INTRODUCTION TO THE NATIONAL AEROSOL CHEMICAL COMPOSITION MONITORING NETWORK OF CHINA

Objectives, Current Status, and Outlook

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NACMON observations provide valuable insights to understand chemical characterizations and source apportionments of aerosols in the Northern China Plain.

China is a big anthropogenic emission source in the world and contributes approximately 18%–35% of the global air pollutant emissions (Hosely et al. 2018). A large quantity of anthropogenic emissions released into atmosphere results in deterioration of air quality and threat of human health (Nel 2005). The Northern China Plain (NCP) is one of the major emission regions in China and extremely high $PM_{2.5}$ pollution (haze; $PM_{2.5}$ is particulate matter that is

smaller than $2.5\ \mu m$ in diameter) frequently occurs in winter. Over the last decades, many studies characterized chemical compositions and physical properties of aerosols, and quantified the potential sources of $PM_{2.5}$ in haze events (Sun et al. 2006; Guo et al. 2014; Huang et al. 2014; Li and Zhang 2014; Zhao et al. 2013; An et al. 2019). These studies concluded that haze in the NCP region is a complex air pollution problem and its formation was attributed to many reasons. High

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anthropogenic emissions such as coal combustion for power supply and residential heating were dominant sources of $\text{PM}_{2.5}$ in winter, which contributed 54% to $\text{PM}_{2.5}$ (Zhang et al. 2013). A large fraction (~50%) of secondary aerosols in Beijing during the 2013 winter haze suggested that efficient transformation of secondary aerosols from their precursors was also an important way of haze formation, especially under high relative humidity (RH) conditions (Huang et al. 2014; Xie et al. 2015; Cheng et al. 2016). On the other hand, regional transport of polluted air and calm conditions, which favored accumulation of air pollution, also resulted in extremely high $\text{PM}_{2.5}$ pollution in this region (Guo et al. 2014; Sun et al. 2016).

Previous many studies investigated the aerosol compositions and source apportionments along with the formation mechanisms of haze in the NCP region. These studies were regularly conducted at individual cities (often one or two such as Beijing and Tianjin). However, haze is a regional-scale air pollution and regional transport can play an important role in forming regional haze pollution in the NCP. Limited to previous results without long-term and comprehensive observation-based network, the regional effects and evolutions of chemical characteristics in aerosols during haze events are not well understood. Therefore, the source apportionment results are often limited to those megacities and the understanding of nearby cities is scarce. To improve the air quality in the NCP region, Chinese government has implemented the “2 + 26” regional strategy since early 2017 (Chen et al. 2019). In the “2 + 26” regional strategy, Beijing and other 27 major cities in NCP were selected as the target places to strictly reduce the airborne $\text{PM}_{2.5}$ concentrations (details of the “2 + 26” strategy can be seen in the online supplementary section “‘2 + 26’ regional strategy”). To evaluate the effects of “2 + 26” regional strategy for air quality improvement, China’s National Aerosol Composition Monitoring Network (NACMON) selected 31 sites, which cover the “2 + 26” cities, to conduct daily aerosol sampling and online measurements of other air pollutants (Table 1 and next section). All the daily aerosol sampling has been set up since March 2017 and online measurements have been gradually installed since late 2018. Based on the NACMON observations, the objectives of scientific issues are 1) to investigate the source and formation process of $\text{PM}_{2.5}$ -driven haze pollution, 2) to establish long-term databases to access the trends in aerosol chemical compositions at a national and regional scale, 3) to provide the validation of model results, 4) to help the Chinese government

make better decisions on air quality control and management, and 5) to evaluate the implemented effects of “2 + 26” strategy. Combining these observations with data analysis and model simulations, the ultimate objective is to reduce the frequency of haze formation and improve air quality in the NCP region. In this paper, we give a brief introduction of instrumentations and methodologies as well as the preliminary results of offline measurements in the NCP region from October 2017 to September 2018. Moreover, chemical characteristics and potential sources of $\text{PM}_{2.5}$ in the NCP are reported. The evolutions of chemical compositions during a haze event are also discussed. The results will help the Chinese government develop strategies for controlling anthropogenic emissions and improving air quality over the NCP region.

NACMON OBSERVATIONS. To implement the “2 + 26” strategy, NACMON conducted comprehensive online (since March 2017) and offline measurements (since late 2018) in 31 cities over the NCP region to obtain the chemical compositions and optical properties of aerosols (Chen et al. 2019). In terms of online measurements, water-soluble ions, organic carbon/elemental carbon (OC/EC), and aerosol optical properties were monitored at all the stations while size distributions, trace metals, and mixing states of aerosols were determined in seven cities (Table 1).

