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Characteristics of the trace elements and arsenic, iodine and bromine species in snow in east-central China

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ABSTRACT

Fifty-five snow samples were collected from 11 cities in east-central China. These sampling sites cover the areas with the most snowfall in 2014, there were only two snowfalls from June 2013 to May 2014 in east-central China. Twenty-three trace elements in the filtered snow samples were measured with inductively coupled plasma-mass spectrometry (ICP-MS). Statistical analysis of the results show that the total concentrations of elements in the samples from different cities are in the order of SJZ > LZ > XA > ZZ > GD > NJ > QD > JX > WH > HZ > LA, which are closely related to the levels of AQI, $PM_{2.5}$ and PM_{10} in these cities, and their correlation coefficients are 0.93, 0.76 and 0.93. The concentration of elements in snow samples is highly correlated with air pollution and reflects the magnitude of the local atmospheric deposition. The concentrations of Fe, Al, Zn, Ba, and P are over 10.0 μg/L, the concentrations of Mn, Cu, Pb, As, Br and I are between 1.0 μg/L to 10.0 μg/L, the concentrations of V, Cr, Co, Ni, Se, Mo, Cd and Sb are less than 1.0 μg/L in snow samples in east-central China, and Rh, Pd, Pt, Hg were not detected. Iodine and bromine species in all samples and arsenic species (As(III), As(V), dimethylarsinic acid (DMA) and monomethyl arsenic (MMA)) in some samples were separated and measured successfully by HPLC-ICP-MS. The majority of arsenic in the snow samples is inorganic arsenic, and the concentration of As(III) (0.104–1.400 μg/L) is higher than that of As(V) (0.012–0.180 μg/L), while methyl arsenicals, such as DMA and MMA, were almost not detected. The concentration of I^- (Br^-) is much higher than that of IO_3^- (BrO_3^-). The mean concentration of soluble organic iodine (SOI) (1.64 μg/L) is higher than that of I^- (1.27 μg/L), however the concentration of Br^- (5.58 μg/L) is higher than that of soluble organic bromine (SOBr) (2.90 μg/L). The data presented here shows that SOI is the most abundant species and the majority of the total bromine is bromide in snow sampled at east-central China. Using Fe as the reference element to calculate the EFs, the enrichment factors of V, Cr, Co, Ni, Mn, Ba and P are between 12.3 and 82.8, and the enrichment factors of Cu, Pb, Mo, Zn, Cd, As, Sb, Br, I and Se are between 189.4 and 27667.9, indicating that these elements are contributed by artificial sources. Results of principal component analysis (PCA) on the elements showed that most of trace elements (e.g. V, Cr, Mn, Co, Ni, Cu, As, Mo, Sb, Se, Br, I, Ba and P)were from the combustion of fossil fuels, traffic and ocean sources and some other elements (e.g. Zn, Cd and Pb) were mainly originated from industrial activities.

1. Introduction

Air pollution has become one of the most important concerns in China in recent years, especially haze. Hazy weather is the most direct indicator of the air quality in China. In 2013, haze covered twenty-five provinces and more than one hundred cities, and the national average days of hazy weather reached 29.9 days, which was the highest recorded in more than 52 years. In 2015, there were 11 large-scale and continuous haze processes in China, which were concentrated at the end of the last two months, especially from November 27 to December 1, and involved North China, Shandong, Henan. Hazy days have the characteristics of strong intensity, wide range, and low visibility and have great influence. They are the most severe hazy days in 2015. Haze is intrinsically caused by a high load of fine particulate matter (PM). Trace elements are important constituents in particulate matter (PM) and of great public concern due to their toxic characters (Chen and Lippmann, 2009; Niu et al., 2015). Trace elements, especially heavy metal elements, can accumulate in human tissues through the food chain and cause damage to the human nervous system and internal organs (An et al., 2008). They also act as potentially derivational factors

