High-time-resolution chemical composition and source apportionment of $PM_{2.5}$ in northern Chinese cities: implications for policy

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Abstract: Fine particulate matter ($PM_{2.5}$) pollution is still one of China's most important environmental issues, especially in northern cities during wintertime. In this study, intensive real-time measurement campaigns were conducted in Xi'an, Shijiazhuang, and Beijing to investigate the chemical characteristics and source contributions of $PM_{2.5}$ and explored the formation of heavy pollution for policy implications. The chemical compositions of $PM_{2.5}$ in three cities were all dominated by organic aerosol (OA) and nitrate (NO_3^-). Results of source apportionment analyzed by hybrid environmental receptor model (HERM) showed that the secondary formation source contributed higher to $PM_{2.5}$ compared to other primary sources. Biomass burning was the dominant primary source in three pilot cities. The contribution of coal combustion to $PM_{2.5}$ is non-negligible in Xi'an and Shijiazhuang but is no longer an important contributor in the capital city of Beijing due to the execution of a strict coal-banning policy. The potential formation mechanisms of secondary aerosol in three cities were further explored by establishing the correlations between the secondary formation source and aerosol liquid water content (ALWC), and O_x ($O_3 + NO_2$), respectively. The results showed that photochemical oxidation and aqueous-phase reaction were two important pathways of secondary aerosol formation. According to sources variations, air pollution events that occurred in campaigns were classified into three types: biomass combustion dominated, secondary formation source dominated, and a combination of primary and secondary sources. Additionally, this study compared the changes in chemical composition and source contributions of $PM_{2.5}$ in past decades. The results suggested that the clean energy replacements for rural household should be urgently encouraged to reduce the primary source emissions in northern

China, and collaborative control on ozone and particulate matter need to be continuously promoted to weaken the atmosphere oxidation capacity for the sake of reducing secondary aerosol formation.

1. Introduction

Fine particulate matter ($PM_{2.5}$, aerodynamic diameter <= 2.5 μm) is of large concern because of its adverse effects on both natural environment (Kuniyal and Guleria, 2019; Kuo et al., 2013) and human health (Pöschl, 2005; Shen et al., 2021; Zeng and He, 2019). With the soaring economic growth and urbanization in China, $PM_{2.5}$ pollution has been a most serious environmental issue in recent decades (Chan and Yao, 2008; He et al., 2002; Pui et al., 2014; Zhang et al., 2013). The most impressive case is that an extremely severe haze pollution episode occurred in eastern and central China in January 2013 with peak value of $PM_{2.5}$ concentration over 500 μg m^{-3}. This month had been reported as the haziest month in the past 60 years in Beijing, China (Wang et al., 2014; Huang et al., 2014). Thereafter, aiming to improve air quality, the China central government issued the Air Pollution Prevention and Control Action Plan (APCAP) in September 2013 (http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm, in Chinese), and the Three-year Action Plan to Fight Air Pollution (TAPFAP) in June 2018 (http://www.gov.cn/zhengce/content/2018-07/03/content_5303158.htm, in Chinese). With the implementation of strict pollution controls, air quality in northern China has improved significantly over the past decade (Wang et al., 2020a, 2017; Li et al., 2020). Previous studies show that $PM_{2.5}$ concentration decreased notably in past two decades, and the composition of organic aerosol (OA), black carbon (BC) and sulfate (SO_4^{2-}) decreased as well, while the ammonium (NH_4^+) slightly increased and nitrate (NO_3^-) increased obviously. In terms of $PM_{2.5}$ sources, contribution of secondary source increased obviously while contribution of industrial emission and coal combustion decreased due to elimination of industries and enterprises with high pollutant emissions, promotion of desulfurization in industrial facilities, replacement of clean energy, and optimization of industrial and energy structures (Lu et al., 2021; Ma et al., 2022;Tao et al., 2017; Wang et al., 2019). However, there is still a significant gap between the $PM_{2.5}$ concentration in northern China and its latest recommendations on air quality guideline (5 μg m^{-3}) by the World Health Organization (https://apps.who.int/iris/bitstream/handle/10665/345329/9789240034228-eng.pdf, page 78). In addition, severe $PM_{2.5}$ pollutions still frequently occurred in northern China during wintertime (Guo et al., 2021; Li et al., 2017a, 2021b). To figure out the causes behind the pollutions and further improve air quality in northern China, it is essential to use online high-time-resolution source apportionment technology to understand the chemical composition and source contribution of $PM_{2.5}$ in those pollution events.

Recently, more research on measurements of $PM_{2.5}$ and its source apportionments were conducted using online high-time-resolution technologies (Li et al., 2017c; Wang et al., 2021a; Elser et al., 2015). Compared to traditional offline filter-based approach, online methods characterize the short-time variation of $PM_{2.5}$. It allows for distinguishing the rapid changes and evolutions of chemical components, and is particularly profitable to gain knowledge on the formations of heavy air pollution or episode events (Liu et al., 2016; Ouyang et al., 2019; Zheng et al., 2016; Elser et al., 2015). For instance, Lv et al. (2021) employed a Positive matrix factorization (PMF) model with high-time-resolution online $PM_{2.5}$ data to accurately quantify and distinguish the source distributions in Beijing during two haze episodes in January 2019. Liu et al. (2019) recognized the main drivers of haze event occurred in winter Beijing in 2016 according to high-time-resolution source apportionment of $PM_{2.5}$ with multiple models. Furthermore, Wang et al. (2021b) found that vehicle emission contributed most to $PM_{2.5}$ during pollution episodes in downtown Lanzhou based on high-resolution online data source apportionment. Currently, to fully understand and solve heavy pollution events in winter that troubles local governments in northern cities of China (Wang et al., 2022b; Xu et al., 2022; Zhou et al., 2022), more advanced online measurement, and source apportionment is a better choice (Tao et al., 2015). It should be pointed out that previous researches were mainly focused on individual cities, and those results have some limitations in guiding the improvement of air quality in the entire northern region of China. Therefore, it is necessary to conduct comparative research among multiple cities.

Considering the differences in geographical location, population, economy, industrial/energy structure, air quality, and depth of air pollution control measures among different cities, three cities in northern China including Beijing, Shijiazhuang and Xi'an were chosen as pilot research subjects. The cities of Beijing and Shijiazhuang are located in the North China Plain, which is one of the most polluted regions in China (Chan and Yao, 2008). Beijing is the capital of China and its air quality has significantly improved under the implementation of the strictest clean air policy since 2013 (Li et al., 2021a; Pang et al., 2021; Vu et al., 2019; Zhang et al., 2020). However, the city was still plagued by pollution events in wintertime (Wang et al., 2020b; Yang et al., 2022c; Zhou et al., 2022). Shijiazhuang was recognized as one of the most serious air pollution cities worldwide (Liu et al., 2018b; Huang et al., 2019). Its air quality had also improved under the implementation of the Clean Air Plan, whereas its annual $PM_{2.5}$ concentration was still unable to meet the China's National Ambient Air Quality Standards (NAAQS-II) of 35 $\mu g\ m^{-3}$ until 2021 (Fig. S1). Xi'an is located in the Fenwei Plain, which is a region that suffered from heavy pollution and was designated as a key region for TAPFAP in 2018 (Cao and Cui, 2021). Compared with Beijing and Shijiazhuang, high-intensity air pollution controls in Xi'an started late due to a lack of financial support. And the annual $PM_{2.5}$

concentration in Xi'an could not meet the NAAQS-II until 2021 as well (Fig. S1). Meanwhile, it is still unclear about the actual causes of the pollution, either topography, meteorological conditions, or local emissions (Chen et al., 2021; Tian et al., 2022; Wang et al., 2015, 2022b). In this study, we conducted intensive real-time observation of $PM_{2.5}$ chemical components in Xi'an, Shijiazhuang, and Beijing during wintertime. The objectives are 1) to determine the characteristics of $PM_{2.5}$ and its chemical components in the three typical northern China cities during wintertime; 2) to quantify the source contribution and explore the potential formation mechanism of secondary aerosols; 3) to explore the unique causes of heavy pollution events in different cities; and 4) to provide suggestions on establishment of efficient policies for air quality continuous improvement. This study provides scientific guidance for developing policy on air quality improvement for northern China cities.