Water-soluble ions were monitored by an ambient ion monitor–ion chromatography (AIM-IC) instrument with a time resolution of 1 h. In addition to ions, AIM-IC also provides inorganic gases (SO_2 , NH_3 , and HNO_3). In general, multiple-ion standards were used to calibrate AIM-IC every week and the daily accuracy check was carried out to ensure the recoveries of all ions within 80%–120%. Hourly carbonaceous aerosols, including OC and EC, were measured by a semicontinuous OC/EC analyzer. The analyzer followed NIOSH 5040 procedure and employed the thermal optical transmittance (TOT) method to determine the OC/EC concentrations in aerosol samples (Yu et al. 2009). Both detection limits of OC and EC were $0.5 \mu\text{g m}^{-3}$ with precision of less than 5%. Aerosol optical properties were measured by lidar techniques. It operated at a 532-nm polarized laser with energy of 20 mJ and a pulse repetition rate of 20 Hz. The time and vertical resolutions are 1 min and 7.5 m, respectively. Based on the vertical and horizontal polarization signal detection, we can obtain several pieces of information such as atmospheric extinction coefficient, polarization ratio profile, and

TABLE I. Information of observed items in NACMON sites over the NCP region.								
Cities/ observations	Optical properties	Online measurements				Offline measurements		
		OC/EC	Ions	Metals	Mixing states	OC/EC	Ions	Metals
Beijing	•	•	•	•	•	•	•	•
Tianjin	•	•	•	•	•	•	•	•
Hebei Province								
Shijiazhuang	•	•	•	•	•	•	•	•
Baoding	•	•	•			•	•	•
Langfang	•	•	•			•	•	•
Zhangjiakou	•	•	•			•	•	•
Xiongan	•	•	•	•	•	•	•	•
Cangzhou								
Tangshan	•	•	•			•	•	•
Qinhd	•	•	•			•	•	•
Henshui	•	•	•			•	•	•
Xingtai	•	•	•			•	•	•
Handan	•	•	•			•	•	•
Henan Province								
Zhengzhou	•	•	•	•	•	•	•	•
Jiaozuo	•	•	•			•	•	•
Xinxiang	•	•	•			•	•	•
Kiafeng	•	•	•			•	•	•
Hebi	•	•	•			•	•	•
Puyang	•	•	•			•	•	•
Shandong Province								
Jinan	•	•	•	•	•	•	•	•
Heze	•	•	•			•	•	•
Jining	•	•	•			•	•	•
Liaochen	•	•	•			•	•	•
Dezhou	•	•	•			•	•	•
Zibo	•	•	•			•	•	•
Binzhou	•	•	•			•	•	•
Shanxi Province								
Taiyuan	•	•	•	•	•	•	•	•
Yangquan	•	•	•			•	•	•
Changzi	•	•	•			•	•	•
Jingchen	•	•	•			•	•	•
Anyang	•	•	•			•	•	•

mixing-layer height after retrieval of lidar-observed datasets. Hourly trace metal concentrations were continuously monitored by an online multielement monitor, which was based on nondestructive X-ray fluorescence spectroscopy (XRF). On the other hand, size distributions and mixing states of aerosols were measured through a single particle aerosol mass spectrometer (SPAMS). Utilizing the mixing states of

aerosols, the potential source and formation processes of aerosols would be identified and assessed (Murphy et al. 2006; Moffet and Prather 2009).

For offline measurements, daily concentrations of $PM_{2.5}$ mass, water-soluble ions, OC/EC, and metallic elements were obtained. The procedure of $PM_{2.5}$ sampling followed the regulation of “Determination of atmospheric particles PM_{10} and $PM_{2.5}$ in ambient

air by gravimetric method (HJ 618-2011).” Typically, the aerosol samples were collected using a low-flow air sampler (5 L min^{-1}), and quartz and polypropylene filters were used as filtration substrates. The former filter was used to analyze water-soluble ions and OC/EC, and the latter one was used for the determination of elements. Water-soluble ions, OC/EC, and trace metals in aerosol samples were analyzed by ion chromatography, OC/EC analyzer, and XRF, respectively. All measurements have their own standard operating procedures (SOP), which followed the national operating standard of China. To be able to provide accurate and precise data, both online and offline measurements within NACMON requires strict quality assurance (QA) and quality control (QC). The detailed information of these measurements in NACMON can be found in the online supplementary section “Measurements in NACMON.” Due to the comprehensive and long-term aerosol measurements, the formation mechanisms, source apportionments, regional effects of haze in the NCP, and the interactions between haze pollution and climate change will be explored in the future.

