Size-resolved aerosol water-soluble ions at a regional background station of Beijing, Tianjin, and Hebei, North China

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ABSTRACT

The characteristics of water-soluble ions in size-resolved particulate matter were investigated using ion chromatography at Shangdianzi, a regional background station of Beijing, Tianjin, and Hebei. Seasonal total concentrations of ions (Na+, Mg2+, K+, Ca2+, NH4+, Cl−, SO42− and NO3−) were 75.5 ± 52.9 μg/m3 in spring, 26.5 ± 12.3 μg/m3 in summer, 22.7 ± 20.4 μg/m3 in autumn, and 31.1 ± 23.9 μg/m3 in winter, respectively. The secondary ions (NO3−, SO42− and NH4+) mainly associated with fine particles, accounted for 84.2% in spring, 82.1% in summer, 81.5% in autumn and 76.3% in winter of all ions. Strong correlations were found between NH4+ and SO42− (r = 0.95, p < 0.01) as well as NH4+ and NO3− (r = 0.90, p < 0.01) in fine particles; while in coarse particles, correlations between Mg2+ and NO3− (r = 0.80, p < 0.01), and Ca2+ and NO3− (r = 0.85, p < 0.01) were found. The concentrations of Na+, K+, Mg2+, Ca2+, NH4+, Cl−, NO3− and SO42− were 2.02, 0.81, 0.36, 1.65, 9.58, 4.01, 18.9, and 18.4 μg/m3 in particulate matter from southeast-derived air masses, which were typically 1.58–3.37 times higher than in northwest trajectories. Thus, concentrations of water-soluble ions at this background station were heavily influenced by regional transport of serious pollution derived from biomass burning, coal combustion, industrial and vehicle exhaust emissions from Beijing, Tianjin, and Hebei.

Introduction

Over the past decades, a specific interest in water-soluble ions (WSIs) has developed because they are ubiquitous with high mass concentrations (Wang and Shooter, 2001) in particulate matter (PM) over large regions of the Earth; they affect climate and other environments (Cheng et al., 2011; Jung et al., 2009; Khoder and Hassan, 2008). WSIs, such as NH4+, Ca2+, Na+, Mg2+, K+, SO42−, NO3− and Cl−, are significant components of atmospheric particles (Satsangi et al., 2013), with concentrations dependent upon the particle source (Kawashima and Kurahashi, 2011). WSIs can scatter or absorb both incoming solar radiation and thermal radiation emitted from the Earth’s surface, directly changing the radiation balance (Bellouin et al., 2005). In regional haze episodes, SO42−, NO3−, and NH4+ in PM2.5 significantly contributed to the acidity of aerosols and to degradation of visibility (Pathak et al., 2005; Li et al., 2013a; Wang et al., 2012).

The region of Beijing, Tianjin, and Hebei (BTH) is one of the most important city agglomerations in China, yielding 10.4% of the national Gross Domestic Product (GDP) and having 8.1% of
the national population in 2014. As a result of its rapid economic development, an increase in the number of polluted and hazy days resulting from reactive gas and fine particles has occurred. Such pollution reflects the increase in emissions from motor vehicles and fossil fuel consumption in BTH. In many urban areas of BTH, more than 100 days per year have haze, causing the annual average visibility to be lower than 15 km in recent years (P. Zhao et al., 2011). Many studies on the chemical composition of PM in Beijing (He et al., 2001; Song et al., 2012; Sun et al., 2012) and Tianjin (Gu et al., 2011) have been carried out in the last few years. More detailed studies of the seasonal size distributions of WSIs could help us understand the transformation, transport, and fate of these aerosols. A few studies have focused on seasonal variations and size distributions of atmospheric particles in BTH (Guo et al., 2010; Li et al., 2012; Yang et al., 2015). However, most of these studies are concentrated on urban areas of Beijing and Tianjin.

The investigation of the variation in atmospheric composition related transport pathways in background areas assists us in determining the influence of long-range transport of pollutants on the atmospheric environment (Schichtel et al., 2006; Salvador et al., 2010; Das and Jayaraman, 2012; Tang et al., 2014). The Shangdianzi (SDZ), one of the regional Global Atmosphere Watch (GAW) stations in China, is an ideal site for characterizing the regional signature of pollutants. To date, observations at this site have targeted gaseous species, while aerosol observations have been mostly short-term, mainly involving measurement of PM mass concentrations (Zhao et al., 2009; Yao et al., 2012; Pu et al., 2015). Few measurements with seasonal and size resolution have been carried out at SDZ. Such measurements will provide better insight into aerosol formation, transport, removal and chemical reactions in the atmosphere.

