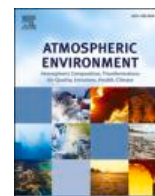




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journal homepage: www.elsevier.com/locate/atmosenv

Quantifying ambient concentrations of primary and secondary organic aerosol in central Los Angeles using an integrated approach coupling source apportionment with regression analysis

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HIGHLIGHTS

- The correlations between carbonaceous PM_{2.5} and criteria gaseous pollutants were explored.
- The tailpipe emissions decreased during the study period in central Los Angeles.
- SOA and O₃ were highly correlated as they have the same precursors and formation pathways.
- The decrease in NO₂ levels were faster than EC levels due to California regulations.

ARTICLE INFO

Keywords:

Carbonaceous PM Species
 Primary and secondary organic aerosol (POA
 SOA)
 Criteria gaseous pollutants
 Regression analysis
 Positive matrix factorization (PMF)

ABSTRACT

The main objective of this study was to explore the correlations between carbonaceous PM_{2.5} species (i.e., primary and secondary organic aerosols (POAs and SOAs) and elemental carbon (EC)) with various criteria gaseous pollutants over the 2015–2019 period in order to devise a simple way of estimating the 24-hr concentration of these PM species in central Los Angeles. The Positive Matrix Factorization (PMF) model was used to determine the contribution of various sources to organic carbon (OC) and its volatility fractions (OC₁–OC₃) in the atmosphere in years of 2015, 2017, and 2019. A 5-factor solution that includes vehicular exhaust emissions (traced by EC and OC₁), non-tailpipe emissions (characterized by Ti, Cu, and Zn), SOAs (identified by sulfate and ozone), biomass burning (associated with high loadings of K+/K), and local industrial activities (traced by Cr) was identified as the most physically interpretable solution for each of the investigated years. The SOA concentrations determined using the PMF model output and the POA concentrations, calculated by subtracting the SOA concentrations from the total OC mass, were employed in our regression analysis with criteria gaseous pollutants, the concentrations of which are routinely reported by the air quality agencies. The result of the regression analysis revealed high correlation between the concentrations of SOA and ozone ($R^2 > 0.74$) in different years, probably because they are originated from synchronized photochemical reactions in the atmosphere. The SOA/O₃ ratios were between ~47–56 µg/m³/ppm in the time period of 2015–2019. A strong correlation between POA and CO was also observed ($R^2 > 0.70$); the POA/CO ratios decreased from about 6.5 to 5 µg/m³/ppm from 2015 to 2019, which is consistent with the trend of tailpipe emission contributions to total OC concentrations in our monitoring site. EC was also strongly correlated with NO₂ ($R^2 > 0.73$) and CO ($R^2 > 0.70$) because they are emitted from the same combustion sources (e.g., vehicular emissions in central Los Angeles). Our results show persistent and significant correlations between the concentrations of criteria gaseous pollutants and carbonaceous PM_{2.5} species (i.e., POA, SOA, and EC) in different years, offering a straightforward approach to estimate the 24-h average concentration of these PM species in central Los Angeles.

1. Introduction

Numerous toxicological and epidemiological studies have provided

compelling evidence linking exposure to ambient particulate matter (PM) with adverse health outcomes, including neurodegenerative effects as well as respiratory and cardiovascular diseases (Berger et al., 2018;

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<https://doi.org/10.1016/j.atmosenv.2021.118807>

Received 17 May 2021; Received in revised form 28 September 2021; Accepted 20 October 2021

Available online 23 October 2021

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Davis et al., 2013; Gauderman et al., 2015; Mabahwi et al., 2014; Pope et al., 2004; San Tam et al., 2015). Among different size ranges of PM, ambient PM_{2.5} (i.e., particles with aerodynamic diameters <2.5 μm) is of particular interest because of its diverse physicochemical characteristics, various sources, and higher oxidative capacity (Apte et al., 2018; Davidson et al., 2007; Pope et al., 2004; Wang et al., 2016). As strong associations between different PM_{2.5} chemical components and distinct health outcomes have been documented earlier (Crilley et al., 2017; Song et al., 2007; Taghvaei et al., 2018b), many studies have focused on specific PM components to investigate the toxicity and endpoint health impacts of ambient PM (Akhtar et al., 2010; Bae et al., 2017; Fang et al., 2016; Saffari et al., 2015; Tohidi et al., 2020).

Total particulate carbonaceous material including organic carbon (OC), elemental carbon (EC), and carbonate carbon (CC) constitute a significant portion of the PM_{2.5} mass in different urban environments (Karanasiou et al., 2011). Several studies in the literature have shown strong associations between increased levels of carbonaceous aerosols in ambient air and severe impacts on human health as well as the climate (Atkinson et al., 2015; Bates et al., 2019; Chylek et al., 2006; Grahame and Schlesinger, 2010; Ning et al., 2008). EC is emitted to the atmosphere due to incomplete combustion of carbonaceous fuels, biomass burning, and cooking (Hasheminassab et al., 2014b; Healy et al., 2017; Herich et al., 2011; Schauer, 2003; Zotter et al., 2017). OC is chemically mixed with other elements and can either be in the form of primary organic aerosol (POA) or secondary organic aerosols (SOA). POA is originated directly from primary sources (e.g., traffic emissions and biomass burning) while SOA is formed indirectly through photooxidation of volatile and semi-volatile organic compounds in the presence of sunlight (Chung and Seinfeld, 2002; Jimenez et al., 2009; Ng et al., 2007; Plaza et al., 2011; Saylor et al., 2015; Zhang et al., 2007).

Although investigating the formation mechanisms, emission rates, and ambient concentrations of POAs and SOAs is important due to their distinct physicochemical characteristics and documented health impacts (Delfino et al., 2010; Künzi et al., 2015; Liu et al., 2020; Wang et al., 2017; Xu et al., 2020), there is no straightforward approach to determine and quantify their concentrations in the atmosphere. Receptor models, including the chemical mass balance (CMB), which requires a priori knowledge of primary sources, and PMF that needs a large number of ambient samples compared to CMB model, have widely been used in the literature to identify the contribution of POA and SOA sources to the total OC concentrations (Altwayjiri et al., 2021; Soleimani et al., 2019a; Stone et al., 2008). It has been reported that using constraints in the PMF model significantly reduces the rotational ambiguity of the resolved solutions (Norris et al., 2009; Paatero et al., 2002). As the PMF software has built-in constraints, it is sometimes deficient in finding appropriate factor profiles and contributions (Norris et al., 2009; Paatero et al., 2002). In such cases, a priori information about source contributions, source profiles, or chemical species ratios can serve as additional constraints (Norris et al., 2009). Many studies (Amato and Hopke, 2012; Amato et al., 2009; Sturtz et al., 2014) used measured profiles from potential source-types to constrain the PMF source factors, resulting in better correspondence between the calculated and measured abundances. Crespi et al. (2016) and Liao et al. (2015) have also used partially-constrained PMF model to constrain a small number of species instead of the entire species with multiple time resolutions. Bae et al. (2019) employed CMB and PMF models to determine PM_{2.5} source contributions to ambient OC at two urban locations in California's San Joaquin Valley. Shirmohammadi et al. (2016) also applied a hybrid molecular marker-based chemical mass balance (MM-CMB) model to investigate the source contributions to PM_{0.25}-bound and PM_{2.5}-bound OC concentrations in central Los Angeles (CELA).

Another approach to identifying the POA and SOA concentrations has been the EC/OC tracer method in earlier studies (Cabada et al., 2004; Masiol et al., 2017; Yu et al., 2004). For example, Lim and Turpin (2002) investigated the OC and EC hourly data to determine the concentrations of POA and SOA in Atlanta, GA. They reported that SOA

contributed up to 46% of measured OC in Atlanta, consistent with the observations in the Los Angeles basin. However, these studies are expensive and time-consuming since they generally require notably large datasets from various species (e.g., organic compounds, metals and trace elements, inorganic ions, and gaseous pollutants) with acceptable uncertainties in order to obtain statistically robust and physically interpretable results from source apportionment models (Manousakas et al., 2015). They also require expensive instrumentation and analytical costs for the chemical analysis of different species.

