



Variability of sources
of ambient fine
particular matter

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Spatial and temporal variability of sources of ambient fine particular matter (PM_{2.5}) in California

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Abstract

To identify major sources of ambient fine particulate matter ($PM_{2.5}$, $dp < 2.5\mu m$) and quantify their contributions in the state of California, positive matrix factorization (PMF) receptor model was applied on speciation trends network (STN) data, collected between 2002 and 2007 at 8 distinct sampling locations, including El Cajon, Rubidoux, Los Angeles, Simi Valley, Bakersfield, Fresno, San Jose, and Sacramento. Between five to nine sources of fine PM were identified at each sampling site, several of which were common among multiple locations. Secondary aerosols, including secondary ammonium nitrate and ammonium sulfate, were the most abundant contributor to ambient $PM_{2.5}$ at all sampling sites, except for San Jose, with an annual average cumulative contribution of 26 to 63 %, across the state. On an annual average basis, vehicular emissions (including both diesel and gasoline vehicles) were the largest primary source of fine PM at all sampling sites in southern California (17–18 % of total mass), whereas in Fresno and San Jose, biomass burning was the most dominant primary contributor to ambient $PM_{2.5}$ (27 and 35 % of total mass, respectively), in general agreement with the results of previous source apportionment studies in California. In Bakersfield and Sacramento, vehicular emissions and biomass burning displayed relatively equal annual contributions to ambient $PM_{2.5}$ mass (12 and 25 %, respectively). Other commonly identified sources at all sites included aged and fresh sea salt as well as soil, which contributed to 0.5–13 %, 2–27 %, and 1–19 % of the total mass, respectively, across all sites and seasons. In addition, few minor sources were exclusively identified at some of the sites (e.g. chlorine sources, sulfate-bearing road dust, and different types of industrial emissions). These sources overall accounted for a small fraction of the total PM mass across the sampling locations (1 to 15 %, on an annual average basis).

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1 Introduction

Exposure to ambient airborne particulate matter (PM) is one of the leading causes of morbidity and mortality, contributing to more than 3 million premature deaths in the world annually, based on a recent global burden of disease study (Lim et al., 2013). PM inhalation has been linked to a wide range of adverse health effects, such as respiratory inflammation (Araujo et al., 2008), cardiovascular diseases (Delfino et al., 2005; Ostro et al., 2014), and most recently neurodegenerative and neurodevelopmental disorders (Davis et al., 2013a, b). During the past few decades, California has been constantly suffering from high concentrations of ambient PM, among the highest levels recorded within the United States, with estimated rates of PM-related morbidity and mortality exceeding any other state in the country (Fann et al., 2012).

Ambient PM in California originates from a large number of diverse sources (Hu et al., 2014) and is a complex mixture of different chemical components, the composition of which may change drastically with PM size (Hu et al., 2008), location, and season (Cheung et al., 2011; Daher et al., 2013). Current PM regulations in California target PM₁₀ and PM_{2.5} (particles with aerodynamic diameter less than 10 and 2.5 μm, respectively) mass concentrations, with PM_{2.5} being of major concern due to the higher rate of PM_{2.5}-related morbidity and mortality in the state compared to PM₁₀ (Ostro et al., 2006; Woodruff et al., 2006). These regulations only target PM mass concentration, regardless of their source of emission and/or toxico-chemical characteristics. There is, however, strong evidence that the level of toxicity and health-related characteristics of PM are significantly affected by their chemical composition and therefore by their emission sources (Rohr and Wyzga, 2012; Stanek et al., 2011; Zhang et al., 2008; Saffari et al., 2013). Recently, there has been growing interest in using source apportionment data in epidemiological health studies (Sarnat et al., 2008; Özkaynak and Thurston, 1987; Laden et al., 2000; Mar et al., 2000; Ostro et al., 2011). These studies have provided significant evidence that exposure to PM from certain sources is linked to mortality. In a recent study in Barcelona, Ostro et al. (2011) found that exposure to sev-

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5 eral sources, including traffic emissions, sulfate from ship emissions and long-range transport, as well as construction dust, is statistically significantly associated with all-cause and cardiovascular mortality. Nonetheless, to draw firm conclusions and develop more effective control strategies to reduce population exposure to harmful sources of airborne PM, further epidemiological studies that use source apportionment data are warranted.

To date, several source apportionment studies have been conducted in California, using source-oriented (Hu et al., 2014; Kleeman and Cass, 2001; Zhang et al., 2014; DeNero, 2012) and receptor models (Hasheminassab et al., 2013; Hwang and Hopke, 2006; Ham and Kleeman, 2011; Kim and Hopke, 2007; Kim et al., 2010; Schauer and Cass, 2000). Source-oriented models focus on the transport, dilution, and transformation of pollutants from the source of emission to the receptor site; thereby providing an overall estimation regarding the spatial distribution of source contributions. Receptor models, on the other hand, focus on the behavior of ambient environments at the point of impact (Hopke, 2003). Even though these studies have provided important insights on the characteristics of sources of ambient PM as well as their relative contributions, they have been mostly conducted in a limited number of sampling locations and/or within a relatively short period of time. As a result, spatial and temporal variability of the identified sources has not been extensively examined.

20 In this study, positive matrix factorization (PMF), one of the most widely-used receptor-oriented source apportionment techniques (Paatero and Tapper, 1994), was employed in order to provide a detailed and long-term (from 2002 to 2007) quantification of the contributions of different emission sources to ambient PM_{2.5} mass concentration in California, at 8 distinct locations spanning southern, central, and northern regions of the state. The association between PM-related mortality and PM_{2.5} mass concentration as well as individual PM_{2.5} chemical components has been investigated in previous epidemiological studies in California (Ostro et al., 2006, 2007). The results of this study will be used as an input for future epidemiological studies conducted by California Environmental Protection Agency (CAL EPA), in order to further expand

the current epidemiological knowledge, by establishing the relationship between PM-related adverse health effects and specific source contributions. These findings will be crucial in establishing targeted and cost-effective regulations of PM_{2.5} emissions in the state of California.

2 Methodology

2.1 Sampling sites

Sampling was conducted at eight Speciation Trends Network (STN) sampling sites, established by the United States Environmental Protection Agency (US EPA), located in distinctly different cities all over California, including El Cajon, Rubidoux, Los Angeles, Simi Valley, Bakersfield, Fresno, San Jose and Sacramento. The studied sampling sites comprise a mixture of urban and semi-rural communities, with El Cajon and Rubidoux being located in semi-rural areas, while the rest of sampling sites being situated in densely developed urban regions of the state. Figure S1 in the Supplement shows the location of all sampling sites.