2. Methods

2.1 Sampling sites and periods

In this study, intensive online measurements of $PM_{2.5}$ were conducted at three pilot cities of Xi'an, Shijiazhuang, and Beijing during wintertime (Fig. 1). The sampling sites in Xi'an and Beijing are located at two Chinese Academy of Sciences (CAS) stations. The one in Xi'an is the Guanzhong Plain Ecological Environment Change and Comprehensive Treatment National Observation and Research Station, Institute of Earth Environment (IEE) (34.24°N, 108.87°E), and another one in Beijing is Tower Branch of the Institute of Atmospheric Physics (IAP) (39.98°N, 116.39°E). Both two sites are surrounded by commercial and residential buildings without intense industrial emissions nearby. Previous studies indicated that these two sites were influenced by biomass and coal burning for heating and cooking during wintertime as well as usual local traffic emissions (Tian et al., 2021; Xu et al., 2021). The sampling site in Shijiazhuang is situated in the courtyard of Hebei Sailhero Environmental Protection High-tech Co., Ltd. (38.04°N, 114.65°E), which is surrounded by pharmaceutical and machine-building industries and close to the streets. The intensive campaigns were continuously conducted for ~1 month in each city (i.e., 12 December 12th 2020 to January 7th 2021 in Xi'an, December 20th 2021 to January 24th 2022 in Shijiazhuang, and January 17th 2021 to February 20th 2021 in Beijing).

2.2 Online measurements of $PM_{2.5}$ chemical components

2.2.1 Organic aerosol and inorganic ions

Concentrations of OA, NO_3^-, SO_4^{2-}, ammonium (NH_4^+), and chloride (Cl^-) in $PM_{2.5}$ at a 15-minute time resolution

were monitored by a quadrupole aerosol chemical speciation monitor (Q-ACSM, Aerodyne Research Inc., Billerica, Massachusetts, USA) equipped with a $PM_{2.5}$ lens. The detailed operational principles and calibration method of the Q-ACSM are described elsewhere (Ng et al., 2011; Hu et al., 2017). First, the sampled ambient air stream passed through a PM_{10} impactor inlet and a Nafion® dryer (MD-700-24F-3; Perma Pure, Inc., Lakewood, NJ, USA) with a flowrate of 5 L min^{-1} before entering the Q-ACSM chamber. Then, the pre-treatment particles passed through a 100 μm critical orifice at 0.1 L min^{-1} and were focused into a narrow beam by an aerodynamic intermediate pressure lens. The focused particle beam was flash vaporized by a capture vaporizer (CV) at ~600 C°. The vaporized compounds were then ionized by an electron impactor (EI) ionization source at 70 eV and subsequently analyzed by the quadrupole mass spectrometer.

Based on calibration system consists of an atomizer (Model 9302, TSI Inc., Shoreview, MN, USA), a differential mobility analyzer (DMA, TSI model 3080, TSI Inc.), and a condensation particle counter (CPC, TSI model 3772, TSI Inc.), ammonium nitrate (NH_4NO_3) and ammonium sulfate ($(NH_4)_2SO_4$) aerosol were used for calibration. The raw data of Q-ACSM were analyzed by the ACSM local tool (V1.5.3.5, Aerodyne Research Inc., Billerica, Massachusetts, USA) compiled with Igor Pro 6.37 (Wavemetrics, Lake Oswego, OR, USA). The response factors (RFs) for NO_3^- in Xi'an, Shijiazhuang, and Beijing were set as 2.03×10^{-11}, and 5.9×10^{-11}, 2.20×10^{-11}, respectively, and the relative ionization efficiencies (RIEs) for NH_4^+ and SO_4^{2-} were set as 8.06 and 0.83 in Xi'an, 5.82 and 0.30 in Shijiazhuang, 6.31 and 0.38 in Beijing, respectively. Other RIEs for NO_3^-, OA, and Cl^- were set as default values of 1.4, 1.1, and 1.3, respectively (Ng et al., 2011). In addition, the collection efficiency (CE) value of Q-ACSM equipped with a $PM_{2.5}$ lens was recommended as 1 based on laboratory simulation experiments by Xu et al. (2017). Finally, the chemical components monitored by Q-ACSM was corrected by the results of offline filter sampling experiments during the same periods (Fig. S2).

2.2.2 Black carbon

BC concentration in $PM_{2.5}$ was obtained by an Aethalometer (Model AE33, Magee Scientific Inc., Berkeley, CA, USA) with a 1-minute time resolution. The AE33 monitors the light attenuation of seven wavelengths (λ = 370, 470, 525, 590, 660, 880, and 940 nm), and the light attenuation at λ = 880 nm was used to calculate BC concentration (Wang et al., 2019; Drinovec et al., 2015). Briefly, the ambient air was first sampled on a filter tape inside the instrument through a $PM_{2.5}$ cyclone (SCC-1.829, BGI Inc., USA) at a flowrate of 5 L min^{-1}. The entering particles were divided into two sample spots on the filter through two channels with different follows. Then the light

attenuation transmitted through two parallel spots was detected. For quality accuracies of monitoring, the sampled particles were desiccated with a Nafion® dryer (MD-700-24F-3; Perma Pure, Inc., Lakewood, NJ, USA) before entering the AE33. Furthermore, a real-time loading effect compensation algorithm based on two spots measurement was used to eliminate the nonlinear loading effects of the Aethalometer. A detailed description of the Model AE33 principle can be found in Drinovec et al. (2015).

2.2.3 Elements

Twenty-four elements, including Si, K, Ca, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Ag, Cd, Sn, Ba, Au, Hg, Th, Pb, and Pd in $PM_{2.5}$, were analyzed by a Xact625 Ambient Metals Monitor (Cooper Environmental Services, Tigard, Oregon, USA) with a 1-hour time resolution. Si, K, Ca, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Ba, and Pb were selected for further analysis in Xi'an and Beijing, while other elements were excluded due to most of their concentration below the method detection limit. In Shijiazhuang, S, Cl, and Ti were analyzed by replacement of Ga, Ag and Au, respectively. Finally, Si, K, Ca, Ti, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Ba, and Pb were selected for further analysis. The description and detection principles of Xact625 were introduced by Furger et al. (2020) and Rai et al. (2020). In brief, the ambient air stream was firstly sampled on a Teflon filter tape inside the instrument through a $PM_{2.5}$ cyclone inlet at a constant flow rate of 16.7 L min^{-1}, and then the sample was automatically analyzed by nondestructive energy-dispersive X-ray fluorescence (XRF) to determine the mass of the species. For quality control and assurance, the Xact625 performed automatic internal quality control by testing the Pd rod every hour to ensure the stability of the instrument. Energy calibration was performed daily from 00:00 to 00:15 and a range calibration from 00:15 to 00:30 local standard time (LST) to monitor any possible shift and instability of the XRF (Liu et al., 2019). During our sampling periods, the concentration of Pd varies within 3 standard deviations (Fig. S3), illustrating the reliable and stable performance of the Xact625.

2.2.4 Complementary data

Online hourly concentrations of $PM_{2.5}$ and gas pollutants (i.e., NO_x, NO_2, CO, SO_2, and O_3) were acquired from the National Air Quality Monitoring Station (https://air.cnemc.cn:18007/). Meteorological parameters, including wind speed (WS), wind direction (WD), relative humidity (RH), and temperature (T) were obtained from National Meteorological Station (http://data.cma.cn/). The detailed information for complementary data was listed in Table S1.

2.3 Data analysis

2.3.1 $PM_{2.5}$ mass reconstruction

Chemical closure was utilized to assess whether chemical compositions can be representative of $PM_{2.5}$. The sum of OA, NO_3^-, SO_4^{2-}, NH_4^+, Cl^-, BC, mineral dust (MD), and trace elements (TE) was considered as the reconstructed $PM_{2.5}$, where MD and TE were calculated as follows (Chow et al., 2015).

$$[MD] = 2.20\times[Al] + 2.49\times[Si] + 1.63\times[Ca] + 2.42\times[Fe] + 1.94\times[Ti] \quad (1)$$

$$[TE] = [K] + [Cr] + [Mn] + [Ni] + [Cu] + [Zn] + [As] + [Se] + [Ba] + [Pb] \quad (2)$$

where [ ] represents the chemical species concentration; [Al] and [Ti] were calculated by the concentration of Ca ([Al] = 4.3×[Ca] and [Ti] = 0.25×[Ca]) (Wei et al., 1991). Good correlations between online and reconstructed $PM_{2.5}$ mass (slope = 0.87–1.10, R^2 = 0.82–0.93) in three pilot cities (Fig. S4) indicated that our measurements could detect major components of $PM_{2.5}$. The $PM_{2.5}$ concentration used in the following discussion referred to the reconstructed $PM_{2.5}$ concentration.

