Effects of Regional Transport on Haze in the North China Plain: Transport of Precursors or Secondary Inorganic Aerosols

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Abstract Most previous studies treat regional transport of aerosols as a whole, without distinguishing the transport of secondary aerosols and that of their precursors. A new method of quantifying the transport forms of secondary inorganic aerosols (SIA) using the Nested Air Quality Prediction Modeling System was proposed. The contribution of nonlocal emissions to SIA in the receptor region was divided into three parts: (1) SIA chemically formed by nonlocal emissions in their source regions; (2) SIA chemically formed by nonlocal emissions during transport; and (3) SIA chemically formed by nonlocal emissions in the receptor region, representing transport of precursors. In the North China Plain, the transport of precursors and SIA produced during transport are the two main transport forms. Furthermore, the contribution from transport of precursors increased under polluted conditions in most cities. The results indicate that joint control of precursors is paramount for mitigating air pollution.

Plain Language Summary Uncertainties still exist about the sources of secondary inorganic aerosols (SIA), which are one of the crucial drivers of haze pollution. The spatial distribution of precursors is not exactly consistent with that of aerosols, which is puzzling to policymakers. Most existing studies only treat contribution from regional transport of nonlocal emissions as a whole but do not distinguish the transport of secondary aerosols themselves and that of their precursors. Therefore, it is necessary to quantify the contribution from transport of precursors and secondary aerosols. The Nested Air Quality Prediction Model System with an online tracer-tagging module was used to investigate the regional sources of SIA in Beijing and surrounding cities. The regional contribution of nonlocal emissions to SIA in the receptor region was divided into three parts: (1) SIA chemically formed by nonlocal emissions in their source region; (2) SIA chemically formed by nonlocal emissions in transport pathway; and (3) SIA chemically formed by nonlocal emissions in the receptor region, indicating transport of precursors. The results showed that transport of precursors and the SIA produced during transport are two main transport forms. The results strongly suggest that the joint control of precursors is important for mitigating air pollution.

1. Introduction

Pollution episodes have frequently occurred in the North China Plain in recent years (Li et al., 2017; Sun et al., 2014, 2016; Wang et al., 2014, 2018). High concentrations of aerosol particles lead to air pollution. This affects visibility and human health (Chen et al., 2018; Hyslop, 2009; Ouyang, 2014; Peng et al., 2016) and has attracted wide concern. Secondary inorganic aerosols (SIA) are critical drivers of severe haze in China (Huang et al., 2014; Wang et al., 2016; Zheng et al., 2015). From clean to polluted conditions, the ratio of SIA increases from 40% to 59% (Wang et al., 2016). Extensive studies have been conducted on the formation and regional sources of SIA, but debate and uncertainty still exist on the sources of SIA during periods of heavy pollution (Itahashi, Hatakeyama, et al., 2017; Itahashi, Uno, et al., 2017; Li et al., 2017). Local chemical conversion was thought to lead to an increase of SIA (Ma et al., 2017; Zhu et al., 2016), but researchers...
have also found that regional transport contributes substantially to SIA (Itahashi, Hatakeyama, et al., 2017; Li et al., 2017; Sun et al., 2014; Wu et al., 2017).

Studies have revealed that the concentration of particulate matter at receptor sites is higher than that in source regions (Li et al., 2017). The higher concentrations in the receptor region may be caused by the transport of precursors from the regional source region followed by chemical conversion in the receptor region or by the transport of secondary particles from the source region. Clarification is required on whether SIA are transported by precursors or as particles in order to further investigate the formation of SIA, but scant research has covered this topic (Xing et al., 2017). Furthermore, different source apportionment methods result in different regional contributions (Thunis et al., 2019). This is problematic as it means that regional sources may not be properly represented in the atmospheric models used for prediction and regulation.

