



Water-soluble ions in atmospheric aerosols measured in five sites in the Yangtze River Delta, China: Size-fractionated, seasonal variations and sources



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HIGHLIGHTS

- The seasonal variations and size distributions of WSIs were discussed.
- The impacts of monsoon on the seasonal distributions of WSIs.
- The geographic locations and emission sources caused discrepancies of WSIs.
- The seasonal changes in WSIs sources were discussed.

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ABSTRACT

In order to investigate the regional variations of water-soluble ions (WSIs), size-resolved measurement of aerosol particles and WSIs was conducted by using Anderson Sampler and Ion Chromatography at five sites (Nanjing, Suzhou, Lin'an, Hangzhou and Ningbo) in the Yangtze River Delta (YRD) region, China in the Autumn of 2012 and Winter, Spring and Summer of 2013. WSIs exhibited obvious seasonal variations due to the monsoon conversion, with the highest level in winter and lowest level in summer. The aerosol mass concentrations and WSIs in different size segments varied with four seasons. The dominant ions concentrations in $PM_{2.1}$ ranked in the order of $SO_4^{2-} > NO_3^- > NH_4^+ > Cl^- > K^+ > Ca^{2+}$, and the dominant ions concentrations in $PM_{2.1-10}$ ranked in the order of $Ca^{2+} > NO_3^- > SO_4^{2-} > Cl^- > NH_4^+ > Na^+$. The size spectra of mass and WSIs concentration peaked mostly at 0.43–0.65 μm in four seasons. The concentration discrepancies of WSIs in different cities were caused by the geographic locations and emission source. It's belonged to ammonium-rich distribution in $PM_{2.1}$ and ammonium-poor distribution in $PM_{2.1-10}$ in the YRD region. The impact of temperature on mass concentrations of NO_3^- and NH_4^+ in $PM_{2.1}$ were stronger than those in $PM_{2.1-10}$. PCA analysis shows that the sources of WSIs dominant by anthropogenic sources, soil particles or falling dust, sea salt and burning process.

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1. Introduction

Aerosol particles play an important role in global climate change by their direct and indirect radiation effects (Bellouin et al., 2005;

Haywood and Boucher, 2000; Ramanathan and Carmichael, 2008), and can endanger human health and impair visibility as well (Andreae et al., 1998; Deng et al., 2011; Kang et al., 2013; Tie and Cao, 2009; Tie et al., 2009; Zhou et al., 2010). The chemical compositions of aerosol particles determine their hygroscopicity, solubility and the particle light extinction efficiency (Henning et al., 2003; Lin et al., 2013). As the main chemical component in aerosols, WSIs compose up to 60–70% of the total mass of suspended

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particulate matter (Ali-Mohamed, 1991). Therefore, it is scientifically important to investigate the WSI characteristics for full understanding aerosols' physical and chemical properties, sources, behaviors and formation mechanisms.

With the rapid development of industrialization and urbanization, the contributions of vehicle exhaust and industry emission to aerosol particles grow increasingly (Safai et al., 2010). Related studies found that WSIs constituted 30%–80% of aerosol mass in urban areas (Contini et al., 2014; Shen et al., 2009; Shi et al., 2014; Tan et al., 2009). The ratio of WSIs in PM_{1.0} was 64.4% in Beijing (Sun et al., 2010) and 79.2% in Shanghai (Huang et al., 2012), and was 38.9% in Xi'an (Zhang et al., 2011) and 68.8% in Guangzhou (Lai et al., 2007) for that in PM_{2.5}. The proportions of WSIs were high even in clean background areas, which was 44.0% in PM_{2.0} mass in European background site (K-puszta) (Ocskay et al., 2006), and 55.1% in PM_{2.1} mass at Mt. Huang (Li et al., 2014). Zhang et al. (2011) observed that the ratios of WSI were higher in fine particles, with major ions of SO₄²⁻, NO₃⁻ and NH₄⁺. Therefore, the concentrations, relationships and behaviors of ions are regarded as valuable indicators for estimating pollution (Tian et al., 2013; Yao et al., 2002).

At present, most of WSI studies have focused on the concentrations, species, temporal variations, source identification and size distributions. Li et al. (2012) and Miyazaki et al. (2012) discovered that non-sea salt sulphate and ammonium are found to be predominantly in the fine mode, while sea spray SO₄²⁻, Cl⁻, Na⁺, Mg²⁺ and Ca²⁺ are more abundant in the coarse fraction. It's found that the mass size distributions can provide evidence for examining the formation pathways of WSIs (Guo et al., 2010; Zhang et al., 2008). Li et al. (2013) considered that NH₄⁺ played an important role in the size distributions and the formations of SO₄²⁻ and NO₃⁻. The WSIs were mainly from anthropogenic sources in Xi'an (Zhang et al., 2011), and had obvious seasonal variations (Cao et al., 2009; Cheng et al., 2012; Hu et al., 2002; Wang et al., 2001). The vertical distribution of WSIs was discussed as well by Li et al. (2014) and Tian et al. (2013). The distribution characters of WSIs under different pollution conditions of haze, dust, and biomass burning had significantly distinctions (Cheng et al., 2011; Park et al., 2004, 2013; Yamasoe et al., 2000).

