Measurement of $PM_{2.5}$ and Water-Soluble Ions at Central Tokyo, Japan and Source Apportionment

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Abstract: Air pollution by fine particulate matter, $PM_{2.5}$ has been inviting considerable concerns in East Asia. This study then aimed to characterize $PM_{2.5}$ and its water-soluble ions collected at Shibuya, central Tokyo, Japan from July 2013 to May 2015, in order to better understand the air pollution mechanism and potential sources of $PM_{2.5}$ of the city. Analytical results showed the $PM_{2.5}$ was a mixture of aggregates of formless particles and its concentration ranged from 5.4 to 39μg/m^3, with a mean of 14 ± 6.7μg/m^3 (n=46). SO_4^{2-} and NH_4^+ were abundant in $PM_{2.5}$ and their presence mostly determined the variation of $PM_{2.5}$ level. High concentration episodes were found in the season when air masses came from Asian continent, and a long-range transport of urban aerosols and soil-derived particles was suggested by changes in the chemical compositions.

Keywords: $PM_{2.5}$, water-soluble ions, source apportionment, Tokyo.

1. INTRODUCTION

Air pollution by fine particulate matter, $PM_{2.5}$ (defined as PM less than 2.5μm in aerodynamic diameter) has been inviting considerable concerns in East Asia because of severe pollution levels in China [1, 2], and its adverse effects on human health. Increased mortality by pulmonary and/or cardiovascular diseases due to exposure to $PM_{2.5}$ has been documented in a number of cohort studies such as Harvard Six Cities Study [3-5], American Cancer Society (ACS) cohort study [6-8], Great Britain Study [9] and other valuable studies summarized in ref [10]. Association of increased daily mortality with short-term exposure to $PM_{2.5}$ has been also reported by epidemiological studies carried out in Chinese cities [11, 12]. However, unlike other countries, no remarkable relationship has been found between fine particles and excess mortality in Japan [10]. $PM_{2.5}$ is a mixture of various particles emitted and/or generated from natural and anthropogenic sources, and hence appearance of adverse health effects may depend on the chemical composition of $PM_{2.5}$ at the receptor site. Therefore, understanding chemical components and source apportionment of $PM_{2.5}$ using the chemical fingerprint is important for pollution control of the complex media.

Water-soluble components are known to account for majority of the $PM_{2.5}$ mass [13, 14]. The major ions such as sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium ion (NH_4^+) have effects on the hygroscopic nature and acidity of aerosol, while their concentrations in air vary with seasons and geological locations [13]. Then, this study aimed to characterize $PM_{2.5}$ and its water-soluble ions collected at Shibuya, central Tokyo, Japan from July 2013 to May 2015, in order to better understand the air pollution mechanism and potential sources of $PM_{2.5}$ of the city, employing correlation analysis and trajectory analysis. The correlation analysis has been widely used for source apportionment of particulate matters, because significant mutual correlations between seasonal variations of air concentrations in PM and its components may show a possibility of sharing the common emission source by the paired variables [15]. Air trajectories are also used to study source–receptor relationship in environmental research by calculating airflow pattern from an emission source (forward) or from a sampling site (backward) [16, 17]. In this study, the back trajectory analysis was used to investigate the potential source area of high concentration episodes of $PM_{2.5}$ at central Tokyo.

2. METHODS

2.1. Sampling Site

Figure **1** shows the geographical location of Shibuya, Tokyo, Japan. Routine sampling of $PM_{2.5}$ was conducted at the open-air space of the school building