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of cardiovascular diseases, reproductive impairments, and cancers (Dockery and Pope, 1996). In recent years, many studies have been conducted in megacities of east-central China region addressing the chemical characteristics and potential sources of PM and associated trace elements (Waheed et al., 2011; Song et al., 2012; Yang et al., 2013; Niu et al., 2015; Zhang et al., 2015; Tan et al., 2017; Feng et al., 2016). These studies revealed that the trace elements favoring in fine particles, such as Pb, Zn, Ni, Cu, Mn, etc., were mainly from anthropogenic sources. Air pollution in this area is greatly affected by human activities.

The snow (chemistry) reflects the chemical composition of its parent aerosol, dry fallout and surface adsorbed material. Atmospheric pollution information can be stored in snow and ice in high altitude regions. Snow cover has the chemical composition of the atmosphere during formation and subsequent accumulation (Osada et al., 2010). Therefore, snow cover is widely used as a reliable indicator to judge the type of the air pollution and is used to trace the relative contribution of different sources of atmospheric pollutants (Hiromitsu et al., 1988; ELİK, 2002). Thus, analysis of the content and distribution of trace elements in snow cover can provide information on the behaviors and sources of atmospheric pollutants. Therefore, various studies have been carried out on the chemical composition of snow around the world, especially on polar snow and mountain snow. Research carried out on trace elements in snow and ice from South America, Arctic, Greenland, Antarctic, Alps and Himalayas has allowed us to obtain important information to identify their source attributions, on the other hand, the data of the research documented the atmospheric pollution for various trace elements and also provided distinct evidence that atmospheric pollution of various trace elements exists on regional, hemispheric and global scales, as a result of long-range transport and dispersion. These data play an important role to assess with great clarity the ancient and modern atmospheric pollution for trace elements at a regional scale (Boutron et al., 1995; Van et al., 1998; Rosman et al., 2000; Barbante et al., 2003; Schwikowski et al., 2004; Hong et al., 2004; Hegg et al., 2009; Liu et al., 2011; Dong et al., 2015; Wang et al., 2015). However, there were a few studies in the experiments of measuring the trace elements in snow from cities (An et al., 2008; Hu et al., 2012; ELİK, 2002; Kazuhiko et al., 2000), and there is almost no information available from the published literature surveying chemical species in snow from east-central China region.

As one of the most industrialized and densely populated areas, the atmospheric quality in the east-central China region is strongly influenced by human activities, such as industrial production. From June 2013 to May 2014, snow samples were collected in eleven cities in the east-central China region. During this period, there were only two snowfalls in this region. Therefore, 55 snow samples were collected from 11 cities in east-central China, and 23 trace elements (e.g., Al, Mn, Fe, Zn, Ba, P, Cu, Pb, Br, V, Cr, Co, Ni, As, Se, Mo, Cd, Sb, I, Pt, Pd, Rh and Hg) and the arsenic, iodine and bromine species in the filtered snow samples were measured. The main purpose of the present work is to expand the knowledge of the spatial characterizations of trace elements in east-central China to evaluate trace element pollution related to anthropogenic sources, and to explore relationships between the concentration of trace element and Air Quality Index (AQI) that are less commonly considered, and to identify the forms of atmospheric arsenic, bromine and iodine, and finally to evaluate that the snow chemistry in east-central China region atmosphere probably reflects the magnitude of the local atmospheric deposition and the influence of atmospheric pollution of trace elements.

