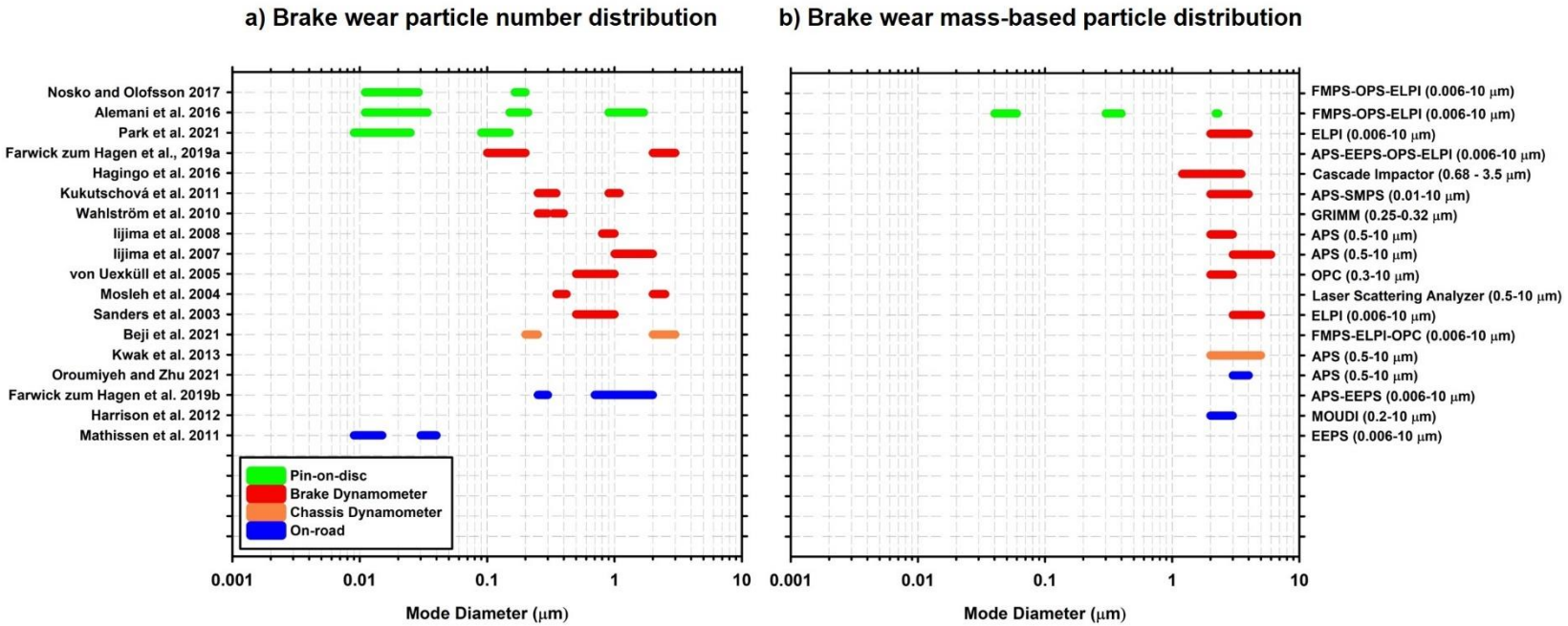

Figure 1.31: Overview of the reported mode diameters of size distributions in the previous studies based on different measurement methods for (a) brake wear particle number distribution and (b) brake wear mass-based particle size distribution. The reported numbers in the parentheses show the detection size range of the measurement instruments: Engine Exhaust Particle Size (EEPS), Aerodynamic Particle Sizer (APS), Electrical Low Pressure Impactor (ELPI), Fast Mobility Particle Sizer (FMPS), Optical Particle Counter (OPC), Laser Scattering Analyzer, Optical Particle Sizer (OPS), Scanning Mobility Particle Sizer (SMPS)

Previous on-road and laboratory studies have shown that beyond a specific brake temperature (140 °C <T_{crit} < 240 °C), ultrafine brake wear particles were generated, while at lower temperatures, ultrafine particles were not observed (Alemani et al. 2016, 2018; Farwick zum Hagen et al. 2019a, 2019b; Garg et al. 2000; Niemann et al. 2020; Nosko and Olofsson 2017). T_{crit} has been shown to increase during multiple runs of laboratory analysis of brake wear particles, presumably due to the differences in the volatilization onset temperatures of the brake wear organic material (Farwick zum Hagen et al. 2019b). For instance, T_{crit} for brake pads with organic binder content (i.e., phenolic resin) has been shown to be 180 °C, while higher T_{crit} of 240 °C has been reported for brake pads with inorganic binder content, underscoring the effect of organic content of brakes on reducing Tcrit (Alemani et al. 2018; Niemann et al. 2020).

1.3.1.3 Emission factors

Brake wear PM emission factors (EFs) have been estimated based on different sampling approaches introduced in section 1.2. The brake wear $PM_{2.5}$ EFs have been reported to be in the range of 0.5 - 5.5 mg km^{-1} Veh^{-1} in previous dynamometer studies (Garg et al. 2000; Hagino et al. 2016; Iijima et al. 2008), while PM_{10} EFs have been estimated to be 2.9 – 8.1 mg km^{-1} Veh^{-1} (Farwick zum Hagen et al. 2019b; Garg et al. 2000; Hagino et al. 2016; Iijima et al. 2008; Sanders et al. 2003). While previous source apportionment studies reported PM_{10} EFs of 8.0 – 80.0 mg km^{-1} Veh^{-1} (Abu-Allaban et al. 2003; Bukowiecki et al. 2010, 2009; Luhana et al. 2004), a lower PM_{10} EFs of 1.4 – 2.1 mg km^{-1} Veh^{-1} have been reported in an on-road study based on Los Angeles City Traffic cycle (Farwick zum Hagen et al. 2019a). The reported brake wear PM EFs are largely variable and comparing them is challenging due to the differences in sampling approaches and experimental variables. Therefore, the updated emission inventories, which take into account multiple factors including braking activity and vehicle miles traveled, are required for providing a more holistic insight into the emission factors of brake wear particles.

The environmental organizations and governmental agencies have provided various methods for estimating the brake wear $PM_{2.5}$ and PM_{10} EFs. United Kingdom National Atmospheric Emissions Inventory (NAEI) reported the average brake wear $PM_{2.5}$ and PM_{10} EFs to be 3 and 7 mg km^{-1} Veh^{-1}, respectively (NAEI 2018). Moreover, United States Environmental Protection Agency (USEPA), under Motor Vehicle Emission Simulator (MOVES) program, provided the brake wear emission factors for different vehicle classes (Figure 1.2) (USEPA 2020). Various factors, including vehicle braking activity, average deceleration rate, and vehicle weight have been included for estimating the brake wear emission factors. According to MOVES emission inventory, brake wear $PM_{2.5}$ EFs are in the range of 1.0 - 9.6 mg km^{-1} Veh^{-1}, while brake wear PM_{10} EFs are 7.8 – 77.0 mg km^{-1} Veh^{-1}. As shown in Figure 1.2, intercity bus and refuse trucks have the highest brake wear $PM_{2.5}$ and PM_{10} emission factors, despite their lower weights compared to vehicles in higher weight classes (e.g., combination trucks). This clearly shows the effect of vehicle braking activity as both refuse trucks and intercity buses have more frequent braking events compared to the vehicles in other weight classes.

European Environment Agency (EEA) has provided a more detailed method for calculating brake wear $PM_{2.5}$ and PM_{10} EFs at different vehicle speeds as previously detailed (European Environment Agency 2019). In summary, brake wear PM EFs can be estimated based on EF values of brake wear total suspended particles (TSP) using Eqs (1.1) and (1.2) (European Environment Agency 2019):

$$EF_{PM_{2.5}} = 0.39 * EF_{TSP,B} * S_B(V) \quad (1.1)$$

$$EF_{PM_{10}} = 0.98 * EF_{TSP,B} * S_B(V) \quad (1.2)$$

Where EFTSP,B is the EF of brake wear TSP, and SB (V) is the correction factor for average vehicle velocity and is calculated from Eq (1.3). SB (V) is equal to 1.67 and 0.185 for vehicle speeds below 40 km/h and above 95 km/h, respectively. For vehicle speeds in the range of 40-95 km/h, SB (V) is calculated using a linear equation:

$$S_B(V) = -0.027 * V + 2.75 \quad (1.3)$$

Table 1.2 presents $EF_{TSP,B}$ for LDVs for use in Eqs (1) and (2):

Table 1.31 Brake wear TSP emission factors for different vehicle classes (European Environment Agency 2019)

Vehicle category	Brake wear TSP emission factor (µg/km)	Range
Two-wheel vehicles	3.7	2.2 - 5.0
Passenger cars	7.5	4.4 - 10.0
Light-duty trucks	11.7	8.8 - 14.5

$EF_{TSP,B}$ for heavy-duty vehicles (HDVs) can be calculated using adjusted values from Eq (1.4):

$$EF_{TSP,B,HDV} = 3.13 * LCF_B * EF_{TSP,B,PC} \quad (1.4)$$

Where $EF_{TSP,B,HDV}$ and $EF_{TSP,B,PC}$ are TSP EFs for HDV and passenger car, respectively, and LCF_B is the load correction factor for brake wear particles, and it is calculated using Eq (1.5):

$$LCF_B = 1 + 0.79 * LF \quad (1.5)$$

Where LF is the loading factor between zero (empty truck) to one (fully loaded truck).

As shown in Figure 1.2, for a similar vehicle category, higher brake wear $PM_{2.5}$ EFs have been reported by EEA than USEPA, while EEA PM_{10} EFs were generally lower. For instance, the reported brake wear $PM_{2.5}$ EFs of passenger vehicles by EEA are 1.9 times higher than USEPA brake wear EFs for passenger cars. On the other hand, USEPA brake wear PM_{10} EFs are 1.6 times higher than EEA for passenger cars. Similar differences are observed between the reported brake wear EFs for light-duty trucks (light commercial trucks in EEA), and different classes of heavy-duty trucks by the two models. This is likely due to the fact that the reported brake wear EFs in the MOVES model are estimated based on a limited number of older studies of brake wear emissions, assuming brake wear PM_{10} EFs to be approximately 8.0 times higher than $PM_{2.5}$ (Sanders et al. 2003; USEPA 2020). Since more recent studies have shown that a larger fraction of brake wear particles is confined in fine PM, an updated MOVES model based on more recent findings in the literature is crucial for improving the accuracy of vehicle brake wear PM emission inventory in the US.

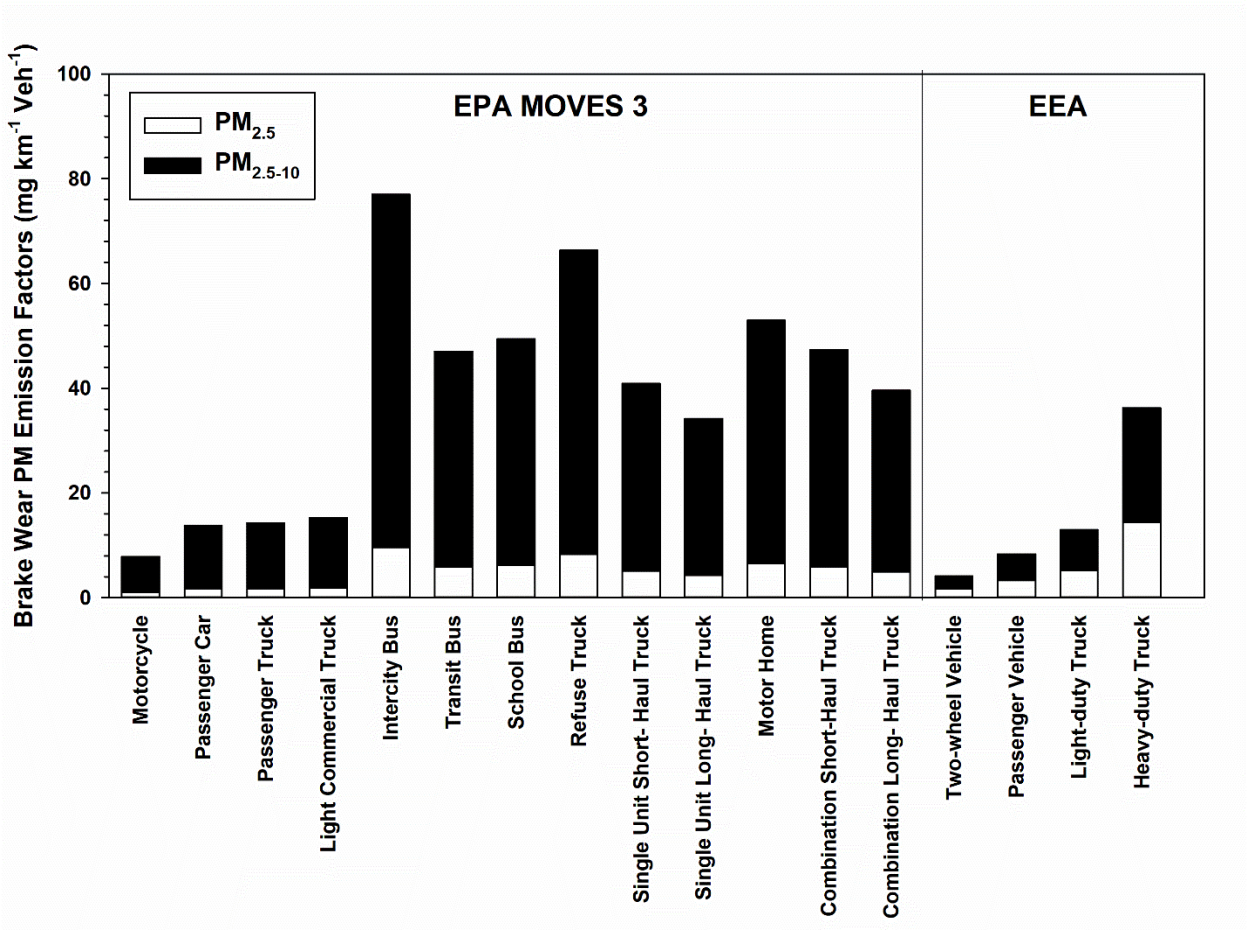

Figure 1.31: Average brake wear PM emission inventory based on the estimated values by United States Environmental Protection Agency (USEPA) (vehicles sorted by weight for calendar year 2017) and European Environment Agency (EEA). EEA brake wear PM EFs are estimated for vehicle speed of 60 km/h. EEA heavy-duty truck brake wear PM EFs were estimated for a half full truck with 4 axles.

1.3.2 Tire wear emissions

In real-life driving conditions, tire wear particles are generated through the contact and friction between the tire and pavement material during different modes of driving as a result of mechanical and thermomechanical processes. Tire treads are generally made of a mixture of rubbers, fillers, vulcanizing agents, anti-degradants, and plasticizers (Grigoratos and Martini 2014). The global annual emissions of tire treads have been estimated to be approximately 5.9 million tons year^{-1}, with population-normalized values in the range of 0.2 to 5.5 kg year^{-1} capita^{-1} (Baensch-Baltruschat et al. 2020). Since the physical and chemical characteristics of generated tire wear particles are different from the original polymeric tread particles due to heat, pressure, and mixing with road dust, tire and road wear particles (TRWP) is a more representative term for referring to the wear products of tire and road interaction (Baensch-Baltruschat et al. 2020; Beji et al. 2020).

TRWPs can become airborne or deposit on the road and further be transported to aquatic environments, based on their physical and chemical properties and local geographical features (Gill et al. 2017; Jan Kole et al. 2017; Panko et al. 2018; Wagner et al. 2018). A recent study in Germany has estimated that 66-76% of total tire wear emissions deposit on near-roadway soils, while 12-20% of the emissions are directed to surface waters and aquatic environments

(Baensch-Baltruschat et al. 2021). On the other hand, a smaller fraction of tire wear emissions has been previously reported to be airborne. While the average airborne fraction of TRWPs has been reported to be less than 2% (Panko et al. 2013; Park et al. 2018a), some studies have documented a higher fraction of airborne TRWPs in the range of 5-7% (Gualtieri et al. 2008; Wik and Dave 2009).

1.3.2.1 Chemical composition

A limited number of studies have investigated the chemical composition of tire tread particles. The distinction between TRWPs and tread particles should be considered when comparing the results of the previous studies. For instance, it has been shown that tread particles have a higher polymer content than TRWPs, while the concentrations of minerals have been shown to be higher in TRWPs, indicating the effect of road dust on TRWPs (Kreider et al. 2010; Piscitello et al. 2021). Road simulator results from testing various studded and non-studded tires on different road materials associated S and Zn to tread particles, while the presence of Al and Si was linked to pavement wear despite showing a high contribution to TRWPs (Alves et al. 2020; Beji et al. 2020; Gustafsson et al. 2008). Alves et al. (2020) reported the abundance of other elements, including Ca, Fe, K, Mg, Na, Ti in TRWPs and tire treads to be in the range of 1400 – 37700 and 48 – 4300 $\mu g\ g^{-1}$, respectively. On the other hand, a few other elements, including rare-earth elements As, Se, Sn, Cd, and Sb were only observed in TRWPs, indicating their association with the road dust rather than tire tread (Alves et al. 2020).

Kreider et al. (2010) also showed that while TRWPs have higher Si and Al content due to the mixing with road dust compared to tread particles, the concentration of Zn is higher in tread particles, presumably due to the wide usage in tire production in the form of ZnO (Milani et al. 2004a). The association of Zn with tread particles was confirmed during sampling on a chassis dynamometer (Kwak et al. 2013). Zn has been frequently used as a tracer for TRWPs for source apportionment in many of the previous studies of ambient PM measurement (Harrison et al. 2012; Lawrence et al. 2013; Lin et al. 2015; Querol et al. 2008; Srimuruganandam and Shiva Nagendra 2012; Thorpe and Harrison 2008; Yu and Park 2021). However, since has been linked to sources, including brake wear emissions (Bukowiecki et al. 2009; Schauer et al. 2006a), lubrication oil (Sternbeck et al. 2002; Wang et al. 2021), and industrial emissions (Jeong et al. 2019), it cannot be used an exclusive tracer for tire wear emissions.

Tire tread materials are manufactured from various organic materials. It has been reported that carbonaceous species constitute 65-72% of total tire tread material (Aatmeeyata and Sharma 2010b; Park et al. 2017). EC and OC have been shown to account for 18-40% and 54% of tire material weight, respectively (Jan Kole et al. 2017; Park et al. 2017). A recent study has found approximately 300 organic species in conventional tire treads and TRWPs, including phthalates, PAHs, n-alkanols, phenolic compounds, levoglucosan, steranes, and various aliphatic species (Alves et al. 2020). Alves et al. (2020) reported n-undecane as the dominant aliphatic compound, while naphthalene was shown to have the highest concentration among the 22 extracted PAH compounds. On the other hand, Pyrene and fluoranthene have been shown to be the dominant PAHs in TRWPs in other studies (Boonyatumanond et al. 2007; Depaolini et al. 2017; Kreider et al. 2010; Llompart et al. 2013; Markiewicz et al. 2017; Sadiktsis et al. 2012). While the abundance of PAHs in tire tread has been documented, the concentrations of PAHs have been found to be substantially higher in road wear particles than tread particles (Alves et al. 2020; Kreider et al. 2010). Therefore, the concentration of PAHs is expected to be higher in TRWPs than tire tread particles.

Due to the documented toxicity of PAHs (Bortey-Sam et al. 2017; Marris et al. 2020; Ren et al. 2011; Vardoulakis et al. 2020), European Union banned the usage of highly aromatic (HA) oils in tire manufacturing for tires produced after 2010 under the REACH legislation, which has led to a

gradual decrease in PAH content of tires (Diekmann et al. 2019). In fact, a statistically significant reduction in the PAH content of European-made tires has been observed between the tires produced before and after 2010 (Depaolini et al. 2017). On the other hand, the PAH content of tires manufactured outside Europe did not show a significant reduction during the same period (Depaolini et al. 2017). An analysis of PAH emission factors from traffic sources has demonstrated that tires are still one of the leading non-exhaust emission source of PAHs in Europe (Markiewicz et al. 2017). Markiewicz et al. (2017) estimated the range of emission factors for 16 USEPA priority PAHs from traffic sources to be 900-3900 μg km^{-1} Veh^{-1} in Sweden. While vehicle exhaust (24-2300 μg km^{-1} Veh^{-1}) has been reported to be the dominant traffic-related PAH source, tire wear (1.5-630 μg km^{-1} Veh^{-1}) was the most significant non-exhaust emission source of PAHs (Markiewicz et al. 2017). The tire wear PAH emission factors have been reported to be largely variable since tire particles can be impacted by various factors. For instance, the collected tire wear samples during studded tire season showed a higher correlation (r=0.96) with PAH than the samples collected during non-studded tire season (r=0.88), due to the differences in tire material and temperature (Järlskog et al. 2021).

1.3.2.2 Size distributions

The reported tire wear particle sizes in the previous studies show a considerable variation from 1 nm to 1000 μm (Klöckner et al. 2021; Park et al. 2017). While less than 10% of tire wear particles have been reported to be airborne, many studies have focused on the airborne fraction of tire wear particles due to its potential contribution to adverse health effects. Figure 1.3 shows the previously reported mode diameters of tire particle size distributions in the airborne size range (Aatmeeyata et al. 2009; Alves et al. 2020; Beji et al. 2020; Dahl et al. 2006; Dall'Osto et al. 2014; Foitzik et al. 2018; Grigoratos et al. 2018; Gustafsson et al. 2008; Hussein et al. 2008; Kim and Lee 2018; Mathissen et al. 2011; Olofsson et al. 2018; Oroumiyeh and Zhu 2021; Panko et al. 2009; Sjödin et al. 2010). The majority of these studies have reported unimodal mass-based particle size distributions with mode diameters in the range of 1-10 μm for tread wear particles and TRWPs (Alves et al. 2020; Grigoratos et al. 2018; Hussein et al. 2008; Kim and Lee 2018; Kupiainen et al. 2005; Kwak et al. 2013; Park et al. 2018a; Sjödin et al. 2010), while bimodal mass-based particle size distributions have also been reported with a second mode in the fine and ultrafine size ranges (Figure 1.3) (Beji et al. 2020; Olofsson et al. 2018; Panko et al. 2009).

As shown in Figure 1.3, the reported tire particle size distributions are largely variable. The reported tire wear particle size distributions have been unimodal, bimodal, and multimodal, with mode diameters in the range of 0.01 to 5 μm. The majority of the studies have observed at least one peak in the ultrafine size range. It has been reported that the generation of ultrafine tire wear particles is enhanced at higher driving velocities and larger slip angles, as well as longitudinal acceleration and deceleration (Foitzik et al. 2018). Other studies have reported the impact of tire condition, including tire type, tire studding, tire tread rating, tire travel distance, and tire temperature, as well as the vehicle mass and deceleration rate on the generated tire particles (Alves et al. 2020; Grigoratos et al. 2018; Oroumiyeh and Zhu 2021; Park et al. 2017). For instance, Park et al. (2017) reported that at tire surface temperatures of 160 ºC and above, the generation of nanoparticles is initiated, and it is gradually enhanced at higher temperatures up to 400 ºC.

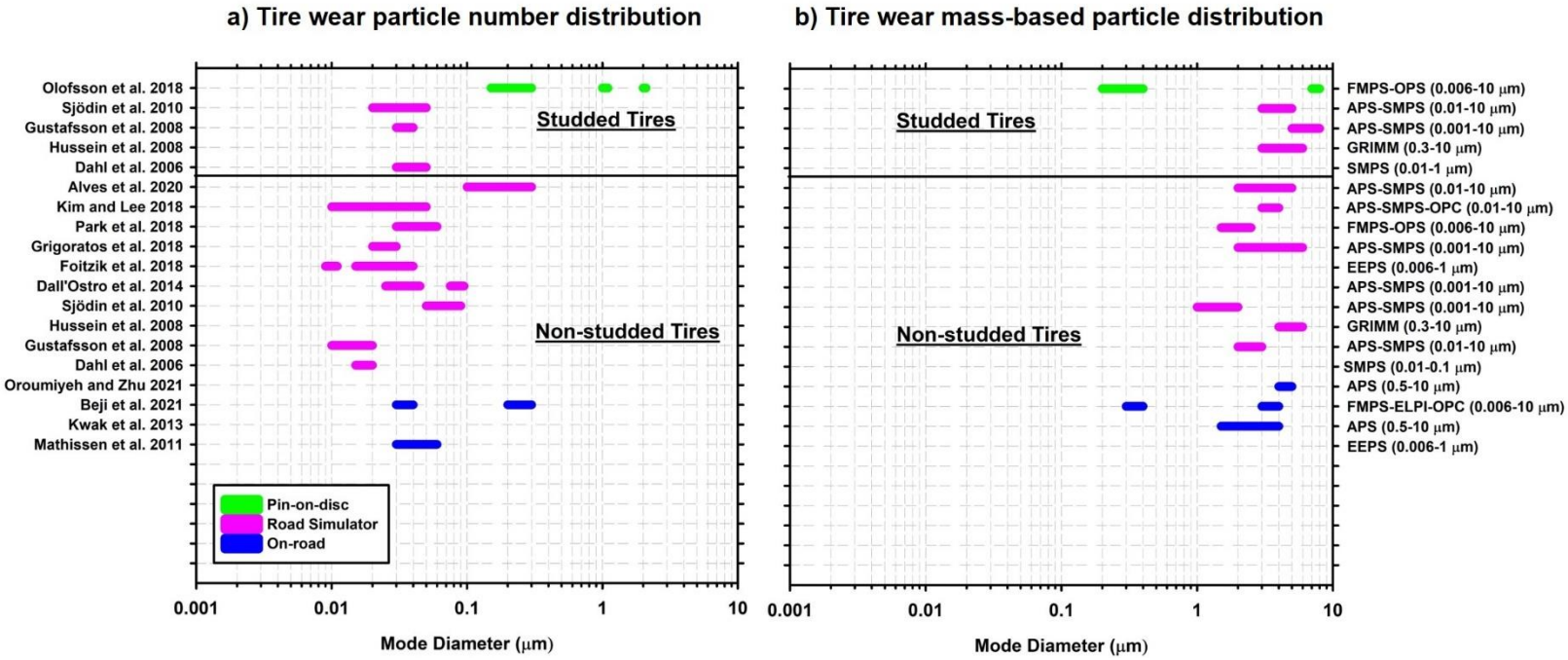

Figure 1.32: Overview of the reported mode diameters of size distributions in the previous studies based on different measurement methods for (a) tire wear particle number distribution and (b) tire wear mass-based particle size distribution. The reported numbers in t the parentheses show the detection size range of the measurement instruments: Engine Exhaust Particle Sizer (EEPS), Aerodynamic Particle Sizer (APS), Electrical Low Pressure Impactor (ELPI), Fast Mobility Particle Sizer (FMPS), Optical Particle Counter (OPC), Laser Scattering Analyzer, Optical Particle Sizer (OPS), Scanning Mobility Particle Sizer (SMPS)

As mentioned in section 1.3.2, only a limited fraction of tread particles have been reported to be airborne. Moreover, tire wear particles are one of the major sources of microplastics in the environment with a wide range of particle diameters (1-1000 μm) (Hartmann et al. 2019; Sommer et al. 2018). A significant correlation ($P < 0.05$) has been observed between the microplastic concentration in urban areas and traffic density (Kitahara and Nakata 2020). In fact, tire particles have been reported the dominant source of microplastics in the environment contributing to 50-60% of annual microplastics emissions (Boucher and Friot 2017; Hüffer et al. 2019; Lassen et al. 2015). Therefore, many studies have investigated the tire particle size distribution in the super-coarse (aerodynamic diameter ≥ 10 μm) size range to investigate the fate and transport of tire wear particles in the soil, vegetation, and aquatic environments.

Both tread wear particles and TRWPs have been reported in a wide size range up to 1000 μm (Klöckner et al. 2021). For instance, the size distribution of TRWPs has been reported to be unimodal with a mode diameter of 25 μm (Klöckner et al. 2021; Kovochich et al. 2021). On the other hand, Kreider et al. (2010) reported a bimodal TRWP size distribution with mode diameter of 5 and 25 μm while observing a unimodal distribution for tread wear particles with mode diameter of 25 μm. A few factors, including vehicle load, payment roughness, and temperature been reported to affect the tire wear particle size distribution in the super-coarse size range (Grigoratos et al. 2018; Gustafsson et al. 2008; Järlskog et al. 2021; Sjödin et al. 2010). For instance, the wear mechanism during driving on a surface with higher roughness is dominated by mechanical abrasion, which leads to the generation of larger particles, while lower particles are generated due to fatigue wear at lower roughness conditions (Chang et al. 2020).

1.3.2.3 Emission factors

Previous studies have estimated the tire wear $PM_{2.5}$ and PM_{10} EFs using various sampling approaches discussed in section 1.2. Tire wear $PM_{2.5}$ EFs have been reported to be in the range 0.3 – 11 mg km^{-1} Veh^{-1} (Aatmeeyata et al. 2009; Kupiainen et al. 2005; Luhana et al. 2004; Panko et al. 2013). While road simulator studies have observed a relatively wide range of 1.9 – 11 mg km^{-1} Veh^{-1} for tire wear PM_{10} EFs (Aatmeeyata et al. 2009; Alves et al. 2020; Dahl et al. 2006; Kupiainen et al. 2005; Sjödin et al. 2010), studies based on ambient measurements have reported tire wear PM_{10} EFs to be in the range of 7 – 7.4 mg km^{-1} Veh^{-1} (Luhana et al. 2004; Panko et al. 2013). It is important to note that tire wear concentrations and EFs are a function of road conditions, including surface wetness and temperature (Gustafsson et al. 2008; Mathissen et al. 2011), as well as the vehicle type and size (Beddows and Harrison 2021; Kim and Lee 2018; Oroumiyeh and Zhu 2021; Salminen 2014). Therefore, the reported EFs in the previous studies are not always comparable. Instead, emission inventories can be used for a more comprehensive understanding of the tire wear EFs.

UK NAEI has estimated tire wear $PM_{2.5}$ and PM_{10} EFs to be 5 and 7 mg km^{-1} Veh^{-1}, respectively (NAEI 2018). MOVES model by USEPA included a few factors, including vehicle speed and number of axles to estimate the tire wear $PM_{2.5}$ and PM_{10} EFs (Figure 1.4). However, vehicle weight which has been shown to be an important variable in determining tire wear emissions has not been considered in the MOVES model. As shown in Figure 1.4, tire wear $PM_{2.5}$ EFs are in the range of 0.4 – 2.6 mg km^{-1} Veh^{-1}, while tire wear PM_{10} EFs are estimated to be 2.7 – 17.1 mg km^{-1} Veh^{-1}. The combination long-haul and short-haul trucks, as well as intercity bus and refuse truck were shown to have the highest tire wear $PM_{2.5}$ and PM_{10} EFs.

The most comprehensive method for estimating tire wear $PM_{2.5}$ and PM_{10} EFs has been proposed by EEA (European Environment Agency 2019). Tire wear PM EFs are estimated based on the concentration of tire wear total suspended particles (TSPs) (European Environment Agency 2019). In summary, tire wear PM EFs can be calculated using Eqs (1.6) and (1.7):

$$EF_{PM_{2.5}} = 0.42 * EF_{TSP,T} * S_T(V) \quad (1.6)$$

$$EF_{PM_{10}} = 0.60 * EF_{TSP,T} * S_T(V) \quad (1.7)$$

Where $EF_{TSP,T}$ and $S_T\ (V)$ are the tire wear TSP EF and average vehicle velocity correction factor. $S_T\ (V)$ is equal to 1.39 and 0.90 for vehicle speeds below 40 km/h and above 90 km/h, respectively, while it is calculated from Eq (1.8) for velocities in the range of 40-90 km/h:

$$S_T(V) = -0.00974 * V + 1.78 \quad (1.8)$$

Table 1.3 presents $EF_{TSP,T}$ for LDVs for use in Eqs (6) and (7):

Table 1.32: Tire wear TSP emission factors for different vehicle classes (European Environment Agency 2019)

Vehicle category	Tire wear TSP emission factor (µg/km)	Range
Two-wheel vehicles	4.6	4.2 – 5.3
Passenger cars	10.7	6.6 – 16.2
Light-duty trucks	16.9	9.0 – 21.7

For HDVs, $EF_{TSP,T}$ is calculated by Eq (1.9):

$$EF_{TSP,T,HDV} = {}^{N_{axle}}/_{2} * LCF_T * EF_{TSP,B,PC} \quad (1.9)$$

Where $EF_{TSP,T,HDV}$ and $EF_{TSP,B,PC}$ represent EFs for HDV and passenger car, respectively, and LCF_B is the load correction factor for tire wear particles from Eq (1.10):

$$LCF_T = 1.41 + (1.38 * LF) \quad (1.10)$$

Where LF is the loading factor is in the range of zero to one for empty to fully loaded HDVs.

The EEA tire wear PM_{10} EFs for passenger vehicle and light-duty truck are 1.5 and 2.3 times higher than MOVES tire wear EFs, respectively. In addition, tire wear $PM_{2.5}$ EFs of passenger vehicles and light-duty trucks are 6.0 and 10.6 times higher in EEA emission inventory than MOVES, respectively. Similar to MOVES brake wear EFs, since the model has not been updated in the past decade, a large discrepancy between the EEA and MOVES tire wear EFs is observed. While MOVES model has used a tire wear $PM_{10}/PM_{2.5}$ ratio of 6.7, the MOVES report discusses that more recent studies have shown a smaller tire wear $PM_{10}/PM_{2.5}$ ratio in the range of 2.0-2.5, which is expected to be applied in the next versions of the model (Grigoratos 2018; USEPA 2020).

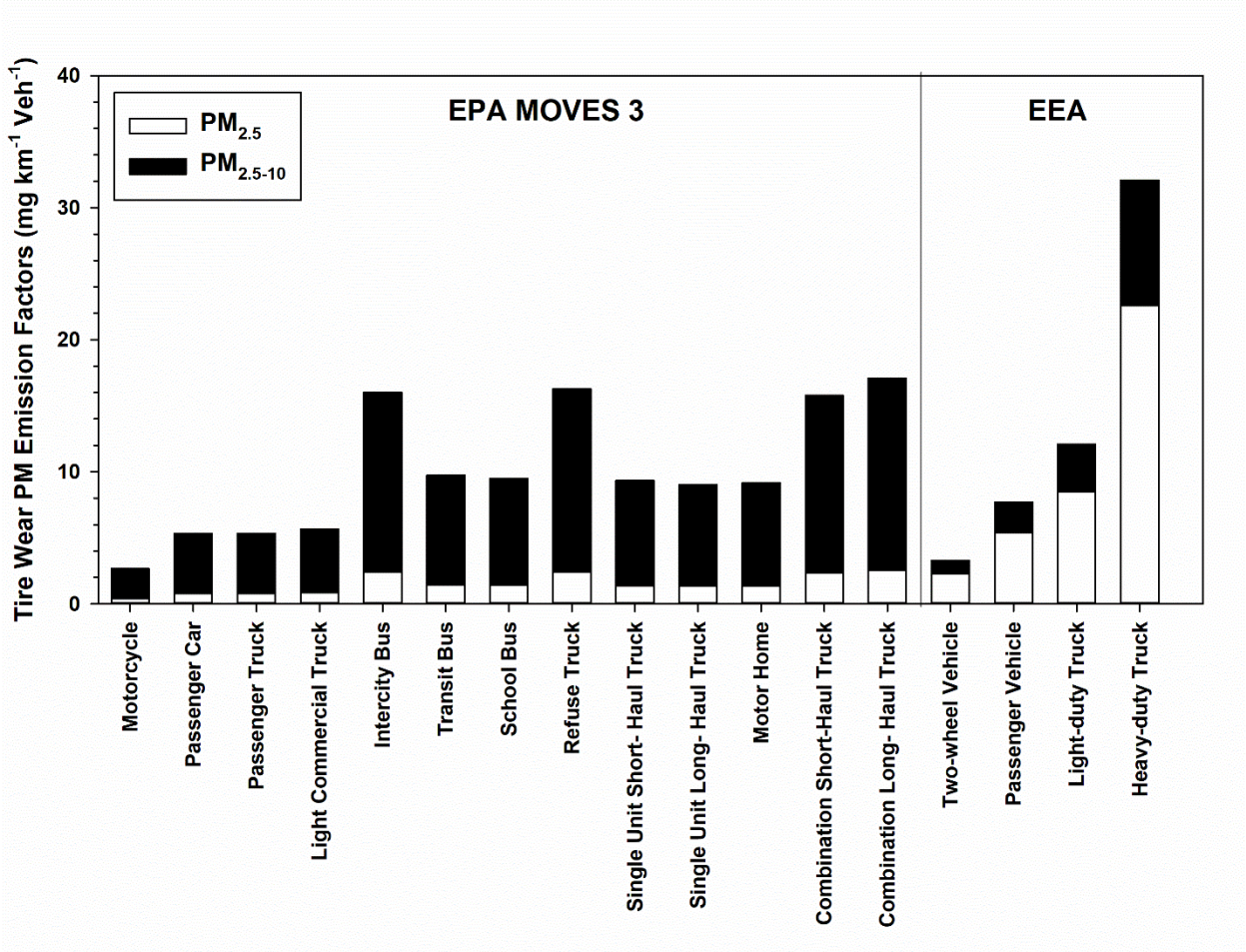

Figure 1.33: Average tire wear PM emission factors based on MOVES 3 (vehicles sorted by weight for calendar year 2017) and EEA. EEA tire wear PM EFs are estimated for vehicle speed of 60 km/h. EEA heavy-duty truck tire wear PM EFs were estimated for a half full tru ck with 4 axles.

1.4 Effect of auto electrification on the vehicular non-exhaust emissions

The global vehicle fleet has been changing in the past few years with the advancements in electric vehicles (EV) technology. EVs are becoming more affordable, and the EV market has been expanding at a higher rate. While the total share of EVs from the global vehicle market was only 2.5% in 2019 (Barkenbus 2020), the global EV market is expected to reach 30% by 2035 and 42.5% by 2035 (Rietmann et al. 2020; Rietmann and Lieven 2019). The expansion in the EV market is occurring at a faster rate in some of the countries due to governmental policies and significant national and local incentives to encourage auto shoppers (Zhou et al. 2015). For instance, the EV market in Sweden is predicted to exceed 50% prior to 2035 (Rietmann et al. 2020), while the EV sales in Norway exceeded 50% in 2021, according to the International Energy Agency (IEA) (IEA 2021).

EVs have an intrinsic environmental advantage over internal combustion engine vehicles (ICEVs) due to their zero exhaust emissions. While the impact of auto electrification on reducing exhaust emissions has been shown to be dependent on various factors such as electricity source (Wang et al. 2020b), EVs have the potential to mitigate traffic-related pollutants, including NO_x, SO_2, and CO_2, in the countries with more sustainable energy sources (Buekers et al. 2014; Burchart-Korol et al. 2018; de Souza et al. 2018).

Despite the advantage of EVs over ICEVs in reducing exhaust emissions, EVs can still contribute to non-exhaust emissions. EVs have been reported to be 24-56% heavier than ICEVs in an equal class, due to the application of heavy EV batteries for achieving a longer range per charge (Moawad et al. 2011; Timmers and Achten 2016, 2018). EVs have been reported to have higher brake and tire wear EFs than ICEVs in an equal output power level due to the extra weight of the batteries (Beddows and Harrison 2021). However, it is important to note that brake wear EFs of EVs have been reported to decrease with the application of regenerative braking system, which minimizes the vehicle's frictional braking force and converts the wheels' kinetic energy into electricity to power the vehicle (Beddows and Harrison 2021; Hall 2017). Moreover, advancements in battery technology to produce high-capacity EV batteries as well as other lightweight design strategies, including the application of lightweight composite material, carbon fiber, and aluminum alloys in vehicle manufacturing, can reduce EV non-exhaust emissions in the future (Delogu et al. 2017; Xu et al. 2020).

The Battery is one of the most important components of EVs because of its crucial role in generating power for the vehicle. With the substantial rate of auto electrification, emerging battery emissions are expected to be introduced to the environment, which have been rarely investigated, and their contribution to airborne traffic emissions has never been estimated. In this section, the potential contribution of EV batteries to non-exhaust emissions is briefly discussed.

Lithium-ion batteries (LIBs) are currently the most common energy source of EVs. They are categorized into various types based on their positive and negative electrode components (i.e., Lithium Nickel Manganese Cobalt Oxide (NMC), Lithium Manganese Oxide (LMO), and Lithium Nickel Cobalt Aluminum Oxide (NCA)) (Miao et al. 2019). The most common LIB electrolytes include Lithium salts such as $LiPF_6$, organic carbonates such as dimethyl carbonate (DEC), as well as polymer and ceramic electrolytes (Essl et al. 2020; Miao et al. 2019; Que et al. 2016). In addition, a porous membrane is integrated inside the battery to prevent a short circuit between the electrodes while allowing for the permeation of Li ions (Hannan et al. 2018).

Upon the first usage of the battery, a layer (solid-electrolyte interface (SEI)) formed by the dissociation of battery electrolytes covers the separating membrane, which can affect the battery performance and battery aging over time (Wang et al. 2018; Yang et al. 2018). At elevated temperatures, SEI undergoes a series of chain reactions, which can eventually lead to the emission of flammable gasses and toxic materials from LIBs and thermal runaway (Essl et al. 2020; Golubkov et al. 2018; Huang et al. 2015).

The majority of the previous studies on hazard analysis of LIBs are based on extreme or abnormal conditions associated with the battery failure, including overheating and thermal runaway conditions. Particle measurements during the extreme conditions such as thermal runaway are logistically challenging due to the high battery temperature of up to 1000 ºC (Golubkov et al. 2015). The limited number of studies on the battery emissions were not focused on airborne fraction of PM due to the extreme battery temperature and safety concerns for aerosol sampling. Instead, deposited PM samples were collected upon returning the battery to normal temperature and further analyzed for size distribution and chemical speciation. Therefore, the collected PM has been reported to be in the size range of 0.1-8.0 mm, without accounting for airborne battery particles (Zhang et al. 2019b). Approximately 44% and 36% of the collected battery PM have

been reported in the size range of 0.1-0.8 mm and 0.8-1.7 mm, respectively (Zhang et al. 2019b). Moreover, it has been reported that 11-29% of the cell mass can be lost in the form of particles and gas, depending on the battery charging level and battery material (Lai et al. 2021; Zhang et al. 2019a, 2019b).

The chemical composition of the generated PM depends on many factors, including the battery type, electrolyte material, and state-of-charge (SOC) (Essl et al. 2020). It has been reported that the major elemental components of the emitted particles from an NMC/LMO battery are C, Li, Co, Ni, Mn, and Al (Lai et al. 2021). Other elements including Cu, Fe, K, S, as well as trace amounts of Ba, Ca, Cr, Mg, Mo, Na, Sb, Sn, Sr, Si, Ti, V, Zn, and Zr, have also been reported in chemical speciation of the emitted particles during the thermal runaway of NMC batteries (Zhang et al. 2019b). Metallic elements have been reported to constitute 43% of the total emitted PM mass, indicating that EV LIBs could be among the important contributors of traffic-related PM metals (Zhang et al. 2019b).

At the lower temperatures, as a result of SEI decomposition, the reaction between electrolyte and lithium generates heat and produces gaseous compounds, which are continuously produced through the thermal runaway conditions (Essl et al. 2020). The primary gaseous compounds produced during this process are CH_4, C_2H_4, CO, CO_2, H_2, and HF, depending on the LIB material (Fernandes et al. 2018; Larsson et al. 2016; Sturk et al. 2019). During this stage of LIB degradation, solid materials including LiF, Li_2CO_3, and $LiPF_6$ can also be generated, which can contribute to particle generation upon venting from the battery (Fernandes et al. 2018; Wang et al. 2006; Wilken et al. 2013).

The results of the previous studies clearly show that EV batteries could generate large particles with high metallic content during battery failure conditions. However, none of the previous studies have investigated the contribution of battery emissions to airborne PM. Moreover, there is a significant lack of knowledge about the potential battery emissions during normal operating conditions of the batteries. Therefore, future research is necessary to (i) investigate and quantify EV battery airborne PM emissions in temperatures below the thermal runaway, and (ii) investigate the potential contribution of evaporation/condensation processes in generating particles from the emitted battery gaseous compounds.

1.5 Health effects associated with vehicular non-exhaust emissions

The chemical composition of fine and coarse PM has been shown to affect PM toxicity (Rönkkö et al. 2018; Weichenthal et al. 2019). Non-exhaust PM metals have been associated with various health outcomes, including cancer (Chen et al. 2021; Raaschou-Nielsen et al. 2016), reduced lung function (Huang et al. 2018), cardiovascular and cardiopulmonary diseases (Magari et al. 2002; Samoli et al. 2016; Ye et al. 2018), birth defects (Basu et al. 2014a; Pedersen et al. 2013), and increased mortality (Basagaña et al. 2015; Ostro et al. 2007, 2010, 2015; Valdés et al. 2012; Wang et al. 2017). These metals can trigger oxidative stress by the disproportionate generation and consumption of reactive oxygen species (ROS), including hydroxyl radicals, oxygen radicals, peroxides, and oxygen superoxide (André Nel 2005; Bates et al. 2019; Hadei and Naddafi 2020; Park et al. 2018b; Wei et al. 2009). The oxidative potential is defined as PM capacity for producing ROS, which is substantially dependent on PM chemical composition, and has been used for determining PM toxicity (Fang et al. 2019; Weichenthal et al. 2019; Yadav and Phuleria 2020).

Oxidative potential can be measured by various cellular and acellular assays and have been associated with different health outcomes, including asthma (Yang et al. 2016), diabetes (Strak et al. 2017), and hospital admissions for cardiovascular and respiratory causes (Abrams et al. 2017; Bates et al. 2015; Gao et al. 2020b). PM transition metals can contribute to oxidative

potential since they can catalyze radical reactions and facilitate the oxidation process due to their electronic structure and multiple valences (Miller et al. 1990). Among the transition metals, Cu and Fe, which are both associated with brake wear emissions, have been frequently reported to be correlated with oxidative potential in different studies based on various acellular assays (Gao et al. 2020a, 2020b; Hakimzadeh et al. 2020; Mousavi et al. 2019; Perrone et al. 2019; Rao et al. 2020; Shirmohammadi et al. 2017; Weichenthal et al. 2019; Yang et al. 2015a). Other metals associated with brake and tire wear emissions, including Ba (Mousavi et al. 2019; Perrone et al. 2019; Shirmohammadi et al. 2017; Weichenthal et al. 2019; Yang et al. 2015a), Cr (Crobeddu et al. 2017; Liu et al. 2018), Mn (Gao et al. 2020a; Perrone et al. 2019), Mo (Calas et al. 2018), Sb (Crobeddu et al. 2017; Shirmohammadi et al. 2017), Sn (Calas et al. 2018), Zn (Liu et al. 2018; Shirmohammadi et al. 2017), and Zr (Calas et al. 2018) have also been reported to be associated with oxidative potential. While some studies reported the association of total metals with oxidative potential, other studies documented the association of water-soluble (WS) fraction of metals with oxidative potential as it has been reported that solubility of metals can enhance their bioavailability (Mukhtar and Limbeck 2013). The association of the WS fraction of some of the brake and tire wear metals, including WS- Ba, WS-Cr, WS-Cu, WS-Fe, WS-Mo, and WS-Zn have been documented in a number of studies (Cheung et al. 2012; Hu et al. 2008; Pietrogrande et al. 2018; Shirmohammadi et al. 2015; Strak et al. 2012).

The impact of organic content of vehicular non-exhaust emissions on PM health effects should also be considered. In general, exhaust and non-exhaust traffic emissions both contribute to organic carbon (OC) in urban areas (Amato et al. 2016b; Gianini et al. 2013; Hasheminassab et al. 2014; Taghvaee et al. 2019; Veld et al. 2021; von Schneidemesser et al. 2010). In Los Angeles, the relative contribution of non-exhaust emissions to OC has been reported to increase from 14% in 2005 to 28% in 2015, while the contribution of exhaust emissions to OC has decreased (Altuwayjiri et al. 2021). A considerable fraction of OC has been attributed to road dust, which can also be linked to non-exhaust emissions (Mousavi et al. 2018). As mentioned in previous sections, organic materials have been shown to constitute both brake and tire wear particles (Aatmeeyata and Sharma 2010a; Alves et al. 2020, 2021; Dall'Osto et al. 2014; Gadd and Kennedy 2000; Jan Kole et al. 2017; Park et al. 2017; Plachá et al. 2015). OC, water-soluble OC (WSOC), and volatile organic compounds, including PAHs have been reported to be associated with oxidative potential (Calas et al. 2018; Hakimzadeh et al. 2020; Janssen et al. 2015; Kramer et al. 2021; Liu et al. 2020; Lovett et al. 2018; Mousavi et al. 2019; Perrone et al. 2019; Pietrogrande et al. 2018; Strak et al. 2012, 2017; Taghvaee et al. 2019; Zhang et al. 2016). Some of the organic constituents of brake and tire wear particles, including PAHs, glycerol compounds, heavy alkanes, and phenolic compounds are toxic and contribute to overall PM health effects (Alves et al. 2020, 2021; Markiewicz et al. 2017). For instance, PAHs have been associated with various health outcomes, including birth defects (Ren et al. 2011), cancer (Armstrong et al. 2004; Vardoulakis et al. 2020), and cardiovascular diseases (Marris et al. 2020; Xu et al. 2010) and respiratory diseases (Bortey-Sam et al. 2017). Moreover, it has been shown that the presence of organic material can increase the toxicity of transition metals by increasing the solubility of metals (Gao et al. 2020b; Tapparo et al. 2020).

The majority of previous studies have not separated the contribution of exhaust and non-exhaust emissions to oxidative potential, and they have estimated the overall contribution of vehicular emissions, including gasoline and diesel vehicles (Bates et al. 2015; Hakimzadeh et al. 2020; Liu et al. 2018; Mousavi et al. 2019; Taghvaee et al. 2019). On the other hand, a few studies have distinguished the contribution of exhaust and non-exhaust emission sources through source apportionment techniques (Jeong et al. 2020a; Shirmohammadi et al. 2015, 2016). Jeong et al. (2020) reported a higher correlation between the $PM_{2.5}$ oxidative potential and non-exhaust emissions than exhaust emissions in Toronto, highlighting the role of non-exhaust emissions in

contributing to $PM_{2.5}$ oxidative potential. Shirmohammadi et al. (2016) estimated the contribution of various sources to $PM_{2.5}$ and $PM_{0.18}$ oxidative potential in southern California and reported that while exhaust emissions were a significant source of $PM_{0.18}$ oxidative potential, vehicular abrasion source, including brake and tire wear particles was a significant source of $PM_{2.5}$ oxidative potential. Shirmohammadi et al. (2015) studied the contribution of water-soluble and water-insoluble metals to $PM_{2.5-10}$ oxidative potential and reported vehicular abrasion and resuspended dust as the primary contributors to $PM_{2.5-10}$ oxidative potential in southern California. Future ambient measurement studies with source apportionment can provide a better understanding of the relative contribution of exhaust and non-exhaust emissions to $PM_{2.5}$ and $PM_{2.5-10}$ and their oxidative potential.

1.6 Conclusions and future directions

Exhaust emissions have significantly decreased in different parts of the world, due to the prohibitive environmental policies, as well as scientific improvements in the auto industry that have led to the manufacturing of fuel-efficient vehicles with low exhaust emissions. On the other hand, the role of non-exhaust emissions has been underestimated in the emission inventories, while their relative contribution to traffic-related emissions is on the rise.

Brake components have been reported to have a high metallic content. While Fe has been shown to be the dominant element in brake rotors and brake pads, many other elements, including Al, Ca, Cd, Cr, K, Mn, Mo, Ni, Pb, Si, Ti, Zn, and Zr have also been observed in brake wear particles in fine and coarse PM. Overall, Ba, Cu, Sb, and Sn were found to be the most appropriate brake wear tracers. Moreover, carbonaceous species have been reported to have a considerable contribution to the total weight of brake wear particles. Approximately 150 organic compounds, including n-alkanes, n-alkenes, n-alkanols, glycerol compounds, phenolic compounds, and polycyclic aromatic hydrocarbons (PAHs) have been observed in brake wear particles.

The mass-based particle size distribution of brake wear particles has been reported to be unimodal with mode diameter in the range of 1-10 μm. The reported brake wear particle size distributions have been reported to be variable, depending on the sampling approach and instrumentation. A unimodal, bimodal, and multimodal brake wear particle size distributions with mode diameters in the ultrafine and fine size ranges have been reported in the previous studies. Brake temperature has been shown to have a significant impact on the brake wear particle size distribution. Beyond a specific brake temperature (T_{crit}) in the range of 140-240 ºC, a considerable amount of ultrafine particles are generated, which can impact the brake wear particle size distribution.

It has been shown that approximately 2-7% of the tire tread particles are airborne, while the remainder transport to the aquatic environment or deposit on the roads. Many elements, including Al, Si, Zn, Ca, Fe, K, Mg, Na, K, S, and Ti have been reported to be abundant in tire-road wear particles (TRWPs). Zn and S have been shown to be highly associated with tire treads, while some of the reported elements in TRWPs, including Al, Si, and Ti have been associated with other emission sources, including road dust. Despite the association of Zn with tire tread particles, selecting Zn as a tire wear tracer should be taken with caution due to the association of Zn with other emission sources. Carbonaceous species constitute up to 72% of tire tread weight, and various organic species, including PAHs, n-alkanols, phenolic compounds, levoglucosan, steranes, and aliphatic species have been reported in TRWPs. Overall, TRWPs have shown to be the leading non-exhaust emission source of PAHs. Among the reported PAHs, pyrene, fluoranthene, and naphthalene have been reported to have the highest concentrations.

The airborne tire wear particle size distributions have been reported to be unimodal, bimodal, and multimodal, with mode diameters in the range of 0.01 to 5 μm. Moreover, the majority of the studies reported a minimum of one peak in the ultrafine size range. Tire wear particles can contribute to up to 60% of the microplastics with diameters in the range of 1-1000 μm. The mass-based particle size distributions of larger tire tread particles and TRWPs have been reported to be unimodal with mode diameters of 25 μm, while a bimodal TRWP size distribution has been documented.

Various studies have reported the brake wear $PM_{2.5}$ and PM_{10} emission factors (EFs) to be in the range of 0.5-5.5 and 1.4-80.0 mg km^{-1} Veh^{-1} , respectively. In addition, tire wear $PM_{2.5}$ EF has been shown to be 0.3-11.0 mg km^{-1} Veh^{-1}, while tire wear PM_{10} EFs were 1.9-11.0. Due to the large variation in $PM_{2.5}$ and PM_{10} EFs, emission inventories can be used for comparing the brake and tire wear PM EFs of vehicles. While the United States Environmental Protection Agency (USEPA) estimated lower brake and tire wear $PM_{2.5}$ and PM_{10} EFs, more updated emission inventory models from the European Environment Agency (EEA), estimated higher $PM_{2.5}$ and PM_{10} EFs for the vehicles in different size classes. Moreover, a lower $PM_{2.5}/PM_{10}$ EF ratio is reported by USEPA than EEA, which can have significant policy implications as most of the PM health effects have been documented in the fine size.

The environmental organizations have estimated the brake and tire wear PM EFs of the heavy-duty vehicles (HDVs) to be considerably higher than light-duty vehicles (LDVs) based on their developed emission inventories. It has been shown that vehicle load, number of axles, and vehicle speed can affect the brake and tire wear EFs. Moreover, larger HDVs are mostly equipped with air brakes, which use compressed air instead of brake fluid. Air brakes are generally more sophisticated, and they have a different mechanical design from hydraulic brakes that are used in LDVs. In addition, large HDVs mostly use larger studded tires that can have different characteristics from the tires used in LDVs. Therefore, the brake and tire wear particles from HDVs are expected to have different physical and chemical characteristics. Despite the high brake and tire wear EFs of HDVs, and the potential differences in the characteristics of their tires and brakes, none of the previous studies have directly investigated their brake and tire wear emissions through the on-road sampling or laboratory measurements. This clear gap of knowledge can prohibit building a more detailed emission inventory for non-exhaust traffic emission sources, and it should be addressed in the forthcoming studies in this area.

The vehicle fleet is expected to undergo a dramatic change in the upcoming decade with the fast growth in the electric vehicle (EV) market. While the positive effects of auto electrification in reducing exhaust emissions are undeniable, the contribution of EVs to non-exhaust emission should be taken into consideration. The current EVs are equipped with relatively heavy lithium-ion batteries (LIBs), which can increase the EV brake and tire wear emissions. While regenerative braking technology can reduce the generation of brake wear particles, the decrease in the EV brake wear emissions due to the regenerative braking should be quantified through the laboratory or on-road experiments in the future. Moreover, the previous studies have shown that under abnormal battery conditions such as thermal runaway, LIBs generate toxic gases and large particles with high metallic content, and therefore, LIBs can contribute to non-exhaust emissions. However, previous studies have been limited to investigating battery emissions under abnormal conditions, and they were focused on super-coarse particles. The potential contribution of LIB PM emissions to $PM_{2.5}$ and PM_{10} under normal operation condition should be estimated.