2.3.2 Hybrid environment receptor model

Source apportionment of $PM_{2.5}$ was analyzed with a bilinear model named the hybrid environment receptor model (HERM). HERM is developed by the IEECAS and the University of Nevada, Las Vegas (Chen and Cao, 2018). Like other receptor models, the speciation of pollutants at a receptor site can be separated into emission sources and the chemical compositions of the sources. To solve the mass balance of $PM_{2.5}$, the bilinear HERM in matrix notation is defined as follows

$$C_{mn} = \sum_{i=1}^{I} F_{mi} G_{in} + Q_{mn} \quad (3)$$

where C_{mn} is the measured concentration of chemical species m during time n; F_{mi} is the source profile, that is the fractional quantity of species m in source i emission; G_{in} represents the contribution of source i during time n; and Q_{mn} is the model residual for species m concentration measured during time n. Based on an iterative conjugate gradient algorithm, the HERM solves G_{in} and unknown F_{mi} by minimizing the Q_{mn}, which is defined as follows.

$$Q_{mn} = \sum_{m=1}^{M} \sum_{n=1}^{N} \frac{(C_{mn} - \sum_{i=1}^{I} F_{mi} G_{in})^2}{\sigma_{C_{mn}}^2 + \sum_{i=1}^{I} (\sigma_{F_{mi}}^2 G_{in}^2 + \delta_{mi} \sigma_{C_{mn}}^2)} \quad (4)$$

where M, N, and I are the number of samples, chemical species, and sources, respectively; $\sigma_{F_{mi}}$ represents the error in the variability in the constrained factor profile. δ_{mi} was set to 0 or 1 depending on whether the i^{th} factor profile is constrained or unconstrained, respectively.

The HERM input data included the concentration and uncertainty data of chemical species. 19 chemical species in

198 Xi'an and Shijiazhuang and 20 chemical species in Beijing were selected for source apportionment, respectively.
199 Details of selected chemical species and its uncertainty calculation was described in Text S1 in the Supplement. A
200 range from two to ten factors solutions was investigated by HERM with completely unconstrained factor profiles to
201 search for optimal solutions. The detailed diagnostics can be seen in Text S2 in the Supplement. A six-factor solution
202 for Xi'an and Shijiazhuang and an eight-factor solution for Beijing were found to be the optimal solution based on
203 multiple criteria including 1) variations in Q/Q_{exp} which can be used to choose the optimal number of resolved factors,
204 2) physical meaningfulness of distinct factor profiles and explained variation (EV) values of variables, 3) good
205 correlations between sources contribution and external and internal tracers, and 4) agreement between the measured
206 and modeled $PM_{2.5}$ mass. More detailed information on the final selected factor profiles and contributions is presented
207 in Sect. 3.2.

208 **2.3.3 Aerosol liquid water content**

209 Aerosol liquid water content (ALWC) was calculated by ISORROPIA-II thermodynamic equilibrium model
210 (http://isorropia.eas.gatech.edu/) based on data of $PM_{2.5}$ chemical species (including NO_3^-, SO_4^{2-}, NH_4^+, and Cl^-) and
211 meteorological parameters including relative humidity (RH) and temperature (T), more model information can be
212 found in Fountoukis and Nenes (2007). It should be noted that the ISORROPIA-II model does not consider the
213 contribution of the organic, as inorganic aerosols are the most hygroscopic species and dominant contributor to
214 ALWC (Huang et al., 2020).

215 **3. Results and discussion**

216 **3.1 Characteristics of $PM_{2.5}$ and its chemical components**

217 Figure 1 illustrates the mass composition of $PM_{2.5}$ in three pilot cities during the sampling periods, and their
218 concentrations levels are summarized in Table S4. The average $PM_{2.5}$ concentrations in Xi'an, Beijing, and
219 Shijiazhuang were 77±47 μg m^{-3}, 64±57 μg m^{-3}, and 60±39 μg m^{-3}, respectively. It is noted that the average $PM_{2.5}$
220 concentrations in Xi'an, Beijing, and Shijiazhuang did not meet the second level of the NAAQS, indicating that it is
221 necessary to establish more particular and efficient pollution reduction measures. As shown in Fig. 1, the chemical
222 compositions of $PM_{2.5}$ were similar in Beijing and Shijiazhuang (Fig. 1b and c) which was mainly composed of OA
223 (26.9–34.2%), followed by NO_3^- (23.6–26.5%), SO_4^{2-} (11.8–15.0%), NH_4^+ (11.8–14.8%), MD (7.4–10.1%), BC (2.9–
224 6.5%), and Cl^- (1.1–4.8%). However, in Xi'an, MD contributed in comparison more to $PM_{2.5}$ (17.3%), while SO_4^{2-}

had a smaller contribution (6.8%). This could be explained by more construction activities and MD transport from the Loess Plateau to Xi'an (Long et al., 2016; Yan et al., 2015). Meanwhile, the lowest sulfur oxidation ratio (SOR) was observed in Xi'an (0.18 ± 0.08, see Table S5), indicating weak efficiency of the second generation of SO_4^{2-}. The sum of SO_4^{2-}, NO_3^-, and NH_4^+ accounted for 39.0–53.0% of $PM_{2.5}$ in three pilot cities, highlighting the importance of the secondary inorganic components in northern China. In addition, the fractions of BC, Cl^-, and TE in $PM_{2.5}$ were lower in Beijing than those in the other two cities, which can be explained by the stricter local control policies on solid fuels combustion and tightening the industrial emission standards in and near the capital city of China (Li et al., 2021a; Pang et al., 2021).

To better understanding the impact of the chemical components, the mass fraction of each component was plotted as a function of the $PM_{2.5}$ mass concentration (Fig. 2a–c). The two dominant components of $PM_{2.5}$ were OA (25.7–38.0%) and MD (19.9–37.1%) while the $PM_{2.5}$ concentrations were below 40 μg m^{-3}. The faction of OA in $PM_{2.5}$ was the highest in Shijiazhuang and Beijing, while MD contributed most to $PM_{2.5}$ in Xi'an. This is potentially related to more emissions and higher backgrounds of local dust. With increases of the $PM_{2.5}$ mass concentration, the fractions of chemical components in Xi'an and Shijiazhuang changed notably. The factions of OA and NO_3^- increased the most and reached the peaks of 40.1% and 28.7%, respectively, when the $PM_{2.5}$ concentration reached ~196 μg m^{-3} in Xi'an. On the contrary, NO_3^- and SO_4^{2-} were two dominant drivers of increasing $PM_{2.5}$ concentrations in Shijiazhuang, showing peak contributions of 32.5% and 18.7%, respectively, when the $PM_{2.5}$ concentration was over 100 μg m^{-3}. Compared to Xi'an and Shijiazhuang, Beijing had relatively stable fractions of each chemical component with increasing $PM_{2.5}$ concentrations. Particularly, the fractions of OA and NO_3^- contributed dominantly with averages of 33.3±3.0% and 25.3±2.5%, respectively, when the $PM_{2.5}$ > 40 μg m^{-3}.