2. Methodology

2.1. Snowfall events and sample site

There were only two snowfalls in east-central China from 2013 to winter of 2014. The first snowfall, with a depth of 5–16 cm, occurred on February 4-5, 2014, covering northwest of Hubei, northern Jiangsu and Anhui, southern Shandong, most of Henan, southeast of Shanxi, Shanxi and other places. Among these areas, there were heavy snowfalls, with a depth of 10–16 cm, in northern Jiangsu and Anhui, southern Shandong and eastern Henan. The second snowfall, with a depth of 3–10 cm, occurred on February 17-18, 2014, covering southern Shanxi, eastern Hubei, northeast of Hunan, south-central Anhui, northwest of Zhejiang, north-central Jiangxi, eastern Guizhou and northeast of Yunnan. Among these areas, there were heavy snowfalls, with a depth of 10–16 cm, in southeastern Hubei, southern Anhui, northwestern Zhejiang, and northwestern and northeastern Jiangxi.

This study selected 11 representative cities (almost all cities, except for Lanzhou and Xi'an, are in east-central China, including Zhengzhou, Shijiazhuang, Jiaxiang, Qingdao, Hangzhou, Wuhan, Nanjing, Lin'an, Guangde) to sample. These sample sites cover areas that had snowfall in east-central China during the winter of 2014. These 11 cities include five cities in the areas of the first snowfall, Zhengzhou (ZZ), Shijiazhuang (SJZ), Qingdao (QD), Jiaxiang (JX) and Lanzhou (LZ) (see the left side of Fig. 1), and six cities in the areas of the second snowfall, Nanjing (NJ), Wuhan (WH), Hangzhou (HZ), Guangde (GD), Lin'an (LA) and Xi'an (XA) (see the right side of Fig. 1).

2.2. Sample collection

Fifty-five snow samples were collected from these 11 cities, 5 areas for each city, including the central business district, residential areas, cultural areas, industrial areas and suburbs (Junge and Scheich, 1971). Sampling points were set in each area. All the samples from one area were mixed to obtain the final sample for each area, and thus, there were 5 snow samples for each city. The sampling points were set in open spaces, far away from pollution sources, such as traffic arteries and chimneys, and from tall buildings and trees. The snow at the sampling sites should be as deep as possible. The surface and the bottom of the snow were removed, and only the middle section was collected in a new 250 mL low-density polyethylene (LDPE) bottle. Each bottle was washed by the middle-section snow three times before use. When the snow was fully compacted, the LDPE bottles were put into shading bags and stored in incubators. The incubators were kept outdoors in the cold environment.

2.3. Sample analysis

After dissolution, the snow water samples were first filtered through a 0.45 μm cellulose acetate filter to remove suspended matter and were then transferred to ultraclean 50 mL bottles. The sample was divided into two parts. In one part, ultrapure HNO_3 was added, and the sample was acidified to pH $<$ 2.0 to inhibit microbial activities and reduce the adsorption of heavy metals. The sample was stored in the dark under 4 °C and was used to analyze the metal elements. The other part was kept at under 4 °C in the refrigerator and was used to analyze the nonmetal elements, i.e., the arsenic, bromine and iodine species.

The trace elements, total soluble arsenic (TSAs), total soluble iodine (TSI) and total soluble bromine (TSBr) were analyzed by inductively coupled plasma-mass spectrometry (PerkinElmer, NexION 300D, America), arsenic species (As(III), As(V), DMA and MMA) were analyzed by HPLC-ICP-MS (PerkinElmer, Flexar and NexION 300D, America), where a PRP-X100 column (150 mm × 4.6 mm, 10 μm) (Hamilton) was used as the anion exchange column, the mobile phase was 10 mM/L $(NH_4)_2HPO_4$, and the flow rate was 1.0 mL/min. Analysis of the trace elements and arsenic species was completed within 2 months. The iodine and bromine species (I^{-1}, IO_3^-, Br^- and BrO_3^-) were analyzed by HPLC-ICP-MS (PerkinElmer, Flexar and NexION 300D, America), where a Dionex IonPac AS14 (4 mm × 250 mm) + AG14 (4 mm × 50 mm) column was chosen as the chromatographic column, the mobile phase was 50 mM/L NH_4NO_3 gradient elution, and the flow rate was 1.2 mL/min. The analysis of iodine and bromine

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