Chemical Composition Characteristics and Source Apportionment of PM$_{2.5}$ in Ceramic Industrial Base during Winter

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ABSTRACT

The national architectural ceramic industrial center in east China is suffering from serious ambient fine particle pollution. The study reported herein describes an effort to ascertain the degree and sources of the PM$_{2.5}$ collected in a ceramic industrial base during winter. The major chemical components in PM$_{2.5}$ were analyzed, including carbonaceous aerosols, water-soluble ions, and inorganic elements. The chemical mass balance (CMB) model, backward trajectory method and potential source contribution function model, etc. were used to track and identify possible sources and contributions in the formation of the PM$_{2.5}$. The results showed that the average PM$_{2.5}$ concentration during sampling period was 134 $\pm$ 74.7 $\mu$g m$^{-3}$, which exceeding World Health Organization (WHO) Air Quality Guidelines levels. The dominant components in the PM$_{2.5}$ at this sampling site were found to be secondary ions (sulfate and nitrate) and carbon fractions. Water-soluble ions and total carbon contributed about 48.7% and 13.9% of the PM$_{2.5}$ mass, respectively. In addition, the $\text{SO}_4^{2-}/\text{NO}_3^-$ ratio in the ambient PM$_{2.5}$ during the sampling period was 1.16, indicating that it was the result of primarily emissions from stationary sources. Furthermore, source apportionment using the CMB model indicated that a ceramic industry source was the main contributor to the PM$_{2.5}$ mass, which accounted for about 27.9%, and this was followed by secondary formation dust sources, and gasoline/diesel vehicle exhaust emissions and motor vehicle non-exhaust emissions. Based on the backward trajectory analysis and potential source apportionment, it was found that PM$_{2.5}$ regional transmission existed, but it originated primarily from local sources and surrounding areas. Hence, this study provided a scientific basis for identifying the sources of PM$_{2.5}$ pollution during a typical pollution period and provided important input for PM$_{2.5}$ control strategies in a typical industrial area.

Keywords: Ceramic industry bases, PM$_{2.5}$, Chemical composition characteristics, Source apportionment, CMB

1 INTRODUCTION

Although China’s air quality has improved in recent years as a result of the implementation of the Air Pollution Prevention and Control Action Plan, the ambient PM$_{2.5}$ (particulate matter with an aerodynamic diameter of less than 2.5 $\mu$m) levels in most industrial areas and cities far exceeds...
the air quality guidelines of the World Health Organization (WHO) in which the daily average PM$_{2.5}$ concentration is set at the level of 15 $\mu$g m$^{-3}$ (Cai et al., 2017; Lin et al., 2018). Moreover, over 80% of the Chinese population has been exposed to a harmful environment in which PM$_{2.5}$ levels have exceeded the 35 $\mu$g m$^{-3}$ standard (Li et al., 2017). PM$_{2.5}$ is associated with many adverse health effects (Bai et al., 2018). It also affects the formation processes of clouds and rainfall, and indirectly affects climate change (Nguyen et al., 2019; Zhao et al., 2020). The chemical composition of PM$_{2.5}$ is as complex as its sources (Heo et al., 2013). It is composed primarily of water-soluble inorganic ions, organic carbon (OC), elemental carbon (EC), crust and trace inorganic elements and other components. Therefore, studying the chemical composition and source apportionment of ambient PM$_{2.5}$ in a select area is a critical way to provide a sound scientific basis for human health protection and air pollution control. On the other hand, due to air pollution in economically developed cities in recent years, some heavily polluting industries have been transferred to underdeveloped areas. Exploring the impact of these industries on local air quality pollution is very important and is urgently needed to control of regional air pollution and improvement the quality of ambient air for local residents (Heo et al., 2013; Cai et al., 2017).

Particulate matter (PM) from the ceramic industry has accounted for a relatively high proportion of industrial source emissions (da Silva et al., 2019), so that the ceramic industry is one of the key focal points for the control and prevention of air pollution in China. The main exhaust gases in the ceramic industry contain a higher amount of particulates, particularly in the case of the drying process. From either the perspective of emission factor or source analysis, the impact of ceramic industry pollutant emissions on ambient air quality is significant (Bai et al., 2018; Zhou et al., 2018; da Silva et al., 2019). The high degree of PM pollution from the ceramic industry has a detrimental effect on human health (da Silva et al., 2019; Bessa et al., 2020). da Silva et al. (2019) found that the PM emitted from the ceramic industry caused human DNA damage and produced a genotoxic effect in the meiotic pollen cells of Tradescantia pallida. Bessa et al. (2020) also found that the ceramic manufacturing had a significant potential to generate airborne particles, particularly ultrafine particles (UFP; < 100 nm), which placed workers in this industry at great risk due to exposure to these particles. In addition, the composition of PM emitted by the ceramic industry has significant regional characteristics as a result of the types of raw materials used and local soil. Therefore, the study of source analysis related to ceramic industry is very important for local source apportionment and pollution prevention and control.

