Seasonal variation and source apportionment of inorganic and organic components in PM$_{2.5}$: influence of organic markers application on PMF source apportionment

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Abstract
PM$_{2.5}$ samples collected over a 1-year period in a Chinese megacity were analyzed for organic carbon (OC), elemental carbon (EC), water-soluble ions, elements, and organic markers such as polycyclic aromatic hydrocarbons (PAHs), hopanes, steranes, and n-alkanes. To study the applicability of organic markers in source apportionment, the relationship between organic and inorganic components was analyzed, and four scenarios were implemented by incorporating different combinations of organic and inorganic tracers. The consistent temporal variations trend of 4-ring PAHs and SO$_4^{2-}$ prove that coal burning directly emits a portion of sulfate. The concentrations of ∑5–7-ring PAHs, NO$_3^-$, and NO$_2^-$ show a trend of simultaneous increase and decrease, implying collective impacts from the vehicle source. The concentrations of OC and EC positively correlate with the 5–7-ring PAHs and Cu and Zn, which proves that part of Cu and Zn comes from vehicle emissions. Five factors were identified by incorporating only conventional components, including secondary source (SS, 30%), fugitive dust (FD, 14%), construction dust (CD, 4%), traffic source (TS, 19%), and coal combustion (CC, 14%). Six factors were identified by incorporating conventional components and PAHs, including SS (28%), FD (15%), CD (4%), CC (13%), gasoline vehicles (GV, 12%), and diesel vehicles (DV, 10%). Eight factors were identified by incorporating conventional components, PAHs, hopanes, and n-alkanes, including SS (26%), FD (17%), CD (3%), GV (14%), DV (8%), immature coal combustion (ICC, 5%), mature coal combustion (MCC, 10%), and biogenic source (BS, 1%).

Keywords Organic markers · PMF · PM$_{2.5}$ · Source apportionment

Introduction
Source apportionment of particulate matters (PMs) is the process to identify and quantify the contribution of the emission sources, and it is vital for formulating emission reduction policies for air pollution (Liu et al. 2017, 2016; Lu et al. 2018). However, it is more difficult to identify specific sources based on only the inorganic markers (Galvao et al. 2019; Lin et al. 2010; Xue et al. 2019). Organic compounds in source apportionment can be utilized to identify specific sources. For example, PAHs (polycyclic aromatic hydrocarbons) are generated due to incomplete combustion of organic material, which can be used as markers for the vehicle exhausts, combustion of coal and biomass (Chen et al. 2016; Esmaeilrad et al. 2020; Fraser et al. 2003; Galvao et al. 2019; Lin et al. 2010; Liu et al. 2012; Oros and Simoneit 2000; Pereira et al. 2017; Xue et al. 2019). Hopanes and steranes are emitted by using lubricating oils in gasoline and diesel vehicles, while hopanes are also present in the smoke of coal combustion. Configurations of hopanes can also be employed to identify fossil fuels of different maturity (Krumal et al. 2013; Liu et al. 2012; Pereira et al. 2017). Alkanes with different carbon numbers...
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There are some studies that use both organic and inorganic markers in PMF to improve factors interpretation (Belis et al. 2019; Choi et al. 2015; Esmaeilirad et al. 2020; Galvao et al. 2019; Wang et al. 2015, 2012; Xue et al. 2019). Some studies applied organic markers to identify the sources of OC (Kang et al. 2018; Krumal et al. 2013; Wang et al. 2009, 2017). Several approaches have previously been summarized by using organic markers to distinguish the collinearity sources of PMs, e.g., molecular marker chemical mass balance (MM-CMB) models (Ke et al. 2008; Schauer and Cass 2000; Zheng et al. 2002) and hybrid of positive matrix factorization (PMF) and MM-CMB models (Ke et al. 2008; Lu et al. 2018). PMF works by using sufficient identification information to apportion sources of PM (Dai et al. 2020; Ke et al. 2008). However, when the variables included model increase, the demand for sample size is also greater (Ke et al. 2008). Thus, the sample size is a limitation that has to be considered. Besides, PMF is unable to apportion particulate matter sources with large fractions of low signal-to-noise organic components and few inorganic makers well (Christensen and Schauer 2008; Lu et al. 2018). Simultaneously inputting a large number of organic components and a small number of inorganic components in the PMF model without screening may cause obvious deviations due to the collective influence of element and ions (Lu et al. 2018). As the organics data set increases, the influence of organics closely resembles (Christensen and Schauer 2008). However, how to better incorporate organic and inorganic markers into the PMF model for the purpose of source apportionment has been less investigated.

