



Seasonal ion characteristics of fine and coarse particles from an urban residential area in a typical industrial city



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ABSTRACT

This study investigated seasonal ion variation and correlation characteristics in size-fractionated particles collected for four seasons using an eight-stage cascade impactor sampler from an urban residential area in the largest industrial city, Korea. The identified ionic species accounted for 47.4, 52.5, 35.8, and 44.5% of $PM_{1.1}$, $PM_{2.1}$, $PM_{10-2.1}$, and PM_{10} ($PM_{2.1} + PM_{10-2.1}$), respectively. Sulfate (SO_4^{2-}) was the most abundant species and had the highest relative concentrations of both fine and coarse particles in summer. However, nitrate (NO_3^-) and ammonium (NH_4^+) showed the highest fractions in fine particle mass concentration in winter. NH_4^+ showed the highest mass fraction in coarse particles in spring. The highest ratio of NO_3^-/SO_4^{2-} in fine particle in winter was due to the decreased sulfate and increased nitrate formation by low winter temperature. Most ions, such as SO_4^{2-} , NH_4^+ , K^+ , Cl^- and NO_3^- , accumulate in fine particles ($PM_{2.1}$); while the concentrations of Na^+ , Ca^{2+} and Mg^{2+} were greater in the coarse particles ($PM_{10-2.1}$). In the cluster analyses of the ion components, road and soil dust, secondary aerosols and sea salt were shown to be possible major airborne PM sources in residential areas of the industrial city.

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1. Introduction

In the last few decades, atmospheric aerosols have received much attention due to their impacts on human health, visibility and global climate change (Cheng et al., 2011; Jung et al., 2009; Chu et al., 2008; Gilli et al., 2007; Neuberger et al., 2004; Sheffield et al., 2011). Atmospheric aerosols are analytically complex matrices that contain water-soluble inorganic species (ionic species), organic and elemental carbon, and metals and metalloids (Onat et al., 2012; Kim et al., 2002, 2004). According to the relevant studies on aerosols, airborne ionic species are responsible for a large proportion of the visibility loss and cloud formation in the air (Penner et al., 2011; Cheng et al., 2011; Jung et al., 2009; Lee and Sequerira, 2002). They can also play a major role in the acidification of precipitation and may affect climate change (Khoder and Hassan, 2008). Ionic species, such as sulfate, nitrate and ammonium, are the most common components of secondary airborne particles and can comprise a large fraction of the particulate matter (PM) in the

atmosphere (Wang et al., 2005, 2006; Sun et al., 2006; Hueglin et al., 2005; Yao et al., 2002).

Secondary particles are formed in the atmosphere through a combination of chemical and physical reactions among primary pollutants such as the direct emissions of sulfur dioxides (SO_2), nitrogen oxides (NO_x), and ammonia (NH_3) gases. It is difficult to predict ambient concentrations of these secondary air pollutants because they are also affected by long-range transport, and their formation is dependent on unstable factors such as ambient temperature and humidity. Acidic aerosols produced from acidic gaseous pollutants, such as SO_x and NO_x , have drawn much attention of the investigators and technologists who are studying the atmospheric environment and public health (Verma et al., 2010; Gilli et al., 2007). Human exposure to acidic aerosols in ambient air can cause respiratory irritation, pulmonary disease and asthma (Fang et al., 2005). In particular, sulfate, one of the major acidic aerosol components, plays the most important role in visibility restriction among the soluble ions (Cheng et al., 2011; Jung et al., 2009; Lin et al., 2011) because sulfate usually has a high concentration in the particle size range of 0.4–1.0 μm which is largely responsible for the reduction in atmospheric visibility.

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There have been many studies which investigated ionic components in fine particles in Korea (Park and Kim, 2004; Hong et al., 2008; Han et al., 2008; Lee et al., 2008). Park and Kim (2004) reported that different sampling sites (cities) had different atmospheric behavior and formation mechanism of aerosols. Hong et al. (2008) reported that different photochemical conditions and meteorology can affect ionic compositions differently. Han et al. (2008) published that concentrations of airborne ionic species with different airborne trajectories, measured in different winter seasons, were statistically different from each other. However, most of the studies focused on limited ionic species, mainly including SO_4^{2-} , NO_3^- , and NH_4^+ , from general urban areas during short study periods. The general public who live near industrial areas or in even residential areas in industrial cities have greatly concerned on environmental impacts or exposure to anthropogenic aerosols or ionic species. However, only limited studies are available for airborne ionic component information in industrial cities (Lee and Lee, 2005; Han et al., 2008) in Korea. Fine and coarse particles vary in their mechanisms of generation, seasonal concentration, visibility reduction, deposition into the lung, human health effects, etc. (Parmar et al., 2001; Kim et al., 2006). Thus, the objective of this study is to investigate the concentration, distribution, seasonal variations and correlation of ionic species in the fine and coarse airborne particles (Almeida et al., 2008; Kim et al., 2006; Hueglin et al., 2005; Manoli et al., 2002) in an urban residential area of a typical industrial city, Korea.