To overcome the above-mentioned challenges, in this study, we investigated the correlation between carbonous species in the atmosphere obtained from the PMF model and concentration of criteria gaseous pollutants reported by air quality agencies, as the means to estimate the 24-hr average ambient concentrations of POA and SOA in CELA. We utilized the outputs of our comprehensive PMF model for different years (i.e., 2015, 2017, and 2019) at CELA to derive linear regressions between the carbonaceous species and criteria pollutants. The input to our model was provided by the United States Environmental Protection Agency (US EPA) through the Chemical Speciation Network (CSN).

2. Methodology

2.1. Sampling location, period, and instrumentation

Fig. 1 shows the location of the monitoring site located in CELA (34°03'59.7"N, 118°13'36.8"W). CELA is in the heart of an 18 million urban area (i.e., Greater Los Angeles Area, the largest urban area in the United States) and is impacted by various types of emission sources such as vehicular and industrial (Heo et al., 2013; Mousavi et al., 2018a, 2019; Shirmohammadi et al., 2017). Previous studies have documented that CELA site is representative of a typical urban area in Los Angeles (Hasheminassab et al., 2014a).

The comprehensive PM_{2.5} chemical composition data as well as different air pollutants used in our study were obtained from the experimental monitoring conducted by the US EPA as part of the Air Quality System (AQS) and CSN database (US EPA, 2019a) for the entire years of 2015, 2017, and 2019 from January through December. In our study, we focused on these recent years (i.e., 2015–2019) because the correlations between different particulate and gaseous pollutants strongly depend on the emission sources in the area, and earlier studies have documented that due to the implementation of strict air quality regulations in California such as development of aftertreatment technologies, chemical composition of air pollutants emitted from the sources has significantly changed over the recent years (Biswas et al., 2009; Herner et al., 2011; Pakbin et al., 2009).

According to the CSN database, every six days, 24-hr time-integrated PM_{2.5} samples were collected on quartz filters using the URG 3000N Carbon Sampler (URG-3000N Carbon Sampler, URG Inc., 3000N (module C), USA) with an operational flow rate of 22 L per minute and on polytetrafluoroethylene (PTFE) and nylon filters employing a low volume Met One Speciation Air Sampling System (SASS, Met One Instruments Inc., 131 OR, USA) with a flow rate of 6.7 L per minute (SCAQMD, 2014, 2015). The concentration of EC, OC as well as OC volatility fractions were measured utilizing the Desert Research Institute (DRI) thermal/optical Carbon Analyzer (DRI thermal/optical carbon analyzer, Atmoslytic Inc., model 2001, USA) applying the Interagency Monitoring of Protected Visual Environments (IMPROVE_A) thermal protocols. The limits of detection (LOD) were equal to 0.45 μg/m³ and 0.06 μg/m³ for OC and EC, respectively (Desert Research Institute, 2005). According to this protocol, the OC fractions of the collected PM_{2.5} are gradually desorbed from quartz filters as temperatures are ramped through different stages: OC₁ (<140 °C), OC₂ (140–280 °C), OC₃ (280–480 °C). The OC fractions mainly consist of semi-volatile organic compounds with different vapor pressures, and their volatility decreases from OC₁ to OC₃ (Chow et al., 1993, 2007).

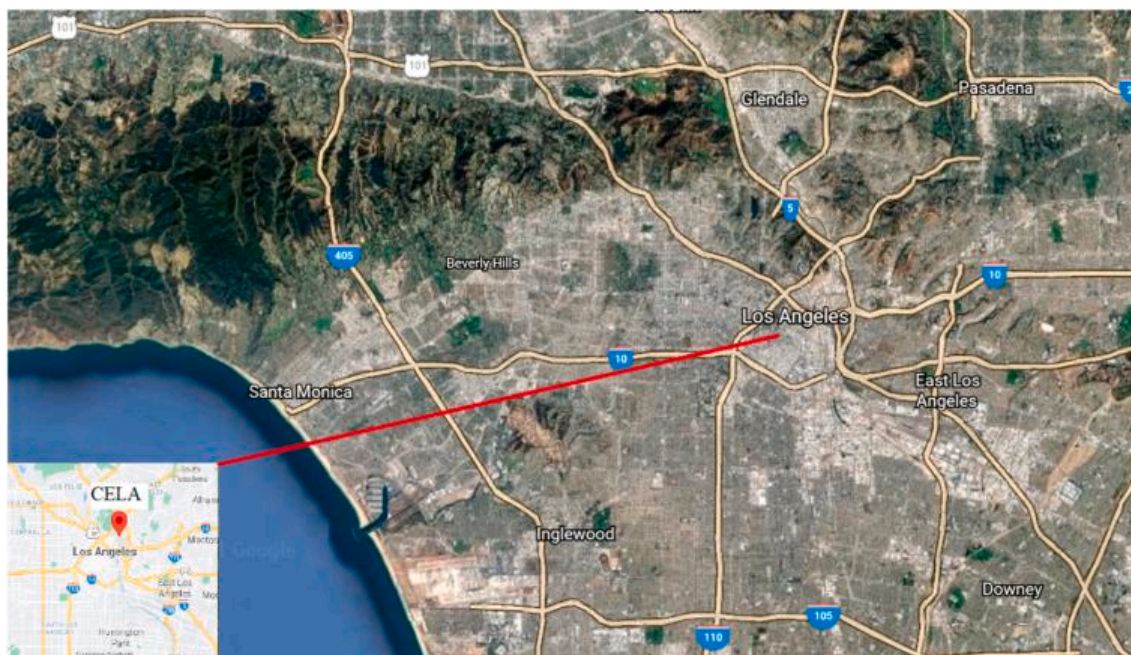


Fig. 1. Location of the monitoring site in central Los Angeles (CELA).

Inorganic Compendium Method IO-3.3 (US EPA, 1999) was used applying the energy dispersive X-ray fluorescence (EDXRF) to quantify the trace element and metal content of PTFE filters. Ion chromatography (IC) was used to determine content of inorganic ions in $PM_{2.5}$ samples collected on nylon filters (US EPA, 1999). In addition to the above-mentioned $PM_{2.5}$ chemical components, the concentrations of carbon monoxide (CO) and ozone (O_3) were continuously recorded by means of non-dispersive infrared photometry (NDIR) analyzer (AQMS-400, Focused Photonics Inc.) and ultraviolet (UV) continuous monitor (49, Thermo Environmental Instruments Inc.) (US EPA, 2019b), respectively, while the chemiluminescence method was implemented for the determination of NO_2 as indicated by the US EPA (Demerjian, 2000), which has been discussed in detail elsewhere (Maeda et al., 1980).

As the above-mentioned data are provided by the US EPA, highest standards of quality assurance and control are employed in field and lab audits to ensure the quality of the data (Solomon et al., 2014). The field audits consist of six parts: (1) determining detailed responsibilities for the site operations, (2) safety inspection, (3) confirming the quality of the selected site as well as the sampling tools according to EPA standards, (4) maintenance inspection of the sampling site and logbooks, (5) quality insurance of the sample handling and proper chain of custody, (6) validating appropriate procedures for storage and delivery. Moreover, all laboratories analyzing the CSN samples are annually evaluated for their chemical analysis reliability using performance evaluation (PE) samples provided by the National Analytical Radiation Environmental Laboratory (NAREL). These PE samples include the filters and solutions with a known quantity of the analyte loadings prepared by NAREL as reference. Solutions of anion and cation with known concentrations are also used for ion chromatography analysis. Performance audit (PA) samples, including National Institute of Standards and Technology (NIST) traceable metal weights, are also sent to analytical laboratories.

Regarding trace elements and metals, PM samples are analyzed independently by the US EPA's National Exposure Research Laboratory (NERL) EDXRF facility. Once the audited laboratories analyze the filters, they are sent back to NERL for reanalysis to certify that the level of elements on filters has not been affected by handling or delivery. In addition to the above-mentioned quality assurance procedures, NAREL conducts on-site laboratory technical systems audits (Solomon et al., 2014).