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E-ISSN: 2410-3624/15

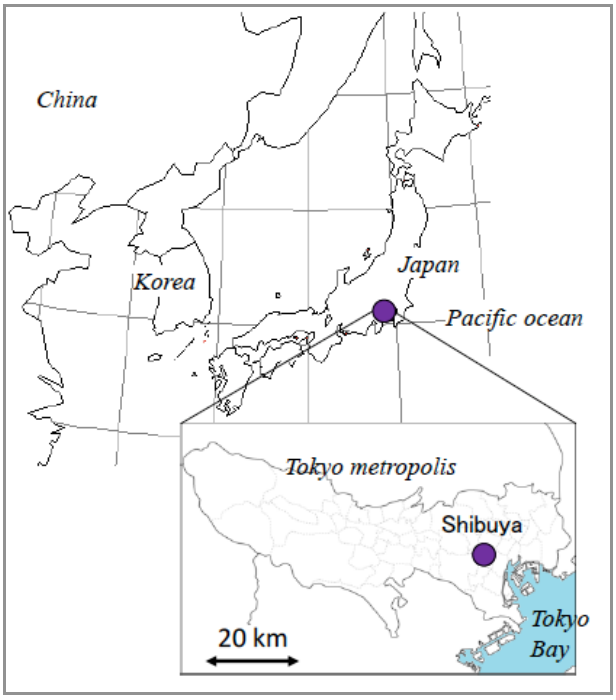

Figure 1: Location of sampling site, Shibuya, central Tokyo, Japan.

of Tokai University Bosei Senior High School located at Shibuya, Tokyo. Shibuya is one of the biggest cities in central Tokyo, Japan, having a population density of 13,540 people per km². The city is used to refer to a fashion center of Japan, particularly for young people. The sampling site locates in cultural and residential area of the city.

2.2. Collection of $PM_{2.5}$ Samples

$PM_{2.5}$ was collected on a quartz fiber filter (QMA 47mmφ, Whatman) at a flow rate of 16.7L/min for 7 days by $PM_{2.5}$ sampler (Thermo Fisher Scientific, Partisol® - FRM Model 2000). Both before and after sampling, filters were conditioned more than 24 hours under constant temperature (20℃) and relative humidity (50 ± 5%), then the weight of filter was quantified by using a microbalance. The concentration of $PM_{2.5}$ was determined by the weight difference and total sampling volume of air. Note that routine collection of $PM_{2.5}$ and measurement of $PM_{2.5}$ concentration were carried out by high school students, and subsequent chemical analysis was conducted by staffs of Tokai University.

2.3. FE-SEM and EPMA Analysis

Morphology of individual fine particles consisting of the $PM_{2.5}$ at Shibuya was observed by Field Emission Scanning Electron Microscope (FE-SEM, Hitachi, S-4800) at Tokai University (Kanagawa, Japan), after coated with a thin Au film to achieve higher quality secondary electron images. Distribution of major ions as particles on the quartz fiber filter was also analyzed by using Electron Probe Micro Analyzer (EPMA, Shimadzu, EPMA-1610) at Tokai University. EPMA is a non-destructive analytical tool used to determine the chemical composition of small volumes of solid materials within 10~30 cubic micrometers. The sample is bombarded with an electron beam, and signals that come from the sample are collected and displayed as like a map.

2.4. Water-Soluble Ion Analysis

After measurement of $PM_{2.5}$ concentration, the filters were cut into 14mmφ pieces by a pierce punch. The two pieces were transferred into a test tube together and water-soluble ions in $PM_{2.5}$ were extracted in 10mL ultrapure water (Milli-Q) with a mild shaking for 90 min. After filtration by a disposable cellulose acetate membrane filter having 0.20μm of pore size (Advantec, DISMIC-25CS), the filtrate was served for ion analysis. Concentrations of the water-soluble ions (Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, NO_3^-, SO_4^{2-}) were determined by ion chromatography (IC) systems.

The IC system for the determination of cation consisted of Shimadzu LC-20AD with a conductivity detector COD-10AVP. The following conditions were used: column, φ 4.6×150mm, 7μm, Shim-pack IC-C4 (Shimadzu); eluent, 2.5mM oxalic acid solution at 1.0ml/min (isocratic); oven temperature, 40℃; injection volume, 20μL. The IC system for anion consisted of Dionex ICS-90 with a chemical suppressor AMMS III. The following conditions were used: separation column, 4.0 × 250mM, IonPac AS9-HC (Dionex) with a guard column IonPacAG4 (Dionex); eluent, 5mM sodium carbonate at 1.0ml/min (isocratic), scavenger for the chemical suppressor, 15mM sulfuric acid solution at 1.0ml/min; oven temperature, 40℃; injection volume, 25μL. Dilution series of reagent grade NaCl, NH_4Cl, KCl, $MgCl_2$, $CaCl_2$, NaCl, $NaNO_3$ and Na_2SO_4 in ultrapure water were used for calibration and determination. All the reagents were obtained from Kanto Chemical, Japan.

