JOURNAL OF ENVIRONMENTAL SCIENCES XX (2016) XXX–XXX

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JES JOURNAL OF ENVIRONMENTAL SCIENCES

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

Yongjie Yang[1], Rui Zhou[1], Yue Yu[1], Yan Yan[1], Yan Liu[1], Yi'an Di[1], Dan Wu[2], Weiqi Zhang[3,*]

1. National Research Center for Environmental Analysis and Measurement, Beijing 100029, China. E-mail: yyj800308@163.com
2. Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, School of Environmental Science and Engineering, Nanjing University of Information Science and Technology, Nanjing 210044, China
3. State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing 100012, China

ARTICLE INFO

Article history:
Received 8 April 2016
Revised 20 June 2016
Accepted 21 July 2016
Available online xxxx

Keywords:
Regional transport
Size-resolved fractions
Seasonal variation
Particulate matter
Water-soluble ions

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^+, Mg^{2+}, K^+, Ca^{2+}, NH_4^+, Cl^-, SO_4^{2-} and NO_3^-) were 75.5 ± 52.9 μg/m^3 in spring, 26.5 ± 12.3 μg/m^3 in summer, 22.7 ± 20.4 μg/m^3 in autumn, and 31.1 ± 23.9 μg/m^3 in winter, respectively. The secondary ions (NO_3^-, SO_4^{2-} and NH_4^+), 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 NH_4^+ and SO_4^{2-} (r = 0.95, p < 0.01) as well as NH_4^+ and NO_3^- (r = 0.90, p < 0.01) in fine particles; while in coarse particles, correlations between Mg^{2+} and NO_3^- (r = 0.80, p < 0.01), and Ca^{2+} and NO_3^- (r = 0.85, p < 0.01) were found. The concentrations of Na^+, K^+, Mg^{2+}, Ca^{2+}, NH_4^+, Cl^-, NO_3^-, and SO_4^{2-} were 2.02, 0.81, 0.36, 1.65, 9.58, 4.01, 18.9, and 18.4 μg/m^3 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 NH_4^+, Ca^{2+}, Na^+, Mg^{2+}, K^+, SO_4^{2-}, NO_3^- 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, SO_4^{2-}, NO_3^-, and NH_4^+ in $PM_{2.5}$ significantly contributed to the acidity of aerosols and to degradation of visibility (Pathak et al., 2009; 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

* Corresponding author. E-mail: zhangwq@craes.org.cn (Weiqi Zhang).

http://dx.doi.org/10.1016/j.jes.2016.07.012
1001-0742/

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.

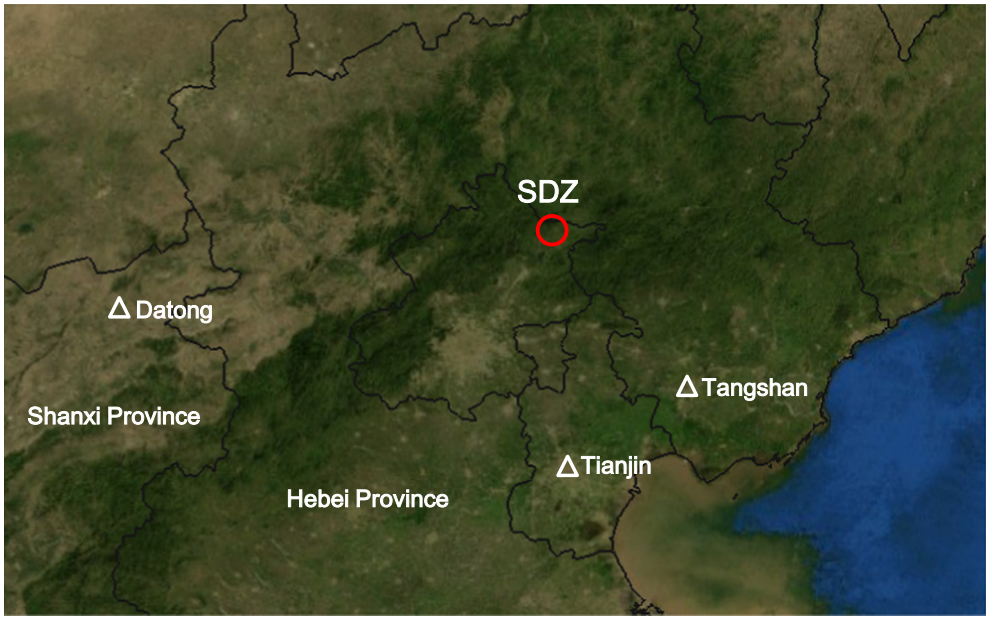

Fig. 1 – Location of sampling site. SZD: Shangdianzi.

Please cite this article as: Yang, Y., et al., Size-resolved aerosol water-soluble ions at a regional background station of Beijing, Tianjin, and Hebei, North China, J. Environ. Sci. (2016), http://dx.doi.org/10.1016/j.jes.2016.07.012