3.2 Source apportionment of $PM_{2.5}$

Six potential sources, including biomass burning, fugitive dust, industrial emission, coal combustion, vehicle emission, and secondary formation source, were resolved by the HERM analysis. In Beijing, secondary formation source was furtherly divided into secondary nitrate plus OA and secondary sulfate plus OA. A special pollution source of firework was separated due to the Chinese Spring Festival (from New Year's Eve to January 3rd in the lunar calendar). Figures S6 – S8 present the sources profiles and contributions in Xi'an, Shijiazhuang, and Beijing, respectively. Biomass burning features high Explained Variation (EV) for the two tracers Cl^- (33–58%) and K (30–44%) in the three cities (Ni et al., 2017; Zhao et al., 2021). The fugitive dust is characterized by high EV values for

Si (60–90%) and Ca (34–54%), which are the dominant chemical species in the fugitive dust profiles in northern China (Shen et al., 2016; Zhao et al., 2006). The fractions of industrial emission vary among the cities, showing high EV for Ni (55% and 87%) and Cr (25% and 70%) in Xi'an and Shijiazhuang, and high EV for Cr (26%), Mn (40%), and Pb (27%) in Beijing. Ni is possibly emitted from the semiconductor industry (Simka et al., 2005). Cr, Mn, and Pb could originate from the steel manufacturing and incinerator fly ash (Duan and Tan, 2013; Ledoux et al., 2017). Coal combustion is characterized by high EV values for As (38–75%), Se (40–50%), and Pb (31–57%). These elements are enriched in coals, which are reliable indicators of coal combustion (Tian et al., 2013; Xu et al., 2012). The vehicle non-exhaust emissions could be identified by the elements Ba, Cu, Ca, Fe, and Mn. Cu and Ba can be released from brake and tire wear of vehicles (Adachi and Tainosho, 2004; Thorpe and Harrison, 2008). Moreover, Fe and Mn could be emitted from the combustion of lubricating oil and fuel additives (Ålander et al., 2005; Lewis et al., 2003). Relatively high EV values for Ba (68%), Cu (36%), and Ca (35%) are seen in Xi'an, significantly high EV values of Mn (68%), Fe (65%), Cu (53%), and Ba (80%) are characterized in Shijiazhang and relatively high EV values of Fe (34%) and Cu (39%) are featured in Beijing, respectively. Moreover, moderate EV values for BC (18–27%) and OA (13–22%) are commonly regarded as contributions of vehicles engine exhaust, while the temporal variations of VE are well correlated with gaseous NO_x or NO_2 in three cities (R^2 = 0.45–0.78), which is the good tracer of traffic-related emissions (Huang et al., 2017; Li et al., 2017b). The secondary sources resolved by HERM are different among the three cities. In Xi'an and Shijiazhuang, this factor are characterized by high EV values for SO_4^{2-} (62–75%), NO_3^- (55–53%), NH_4^+ (60–56%) and a medium EV value for OA (23–29%), which showed good correlations with SO_4^{2-} (R^2 = 0.85–0.90) and NO_3^- (R^2 = 0.85–0.92) (Dai et al., 2020; Tian et al., 2022). In addition, The OA concentration in this factor was calculated by EV value of OA, which was close to the secondary OA (SOA) concentration estimated by BC-trace method (see Text S3 and Table S6). This means that SOA was mixed in this factor, therefore, this factor was identified as secondary formation source. In Beijing, two secondary sources were resolved. The first one was characterized by high EV value for NO_3^-(58%), NH_4^+ (42%) and medium values for OA (21%), another one was characterized by high EV value for SO_4^{2-}(58%), and medium values for OA (16%), NH_4^+ (30%). The OA concentration in those two factors was also comparable to that estimated by BC-trace method (see Text S3). So, those two sources were identified as secondary nitrate plus OA and secondary sulfate plus OA, respectively. The combination of secondary nitrate plus OA and secondary sulfate plus OA is equivalent to the secondary formation source for next discussion. Additionally, the source of firework emission is characterized by high EV values of Ba (83%), Cu (45%), and K (38%), which are recognized as common indication in fireworks (Rai et al., 2020; Tian et al., 2014).

The modeled $PM_{2.5}$ mass was well correlated with the reconstructed $PM_{2.5}$ mass (R^2 = 0.99, slope = 0.90–1.01, Fig. S10) in three pilot cities, indicating the established models are reasonable. As shown in Fig. 1d and e, the contributions of primary sources (i.e., the sum of biomass burning, fugitive dust, industrial emission, coal combustion, and vehicle emission) in $PM_{2.5}$ were significantly higher than those of the secondary formation source in Xi'an and Shijiazhuang, indicating the $PM_{2.5}$ in these two cities are mainly influenced by the primary source emissions during wintertime. Particularly, biomass burning and coal combustion were two dominant contributors to $PM_{2.5}$ with contributions of 24.6% and 15.1%, respectively, in Xi'an; and 24.4% and 16.0%, respectively, in Shijiazhuang. These suggest that controls of solid fuel combustion are critical to reducing $PM_{2.5}$ pollution in these cities. In contrast, the contribution of secondary formation source to $PM_{2.5}$ in Beijing was highly dominant (> 50%), potentially attributed to strict control of primary emissions under the execution of a series of pollution control policies (Lv et al., 2016; Pang et al., 2021), and more regional transportation of secondary pollutants (Liu et al., 2019; Wang and Zhao, 2018). Among the primary sources, the contributions of biomass burning and vehicle emission were only 18.4% and 11.3%, respectively, further reflecting the benefits of reductions of all primary emissions. Due to the Chinese Spring Festival, the contribution of firework (7.9%) to $PM_{2.5}$ ranked second in primary sources (Fig. S11). Which indicates more refined control schemes need to be encouraged to deal with such special event in the future. It should be noted that contribution of fugitive dust was all lower than fraction of mineral dust in the three pilot cities (Fig. 1). This is because fugitive dust defined here mainly refers road and construction dust emission. While mineral dust represents material assumed oxides of mineral elements such as Al, Si, Ca, Ti and Fe (Chow et al., 2015). These mineral elements in $PM_{2.5}$ comes from more emission sectors including industry, crust, and transportation, construction, combustion (Liu et al., 2018a; Lu et al., 2014; Pant and Harrison, 2013; Shen et al., 2016).

Figures 2d–f shows variations of source contribution with increases in $PM_{2.5}$ mass concentrations in three pilot cities. The most two dominant sources were secondary formation source (32.1%) and fugitive dust (31.4%) in Xi'an, coal combustion (24.9%) and vehicle emission (21.3%) in Shijiazhuang, and secondary formation source (24.3%) and fugitive dust (23.8%) in Beijing, when the $PM_{2.5}$ mass concentration <40 μg m^{-3}. In Xi'an, when the $PM_{2.5}$ mass concentrations exceeded 180 μg m^{-3}, the contribution of biomass burning raised mostly and reached the peak of 38.4%, demonstrating that biomass burning plays an important role in worsening of air quality in Xi'an. On the contrary, the contributions of secondary formation source increased mostly in comparison to other sources in Shijiazhuang and Beijing, indicating the $PM_{2.5}$ pollution was mainly dominated by the secondary aerosol formations during the wintertime. And the peak contributions of secondary nitrate and sulfate were 66.5% and 74,7% while the

312 $PM_{2.5}$ mass concentration increased to 113 μg m^{-3} and 223 μg m^{-3} in Shijiazhuang and Beijing, respectively.

313 **3.3 Formation of secondary aerosols**

314 Using the high-time-resolution data, we further explored the possible formation mechanisms of secondary formation
315 source. The concentration of secondary formation source is standardized by dividing background corrected CO (ΔCO)
316 to weaken impact of planetary boundary layer height (PBLH) (DeCarlo et al., 2010). In this study, ΔCO is defined as
317 the 1.25th percentile of CO concentration during the campaign, which are 0.17, 0.15, and 0.16 ppm in Xi'an,
318 Shijiazhuang, and Beijing, respectively. O_x (NO_2 + O_3) is an indicator of the photochemical oxidation degree (Wood
319 et al., 2010). The function between secondary formation source/ΔCO ratio and O_x during the daytime (i.e., 08:00–
320 17:00 LST) (Fig. S12) was plotted to explain the effect of photochemical oxidations in three pilot cities. As shown
321 in Fig. 3, good linear correlations of secondary formation source/ΔCO and O_x (R^2 = 0.83–0.99) suggest that
322 photochemical oxidations play an important role in the formation of secondary aerosol during the daytime. Compared
323 to the low-level O_x, formation of secondary aerosol significantly enhanced at high-level O_x (>50 ppb) in Xi'an and
324 Beijing, characterized by larger slopes of 17.2 and 38.9, respectively (Fig. 3a and c). Furthermore, the highest
325 atmospheric oxidation capacity was found in Beijing, inferring by the highest fraction of O_3 to O_x. This is consistent
326 with the highest contribution of secondary formation source to $PM_{2.5}$ in Beijing during the daytime (Fig. S13a–c).