CHEMICAL COMPOSITIONS OF $\text{PM}_{2.5}$

In this section, we present the results of offline measurements of aerosols collected in the NCP region from 2017 October to 2018 September. During the sampling period, the annual average $\text{PM}_{2.5}$ mass

concentrations at the various sampling sites ranged from 53.5 to $112.5 \mu\text{g m}^{-3}$ (Fig. 1). Compared with the annual guideline ($35 \mu\text{g m}^{-3}$) suggested by the World Health Organization (WHO), the average $\text{PM}_{2.5}$ concentrations at different sites were 1.2–3.5 times higher. The Chinese government have made big efforts toward the reduction of industry and vehicle emissions for many years (Zheng et al. 2018), but the $\text{PM}_{2.5}$ mass concentrations at all monitoring sites still exceeded the Chinese grade I standard (annual mean is $35 \mu\text{g m}^{-3}$) by factors of 1.5–3.2, reflecting that PM pollution in the NCP was a serious problem. As shown in Fig. ES1 in the online supplement, higher $\text{PM}_{2.5}$ levels were usually observed in the wintertime, whereas lower $\text{PM}_{2.5}$ concentrations were found in the summertime, except for Beijing and Tianjin. The winter maximum and summer minimum of $\text{PM}_{2.5}$ was a typical seasonal cycle in China (Zhang and Cao 2015). The higher $\text{PM}_{2.5}$ levels observed in the cold season were attributed to the local meteorological conditions (e.g., stagnant weather and temperature inversion), which were unfavorable for dilution of air pollution, and enhancements of additional emission sources from fossil fuel combustion and biomass burning (Zhang and Cao 2015). Interestingly, in Beijing, the average $\text{PM}_{2.5}$ concentration in the wintertime was $43 \mu\text{g m}^{-3}$ (see Fig. ES1), which was much lower than that ($65 \mu\text{g m}^{-3}$) during

the summer. In terms of Tianjin, the average $\text{PM}_{2.5}$ mass during the winter was equal to that during the summer. These seasonal patterns were different from those observed in other cities. To improve air quality and decrease haze formation in Beijing, the Chinese government developed some strategies to reduce anthropogenic emissions, especially for coal combustion. For example, Beijing Gas Group has reduced coal consumption for residential heating year by year since 2013. According to the statistical data, 10 million tons of coal consumption was cut down in 2017; instead, natural gas was used as a substitute energy for

FIG. 1. The average concentrations of $\text{PM}_{2.5}$ mass and chemical compositions at 31 NACMON stations observed from October 2017 to September 2018.

residential heating (Zheng et al. 2018). The decrease of coal consumption might be a reasonable explanation for lower $\text{PM}_{2.5}$ concentrations in Beijing during the 2017 wintertime. Similarly, high $\text{PM}_{2.5}$ concentrations in Tianjin were found during wintertime in the previous studies (Zhou et al. 2017). The high $\text{PM}_{2.5}$ mass accompanying with dramatic enhancements of sulfate suggested that coal combustion was a major source (Zhou et al. 2017). In this work, the $\text{PM}_{2.5}$ concentrations in Tianjin during the winter season ($55 \mu\text{g m}^{-3}$) was very similar to those during the summer ($53 \mu\text{g m}^{-3}$) and autumn ($50 \mu\text{g m}^{-3}$) seasons, but lower than that in the spring ($64 \mu\text{g m}^{-3}$). The seasonal variations of $\text{PM}_{2.5}$ were attributed to complex conditions, such as meteorology, emission intensity, and regional transport of air pollution (Zhang and Cao 2015; Zhong et al. 2019). However, the reduction of coal consumption (reduced 2.6 million tons in 2017) in Tianjin might also partly explain the low $\text{PM}_{2.5}$ concentrations during the wintertime in this coastal city.