Non-exhaust emissions are an important source of PM metals with various health effects. Metals associated with the brake and tire wear emissions could induce various health outcomes, including cancer, reduced lung function, cardiovascular and cardiopulmonary diseases, birth defects, and increased mortality. Metals associated with brake and tire wear emissions, including

Ba, Cr, Cu, Fe, Mn, Mo, Sb, Sn, Zn, and Zr have been associated with oxidative potential. Moreover, the brake and tire wear organic content includes toxic organic compounds such PAHs, phenolic compounds, and glycerol compounds that can contribute to overall PM health effects.

Despite the growing body of research that emphasizes the role of non-exhaust emissions in contributing to traffic-related air pollution, non-exhaust emissions have not been directly regulated in different parts of the world. While few attempts have been made in Europe and the US to regulate the chemical composition of brake pads and tire treads, the lack of a comprehensive regulation strategy has prevented mitigation of these important traffic sources. The current work aimed to provide a comprehensive understanding of the vehicular non-exhaust emissions, their physical and chemical characteristics, and their associated health effects. This literature review study also provided insight into the future trend of non-exhaust emissions with the growth of auto electrification.

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2 Elemental composition of fine and coarse particles across the Greater Los Angeles area: spatial variation and contributing sources*

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2.1 Abstract

The inorganic components of particulate matter (PM), especially transition metals, have been shown to contribute to PM toxicity. In this study, the spatial distribution of PM elements and their potential sources in the Greater Los Angeles area were studied. The mass concentration and detailed elemental composition of fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) particles were assessed at 46 locations, including urban traffic, urban community, urban background, and desert locations. Crustal enrichment factors (EFs), roadside enrichments (REs), and bivariate correlation analysis revealed that Ba, Cr, Cu, Mo, Pd, Sb, Zn, and Zr were associated with traffic emissions in both $PM_{2.5}$ and $PM_{2.5-10}$, while Fe, Li, Mn, and Ti were affected by traffic emissions mostly in $PM_{2.5}$. The concentrations of Ba, Cu, Mo, Sb, Zr (brake wear tracers), Pd (tailpipe tracer), and Zn (associated with tire wear) were higher at urban traffic sites than urban background locations by factors of 2.6 to 4.6. Both $PM_{2.5}$ and $PM_{2.5-10}$ elements showed large spatial variations, indicating the presence of diverse emission sources across sampling locations. Principal component analysis extracted four source factors that explained 88% of the variance in the $PM_{2.5}$ elemental concentrations, and three sources that explained 86% of the variance in the $PM_{2.5-10}$ elemental concentrations. Based on multiple linear regression analysis, the contribution of traffic emissions (27%) to $PM_{2.5}$ was found to be higher than mineral dust (23%), marine aerosol (18%), and industrial emissions (8%). On the other hand, mineral dust was the dominant source of $PM_{2.5-10}$ with 45% contribution, followed by marine aerosol (22%), and traffic emissions (19%). This study provides novel insight into the spatial variation of traffic-related elements in a large metropolitan area.

2.2 Introduction

Exposures to particulate matter (PM) have been linked to a wide range of chronic and acute health conditions, including cardiovascular diseases, lung cancer, and adverse birth outcomes (Burnett et al. 2018; Gharibvand et al. 2017; Orach et al. 2021; Sapkota et al. 2012). Both fine particles ($PM_{2.5}$, aerodynamic diameter less than or equal to 2.5 μm) and coarse particles ($PM_{2.5-10}$, aerodynamic diameter ranging from 2.5 to 10 μm) have been associated with increased mortality (Wang et al. 2020a; Zhang et al. 2017). While most studies addressing the health effects of PM have focused on undifferentiated PM mass, there are a growing number of studies examining PM chemical components, including metals (Franklin et al. 2008; Rönkkö et al. 2018). It has been reported that the metal components are among the major contributors to the overall health effects of PM (Badaloni et al. 2017; Wallenborn et al. 2009). Metals are known to induce oxidative stress (Araujo and Nel 2009; Gao et al. 2020b), which is one of the most significant physiological mechanisms of PM toxicity (Bates et al. 2019; Miller 2020). A better understanding of the major sources of PM elements in the environment is required for mitigating their associated health effects.

In Southern California, traffic emissions have been estimated to contribute to 32% and 18% of ambient $PM_{2.5}$ and $PM_{2.5-10}$, respectively (Habre et al. 2020a), and they are one of the dominant sources of PM elements (Mousavi et al. 2018). Significant steps have been taken to control traffic emissions through technological advancements and restrictive policies, which have led to

remarkable reductions in exhaust emissions in the past few decades (Pitiranggon et al. 2021; Thorpe and Harrison 2008). With the continuous reductions in exhaust emissions in light of the previous emission control strategies, the relative importance of non-exhaust emission sources is increasing in the metropolitan areas (Jeong et al. 2020a; Oroumiyeh and Zhu 2021).

Brake and tire wear particles and resuspended dust are among the primary contributors to non-exhaust PM (Piscitello et al. 2021). Specific elements have been used as metalss for identifying non-exhaust emission sources (Grigoratos and Martini 2015a; Pant and Harrison 2013a). For brake wear particles, Ba, Cu, and Sb are the most common tracers (Gietl et al. 2010; Schauer et al. 2006a; Sternbeck et al. 2002), while Cr, Fe, Mo, Sn, Ti, and Zr have also been reported as brake wear tracer (Amato et al. 2011a; Apeagyei et al. 2011). For tire wear particles, Zn has been widely used as a tracer since zinc oxide can contribute up to 1% of tire mass (Farahani et al. 2021; Harrison et al. 2012; Milani et al. 2004a; Morillas et al. 2020a). However, Zn is not specific to tire wear because high Zn concentrations have also been reported for other emission sources, including brake wear particles (Lough et al. 2005a) and industrial emissions (Jeong et al. 2019; Morillas et al. 2019). In addition, platinum-group elements (PGEs), including Rh, Pd, and Pt, are primarily generated by catalytic converters in light-duty vehicles (LDVs) and can be used as tailpipe tracers (Bozlaker et al. 2014; Das and Chellam 2020). Moreover, other elements, including Bi, Ce, Ni, and V have been linked to marine fuel emissions (Auffan et al. 2017; Crosignani et al. 2021; Morillas et al. 2019; Spada et al. 2018). On the other hand, using elemental tracers for identifying resuspended road dust should be taken with caution since both crustal and vehicle abrasion sources contribute to resuspended road dust (Denby et al. 2018).

Los Angeles is a megacity and one of the largest metropolitan regions in the United States. Due to its sprawling landscape, Los Angeles is heavily impacted by traffic emissions (Jerrett et al. 2005b; Su et al. 2016). Chemical speciation in the Los Angeles area was first studied a few decades ago (Chow et al. 1994). With advancements in analytical measurement technologies, more recent studies have investigated the spatial variation of a larger spectrum of PM elements in the Los Angeles area (Arhami et al. 2009; Cheung et al. 2011; Habre et al. 2020a; Hasheminassab et al. 2020). While these studies provided valuable insights into the chemical speciation and spatial variation of PM elements, they did not cover a large area with a spatial resolution high enough to facilitate research on the association between PM elements and adverse health outcomes. The current study aims to develop a spatially-resolved chemical speciation dataset for $PM_{2.5}$ and $PM_{2.5-10}$ to investigate spatial variations of traffic-related elements in the Greater Los Angeles area to enable future health effect studies.

2.3 Material and methods

2.3.1 Site selection

This study is part of a larger effort to evaluate the association between brake and tire wear emissions and adverse birth outcomes in Los Angeles. The sampling sites were selected with the goal of maximizing the variability in PM mass concentration and chemical speciation to facilitate exposure modeling across the Los Angeles basin. To select monitoring locations, we developed a multi-criteria selection method that included variables likely to influence the spatial distribution of such as intersection density, traffic density, and slope gradients. We also included the density of PurpleAir sensors to support the co-kriging model. Potential monitoring locations included homes of the women in the PARENT study (described in Chapter 6), homes of the investigators, and government monitoring locations. Each site was ranked based on the multi-criteria selection method. After ranking, sites were divided into quadrants and potential locations were selected for detailed field evaluation that restricted locations to those no more than two stories high, those that

had power, those that had sufficient space for the monitors, and those that were absent other sources of particles such as barbeques.

While the effect of traffic intensity on the $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations was the main focus of the current study, other factors such as road slope and intersection density which likely affect brake and tire wear particles were also considered for sampling site selection (Abu-Allaban et al. 2003; Harrison et al. 2012). Overall, PM samples were collected at 46 locations in the Greater Los Angeles region (Figure S2.1) during two periods, in September 2019 and February 2020. In each field campaign, samples were collected concurrently over a two-week integrated period with four replicate sampling locations between the two periods. While most of the sampling sites were within Los Angeles County, the study also covered the adjacent parts of Riverside, San Bernardino, and Ventura counties.

The sampling sites can be categorized into four groups: **(a)** urban traffic, **(b)** urban community, **(c)** urban background, and **(d)** desert (see Table S2.1 in the supplementary information (SI) for more details). The annual average daily traffic (AADT) data from 2018 were collected from the Federal Highway Administration to compare the traffic volume at different urban sampling sites (Table S2.1). The mean AADT values for each sampling location were calculated based on the AADT values of the major surrounding roads and highways following the buffering method proposed in the previous studies (Henderson et al. 2007a).

2.3.2 Sample collection and analytical methods

Ambient $PM_{2.5}$ and $PM_{2.5-10}$ samples were collected using Harvard cascade impactors (Lee et al. 2006). The cascade impactors were configured for 3-stage collections and were contained inside custom-made pump boxes that operated at 5 LPM. The first stage of the impactor collected super-coarse particles (aerodynamic diameter larger than 10 μm) using pre-cleaned 3/4" diameter Polyurethane foam (PUF) substrates, the second stage collected coarse particles using 3/8" diameter pre-cleaned PUF, and the final stage was configured with pre-cleaned Teflon membrane filters (Teflo, 2 μm, Pall Life Sciences, 37 mm) to collect $PM_{2.5}$. The filters, PUF substrates, and the components of the impactors were rigorously pre-cleaned in the trace element laboratory at the University of Wisconsin-Madison State Laboratory of Hygiene (WSLH) prior to use, following protocols established in previous studies (Dillner et al. 2007; Lough et al. 2005a). The impactors were pre-loaded in the laboratory to minimize potential field-handling contamination.

The PM mass loading on all three substrates was determined by micro-gravimetry (weighting precision = 0.001 μg) at the WSLH. The substrates were equilibrated in the dedicated temperature (21±1.5 °C) and humidity-controlled (40±3%) weighing room for 30 hours before taring and post-weighing. Active ionization sources, including Po strips were installed on the microbalances to remove static charges during weighing. The concentrations of 55 chemical elements in $PM_{2.5}$ and $PM_{2.5-10}$ were determined by Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS) (Thermo-Finnigan Element 2XR) as previously detailed (Dillner et al. 2007; Herner et al. 2006a; Lough et al. 2005a; Pakbin et al. 2011) (For more information about the quality assurance/quality control procedure and the effect of relative humidity on $PM_{2.5}$ and $PM_{2.5-10}$ concentrations, please refer to section S1.1 in the SI). The signal-to-noise (S/N) ratios were calculated based on the previously suggested method (Norris et al. 2014). In this study, elements with concentrations exceeding the method detection limit (MDL) in at least 80% of the samples and S/N ratios greater than two were included in subsequent data analysis. Overall, 43 elements were included for data analysis (see Table S2.2 in SI).

2.3.3 Statistical analysis

Bivariate Spearman correlation analysis was performed to determine the associations between measured elements. The elements with large anthropogenic and crustal contributions were

identified by the crustal enrichment factors (EFs) (Harrison et al. 2003; Salomons and Förstner 1984), calculated using Eq (2.1):

$$EF_x = \frac{\left(\frac{C_x}{C_{ref}}\right)_{Sample}}{\left(\frac{C_x}{C_{ref}}\right)_{Crust}} \quad (2.1)$$

Where C_x is the concentration of the x element in the field sample, and C_{ref} represents the concentration of the reference element. The elemental concentrations of the upper continental crust (UCC) were used as the crustal concentrations in Eq (1) for estimating EFs (Taylor and McLennan 1985), and Al was used as the reference element (Birmili et al. 2006; Gao et al. 2002).

In order to assess the impact of traffic emissions on $PM_{2.5}$ and $PM_{2.5\text{-}10}$ elements, roadside enrichments (REs) were calculated based on Eq (2.2):

$$RE_i = \frac{C_i^H - C_i^L}{C_i^H} \quad (2.2)$$

Where C_i^H and C_i^L, represent the concentration of the i^{th} element at high and low traffic sites, respectively (Amato et al. 2011c; Oliveira et al. 2010). The calculated REs were based on concentrations at three pairs of sampling locations: **(a)** urban community vs. urban background sites, **(b)** urban traffic vs. urban background sites, and **(c)** urban traffic vs. urban community sites. A similar approach was used in a previous study for identifying metals associated with traffic emissions (Amato et al. 2011c). All elements were categorized into three different groups representing strong, weak, and inconsistent roadside enrichment levels (i.e., Types I, II, and III) (see Table S2.3 for details).

Principal component analysis (PCA) was separately applied to the $PM_{2.5}$ and $PM_{2.5\text{-}10}$ elemental concentration datasets, each with 50 samples to identify the major sources of PM elements. Overall, the standardized format of 43 elements was included in PCA. Varimax orthogonal rotation was used to optimize the variance between the components, and an eigenvalue of unity or larger was used as a threshold for the inclusion of the extracted source factors in PCA. Based on the resulting principal component loadings, the $PM_{2.5}$ and $PM_{2.5\text{-}10}$ sources were identified using the bivariate correlation analysis results, EFs, and REs.

Two separate linear regression models were further developed to compare the sources contributing to $PM_{2.5}$ and $PM_{2.5\text{-}10}$ based on methods suggested in a previous study (Larsen and Baker 2003). In each model, ambient $PM_{2.5}$ and $PM_{2.5\text{-}10}$ concentrations were treated as dependent variables, and their corresponding principal component scores were the independent variables. The linear regression models were developed in a stepwise sequence with the aim of maximizing the coefficient of determination (R^2 values) by including noncolinear and statistically significant source factors (p<0.05).

Descriptive statistical analysis, PCA, multi-variable linear models, and significance tests were performed using SPSS 27.0 (IBM, Armonk, NY). Sigmaplot 12.5 (Systat, San Jose, CA) was used for plotting figures.

2.4 Results and Discussion

2.4.1 Spatial variation of PM mass concentrations

The $PM_{2.5}$ and $PM_{2.5-10}$ mass concentrations at the urban traffic, urban community, urban background, and desert sampling sites are presented in Figure 2.1. The average concentrations of $PM_{2.5}$ at those sites were 10.9 μg/m^3, 8.9 μg/m^3, 6.8 μg/m^3, and 10.5 μg/m^3, respectively, while the average concentrations of $PM_{2.5-10}$ at those sites were 9.6 μg/m^3, 8.3 μg/m^3, 7.4 μg/m^3, and 12.4 μg/m^3, respectively. The $PM_{2.5}/PM_{2.5-10}$ ratios were slightly higher at the urban traffic (0.53) and urban community (0.52) sites than at the urban background (0.48) and desert (0.46) sites. Overall, a statistically significant difference was observed between the $PM_{2.5}/PM_{2.5-10}$ ratios across various sampling locations (Kruskal-Wallis method ($p < 0.05$)). Higher $PM_{2.5}/PM_{2.5-10}$ ratios at the urban traffic and urban community sites were presumably due to fresh emissions from traffic and industrial sources. In Europe, lower $PM_{2.5}/PM_{2.5-10}$ ratios were reported at locations with dry weather conditions or in close proximity to desert dust (Eeftens et al. 2012; Querol et al. 2008), while higher $PM_{2.5}/PM_{2.5-10}$ ratios were reported at urban locations with high traffic volumes (Amato et al. 2016a). Similar findings were reported in Southern California, showing that desert sampling sites had the lowest $PM_{2.5}/PM_{2.5-10}$ ratios, while urban sampling locations had higher $PM_{2.5}/PM_{2.5-10}$ ratios (Kim et al. 2000; Motallebi et al. 2003).

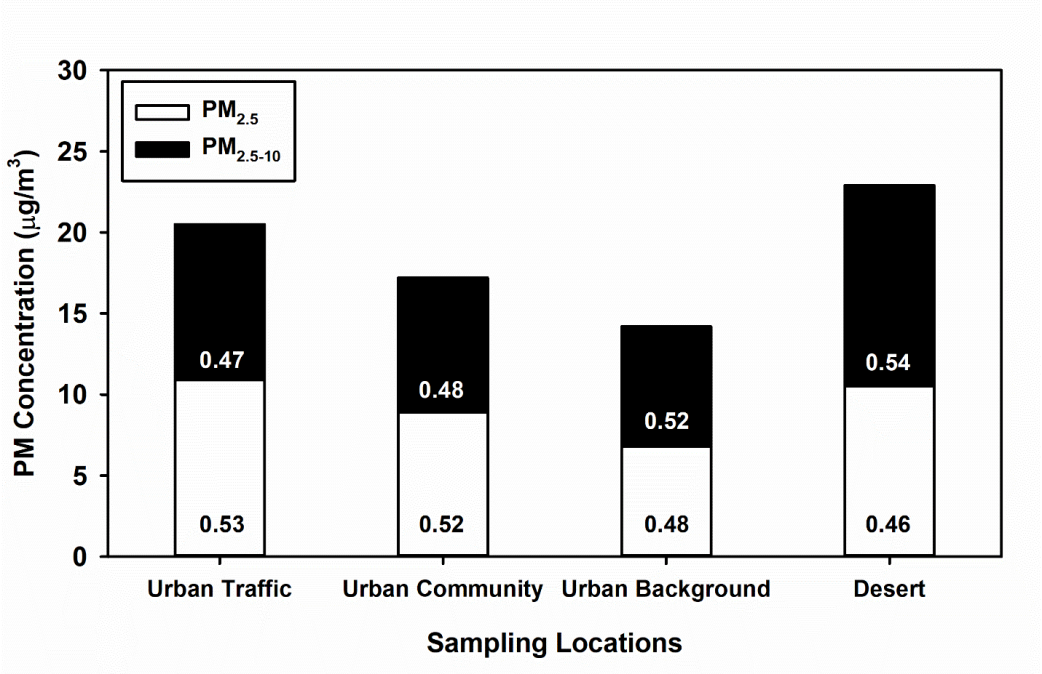

Figure 2.41: PM2.5 and PM¬2.5-10 concentrations at different sampling locations. Denoted numbers represent PM2.5/PM10 and PM2.5-10/PM10 ratios.

2.4.2 $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations and their spatial variation

Figure 2.2 presents absolute average elemental concentrations in $PM_{2.5}$ and $PM_{2.5-10}$ at different sampling locations. The most abundant elements in both $PM_{2.5}$ and $PM_{2.5-10}$ were Na, S, Fe, Ca, Al, Mg, K, Ba, and Ti. The majority of these elements are naturally abundant in the crust of the earth, while S is also present in the secondary inorganic sulfate. As presented in Figure 2.2, while the concentrations of crustal elements such as Al, Ca, Rb, K, Ti, and Fe were higher in coarse particles, the levels of many of the brake and tire wear tracers (Mo, Cr, Sb, Zn), as well as S, Pb,

and Ni were higher in PM2.5. The concentrations of Ba, Cu, and Zr in $PM_{2.5}$ and $PM_{2.5-10}$ were comparable. These results are in agreement with findings of previous studies that reported crustal elements such as Al, Ca, Mg, Na, and Si were more abundant in $PM_{2.5-10}$ (Kim et al. 2000; Lough et al. 2005a). In addition, other trace elements, including Cd, Pb, and Ni were shown to have higher concentrations in $PM_{2.5}$ than $PM_{2.5-10}$ in previous studies (Poulakis et al. 2015; Querol et al. 2008). Tables S2.4 and S2.5 in SI summarizes the absolute and normalized elemental concentrations in $PM_{2.5}$ and $PM_{2.5-10}$, respectively. Figure S2.2 presents the relative contributions of $PM_{2.5}$ elements to the elemental concentrations in PM_{10}.

Figure 2.3 represents concentration ratios (panels a and b) and crustal enrichment factor (EF) ratios (panels c and d) of selected traffic (i.e., Ba, Cr, Cu, Mo, Pd, Sb, Zn, and Zr) and crustal (i.e., Ca, Fe, K, Li, Mn, Rb, Ti) elements at the urban traffic, urban community, and desert sites for both fine and coarse particles (Amato et al. 2011a; Apeagyei et al. 2011; Gietl et al. 2010; Pakbin et al. 2011; Schauer et al. 2006a; Sternbeck et al. 2002). To calculate the concentration and EF ratios, elemental concentrations and EFs at different sampling sites were normalized by the average concentrations and EFs at the urban background sites, respectively. As shown in Figure 2.3 (panels a and b), urban traffic sites had higher levels of brake and tire wear tracers in comparison to other sampling sites. At the urban traffic sites, the average concentrations of Ba, Cu, Mo, Sb, and Zr in $PM_{2.5}$ and $PM_{2.5-10}$ were 3.3-4.6 and 2.6-4.4 times higher than urban background sites, respectively. In previous studies, the concentrations of traffic tracers such as Ba and Cu in $PM_{2.5}$ were shown to be 3.7 and 4.4 times higher at a freeway site compared to background downtown sites (Jeong et al. 2019). Moreover, the concentrations of the heavy metals in $PM_{2.5-10}$ have been previously reported to be up to 10 times higher at locations with high traffic intensity compared to rural sites (Pakbin et al. 2011). The higher concentrations of Ba, Cu, Sb, and Zn at the urban locations have been attributed to traffic sources, and particularly brake and tire wear emissions (Querol et al. 2007). On the other hand, a large increase in the concentrations of crustal elements was not observed at the urban traffic sites. In fact, average concentrations of crustal elements were only increased by a factor of 1.1-2.1 for $PM_{2.5}$, and 1.1-1.5 for $PM_{2.5-10}$. In contrast, desert sites had the highest concentrations of crustal elements.

The elements with significant anthropogenic sources were identified by EF ratios. As shown in Figure 2.3, in both $PM_{2.5}$ and $PM_{2.5-10}$, the increase in concentrations of traffic tracers at the urban traffic locations led to higher EF ratios, indicating a strong influence of traffic activity on these elements. A similar effect was observed for EF ratios of traffic elements at the urban community locations, while EF ratios of these elements were consistently lower than unity at the desert sites with low traffic activity. On the other hand, while the concentrations of crustal elements at desert sites were up to 3 times higher than those at the background locations, the normalized EFs remained close to unity. Among crustal elements, Fe, Li, Mn, and Ti at the urban traffic sites showed EF ratios greater than unity in $PM_{2.5}$, while their EF ratios were closer to unity in $PM_{2.5-10}$. This indicates that while Fe, Li, Mn, and Ti in $PM_{2.5-10}$ mostly originated from geogenic activities, there were also anthropogenic sources for these elements in $PM_{2.5}$ at the urban traffic locations. Figure S2.3 in SI presents the calculated EFs for $PM_{2.5}$ and $PM_{2.5-10}$ elements.

Figure S2.4 presents the Spearman correlation coefficients between AADT values and selected $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations. As shown in Figure S2.4, traffic elements showed a moderate correlation ($\rho > 0.5$) with AADT, except for Zn in $PM_{2.5}$ and Pd in $PM_{2.5-10}$. On the other hand, the crustal elements were poorly correlated ($\rho < 0.4$) with AADT, except for Ti and Fe. It is important to note that while AADT can affect the concentrations of traffic elements, other factors, including wind direction and proximity to areas with high braking activity, including freeways exit ramps, and locations with stop-and-go traffic can also affect the traffic-related elemental concentrations (Abu-Allaban et al. 2003; Harrison et al. 2012). Table S2.6 in the SI section

presents the $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations of the selected traffic elements in all of the collected samples.

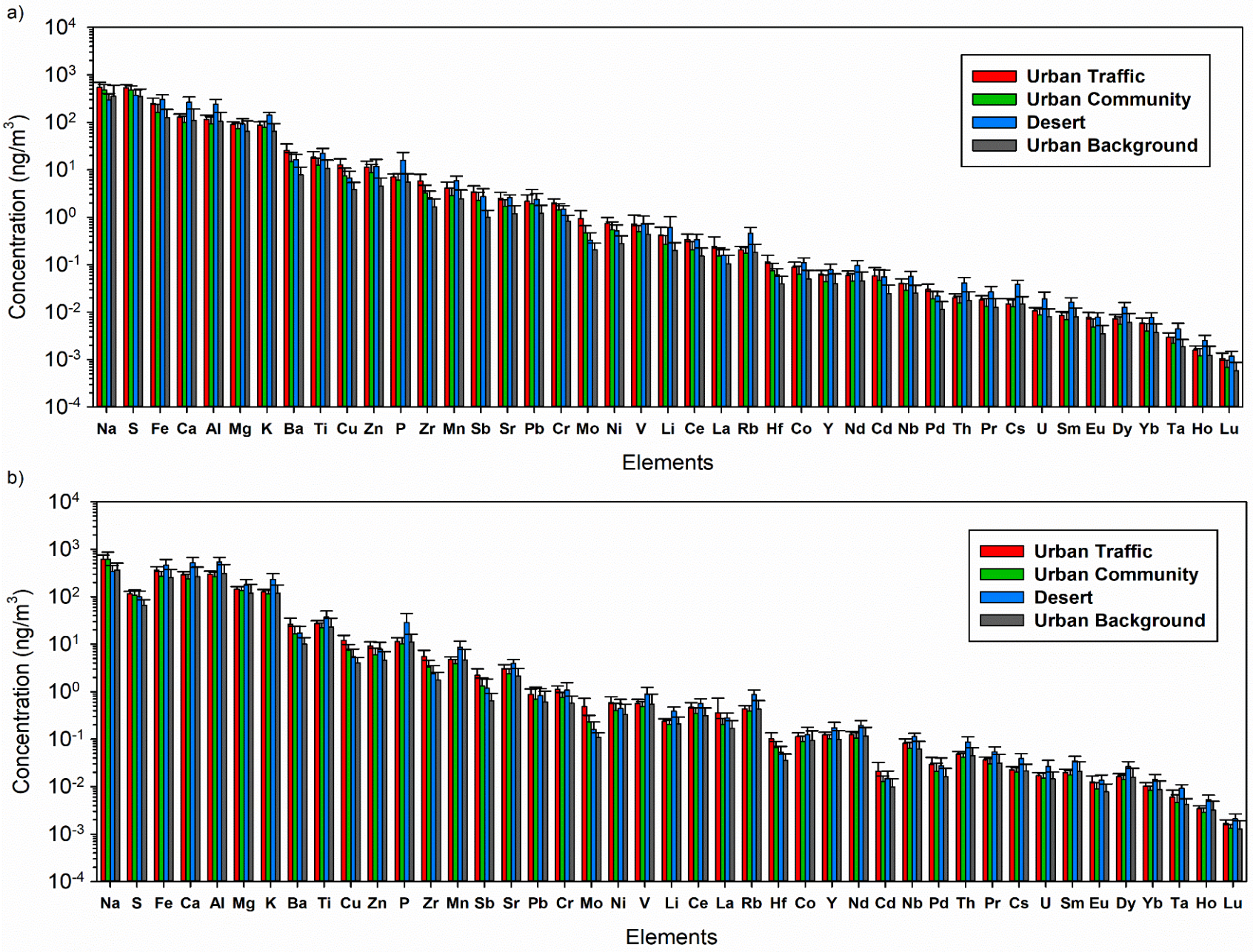

Figure 2.2. Absolute elemental concentrations in (a) $PM_{2.5}$ and (b) $PM_{2.5-10}$ at different sampling locations

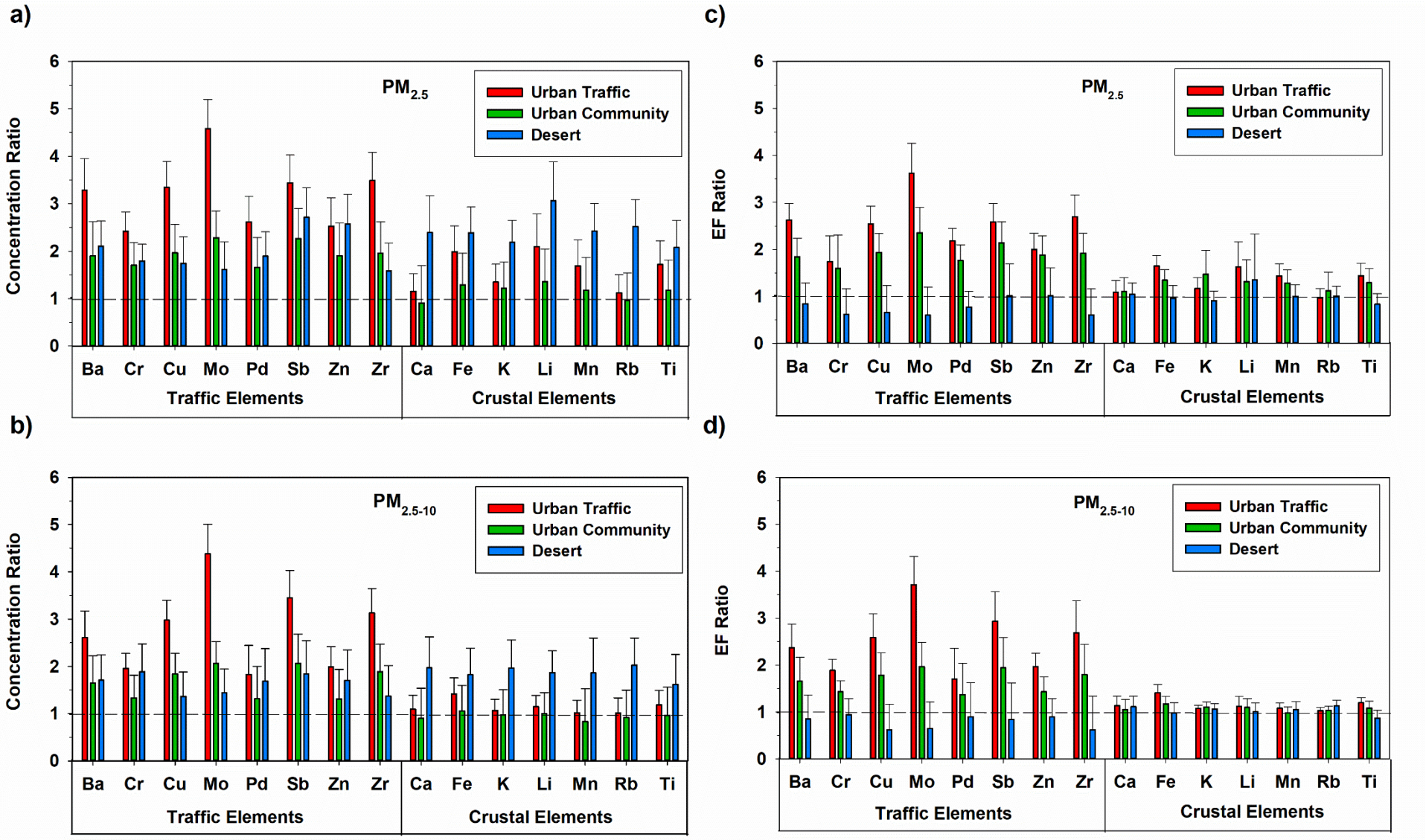

Figure 2.3: Concentration and crustal enrichment factor (EF) ratios at the urban traffic, urban community, and desert sites normalized by the values in urban background sites for the selected crustal and traffic elements (a) PM2.5 elemental concentrations, (b) $PM_{2.5-10}$ elemental concentrations, (c) EF values of $PM_{2.5}$ elements, (d) EF values of $PM_{2.5-10}$ elements

2.4.3 Variability in the chemical composition of $PM_{2.5}$ and $PM_{2.5-10}$

Figure 2.4 shows the coefficient of variation (CV = standard deviation/mean) for $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations. In the literature, a CV > 20% has been used as a threshold to indicate that an element is heterogeneous within an urban environment (Wilson et al. 2005). As shown in Figure 2.4, the elemental concentrations showed a notably higher variation than $PM_{2.5}$ and $PM_{2.5-10}$ mass concentrations across different sampling sites. Overall, elemental concentrations were more variable in fine particles than in coarse particles, except for a few elements such as Mo, La, P, and Ta.

Figure S2.5 compares the concentrations of selected $PM_{2.5}$ and $PM_{2.5-10}$ elements in the current study and other cities, including Barcelona, Florence, Lecce, and Taipei City (Amato et al. 2016a; Hsu et al. 2019; Perrone et al. 2019). As shown in Figure S2.5, while the $PM_{2.5}$ elemental concentrations of Al, Ba, Cu, and Sb were higher in Los Angeles, the levels of Cr, Mo, V, and Zn were relatively lower. In comparison, the $PM_{2.5-10}$ elemental concentrations in Los Angeles were within the range of the reported concentrations in other cities. It should be noted that the differences across the studies in terms of the type and number of the sampling locations, as well as the sampling seasons, could also contribute to the observed differences.

Figure S2.6 presents the normalized concentrations of selected elements based on the mean concentrations reported by a previous study of Southern California communities in 2008-2009 (Habre et al. 2020a) (see the description of Figure S2.6 in the SI section for more information about the methodology comparison between the previous and current studies). With respect to the concentrations reported by Habre et al. (2020), the average $PM_{2.5}$ concentration is approximately 35% lower while the majority of $PM_{2.5}$ elemental concentrations present higher values. For instance, at the urban traffic sites, $PM_{2.5}$ elemental concentrations of Ba, Cr, Cu, Mo, Sb, and Pd were 3.2 to 6.8 times higher than previously reported concentrations. Moreover, concentrations of Al, Ca, Dy, Fe, K, Li, Mn, Rb, and Ti in $PM_{2.5}$ were 3.1 to 5.2 times higher at the desert locations compared to the reported concentrations by Habre et al. (2020). In contrast to $PM_{2.5}$, $PM_{2.5-10}$ elemental concentrations were not substantially different from the previously reported concentrations.

At the urban traffic sites, the concentration of Li in $PM_{2.5}$ was 3.7 times higher than the previous findings, showing the largest concentration increase among the crustal elements. Moreover, crustal enrichment factors (EFs) of Li were compared to the reported average EFs at industrial and near-roadway sampling locations close to the Port of Los Angeles (Arhami et al. 2009). The EFs of Li in $PM_{2.5}$ and $PM_{2.5-10}$ have increased by factors of 5.3 and 3.0, respectively, compared to the previously reported EFs (Arhami et al. 2009). In fact, Li had the largest increase in EFs among all of the reported elements by Arhami et al. (2009). This suggests that new anthropogenic sources for Li might be introduced in the Greater Los Angeles area during the past few years. The vehicle fleet composition in Los Angele County shows that the number of electric vehicles has substantially increased from approximately 600 vehicles in 2010 to 369,300 vehicles in 2020 (California Energy Commission 2021). Therefore, one of the potential sources of Li at the urban traffic locations could be emissions from the venting of Li-ion batteries used in electric vehicles. Li-ion batteries could degrade over time and generate gaseous chemicals (e.g., HF, C_2H_5F) and solid species (LiF) as decomposition byproducts (Sturk et al. 2019; Wilken et al. 2013). The battery degradation can be accelerated at high temperatures or with battery aging (Yang et al. 2018). Given the foreseeable surge in electric vehicle adoption, future studies are needed to focus on Li emissions from Li-ion batteries.

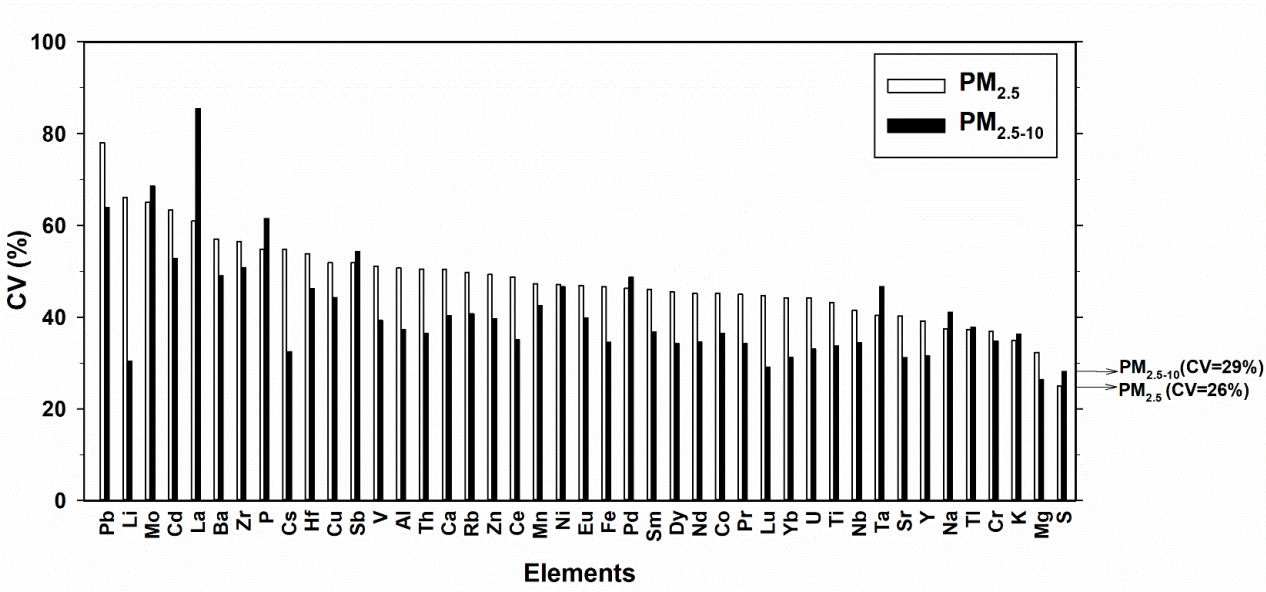

Figure 2.2: Coefficient of variation (CV) of PM2.5 and PM2.5-10 elements based on absolute elemental concentration

2.4.4 Associations between $PM_{2.5}$ and $PM_{2.5-10}$ chemical elements

Spearman correlation coefficients (ρ) for $PM_{2.5}$ and $PM_{2.5-10}$ elements are shown in Figure S2.7. The majority of elements associated with exhaust (Pd) and non-exhaust roadway emissions (Ba, Cr, Cu, Sb, Zn, Zr) showed relatively strong inter-element correlations (ρ >0.70) in $PM_{2.5}$ and $PM_{2.5-10}$. In particular, Ba, one of the common brake wear tracers, was strongly correlated (ρ >0.90) with other brake wear tracers, including Cu, Sb, and Zr in $PM_{2.5}$ and $PM_{2.5-10}$. Similarly, Pd, a tailpipe tracer, showed a strong correlation (ρ >0.90) with Ba in both $PM_{2.5}$ and $PM_{2.5-10}$. In addition, Mo showed moderate positive correlations with other traffic-related elements (ρ >0.50), except for Pd in $PM_{2.5-10}$ (ρ <0.50).

In general, crustal elements, including Al, Ca, and K, showed relatively strong inter-element correlations (ρ >0.70) in both $PM_{2.5}$ and $PM_{2.5-10}$. Relatively strong correlations (ρ >0.70) were observed for Fe with most of the traffic-related elements in $PM_{2.5}$ and $PM_{2.5-10}$. A few other elements, including Li, Mn, and Ti showed moderate to strong correlations with traffic-related elements in $PM_{2.5}$ except for Mo and Cr. However, the correlations of Li, Mn, and Ti with traffic-related elements in $PM_{2.5-10}$ were generally weaker. This agrees with the findings in section 2.2, which discussed the effect of traffic emissions on Li, Mn, and Ti in $PM_{2.5}$.

Correlations among Mg, Na, and V were fairly strong in $PM_{2.5}$. While V has been attributed to marine fuel emissions, especially near the ports, Na and Mg are associated with sea salt aerosol (Amato et al. 2016a; Pakbin et al. 2011). The high correlation between Na, Mg, and V likely indicates mixed marine aerosols with emissions from the ports of Long Beach and Los Angeles. On the other hand, in $PM_{2.5-10}$, V was not correlated with Na, and both Mg and V showed stronger correlations with crustal elements.

It should be noted that compared with the average concentrations reported by Habre et al. (2020), the concentrations of Ni and V in $PM_{2.5}$ were lower in the current study, except for Ni at the urban traffic locations (Figure S2.6). During the past decade, a few regulations were implanted to control sulfur in marine fuel, which led to a decrease in Ni and V content of the fuel oils (Spada et al. 2018). These results show that while Ni and V concentrations have decreased during the past decade as a result of the implemented regulations on fuel sulfur content, their concentrations near

the ports are still higher than other sampling locations. As shown in Figure S2.8, the majority of locations with V concentrations of 1.0 ng/m^3 and greater in fine particles were located within 8 miles of the Port of Long Beach. A similar effect was observed for Ni concentrations, which were constantly above 0.6 ng/m^3 at the sampling sites near the ports.

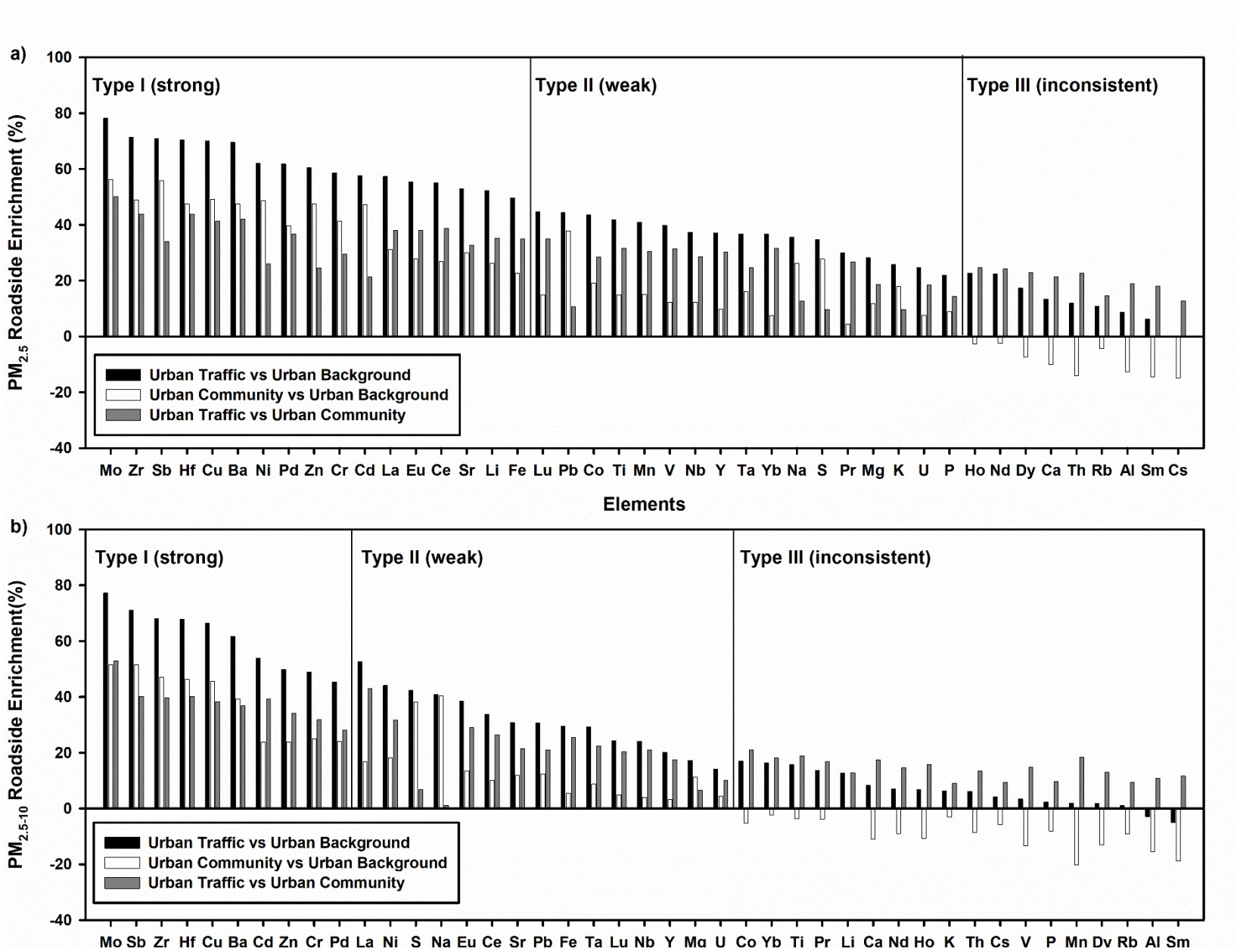

Figure 2.3: Roadside enrichments for different types of elements (a) $PM_{2.5}$, (b) $PM_{2.5-10}$

2.4.5 Identification of traffic-related elements using roadside enrichments (REs)

Figure 2.5 and Table S2.7 present the estimated roadside enrichments (REs) of the $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations. The influence of traffic on $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations was determined by categorizing the elements into three different classes representing strong, weak, and inconsistent REs (see Table S2.3). All traffic-related tracers, including Ba, Cr, Cu, Zn, Zr, Mo, Pd, and Sb were categorized as Type I elements with strong RE (RE>20% for all pairs) in both fine and coarse particles. Among traffic tracers in $PM_{2.5}$, the REs of Ba, Cu, Mo, and Zr were higher than 40% for each pair of sampling location types. Type II elements that showed weak RE (positive RE with RE<20% for at least one pair) included elements associated with marine aerosol (Na, Mg), as well as S, Pb, and some of the lanthanoid elements (Lu, Y, Eu) in fine and coarse particles. Crustal elements such as Al, Ca, Rb, as well as some of the rare-earth elements such as Nd, Sm, Dy, Ho were categorized as Type III elements with inconsistent RE (RE<0 for at least one pair).

While for $PM_{2.5}$ most of the elements were categorized as Type I, for $PM_{2.5-10}$ the majority of the elements were classified as Type III. This indicates that while several elements are dominated by traffic emissions in $PM_{2.5}$, they are likely from multiple sources in $PM_{2.5-10}$. For instance, Fe and Li were assigned to Type I in $PM_{2.5}$ due to their roadside enrichments, however in $PM_{2.5-10}$ they met the criteria for Type II and Type III elements, respectively.

Analyses of crustal enrichment factor ratios, bivariate correlation, and roadside enrichments revealed that Ba, Cr, Cu, Mo, Pd, Sb, Zn, and Zr were largely impacted by traffic emissions in both $PM_{2.5}$ and $PM_{2.5-10}$. Previously, Ba, Cr, Cu, Mo, Sb, and Zr have been associated with brake wear particles (Amato et al. 2011a; Apeagyei et al. 2011; Gietl et al. 2010), while Zn has been mostly used as a tracer of tire wear particles (Harrison et al. 2012; Wagner et al. 2018). In addition, Pd is used in catalytic converters in internal combustion engine vehicles and has been used as a tracer for exhaust emission (Bozlaker et al. 2014; Das and Chellam 2020). Moreover, based on the findings of the previous sections, it is inferred that Fe, Mn, Li, and Ti show a different behavior in fine and coarse particles. The previous studies have linked Mn to crustal (Cesari et al. 2016), traffic (Lawrence et al. 2013; Schauer et al. 2006a; Wang et al. 2021), and industrial sources (Querol et al. 2007). While Fe has been linked to traffic (Amato et al. 2009; Schauer et al. 2006a) and road dust (Pakbin et al. 2011), Ti has also been associated with mineral dust (Amato et al. 2011c) and non-exhaust traffic sources (Apeagyei et al. 2011). Therefore, these elements are associated with traffic emissions in $PM_{2.5}$, while they can be generated by other emission sources in $PM_{2.5-10}$.

2.4.6 Source identification using principal component analysis (PCA)

The major sources of PM elements in the region were determined by applying PCA to $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentration datasets. The analyses of Spearman correlation, EFs, and REs were used to help assign sources to PCA factors, and elements with PCA loadings of 0.5 and higher were used for source identification. Table S2.8a shows PCA results for $PM_{2.5}$ chemical speciation data. For $PM_{2.5}$, four principal components explained 88% of the total variance. The first principal component (PC1), responsible for 42% of the total variance, has substantial contributions from the crustal elements such as Rb, Al, Ca, and K, and it potentially indicates mineral dust. Moreover, this factor has robust loadings from the rare-earth elements such as Dy, Ho, Lu, Nd, Pr, Sm, Y, and Yb that are typically found in mineral dust (Amato et al. 2016a; Pakbin et al. 2011). The common traffic tracers, including brake wear tracers (Ba, Cu, Mo, Sb, and Zr), tire wear tracer (Zn), and tailpipe tracer (Pd), largely contributed to PC2, which accounts for 31% of the total variance. The elements associated with marine aerosol, including Na and Mg had a substantial contribution to PC3. This factor also explains most of the variability for V and S. While S can have various sources, including marine aerosol, V is indicative of marine fuel emissions (Amato et al. 2016a; Farahani et al. 2021; Morillas et al. 2020b; Spada et al. 2018), and therefore,

PC3 is a mixed source with contributions from marine aerosol and fuel combustion, accounting for 11% of the total variance. The fourth principal component (PC4) accounts for 5% of the total variance, and it has a high contribution from Cd with lower contributions from Pb, Ni, and Zn. The association of Cd with combustion emissions and metal industries has been previously reported (Liang et al. 2017; Pacyna and Pacyna 2001; Rossini et al. 2005). In addition, Ni and Pb have also been associated with steel production emissions and industrial fuel combustions (Querol et al. 2007), suggesting that PC4 potentially represents industrial emissions.

Table S2.8b presents the PCA results for $PM_{2.5-10}$ elements. Three components were extracted, which accounted for 86% of the total variance. The majority of crustal elements such as Al, Rb, Ca, as well as rare-earth elements are contained in PC1, presumably indicating mineral dust. The brake, tire, and tailpipe tracers such as Ba, Cu, Mo, Pd, Sb, Zn, and Zr are included in PC2 , which can be attributed to traffic emissions. The contribution of Na, S, and Mg to PC3 likely represents marine aerosol with some contributions from fuel combustion. The mineral dust, traffic, and marine aerosol source factors are responsible for 52%, 26%, and 8% of the total variance, respectively.

While Fe, Li, Mn, and Ti, showed comparable loadings in the mineral dust and traffic source factors in $PM_{2.5}$, they were largely attributed to the mineral dust source factor in $PM_{2.5-10}$. This agrees with the findings in the previous sections, showing that Fe, Li, Mn, and Ti originated from traffic emissions in $PM_{2.5}$, while their concentrations in $PM_{2.5-10}$ are associated with other sources. Similarly, while V was largely attributed to marine aerosol in $PM_{2.5}$, the mineral dust source factor had the highest V loading in $PM_{2.5-10}$.

2.4.7 Comparison of the major sources contributing to $PM_{2.5}$ and $PM_{2.5-10}$

To better understand the major sources of $PM_{2.5}$ and $PM_{2.5-10}$, multiple linear regression analysis was used to investigate the association of the extracted PCA source factors with $PM_{2.5}$ and $PM_{2.5-10}$ mass concentrations. Using the extracted principal component scores as independent variables and the mass concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ as dependent variables, two separate stepwise linear regression models were examined. Eq (2.3) shows the resulting equation for predicting $PM_{2.5}$ concentrations:

$$\hat{Z}_1 = 0.49\,FS_1 + 0.60\,FS_2 + 0.39\,FS_3 + 0.18\,FS_4 \quad (R^2 = 0.76) \tag{2.3}$$

Where $\hat{Z}_1$ is the standardized format of $PM_{2.5}$ mass concentration, and *FS1 – FS4* represent the factor scores for mineral dust, traffic, marine aerosol, and industrial emissions, respectively (Tables S2.9a) (Larsen and Baker 2003). Based on the standardized regression coefficients and coefficients of determination (R^2), the relative contribution of various sources to $PM_{2.5}$ was estimated (Larsen and Baker 2003; Mousavi et al. 2019; Taghvaee et al. 2019). Traffic emissions had the largest contribution (27%) to $PM_{2.5}$ mass concentrations, followed by mineral dust (23%), marine aerosol (18%), and industrial emissions (8%), while the source of 24% of $PM_{2.5}$ mass was undetermined.

Likewise, Eq (2.4) represents the linear relationship for $PM_{2.5-10}$

$$\hat{Z}_2 = 0.78\,FS_1 + 0.33\,FS_2 + 0.38\,FS_3 \quad (R^2 = 0.86) \tag{2.4}$$

Where $\hat{Z}_2$ represents the standardized format of $PM_{2.5-10}$ concentration, and *FS1 – FS3* represent the factor scores for mineral dust, traffic, and marine aerosol, respectively (Tables S2.9b). Mineral dust was the dominant source of coarse particles with 45% contribution to $PM_{2.5-10}$, while the

contribution of marine aerosol and traffic emissions were 22% and 19%, respectively, and 14% of $PM_{2.5-10}$ mass had an undetermined source.

It is important to note that the presented source apportionment results are solely based on the $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations, and other important components of fine and coarse particles, including carbonaceous species, have not been included in this model. Therefore, the presented source apportionment results might not provide a comprehensive picture of the factors contributing to $PM_{2.5}$ and $PM_{2.5-10}$ concentrations in the region. To assess the performance of the model at different sampling locations, an analysis was performed on the model residual values. As shown in Figure S2.9, the mean absolute percentage error (MAPE) values were lower at the urban traffic and desert locations, but higher at the urban background and urban community locations (Bai et al. 2019). This finding indicates that the model has a better performance at locations with high elemental concentrations (urban traffic and desert locations), while showing a reduced prediction accuracy at locations with lower elemental concentrations. This could be due to the contribution of other sources, including secondary aerosols, which have not been accounted for in this model.

2.5 Conclusions

$PM_{2.5}$ and $PM_{2.5-10}$ samples were collected at 46 locations in the Greater Los Angeles area representing urban background, urban community, urban traffic, and desert sites. For $PM_{2.5}$, traffic emissions had the largest contribution (27%), followed by mineral dust (23%), marine aerosol (18%), and industrial emissions (8%). For $PM_{2.5-10}$, mineral dust was the dominant source with 45% contribution followed by marine aerosol (22%) and traffic emissions (19%). For elements, Ba, Cr, Cu, Mo, Pd, Sb, Zn, and Zr were affected by traffic emissions in both $PM_{2.5}$ and $PM_{2.5-10}$. However, Fe, Li, Mn, and Ti were associated with traffic emissions mostly in $PM_{2.5}$. Comparison of PM elemental concentrations to previously reported levels in the region showed a decrease in Ni and V in $PM_{2.5}$, presumably due to the regulations on fuel oil sulfur content. In contrast, large increases in crustal enrichment factors of Li in $PM_{2.5}$ and $PM_{2.5-10}$ were observed, which might be associated with Li emissions from the degradation of electric vehicle Li-ion batteries. In addition, the contribution of other potential Li sources, including lubrication oils and greases should be investigated in future research. Overall, these results provide insight into the spatial distribution of PM elements across a large metropolitan area and underscore the role of both exhaust and non-exhaust traffic emissions in generating PM elements.