2.2. Source apportionment analysis

2.2.1. PMF model

PMF is a receptor model which has widely been used to identify the sources and quantify their contributions to the target variable (here, ambient $PM_{2.5}$ -bound OC) (Paatero and Tapper, 1994; Wang et al., 2019). This multivariate model is used for solving the chemical mass balance equation:

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

where X_{ij} , the mass concentration, refers to the i_{th} sample and the j_{th} species and number of factors p . g_{ik} stands for the airborne mass concentration contributed by k_{th} factor to i_{th} sample. f_{kj} indicates j_{th} species resolved factor of each source. e_{ij} is the model residual error in i_{th} sample for j_{th} specie.

The main goal of the PMF model is to find out the most appropriate factor profile and contribution by minimizing the objective function, Q , based on the following equation:

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{u_{ij}} \right)^2 \quad (2)$$

where n and m represent the number of samples and species, u_{ij} refers to the uncertainty of the measured mass concentration for the j_{th} species and the i_{th} sample.

The above-mentioned minimization is conducted by assigning non-negative values to the factor profiles and contributions as the constraints of the optimization process (Norris et al., 2014b). The following equation was utilized to determine the uncertainties of the input species to our PMF model (Paatero et al., 2014):

$$\sigma_{ij} = (0.05 \times X_{ij}) + DL_j \quad (3)$$

in which σ_{ij} is the calculated uncertainty of the i_{th} sample and the j_{th} species. DL_j indicates the detection limit assigned to the j_{th} species.

The mass concentration of the species as well as the above-mentioned user-defined uncertainty were employed as input to the US EPA's PMF model version 5.0 and the OC concentration was chosen as

the “total variable”. The PMF runs were conducted using the robust mode in which the impact of samples with significant uncertainties are minimized. To further validate the PMF outputs, we performed different uncertainty analyses including the Bootstrap (BS), Displacement (DISP), and BS-DISP (Bootstrap + Displacement) tests.

DISP analysis investigates the effects of rotational ambiguity by evaluating the largest range of source profile values without a notable increase in PMF objective function (Q), and does not capture the uncertainty of PMF solutions caused by random errors in the data. On the other hand, BS analysis includes effects from random errors and partially includes effects of rotational ambiguity. Unlike DISP and BS, BS-DISP analysis covers to a great extent the effects of random errors and rotational ambiguity. Therefore, for modeling errors (e.g., variation of source profiles with time, incorrect number of factors, etc.), DISP intervals are directly affected; however, BS results are generally robust. In combined mode, the results of BS-DISP analysis are more robust compared to DISP because the displacements in DISP analysis of BS-DISP are not as strong as when performing DISP by itself. (Brown et al., 2015; Norris et al., 2014a; Paatero et al., 2014; Reff et al., 2007).

Based on the results of the BS analysis, our PMF outputs were verified because for all the PMF-resolved factors, around 90% of the results were re-mapped. Regarding the DISP analysis, our PMF solutions were considered reliable without any rotational ambiguity due to the <1% drop in the Q value and absence of any factor swap for the $dQ_{\max} = 4$. Also, a sensitivity test for different PMF runs with different number of factors (Fig. S1) validated our number of factor selection (i.e., 5 factors).

2.2.2. PMF input

In the PMF model, different numbers of factors and various extra modeling uncertainty values were investigated following a trial-and-error approach to identify the most interpretable and statistically robust solution for emission sources contributing to the total OC mass concentrations. The final number of factors (i.e., 5) were determined according to several criteria: 1) Strong correlation (i.e., high linear regression R^2 value) of predicted versus measured total metal mass concentrations, 2) Physically interpretable PMF-resolved source profiles, 3) Evaluation of the uncertainty analyses on the PMF outputs (BS, DISP, and BS-DISP).

The optimum solution in our model included EC, OC, OC_x (i.e., OC_1 , OC_2 , and OC_3), sulfate (SO_4^{2-}), O_3 , potassium ion to potassium ratio (K_+/K), and metal elements such as potassium (K), zinc (Zn), titanium (Ti), copper (Cu) and chromium (Cr). Numerous studies have documented EC, OC_1 , OC_2 , and OC_3 as indicators of gasoline and diesel exhaust emissions (Cao et al., 2005; Schauer, 2003; Zong et al., 2016), O_3 and SO_4 as chemical markers of the photochemical reactions and secondary aerosols (Heo et al., 2015; Jacob, 1999; Taghvaei et al., 2018a), K^+/K as a frequently used tracer of biomass burning (Lee et al., 2007; Zhu et al., 2017), Cu and Ti as surrogates of road dust and brake abrasion particles (Adamiec et al., 2016; Harrison et al., 2012), and Cr as a marker of industrial activities in the area (Mousavi et al., 2018b; Propper et al., 2015).

2.2.3. Linear regression analysis

The SOA concentrations were determined based on the contribution of the “SOA” factor to total OC mass concentrations resolved by the PMF model. The POA concentrations were calculated as the difference of total OC and PMF-resolved SOA mass concentrations. We then conducted linear regression analysis between SOA and POA concentrations and different criteria gaseous pollutants reported frequently by air quality monitoring stations. The time-integrated data of the gaseous pollutants for different years were extracted from CSN database as mentioned earlier. We also conducted linear regression analysis between EC and gaseous pollutants including CO and NO_2 . Table S2 shows the standard error for SOA and POA linear regression analysis.

3. Results and discussions

3.1. PMF source apportionment results

3.1.1. Number of factors

Based on the correlation coefficient (i.e., R^2) values between the predicted and actual total OC mass concentration ($R^2 > 0.90$), our PMF model quantified the contributions of 5 factors to the total OC in CELA. As it will be elaborately discussed in the following sections, the PMF-resolved factors were tailpipe emissions, non-tailpipe emissions, biomass burning, SOA, and local industrial activities. The PMF-resolved factor profiles for the years 2015, 2017, and 2019 at our monitoring site are shown in Fig. 2. Figs. 3 and 4 also represent the relative and absolute contributions of the identified sources to the total OC mass concentrations for the study location.

3.1.2. Factor identification

3.1.2.1. Factor 1: tailpipe emissions. The first factor was associated with ~50–60% loadings of EC and high loadings of OC_1 (i.e., 50–90%). This factor also demonstrated ~40–50% and ~30–40% contributions of OC_2 and OC_3 , respectively (Fig. 2). EC is a well-known tracer of vehicular emissions (Mooibroek et al., 2011; Zong et al., 2016), and OC_1 has also been associated with tailpipe emissions (Cao et al., 2006; Zong et al., 2016). Moreover, OC_2 and OC_3 have been reported as the significant components of gasoline exhaust (Cao et al., 2006; Zhu et al., 2010), which corroborates the vehicular origin of this factor. The factor also has a significant contribution to total OC mass in CELA during the whole study period, accounting for ~35–45% of total OC mass in the investigated site (Fig. 3). Furthermore, the absolute contribution of this factor to total OC (Fig. 4 and Table S1) decreased significantly ($P_{\text{value}} < 0.05$) from $1.5 \pm 0.20 \mu\text{g}/\text{m}^3$ to $1.0 \pm 0.10 \mu\text{g}/\text{m}^3$ over the 2015–2019 period, which could be due to the adopted strict air quality regulations targeting tailpipe emissions in California during the recent years as elaborately discussed elsewhere (Altwayjiri et al., 2021).

3.1.2.2. Factor 2: non-tailpipe emissions. The second factor was characterized by high loadings of Ti, Cu, and Zn (i.e., ~60–80%) (Fig. 2), and contributed to a large portion of total OC mass concentration (i.e., 28–33%) according to Fig. 3. Previous studies documented Ti, Cu, and Zn as chemical tracers of particle brake wear, tire wear, and engine abrasion (Adamiec et al., 2016; Harrison et al., 2012; Peltier et al., 2011; Thorpe and Harrison, 2008). It should be noted that in recent years, electric vehicles (EVs) are replacing internal combustion engine vehicles. EVs are much heavier than other vehicles, which increases the friction between their tires and road surfaces, resulting in higher re-suspension of road dust particles (Timmers and Achten, 2016). Beddows and Harrison (2021) reported that EVs have approximately 5% higher $PM_{2.5}$ emission factors than euro-6 diesel and petrol equivalent. Moreover, Farahani et al. (2021) showed an association between the growing use of EVs and the increase of resuspended road dust emissions in the area. Therefore, a fraction of non-tailpipe emissions could be attributed to EVs in the Los Angeles basin (Kapustin and Grushevenko, 2020). As shown in Fig. 3, the relative contribution of non-tailpipe particles to total OC were $28 \pm 2.5\%$ in 2015, $28 \pm 2.2\%$ in 2017, and $33 \pm 2.6\%$ in 2019. According to Fig. 4 and Table S1, the absolute contributions of non-tailpipe emissions to total OC mass concentrations were comparable throughout the study period (P_{value} of 0.12). Altwayjiri et al. (2020) reported that the relative (fractional) contribution of non-tailpipe emissions to the total OC mass increased during the 2005–2015 period in CELA. Our findings also demonstrate a similar trend from 2015 to 2019, which is most likely attributed to the lack of local regulations controlling the non-exhaust emissions in California.