Identifying and quantifying the main sources of PM$_{2.5}$ can provide a scientific and technological basis for air quality management and air pollution control. There are a number of widely used receptor models that can be used to achieve this goal (Watson et al., 1984; Bullock et al., 2008; Wang et al., 2021a). These include CMB model (Pipalatkar et al., 2014), enrichment factor method (EF) (Anil et al., 2019), principal factor analysis (PCA) (Wu et al., 2019) and positive definite matrix factor (PMF) model (Tian et al., 2020; Hassan et al., 2021). Police et al. (2018) used the PMF model for source analysis and found that anthropogenic sources were clearly the major contributor to PM$_{2.5}$, but natural sources were reported to be the chief contributor to PM$_{2.5}$-10. Wang et al. (2019) found that amended CMB method could separate different fugitive dusts and obtain good results: secondary sulfate contributes the most to PM$_{2.5}$ pollution in Chinese eastern coastal cities at 44.50%. Liang et al. (2019) also used the PMF model and identified the four sources of PM$_{2.5}$ at a sampling site and the OC and EC accounted for the main proportion of the PM$_{2.5}$ mass, followed by secondary inorganic aerosols and coal combustion. Although the CMB and PMF receptor models have been commonly used to investigate the origin of ambient PM (Police et al., 2018; Liang et al., 2019; Tian et al., 2020; Hassan et al., 2021), to the best of our knowledge, there are few reports on the use of CMB model to determine the major sources of PM$_{2.5}$ sampled in a typical Ceramic Industrial Base. Moreover, the backward trajectory method, potential source contribution function model, etc. can also be used to analyze if the local ambient air pollution is effect more by local sources contribution or by transmissions.

The goal of this present study was to characterize the ambient PM$_{2.5}$ of a typical ceramic industry base, and determine the contributions by the significant local sources and transmissions to the ambient PM$_{2.5}$ in this ceramic industrial base. To accomplish this, the CMB model and backward trajectory method was used together with the potential source contribution functions model, etc. The results of this study could provide an important scientific basis for health protection and air pollution control in a typical ceramic industrial base.
2 MATERIALS AND METHODS

2.1 Ambient Samples

In this reported study, the roof of the Gao’an Mingzhu business hotel (a height of 28 m above ground; Table 1) near the management committee of selected ceramic industry was chosen as the sampling site (Fig. 1). Air samples were collected from December 23, 2020 to January 06, 2021, and the sampling period was from 09:00 AM to 09:00 AM the next day. The samples were collected using two PM$_{2.5}$ samplers (Qingdao Laoshan Institute of Applied Technology, Qingdao, China), with a sampling flow rate of 100 L min$^{-1}$, and a total of 15 PM$_{2.5}$ samples were collected. The daily average sampling volume of PM$_{2.5}$ samples is 107 ± 34.1 m$^3$. It is noting that the Quartz fiber have long been used to collect airborne particulate matter for subsequent determination of carbon content by thermal analysis. And the Teflon filter membrane have long been used to collect airborne particulate matter for subsequent determination of inorganic components (Si element), which can accurately obtain the concentration of inorganic components of PM$_{2.5}$. Quartz fiber filter and Teflon filter membranes were used in parallel to collect particulate matters.

Table 1. Sampling points in Gao’an ceramic industrial base.

<table>
<thead>
<tr>
<th>Name of sampling point</th>
<th>Coordinate position</th>
<th>Number of sampling medium</th>
<th>Functional positioning</th>
<th>Height from the ground (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Roof of the Mingzhu Hotel</td>
<td>115°26'4.68&quot;E</td>
<td>Quartz fiber membrane</td>
<td>industrial base</td>
<td>28</td>
</tr>
<tr>
<td></td>
<td>28°10'57.66&quot;N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Roof of the Mingzhu Hotel</td>
<td>115°26'4.68&quot;E</td>
<td>Teflon membrane</td>
<td>industrial base</td>
<td>28</td>
</tr>
<tr>
<td></td>
<td>28°10'57.66&quot;N</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 1. Sampling site.
The Quartz filter membranes were purified in a muffle furnace at 550°C for 4 hr before being used. The Teflon filter membranes were dried in muffle furnace at 60°C for 2 hr. Then the filter membranes were weighed before and after sampling using a balance after being exposed to 48 hr of constant temperature and humidity (temperature of 25°C, humidity of about 25%).

2.2 Chemical Analysis

The mass concentration of PM$_{2.5}$ was determined using the gravimetric method (Liang et al., 2019). The OC and EC in PM$_{2.5}$ were determined using a DRI-2015 thermal/optical carbon analyzer (Atmoslytic Inc., USA). The water-soluble ions (NO$_3^-$, SO$_4^{2-}$, Cl$^-$, F$^-$) in PM$_{2.5}$ were detected using an ion chromatograph (Ion Chromatography, IC, Dionex 600). The Na$^+$, Ca$^{2+}$, K$^+$, and Mg$^{2+}$ ions were detected using an inductively coupled plasma emission spectrometer. NH$_4^+$ was determined by using a spectrophotometer. The elements (Zr, Al, Sr, Mg, Ti, Ca, Fe, Ba, and Si) were analyzed using a VISTA-MPX plasma spectrometer; while Li, Be, Na, P, K, Sc, V, Cr, Mn, etc. were distinguished using an Agilent 7500a ICP-AES.

2.3 Quality Control

Quality control during sampling consisted of: 1) before sampling, the flow rate of the sampling device was calibrated to ensure its normal operation during sampling; 2) it is critical to ensure that the filter membrane is complete and there are no membrane impurities; 3) each membrane was weighed 3 times in parallel with the error margin of 50 µg.

2.4 Data Handling for Models

To determine the sources of ambient PM$_{2.5}$ of the ceramic center during the sampling period, the principal component analysis (PCA) method was employed to conduct a semi-quantitative identification analysis (Wang et al., 2012). This method analyzes the chemical components contained in ambient PM$_{2.5}$ based on the concept of dimensionality reduction. First, Z-score standardized processing was performed of the various components in ambient PM$_{2.5}$, with the calculation performed using the following Formula (1) (Wang et al., 2012). Then the maximum variance rotation method was utilized to reduce the components of ambient PM$_{2.5}$ into several major factors and extract the characteristic value and variance of each factor. The component with a greater load value for each factor enjoys greater representation. The communality of each component is the contribution of all the extracted factors to the variance in this component, indicating that all factors contribute to the variance of this component.

$$Z_{ik} = \frac{C_{ik} - C_i}{S_i}$$

where, $Z_{ik}$ is the standard score of component $i$ in sample $k$; $C_i$ is the mass concentration of component $i$ in sample $k$; $C_i$ is the arithmetic mean value of component $i$; $S_i$ is the standard deviation of components of sample $i$.