Significant contamination by field handling and during storage was slightly found in storage blanks for anions, limit of detection (LOD) was defined as three-fold the standard deviation of the blank for anions and of 1.0μL ml^{-1} of standard solutions for cations (3σ). The LODs resulted in 0.0053μg/m^3 for Na^+, 0.0030μg/m^3 for

NH_4^+, 0.0023μg/m^3 for K^+, 0.0032μg/m^3 for Mg^{2+}, 0.013μg/m^3 for Ca^{2+}, 0.0010μg/m^3 for Cl^-, 0.0029μg/m^3 for NO_3^- and 0.0036μg/m^3 for SO_4^{2-}.

2.5. Back Trajectory Analysis

In order to characterize the general behavior of air masses in a given sampling period and to evaluate the relative contributions by local sources and long-range transportation to the total $PM_{2.5}$, three-day backward trajectories from the sampling site (longtitude: 139.7, latitude: 35.6) were computed beginning every 24h during the sampling duration using the Meteorological Data Explorer (METEX) program (Center for Global Environmental Research, National Institute for Environmental Studies) developed by Zeng *et al*. [16, 17] and available from the METEX website. The altitude above ground level was set to 500m.

3. RESULTS AND DISCUSSION

3.1. Morphology and Elemental Map of $PM_{2.5}$

Figure **2** shows a typical FE-SEM image of fine particles collected on the quartz fiber filter at Shibuya, Tokyo. The aggregates of formless particles were attached on the surface of fiber webs. These particles seemed a mixture of different particles and the shape and structure was not uniform, so it was very difficult to identify the particles by visual observations.

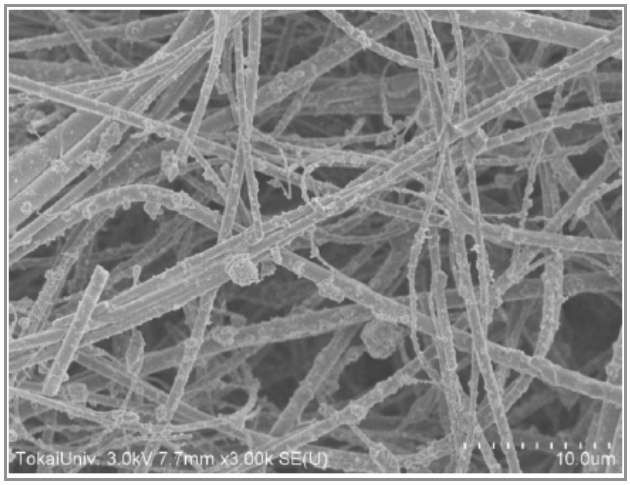

Figure 2: A typical FE-SEM image of the $PM_{2.5}$ sample collected at Shibuya, Tokyo (x3000).

EPMA analysis showed significant abundances of major elements such as Al, C, Ca, Fe, K, Mg, N, Na, O, S, Si and Ti in the $PM_{2.5}$ sample. Among the elements, Si and O are major components of the quartz fiber itself, and certain amount of Al, Ca, Mg and Na are contained as impurities of the filter [18]. Excluding these elements, relatively higher abundance was found for C and S. Figure **3** shows elemental distribution maps of C and S in a focus window of 480×480μm length. Carbon (red dot) was widely distributed in all over the sample with higher content. This means fine particles containing carbon, usually referred to elemental carbon (EC) and organic carbon (OC), are major components of $PM_{2.5}$ collected at Shibuya, Tokyo. Meanwhile, the picture also shows the presence of S (red dot) with very fine and dispersed signals. Comparing with carbon, S-containing particles were much finer than those containing carbon, probably because of most of S-containing aerosols such as water-soluble sulfate are secondary particles converted from gases.