As one of the fastest growing economic regions in East Asia, aerosol particles in the YRD has become one of the most harmful pollutants (Brauer et al., 2012; Zhou et al., 2010). Large numbers of researches, concerning the mass concentration, chemical component, sources of aerosols, their impacts on visibility and haze mechanism, have been carried out in the YRD region (Cao et al., 2009; Cheng et al., 2014; Deng et al., 2011; Huang et al., 2011; Kang et al., 2013; Tie et al., 2009). However, these studies are mostly limited in one city of the YRD region, such as Nanjing (Deng et al., 2014; Ding et al., 2013; Kang et al., 2013), Hangzhou (Cao et al., 2009) and Shanghai (Du et al., 2011; Wang et al., 2006; Xiu et al., 2004). Although air pollution tends to be regional (Wang et al., 2012), the investigation concerning the size distributions of WSIs in aerosols in the YRD region were rare. In this study, the size distributions of WSIs in five cities (Nanjing, Suzhou, Lin'an, Hangzhou and Ningbo) were obtained by using Anderson Sampler and Ion Chromatography in October 2012, January, April, and July 2013. This paper presents concentration levels and the seasonal variations of these measured species. We analyses the geographic locations and monsoon caused discrepancies of WSIs. In addition, seasonal differences in major sources of WSIs were discussed by the PCA model.

2. Instruments and experiments

2.1. Observation stations and experiment descriptions

The five stations were located in Nanjing, Suzhou, Hangzhou,

Lin'an, and Ningbo, which are the major cities in the YRD region (Fig. 1). One regional background station was located in Lin'an (Table 1). The information of these stations and their surroundings were listed in Fig. 1 and Table 1. The measurements were carried out on Oct. 17–24, 2012, Jan. 17–24, April 9–16 and July 8–14, 2013 in five cities. While the observation was conducted on April 10–19, 2013 in spring and July 8–18, 2013 in summer of Nanjing, April 9–18, 2013 in spring of Suzhou, and July 8–12, 2013 in summer of Ningbo. Besides, some of the observation data were invalid on Oct. 17–24, 2012 in Ningbo and Oct. 21–24, 2012 in Hangzhou due to the man-made mistakes. After excluding the invalid data, 149 samples were obtained during the observation period, there were 44, 37, 28 and 40 samples in spring, summer, autumn and winter, respectively. Every sample contained data about WSIs of 9 size segments in PM₁₀. Main meteorological factors' changes were recorded by automatic weather station. The size-segregated aerosol particles were continuously collected for 23 h at each site.

2.2. Instrumentation

Aerosol particle observation was carried out by a 9-stage Anderson-type aerosol sampler (Anderson 2000 Inc., USA) with size ranges of <0.43, 0.43–0.65, 0.65–1.1, 1.1–2.1, 2.1–3.3, 3.3–4.7, 4.7–5.8, 5.8–9.0 and 9.0–10.0 μm for Water-soluble ionic components. As the flow rate required by the Anderson-type aerosol sampler is 28.3 L/min. The sampler was operated with an 80 mm Teflon filter (Whatman, Clifton, England) for water-soluble ionic components, and the membranes were weighted by Mettler Toledo MX-5 microbalance after constant temperature (25 °C) and humidity (50%) treatment for 48 h before and after sampling, the microbalance was calibrated using standard weight. The weight difference before and after sampling is particle weight.

Water-soluble ions were measured by 850 professional Ion Chromatography (IC) (Metrohm, Switzerland), NH₄⁺, Ca²⁺, Mg²⁺, Na⁺, K⁺, Cl⁻, NO₃⁻, SO₄²⁻, F⁻ and NO₂⁻ were analyzed. Chromatography includes column oven, conductivity detector, with an 858 auto-injector and MagIC Net chromatography workstation (Metrohm, Switzerland); Column; Metrosep C 4150/4.0 separation column and Metrosep A Supp 5150/4.0 separation column; eluent: 3.2 mmol/L Na₂CO₃+1.0 mmol/L NaHCO₃ (anions), 1.7 mmol/L HNO₃ + 0.7 mmol/L pyridine carboxylic acid (cations); column temperature: 30 °C; flow-rate: 1.0 mL/min; inject volume: 20 μL. The detection limits for Na⁺, NH₄⁺, K⁺, Mg²⁺, Ca²⁺, F⁻, Cl⁻, NO₂⁻, NO₃⁻ and SO₄²⁻ were 0.001, 0.005, 0.001, 0.002, 0.001, 0.04, 0.01, 0.04, 0.006, 0.05 mg/L, respectively. Detailed information can be referred to Li et al. (2014). Half of each Teflon filter was cut and placed into a 15 mL PET vial, the vial was then ultrasonically extracted for 1 h and mechanically oscillated for 1 h with 10 mL deionized water (18.2 MU). PTFE filters and filter chips were used to remove the insoluble particles before samples analyzed by IC.

3. Results and discussion

3.1. Ions balance of seasonal and size-fractionated

Fig. 2a shows the correlation coefficients of Anion and Cation equivalent were between 0.72 and 0.94. Ratios of Anion/Cation were in the range of 0.65–1.15, which was similar to the results in Guangzhou (Hu et al., 2008) and Thessaloniki (Voutsas et al., 2014). The presence of water-soluble organic anions (such as organic acidic ions) and carbonates may be of importance for the charge balance (Arsene et al., 2011; Contini et al., 2014; Kerminen et al., 2001). However, we didn't have data about organic components and carbonates, thus we didn't consider the impacts of organic and carbonates ions on WSIs. Ratios of Anion/Cation varied greatly with

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