Sources of the ambient PM$_{2.5}$ in ceramic industrial base during the sampling period were allocated using the CMB software (NKCMB1.0) developed by the National Key Laboratory of Prevention and Control of Urban Air Particle Pollution for Environmental Protection (Shi et al., 2011; Fang et al., 2017). The CMB program produces an effective-variance-least-squares solution to the linear combination of the product of the source contribution and its concentration (Watson et al., 1984). Molecular marker species employed in this analysis were assumed to be stable during transport from source to receptor. Since NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$ are present primarily in the form of NH$_4$NO$_3$ and (NH$_4$)$_2$SO$_4$, these species were used as the source profiles of the "pure" secondary sulfates and secondary nitrates in the secondary inorganic aerosol. The sources profiles of gasoline vehicle exhaust emissions, diesel vehicle exhaust emissions, motor vehicle non-exhaust emissions and dust sources were based on the PM$_{2.5}$ component profiles of these sources reported in the literatures (Watson et al., 1984; Shi et al., 2011; Fang et al., 2017).

To analyze the regional transport of PM$_{2.5}$ during the sampling period, the Backward Trajectory (HYSPLIT) analysis was used to calculate and analyze the transport and diffusion trajectories of atmospheric pollutants. The HYSPLIT model was developed by the United States National Oceanic
and Atmospheric Administration (NOAA) (Louie et al., 2005; Wang et al., 2018). Simultaneously, the sampling period was divided into polluted days and non-polluted days according to the local AQI, and cluster analysis was then performed using the HYSPLIT model.

Long-range transported PM$_{2.5}$ was also identified to explain the effects of transported PM$_{2.5}$ over time. In this paper, potential source contribution function (PSCF) was applied to determine possible source areas. PSCF can be interpreted as a conditional probability that describes the spatial distribution of probable geographical source locations that are incidental to their trajectories at the sampling site. The Reanalysis Data Set was employed in these analyses. The “Reanalysis” is to use HYSPLIT model to run the air mass trajectory data set during sampling. A trajectory end point lies in a single grid cell of latitude–longitude coordinates, i–j. The PSCF value is then defined as (Nicolas et al., 2011; Liu et al., 2016):

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}}$$  \hspace{1cm} (2)

In this study, the selected research area was divided into 0.25° × 0.25° horizontal grid cells (i, j), and the probability of occurrence of pollution trajectory in each grid cell was then calculated. The mesh size (0.25° × 0.25°) is set according to the actual needs, which can better identify the potential source transport area. To reduce the error caused by the smaller $n_{ij}$ value in the PSCF calculation, a weight coefficient ($W_{ij}$) is introduced (Nicolas et al., 2011; Liu et al., 2016), as follows:

$$W_{ij} = \begin{cases} 1.0 & (n_{ij} > 80) \\ 0.7 & (20 < n_{ij} \leq 80) \\ 0.42 & (10 < n_{ij} \leq 20) \\ 0.05 & (n_{ij} \leq 10) \end{cases}$$  \hspace{1cm} (3)

Weighted calculation for PSCF$_{ij}$:

$$WPSCF_{ij} = PSCF_{ij} \cdot W_{ij}$$  \hspace{1cm} (4)

### 3 RESULTS AND DISCUSSION

#### 3.1 Chemical Composition of PM$_{2.5}$

#### 3.1.1 Ambient PM$_{2.5}$ Concentration

Fig. 2(a) illustrates the changes in ambient PM$_{2.5}$ concentration during the sampling period at the select ceramic industrial site. It was found that the average concentration of PM$_{2.5}$ was 134 ± 74.7 µg m$^{-3}$, which far exceeded China’s ambient air quality secondary standard for the 24-hr mean concentration level (75 µg m$^{-3}$) (GB3095-2012) (Guo and Shen, 2019; Wang et al., 2021b). Also, the average PM$_{2.5}$ concentration (134 µg m$^{-3}$) was 8.93 times greater than the 24-hr mean PM$_{2.5}$ recommended by the WHO (15.0 µg m$^{-3}$) (Guo and Shen, 2019; Wang et al., 2021b). These results suggested that the local aerosol pollution was quite serious during the sampling period. For comparison, the ambient 24-hr average air quality standard of 75 µg m$^{-3}$ for PM$_{2.5}$ is also shown in Fig. 2(a). It can be seen that PM$_{2.5}$ concentrations were more than 75 µg m$^{-3}$ during the first 8 days of the sampling period and all PM$_{2.5}$ concentrations were much higher than 15 µg m$^{-3}$. The highest value was attained on December 24th, 2020 with 196 µg m$^{-3}$. It was found that the concentration of PM$_{2.5}$ was 8.93 times greater than the 24-hr standard for PM$_{2.5}$, which also demonstrated serious particle-related pollution at our sampling site. The PM$_{2.5}$ concentration level exhibited a downward trend from December 29th to January 1st, where the PM$_{2.5}$ concentration decreased to 59.1 µg m$^{-3}$, which may have been the result of the removal of fine particulates by rain (Fig. 2(b)). After the weather cleared, the concentration of the ambient PM$_{2.5}$ increased to about 125 µg m$^{-3}$ and remained relatively stable at that level. As a northeast wind prevailed, it rained which was one of the reasons for the decline in pollution of PM$_{2.5}$ during this period. This result
Fig. 2. (a) Variation of PM$_{2.5}$ mass concentration during the sampling period; (b) Variation of meteorological factors at state-controlled sites.

illustrates that changes in the concentration of ambient PM$_{2.5}$ were somewhat related to local weather conditions and the local aerosol pollution was quite serious during the sampling period.