Chemical components of $\text{PM}_{2.5}$ were reconstructed through the chemical mass closure (CMC) approach, which has been widely employed for airborne PM samples in China (Zhang et al. 2013; Lin et al. 2016). Here, we constructed CMC by considering organic matter (OM), nitrate, sulfate, ammonium, mineral dust, elemental carbon, as well as trace elements (see Table ES1). OM, which is mainly emitted from either combustion sources or secondary transformation by VOCs, constituted a major fraction (17%–30%) of the total $\text{PM}_{2.5}$ mass in all cities, followed by nitrate (7%–23%), sulfate (9%–18%), ammonium (4%–13%), elemental carbon (3%–7%), mineral dust (2–8%), trace element (2%–4%), and chloride (1%–4%). Secondary inorganic aerosol (SIA; summation of SO_4^{2-} , NO_3^- and NH_4^+) accounted for 19%–53% of $\text{PM}_{2.5}$ mass. Here, we calculated 48-h backward trajectory arriving at the six selected cities (Beijing, Tianjin, Shijiazhuang, Zhengzhou, Jinan, and Taiyuan) to investigate the evolutions of chemical components depending on different air clusters. All air masses in each city were categorized into four clusters (see Fig. ES2 and online supplementary section “Backward trajectory”). As expected, the higher $\text{PM}_{2.5}$ mass with enriched secondary aerosol was observed when the air parcels were from southwestern (SW), south (S), and southeastern (SE) regions (Table ES2). The air of these clusters was usually characterized by low wind speed, high RH, and passed over high anthropogenic emission region (Fig. ES3), facilitating the transformation of gas to particles and therefore resulted in high $\text{PM}_{2.5}$ mass with high loadings of secondary aerosols (Sun

et al. 2006; Zhang et al. 2013; Zhao et al. 2013; Guo et al. 2014).

Furthermore, we compared the chemical compositions of $\text{PM}_{2.5}$ in Beijing between this work and earlier studies since the anthropogenic emissions of both primary PM and precursor gases have changed over the last two decades (Zheng et al. 2018). Figure ES4 shows the fraction of each chemical component of $\text{PM}_{2.5}$ in Beijing observed in this study and the literatures (He et al. 2001; Sun et al. 2004; Duan et al. 2006; Song et al. 2006; Zhang et al. 2013; Zhao et al. 2013; Park et al. 2018; Zheng et al. 2019). We attempted to investigate the trend of each chemical component by the comparisons. OM constituted a major fraction (21%–51%) of $\text{PM}_{2.5}$ mass, but no significant annual trends can be found. In addition, SIA was also a main chemical component in $\text{PM}_{2.5}$. The contribution of SIA to $\text{PM}_{2.5}$ mass varied from 23% to 45%. Before 2014, SO_4^{2-} was the most predominant species of SIA. However, nitrate has dominated SIA since 2015, with an annual average change rate of 0.6% ($p < 0.05$). Mineral dust seemed to be the third largest contributor to $\text{PM}_{2.5}$ with abundances of 2%–24%. Elemental carbon was mainly produced from combustion sources and contributed 2%–11% to $\text{PM}_{2.5}$ mass. Unlike nitrate, mineral dust and EC did not show any trends in the past 20 years. Due to an upward trend of nitrate from 2000 to 2018, enhancements of nitrate-to-sulfate mass ratios were also found over the NCP region. Several recent studies indicated that the mass ratio of nitrate to sulfate in $\text{PM}_{2.5}$ was higher than 1.0 in the NCP region (Li et al. 2018; Wei et al. 2018; Zou et al. 2018), which was different from those (nitrate-to-sulfate mass ratio ranged from 0.3 to 0.9) obtained in the last decade (Yao et al. 2002; He et al. 2012; Zhang et al. 2013; Zhao et al. 2013). The Chinese government has implemented clean-air actions since 2010. SO_2 emissions decreased through installation of flue gas desulfurization in coal-fired power plants and replacement of residential coal use with natural gas (Li et al. 2017; Zheng et al. 2018). In the NCP region, the annual SO_2 emission was cut down from ~1000 to ~400 Gg between 2008 and 2016, resulting in a decrease (~35%) of sulfate aerosols (Liu et al. 2018). Although NO_x emissions were also reduced due to clean-air actions, its change rate (–17%) was much smaller than that (–62%) of SO_2 (Zheng et al. 2018). As a consequence, the declined rate of nitrate was lower than sulfate, causing enhanced nitrate-to-sulfate ratios in airborne $\text{PM}_{2.5}$ in China recently. The elevated nitrate-to-sulfate ratio also highlighted that sulfate-dominant SIA has shifted to nitrate-dominant one, and how to control NO_x emissions and suppress nitrate aerosol formation will be an important issue in China.

FIG. 2. Relative abundance of each chemical component in $PM_{2.5}$ observed in (a) Beijing, (b) Tianjin, (c) Shijiazhuang, (d) Zhengzhou, (e) Jinan, and (f) Taiyuan cities under different $PM_{2.5}$ conditions. The definitions of various $PM_{2.5}$ conditions are described in the text.

NITRATE WAS A CONTRIBUTING SPECIES ON HAZE DAYS.