The Sacramento sampling site is located next to a park in a residential area with commercial establishments and high-density residential homes in the surrounding neighborhood. It is also about 3 km southeast of a major freeway (I-80). The sampling site in San Jose is located 46 km east of the Pacific Ocean and 14 km southeast of the San Francisco Bay. It is also surrounded by primary commercial facilities (Hwang and Hopke, 2006). Cities of Fresno and Bakersfield are located in California's heavily polluted San Joaquin Valley (SJV) (Zhao et al., 2011). These two cities are relatively far from the Pacific Ocean and are mostly impacted by secondary aerosols formed by emissions from upwind areas (Ying and Kleeman, 2006). Moreover, this part of the state usually suffers from severe particulate pollution, especially during the colder seasons (Kleeman et al., 2009). The northern parts of the SJV are dominated by agricultural activities, while the southern regions are mostly impacted by oil production

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(Held et al., 2004). The sampling site in Bakersfield is located about 4 miles southwest of downtown, in a residential neighborhood and 2 km away from the nearest freeway (State Route (SR) 99). The sampling site in Fresno is about 5.5 km northeast of the downtown commercial district (Watson et al., 2000), next to a four-lane artery with moderate traffic level. Simi Valley is located 50 km northwest of downtown Los Angeles, in Ventura county, and the sampling site in this city is situated 500 m south of SR 118 (Kim and Hopke, 2007). Two sampling locations in the South Coast Air Basin (SoCAB) were considered in this study; Los Angeles (LA) and Rubidoux. The sampling site in downtown LA is surrounded by three major freeways (i.e. I-110, I-5, and US-101) and is 30 km away from the ports of LA and Long Beach, both of which are the busiest ports in the US (Minguillón et al., 2008). This sampling site is therefore heavily impacted by primary emissions (Moore et al., 2007). Rubidoux is situated 60 km inland from downtown LA and is typically subject to aged and photo-chemically processed particulate plumes advected from upwind regions (Sardar et al., 2005). Previous studies have reported high concentration of ammonium nitrate in this region, which is mostly formed by the atmospheric reaction of nitric acid with ammonia from Chino dairy farms and livestock in upwind regions (Hughes et al., 1999). Lastly, the El Cajon sampling site is located in an inland valley, downwind of a heavily populated coastal zone, in San Diego County. This site is also impacted by emissions from I-8 freeway, situated 500 m to its north.

2.2 Sampling schedule and chemical analysis

Time-integrated 24 h PM_{2.5} samples were collected between 2002 and 2007 at all sampling sites, except for Los Angeles and Rubidoux, at which PMF analyses were conducted by a larger data set, collected between 2002 and 2013, as discussed in detail by Hasheminassab et al. (2014). For this study, however, average source contributions between 2002 and 2007 were calculated in order to compare the results with those obtained for the rest of the sampling sites. During the studied period (i.e. 2002 to 2007), PM_{2.5} samples were collected every third day in Sacramento, San Jose, Fresno, Bak-

ersfield, Rubidoux, and El Cajon sites, while every sixth day in Simi Valley and Los Angeles sites.

$PM_{2.5}$ mass concentration was determined gravimetrically by pre- and post-weighing the Teflon filters. Chemical analyses were performed based on the EPA protocol adopted for the STN field sampling. Concentration of elements on Teflon filter samples was quantified by energy-dispersive X-ray fluorescence (EDXRF) (RTI, 2009a). Major ions, including nitrate, sulfate, ammonium, sodium, and potassium, were measured by Ion Chromatography (IC) (RTI, 2009b, c). Elemental carbon (EC) and organic carbon (OC) were quantified from quartz filters, using Thermal Optical Transmittance (TOT) NIOSH 5040 carbon method (Birch and Cary, 1996).

2.3 Source apportionment

In this study, to identify the major sources of ambient $PM_{2.5}$ and quantify their relative contributions to the total mass at each site, the EPA positive matrix factorization (PMF) receptor model (version 3.0.2.2) was used. PMF is a factor analysis model that solves the chemical mass balance equations using a weighted least-squares algorithm and by imposing non-negativity constraints on the factors (Reff et al., 2007).

2.3.1 Data screening

The first step of data screening was correcting the OC data to account for sampling artifacts, caused by adsorption and/or desorption of organic vapors on quartz filters (Chow et al., 2010). For each sampling site, the OC artifact was estimated using the intercept of the linear regression of OC against $PM_{2.5}$ mass concentration (Kim et al., 2005). OC concentrations were then corrected by subtracting the OC artifact concentrations. The estimated OC artifact values (\pm standard errors) at each site are presented in Table S2.

To avoid double-counting of species, the linear correlations in each pair of S/SO_4^{2-} , Na/Na^+ , and K/K^+ were examined. Depending on the goodness of fit and the percent number of samples below detection limit (BDL) (threshold of 70%), either IC SO_4^{2-} ,

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Na⁺, K⁺ or XRF S, Na, K data were included in the PMF analyses. Measured concentrations below the detection limit (BDL) were replaced by half of the detection limit (DL) values, and their uncertainties were set as 5/6 of the DL values (Polissar et al., 1998). Missing values were replaced by the geometric mean of the species concentration, and their accompanying uncertainties were set as four times this geometric mean concentration. Species with more than 70 % BDL values as well as samples with missing mass and/or all of the elemental concentrations were excluded from the model. Lastly, occasional samples with unusually high concentrations of a few chemical species, such as those collected around 4 July and/or New Year eves with extremely high concentrations of K and/or K⁺, were discarded.

2.3.2 PMF model

Species with a signal-to-noise (*S/N*) ratio between 0.2–2, as well as those that have BDL values more than 50 % were considered as weak variables and their uncertainties were increased by a factor of 3. In order to directly apportion the total PM mass, PM_{2.5} mass concentrations were included in the data matrix as a total variable in the PMF model (Lee et al., 2011). To ensure that the inclusion of total PM mass concentration does not affect the resulting PMF solution, their uncertainties were increased by a factor of 3, similarly to a weak variable (Reff et al., 2007). The model was performed in the default robust mode to diminish the influence of extreme values on the PMF solution, and the FPEAK parameter was applied to control rotational ambiguity (Paatero et al., 2002). Furthermore, a value of 5 % extra modeling uncertainty was applied.

Moreover, uncertainties in the source profiles were estimated by a bootstrap procedure (Norris et al., 2008). 500 runs were considered for the bootstrap analysis in this study, and a solution was considered valid when the occurrence of unmapped factors was less than 10 % of the total runs. The final solutions were chosen based on the evaluation of the deduced source profiles and the quality of the chemical species fits by testing different numbers of factors.