The center of the Chinese architectural ceramic industry is located 60 km southwest from the city of Nanchang (Fig. 1, Nanchang city is the capital of Jiangxi Province), located in a subtropical monsoon climate. For the purpose of the air-quality assessment, the pollution levels in different cities or industrial areas needed to be compared. A summary of the ambient PM$_{2.5}$ concentration statistics for different cities during the sampling period and industrial areas reported in the literature is provided in Table 2. In this study, the mean mass concentration of PM$_{2.5}$ was $134 \pm 74.7$ µg m$^{-3}$. Compared to the PM$_{2.5}$ mass concentrations in two cities near the sampling site during the sampling period (data from China’s Air Quality Online Monitoring and Analysis Platform), the mean PM$_{2.5}$ concentration in Nanchang city ($96.7 \pm 63.3$ µg m$^{-3}$) and Yichun city ($71.5 \pm 94.5$ µg m$^{-3}$) were lower than the $134$ µg m$^{-3}$ reported for the Chinese architectural ceramic industrial base. However, these PM$_{2.5}$ concentrations were much lower than the mean PM$_{2.5}$ concentrations reported in Ningdong National Energy and Chemical Industrial Base ($312 \pm 127$ µg m$^{-3}$) (Liang et al., 2019) and Petrochemical industry city of northern China ($165 \pm 79.1$ µg m$^{-3}$) (Luo et al., 2018). However they were higher than the mean PM$_{2.5}$ concentrations reported in the industrial city of Dongguan in southern China ($53$ µg m$^{-3}$) (Zou et al., 2017) and the coastal industrial area of India ($31.8 \pm 14.9$ µg m$^{-3}$) (Police et al., 2018). The PM$_{2.5}$ concentrations reported for the Chinese architectural
ceramic industrial center were relatively high during the sampling period. The high PM$_{2.5}$ concentration could be attributed to the large amounts of fossil fuels used by ceramic enterprises that resulted in high emissions of boiler, which increased pollutant emissions.

In this reported study, the concentrations of the chemical components in the test samples were found to be substantially different. The average percentage of each chemical component in PM$_{2.5}$ was divided into four intervals, < 0.1%, 0.1–1%, 1–10%, and >10%, as depicted in Fig. 3. The chemical species that accounted for >10% of the components of PM$_{2.5}$ were NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$. The chemical species that accounted for 1–10% of the components of PM$_{2.5}$ were OC, EC, Cl$^-$, F$^-$, Na$^+$, Na, Mg, Si, etc.

### 3.1.2 Water-soluble ionic species

Fig. S1 shows the proportion of water-soluble ions in PM$_{2.5}$ and proportion of specific water-soluble ion components in total water-soluble ions. Among the water-soluble ions, NO$_3^-$, SO$_4^{2-}$ and NH$_4^+$ ions were the main water-soluble ions in PM$_{2.5}$, which comprised 33.8%, 30.5% and 24.6% of the mean concentration of total water-soluble ions (NWI for short). The mean concentration of the total water-soluble ions was 65.3 ± 45.1 µg m$^{-3}$ accounting for 48.7% of the total PM$_{2.5}$. It was found that SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$ were the most abundant ions in the PM$_{2.5}$ accounting for about 88.9% of the total water-soluble ions. The water-soluble ions SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$ were from their gaseous precursors resulting from photochemical reactions, and are known to be significant secondary pollution particulates mainly emanating from the Chinese architectural ceramic industry during the sampling period. The content of other water-soluble ions in PM$_{2.5}$ was very low, illustrating that the contribution of other water-soluble ions to PM$_{2.5}$ was relatively small. In addition, the ratio of SO$_4^{2-}$/NO$_3^-$ was used to determine whether the contribution of ambient PM$_{2.5}$ was dominated by stationary sources or mobile sources (Huang et al., 2012). The

### Table 2. Average concentration of ambient PM$_{2.5}$ during sampling period in some cities and typical industrial base.

<table>
<thead>
<tr>
<th>Region</th>
<th>Mean concentration of PM$_{2.5}$ (µg m$^{-3}$)</th>
<th>SO$_4^{2-}$/NO$_3^-$</th>
<th>OC/EC</th>
<th>Data sources</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gao’an ceramic industrial base of China</td>
<td>134 ± 74.7</td>
<td>1.16</td>
<td>1.94</td>
<td>This work</td>
</tr>
<tr>
<td>Urban area of Yichun City, China</td>
<td>71.5 ± 94.5</td>
<td>-</td>
<td>-</td>
<td>China’s air quality online monitoring and analysis platform</td>
</tr>
<tr>
<td>Urban area of Nanchang City, China</td>
<td>96.7 ± 63.3</td>
<td>-</td>
<td>-</td>
<td>China’s air quality online monitoring and analysis platform</td>
</tr>
<tr>
<td>Ningdong National Energy and Chemical Industrial Base, China</td>
<td>312 ± 127</td>
<td>1.12</td>
<td>1.54</td>
<td>(Liang et al., 2019)</td>
</tr>
<tr>
<td>Industrial city in southern China</td>
<td>53.0</td>
<td>2.95</td>
<td>2.71</td>
<td>(Zou et al., 2017)</td>
</tr>
<tr>
<td>Petrochemical industry city of northern China</td>
<td>165 ± 79.1</td>
<td>2.21</td>
<td>8.31</td>
<td>(Luo et al., 2018)</td>
</tr>
<tr>
<td>A coastal industrial area of India</td>
<td>31.8 ± 14.9</td>
<td>-</td>
<td>-</td>
<td>(Police et al., 2018)</td>
</tr>
</tbody>
</table>
**Table 3.** Correlation coefficient matrix for water-soluble ions in ambient PM$_{2.5}$ during sampling period.