In this study, we conducted a four-season monitoring program to measure WSIs in PM and to investigate the influence of long-range transport of regional emissions on ionic episodes in downwind rural atmospheres. We also provide useful information for establishing aerosol pollution control strategies for mitigating the effects of WSIs on regional environment and climate.

1. Materials and methods

1.1. Sampling

The SDZ background station (40°39′N, 117°07′E, 293.9 m a.s.l.) is located in the northern part of the North China Plain, in the Miyun County of Beijing, about 100 km and 55 km northeast of the urban area of Beijing and the Miyun Township, respectively (Fig. 1). At this mountainous site, there are only small villages with sparse populations having insignificant anthropogenic emission sources. Therefore, atmospheric pollution levels at SDZ represent background concentrations of atmospheric pollutants for economically-developed regions of BTH.

PM samples were collected using an eight-stage low pressure impactor (Andersen Series 20-800, USA) in spring (9–22 April, 2012), summer (3–16 August, 2012), autumn (13–25 October, 2012), and winter (14–27 January, 2013) from 2012 to 2013. A total of 477 PM samples and 53 blank samples were collected. Mixed cellulose ester filter substrates (Thermo-Electron Corporation, USA) were used on all stages, with a flow rate of 28.3 L/min. The 50% cut-off diameters (D50) for our stages were 9.0, 5.8, 4.7, 3.3, 2.1, 1.1, 0.65 and 0.43 μm. Sampling time was for 24 hr (from 8:00 am to 8:00 am on the following day). After sampling, the filters were individually placed in plastic bags and stored in a freezer at −20°C until subsequent analysis.

1.2. Analytical methods

1.2.1. Ion analysis

The analysis of WSIs in PM samples was carried out using two ion chromatography (IC) systems (ICS-90, Dionex, USA) at
National Research Center for Environmental Analysis and Measurement, following procedures outlined in Yang et al., 2015. The detection limits for the eight ions (SO$_4^{2-}$, NO$_3^-$, Cl$^-$, NH$_4^+$, Ca$^{2+}$, Na$^+$, Mg$^{2+}$, K$^+$), calculated the three times the standard deviation of seven replicate blank samples, were all lower than 0.05 $\mu$g/m$^3$. Quality assurance was routinely conducted using standard reference materials (125052, Merck Co., USA). Data from blank samples was subtracted from the corresponding sample data after analysis. The experimental uncertainties were ±0.04 for SO$_4^{2-}$ and NO$_3^-$, but ±0.03 for Cl$^-$ and NH$_4^+$, ±0.02 for Ca$^{2+}$, Mg$^{2+}$, Na$^+$ and K$^+$. 

1.2.2. Meteorological data
An automatic weather station was placed 10 m away from the sampler to record meteorological data simultaneously. Seasonal meteorological parameters (Fig. 2), including air temperature, wind speed, air pressure, and relative humidity, were characterized from these data. The average temperature was 15.4°C in spring, 24.4°C in summer, 9.8°C in autumn, and 6.1°C in winter. The average wind speed was 3.4 m/sec in spring, 2.1 m/sec in summer, and 2.0 m/sec in autumn, and 2.5 m/sec in winter. The average atmospheric pressure was 975.8 hPa in spring, 974.6 hPa in summer, 983.3 hPa in autumn, and 990.9 hPa in winter. The average relative humidity was 47.9% in spring, 77.7% in summer, 58.1% in autumn, and 61.8% in winter. Generally, the nearby valley topography at SDZ (Fig. 3) influenced prevailing wind directions. In autumn and winter, winds are from the east-northeast; while in spring, wind flow is mainly from the west-southwest.