3 Meteorology

Select meteorological parameters data, including temperature, relative humidity (RH), precipitation, as well as vector-average wind speed and direction were acquired from the online database of the California Air Resources Board (CARB). Table S1 presents the seasonal average of these parameters at all studied sampling sites. In this study, seasons were defined as spring (March–May), summer (June–August), fall (September–November) and winter (December–February), and seasonal/annual averages of all parameters reported in the following sections and shown in the figures and tables were calculated over all 6 years (i.e. 2002 to 2007). In addition, in all of the figures and tables presented in this study, sampling sites were ranked according to their latitude, from south to north (i.e. from El Cajon to Sacramento).

Most intense seasonality in temperature and relative humidity was observed at the inland areas of the SJV, in Fresno and Bakersfield. These two sites experience the hottest and driest summertime weather across the state (temperature over 25 °C and RH below 40 %), while during winter, the mean temperature in these cities is within the lowest levels among all sites (below 10 °C) and the relative humidity reaches about 75 %, comparable to levels in other sites in the northern region of the state (i.e. San Jose and Sacramento). Unlike northern areas, relative humidity exhibited more moderate seasonality in southern California, displaying minima in fall/winter (50–71 %) and maxima in spring/summer (59–77 %). At all sampling locations, the average of yearly total precipitation was negligible in summer, but greatest in winter. During the studied period, Sacramento showed the highest total precipitation in winter, followed by Los Angeles, San Jose, and Simi Valley (23.4 ± 7.1 , 21.7 ± 17.1 , 16.3 ± 3.9 , and 14.1 ± 13.0 cm, respectively). Additionally, wind speeds were generally much stronger in summer compared with fall/winter. During spring and summer, wind blows mostly from coast to inland in the southern part of the state (i.e. El Cajon, Rubidoux, LA, and Simi Valley), with a predominant westerly/southwesterly direction, while it shifts in winter and has a predominantly northerly origin at all sites, with the exception of El Cajon. In Bak-

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ersfield and Fresno, wind constantly blows from northwest throughout the year, except for Fresno in winter, when wind has an easterly direction. Lastly, in Sacramento, the prevailing wind direction is southerly/southwesterly throughout the year.

4 Results and discussion

4.1 Particulate mass

Seasonal average mass concentration of ambient $PM_{2.5}$, across all 6 years, at each sampling site is presented in Table 1. Overall, mass concentrations spanned a broad range of 8.2 to 36.6 $\mu\text{g m}^{-3}$ across the studied sites and all seasons. $PM_{2.5}$ mass concentration showed a very strong seasonality in central and northern parts of the state (i.e. Bakersfield, Fresno, San Jose, and Sacramento), with 2 to 4 times higher concentrations in winter compared with summer. This trend is typical of the California's Central Valley, which usually experiences the most severe particulate pollution during winter in the US (Ying and Kleeman, 2009). In winter, ambient $PM_{2.5}$ mass concentrations peaked at Bakersfield and Fresno (32.0 ± 1.8 and 36.6 ± 1.5 $\mu\text{g m}^{-3}$, respectively). Severe stagnation periods and decreased mixing height are mostly responsible for elevated particulate pollution during winter in this part of the state. As it will be discussed in the following section, secondary ammonium nitrate and emissions from biomass burning were mainly responsible for elevated $PM_{2.5}$ mass concentrations in these two cities during winter. In summer, on the other hand, highest mass concentrations were observed in sampling sites located in the Los Angeles Basin (i.e. Los Angeles and Rubidoux). Rubidoux displayed highest mass concentration in fall, followed by summer and spring. In addition to local sources, this region of the state is typically subject to transported plumes from upwind regions in west and central Los Angeles (Daher et al., 2013; Sardar et al., 2005), particularly during the warm seasons when the westerly wind prevails (Table S1).

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4.2 Source characterization and apportionment

4.2.1 Overview

Five to nine particle sources were identified at each sampling site. Resolved source profiles along with the explained variation (EV) of each species are shown in Fig. S2a–h, for all studied sampling sites. Gray bars represent the normalized concentration of each species to the mass concentration of PM_{2.5} apportioned to that factor, while the black dots represent the percent of each species apportioned to that factor (Lee et al., 1999). Table 2 summarizes the marker species which were used to identify each source profile. Several sources, including secondary ammonium nitrate, secondary ammonium sulfate, vehicular emissions, biomass burning, soil, fresh and aged sea salt were commonly identified at multiple sites. Few minor sources were exclusively identified at some of the sites, depending on the site location and nearby emission sources. These sources, however, accounted for a small fraction of the total mass (1–15% across the state, on an annual average basis).

Table 3 presents the slope, intercept and R^2 of the linear correlations between daily-resolved measured ambient PM_{2.5} and estimated PM_{2.5} mass concentrations obtained from PMF. It can be inferred that the PMF model was able to effectively estimate the measured PM_{2.5} mass concentrations at all sites (slopes varying from 0.83–0.91 and R^2 ranging from 0.85–0.96).

Year-to-year variability in the source contributions was overall quite small for almost all identified sources. This can be deduced from the relatively small standard errors in the 6 year seasonal average source contributions, as shown in Table S3a–d (median relative standard error of 8%, across all sites, seasons, and sources). Identified sources, on the other hand, displayed distinct seasonal and spatial variability. The percent contributions from these sources to PM_{2.5} mass are presented in Fig. 1. Overall, secondary aerosols (including secondary ammonium nitrate and ammonium sulfate) collectively comprised the largest fraction of ambient PM_{2.5} at all sampling sites (ex-

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tained at other sampling sites, the contributions from diesel and gasoline vehicles were combined together at Rubidoux and referred to as vehicular emissions throughout the discussion.

As can be seen in Fig. 2, across the state, estimated $PM_{2.5}$ mass attributed to vehicular sources (including diesel and gasoline vehicles) displayed highest levels at Rubidoux, Los Angeles, and Sacramento, with annual average (\pm standard error) contributions of 4.3 ± 0.1 , 3.6 ± 0.1 , and $3.5 \pm 0.1 \mu g m^{-3}$, respectively. Spatial pattern of $PM_{2.5}$ emissions from mobile sources across the state is in a good agreement with the findings of a recent study by Hu et al. (2014), in which they applied a source-oriented air quality model to predict primary $PM_{2.5}$ source contributions across the state of California from 2000 to 2006.

Vehicular emissions displayed similar seasonal patterns at all sampling sites with higher contributions in fall and winter compared to spring and summer. In spring, summer, and fall, highest vehicular emissions source contributions were observed at Rubidoux. In contrast, during winter, when particulate pollution is confined within the emission area due to higher atmospheric stability and lower mixing height, vehicular sources contribution exhibited the highest value in downtown Los Angeles. This trend is typical of the LA Basin, in which downwind “receptor” areas are generally impacted by emissions from upwind “source” regions, when westerly/south-westerly onshore winds prevail (Table S1) (Daher et al., 2013). Several previous studies have reported similar trends in the LA Basin (Hasheminassab et al., 2013; Heo et al., 2013; Sardar et al., 2005). It should be noted that after 2007 until 2012, contributions of vehicular emissions to ambient $PM_{2.5}$ in the LA Basin statistically significantly decreased by 20 to 25 %, following the implementation of major federal, state, and local regulations on vehicular emissions, particularly on diesel trucks (Hasheminassab et al., 2014).