<table>
<thead>
<tr>
<th>Ions</th>
<th>NO$_3^-$</th>
<th>SO$_4^{2-}$</th>
<th>NH$_4^+$</th>
<th>Cl$^-$</th>
<th>F$^-$</th>
<th>Na$^+$</th>
<th>Ca$^{2+}$</th>
<th>K$^+$</th>
<th>Mg$^{2+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO$_3^-$</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SO$_4^{2-}$</td>
<td>0.611*</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>0.906**</td>
<td>0.843**</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cl$^-$</td>
<td>-0.329</td>
<td>0.043</td>
<td>-0.161</td>
<td>-0.092</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na$^+$</td>
<td>-0.139</td>
<td>-0.076</td>
<td>-0.075</td>
<td>-0.036</td>
<td>0.772**</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ca$^{2+}$</td>
<td>0.628**</td>
<td>0.712**</td>
<td>0.821**</td>
<td>0.633*</td>
<td>0.085</td>
<td>-0.323</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>K$^+$</td>
<td>0.282</td>
<td>0.674**</td>
<td>0.529*</td>
<td>0.547*</td>
<td>0.439</td>
<td>0.523*</td>
<td>0.832**</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Mg$^{2+}$</td>
<td>-0.200</td>
<td>0.151</td>
<td>-0.026</td>
<td>-0.014</td>
<td>0.849**</td>
<td>0.923**</td>
<td>0.352</td>
<td>0.657**</td>
<td>1</td>
</tr>
</tbody>
</table>

Note: *Correlation is significant at the 0.05 level. ** Correlation is significant at the 0.01 level.

The ratio of SO$_4^{2-}$/NO$_3^-$ in this ceramic industrial base was 1.16, suggesting that the contribution of stationary source emissions to PM$_{2.5}$ was higher than mobile source emissions.

Furthermore, as listed in **Table 2**, with the exception of the Ningdong National Energy and Chemical Industrial base (1.12) (Li et al., 2019), the ratio of SO$_4^{2-}$/NO$_3^-$ in PM$_{2.5}$ in the reported ceramic industrial base was much lower than the ratio of SO$_4^{2-}$/NO$_3^-$ reported in petrochemical industrial city of northern China (Zibo city, 2.21) (Luo et al., 2018) and industrial city (Dongguan) in southern China (2.95) (Zou et al., 2017).

The correlation coefficient matrix for water-soluble ions in PM$_{2.5}$ is shown in **Table 3**. As these results show, the concentrations of NO$_3^-$ and NH$_4^+$ were significantly correlated ($R = 0.906$, $P < 0.01$). Similarly, a strong correlation ($R = 0.843$, $P < 0.01$) was observed for the concentrations of SO$_4^{2-}$ and NH$_4^+$. These good correlations suggested that the species were associated with ambient air and primarily existed as ammonium sulfate ([NH$_4$]$_2$SO$_4$), ammonium bisulfate (NH$_4$HSO$_4$), and ammonium nitrate (NH$_4$NO$_3$) (Zhang et al., 2011; Huang et al., 2012). The NO$_3^-$ and SO$_4^{2-}$ concentrations were highly correlated with a coefficient of 0.611 ($P < 0.05$), suggesting that the precursors of the two species were released from a similar emission source or sources. It was found that the ratio of NH$_4^+$ to NO$_3^-$ was 2.11, and the ratio of NH$_4^+$ to SO$_4^{2-}$ was 4.66. This suggested that NH$_4^+$ was mainly combined with NO$_3^-$ and SO$_4^{2-}$ in the form of (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$. (Zhang et al., 2011) also found that the NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$ in PM$_{2.5}$ are also present in the form of (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$. In addition, Mg$^{2+}$ and Na$^+$ also had a high correlation ($R = 0.923$, $P < 0.01$), which indicated that they were from similar sources, possibly soil or dust.

### 3.1.3 Carbonaceous aerosols

Generally speaking, carbonaceous aerosols can be divided into elemental carbon (EC) and organic carbon (OC). EC is a primary pollutant emitted directly from incomplete combustion (anthropogenic and natural) of carbon-containing materials (Huang et al., 2012). OC is the carbon content of a mixture of nonvolatile and semi-volatile organic compounds in various oxidation states. Both of them are important parameters used to identify the source of air pollution (Zhang et al., 2011; Huang et al., 2012).

**Table S1** lists the mass concentration levels of TC, OC and EC in PM$_{2.5}$ of the selected ceramic base during the sampling period. The mean concentration of TC in PM$_{2.5}$ was 18.6 ± 19.5 µg m$^{-3}$, which accounted for about 13.9% of PM$_{2.5}$. The contents of TC in PM$_{2.5}$ were significantly lower than the concentration of water-soluble ions. The mean concentration of OC was 12.4 ± 19.5 µg m$^{-3}$, and the mean concentration of EC was 6.18 ± 4.21 µg m$^{-3}$. The mean OC/PM$_{2.5}$ and EC/PM$_{2.5}$ ratios of the ceramic base were 9.28% and 4.62%.