2. Results and discussion

2.1. Seasonal variations of WSIs
In this study, a particle size of 2.1 $\mu$m was used to separate fine and coarse particles, reflecting the size specifications of our sampler (Yang et al., 2010). Seasonal variations of WSIs in fine and coarse particle fractions are shown in Fig. 4. The total concentrations of ions, including Na$^+$, Mg$^{2+}$, K$^+$, Ca$^{2+}$, NH$_4^+$, Cl$^-$, SO$_4^{2-}$, and NO$_3^-$, were 38.9 ± 38.0 $\mu$g/m$^3$ in all PM, with 13.0 ± 11.0 $\mu$g/m$^3$ in fine particle fractions, and 25.8 ± 18.5 $\mu$g/m$^3$ in coarse particle fractions. This finding, in accordance with other studies (Li et al., 2012; Yang et al., 2015; Li et al., 2013b), indicates that PM pollution in China is serious, especially in the target area (Zhao et al., 2013). Surprisingly for this rural site, the levels of WSIs are relatively high, in some cases comparable to urban sites in BTH (Yang et al., 2015; Li et al., 2013b). Clearly, human activities in the target area have greatly affected PM composition in the air.

The seasonal total concentrations of ions in the PM, including Na$^+$, Mg$^{2+}$, K$^+$, Ca$^{2+}$, NH$_4^+$, Cl$^-$, SO$_4^{2-}$, and NO$_3^-$, were 75.5 ± 52.9 $\mu$g/m$^3$ in spring, 26.5 ± 12.3 $\mu$g/m$^3$ in summer, 22.7 ± 20.4 $\mu$g/m$^3$ in autumn, and 31.1 ± 23.9 $\mu$g/m$^3$ in winter, respectively. The seasonal total concentrations of ions in the coarse particles were 25.6 ± 14.3 $\mu$g/m$^3$ in spring, 11.4 ± 4.2 $\mu$g/m$^3$ in summer, 7.7 ± 4.9 $\mu$g/m$^3$ in autumn, and 7.6 ± 4.5 $\mu$g/m$^3$ in winter. Specifically, ion concentrations in fine particle fractions were 49.9 ± 40.9 $\mu$g/m$^3$ in spring, 15.1 ± 9.3 $\mu$g/m$^3$ in...
summer, 15.0 ± 16.0 μg/m³ in autumn, and 23.5 ± 19.8 μg/m³ in winter. High PM mass concentrations in spring were also reported by other researchers at SDZ (Zhao et al., 2009; Zhao et al., 2013). The combination of relatively high wind speed (Fig. 2) and large tracts of barren land led to high contributions of fugitive dust in spring. In addition, new particle formation occurred most frequently in spring. However, the number concentrations for nucleation mode particles were all highest in spring (Shen et al., 2011), suggesting additional sources.

The seasonal average Ca²⁺ and Mg²⁺ concentrations both followed the order: spring > winter > summer and autumn, especially in coarse particle fractions. The springtime peaks in Ca²⁺ and Mg²⁺ were attributed to dust from roads and construction activities, suspended by relatively high wind speed in spring in BTH (Pan et al., 2013). Our weather records indicate that three days (April 10, 17, and 18) in particular were dominated by windblown dust during the spring sampling period. PM on samples from these days consisted of soil particles from local and regional origins. The average seasonal concentrations of Na⁺ in the PM were in the order: spring (2.63 ± 1.41 μg/m³) > winter (1.52 ± 0.48 μg/m³) > autumn (1.41 ± 0.44 μg/m³) > summer (1.08 ± 0.26 μg/m³). The magnitude of the seasonal Cl⁻ concentration was found to be spring (3.04 ± 1.41 μg/m³) > winter (0.66 ± 0.52 μg/m³) > autumn (0.56 ± 0.54 μg/m³) > summer (0.32 ± 0.14 μg/m³). Non-sea-salt K⁺ concentrations in the PM had seasonal averages decreasing in the order: spring (0.82 ± 0.65 μg/m³) > winter (0.66 ± 0.52 μg/m³) > autumn (0.56 ± 0.54 μg/m³) > summer (0.32 ± 0.14 μg/m³). Non-sea-salt K⁺ (nss-K⁺) is often used to identify a biomass burning source for PM in the atmosphere (Duan et al., 2004). However, for better accuracy in this study, nss-K⁺ was evaluated using the sea salt Na⁺ fraction (Becagli et al., 2005; Boreddy And Kawamura, 2015). Thus, the nss-K⁺ contributions to water-soluble potassium were 98% in spring, 91% in summer, 98% in autumn, and 99% in winter. The relatively high K⁺ concentrations at SDZ suggest that burning of wood, grass or crop residues was an important source of PM in this region.