Despite the benefits of using organic markers in the interpretation of PMF factors, some uncertainty still remains in the source apportionment due to similar temporal variations of the organic markers. Compared with inorganics, organics are more susceptible to meteorological factors (Al-Naiema et al. 2018; Kim et al. 2013). PAHs will undergo reactions such as photolysis, oxidation, desorption, and resorption under the influence of temperature and luminosity (Al-Naiema et al. 2018; Huang et al. 2014; Kim et al. 2013; Mu et al. 2018). Therefore, the reaction activity of organics will affect whether it can be used as a marker (Lin et al. 2010).

In view of the above issues, one-year monitoring of PM$_{2.5}$ components was carried out. Conventional components including elements, ions, OC, EC, and organic components including PAHs, hopanes, steranes, and n-alkanes were analyzed. Four main objectives were established in this paper: (1) to explore the seasonal variation in concentrations and sources of inorganic and organic components; (2) to identify the relations among organic and inorganic markers, as well as the relationship between components and meteorological factors, through their temporal trends and correlation coefficients; and (3) to study the applicability of organic and inorganic markers in the PMF model by incorporating different combinations of organic and inorganic tracers.

**Materials and methods**

**Particulate matter sampling**

The sampling site is located on the 7th floor in the Environmental Protection Building (104°04′E, 30°35′N), which is located in the central city of Chengdu and belongs to residential areas and office areas. There is no obvious pollution source around except for some subway construction around during in the sampling period. PM$_{2.5}$ samples were collected using 90-mm quartz and polypropylene fiber filters by a medium-volume air sampler. The sampling was stopped during rainy and snowy days. There were 80 daily PM$_{2.5}$ samples that were collected in four seasons in 2018, including spring (April), summer (July and August), autumn (October and November), and winter (January, February, and December). Each sampling lasted for 22 h, with a flow rate of 100 L min$^{-1}$. Other conditions of quality control and quality assurance of sampling were the same as those in our previous studies (Xue et al. 2019).

**Filter analysis**

The detailed analysis method and instrument of 7 ions ($\text{NH}_4^+$, $\text{Cl}^-$, $\text{NO}_3^-$, $\text{SO}_4^{2−}$, $\text{Na}^+$, $\text{K}^+$, and $\text{Ca}^{2+}$), 18 inorganic elements (Al, As, Ca, Cd, Co, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, Pb, Si, Ti, V, and Zn), organic carbon (OC), and elemental carbon (EC) are provided in Supplementary context I and Table S1. For organic components, each filter was cut into a quarter for analysis of 18 PAHs, 7 hopanes, 2 steranes, and 20 n-alkanes. Samples were pretreated by ultrasound extraction for 15 min and repeated twice. Then, 10 ml of dichloromethane (DCM) and 10 ml of hexane (1:1,v:v) were added to the tubes. Florisil solid-phase extraction column (1 g/12 mL) was used to clean up the extract and the above fractions and then were eluted with 20 mL of DCM/hexane (1:1). The collected solution after the solid-phase extraction was combined and reduced to less than 5 mL (about 2 mL or 3 mL) using the rotary evaporator. The extract was then solvent-exchanged to hexane, and the volume was finally condensed to 1 mL. The mixed recovery surrogates (naphthalene-d$_8$, acenaphthene-d$_{10}$, phenanthrene-d$_{10}$, chrysene-d$_{12}$, perylene-d$_{12}$, hexamethylbenzene, and n-Tetracosane-d$_{50}$) were spiked into the sample vial. The prepared samples need to be refrigerated at −20 °C and analyzed within 30 days. The organic compounds were analyzed by a gas chromatography/mass spectrometer (GC/MS), which was equipped with a DB-5MS fused-silica capillary
column (30 m × 0.25 mm, 0.25 mm film thickness, Agilent Technology); 1 μL was injected into a splitless injector, and pure helium (purity of 99.99% or more) was used as a carrier gas at a constant flow rate of 1.0 mL min⁻¹.

**Analytical procedure of GC/MS**

To analyze PAHs, hopanes, and steranes, inlet and temperature transmission line temperatures were set to 230 °C and 280 °C, and the initial ionization temperature was set to 280 °C. The chromatography temperature program was set as follows: The column oven temperature was initially held at 50 °C for 2 min, increased to 240 °C at a rate of 10 °C min⁻¹, and finally increased to 280 °C at 5 °C min⁻¹ and then held for 20 min. Mass spectrometry conditions were set as follows: The EI mode was selected, and the ionization energy level was 70 eV. An internal standard (IS) (naphthalene-D8, acenaphthylene-D10, phenanthrene-D10, phenanthrene-D12, pyrene-D10, hexamethylbenzene) was applied to the samples to qualify the actual volumes of the target compounds present. As for n-alkanes, inlet and temperature transmission line temperatures were 300 °C; the initial ionization temperature was 300 °C. Chromatography temperature program followed the steps; column oven temperature was initially held at 50 °C, increased to 60 °C at a rate of 2.5 °C min⁻¹, and then increased to 280 °C at a rate of 22 °C min⁻¹, and finally increased to 325 °C at 30 °C min⁻¹ and held for 11 min.