327 The aqueous-phase reaction is another important pathway for secondary aerosol formation in the atmosphere (Wang
328 et al., 2018; Xue et al., 2014). ALWC is considered an indicator of an aqueous-phase reaction (Ervens et al., 2011).
329 Considering that the aqueous-phase reaction occurs both during the daytime and nighttime characterized by good
330 correlations between secondary formation source/ΔCO and ALWC (R^2 = 0.81–0.98, Fig. S14). The correlations of
331 secondary formation source/ΔCO ratio and ALWC during all sampling periods were re-established in three pilot cities
332 to assess the implications of aqueous-phase chemistry for secondary aerosol production. As shown in Fig. 4, the
333 secondary formation source/ΔCO showed a significant linear correlation to ALWC (R^2 = 0.92–0.99) when RH < 80%,
334 indicating an obvious effect of aqueous-phase reaction on the secondary aerosol formation during the sampling
335 periods. However, when RH >80%, the secondary formation source/ΔCO showed no notable increase with ALWC
336 in Shijiazhuang (Fig. 4b), whereas a tiny increase with ALWC in Beijing (Fig. 4c). The higher ALWC at RH > 80%
337 probably inhibits secondary aerosol formation due to the decrease in aerosol acidity (Huang et al., 2019; Meng et al.,
338 2014). Khan et al. (2008) found that NO_3 radicals can rapidly generate from the reaction between NO_2 and O_3 with
339 unsaturated organic species during nighttime. The value of $O_3 \times NO_2$ can thus represent its production reaction rate or

be used as a proxy for the NO_3 radical. The highest NO_3 radical production rate was found in Beijing, followed by Xi'an and Shijiazhuang, when RH<80%. This could be used to explain the highest contribution of secondary formation source to $PM_{2.5}$ in Beijing during the daytime and nighttime (Fig. S13). Moreover, the results showed that both photochemical oxidation and aqueous-phase reaction play more important roles in Beijing, where the primary sources have been better controlled. This reflects that pollution control policies need to be focused on the suppression of secondary formations.

3.4 Elaborations of different episode cases

During the sampling periods, the concentration of $PM_{2.5}$ and its chemical components accumulated within a short period in a few cases (Fig.S15a–c). We define such a rapid rise in $PM_{2.5}$ mass concentration as a pollution episode. As shown in Table 1, meteorological conditions, the concentration levels of gaseous pollutants, chemical compositions, and source contributions of $PM_{2.5}$ during pollution episodes in three pilot cities are summarized. The episodes were accompanied by low wind speed (< 2 m s^{-1}), leading to weak dispersions of fresh emissions and accumulated pollutants (Chen et al., 2020b). OA and NO_3^- were the two dominant chemical components in $PM_{2.5}$ during all pollution episode cases with fractions of 26–40% and 23–32%, respectively. Their high abundances could be explained by the significant reduction of SO_2 emissions because of the prohibiting of burning bulk coals and executing the "Coal-to-Natural Gas" policy in recent years (Meng et al., 2022). In this study, eight pollution episodes (donated as EP1-EP8) were classified into three types: The first type was dominated by biomass burning (30–40%) (EP1, EP4, and EP8). The second type was dominated by secondary formation source (61–70%) (EP5, EP6, and EP7). The two remaining pollution episodes were mutually contributed by both primary and secondary sources (EP2 and EP3), in which secondary formation source (34–39%) and biomass burning (23–24%) were the two dominant contributors to $PM_{2.5}$.

To gain insights into the process of pollution episodes, three typical pollution events were chosen for detailed discussion (i.e., EP2 in Xi'an, EP4 in Shijiazhuang, and EP7 in Beijing) based on the validity and integrity of the data and the representativeness of the selected pollution events. The two-stage evolution was distinguished for EP4 as an example of the first type of episode (Fig. S16). At Stage 1, the $PM_{2.5}$ mass concentrations rapidly increased from 7 to 82 μg m^{-3} under stable weather conditions inferring by low wind speed (1.8±0.8 m s^{-1}, Fig. S16b), in which the concentrations (fractions) of biomass burning increased from 0.6 μg m^{-3} (7%) to 36.7 μg m^{-3} (55%) due to heating activities during nighttime. Meanwhile, the chemical composition was relatively stable and dominated by OA (31±5%)

and NO_3^-(21±5%). At Stage 2, the $PM_{2.5}$ mass concentration continuously increased to 105 μg m^{-3} in a few hours along with the most notable abundance of the source of secondary formation source, which concentration (contribution) rapidly increased from 2.3 μg m^{-3} (4%) to 54.4 μg m^{-3} (52%) (Fig. S16g and h). This is due to the aqueous-phase reactions effect inferring by the rapid increase in ALWC (from 16 μg m^{-3} to 78 μg m^{-3}, Fig. S16c) and RH (from 51% to 79%, Fig. S16a).

In contrast, a three-stage evolution was discriminated for the second type of episode, using EP7 as an example (Fig. S17). At Stage 1, the $PM_{2.5}$ concentration gradually increased from 11 to 30 μg m^{-3}, as well as NO_2 (from 15 to 59 μg m^{-3}, Fig. S17d) due to the boosts of the predominant contributions of vehicle emission and biomass burning (Fig. S17g and h). In the meantime, the contribution of coal combustion also slowly increased along with SO_2 (Fig. S17d and h). At Stage 2, under the lowest average wind speed in the study period (0.7±0.4 m s^{-1}, Fig. S17b), the $PM_{2.5}$ mass concentrations moderately increased from 30 to 91 μg m^{-3} with relatively stable chemical composition and source contribution (Fig. S17f and h). Compared to Stage 1, the fractions of NO_3^- increased mostly from 9±3% to 23±3%, this is probably influenced by photochemical oxidations inferring by relative high O_x and NO_2 concentration (Fig. S17c and d). At Stage 3, the $PM_{2.5}$ mass concentration rapidly rose to 142 μg m^{-3} and then remained stable. Furthermore, the greatest increase of sources concentrations (contribution) was secondary formation source from 18.9 μg m^{-3} (48%) to 120.6 μg m^{-3} (80%). This might be due to the occurrence of an aqueous-phase reaction, which was indicated by the elevation of RH and ALWC (Fig. S17a and c).

Figure S16 illustrates the third type of episode using EP2 as an example, in which a four-stages evolution was resolved. At Stage 1, the $PM_{2.5}$ mass concentration (14±3 μg m^{-3}) was relatively low and dominated by the contributions of secondary formation source (43±17%) and fugitive dust (24±8%), as well as MD (28±7%) and OA (26±7%). At Stage 2, the $PM_{2.5}$ mass concentrations promptly increased from 21 to 82 μg m^{-3}, with the two dominant chemical components of OA (21.7 μg m^{-3}) and NO_3^- (17.1 μg m^{-3}). The $PM_{2.5}$ increases can be also attributed to the raise of secondary formation source (25.3 μg m^{-3}) and biomass burning (14.4 μg m^{-3}). The enhancement of secondary aerosol was probably generated through the aqueous-phase reaction evidenced by the increase of ALWC and NO_2 (Fig. S18c and d). At Stage 3, $PM_{2.5}$ mass continuously increased to 139 μg m^{-3} with a dominant increase of primary sources emission including biomass burning (29.0 μg m^{-3}), vehicle emission (21.5 μg m^{-3}) and coal combustion (16.5 μg m^{-3}) along with the increases of SO_2 and NO_2 as well (Fig. S18d). The three primary sources contributed >60% of the total resolved sources. Meanwhile, the secondary formation source also increased slowly through aqueous-phase

reaction inferring by increase of ALWC (Fig. S18c). At the final Stage 4, the $PM_{2.5}$ mass concentration maintained relatively stable with an average of 142±11 μg m^{-3}, dominated by secondary formation source (34±6%) and biomass burning (28±6%); and chemical components of OA (36±4%) and NO_3^- (25±1%).

In summary, the pollution events occurred in Xi'an was mainly derived by stronger emissions of primary sources under adverse meteorological conditions, even though the aqueous-phase reaction also contribute to secondary aerosol formation. In contrast, pollution events occurred in Shijiazhuang and Beijing were mainly influenced by formation of secondary aerosols through both of aqueous-phase reaction and photochemical oxidation. What's more, aqueous-phase reaction plays a more important role than photochemical oxidation. Hence, to further improve the air quality in the north of China, primary source emissions should be prioritized for control in the northwest region, with a focus on biomass burning and coal combustion. In the North China Plain, the priority should be given to reduce emissions of the precursors from secondary sources, with a focus on NO_x and volatile organic compounds (VOCs).