Among the studied locations in the California’s Central Valley, vehicular emissions displayed the highest levels in Sacramento, while lowest in San Jose, accounting for nearly 30 and 10 % of total mass, respectively, on an average over 6 years. Vehicular emissions were comparable at Bakersfield and Fresno during spring and sum-

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mer, whereas levels were slightly higher at Bakersfield in fall and winter. Schauer and Cass (2000) conducted a 4-day sampling in Bakersfield during the winter of 1995 to quantify the sources of ambient $PM_{2.5}$, using chemical mass balance (CMB) receptor model. Average wintertime level of vehicular emissions in our study at Bakersfield ($3.0 \pm 0.2 \mu g m^{-3}$) was about half of that reported by Schauer and Cass (2000) ($6.3 \pm 0.4 \mu g m^{-3}$), whereas the percent contributions of this source to total mass were comparable in both studies (10 and 12 %, respectively). This finding suggests that vehicular emissions have decreased by almost half after almost a decade in Bakersfield.

4.2.3 Secondary aerosols

Secondary ammonium nitrate source profile was identified by high concentrations of NO_3^- and NH_4^+ (Fig. S2a–h). Its contribution ranged from 0.2 to $16.8 \mu g m^{-3}$, accounting for 3 to 55 % of ambient $PM_{2.5}$, among all sites and seasons, as displayed in Fig. 3 and tabulated in Table S3a–d. Seasonally, the contribution of secondary ammonium nitrate was largest in winter while lowest during summer, with statewide average contribution of 8.4 and $3.2 \mu g m^{-3}$, respectively. Elevated concentration of secondary ammonium nitrate during the cold seasons is mainly due to the increased partitioning of ammonium nitrate into the particle phase, favored by lower wintertime temperatures and higher relative humidity (Ying, 2011). This source displayed considerably higher contribution at Fresno and Bakersfield in winter (16.8 ± 1.3 and $15.8 \pm 1.0 \mu g m^{-3}$, respectively). Ying and Kleeman (2006) stated that diesel engines and catalyst equipped gasoline engines are important local sources that contribute to secondary nitrate in the SJV. Unlike all other sites, the seasonal trend of secondary ammonium nitrate was reverse at Rubidoux, with higher concentration in summer compared to winter (12.5 ± 0.8 and $8.9 \pm 0.8 \mu g m^{-3}$, respectively). This is probably due to increased advection of ammonia from the upwind Chino area, caused by stronger westerly/southwesterly wind speed during summer in the LA Basin (Hasheminassab et al., 2013) combined with the increased photochemical production of nitric acid in summer, which reacts with fugitive

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ammonia to produce high concentrations of ammonium nitrate in summer in this area (Hughes et al., 2002; Sardar et al., 2005).

The characterized secondary ammonium sulfate source profiles have high loadings of SO_4^{2-} and NH_4^+ (Fig. S2a–h). This source was identified at all sites, except at Fresno, where sulfate largely partitioned in a source named “sulfate-bearing road dust” along with a few other components, which will be discussed in further detail below. Annual average contributions of this source ranged from 1.3–4.6 $\mu\text{g m}^{-3}$ (or 10–24 % of total mass) among all sites, indicating that this source is a smaller contributor to total mass compared with secondary ammonium nitrate. Secondary ammonium sulfate exhibited a similar seasonal trend at all monitoring sites, displaying wintertime minima while summertime peaks due to increased photochemical activity that forms this species. Levels were also overall higher in the southern part of the state, compared to the upper regions. As argued by Ying and Kleeman (2006), the majority of secondary aerosols formed in southern California are formed from locally emitted precursors, whereas in the SJV secondary PM is mostly impacted by emissions from upwind areas (i.e. regional sources).

4.2.4 Biomass burning

Identified biomass burning source profiles consisted primarily of EC, OC, and either K or K^+ (Fig. S2a–h). Biomass burning includes emissions from wildfires and residential wood combustion. This source showed distinct seasonal and spatial variability, with highest levels observed during winter and also in upper parts of the state. Higher concentrations associated with biomass burning in winter are mainly due to the higher residential wood burning during the colder seasons. Central and northern parts of the state usually experience colder winters compared to southern regions (Table S1), therefore higher biomass burning is expected in these geographical locations, as shown in many previous studies (Hu et al., 2014; Chen et al., 2007). Biomass burning was the major primary source of $\text{PM}_{2.5}$ at Fresno and San Jose during all seasons, with levels ranging from 2.4 to 10.4 $\mu\text{g m}^{-3}$ (or 22 to 30 % of $\text{PM}_{2.5}$) at Fresno and from 2.2 to 8.0 $\mu\text{g m}^{-3}$

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(or 22 to 43 % of $PM_{2.5}$) in San Jose (Fig. 5). This source was also the dominant primary contributor to ambient $PM_{2.5}$ in Bakersfield and Sacramento during winter (12 and 31 % of $PM_{2.5}$, respectively), consistent with the findings of many previous studies in this area (Chow et al., 2007; Gorin et al., 2006; Schauer and Cass, 2000).

4.2.5 Soil

Resolved soil source profiles were dominated by crustal elements such as Al, Ca, Fe, Si, and Ti (Fig. S2a–h). These profiles generally lacked the contributions from EC and OC, indicating that they are not majorly impacted by emissions of road dust. As stated above, road dust was partially apportioned in the resolved vehicular emissions source profiles. A distinct source profile attributable to soil was not identified at Fresno. Instead, crustal elements partitioned in a separate source profile, along with high loadings of sulfate, EC, and OC, which was characterized as “sulfate-bearing road dust”. Across the state, soil exhibited lower concentrations in northern regions, namely at San Jose and Sacramento (Fig. 6). This is likely attributed to increased precipitation and higher relative humidity in this part of the state (Table S1), which limit the wind-induced resuspension of soil (Harrison et al., 2001). Soil, in contrast, accounted for a large fraction of $PM_{2.5}$ at Bakersfield, in concert with the findings of Chen et al. (2007). During summer, in particular, contribution of soil to total mass was near 20 % at Bakersfield, which could be mainly due to the lack of precipitation and low relative humidity in this area (Table S1). As discussed by Chen et al. (2007), farm lands, pasture lands, and unpaved roads are major sources of soil and windblown dust in the SJV.