This study investigated SIA transport forms using an air quality prediction model system by tagging both the emission region of the precursors and the formation region of the secondary particles during the period from 1 November to 31 December 2017. SIA in the receptor region is formed from local emission and regional transport of nonlocal emissions. In this study, based on the chemical conversion regions of nonlocal emissions, the contribution of nonlocal emissions to SIA was then divided into three parts: chemical conversion of nonlocal emissions in the source regions, during transport, and in the receptor region. The goal of this study is to distinguish the transport of precursors from the transport of secondary aerosols. The results will facilitate the identification of specific SIA source regions and provide a useful reference for the formulation of pollution control policies.

2. Materials and Methods

2.1. Model Description

The Nested Air Quality Prediction Model System (NAQPMS) was employed in this study (Wang et al., 2001). The 3-D Eulerian terrain-following air quality model has been widely used to investigate the sources and formation of regional dust, haze, and ozone pollution (Li et al., 2016, 2017; Wang et al., 2017). The composition and phase state of an inorganic aerosol system was calculated using a thermodynamic equilibrium model (ISORROPIA v1.7) (Nenes et al., 1998). Further technical details about the NAQPMS can be found in Li et al. (2012, 2011) and Du et al. (2019).

Two nested modeling domains were used in this study. The first domain covered most of eastern China, and the second domain covered Beijing and surrounding areas including Hebei, Tianjin, Henan, Shandong, Shanxi, and northern of Anhui and Jiangsu provinces (Figure S1a in the supporting information). The horizontal resolutions of the two domains were 27 km and 9 km, respectively, and there were 20 layers in the vertical direction. The meteorological field for the NAQPMS was provided by the Weather Research and Forecasting (version 3.6.1) model (Skamarock et al., 2008) driven by the European Centre for Medium-Range Weather Forecasts data. Observation nudging was used to minimize bias. The global chemistry transport model MOZART version 2.4 provided initial and lateral boundary conditions for the NAQPMS. The anthropogenic emission inventory entered into the model was obtained from the Multiresolution Emission Inventory for China with 2016 as the base year (http://www.meicmodel.org). The emission inventory was updated according to the observation data based on the latitude and longitude of the state control sites. November and December 2017 were chosen to represent autumn and winter, respectively. Each month was simulated separately, and the first 10 days of each simulation were set aside as a spin-up period to reduce the influence of the initial conditions.

Hourly surface PM$_{2.5}$ data for the state control sites were provided by the China National Environmental Monitoring Center (http://www.cnemc.cn/). The surface meteorology parameters were obtained from the China Meteorological Administration (http://data.cma.gov.cn/). The hourly concentrations of water-soluble ions in PM$_{2.5}$ in Beijing were monitored by an ambient ion monitor, and the daily water-soluble ions in surrounding cities of Beijing were measured by ion chromatography (Dao et al., 2019).

2.2. Source Apportionment Method

In this study, an online tracer-tagging module was coupled into the NAQPMS to quantify the contributions from the transport of secondary aerosols themselves and the SIA precursors. The source-tagging module is
able to quantify the contribution from chemical transformation of emissions in each tagged region. In this module, we assume that each chemical species is mixed well in each grid and shares the same loss coefficients. Therefore, the fraction of each tagged source in the SIA concentration at a given grid was changed by inflows from its adjacent grid cells and chemical transformation in the grid. For inflows from the upstream grid cell, the change in the fraction of each tagged source in the total SIA concentration is allocated proportionately to the source in the upstream region. For chemical processes, the module tagged the emission regions of precursors and chemical formation regions of SIA. And the chemical formation of SIA was apportioned to their precursors (e.g., sulfate to SO$_2$, nitrate to NO$_X$, and ammonium to NH$_3$) at each time step. More technical details about the method are presented in the studies of Wu et al. (2017) and Li et al. (2014).

\[
C_{ij}(t + \Delta t) = C_{ij}(t) + \Delta C_j \times \frac{\text{Precursor}_i}{\sum \text{Precursor}_i}
\]

where \(i\) is the \(i\)th emission region; \(j\) is the \(j\)th chemical conversion region; \(i\) and \(j\) can be 1 to \(n\); \(n\) is the total number of regions (\(n\) equals 34 in this study); \(\Delta C_j\) is the chemical conversion of precursors in region \(j\); \(\text{Precursor}_i\) is concentration of precursors from region \(i\); and \(C_{ij}\) means the SIA formed in region \(j\) by precursors from source region \(i\).