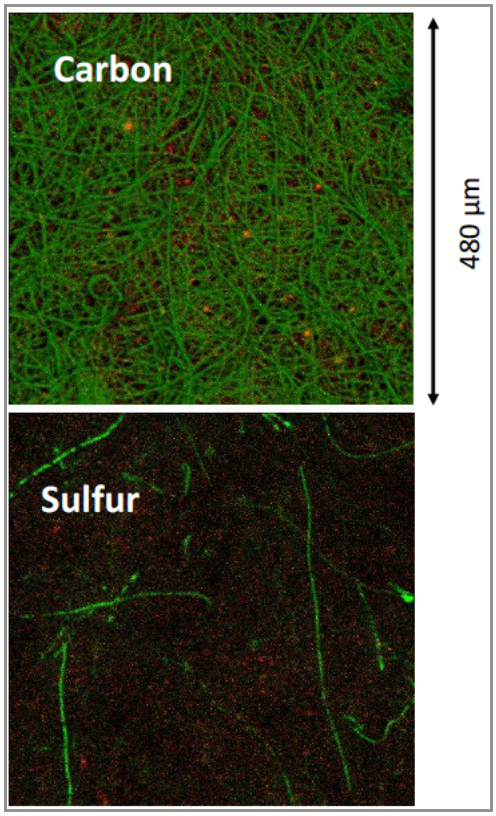

Figure 3: Elemental distribution maps of C and S on the $PM_{2.5}$ sample collected at Shibuya, Tokyo.

3.2. Concentration of $PM_{2.5}$ and Water-Soluble Ions

Mean concentrations and standard deviations of $PM_{2.5}$ and water-soluble ions were summarized in Table **1**. For the convenience in investigating seasonal differences, the results were classified into four seasons; spring (March to May), summer (June to August), autumn (September to November), and winter (December to February). The mean values and standard deviations at each season were also

Table 1: Concentrations of $PM_{2.5}$ and Water Soluble Ions Observed at Shibuya, Tokyo, Japan from July 2013 to May 2015 (Mean ± Standard Deviation in μg/m^3)

	Annual (*n*=46)	Spring (*n*=14)	Summer (*n*=6)	Autumn (*n*=15)	Winter (*n=12*)
PM2.5	14 ± 6.7	18 ± 7.6	12 ± 4.3	11 ± 3.3	14 ± 8.3
Na^+	0.15 ± 0.075	0.19 ± 0.068	0.067 ± 0.056	0.14 ± 0.077	0.14 ± 0.072
$NH4^+$	1.2 ± 0.93	2.0 ± 1.2	1.4 ± 0.24	3.0 ± 0.39	0.87 ± 0.60
K^+	0.17 ± 0.23	0.28 ± 0.39	0.13 ± 0.026	0.13 ± 0.04	0.12 ± 0.077
Mg^{2+}	0.025 ± 0.014	0.030 ± 0.015	0.036 ± 0.018	0.026 ± 0.012	0.015 ± 0.0066
Ca^{2+}	0.078 ± 0.032	0.092 ± 0.044	0.16 ± 0.033	0.07 ± 0.02	0.06 ± 0.021
Cl^-	0.018 ± 0.031	0.016 ± 0.023	0.0093 ± 0.0064	0.010 ± 0.007	0.031 ± 0.054
NO_3^-	0.24 ± 0.59	0.18 ± 0.34	0.088 ± 0.0068	0.047 ± 0.036	0.63 ± 1.0
SO_4^{2-}	3.5 ± 2.3	4.9 ± 2.4	3.8 ± 1.1	2.5 ± 1.4	3.0 ± 2.7
Others	8.6 ± 4.1	9.7 ± 4.2	7.9 ± 2.5	7.5 ± 3.0	9.1 ± 5.5

presented in the table. The 7-day average concentration of $PM_{2.5}$ ranged from 5.4 to 39μg/m^3, with a mean of 14 ± 6.7μg/m^3 (*n*=46). The $PM_{2.5}$ concentration was observed higher in spring and winter due in part to high concentration episodes described below, and relatively low in summer and autumn. Since the annual mean was somewhat less than the Japanese environmental quality standard (EQS) for $PM_{2.5}$ (15μg/m^3), status of air pollution by $PM_{2.5}$ is not so severe at present.

The concentrations of water-soluble ions in $PM_{2.5}$ followed the order of SO_4^{2-}> NH_4^+ > NO_3^- > K^+ = Na^+ > Ca^{2+}> Mg^{2+}> Cl^-. However, this order slightly changed with seasons; NO_3^- was relatively low in spring to autumn, and NH_4^+ was higher in autumn. Total mass of the eight kinds of ions accounted for about 40% of the $PM_{2.5}$ mass in any seasons.