**Quality assurance and quality control**

Strict quality control procedures were implemented during the experiment. All samples were collected by one instrument and analyzed by the same methods. All instruments and glassware were pretreated before use. When analyzing each batch of samples, method/lab blanks and parallel samples were used to control the data quality. The concentration range of the calibration curve of the studied substance was 0.005–20 mg mL⁻¹, with the correlation coefficients (r) exceeding 0.99. The purity of the standards used were all above 98%, and other basic information is shown in Supplementary Table S2. The recovery rates of most PAHs, hopanes, steranes, and n-alkanes were within 100 ± 20% (Supplementary Table S3). However, the recovery rates of low-molecular-weight PAHs and alkanes were low; due to the higher volatility of low molecular weight PAHs and alkanes, there may be losses during the pretreatment process such as ultrasonic extraction and rotary evaporation steps (Famiyeh et al. 2021; Xue et al. 2019). The limit of detection (LOD) for PAHs, hopanes, steranes, and n-alkanes is given in Supplementary Table S4.

**Data analysis**

SPSS19.0 performs descriptive analysis, principal component analysis, and cluster analysis, and R language performs correlation analysis and visualization. The United States Environmental Protection Agency Positive matrix factorization (US EPA PMF5.0) was used to identify the source categories and estimate their contributions in our study, whose principle has been well introduced in other studies (Ikemori et al. 2021; Liu et al. 2017; Tian et al. 2016, 2014; Yu and Park 2021).

To study the applicability of organic markers in source apportionment, four scenarios were implemented. The EPA PMF5.0 was applied for the source apportionment of PM₂.₅ during the four scenarios. The detailed source apportionment procedure is reported in Supplementary context 2.

1. PMFtra: The first scenario was set by incorporating only conventional components (elements, ions, OC, and EC) into the PMF, which was defined as PMFtra. A total of 18 chemical species of 80 samples were simulated.

2. PMFmore: The second scenario was set by incorporating conventional components and all organic markers (PAHs, hopanes, and n-alkanes) into the PMF, which was defined as PMFmore. The purpose of the second scenario is to investigate how to include as many organic markers as possible, and whether there is a difference between the source apportionment results. A total of 57 chemical species of 80 samples were simulated.

3. PMFless: To study whether the part organic markers can play a role in source apportionment, the third scenario was set by incorporating conventional components and PAHs into the PMF, which was defined as PMFless. A total of 29 chemical species of 80 samples were simulated.

4. PMFscr: In order to minimize the variables included in the PMFscr. This scenario was set by incorporating conventional components and organic markers (PAHs, hopanes, and n-alkanes) after screening into the PMF, which was defined as PMFscr. A total of 40 chemical species of 80 samples were simulated.

**Results and discussion**

**Seasonal characteristics of PM₂.₅ levels and components**

Seasonal trends of particulate and components with concentration during four seasons have been studied. The measured concentrations of PM₂.₅, WSI, elements, OC, EC, and the
sum of organic species are shown in Fig. 1. Concentrations of PM$_{2.5}$ are highest in the winter (114.2 ± 58.0 μg m$^{-3}$) and lowest in the summer (48.2 ± 14.3 μg m$^{-3}$). Concentrations of WSI in the autumn and winter are higher than those in summer and spring. The average concentrations of the WSI in the four seasons are 32.2 ± 20.2 μg m$^{-3}$ (winter), 32.0 ± 17.6 μg m$^{-3}$ (autumn), 20.4 ± 13.6 μg m$^{-3}$ (spring), and 13.6 ± 5.7 μg m$^{-3}$ (summer). Concentrations of NO$_3^-$ are higher in autumn and winter than in spring and summer (Supplementary Fig. S1), which might be caused by the NH$_4$NO$_3$ volatilization in the warmer spring and summer. The total concentrations of the inorganic elements are the highest in spring (14.5 ± 5.1 μg m$^{-3}$ and 4.6 ± 1.6 μg m$^{-3}$) and winter (14.7 ± 7.5 μg m$^{-3}$ and 4.5 ± 2.8 μg m$^{-3}$) are higher than those in summer (13.6 ± 5.8 μg m$^{-3}$ and 4.1 ± 1.6 μg m$^{-3}$) and spring (14.0 ± 7.5 μg m$^{-3}$ and 3.9 ± 2.9 μg m$^{-3}$). However, a slight difference was found in the seasonal variation of the sum of organic species (PAHs, hopanes, steranes, and n-alkanes), which the lowest concentration is observed in autumn. The annual average concentrations of the sum of organic species are approximately equal to 0.6 μg m$^{-3}$.