\[
C_i = \sum_{j = 1}^{n} C_{ij}
\]

\[
C_j = \sum_{i = 1}^{n} C_{ij}
\]

where \(C_i\) is the SIA formed by precursors emitted in source region \(i\). \(C_j\) is the SIA formed in region \(j\).

\[
F_{ij} = \frac{C_{ij}}{\sum_{i = 1}^{n} \sum_{j = 1}^{n} C_{ij}} \times 100\%
\]

where \(F_{ij}\) is the relative contribution of the SIA formed in region \(j\) by precursors from source region \(i\), and the sum of \(F_{ij}\) equals 100%. The “contribution” mentioned later in the context refers to relative contribution. Therefore, the contribution of precursors from any source region forming SIA in any region to the SIA in any grid cell can be calculated. To better analyze the sources of SIA, we defined the contribution from local

Figure 1. Schematic diagram of the sources of secondary inorganic aerosols (SIA). LOC_LOC is SIA formed by local emissions in the receptor region; LOC_TRA is SIA formed by local emissions during the transport pathway; REG_SOURCE is SIA chemically formed by nonlocal emissions in their source regions; REG_TRA is SIA chemically formed by nonlocal precursors during transport pathway to the receptor region; REG_REC is SIA chemically formed by nonlocal precursors in the receptor region.
emissions as local contribution, and the contribution from nonlocal emissions as regional transport. According to the chemical conversion regions (emission region, transport pathway, and receptor region) of local and nonlocal emissions, the total concentration of SIA at the receptor region was divided into five parts: ① LOC_LOC is the contribution of precursors from the receptor region forming SIA in the receptor region, representing local chemical conversion of local emissions; ② LOC_TRA is the contribution of precursors from the receptor region forming SIA in regions outside the receptor region and then flowing back to the receptor region, representing chemical conversion of local emission during transport; ③ REG_SOURCE is the contribution of precursors from regions outside the receptor region that form SIA in their source region, representing the SIA chemically formed by nonlocal emissions in their source regions, namely, regional transport of SIA; ④ REG_REC is the contribution of precursors from regions outside the receptor region forming SIA in the receptor region, representing the SIA chemically formed by nonlocal emissions in receptor region, namely, the regional transport of precursors; and ⑤ REG_TRA is the contribution of precursors from regions outside the receptor region forming SIA during the transport pathway to the receptor region, representing the SIA chemically formed by nonlocal emissions during the transport pathway, namely, the SIA produced during regional transport. The schematic diagram is presented in Figure 1. The sum of LOC_LOC and LOC_TRA is the local contribution, and the sum of REG_SOURCE, REG_TRA, and REG_REC is the contribution of regional transport (Table S1). Therefore, transport forms of SIA can be clarified by quantifying the contribution of REG_SOURCE, REG_TRA, and REG_REC, because they mean regional transport of secondary aerosols themselves, SIA produced during regional transport and regional transport of precursors, respectively.

In this study, 34 regions, according to administrative divisions, were selected for source tagging (Figure S1c). A total of 33 regions were located in Beijing, Tianjin, and other surrounding cities—Tangshan, Baoding, Shijiazhuang, Langfang, Cangzhou, Hengshui, Xingtai, Handan, and other cities in Hebei Province (HBOT); Taiyuan, Yangquan, Changzhi, Jincheng, and other cities in Shanxi Province (SXOT); Jinan, Zibo, Jining, Dezhou, Liaocheng, Binzhou, Heze, and other cities in Shandong Province (SDOT); and Zhengzhou, Kaifeng, Anyang, Hebi, Xinxian, Jiaozuo, Puyang, and other cities in Henan Province (HNOT). The region outside the 33 regions above is designated as other cities (OT).