4.2.6 Fresh and aged sea salt

Sources with high concentrations of Na^+ and Cl^- were characterized as fresh sea salt (Fig. S2a–h). Aged sea salt source profiles, on the other hand, were dominated by loadings of Na^+ , SO_4^{2-} , and NO_3^- . Unlike fresh sea salt, chlorine has a negligible or near-zero contribution to aged sea salt source profile. Chlorine is typically depleted

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due to reactions of sea salt with acidic gases during the long range transport of sea salt aerosols from the point of emission (Song and Carmichael, 1999). Aged sea salt overall accounted for a larger fraction (2–27 %) of ambient PM_{2.5} compared to fresh sea salt (1–13 %), in all sites and seasons (Figs. 1, S3, and S4). Aged sea salt showed a clear seasonal pattern at all sites, with higher concentrations in summer, consistent with increasing onshore winds (Table S1), while lowest during winter.

It is also noteworthy that the PMF model did not apportion a separate factor for ship emissions or a source related to ocean goods transport. However, high loadings of Ni and V (tracers of ship emissions, Arhami et al., 2009) in secondary ammonium sulfate and aged sea salt source profiles for the sampling sites in the LA Basin, suggest that these sources are affected in part by emissions from ships serving the ports of LA and Long Beach (Hwang and Hopke, 2007).

4.2.7 Other sources

As noted above, few sources were exclusively identified at some sites, with relatively low annual contributions to total mass (1–15 %, across the sites). At Rubidoux, a source profile was deduced with high loadings of Zn, Pb, EC, and OC (Fig. S2b), which is most likely attributed to local “mixed industrial” emissions in the surrounding areas. A similar source profile was also obtained in previous studies in this area (Kim and Hopke, 2007; Kim et al., 2010). At San Jose, a source profile dominated by Ni was identified, which likely indicates the contribution from nearby Ni-related industrial sources. Hwang and Hopke (2006) reported similar findings at the same sampling location, by application of the PMF model on STN data, collected between 2002 and 2005. This source, nonetheless, accounted for less than 2 % of the total mass, on an annual average basis. Copper smelters source profile, with a very high loading of Cu (> 80 %) and a slight contribution of EC, was identified in El Cajon and Bakersfield sampling sites (Fig. S2a, e). This source accounted for about 1 and 4 % of total mass, over all years, in Bakersfield and El Cajon, respectively.

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Figure 7 shows the seasonal trends of industrial emissions in locations where these sources were identified. In El Cajon and Rubidoux, contributions of the identified industrial sources peaked in winter, while in Bakersfield and San Jose, maximum emissions from copper smelters and Ni-related sources were observed in summer. It is important to note that even though the contributions from the identified industrial sources to total PM mass were overall trivial ($< 4\%$), these sources and the related elements may be important contributors to the overall particle toxicity (Toledo et al., 2008; von Schneidmesser et al., 2010; Dall'osto et al., 2008; Saffari et al., 2013).

At Fresno, a source profile with a high loading of sulfate along with road dust tracers, such as OC, EC, Fe, Ca, Mn, Si and Ti, was resolved (Fig. S2f). These road dust tracers most likely originate from the re-suspension of deposited soil and road dust enriched with vehicular emissions and lubricating oils (Pant and Harrison, 2013; Dall'osto et al., 2014). This source was therefore named “sulfate-bearing road dust” (Katrinak et al., 1995). As mentioned above, separate source profiles for secondary ammonium sulfate and soil were not identified at Fresno. Nonetheless, the relatively high loadings of sulfate and a few crustal elements (e.g. Al, Ca, Fe, Si), along with the modest contribution of ammonium, suggest that these two sources are partially apportioned into this source profile. On an average basis over all 6 years, “sulfate-bearing road dust” accounted for about 15% of total mass at Fresno and its contribution was highest in summer among all seasons ($2.7 \pm 0.1 \mu\text{g m}^{-3}$).

Relatively similar source profiles, with high loadings of chlorine, were obtained at Fresno, Bakersfield, and Sacramento, with annual average contributions of about 5, 2, and 1% to total mass, respectively (Fig. S2e, f, and h). This source, which was denoted as “chlorine sources”, was mostly detected during fall and winter at Fresno and Bakersfield, in the SJV, while it displayed the maximum seasonal average value in summer at Sacramento (Fig. 8).

5 Summary and conclusions

Source apportionment analyses were conducted using PMF receptor model applied on chemical speciation datasets, obtained from 8 different STN sampling sites throughout the state of California, between 2002 and 2007. Five-to-nine major sources contributing to ambient PM_{2.5} were identified at each site, with several of which being common in multiple locations. Overall, secondary aerosols (including secondary ammonium nitrate and ammonium sulfate) were collectively the main contributor to PM_{2.5} mass at all sampling sites, except for San Jose. Annual average source contribution of secondary ammonium nitrate and ammonium sulfate ranged from 3.1 to 12 µg m⁻³ (or 16 to 50 % of total mass) and 1.3 to 4.6 µg m⁻³ (or 10 to 23 % of total mass) across the state, respectively. On an annual average basis, vehicular emissions (including both diesel and gasoline vehicles) were the largest primary sources of PM_{2.5} at all sampling sites in the southern part of the state (i.e. El Cajon, Rubidoux, LA, and Simi Valley), with 17–18 % contribution total PM mass. In Fresno and San Jose, on the other hand, biomass burning was the dominant primary source of ambient PM_{2.5}, contributing to 27 and 35 % of total mass, on average over all years. In Bakersfield and Sacramento, biomass burning and vehicular emissions equally contributed to PM_{2.5} mass with near 12 and 25 % annual contributions, respectively. Other sources commonly identified at all sites were minor contributors to PM_{2.5}, including aged and fresh sea salt as well as soil, which contributed to 0.5–13 %, 2–27 %, and 1–19 % of total mass, respectively, across all sites and seasons. Furthermore, a few sources (including chlorine sources, sulfate-bearing road dust, and different types of industrial emissions), which overall accounted for a small fraction of total mass (1 to 15 %, on an annual average basis), were solely identified at some of the sites.

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Table 1. Seasonal average mass concentration (\pm standard error) ($\mu\text{g m}^{-3}$) of ambient $\text{PM}_{2.5}$ at the 8 sampling sites in the period between 2002 and 2007.

	El Cajon	Rubidoux	Los Angeles	Simi Valley	Bakersfield	Fresno	San Jose	Sacramento
Spring	12.0 \pm 0.5	23.6 \pm 1.3	18.1 \pm 1.5	12.8 \pm 0.8	11.8 \pm 0.5	16.4 \pm 1.1	9.7 \pm 0.4	8.2 \pm 0.3
Summer	13.1 \pm 0.4	25.6 \pm 0.9	20.2 \pm 0.7	15.9 \pm 0.5	13.5 \pm 0.4	9.7 \pm 0.3	9.6 \pm 0.4	9.2 \pm 0.4
Fall	14.5 \pm 0.5	27.4 \pm 1.5	20.8 \pm 1.2	14.4 \pm 0.9	24.6 \pm 1.7	13.7 \pm 0.6	14.8 \pm 0.8	15.1 \pm 0.9
Winter	17.1 \pm 0.7	20.0 \pm 1.1	20.4 \pm 1.6	9.8 \pm 0.8	32.0 \pm 1.8	36.6 \pm 1.5	18.6 \pm 1.2	23.5 \pm 1.2

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Table 2. Summary of the marker species for identified PM_{2.5} sources, resolved by the PMF model.