In addition, the OC/EC ratio can be used to assess the source of air pollution. To this end, it can be seen from **Table S1** that the mean OC/EC ratio was 1.94, indicating that the most important sources were vehicle related emissions and coal-burning source emissions. Moreover, a strong correlation was found between OC and EC ($R = 0.97$, Fig. S2) indicating that OC and EC may have similar pollution sources. Generally speaking, the OC/EC ratio of gasoline vehicle exhaust in motor vehicle exhaust is 2.2, and the OC/EC ratio of diesel vehicle exhaust is 0.8 (Geng et al., 2013). Geng et al. (2013) studied the chemical composition characteristics of PM$_{2.5}$ in an industrial park.
in Zhengzhou. These authors found that the mean OC/EC ratio (5.6) in PM$_{2.5}$ of the park was much higher than the OC/EC ratio (1.94) of this study’s ambient PM$_{2.5}$ at the ceramic industry center (Geng et al., 2013). Furthermore, as listed in Table 2, with the exception of the Ningdong National Energy and Chemical Industrial center (1.54) (Liang et al., 2019), the ratio of OC/EC in ceramic industry center’s PM$_{2.5}$ was much lower than the ratio of OC/EC reported in petrochemical industry city of northern China (8.31) (Luo et al., 2018) and the industrial city (Dongguan) in southern China (2.71) (Zou et al., 2017).

3.1.4 Inorganic elements

The inorganic elements contained in PM$_{2.5}$ can be roughly divided into two types, crustal elements and tracer elements of human activities. The crustal elements include Si, Na, Al, K, Mg, Ca, Fe, Ti, Mn, and other chemical elements, while V, Cr, Cd, Ni, Cu, Pd, Zn, As, Sn, Se, etc. are human generated trace elements, some of which are trace heavy metals. High contents of such inorganic elements may pose a serious threat to human health.

The 39 identified elements comprise about 17.5% of the PM$_{2.5}$ mass (Fig. S1(a)), and all the elements have an average concentration of 23.4 $\mu$g m$^{-3}$. The total contribution of these elements to PM$_{2.5}$ varied with the season. The concentrations of the most abundant species followed the order Si > Al > Ti > Ca > Fe > K > Na > Mg > Zn > Pb > Ba > Mn > Sb > Zr > Cu > Sr > Cr > Rb > As > Li > K > Bi > Sn > Cd > V > Mo > Ti > Ce > Cs > W > La > Y > Co > Th > Be > U > Sc > Sm. In this reported study, Si was the most abundant element with an average concentration of 9.60 $\mu$g m$^{-3}$ and a standard deviation of 3.12 $\mu$g m$^{-3}$. The second most abundant element was Al which with a concentration of 4.39 $\pm$ 1.30 $\mu$g m$^{-3}$. As shown in Fig. S3, the inorganic elements in the PM$_{2.5}$ were dominated by Si, Al, Ti, Ca, K, and Fe, accounting for about 90.2% of all inorganic elements. Other trace elements were mostly related from human activities, accounting for only 9.79%.

The enrichment factor has been widely used to evaluate the sources of elemental components in PM$_{2.5}$ (Guo et al., 2018). It can be calculated as follows:

$$EF_i = \frac{(C_i/C_r)_{aerosol}}{(B_i/B_r)_{soil}}$$

where $C_i/C_r$ is the concentration ratio of i to r in the aerosol, and $B_i/B_r$ is the concentration ratio of i to r in the soil. The term i represents a subject element in the aerosol, and r is the reference element.

In this study, Al was chosen as the reference element. The average composition of elements in the soil was obtained from references (Zhao et al., 2017a, b). The elemental components in the PM$_{2.5}$ originate from the soil when $EF_i$ is close to 1. When $EF_i$ is between 1 and 10, natural sources are the main source of elements although human activities do provide some contribution to the mass of the PM$_{2.5}$. When $EF_i$ is greater than 10, anthropogenic sources dominate (Guo et al., 2018).

Fig. 4 shows EFs of the elements in the ceramic center’s PM$_{2.5}$ during the sampling period. As these data show, the EFs of Ti and Ca were primarily greater than 10, indicating they were related to human activities. The EFs of Na, Mg and K were between 1 and 10, indicating that these elements originated from crustal sources and human activities provided some contribution to PM$_{2.5}$ mass. The EFs of Zn, Pb, Sb, Zr, V and other elements did not exceed 1, indicating that these elements in the PM$_{2.5}$ originated from surrounding industries or human activities and their contribution were not great.

3.2 Source Apportionment

3.2.1 Principal component analysis

The principal component analysis was performed using statistical SPSS software and the results were listed in Table S2. Due to the relatively low content of trace elements such as P, Ba and Mn, 26 conventional components were selected for principal component analysis and other components were not included in the analysis.

In the case of the conventional components (carbon, water-soluble ions and inorganic elements),
within the 95% confidence interval, 6 factors were extracted from ambient ceramic industrial center’s PM$_{2.5}$. This provided an explanation for 90.83% of the information included in analysis concentration matrix of its PM$_{2.5}$. The commonality of all components included in PCA analysis was close to or higher than 0.60, indicating that these factors detailed these components. These components included OC, EC, Ca$^{2+}$, K$^+$, NH$_4^+$, NO$_3^-$, SO$_4^{2-}$, Cl$^-$, Ti, Cu, As, Pb, Mn, and Ni, which had a greater load value for factor 1. This factor accounted for about 33.90% of the total variance. The OC and EC are the primary markers for motor vehicle sources, while NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$ were primarily markers of secondary particles, and Cl$^-$, Cu, As, Mn and Ni were markers for industrial sources (Pant and Harrison, 2013). Based our investigation on particulate matter emission from the ceramic industries, it was found that Ca$^{2+}$, K$^+$, SO$_4^{2-}$ and Ti appeared to be markers for the ceramics industry source. Therefore, this factor (Factor 1) represented a mixed source, which was dominated by motor vehicles, secondary formation and ceramic industry sources. The components with high load values in factor 3 included Na and Cr, K and Cl$^-$, which accounted for about 12.0% of the total variance. The component with a relatively high load value for factor 4 was Ni, which represented about 8.81% of the total variance. The components with a relatively high load values for factor 5 included Fe and NO$_3^-$, accounting for about 7.65% of the total variance. Cr, Cl$^-$, Fe, and Ni are the markers of industrial sources. Therefore, all these results suggested that ceramic industrial sources were responsible for three factors (Factor 3, 4 and 5). The element Zn was far more prevalent in the pollutant with a relative load value for factor 6 that accounted for about 4.38% of the total variance. Zn is a marker for non-exhaust emission sources from motor vehicles (Pant and Harrison, 2013).