3.5 Policy implications

In past decades, the air quality in China improved notably under the implementation of air pollution control policies including APCAP and TAPFAP. The $PM_{2.5}$ mass in Xi'an, Shijiazhuang and Beijing were the lowest during campaigns compared with those in last decades (Table S7). The variations of the chemical composition and the source contribution of $PM_{2.5}$ in the three pilot cities are displayed in Fig. 5. As shown, the dominant chemical components of $PM_{2.5}$ changed from OA and SO_4^{2-}, to OA and NO_3^- (Fig. 5a–c). This could be attributed to the reduction in coal consumption due to clean energy replacement and the increase of vehicle ownership, which lead a decrease of the SO_2 and an increase of NO_2 (Wang et al., 2013). Since the atmospheric oxidation reaction (i.e., aqueous-phase reaction and photochemical oxidation) of the precursors (i.e., NO_2, VOCs) is the primary source for the OA and NO_3^- in the atmosphere (Feng et al., 2018; Li et al., 2022; Tao et al., 2016; Yang et al., 2022b; Ziemann and Atkinson, 2012), and it is impossible to avoid, thus, the precursors of OA and NO_3^- should be reduced from the combustion and transportation sectors (Fermo et al., 2021; Liu et al., 2022; Wang et al., 2021c; Zhang et al., 2019). In addition, the fraction of NH_4^+ in $PM_{2.5}$ increased with an alarming rate. This is coincidentally in a similar trend of NH_3. Studies have reported that controls of NH_4^+ is more effective than that of NO_x in the reduction of $PM_{2.5}$ mass concentrations (Gu et al., 2021; Zheng et al., 2022). Therefore, collaborative control measures for the emissions of precursors including NO_x, VOCs, and NH_3 are necessary.

As shown Table S8 and Fig. 5d-f, coal combustion decreased remarkably due to the coal-related policies implementation including the strength of emissions standards for coal-fired power plants, the change of energy sources from coal to natural gas in some industrials, and the coal burning was forbidden in the main urban areas (Shen, 2016; Yang and Teng, 2018). The similar trend was also founded in the results of $PM_{2.5}$ source apportionment in Beijing released by Beijing Municipal Ecology and Environment Bureau (Fig. S19). Meanwhile, the contribution of industrial emission and vehicle emission decreased slightly because of the improvement of industrial emission standards (He et al., 2020; Wang et al., 2020a) and the traffic-related policy implementation such as the strength of vehicle emission standards, improvement of fuel quality, and elimination of high-emission-vehicles. This resulted in the reduction of the precursor gases and $PM_{2.5}$ from vehicles (Feng et al., 2021; Fontaras et al., 2012; Jin et al., 2012). However, the emission of biomass burning did not show a significant reduction in recent years, and its contribution increased from 9% in 2014 to 25% in 2020 (Xi'an), from 3% in 2015 to 24% in 2022 (Shijiazhuang), and from 6% in 2013 to 18% in 2021 (Beijing) (Fig. 5d-f). This is likely because biomass burning is an open source, which makes it more difficult to control compare with other primary sources. Biomass used for residential heating in rural areas is still frequently occurred (Ren, 2021; Tian et al., 2022; Yang et al., 2022a; Zhang et al., 2017). Hence, the clean energy revolution should be promoted urgently especially in the entire regions in northwest China. Moreover, the contributions of secondary formation source increased, it is potentially explained by the high reduction rate of primary emissions and the improvement of atmospheric oxidation capacity (Chen et al., 2020a; Feng et al., 2020). Therefore, more control measures should focus on weakening the atmospheric oxidation capacity, such as reduction of O_3 formation, to reduce the formation of secondary pollutants which are now identified as the most critical drivers of pollution. Considering those factors, it is also important to promote the mitigation of *both* $PM_{2.5}$ and O_3.

4. Conclusion

The intensive real-time measurement campaigns about $PM_{2.5}$ chemical components were conducted in Xi'an, Shijiazhuang, and Beijing during the wintertime respectively. Chemical compositions of $PM_{2.5}$ in the three cities were all dominated by OA (26.9–34.2%) and NO_3^- (23.6–26.5%). Six sources of $PM_{2.5}$ in Xi'an and Shijiazhuang were resolved by HERM and their contributions were similar, with a descending order of secondary formation source (32.2–37.6%), biomass burning (24.4–24.6 %), coal combustion (15.1–16.0%), vehicle emission (12.2–12.5 %), industrial emission (5.5–7.7%) and fugitive dust (4.4–7.8%). However, the secondary nitrate (29.0%) and the secondary sulfate (23.0%) were separately resolved and relatively more important in Beijing. In addition, the contribution of firework (7.9%) to $PM_{2.5}$ was found during the Chinese Spring Festival.

The possible formation mechanism of secondary formation source in three pilot cities was explored. The results showed that secondary aerosols were generated by both photochemical oxidation and aqueous-phase reaction. Meanwhile, the formation rate of secondary aerosols in Beijing was higher than that in Xi'an and Shijiazhuang. Furthermore, the eight pollution episodes within the sampling periods were categorized three types and characterized respectively. The dominant chemical compositions of $PM_{2.5}$ were OA (26–40%) and NO_3^- (23–32%) during all pollution episodes. Furthermore, secondary formation source and biomass burning were two major derivers of the pollution.

The dominant chemical components of $PM_{2.5}$ in pilot cities have changed from OA and SO_4^{2-} to OA and NO_3^- under the implementation of a clean air plan in past decades. This indicates that reduction of precursors including NO_2 and VOCs should be a key task in the future. In addition, the contribution of biomass burning increased, especially in Xi'an. This indicates that clean energy for heating activities in rural areas in northwest China is still insufficient. Furthermore, to weaken the atmospheric oxidation capacity for reducing the contribution of secondary formation source, it is necessary to promote the collaborative control on ozone and particulate matter.

Data availability. Data used to support the findings in this study are archived at the Institute of Earth Environment, Chinese Academy of Sciences, and are publicly available at https://doi.org/10.5281/zenodo.8106655.

Competing interest. The authors declare that they have no conflict of interest.

Author contributions. QW, YH, JC designed the campaigns. WR and YZ conducted the field measurements. YZ, JT, HL, LQ and TC performed data analysis and interpretation. MM, KRD, JGS and ASHP were involved supervision and review. YZ wrote the paper with contributions from all co-authors.

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852 **Table 1**. Meteorological conditions, gas pollutants, chemical composition, and source contribution of $PM_{2.5}$ during

853 pollution episodes in Xi'an, Shijiazhuang, and Beijing

Parameters	Xi'an		Shijiazhuang			Beijing		
	EP1	EP2	EP3	EP4	EP5	EP6	EP7	EP8
T (℃)	4.9 ± 2.6	1.5 ± 3.5	0.4 ± 3.4	-0.2 ± 3.3	-2.7 ± 3.3	0.3 ± 3.2	-1.6 ± 3.3	5.1 ± 3.6
RH (%)	52 ± 10	45 ± 10	61 ± 15	40 ± 13	75 ± 11	57 ± 19	45 ± 24	36 ± 10
WS ($m\ s^{-1}$)	0.5 ± 0.2	0.7 ± 0.3	1.4 ± 0.6	1.9 ± 0.8	1.4 ± 0.7	1.0 ± 0.6	1.1 ± 0.6	1.0 ± 0.6
Dominant WD[a]	WSW, WNW	WSW	NNW	NNW	NNW	NNE	NNW, NNE	NNE, ENE
CO ($mg\ m^{-3}$)	1.39 ± 0.40	1.15 ± 0.56	1.47 ± 0.62	0.60 ± 0.30	0.43 ± 0.33	1.04 ± 0.56	0.81 ± 0.32	1.00 ± 0.55
SO_2 ($\mu g\ m^{-3}$)	15 ± 3	15 ± 5	9 ± 4	8 ± 4	3 ± 1	6 ± 5	4 ± 3	6 ± 4
NO_2 ($\mu g\ m^{-3}$)	74 ± 22	63 ± 32	63 ± 14	47 ± 21	27 ± 11	54 ± 22	46 ± 17	42 ± 21
O_x (ppm)	47 ± 8	42 ± 10	36 ± 7	32 ± 6	27 ± 3	36 ± 6	33 ± 4	43 ± 9
ALWC ($\mu g\ m^{-3}$)	15 ± 11	8 ± 8	42 ± 37	12 ± 11	59 ± 448	28 ± 47	23 ± 8	11 ± 13
Dominant Chemical composition	OA (38%) NO_3^- (24%)	OA (34%) NO_3^- (24%)	NO_3^- (27%) OA (26%)	OA (30%) NO_3^- (23%)	NO_3^- (32%) OA (26%)	OA (32%) NO_3^- (26%)	OA (32%) NO_3^- (26%)	OA (40%) NO_3^- (23%)
Dominant source contribution[b]	BB (30%) SF (25%) CC (17%)	SF (34%) BB (24%) VE (16%)	SF (39%) BB (23%) CC (16%)	BB (40%) CC (16%) VE (16%)	SF (70%) BB (16%)	SF (62%) BB (13%)	SF (61%) BB (14%)	BB (38%) SF (27%) VE (15%)