Haze pollution in China is mainly caused by stagnant weather and high RH conditions, and is a well-known regional-scale phenomenon that frequently occurs in winter (Li and Zhang 2014). To better understand the evolutions of chemical species in $PM_{2.5}$ from clear days to haze events, we classified the aerosol samples collected in the wintertime into low ($PM_{2.5} \leq 35 \mu g m^{-3}$), moderate ($35 < PM_{2.5} \leq 75 \mu g m^{-3}$), high ($75 < PM_{2.5} \leq 150 \mu g m^{-3}$), and haze PM ($PM_{2.5} > 150 \mu g m^{-3}$) categories. Nevertheless, in Shijiazhuang and Zhengzhou, the aerosol samples of low $PM_{2.5}$ category were only a few cases, and therefore, we extended the threshold value of low PM category to $45 \mu g m^{-3}$ at the both stations. As expected, the increased concentrations of all chemical components coincided with increasing $PM_{2.5}$ levels (Fig. ES5). If we checked the relative contribution of each component to $PM_{2.5}$ mass in

different air quality regimes, we would find some interesting results. The relative abundances of OM, EC, chloride, and mineral dust as well as trace elements decreased significantly during the haze days though the increases of these compositions were observed in some cities (Figs. 2 and 3). This suggested that OM, EC, and chloride were not the dominant contributing species to $PM_{2.5}$ haze in the NCP. The decrease of relative contribution in mineral dust demonstrated that fugitive dust emissions from paved and unpaved road dust and natural soil were not major sources of haze either. In terms of sulfate, the relative contribution to $PM_{2.5}$ typically kept constant levels or decreased during PM haze. In some cities, like Jinan (see Fig. 2), the sulfate contribution enhanced apparently (haze/low > 1.2) during haze days, indicating that sulfate might result in formation of PM haze in these cities (the details will be discussed in the following section). For nitrate and ammonium, enhancements of both

FIG. 3. Station numbers of haze/low ratios of different chemical components in $PM_{2.5}$ observed over the NCP region.

absolute concentrations and relative abundances were observed in most cities during the haze days; their average enhanced ratios were 2.2 and 1.9, respectively. This did demonstrate that nitrate and ammonium were major species that can stimulate the formation of PM haze in the NCP. Hydrolysis of N_2O_5 on water surface in the preexisting aerosols with high relative humidity conditions was considered as important pathway for nitrate formation in a very polluted NCP region (Wang et al. 2017; He et al. 2018).

Previous several studies have suggested that significant enhancements of sulfate and nitrate were found in haze events in northern China and the increased ratio of sulfate was much faster than that of nitrate (He et al. 2014; Wang et al. 2013; Sun et al. 2016). However, we found that NO_3^- and NH_4^+ were major contributing species to $\text{PM}_{2.5}$ during the haze days. Again, this implied that reduction of NO_x and NH_3 emissions was a crucial way to alleviate the formation of PM haze and improve air quality in the NCP.

SOURCE APPORTIONMENTS OF $\text{PM}_{2.5}$ IN POLLUTED SEASON.

We quantified source apportionments of $\text{PM}_{2.5}$ in the six major cities by the positive matrix factorization (PMF) model. The determined 16 species served as input data of PMF. We tried to resolve four to nine potential sources of ambient $\text{PM}_{2.5}$. According to the $Q_{\text{true}}/Q_{\text{expected}}$ values (Fig. ES6 and online supplementary section “PMF approach”), we thought six factors were the optimum solution in our case. The modeled $\text{PM}_{2.5}$ mass was in line with that of the observation ($R^2 = 0.95$; Fig. ES7). On average, the model underestimated approximately 5% of the observed $\text{PM}_{2.5}$ concentrations. By using the PMF model, traffic emission, secondary aerosols, coal combustion, biomass burning, and steel industry along with mineral dust were quantified (see Fig. ES8). The relative contributions of various sources fluctuated evidently during the wintertime (Fig. ES9). In general, secondary aerosols were the most predominant source, contributing 23%–39% to $\text{PM}_{2.5}$ (see Fig. 4), followed by traffic emissions (17%–27%), mineral dust (11%–18%), coal combustion (10%–16%), steel industry (6%–21%), and biomass burning (7%–19%). Numerous studies have estimated the source apportionments of $\text{PM}_{2.5}$ in Beijing by the PMF/chemical mass balance (CMB) model over the last two decades (Zheng et al. 2005; Zhang et al. 2007; Zhang et al. 2013; Zíková et al. 2016; Zheng et al. 2019; Huang et al. 2014; Wang et al. 2016; Gao et al. 2018). We attempted to compare our PMF results with the earlier studies and to check if any trends of source apportionments can