Source	Marker species
Vehicular emissions	EC, OC, Fe, Cu, Zn, Pb, Mn
Secondary ammonium nitrate	NO ₃ ⁺ , NH ₄ ⁺
Secondary ammonium sulfate	SO ₄ ²⁻ , NH ₄ ⁺
Soil	Al, Si, Ca, Fe, Ti
Fresh sea salt	Na ⁺ , Cl ⁻
Aged sea salt	Na ⁺ , NO ₃ ⁺ , SO ₄ ²⁻
Biomass burning	EC, OC, K/K ⁺
Copper smelters	Cu, EC
Mixed industrial	EC, OC, Zn, Pb
Chlorine sources	Cl ⁻
Sulfate-bearing road dust	EC, OC, SO ₄ ²⁻ , Fe, Ca, Mn, Si, Ti
Ni-related industrial sources	Ni, Mn, Mg

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Table 3. Summary statistics of the linear correlations between daily-resolved measured ambient PM_{2.5} and estimated PM_{2.5} mass concentrations obtained from the PMF model. Errors correspond to one standard error.

	R^2	Slope	Intercept ($\mu\text{g m}^{-3}$)
El Cajon	0.85	0.91 ± 0.02	0.89 ± 0.26
Rubidoux	0.96	0.91 ± 0.01	1.30 ± 1.22
Los Angeles	0.86	0.88 ± 0.02	1.58 ± 0.47
Simi Valley	0.91	0.91 ± 0.02	0.84 ± 0.23
Bakersfield	0.95	0.91 ± 0.01	0.95 ± 0.24
Fresno	0.94	0.91 ± 0.01	1.01 ± 0.23
San Jose	0.88	0.85 ± 0.01	1.35 ± 0.23
Sacramento	0.91	0.83 ± 0.01	1.47 ± 0.18

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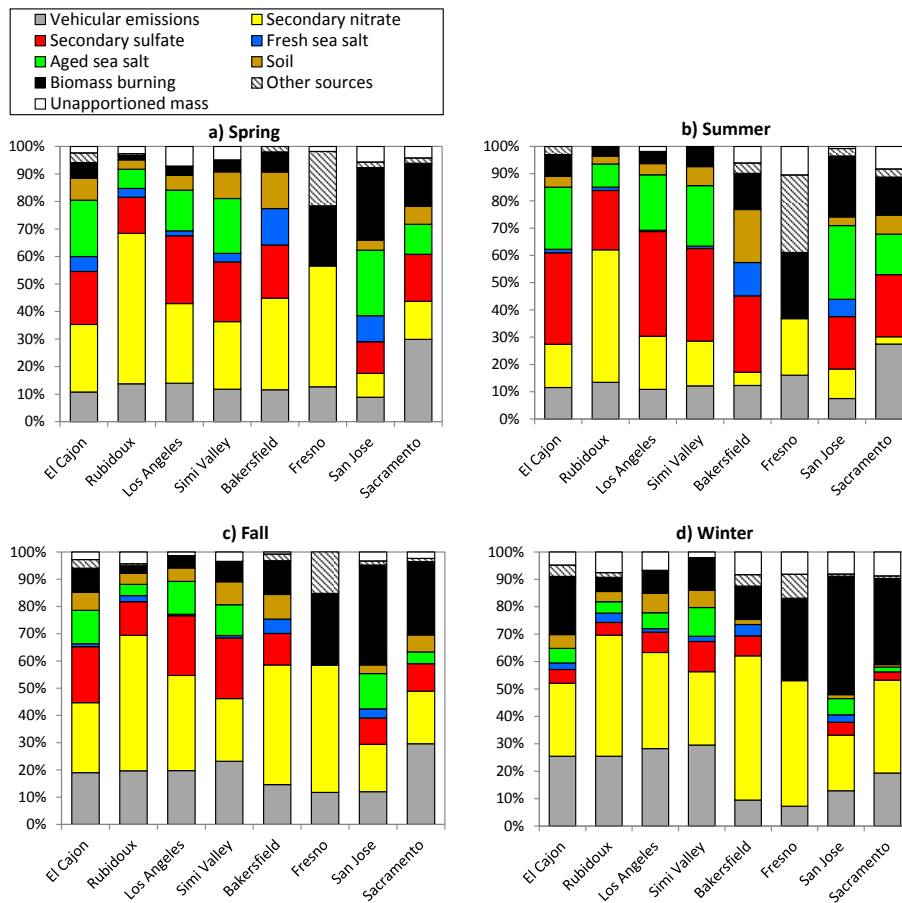


Figure 1. (a–d) Seasonal variation in the percent contribution of identified sources to ambient $PM_{2.5}$, by site.

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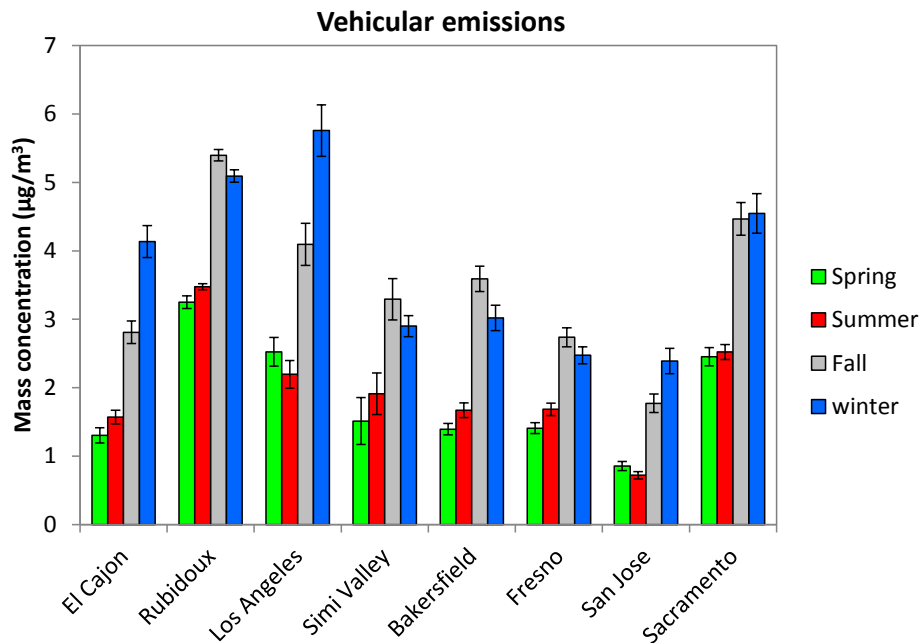