2.3. Model Validation

The NAQPMS was able to simulate sulfate, nitrate, and ammonium well in the North China Plain (Figure 2). The correlation coefficient (R) between observations and simulations was 0.54–0.84. The normalized mean bias was low (~0.36 to 0.81). The simulation performance for PM$_{2.5}$ in Beijing, Tianjin, Dezhou, and Jinan was acceptable, and the performance in Baoding, Cangzhou, Handan, and Zhengzhou was close to optimal according to the criterion of Boylan (Boylan & Russell, 2006) (Figure S2). Four regional haze episodes were identified based on the time series of PM$_{2.5}$ according to the method of G. Zheng, Duan, et al. (2016). The peak concentration of each episode had to be higher than 150 μg/m$^3$ (Figure S2). The following four periods were included: 4–7 November 2017 (Ep1), 18–21 November 2017 (Ep2), 30 November to 3 December 2017 (Ep3), and 26–30 December 2017 (Ep4). The NAQPMS reproduced the spatial distribution of PM$_{2.5}$ during Episodes 1 to 4 (Figure S3). The favorable model performance for pollutants and components was related to the good simulation of meteorological parameters. The correlation coefficients of wind speed, temperature, and relative humidity in Beijing were 0.6, 0.9, and 0.8, respectively (Figure S4). Some underestimations of relative humidity and overestimations of wind speed were related to the choice of the planetary boundary layer scheme (Y. G. Zheng, Liu, et al., 2016).

3. Results

Ammonium sulfate (NH$_4$)$_2$SO$_4$ and ammonium nitrate (NH$_4$NO$_3$) contributed a large proportion of the particle extinction (Han et al., 2017; Wang et al., 2015). In this study, the sources of ammonium sulfate and ammonium nitrate in Beijing and surrounding cities were divided into local contributions (LOC_LOC and LOC_TRA) and regional transport (REG_SOURCE, REG_TRA, and REG_REC) according to the method shown in section 2.2 to distinguish between the transport of precursors and SIA.
3.1. Sources of Ammonium Sulfate and Ammonium Nitrate in Beijing

As the core city of the North China Plain city cluster, Beijing's simulated average PM$_{2.5}$ concentration during the study period was 45 μg/m$^3$, and peak PM$_{2.5}$ values frequently reached 150 μg/m$^3$. The mean ratio of SIA to PM$_{2.5}$ was 43%, and the ratio was 39% under relatively clean conditions (no higher than 115 μg/m$^3$) and 46% under polluted conditions (higher than 115 μg/m$^3$). Figure 3 shows the daily sources of (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ concentrations in Beijing during the study period. On average, the contribution of regional transport (the sum of REG_SOURCE, REG_TRA, and REG_REC) was more substantial than that of local emissions, and it was 58% and 56% to (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$, respectively. This result was consistent with those of other studies in which regional transport accounted for a large proportion of SIA (Itahashi, Hatakeyama, et al., 2017; Li et al., 2017; Sun et al., 2014). Regarding contribution of regional transport, the contribution of REG_SOURCE was low (approximately 7%). Contribution from SIA chemically formed by non-Beijing precursors in Beijing (REG_REC) to (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ was substantial, reaching 28% and 24%, respectively. The contribution from SIA chemically converted by non-Beijing precursors during transport pathway to Beijing (REG_TRA) was not negligible, with values of 23% and 27% to (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$, respectively. This result indicates that chemical conversion occurred during regional transport.

The local contribution was mainly from SIA generated by local precursors in Beijing, and it was 34% and 32% to (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$, respectively, in Beijing. Some precursors emitted in Beijing were blown away from and formed SIA in other regions. And the SIA can flow back to Beijing and accounted for 8% and 12% of (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$, respectively. A small difference was noted between the contribution to (NH$_4$)$_2$SO$_4$ and that to NH$_4$NO$_3$ from regional transport (58% vs. 56%, respectively). However, SIA chemically converted by non-Beijing precursors during transport pathway to Beijing contributed more to NH$_4$NO$_3$ than to (NH$_4$)$_2$SO$_4$ (27% vs. 23%, respectively). The contribution difference between NH$_4$NO$_3$ and (NH$_4$)$_2$SO$_4$ could be related to their precursors, formation mechanisms, and physical characteristics.
The average local contribution of SO$_2$, NO$_2$, and NH$_3$ in Beijing during the study period was 76.1%, 85.8%, and 89.3%, respectively, which was consistent with the results of Wu et al. (2011), who found that the local contribution of NO$_2$ was larger than that of SO$_2$.