**Seasonal characteristics of organic components**

The PAHs concentrations of four seasons are presented in Fig. 2. The annual average concentration of total PAHs is 7.4 ng m$^{-3}$. The total PAHs are higher in winter (13.0 ng m$^{-3}$) than in spring (4.9 ng m$^{-3}$), autumn (3.9 ng m$^{-3}$), and summer (2.8 ng m$^{-3}$). DBA is the most abundant PAH (17–23% of total PAHs) in four seasons, followed by BbF (6–16%) and BghiP (9–13%). These results echo those of Ma et al. (2018), who found a higher concentration of PAHs in the winter period than in the spring and summer.

The mean concentrations of hopanes, ααα(20R)-cholestane, and ααα(20S)-cholestane in four seasons are shown in Fig. 3. The sum concentrations of hopanes and steranes are 7.7 ng m$^{-3}$, 7.6 ng m$^{-3}$, 5.5 ng m$^{-3}$, and 4.6 ng m$^{-3}$ in spring, winter, summer, and autumn, respectively. Like
PAHs, the high concentration in winter is mainly due to larger emissions from coal combustion and poor dispersion (e.g., weaker winds and lower temperature inversions). The concentration of hopanes is approximately 10 times that of steranes (Supplementary Fig. S3). \(17\alpha(H),21\beta(H)\)-Hopane is the most abundant hopane, followed by \(17\alpha(H),21\beta(H)-30\text{-norhopane}\). \(17\alpha(H),21\beta(H)\)-Hopane can be used as a marker for mature fossil fuels, and \(17\alpha(H),21\beta(H)-30\text{-norhopane}\) can indicate the maturity of coal combustion sources, such as anthracite.

Concentrations of the n-alkanes in four seasons are plotted in Fig. 4. A total of 20 n-alkanes (ranging from C14 to C33) are detected. The total n-alkane concentrations in PM2.5 range from 161.3 to 2020.4 ng m\(^{-3}\). The total concentrations of n-alkanes (Supplementary Fig. S4) in summer (854.3 ng m\(^{-3}\)) are higher than those in spring (652.9 ng m\(^{-3}\)), autumn (651.2 ng m\(^{-3}\)), and winter (586.6 ng m\(^{-3}\)). Concentrations of C14 to C21 n-alkanes are higher in spring and winter than those in summer and autumn (Fig. 4). Studies have shown that n-alkanes with lower carbon numbers (C \(\leq\) 24) are mainly from the combustion of fossil fuels, such as vehicle emission and coal combustion (Ling et al. 2008; Lyu et al. 2017; Tian et al. 2021; Wang et al. 2009). Concentrations of C22–C33 n-alkanes are higher in summer than those in autumn, spring, and winter (Fig. 4). High-carbon n-alkanes (C \(\geq\) 25) are mainly from higher plant wax emissions and other biogenic sources such as pollen and spores (Lyu et al. 2017; Wang et al. 2009).

**Diagnostic ratios of organic components**

The PAHs diagnostic ratios for four seasons (Table 1) are obtained since they can prove some emission sources, such as gasoline vehicles, diesel vehicles, coal, or wood combustion (Famiyeh et al. 2021; Krumal et al. 2013). The values of PAHs ratios can also be changed by the phase, transport, and degradation of PAHs (Ma et al. 2018). The ratio of BeP/BaP is related to aerosol photolysis. BeP and BaP emissions from the local PAHs contain are similar. However, BaP is more unstable and is likely to undergo photolysis or oxidation (Famiyeh et al. 2021; Kong et al. 2018; Jin et al. 2019; Iakovides et al. 2019; Oliveira et al. 2011; Pacheco et al. 2017; Pereira et al. 2017). The average BeP/BaP ratio is 1.4 in winter, 1.8 in autumn, 2.0 in spring, and 2.1 in summer, respectively. The concentration of BeP is higher than that of BaP, especially in spring and summer, mainly due to the photolysis or oxidation of the BaP in summer and spring caused by the high temperature and sufficient sunlight (Pereira et al. 2017). This suggested that the PAHs found at the site are aged. The BaP/BghiP and IcdP/(IcdP + BghiP) ratios are close to 0.4–0.7 and 0.2–0.4, similar to the ratio for the gasoline vehicles and diesel vehicle presented in Table 1. The average Phe/Ant ratio ranged between 5.6 and 7.7 in the four seasons, also approaching that of the gasoline vehicles and diesel vehicle (Cao et al. 2019; Fang et al. 2020; Famiyeh et al. 2021; Gune et al. 2019; Hu et al. 2019).

