

## Investigation type of distribution of aerosol and its impact on air pollution

Mahdi Ojaghi<sup>1</sup>, Ziba Beheshti<sup>2</sup>, Karim Isazadeh<sup>3</sup>, Mohammad hossein Mohammadi ashnani<sup>4</sup>

<sup>1</sup>. MSc in Planning, management & training of Environment, Faculty of Environment, Tehran University.

<sup>2</sup>. MSc in Assessment and Land Use Planning, Faculty of Environment and Energy, Islamic Azad University, Tehran

<sup>3</sup>. MSc in Agero Ecology, Islamic Azad University, Tehran

<sup>4</sup>. MSc in Planning, management & training of Environment, Tehran University

**Abstract:** Air pollution started to become a problem for human beings with the industrial revolution, but nowadays, with the introduction of laws against emissions. The atmosphere is a complex natural gaseous system that is essential to support life on planet Earth. The size distributions of K and Mg are also discussed in relation to particular sources. A survey of atmospheric aerosols in a suburban area near Tokyo, Japan was conducted using an Andersen sampler. Significant amounts of Na<sup>+</sup> and Cl<sup>-</sup> collected were considered to be derived from sea salt. The difference between the Na<sup>+</sup>/Cl<sup>-</sup> ratio in the area and that in sea salt indicated a considerable loss of Cl<sup>-</sup>. This is assumed to be caused by the formation of NaNO<sub>3</sub>, which is one of the main nitrate species present. Most of the sulfate in the sample was found to be (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in the form of fine particles, which is different from the sulfate derived from sea salt and soil.

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### 1. Introduction

Air pollution started to become a problem for human beings with the industrial revolution. Air pollution has been recognized as a threat to human health as well as to the Earth's ecosystems. Air pollutants emitted into the atmosphere from various sources can be classified as either gaseous or particulate matter (PM). These types of pollutants are closely related to each other through gas-to-particle conversion, called condensation, and particle-to-gas conversion, called evaporation. An aerosol can be defined as a system of solid or liquid particles suspended in an ambient environment. Aerosols, which can be manmade or occur naturally, vary in size and composition. From the viewpoint of aerosol science, a particle is described as primary when it is emitted directly from a source into the atmosphere.

On the other hand, a particle that is initially emitted as gaseous matter and then converted to PM through ambient chemical and physical processes is described as a secondary particle. Understanding the behaviors of PM and the complex influences it has on human beings, animals, and plants requires analysis of size distribution, mass concentration, and chemical composition. Size distribution and chemical composition analysis detail basic characteristics of atmospheric PM. These characteristics are closely related to the effects PM has on human health. This study investigates the size distribution of anion species (Cl) and cation species (4NO<sub>2</sub> 24SO<sub>2</sub> NH<sub>3</sub>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, and K<sup>+</sup>) of PM in the atmosphere.

### 2. Material and Methods

After ambient air was collected, the backup filter and the quartz filters from the eight stages were removed from the Andersen sampler and soaked in a controlled environment at 35°C and 50% relative humidity for 24 h. After soaking, the PM mass concentrations of each stage were determined gravimetrically using an electronic microbalance in a temperature and humidity controlled room. An experiment was conducted to evaluate ion composition.

The quartz filter from each stage was cut into a piece of a quarter (9 pieces in total). Aerosols in ambient air were collected at the Center for Environmental Science at Saitama (CESS), Kazo, and Saitama Prefecture, Japan. CESS is located in a suburban area surrounded by paddy fields, vegetable gardens, and roads, as illustrated by it.

An Andersen sampler was placed on the roof of the CESS air-monitoring station with an intake located approximately 4 m above ground level. Ambient aerosols were collected continuously for one week from October 23 to 30, 2009 using a 47 mm quartz filter and an eight-stage Andersen impactor (Model AN- 200) with a backup filter at an air flow rate of 28.3 L min<sup>-1</sup>.