Na^+ and Cl^- are major components of sea-salt particles. However, Cl^- concentration was much lower than that of Na^+ with 0.058~0.095 of molar ratio of Cl^- /Na^+ in every season, probably because of "chlorine loss". This is a phenomenon that volatile HCl is released by a heterogeneous reaction of sea salt particles with acidic gases such as SO_2, H_2SO_4 and HNO_3 in air [19, 20]. Considering the contribution from sea salt particles, concentration of sea-salt sulfate (ss-SO_4^{2-}) was estimated using Na^+ as an indicator of sea salt particles by equation (1),

$$[ss\text{-}SO_4^{2-}] = [Na^+] \times 0.251 \quad (1)$$

where, 0.251 is a weight ratio of SO_4^{2-}/Na^+ in sea water. The results showed concentration of ss-SO_4^{2-} ranged from 0.0043 ~ 0.091μg/m^3 with an average of 0.038μg/m^3 (*n*=46) which corresponded to only 1.4% of total SO_4^{2-}. Even though the sampling site locates only 5 km apart from the seashore of Tokyo bay, sea salt particles were not important as a source of SO_4^{2-} in $PM_{2.5}$ collected at Shibuya, Tokyo.

3.3. Correlation Analysis

Figure **4** shows variations of air concentrations of $PM_{2.5}$ and water-soluble ions at Shibuya, Tokyo. The date shown in the X-axis is start day of the 7-day samplings. The $PM_{2.5}$ concentration greatly varied with samplings with some peaks. The variation of SO_4^{2-} concentration was similar, whilst variations of NH_4^+ and K^+ were not similar to that of $PM_{2.5}$. Then, Pearson's correlation coefficients between paired data on the variation of air concentrations were calculated using a software, IBM SPSS® Statistics ver.19 and shown in Table **2**.

$PM_{2.5}$ concentration significantly correlated with those of NH_4^+ (*r* = 0.76) and SO_4^{2-} (*r* = 0.79). This means variation of $PM_{2.5}$ was mostly determined by the presence of ammonium sulfate such as $(NH_4)_2SO_4$ and NH_4HSO_4, typically most abundant secondary particles in air. Influence of sea salt particle (Na^+) and combustion effluent (NO_3^-) was also suggested on the $PM_{2.5}$ concentration with significant correlations.

A pair of Mg^{2+} and Ca^{2+} showed a significant correlation with *r* = 0.54. The both mineral ions are well known to enrich in a desert soil by soil salinity, and hence they are good indicators of yellow dust storm "Kosa" [21]. NH_4^+ is also known to be rich in soil of