The homohopane index (22S/(22S + 22R)) of hopanes is shown in Table 1, which is much greater in summer (0.5) than in spring (0.2), autumn (0.1), and winter (0.2). The homohopane index is in the range of 0.2–0.3 for winter samples in Brno, Czech Republic, while which is higher (0.4–0.5) for summer (Krumal et al. 2013). The homohopane index is 0.3 for winter and 0.6 for summer in Baoji, China (Wang et al. 2009). It was reported that the homohopane index (22S/(22S + 22R)) for coal smoke samples increases with coal rank (lignite 0.05; brown coal 0.09; sub-bituminous coal 0.20; bituminous coal 0.35) (Oros and Simoneit 2000), and the homohopane index (22S/(22S + 22R)) for traffic emissions is about 0.5–0.6 (Tian et al. 2021). This implies vehicle exhausts contributed most to hopanes in summer; however, coal combustion also contributed to hopanes in
other seasons (Tian et al. 2021). Diagnostic ratios of C29αβ/C30αβ are 0.6–0.7 in four seasons, which are in the range of traffic emissions. As shown in Fig. 4 and Table 1, the Cmax of n-alkanes are C16, C18, and C28 in winter, while the Cmax of n-alkanes are at C28 and C30 in summer. Above all, n-alkanes in Chengdu during spring and winter mainly come from coal combustion and vehicle emissions, while n-alkanes are partly influenced by biogenic sources in summer and autumn. Previous studies showed that C29 and C31 mainly come from the emission of plant wax (Ling et al. 2008; Wang et al. 2009). The Cmax of gasoline vehicles are mainly C18 and C25, and the Cmax of diesel vehicles is C20 (Ling et al. 2008; Lyu et al. 2017; Wang et al. 2009).

Correlations of components in PM$_{2.5}$

Temporal variations of concentrations of conventional and organic components and the meteorological factors (T (temperature), relative humidity (RH), and wind speed (WS)) during the sampling period were presented in Supplementary Figs. S5 and S6. The meteorological parameters were downloaded from a website (https://rp5.ru/). Supplementary Fig. S5a presents that the concentrations of $\Sigma$5–7-ring PAHs, NO$_3^-$, and NO$_2$ have a consistent temporal variations trend, showing a trend of simultaneous peak and valley concentrations. The results show that 5–7-ring PAHs are potentially affected by the vehicle source in this study (Lin et al. 2010; Pereira et al. 2017; Xue et al. 2019). Supplementary Fig. S5b presents that the concentrations of $\Sigma$4-ring PAHs and SO$_4^{2-}$ have consistent temporal variations. Four-ring PAHs (like pyrene and benzo(a)anthracene) have been used as markers for coal emissions (Lin et al. 2010; Liu et al. 2012; Oros and Simoneit 2000). Additionally, it can be reported that burning coal can directly emit a portion of sulfate (Dai et al. 2019).

The concentrations of OC and EC exhibit a weak correlation with the sum of 5–7-ring PAHs (0.52, p value < 0.05) and Cu and Zn (0.51 and 0.70, p value < 0.01) (Supplementary Table S5), which prove that part of Cu and Zn comes from vehicle emissions; 5–7-ring PAHs are markers for traffic (Lin et al. 2010; Pereira et al. 2017; Xue et al. 2019), which are released by incomplete combustion of fossil fuels. Unlike 5–7-ring PAHs, Cu and Zn are mainly from tires and brakes wear or other vehicle parts, not fuel combustion (Birmili et al. 2006; Guan et al. 2018).

Supplementary Fig. S6 presents that the concentrations of ions and PAHs have an opposite temporal variations trend with temperature. Cl$^-$, NO$_3^-$, NH$_4^+$, and 2–4-ring PAHs are more significantly affected by temperature (Zang et al. 2021). The photochemical and secondary reactions of the most reactive species (such as 2–4-ring PAHs, NO$_3^-$, and NH$_4^+$) are more easily occurring at high temperatures (Hsu et al. 2019). Besides, due to the higher volatility of low molecular weight PAHs (2–4-ring PAHs), as the temperature increases, the G/P distribution behavior leads it to more favorably partition into the gas phase (Hsu et al. 2019). However, high-carbon n-alkanes (C29-C33) have a consistent temporal variations trend with temperature, which is consistent with that the high-carbon n-alkanes are mainly from higher plant wax emissions and other biogenic sources such as pollen and spores, which are mainly strong in the higher temperature season (Lin et al. 2008; Tian et al. 2021).
Table 1 Diagnostic ratios of organic components