The transport form of SIA exhibited different characteristics during different pollution episodes (Figure 3). The contribution from regional transport dominated (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ levels and was more than twice the local contribution during Episodes 1–3. Local emissions dominated (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ during Ep4, contributing 59% and 61%, respectively. The chemical conversion of non-Beijing precursors in Beijing was the most important aspect of regional transport during Ep4. This was related to the weather.

Figure 3. Sources of (a) ammonium sulfate [(NH$_4$)$_2$SO$_4$] and (b) ammonium nitrate (NH$_4$NO$_3$) in Beijing from 1 November to 31 December 2017. The second chart from the right is the average of the whole study period; the bars indicate the daily source, and the charts above them are the mean values in each haze episode. LOC_LOC is SIA formed by local emissions in the receptor region; LOC_TRA is SIA formed by local emissions during the transport pathway; REG_TRA is SIA chemically formed by nonlocal precursors during transport pathway to the receptor region; REG_REC is SIA chemically formed by nonlocal precursors in the receptor region; and REG_SOURCE is SIA chemically formed by nonlocal emissions in their source regions.
circulation system. Surface weather charts showed that the Beijing-Tianjin-Hebei area was controlled by high pressure during Ep4 (Figure S5). Lower wind speeds and higher relative humidity were observed during Ep4 (Figure S4). The potential source region analysis by FLEXPART revealed that Beijing was mainly affected by Beijing and surrounding cities during Ep4. Relatively obvious transport paths can be observed in other episodes (Figure S6). In summary, SIA chemically converted by non-Beijing precursors during transport pathway to Beijing and SIA chemically formed by non-Beijing precursors in Beijing were the two vital elements of regional transport during Episodes 1 to 4. The results indicate that the collaborative control of precursors among cities is imperative.

3.2. Sources of Ammonium Sulfate and Ammonium Nitrate Under Different Pollution Levels

The PM$_{2.5}$ concentration is divided into two classes: 0–75 $\mu$g/m$^3$ (clean) and higher than 75 $\mu$g/m$^3$ (polluted) according to the Technical Regulation on Ambient Air Quality Index (http://kjs.mee.gov.cn). Five cities, namely, Beijing, Shijiazhuang, Dezhou, Zhengzhou, and Taiyuan, were selected to explore the relationships between the SIA sources and pollution levels (Figure 4).

The source of SIA varied under different pollution levels. For Beijing, the average local contribution of (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ increased from 34% and 31% to 45% and 49% from clean to polluted periods, respectively. The mean contribution of REG_TRA to (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ decreased by 22% and 21%, respectively, from clean to polluted periods, whereas the mean contribution from REG_REC to (NH$_4$)$_2$SO$_4$ increased by approximately 6%, and the contribution of REG_REC to NH$_4$NO$_3$ did not change much. The variation trends in local contribution and REG_TRA from clean to polluted conditions in Shijiazhuang and Zhengzhou were similar to those in Beijing. Conditions in Dezhou and Taiyuan were quite different, with a decrease in the local contribution. Furthermore, the contributions from REG_REC and REG_TRA