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2.7 Appendix A.1 Supplementary information of chapter 2

Table S2.1 Detailed description of the sampling sites including 24 locations in summer (September 2019) and 26 locations

***Four sites repeated between summer and winter**

Type	Description	Season	Number of sampling Locations*	Cities included in sampling	Annual average daily traffic (AADT)	Average relative humidity
Urban Traffic	Near-roadway and urban locations near major roads and intersections with high traffic volume	Summer	6	Compton, Long Beach, Los Angeles	50,400 ± 32,000	60%
		Winter	7	Compton, Long Beach, Los Angeles		
Urban Community	Urban residential locations and suburban areas with moderate traffic	Summer	13	Chino Hills, Hawthorne, Pasadena, La Miranda, Los Angeles, Santa Monica	18,100 ± 17,700	61%
		Winter	15	Altadena, El Monte, Gardena, Lakewood, Los Angeles, Rancho Palos Verdes, Torrance, West Covina, Whittier		
Urban Background	Residential sites far from traffic in more secluded areas	Summer	3	Artesia, Bell Canyon, Los Angeles	7,400 ± 6,900	55%
		Winter	2	Los Angeles		
Desert	Located inland, far from the ports, in desert-dominated environment	Summer	2	Riverside, San Bernardino	N/A	39%
		Winter	2	Palmdale, Riverside		

S1.1 Sample analysis

PM collected on PUF and Teflon filter substrates was completely solubilized using an automated microwave-assisted acid digestion protocol (Milestone Ethos Easy with SK15 rotor), which has been discussed in detail in the previous studies (Dillner et al. 2007; Herner et al. 2006a; Lough et al. 2005a). For collected Teflon filter samples, a mixture of ultra-high purity acids (16 M HNO_3, 1.50 mL; 12 M HCL, 0.50 mL; and 28 M HF, 0.20 mL) was used to solubilize the PM (Herner et al. 2006a). For the PUF samples (coarse fraction), the microwave chemistry used a mixture of acids and hydrogen peroxide (16 M HNO_3, 1.50 mL;12 M HCL, 0.38 mL;28M HF, 0.10; and 30% H_2O_2, 0.50 mL) to bring PM and PUF into complete solution (Dillner et al. 2007). The microwave digests were diluted to 15.0 ml (fine fraction) or 30.0 ml (coarse fraction) for SF-ICP-MS analysis. Included in each digestion batch of 33 samples were five method blanks, five Certified Reference Material (CRM) (National Institute of Standards and Technology (NIST); Urban Particulate Material-2x, San Joaquin Soil, Marine Sediment, Auto Catalyst), and two spiked blanks as previously detailed (Lough et al. 2005a; Pakbin et al. 2011).

The overall uncertainty of each reported analytical measurement was estimated by error propagation of the three primary sources of uncertainty: (a) SF-ICP-MS measurement - from the standard deviation of quadruplicate measurements on each sample, (b) method blank subtraction – from the standard deviation of multiple blanks, and (c) digestion recovery - long term running precision of replicate measurements NIST CRMs (Lough et al. 2005a; Pakbin et al. 2011).

The effect of relative humidity (RH) on $PM_{2.5}$ and $PM_{2.5-10}$ concentrations have been previously reported (Lou et al. 2017). As shown in Table S2.1, while RH at the urban traffic (60%), urban community (61%), and urban background (55%) locations were generally similar, RH values at the desert locations (39%) were lower than other sites. At the urban sites, the effect of RH on $PM_{2.5}$ and $PM_{2.5-10}$ mass concentrations is expected to be minimal due to the limited variation in RH. While RH could have an impact on the measured concentrations at the desert sites, the $PM_{2.5}$ and $PM_{2.5-10}$ concentration values were not adjusted for RH since only a small fraction of the samples (4 out of 50 samples) were collected at the desert locations. In addition, all samples were conditioned at the RH of 40±3% and the temperature of 21±1.5 ºC before the chemical speciation analysis in the lab to minimize the effect of relative humidity.

Table S2.2 Method detection limits (MDLs) for the measured elements

Symbol	Element	MDL (ng/m^3)	% of values below MDL	Symbol	Element	MDL (ng/m^3)	% of values below MDL
Li	Lithium	3.5E-02	0	Mo	Molybdenum	5.5E-02	2
Na	Sodium	1.4E+00	0	Pd	Palladium	2.5E-03	2
Mg	Magnesium	3.5E-01	0	Cd	Cadmium	3.0E-03	0
Al	Aluminum	9.0E-01	0	Sb	Antimony	8.5E-03	0
P	Phosphorus	2.9E-01	0	Cs	Cesium	4.0E-04	0
S	Sulfur	1.5E+00	0	Ba	Barium	2.5E-02	0
K	Potassium	6.5E-01	0	La	Lanthanum	5.5E-03	0
Ca	Calcium	1.5E+00	0	Ce	Cerium	5.5E-03	0
Ti	Titanium	1.9E-01	0	Pr	Praseodymium	1.5E-04	0
V	Vanadium	2.5E-03	0	Nd	Neodymium	6.0E-04	0
Cr	Chromium	7.0E-02	0	Sm	Samarium	2.5E-04	0
Mn	Manganese	3.0E-02	0	Eu	Europium	4.0E-04	0
Fe	Iron	2.0E-01	0	Dy	Dysprosium	2.5E-04	0
Co	Cobalt	2.5E-03	0	Ho	Holmium	4.5E-04	6
Ni	Nickel	2.2E-01	14	Yb	Ytterbium	4.5E-04	0
Cu	Copper	9.5E-02	0	Lu	Lutetium	2.0E-04	6
Zn	Zinc	4.8E-01	0	Hf	Hafnium	5.0E-03	0
Rb	Rubidium	7.5E-03	0	Ta	Tantalum	2.0E-03	0
Sr	Strontium	7.0E-02	0	Pb	Lead	2.9E-03	0
Y	Yttrium	2.5E-03	0	Th	Thorium	7.5E-04	0
Zr	Zirconium	1.6E-03	0	U	Uranium	2.5E-03	2
Nb	Niobium	2.0E-03	0				

Table S2.3 shows the criteria for classifying PM elements based on roadside enrichment (RE). Elements that consistently showed strong roadside enrichments (REs>20%) with traffic volume across different pairs of sampling locations were categorized as Type I elements. The RE for Type II elements were always positive, indicating an impact from traffic. However, the REs of Type II elements were not consistently strong (20% or higher), at least between one pair of the sampling sites, and therefore, these elements may originate from other anthropogenic sources as well as traffic. The RE for Type III elements were negative between at least one pair of the sampling sites, and elements in this category were not consistently impacted by traffic.

Table S2.3. Criteria for classifying PM elements based on roadside enrichment (RE)

	Enrichment with traffic	**UC vs UB**	**UT vs UB**	**UT vs UC**
Type I	Strong	RE>20% for all pairs		
Type II	Weak	Positive RE with RE<20% for at least one pair		
Type III	Inconsistent	RE<0 for at least one pair		

Table S2.4. Average absolute concentrations of metals and trace elements in $PM_{2.5}$ and $PM_{2.5-10}$ at different sampling sites (ng/m^3)

	$PM_{2.5}$				$PM_{2.5-10}$			
	Urban traffic	Urban community	Urban background	Desert	Urban traffic	Urban community	Urban background	Desert
PM mass	10.9±1.3	8.9±2.1	6.8±2.7	10.5±2.6	9.6±0.8	8.3±2.0	7.4±3.7	12.4±3.8
Na	550.1±147.4	480.5±152.3	354.6±249.1	296.7±105.4	621.1±136	614.2±257	367±145.5	344.2±108.8
S	535.4±80.8	483.8±101.6	349.2±155.3	375.3±105.1	116±13.8	108.1±30.4	66.9±19.8	99±32.9
Fe	251.3±73.3	163.5±75.1	126.4±60	302±78.3	361.6±70.3	269.5±73.9	254.7±118.9	464.6±144.9
Ca	127.5±22	100.3±33.4	110.4±78.9	264.4±77.3	288.9±49.8	238.7±58.9	264.9±155.8	521.4±150.4
Al	114.7±26.1	93±38.8	104.8±57.4	240.7±62.9	298.4±46.5	266±63	307±172.1	541.9±136.2
Mg	90.2±13.1	73.3±22.1	64.7±42	91.6±27.7	144.7±17.4	135.1±30	120±61.3	176.9±53.2
K	87.6±15.7	79.1±26.1	64.9±29	141.7±19	126.2±16.4	114.8±22.9	118.3±58.7	232.2±74.9
Ba	25.7±9.4	14.9±8.3	7.8±3.6	16.5±4.6	26.2±9.3	16.5±7.3	10±3.7	17.1±6.5
Ti	18.4±5.8	12.6±4.9	10.7±5.4	22.3±5.9	27.3±4	22.2±5.9	23±12.2	37.3±12.8
Cu	12.8±4.2	7.5±3.3	3.8±1.6	6.7±2.6	12.1±3.2	7.5±2.4	4.1±1.2	5.5±2.3
Zn	11.5±3.6	8.7±4.4	4.6±2.1	11.8±4.9	9.2±2.1	6±2.2	4.6±2.3	7.8±3.1
P	7.1±1	6.1±2.2	5.5±2.7	16±7.2	11.3±2.5	10.3±3.4	11.1±5.2	28.6±16
Zr	5.8±2.2	3.2±1.5	1.7±0.8	2.6±0.9	5.5±2	3.3±1.2	1.8±0.8	2.4±1.1
Mn	4.1±1.4	2.9±1.3	2.4±1.3	5.9±1.4	4.8±0.6	3.9±0.9	4.7±3.1	8.7±2.9
Sb	3.4±1.1	2.3±1.1	1±0.4	2.7±1.3	2.2±0.8	1.3±0.6	0.6±0.3	1.2±0.7
Sr	2.6±0.8	1.7±0.6	1.2±0.6	2.6±0.4	3.1±0.6	2.4±0.6	2.1±0.9	3.9±0.8
Cr	2.0±0.4	1.4±0.5	0.8±0.3	1.5±0.2	1.1±0.2	0.8±0.2	0.6±0.2	1.1±0.5
Pb	2.2±0.8	1.9±1.9	1.2±0.6	2.4±0.8	0.9±0.3	0.7±0.6	0.6±0.4	0.8±0.3
Co	0.09±0.03	0.03±0.06	0.06±0.03	0.03±0.05	0.11±0.02	0.02±0.09	0.09±0.02	0.02±0.09
Mo	0.9±0.4	0.5±0.2	0.2±0.1	0.3±0.1	0.5±0.2	0.2±0.1	0.1±0.1	0.2±0.1
V	0.7±0.4	0.5±0.2	0.4±0.3	0.7±0.3	0.6±0.1	0.5±0.1	0.5±0.3	0.9±0.3
Ni	0.7±0.2	0.5±0.3	0.3±0.1	0.5±0.2	0.6±0.2	0.4±0.2	0.3±0.2	0.4±0.2
Li	0.42±0.20	0.27±0.14	0.20±0.10	0.61±0.42	0.23±0.03	0.21±0.04	0.21±0.08	0.39±0.09
Ce	0.3±0.1	0.2±0.1	0.2±0.1	0.3±0.1	0.5±0.1	0.3±0.1	0.3±0.1	0.6±0.1
Rb	0.2±0.1	0.2±0.1	0.2±0.1	0.5±0.1	0.4±0.1	0.4±0.1	0.4±0.2	0.9±0.2
La	0.2±0.1	0.2±0.1	0.1±0.1	0.2±0.1	0.4±0.4	0.2±0.1	0.2±0.1	0.3±0.1
Hf	0.13±0.05	0.08±0.03	0.04±0.02	0.06±0.02	0.11±0.03	0.03±0.07	0.07±0.02	0.02±0.04
Cd	0.058±0.029	0.047±0.032	0.025±0.013	0.056±0.021	0.021±0.011	0.013±0.003	0.010±0.005	0.015±0.006
Y	0.064±0.012	0.045±0.016	0.040±0.024	0.079±0.024	0.123±0.017	0.101±0.023	0.098±0.051	0.172±0.054
Nd	0.059±0.014	0.045±0.019	0.046±0.024	0.096±0.026	0.124±0.020	0.106±0.029	0.116±0.061	0.193±0.052
Nb	0.041±0.010	0.029±0.011	0.025±0.011	0.056±0.016	0.082±0.019	0.064±0.020	0.062±0.029	0.113±0.019
Pd	0.030±0.009	0.019±0.008	0.012±0.005	0.022±0.005	0.029±0.011	0.021±0.010	0.016±0.008	0.027±0.013
Pr	0.018±0.004	0.013±0.006	0.013±0.007	0.027±0.008	0.036±0.006	0.030±0.008	0.031±0.016	0.054±0.015
Th	0.020±0.004	0.016±0.006	0.018±0.009	0.041±0.012	0.048±0.007	0.041±0.009	0.045±0.021	0.088±0.023
Cs	0.015±0.004	0.013±0.005	0.015±0.006	0.039±0.008	0.022±0.003	0.020±0.004	0.021±0.008	0.039±0.010
Sm	0.009±0.002	0.007±0.003	0.008±0.004	0.016±0.004	0.020±0.003	0.018±0.004	0.021±0.012	0.034±0.009
Eu	0.008±0.002	0.005±0.002	0.004±0.002	0.008±0.002	0.013±0.004	0.009±0.003	0.008±0.004	0.013±0.004
Dy	0.007±0.002	0.006±0.002	0.006±0.003	0.013±0.003	0.016±0.003	0.014±0.003	0.016±0.008	0.026±0.007
Ho	0.002±0.001	0.001±0.001	0.001±0.001	0.003±0.001	0.003±0.001	0.003±0.001	0.003±0.002	0.005±0.001
Yb	0.006±0.002	0.004±0.002	0.004±0.002	0.008±0.002	0.010±0.002	0.008±0.002	0.009±0.001	0.014±0.001
Lu	0.001±0.001	0.001±0.001	0.001±0.001	0.001±0.001	0.002±0.001	0.001±0.001	0.001±0.001	0.002±0.001
Ta	0.003±0.001	0.002±0.001	0.002±0.001	0.004±0.001	0.006±0.002	0.005±0.002	0.004±0.001	0.009±0.002
U	0.011±0.002	0.009±0.003	0.008±0.004	0.019±0.007	0.017±0.003	0.015±0.004	0.015±0.006	0.027±0.009

Average concentration ±s standard deviation (ng/m^3)

Table S2.5. Average normalized concentrations of metals and trace elements in $PM_{2.5}$ and $PM_{2.5-10}$ at different sampling sites (µg/g)

	$PM_{2.5}$				$PM_{2.5-10}$			
	Urban traffic	Urban community	Urban background	Desert	Urban traffic	Urban community	Urban background	Desert
Na	28078.3±5774	48553.8±16616.5	57801.8±24526.8	51243.9±15915.5	27567.9±2491.3	52220.1±7323.9	78505.7±36421.8	64859.4±15190.3
S	35863.1±4092.3	51142±7268.3	58044.6±21031.4	49758.1±10163.9	8075.4±1514.8	9872.6±1912.5	13565.4±4006	12102.8±1569.1
Fe	28737.2±1256.2	17587.5±2661.7	18359.4±6861.2	22729.1±5151.1	37374.1±3106.2	34845.2±2703.2	34854.8±16272.6	37406.2±5712.7
Ca	25176.3±3059.3	14429.4±5054.8	11415.7±3169.1	11680.5±1586	42367.1±2950.4	34717.9±5087	30801±13133.3	29915.1±4093.9
Al	23577.3±4511.8	14014.5±3709.5	10480.2±3619.4	10455.9±1881.5	45171.3±5767.1	40608.1±3846.9	35085.9±18712.9	30967.7±3955.9
Mg	8734±1193.2	8595±2604.9	8514.4±2689.2	8298±1118	14311.1±478.1	16209.3±891.9	17535.7±7482.4	15081±1826.4
K	14253.4±3159.9	9052.9±1520.1	9810.1±7589.9	8015.4±987	18743.4±886.1	15995.2±1053	15202.9±8309.6	13107.8±1366.1
Ba	1568.2±167.4	1102.8±265.2	1625.3±672.6	2318.4±716.9	1387.5±333.5	1475.4±455	2099.2±1084.1	2695.1±887.5
Ti	2147.1±304.5	1485.4±234	1438.3±532.9	1665.3±412	2971.7±337.2	3115.1±353.3	2871.3±1290.7	2839.5±347.9
Cu	614.4±147.4	557.9±98.4	843.5±286.6	1165.3±330.9	426.5±120.6	625.4±196.7	968±484.3	1252.7±327.9
Zn	1081.8±293.6	647.5±125.7	964.2±374.2	1038.1±228.5	612.1±163.3	631.7±125.5	753.7±277	946.6±180.5
P	1480.3±417	781.8±157.2	703.8±267	652.8±100.3	2132.9±635	1546.4±379.4	1326.9±653.9	1187.4±286.5
Zr	242.7±44.2	238.4±73.7	368.3±156.1	525.1±184.4	187.9±64.2	271.9±122.2	444.4±308.6	569.9±202.1
Mn	569.1±46	334.7±63.6	323.3±113.2	372.2±90.2	699.9±94	605.5±100.1	505±223.9	495.2±52.7
Sb	245.3±86.7	146.6±28.7	247.9±84.6	310.3±80.1	91.9±42.7	99.7±43	169.6±92.3	229.1±79.2
Sr	255.3±38.1	165.5±30.8	211.7±158.6	232.3±60.1	331.6±46.8	294±41.6	315.9±158.8	318.3±52
Pb	223.7±36.9	170.1±48.8	211.6±162.3	196.6±65.8	65±20.1	79.8±24.3	82.5±50.3	91.2±26.7
Cr	147.4±21.8	128.5±15.8	183.1±175.8	184.7±31.8	82.4±17.5	80.6±7.8	95.5±28.5	115.7±16.1
Mo	30.1±8.8	30±3.6	52.3±14.8	85±35.6	12.2±3.9	17.3±5.4	28.6±11.8	50±24.4
Ni	48.7±6.1	41.7±5.2	62.6±27.6	66.8±17.9	32.4±16.7	48.7±28.3	51.3±25.2	61.6±22.6
V	69.1±19.3	66±27.4	58.1±19.9	66.4±34.1	71.1±9.7	72.5±14.2	62.3±24.6	59.5±14.3
Li	78.6±82.5	28.9±7.9	30.6±13.8	38.3±17.7	32.3±4.2	28.7±3.1	27.5±15.1	24.6±2.8
Ce	31.6±4.4	21.2±3.3	22.9±7.8	31±9	47.1±7.8	43.2±2.4	45±21.5	49.5±13.6
La	14.8±2.1	14.9±4.8	17.4±9	23.3±16.2	23±4.1	23.9±3.4	26.4±12.4	38.3±40.9
Rb	44±8.4	25.4±5.3	21±12.3	18.7±3.1	71.6±6.6	57.3±4.3	51.5±27.5	44.9±5.5
Hf	5.5±1	5.7±1.4	8.4±3.3	12.1±3.8	4.3±0.8	5.5±2.1	8.8±5.5	11.5±3.5
Co	10.5±0.4	6.9±1	7±2.6	7.9±1.5	12.4±1.2	12.4±1.8	11.4±4.7	11.7±1.8
Y	7.3±0.6	5.3±1.6	4.9±1.3	5.8±0.7	13.8±2	13.1±0.9	13.1±5.8	12.7±1.5
Nd	9.21±1.23	6.28±1.33	5.07±1.82	5.39±1.02	15.98±2.54	15.57±0.97	13.85±6.85	12.9±1.78
Cd	5.46±1.71	3.55±1.24	6.62±11.37	5.15±2.16	1.16±0.38	1.33±0.15	1.72±1.19	2.16±0.95
Nb	5.5±1.21	3.59±0.41	3.33±1.23	3.69±0.69	9.84±2.32	8.52±0.71	8.54±5.19	8.43±1.6
Pd	2.08±0.09	1.6±0.3	2.21±1.1	2.73±0.6	2.28±0.89	2.26±1.04	2.78±1.88	3.03±1.1
Th	3.96±0.68	2.43±0.72	1.77±0.63	1.85±0.33	7.39±1.58	6.15±0.78	5.52±3.17	4.95±0.55
Pr	2.56±0.38	1.73±0.38	1.48±0.5	1.64±0.28	4.48±0.83	4.23±0.21	3.92±1.9	3.77±0.49
Cs	3.77±0.28	2.18±0.52	1.54±0.81	1.36±0.37	3.21±0.29	3±0.41	2.77±1.88	2.32±0.31
U	1.83±0.52	1.17±0.33	1±0.34	0.99±0.2	2.16±0.54	2.08±0.45	2±0.98	1.77±0.3
Sm	1.58±0.22	1.07±0.25	0.8±0.32	0.77±0.14	2.85±0.49	2.76±0.28	2.34±1.32	2.08±0.32
Eu	0.75±0.02	0.48±0.07	0.53±0.19	0.71±0.15	1.1±0.19	1.05±0.18	1.16±0.62	1.3±0.38
Dy	1.23±0.14	0.81±0.21	0.64±0.23	0.66±0.13	2.15±0.19	2.13±0.17	1.85±0.95	1.68±0.24
Yb	0.72±0.04	0.5±0.11	0.45±0.15	0.53±0.13	1.15±0.05	1.14±0.02	1.11±0.59	1.06±0.16
Ta	0.42±0.07	0.27±0.04	0.26±0.11	0.26±0.04	0.9±0.52	0.64±0.22	0.63±0.5	0.61±0.24
Ho	0.24±0.02	0.16±0.04	0.13±0.04	0.14±0.02	0.43±0.03	0.42±0.02	0.37±0.19	0.35±0.04
Lu	0.11±0	0.07±0.02	0.07±0.02	0.09±0.02	0.17±0.01	0.17±0	0.19±0.2	0.17±0.03

Average concentration ±s standard deviation (µg/g)

Table S2.6. Elemental concentrations of selected traffic elements in the collected samples

Table S2.6a. Selected $PM_{2.5}$ elemental concentrations (ng/m^3)

Sample	Sampling Location	Ba	Cr	Cu	Mo	Pd	Sb	Zn	Zr
1	Urban Traffic	19.105	1.493	8.067	0.563	0.027	2.131	9.197	3.608
2	Urban Traffic	16.565	1.546	9.181	0.669	0.023	3.093	8.726	3.771
3	Urban Traffic	30.497	1.959	15.237	0.646	0.031	3.690	11.530	7.838
4	Urban Traffic	13.347	2.383	7.890	0.488	0.017	2.686	6.624	3.060
5	Urban Traffic	32.198	1.955	17.050	0.811	0.036	3.467	12.801	8.224
6	Urban Traffic	27.276	2.179	15.009	0.781	0.028	2.910	11.159	7.342
7	Urban Traffic	30.085	2.881	14.418	1.382	0.039	4.999	18.671	5.254
8	Urban Traffic	40.339	2.176	19.516	1.462	0.037	4.663	16.293	8.896
9	Urban Traffic	40.656	2.681	17.033	1.780	0.051	5.685	16.246	7.574
10	Urban Traffic	33.416	1.815	16.152	0.597	0.036	3.816	12.063	8.174
11	Urban Traffic	19.505	1.579	11.113	0.451	0.024	2.404	7.745	4.620
12	Urban Traffic	20.211	2.079	10.744	1.121	0.025	3.529	11.525	4.166
13	Urban Traffic	11.127	1.517	5.473	1.489	0.018	1.563	7.340	2.567
14	Urban Community	18.001	1.631	8.694	0.661	0.023	2.740	10.559	3.932
15	Urban Community	12.730	1.358	6.193	0.453	0.018	2.637	10.461	2.528
16	Urban Community	18.315	1.175	9.670	0.366	0.023	2.246	6.590	4.292
17	Urban Community	10.585	1.318	6.497	0.400	0.014	1.866	5.120	2.729
18	Urban Community	8.944	1.462	5.323	0.457	0.013	1.510	5.316	2.042
19	Urban Community	28.317	1.820	13.376	0.874	0.031	3.745	17.894	5.602
20	Urban Community	7.689	0.916	5.044	0.349	0.011	1.621	7.130	2.020
21	Urban Community	5.116	2.138	3.112	0.152	0.014	0.670	3.931	1.705
22	Urban Community	5.693	0.939	4.149	0.265	0.009	0.959	4.704	1.623
23	Urban Community	18.332	1.586	8.844	0.521	0.022	2.708	9.357	4.118
24	Urban Community	12.804	1.856	6.821	0.593	0.019	2.366	8.731	2.609
25	Urban Community	2.918	0.212	2.691	0.278	0.006	0.793	2.703	0.792
26	Urban Community	15.281	1.456	8.747	0.375	0.021	2.348	7.626	4.104
27	Urban Community	15.925	1.413	8.786	0.494	0.021	2.380	7.843	4.073
28	Urban Community	16.978	1.311	9.881	0.403	0.021	1.997	8.877	4.404
29	Urban Community	12.387	1.783	6.823	0.472	0.015	2.002	8.240	2.624
30	Urban Community	6.469	0.914	3.737	0.377	0.010	1.079	3.843	1.409
31	Urban Community	11.863	1.524	5.310	0.443	0.017	2.692	9.668	2.403
32	Urban Community	11.848	1.567	6.188	0.317	0.014	2.242	8.277	2.569
33	Urban Community	31.563	2.234	13.119	0.806	0.034	4.757	19.701	5.392
34	Urban Community	30.995	1.746	11.568	0.735	0.034	3.489	14.285	5.255
35	Urban Community	1.579	0.220	1.776	0.087	0.003	0.356	1.466	0.555
36	Urban Community	26.892	2.143	12.057	0.695	0.031	4.203	15.726	4.709
37	Urban Community	13.209	1.195	7.100	0.362	0.020	1.659	6.929	3.763
38	Urban Community	11.526	1.115	5.609	0.334	0.016	1.564	7.638	2.533
39	Urban Community	8.286	1.010	4.869	0.408	0.013	1.212	4.129	2.042
40	Urban Community	23.854	1.613	10.962	0.816	0.029	4.247	12.979	4.915
41	Urban Community	29.081	2.173	14.017	0.646	0.035	3.276	13.701	6.054
42	Urban Background	10.125	1.099	4.761	0.273	0.017	1.154	6.764	1.754
43	Urban Background	5.866	0.801	3.212	0.175	0.009	0.833	3.072	1.239
44	Urban Background	1.736	0.340	1.058	0.061	0.002	0.289	1.222	0.396
45	Urban Background	10.119	0.959	5.221	0.260	0.014	1.333	5.177	2.658
46	Urban Background	11.270	0.977	4.938	0.259	0.014	1.386	6.582	2.232
47	Desert	17.770	1.612	6.917	0.330	0.024	2.739	11.492	2.837
48	Desert	17.047	1.496	7.251	0.390	0.024	2.690	12.490	2.817
49	Desert	9.222	1.098	2.634	0.111	0.013	0.878	4.669	1.141
50	Desert	21.846	1.766	9.938	0.497	0.026	4.540	18.364	3.679

Table S2.6b. Selected $PM_{2.5-10}$ elemental concentrations (ng/m^3)

Sample	Sampling Location	Ba	Cr	Cu	Mo	Pd	Sb	Zn	Zr
1	Urban Traffic	17.537	0.978	7.779	0.312	0.019	1.355	6.644	3.181
2	Urban Traffic	18.455	1.019	10.268	0.544	0.020	1.667	8.108	3.920
3	Urban Traffic	31.985	1.279	14.524	0.380	0.032	2.831	9.184	7.690
4	Urban Traffic	15.988	0.905	10.516	0.354	0.014	1.407	6.534	4.176
5	Urban Traffic	32.600	1.190	15.633	0.474	0.038	2.833	10.150	7.974
6	Urban Traffic	28.380	1.033	13.916	0.497	0.032	2.557	8.927	7.095
7	Urban Traffic	27.802	1.177	10.178	0.363	0.034	2.287	9.599	4.729
8	Urban Traffic	35.466	1.255	15.183	1.037	0.039	3.183	11.747	7.142
9	Urban Traffic	38.023	1.504	13.368	0.462	0.046	2.978	12.356	6.270
10	Urban Traffic	42.559	1.351	17.721	0.381	0.053	3.582	12.521	8.670
11	Urban Traffic	19.188	0.930	10.808	0.274	0.019	1.450	8.015	3.879
12	Urban Traffic	20.632	1.161	11.404	0.963	0.020	1.795	9.216	3.950
13	Urban Traffic	11.406	0.749	5.680	0.234	0.017	0.939	6.018	2.719
14	Urban Community	23.658	0.902	9.099	0.285	0.030	1.997	7.204	4.547
15	Urban Community	11.385	0.713	5.501	0.181	0.014	0.926	5.235	2.200
16	Urban Community	17.279	0.886	9.211	0.230	0.017	1.502	7.620	3.980
17	Urban Community	11.750	0.731	7.460	0.232	0.010	0.934	4.722	3.043
18	Urban Community	13.598	0.839	8.531	0.351	0.015	1.129	5.990	3.350
19	Urban Community	36.286	1.160	14.220	0.518	0.043	2.835	11.469	6.541
20	Urban Community	15.344	0.756	6.090	0.155	0.022	1.201	5.740	3.053
21	Urban Community	6.639	0.231	3.225	0.072	0.011	0.553	1.894	1.930
22	Urban Community	16.499	0.904	8.895	0.367	0.017	1.393	7.745	3.160
23	Urban Community	19.839	0.698	8.300	0.258	0.024	1.684	6.314	4.113
24	Urban Community	7.462	0.526	4.290	0.121	0.010	0.519	3.635	1.639
25	Urban Community	7.205	0.451	4.170	0.147	0.010	0.570	3.062	1.521
26	Urban Community	24.252	0.801	9.290	0.234	0.031	1.909	6.772	4.739
27	Urban Community	18.842	0.753	8.366	0.229	0.023	1.483	5.399	4.121
28	Urban Community	13.721	0.747	8.062	0.222	0.014	1.161	6.009	3.390
29	Urban Community	14.330	0.762	7.822	0.319	0.014	1.186	7.655	3.041
30	Urban Community	8.648	0.742	5.365	0.221	0.011	0.717	3.749	1.877
31	Urban Community	11.329	0.562	4.246	0.115	0.022	0.897	4.099	1.918
32	Urban Community	13.016	0.775	6.662	0.192	0.015	1.049	5.397	2.593
33	Urban Community	24.541	1.167	8.332	0.228	0.034	1.843	9.426	3.659
34	Urban Community	27.589	0.918	8.862	0.222	0.033	1.984	6.808	4.348
35	Urban Community	7.119	0.576	3.875	0.104	0.009	0.395	3.197	1.566
36	Urban Community	23.119	1.071	7.907	0.207	0.035	1.599	9.520	3.450
37	Urban Community	14.695	0.652	7.081	0.181	0.022	1.125	4.591	3.644
38	Urban Community	13.685	0.570	5.978	0.150	0.021	1.029	5.121	2.619
39	Urban Community	10.231	0.658	6.299	0.237	0.013	0.904	4.300	2.363
40	Urban Community	23.833	0.880	10.109	0.253	0.038	2.150	7.738	4.645
41	Urban Community	26.662	0.906	11.588	0.335	0.036	2.524	8.581	5.727
42	Urban Background	12.380	0.979	4.191	0.111	0.020	0.567	8.679	1.462
43	Urban Background	6.874	0.504	3.332	0.098	0.009	0.400	2.874	1.193
44	Urban Background	4.542	0.276	2.197	0.069	0.005	0.325	2.193	0.831
45	Urban Background	11.642	0.507	5.394	0.140	0.022	0.993	3.611	2.925
46	Urban Background	14.728	0.592	5.159	0.133	0.025	0.932	5.640	2.356
47	Desert	17.957	1.560	6.467	0.171	0.022	1.030	8.864	2.470
48	Desert	15.607	1.095	5.989	0.175	0.021	1.136	9.925	2.347
49	Desert	8.334	0.340	1.673	0.046	0.016	0.347	2.549	0.810
50	Desert	26.651	1.328	7.989	0.242	0.050	2.217	10.015	3.998

Table S2.7. Roadside enrichments (REs) of (a) $PM_{2.5}$ elements, and (b) $PM_{2.5-10}$ elements

Table S2.7a. $PM_{2.5}$ RE			
Type I	UC vs. UB	UT vs. UB	UT vs. UC
Li	26%	**52%**	35%
Cr	**41%**	**59%**	30%
Fe	23%	**50%**	35%
Ni	**49%**	**62%**	26%
Cu	**49%**	**70%**	**41%**
Zn	**48%**	**60%**	25%
Sr	30%	**53%**	33%
Zr	**49%**	**71%**	**44%**
Mo	**56%**	**78%**	**50%**
Pd	40%	**62%**	37%
Cd	**47%**	**58%**	21%
Sb	**56%**	**71%**	34%
Ba	**47%**	**70%**	**42%**
La	31%	**57%**	38%
Ce	27%	**55%**	39%
Eu	28%	**55%**	38%
Hf	**47%**	**70%**	**44%**
Type II	UC vs UB.	UT vs. UB	UT vs. UC
Na	26%	36%	13%
Mg	12%	28%	19%
P	9%	22%	14%
S	28%	35%	10%
K	18%	26%	10%
Ti	15%	**42%**	32%
V	12%	40%	31%
Mn	15%	**41%**	30%
Co	19%	**44%**	29%
Y	10%	37%	30%
Nb	12%	37%	29%
Pr	4%	30%	27%
Yb	7%	37%	32%
Lu	15%	**45%**	35%
Ta	16%	37%	25%
Pb	38%	**44%**	11%
U	8%	25%	19%
Type III	UC vs UB.	UT vs. UB	UT vs. UC
Al	-13%	9%	19%
Ca	-10%	13%	21%
Rb	-4%	11%	15%
Cs	-15%	0%	13%
Nd	-2%	22%	24%
Sm	-14%	6%	18%
Dy	-7%	17%	23%
Ho	-3%	23%	25%
Th	-14%	12%	23%

REs> 40% are bolded

UT: Urban traffic, UC: Urban community, UB: Urban background

Table S2.7b. $PM_{2.5-10}$ RE			
Type I	UC vs UB.	UT vs. UB	UT vs. UC
Cr	25%	**49%**	32%
Cu	**46%**	**66%**	38%
Zn	24%	**50%**	34%
Zr	**47%**	**68%**	40%
Mo	**52%**	**77%**	**53%**
Pd	24%	**45%**	28%
Cd	24%	**54%**	39%
Sb	**52%**	**71%**	**40%**
Ba	39%	**62%**	37%
Hf	**46%**	**68%**	**40%**
Type II	UC vs UB.	UT vs. UB	UT vs. UC
Na	**40%**	**41%**	1%
Mg	11%	17%	7%
S	38%	**42%**	7%
Fe	5%	30%	25%
Ni	18%	**44%**	32%
Sr	12%	31%	21%
Y	3%	20%	18%
Nb	4%	24%	21%
La	17%	**53%**	**43%**
Ce	10%	34%	26%
Eu	13%	39%	29%
Lu	5%	24%	20%
Ta	9%	29%	22%
Pb	12%	31%	21%
U	4%	14%	10%
Type III	UC vs UB.	UT vs. UB	UT vs. UC
Li	0%	13%	13%
Al	-15%	-3%	11%
P	-8%	2%	10%
K	-3%	6%	9%
Ca	-11%	8%	17%
Ti	-4%	16%	19%
V	-13%	3%	15%
Mn	-20%	2%	18%
Co	-5%	17%	21%
Rb	-9%	1%	9%
Cs	-6%	4%	9%
Pr	-4%	14%	17%
Nd	-9%	7%	15%
Sm	-19%	-5%	12%
Dy	-13%	2%	13%
Ho	-11%	7%	16%
Yb	-2%	16%	18%
Th	-8%	6%	13%

REs> 40% are bolded

UT: Urban traffic, UC: Urban community, UB: Urban background

Table S2.8. Varimax normalized principal component loadings (a) $PM_{2.5}$, and (b) $PM_{2.5-10}$

Table S2.8a. Varimax normalized principal component loadings in $PM_{2.5}$

	Principal component loadings			
	Mineral dust	Traffic	Marine aerosol	Industrial
Rb	**0.97**	0.08	0.01	0.14
Th	**0.97**	0.17	0.07	0.02
Sm	**0.97**	0.21	0.03	0.07
Al	**0.96**	0.19	0.03	0.08
Ca	**0.95**	0.15	0.13	0.06
Dy	**0.94**	0.32	0.05	0.04
P	**0.91**	0.06	0.14	0.12
Nd	**0.91**	0.38	0.07	0.09
Ho	**0.90**	0.39	0.10	0.04
U	**0.90**	0.17	0.09	-0.11
Cs	**0.90**	0.14	-0.27	0.08
Pr	**0.86**	0.45	0.11	0.13
Nb	**0.81**	0.52	0.06	0.11
Ta	**0.78**	0.35	0.20	0.18
Yb	**0.78**	0.60	0.07	-0.01
Y	**0.76**	0.48	0.37	0.05
K	**0.76**	0.33	0.04	0.36
Mn	**0.76**	0.53	0.09	0.32
Ti	**0.72**	0.58	0.14	0.19
Fe	0.69	0.69	0.07	0.16
Co	0.62	0.42	0.23	0.40
Zr	0.15	**0.97**	0.01	-0.10
Hf	0.16	**0.97**	0.05	-0.12
Cu	0.20	**0.97**	0.09	0.00
Ba	0.29	**0.94**	0.06	0.09
Pd	0.34	**0.88**	0.17	0.18
Sb	0.32	**0.82**	0.16	0.33
Eu	0.58	**0.78**	0.13	0.07
Zn	0.43	**0.74**	0.09	0.41
Sr	0.49	**0.74**	0.15	0.23
Cr	0.21	**0.74**	0.25	0.35
Mo	0.00	**0.73**	0.44	0.30
Lu	0.68	**0.72**	0.06	-0.04
Cd	0.21	0.65	-0.22	0.61
Ce	0.51	0.63	0.30	0.19
Pb	0.27	0.53	-0.13	0.45
Li	0.35	0.51	-0.30	-0.13
Ni	0.15	0.50	0.41	0.41
Na	-0.14	-0.01	**0.94**	-0.01
S	-0.01	0.17	**0.92**	0.02
V	0.33	-0.04	**0.80**	0.18
Mg	0.51	0.28	**0.72**	0.01
La	0.07	0.13	0.69	-0.13
% of variance	41.67	30.92	10.61	4.77
Cumulative %	41.67	72.59	83.20	87.97
Eigenvalue	17.92	13.30	4.56	2.05

Bold: loadings larger than 0.7, Underlined: loadings between 0.5 and 0.7

Table S2.8b. Varimax normalized principal component loadings in $PM_{2.5-10}$

	Principal component loadings		
	Mineral dust	Traffic	Marine aerosol
Al	**0.99**	0.05	0.08
Rb	**0.98**	0.10	0.00
Sm	**0.98**	0.04	0.08
Dy	**0.98**	0.12	0.09
Mn	**0.96**	0.07	0.15
Nd	**0.96**	0.21	0.07
Ho	**0.96**	0.24	0.07
Th	**0.95**	0.14	0.01
K	**0.95**	0.03	0.21
Cs	**0.93**	0.20	-0.05
Pr	**0.93**	0.29	0.07
Li	**0.93**	0.25	0.06
Ti	**0.90**	0.30	0.17
Yb	**0.90**	0.38	0.08
Y	**0.89**	0.37	0.16
Ca	**0.89**	0.30	0.03
P	**0.88**	-0.07	0.13
Co	**0.87**	0.25	0.29
U	**0.87**	0.08	0.16
V	**0.86**	-0.15	0.43
Lu	**0.82**	0.48	0.02
Fe	**0.80**	0.57	0.09
Nb	**0.77**	0.56	-0.12
Sr	**0.72**	0.62	0.11
Ce	**0.70**	0.50	0.15
Sb	0.06	**0.99**	0.03
Ba	0.18	**0.97**	-0.01
Zr	-0.04	**0.97**	0.03
Cu	0.00	**0.94**	0.19
Hf	-0.04	**0.93**	0.11
Pd	0.28	**0.88**	-0.18
Zn	0.48	**0.83**	0.18
Eu	0.54	**0.82**	-0.09
Cr	0.55	**0.72**	0.31
Mo	-0.07	**0.70**	0.35
Cd	0.37	0.55	0.31
Pb	0.47	0.50	-0.01
Ta	0.43	0.45	-0.18
Na	-0.22	0.02	**0.89**
S	0.04	0.34	**0.78**
Ni	0.30	0.11	**0.73**
Mg	0.61	0.07	**0.72**
La	0.25	-0.04	0.33
% of variance	51.68	25.60	8.27
Cumulative %	51.68	77.28	85.55
Eigenvalue	22.22	11.01	3.56

Bold: loadings larger than 0.7, Underlined: loadings between 0.5 and 0.7

Table S2.9. Results of multiple linear regression analysis for mass concentrations of (a) $PM_{2.5}$ and (b) $PM_{2.5-10}$

(a) $PM_{2.5}$

	Unstandardized coefficients	Standardized coefficients	t	Sig.	R^2
	B	Beta			
Constant	9.35		33.71	<0.001	0.76
Traffic	1.44	0.60	8.53	<0.001	
Mineral dust	1.19	0.49	7.07	<0.001	
Marine aerosol	0.94	0.39	5.60	<0.001	
Industrial	0.43	0.18	2.57	0.014	

(b) $PM_{2.5-10}$

	Unstandardized coefficients	Standardized coefficients	t	Sig.	R^2
	B	Beta			
Constant	8.91		64.91	<0.001	0.86
Mineral dust	2.01	0.78	14.50	<0.001	
Marine aerosol	0.97	0.38	6.98	<0.001	
Traffic	0.85	0.33	6.15	<0.001	

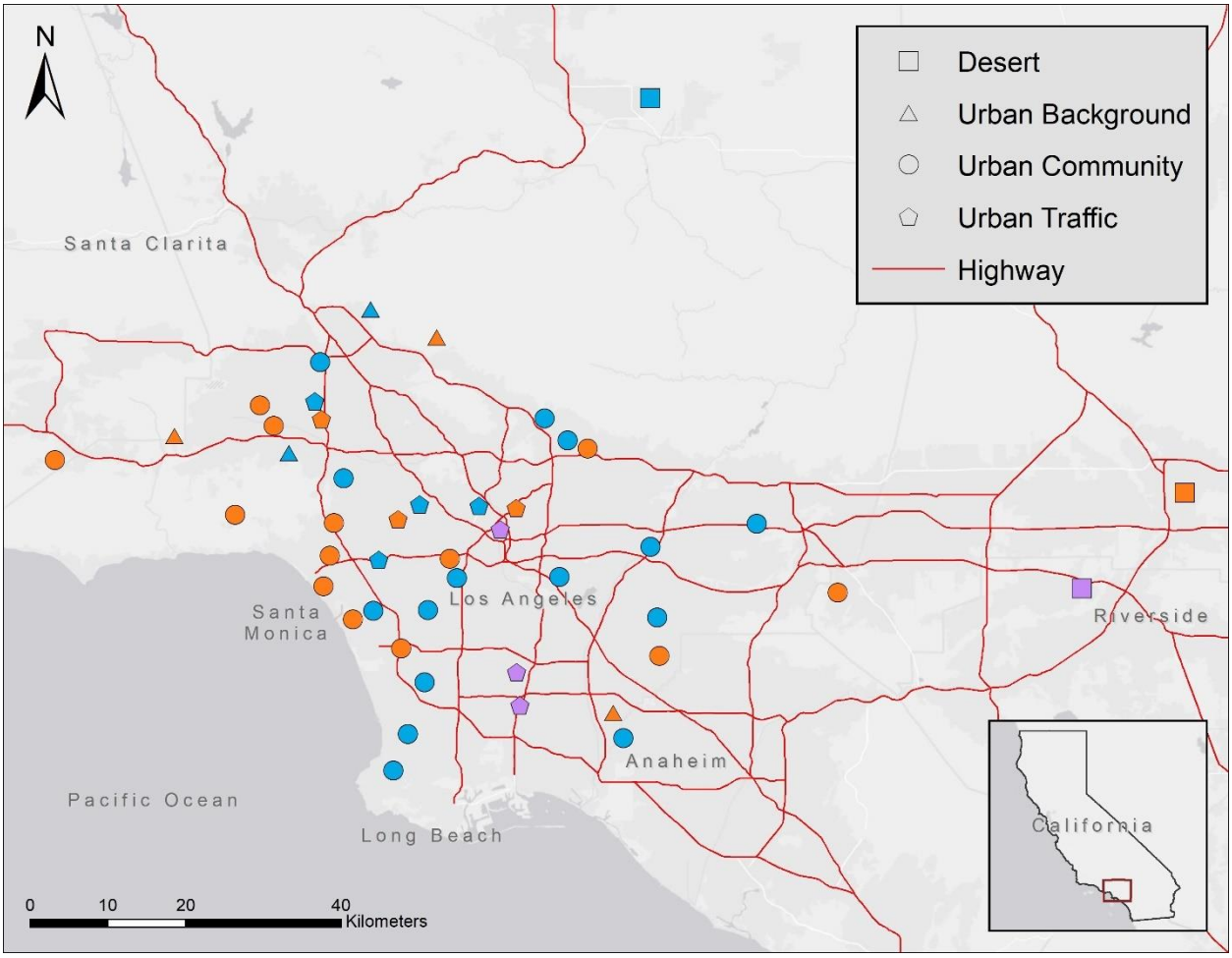

Figure S2.1. Location of the sampling sites in the Greater Los Angeles area for summer (orange), winter (blue), and repeated samples in summer and winter (purple)

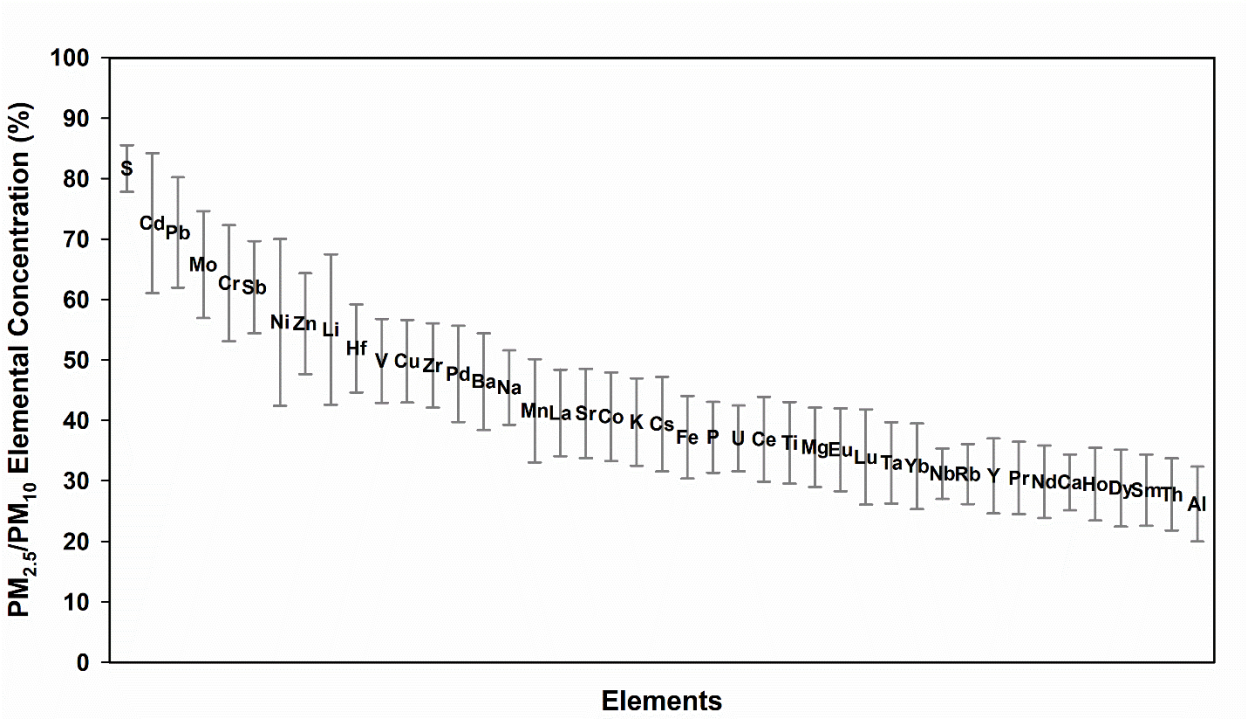

Figure S2.2. Percentage of metals and trace elements in PM_{10} that is confined in $PM_{2.5}$. Symbol of the chemical elements represent the average values, and error bars show the standard deviation across all of the sampling sites.

Figure S2.3 shows average crustal enrichment factors (EFs) for selected elements and trace metals in both fine and coarse particles across the sampling sites. The calculated EFs were generally higher in $PM_{2.5}$ than $PM_{2.5-10}$, presumably because of the different physical and chemical processes that selectively enhanced these elements in smaller particles. This is in agreement with the findings of previous studies that reported higher EFs in $PM_{2.5}$ compared to $PM_{2.5-10}$ (Arhami et al. 2009). Pd, Sb, and S were shown to have the largest average EFs among $PM_{2.5}$ and $PM_{2.5-10}$ elements. Some of the traffic tracers including Mo, and Zn as well elements such as Cd and Li also showed substantial average EF values (EF>5) in $PM_{2.5}$ and $PM_{2.5-10}$. Other traffic tracers such as Ba, Cr, Pb, and Zr showed high EFs only in $PM_{2.5}$. In contrast to traffic tracers, crustal elements such as Ca, K, Mg were shown to have low EFs (EF<5) in fine and coarse particles, indicating a lack of major anthropogenic sources for these elements.

Previously, Sb, S, Cd, Mo, and Zn were reported to have the largest EF values in $PM_{2.5}$ and $PM_{2.5-10}$ (Arhami et al. 2009). Other studies have reported high EF values for Cu and Zn (Birmili et al. 2006; Cheung et al. 2011). More recent studies reported very high EF values for Pd (10^3 <average EF< 10^4) in $PM_{2.5}$ and PM_{10} (Das and Chellam 2020). While a wide range of sampling sites with diverse local emission sources were covered in this study, our findings are in reasonable agreement with the results of the previous studies.

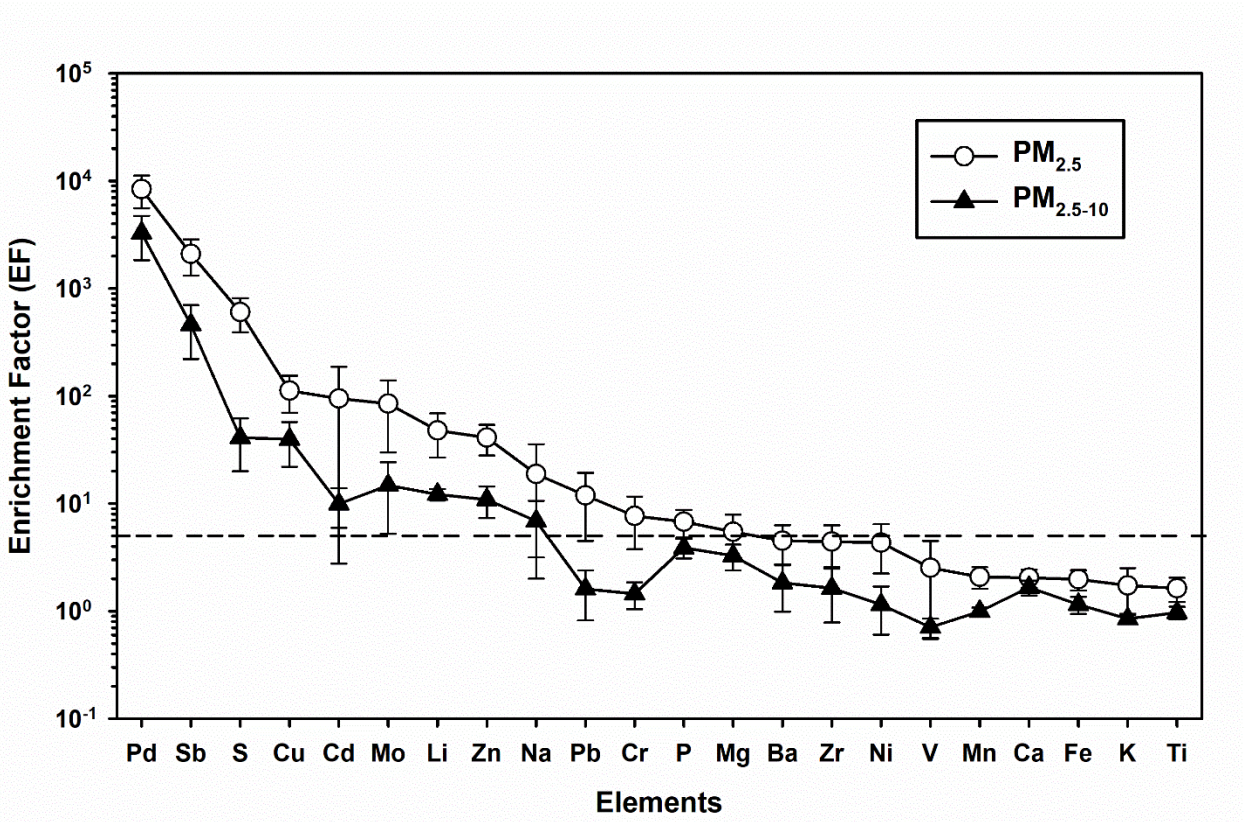

Figure S2.3. Crustal enrichment factors (EFs) for selected elements. The horizontal line represents anthropogenic EF threshold (EF=5).

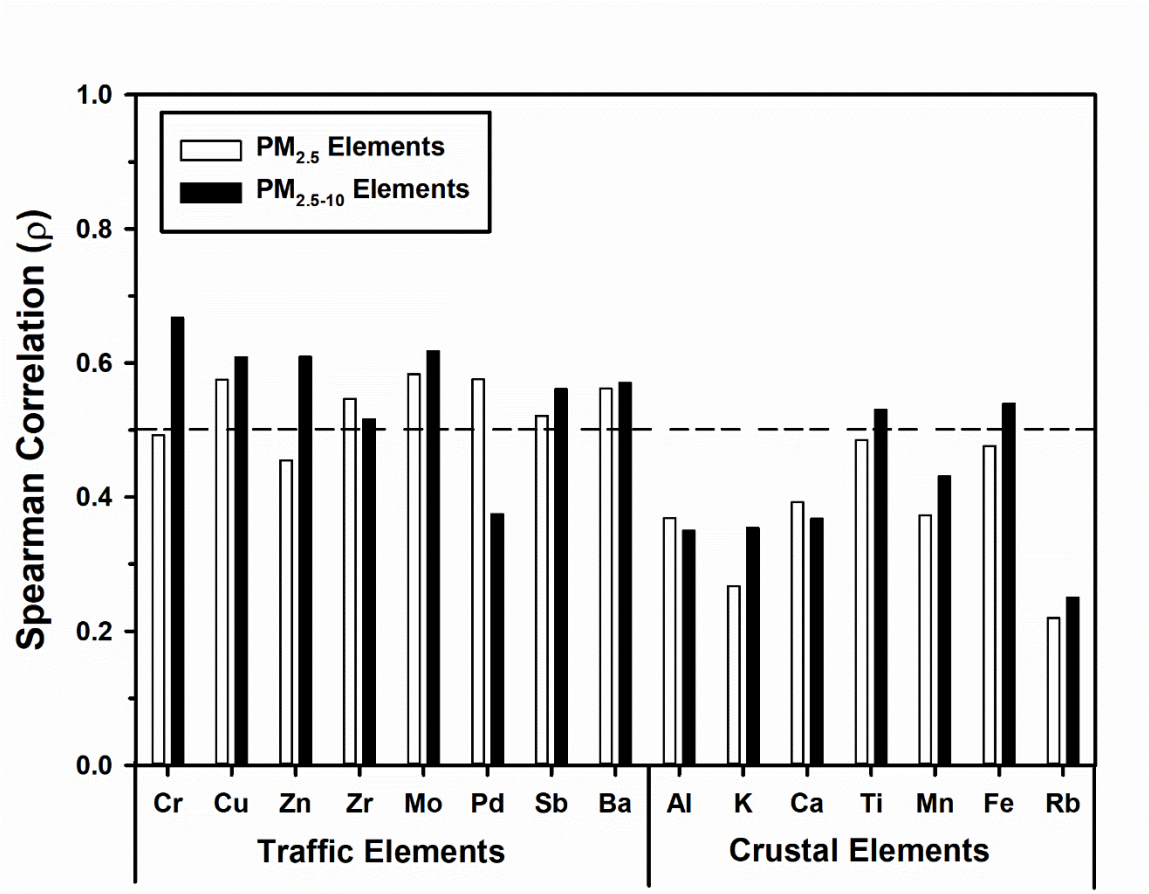

Figure S2.4. Spearman correlation coefficients of selected $PM_{2.5}$ and $PM_{2.5-10}$ traffic and crustal elements with the annual average daily traffic (AADT)

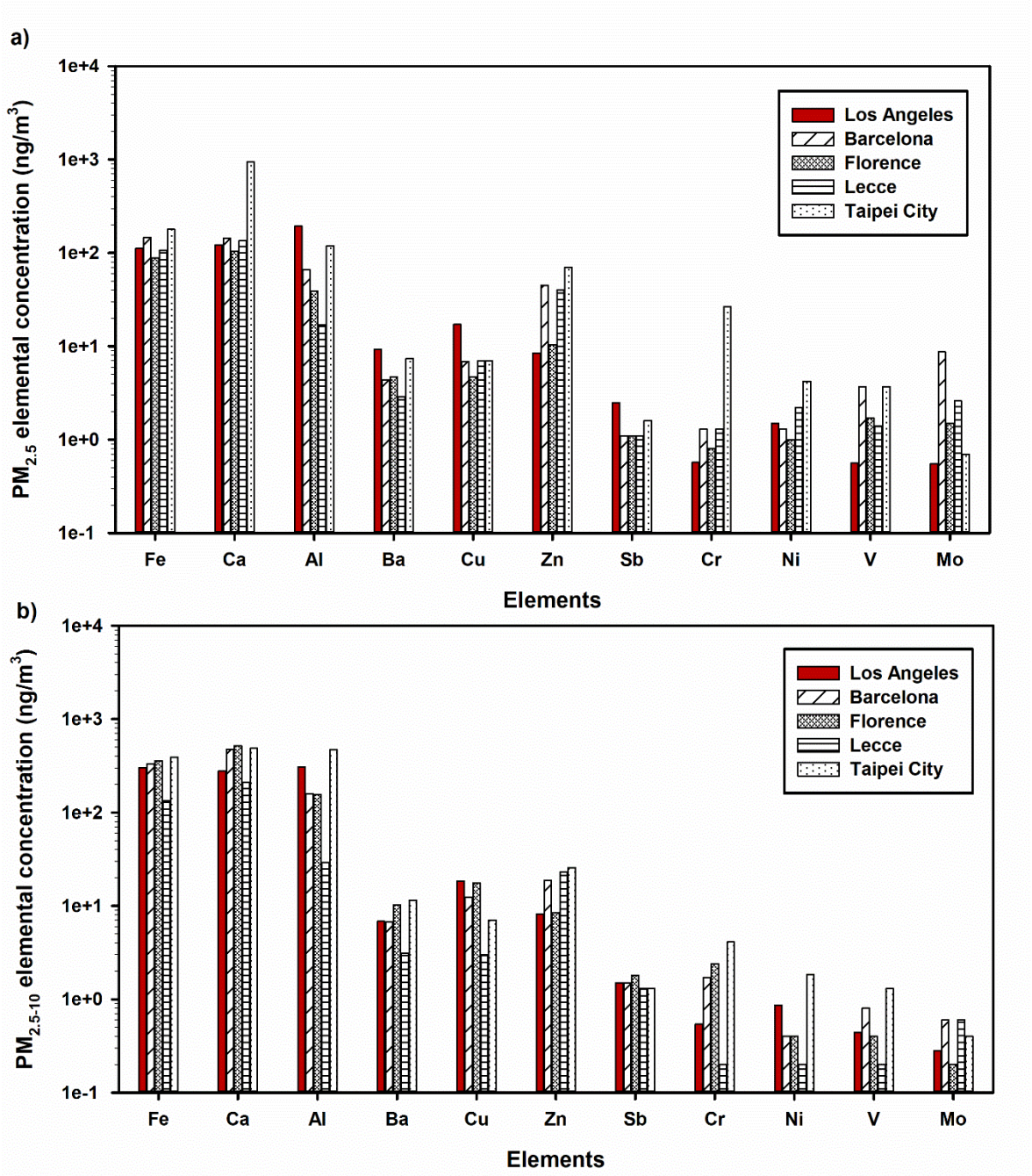

Figure S2.5. Concentrations of selected elements in Los Angeles (current study), Barcelona (Amato et al. 2016a), Florence (Amato et al. 2016a), Lecce (Perrone et al. 2019), Taipei city (Hsu et al. 2019) (a) $PM_{2.5}$ elemental concentrations, (b) $PM_{2.5-10}$ elemental concentrations

The normalized concentrations of selected elements based on the reported values by Habre et al. (2020) are shown in Figure S2.6. In both the current and previous studies, two-week integrated samples were collected in both warm and cold times of the year, and the chemical speciation analysis was performed at Wisconsin State Laboratory of Hygiene (WSLH) through Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS) using similar filter and Polyurethane foam (PUF) substrates. The sampling locations of the previous studies were also bounded to a similar area in Southern California, covering coastal, urban, and suburban locations with various emission sources, including traffic, industrial, and combustion sources. Therefore, we expect the comparison between the elemental composition of the two studies to be meaningful.