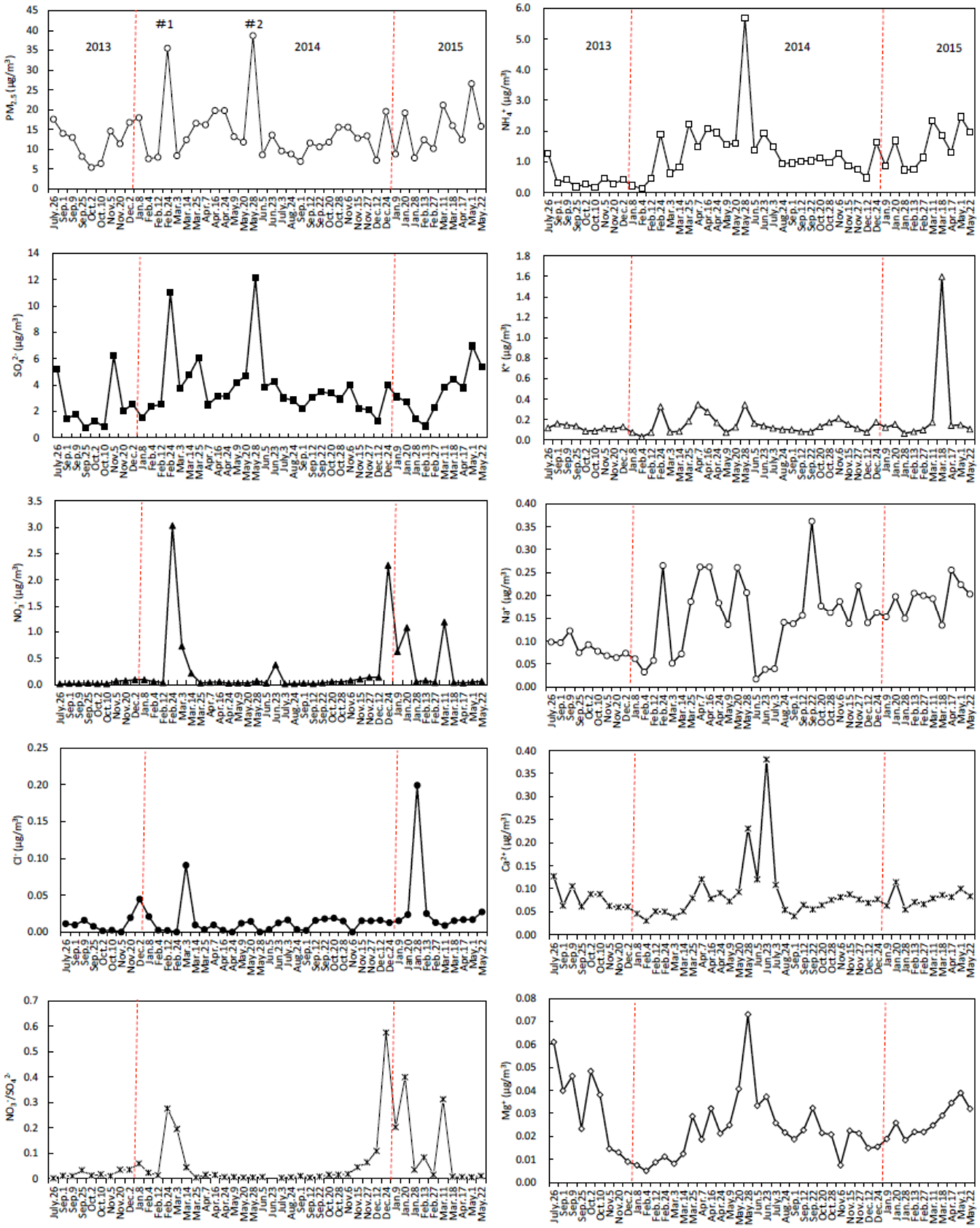

Figure 4: Variations of air concentrations of $PM_{2.5}$ and water soluble ions and nitrate/sulfate ratio observed at Shibuya, Tokyo, Japan from July 2013 to May 2015 (*n*=46, each plot shows 7-day mean).

Table 2: Correlation Coefficients between Paired Data of $PM_{2.5}$ and Water-Soluble Ions

	$PM_{2.5}$	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	NO_3^-	SO_4^{2-}
$PM_{2.5}$	1								
Na^+	**.41**	1							
NH_4^+	**.76**	**.43**	1						
K^+	.27	.11	.30	1					
Mg^{2+}	.28	.18	**.52**	.12	1				
Ca^{2+}	.29	-.07	**.52**	.11	**.54**	1			
Cl^-	-.19	-.05	-.15	-.09	-.17	-.14	1		
NO_3^-	**.47**	.19	.17	.06	-.23	-.04	-.02	1	
SO_4^{2-}	**.79**	.29	**.77**	.23	.32	.31	-.19	.39	1

Values expressed in bold are significant at 1% significance level.

China. Therefore, the significant correlation of NH_4^+, Ca^{2+} and Mg^{2+} indicates a possible long-range transportation of soil dust from China to central Tokyo.

3.4. High Concentration Episodes

As can be seen in Figure **4**, two remarkable peaks were found in $PM_{2.5}$ and SO_4^{2-} concentrations at the samples collected at Feb.24~Mar.2, 2014 (#1) and May 28~June 3, 2014 (#2). Figure **5** shows 3-day backward air trajectories corresponding to #1 and #2. As for #1, air trajectories came from Asian continent including northern part of China with remarkable peaks of SO_4^{2-} and NO_3^- concentrations. Since the mass ratio of NO_3^-/ SO_4^{2-} has been used as an indicator of relative importance of mobile versus stationary sources in the air pollution [13], there were previously several reports on the ratio: 0.53 at Jinan in winter [13], 0.63 at Beijing [22], 0.60 at Shanghai [23], 0.95 at Seoul, Korea [24]. Since the mass ratio apparently increased at #1 with 0.28, this suggests #1 sample is a mixture of urban aerosol particles transported from urban cities in Asian continent and fine particles originated from domestic sources of Japan.