be found since the anthropogenic emission changed significantly in the past 10 years. Secondary aerosols were a dominant source of $\text{PM}_{2.5}$ in this capital city all along. A positive trend was found with an annual change rate of $2.2\% \text{ yr}^{-1}$ ($p < 0.05$). Inorganic species dominated the secondary aerosol sources (see Fig. ES8), demonstrating that control of NO_x and SO_2 emissions is important way to decrease PM levels. For primary sources, coal combustion and traffic emissions were major contributors of $\text{PM}_{2.5}$ in Beijing. Previously, several studies showed that coal combustion was the largest primary source with relative contribution being usually higher than 26% (Fig. ES10). Coal is the primary energy source in China, and the use of coal ranges from power plants and industries to domestic households. In this work, the contribution of coal combustion was 18%, which was half of that in 2014 and 2015 (Gao et al. 2018; Zheng et al. 2019). This might reflect the effectiveness of control of SO_2 emissions in Beijing in 2017 as mentioned above. Biomass burning was another emission source of $\text{PM}_{2.5}$. As shown in Fig. ES10, a significant negative trend with a slope of $-1.3\% \text{ yr}^{-1}$ ($p < 0.05$) was found. This might be because the Chinese government prohibited burning harvest crop residuals in the 2010s. Mineral dust (e.g., paved and unpaved road dust, soil) and industries were minor sources of $\text{PM}_{2.5}$; their contributions were regularly lower than 15% in Beijing. However, mineral dust contained enriched transition metal ions, which are an important catalyst of sulfate formation during in-cloud processes and resulted in indirect effects on climate change (Harris et al. 2013). In this work, steel manufacturing was the only industrial source estimated by PMF, and its contribution was 7% in Beijing. However, quantities of SO_2 , NO_x , and VOC emitted from industry emissions would transform to secondary aerosols in the atmosphere. Unfortunately, the sources of secondary aerosols could not be estimated by the PMF model. Consequently, the relative contribution of industry emissions to $\text{PM}_{2.5}$ might be underestimated, and a new approach to exactly partition the potential sources of secondary aerosols will be needed in the future.

We also investigated source apportionments of $\text{PM}_{2.5}$ under various air quality conditions. As shown in Fig. 5, the contributions of mineral dust decreased evidently with increasing $\text{PM}_{2.5}$ levels in all cities. The contributions of biomass burning and traffic emissions stayed at constant levels or decreased in high $\text{PM}_{2.5}$ levels. For coal combustion, its contribution to $\text{PM}_{2.5}$ decreased obviously with increasing $\text{PM}_{2.5}$ levels in most cities. However, in Beijing, the

FIG. 4. Source apportionments of $PM_{2.5}$ in the six selected cities over the NCP region from December 2017 to February 2018.

FIG. 5. Source apportionments of $PM_{2.5}$ in the six selected cities under various air quality conditions.

contribution of coal combustion increased from 15% on clear days to 22% on haze days. Although enhanced coal combustion was found on PM haze days in Beijing, the absolute contribution was much lower compared to the earlier study (57%; Zhang et al. 2013). This indicated that efficient control strategies decreased the contribution of coal combustion source to $PM_{2.5}$ in the capital of city. In terms of the steel industry, significant enhancements (haze/low ratio = 3.0) of steel industry pollution was only found in Shijiazhuang, suggesting that the controlled emission of steel manufacturing could decrease $PM_{2.5}$ concentrations in this city. The fraction of secondary aerosols increased substantially with increasing $PM_{2.5}$ levels in all selected cities. For instance, in Beijing, secondary aerosols contributed 14% to airborne $PM_{2.5}$ during low PM days, then increased to 19% under a moderate air quality condition. When $PM_{2.5}$ concentrations exceeded

75 and $150 \mu g m^{-3}$, the contributions of secondary aerosols rose up to 38% and 58%, respectively. The dramatic enhancements of secondary aerosols in the haze days were also observed in other cities (the haze/low ratio ranged from 1.8 for Taiyuan to 3.0 for Zhengzhou), highlighting that secondary aerosols were the dominant source of $PM_{2.5}$ during the PM haze events over the NCP region. The elevated secondary aerosols during the haze events might be attributed to both local and long-range transported air pollution (Huang et al. 2014). Indeed, secondary aerosols, especially for SIA, was a major source of fine PM during haze days throughout China. SIA is mainly transformed from NO_x and SO_2 emitted by vehicle and industry emissions and its emission sectors are not well understood yet. Thus, in the future, identification of potential sources of secondary aerosols will be an important issue for alleviation of PM haze formation.

EVOLUTIONS OF AEROSOL COMPONENTS IN A HAZE CASE.