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**Figure 4.** Sources of ammonium sulfate (NH$_4$)$_2$SO$_4$ and ammonium nitrate (NH$_4$NO$_3$) in Beijing, Shijiazhuang, Dezhou, Zhengzhou, and Taiyuan under clean and polluted (PM$_{2.5}$ higher than 75 $\mu$g/m$^3$) conditions. Boxes with upward-sloping stripes represent clean conditions, and boxes with downward-sloping stripes represent polluted conditions. The black horizontal lines above and below each box are the 95th and 5th percentiles, respectively, and the top, middle, and bottom horizontal lines of the boxes indicate the 75th, 50th, and 25th percentiles of the data. The blue circles on each box represent the mean values.
in Dezhou increased from clean air to high pollution conditions. The results indicate that an increase in the local contribution aggravated the pollution level in Beijing, Shijiazhuang, and Zhengzhou. However, an increase in the contribution of regional transport, especially REG_TRA and REG_REC, led to heavier pollution in Dezhou and Taiyuan; this trend can be attributed to the topography and emission intensity of the cities. The results also revealed the complexity and different sources of SIA among cities. In general, the contribution of REG_REC was influential in regional transport regardless of the pollution level and city. This result emphasizes the importance of the transport of nonlocal precursors to pollution accumulation.

As the two most important components of regional transport, the spatial distribution of contribution from REG_TRA and REG_REC to ammonium sulfate and ammonium nitrate under clean and polluted conditions needs to be analyzed. Figure 5 illustrates the difference in contribution of regional transport, REG_TRA, and REG_REC between polluted and clean conditions. The results showed that from clean to polluted conditions, the relative contribution of REG_TRA decreased in most cities, while the relative contribution of REG_REC increased in most cities. The results imply that the contribution from REG_REC (the transport of precursors) is very important under polluted conditions.

4. Conclusion

The contribution of local emissions and regional transport to haze episodes over Beijing and surrounding cities is little understood. The NAQPMS with an online tracer tagging module that tagged both precursor emission regions and SIA formation regions was used in this study. SIA at the receptor region is the sum of contributions from local emission and regional transport of nonlocal emissions. The local contribution was divided into local chemical conversion of local emission and chemical conversion during the transport of local emission. The contribution of regional transport to SIA was divided into three parts: chemical conversion of nonlocal emissions in their emission regions, chemical conversion of nonlocal emissions during transport pathway, and chemical conversion of nonlocal emissions in the receptor region. The transport forms of ammonium nitrate and ammonium sulfate in Beijing and surrounding cities were investigated.

**Figure 5.** (a and d) Average differences in the contribution of regional transport, (b and e) REG_TRA (SIA chemically converted by nonlocal precursors during transport pathway to the receptor region), and (c and f) REG_REC (SIA chemically converted by nonlocal precursors in the receptor region) to ammonium sulfate and ammonium nitrate in Beijing and surrounding cities ("2 + 26 cities") between polluted and clean conditions.
For Beijing, regional transport contributed substantially to (NH$_4$)$_2$SO$_4$ and NH$_4$NO$_3$ (58% and 56%, on average, respectively). For regional transport, SIA formed by non-Beijing emissions in Beijing and SIA formed by non-Beijing emissions during the transport pathway were the two main sources. The contribution from chemical conversion of non-Beijing emissions in their source region was low (approximately 7%). For the local contribution, the local chemical conversion of local emission in Beijing was crucial, and the chemical conversion of local emission during transport that then moved back into Beijing cannot be ignored. The transport forms of SIA exhibited different characteristics during different pollution episodes. During episodes with high relative humidity and low wind speed, local contribution was dominant and chemical conversion of non-Beijing precursors in Beijing was the largest contributor among regional transport contributors.

SIA sources varied under different pollution conditions. An increase in the local contribution and the chemical conversion of non-Beijing precursors in Beijing aggravated the pollution level in Beijing. The contribution from SIA formed by nonlocal emissions during transport decreased, while the contribution from SIA formed by nonlocal emissions in receptor region increased from clean to polluted conditions in most cities surrounding Beijing. Quantifying the contribution of local emissions and regional transport forms of nonlocal emissions may provide a reference for policymakers in the formulation of effective SIA control measures. The results suggest that the joint control of local and regional precursor emissions is essential.

Data Availability Statement

Observation data and NAQPMS simulation results used for the figures are publicly available at https://zenodo.org/10.1029/2020GL087461

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