3.1.2.3. Factor 3: secondary organic aerosols (SOA). The third factor

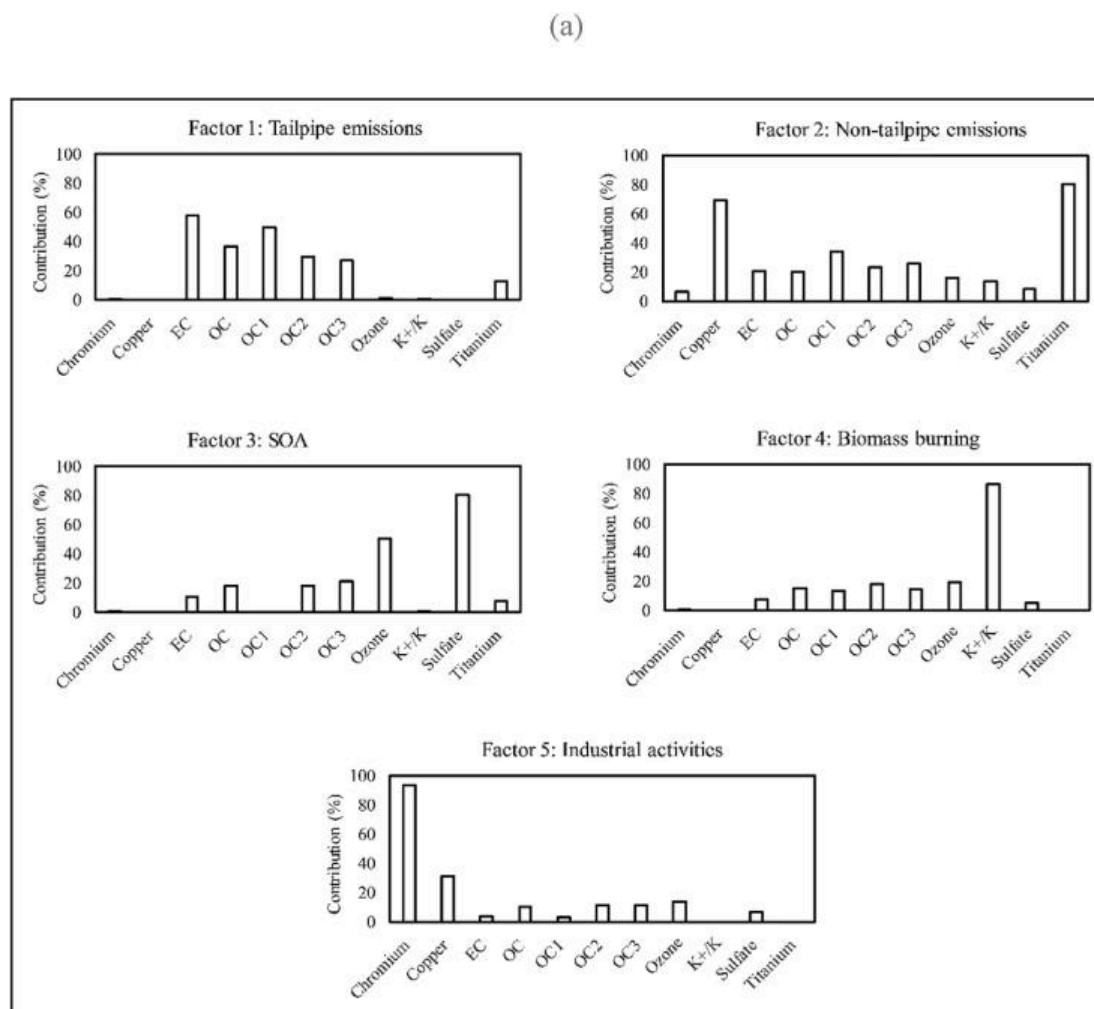


Fig. 2. PMF-resolved factor profiles for (a) 2015; (b) 2017; and (c) 2019.

demonstrated 80% contributions of sulfate and approximately between 45 and 75% contributions of O_3 according to Fig. 2, and it is another major contributor (>18%) to total OC concentrations in CELA (Fig. 3). Based on earlier studies, sulfate (in the form of ammonium sulfate), O_3 , and SOA are formed through concurrent photochemical reactions involving hydroxyl radicals (OH^\cdot) in the ambient atmosphere (Carlton et al., 2009; Jacob, 1999); thus, O_3 and sulfate are regarded as surrogates of SOA formations in the atmosphere (Heo et al., 2009; Taghvaei et al., 2018b). For example, Yuan et al. (2006) have illustrated significant correlations between the SOA and secondary sulfate concentrations at multiple environments in different seasons in Hong Kong. Moreover, it has been documented that organic and inorganic secondary aerosols, including ammonium sulfate and SOA, are internally mixed in the atmosphere (Harrison et al., 2016; Huang et al., 2014; Turpin et al., 1991). We, therefore, concluded that “SOAs” is an appropriate label for this factor. According to Fig. 3, the SOA fractional contribution to total OC over the study period remained constant. However, our findings revealed that the absolute contribution of this factor to total OC decreased overall from 2015 ($\sim 0.83 \mu\text{g}/\text{m}^3$) to 2019 ($\sim 0.46 \mu\text{g}/\text{m}^3$) (Fig. 4 and Table S1). This trend is likely due to the implementation of numerous air quality regulations during the study period, which limited the emissions of primary organic precursors of secondary aerosols. These regulations are elaborately discussed in sections 3.3 and 3.5.

3.1.2.4. Biomass burning. Other significant contributors to primary OA

in urban areas are biomass burning and cooking emissions (Crippa et al., 2013; Mohr et al., 2015; Shah et al., 2018; Sun et al., 2011). The fourth factor is represented by high loadings of K^+/K (i.e., ~ 70 – 80%). The ratio of K^+/K has previously been used as a tracer of biomass burning emissions in metropolitan environments, including CELA (Jung et al., 2014; Soleimanian et al., 2019b; Yu et al., 2018). Furthermore, meat cooking sources may also emit K^+ (Simoneit, 2002). So it is conceivable that cooking emissions may also be contributing to this factor since they are comparable in magnitude and chemical signature to vehicular emissions of POA (e.g., Shah et al. (2018); Mohr et al. (2015); Sun et al. (2011)). While it is common to expect higher contributions of biomass burning emissions to OC in winter, our results (Fig. S2) showed comparable contributions during the warm and cold periods in CELA ($P_{\text{value}} > 0.16$). A potential reason for this observation may be attributed to the frequent summer time wildfire events in the area (Okoshi et al., 2014; Warneke et al., 2012), counteracting the influence of higher wood burning emissions during the winter time (Heo et al., 2013; Lee et al., 2007). This factor accounts for less than 10% of the total OC mass concentrations throughout the study period, according to Fig. 3.