The secondary ions [NO₃⁻, SO₄²⁻ and NH₄⁺] were the major ionic components in PM samples, comprising 84.2%, 82.1%, 81.5%, and 76.3%, of all ions in spring, summer, autumn and winter, respectively. This suggests that secondary WSIs play an important role in PM formation in BTH. The magnitude of the seasonal SO₄²⁻ concentration was found to be spring (25.9 ± 21.3 μg/m³) > winter (10.7 ± 10.3 μg/m³) > summer (8.43 ± 5.82 μg/m³) > autumn (4.05 ± 4.00 μg/m³). The magnitude of the seasonal NO₃⁻ concentration was found to be spring (26.6 ± 21.7 μg/m³) > autumn (9.94 ± 9.60 μg/m³) > winter (9.04 ± 7.91 μg/m³) > summer (6.42 ± 4.00 μg/m³). The mass ratios of NO₃⁻/SO₄²⁻ during the sampling periods were 1.03 in spring, 0.76 in summer, 2.45 in autumn, and 0.85 in winter. Typically, the ratio of NO₃⁻/SO₄²⁻ in PM can be used to analyze the contribution of mobile sources (such as vehicle exhaust) to stationary ones (such as boilers) (Yao et al., 2002). The ratio of NO₃⁻/SO₄²⁻ in the present study was higher than that was measured at SDZ in February and October 2004 (Yan et al., 2012). The total number of motor vehicles in BTH rose from 2.56 million in 2000 to 11.01 million in 2010, while coal consumption decreased over the same period (Zhao et al., 2013). These corresponding changes in emission sources support our premise that the vehicle exhaust emissions are now making an important contribution to particulate pollutants in BTH. The average seasonal NH₄⁺ concentrations were in the order: spring (11.0 ± 7.2 μg/m³) > summer (6.89 ± 2.38 μg/m³) > autumn (4.51 ± 3.22 μg/m³) > winter (3.98 ± 3.05 μg/m³). Ammonia is primarily emitted from agricultural activities, related to the application of nitrogen-based fertilizers and the volatilization of livestock waste. Agricultural fertilizer usage, comprising 489.0 million tons in Shijiazhuang, compared with 136.7 million tons in Beijing and 244.5 million tons in Tianjin in 2012 (Dao et al., 2014), was clearly one of the primary sources of NH₄⁺ at SDZ.

2.2. Acidity of PM over four seasons

Ion balance calculations are frequently used to investigate the acid–base balance of ions in aerosols; these are determined from values for cation equivalents (CE) and anion equivalents (AE), calculated using the following equation:

\[
\frac{\text{CE}}{\text{AE}} = \left( \frac{[\text{Na}^+]_{23}}{[\text{NH}_4^+]_{18}} + \frac{[\text{K}^+]_{39}}{[\text{Mg}^{2+}]_{12}} + \frac{[\text{Ca}^{2+}]_{20}}{[\text{SO}_4^{2-}]_{48} + [\text{NO}_3^-]_{62} + [\text{Cl}^-]_{35.5}} \right)
\]

Variation of CE/AE with PM size over four seasons is shown in Fig. 5. The average CE/AE ratios were in the order: summer > autumn > spring > winter. The magnitude of the seasonal CE/AE ratio for fine particles was found to be summer (1.61) > autumn (1.50) > winter (0.95) > spring (0.90). The magnitude of the seasonal CE/AE ratio for coarse particles was found to be autumn (2.54) > summer (2.21) > spring (1.46) > winter (1.20). Notably, the CE/AE for fine particles were less than 1 in both winter and spring. This is because H⁺ was not included in our calculation, giving low CE/AE ratios. This also indicates that the fine particle fraction at SDZ is highly acidic during these two seasons. Generally, the CE/AE ratios were higher than at an urban site measured during the same period (Yang et al., 2015), especially for coarse particles. This is
consistent with neutralization by alkaline dusts during regional transport.

The relationships between CE and AE are shown in Fig. 6. Good correlation between CE and AE was found for all samples, indicating that the relationship between measured cations and anions remained constant during neutralization (Lai et al., 2007). The correlation coefficient between CE and AE for fine particles ($R^2 = 0.92$) was higher than for coarse particles.