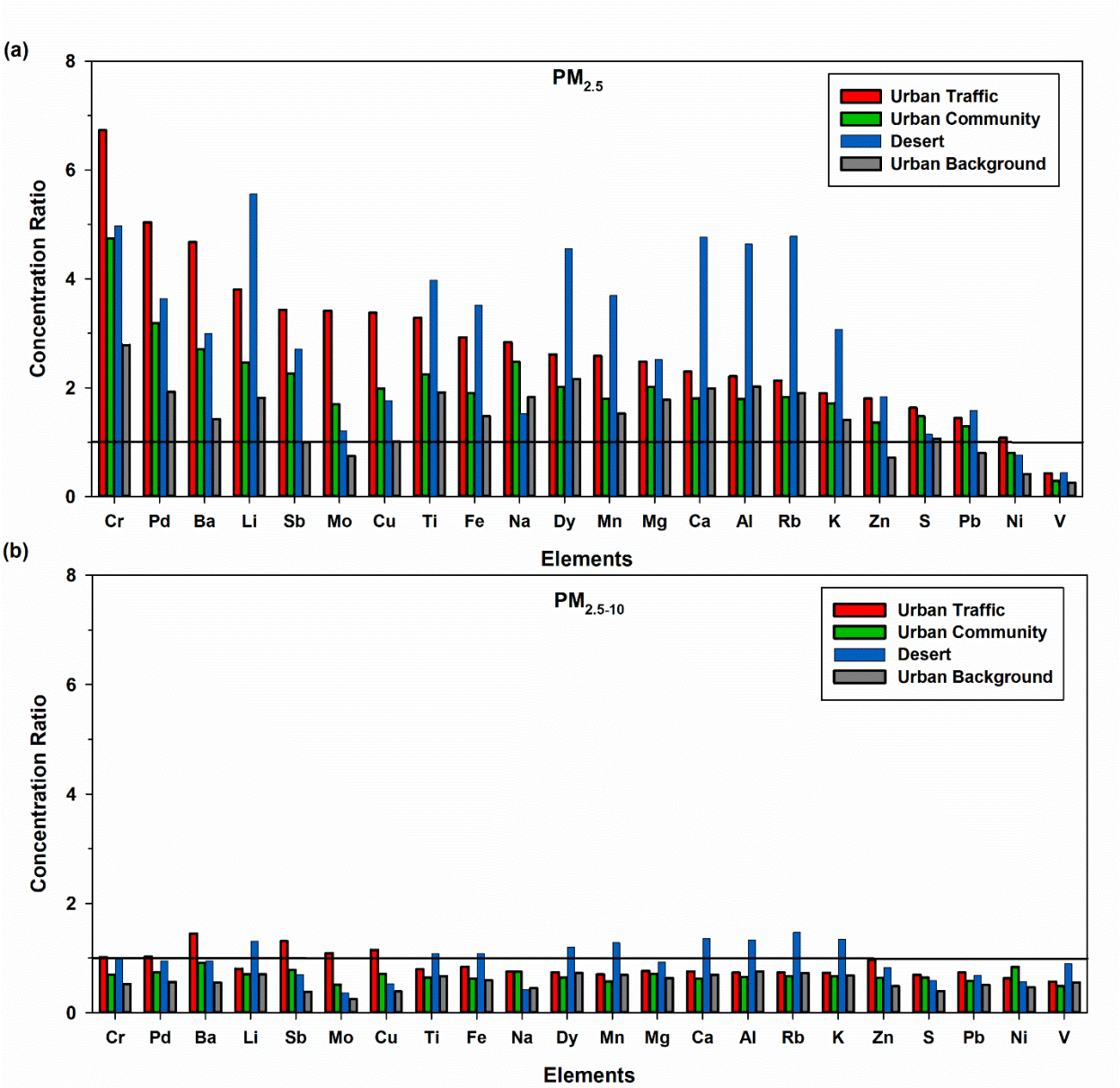

Figure S2.6. Concentration ratios of selected elements between this study and a previous study of Southern California communities in 2008-2009 (Habre et al. (2020)) (a) $PM_{2.5}$ elements (b) $PM_{2.5-10}$ elements

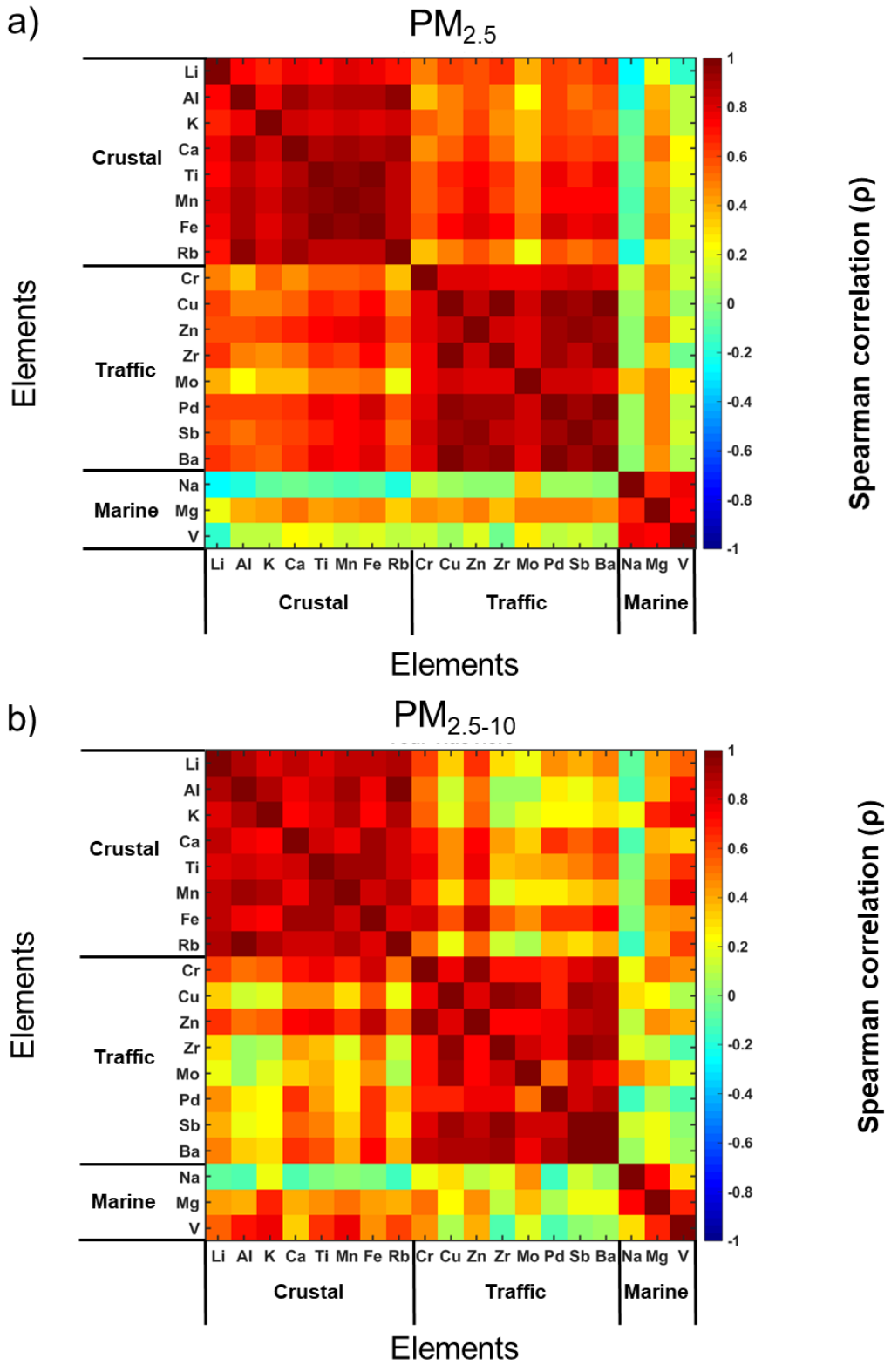

Figure S2.7. Spearman correlation matrix for selected trace elements representing marine, traffic, and crustal sources (a) $PM_{2.5}$, (b) $PM_{2.5-10}$

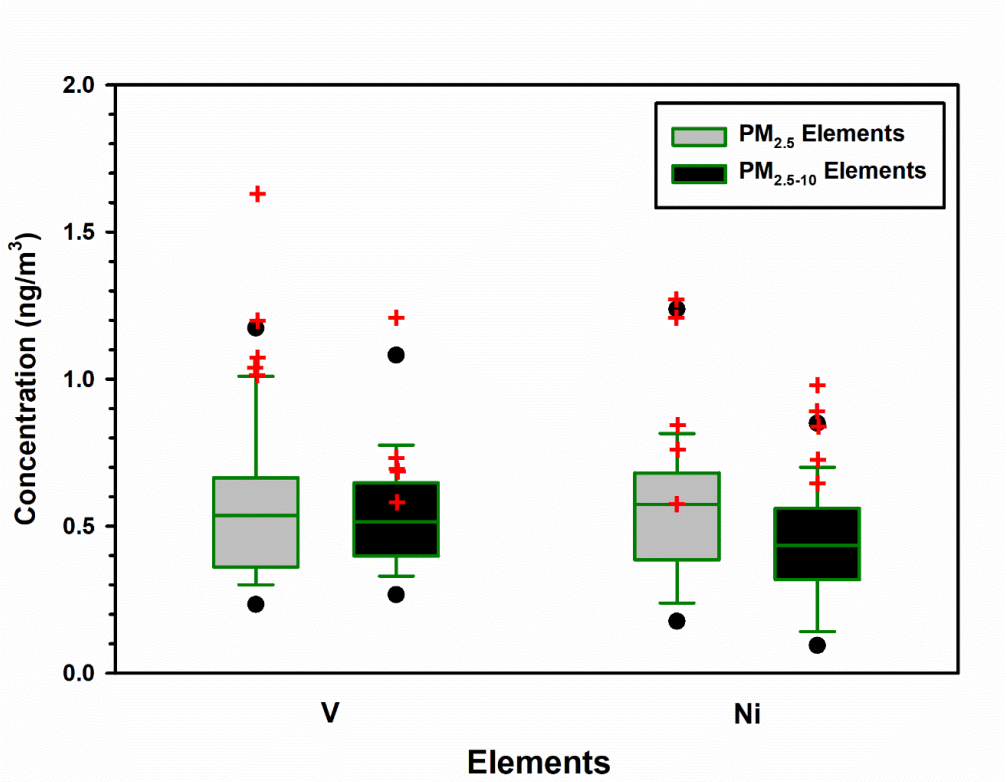

Figure S2.8. The distribution of $PM_{2.5}$ and $PM_{2.5-10}$ concentrations of V and Ni. The red marks denote the elemental concentrations of V and Ni at the sampling locations within 8 miles of Ports of Long Beach and Los Angeles

To assess the performance of the model at different sampling locations, an analysis was performed on model residual values. Mean absolute percentage error (MAPE) was calculated based on Eq (S.1) (Bai et al. 2019):

$$MAPE = \frac{100}{n} \sum_{t}^{n} \left| \frac{A_t - F_t}{A_t} \right| \quad \text{(S.1)}$$

Where *n* is the number of samples, and A_t and F_t are the observed and predicted values.

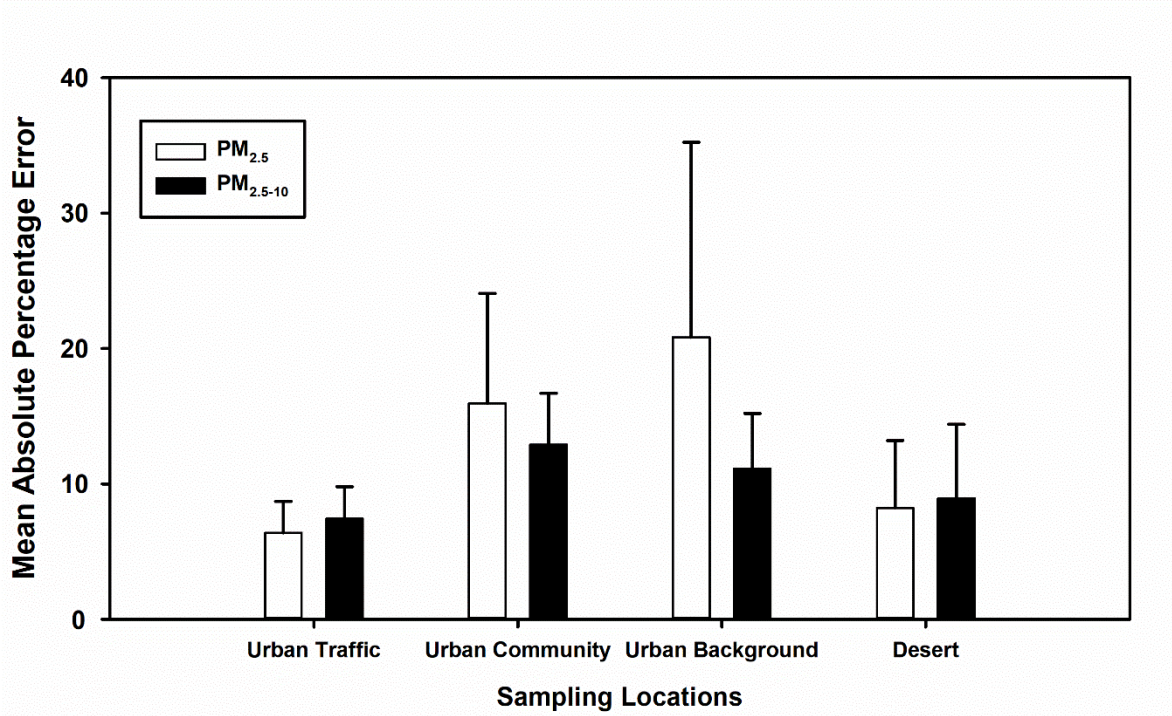

Figure S2.9. Mean absolute percentage error (MAPE) at various sampling locations

3 Associations between Aerosol Oxidative Potential Measured with the Dithiothreitol and Hydroxyl Radical Assays and Metals, Emissions Sources and Socioeconomic Status in the Greater Los Angeles Area

3.1 Abstract

It has been proposed that oxidative potential (OP) might be a better metric than $PM_{2.5}$ mass to consider in studies on the adverse health effects of particulate air pollution exposure. Here, we investigate how OP depends on chemical components and sources and how exposures to different levels of OP vary among land use types and by socioeconomic status of residents in the Greater Los Angeles California area. We measured OH formation (OP^{OH}) and dithiothreitol loss (OP^{DTT}) for $PM_{2.5}$ samples that were collected during two two-week periods during September 2019 and February 2020 at 51 sites, with 4 replicate sites. Our results show that mass-normalized OP^{OH} and OP^{DTT} were most strongly correlated with Zr, Ba, Cu and Pb, Ba, Zr, respectively. We then measured OP^{OH} and OP^{DTT} for the metals that showed the highest correlations with OP individually and in some cases, pairs of metals. Our laboratory measurements confirmed the high activity for Cu(II) in both assays, and Fe(II) and to a lesser degree Fe(III) in the OH assay, and uncovered an antagonistic effect for Pb. Ba showed no activity, and thus the strong association with OP was likely due to its correlation with Cu. A Positive Matrix Factorization source apportionment model was applied to identify sources that may contribute to OP^{OH} and OP^{DTT}. OP^{OH} was attributed to four sources: tailpipe, brake and tire wear, marine (including a contribution from shipping ports), and soil mixed with industrial emissions. Tailpipe emissions contributed more than half of total OP^{OH} signals, followed by 16% from brake and tire wear. Taking extra modeling uncertainty, the source apportionment for OP^{DTT} produced similar source profiles and contributions. Finally, we linked the OP data with the CalEnviroScreen database, a census tract-level compilation of several socioeconomic, pollutant exposure, and health outcome metrics. Residents living in the most socioeconomically disadvantaged census tracts (lowest quartile) were exposed to about 27, 37 and 62% higher levels of $PM_{2.5}$ mass, volume-normalized OP^{DTT}, and OP^{OH} than residents in the highest socioeconomic status group (highest quartile), respectively. The large variation of volume-normalized OP^{OH} across different socioeconomic status groups was not only because more social disadvantaged areas were exposed to higher particle mass concentration, but also because those particles had higher intrinsic toxicity.

3.2 Introduction

Airborne particulate matter smaller than 2.5 microns in diameter ($PM_{2.5}$) is widely recognized as contributing to a wide range of adverse health outcomes, including all-cause mortality, cardiovascular mortality, cardio-respiratory morbidity, metabolic diseases such as diabetes, cognitive decline, and adverse birth outcomes (Coker et al. 2015, Burnett et al. 2018, Pope et al. 2018). A leading hypothesis of why particulate matter (PM) could be responsible for some of these adverse health effects is by inducing oxidative stress. Epithelial lining fluid (ELF), a thin layer extending from the nasal cavity to the pulmonary alveoli, acts as the first line of defense against PM inhalation. Ideally, reactive oxygen species (ROS) and antioxidants in ELF remain in balance, but inhaled particulate matter can catalyze the reduction of oxygen and produce excess ROS. If this overwhelms the antioxidant defenses, oxidative stress occurs (Birben et al. 2012). A handful of studies have shown that the oxidative potential of aerosol – a suspension of particles in liquid

– to be more strongly associated with health outcomes than aerosol mass concentration (Borm et al. 2007, Maikawa et al. 2016, Weichenthal et al. 2016, Bates et al. 2019).

A range of acellular oxidative potential (OP) assays have been developed to quantify different aspects of particles' ability to generate ROS and/or induce oxidative stress. These assays can be divided into assays that measure oxidant production and those that measure the depletion of common lung antioxidants or other organic reductants. The oxidant production assays include the hydroxyl radical (OH) assay (Gonzalez et al. 2018), where OH is the most reactive ROS species formed in surrogate lung fluid, and the electron paramagnetic resonance (EPR) assay (Gehling et al. 2014) that measures particle-bound free radicals. Depletion assays include the ascorbic acid (AA) and glutathione (GSH) assays (Mudway et al. 2001, Ayres et al. 2008, Campbell et al. 2019) and the dithiothreitol (DTT) assay (Kumagai et al. 2002, Cho et al. 2005). While AA and GSH are important cellular antioxidants, DTT acts as a surrogate for biological reductants. OP assays have been consistently positively associated with concentrations of a subset of transition metals, especially soluble copper and iron (Shirmohammadi et al. 2016, Kuang et al. 2019, Yu et al. 2019, Gao et al. 2020). Many studies have also reported strong associations with certain organics, such as quinones or polyaromatic hydrocarbons (Fang et al. 2015) and several have shown strong synergistic interactions between complex organics such as biomass burning organic aerosol and iron (Gonzalez et al. 2017). Many questions remain, however, about which assays are most strongly related to health outcomes, and which components in particles produce a signal in the assays.

Tailpipe emissions have been the subject of numerous regulations since the 1970's, and these have resulted in substantial reductions emissions from tailpipes, even after accounting for the large increases in vehicle miles travelled over the past decades (Parrish et al. 2011). During the same period, increasing vehicle use has increased emissions from brake, tire and engine wear particles, increasing their relative contributions to particulate matter from traffic sources. Consistent with these changes, there has been observed evidence of simultaneous decreases in elemental carbon and polycyclic aromatic hydrocarbons (PAHs) and increasing concentrations of metals in $PM_{2.5}$ over time, many of which are considered tracers for brake and tire wear and road dust (Liacos et al. 2012, Shirmohammadi et al. 2017). Recently, Shirmohammadi et al. (2016) observed a small increase in the mass-normalized aerosol oxidative potential measured by the DTT assay for aerosols collected in the Los Angeles area between 2002 and 2012.

Lower socioeconomic status (SES) has often been found to increase the likelihood that individuals will experience higher exposures and suffer worse health impacts than people of higher socioeconomic status. People of lower socioeconomic status often face a double jeopardy, whereby they have worse environmental exposures and heightened susceptibility to those exposures due to higher rates of pre-existing conditions and other lifestyle factors such as poor nutrition that put them at higher risk (Winkleby et al. 1992, Justice 1999, Morello-Frosch et al. 2001, Morello-Frosch et al. 2002, Brown et al. 2004, Forastiere et al. 2007, Charafeddine and Boden 2008). Less common are studies that describe variations in intrinsic toxicity of air pollution in neighborhoods according to socioeconomic status of its residents.

In this study, we collected 55 $PM_{2.5}$ samples across the Los Angeles Metropolitan area, California, for two 2-week periods during September 2019 and February 2020 and measured their oxidative potential with the OH and DTT assays (OP^{OH} and OP^{DTT}), and concentrations of black carbon (BC) and 52 elements, mainly metals. We (1) investigated the relationships of volume- and mass-normalized OPs with $PM_{2.5}$ mass, BC and elements, (2) performed lab measurements of OP^{OH} and OP^{DTT} for several individual metals; (3) used positive matrix factorization (PMF) model to identify important sources that contribute to OP, including tailpipe, brake and tire wear, marine, soil and industrial sources; (4) explored differences in $PM_{2.5}$ mass, OP^{OH} and OP^{DTT} exposures

by socioeconomic status, environmental exposure, and health outcome metrics at the census tract level as defined by CalEnviroScreen, a database of health, economic and exposure metrics maintained by California's Office of Environmental Health Hazard Assessment (OEHHA).

3.3 Method

3.3.1 Sample Collection

Ambient $PM_{2.5}$ samples were collected at 51 sites with 4 locations as seasonable repeats for a total of 55 samples (Figure 3.1) across the Los Angeles Metropolitan area, California, for approximately 2 weeks each during September 2019 and February 2020. Sampling locations included background, desert, community and traffic sites (Figyre. 3.1). Particles were collected on pre-cleaned 37 mm Teflon filters (Pall Inc.) with the BGI H-PEM (Harvard-Personal Exposure Monitor) at 1.78 ± 0.02 Lpm.

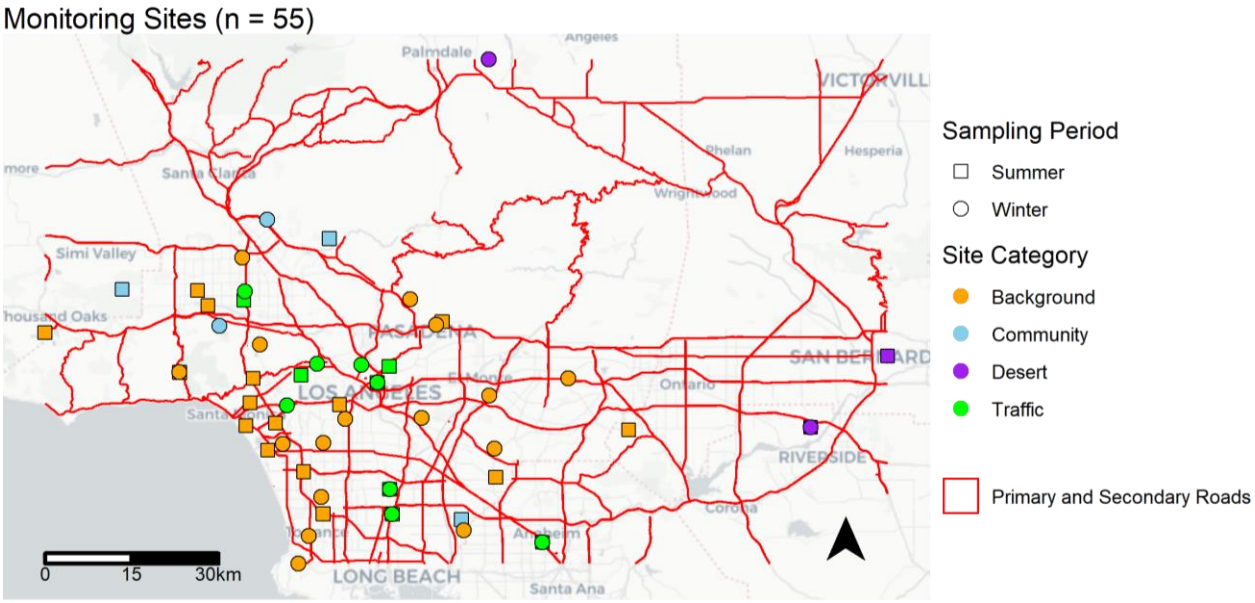

Figure 3.1: Sample site locations and types during two seasons in September 2019 and February 2020 in greater Los Angeles

3.3.2 Chemicals

We purchased 10% trichloroacetic acid, disodium terephthalate and 2-hydroxyterephthalic acid from Fisher Scientific and tris base from Promega. All other chemicals were purchased from Sigma-Aldrich. The highest available purity was selected for all purchases.

3.3.3 Aerosol oxidative potential measurements

After measuring $PM_{2.5}$ mass and BC concentration, we cut the filters in half for the OH and DTT assays, respectively. We added 25 µL of 50% v/v 2,2,2-trifluoroethanol and water mixed solvent to wet half of these filters before putting them into incubation solutions. Buffers used in the measurements were treated with Chelex 100 Resin (Bio-Rad Laboratories, Inc.) to remove trace metal contamination.

3.3.3.1 OH assay

The OH assay measures the OH radical formation during a 2-hour incubation of samples in surrogate lung fluid (SLF). The SLF consists of 200 μM Ascorbate, 100 μM each reduced glutathione and uric acid sodium salt dissolved in 10 mM phosphate buffer (114 mM NaCl, 7.8 mM sodium phosphate dibasic and 2.2 mM potassium phosphate monobasic). Solutions were made freshly before each experiment. We incubated the filters in SLF at 37 °C in 15 mL Falcon tubes (Corning, Falcon®) and adjusted the volume of SLF for each sample to maintain $PM_{2.5}$ incubation concentration at 25 μg/mL and avoid any concentration-dependent effects on the measurements. We quantified the OH radical by adding 10 mM disodium terephthalate (TA) to react with OH and form 2-hydroxyterephthalic acid (TAOH). TAOH is highly fluorescent and was quantified at $\lambda_{ex}/\lambda_{em}$ of 320/420 nm using the Lumina Fluorescence Spectrometer. The yield of TAOH is pH dependent and 33.14% at pH 7.3 (Gonzalez et al. 2018). A calibration curve for TAOH ranging from 0-800 nM was constructed daily.

3.3.3.2 DTT assay

For the DTT assay, we followed the procedures of Cho et al. (2005). The DTT solution used in the assay was 100 μM DTT in 100 mM phosphate buffer (78 mM sodium phosphate dibasic and 22 mM potassium phosphate monobasic). Half filters were incubated in the 100 μM DTT solution at 37 °C in 50 mL polypropylene centrifuge tubes (Thermo Scientific). The $PM_{2.5}$ incubation concentration was fixed at 10 μg/mL. At 8, 16, 24 and 32 min, we took 0.25 mL aliquots of the reaction mixture and added it to 0.25 mL of 10% trichloroacetic acid to quench the reactions. When all time points were quenched, we added 25 μL of 10 mM dithiobisnitrobenzoic acid (DTNB) to the reaction mixture and waited for 5 min to allow the reactions to fully proceed. We then added 1 mL of 0.40 M Tris-Base (pH 8.9) with 20 mM of EDTA. The reaction of DTT and DTNB forms 2-nitro-5-thiobenzoic acid (TNB), which has a molar absorption coefficient of 14150 M^{-1} cm^{-1} at 412 nm (Eyer et al. 2003) over the pH range 7.6 to 8.6. Absorbance was measured in a 96 well microplate in a Tecan M1000 Plate Reader at 25 °C. We found a small increase in the absorption signal for the final solutions during 2 hours, which was possibly due to the alkaline hydrolysis of DTNB which decomposes DTNB at a rate of 0.2% per hour at room temperature and at pH 8 and forms TNB at the same time (Riddles et al. 1979). Therefore, we measured the absorbance of the final solution immediately to maintain consistency across all samples. Finally, a DTT consumption rate was calculated based on the measured DTT concentration at different time points.

3.3.4 Black carbon (BC) quantification

We estimated BC on sample filters with optical absorption at 370 and 880 nm (Magee Scientific Optical Transmissometer). We placed 37mm Teflon filters on quartz filters to obtain an even light on the detector. The instrument reports incident and transmitted light (I_0 and I, respectively). For a filter sample with sampled volume of air (V) and filter collecting area (S), we calculated the absorption coefficient based on Beer's law using the following equation:

$$b_{ATN} = ln\frac{I_0}{I}\frac{S}{V}. \quad (3.1)$$

There are two dominant artifacts associated with filter-based absorption techniques. One is the scattering by the filter fibers, leading to increased light attenuation; the other is the shadowing by the deposited particles upon one another, as particles are not perfectly loaded in a single layer, leading to a reduction in the measured attenuation. Here, we corrected the multi-scattering issue and loading effect for the absorption coefficient using the following expression (Weingartner et al. 2003):

$$b_{ATN,corrected} = ln\frac{I_0}{I}\frac{S}{V}\frac{1}{C_{ref}\cdot\left\{\left(\frac{1}{f_\lambda}-1\right)\frac{ln\left(In\frac{I_0}{I}\right)-ln(10\%)}{ln(50\%)-ln(10\%)}+1\right\}}, \quad (3.2)$$

where C_{ref} corrects the overestimated attenuation from multiple-scattering and the other term compensates for the loading effect. C_{ref} is a constant for all wavelength and a value of 2.14 is commonly used for quartz filters (Weingartner et al. 2003, Ajtai et al. 2011, Zotter et al. 2017). The light absorption coefficient for PTFE has been reported to be 59% of quartz-fiber filters (Vecchi et al. 2014). Therefore, we used 2.14 × 0.59 for C_{ref} for PTFE filters. f_λ is wavelength-dependent, with value of 1.155 and 1.064 for 370 and 880 nm, respectively (Zotter et al. 2017).

Finally, BC concentration is calculated at λ = 880 nm from:

$$BC = \frac{b_{ATN,corrected,\lambda}}{\sigma_\lambda}, \quad (3.3)$$

where 16.6 m^2 g^{-1} @ 880 nm was assumed for σ_λ. This value is recommended by the manufacturer for urban traffic-related BC.

IR only measures blank carbon, thus measurements at 880 mm are usually associated with BC from fossil fuel burning (Gali et al. 2021). UV measures both black and brown carbon. To have a better idea of the relative abundance of BC from fossil fuel burning vs biomass burning, we also calculated AAE (Ångström exponent):

$$AAE = \frac{ln\left(b_{ATN,corrected,370}/b_{ATN,corrected,880}\right)}{ln\left(880/370\right)}. \quad (3.4)$$

3.3.5 Element analysis

We measured 52 elements (mainly metals) using Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS, Thermo-Finnigan Element 2XR), following the microwave-assisted dissolution protocol as previously detailed (Lough et al. 2005, Herner et al. 2006, Dillner et al. 2007). A mixture of 1.5 mL 16 M HNO_3, 0.5 mL 12 M HCl, and 0.2 mL 28 M HF was used for dissolution of PM on Teflon filters and the digestates were finally diluted to 15 mL before measuring within the SF-ICP-MS.

3.3.6 Laboratory measurements of metal activity in the DTT and OH assays

Metal stock solutions from copper(II) sulfate pentahydrate, copper(I) chloride, iron(III) chloride hexahydrate, iron(II) sulfate heptahydrate, zinc(II) sulfate heptahydrate, barium(II) chloride, tin(II) chloride, mercury(II) chloride, lead(II) nitrate, antimony(III) chloride, and antimony(V) chloride were prepared daily by dissolving metals in milliQ water, and then diluted so that the final solutions had concentrations of 1 μM in the OH and DTT assays. MilliQ water analyzed in the same manner as our samples was used as a blank; all reported values were blank corrected.

3.3.7 Data analysis

The measured OH formation rate, DTT loss rate and BC concentration data were further converted to mass or volume normalized data. Before analyzing the data, missing values were replaced with the geometric mean of that data set, and for each metal species concentrations below the detection limit were replaced by half of the detection limit. Spearman's correlation analysis was performed with the SPSS software (SPSS Inc., version 27, USA). Although some

sites were excluded from exposure modeling and description data due to relatively large uncertainties in metal data, we decided to keep them because the main focus of this chapter is oxidative potential instead of metal. We are more interested in the contribution of metals to OP and uncertainty in one metal concentration does not necessarily have an impact. In addition, the data size required for PMF is larger and only 15 elements (including BC) were selected in the PMF model and thus it is unnecessary to exclude sites with uncertainties in other elements. Overall, we used a slightly less stringent inclusion criterion to maximize our available sample.

We used the US Environmental Protection Agency's positive matrix factorization (PMF) model (version 5.0) to identify the major sources for the volume normalized OP^{OH} and OP^{DTT}. PMF is a multivariate factor analysis tool that decomposes a matrix of speciated sample data into factor contributions and factor profile matrices and the model is governed by the chemical mass balance equation:

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \quad (3.5)$$

where x_{ij} refers to a speciated set with i number of samples and j number of species; p refers to the number of factors; g_{ik} is the contribution of the k^{th} factor to i^{th} sample; f_{kj} is the loading of j^{th} species in the k^{th} factor; and e_{ij} represents the residual error for the i^{th} sample and j^{th} species.

PMF also requires uncertainty data. Uncertainty of the OP^{OH}, OP^{DTT} and BC experimental data for each season was assumed to be three times the standard deviation (corresponding to a confidence interval of 99.7%) of multiple measurements of blanks. The experimental uncertainty was then converted into uncertainty for the volume normalized OP^{OH}, OP^{DTT} and BC concentrations using the general laws of uncertainty (Farrance and Frenkel 2012). Uncertainty for element measurements was the propagation of three major sources of analytical uncertainty (SF-ICPMS measurement uncertainty, method blank uncertainty and digestion uncertainty). For data below detection limit, uncertainty was set to be $5/6^{ths}$ of the detection limit (Reff et al. 2007). For missing data, the uncertainty was set to be 5 times the mean uncertainty of that data set. We used 15 elements including BC, Na, Mg, Al, S, K, Ca, Ti, Cr, Mn, Fe, Cu, Zn, Sb and Ba in the OH and DTT source apportionment model. The signal to noise ratios for these species and the target species are >1, and thus they were all categorized as "strong" species.

Three base model error estimation methods were applied to evaluate the rotational ambiguity and random errors of selected PMF runs. The base model displacement (DISP) analysis requires that the decrease in Q is less than 1% and no factor swaps occur for the smallest dQmax. Additionally, to be considered valid, PMF runs were required to have at least 80% of the factors mapped in the Bootstrap (BS) analysis and the change in Q associated with the displacement smaller than 0.5% in the BS-DISP analysis.

3.3.8 CalEnviroScreen Data

CalEnviroScreen 4.0 is the latest iteration of the California Communities Environmental Health Screening Tool released in 2021 by the California Office of Environmental Health Hazard Assessment (Sacramento, CA, https://oehha.ca.gov/calenviroscreen/report/draft-calenviroscreen-40). This database consists of quantitative metrics describing pollution exposure, environmental effects, health outcomes, and socioeconomic status at the census-tract level (generally 2500 to 8000 people). To compare $PM_{2.5}$ mass concentration and aerosol oxidative potential to socioeconomic status and other factors, we first removed the seasonal influence of $PM_{2.5}$/OP data by subtracting the seasonal average from each sample and dividing by seasonal standard deviation (recognizing that $PM_{2.5}$ mass and OP had similar patterns in summer and

winter). We then linked the deseasonalized $PM_{2.5}$ and volume normalized OP data with the CalEnviroScreen by assigning the $PM_{2.5}$/OP data to the corresponding census tract and matching them with underlying CalEnviroScreen indicators (8 exposure indicators, 5 environmental indicators, 5 socioeconomic factor indicators, and 3 health outcomes indicators) in the matching census tract. Percentiles for those indicators were used for correlation analysis in SPSS.

3.4 Results and Discussion

3.4.1 Seasonal variability and relationships between $PM_{2.5}$ mass, OP^{OH} and OP^{DTT}

Averages and ranges for $PM_{2.5}$ mass, volume-normalized OP^{OH} and OP^{DTT} (OP_v^{OH} and OP_v^{DTT}) measured during the summer and winter campaigns are summarized in Table 3.1. To better understand the intrinsic oxidative potential of $PM_{2.5}$, we also investigated the mass-normalized OP^{OH} and OP^{DTT} (OP_m^{OH} and OP_m^{DTT}) (Table 3.1). The intrinsic DTT activity in this study (an average of 0.05 and 0.07 nmol min^{-1} μg^{-1} for summer and winter) falls in the range of intrinsic DTT measured from traffic emissions and is above the range for ambient fine particles summarized elsewhere (Bates et al. 2019). Several earlier studies in Los Angeles found much lower OP_m^{DTT}, at around 0.03 nmol min^{-1} μg^{-1} (Hu et al. 2008, Shirmohammadi et al. 2016, Shirmohammadi et al. 2017). The largest source for this discrepancy may be that the earlier studies did not control for the mass concentration of particles in the DTT solutions; Charrier et al. (2016) showed a strongly asymptotic relationship of the DTT response on the concentration of particles in the extraction solution. These studies report they used the method of Cho et al. (2005), who used PM concentrations from 5 – 40 µg/mL; the mass normalized response in this range decreases by a factor of about three. We used a constant value (10 µg/mL) at the lower end of this range, thus, higher values might be expected. Our average OP_v^{DTT} is 0.43 and 0.67 nmol min^{-1} m^{-3} for summer and winter, respectively, which is also higher than the values in the above cited literature, likely for the same reason.

In our study, $PM_{2.5}$ mass, OP_v^{OH} and OP_v^{DTT} are higher in winter, by 17, 53 and 56%, respectively. Average winter OP_m^{OH} and OP_m^{DTT} are 31% and 32% higher than those measured in summer samples. Winter is characterized by less photochemically-generated secondary aerosol formation, more partitioning of semi-volatile organic compounds into the particle phase (Saffari et al. 2014) and less vertical mixing height of the atmosphere in winter compared to summer. This results in somewhat higher $PM_{2.5}$ mass concentrations and lower contributions from inorganic ions and secondary organics in winter. As expected, given the higher mass concentrations in winter, volume-normalized OP was higher, but even after controlling for mass, OP was higher in the winter, consistent with the lower contribution from secondary aerosol materials that appear to be less active in the OP assays. This is also supported by the larger mass fractions of some key metals in the DTT or OH assay, e.g. Cu, Fe and Mn in winter.

Table 3.1. A summary of PM mass, OP and selected metal data for both seasons

	$PM_{2.5}$ ($\mu g/m^3$)	OP_v^{OH} (pmol/min /m^3)	OP_v^{DTT} (pmol/min /m^3)	OP_m^{OH} (pmol/min /µg)	OP_m^{DTT} (pmol/min /µg)	Cu (ng/m^3)	Fe (ng/m^3)
Summer average	8.0	3.9	428	0.48	52.8	7.0	151.0
Summer standard deviation	1.5	1.3	103	0.10	5.4	4.8	86.4
Winter average	9.4	6.0	666	0.63	69.8	10.6	229.1
Winter standard deviation	2.5	2.2	216	0.13	10.1	5.6	94.5

Correlations between $PM_{2.5}$ mass and volume normalized OP^{OH} and OP^{DTT} are shown in Fig. 3.2. Both OP_v^{OH} and OP_v^{DTT} are strongly ($r_s > 0.6$) correlated with $PM_{2.5}$ mass, but the OP_v^{DTT} ($r_s = 0.86 – 0.88$) correlation is markedly stronger than for OP_v^{OH} ($r_s = 0.75 - 0.80$), with slightly larger correlations in winter. OP_v^{OH} and OP_v^{DTT} also strongly correlate ($r_s = 0.80 - 0.81$).

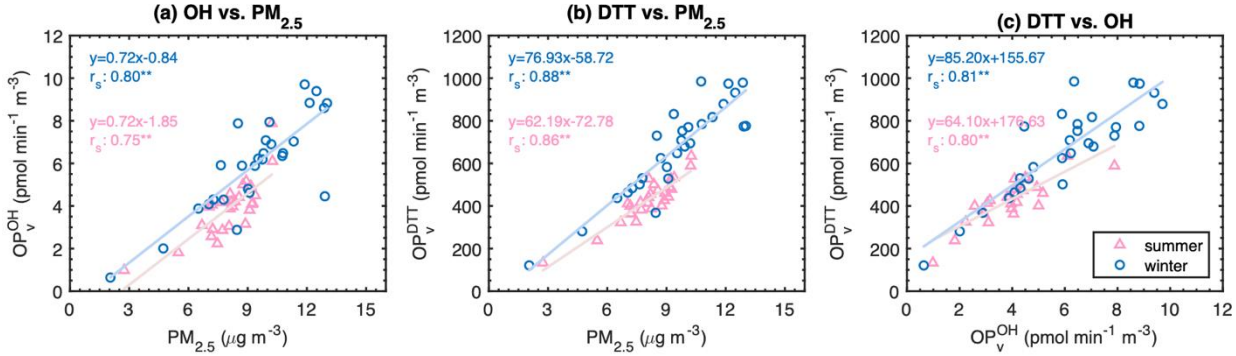

Figure 3.2: Relationships between PM2.5 mass concentration and OP_v^{OH} and OP_v^{DTT}.

Fig. 3.3 shows correlations of mass-normalized OP^{OH} and OP^{DTT} with $PM_{2.5}$ mass and with one another. OP_m^{OH} and OP_m^{DTT} are less correlated ($r_s = 0.53 - 0.60$, both significant at a 99% confidence interval). OP_m^{DTT} is not statistically significantly correlated with $PM_{2.5}$ mass in summer and is moderately ($0.4 < r_s < 0.6$) correlated ($r_s = 0.42$, 95% confidence) in winter. OP_m^{OH} has a moderately positive significant association with $PM_{2.5}$ mass concentration in both seasons. This trend contrasts with many other studies (Visentin et al. 2016, Li et al. 2018, Yu et al. 2019, Campbell et al. 2021), in which an inverse relationship between $PM_{2.5}$ mass and mass-normalized OP was observed, a phenomenon that has been attributed to OP-inactive or low-active components such as inorganic ions that add to the PM mass on highly polluted days (Li et al. 2018). The $PM_{2.5}$ mass concentration in these studies reached 100 μg m^{-3} or even higher, in contrast to our study, for which two-week averages were below 13 μg m^{-3}. Additionally, the mass fractions (or mass-normalized concentrations) of Fe and Cu were positively correlated to the $PM_{2.5}$ mass. Our observed positive correlation between OP_m^{OH}, OP_m^{DTT}, Fe, and Cu, with $PM_{2.5}$ may also reflect an increasing contribution of urban particles with higher available metals and more active organics relative to background aerosols as the $PM_{2.5}$ mass concentration increases.

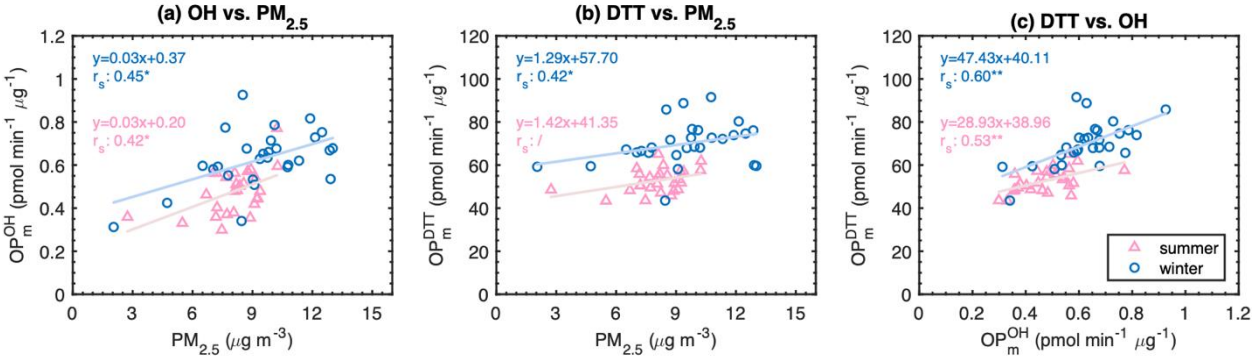

Figure 3.3: Relationships between OP_m^{OH} and OP_m^{DTT} with $PM_{2.5}$ mass and each other

3.4.2 Correlations among OP, BC, and elements

The average AAE measured in this study was about 0.8, indicating that brown carbon from biomass burning was not an important particle source in our samples and BC is predominantly from tailpipe emissions. Los Angeles is sometimes impacted by wildfires, but wildfires were

absent during the measurement periods of this study and residential wood burning is a relatively minor contributor. The dominant role of fossil fuel combustion in total BC in Los Angeles is consistent with earlier studies (Pratsinis et al. 1984, Mousavi et al. 2018).

To understand how OP depends on different chemical components, we calculated Spearman's correlations (r_s) of both volume-normalized and mass-normalized OP^{OH} and OP^{DTT} with BC and elements. For the volume-normalized data, statistically significant correlations were observed between OP and most of the elements, many of which likely result from strong correlations between the element and $PM_{2.5}$ mass concentration, making the data difficult to interpret. Mass-normalized OP^{OH} and OP^{DTT} correlations with measured elements are less strong. OP_m^{OH} and OP_m^{DTT} showed strong (r_s > 0.6) correlations with 14 and 15 out of a total of 53 elements, respectively (Fig. 3.4). Fig. 3.4 also shows Spearman's correlation for selected dust and sea salt tracers.

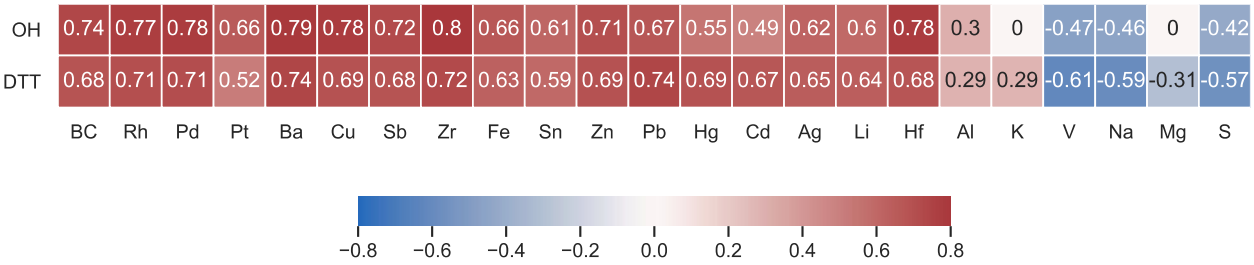

Figure 3.4: Correlation heatmap for mass-normalized OP and selected elements.

The elements commonly associated with brake and tire wear are Ba, Cu, and Sb (Adachi and Tainosho 2004), followed by Zr, Fe, Sn, and Zn, although these four have multiple sources. OP_m^{OH} was mostly strongly correlated with Zr, Ba and Cu (r_s = 0.78 – 0.80), and was also strongly correlated with Sb, Zn, Fe and Sn (r_s = 0.61 – 0.72). Likewise, among the top three elements showing the largest correlations with OP_m^{DTT}, two are brake wear tracers: Ba and Zr (r_s = 0.72 – 0.74). In addition, OP_m^{DTT} also showed fairly strong correlations with Cu, Zn, Sb, Fe and Sn (r_s = 0.59 – 0.69). The strong association between OP and brake and tire wear tracers suggests a role for non-tailpipe emissions in contributing to aerosol oxidative potential.

Mass-normalized OP^{OH} and OP^{DTT} also showed moderate to high correlations (r_s = 0.52 – 0.78) with elements associated with tailpipe emissions, such as BC, Rh, Pd and Pt. The positive correlation between OP and tailpipe tracers has also been observed in many previous studies (Cheung et al. 2010, Shirmohammadi et al. 2016, Lovett et al. 2018). Additionally, OP is also associated with other anthropogenic metals, including Pb, Hg, Cd, Ag, Li, and Hf. Airborne Pb has been attributed to leaded aviation fuel in small piston aircraft, a source that was reported as one of the largest sources of airborne Pb emissions in the United States by 2005 (Murphy et al. 2007), and it still persists in soil. Resuspended road dust has been shown to be another important source of airborne Pb. Higher traffic levels were thought to have the potential to re-entrain larger amount of historically deposited Pb in soil (Cho et al. 2011). Hg is believed to originate from road dust and non-ferrous metal smelting (Huang et al. 2015, Yu et al. 2019). Cd is found to be emitted from multiple sources, such as tire wear, fuel combustion and metal industries (Tchounwou et al. 2012, Nayebare et al. 2018). Ag, Li and Hf are mostly associated with industrial emissions (Bein et al. 2005, Viana et al. 2008, Furutani et al. 2011).

Clean sea salt particles contain Na, Mg, and S (Ebert et al. 2000, Adachi and Buseck 2015) and V is a tracer for heavy fuel oil combustion by ships (Corbin et al. 2018). Na, S and V were all negatively correlated with OP_m^{OH} and OP_m^{DTT} (r_s = -0.42 to -0.61); Mg had no correlation with

OP_m^{OH} (r_s = 0) and only a weak (r_s < 0.4) negative correlation with OP_m^{DTT} (r_s = -0.31). The negative correlations between mass normalized OP and sea salt aerosols suggest the smaller toxicity of sea salt aerosols compared with aerosols emitted from traffic or industries and a dilution effect on OP_m^{OH} and OP_m^{DTT}.

3.4.3 Laboratory measurements of OP^{OH} or OP^{DTT} for individual and combinations of metals

In our aqueous oxidative potential analyses, chemical components need to be dissolved in the solution to be detected, thus soluble concentration is an important factor influencing the toxicity of aerosol chemical components. Combining our measurements of total metals with solubility obtained from the literature, we can estimate metal concentrations in the solutions. The top 10 metals with the highest correlations with OP_m^{OH} or OP_m^{DTT} are Zr, Ba, Cu, Pd, Rh, Sb, Zn, Pb, Hf, Hg and Fe. Of these, Pd, Rh and Hf have low concentrations (< 0.09 ng m^{-3}) and Zr is only slightly soluble (< 10%), and thus, they are unlikely to contribute much activity. Figure 3.5 shows oxidative potential from individual metals or combinations of the remaining metals (Ba, Cu, Sb, Zn, Pb, Fe, and Hg).

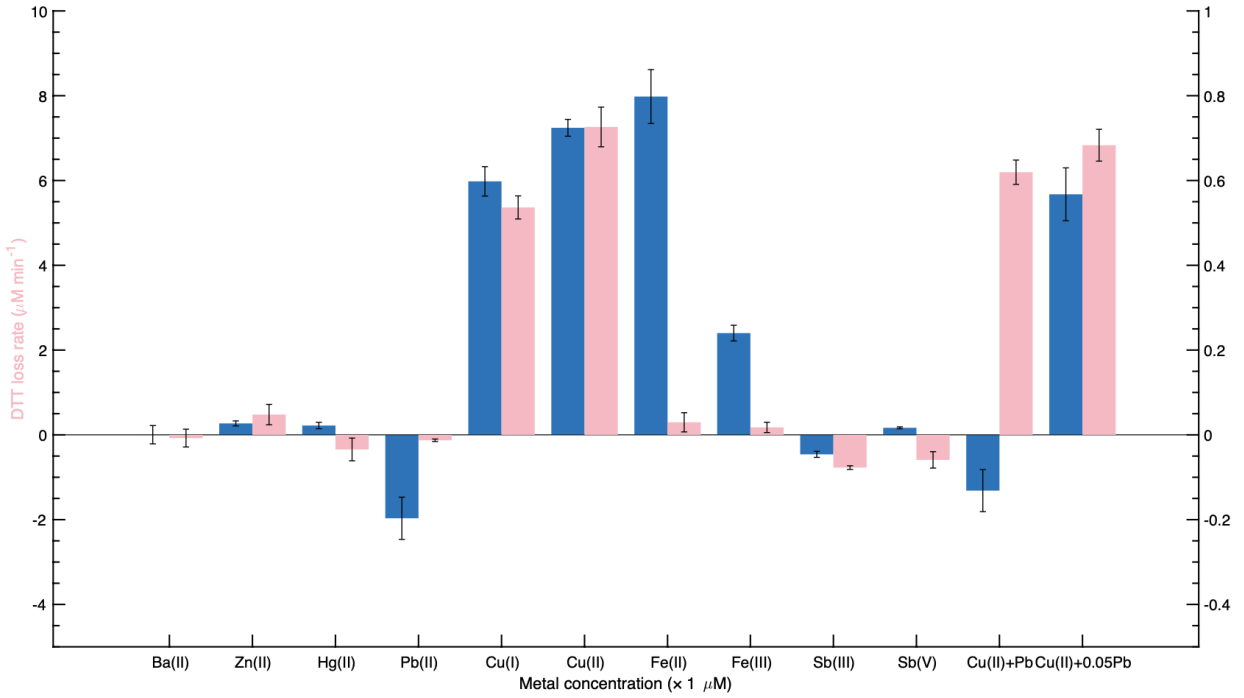

Figure 3.5: Measured OH formation and DTT loss rates from 1 μM individual metals, 1 μM Cu + Pb, and 1 μM Cu + 0.05 μM Pb

The OH activities we measured for Cu(II) and Fe(II) are about 1.6 and 0.8 times the values measured by Charrier and Anastasio (2015), possibly because we did not add citric acid in the surrogate lung fluid used for our measurements.