854 [a] WSW: west-southwest; WNW: west-northwest; NNW: north-northwest; NNE: north-northeast; ENE: East-northeast

855 [b] BB: biomass burning; SF: secondary formation source; CC: coal combustion; VE: vehicle emission

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Figure captions:

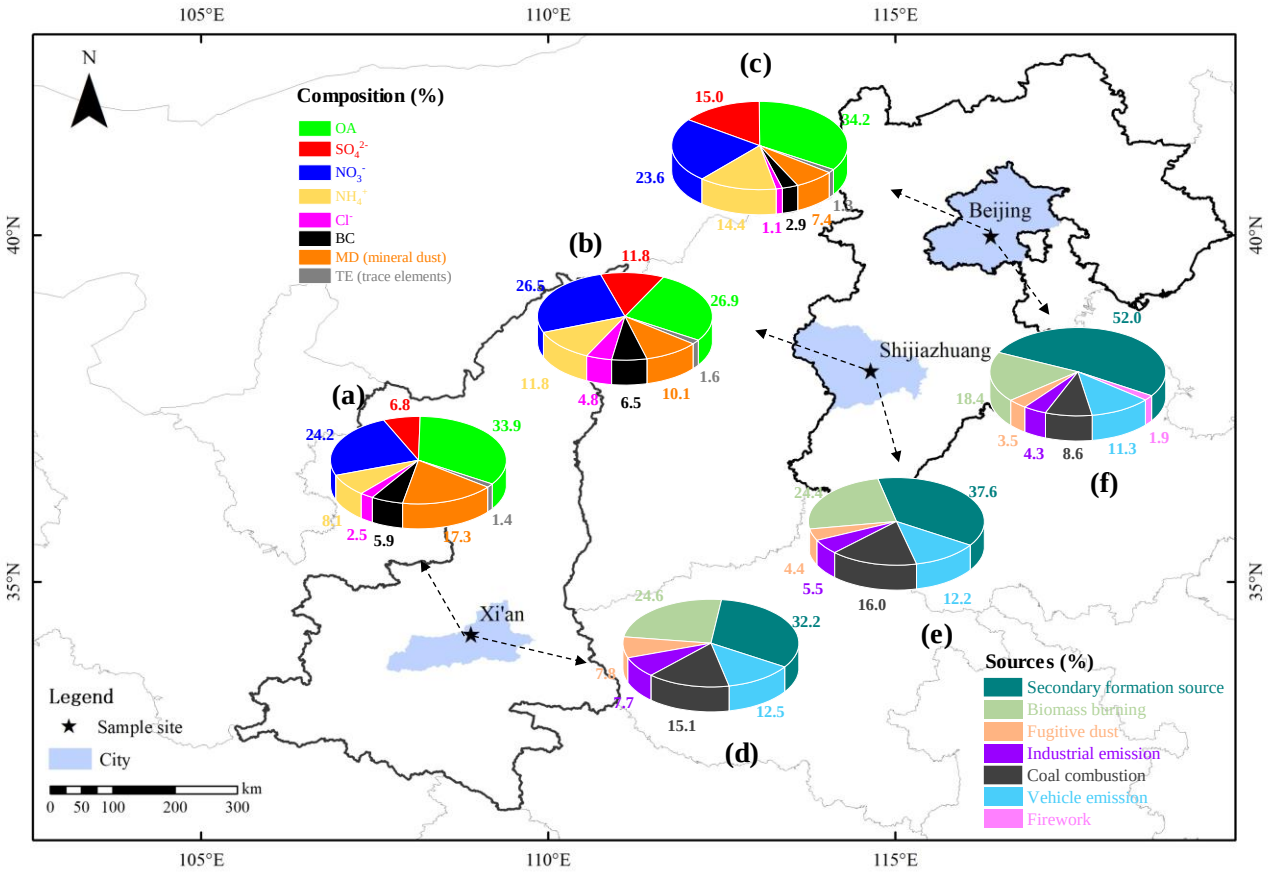
Figure 1. Chemical composition and source apportionment results of $PM_{2.5}$ in three pilot cities of northern China during the sampling period.

Figure 2. Mass fractions of chemical components (a-c) and sources contribution (d-f) with reconstructed $PM_{2.5}$ concentration in Xi'an, Shijiazhuang, and Beijing.

Figure 3. Correlations of secondary formation source/ΔCO and O_x mixing ratio in (a) Xi'an, (b) Shijiazhuang, and (c) Beijing. Each point and its error bar represent the mean and standard deviation in each bin (ΔO_x = 5 ppb).

Figure 4. Correlation of secondary formation source/ΔCO and ALWC during sampling periods in (a) Xi'an, (b) Shijiazhuang, and (c) Beijing, respectively. The points and error bar represent the mean values and standard deviation values of secondary formation source/ΔCO and ALWC in each bin. In Xi'an, each bin is 5 μg m^{-3} (ΔALWC = 5 μg m^{-3}). In Shijiazhuang, each bin is 5 μg m^{-3} (ΔALWC = 5 μg m^{-3}) when ALWC ranged from 0 to 75μg m^{-3}, but 25 μg m^{-3} (ΔALWC = 25 μg m^{-3}) for ALWC ranged from 75 to 200 μg m^{-3} due to limitations in data. In Beijing, each bin is 5 μg m^{-3} (ΔALWC = 5 μg m^{-3}) when ALWC ranged from 0 to 50 μg m^{-3}, but 100 μg m^{-3} (ΔALWC = 100 μg m^{-3}) for ALWC ranged from 50 to 900 μg m^{-3} due to limitations in data.

Figure 5. Summary of $PM_{2.5}$ and its composition (a, b, c) and source contribution (d, e, f) in Xi'an, Shijiazhuang, and Beijing in winter in past decades. Where * represents the result of this study. The data and references used for this figure are listed in Table S5 and S7.



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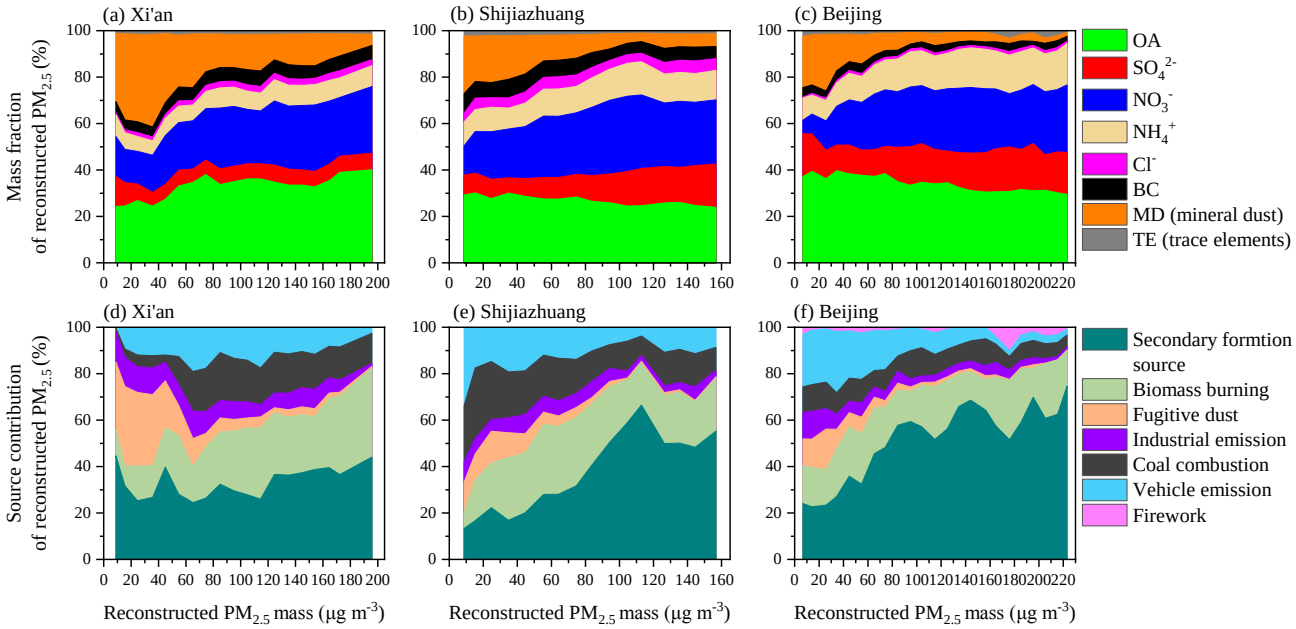