**Fig. 4** – Seasonal variations of water-soluble ions (WSIs) in fine and coarse particle fractions at SDZ. SDZ: Shangdianzi.
particles ($R^2 = 0.85$), showing that cations and anions in fine particle fractions remained in better equilibrium during neutralization than coarse particles.

### 2.3. Size distributions for WSIs over four seasons

The typical size distributions obtained for WSIs over four seasons are illustrated in Fig. 7. Mg$^{2+}$ and Ca$^{2+}$ have similar bimodal size distributions that dominated the coarse mode over four seasons. The correlation coefficient between Mg$^{2+}$ and Ca$^{2+}$ was 0.95 ($p < 0.01$), indicating they have the same source, possibly dust or mechanical abrasion products (Zhang et al., 2008; J. Zhao et al., 2011).

In this study, the size distributions of Na$^+$ and Cl$^-$ are both bimodal over all four seasons. In coarse particles, Na$^+$ was strongly correlated with Cl$^-$ ($r = 0.93$, $p < 0.01$), representing marine sources from the Bohai Sea, and reflecting long-range transportation. The size distributions of Cl$^-$ displayed seasonal variations, with much higher concentrations in winter and spring, indicating that coal combustion contributed to a major Cl$^-$ peak in the fine mode. K$^+$ had a bimodal size distribution that dominated the fine mode over all four seasons. The relatively high K$^+$ concentrations were observed in the size range from 0.43 μm to 1.1 μm. Johansson et al. (2003) reported that the particle emission from biomass burning was dominated by submicron particles (size < 1 μm), their main inorganic components being K, S, Cl and O. Thus, the presence of these ions at our study site indicates a serious problem with biomass burning in BTH.

Size distributions of secondary inorganic ions including NO$_3^-$, SO$_4^{2-}$ and NH$_4^+$ were bimodal, dominating the fine mode in all four seasons. These ions had peaks in the size ranges 0.43–1.1 μm and 4.7–5.8 μm. NH$_4^+$ is formed from its gaseous precursor (NH$_3$) through gas-phase and aqueous-phase reactions with acidic species. The mean molar ratios of NH$_4^+$ to SO$_4^{2-}$ were 1.90, 2.96, 4.53 and 1.88 in spring, summer, autumn and winter, respectively. This indicates that SO$_4^{2-}$ and NO$_3^-$ are sufficiently neutralized by NH$_4^+$ in the ammonia-rich atmosphere at SDZ. The strong correlation between NH$_4^+$ and SO$_4^{2-}$ ($r = 0.95$, $p < 0.01$) and NH$_4^+$ and NO$_3^-$ ($r = 0.90$, $p < 0.01$) in fine particle fractions suggests that these three ions primarily existed as ammonium sulfate ((NH$_4$)$_2$SO$_4$), ammonium bisulfate (NH$_4$HSO$_4$) and ammonium nitrate (NH$_4$NO$_3$). Particulate SO$_4^{2-}$ and NO$_3^-$ could also be formed by gas-phase reactions of acid precursors with NH$_3$ and could be enriched in the fine mode. This would indicate that NH$_4^+$ played an important role in determining the size distributions of SO$_4^{2-}$ and NO$_3^-$.

In coarse particles, strong correlations between Mg$^{2+}$ and NO$_3^-$ ($r = 0.80$, $p < 0.01$) as well as Ca$^{2+}$ and NO$_3^-$ ($r = 0.85$, $p < 0.01$) were found at SDZ, suggesting that Ca(NO$_3$)$_2$ or Mg(NO$_3$)$_2$ was produced by heterogeneous reactions of alkaline minerals with acidic HNO$_3$ gas (Li and Shao, 2009). There also was strong correlation between coarse NH$_4^+$ and SO$_4^{2-}$ ($r = 0.71$, $p < 0.01$), indicating that coarse ammonium sulfate was most likely produced by heterogeneous reaction of NH$_3$ and H$_2$SO$_4$ on the surfaces of coarse particles.
Fig. 7 – Size distributions for WSIs over four seasons at SDZ. WSIs: water-soluble ions; SDZ: Shangdianzi.
2.4. Imprints of regional transportation