Charrier and Anastasio (2012) quantified OP^{DTT} for a series of metals and Yu et al. (2018) measured OP^{DTT} for Cu(II) and Fe(II), the values reported in our study for Cu(II) and Fe(II) are at levels between the measurements reported in these two studies. Both our study and Charrier and Anastasio (2012) measured OP^{DTT} for Fe(III), Zn(II) and Pb(II), with Fe(III) and Zn(II) in roughly

good agreement and a discrepancy for Pb(II) activity. Unlike the positive DTT activity for Pb(II) in their study, we observed an inhibitory effect of Pb(II).

Among all the tested metals, Cu and Fe are the most active metals in the OH assay, with both redox states (I and II) of Cu and (II and III) of Fe playing a role. The Fenton reaction Fe(II) + H_2O_2 → Fe(III) + OH + OH^- is an important pathway for generating OH radicals, which can be seen in the high signal of Fe(II) in the OH assay. Antioxidants in lung fluid such as ascorbic acid and glutathione readily react with OH radicals and thus slow down the radical chain reactions and protect cells from oxidative stress. However, Fe(III) and Cu(II) also act as catalysts in the oxidation of ascorbic acid and the reaction produces H_2O_2 (Khan and Martell 1967, Shen et al. 2021). Fe(III) can also react with glutathione, converting the reduced glutathione to the oxidized form and producing Fe(II) at the same time, which can produce OH via the Fenton reaction (Da et al. 2020). As a result, Fe(III) in lung fluid is fairly efficient at producing OH. Furthermore, we observed a synergism between Cu(II) and Fe(II) or Fe(III).

In terms of the DTT assay, Cu is by far most active metal. Both Fe(II) and Fe(III) have modest activity in the DTT assay (Charrier and Anastasio 2012, Yu et al. 2018) and Fig. 3.5. Ba was strongly correlated with both OP_m^{OH} and OP_m^{DTT} (Fig. 3.4), however, it does not show any signal in the OH or DTT experiments (Fig. 3.5). Ba is very strongly correlated with Cu (r_s = 0.95 - 0.96), which likely explains its strong correlation with OP_m^{OH} and OP_m^{DTT}. Likewise, Zn(II) and Sn(II) also contribute little to OP_m^{OH} or OP_m^{DTT}, suggesting that their positive correlations may also be due to collinearity with other metals from brake and tire wear emissions. Pb(II) and Hg(II), both associated with road dust, are strongly correlated with each other, and they are both correlated with other metals including Fe and Mn. Although not tested in our study, Mn has been found to be a very active metal in the DTT assay (Charrier and Anastasio 2012). The positive correlations between Pb, Hg and OP likely result from the association of Pb and Hg with other more redox active metals.

While Pb(II) and Sn(II) positively correlate with OP_m^{OH} and OP_m^{DTT}, they show less of an OP signal than the blanks, especially for OH formation, indicating antagonistic activity. We found and antagonistic effect of Pb(II) on Cu(II) in both the OH formation and DTT consumption measurements of mixtures of these two metals. A 1 μM Cu(II) in SLF results in about a 7.2 nM min^{-1} OH production and a 0.73 μM min^{-1} DTT loss, while adding 1 μM Pb(II) to the solution decreases the DTT signal by 15% and completely suppresses the OH signal. In the ambient air samples we collected in this study, the Cu concentration was approximately 20 times higher than Pb, while their solubilities are comparable for $PM_{2.5}$ (Heal et al. 2005, Sarti et al. 2015). Combining Cu and Pb in a 20:1 ratio results in 22% and 6% decreases due to Pb in the OH and DTT responses, respectively. These synergistic or antagonistic effects illustrate the potential for complex interactions between different metals; organics can have similarly large moderating effects (Gonzalez et al. 2017). Underlining the need for either direct measurements of OP or more work to characterize the complexities underlying OP generation by different components of $PM_{2.5}$.

3.4.4 Source apportionment of aerosol oxidative potential

The PMF OP^{OH} model identified four sources that contribute to OP^{OH} with an R^2 of 0.91, as shown in Figure 3.6. Factor 1 is mainly characterized by high loadings of BC, a tracer for tailpipe emissions (Gali et al. 2021). This factor also contains some sulfur, Ti, Cr and Mn; fossil fuel combustion is usually associated with sulfur oxide emissions, and these three metals can be emitted by diesel vehicles (Liu et al. 2018). A small amount of Al loads on this factor, a metal associated with road dust (Cheng et al. 2015, Zhongming et al. 2020), along with Fe, Ti, Cr and Mn, metals associated with brake wear (Khairy et al. 2011, Gunawardana et al. 2012, Zhongming et al. 2020). Altogether, this may represent a mixture of road dust in this source. Overall, this factor contributes 60% of the total oxidative potential quantified by the OH assay (Fig. 3.7 (a)).

Factor 2 is dominated by Cu, Zn, Sb and Ba. This factor mainly represents brake and tire wear emissions. Cu is a high-temperature lubricant commonly used in brake pads (Charron et al. 2019, Jeong et al. 2019, Huang et al. 2021). Sb is another typical lubricant used in the brake lining to reduce vibrations and improve friction stability (Varrica et al. 2013, Hagino et al. 2016). Ba is used as filler in brake pads (Gietl et al. 2010, Jeong et al. 2019). Zn is believed to originate largely from tire wear and also to be originating from brake wear and engine lubrication oil (Hjortenkrans et al. 2007, Wang et al. 2021). Fe is found to have multiple anthropogenic and geogenic sources, including tailpipe emission (Wang et al. 2021). It is more commonly associated, however, with brake and engine wear (Charron et al. 2019, Huang et al. 2021, Silva et al. 2021). Ti, Cr, and Mn are also associated with brake wear to some extent (Amato et al. 2011, Apeagyei et al. 2011, Nayebare et al. 2018). Compared with more consistent brake wear tracers, they are less strongly loading on this factor. Brake and tire wear is the source, with the second largest contribution to OPOH, i.e. a contribution of 16%.

Factor 3 is characterized by high loadings for Al, Ca, Mn, Fe and Ti, all of which are crustal elements. However, the relatively high loading of Cr, Zn and Ba, and to a lesser extent, Cu and Sb, suggests that this factor may be representing mixed sources of soil and industrial emissions. This factor contributes 14% to OP^{OH}.

The last factor identified in the PMF is associated with marine emissions, and is dominated by Na, Mg and S; loadings of other elements are almost negligible. The factor does contain some BC, consistent with a contribution from emissions by the very active ports of Los Angeles and Long Beach. As expected, this factor makes a small contribution to OP^{OH}, about 9%.

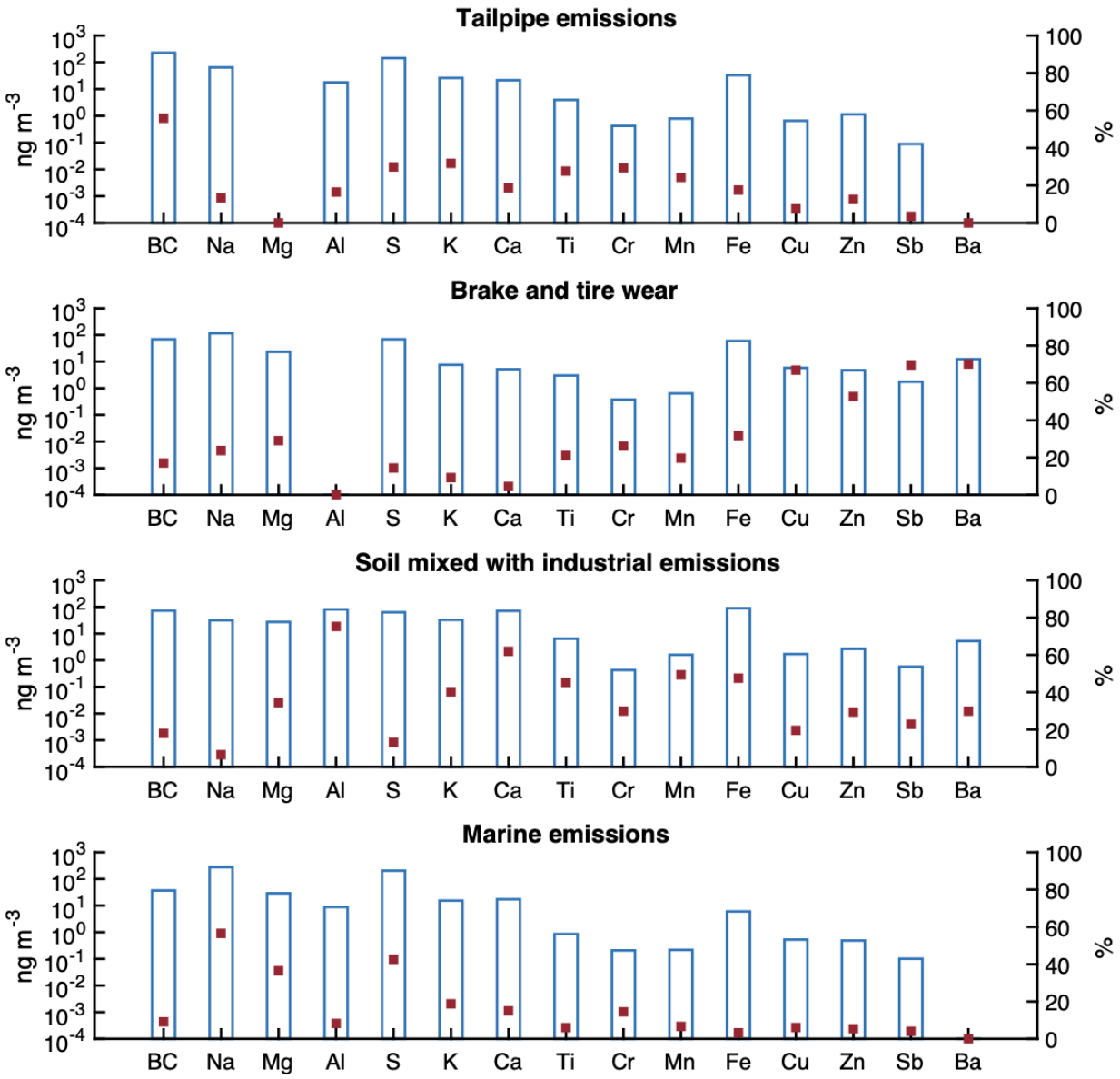

Figure 3.6: Factor profiles of the OP^{OH} PMF model. The bars (left axis) represent the concentration of species and the dots (right axis) denote the percentage of species on each factor

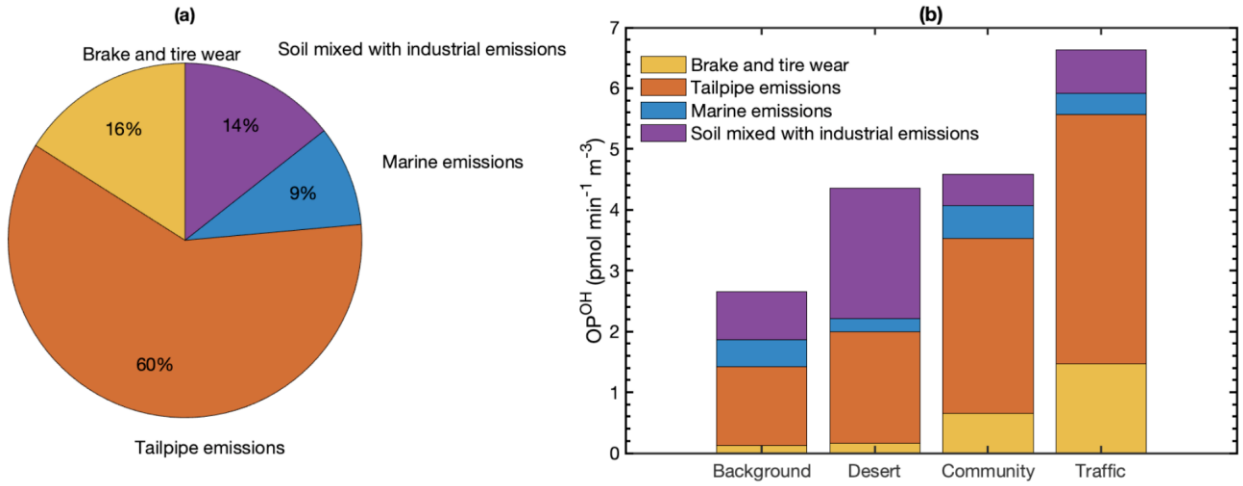

Figure 3.7: Average source contribution of the the OP^{OH} PMF model for all sites (a) and for each site category (b).

Biomass burning was not identified as a source, consistent with the AAE data and despite including potassium (K) in the PMF model. In addition, our PMF model does not identify a secondary aerosol formation source, an outcome that may result from a lack of secondary tracers including inorganic salts and organic components. Besides, our sampling sites are more concentrated on the west side of the Los Angeles Basin, where a sea breeze starting from the coast has not traveled long enough for photochemistry to happen such that secondary aerosols can accumulate.

To understand a picture of the spatial variability of OP^{OH}, we divided the sites into four categories: background, desert, community, and traffic sites based on the environments around each site and their proximity to main roads (Oroumiyeh et al. 2021). The contribution of each source factor resolved by the PMF to total OP^{OH} is shown in Fig. 3.7 (b). The sites have large differences in OP^{OH}; differing by a factor of 2.5 between the traffic and background sites, with the desert and community sites falling in between. Overall, the contributions of each of the factors to each site type are consistent with expectations. Tailpipe emissions make a major contribution to all of the sites, but is largest for the traffic sites, followed by community sites. Brake and tire wear also contributes heavily to the traffic sites, followed by the community sites, but makes very small contributions to the background and desert sites. This is consistent with the fact that mechanically generated $PM_{2.5}$ from brake and tire wear particles are expected to be larger than combustion generated tailpipe particles and thus are expected to travel further smaller than tailpipe emissions. The desert sites in this study were located on the east and north edges of Los Angeles Basin, and are therefore farthest from the Pacific Ocean, and the marine factor contributes very little at the desert sites. At the desert sites we see the largest contributions (both in absolute and fractional terms) from soil.

Despite the limited number of samples, our PMF model was able to separate non-tailpipe and tailpipe emission sources. The PMF model indicates that from an exposure perspective, tailpipe emissions are still the largest source of OP, followed by brake and tire wear emissions. It is noteworthy that the contribution to OP reported in this study is an outcome of both the mass concentration and the intrinsic toxicity of the sources, two contributions that can be disentangled by knowing either of these quantities. Unfortunately, a PMF analysis of the $PM_{2.5}$ mass could not be performed reliably because we do not have measurements of the main contributors to particle mass; BC and metals contribute only about 24 ± 6%. Many studies have reported a steady

decrease in the ratio of tailpipe emissions to non-tailpipe emissions (Jeong et al. 2020, Zhongming et al. 2020), but their relative contributions to PM mass have shown fairly large variations across different studies and sites (Habre et al. 2021). Interestingly, Fe and Cu, the two most active metals in the OH assay, are both larger contributors to the brake and tire wear source than the tailpipe source. However, our measurements were of total metals, not soluble metals, and soluble metals may be different for traffic and brake and tire wear particles. Further, organic chelators, also not characterized here, can increase or decrease metal activity.

Our PMF model for OP^{DTT} produced similar sources and contributions as the OP^{OH} model. Taking extra modeling uncertainties was necessary, however, to produce a stable and reliable source apportionment result. While both measurements of OP respond strongly to soluble Cu, other responses differ. The relationship between metals and DTT activity remains controversial. Some studies have claimed that in this assay organics are more important than metals (Cho et al. 2005; Fang et al. 2016) as these studies typically observed stronger correlations between DTT activity and organics. Other studies have argued for a metal dominance, going as far as proposing a simple additivity for individual metal responses to generate the oxidative potential (Charrier and Anastasio 2012; Charrier et al. 2015); yet this is unlikely to be accurate even from the point of view of metals alone. Organics clearly play a role in the DTT assay, both directly and by modifying the activity of metals. Taken together, more measurement data that also includes water soluble organics and metals is needed to for a more reliable source apportionment for DTT activity.

3.4.5 Exploratory environmental justice analysis

3.4.5.1 Association of $PM_{2.5}$, OP_v^{OH} and OP_v^{DTT} with pollution burden

To understand more fully whether the OP indicators were associated with social disadvantage or other exposures, we conducted exploratory correlational analyses with various elements of the CalEnviroScreen 4.0. Tab. 3.2 shows the bivariate Spearman's correlations between $PM_{2.5}$ mass/OP and pollution burden indicators from CalEnviroScreen 4.0. $PM_{2.5}$ mass, OP_v^{OH}, OP_v^{DTT}, OP_m^{OH} and OP_m^{DTT} all showed weakly to moderately positive correlations with exposure indicators related to traffic-related air pollution ($PM_{2.5}$ concentrations, diesel particulate matter emissions, traffic impacts) with the exception that $PM_{2.5}$ mass was not correlated with traffic impacts. Children's lead risk from housing, a metric based both on the likelihood of lead-based paint in housing and the percentage of low-income households with children was also moderately correlated with $PM_{2.5}$ mass, OP_v^{OH}, OP_v^{DTT} and OP_m^{OH}. The toxic releases from facilities indicator was moderately correlated with $PM_{2.5}$ mass and weakly correlated with volume-normalized OP. Factors that may reduce correlations include differences in the time periods sampled (our study was conducted in 2019/20, the CalEnviroScreen air data was from various dates before 2018), the fact that we sampled in only one location within a census tract, and other factors such as measurement and estimation errors.

Table 3.2. Correlation matrix of $PM_{2.5}$/OP and exposure indicators from CalEnviroScreen

		This study					CalEnviroScreen exposure indicators					
		$PM_{2.5}$	OP_v^{OH}	OP_v^{DTT}	OP_m^{OH}	OP_m^{DTT}	Ozone	$PM_{2.5}$	Diesel PM	Toxic releases	Traffic	Children's lead risk from housing
CalEnviScreen exposure indicators	Ozone	-0.28*	-0.09	-0.12	0.15	0.15	--					
	$PM_{2.5}$	0.45**	0.51**	0.50**	0.49**	0.33*	-0.14	--				
	Diesel PM	0.47**	0.56**	0.50**	0.50**	0.36**	-0.23	0.61**	--			
	Toxic releases	0.40**	0.32*	0.28*	0.17	0.01	-0.60**	0.50**	0.38**	--		
	Traffic	0.24	0.34*	0.35**	0.30*	0.36**	-0.24	0.28*	0.64**	0.06	--	
	Children's lead risk from housing	0.46**	0.46**	0.44**	0.42**	0.23	-0.32*	0.47**	0.50**	0.50**	0.20	--

** denotes correlations are at a 99% confidence interval and * for 95% confidence interval.

3.4.5.2 $PM_{2.5}$/oxidative potential and socioeconomic factors

Tab. 3.3 shows the Spearman's correlations of $PM_{2.5}$ and OP with five socioeconomic factors (educational attainment, housing-burdened low-income households, linguistic isolation, poverty, and unemployment). Socioeconomic factors all either weakly or moderately correlated (r_s = 0.34 – 0.53) with $PM_{2.5}$ mass and volume-normalized OP, and they were correlated with each other. Mass-normalized OP^{OH} showed similar correlations with socioeconomic factors as volume-normalized OP^{OH}, while mass-normalized OP^{DTT} exhibited weaker correlations compared with volume-normalized OP^{DTT}. Poverty and housing-burdened low-income households were the two factors that were mostly associated with $PM_{2.5}$ mass and OP compared with the remaining factors.

Table 3.3. Correlations of $PM_{2.5}$ mass/OP with socioeconomic factors

		This study					CalEnviroScreen socioeconomic indicators				
		$PM_{2.5}$	OP_v^{OH}	OP_v^{DTT}	OP_m^{OH}	OP_m^{DTT}	Educational attainment	Linguistic isolation	Poverty	Unemployment	Housing burden
CalEnviroScreen socioeconomic indicators	Educational attainment	0.34*	0.37**	0.38**	0.42**	0.29*	--				
	Linguistic isolation	0.38**	0.42**	0.42**	0.46**	0.28*	0.76**	--			
	Poverty	0.44**	0.49**	0.44**	0.50**	0.28*	0.88**	0.74**	--		
	Unemployment	0.53**	0.44**	0.37**	0.31*	0.05	0.45**	0.32*	0.58**	--	
	Housing burden	0.45**	0.50**	0.43**	0.49**	0.31*	0.66**	0.61**	0.77**	0.41**	--

*95% confidence interval; **99% confidence interval.

We divided sites into different SES groups based on the grouped socioeconomic factor defined in the CalEnviroScreen, and we found that summer and winter samples were almost evenly distributed in each group. $PM_{2.5}$ mass, OP_v^{OH}, OP_v^{DTT}, OP_m^{OH} and OP_m^{DTT} before deseasonalization are plotted against the socioeconomic group quartiles in Fig. 3.8. We can see from the figure that $PM_{2.5}$, OP_v^{OH} and OP_v^{DTT} levels consistently increased as socioeconomic disadvantages intensified, and the differences increased in order of $PM_{2.5}$, OP_v^{DTT} and OP_v^{OH}. People in the highest socioeconomic disadvantage quartile were facing the highest levels of pollution, compared with people in the lowest quartile, on average they were exposed to 27%, 62% and 37% more $PM_{2.5}$ mass, OP_v^{OH} and OP_v^{DTT}, respectively. Both volume-normalized indicators of OP showed greater relative exposure gradients over SES gradients than $PM_{2.5}$ mass, although our relatively limited sample prevents us from drawing definitive solutions. OP_m^{OH} showed similar correlations with individual socioeconomic indicators as OP_v^{OH}, but it had less variation among different SES groups, with similar values in the second and third quartile, while

the difference in the lowest and highest quartiles was large. Altogether this indicates that the higher OP^{OH} people in more disadvantaged community experienced was not only a result of higher particle mass concentrations, but also because of a more toxic airborne environment they were exposed to. The variations among different groups for OP_m^{DTT} was much smaller compared with the oxidative potential quantified by the OH assay, suggesting that the chemical components that were responding to the DTT assay did not have much variability across different groups, and thus the trend of OP_v^{DTT} was similar to $PM_{2.5}$ mass, in line with the strong correlations we saw between these two.

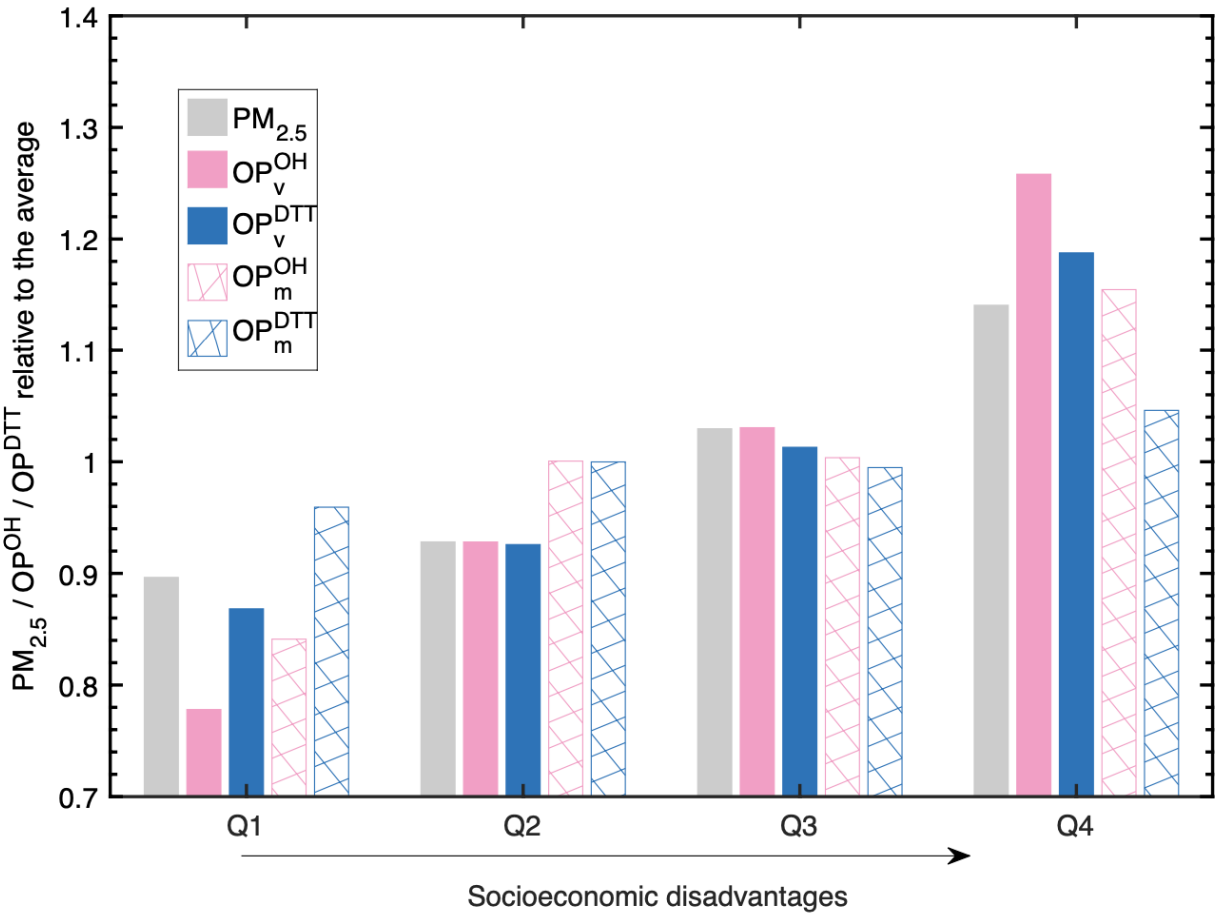

Figure 3.8: PM mass and oxidative potential relative to their average in different socioeconomic groups. The bars denote the average $PM_{2.5}$ mass or OP in each group.

People with low income and housing affordability are more likely to live near high-traffic areas and thus they were exposed to more pollutants, especially vehicular emissions, found to be the largest contributor to aerosol oxidative potential in this study. Although the industry mixed with soil source only contributed 14% to OP_v^{OH}, the contribution was also higher for more socioeconomically disadvantaged groups, suggesting that this source might contribute to the disproportionate burden of oxidative potential as well.

3.4.5.3 3.4.5.3. $PM_{2.5}$/oxidative potential and adverse health outcomes

Table 3.4. shows the Spearman's correlations between $PM_{2.5}$ mass/OP and three adverse health outcomes (asthma, cardiovascular disease, and low birth-weight infants) at the census tract level. OP_v^{OH} and OP_m^{OH} were statistically significantly correlated with the census tract group prevalence

of all three health outcomes (Tab. 3.4), with correlations that were weak to moderate (r_s = 0.30 – 0.44). The relationships of OP_v^{DTT} with these health outcome indictors were almost the same as those of $PM_{2.5}$ mass; both were more strongly correlated with the prevalence of low birth-weight infants (r_s = 0.39 – 0.4), while OP_m^{DTT} did not correlate any health outcomes. The overall relatively low correlation coefficients are likely partly due to exposure variation within census tracts that this ecologic measure cannot detect, a lack of coincidence in timing (our exposure data was from 2019-20; while the asthma and cardiovascular data was from 2015 – 2017 and the birth outcome data from 2009 - 2015); the r_s for our measured $PM_{2.5}$ mass concentration with the CalEnviroScreen $PM_{2.5}$ value at the census tract was only 0.45 (Tab. 3.2). In contrast, earlier studies found statistically significant associations between OP^{DTT} and asthma (Bates et al. 2015, Yang et al. 2016, Abrams et al. 2017), and cardiovascular disease (Bates et al. 2015, Abrams et al. 2017) while the association between asthma, cardiovascular disease and PM mass was weaker or absent. OP^{OH} has not previously been investigated in an epidemiological context. Our results suggest that the OH assay may be better at predicting adverse health outcomes.

Table 3.4. Associations of $PM_{2.5}$ mass/OP with adverse health outcomes

		This study					CalEnviroScreen health indicators		
		$PM_{2.5}$	OP_v^{OH}	OP_v^{DTT}	OP_m^{OH}	OP_m^{DTT}	Asthma	Cardiovascular disease	Low birth-weight infants
CalEnviroScreen health indicators	Asthma	0.20	0.30*	0.19	0.39**	0.11	--		
	Cardiovascular disease	0.26	0.32*	0.22	0.37**	0.12	0.85**	--	
	Low birth-weight infants	0.40**	0.44**	0.39**	0.35**	0.21	0.59**	0.46**	--

*95% confidence interval; **99% confidence interval.

3.5 Conclusions

In this study we measured oxidative potential (OP^{OH} and OP^{DTT}) for 55 samples in the Greater Los Angeles Area during late summer 2019 and winter 2020. Correlation analysis showed that oxidative potential was strongly associated with tracers of tailpipe emissions and brake and tire wear. A PMF source apportionment analysis further identified four emission sources contributing to volume-normalized OP^{OH}, with tailpipe and brake and tire wear being the two largest contributors, followed by soil mixed with industrial emissions and marine emissions. The PMF results for OP^{DTT} were similar but weaker. Linking the oxidative potential data with CalEnviroScreen, revealed a disproportionate burden of $PM_{2.5}$/both oxidative potential metrics for people in lower socioeconomic status groups. However, the largest difference was for OP^{OH}, indicating that the lower socioeconomic groups are both exposed to more particle mass, and that these particles may be more toxic. OP^{OH} was also significantly correlated with the CalEnviroScreen health outcome data, $PM_{2.5}$ and OP^{DTT} were less so. Overall, this may suggest a role for the OH assay in predicting PM-induced adverse health outcomes.

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4 Co-kriging with external drift using a low-cost sensor network to model spatial variation of brake and tire-wear related heavy metals and reactive oxygen species in Southern California

4.1 Abstract

Due to regulations and technological advancements reducing tailpipe emissions, there is an increased focus on non-exhaust automobile emissions, including break and tire wear particulate matter (PM) which contain heavy metals capable of generating oxidative stress in exposed organisms. At the same time, improvements in electrical engineering, internet connectivity, and an increased public concern over air pollution have led to a proliferation of dense low-cost air sensor networks such as the PurpleAir monitors, which primarily measure unspeciated fine particulate matter ($PM_{2.5}$).

In this study, we model the concentrations of barium, zinc, 2-hour reactive oxygen species (ROS) formation, and black carbon alongside DTT loss and OH formation. We use a cokriging approach, incorporating data from the PurpleAir network as a secondary predictor variable and a land-use regression (LUR) as an external drift. We obtained land-use variables such as traffic, business density, tree canopy cover, and impervious surfaces.

Our LUR models are associated with adjusted R^2 values between 0.37 and 0.68, depending on outcome. Adding in PurpleAir data with co-kriging improved predictive accuracy in many of our outcomes, with significant increases predictive accuracy ranging between 2.1% to 14.1%. We also saw improvements in precision, with decreases in the standard deviation of error ranging between 3.1% to 36%.

4.2 Introduction

A well-established correlation exists between exposure to particulate matter in fine ($PM_{2.5}$) and coarse (PM_{10}) size ranges exposure and a wide array of adverse human health effects (Sharma et al. 2020). The composition of PM, as a complex, multisource pollutant, varies depending on factors such as season (Wang et al. 2020), geography (Wang et al. 2019), and time of day (de Jesus et al. 2019). Traffic is significant source of ambient $PM_{2.5}$ and is a dominant contributor to metals found in PM (Grieshop et al. 2006; Habre et al. 2020). Recent vehicle emissions regulations have decreased average tailpipe emissions, but do not target brake and tire wear, a significant source of metals in PM (CARB 2020). As a result, in $PM_{2.5}$ and PM_{10}, the relative share of non-exhaust vehicle emissions, namely brake and tire wear, has increased in recent years (Jeong et al. 2020b; Pant and Harrison 2013b).

Prior studies in individual roadways and the laboratory have identified specific metals and elements that can serve as tracers of varying specificity for identifying non-exhaust emissions sources, including barium, copper, and antimony for brake wear particles and zinc for tire wear particles (Amato et al. 2011; Grigoratos and Martini 2015; Milani et al. 2004). Knowledge on the spatial distribution of such elements, however, remains limited.

Land-use regression (LUR) is a well-established technique in the exposure assessment of intraurban air pollution, where regression models include land use, traffic, physical geography, and business density as predictors of measured air pollution concentrations (Briggs 2005; Hoek et al. 2008; Jerrett et al. 2005a). Resulting model coefficients can then be applied to interpolate a predictive surface for an entire study area.

LUR has been used at length in the study of traffic-related pollutants including unspeciated particulate matter and NO_2 (Henderson et al. 2007; Sahsuvaroglu et al. 2006), but relatively few studies have applied such methods to particulate matter constituents. Earliest studies applying LUR to PM constituents came out of Europe, where de Hoogh et al. (2013) analyzed spatial data from 400 sampling locations in 20 study areas across Europe, constructing LUR models for each site and each metal, finding that traffic-related variables were often associated with metals identified as brake and tire tracers, such as copper and zinc (de Hoogh et al. 2013). Zhang et al. (2015) collected $PM_{1.0}$ collected data at 54 sampling locations (25 in the summer, 29 in winter) in Calgary, Alberta and found that for barium, zinc, copper, and antimony, auto- and traffic-related variables contributed significantly (Zhang et al. 2015). Ito et al. (2016) sampled at 99 locations in New York City used speciated data to produce LUR models, identifying copper, iron, and titanium as significant markers of near-road pollution (Ito et al. 2016). Weichenthal et al. (2018) used LUR to model copper, iron, and a reactive oxygen species measure modeled with the two preceding metals using 67 sites monitored across two seasons (Weichenthal et al. 2018a). They found strong associations between road and traffic-related variables and all three of their dependent variables, while also finding strong correlations between copper, barium, and iron.

Later studies expanded beyond traditional LUR approaches by altering either the statistical methodology or including novel independent variables outside of typical LUR. Brokamp et al. (2017) collected $PM_{2.5}$ samples at 24 sites in Cincinnati, Ohio, and modeled metal concentrations using both linear land use regression models and machine learning-based random forest regression models. Compared to previous models, they found better model fit using with the latter approach. In brake and tire wear-related metals, such as copper and zinc, they found that traffic variables such as length of road and truck traffic were significant predictors of airborne concentrations (Brokamp et al. 2017). Tripathy et al. (2019) continued with a linear regression approach but integrated a gaussian atmospheric dispersion model into their predictions, also finding improved model fit compared to only using land-use variables.

A general limitation of measuring speciated $PM_{2.5}$ elements concerns the time and labor involved in the data collection and analysis stages. Gravimetric collection of air samples on filters is the current gold standard in metals, offering precise information on multiple metal constituents and isotopes. Due to limitations in time, human capital and monitoring equipment, studies that model over large areas encounter challenges when scheduling simultaneous sampling over dozens of locations. As a result, solely using gravimetric sampling might be infeasible in many studies.

A potential solution to this problem likes in the use of low-cost air sensor networks, such as PurpleAir (PurpleAir 2021b). A recent phenomenon following increased public concern over air quality issues, improvements in wireless connectivity technology, and decreased cost of components, low-cost air sensor networks have experienced increased attention from government regulatory agencies, such as the U.S. Environmental Protection Agency and the California Air Quality Management District (Johnson et al. 2020). The PurpleAir device consists of two Plantower laser particle counters, the PMS5003 and PMS1003, which uses light scattering of a laser to count suspended particulates in the air at various sizes and provides the mass concentration of $PM_{1.0}$, $PM_{2.5}$, and PM_{10} (PurpleAir 2021a). Devices also include wireless internet connectivity, and with the consent of the user, uploads time series data to the PurpleAir website, which provides said data free of charge. PurpleAir sensors are not reference grade, although they correlate well with federal reference monitors (Kosmopoulos et al. 2020). In addition, these low-cost sensors do not measure speciated $PM_{2.5}$ constituents. The network, however, is far denser compared to regulatory monitors and monitors deployed in academic studies, and information is collected and stored real-time.

In this study we leverage the PurpleAir network to enhance the prediction of metal species and indicators of oxidative stress. We implement a geostatistical modeling with the co-kriging method, which attempts to predict a target, gold standard variable (i.e., filter-based particle samples) with an auxiliary variable, which is more widely distributed (i.e., PurpleAir sensors). Widely used in mining and petroleum engineering, cokriging in air pollution studies has previously seen limited, but effective use (Li et al. 2012). With a focus on outcomes associated with brake and tire wear, this study is the first to model speciated $PM_{2.5}$ implementing information from a low-cost air sensor network.

4.3 Methods

4.3.1 Study overview

The following study presents an analysis of 50 speciated $PM_{2.5}$ samples collected across two sampling campaigns in the Los Angeles metropolitan area. In the following study, we implement (1) a machine learning-based deletion/substitution/addition algorithm, which selects variables based on predictive accuracy, and (2) co-kriging with external drift (CED) by introducing data from the PurpleAir low-cost sensor network to improve predictions potentially biased by a small sample of gold-standard equipment. In doing so, we aim to provide more precise, less biased estimates of speciated $PM_{2.5}$ metals and associated oxidative stress markers. In our study, our exposure surface – the largest of its kind in Los Angeles - specifically targets metals identified as tracers of brake and tire wear and present exposure surfaces to be used in health studies.

4.3.2 Site selection, sample collection and analysis

Previous literature have detailed the site selection and sampling methods of this study, itself a part of a larger effort to characterize the association between brake and tire wear particulates and adverse birth outcomes (Oroumiyeh et al. 2021). Our study targets the greater Los Angeles area, a geographically diverse and heavily populated region of 11.4 million people.

The study analyzes data on ambient fine and coarse particulate matter (PM) samples using Harvard cascade impactors (CI) and personal environmental monitors (PEM). PEMs and CIs were installed in a pump box connected to two pumps running at 1.8 liters per minute and 5 liters per minute, respectively. CIs collected particulates at both fine (aerodynamic diameter smaller than 2.5 μm), coarse (aerodynamic diameter between 2.5 and 10 μm), and super coarse (aerodynamic diameter greater than 10 μm) size fractions, while PEMs collected PM in the fine fraction. We deployed over two sampling campaigns, one in summer 2019 and the other in winter 2020. In each campaign, monitors simultaneously collected two-week samples at 46 different locations in the Greater Los Angeles region, with four sites serving as repeat locations. Mapped out in figure 4.1, our sites comprised of a mix of individual homes and government regulatory air monitoring locations. Our site selection methods, detailed in a prior report, involves a Multi-Criteria Decision Analysis, combining variables hypothesized a priori to be associated with brake and tire wear such as traffic intensity, road slope variance, and intersection density, as well as expert understanding of local geographies and PurpleAir densities to improve co-kriging (Jerrett 2021).

After data collection, we sent CI samples to be analyzed at the University of Wisconsin-Madison State Laboratory of Hygiene (WLSH). WLSH determined the mass concentrations and PM mass-normalized concentration of 55 chemical elements with Sector Field Inductively Coupled Plasma Mass Spectrometry (SF-ICP-MS), in line with previous studies (Herner et al. 2006b). After filtering out elements that were measured above the detection in more than 80% of samples and a signal-to-noise ratios above two, we obtained a dataset of 43 elements.

PEM samples were analyzed in-house for black carbon and oxidative stress activity. Black carbon was analyzed with the Magee Scientific Optical Transmissometer, while oxidative stress activity was measured with two outcomes: OH radical formation and dithiothreitol (DTT) loss. We measured OH formation by incubating samples in surrogate lung fluid and quantified OH radical formation by adding disodium terephthalate, following the procedure set forth by prior studies (Gonzalez et al. 2017). DTT loss was measured by incubating filters in DTT solution prepared in a phosphate buffer and stopping the reaction at fixed time points, following the procedure detailed by prior studies (Cho et al. 2005).

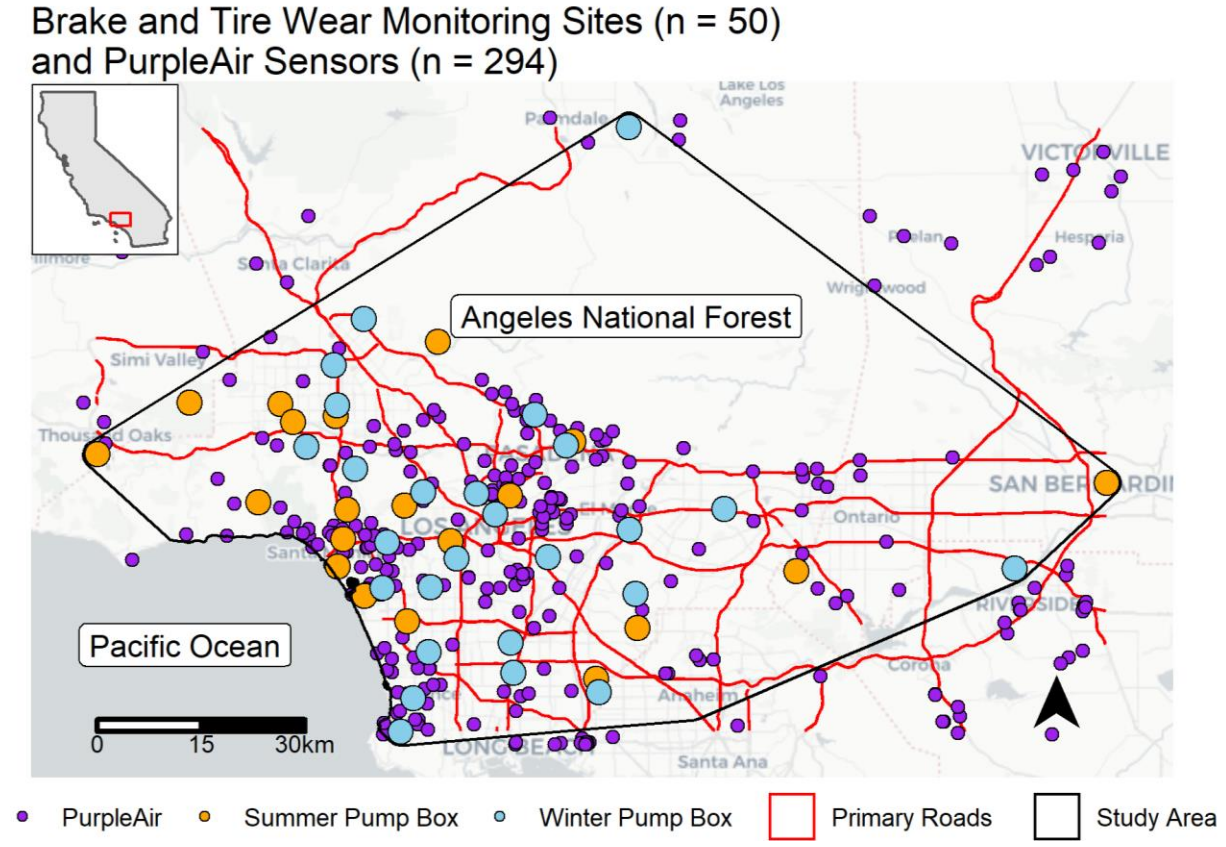


Figure 4.1: Locations of sampling locations (green and yellow) and PurpleAir sensors (purple). The study location within the state of California is illustrated on the top-left, with the study location outlined in red. The area to be interpolated, defined by the convex hull of the pump boxes is outlined in black. Primary roads in the study area's bounding box are shown in red.

4.3.3 Preliminary analysis

Prior to deployment, a preliminary list of metals hypothesized to serve as specific tracers for different sources (brake wear, tire wear, tailpipe emissions, and dust and soil) based on a prior literature search. To finalize a list of potential dependent outcomes, we calculated Spearman correlations between hypothesized tracers (Figure 4.2). Understanding correlations between different potential tracers, we narrowed down our dependent outcomes unspeciated $PM_{2.5}$ alongside six different variables in the fine PM fraction: barium (brake wear), zinc (tire wear), DTT loss, OH formation, and black carbon, and a reactive oxygen species (ROS) measurement based on a toxicokinetic model using copper and iron concentrations (Lakey et al. 2016). The toxicokinetic model uses concentrations of copper and iron, metals both associated with brake and tire wear, to calculate the rate of production of ROS in the epithelial lining fluid. Apart from gravimetric $PM_{2.5}$ and this measurement, where we modeled the rate of ROS production in the epithelial lining, we generated predictive surfaces for both volume-normalized and mass-normalized concentrations for each dependent variable.

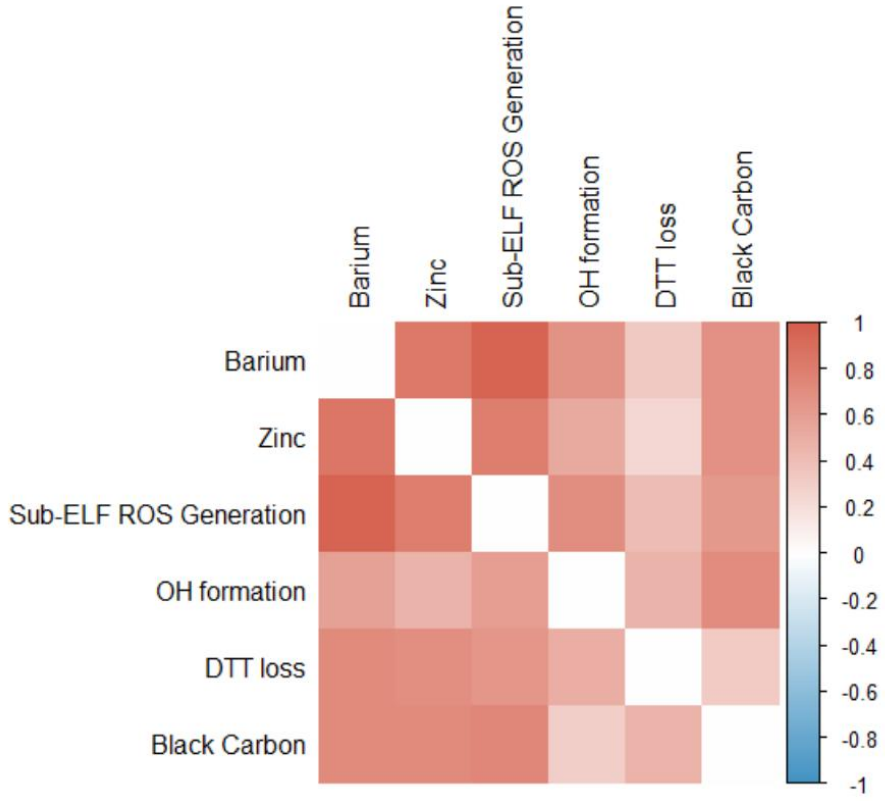


Figure 4.2: Spearman correlation matrix between volume-normalized study outcomes during the summer (upper) and winter (lower) sampling campaigns.

4.3.4 Land-use data

We downloaded land-use data from the following sources: (1) raster data on impervious surfaces, tree canopy, and green space data from the 2016 National Land Cover Database (NLCD) (MRLC 2016); (2) 2018 traffic data from the federal highway administration (Roff 2020); (3) road network and rail network information from the U.S. Census Bureau (Bureau); and (4) brake and tire wear-related businesses, such as auto repair shops, from the ESRI business analyst (ESRI 2018).

NLCD data came in three raster files, each at a resolution of 30 by 30 meters: impervious percent, tree canopy percent, and a dataset which classified each pixel into type of land use (MRLC 2016). Following a previous study, we created green space by grouping the following variables together: open water, ice/snow, developed/open space, developed/low intensity, deciduous forest, evergreen forest, mixed forest, dwarf scrub, shrub/scrub, grassland/herbaceous, sedge/herbaceous, pasture/hay, and cultivated crops (Akpinar et al. 2016). We classified the following as non-green space: medium intensity land cover, high intensity land cover, and barren land (rock/sand/clay) (table S4.1)

Information on all businesses in the study area were downloaded from the ESRI business analyst. We then selected businesses that would potentially emit heavy metals and those related to brake and tire wear such as auto shops and brake and tire manufacturers based on NAICS code. Subsections for NAICS codes selected are available in table S4.2.

4.3.5 Buffering and zonal statistics

We assigned land-use data to our sampling sites using nested circular buffers (Henderson et al. 2007b; Klot 2011). Around each monitoring each site, we constructed circle-shaped areal units of 100-, 200-, 300-, 400-, 500-, 750-, and 1000-meter radii. In each circle, we calculate the either the average value for land use for NLCD and traffic data or the density of independent variables for intersection and businesses data. The complete list of potential variables as well as well as their sources are found in table 4.1.

Table 4.1: List of dependent variables in LUR model

Variable Category	Covariates	Data Sources
Land Use	Tree Canopy (%) Impervious (%) Green space (%) Non-green space (%)	2016 NLCD (MRLC 2016)
Traffic and/or road	Intersection density (#/m^2) Slope variance (%) Road length (m) Annual average daily traffic (Veh/day) Annual average daily heavy-duty traffic (Veh/day)	US Census Bureau (Bureau) Federal Highway Administration (Roff 2020)
Distance	Distance to rail (m) Distance to coast (m) Distance to major road (m)	US Census Bureau
Commercial	Brake and tire-related businesses (count)	ESRI Business Analyst (ESRI)

4.3.6 Land-use regression model building and validation

In line with other studies, we approach our data assuming a linear relationship between independent and dependent variables (Zhang et al. 2015). We began by plotting histograms and univariate plots to investigate the distribution of dependent and independent variables. If a histogram revealed the distribution of a variable to be skewed, or if univariate plots showed a non-linear relationship between independent and dependent variable, we log transformed our variables. To reduce collinearity between variables of the same class but different buffer radii, for each class of variable, we selected the buffer distance with the highest univariate correlation with the outcome of interest. After generating variables for each point, we used a method previously used to predict $PM_{2.5}$ concentrations in the United States, selecting variables using a deletion-substitution-addition (DSA) algorithm (Beckerman et al. 2013). The DSA algorithm selected variables based on prediction accuracy with a L2 loss cross-validation function (Neugebauer and Bullard 2010). After the DSA algorithm chose the optimal set of predictors of sizes 1-10 variables, we conducted another round of K-fold cross-validation with five folds to select an optimal number of variables in the final model based on the mean square predictor error.

To adjust for seasonality, we included a dummy variable indicating the sampling season (0 = summer, 1 = winter). In our final prediction surfaces, we calculated the average of the summer and winter surfaces by dividing the season coefficient by two and adding the resultant term to the remainder of the model.

4.3.7 PurpleAir Data

Within the study area, we downloaded hourly-averaged PurpleAir data temporally aligned with both study periods from the PurpleAir website. We restricted our sample to outdoor sensors. Downloaded data contained date and time of measurement, particle matter mass concentration at 1, 2.5, and 10 microns, particle concentrations at various sizes, temperature, and relative humidity. We removed data from sensors that had 10% or greater rate of missing data and any observations that did not report temperature. We verified that our sensors were outdoor by creating time series temperature plots. If sensors reported a very low range of temperature, we assumed that they were mislabeled indoor sensors and subsequently deleted the sensor.

Among the remaining sensors, we conducted further QA/QC per Plantower's factory standards (AQMD 2015), which includes the following criteria:

- $PM_{2.5}$ mass concentrations above 500 $\mu g/m^3$
- For observations below 100 $\mu g/m^3$, remove rows if the difference between A and B exceeds 10 $\mu g/m^3$
- For observations greater than 100 $\mu g/m^3$, remove rows if the difference between A and B exceeds 10%

In addition, we also removed a limited number of rows with extreme temperature and relative humidity values ($0 \leq RH\% \leq 100$; $-200 \leq$ temperature ≤ 1000). Once cleaned, for each sensor, we obtained a final measurement by averaging measurements from the A and B sensors.

We confirmed the use of $PM_{2.5}$, the same size fraction as our target outcome, as the auxiliary variable of choice by calculating the correlation coefficient between different particle sizes and our outcomes of interest. While different particle size data tended to be highly correlated with one another, we found that $PM_{2.5}$ mass concentration, on average, had the highest levels of correlation with the target outcomes.

4.3.8 Co-kriging with external drift (CED)

To avoid singularity issues, for co-located points in both speciated metals and PurpleAir data, we shifted longitude and latitude coordinates by a random distance between 0 and 5 meters. Once

we confirmed the absence of co-located points, we generated empirical semi-variograms for the LUR residuals and PurpleAir $PM_{2.5}$ measurements. We also generated empirical cross co-variograms between PurpleAir $PM_{2.5}$ and LUR model residuals, implementing our LUR as an external drift within the co-kriging model (cokriging with external drift, or CED). After fitting initial theoretical variograms, we evaluated the performance by simultaneously conducting 1000 iterations of 10-fold cross-validation for both the original LUR model as well as the CED model. We then used two-sample t-tests to compare the mean and standard deviation of the 1000 subsequent mean square errors generated and made manual adjustments to the cross co-variogram parameters. We repeated this process with the goal of minimizing both the mean and standard deviation of the MSE as much as possible.

4.3.9 Generating exposure surfaces

After completing LUR model building, we created raster surfaces for predictor variables using the focal statistics function in ArcMap 10.8 (ESRI, Redlands, CA). In our convex hull, we used R 3.6.3 (R Foundation, Vienna, Austria) to generate a 30-meter by 30-meter grid of our study area, extract independent variable values at each point in the grid, and predict with both the LUR and CED models. We avoided predicting in the Angeles National Forest, a national forest with a small human population and whose conditions were not represented in our study sample.

4.4 Results and Discussion

4.4.1 Summary statistics

We performed analysis and model-building on six different potential outcomes in the $PM_{2.5}$ size fraction: barium, zinc, OH formation, DTT loss, black carbon, and 2-hour modeled ROS based on copper and iron measurements. Our analytical samples were of acceptable quality, based on the percentage of measurements above the limit of detection and a high signal-to-noise ratio. Table 4.2 summarizes both the volume-normalized concentration and mass-normalized (or equivalent) measurement for each model outcome, if available.

Our summary statistics are mostly in-line with findings from previous studies. Our mean gravimetric $PM_{2.5}$ concentration of 9.4 ug/m^3 was slightly lower, but overall comparable to 2019 Los Angeles average of 12.7 ug/m^3, likely because our sampling periods did not overlap with acute events affecting particulate matter concentrations such as wildfires. Zinc measurements accord with the lower end of measurements published by de Hoogh et al. (2013) and Brokamp et al. (2017) but are much lower than those reported by Tripathy et al. (2019), despite all studies taking place in either European or American metropolitan areas. A reason for this might be that zinc, while found in car tires, is also produced during steel production. Tripathy et al. collected samples from Pittsburgh, PA, which has a higher-than-average concentration of steel mills compared to the rest of the U.S., which would likely increase ambient zinc exposure (Brokamp et al. 2017; Tripathy et al. 2019).

For black carbon, our study measured lower concentrations compared to a study of air samples from 2016 in a similar study area (Jones et al. 2020). Such a discrepancy in black carbon levels is likely due differences in sampling strategy, where Jones et al. (2020) focused on clustering monitoring sites around major freeways in the Los Angeles area, while we sampled in both high- and low-traffic areas. Additionally, during each sampling campaign, we took a single 14-day sample which included sampling during the day and night, the latter featuring highly reduced levels of traffic. Jones et al. (2020) alternatively took short measurements during the daytime, albeit outside of rush hour traffic. Regarding DTT loss, our average loss rate of 0.41 ug/m^3 is comparable to findings published by Charrier et al. (2012), who analyzed $PM_{2.5}$ samples from the San Joaquin Valley in California, US. DTT was also studied by Yang et al. (2015), but due to differences in assays and units reported, our results are not easily comparable (Yang et al. 2015b). Our remaining pollutants, barium, sub-ELF 2-hour ROS generation, and OH formation in ambient $PM_{2.5}$ has not been studied extensively, making comparison with other studies infeasible.

We have also calculated univariate Spearman correlation coefficients between our six outcome variables, summarized in figure 4.2. Due to (1) atmospheric conditions altering $PM_{2.5}$ levels during different seasons and (2) a different spatial distribution of sampling locations between summer and winter campaigns, sampling period likely confounds our pollutant measurements. Consequently, we have divided our correlation matrix into summer (upper) and winter (lower) and added a dummy variable indicating sampling period (0 for summer, 1 for winter) to our land use regression. We find that between brake and tire-wear metal-based variables (Ba, Zn, and 2-hour modeled ROS), Spearman correlations are very high (ρ: 0.8-0.98). Such high correlations between brake and tire-wear associated metal-based variables are consistent with a study of speciated $PM_{1.0}$ by Zhang et al. (2015) (Zhang et al. 2015). We find that between non-metal-based variables, correlations are positive, but differ between summer and winter. DTT loss is more highly correlated with other variables in the winter sample (ρ: 0.51-0.73) compared to the summer (ρ: 0.24-0.47), which are consistent with Yang et al.'s results (Yang Aileen et al. 2015). In contrast, OH formation had slightly higher correlations with other variables during the summer (ρ: 0.53-0.71) compared to the winter (0.46-0.60).

Table 4.2: Summary statistics for dependent variables. ** concentration of ROS generated in the epithelial line, not concentration in the air.