Figure 2. Seasonal average source contribution ($\mu\text{g m}^{-3}$) of vehicular emissions to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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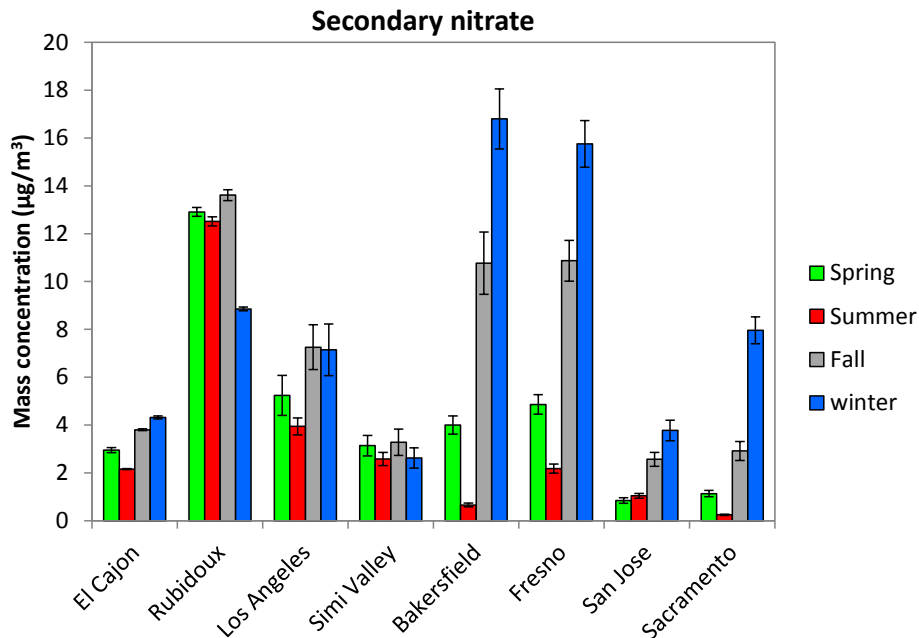


Figure 3. Seasonal average source contribution ($\mu\text{g m}^{-3}$) of secondary ammonium nitrate to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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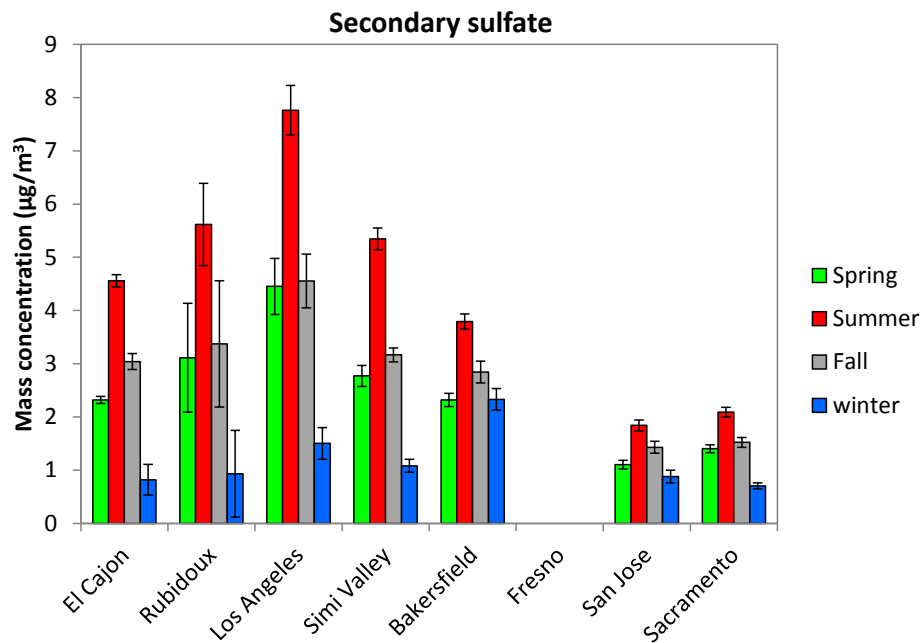


Figure 4. Seasonal average source contribution ($\mu\text{g m}^{-3}$) of secondary ammonium sulfate to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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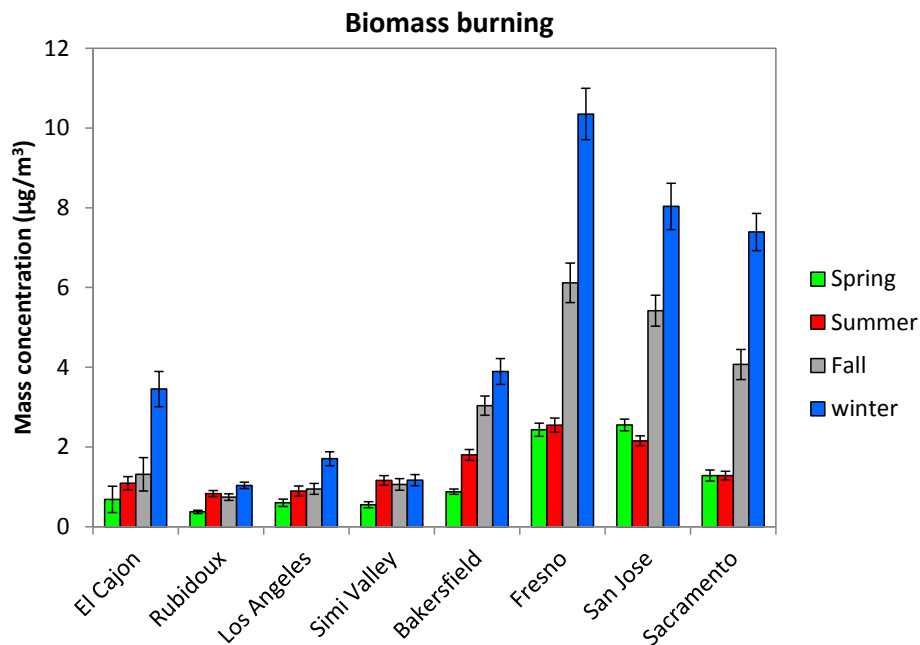