During 13–18 January 2018, a regional-scale haze pollution deteriorated the air quality of the NCP region; this haze event lasted for one week in some cities. As shown in Fig. ES11, $\text{PM}_{2.5}$ concentrations in the selected cities began increasing on 13 January, remaining at high $\text{PM}_{2.5}$ levels in some cities (Shijiazhuang, Zhengzhou, Jinan, and Taiyuan) from 13 to 18 January, and then decreased after 19 January. During this period, the temperature was regularly below 0°C , and the wind speed was usually lower than 2 m s^{-1} over the NCP region (Fig. 6a shows the results in Beijing). Meanwhile, the humid air ($\text{RH} > 65\%$) was predominantly from eastern and southeastern regions on 12 and 13 January (Fig. ES12a), picking up polluted air and transporting it to the receptor sites. Consequently, dramatic increases of $\text{PM}_{2.5}$ concentrations were observed. Since 15 January, the air masses mainly originated from northwestern regions, crossing low emission areas, and brought drier air to the receptor site. As a result, apparent decreases of $\text{PM}_{2.5}$ were found in some cities.

In northern China, heterogeneous reactions are important ways to produce particulate sulfate and nitrate salts during the haze events (Wang et al. 2016; Wang et al. 2017). Aerosol liquid water content (ALWC) plays an important role in particulate sulfate and nitrate formation. ALWC can serve as a reactor for transformation of SO_2 to sulfate, thereby facilitating SO_4^{2-} formation (Cheng et al. 2016; Wang et al. 2016). In addition, a high

FIG. 6. Absolute concentration and relative abundances of chemical compositions in $\text{PM}_{2.5}$ along with weather parameters in (a) Beijing and (b) Zhengzhou during 8–24 Jan. The shadow denotes a regional-scale haze event in the NCP.

ALWC speeds up the uptake coefficient of N_2O_5 , enhancing nitrate formation during the haze pollution (Thornton et al. 2003; Bertram et al. 2009). In general, ALWC increases with increasing of ambient RH (Wu et al. 2018). Consequently, ambient RH would accelerate sulfate and nitrate formation (Sun et al. 2018). On the other hand, nitrate and sulfate aerosols will deliquesce when the ambient RH is higher than their own deliquescent RH (DRH; 80% for SO_4^{2-} and 62% for NO_3^- under 1-atm and 20°C conditions), resulting in increases of their sizes and concentrations (Jing et al. 2018; Sun et al. 2018). In this event, both absolute concentrations and relative abundances of nitrate increased significantly. Low ambient temperature facilitated the gas-to-particle conversion of nitrate, and 65% of RH explained enhanced nitrate in the haze event (An et al. 2019). Notably, sulfate concentrations increased evidently during the high $\text{PM}_{2.5}$ levels, but its contribution to $\text{PM}_{2.5}$ stayed at constant levels. Several studies suggested that heterogeneous reaction of SO_2 frequently occurred when the ambient RH was >80% in haze event over China (Xie et al. 2015; Cheng et al. 2016). In this event, the hourly RH in Beijing during this haze event was 60%–70%. The “relatively low” RH might not facilitate SO_2 to SO_4^{2-} via $\text{SO}_2 + \text{NO}_2$ reaction. Moreover, the ambient RH was lower than the DRH of sulfate, restricting dramatic buildup of sulfate aerosols. Therefore, the fraction of sulfate in $\text{PM}_{2.5}$ did not increase during the haze event. Similar results were also found in Tianjin and Taiyuan (Fig. ES13). During the haze period, the RH in both cities was lower than 80%. Although SO_4^{2-} concentrations rose, its contribution to $\text{PM}_{2.5}$ did not increase. On the contrary, both enhancements of absolute concentrations and relative abundances of NO_3^- were found.

Different evolutions of aerosol compositions were found in Zhengzhou during the same haze event (Fig. 6b). On 8–10 January, the elevated mineral dust was found with contribution of 29% in $\text{PM}_{2.5}$. The backward trajectory analysis revealed the air parcels mainly passed over the northwest region with high wind speed ($>4 \text{ m s}^{-1}$), bringing dry air and dust-enriched particles to the receptor site (Fig. ES12b). During this period, the $\text{PM}_{2.5}$ concentration was as low as $40 \mu\text{g m}^{-3}$. Because of low RH, we found low fractions of SIA in $\text{PM}_{2.5}$. Since 12 January, the air masses were primarily from west, south, and northeast directions and spent much time ($\text{WS} < 2 \text{ m s}^{-1}$) in traveling the neighboring areas of Zhengzhou with a large quantity of anthropogenic emission. Meanwhile, the ambient RH increased and kept at 80% in the next several days. Like Beijing, significant enhancements of nitrate concentration were found during this haze events. Also, both absolute concentrations