|                      | Gasoline vehicle | Diesel vehicle | Brown coal | Sub-bituminous coal | Bituminous coal | Wood burning | Road dust | References                                                                 |
|----------------------|------------------|----------------|------------|---------------------|----------------|--------------|----------|-----------------------------------------------------------------------------|
| BaP/BghiP            | 0.5–0.8          | 0.3–0.4        |            |                     |                |              |          | Famiyeh et al. (2021); Gao and Ji (2018); Gune et al. (2019)               |
| BeP/BaP              | 1.1–1.3          | 2.0–2.5        | 0.8–1.6    | 0.4                 | 1.4            |              |          | Famiyeh et al. (2021); Kong et al. (2018); Jin et al. (2019); Iakovides et al. (2019); Oliveira et al. (2011); Pacheco et al. (2017); Pereira et al. (2017) |
| Phe/Ant              | 3.4–8.0          | 7.6–8.8        | 3          |                     | 8              |              |          | Cao et al. (2019); Fang et al. (2020); Famiyeh et al. (2021); Gune et al. (2019); Hu et al. (2019) |
| IcdP/                 | 0.1–0.2          | > 0.3          | 0.58       | 0.6                 |                |              |          | Fang et al. (2020); Famiyeh et al. (2021); Gurkan Ayyildiz and Esen (2020); Iakovides et al. (2019); Kubo et al. (2020); Zhang et al. (2018) |

|                      | Spring | Summer | Autumn | Winter |
|----------------------|--------|--------|--------|--------|
| BaP/BghiP            | 0.4    | 0.6    | 0.5    | 0.7    |
| BeP/BaP              | 2      | 2.1    | 1.8    | 1.4    |
| Phe/Ant              | 6.6    | 6.7    | 5.6    | 7.7    |
| IcdP/                 | 0.4    | 0.4    | 0.3    | 0.2    |
| Table 1 (continued) | Diesel vehicle | Brown coal | Sub-bituminous coal | Bituminous coal | Wood burning | Road dust | References |
|---------------------|----------------|------------|---------------------|----------------|--------------|-----------|------------|
| Fluo/Fla+pyr        | 0.4            | 0.6–0.7    | 0.5                 | 0.5            | 0.5          | Famyeh et al. (2021); Iakovides et al. (2019); Tian et al. (2013); Wang et al. (2017) |
| Cmax                | C10, C25       | C20        | >C29                | C24            | C29αβ/C30αβ  | Iakovides et al. (2021); Krumholz et al. (2016); Oros and Simoneit (2000); Tian et al. (2013); Wang et al. (2017) |
| CPI                 | 0.6–0.7        | 0.4        | 0.9–1.2             | 0.9            | 0.7          | Iakovides et al. (2021); Krumholz et al. (2016); Oros and Simoneit (2000); Tian et al. (2013); Wang et al. (2017) |
| 22S/(22S+22R)       | 0.6            | 0.5        | 0.1–0.4             | 0.2            | 0.5          | Iakovides et al. (2021); Krumholz et al. (2016); Oros and Simoneit (2000); Tian et al. (2013); Wang et al. (2017) |
2021; Lyu et al. 2017; Wang et al. 2009). All components present opposite temporal variations trend with wind speed in winter, showing a trend of higher concentrations with lower speed, and low concentrations with high speed.

Source characterization

Identification of PMF factors

US EPA PMF5.0 was used to realize source apportionment in four scenarios. Bootstrap (BS) and displacement of factor elements (DISP) were run to estimate the uncertainty of receptor modeling (Supplementary Table S6). The results of scenario 2 show that when a large number of components (conventional components and all organic markers without screening) are included, the co-linearity of the sources increases due to the large correlation between some organic components (Supplementary Fig. S7). That is, the model extracts organic components uniformly to one or two factors, thereby masking the identity of the organic components, making it impossible to distinguish the sources by its identity (Supplementary Fig. S8). Therefore, based on the analysis of the correlation between organic and inorganic markers and between organic and organic markers, the components included in the PMF were screened. Figure 5 and Supplementary Fig. S9 show the source profiles in three scenarios (PMFtra, PMFscr, and PMFless). The source categories were identified as follows.

1. Secondary source

Factortra1 (PMF model by incorporating only conventional components), Factorscr1 (PMF model by incorporating conventional components and organic markers (PAHs, hopanes, and n-alkanes after screening), and Factorless1 (PMF model by incorporating conventional components and organic markers (only PAHs)) are characterized by high loadings of SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and OC. Secondary nitrate formed via the ammonia and NO$_3^-$, and secondary sulfate was produced by ammonia and SO$_4^{2-}$ (Dai et al. 2021; Zang et al. 2021). Photochemical reactions along with the precursors like NOX and volatile organic compounds lead to the formation of secondary organic aerosol (Bari and Kindzierski 2017). Thus, SO$_4^{2-}$, NO$_3^-$, NH$_4^+$, and OC are considered as markers of sulfate and nitrate and SOA (Dai et al. 2020; Xue et al. 2019). Therefore, Factortra1, Factorscr1 and Factorless1 are called as secondary source in this study.