Figure 5. Seasonal average source contribution ($\mu\text{g m}^{-3}$) of biomass burning to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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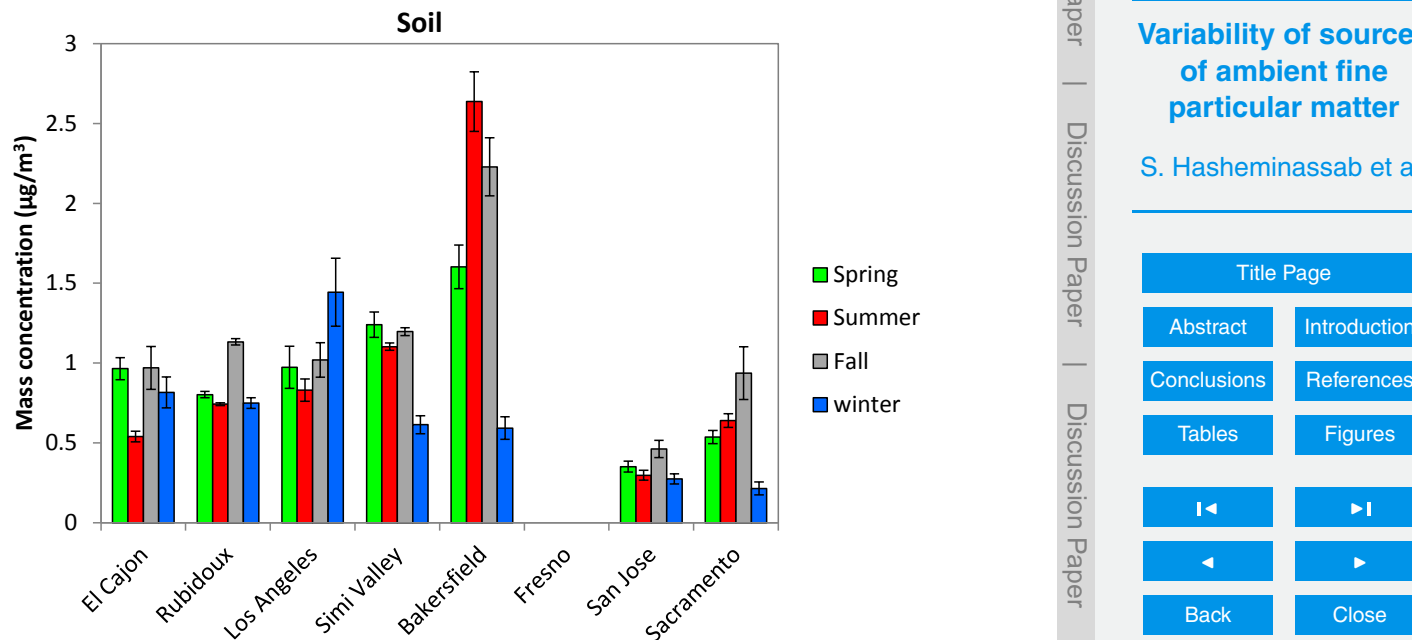


Figure 6. Seasonal average source contribution ($\mu\text{g m}^{-3}$) of soil to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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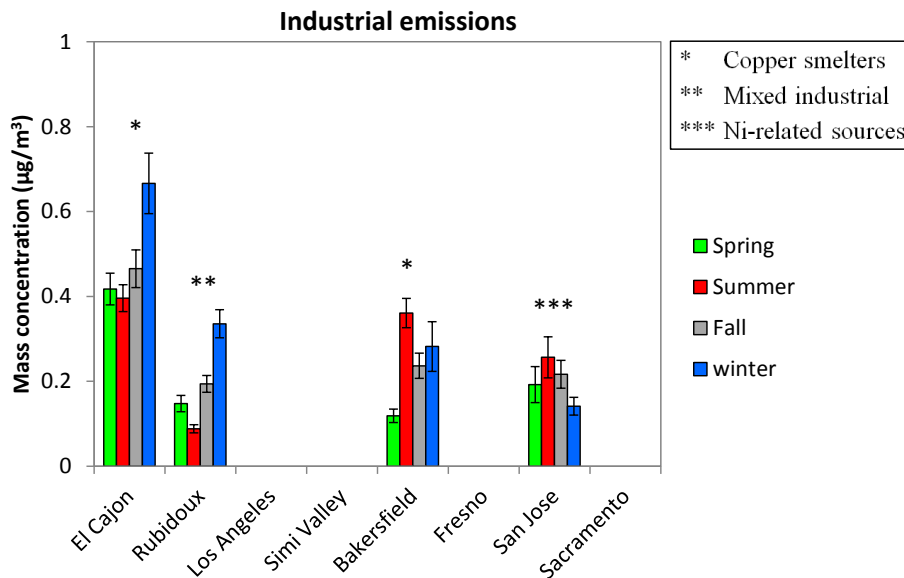


Figure 7. Seasonal average source contribution ($\mu\text{g m}^{-3}$) of industrial emissions to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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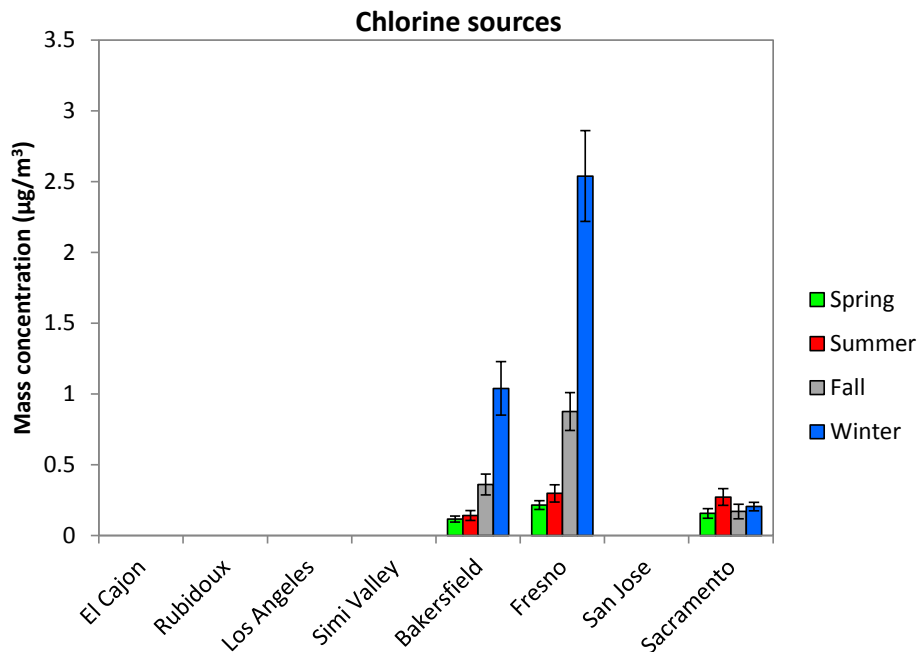


Figure 8. Seasonal average contribution ($\mu\text{g m}^{-3}$) of chlorine sources to ambient $\text{PM}_{2.5}$, by site. Error bars correspond to one standard error.

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