3.1.2.5. Factor 5: local industrial activities. This factor has ~ 80 – 90% loading of Cr in its profile and has a negligible contribution to the total OC over the investigated period. Previous studies in the literature have identified Cr as a tracer of industrial activities (e.g., electroplating, refractory, metallurgy, and foundry industries) (Mansha et al., 2012;

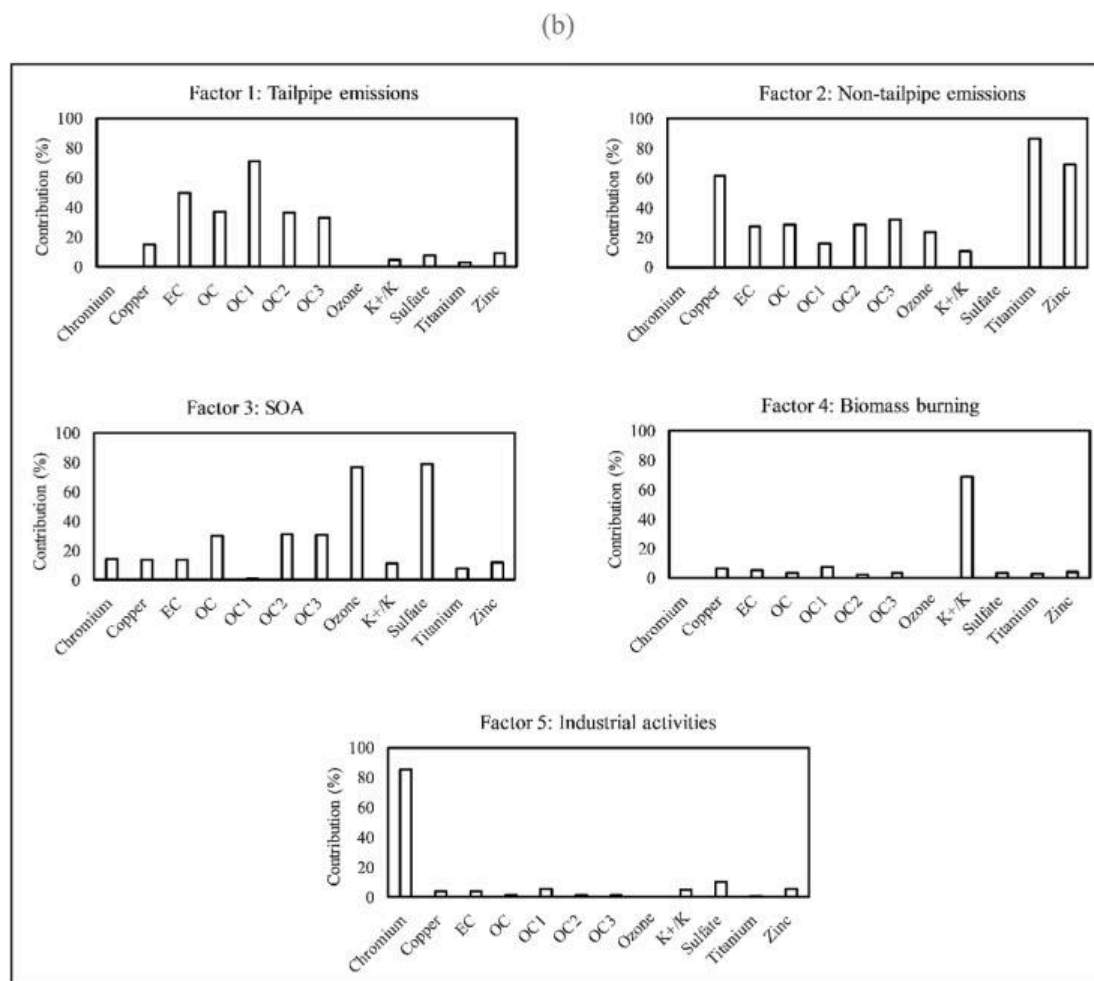


Fig. 2. (continued).

Morrison and Murphy, 2010; Tositti et al., 2014). Similar to the observed trend in tailpipe emissions, absolute contributions of this factor to the total OC mass decreased from $0.37 \pm 0.1 \mu\text{g}/\text{m}^3$ to $0.06 \pm 0.01 \mu\text{g}/\text{m}^3$ over the 2015–2019 period.

3.2. Linear regression between SOA and O_3

The results of the regression analysis for 2015, 2017, and 2019 (Fig. 5) displayed positive associations between PMF-resolved SOA factor and O_3 in CELA. Our findings revealed that although the SOA concentration resolved by the PMF model varied significantly over the 2015–2019 period, the average SOA/ O_3 values remained almost constant (approximately between ~ 47 and $55 \mu\text{g}/\text{m}^3/\text{ppm}$) over the investigated period (i.e., 2015 to 2019).

According to the figure, there was a high correlation ($R^2 > 0.74$) between the PMF-resolved SOA concentrations and O_3 values, probably due to synchronized photochemical reactions producing these species in the atmosphere (Carlton et al., 2009; Jacob, 1999). Numerous studies in the literature reported that a significant portion of O_3 and SOA are originated from the same volatile organic compounds (VOCs) in different environments (Cui, 2013; Lin et al., 2015; Shin and Jo, 2013; Wu et al., 2017). For example, Wu et al. (2017) investigated the O_3 and SOA formation potential from anthropogenic VOC emissions and reported that alkylbenzenes (i.e., a known VOC from anthropogenic emissions) accounts for about 40–50% of the total ozone and SOA formation in various locations in China during the year 2010. The California Research at the Nexus of Air Quality and Climate Change (CalNex) campaign at Pasadena in Los Angeles county, reported an

increase in total particulate carbon when the primary emissions in the area were transported to Pasadena and coincided with an increase in secondary pollutants such as acetaldehyde (Bahreini et al., 2012). It has been shown that vehicle emissions (especially emissions from gasoline vehicles) are the predominant source of light VOCs, including benzene (Marr and Harley, 2002; Warneke et al., 2007) in California. Once emitted in the air, VOCs undergo reactions with atmospheric oxidants and form SOA. In addition, in an air mass, O_3 generation arises from OH reactions with CO and VOCs (Hayes et al., 2013). While there are mitigating policies to decrease the vehicular VOCs, some recent studies pointed to the growing importance of volatile chemical products (VCPs) as significant contributors to the formation of ozone and SOA due to higher reactivity with OH (Li et al., 2018; McDonald et al., 2018). Shah et al. (2019) showed that the potential of SOA formation from VCPs is larger than that from vehicles (with a ratio of 1.3) in urban environments. In a similar study in Los Angeles, McDonald et al. (2018) reported that SOA formed from VCPs (e.g., personal care products) to vehicular VOCs has a ratio of 1.4. Consequently, the similarity in formation mechanisms of SOA and ozone provides a possible platform for quantifying the SOA concentration based on ozone concentrations. Earlier studies in the Los Angeles basin showed that ozone exhibited higher concentrations on weekends in comparison to weekdays, which stems from lower vehicular emissions than non-methane VOC emissions on weekends, resulting in higher ozone production and lower ozone destruction by nitrogen oxides (Pollack et al., 2012; Warneke et al., 2013). Similarly, Heo et al. (2015) showed higher SOA formations on weekends than weekdays in CELA, corroborating our correlations. These observations are expected to be similar to other urban areas in the US,

(c)