2.4.1. Trajectory cluster analysis

Backward air mass trajectory analysis (three-day with 1 hr time resolution) was used to identify the long-range transport of air masses from different source regions (Draxler and Rolph, 2013; Rolph, 2013; Yang et al., 2015). Trajectory clusters occurring in the present study are shown in Fig. 8. These backward trajectories could be divided into two major types (northwest and southeast types), based on their direction, and the regions through which they traveled. The northwest backward trajectory type accounted for 43.6% of all trajectories, while the southeast backward trajectory type represented 56.4% of all trajectories. The total concentrations of WSIs in PM were 19.9 ± 4.3 μg/m³ and 55.7 ± 42.5 μg/m³ in northwest and southeast backward trajectory types (Fig. 9). The concentrations of Na⁺, K⁺, Ca²⁺, Mg²⁺, NH₄⁺, Cl⁻, NO₃⁻, and SO₄²⁻ were 2.02, 0.81, 0.36, 1.65, 9.58, 4.01, 18.9, and 18.4 μg/m³ in PM with southeast backward trajectories, respectively. These values were 1.58–3.37 times higher than for northwest backward trajectories. Markedly elevated levels of all ions were associated with southeast derived air masses. Influenced by monsoon and local valley topography, clean air masses often arrive at the site via sparsely populated regions of Siberia, and the Mongolian Plateau from the northeast, while polluted air from Beijing and other industrialized areas on the North China Plain comes from the southeast (Vollmer et al., 2009).

![Fig. 8 – Air mass backward trajectories at SDZ during sampling period. SDZ: Shangdianzi.](image)

![Fig. 9 – The average concentrations of WSIs at SDZ for different types of air masses. WSIs: water-soluble ions; SDZ: Shangdianzi.](image)
2.4.2. Size distributions of WSIs for different trajectory types

The difference in the size distributions for different types of air masses is shown in Fig. 10. Mg\textsuperscript{2+} and Ca\textsuperscript{2+} exhibited higher concentrations in coarse particles having a southeast than northwest backward trajectory, suggesting the source of road dust and construction dust in BTH is from long-range transport.

Fig. 10 – Size distributions of WSIs at SDZ for different types of air masses. WSIs: water-soluble ions; SDZ: Shangdianzi.
transport. Na⁺, Cl⁻ and NH₄⁺ exhibited higher concentrations in both fine and coarse particles in air masses with a southeast backward trajectory compared with a northwest backward trajectory. Na⁺ and Cl⁻ concentrations enhanced by sea-salt from Bohai Sea were from southeast derived air masses. Likewise, more intense agricultural activities in BTH, south of SDZ, favored higher NH₄⁺ concentrations in PM in southeast-derived air masses. The concentrations of NO₃⁻, SO₄²⁻ and K⁺ mainly increased in fine particles having a southeast backward trajectory, indicating serious pollution related to biomass burning, coal combustion, industrial emission and vehicle exhaust in BTH.

3. Conclusions

The concentrations of eight WSIs in size-segregated PM were measured at SDZ, a regional background site for BTH, during four seasons from 2012 to 2013. Using meteorological data, variation in size distributions, and backward trajectory paths, our field measurements were used to evaluate long-range transport of WSIs in BTH. Our major conclusions are outlined below.

(1) Secondary WSIs (NH₄⁺, SO₄²⁻, and NO₃⁻) are the dominant ions in all four seasons, especially in fine particle fractions. Particulate SO₄²⁻ and NO₃⁻ could be formed by gas-phase reactions of acid precursors with NH₃, enriched in the fine mode, indicating that NH₃ played an important role in determining the size distributions and formation of SO₄²⁻ and NO₃⁻ in the ammonia-rich atmosphere. Control of ammonia emissions is therefore very important for abatement of pollutant emissions in BTH.

(2) In coarse particles, strong correlations between Mg²⁺ and NO₃⁻, as well as Ca²⁺ and NO₃⁻ were found in this study, suggesting that Ca(NO₃)₂ or Mg(NO₃)₂ were produced by heterogeneous reactions of alkaline minerals with acidic HNO₃ gas. There also was strong correlation between coarse mode NH₄⁺ and SO₄²⁻, indicating that coarse ammonium sulfate was most likely produced by heterogeneous reaction of NH₃ and H₂SO₄ on the surfaces of coarse particles.

(3) Backward trajectory cluster analysis showed that the concentrations of WSIs at SZD were heavily influenced by regional transport of serious pollution from biomass burning, coal combustion, industrial emission and vehicle exhaust in BTH.

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