Meanwhile, air trajectories of #2 came from China or around national borders on the sea, and passed through western part of Japan. According to Aeolian Dust Information by Japan Meteorological Agency [25], a long-range transport of yellow sand was observed in wide areas of western Japan during the sampling. Therefore, soil-derived particles from dessert areas in the continent were involved in the $PM_{2.5}$ sample with peaks of NH_4^+, Ca^{2+} and Mg^{2+}. Then, SO_4^{2-} concentration became also high because of formation of salt with these cations.

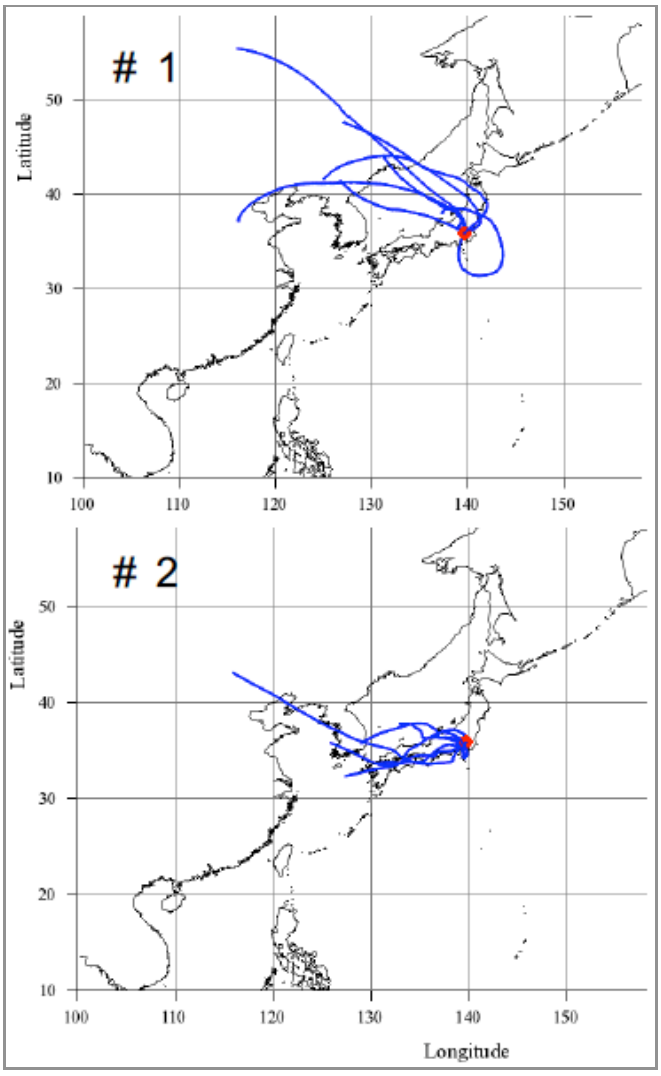

Figure 5: Three-day backward air trajectories during 7-day samplings, calculated for #1 (Feb.24~Mar.2, 2014) and #2 (May 28~June 3, 2014).

4. CONCLUSION

By investigating the concentrations of $PM_{2.5}$ and water-soluble ions at Shibuya, central Tokyo for 2 years, the following findings were obtained. Present status of air pollution by $PM_{2.5}$ was not so severe at the sampling site with a mean concentration of 14 ± 6.7μg/m^3 during the observation. SO_4^{2-} and NH_4^+ were abundant in $PM_{2.5}$ and their presence mostly determined the variation of $PM_{2.5}$ level. High concentration episodes were found in the season when air masses came from Asian continent, and a long-range transport of urban aerosols and soil-derived particles was suggested by correlations with indicator ions.

ACKNOWLEDGEMENT

This research activity was supported by Japan Science and Technology Agency (JST) Supporting Science Club program and JSPS KAKENHI Grant number 26410198. Authors awfully thank Mr. Yoshio Yamashita and Science Club members of Bosei Senior High School.

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Received on 12-11-2015 Accepted on 27-11-2015 Published on 31-12-2015

DOI: http://dx.doi.org/10.15377/2410-3624.2015.02.02.4