This system was used to obtain information on size-segregated particles and to classify particles into nine size ranges (0.08 to 30 μm). The Andersen sampler can selectively trap different sized particles according to their momentum. Cations and anions were extracted from

the pieces of quartz filter using ultrapure water in an ultrasonic bath for 20 min. The cations and anions ( $\text{Cl}^-$ ) were analyzed by ion chromatography (Dionex IC-20) using the following materials and conditions. Anion chromatography was performed with an AS12A column using 2.7 mM  $\text{Na}_2\text{CO}_3$  and 0.3 mM  $\text{NaHCO}_3$  as eluents at 35°C, a flow rate of 1.2 mL/min, and an injection volume of 25  $\mu\text{L}$ . Cation chromatography was performed with a CS12A column using 20 mM methane sulfuric acid as the eluent at 35°C, a flow rate of 1.0 mL/min, and an injection volume of 50  $\mu\text{L}$ . The detection limits in millimolar were 0.003 for  $\text{Na}^+$ , 0.004 for  $\text{K}^+$ , 0.004 for  $\text{Mg}^{2+}$ , 0.015 for  $\text{Ca}^{2+}$ , 0.021 for  $\text{Cl}^-$ , 0.028 for  $\text{SO}_4^{2-}$ , and 0.019. The calibration curves of the anions and cations were determined using standard solutions. The blank filters were also extracted and analyzed for operation blanks.

### 3. Results and Discussion

The distribution is bimodal; the fine particles peak at approximately 1  $\mu\text{m}$  and the coarse particles peak at approximately 4  $\mu\text{m}$ . The mass concentrations of the fine and coarse particles were 20.7 and 18.8  $\mu\text{g}/\text{m}^3$ , respectively. The total concentration was 39.5  $\mu\text{g}/\text{m}^3$ .

Typically, aerosols in the coarse particle range ( $>2\mu\text{m}$ ) originate from natural sources, while those in the fine particle range ( $<2\mu\text{m}$ ) are derived from anthropogenic sources [1-5]. The size distribution shown in it was influenced by both natural and anthropogenic sources in the study area.  $\text{Cl}^-$  was distributed bimodal with one peak in the fine particle range and another in the coarse particle range. However,  $\text{Na}^+$  appears to be distributed in a monomial form with a peak in the coarse particle range. Sea salt contains  $\text{Na}^+$  and  $\text{Cl}^-$  at a mole ratio of approximately 0.89 because it is composed of species other than  $\text{Na}^+$  and  $\text{Cl}^-$ . Although Saitama City is located inland, sea spray was a fairly large contributor of coarse particles. It is commonly known that coarse  $\text{Na}^+$  and  $\text{Cl}^-$  particles originate from sea salt. However, the mole ratio of  $\text{Na}^+$  to  $\text{Cl}^-$  in the sample does not coincide with that of sea salt, showing a lower than expected level of  $\text{Cl}^-$ . The level of  $\text{Cl}^-$  observed in this study gave a mole ratio of 3.60. This is thought to be due to sea salt particles reacting with ambient  $\text{HNO}_3$  and/or  $\text{NO}_2$  to form  $\text{NaNO}_3$  and releasing  $\text{HCl}$  ( $\text{NaCl} + \text{HNO}_3 = \text{NaNO}_3 + \text{HCl}$ ). Accordingly, in the coarse range, it is necessary to examine the mole ratio of  $\text{Na}^+ / (\text{Cl}^- + \text{NO}_3^-)$  rather than that of  $\text{Na}^+ / \text{Cl}^-$ . The recalculated value was 0.72, which is relatively close to the expected value of 0.89. This indicates that a moderately rich  $\text{Cl}^-$  condition was observed. However, this appears consistent with experimental and expected values.

Moreover, a small peak of  $\text{Cl}^-$  was observed in the fine particle range. This peak is assumed to be derived from  $\text{HCl}$  emitted from the incineration of garbage that included polychlorinated hydrocarbons. Prior to the implementation and enforcement of strict controls,  $\text{HCl}$  emissions in Japan were significant. This study confirms that  $\text{HCl}$  remains at a noticeable level. The counter ion of  $\text{Cl}^-$  is considered to be after the neutralization of  $\text{HCl}$  with ammonia gas in ambient air. The dominance of  $\text{NaNO}_3$  could be caused by the evaporation of  $\text{NH}_4\text{NO}_3$  due to the moderate temperature (20°C - 25°C during the day) at the time of sample collection. Nitrate can be formed as a secondary pollutant through photochemical reactions in the atmosphere. It also shows the size distribution of  $\text{Ca}^{2+}$ , which has a strong peak in the coarse particle range.  $\text{Ca}^{2+}$  concentration was found to be at a relatively high level, next to  $\text{Na}^+$ , in the coarse particle range. This may be attributed to the counter ion of sulfate in the coarse particle range because  $\text{CaSO}_4$  would be derived from sea salt and soil. Calcium carbonate derived from soil could be a source of  $\text{Ca}^{2+}$  in the coarse particle range. Previous studies have shown that  $\text{Ca}^{2+}$  is more abundant in coarse particles during dust storms.