and relative abundances of sulfate in $\text{PM}_{2.5}$ increased apparently due to the high RH conditions and resulted in elevated $\text{PM}_{2.5}$ levels. After 23 January, the air masses came from the eastern region and were accompanied with high wind speed ($\text{WS} > 4 \text{ m s}^{-1}$) and lower RH (~60%). The high wind speed favored dilution of PM pollution, and lower RH did not facilitate the conversion of SO_2 to SO_4^{2-} , resulting in low concentrations of both $\text{PM}_{2.5}$ and sulfate. Although the nitrate concentrations were low, the relative contribution of nitrate was as high as during the haze. In Shijiazhuang and Jinan, the apparent increases of abundance in SO_4^{2-} were also attributed to the high RH (Fig. ES13). These findings also reflected that a threshold RH value of nitrate formation was lower than that of sulfate. This can partly explain why increases of nitrate contribution were in general found during the haze period over the NCP region. In conclusion, regional transport, meteorological conditions, and emission sources influenced the evolutions of aerosol compositions. Secondary aerosols were the dominant component during the haze events. In addition to primary PM emissions, control of the emissions of gases will be an important strategy to decrease secondary aerosol formation and improve air quality in the NCP region.

IMPLICATIONS AND FUTURE PERSPECTIVES.

In the NCP region, PM haze pollution frequently occurs during winter season. Emission sources, weather conditions, and transformation process were the main factors to trigger haze formation (Guo et al. 2014; An et al. 2019). In this paper, we, for the first time, systematically summarized the chemical compositions and source apportionments of aerosols in the NCP region through multistation observations. During the haze pollution, nitrate-dominant or sulfate-dominant aerosol in haze pollution was driven by ambient RH other than their precursor gases. Compared with previous studies, coal combustion and biomass burning revealed downward trends, whereas secondary aerosols, especially for NO_3^- , showed upward trends over the last two decades. This implied that decreasing nitrate concentrations played an important role in alleviation of haze formation. However, almost 40%–50% of nitrate and ammonium were in the presence of a secondary product that consists mainly of NH_4NO_3 . Therefore, control of NO_x and NH_3 emissions would be a way to decrease particulate nitrate ammonium concentrations. Coal burning, industries, and traffic emissions were dominant sources of NO_x in the NCP region (Zhao et al. 2012). These anthropogenic NO_x emissions have been reduced by ~20% in recent years (Zheng et al. 2018). For NH_3 , isotope techniques gave direct evidence that fossil

fuel combustion could be an important source for gaseous and particulate NH_3 in urban polluted atmosphere (Pan et al. 2016; Chang et al. 2018). This indicated that control of NH_3 emissions from nonagriculture activities has to be paid more attention for improving air quality in the NCP region.

Haze is a severe air pollution in China that is characterized by high concentrations of $\text{PM}_{2.5}$. The comprehensive aerosol measurements by NACMON provide us insights to understand the temporal and spatial distributions of aerosol chemical compositions. By using the PMF model, source apportionments of aerosols can be identified and help the Chinese government to develop strategies for improving air quality in this region. In future studies, some scientific topics need to be studied through aerosol observations combined with model simulation. For example, due to the long-term aerosol measurements, the effectiveness of the control of emissions on reduction of $\text{PM}_{2.5}$ concentrations can be evaluated (Chen et al. 2019). However, regional transport is one of the major reasons for haze formation in the NCP region (Guo et al. 2014; Huang et al. 2014; Zheng et al. 2015; Cheng et al. 2016). In this study, we cannot quantitatively estimate the influence of regional transport on high $\text{PM}_{2.5}$ pollution based on field observations. But this will be achieved through aerosol observations and model simulations in the future. Haze provides a scientific platform to improve many aspects of atmospheric chemistry and physics processes associated with extremely high PM concentrations (An et al. 2019). Some atmospheric processes occur on hourly scales (Wang et al. 2016). By using the online measurements, the evolutions of aerosol chemical compositions and atmospheric chemical and physics processes during haze events will be explored. Indeed, atmospheric aerosols, meteorological conditions, and climate change are linked together (Wang et al. 2014; Cai et al. 2017). Thus, studies of interactions and feedbacks between haze pollution and meteorological/climate change are needed. Finally, the human health effects of aerosols need to be evaluated since many studies showed that $\text{PM}_{2.5}$ increases human mortality. All these important issues cannot be investigated by “single point” aerosol observation. The NACMON provides comprehensive aerosol observed data in many cities over the NCP region. Combining with model simulation, we believe the scientific questions as mentioned above will be resolved in the future.

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