Outcome	Volume-Normalized Mean (SD)	Mass-Normalized Mean (SD)
$PM_{2.5}$	9.4 (2.4) ug/m3	-
Barium	17 (9.9) ng/m3	1700 (750) ng/g
Zinc	9.3 (4.6) ng/m3	930 (320) ng/g
Sub-ELF ROS Generation**	100 (26) nmol/L	-
DTT loss	0.62 (0.12) uM/min/L	0.62 (0.12) uM/min/ug
OH formation	14 (3.5) nM/min/L	0.56 (0.14) nM/min/ug
Black Carbon	0.41 (0.17) ug/m3	4.6 (1.2) %

4.4.2 Variable selection and LUR model results

Figure S4.1 shows the expected cross validation errors for models of each side chosen by the DSA search algorithm resulting from a 5-fold out-of-sample cross validation process. For each pollutant, we then chose the model with the minimum error, regardless of model size or fit. Table 4.3 summarizes the results of our variable selection and the building of 11 different land use regression models for six pollutants, including predictors selected, regression coefficients, and adjusted R^2. The most common predictor, which appears in all LUR models except for $PM_{2.5}$, is the dummy period variable. All models featured at least one traffic-related variable, either distance to railway, heavy duty traffic, or both. Other common variables included the land use variables tree canopy cover percent, impervious surface percent, and number of businesses within the buffer distance. We find that the pollutants with the greatest model fit are barium mass concentration, black carbon mass concentration, and DTT mass concentration, with adjusted R^2 statistics of 0.62, 0.64, and 0.68, respectively. Conversely, the mass-normalized OH formation, $PM_{2.5}$, zinc normalized mass concentration models fit the worst, with adjusted R^2 statistics of 0.37, 0.41, and 0.43, respectively. Although we did not consider model fit in our variable selection process, our model R^2 values are similar to those obtained in prior literature (Yang et al. 2015b; Zhang et al. 2015). However, in contrast to prior published studies, the aggressive search algorithm employed focuses on predictive accuracy, rather than model fit. Our CV method has previously been shown to be optimized for predictive accuracy, which allows us to minimize the risk of overfitting in our modelling common among studies that choose models based on an R^2 statistic (Davies and van der Laan 2016).

Across our 12 LUR models, the direction of effect associated with each variable remained consistent. A positive period variable indicates that pollutant concentrations were greater in the winter compared to the summer. Negative distance to rail coefficients indicates that higher levels of pollutant closer to railways. Business count coefficients, regardless of buffer distance, are uniformly positive, matching the expectation that auto-repair and related businesses are positively correlated with brake and tire wear metals or that they may also be destinations with heavier traffic in the surrounding area. Similarly, the coefficient for heavy duty traffic, itself very highly correlated with passenger traffic, is uniformly positive across our 12 models. Interestingly, tree canopy percentage coefficients were not only commonly selected, but were also uniformly positive. Although tree canopy cover may serve as a marker for greenspace, our study almost exclusively monitored in urban or residential areas, where it is common to plant trees alongside sidewalks. Prior studies have found that the increased tree canopy cover does not necessarily correlate with increased carbon density, which measures the carbon storage per unit area, particularly in urban

settings (Chen et al. 2020). Therefore, it is possible in our study area, a large, highly developed urban setting, tree canopy serves as a proxy not for density of green space, but instead proximity to the road.

Table 4.3: Model summaries for mass concentration (left) and normalized mass concentration (right). * indicates that the dependent variable was log-transformed. ** concentration of ROS generated in the epithelial line, not concentration in the air.

	Volume-normalized concentration			Mass-normalized concentration		
Pollutant	Adj R^2	Model Parameters	Coeff.	Adj R^2	Model Parameters	Coeff.
$PM_{2.5}$	0.41	Log annual average daily traffic (500) Log distance to rail Log distance to major road Impervious % (1000) Tree canopy % (400)	0.32 -0.75 -0.38 0.05 0.08	-	-	-
Barium	0.60	Period Tree canopy % (400) Log distance to rail Impervious % (200) Business count (750)	9.146 0.763 -3.40 0.259 0.313	0.56	Period Tree canopy % (750) Log distance to rail Impervious % (200) Business count (750) Log annual average daily heavy-duty traffic (400)	725.79 47.42 -162.9 10.55 25.9 59.8
Zinc	0.47	Period Log distance to rail Log annual average daily traffic (300)	3.69 1.40 0.64	0.42*	Period Tree canopy % (750) Log distance to rail Impervious % (100)	6.95 0.026 -0.12 0.0056
Sub-ELF ROS Generation**	0.58	Period Tree canopy % (750) Log distance to rail Impervious % (200) Business count (750) Log annual average daily heavy-duty traffic (400)	81.77 1.57 -5.49 0.55 0.87 2.42	-	-	-
OH formation	0.56	Period Business count (750) Log annual average daily heavy-duty traffic (400) Tree canopy % (1000) Impervious % (200)	1.88 0.07 0.28 0.07 0.04	0.37	Period Business count (750) Log annual average daily heavy-duty traffic (400)	0.38 0.01 0.02
DTT loss	0.60	Period Business count (300) Log annual average daily heavy-duty traffic (400) Tree canopy % (1000) Impervious % (1000)	215.1 34.1 22.1 10.16 4.26	0.68	Period Business count (300) Log annual average daily heavy-duty traffic (400) Tree canopy % (1000)	17.49 2.02 1.04 0.34
Black Carbon	0.64	Period Impervious % (1000) Tree canopy % (400) Log distance to rail	0.15 0.01 0.01 -0.06	0.56	Period Impervious % (1000) Tree canopy % (400) Log distance to rail	1.25 0.02 0.06 -0.46

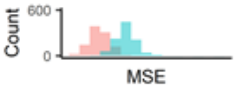
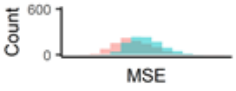
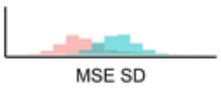
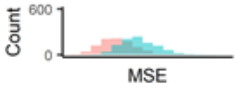
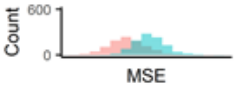
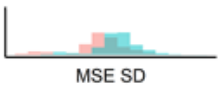
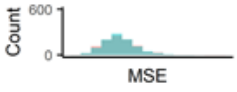
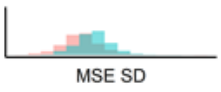
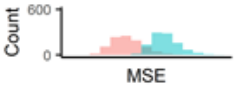
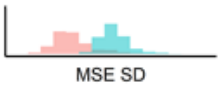
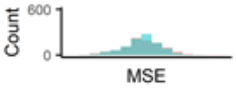
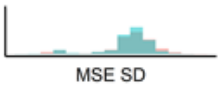
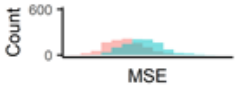
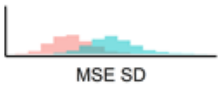
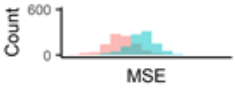
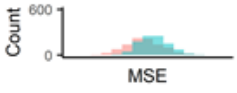
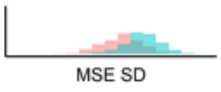
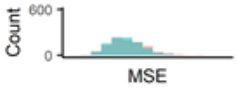
4.4.3 Comparing LUR and CED

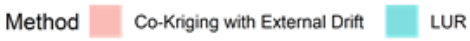
We found that based on the semi-variogram of LUR residuals, our LUR models explain most, if not all, of the spatial variability in our samples. Based on the cross covariograms between LUR residuals and PurpleAir $PM_{2.5}$, however, we find that there exists a degree of cross-covariance between the main outcomes and auxiliary variable (Figures S4.4 and S4.5). We find this reflected in the results of iterated 10-fold cross validation, where after optimizing the cross covariogram parameters, we see either no effect or a statistically significant improvement in prediction accuracy.

After conducting 1000 iterations of 10-fold cross validation comparing CED and LUR, we found statistically significant reductions in the mean and standard deviation of MSE for 8 out of the 11 the study's outcomes, as shown on table 4.4. There were statistically significant changes in neither mean or standard deviation of MSE for mass-normalized zinc concentrations and volumized DTT loss. While black carbon percentage had a small but statistically significant increase in mean MSE, standard deviation of MSE decreased, suggesting increased precision but slightly reduced accuracy. For other outcomes, we found mean reductions in MSE ranging between 2.1% and 14.1%, and reductions in MSE standard deviation between 3.1% and 36%. These results suggest that for most brake and tire wear-related outcomes, we see that incorporating $PM_{2.5}$ measurements via co-kriging leads to improved predictive accuracy and precision. While cokriging methods have been used in the past to improve PM_{10} estimates with wind speed curves, to our knowledge our study is the first to show that compared to conventional LUR methods, cokriging with a low-cost air sensor network improves predictions of $PM_{2.5}$ species and markers of oxidative stress (Giraldo et al. 2020).

Modeled surfaces for selecting dependent variables comparing CED and LUR are illustrated in Figure 4.3. In addition to generally lower MSE and MSE standard deviation, visual inspection reveals that CED appears to act as a smoother for the LUR model. This is particularly noticeable when mapping out the volumized concentration of zinc (Figure 4.3, middle), where the LUR model shows noticeable hotspots along rail lines in the study area that result from the distance to rail predictor. Incorporating information from the PurpleAir network noticeably smoothens out the surface, to a seemingly greater degree compared to other brake and tire wear-related metal outcomes that were highly correlated in the dataset. This might be because zinc, considered a tire wear tracer due to its presence in car tires, is a multisource pollutant whose emissions is not exclusive to tire wear (Jeong et al. 2020). This is in contrast with barium, copper, and iron – the latter two being the inputs that generate the sub-ELF 2-hour ROS generation estimates – whose emissions are much more specific to brake wear (Lough et al. 2005). Changes in estimated spatial distribution between zinc's LUR and CED models, as a result, may indicate the ability for the PurpleAir network to overcome a potential limitation present in this study, where information on potential point sources may be incomplete for outcomes less specific to brake and tire wear.

Table 4.4: Percent changes in mean and standard deviation of MSE after implementing CED compared to LUR for volume-normalized (left) and mass-normalized (right) concentrations. Under each estimate are histograms illustrating the 10-fold cross-validation performance differences between LUR (blue) and CED (pink) modelling approaches after 1000 iterations. A darker area indicates overlap between LUR and CED results. ** concentration of ROS generated in the epithelial line, not concentration in the air.

	Volume-Normalized		Mass-Normalized	
Pollutant	Mean MSE Change (95% CI)	Mean MSE SD Change (95% CI)	Mean MSE Change (95% CI)	Mean MSE SD Change (95% CI)
PM2.5	**-12.2% (-12.7%, -11.8%)** 	**-21.9% (-22.4%, -21.4%)** 	-	-
Barium	**-3.1% (-3.6%, -2.6%)** 	**-7.9% (-8.4%, -7.4%)** 	**-5.7% (-6.2%, -5.2%)** 	**-8.8% (-9.4%, -8.2%)** 
Zinc	**-6.9% (-7.3%, -6.4%)** 	**-4.9% (-5.5%, -4.3%)** 	**-0.1% (-0.6%, 0.4%)** 	**-3.5% (-4.0%, -3.0%)** 
Sub-ELF ROS Generation**	**-14.1% (-14.6%, -13.6%)** 	**-14.3% (-14.8%, -13.7%)** 	-	-
DTT Loss	**0.3% (-0.2%, 0.8%)** 	**0.2% (-0.4%, 0.8%)** 	**-6.1% (-6.6%, -5.5%)** 	**-13.8% (-14.5%, -13.2%)** 
OH Formation	**-6.3% (-6.7%, -5.9%)** 	**-15.8% (-16.2%, -15.3%)** 	**-2.1% (-2.4%, -1.8%)** 	**-3.1% (-3.4%, -2.9%)** 
Black Carbon	**-12.6% (-12.9%, -12.2%)** 	**-36.0% (-36.3%, -35.7%)** 	**0.7% (0.3%, 1.0%)** 	**-8.1% (-8.4%, -7.9%)** 



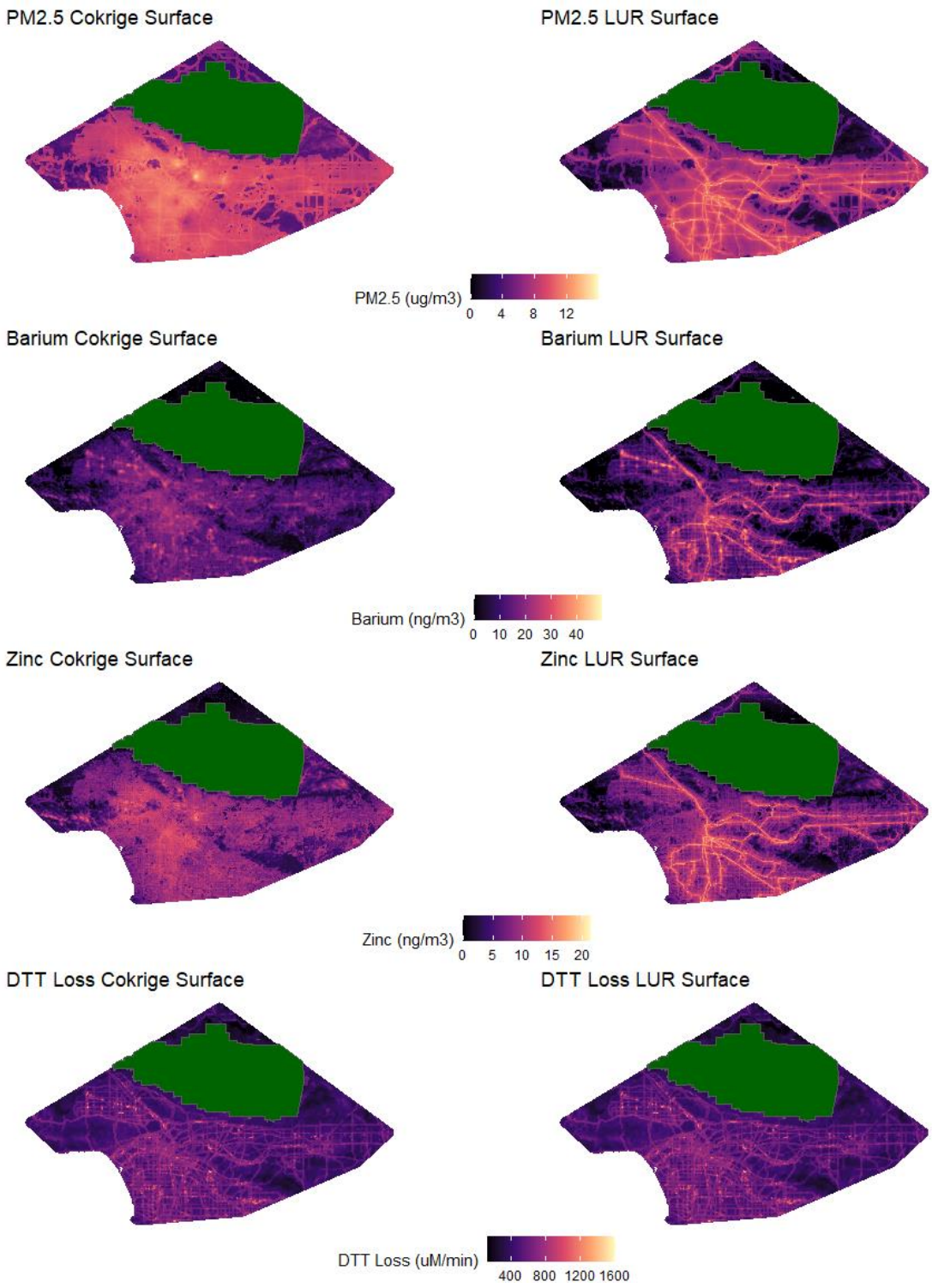


Figure 4.3: Exposure surfaces generated by CED (left) and LUR (models)) for (from top to bottom) volumized estimates of $PM_{2.5}$, barium concentration, zinc concentration, and DTT loss rates, which represent brake wear, tire wear, and oxidative stress potential, respectively. The Angeles National Forest, where we do not interpolate, is labeled and shown in green.

4.5 Conclusion

Here, we have presented an analysis of brake and tire wear across Southern California. Conducting a LUR model on data over two fieldwork campaigns, we have constructed several exposure surfaces over a large area for use in population health studies. We have demonstrated an increase in prediction accuracy by integrating a consumer-grade low-cost air sensor network into our modelling. Finding reduced prediction error and increased precision of predictions when adding in data from the low-cost sensor network, our study demonstrates that as sensor networks continue to expand, the ease of data collection and high spatial resolution associated with such networks has potential to improve exposure modeling and subsequent air pollution epidemiology. With PurpleAir sensors seemingly acting as a smoother near roadways, railways, and brake and tire wear-associated businesses when added on to an LUR model, our exposure model may provide greater levels of nuance to predicted pollutant levels across a study area. Lastly, our approach may be able to overcome time-related data collection challenges when modeling speciated $PM_{2.5}$. While gravimetric data is collected at single time points, low-cost sensor networks collect and report real-time information. Seasonal changes in the cross covariance between brake and tire wear metals and low-cost sensor network $PM_{2.5}$ concentrations are not yet known, but if it either (1) remains somewhat constant or (2) can be separately modeled, then the methods in our study may be used to create seasonalized estimates of brake and tire wear, measurements which would otherwise be highly resource intensive. Another strength in our study is that our samples reflect normal conditions, as our monitoring did not take place during major recent events affecting air quality, namely the California wildfires and the COVID-19 pandemic.

Our study does have a few limitations. Although the number of sensors deployed matches that of some prior studies, such as Zhang et al. (2015), our study area is much larger than that of prior studies, and the number of monitoring sites is less than ideal. We decided to prioritize covering a large study area over having many repeated monitoring sites, thus have very low temporal resolution, repeating measurements at only four sites. Although adding in the PurpleAir network improves cross-validation for most outcomes and improves the spatial resolution of our data, there may be concerns surrounding the network itself. While low-cost, the PurpleAir sensor remains a relatively luxury item, and density is consequently biased towards high-income households. Similarly, most of our gravimetric samplers were placed in subjects of an existing birth cohort, whose members largely came from relatively well-off families who own their homes. Lack of data support around disadvantaged communities would increase measurement error, which would mean that we would likely have higher measurement error in low-income communities that lacked the low-cost sensors. This would likely bias the health effects toward the null overall. It is difficult to know, however, whether this would influence the epidemiological findings or whether this might be the reason that some subgroups such as Black women tended to have smaller effects than other groups. Additionally, with the improvements in our predictive models by adding in PurpleAir data, our study suggests that adding in temporally aligned data may help address a fundamental issue among LUR studies.

Despite the above limitations, our model has identified the effects of various land use variables, such as traffic, business density, and impervious surfaces on brake and tire wear metals and ROS outcomes in a large study area. As the first study to incorporate low-cost sensor data to model speciated $PM_{2.5}$, as well as the largest-scale study covering the Los Angeles metropolitan area, our study results suggest that such sensor networks, particularly when further developed, have potential for use in modeling research. When dense enough, they have the potential to complement existing research methods, particularly when studying relatively difficult-to-measure outcomes, such as the constituents of $PM_{2.5}$ or oxidative stress markers. Future directions include

applying exposure estimates to health studies, including existing cohorts and passively collected state record birth outcome data.

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4.7 Appendix A.2 Supplementary information of chapter 4

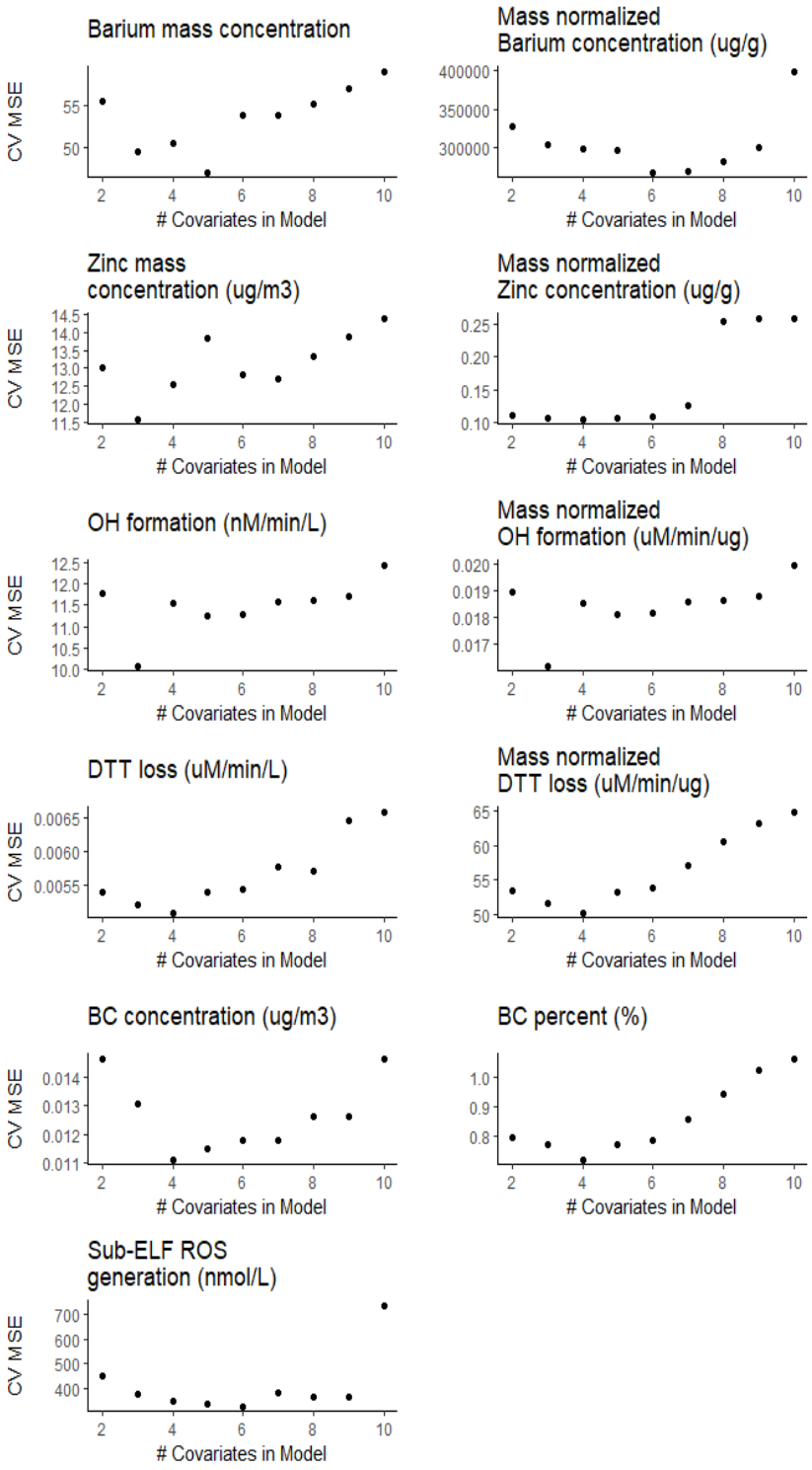


Figure S4.1: Cross validation (CV) error plots for each outcome as a function of model size.

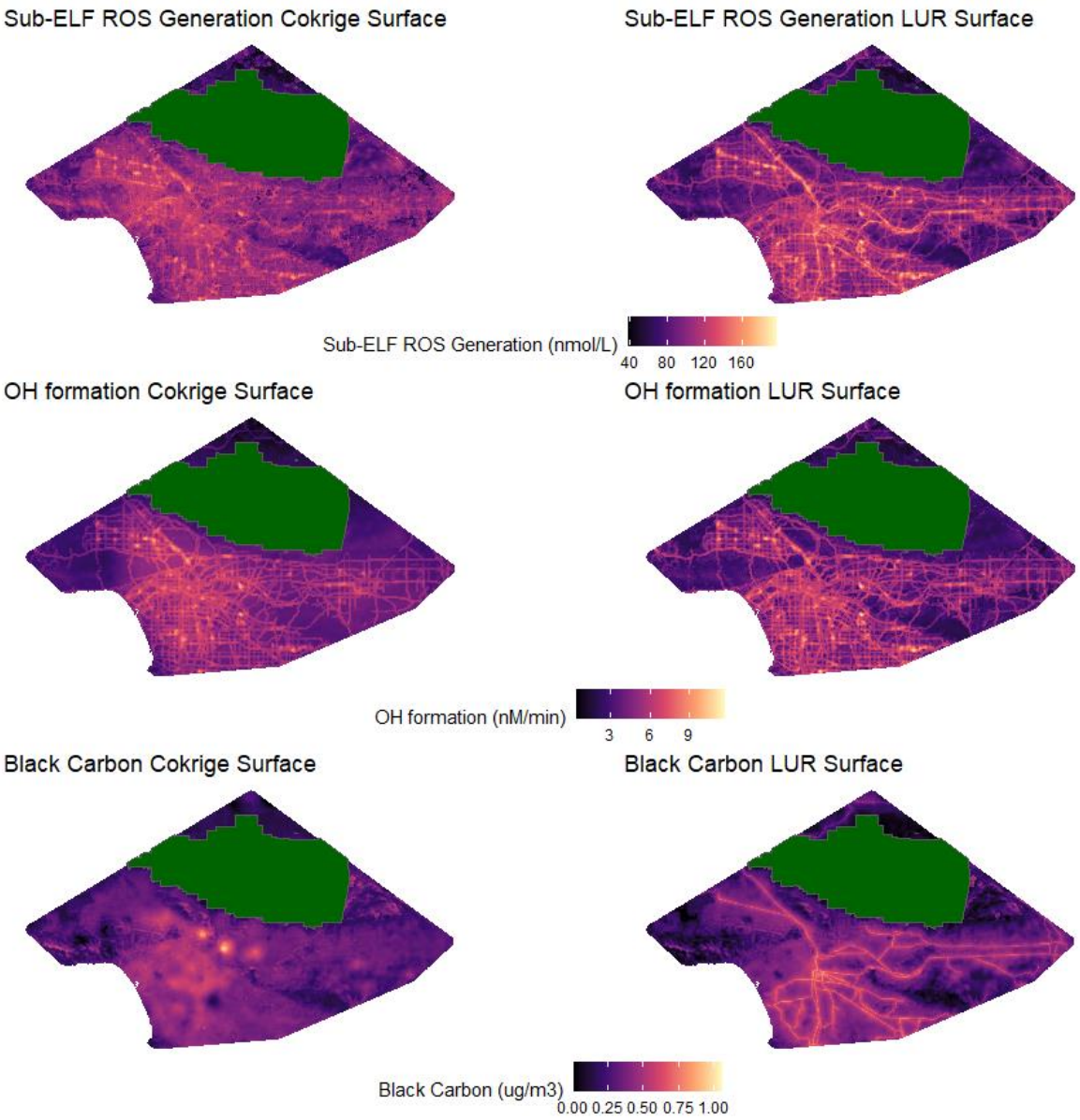


Figure S4.2: Exposure surfaces, volume-normalized concentrations for outcomes whose surfaces were not shown in main text

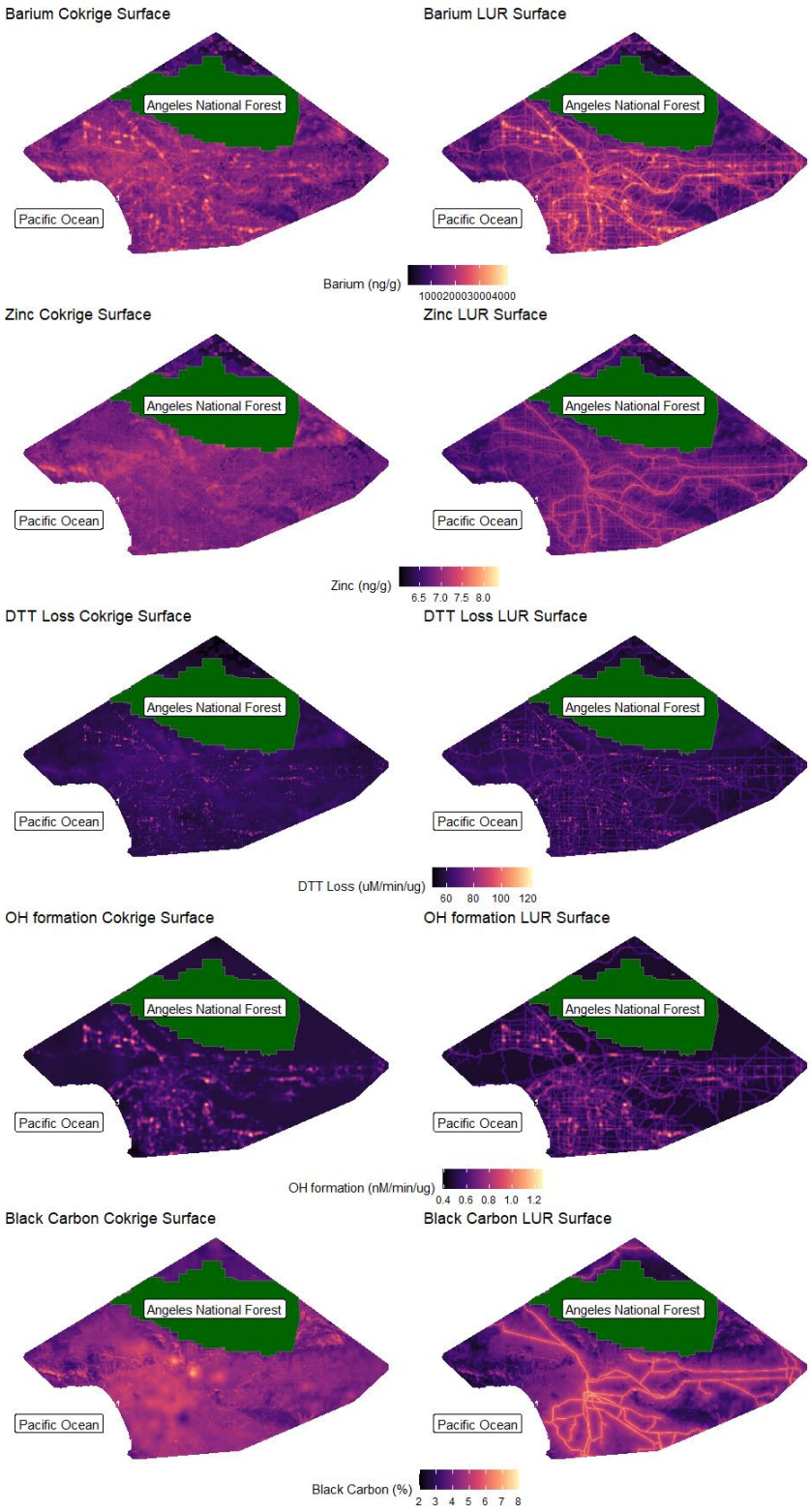
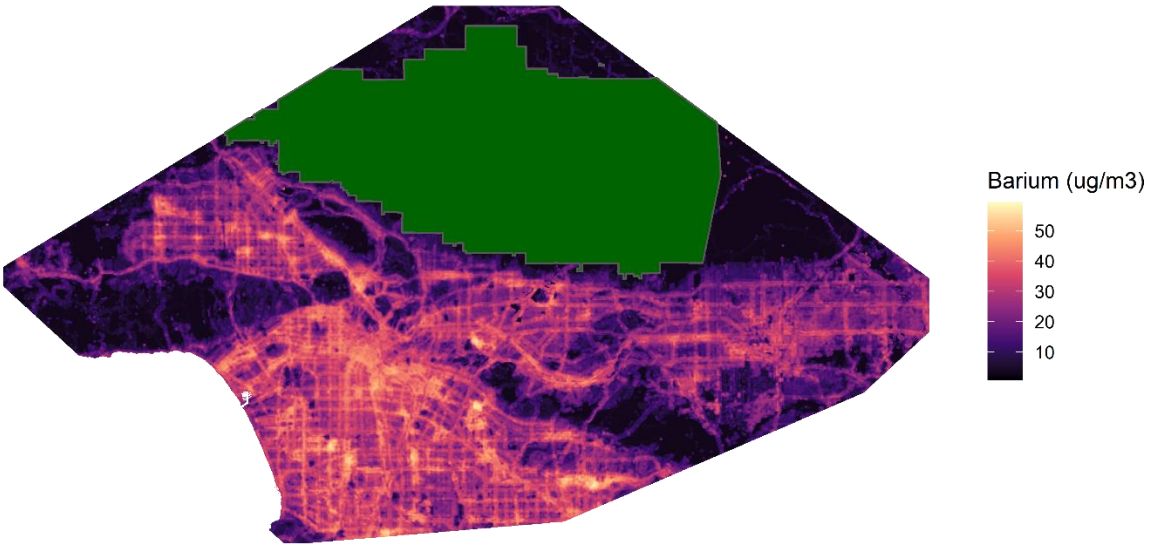


Figure S4.3: Exposure surfaces, mass-normalized concentrations

Barium LUR Surface, Coarse Fraction



Zinc LUR Surface, Coarse Fraction

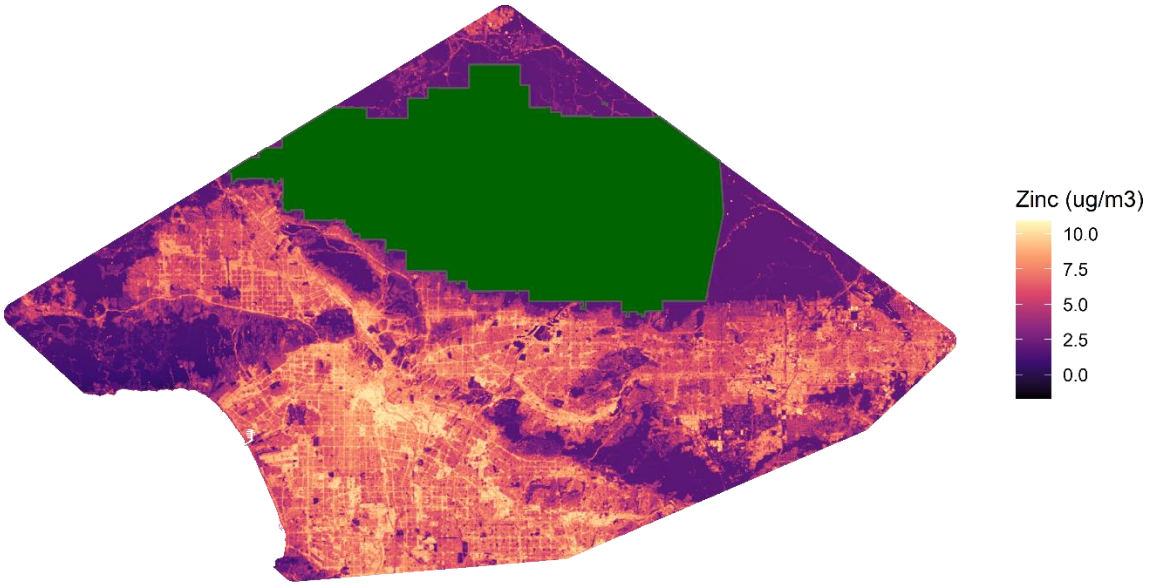


Figure S4.4: Exposure surfaces, volume-normalized concentrations of barium and zinc in the coarse fraction

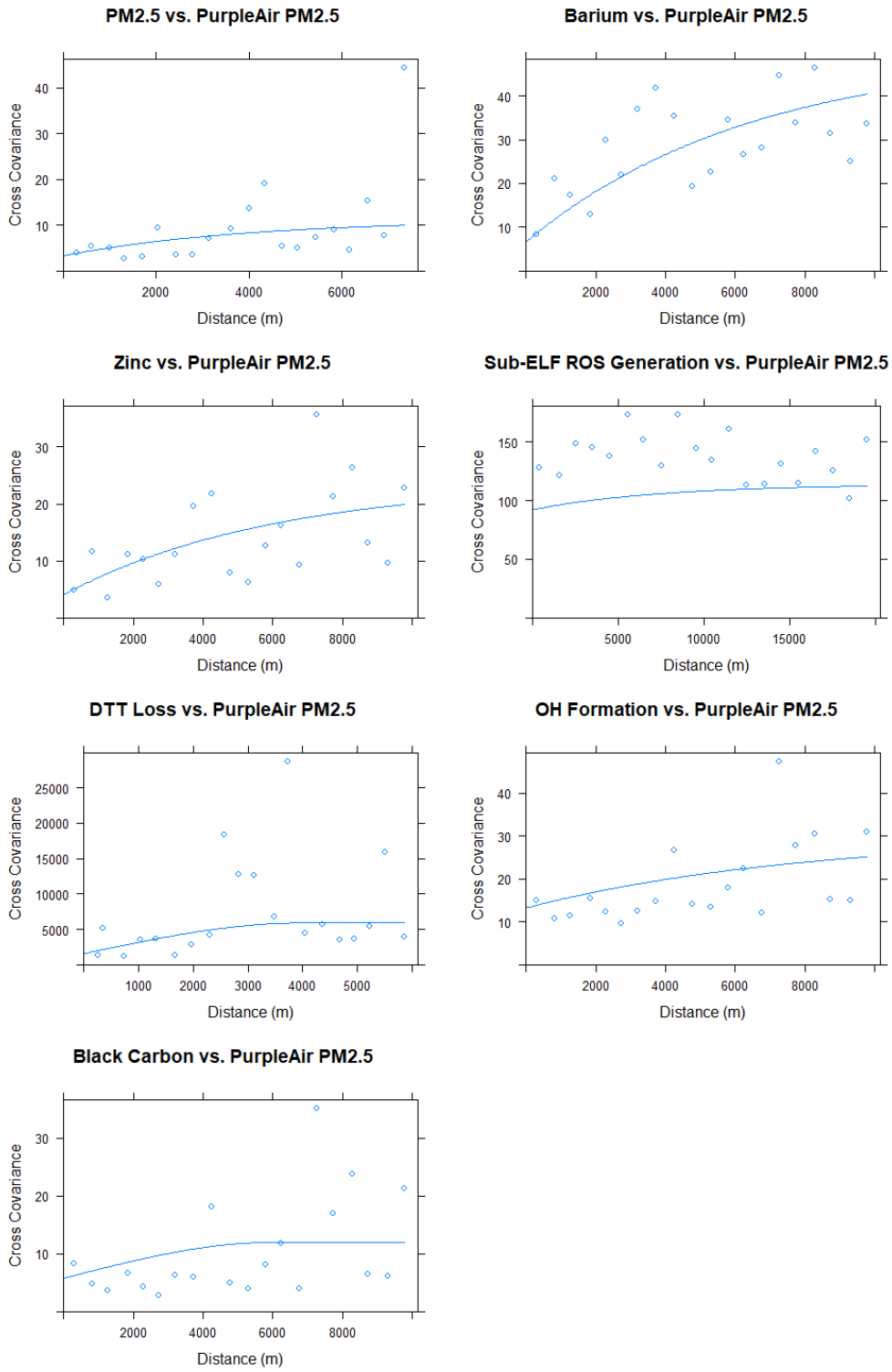


Figure S4.5: Cross covariograms between volume-normalized concentrations and PurpleAir. The theoretical (modeled) variogram is represented by the solid blue line, whereas the empirically derived variogram is represented by individual dots.

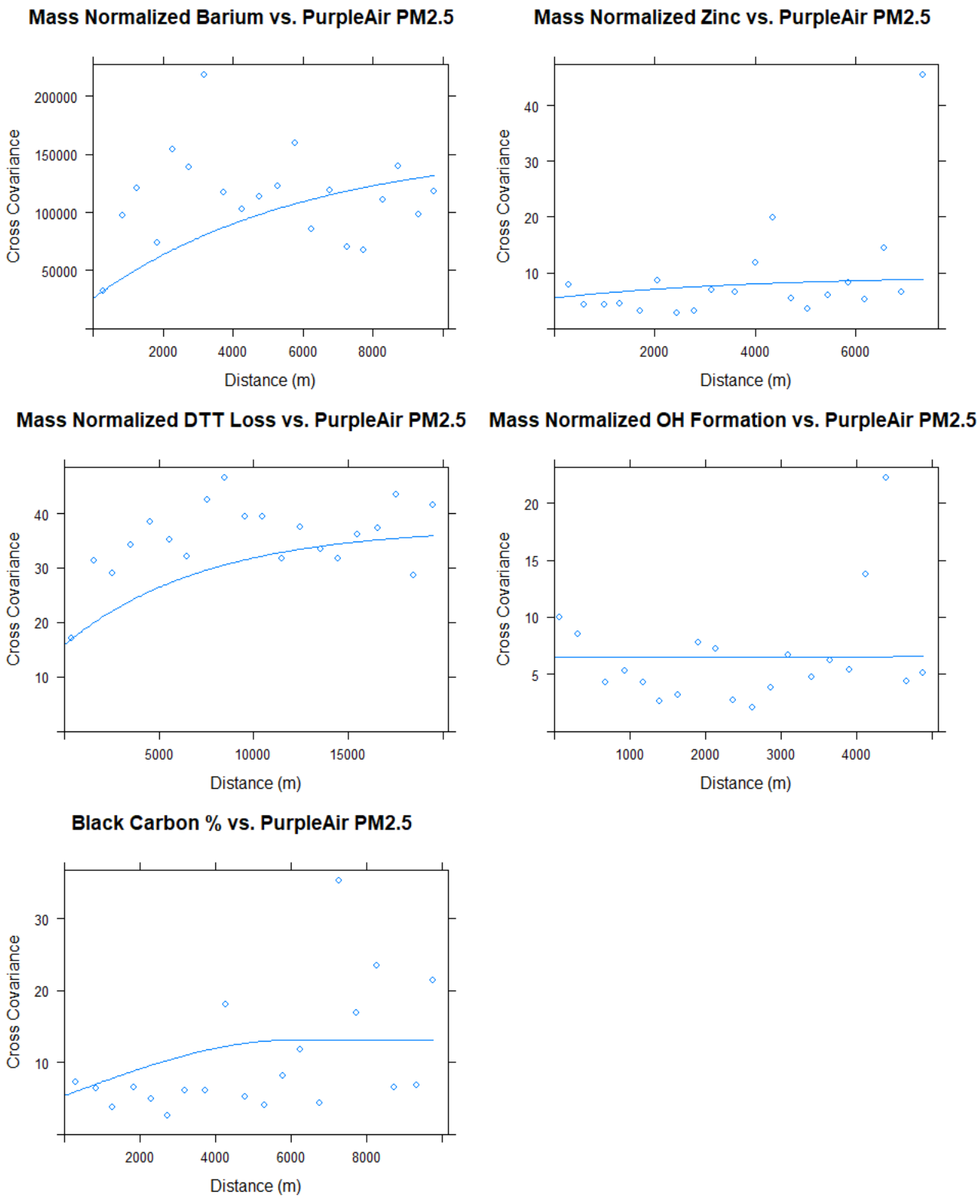


Figure S4.6: Cross covariograms between mass-normalized concentrations and PurpleAir. The theoretical (modeled) variogram is represented by the solid blue line, whereas the empirically derived variogram is represented by individual dots.

Table S4.1: NLCD classifications

	NLCD Classification	Spatial resolution	Year
Green Space	11, 12, 21, 22, 41, 42, 43, 51, 52, 71, 72, 73, 74, 81, 82	30 x 30 meters	2016
Non-green space	23, 24, 31	30 x 30 meters	2016

Table S4.2: First three digits of NAICS codes used to identify businesses potentially associated with tire-wear related metals

NAICS 3-digit code	Classification
212	Mining (except Oil and Gas)
213	Support Activities for Mining
221	Utilities
325	Chemical Manufacturing
326	Plastics and Rubber Products Manufacturing
331	Primary Metal Manufacturing
332	Fabricated Metal Product Manufacturing
333	Machinery Manufacturing
335	Electrical Equipment, Appliance, and Component Manufacturing
336	Transportation Equipment Manufacturing
339	Miscellaneous Manufacturing
423	Merchant Wholesalers, Durable Goods
811	Repair and Maintenance

5 The association of heavy metals and oxidative stress with adverse birth outcomes based on the Los Angeles birth records

5.1 Introduction

5.1.1 Preterm Birth and Term Low Birth Weight

Preterm birth (PTB) is defined by the World Health Organization (WHO) as childbirth occurring at less than 37 completed weeks or 259 days of gestation (WHO); it is estimated that approximately 10% of all deliveries in the United States are preterm (National Vital Statistics Reports Volume 68, Number 3, November 30, 2019, Births: Final Data for 2018).

Preterm birth is the leading cause of childhood mortality. Approximately 1 million babies die every year due to complications of preterm birth (Walani 2020). In addition, compared to children born at term, children born prematurely are at higher risk of showing cerebral palsy, sensory deficits, learning disabilities and respiratory illnesses (Barker 2004; Beck et al. 2010; Gopinath et al. 2010; Howson et al. 2013; Institute of Medicine (US) Committee on Understanding Premature Birth and Assuring Healthy Outcomes 2007; Lieshout et al. 2015; Mathewson et al. 2017; Osmond and Barker 2000; Wilcox 2001).

Many preterm births are also low birth weight (LBW), which is defined as birth weight less than 2500g), however, these infants may or may not be growth retarded depending on their gestational age. As a medically defined cut-off, LBW is used as a predictor of infants' risk of morbidities and mortality (Hughes et al. 2017). For example, recent studies reported that children born LBW are at higher risk of developing diabetes and cardiovascular disease later in life (Risnes et al. 2011). However, LBW represents a heterogeneous group of outcomes with different pathogenic mechanisms rendering the interpretation of findings challenging. Specifically, some infants are LBW due to PTB with their weight being adequate for gestational age while others are a result of intrauterine growth restriction (IUGR); birth weight less than expected for a given gestational age), and some might be a result of both mechanisms (Maisonet et al. 2004). Thus, term low birthweight (TLBW), which is defined as LBW among infants with a gestational age ≥ 37 weeks, is a preferred measure for inadequate birth weight. Since this outcome examines birthweight only among term infants, TLBW captures in utero growth retardation(Savitz et al. 2002). Additionally, TLBW has also been reported to be associated with childhood morbidities such as respiratory disease and learning disabilities (Caudri et al. 2007; Wang et al. 2008).

In summary, the consequences of adverse birth outcomes (PTB and TLBW) include fetal and neonatal mortality, morbidity, poor cognitive development, and an increased risk of chronic diseases later in life. Risk factors for PTB and TLBW have been studied, including medical conditions of the mother or fetus, genetic influences, behavioral and socioeconomic factors and environmental exposure, etc. (Bibby and Stewart 2004; Du et al. 2017; Goldenberg et al. 2008). A growing number of studies has assessed associations between exposure to $PM_{2.5}$ and traffic related air pollution during pregnancy and adverse birth outcomes, but only a few studies to date evaluated the effects of $PM_{2.5}$ components on TLBW or PTB (Yuan et al. 2019).

5.1.2 Previous Studies on $PM_{2.5}$, $PM2_{.5}$ compositions and PTB and TLBW

5.1.2.1 PM2.5 mass and sources measures and adverse birth outcomes

Plenty of studies on $PM_{2.5}$ and birth outcomes have been published in the past decades. Meta-analyses and pooled multicenter analyses suggested that particulate matter is associated with low birth weight and preterm birth, although there is some heterogeneity in results among studies

(Dadvand et al. 2014; Stieb et al. 2012). Considering the potential for heterogeneity of the $PM_{2.5}$ effect according to particles composition and sources in certain geographies, we focus our literature review on $PM_{2.5}$ and birth outcomes on studies conducted in California, especially in Los Angeles (LA) County, which is our study location. Overall, studies reported increase in PTB and/or TLBW risk of varying effect sizes and different exposure windows, the later suggesting that the effects of $PM_{2.5}$ may differ for different stages of fetal development.

Apart from the possibility that differences in effect estimates may be due to study design, differences in control of confounders, the inconsistency in results may be mainly attributable to different air pollution measurement/modeling, as well as pollution characteristics. PM has various core compositions and absorbs transition metals, polycyclic aromatic hydrocarbons (PAHs) and other compounds, which are able to generate oxidative stress and inflammation to various extents (Laurent et al. 2014). Therefore, differences in PM composition and sources have been hypothesized to modify the relationship between total PM mass and adverse birth outcomes such as LBW (Bell et al. 2007; Ebisu et al. 2014; Ebisu and Bell 2012).

5.1.2.2 $PM_{2.5}$ constituents and adverse birth outcomes

Exposures to $PM_{2.5}$ are traditionally assigned as particle mass concentrations as described above, while little is known about the specific constituents or sources of $PM_{2.5}$ that are most relevant to health (Weichenthal et al. 2018b). Only a few studies have evaluated the effects of $PM_{2.5}$ constituents on birthweight or preterm birth using data from California (Basu et al. 2014b, 2017; Laurent et al. 2014, 2016), and the results are not consistent. Specifically, Laurent et al. applied the University of California Davis/CIT_Primary (UCD_P) chemical transport model (Hu et al. 2014) to estimate primary ground-level PM element concentrations, reporting that TLBW risk increased with several chemical species in $PM_{2.5}$ exposure such as potassium, iron, chromium, and titanium, which were identified as tracer metals for brake and tire wear. (Schauer et al. 2006b) While for the study using the chemical transport model reported higher risks of PTB associated with per IQR increase of exposures to PM2.5 components such as nitrate and ammonium.(Laurent et al. 2016) A California wide study (Basu et al. 2014b) assessed several $PM_{2.5}$ constituents by calculating the distance between the geocoded US EPA monitor locations and the population weighted centroid of the 2000 US Census zip code tabulation area (ZCTA) associated with the maternal residential zip code reported in the birth record. All births with valid maternal zip codes located within 20km of a monitor were assigned exposures from that monitor, while all other births were excluded. The largest reductions in birthweight were found to be associated with exposure during pregnancy to vanadium, sulfur, sulfate, iron, titanium, manganese, ammonium, zinc and copper. There was also a study on PTB which assessed exposures based on maternal residential ZCTA by Basu et al., suggesting PM2.5 constituents including ammonium, nitrate and bromine were associated with higher risk of PTB, while reverse associations with PTB were found for some PM2.5 metal constituents such as sodium and vanadium. (Basu et al. 2017) A study by Ghosh et al. (Ghosh et al. 2012) in LA also reported higher odds of TLBW associated with exposure to vanadium (likely indication burning of shipping fuels) during pregnancy for women living within 5 miles of a California Air Resources Board (CARB) air toxics monitoring station at North Long Beach in LA County. Moreover, a number of air toxics such as benzene, xylenes, and toluene were found to have a greater impact on TLBW than $PM_{2.5}$ total mass, suggesting that the observed associations between $PM_{2.5}$ constituents and birth weight are not merely a result of their correlations with $PM_{2.5}$ mass.

Although PM-metals are mainly from non-combustion sources such as brake and tire wear in urban areas, few models are available to estimate spatial variations in PM-metal concentrations for use in population-based studies. To our knowledge, there are currently no results from land use regression (LUR) models of $PM_{2.5}$ metal constituents based on dense metal monitoring for

the LA area. In most studies, air pollution measures were based on closest monitor data within a relatively large radius (20 km) or even county level data. If certain $PM_{2.5}$ constituents are driving the health outcome associations and also are more spatially heterogeneous due to their sources than total $PM_{2.5}$ mass, it is possible that associations with those constituents may be underestimated due to higher misclassification of exposure when using a large radius or county-level data. Thus, it might be important to predict $PM_{2.5}$ metal concentrations from specific sources using spatial models when the goal is to estimate the effects of $PM_{2.5}$ -metals on TLBW and PTB so that constituents and/or sources with particularly strong impacts on adverse birth outcomes can be identified.

The observed associations might be misleading if constituents are not responsible for the adverse outcomes but are strongly correlated with other pollutants that increase risk, for instance, because the pollutants share common sources and exhibit similar dispersion patterns. Considering the challenge of distinguishing individual effects of specific metal constituents from each other statistically when they are highly correlated, it has been suggested to instead generate a read-out of the shared mechanism of action on the health outcome such as oxidative stress caused by the mixture. Thus, here we will examine the impact of estimated concentrations of metals barium and zinc as markers of brake wear and tire wear, respectively, and concentrations of reactive oxygen species that generate oxidative stress as measured and modeled by established methods in previous chapters. Besides $PM_{2.5}$, since the lesser extent coarse particles ($PM_{2.5\text{-}10}$) are also associated with adverse birth outcomes, we will also include $PM_{2.5\text{-}10}$ -barium and $PM_{2.5\text{-}10}$ -zinc as exposures of interest in our study. Specifically, we will evaluate the associations between adverse birth outcomes (PTB and TLBW) and two speciated $PM_{2.5\text{-}10}$ constituents (barium, zinc) as well as a total of six different $PM_{2.5}$ related exposures including three speciated $PM_{2.5}$ constituents (barium, zinc, black carbon), two measures of oxidative stress activity (OH radical formation and dithiothreitol (DTT) loss), and a 2-hour modeled reactive oxygen species (ROS) concentrations according to a kinetic multilayer model of surface and bulk chemistry in the lung epithelial lining fluid (KM-SUB-ELF) using the concentrations of the $PM_{2.5}$ constituents iron and copper (Lakey et al. 2016b).

5.2 Methods

5.2.1 Study Population

Birth certificate records for all births occurring from January 1st, 2017 through December 31th 2019 in Los Angeles County, California (N=334,059) were obtained from the California Department of Public Health. We excluded births with missing data for maternal residential address (N=1,321) or gestational length based on the date of the last reported menses (N=7,312), with extreme or implausible gestational ages (<20 weeks or >45 weeks) (N=8,425) or birthweights (<500 g or >6800 g) (N=362), with home addresses outside of California (N=47) and some that failed geocoding (N=72) (see below), as well as multiple births (N=10,524).

The remaining births included 25,674 preterm births, defined as having a gestational age less than 37 weeks, and 283,005 term normal birthweight infants (2500–6800 g) born between 37 and 45 weeks of gestation as the reference. To examine term low birthweight, we identified 6,727 term births with a birthweight of less than 2500 g, indicating intrauterine growth restriction, and used the same reference group of term normal birthweight infants for analyses

We further restricted our study population to those individuals residing at birth within the fields covered by the exposure surfaces we generated for LA county; thus, we included 23,131 preterm births, 6,102 term low birthweight, and 256,381 term normal birthweight infants in our models below.

5.2.2 Air Pollution Estimation

5.2.2.1 $PM_{2.5}$ mass and $PM_{2.5}$ constituents Assessments

We geocoded maternal addresses of residence recorded on birth certificates using the Countywide Address Management System (CAMS) locator (https://cams-lacounty.hub.arcgis.com/pages/cams-geocoder), which is created from the CAMS Datasets (address point and road segments) and maintains geocoder utilizing Esri ArcGIS Address Locator, but with a custom Address Locator Style that developed to accommodate complex addresses within LA County.

For each of the birthyear, more than 85% of the addresses were successfully geocoded to a point address and 10% - 15% of the addresses were geocoded to a street address. For the addresses that failed to be geocoded via CAMS locator (N=6,433), we used the Google sheet Add-on Tool 'Geocode by Awesome Table'. By overlaying the co-kriging surfaces described in previous chapters onto the geocoded addresses, we estimated each mother's exposures to $PM_{2.5}$ mass, speciated $PM_{2.5}$ constituents (barium, zinc, black carbon), measures of oxidative stress (OH formation, DTT loss) by $PM_{2.5}$ and 2-hour modeled ROS based on $PM_{2.5}$ copper and $PM_{2.5}$ iron measurements. We also predicted the exposures of barium and zinc as normalized concentrations according to coarse ($PM_{2.5\text{-}10}$) mass per volume (ng/m^3). $PM_{2.5}$ mass were predicted as ug/m^3, and all of the speciated $PM_{2.5}$ exposures except for the ROS were predicted as both mass concentrations (ng/g for barium and zinc, ug/g for black carbon, uM/min/ug for OH formation and DTT loss) and normalized concentrations according to $PM_{2.5}$ mass per volume (ng/m^3 for barium and zinc, ug/m^3 for black carbon, $uM/min/m^3$ for OH formation and DTT loss). The modeled $PM_{2.5}$ ROS were estimated as continuous measures in nmol/L using the toxicokinetic model based on the copper and iron measurements. These surfaces for the two speciated $PM_{2.5\text{-}10}$ exposures, six speciated $PM_{2.5}$ exposures as well as the $PM_{2.5}$ mass exposure were not different by year; and the surfaces covered the whole pregnancy period for the births in our study population (i.e., 2016-2019) thus, we were estimating the whole pregnancy exposures which were purely spatial contrasts. Besides that, we also generated summer and winter specific estimates for $PM_{2.5}$ barium, $PM_{2.5}$ zinc and for $PM_{2.5}$ ROS by relying on seasonal co-kriging surfaces when estimating exposures at the residence based on address geocodes.