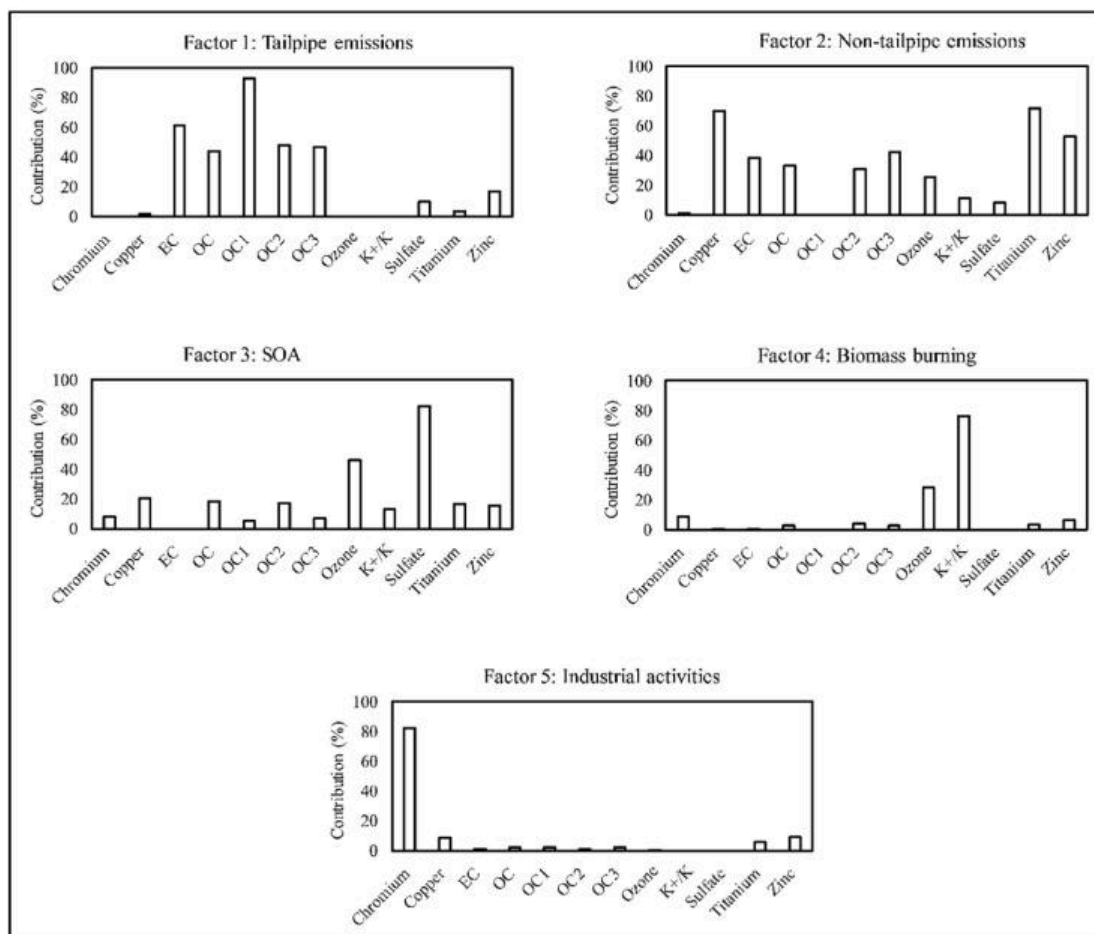


Fig. 2. (continued).

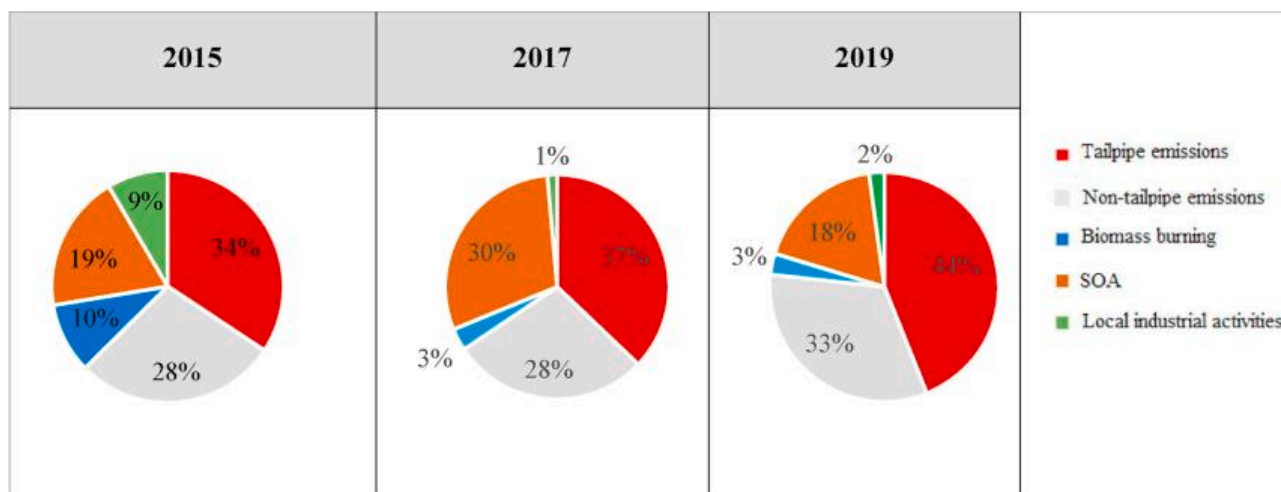


Fig. 3. The relative (fractional) contribution of PMF-resolved sources to ambient OC in CELA over the years of 2015, 2017, and 2019.

given that VOC emissions have similar composition and emission rates in the US and have followed consistent trends over the past several years (Warneke et al., 2007, 2012).

3.3. Linear regression between POA and CO

The contributions of POA to OC mass were estimated by subtracting the PMF-resolved SOA concentrations from the total OC mass (Turpin

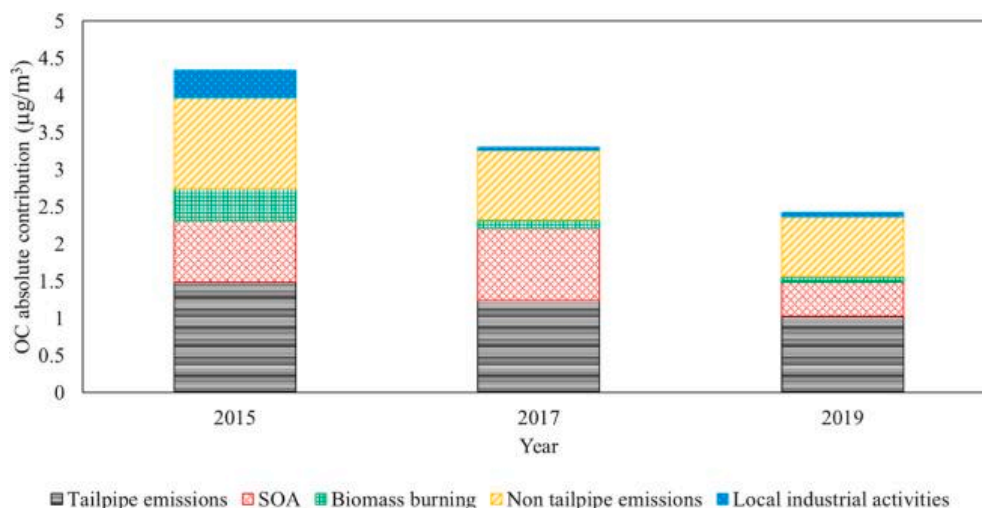


Fig. 4. Absolute source contributions to ambient OC mass concentrations during the years of 2015, 2017 and 2019 in CELA.

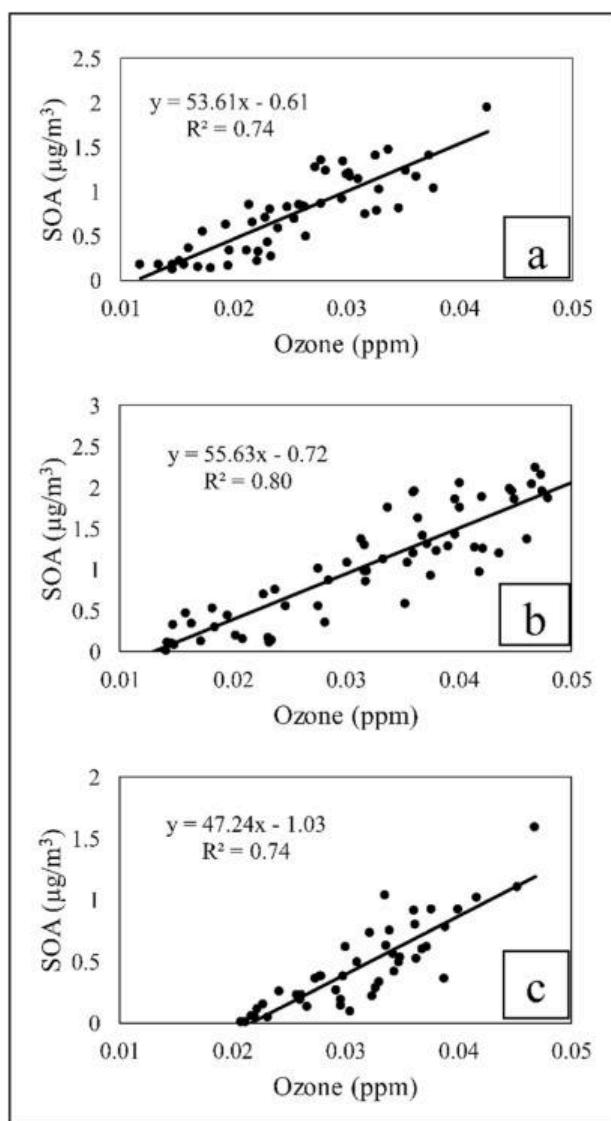


Fig. 5. Linear regression between PMF-resolved SOA and O_3 in: (a) 2015; (b) 2017; and (c) 2019 in central Los Angeles (CELA).