2. Fugitive dust

Factortra2, Factorscr2, and Factorless2 are characterized by large percentage contributions to Si, Ca, and Al, as well as some Fe, Cu, and SO$_4^{2-}$. Fugitive dust

![Fig. 5](image-url)
is entrained into the atmosphere by both natural and anthropogenic sources such as soil dust and other anthropogenic activities (Bi et al. 2019). Elements like Si, Al, Ca, Fe, and Cu are typical markers of fugitive dust (Bi et al. 2019; Dai et al. 2019; Liu et al. 2017). Thus, Factortra3, Factortra4, and Factortra5 are characterized as urban fugitive dust.

(3) Construction dust

Factortra3, Factortra5, and Factortra7 are characterized by the high weights of Si and Ca as well as some Na, Mg, and NO3−. Construction dust is mainly generated by construction activities because the construction site has not been hardened (Yang et al. 2020). Elements like Si, Ca, Na, and Mg are considered as markers of construction dust (Bi et al. 2019). Thus, Factortra3, Factortra5, Factorma1, and Factorma3 are characterized as construction dust.

(4) Traffic source

Factortra4 is characterized by high proportions of OC, EC, Al, Si, Ca, Cu, Fe, and Zn. Vehicle emissions are likely to be the predominant source of PM2.5 in urban cities in China in recent years (Bi et al. 2019; Cui et al. 2016; Zhang et al. 2015). Chengdu’s vehicle population reached more than four million in 2016, rendering Chengdu second only to Beijing in terms of levels of car ownership (Shi et al. 2018; Kong et al. 2020). It has been shown that vehicle emissions have a large amount of OC, EC, NO3−, and NH4+ (Bi et al. 2019; Sun et al. 2021; Tian et al. 2021). The fractions of Al, Si, and Ca may be associated with the resuspension of road dust (Bi et al. 2019). Cu, Fe, and Zn are indicative of tire and brake wear (Birmili et al. 2006; Guan et al. 2018; Bi et al. 2019; Sun et al. 2021; Tian et al. 2021). Thus, Factortra4 is finally identified as the traffic source, which includes exhaust and non-exhaust.

By incorporating PAHs, vehicles can be refined into gasoline and diesel vehicles in PMFscr4 and PMFless4. Factortra4 is characterized by high percentage concentrations of OC, EC, NO3−, BbF, BkF, BaP, 17α(H),21β(H)-hopane, C16, C18, and C25. The concentration of OC to EC ratio is higher than 2 (3.0) in PMFscr4. BbF, BkF, BaP, 17α(H),21β(H)-hopane, C16, C18, and C25 have been regarded as markers of gasoline vehicles (Chen et al. 2016; Choi et al. 2015; Galvao et al. 2019; Lin et al. 2010; Wang et al. 2021). Factorma1 is also characterized by high BbF, BkF, BaP, and IcdP. The concentration of OC to EC ratio is 3.5 in PMFless4. Thus, Factortra4 and Factorma1 are identified as gasoline vehicles.

Factortra5 is characterized by the high weights of OC, EC, Flt, Pyr, BbF, IcdP, DBA, BghiP, Cor, 17α(H),21β(H)-hopane, C16, C18, C20, and C28; the concentration of OC to EC ratio is lower than 1 (0.86) in Factortra5. Diesel vehicles generally emit more EC than OC, and more light PAHs (Flt, Pyr, BbF), and the n-alkanes in highest abundance were found to be C20 (Chen et al. 2016; Fraser et al. 2003; Kang et al. 2018; Lin et al. 2010; Pereira et al. 2017). Factorless5 also presents high fractions of OC, EC, BaA, Ant, and DBA. The concentration of OC to EC ratio is 0.66 in Factorless5. Therefore, Factortra5 and Factorless5 are identified as the diesel vehicles source.

(5) Coal combustion

Factortra5 is characterized by high loadings of OC, EC, and SO42−, as well as some Si and Ca. Coal is the main fuel used in China and is widely used in coal-fired power plants, coal-fired industrial boilers, and residential household stoves (Bi et al. 2019). The source profiles of coal combustion in China mainly consist of crustal materials, OC, EC, and SO42− (Belis et al. 2019; Bi et al. 2019; Xue et al. 2019; Tian et al. 2021). Thus, Factortra5 is identified as coal combustion.