2. Material and methods

2.1. Area description

Ulsan is known as the industrial heart of Korea and is the hometown of the largest industrial corporations in Korea such as Hyundai (automobile and shipbuilding) and SK (petrochemical). Ulsan is also home to other large industrial complexes (ICs) including petrochemical IC, non-ferrous metallic IC, automobiles and parts manufacturing plants, and shipbuilding companies. The sampling site for ambient PM measurement is located on the roof of the Mugeo-dong office (civic center) building (7.5 m height) in an urban residential area of Ulsan, Korea. The sampling site is 100–700 m from 15 to 20 story apartment complexes (42–58 m height) and 120 m from Ulsan highway. In addition, there is a busy traffic rotary connecting five four-lane highways approximately 180 m from the sampling site. Even though the sampling site is located 15–17 km from the ICs (Fig. 1), a substantial quantity of industrial emissions can reach the sampling area particularly under downwind conditions of southeastern winds which pass through heavily industrialized zones (Lee and Hieu, 2011; Ny and Lee, 2011).

2.2. Sample collection and analysis

Daily PM measurements were carried out using an ambient cascade impactor (Model 20-800, Tisch Environmental, Inc.), which can classify the nine particle size intervals (<0.4, 0.4–0.7, 0.7–1.1, 1.1–2.1, 2.1–3.3, 3.3–4.7, 4.7–5.8, 5.8–9.0 and 9.0–10 μm), operated at a constant flow rate of 28.3 L/min. 30 daily (based on 24 h) sets of airborne PM samples (270 filters including backup filters) for all of the four seasons were collected

from the roof of the civic center, located in a representative urban residential area, on 81-mm glass fiber filters using the cascade impactor (Khoder and Hassan, 2008; Pateraki et al., 2008). One daily (weekday) PM sampling was conducted almost every week (excluding a few weeks with frequent rainfall events or heavy rainy days in spring and monsoon periods) from April 2008 to January 2009. To minimize environmental impacts of ionic components by high humidity or moisture on rainy days, PM sampling was avoided on rainy days above 0.5 mm or right after rainfall events. Thus 3–4 daily PM sampling was conducted each month during the study period. After sampling, the filters were kept in Petri dishes and transported to the laboratory. All sample and blank filters were kept in a desiccator in a conditioned room with a relative humidity (RH) of $45 \pm 5\%$ and temperature of $20 \pm 2^\circ\text{C}$ for 48 h before weight measurement using a balance. PM mass concentrations were determined using gravimetric analysis with an electronic microbalance with a sensitivity of 0.01 mg. After balancing, the filters were stored in Petri dishes in a refrigerator until analysis. The effects of heating and cooling sources such as solar radiation and electrical apparatuses were minimized. The ions were extracted from the filters by placing them into 15 mL ultra-pure water and then into an ultra sonification bath for 30 min (Talebi and Abedi, 2005). This study focused on comparison of ionic components in fine ($\text{PM}_{1.1}$ or $\text{PM}_{2.1}$) and coarse ($\text{PM}_{10-2.1}$) particles, which usually have quite different sources and atmospheric behaviors, for easy comparison with the results obtained from other reported studies (Kim et al., 2005). The samples were then kept in a refrigerator at 4°C until they were analyzed. Eight major ionic species (Na^+ , K^+ , Mg^{2+} , Ca^{2+} , Cl^- , NO_3^- and SO_4^{2-}) were measured using ion chromatography (DIONEX DX-80). A CS12A and an AS14 column (4×250 mm) were used for determinations of cations and anions, respectively. The method detection limit (MDL) and relative standard deviation (RSD), provided by the ion chromatography manufacturer (Thomas et al., 2002), of cations were 2, 1, 3, 1 and 1 ng mL^{-1} and 0.97, 0.83, 0.99, 0.93 and 1.22% for Na^+ , NH_4^+ , K^+ , Mg^{2+} and Ca^{2+} , respectively. The MDL and RSD of anions were 13, 9 and 44 ng mL^{-1} and 0.60, 0.59, and 1.76% for Cl^- , NO_3^- and SO_4^{2-} , respectively.

Glass fiber filters can contain some levels of Na, K and Cl (Espinosa et al., 2001). Even though the blank values were subtracted from the values analyzed from PM samples, the concentrations of the ionic species might not be accurately determined. The identified levels of Na^+ , K^+ and Cl^- from 3 repeated measurement of 3 blank glass fiber filters were 5.8 ± 0.4 ng mL^{-1} (RSD: 6.3%), 0.9 ± 0.0 ng mL^{-1} (RSD: 5.1%), and 1.4 ± 0.0 ng mL^{-1} (RSD: 1.8%), respectively. Thus there might be some errors to estimate exact concentrations of Na^+ , K^+ and Cl^- in fine and coarse particle samples. Typical average concentrations of the ionic species in airborne fine particle ($\text{PM}_{2.1}$) and coarse particle ($\text{PM}_{10-2.1}$) samples during the study periods in Ulsan were around 2445 and 3464 ng mL^{-1} for Na^+ , 288 and 408 ng mL^{-1} for K^+ , 1630 and 611 ng mL^{-1} for Cl^- , respectively. The levels of the ionic species identified from the blank filters were less than 0.3 wt.% of those from PM samples collected for this study. Thus the relative errors accompanied by use of glass fiber filters would not be important for evaluating the results for this study. Field blanks (10% of PM samples) were also used to correct the concentrations of ionic species.

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