5.2.2.2 Other Air Pollutant Assessments

We also re-assessed the role of traffic-related exposures represented by NO_2 concentrations generated from land use regression models as a surrogate for combustion emissions as done previously for earlier periods in LA (Ritz 2009). The modeling methods were described elsewhere (Su et al. 2009, 2020). Briefly, 201 locations were selected using a location-allocation algorithm that considered variability in traffic pollution and the spatial distribution of the LA population. In two seasons during September 2006 and February 2007, samplers from Ogawa & Company USA, Inc. were placed at pre-selected sampling sites. Furthermore, in 2012 October and 2013 March, Ogawa monitors were deployed again for NO_2 measurements at a total of 72 of these sites in LA County. Routine government monitoring of NO_2 was also included to add a temporal dimension to the exposures and to increase the data support for the LUR models. The LUR estimates for each address were predicted based on traffic volumes, truck routes and road networks as well as land use data.

We applied the year specific LUR surfaces for 2016-2019 to generate annual NO_2 estimates for each pregnant woman in this study. The correlations among the annual NO_2 estimates from 2016 to 2019 were highly correlated with Pearson $r > 0.99$, i.e., little difference was detected among different years, so we used the annual NO_2 in 2019, which was predicted as part per billion (ppb) to estimate the women's exposure to NO_2 during pregnancy.

5.2.3 Covariates

Potential confounders were selected a priori according to the literature and availability of the data on birth certificates, including gender of child (female, male), maternal age (< 20, 20-24, 25-29, 30-35, > 35), maternal race/ethnicity (non-Hispanic White, Hispanic/Latinx of any race, African-American/Black, Asian/Pacific Islander, Multi-races, Others) and maternal educational attainment (less than 8^{th} grade, 9^{th} to 12^{th} grade, high school graduate/high school diploma, some degree less than college, college or more than college), maternal birthplace (US-born or Foreign-born), parity (1, 2, ≥3), maternal smoking from 3 months before pregnancy until delivery (Yes vs. No) and payment source for prenatal care (no prenatal care, private, MediCal/government/self-pay, other), the later as a proxy for family income.

5.2.4 Statistical Analysis

Correlations of average speciated $PM_{2.5-10}$ exposures (barium and zinc), $PM_{2.5}$ mass, speciated $PM_{2.5}$ exposures (metals, black carbon and oxidative stress markers) and LUR-NO_2 during pregnancy were checked using a correlation matrix and Pearson correlation coefficients. To make the effect estimates comparable among the variable pollutant estimates, we scaled all exposure concentrations according to their interquartile range (IQR) among non-cases for each of the exposures ($PM_{2.5-10}$ metals, $PM_{2.5}$ mass, three $PM_{2.5}$ constituents, three oxidative stress markers for $PM_{2.5}$, LUR-NO_2). In addition, we determined quartiles of the $PM_{2.5}$ exposures ($PM_{2.5}$ mass, three $PM_{2.5}$ constituents, three oxidative stress markers for $PM_{2.5}$) distribution among non-cases to categorize exposures into four groups (< 25^{th} percentiles, 25^{th}-50^{th} percentile, 50^{th}-75^{th} percentile, > 75^{th} percentile).

We conducted unconditional logistic regression to estimate increases in odds of PTB and TLBW per IQR increase in pregnancy-period exposures to $PM_{2.5-10}$-barium, $PM_{2.5-10}$-zinc, $PM_{2.5}$ mass as well as each of the speciated $PM_{2.5}$ constituents/oxidative stress measures/modeled ROS, respectively. Also, we applied logistic regression analyses for exposure quartiles using the < 25^{th} percentile as the reference to assess patterns in the log odds for the $PM_{2.5}$ mass and speciated $PM_{2.5}$ exposures and adverse birth outcomes without assuming linearity.

We adjusted for potential confounders listed above. Furthermore, we also assessed combustion related traffic emissions by including into our models the estimated exposures for NO_2 from our LUR model for LA County built based on the 2006-2007 and 2012-2013 Ogawa monitoring we conducted. Additionally, we conducted stratified analyses by maternal race/ethnicity after scaling exposures according to their IQRs for non-cases in the study population. Sensitivity analyses were conducted for the stratified analyses by also scaling the exposures to the maternal race/ethnicity-specific non-case IQRs. Analyses were performed using R software, version 9.1.3 and SAS 9. 4 (SAS Institute Inc., Cary, NC, USA.).

5.3 Results

5.3.1 Characteristics of Study Population

Compared to infants born term with normal birth weight, preterm births or term low birth weight infants were more likely to have mothers of younger age (less than 20), with a lower education level (high school graduate or lower), lacking prenatal care, or being covered by Medi-Cal or other government programs for prenatal care payment. Also, term low birth weight infants were more likely to be female and a first-born child and to have a mother of Black or Asian origin compared with term normal birth weight infants; While infants born preterm were more likely to be a third or later-born child, or born to mothers with Hispanic or Black race/ethnicity (Table 5.1).

Table 5.1 Characteristics of the cases and non-cases in Los Angeles County, California, 2017-2019.

Characteristics	Preterm Birth		Term Low Birth Weight		Term Normal Birth Weight	
	N= 23,131	%	N= 6,102	%	N= 256,381	%
Infant sex						
Male	12,818	55.4	2,585	42.4	131,290	51.2
Female	10,313	44.6	3,517	57.6	125,091	48.8
Year of birth						
2017	7,886	34.1	2,132	34.9	91,068	35.5
2018	7,194	31.1	2,014	33.0	84,177	32.8
2019	8,051	34.8	1,956	32.1	81,136	31.7
Maternal age (years)						
Less than 20	954	4.1	299	4.9	9,395	3.7
20 - 24	3,304	14.3	985	16.1	37,971	14.8
25 - 29	5,228	22.6	1442	23.6	63,736	24.9
30 - 35	6,604	28.6	1763	28.9	78,352	30.6
Greater than 35	7,041	30.4	1613	26.4	66,927	26.1
Maternal education						
Less than 8th grade	1,261	5.6	264	4.4	10,027	4.0
9th to 12th grade	3,189	14.1	697	11.7	27,086	10.8
High school graduate/ high school diploma	6,104	27.1	1,601	26.8	60,794	24.2
Degree less than college	6,006	26.6	1528	25.6	64,257	25.6
College or more than college	5,993	26.6	1874	31.4	88,767	35.4
Missing	578		138		5,450	
Maternal race/ethnicity						
White, non-Hispanic	7,159	31.4	1,805	30.0	91,757	36.3
Hispanic of any race	9,618	42.2	2,141	35.6	92,667	36.6
Black	2007	8.8	664	11.0	15,924	6.3
Asian or Pacific islander	3091	13.5	1149	19.1	42,833	16.9
Multi-race, non-Hispanic status	321	1.4	108	1.8	4,073	1.6
Others	625	2.7	150	2.5	5,686	2.3
Not stated or unknown	310		85		3,441	
Parity						
1	9005	39.0	3164	51.9	106389	41.5
2	6560	28.4	1562	25.6	83,683	32.7
3 or more	7,548	32.7	1,371	22.5	66,210	25.8
Missing	18		5		99	

Payment type of prenatal care						
No Prenatal Care	362	1.6	55	0.9	810	0.3
MediCal/Govt/self-pay	12938	56.1	3319	54.5	132,747	51.9
Private	9344	40.5	2591	42.6	117304	45.8
Other	419	1.8	125	2.1	5,146	2.0
Missing	68		12		374	
Maternal birthplace						
U.S. Born	13,629	58.9	3,692	60.5	149,462	58.3
Foreign Born	9,496	41.1	2,409	39.5	106,894	41.7
Missing	6		1		25	
Smoking from 3 months before pregnancy till delivery						
Yes	231	1.0	70	1.2	1,535	0.6
Missing	525		130		3,869	

5.3.2 Whole Pregnancy Exposures

Overall, the exposure estimates for $PM_{2.5}$ mass were moderately to highly correlated with the speciated $PM_{2.5}$ exposures (Pearson r=0.33–0.87) (Table 5.2). Specifically, the Pearson correlation coefficients for the brake and tire wear metals ($PM_{2.5}$ barium and $PM_{2.5}$ zinc) with $PM_{2.5}$ mass were 0.66 and 0.78, respectively; and the highest correlation coefficient of 0.87 was calculated for black carbon. In contrast, the correlation of oxidative stress markers and $PM_{2.5}$ mass were relatively lower than for the $PM_{2.5}$ constituents except for OH formation with a correlation coefficient of 0.67. Furthermore, moderate to high correlations (Pearson r=0.33–0.84) were observed among speciated $PM_{2.5}$ exposures. As expected for markers of brake and tire wear, $PM_{2.5}$ barium and $PM_{2.5}$ zinc were found to be highly correlated with each other, and they also correlated well with oxidative stress markers and black carbon, except for zinc and DTT loss. The correlations of $PM_{2.5}$ black carbon and $PM_{2.5}$ oxidative stress markers were relatively low except for OH formation with Pearson r = 0.61 with black carbon. Correlations among the $PM_{2.5}$ oxidative stress markers were also high ranging from 0.61 to 0.78. Meanwhile, we also observed a moderate to high correlation between $PM_{2.5-10}$ metals and the $PM_{2.5}$ mass and speciated $PM_{2.5}$ exposures. Correlations were relatively high for $PM_{2.5-10}$ barium and $PM_{2.5}$ barium, $PM_{2.5}$ OH or $PM_{2.5}$ DTT with Pearson r ranging from 0.75 to 0.80. For $PM_{2.5-10}$ zinc correlations were moderate with $PM_{2.5}$ barium and $PM_{2.5}$ zinc (Pearson r = 0.68 and 0.66, respectively).

We present the summary statistics including mean, standard deviation (SD) and quartiles for all exposures included in our study in Table 5.3. For most of the exposures, the means among cases (PTB or TLBW) were higher than among non-cases. In addition, modeled exposure levels based on winter monitoring were higher than those based for summer.

Table 5.2 Pearson's correlation coefficients for air pollution exposures among births in Los Angeles County, California, 2017-2019.

PTB and Term Normal Birth Weight															
Air pollutants	Unit	No. of Births	Mean	SD	Pearson Correlation Coefficients									
					$PM_{2.5-10}$ barium	$PM_{2.5-10}$ zinc	$PM_{2.5}$ mass	$PM_{2.5}$ barium	$PM_{2.5}$ zinc	$PM_{2.5}$ ROS	$PM_{2.5}$ OH	$PM_{2.5}$ DTT	$PM_{2.5}$ black carbon	NO_2
$PM_{2.5-10}$ barium	ng/m³	277,456	30.9	7.3	1.00									
$PM_{2.5-10}$ zinc	ng/m³	277,499	7.1	1.3	0.83	1.00								
$PM_{2.5}$ mass	ug/m³	277,538	10.0	1.1	0.45	0.44	1.00							
$PM_{2.5}$ barium	ng/m³	279,512	17.1	3.9	0.75	0.68	0.66	1.00						
$PM_{2.5}$ zinc	ng/m³	279,512	9.7	1.7	0.49	0.66	0.78	0.79	1.00					
$PM_{2.5}$ ROS	nmol/L	279,512	113.5	12.1	0.65	0.58	0.33	0.84	0.62	1.00				
$PM_{2.5}$ OH formation	uM/min/m³	279,512	5.6	1.2	0.77	0.52	0.67	0.81	0.62	0.64	1.00			
$PM_{2.5}$ DTT loss	uM/min/m³	279,512	609.5	116.4	0.80	0.52	0.37	0.64	0.39	0.61	0.78	1.00		
$PM_{2.5}$ black carbon	ug/m³	279,512	0.4	0.1	0.43	0.45	0.87	0.69	0.75	0.35	0.61	0.33	1.00	
NO_2	ppb	279,512	16.8	4.6	0.58	0.59	0.44	0.43	0.44	0.23	0.47	0.43	0.42	1.00
TLBW and Term Normal Birth Weight															
Air pollutants	Unit	No. of Births	Mean	SD	Pearson Correlation Coefficients									
					$PM_{2.5-10}$ barium	$PM_{2.5-10}$ zinc	$PM_{2.5}$ mass	$PM_{2.5}$ barium	$PM_{2.5}$ zinc	$PM_{2.5}$ ROS	$PM_{2.5}$ OH	$PM_{2.5}$ DTT	$PM_{2.5}$ BC	NO_2
$PM_{2.5-10}$ barium	ng/m³	260,575	30.9	7.3	1.00									
$PM_{2.5-10}$ zinc	ng/m³	260,614	7.1	1.3	0.83	1.00								
$PM_{2.5}$ mass	ug/m³	260,647	10.0	1.1	0.45	0.44	1.00							
$PM_{2.5}$ barium	ng/m³	262,483	17.1	3.9	0.75	0.68	0.66	1.00						
$PM_{2.5}$ zinc	ng/m³	262,483	9.7	1.7	0.49	0.65	0.77	0.79	1.00					
$PM_{2.5}$ ROS	nmol/L	262,483	113.5	12.1	0.65	0.58	0.33	0.84	0.62	1.00				

$PM_{2.5}$ OH formation	$uM/min/m^3$	262,483	5.5	1.2	0.77	0.52	0.68	0.81	0.62	0.64	1.00			
$PM_{2.5}$ DTT loss	$uM/min/m^3$	262,483	609.3	116.5	0.80	0.52	0.37	0.64	0.39	0.61	0.78	1.00		
$PM_{2.5}$ black carbon	ug/m^3	262,483	0.4	0.1	0.43	0.45	0.87	0.69	0.75	0.35	0.61	0.33	1.00	
LUR-NO_2	ppb	262,483	16.8	4.6	0.58	0.59	0.44	0.43	0.44	0.22	0.47	0.43	0.41	1.00

Table 5.3 Distributions of air pollution exposures among births in Los Angeles County, California, 2017-2019.

		PTB (N= 23,131)						TLBW (N=6,102)						Term Normal Birth Weight (N=256,381)					
Air pollutants	Unit	No. of Births	Mean	SD	25th Pct	50th Pct	75th Pct	No. of Births	Mean	SD	25th Pct	50th Pct	75th Pct	No. of Births	Mean	SD	25th Pct	50th Pct	75th Pct
$PM_{2.5-10}$-Barium	ng/m^3	22,933	31.5	7.0	27.2	32.2	36.5	6,052	31.5	7.1	27.1	32.2	36.7	254,523	30.9	7.3	26.4	31.7	36.2
$PM_{2.5-10}$-Zinc	ng/m^3	22,938	7.2	1.2	6.5	7.3	8.0	6,053	7.2	1.2	6.5	7.3	8.0	254,561	7.1	1.3	6.4	7.2	8.0
$PM_{2.5}$ mass	ug/m^3	22,944	10.1	1.0	9.4	10.1	10.9	6,053	10.0	1.1	9.4	10.1	10.9	254,594	10.0	1.1	9.3	10.0	10.8
Volume normalized $PM_{2.5}$ barium	ng/m^3	23,131	17.3	3.9	14.7	17.4	20.1	6,102	17.4	4.0	14.7	17.4	20.2	256,381	17.1	3.9	14.5	17.1	19.8
$PM_{2.5}$ mass normalized barium	ng/g	23,131	1847.9	296.2	1642.9	1818.7	2012.4	6,102	1858.0	304.8	1643.9	1825.9	2023.5	256,381	1847.2	297.0	1642.4	1815.9	2013.0
Volume normalized $PM_{2.5}$ barium during winter	ng/m^3	23,131	22.1	3.9	19.4	22.1	24.8	6,102	22.1	4.0	19.4	22.1	24.9	256,381	21.8	3.9	19.2	21.8	24.6
Volume normalized $PM_{2.5}$ barium during summer	ng/m^3	23,131	12.6	3.9	10.0	12.6	15.4	6,102	12.6	3.9	10.0	12.6	15.5	256,381	12.4	3.9	9.8	12.3	15.1

Volume normalized $PM_{2.5}$ zinc	ng/m^3	23,131	9.8	1.8	8.8	9.9	11.1		6,102	9.8	1.7	8.7	9.9	11.1		256,381	9.7	1.7	8.7	9.8	11.0
$PM_{2.5}$ mass normalized zinc	ng/g	23,131	6.9	0.1	6.8	6.9	7.0		6,102	6.9	0.1	6.8	6.9	7.0		256,381	6.9	0.1	6.8	6.9	7.0
Volume normalized $PM_{2.5}$ zinc during winter	ng/m^3	23,131	12.3	1.8	11.2	12.4	13.6		6,102	12.2	1.7	11.2	12.3	13.5		256,381	12.2	1.7	11.1	12.2	13.5
Volume normalized $PM_{2.5}$ zinc during summer	ng/m^3	23,131	7.4	1.7	6.3	7.5	8.7		6,102	7.3	1.7	6.3	7.4	8.6		256,381	7.3	1.7	6.2	7.3	8.6
$PM_{2.5}$ ROS	nmol/L	23,131	113.7	11.8	105.7	113.4	121.3		6,102	114.0	12.2	105.7	113.5	121.7		256,381	113.5	12.1	105.1	113.0	121.3
$PM_{2.5}$ ROS during winter	nmol/L	23,131	124.2	11.8	116.2	123.9	131.8		6,102	124.5	12.2	116.2	124.0	132.2		256,381	124.0	12.1	115.6	123.5	131.8
$PM_{2.5}$ ROS during summer	nmol/L	23,131	103.2	11.8	95.2	102.9	110.8		6,102	103.5	12.2	95.2	103.0	111.2		256,381	103.0	12.1	94.6	102.5	110.8
Volume normalized $PM_{2.5}$ OH formation	uM/min/m^3	23,131	5.6	1.2	4.9	5.8	6.5		6,102	5.6	1.2	4.9	5.7	6.5		256,381	5.5	1.2	4.8	5.6	6.4
$PM_{2.5}$ mass normalized OH formation	uM/min/ug	23,131	0.6	0.1	0.5	0.6	0.6		6,102	0.6	0.1	0.5	0.6	0.6		256,381	0.6	0.1	0.5	0.6	0.6
Volume normalized $PM_{2.5}$ DTT loss	uM/min/m^3	23,131	613.0	115.4	536.0	614.8	681.0		6,102	615.2	119.3	533.9	615.4	684.6		256,381	609.1	116.4	527.5	611.8	677.2
$PM_{2.5}$ mass normalized DTT loss	uM/min/ug	23,131	62.5	4.1	60.0	61.6	64.1		6,102	62.6	4.2	60.0	61.7	64.3		256,381	62.4	4.0	60.0	61.6	64.0
Volume normalized $PM_{2.5}$ black carbon	ug/m^3	23,131	0.4	0.1	0.4	0.4	0.5		6,102	0.4	0.1	0.4	0.4	0.5		256,381	0.4	0.1	0.4	0.4	0.5
$PM_{2.5}$ mass normalized	ug/g	23,131	4.8	0.4	4.5	4.8	5.1		6,102	4.8	0.4	4.5	4.8	5.1		256,381	4.8	0.4	4.5	4.8	5.1

black carbon																				
LUR-NO_2	ppb	23,131	17.2	4.6	14.2	17.0	20.1		6,102	17.0	4.6	14.1	16.8	19.8		256,381	16.8	4.6	13.8	16.5	19.6

5.3.3 Association Between Whole Pregnancy Exposures and PTB/TLBW

As the effect estimates for preterm births and TLBW based on seasonal models (winter/summer) for exposure surfaces for $PM_{2.5}$ metals and $PM_{2.5}$ ROS did not differ much from the ones based on the annual exposure model, in the following, we are presenting only results based on the annual spatial estimates used for this 4-year period when modeling whole pregnancy exposures. We estimated small increases in the odds with almost all speciated $PM_{2.5}$ exposures for preterm birth (ORs ranging from 1.01 to 1.04) and TLBW (ORs ranging from 1.02 to 1.06) per IQR exposure increment (Table 5.4). The effect estimate for $PM_{2.5}$ mass and PTB (OR=1.05, 95% CI 1.02, 1.07) is slightly stronger than for any of the speciated $PM_{2.5}$ exposures, while for TLBW the OR of $PM_{2.5}$ mass was weaker than or equal to some of the speciated $PM_{2.5}$ exposures (OR 1.02 (95% CI 0.98, 1.05) per IQR increment). In addition, the effect estimates for $PM_{2.5-10}$ barium and $PM_{2.5-10}$ zinc on both PTB and TLBW were slightly stronger than for $PM_{2.5}$ barium and $PM_{2.5}$ zinc, respectively, except for $PM_{2.5-10}$ barium and TLBW.

Table 5.4 ORs (95% CI) from unconditional logistic regression models for adverse birth outcomes according to per interquartile range (IQR) increase of whole pregnancy exposures to air pollutants.

PTB and Term Normal Birth Weight						
Air pollutants	Unit (IQR)	No. of Births	ORs (95% CI)			
			Crude	Adjusted [a]	Adjusted [b]	Adjusted [c]
$PM_{2.5-10}$						
Volume normalized $PM_{2.5-10}$ barium	ng/m^3	277,456	1.11 (1.09, 1.14)	1.04 (1.02, 1.06)	1.02 (1.00, 1.04)	1.01 (0.99, 1.04)
Volume normalized $PM_{2.5-10}$ zinc	ng/m^3	277,499	1.13 (1.11, 1.15)	1.05 (1.03, 1.07)	1.04 (1.01, 1.06)	1.03 (1.01, 1.05)
$PM_{2.5}$ mass						
$PM_{2.5}$ mass	ug/m^3	277,538	1.10 (1.08, 1.12)	1.05 (1.02, 1.07)	-	1.03 (1.00, 1.05)
Barium						
Volume normalized $PM_{2.5}$ barium	ng/m^3	279,512	1.09 (1.07, 1.11)	1.03 (1.01, 1.06)	1.02 (0.99, 1.04)	1.02 (0.99, 1.04)
$PM_{2.5}$ mass normalized barium	ng/g	279,512	1.00 (0.99, 1.02)	1.00 (0.98, 1.02)	0.99 (0.98, 1.01)	0.99 (0.97, 1.01)
Zinc						
Volume normalized $PM_{2.5}$ zinc	ng/m^3	279,512	1.08 (1.06, 1.10)	1.03 (1.01, 1.05)	1.01 (0.98, 1.04)	1.01 (0.99, 1.04)
$PM_{2.5}$ mass normalized zinc	ng/g	279,512	0.95 (0.93, 0.96)	0.98 (0.97, 1.00)	0.96 (0.94, 0.98)	0.98 (0.96, 0.99)
$PM_{2.5}$ ROS						
$PM_{2.5}$ ROS	nmol/L	279,512	1.03 (1.01, 1.05)	1.01 (0.99, 1.03)	1.00 (0.98, 1.02)	1.00 (0.98, 1.02)
OH formation						
Volume normalized $PM_{2.5}$ OH formation	uM/min/m^3	279,512	1.09 (1.07, 1.11)	1.04 (1.02, 1.06)	1.02 (1.00, 1.05)	1.02 (1.00, 1.04)
$PM_{2.5}$ mass normalized OH formation	uM/min/ug	279,512	1.07 (1.05, 1.09)	1.02 (1.01, 1.04)	1.02 (1.00, 1.04)	1.01 (1.00, 1.03)
DTT loss						
Volume normalized $PM_{2.5}$ DTT loss	uM/min/m^3	279,512	1.04 (1.03, 1.06)	1.01 (0.99, 1.03)	0.99 (0.98, 1.01)	0.99 (0.97, 1.01)
$PM_{2.5}$ mass normalized DTT loss	uM/min/ug	279,512	1.02 (1.01, 1.04)	1.01 (1.00, 1.02)	1.00 (0.99, 1.02)	1.00 (0.99, 1.02)

Black carbon						
Volume normalized $PM_{2.5}$ black carbon annual	ug/m^3	279,512	1.08 (1.06, 1.10)	1.03 (1.01, 1.05)	0.97 (0.93, 1.02)	1.01 (0.99, 1.04)
$PM_{2.5}$ mass normalized black carbon	ug/g	279,512	1.06 (1.04, 1.08)	1.02 (1.00, 1.05)	0.95 (0.91, 0.99)	1.01 (0.99, 1.03)
NO_2						
LUR-NO_2	ppb	279,512	1.13 (1.11, 1.14)	1.05 (1.03, 1.07)	1.04 (1.02, 1.06)	

TLBW and Term Normal Birth Weight

Air pollutants	Unit (IQR)	No. of Births	ORs (95% CI)			
			Crude	Adjusted [a]	Adjusted [b]	Adjusted [c]
$PM_{2.5-10}$						
Volume normalized $PM_{2.5-10}$ barium	ng/m^3	260,575	1.11 (1.07, 1.15)	1.06 (1.02, 1.10)	1.06 (1.02, 1.11)	1.05 (1.01, 1.10)
Volume normalized $PM_{2.5-10}$ zinc	ng/m^3	260,614	1.10 (1.06, 1.14)	1.04 (1.01, 1.08)	1.04 (1.00, 1.09)	1.03 (0.99, 1.08)
$PM_{2.5}$ mass						
$PM_{2.5}$ mass	ug/m^3	260,647	1.07 (1.03, 1.11)	1.02 (0.98, 1.05)	-	1.00 (0.96, 1.04)
Barium						
Volume normalized $PM_{2.5}$ barium	ng/m^3	262,483	1.10 (1.06, 1.14)	1.06 (1.02, 1.10)	1.10 (1.04, 1.15)	1.05 (1.01, 1.10)
$PM_{2.5}$ mass normalized barium	ng/g	262,483	1.05 (1.01, 1.08)	1.04 (1.01, 1.08)	1.05 (1.01, 1.08)	1.04 (1.00, 1.07)
Zinc						
Volume normalized $PM_{2.5}$ zinc	ng/m^3	262,483	1.06 (1.02, 1.10)	1.02 (0.98, 1.06)	1.04 (0.98, 1.10)	1.01 (0.97, 1.05)
$PM_{2.5}$ mass normalized zinc	ng/g	262,483	0.98 (0.95, 1.01)	0.97 (0.95, 1.01)	0.97 (0.93, 1.00)	0.97 (0.94, 1.00)
$PM_{2.5}$ ROS						
$PM_{2.5}$ ROS	nmol/L	262,483	1.06 (1.02, 1.10)	1.05 (1.02, 1.09)	1.06 (1.02, 1.10)	1.05 (1.01, 1.08)
OH formation						
Volume normalized $PM_{2.5}$ OH formation	$uM/min/m^3$	262,483	1.07 (1.04, 1.11)	1.03 (0.99, 1.07)	1.05 (1.00, 1.10)	1.02 (0.98, 1.06)
$PM_{2.5}$ mass normalized OH formation	uM/min/ug	262,483	1.08 (1.05, 1.11)	1.06 (1.03, 1.09)	1.06 (1.03, 1.09)	1.05 (1.02, 1.09)
DTT loss						
Volume normalized $PM_{2.5}$ DTT loss	$uM/min/m^3$	262,483	1.07 (1.04, 1.10)	1.04 (1.00, 1.07)	1.04 (1.00, 1.08)	1.03 (0.99, 1.07)
$PM_{2.5}$ mass normalized DTT loss	uM/min/ug	262,483	1.04 (1.02, 1.07)	1.03 (1.00, 1.05)	1.03 (1.00, 1.06)	1.02 (1.00, 1.05)
Black carbon						
Volume normalized $PM_{2.5}$ black carbon annual	ug/m^3	262,483	1.07 (1.03, 1.11)	1.03 (0.99, 1.07)	1.06 (0.98, 1.15)	1.01 (0.97, 1.06)
$PM_{2.5}$ mass normalized black carbon	ug/g	262,483	1.05 (1.01, 1.09)	1.02 (0.98, 1.06)	1.03 (0.95, 1.11)	1.01 (0.97, 1.05)
NO_2						
LUR-NO_2	ppb	262,483	1.07 (1.04, 1.11)	1.03 (1.00, 1.07)	1.04 (1.00, 1.08)	-

a Adjusted for sex, parity, maternal age, maternal race and maternal birthplace, payment type of prenatal care, maternal education and maternal smoking.

b Additionally adjusted for PM2.5 mass exposure during pregnancy.

c Additionally adjusted for LUR-NO2 exposure during pregnancy.

For most of the exposures, effect estimates for mass normalized concentrations were similar to those for volume normalized concentrations, except for $PM_{2.5}$ zinc for which the ORs for both PTB and TLBW and $PM_{2.5}$ mass normalized zinc were below one while the ORs for volume normalized $PM_{2.5}$ zinc were 1.03 (95% CI 1.01, 1.05) and 1.02 (0.98, 1.06) for PTB and TLBW per IQR increase, respectively. After adjusting for $PM_{2.5}$ mass exposure (scaled to the $PM_{2.5}$ mass IQR among births with term normal birth weight) in the regression model for each of the speciated $PM_{2.5}$ exposures, the ORs estimated for most of the speciated $PM_{2.5}$ exposures and PTB moved towards the null and some even became negative, but we still observed small positive associations for $PM_{2.5}$ barium (ng/m^3), $PM_{2.5}$ zinc (ng/m^3) and $PM_{2.5}$ OH formation (uM/min/m^3) with ORs of 1.02 (95% CI 0.99, 1.04), 1.01 (95% CI 0.98, 1.04) and 1.02 (95% CI 1.00, 1.05), respectively, suggesting an association between brake and tire wear, and oxidative stress potential on PTB beyond the contributions represented by modeled $PM_{2.5}$ mass exposures.

Interestingly, for TLBW, the ORs associated with each IQR increment of the speciated $PM_{2.5}$ exposures remained the same or were slightly higher after adjusting for the modeled $PM_{2.5}$ mass exposures in the same regression model. Specifically, larger size associations were observed for most of the speciated $PM_{2.5}$ exposures including both volume and mass normalized $PM_{2.5}$ barium, $PM_{2.5}$ ROS and $PM_{2.5}$ black carbon, and volume normalized $PM_{2.5}$ zinc and $PM_{2.5}$ OH formation, with ORs ranging from 1.03 to 1.10; while for all the rest of the speciated $PM_{2.5}$ exposures effect estimates did not change after adjusting for $PM_{2.5}$ mass exposure in the same regression model.

For LUR-NO_2 exposures estimated with our original LUR model for combustion related traffic exposures we observed that whole pregnancy exposures were associated with PTB (OR = 1.05 (95% CI 1.03, 1.07) and TLBW (OR = 1.03, 95% CI 1.00, 1.07) per IQR increment. When adjusting for LUR-NO_2 in regression models for speciated $PM_{2.5-10}$ or $PM_{2.5}$ exposures, the effect estimates decreased slightly but did not become null. Also, the effect estimates for exposure to $PM_{2.5}$ mass normalized OH formation and PTB and TLBW changed very little after adjusting for either $PM_{2.5}$ mass or LUR-NO_2 in the same model. These findings suggest that the metals and oxidative stress potential markers exerted an effect largely independent of that from NO_2.

Estimating effects by quartile for each exposure measure (Table 5.5), risks were increased for PTB and TLBW for volume normalized as well as mass normalized speciated $PM_{2.5}$ exposures (barium, zinc, OH formation, DTT loss and black carbon). Furthermore, the analyses for volume normalized speciated $PM_{2.5}$ exposures suggested a monotonic exposure-response. While we observed positive associations with most of the mass normalized speciated $PM_{2.5}$ exposures and PTB, an exposure-response trend was not detected for most except for black carbon. An exposure-response trend was also observed for TLBW with $PM_{2.5}$ black carbon, as well as with $PM_{2.5}$ barium and $PM_{2.5}$ OH formation.

A monotonic exposure-response was also observed for the $PM_{2.5}$ mass exposure and PTB or TLBW. After adjusting for the $PM_{2.5}$ mass exposure in regression models for speciated $PM_{2.5}$ exposures, most showed association with PTB and TLBW, but effect estimates were slightly lower except for volume normalized barium concentration and TLBW for which we estimated a larger effect size (OR of 1.14 (95% CI 1.03, 1.25) > 75th percentile). Associations for black carbon and PTB/TLBW turned negative, and the 95% CIs included the null when adjusting for $PM_{2.5}$ mass, likely because of the high correlation between the two measures, which may have resulted partly from the similar model structure used to estimate both exposure surfaces. When adjusting for quartiles of LUR-NO_2 in each model for speciated $PM_{2.5}$ exposures, effect estimates changed little with a slight decrease in effect estimate size for both outcomes. Similar to the IQR increment

based results, the ORs for $PM_{2.5}$ mass normalized OH formation and PTB or TLBW changed very little after adjusting for $PM_{2.5}$ mass or LUR-NO_2 in the same regression model.

Table 5.5 ORs (95% CI) from unconditional logistic regression models for adverse birth outcomes according to whole pregnancy exposure levels to $PM_{2.5}$ mass and speciated $PM_{2.5}$ exposures.

PTB				
Air pollutants	**ORs (95% CI)**			
	Crude	**Adjusted [a]**	**Adjusted [b]**	**Adjusted [c]**
$PM_{2.5}$ mass				
< 25 pct	reference	reference	-	reference
25pct ~ 50 pct	0.97 (0.94, 1.01)	0.98 (0.94, 1.02)	-	0.96 (0.92, 1.00)
50pct ~ 75 pct	1.09 (1.05, 1.13)	1.05 (1.01, 1.09)	-	1.03 (0.99, 1.08)
> 75 pct	1.20 (1.15, 1.24)	1.08 (1.04, 1.13)	-	1.05 (1.01, 1.10)
Volume normalized $PM_{2.5}$ barium (ng/m^3)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.06 (1.02, 1.10)	1.02 (0.98, 1.06)	1.01 (0.97, 1.06)	1.01 (0.97, 1.05)
50pct ~ 75 pct	1.15 (1.10, 1.19)	1.06 (1.02, 1.11)	1.03 (0.99, 1.08)	1.05 (1.00, 1.09)
> 75 pct	1.19 (1.15, 1.24)	1.07 (1.02, 1.11)	1.02 (0.97, 1.07)	1.04 (1.00, 1.09)
Volume normalized $PM_{2.5}$ zinc (ng/m^3)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.02 (0.99, 1.07)	0.99 (0.95, 1.03)	0.99 (0.95, 1.03)	0.98 (0.94, 1.02)
50pct ~ 75 pct	1.12 (1.07, 1.16)	1.04 (1.00, 1.08)	1.01 (0.97, 1.07)	1.02 (0.98, 1.07)
> 75 pct	1.18 (1.14, 1.22)	1.07 (1.03, 1.12)	1.02 (0.96, 1.08)	1.05 (1.00, 1.09)
$PM_{2.5}$ ROS (nmol/L)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.06 (1.02, 1.10)	1.01 (0.97, 1.05)	1.00 (0.96, 1.04)	1.00 (0.96, 1.04)
50pct ~ 75 pct	1.13 (1.09, 1.17)	1.05 (1.01, 1.09)	1.03 (0.98, 1.07)	1.04 (0.99, 1.08)

Air Pollutants	**ORs (95% CI)**			
	Crude	**Adjusted [a]**	**Adjusted [b]**	**Adjusted [c]**
$PM_{2.5}$ mass normalized barium (ng/g)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	0.99 (0.96, 1.03)	0.99 (0.95, 1.03)	0.98 (0.94, 1.02)	0.99 (0.95, 1.03)
50pct ~ 75 pct	1.02 (0.98, 1.06)	1.00 (0.97, 1.04)	0.98 (0.94, 1.02)	0.99 (0.95, 1.03)
> 75 pct	1.00 (0.97, 1.04)	0.99 (0.95, 1.03)	0.97 (0.93, 1.01)	0.98 (0.94, 1.02)
$PM_{2.5}$ mass normalized zinc (ng/g)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.01 (0.98, 1.05)	1.00 (0.96, 1.04)	0.97 (0.93, 1.01)	0.98 (0.95, 1.02)
50pct ~ 75 pct	1.03 (0.99, 1.07)	1.02 (0.98, 1.06)	0.98 (0.94, 1.02)	1.00 (0.97, 1.05)
> 75 pct	0.90 (0.87, 0.94)	0.97 (0.93, 1.01)	0.92 (0.88, 0.96)	0.96 (0.92, 1.00)

> 75 pct	1.06 (1.02, 1.10)	1.02 (0.98, 1.06)	0.99 (0.95, 1.04)	1.01 (0.97, 1.05)
Volume normalized $PM_{2.5}$ OH formation ($uM/min/m^3$)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.07 (1.03, 1.11)	1.03 (0.99, 1.07)	1.02 (0.98, 1.07)	1.02 (0.97, 1.06)
50pct ~ 75 pct	1.16 (1.12, 1.21)	1.06 (1.02, 1.10)	1.03 (0.99, 1.08)	1.04 (1.00, 1.08)
> 75 pct	1.18 (1.13, 1.22)	1.07 (1.03, 1.11)	1.02 (0.97, 1.08)	1.05 (1.00, 1.09)
Volume normalized $PM_{2.5}$ DTT loss ($uM/min/m^3$)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.06 (1.02, 1.10)	1.01 (0.97, 1.05)	1.01 (0.97, 1.05)	1.00 (0.96, 1.04)
50pct ~ 75 pct	1.06 (1.02, 1.10)	1.01 (0.97, 1.05)	0.99 (0.95, 1.03)	0.98 (0.94, 1.02)
> 75 pct	1.10 (1.06, 1.15)	1.02 (0.98, 1.06)	0.99 (0.95, 1.03)	0.99 (0.95, 1.03)
Volume normalized $PM_{2.5}$ black carbon				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	0.98 (0.94, 1.02)	0.96 (0.92, 1.00)	0.93 (0.88, 0.97)	0.94 (0.90, 0.98)
50pct ~ 75 pct	1.03 (0.99, 1.07)	0.99 (0.95, 1.03)	0.90 (0.85, 0.96)	0.96 (0.92, 1.01)
> 75 pct	1.16 (1.11, 1.20)	1.06 (1.01, 1.10)	0.93 (0.87, 1.01)	1.02 (0.98, 1.07)

$PM_{2.5}$ mass normalized OH formation (uM/min/ug)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.08 (1.04, 1.12)	1.02 (0.98, 1.06)	1.01 (0.97, 1.05)	1.01 (0.97, 1.05)
50pct ~ 75 pct	1.20 (1.16, 1.25)	1.08 (1.03, 1.12)	1.06 (1.01, 1.10)	1.06 (1.02, 1.11)
> 75 pct	1.21 (1.17, 1.26)	1.07 (1.03, 1.11)	1.05 (1.01, 1.10)	1.05 (1.01, 1.10)
$PM_{2.5}$ mass normalized DTT loss (uM/min/ug)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.09 (1.05, 1.13)	1.06 (1.02, 1.10)	1.04 (1.00, 1.08)	1.06 (1.01, 1.10)
50pct ~ 75 pct	1.07 (1.03, 1.11)	1.04 (1.00, 1.09)	1.01 (0.97, 1.06)	1.04 (1.00, 1.08)
> 75 pct	1.10 (1.06, 1.14)	1.05 (1.01, 1.09)	1.02 (0.97, 1.06)	1.04 (1.00, 1.08)
$PM_{2.5}$ mass normalized black carbon				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	0.99 (0.95, 1.03)	0.97 (0.94, 1.01)	0.94 (0.90, 0.99)	0.96 (0.92, 1.00)
50pct ~ 75 pct	1.03 (0.99, 1.07)	1.00 (0.96, 1.04)	0.92 (0.86, 0.98)	0.98 (0.94, 1.02)
> 75 pct	1.13 (1.09, 1.17)	1.05 (1.01, 1.10)	0.93 (0.86, 1.00)	1.03 (0.98, 1.07)

TLBW

Air pollutants	ORs (95% CI)			
	Crude	Adjusted [a]	Adjusted [b]	Adjusted [c]
$PM_{2.5}$ mass				
< 25 pct	reference	reference	-	reference
25pct ~ 50 pct	1.02 (0.95, 1.10)	0.98 (0.91, 1.06)	-	0.96 (0.89, 1.04)
50pct ~ 75 pct	1.08 (1.01, 1.17)	1.00 (0.92, 1.07)	-	0.98 (0.90, 1.06)

Air Pollutants	ORs (95% CI)			
	Crude	Adjusted [a]	Adjusted [b]	Adjusted [c]

> 75 pct	1.17 (1.09, 1.25)	1.07 (0.99, 1.15)	-	1.05 (0.96, 1.13)
Volume normalized $PM_{2.5}$ barium (ng/m^3)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.06 (0.98, 1.14)	0.99 (0.92, 1.07)	1.01 (0.94, 1.10)	0.99 (0.91, 1.07)
50pct ~ 75 pct	1.12 (1.04, 1.20)	1.05 (0.97, 1.13)	1.07 (0.98, 1.17)	1.04 (0.96, 1.12)
> 75 pct	1.21 (1.13, 1.30)	1.12 (1.04, 1.21)	1.14 (1.03, 1.25)	1.11 (1.02, 1.20)
Volume normalized $PM_{2.5}$ zinc (ng/m^3)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.03 (0.96, 1.11)	0.98 (0.91, 1.06)	0.98 (0.91, 1.07)	0.97 (0.90, 1.04)
50pct ~ 75 pct	1.08 (1.01, 1.16)	1.00 (0.92, 1.07)	0.98 (0.90, 1.08)	0.98 (0.91, 1.06)
> 75 pct	1.12 (1.04, 1.20)	1.04 (0.97, 1.13)	1.00 (0.89, 1.11)	1.02 (0.94, 1.11)
$PM_{2.5}$ ROS (nmol/L)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.04 (0.97, 1.12)	1.00 (0.92, 1.07)	1.00 (0.92, 1.08)	0.99 (0.92, 1.07)
50pct ~ 75 pct	1.07 (1.00, 1.15)	1.05 (0.97, 1.13)	1.04 (0.96, 1.12)	1.04 (0.96, 1.12)
> 75 pct	1.12 (1.04, 1.20)	1.09 (1.01, 1.18)	1.08 (1.00, 1.17)	1.08 (1.00, 1.17)
Volume normalized $PM_{2.5}$ OH formation ($uM/min/m^3$)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.02 (0.95, 1.10)	0.99 (0.92, 1.07)	0.99 (0.92, 1.08)	0.98 (0.90, 1.06)
50pct ~ 75 pct	1.10 (1.02, 1.18)	1.03 (0.95, 1.11)	1.02 (0.94, 1.11)	1.01 (0.93, 1.09)
> 75 pct	1.13 (1.05, 1.21)	1.05 (0.97, 1.13)	1.02 (0.93, 1.12)	1.02 (0.94, 1.11)

$PM_{2.5}$ mass normalized barium (ng/g)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	0.95 (0.88, 1.02)	0.94 (0.87, 1.02)	0.94 (0.87, 1.01)	0.94 (0.87, 1.01)
50pct ~ 75 pct	1.03 (0.96, 1.10)	1.02 (0.95, 1.10)	1.01 (0.93, 1.09)	1.02 (0.94, 1.10)
> 75 pct	1.06 (0.99, 1.14)	1.05 (0.97, 1.13)	1.04 (0.96, 1.12)	1.04 (0.97, 1.12)
$PM_{2.5}$ mass normalized zinc (ng/g)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.07 (1.00, 1.15)	1.04 (0.97, 1.12)	1.02 (0.95, 1.10)	1.03 (0.96, 1.11)
50pct ~ 75 pct	1.01 (0.94, 1.08)	0.97 (0.90, 1.05)	0.94 (0.87, 1.02)	0.96 (0.89, 1.04)
> 75 pct	0.97 (0.90, 1.04)	0.95 (0.88, 1.02)	0.91 (0.84, 0.99)	0.94 (0.87, 1.02)
$PM_{2.5}$ mass normalized OH formation (uM/min/ug)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.08 (1.00, 1.16)	1.05 (0.98, 1.14)	1.06 (0.98, 1.14)	1.05 (0.97, 1.13)
50pct ~ 75 pct	1.16 (1.08, 1.25)	1.12 (1.04, 1.21)	1.12 (1.03, 1.21)	1.11 (1.03, 1.20)
> 75 pct	1.23 (1.14, 1.32)	1.16 (1.08, 1.26)	1.16 (1.07, 1.26)	1.15 (1.07, 1.25)

Volume normalized $PM_{2.5}$ DTT loss ($uM/min/m^3$)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.02 (0.95, 1.10)	0.99 (0.92, 1.07)	1.00 (0.93, 1.08)	0.99 (0.91, 1.06)
50pct ~ 75 pct	1.02 (0.95, 1.10)	0.98 (0.91, 1.05)	0.97 (0.90, 1.05)	0.96 (0.89, 1.04)
> 75 pct	1.14 (1.06, 1.22)	1.07 (0.99, 1.15)	1.06 (0.98, 1.14)	1.05 (0.97, 1.14)
Volume normalized $PM_{2.5}$ black carbon				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.01 (0.94, 1.08)	0.96 (0.89, 1.03)	0.95 (0.87, 1.05)	0.94 (0.87, 1.02)
50pct ~ 75 pct	1.06 (0.98, 1.13)	0.99 (0.92, 1.07)	0.96 (0.86, 1.08)	0.97 (0.89, 1.05)
> 75 pct	1.12 (1.04, 1.20)	1.04 (0.96, 1.12)	0.96 (0.83, 1.10)	1.01 (0.93, 1.09)

$PM_{2.5}$ mass normalized DTT loss (uM/min/ug)				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.01 (0.94, 1.08)	0.98 (0.91, 1.05)	0.97 (0.89, 1.04)	0.97 (0.90, 1.05)
50pct ~ 75 pct	0.98 (0.91, 1.05)	0.95 (0.88, 1.02)	0.93 (0.86, 1.01)	0.94 (0.88, 1.02)
> 75 pct	1.10 (1.02, 1.18)	1.05 (0.97, 1.13)	1.02 (0.94, 1.11)	1.04 (0.96, 1.12)
$PM_{2.5}$ mass normalized black carbon				
< 25 pct	reference	reference	reference	reference
25pct ~ 50 pct	1.02 (0.95, 1.10)	0.97 (0.90, 1.05)	0.95 (0.87, 1.05)	0.96 (0.89, 1.03)
50pct ~ 75 pct	1.03 (0.95, 1.10)	0.97 (0.90, 1.05)	0.92 (0.82, 1.03)	0.96 (0.88, 1.03)
> 75 pct	1.09 (1.02, 1.18)	1.03 (0.95, 1.11)	0.91 (0.79, 1.04)	1.00 (0.93, 1.09)

a Adjusted for sex, parity, maternal age, maternal race and maternal birthplace, payment type of prenatal care, maternal education and maternal smoking.

b Additionally adjusted for $PM_{2.5}$ exposure during pregnancy.

c Additionally adjusted for LUR-NO_2 exposure during pregnancy.

5.3.4 Association between Pregnancy Exposures and PTB/TLBW by Race/Ethnicity

In stratified analyses by maternal race/ethnicity, we estimated stronger effects per IQR increase for most of the speciated $PM_{2.5-10}$ and $PM_{2.5}$ exposures for mothers with Hispanic race/ethnicity while non-Hispanic White mothers exhibited lower risks compared to all other race/ethnicity groups, except for $PM_{2.5-10}$ zinc with a higher risk of PTB among non-Hispanic White mothers (Table 5.6). Infants born to Asian/ Pacific Islander mothers had higher odds of preterm birth compared to other race groups with exposures to barium (in both $PM_{2.5-10}$ and $PM_{2.5}$), or our oxidative stress markers ($PM_{2.5}$ ROS, $PM_{2.5}$ OH formation and $PM_{2.5}$ DTT loss). Also, mothers who reported their race/ethnicity as African American had higher odds of preterm delivery related to $PM_{2.5}$ zinc or $PM_{2.5}$ black carbon exposures; however, the point estimates of effect for most other constituents of $PM_{2.5}$ as well as $PM_{2.5-10}$ for African American mothers were below 1 and the 95% CI included null i.e., we did not see statistically significant associations.

We examined the mean and standard deviations for each of the constituents by maternal race/ethnicity and found the highest mean with the lowest standard deviations for the African American women for most of the investigated $PM_{2.5-10}$ and $PM_{2.5}$ component exposures (Table 5.7). Furthermore, we plotted the population density based on maternal residential addresses on a map by maternal race/ethnicity groups (Figure 5.1), and as expected, the African American women did appear to have more clustering of residencies in one area of LA county compared to other race groups. Similar patterns were observed for TLBW. Compared to other race/ethnic groups, again we estimated a higher risk of TLBW with increasing exposure levels of speciated $PM_{2.5}$ exposures including $PM_{2.5}$ barium and $PM_{2.5}$ zinc, our brake and tire wear markers, $PM_{2.5}$ ROS as well as $PM_{2.5}$ black carbon for mothers of Hispanic origin. For Asian/Pacific Islander mothers we estimated higher effect estimates for $PM_{2.5-10}$ barium, $PM_{2.5-10}$ zinc, $PM_{2.5}$ barium, $PM_{2.5}$ ROS, $PM_{2.5}$ OH formation and $PM_{2.5}$ DTT loss for TLBW. When we scaled the exposures to the maternal race/ethnicity specific IQR among non-cases results changed little or not at all.

Considering the potential of selection bias as 2% of all women had missing information for education, and 1.5% of women had missing information for smoking, we conducted additional sensitivity analyses comparing the results for the total population and the population after excluding those with missing values for either education or smoking while adjusting for all potential confounders except for maternal education and smoking. The results were similar with a slight decrease in the point estimates, suggesting that there was not much if any selection bias caused by excluding women without information on education or smoking. We did not assess multiple pollutants in the same model except for what we reported above as the metals/oxidative stress markers were too highly correlated with each other as shown in Table 5.2.

Table 5.6 ORs (95% CI) from unconditional logistic regression models for adverse birth outcomes according to air pollution exposures in the whole of pregnancy, stratified by maternal race/ethnicity.

PTB							
Air pollutants	Unit (IQR)	ORs (95% CI) [a]					
		Total population (N = 279,512)	White, non-Hispanic (N=98,916)	Hispanic of any race (N=102,285)	African American (N=17,931)	Asian/ Pacific Islander (N=45,924)	Other [b] (N=10,705)
$PM_{2.5-10}$							
Volume normalized $PM_{2.5-10}$ barium	ng/m^3	1.04 (1.02, 1.06)	1.03 (1.00, 1.07)	1.03 (1.00, 1.07)	0.98 (0.91, 1.06)	1.05 (1.00, 1.10)	0.98 (0.89, 1.08)
Volume normalized $PM_{2.5-10}$ zinc	ng/m^3	1.05 (1.03, 1.07)	1.06 (1.03, 1.10)	1.05 (1.01, 1.08)	0.98 (0.91, 1.05)	1.03 (0.99, 1.08)	1.01 (0.92, 1.12)
$PM_{2.5}$ mass							
$PM_{2.5}$ mass	ug/m^3	1.05 (1.02, 1.07)	1.02 (0.99, 1.06)	1.07 (1.04, 1.11)	1.04 (0.96, 1.14)	1.01 (0.97, 1.06)	1.02 (0.93, 1.13)
$PM_{2.5}$ Barium							
Volume normalized $PM_{2.5}$ barium	ng/m^3	1.03 (1.01, 1.06)	1.02 (0.98, 1.05)	1.04 (1.01, 1.08)	0.98 (0.91, 1.05)	1.06 (1.00, 1.12)	1.01 (0.92, 1.11)
$PM_{2.5}$ mass normalized barium	ng/g	1.00 (0.98, 1.02)	0.98 (0.95, 1.01)	1.01 (0.99, 1.04)	0.94 (0.88, 1.00)	1.04 (0.99, 1.10)	0.99 (0.91, 1.07)
$PM_{2.5}$ Zinc							
Volume normalized $PM_{2.5}$ zinc	ng/m^3	1.03 (1.01, 1.05)	1.02 (0.99, 1.05)	1.05 (1.02, 1.08)	1.04 (0.97, 1.12)	1.02 (0.97, 1.08)	1.01 (0.92, 1.11)
$PM_{2.5}$ mass normalized zinc	ng/g	0.98 (0.97, 1.00)	0.96 (0.93, 0.99)	1.03 (1.00, 1.05)	1.01 (0.95, 1.07)	0.97 (0.93, 1.01)	0.95 (0.88, 1.03)
$PM_{2.5}$ ROS							
$PM_{2.5}$ ROS	nmol/L	1.01 (0.99, 1.03)	0.99 (0.96, 1.02)	1.03 (1.00, 1.06)	0.94 (0.88, 1.01)	1.06 (1.01, 1.11)	1.01 (0.92, 1.10)
$PM_{2.5}$ OH formation							
Volume normalized $PM_{2.5}$ OH formation	uM/min/m^3	1.04 (1.02, 1.06)	1.02 (0.99, 1.06)	1.05 (1.01, 1.08)	1.03 (0.96, 1.10)	1.04 (0.99, 1.10)	1.01 (0.92, 1.10)
$PM_{2.5}$ mass normalized OH formation	uM/min/ug	1.02 (1.01, 1.04)	1.02 (0.99, 1.05)	1.01 (0.99, 1.04)	0.95 (0.90, 1.01)	1.08 (1.03, 1.13)	1.01 (0.94, 1.09)
$PM_{2.5}$ DTT loss							
Volume normalized $PM_{2.5}$ DTT loss	uM/min/m^3	1.01 (0.99, 1.03)	1.00 (0.97, 1.03)	1.01 (0.98, 1.04)	0.97 (0.91, 1.04)	1.03 (0.98, 1.08)	0.98 (0.90, 1.07)
$PM_{2.5}$ mass normalized DTT loss	uM/min/ug	1.01 (1.00, 1.02)	1.00 (0.98, 1.03)	1.02 (0.99, 1.04)	0.96 (0.91, 1.01)	1.02 (0.98, 1.07)	1.04 (0.98, 1.10)

$PM_{2.5}$ Black carbon							
Volume normalized $PM_{2.5}$ black carbon	ug/m^3	1.03 (1.01, 1.05)	1.01 (0.98, 1.05)	1.06 (1.03, 1.09)	1.04 (0.96, 1.13)	1.00 (0.95, 1.05)	0.99 (0.89, 1.09)
$PM_{2.5}$ mass normalized black carbon	ug/g	1.02 (1.00, 1.05)	1.00 (0.97, 1.04)	1.06 (1.02, 1.09)	1.04 (0.95, 1.12)	0.99 (0.94, 1.05)	0.99 (0.89, 1.09)
TLBW							
PM2.5 component	**Unit**	**ORs (95% CI) [a]**					
		Total population (N = 262,483)	**White, non-Hispanic (N= 93,562)**	**Hispanic of any race (N=94,808)**	**African American (N= 16,588)**	**Asian/ Pacific Islander (N= 43,982)**	**Others [b] (N= 10,017)**
$PM_{2.5-10}$							
Volume normalized $PM_{2.5-10}$ barium	ng/m^3	1.06 (1.02, 1.10)	1.01 (0.95, 1.08)	1.05 (0.98, 1.12)	0.97 (0.86, 1.10)	1.12 (1.04, 1.21)	1.15 (0.96, 1.38)
Volume normalized $PM_{2.5-10}$ zinc	ng/m^3	1.04 (1.01, 1.08)	1.02 (0.96, 1.09)	1.04 (0.97, 1.12)	0.99 (0.87, 1.12)	1.06 (0.98, 1.13)	1.12 (0.94, 1.34)
$PM_{2.5}$ mass							
$PM_{2.5}$ mass	ug/m^3	1.02 (0.98, 1.05)	0.98 (0.92, 1.05)	1.06 (0.99, 1.13)	1.06 (0.92, 1.23)	0.95 (0.88, 1.02)	1.18 (0.98, 1.42)
$PM_{2.5}$ Barium							
Volume normalized $PM_{2.5}$ barium	ng/m^3	1.06 (1.02, 1.10)	1.02 (0.95, 1.09)	1.09 (1.02, 1.16)	0.93 (0.83, 1.05)	1.08 (0.99, 1.18)	1.12 (0.95, 1.33)
$PM_{2.5}$ mass normalized barium	ng/g	1.04 (1.01, 1.08)	1.02 (0.96, 1.08)	1.07 (1.02, 1.13)	0.90 (0.81, 1.00)	1.10 (1.01, 1.19)	1.03 (0.89, 1.19)
$PM_{2.5}$ Zinc							
Volume normalized $PM_{2.5}$ zinc	ng/m^3	1.02 (0.98, 1.06)	1.00 (0.94, 1.06)	1.05 (0.99, 1.12)	1.02 (0.90, 1.16)	0.95 (0.87, 1.03)	1.10 (0.93, 1.31)
$PM_{2.5}$ mass normalized zinc	ng/g	0.97 (0.95, 1.01)	0.95 (0.90, 1.00)	1.05 (0.99, 1.11)	0.99 (0.89, 1.10)	0.89 (0.83, 0.96)	1.04 (0.90, 1.20)
$PM_{2.5}$ ROS							
$PM_{2.5}$ ROS	nmol/L	1.05 (1.02, 1.09)	1.01 (0.95, 1.07)	1.09 (1.03, 1.16)	0.92 (0.82, 1.03)	1.09 (1.01, 1.18)	1.04 (0.89, 1.23)
$PM_{2.5}$ OH formation							
Volume normalized $PM_{2.5}$ OH formation	uM/min/m^3	1.03 (0.99, 1.07)	0.97 (0.91, 1.04)	1.05 (0.98, 1.11)	1.02 (0.90, 1.14)	1.04 (0.96, 1.13)	1.15 (0.97, 1.36)
$PM_{2.5}$ mass normalized OH formation	uM/min/ug	1.06 (1.03, 1.09)	1.06 (1.00, 1.12)	1.04 (0.99, 1.09)	0.90 (0.82, 1.00)	1.16 (1.08, 1.25)	1.04 (0.91, 1.19)
$PM_{2.5}$ DTT loss							
Volume normalized $PM_{2.5}$ DTT loss	uM/min/m^3	1.04 (1.00, 1.07)	1.00 (0.94, 1.06)	1.02 (0.97, 1.08)	1.00 (0.90, 1.12)	1.13 (1.05, 1.22)	1.04 (0.89, 1.21)
$PM_{2.5}$ mass normalized DTT loss	uM/min/ug	1.03 (1.00, 1.05)	1.02 (0.97, 1.06)	1.04 (1.00, 1.08)	0.97 (0.89, 1.05)	1.07 (1.01, 1.15)	0.97 (0.86, 1.09)

$PM_{2.5}$ Black carbon							
Volume normalized $PM_{2.5}$ black carbon	ug/m^3	1.03 (0.99, 1.07)	0.97 (0.90, 1.04)	1.08 (1.01, 1.15)	0.99 (0.87, 1.14)	0.98 (0.90, 1.07)	1.20 (0.99, 1.45)
$PM_{2.5}$ mass normalized black carbon	ug/g	1.02 (0.98, 1.06)	0.96 (0.90, 1.03)	1.08 (1.01, 1.15)	1.00 (0.87, 1.14)	0.97 (0.89, 1.05)	1.20 (0.99, 1.44)

a Adjusted for sex, parity, maternal age, maternal birthplace, payment type of prenatal care, maternal education and maternal smoking.

b. 'Others' included all other race/ethnicities and multi-race/ethnicities.