Factorma4 is characterized by high loadings of OC, EC, SO42−, Si, Ca, Flt, Pyr, BaA, BbF, 17α(H),21β(H)-30-norhopane, 17α(H),21β(H)-hopane, C24, and C27. Four-ring PAHs have been regarded as markers of coal combustion (Belis et al. 2019; Bi et al. 2019; Xue et al. 2019; Tian et al. 2021). 17α(H),21β(H)-30-Norhopane can identify the marker of combustion of maturity coal (Bi et al. 2008; Oros and Simoneit 2000; Tian et al. 2021). The Cmax of n-alkanes emitted by bituminous coal and anthracite are C27 and C24 (Choi et al. 2015; Galvao et al. 2019; Oros and Simoneit 2000). Therefore, Factorma4 is identified as the mature coal combustion. Factorma7 is characterized by high loadings of OC, EC, SO42−, Si, Ca, Flt, Pyr, BaA, BbF, 17β(H),21β(H)-hopane, C16, C18, and C25. 17β(H),21β(H)-hopane, C16, and C18 have been regarded as markers of immature coal combustion source (Bi et al. 2008; Choi et al. 2015; Lin et al. 2010; Wang et al. 2009, 2015). Thus, Factorma7 is identified as immature coal combustion. Factorless5 is characterized by high loadings of OC, EC, SO42−, Si, Ca, Flt, Pyr, BaA, and BbF. Thus, Factorless6 is identified as the coal combustion. Therefore, we can conclude that by incorporating PAHs, hopanes, and n-alkanes, the PMFscr model distinguishes coal combustion source into mature coal combustion and immature coal combustion sources.

Biogenic source

The profiles of Factorma5 are characterized by high proportions of high-carbon n-alkanes, especially C29, C30, and C31. In contrast to n-alkanes derived from fossil fuel combustion, which are dominated by low molecular weight ones (<C25), n-alkanes originated from plant wax are dominated by high molecular weight ones (>C25) (Lin et al. 2010; Wang et al.
Thus, Factor$_{scr}$ is finally identified as the biogenic source. This factor was not recognized by PMF$_{tra}$ and PMF$_{less}$ due to the lack of incorporation of suitable organic markers.

**Comparison of source apportionment results of three scenarios**

Figure 6 presents the average percentage of PM$_{2.5}$ mass concentrations estimated by the PMF for the three scenarios. The secondary sources, urban fugitive dust, and construction dust share the same average percentage of PM$_{2.5}$ in the three scenarios. As for the coal combustion source, the average percentage of PM$_{2.5}$ mass concentrations obtained by PMF$_{tra}$ is 14%; PMF$_{less}$ incorporating PAHs and conventional components is 13%. The summed percentage of mature coal combustion and immature coal combustion source obtained by PMF$_{scr}$ is 14%; that is, the apportionment results of coal combustion source in the three scenarios are relatively consistent. The average percentage of PM$_{2.5}$ mass concentrations of traffic source obtained by PMF$_{tra}$ is 19%; the average percentage of PM$_{2.5}$ in gasoline vehicles and diesel vehicles obtained by PMF$_{less}$ is 22%. The summed percentage of gasoline and diesel vehicles source obtained by PMF$_{scr}$ is 22%; that is, the apportionment results of traffic source in the three scenarios are basically consistent.

PMF$_{tra}$, PMF$_{less}$, and PMF$_{scr}$ input different components to obtain relatively consistent results, indicating that the source apportionment results after incorporating organic markers are reliable. Correlation between modeled and measured PM$_{2.5}$ concentrations of the three scenarios are also exhibited in Supplementary Fig. S10. Comparing the correlation between modeled and measured PM$_{2.5}$ concentrations of the three PMF models, the source apportionment results of the three models are all good. Therefore, by including the organic components such as PAHs, this study can well distinguish motor vehicle sources into diesel vehicle sources and gasoline vehicle sources. By including the organic components such as PAHs, hopane, and n-alkanes, this study can well distinguish motor vehicle sources from diesel vehicle sources and gasoline vehicle sources and at the same time can distinguish coal combustion sources into maturity coal combustion and immaturity coal combustion sources. What is more, the source apportionment results have certain reliability.
Conclusions

This study explored the seasonal variations of inorganic and organic components in PM$_{2.5}$ and identified the relationship among organic markers, inorganic markers, and meteorological factors. What is more, this study carried out research on the applicability of organic markers and inorganic markers in the PMF model.

The highest and lowest concentrations of conventional components (PM$_{2.5}$, WSI, tracer elements, OC, and EC) were observed in winter and summer, respectively. While the highest concentration of the sum of organic species (PAHs, hopanes, steranes, and n-alkanes) was observed in winter and spring, and the lowest concentration was in autumn.

By including the organic markers such as PAHs, this study can well distinguish motor vehicle sources into diesel vehicle sources and gasoline vehicle sources. If the organic markers (such as PAHs, hopane, and n-alkanes) are included, this study can well distinguish motor vehicle sources into diesel vehicle and gasoline vehicle sources and at the same time distinguish coal combustion sources into maturity coal combustion and immaturity coal combustion sources, and the source apportionment results have certain reliability.

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Data availability All relevant data are within the manuscript and available from the corresponding author upon request.

Declarations

Ethics approval Not applicable.

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