The components with high factor 2 load values included Na$^+$, K$^+$, Mg$^{2+}$, F$^-$, Al, Mg, Ca, Si and K, accounting for 24.1% of the total variance. These components were mainly crustal elements, indicating that this factor pointed to fugitive dust as the source of these components.

In sum, the motor vehicle source, secondary source and industrial sources were the main sources of ambient PM$_{2.5}$ in this ceramic industrial center during the sampling period.

### 3.2.2 Source apportionment of PM$_{2.5}$ based on CMB model

The above analysis (the Section of 3.1) of the chemical components in PM$_{2.5}$ can only provide a brief understanding of the source of the PM$_{2.5}$. Therefore, it was necessary to quantitatively calculate the contributions of various sources to the PM$_{2.5}$ using the CMB model to ascertain source apportionment. In this reported work, the CMB model was used to quantitatively analyze the sources of the ambient PM$_{2.5}$ in the ceramic industrial center. The source apportionment results are shown in Fig. 5. The residual sum of squares in the diagnostic indicators of the model operation was 1.08, the correlation coefficient $R^2$ was above 0.8 (0.880), which were indicators of the accuracy of the model simulation results.
Fig. 5. Source apportionment by CMB: CIS, Ceramic Industry Source; FDS, fugitive dust; SS, Secondary sulfate; SN, Secondary nitrate; EEGV, exhaust emissions from gasoline vehicles; EEDV, exhaust emissions from diesel vehicles, NEMV, non-exhaust emissions from motor vehicles

The source apportionment results in Fig. 5 showed that the ceramic industry source (the emissions from the ceramic plants) was the main contributor to the emissions of ambient PM$_{2.5}$. Compared to the mobile sources, the contribution of ceramic industry sources was relatively high, which was consistent with the above analysis results of the SO$_4^{2-}$/NO$_3^-$ ratio (1.16). The second major contributor was secondary aerosols (secondary sulfate and secondary nitrate). And their contributions to PM$_{2.5}$ were about 27.9 $\mu$g m$^{-3}$ and 20.8%. The value of the secondary nitrate was significantly greater than the secondary sulfate, suggesting that the formation of secondary particles may be due primarily to motor vehicle emissions. This result was consistent with the traffic data that reflected a lot of trucks near the sampling site during the sampling period. Due to the rapid aging of vehicle exhaust emissions, primary and secondary particulate matter from motor vehicle exhaust emissions were major sources of PM$_{2.5}$. In this study, gasoline and diesel vehicle exhaust emissions were found to represent about 15.3% of the PM$_{2.5}$ earmarking it major contributors. The contribution from gasoline vehicle exhaust emissions and diesel vehicle exhaust emissions to PM$_{2.5}$ was 3.03% and 12.3%, respectively. Compared to gasoline vehicle exhaust emissions, diesel vehicle exhaust emissions were higher, 4.05 times greater than gasoline vehicle exhaust emissions. This was consistent with the fact that there were many heavy-duty trucks during the sampling period and the location of the sampling site was near a large freight center. Furthermore, the contribution of fugitive dust sources to ambient PM$_{2.5}$ was relatively large, and its contribution to PM$_{2.5}$ was 19.6%. In addition, the motor vehicle non-exhaust emission also contributed to ambient PM$_{2.5}$, and the contribution value to PM$_{2.5}$ was 0.954%.

3.2.3 Backward trajectory method

The backward trajectory cluster of source contributions at different pollution levels is shown in Fig. 6. The analysis results of the backward trajectory during the sampling period are shown in Fig. 6(a). The trajectory of Cluster 1 originated from the northerly direction, transporting particles from the junction of Jiangxi Province, Anhui Province, and Hubei Province to the sampling point. This trajectory was mainly in the Jiangxi Province which represented a very short transport distance, but it had the highest proportion accounting for 78% of the total number of trajectories. This indicated that PM$_{2.5}$ in the ceramic center during the sampling period was susceptible to airflow from the north. Cluster 2 originated from the north-by-east direction, from southern Shandong Province through Jiangsu Province and Anhui Province to the sampling site. This trajectory had a medium transport distance, accounting for 18% of the total trajectories. Cluster 3 came from the northern part of Hebei Province and turns to Liaoning Province. It passed through Bohai Sea, Shandong Province, Northwestern Jiangsu Province and Anhui Province. It had a long transport distance and represented the lowest proportion, accounting for 4%.

The results of the cluster analysis of the backward trajectory of the polluted weather flows are shown in Fig. 6(b). Cluster 1 was from the northeast and moved from the north of Anhui Province...
Fig. 6. HYSPLIT clustering results of the total period (a), pollution days (b) and non-pollution days (c) during the sampling period.