and Huntzicker, 1995; Wu and Yu, 2016). A strong correlation between PMF-derived POA concentrations and CO concentrations in CELA ($R^2 > 0.70$) during the study period is shown in Fig. 6. The average POA/CO values ranged from approximately $6.5 \mu\text{g}/\text{m}^3/\text{ppm}$ in 2015 to about $5 \mu\text{g}/\text{m}^3/\text{ppm}$ in 2019, in agreement with the trend of tailpipe emission contribution to total OC mass in CELA. The ratio of POA/CO in this study is lower than the values reported by an earlier study by de Gouw et al. (2008) in the northeastern United States in 2004 ($9.4 \mu\text{g}/\text{m}^3/\text{ppm}$). Using a quadrupole mass filter, the authors collected their organic matter (OM) onboard a ship and an aircraft employing an Aerodyne aerosol mass spectrometer (AMS). The POA/CO ratios estimated in earlier studies at Tokyo also using an AMS and in Zurich utilizing the solution of 6-factorial PMF were about 11 and $10.4 \mu\text{g}/\text{m}^3/\text{ppm}$, respectively (Lanz et al., 2007; Takegawa et al., 2006). In addition to the different instrumentation and analytical methods employed to estimate POA, the POA/CO ratio is affected by the mixture of specific POA sources in a given area (e.g., traffic vs biomass combustion), since they all emit CO but in different proportions relative to POA, as well as by the time period during which these studies were conducted since studies published earlier may not incorporate the impacts of subsequent air pollution mitigation policies on the CO and POA levels. Therefore, in addition to the different sampling methods, these other factors will need to be considered when comparing the POA/CO ratios among various studies. A possible explanation of the variations between these measured ratios is the implementation of various regulations in California targeting POA emission sources. Of particular note is the LEV II (CARB, 2019), which was implemented between 2004 and 2010 and targeted vehicles with the model year 2004 and above (Lurmann et al., 2015). In addition, other programs including financial incentives for cleaner port trucks (2007) have also been implemented to mitigate the air pollution generated by mobile sources in the area (Haveman and Thornberg, 2008; Lee et al., 2012). Hence, there could be reductions in POA faster than CO over the years in various parts of the world, which is in agreement with the trend of tailpipe emission contributions to total OC mass as discussed in Section 3.1.2.1. An equally important justification is that the relationship between POA and CO is also expected to depend on meteorological factors in different parts around the globe. This is because a major fraction of POA consists of semi-volatile species that partition to particulate phase upon cooling in the atmosphere (Alam et al., 2003), while CO is an inert non-reactive species, often used as a tracer of atmospheric dilution and its mixing height (Gamage et al., 2020; Turnbull et al., 2006). Thus, meteorology plays a major role on the POA/CO ratio, with colder temperatures increasing POA concentrations to a higher degree than CO, so at colder climates one might observe a higher POA/CO ratio because the POA concentration is affected not only

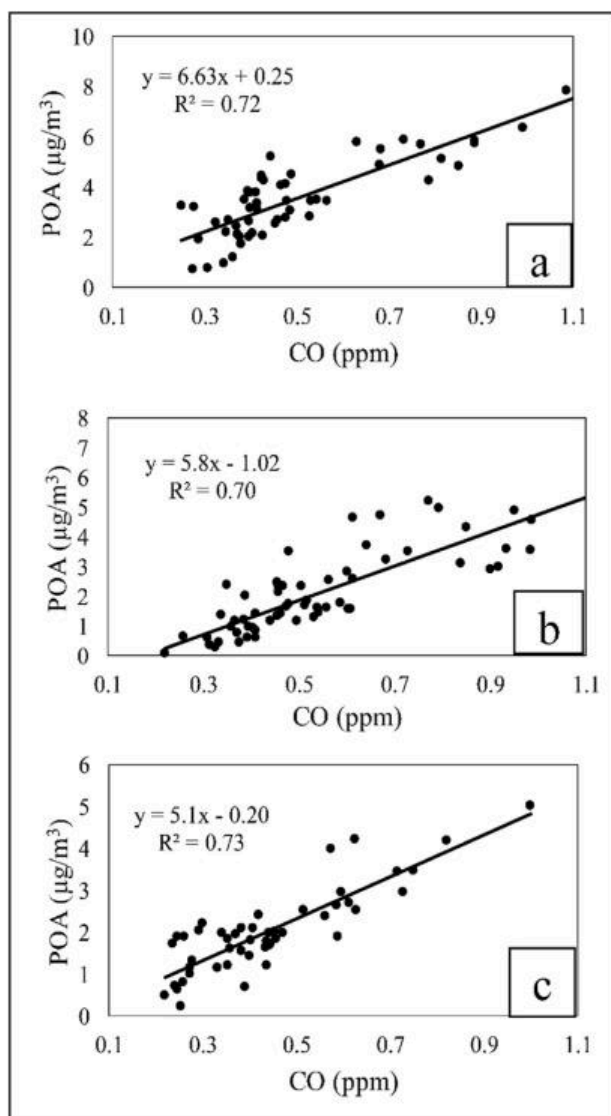


Fig. 6. Linear regression between PMF-resolved POA and CO in: (a) 2015; (b) 2017; and (c) 2019 in central Los Angeles (CELA).

by dilution but also by cooling which favors the partitioning of semi-volatile organic species to the particle phase.

3.4. Linear regression between EC and CO

Regression lines between CO and EC for CELA over the 2015–2019 period are illustrated in Fig. 7. According to the figure, we observed high correlation ($R^2 > 0.70$) between EC and CO values in the study site, corroborating their common origins (i.e., combustion related emissions). Previous studies reported that CO and EC are both products of incomplete combustion, and indicators of combustion emissions (Kirchstetter et al., 1999; Subramanian et al., 2010). We also observed that the EC/CO ratios in CELA during years 2015 and 2019 were comparable (around $2.3 \mu\text{g}/\text{m}^3/\text{ppm}$), and consistent with the findings of Subramanian et al. (2010) who reported an EC/CO ratio of $2.89 \pm 0.89 \mu\text{g}/\text{m}^3/\text{ppm}$ in the Mexico City Metropolitan Area.

3.5. Linear regression between EC and NO₂

Fig. 8 shows the daily correlation of EC and NO₂ concentrations in the study area over the 2015–2019 period. According to the figure, the EC was highly correlated ($R^2 > 0.73$) with NO₂, and EC/NO₂ ratios were

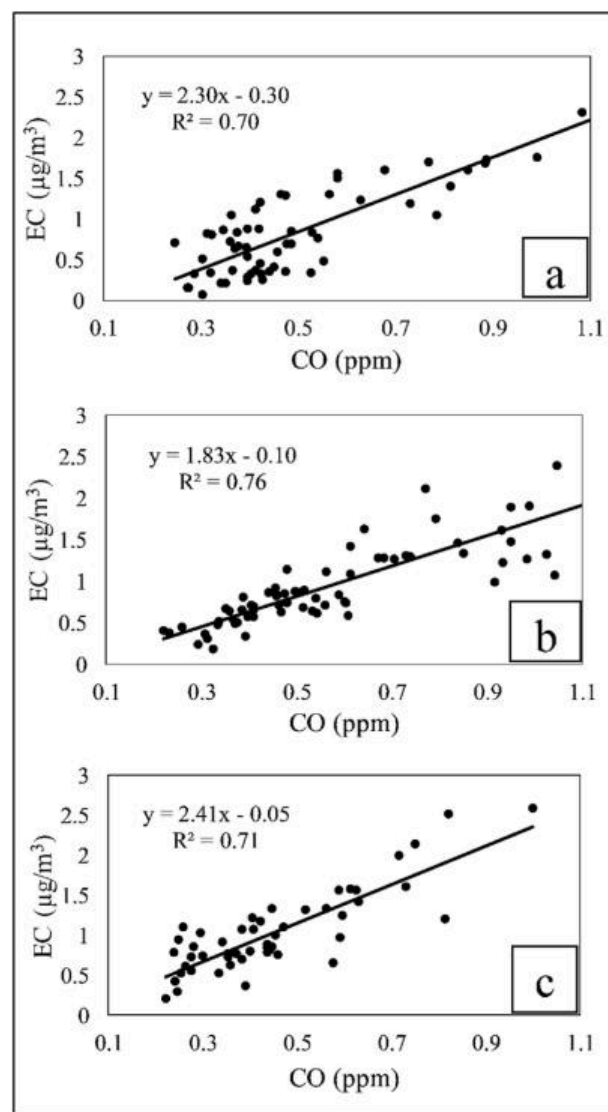


Fig. 7. Linear regression between EC and CO in: (a) 2015; (b) 2017; and (c) 2019 in central Los Angeles (CELA).