Nitric acid may react with ammonia gas to form fine particles and may also react with  $\text{NaCl}$  to form coarse particles. In this study, nitrate is predominantly classified in the coarse particle range. Therefore, it can be concluded that  $\text{NaNO}_3$  was the major species in the sample, and  $\text{NH}_4\text{NO}_3$  was present in lower amounts. Moreover,  $\text{NH}_4\text{NO}_3$  and  $\text{NH}_4\text{Cl}$  are known to be volatile under ambient temperature. It shows the size distribution of atmospheric  $\text{SO}_2$  and shows a monomodal distribution with a strong peak in the fine particle range accompanied by a slight shoulder in the coarse particle range. Sulfate is commonly formed by a slight shoulder in the coarse particle range. Sulfate is commonly formed by the gas-to-particle conversion of  $\text{SO}_2$  through photochemical reactions. Thus, the main peak falls in the fine particle range. The slight shoulder in the coarse particle range could be explained if some sulfate is derived from sea salt and soil. The monomodal size distribution of 4 shown in it has a sharp peak at approximately 0.5  $\mu\text{m}$ , similar to that of  $\text{NH}_4\text{SO}_4$  in the fine particle range. This could imply that 4 exists as a counter ion to 24. Ammonia and sulfuric acid will form ammonium sulfate or ammonium bisulfate, which are stable in ambient air and have a long life.

The magnesium particulates are mainly derived from sea salt. The monomodal distribution with a peak in the coarse particle range is similar to the  $\text{Na}^+$  distribution is shown. Some  $\text{K}^+$  may be

produced by the incineration of plant derivatives, such as paper, wood, and vegetable garbage, which would cause fine particle aerosols. Some studies have shown that  $K^+$  in aerosols can result from biomass burning. Potassium can also originate from sea salt and soil and would fall into the coarse particle range. These two sources result in two size distribution peaks for atmospheric potassium. The orders of the ionic compounds were  $24SO > NO > 3NO > 24SO > 4NH$  in the fine particle range and  $3 > Na^+$  in the coarse particle range. Sulfate was the predominant ion and occupied approximately 45% of the total mass concentration of the eight ionic compounds, followed by nitrate (22%) and ammonium (19%). It shows the ionic balances between the aerosol components from the Andersen sampler stages. The average ratios of total anion equivalence to total cation equivalence were calculated as 1.08 and 1.16 for coarse and fine particles, respectively. That is, a small cationic trend was observed; however, this is considered an acceptable coincidence between ion balances in both particle ranges. In the coarse particle range, the contribution of unevaluated carbonate ions may be a factor in the cation rich calculation. Ionic balances would be improved by evaluating organic hydrocarbon acids, which were not analyzed in the fine particle range.

#### 4. Conclusions

The size distribution of 4 had a sharp monomodal peak in the fine particle range, which is similar to that of sulfate. Most of the sulfate is considered to be ammonium sulfate or ammonium bisulfate. The size distribution of Ca was found to have a strong peak in the coarse particle range. Ca is considered to be a main counter ion of  $NH_4SO$  in the coarse particle range.

Significant amounts of  $Na^+$  and  $Cl^-$  were considered to be derived from sea salt. The difference between the  $Na^+/Cl^-$  ratio in the coarse particles and that in sea salt indicated a considerable loss of  $Cl^-$ . We concluded that this was caused by the formation of  $NaNO_3$ , which is the main nitrate species present. Moreover,  $NH_4NO_3$  was estimated to be in the fine particle range.

Mg is considered to be derived primarily from sea salt and soil and has a peak in the coarse particle range. Some K will originate from sea salt and will also be produced by the incineration of plant derivatives. Size distributions of atmospheric aerosol components in a suburban area of Tokyo, Japan were investigated using an Andersen sampler.

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