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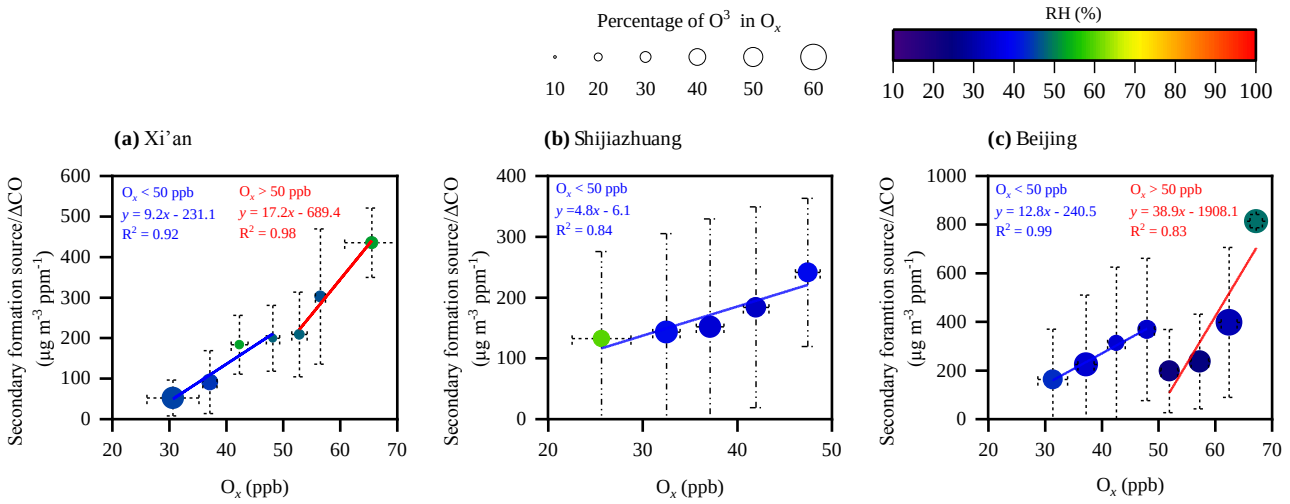

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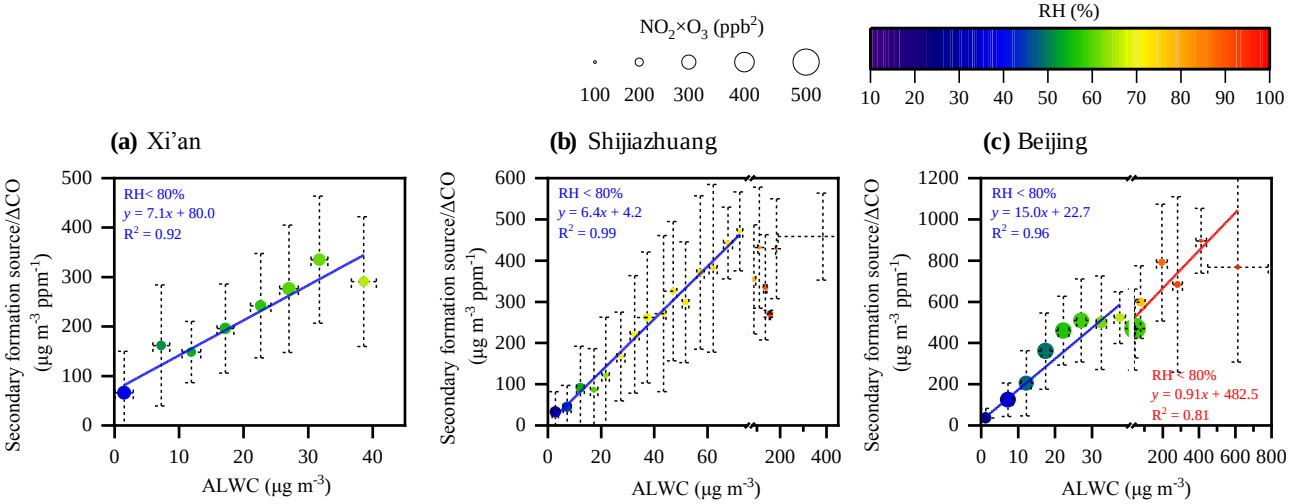

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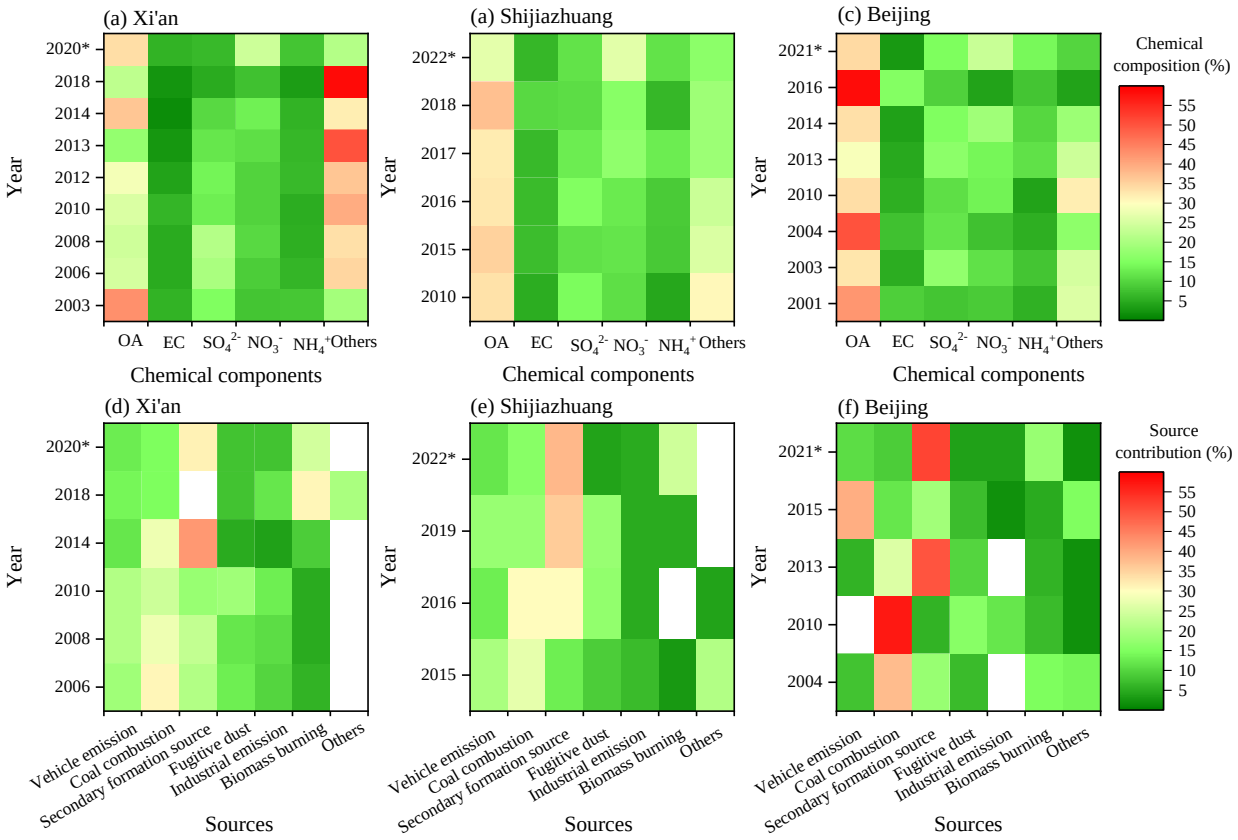

Figure 5. Summary of $PM_{2.5}$ and its composition (a, b, c) and source contribution (d, e, f) in Xi'an, Shijiazhuang, and Beijing in winter in past decades. Where * represents the result of this study, and the empty white area means no data. The data and references used for this figure are listed in Table S7 and S8.