comparable ($\sim 0.04\text{--}0.06 \mu\text{g}/\text{m}^3/\text{ppb}$) over the whole period (i.e., 2015–2019), which is most probably because they are originating from the same sources. Previous studies have documented road traffic (particularly diesel engines) and other combustion activities as major sources of NO₂ and EC (Afzal et al., 2012; Pepe et al., 2019; Zhang et al., 2018). In concert with our observation (Kim et al., 2004), reported $0.034 \mu\text{g}/\text{m}^3/\text{ppb}$ as the EC/NO₂ ratio during their experimental measurements in the vicinity of busy roadways in the San Francisco metropolitan area, while Altuwayjiri et al. (2020) reported a ratio of $0.040 \mu\text{g}/\text{m}^3/\text{ppb}$ in the city of Milan, Italy. It should be noted that there are possible factors for the variations of these estimated ratios, such as temporal and spatial variations of measurements, various measurement instrumentations, available sources in the study area, and established air quality regulations including development of aftertreatment technologies as discussed earlier. In our results, we see an increase of this ratio by $\sim 50\%$ from 2015 to 2019. This could be explained by the faster reduction in NO₂ than EC levels, due to regulatory policies targeting NO₂ that have been implemented in California in recent years (Brauer et al., 2008; Kim et al., 2004). For example, the CA LEV-II program urged reduction of major pollutants (e.g., carbon monoxide (CO) and nitrogen dioxide (NO₂)) emitted from various vehicles (CARB, 2000; Hwang and Doniger, 2004). Furthermore, CA LEV III is planning to further reduce

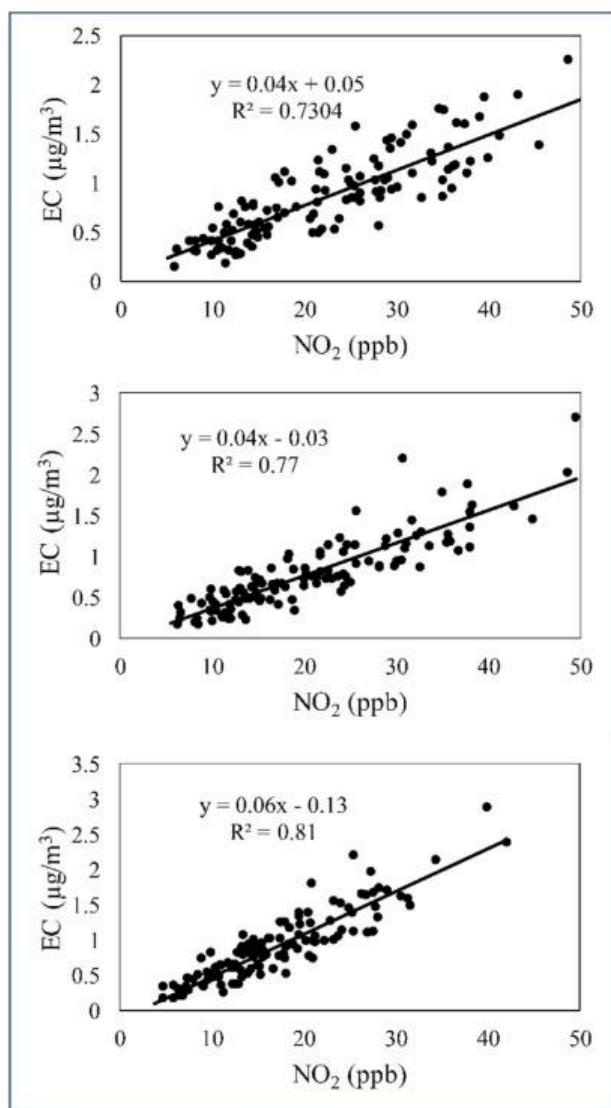


Fig. 8. Linear regression between EC and NO_2 concentrations over the 2015–2019 period in central Los Angeles (CELA).

the NO_2 emissions by 73% by 2025 from 2012 emissions levels (CARB, 2012). Henneman et al. (2021) studied the relationship between various air pollutants and road proximity in the US over long time periods. They reported that NO_2 decreased by ~ 0.29 ppb/yr during 2010–2019, while EC decreased at a relatively constant rate of ~ 0.002 $\mu\text{g}/\text{m}^3$ /yr across 2000–2019 in the proximity of roadsides. They also suggested that NO_2 emission sources, which are far from the roadside, have also declined their emissions since 2010.

4. Summary and conclusions

In this study, the PMF model was implemented to determine the contributing sources to OC mass concentration in central Los Angeles in the period of 2015–2019. We then conducted a regression analysis between elemental, and primary and secondary organic aerosol concentrations resolved by the PMF model with criteria gaseous pollutants in CELA as the means to estimate the 24-hr concentrations of these carbonaceous species. Source apportionment results showed that tailpipe emissions ($38.3 \pm 4.2\%$), non-tailpipe emissions (29.7 ± 2.4), and SOA (22.3 ± 5.4) were the three dominant sources of total OC concentration during the study period in CELA. Moreover, the PMF results showed a decrease in the absolute source contribution of tailpipe

emissions from $\sim 1.5 \pm 0.20$ $\mu\text{g}/\text{m}^3$ to 1.0 ± 0.10 $\mu\text{g}/\text{m}^3$ over the 2015–2019 period, most likely due to the adopted regulations in California. In addition, the regression analysis results revealed a strong correlation between SOA and ozone ($R^2 > 0.74$), which is mainly related to the same precursors (i.e., VOCs) and formation pathways (i.e., photochemical reactions in the atmosphere). The SOA/ O_3 ratios ranged from 47 to 56 $\mu\text{g}/\text{m}^3$ /ppm in the 2015–2019 period. POA concentrations were derived as the difference between total OC and PMF-resolved SOA. According to the regression analysis, the POA/CO ratios decreased from 6.5 to 5 $\mu\text{g}/\text{m}^3$ /ppm from 2015 to 2019, which is the same as the trend of tailpipe emissions to OC mass concentrations in CELA. Lastly, EC was highly correlated with CO ($R^2 > 0.70$) and NO_2 ($R^2 > 0.73$) further corroborating that these species are emitted by the same combustion sources. We should note that our results are based on the available data in a specific site (i.e., CELA) and generalization of these findings to other areas should be done with caution, because the number of factors and emission rates of their sources may vary between different areas. Moreover, the concentrations of these carbonaceous species will also be affected differently by prospective air quality mitigation strategies that might vary among different locations and states. Nevertheless, the correlations explored in this study were intended to provide a simple paradigm for estimating the 24-hr average concentrations of carbonaceous species (i.e., POA, SOA, and EC) based on the reported concentrations of criteria gaseous pollutants; our results could easily be updated or revisited in the future periodically to incorporate the effects of prospective legislative measures and mitigation strategies on the concentrations of primary and secondary organic aerosols in Los Angeles.

CRedit authorship contribution statement

Ramin Tohidi: Conceptualization, Methodology, Data curation, Software, Writing – original draft. **Abdulmalik Altuwayjiri:** Data curation, Methodology, Software, Visualization. **Milad Pirhadi:** Data curation, Methodology, Validation, Writing – review & editing. **Constantinos Sioutas:** Conceptualization, Project administration, Supervision, Writing – review & editing.

Declaration of competing interest

As the first author of the paper, on behalf of all of the co-authors declare that there are no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We would like to acknowledge the financial support by the National Institutes of Health (grant numbers: R01AI065617 and R01ES029395).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2021.118807>.

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