Cluster means - Standard  
120 backward trajectories  
GDAS Meteorological Data

Cluster means - Standard  
17 backward trajectories  
GDAS Meteorological Data

Cluster means - Standard  
104 backward trajectories  
GDAS Meteorological Data

Fig. 6(c) shows the movement trajectory of the clean weather clusters. Cluster 1 was from the northeast in the region of Jiangxi Province. The airflow trajectory was the shortest, accounting for 33% of the total airflow trajectories. Cluster 2 and Cluster 3 both originated from the northern direction and Cluster 2 was from the junction between Hubei Province and Anhui Province to Jiangxi Province, with a short trajectory length, and the largest of all clusters, accounting for 42% of the total trajectories. This indicated that, on non-pollution days, the ceramic center air quality was greatly impacted by the airflow from the junction between Hubei and Anhui. Cluster 3 came from the west of Shandong Province, Henan Province and the east of Hubei Province to Jiangxi Province. The trajectory length was medium, and the number of trajectories accounted for 24%. Cluster 2 originated from the north-by-east direction, mainly from Jiangxi Province, accounting for 24% of the total number of trajectories, and the transport distance was relatively short. The trajectory of Cluster 3 was from the northeast to the southwest, and then to the southeast. This is in the region of Jiangxi Province. The air transport distance was very short, with the highest proportion, accounting for 47%. Cluster 4 was from the west-by-south direction, starting from central Guangxi Province and passing through Hunan Province. It represented long-distance transport. The number of airflow trajectories accounted for 6% of the total number of trajectories. This suggested that the PM$_{2.5}$ concentration of pollution days in sampling site was affected primarily by the airflow in local Jiangxi Province.
Province. The trajectory length was medium, accounting for only 9% of the total trajectories. Cluster 4 started from the northern part of Hebei Province, passed through the western part of Liaoning Province, Bohai Sea, central part of Shandong Province, northwestern corner of Jiangsu Province, and Anhui Province to the Gao'an sampling site. This airflow trajectory was the longest, accounting for only 5% of the total. Cluster 5 originated from the northeast direction, starting from the western part of the Yellow Sea through the central area of Jiangsu Province, Anhui Province to Jiangxi Province. Its transport distance was medium, accounting for 12% of the total number of trajectories.

3.2.4 Potential Source Contribution Function model

The weight potential source contribution function (WPSCF) was used to obtain potential sources and transmission paths of PM$_{2.5}$ during the sampling period. The distribution characteristics of the WPSCF values in the overall period were conducted using the PSCF model (Fig. 7) at the height of 200 m. During the entire sampling period, the potential sources of PM$_{2.5}$ were found to be distributed primarily in the northerly direction. This conclusion was consistent with the results of HYSPLIT cluster analysis. The high value areas of WPSCF (WPSCF > 0.5) were distributed primarily in Chinese ceramic industrial center, east of Jiujiang City and Nanchang City during the overall period, indicating that these locales were the main sources of PM$_{2.5}$ pollution. This was followed by the eastern part of Hubei Province and the southwestern Anhui Province. The WSPCF values

Fig. 7. Analysis results of PSCF in the (a) overall period, (b) on pollution days and (c) non-pollution days during sampling.
of these locales were greater than 0.3, indicating that they also contributed to the concentration of PM$_{2.5}$ in selected ceramic industrial center. The high-value areas of WPSCF on pollution days were concentrated in the industrial center. The WPSCF values in the southwest of Nanchang City, the southeast of Jiujiang City, and the northeast of Yichun City were also high. This suggested that the PM$_{2.5}$ concentration on pollution days in the center was mainly affected by local potential sources and short-distance regional transport. The distribution characteristics of WPSCF on non-pollution days were similar to those of the overall period. The high-value areas of WPSCF were mainly found in the ceramic center, the northeastern region of Yichun City, Nanchang City and the eastern region of Jiujiang. There were also high WPSCF values scattered in the southeast corner of Hubei Province and the southwest corner of Anhui Province.

In summary, the ambient PM$_{2.5}$ of the ceramic industrial base was affected by regional transmission, but it primarily originated from local sources and surrounding areas.

### 4 CONCLUSIONS

1. The average PM$_{2.5}$ concentration at sampling site was 134 µg m$^{-3}$, which far exceeded the air quality guidelines of the World Health Organization (15 µg m$^{-3}$). The dominant components of PM$_{2.5}$ in this sampling site were secondary ions (sulfate and nitrate) and carbon fractions.
2. Water-soluble ions and total carbon contributed about 48.7% and 13.9% to the PM$_{2.5}$ mass, respectively. And the SO$_4^{2-}$/NO$_3^-$ ratio in the ambient PM$_{2.5}$ during the sampling period was 1.16, indicating that it was primarily the result of emissions from stationary sources.
3. The results of EFs indicated that these elements in the PM$_{2.5}$ originated from surrounding industries or human activities and their contributions were not great.
4. The results of PCA showed that the motor vehicle source, secondary source and industrial sources were the main sources of ambient PM$_{2.5}$ in this ceramic industrial center during the sampling period.
5. CMB model was applied to determine the source apportionment of the ambient PM$_{2.5}$. The results showed that the ambient PM$_{2.5}$ at the ceramic industrial center was mainly from ceramic industry sources (27.9%), secondary formation (20.9%), dust sources (19.6%), and gasoline/diesel vehicle exhaust emissions (15.3%) and motor vehicle non-exhaust emissions (0.95%).
6. Based on the backward trajectory analysis and potential source apportionment, it was found that the ambient PM$_{2.5}$ of the ceramic industrial base was affected by regional transmission, but it primarily originated from local sources and surrounding areas.

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### SUPPLEMENTARY MATERIAL

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### REFERENCES


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