Table 5.7 Air pollution exposures distributions among births in Los Angeles County, California, 2017-2019, stratified by maternal race/ethnicity.

Adverse Birth Outcomes	Air Pollutants	Non-Hispanic White			Hispanic of any race			African American/ Black			Asian/ Pacific Islander			Others [a]		
		No. of Births	Mean	SD	No. of Births	Mean	SD	No. of Births	Mean	SD	No. of Births	Mean	SD	No. of Births	Mean	SD
PTB	$PM_{2.5-10}$ barium	7,105	30.7	7.4	9,512	32.4	6.4	1,986	33.1	6.3	3,087	29.2	7.6	935	32.2	6.9
	$PM_{2.5-10}$ zinc	7,108	7.0	1.3	9,514	7.4	1.1	1,986	7.4	1.1	3,087	6.9	1.4	935	7.3	1.2
	$PM_{2.5}$ mass	7,110	10.0	1.1	9,517	10.1	1.0	1,986	10.3	0.8	3,088	9.9	1.1	935	10.1	1.0
	Volume normalized $PM_{2.5}$ barium	7,159	17.1	4.0	9,618	17.6	3.9	2,007	17.7	3.5	3,091	16.5	3.7	946	17.9	4.1
	$PM_{2.5}$ mass normalized barium	7,159	1857.0	295.5	9,618	1849.3	299.1	2,007	1822.2	295.5	3,091	1824.6	278.8	946	1892.1	323.1
	Volume normalized $PM_{2.5}$ zinc	7,159	9.7	1.8	9,618	9.9	1.8	2,007	10.1	1.5	3,091	9.6	1.6	946	9.9	1.8
	$PM_{2.5}$ mass normalized Zinc	7,159	6.9	0.1	9,618	6.9	0.1	2,007	6.9	0.1	3,091	6.9	0.1	946	6.9	0.1
	$PM_{2.5}$ ROS	7,159	114.0	12.3	9,618	113.8	11.5	2,007	113.4	11.0	3,091	112.2	12.0	946	115.9	12.5
	Volume normalized $PM_{2.5}$ OH formation	7,159	5.5	1.2	9,618	5.7	1.2	2,007	5.8	1.1	3,091	5.3	1.2	946	5.7	1.2

	$PM_{2.5}$ mass normalized OH formation	7,159	0.6	0.1		9,618	0.6	0.1		2,007	0.6	0.1		3,091	0.6	0.1		946	0.6	0.1
	Volume normalized $PM_{2.5}$ DTT loss	7,159	607.9	116.8		9,618	619.6	115.7		2,007	624.8	106.3		3,091	593.5	112.6		946	622.1	120.3
	$PM_{2.5}$ mass normalized DTT loss	7,159	62.4	4.0		9,618	62.6	4.2		2,007	62.6	3.8		3,091	62.3	3.7		946	63.0	4.7
	Volume normalized $PM_{2.5}$ black carbon	7,159	0.4	0.1		9,618	0.4	0.1		2,007	0.4	0.1		3,091	0.4	0.1		946	0.4	0.1
	$PM_{2.5}$ mass normalized black carbon	7,159	4.8	0.4		9,618	4.8	0.4		2,007	4.8	0.3		3,091	4.8	0.4		946	4.8	0.4
TLBW	**$PM_{2.5-10}$ barium**	1,790	30.6	7.5		2,120	32.4	6.5		655	33.2	6.2		1,147	29.7	7.6		255	32.7	6.8
	$PM_{2.5-10}$ zinc	1,791	7.0	1.3		2,120	7.3	1.1		655	7.5	1.1		1,147	6.9	1.3		255	7.3	1.2
	$PM_{2.5}$ mass	1,791	9.9	1.1		2,120	10.1	1.0		655	10.3	0.8		1,147	9.8	1.2		255	10.2	1.0
	Volume normalized $PM_{2.5}$ barium	1,805	17.2	4.0		2,141	17.7	4.0		664	17.7	3.5		1,149	16.6	3.8		258	18.1	4.1
	$PM_{2.5}$ mass normalized barium	1,805	1873.2	300.1		2,141	1862.7	315.2		664	1809.5	279.5		1,149	1838.5	297.4		258	1901.9	324.6
	Volume normalized $PM_{2.5}$ zinc	1,805	9.7	1.8		2,141	9.9	1.8		664	10.1	1.5		1,149	9.6	1.6		258	10.0	1.9
	$PM_{2.5}$ mass normalized Zinc	1,805	6.9	0.1		2,141	6.9	0.1		664	6.9	0.1		1,149	6.9	0.1		258	6.9	0.1
	$PM_{2.5}$ ROS	1,805	114.5	12.5		2,141	114.3	12.1		664	113.1	10.6		1,149	112.7	12.4		258	116.1	13.3
	Volume normalized $PM_{2.5}$ OH formation	1,805	5.5	1.2		2,141	5.7	1.2		664	5.8	1.1		1,149	5.4	1.2		258	5.8	1.2

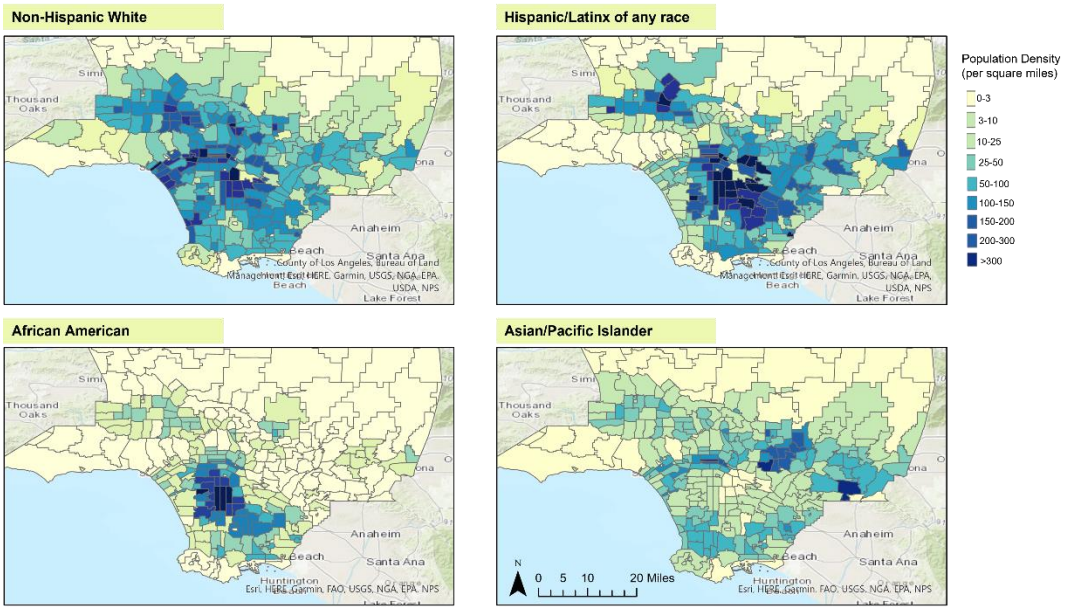
	$PM_{2.5}$ mass normalized OH formation	1,805	0.6	0.1		2,141	0.6	0.1		664	0.6	0.1		1,149	0.6	0.1		258	0.6	0.1
	Volume normalized $PM_{2.5}$ DTT loss	1,805	609.3	121.4		2,141	620.9	120.5		664	626.1	104.6		1,149	603.0	121.9		258	626.0	106.4
	$PM_{2.5}$ mass normalized DTT loss	1,805	62.5	4.2		2,141	62.7	4.4		664	62.6	3.6		1,149	62.5	4.2		258	62.7	3.8
	Volume normalized $PM_{2.5}$ black carbon	1,805	0.4	0.1		2,141	0.4	0.1		664	0.4	0.1		1,149	0.4	0.1		258	0.4	0.1
	$PM_{2.5}$ mass normalized black carbon	1,805	4.8	0.4		2,141	4.8	0.4		664	4.8	0.4		1,149	4.8	0.4		258	4.9	0.4
Term Normal Birth Weight	**$PM_{2.5-10}$ barium**	91,227	30.1	7.7		91,728	32.1	6.5		15,696	33.1	6.4		42,790	28.9	7.7		9,650	31.9	7.2
	$PM_{2.5-10}$ zinc	91,241	6.9	1.3		91,745	7.3	1.1		15,699	7.4	1.1		42,791	6.9	1.4		9,652	7.2	1.2
	$PM_{2.5}$ mass	91,251	9.9	1.1		91,760	10.1	1.0		15,701	10.3	0.8		42,794	9.9	1.2		9,654	10.0	1.0
	Volume normalized $PM_{2.5}$ barium	91,757	16.9	4.0		92,667	17.4	3.9		15,924	17.8	3.6		42,833	16.3	3.6		9,759	17.6	4.1
	$PM_{2.5}$ mass normalized barium	91,757	1861.8	300.9		92,667	1843.3	300.6		15,924	1841.8	306.2		42,833	1813.7	265.6		9,759	1889.8	325.1
	Volume normalized $PM_{2.5}$ zinc	91,757	9.6	1.8		92,667	9.8	1.7		15,924	10.0	1.5		42,833	9.6	1.6		9,759	9.8	1.8
	$PM_{2.5}$ mass normalized Zinc	91,757	6.9	0.1		92,667	6.9	0.1		15,924	6.9	0.1		42,833	6.9	0.1		9,759	6.9	0.1
	$PM_{2.5}$ ROS	91,757	114.0	12.8		92,667	113.4	11.6		15,924	114.0	11.4		42,833	111.6	11.8		9,759	115.4	12.8
	Volume normalized $PM_{2.5}$ OH formation	91,757	5.5	1.2		92,667	5.7	1.2		15,924	5.8	1.1		42,833	5.3	1.2		9,759	5.7	1.2

	$PM_{2.5}$ mass normalized OH formation	91,757	0.6	0.1		92,667	0.6	0.1		15,924	0.6	0.1		42,833	0.5	0.1		9,759	0.6	0.1
	Volume normalized $PM_{2.5}$ DTT loss	91,757	604.9	117.3		92,667	617.5	116.3		15,924	627.1	113.3		42,833	590.4	112.4		9,759	621.4	121.0
	$PM_{2.5}$ mass normalized DTT loss	91,757	62.4	4.0		92,667	62.5	4.2		15,924	62.8	4.1		42,833	62.2	3.5		9,759	62.8	4.5
	Volume normalized $PM_{2.5}$ black carbon	91,757	0.4	0.1		92,667	0.4	0.1		15,924	0.4	0.1		42,833	0.4	0.1		9,759	0.4	0.1
	$PM_{2.5}$ mass normalized black carbon	91,757	4.8	0.4		92,667	4.8	0.4		15,924	4.8	0.3		42,833	4.8	0.4		9,759	4.8	0.4

a. 'Others' included all other race/ethnicities and multi-race/ethnicities.

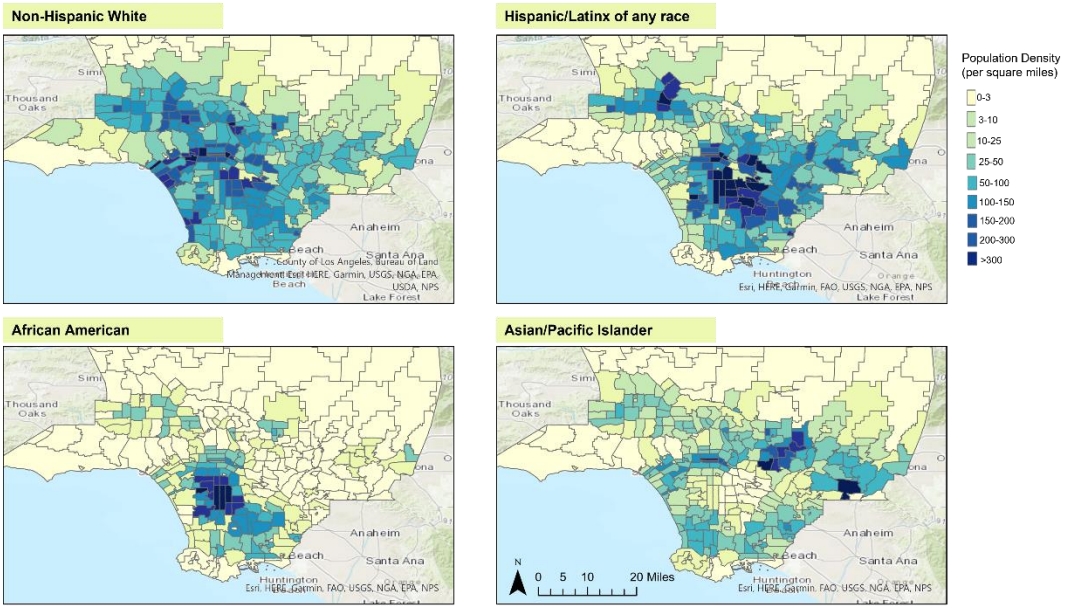
Figure 5.1. Distributions of maternal residential addresses by maternal race/ethnicity.

(a) PTB and term normal birth weight



(b)

TLBW and term normal birth weight



5.4 Discussion

Our results suggest associations of adverse birth outcomes (PTB and TLBW) with $PM_{2.5-10}$- and $PM_{2.5}$- metals from brake wear (barium) and tire wear (zinc) and $PM_{2.5}$-oxidative stress markers, and most of the effects were still seen after co-adjustment for our $PM_{2.5}$ mass exposure measure, suggesting that the $PM_{2.5-10}$- and $PM_{2.5}$-constituents and the oxidative potential of $PM_{2.5}$-metals and particulate mixtures not only vary spatially in a manner different from $PM_{2.5}$ mass but have adverse effects on birth outcomes independent from mass. For some speciated $PM_{2.5}$ exposures especially metals and black carbon effect estimates decreased or were null after adjusting for the $PM_{2.5}$ mass, which likely reflects the high correlation of these constituents with $PM_{2.5}$ mass. Importantly, the correlations between the oxidative potential markers and $PM_{2.5}$ mass were relatively weak and effect estimates did not change when we adjusted for the $PM_{2.5}$ mass suggesting that oxidative stress generation potential of particulate matter is one of the important attributes of particle mass but not the sole one.

In addition, we observed differences in effect estimates for metal components and oxidative stress markers when stratifying by maternal race/ethnicity, with the strongest effects estimated in Hispanic women who contributed the most births in these analyses and for most components and $PM_{2.5}$ mass the smallest effects in Non-Hispanic White women. Given that most exposure averages were similar for white non-Hispanic and Hispanic women this may suggest either greater susceptibility to the effects of these pollutants or higher exposures to outdoor air through housing related characteristics such as lack of air filtration or air conditioning or behaviors that expose Hispanic women more to the local air (such as not leaving the neighborhood for paid employment). For several $PM_{2.5}$ component exposures we estimated effects below the null with wide 95% confidence intervals in African American mothers, which might be due to the generally smaller sample size causing random variations and also the overall smaller variation in exposure levels for women of this ancestry as they mostly lived clustered in one area of South Central LA, i.e., even though they were exposed to the highest levels, the standard deviations of the exposures were smallest. This argument is further supported by the more strongly increased risks we estimated for TLBW and PTB in the small group of 'other' races that included multi-racial parents as they are living more dispersed throughout LA county and exposures vary spatially similar to Hispanics and White women. We would expect the African American women to have lower social support as documented in the social epidemiology literature by Geronimous (Geronimus 1992) with her weathering hypothesis, which posits that Black women experience racism, exposure to violence and less social support, which weathers them down and leads to worse birth outcomes. The higher baseline risk for adverse birth outcomes in African American women could make the detection of a relatively small effect from air pollution more challenging in this group.

Air pollution exposures have also been associated with pregnancy complications such as preeclampsia (Wu et al. 2009), which is a major contributor to preterm birth and are thought to have their origin largely in abnormal early placentation. Specifically, the impaired trophoblastic invasion would lead to maladaptation of uteroplacental arteries, resulting in reduced uteroplacental perfusion and maternal and fetal complications (Kaufmann et al. 2003). Furthermore, oxidative stress, which can cause or be a consequence of increased inflammation, is one of the main hypothesized mechanisms through which air pollution such as $PM_{2.5}$ may cause adverse pregnancy outcomes (Feng et al. 2016; Yan et al. 2019). It has been suggested that maternal air pollution exposure affects pregnancy by inducing oxidative stress and systemic inflammation, in turn causing suboptimal placentation or placental inflammation (Vadillo-Ortega et al. 2014). In addition, $PM_{2.5}$-metals have been reported to induce excessive production of ROS,

which leads to an increased level of oxidative stress and pro-inflammatory effects (Hamad et al. 2016).

This is the first study to assess the effects of speciated $PM_{2.5}$ constituents with oxidative stress potential and adverse birth outcomes. We investigated both effects of $PM_{2.5}$ metals from brake and tire wear as well as the oxidative potential of the entire particle mixture with the aim of identifying $PM_{2.5}$ sources and components with adverse effects on birth outcomes. We evaluated the metals and oxidative stress markers with both volume and mass normalized measures, assuming that concentrations normalized to $PM_{2.5}$ mass per air volume could be more strongly indicative of their toxicity in inhaled air than mass normalized measures that reflect toxicity only per mass weight. Furthermore, we also considered confounding by exhaust emissions from traffic for the speciated $PM_{2.5}$ exposures using an LUR model that predicts NO_2 exposures as markers of tailpipe emissions. The positive associations for metals and oxidative stress markers and PTB/TLBW remained when we added exhausted emission markers into our regression models. Finally, important to note is that using birth certificate data in our study also allowed us to eliminate the possibility of self-selection of participants and its related biases.

Our study has several limitations. We could only estimate air pollution exposures at home addresses provided on the birth certificate, and we are not able to assess time spent by mothers at work, in transit or at other residences prior to birth, which is a problem common to most air pollution health effects studies. In addition, we had no information on whether the mother moved during pregnancy, which could have substantially changed the exposure profiles for those who moved. This would likely cause exposure misclassification bias that most likely is nondifferential and, thus, biases the effect estimates towards the null. A previous study estimated that 9%-32% of all mothers move during pregnancy (Bell and Belanger 2012). Our model only predicted spatial exposures for the whole 4-year period and spatial differences for exposures during the winter or summer periods were the only temporal influences we could assess. Therefore, we were not able to assess exposures specific to each trimester of pregnancy or the actual pregnancy period, nor were we able to investigate a critical window for the effect of speciated $PM_{2.5}$ exposures on adverse birth outcomes. Moreover, the lack of the air pollution samplers or low-cost sensors in low-income communities might induce measurement error, which would bias the effect estimates toward the null. However, it is difficult to know how much this would influence our results or whether this might be the reason that in some subgroups such as African American mothers we estimated smaller effects than in others. Plus, the lower effect estimates for African American mothers could be explained by the clustered living in one area with less exposure variation as discussed earlier. Studies that examine absolute instead of relative risks should be conducted to better understand this issue. Furthermore, adverse pregnancy outcomes can be affected by live birth biases, i.e., a fetus has to survive to be born alive in order to contribute to TLBW or preterm births. It is conceivable that the mostly highly exposed fetuses who would have been born low weight or preterm if they had survived are in fact lost due to early spontaneous abortions or fetal deaths among the most vulnerable and highly exposed which may lead to a lower rate of TLBW or preterm births with higher exposure. Furthermore, studies examining absolute instead of relative risks should be conducted to better understand this issue. Finally, adverse pregnancy outcomes can be affected by live birth biases, i.e., a fetus has to survive to be born alive in order to contribute to TLBW or preterm births. It is conceivable that the mostly highly exposed fetuses who would have been born low weight or preterm if they had survived are in fact lost due to early spontaneous abortions or fetal deaths among the most vulnerable and highly exposed which may lead to a lower rate of TLBW or preterm births with higher exposure.

With the high correlations among the various elements, it is difficult to ascribe specific health effects to each element. In addition, disadvantaged communities typically face numerous burdens

from other pollutants. People living in high air pollution exposure areas also are more likely to have chronic health conditions that would make them more susceptible to the adverse effects of environmental stressors. For example, they are more likely to experience adverse social conditions that increase stress in the family. In the case of childhood asthma, for example, a significant interaction exists between traffic-related air pollution exposures such that children from households with higher household stress had much greater risk of developing asthma than those in household with relatively low stress (Shankardass et al. 2009). Known as the double-jeopardy hypothesis, whereby people living in disadvantaged communities are exposed to higher levels of environmental stressors and they have higher susceptibility to these exposures (Institute of Medicine (US) Committee on Environmental Justice 1999). Such double jeopardy may also affect our epidemiological results. Another example is the co-exposure to noise. We found that co-exposure to both traffic related air pollution and noise from aircraft increased the risk of adverse birth outcomes more than each exposure alone (manuscript under review). Combined with our finding on the positive association between oxidative stress potential and lower socioeconomic position, these findings cast a new light on this issue because people in disadvantaged communities face not only higher levels of several pollutants, but the pollutants that are emitted locally are also more toxic on a per unit basis. Moving forward we recommend that further research should be undertaken to understand social gradients in oxidative stress potential.

5.5 Conclusion

Our results indicated that an increase in the exposure to $PM_{2.5-10}$- and $PM_{2.5}$-metals from brake and tire wear and with higher oxidative potential is associated with higher risks of preterm birth and term low birth weight in Los Angeles County. Infants born to mothers with Hispanic, Asian/Pacific Islander, or multi-racial/mixed origin were at higher risk of preterm or term low birth weight when exposed to $PM_{2.5}$ metals with high oxidative potential. Thus, it is important to evaluate some $PM_{2.5}$ species and their oxidative potential when assessing health effects as these seem to exhibit effects independent of $PM_{2.5}$ mass and may affect subgroups of pregnant women differently dependent on their exposure levels and vulnerability.

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6 The association of heavy metals and oxidative stress with adverse placental outcomes in the Los Angeles birth cohort

6.1 Introduction

From conception to birth, a well-functioning placenta, the organ responsible for the transfer of gases, nutrients, and waste between mother and developing child, is of utmost importance for the health of the pregnant woman, the development and survival of the fetus, and the long-term health of the child. Placenta size has been reported to affect the delivery of oxygen and other nutrients, where studies have shown that low placenta weight is associated with adverse birth outcomes, such as respiratory distress syndrome, neurologic abnormalities, and neonatal death (Hutcheon et al. 2012; Naeye 1987). Consequently, understanding relationships between placental weight and environmental exposures may guide in the prevention of such adverse events.

Pregnancy complications, which include placental complications, are common, occurring in around 30% of all births (Rich-Edwards et al. 2014). Environmental causes behind placental dysfunction and placenta related pregnancy complications are not well understood, but there is evidence that pollutants found in brake and tire wear can (1) cross the blood-placenta barrier and (2) affect the health and development of the newborn after exposure during pregnancy (Bové et al. 2019; Zhang et al. 2018). Prior research has found air pollution, specifically $PM_{2.5}$, to be associated with placental alterations via oxidative stress (Saenen et al. 2019). Metals found in the placenta have been associated with lower placental weight and placental insufficiency (Punshon et al. 2019). The relationship between measured or modeled $PM_{2.5}$ exposures from different sources and particularly speciated $PM_{2.5}$ from brake and tire wear have received relatively little attention to date, and how they affect birth outcomes - possibly through the involvement of the placenta - is not well known.

The following chapter aims to address the some of these gaps in knowledge by applying the exposure estimates of brake and tire wear-related metals and oxidative stress markers from chapter 4 to a study population of 161 mothers and their offspring who live in the study area depicted in chapter 4 and for whom we collected extensive data on pregnancy risk factors and placental function during pregnancy and at the time of birth. This “Imaging Innovations for Placental Assessment in Response to ENvironmenTal pollution study’ (PARENTs) study is a pregnancy cohort study designed to develop novel blood flow-based assessments via MRI-imaging of the placenta to investigate placental function and in turn the relationship of these measures with air pollution exposures measured and modeled at the women’s residences during pregnancy. Here, however, we will focus only on well-established measures of placental health and function for our analyses and explore the contributions of novel exposure surfaces we generated with the funding from CARB.

6.2 Methods

6.2.1 PARENTs study interviews, medical outcome data, and biosample collection

The PARENTs study surveyed pregnant women who sought care and planned to deliver at UCLA hospitals multiple times in pregnancy. Women were recruited if they had a viable singleton gestation, with gestational age (GA) confirmed by study staff. Exclusion criteria were maternal age <18 years, fetal malformation evident before enrollment, known fetal chromosomal abnormality, twin pregnancy, plan to terminate the pregnancy, or inability to provide consent.

These women after providing consent, were surveyed to collect information on demographics, medical history, tobacco use, and other factors. At delivery, the placenta, membranes, and umbilical cord were collected by study staff and newborn information including birth weight was retrieved from medical charts.

The following study assessed two placental outcomes: placental weight and the presence of ischemic placental disease (IPD). Shortly after birth, we collected the mother's placenta. Placental weight Placental weight was obtained via gross assessment of the placental trimmed of membranes and cord. IPD in the mother, assessed using survey data, clinical measurements, and chart abstractions, was defined as one or more of the following outcomes: placental abruption, hypertensive disease of pregnancy (preeclampsia and gestational hypertension), fetal growth restriction (FGR) or a newborn of small-for-gestational age (SGA)

6.2.2 Exposure Estimates

Maternal addresses during the first trimester were recorded during survey collection. We subsequently geocoded all addresses using the Countywide Address Management System (CAMS) locator (ESRI, Redlands, CA), followed by the "Geocode by Awesome Table" tool in Google Sheets for addresses unable to be geocoded by CAMS. In total, we have records on 161 women with geocoded addresses.

Generation of exposure estimates for heavy metal $PM_{2.5}$ constituents and oxidative stress marker were previously detailed in chapter 4. Briefly, gravimetric samplers collected particulate matter on filters across Los Angeles in the summer of 2019 and winter of 2020. We collected 50 samples at 46 different locations, which included some of the homes of PARENTs subjects as well as government regulatory monitoring sites.

Filters were processed in-house at UCLA for oxidative stress markers and at the Wisconsin State Hygiene Laboratory for speciated particulates. After obtaining measurements at each site, we constructed land-use regression models as external drift in a co-kriging model in conjunction with the PurpleAir network, as detailed in chapter 4. After annual and seasonal estimates of each outcome were generated throughout the study area, we assigned exposures to the patient cohort according to their residential address in the first trimester of pregnancy. Here we will rely on only first trimester residences to generate the exposure estimates as this is the most relevant pregnancy for placenta formation and development.

6.2.3 Covariates

Covariates to be entered into our regression models as potential confounders were selected a priori per existing literature on risk factors for adverse birth outcomes and the data available to us. We selected as potential confounders the following covariates: maternal age (continuous), race/ethnicity (Non-Hispanic White or other), fetal sex (male or female), parity (no parity or parity of 1 or more), maternal smoking (never smoker or former smoker), and maternal education (High school or 2-year vocational degree, Bachelor's degree, or advanced degree), and maternal BMI (continuous).

6.2.4 Statistical Analysis

Here we will focus on two endpoints of interest: ischemic placental disease (0 = no, 1 = yes), as a dichotomous variable, and on placental weight in grams. For both outcomes, we present three regression models: a crude/unadjusted model, a minimally adjusted model adjusting for maternal age, race/ethnicity, and fetal sex, and a fully adjusted model which also included parity, maternal smoking, and maternal education.

To estimate the association between pollutants of interest and log odds of ischemic placental disease, we used unconditional logistic regression and for pollutants of interest and placental

weight – a continuous measure - we used multiple linear regression. All exposures were scaled by the inter-quartile range to ensure comparability across different exposures.

6.3 Results and Discussion

In Table 6.1 we present the basic demographic characteristics of our study population, stratified by ischemic placental disease status. Of the original 161 patients with geocoded address data, two subjects were missing data on fetal sex, one was missing data on maternal education, and three subjects were excluded for either NA or current smoking status. With regards to exposure data, six subjects' residences were in areas outside of the exposure modelling area, resulting in their exclusion. Another 13 subjects were missing data on the second outcome, placental weight, resulting in their exclusion from regressions concerning placental weight. Our study population drew mainly form UCLA employees and students and thus it is not surprising that most of the women had high educational attainment, with 141 mothers, or 87.6% of our study population, having a bachelor's degree or higher. A plurality of subjects were first-time mothers, and most subjects were never smokers.

Analysis of location data and exposure prediction variance reveals that although controls were distributed more widely throughout the study area compared to cases (Figure 6.1), there was no statistically significant difference in mean prediction variance. Additionally, we find that the statistical distribution of exposure prediction variances remains the same between cases and controls for each outcome.

Median and IQR values for all exposures stratified by IPD status are summarized in Table 6.2. Although median exposure estimates differed between summer and winter, overall, the IQRs remained for the most part unchanged. For most outcomes, median exposures to outcomes were either very similar between cases and controls or slightly higher among cases. $PM_{2.5}$ mass exposure estimates had a median of 10.16 ug/m^3, slightly lower than the 2019 average of 12.7 ug/m^3 as reported by IQAir and was most similar to the September 2019 mean (10.3 ug/m^3), and lower than the February 2020 mean (13 ug/m^3) the months during which we deployed our samplers across the LA basin (IQAir 2021). This may be because gravimetric sampling periods avoided the 2019 wildfire season and occurred prior to the COVID-19 pandemic, avoiding two significant events that worsened and improved air quality in the region, respectively.

Table 6.1: Study population descriptive characteristics.

	Ischemic Placental Disease		
	No (N=115)	Yes (N=40)	Overall (N=155)
Age			
Median [Min, Max]	33.0 [19.0, 41.0]	34.5 [25.0, 49.0]	34.0 [19.0, 49.0]
Fetal Sex			
Female	64 (55.7%)	18 (45.0%)	82 (52.9%)
Male	51 (44.3%)	22 (55.0%)	73 (47.1%)
Race/Ethnicity			
Non-White	63 (54.8%)	19 (47.5%)	82 (52.9%)
White, non-Hispanic	52 (45.2%)	21 (52.5%)	73 (47.1%)
Parity			
No parity	53 (46.1%)	22 (55.0%)	75 (48.4%)
1 or more	62 (53.9%)	18 (45.0%)	80 (51.6%)
Maternal Smoking			
Never Smoker	93 (80.9%)	28 (70.0%)	121 (78.1%)
Former Smoker	22 (19.1%)	12 (30.0%)	34 (21.9%)
Education			
High school graduate, some college, or 2-year degree	8 (7.0%)	1 (2.5%)	9 (5.8%)
Bachelor’s Degree	50 (43.5%)	11 (27.5%)	61 (39.4%)
Advanced Degree	57 (49.6%)	28 (70.0%)	85 (54.8%)
Maternal BMI			
Median [Min, Max]	23.5 [17.3, 36.0]	24.1 [18.8, 36.9]	23.7 [17.3, 36.9]

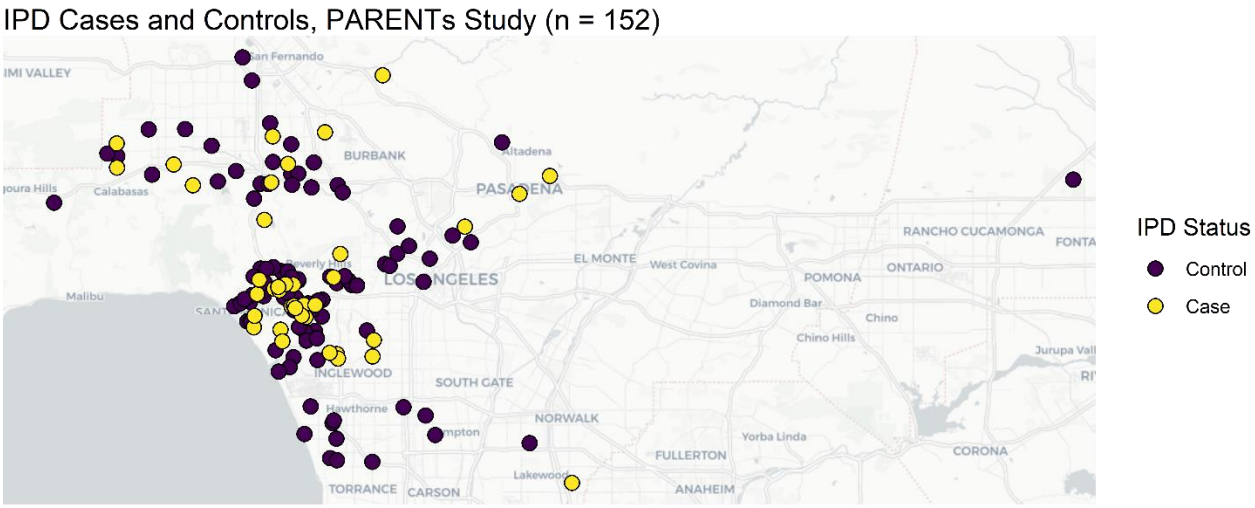

Figure 6.1: Map of study participants in the study area, separated by IPD status

Table 6.2: Median and IQR of exposure estimates among the study population, divided by IPD status.

Outcome	Median (IQR), Controls	Median (IQR), Cases
Barium during summer	12.81 (3.94)	13.41 (5.4)
Barium during winter	22.26 (3.94)	22.86 (5.39)
Barium annual	17.54 (3.94)	18.13 (5.39)
Barium mass normalized	1968.9 (352.84)	2039.94 (463.8)
Zinc during summer	7.34 (2.01)	7.54 (1.84)
Zinc during winter	12.23 (2.01)	12.44 (1.84)
Zinc annual	9.78 (2.01)	9.99 (1.84)
Zinc mass normalized	6.97 (0.17)	6.96 (0.12)
ROS during summer	108.81 (15.54)	110.28 (18.44)
ROS during winter	129.79 (15.55)	131.26 (18.44)
ROS annual	119.3 (15.55)	120.77 (18.44)
DTT loss annual	648.07 (118.41)	663.25 (103.52)
DTT loss mass normalized	61.62 (2.85)	61.62 (4.19)
OH formation annual	5.97 (1.41)	6.13 (1.17)
OH formation mass normalized	0.54 (0.09)	0.55 (0.11)
Black carbon annual	0.45 (0.08)	0.46 (0.11)
Black carbon mass normalized	4.95 (0.36)	4.93 (0.4)
PM25 annual	10.17 (1.13)	10.14 (0.88)

6.3.1 Association between $PM_{2.5}$ constituent exposures and adverse birth outcomes

Odds ratios associated with each exposure for the outcome ischemic placental disease are shown in Table 6.3. Overall, for none of our exposure measures (summer, winter, or annual estimates), did the 95% confidence limits exclude the null value of 1 i.e. none were formally statistically significantly associated with ischemic placental disease. However, for most outcomes, the point estimates were positive, with the highest point estimate being the one for mass normalized OH radical formation. With regards to placental weight as the outcome, presented in Table 6.4, we again find that none of our point estimates is formally statistically significant. Similarly, however, with a few exceptions, most point estimates were negative suggesting a reduction in placental weight with exposures and the mass normalized zinc exposure being most strongly negatively associated with placental weight overall. Thus, even though our current sample size does not allow us to form strong conclusions about the influence of these exposures on placental function related adverse birth outcomes and placental weight the direction of the estimated effects is quite consistent with the results we presented in chapter 5 for preterm births and TLBW, where higher levels of $PM_{2.5}$ components and oxidative stress producing particles were associated with higher odds of these adverse birth outcome.

6.4 Conclusions

In this chapter, we have used a novel method of exposure assessment to analyze the relationship between heavy metals and oxidative potential and placental outcomes in a birth cohort. Unlike chapter 5, this study is not intended as a generalizable study. Rather, it aims to adjust for covariates otherwise unavailable in birth record data, such as maternal smoking and BMI and target more specific, clinically verified outcomes. Though limited by a small sample size, we found similar point estimates after adjusting for confounding. Nonetheless, sample size challenges in the study population resulted in an underpowered study, where none of our point estimates are estimated with the necessary statistical precision especially the adjusted analyses. The PARENTs cohort is drawn from a group of pregnant women that is more educated than women in the general population. Though we find positive associations, the nonrepresentative nature of our sample may bias our result in either direction. On one hand, our subjects tend to be of higher SES and have higher educational attainment compared to the general population, granting a potentially protective effect. On the other, our subjects specifically seek specialized care at UCLA, indicating somewhat higher risk of abnormal pregnancy. Nonetheless, despite our limitations, our exploratory results suggest the possibility of a link between brake and tire wear exposure and adverse placental outcomes. Based on uniformity we observed for the direction of the point estimates, further research that recruits a larger cohort is recommended to follow-up on these novel results.

Table 6.3: Odds ratios (95% confidence interval) from unconditional logistic regression models for $PM_{2.5}$ concentration, black carbon, $PM_{2.5}$ constituents, and oxidative stress markers scaled by IQR and ischemic placental disease (N=150).

Outcome	Unit	Crude	Adjusted[a]	Adjusted[b]
$PM_{2.5}$ Concentration	ug/m^3	0.94 (0.6, 1.5)	0.98 (0.62, 1.6)	0.86 (0.52, 1.4)
Barium during summer	ng/m^3	1.2 (0.8, 1.9)	1.4 (0.87, 2.2)	1.3 (0.8, 2.2)
Barium during winter	ng/m^3	1.2 (0.8, 1.9)	1.4 (0.87, 2.2)	1.3 (0.8, 2.2)
Barium annual	ng/m^3	1.2 (0.8, 1.9)	1.4 (0.87, 2.2)	1.3 (0.8, 2.2)
Barium mass normalized	ng/g	1.3 (0.85, 2.1)	1.5 (0.96, 2.5)	1.5 (0.9, 2.6)
Zinc during summer	ng/m^3	1 (0.63, 1.7)	1.2 (0.7, 2)	1 (0.59, 1.8)
Zinc during winter	ng/m^3	1 (0.63, 1.7)	1.2 (0.7, 2)	1 (0.59, 1.8)
Zinc annual	ng/m^3	1 (0.63, 1.7)	1.2 (0.7, 2)	1 (0.59, 1.8)
Zinc mass normalized	ng/g	0.92 (0.54, 1.6)	1 (0.58, 1.8)	0.84 (0.45, 1.6)
ROS during summer	nmol/L	1.2 (0.75, 2)	1.5 (0.86, 2.5)	1.4 (0.77, 2.5)
ROS during winter	nmol/L	1.2 (0.75, 2)	1.5 (0.86, 2.5)	1.4 (0.77, 2.5)
ROS annual	nmol/L	1.2 (0.75, 2)	1.5 (0.86, 2.5)	1.4 (0.77, 2.5)
DTT loss annual	uM/min/L	1 (0.73, 1.5)	1.1 (0.76, 1.5)	1 (0.71, 1.5)
DTT loss mass normalized	uM/min/g	0.95 (0.73, 1.2)	0.95 (0.72, 1.2)	0.9 (0.67, 1.2)
OH formation annual	uM/min/L	1.1 (0.74, 1.7)	1.2 (0.8, 1.9)	1.2 (0.73, 2)
OH formation mass normalized	uM/min/g	1.3 (0.85, 2)	1.5 (0.92, 2.3)	1.6 (0.98, 2.7)
Black carbon annual	ug/m^3	0.9 (0.6, 1.3)	0.89 (0.59, 1.3)	0.81 (0.53, 1.2)
Black carbon mass normalized	ug/g	0.91 (0.6, 1.4)	0.9 (0.59, 1.4)	0.81 (0.52, 1.3)

[a] Minimally adjusted for maternal age, fetal sex, and race/ethnicity

[b] Additionally adjusted for parity, maternal smoking, maternal education, and maternal BMI.

Table 6.4: Coefficient estimates (95% confidence interval) from multiple linear regression models assessing the relationship between $PM_{2.5}$ concentration, $PM_{2.5}$ constituents, and oxidative stress markers all scaled by IQR and placental weight in grams. (N=139)

Outcome	Unit	Crude	Adjusted[a]	Adjusted[b]
$PM_{2.5}$ Concentration	ug/m^3	-2.4 (-21, 16)	-3.2 (-22, 15)	5.7 (-12, 23)
Barium during summer	ng/m^3	-5 (-22, 12)	-6.7 (-24, 11)	1 (-15, 17)
Barium during winter	ng/m^3	-5 (-22, 12)	-6.7 (-24, 11)	1 (-15, 17)
Barium annual	ng/m^3	-5 (-22, 12)	-6.7 (-24, 11)	1 (-15, 17)
Barium mass normalized	ng/g	-6.8 (-25, 12)	-9.2 (-28, 9.6)	-0.82 (-18, 17)
Zinc during summer	ng/m^3	-9.2 (-29, 11)	-14 (-35, 6.1)	-3.1 (-22, 16)
Zinc during winter	ng/m^3	-9.2 (-29, 11)	-14 (-35, 6.1)	-3.1 (-22, 16)
Zinc annual	ng/m^3	-9.2 (-29, 11)	-14 (-35, 6.1)	-3.1 (-22, 16)
Zinc mass normalized	ng/g	-6.5 (-28, 15)	-12 (-35, 11)	1.3 (-20, 23)
ROS during summer	nmol/L	-9.3 (-30, 11)	-14 (-35, 7.5)	-2.5 (-22, 17)
ROS during winter	nmol/L	-9.3 (-30, 11)	-14 (-35, 7.5)	-2.5 (-22, 17)
ROS annual	nmol/L	-9.3 (-30, 11)	-14 (-35, 7.5)	-2.5 (-22, 17)
DTT loss annual	uM/min/L	-2.3 (-17, 12)	-1.6 (-16, 13)	6 (-7.3, 19)
DTT loss mass normalized	uM/min/g	0.95 (-9.3, 11)	1.4 (-8.8, 12)	4.6 (-5, 14)
OH formation annual	uM/min/L	-2.2 (-19, 15)	-2.7 (-20, 14)	8.2 (-7.7, 24)
OH formation mass normalized	uM/min/g	1.6 (-17, 21)	2.4 (-17, 22)	2.7 (-15, 21)
Black carbon annual	ug/m^3	2.4 (-14, 19)	1.4 (-15, 18)	7.6 (-7.6, 23)
Black carbon mass normalized	ug/g	0.87 (-16, 18)	-0.31 (-18, 17)	5.4 (-10, 21)

[a] Minimally adjusted for maternal age, fetal sex, and race/ethnicity

[b] Additionally adjusted for parity, maternal smoking, and maternal education and maternal BMI.

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6.6 Discussion, Synthesis and Conclusions

Our literature review was presented in Chapter 1. In this review, we aimed to provide a comprehensive understanding of vehicular non-exhaust emissions. We found that while the relative contribution of non-exhaust emission sources to overall traffic-related $PM_{2.5}$ and PM_{10} emissions has been continuously increasing in recent years, large uncertainties exist in the reported non-exhaust emission factors. The reported brake and tire wear mass-based particle size distributions have been shown to be primarily unimodal, while multimodal particle number distributions with mode diameters in the fine and ultrafine size ranges have been documented in the previous studies. We found that the elements associated with brake and tire wear particles, including Ba, Cr, Cu, Fe, Mn, Sb, Sn, Zn, and Zr have been linked to increased OSP and various health outcomes, including cancer, reduced lung function, birth defects, and cardiovascular diseases. Moreover, we found the organic content of the brake and tire wear includes toxic compounds such as PAHs, phenolic compounds, and glycerol compounds that can contribute to overall PM health effects. The effects of auto electrification on non-exhaust emission sources in the future were also discussed in Chapter 1. Electric vehicle (EV) batteries have been introduced as one of the potential non-exhaust sources, whose emissions have been rarely investigated in previous studies.

In Chapter 2, we did an extensive analysis on the spatial variation of the elemental concentrations of the collected fine and coarse particles at various sampling locations, including urban traffic, urban community, urban background, and desert sites. The concentrations of traffic-related elements, including Ba, Cu, Mo, Sb, Zr, Pd, and Zn were 3 to 5 times higher at the urban traffic sites than urban background locations. In addition, our results also showed a decrease in Ni and V in $PM_{2.5}$ compared to the previously reported levels in the region, presumably due to the regulations on fuel oil sulfur content.

Principal component analysis was used to extract the source factors that explained the variance in the $PM_{2.5}$ and $PM_{2.5-10}$ elemental concentrations. Based on multiple linear regression analysis, the contribution of traffic emissions (27%) to $PM_{2.5}$ was found to be highest, followed by mineral dust (23%), marine aerosol (18%), and industrial emissions (8%). Mineral dust was the dominant source of $PM_{2.5-10}$ with a 45% contribution, followed by marine aerosol (22%), and traffic emissions (19%).

In Chapter 3, we presented our findings from the OSP analysis. We successfully conducted two assays to assess the oxidative stress potential of the particles: hydroxyl radical production (OPOH) and dithiothreitol consumption (OPDTT). Both OSP assays showed more spatial variability among the monitoring sites than did particle mass. We used a Positive Matrix Factorization (PMF) source apportionment model analysis to identify sources contributing to oxidative potential. We found that tailpipe emissions and brake and tire wear were the two largest contributors of OPOH. Sources contributing to OPDTT were similar, but the results were less stable, potentially due to a lack of measurements of organic species, which may play a substantial role in the DTT assay.

Additionally, we measured the OPOH and OPDTT activity of the metals most strongly correlated to the field measurements of OPOH and OPDTT individually and in combination in the laboratory. We found a strong association between OSP and brake and tire wear tracers. This finding suggests a role for non-tailpipe emissions contribute to aerosol OSP.

Finally, we explored relationships between the OSP assays and several socioeconomic factors, exposure estimates, and adverse health outcome indicators from the CalEnviroScreen developed by the California Office of Environmental Health Hazards Assessment. Generally, we found

positive correlations between measures of social disadvantage and oxidative stress potential of the particles, indicating that people living in more disadvantaged neighborhoods were exposed to particles with higher OSP, not only because particle mass concentrations were higher, but also because the particles themselves had higher intrinsic toxicity. There also were positive correlations between OPOH and all three health outcome indicators (adverse birth outcomes, cardiovascular events and asthma) in CalEnviroScreen, and between OPDTT and birth outcomes aggregated to the census tract. These results suggest a useful role for the OSP assays, particularly the OH assay, in predicting adverse health outcomes, which following this exploratory ecological analysis were assessed with individual birth record data and with measures of placental health in an ongoing cohort study as described below.

Methods and results of our exposure modeling approach were presented in Chapter 4. We successfully fit land use regression (LUR) models for 6 elements, 1 measure of reactive oxygen species (ROS) derived from a toxicokinetic model, and the two assays of OSP. Model performance as measured by leave-out cross-validation R^2 ranged from 0.37-0.68. To improve the model prediction, we also fit co-kriging models that used the filter-based particle elements or oxidative stress markers as the gold-standard measurement along with 294 Purple Air measurements taken from a publicly-available website. Co-kriging model predictions outperformed the LUR models. Predictive surfaces for all models were developed at 30 m resolution. We also fit two models of $PM_{2.5-10}$ with land use regression. Results showed reasonably good model fits. All exposure models were then assigned to the two health data sets described below for epidemiological analyses. PTB and/or TLBW

We then modeled health outcomes relying on the exposure predictions from Chapter 4; the results are presented in Chapter 5 for adverse birth outcomes documented on State of California birth records. We first geocoded the mother's address at the time of the birth provided on the birth record and then modeled exposure associations with two health outcomes: pre-term birth (PTB) and term low birthweight (TLBW). We also included NO_2 predictions from a land use regression model supplied to us by Dr. Jason Su of UC Berkeley. These exposures were developed in another CARB-funded study. Overall, associations between most of the metals and oxidative stress potential estimates and health outcomes were positive and statistically significant. Effect sizes were small for the most part based on an IQR exposure increment. We also examined concentration-response relationships by breaking the exposures into quartiles. Most of these analyses suggested a monotonic dose-response, with higher exposures related to higher risks, although the ROS model suggested some potentially non-linear associations. We also estimated health effects from the coarse fraction $PM_{2.5-10}$ for Ba and Zn, with the results being similar to the same elements of the $PM_{2.5}$ mass effects for those same elements. We also observed positive associations for outdoor land-use regression derived NO_2 concentrations and both indicators of birth outcomes.

We also analyzed two pollutant models. This involved adding either NO_2 or $PM_{2.5}$ to an epidemiology model with one of the metals or OSP estimates. The addition of NO_2 suggested very little confounding as the parameter estimates for the metal species or ROS potential changed only minimally. In contrast, when we included $PM_{2.5}$ mass into the regression models, we observed an attenuation of the estimated effect sizes for the metal and oxidative stress potential markers, probably due to the relatively high correlation between $PM_{2.5}$ mass and its elements.

For placental-based health outcomes, we were hampered by problems due to data and sample collection, which were beyond our control, as the study was funded by NIH and the fieldwork was under the control of other collaborators. Unfortunately, this resulted in a smaller sample size (N = 161) than we initially anticipated (the study was expected to enroll 300 pregnant women). With this sample of 161, we conducted unconditional logistic regression analyses for IPD and linear

regression for placental weight. Due to the small sample size, we were underpowered to detect formally statistically significant associations, but we observed mostly positive associations between the exposures and IPD. We also observed associations between increased exposure and lower placental weight, but these also failed to reach conventional significance levels. Thus, the overall pattern of associations suggested that these air pollutant markers were associated with IPD and lower placental weight. Associations with IPD were strongest and borderline significant for mass normalized OH formation.

6.7 Limitations

This study suffered from two major shortcomings, which resulted from limited budget and reliance on health data collected by other collaborators funded from another grant. First, due to budgetary constraints, we were unable to collect sufficient PM samples to support the small-area exposure modeling. This may have resulted in sub-optimal model performance with a tendency to over-smooth the exposure surfaces. We compensated for this limitation by using auxiliary land use and traffic data in our land use regression models and by fitting co-kriging models that augmented our sparse monitoring with data from the PurpleAir network, but it remains possible that measurement error in the exposures surfaces might have attenuated effects toward the null. Second, due to problems with data collection on the placental outcomes resulting in a diminished sample size, these analyses lacked statistical power to detect significant associations for all exposures assessed. However, we were able to see the overall patterns of positive association for this small cohort, and the results agreed with those from the large LA birth cohort.

6.8 Policy Implications

Our findings have several policy implications. With the climate imperative of reducing CO_2 emissions, one strategy is to simultaneously increase the renewable component of the energy mix while also shifting driving toward electric and hydrogen vehicles. If fleet composition continues to shift toward vehicles with no tailpipe emissions, it is likely that transportation-related particle emissions will decline, but the remaining particles may have higher oxidative stress potential due to the relatively larger contribution from brake and tire wear. In such an instance, health effects from traffic-related air pollution may not decline as much as would be expected if based solely on particle mass reductions. Other measures to reduce vehicle miles travelled through changes to urban design that promote walking and biking as well as improvements to public transit may also be needed to reduce emissions from brake and tire wear. In addition, technology-forcing regulations may also be required to minimize emissions from brake and tire wear. Measures such as particle traps around braking systems might be needed. Regulations could also be put in place to change the matrix of materials in brakes and tires so the emitted particles become less toxic. On monitoring, given the high cost of assessing elemental compositions with gravimetric filters, other modeling approaches that increase exposure assessment precision, such as the co-kriging methods developed here will likely be needed. This approach requires widespread data support from low costs sensors such as the Purple Air monitors. Although we did not quantitatively assess the spatial patterns of Purple Air monitors, our visual inspection revealed that many disadvantaged areas have either no or much less dense monitoring than those areas with more economic resources. State and local air agencies may have to augment these low-cost sensor networks with monitors to increase the density of sensors. This will be essential for developing exposure models that predict accurately in all areas, regardless of socioeconomic position.

6.9 Conclusions

Using the PMF model with markers of oxidative stress potential revealed exhaust and non-exhaust sources were contributing to aerosol oxidative potential separately, with both sources being important contributors. Linking the oxidative potential data with CalEnviroScreen indicators suggested positive correlations between measures of social disadvantage and OP of particles. The OP analyses also suggested that there were positive correlations with other adverse exposure and several health outcome indicators included in the CalEnviroScreen, although we caution that due to the ecological nature of the health outcome data, these findings are more exploratory than definitive.

Although we had limited monitoring data support to fit LUR and co-kriging models, we were able to fit models that performed well overall, with a few exceptions. We were also able to integrate for the first time information from the crowd-sourced PurpleAir network to our models and this improved model fit significantly. In our visual assessment of the locations of the Purple Air monitors, there appeared to be fewer samples in socially disadvantaged neighborhoods. This could have introduced differential exposure measurement error, such that exposures were predicted less accurately in these disadvantaged neighborhoods than in the more advantaged neighborhoods. This might have attenuated effects of the particles from some groups living in these disadvantaged neighborhoods. Going forward we recommend that government agencies may wish to intervene to ensure equitable coverage of the montiors among all social groups. This would also assist with developing better exposure estimates for all social groups, which would likely reduce biases in subsequent epidemiological analysis.

Results from the epidemiological analyses of birth outcomes and our various exposure measures found associations of metals and OSP markers, both in fine and coarse mode sizes, with TLBW and PTBs. Effects sizes estimates for particle components were similar to those for $PM_{2.5}$ mass modeled with input from the PurpleAir monitors. Importantly, given its widespread use in epidemiological studies, we note that NO_2 seemed to exert little or no confounding bias on the estimates for metal species or markers of OSP. In contrast, when including $PM_{2.5}$ mass, we observed attenuation in the metal and oxidative stress potential markers estimated effect sizes. We attribute this attenuation to the relatively high correlation between $PM_{2.5}$ mass and its elements or oxidative stress potential markers, likely due to the similar model structure and data inputs used to model both undifferentiated $PM_{2.5}$ mass and speciated $PM_{2.5}$.

Our analysis of IPD and placental weight, while underpowered, revealed consistent patterns of association with all estimated coefficients having the expected sign. We interpreted this result as suggestive of an effect of metals and OSP markers on IPD and placental weight.

Abbreviations

PM, particulate matter; EV, electric vehicle; LDV, light-duty vehicle; HDV, heavy-duty vehicle; NAO, non-asbestos organic; NAEI, National Atmospheric Emission Inventory; EEA, European Environment Agency; USEPA, United States Environmental Protection Agency; TSP, total suspended particles; TRWP, tire-road wear particle; PAH, Polycyclic aromatic hydrocarbon; AADT, annual average daily traffic; s/n, signal-to-noise ratio; PCA, Principal component analysis; CV, coefficient of variation; EF, crustal enrichment factor; RE, roadside enrichment; OP, oxidative potential; DTT, dithiothreitol; DTNB, dithiobisnitrobenzoic acid; TNB, 2-nitro-5-thiobenzoic acid; OP^{OH}, oxidative potential measured by the production of hydroxyl radicals; OP^{DTT}, oxidative potential measured by depletion of dithiothreitol; OP_v^{OH}, volume-normalized OP^{OH}; OP_v^{DTT}, volume-normalized OP^{DTT}; OP_m^{OH}, mass-normalized OP^{OH}; OP_m^{DTT}, mass-normalized OP^{DTT}; BC, black carbon; SLF, surrogate lung fluid.