Preliminary Study on the Measurement of the Electrostatic Charging State of PM$_{2.5}$ Collected on Filter Media

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ABSTRACT

This study focused on the measurement of the actual charging state of ambient aerosol particles, which is important for understanding the intricate process of adverse health effects caused by particulate matter (PM). The net electrostatic charging state of PM$_{2.5}$ collected on filter media was measured in this study. The Faraday cage method and surface potential measurements were used in this study. The results showed that the polarities of the net charging state measured using these two methods were in agreement for 42 out of 48 samples (87.5%), and 36 samples (75%) were negatively charged. The filters were not significantly charged by friction between the filters and air not containing PM. Charge addition to or leakage from the filters was not observed over a two-month storage period. Net charging state of PM$_{2.5}$ collected on the filters was concluded to be negative in most cases, based on data's support of the assumption that aerosol charging state is not altered by the process of PM collection using filter.

Key words: Aerosols, Electrical charge, Faraday cage, PTFE filter, Surface potential

1. INTRODUCTION

Exposure to atmospheric aerosols is considered a serious concern for human health (IARC, 2013; Pope *et al*., 1995; Dockery *et al*., 1993). Particulate matter (PM) that passes through a size-selective inlet with a 50% efficiency cut-off in an aerodynamic diameter of 2.5 μm (PM$_{2.5}$) has been regulated worldwide for the last ten to twenty years (USEPA, 2013; Ministry of Environment, Japan, 2009; European Parliament and of the Council, 2008). To understand the adverse effects of PM more accurately, we need to know its physico-chemical properties such as agglomeration/aggregation state, chemical composition, particle size, shape, solubility, dispersibility, surface chemistry, specific surface area, and charging state (ISO, 2012). In particular, it has been reported that the deposition of particles in the respiratory tract could be enhanced by the surface charge of PM (Majid *et al*., 2012; Ali *et al*., 2008; Cohen *et al*., 1998). For example, Cohen *et al*. (1998) showed that singly-charged 125 nm particles were deposited to human airway replica 5-times more than zero-charged particles. Therefore, it has been suggested that the charging state of PM is important in understanding the intricate process of the effect of particle deposition on the human body. Furthermore, bio-activities of particles, such as particle intake and cyto-toxicity, could vary depending on the charging state even if their chemical composition and PM size were similar (Fröhlich, 2012; Yang *et al*., 2012; Patil *et al*., 2007).

It is known that the charging state of aerosol under a steady state is almost the same as the Boltzmann equilibrium charge state having near symmetric distribution between negative and positive polarities (Fuchs, 1963; Gunn, 1956). The Boltzmann equilibrium charge state is achieved after long exposure to sufficient amount of such bipolar ions (Flagan, 2011). Wiedensohler (1988) suggested that net charging state of aerosol particles was negative because the mobility of negative ions was larger than positive ions. Recently, Jayaratne *et al*. (2014) showed that particles carried more than double the charge they would normally carry in equilibrium under a certain condition, and had even net positive charge in some cases. The number of field observation studies that measure the actual charging state of aerosol particles is quite limited. Therefore, it is still open for discussion. This study aimed to measure the net electrostatic charging state of PM$_{2.5}$ collected on filter media using two independent methods.
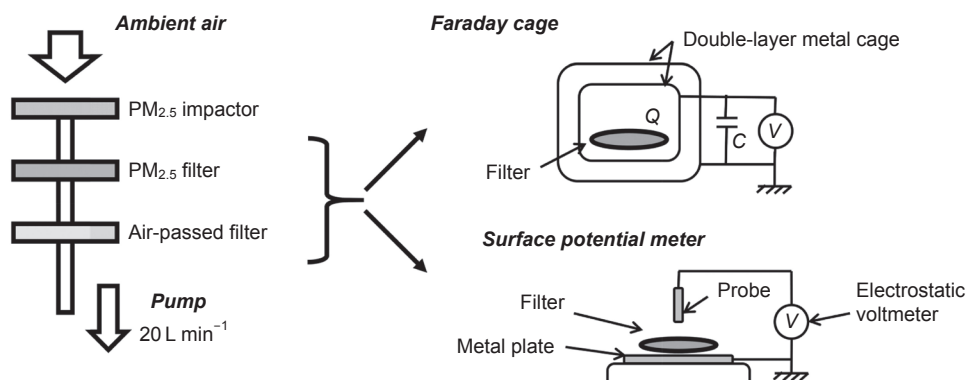
Fig. 1. Schematics of the experiments conducted in this study.

2. METHODS

2. 1 Concept of the Experiments

It would be convenient to measure the electrostatic charging state of PM collected on filter media since measurements of the chemical composition of PM are frequently conducted based on filter sampling methods. Therefore, we aimed to develop analytical procedures to measure the net charging state of filter samples. Determining the net charging state using two (or more) principles would confer analytical robustness. Therefore, in this study, two independent methods—, i.e., the Faraday cage method and surface potential measurements— were employed. The experimental strategy used in this study is shown in Fig. 1.

2. 2 Filter Sampling of PM$_{2.5}$

Ambient air PM$_{2.5}$ samples were collected on poly-tetrafluoroethylene (PTFE) filters (Whatman 2 µm PTFE 46.2 mm Filter, PP ring Supported for PM$_{2.5}$) with an airflow rate of 20 L min^{-1}. The charge amount of the filter was measured using the Faraday cage method described below, then the filter was set in the sampling train. A multinozzle cascade impactor (MCI, NL-20-2.5A, Tokyo Dylec. Corp.) was used as the PM$_{2.5}$ impactor (Okuda *et al.*, 2007a, b). A fluorocarbon coated glass fiber filter (Pallflex T60A20), which was immersed in silicone oil (Dow Corning 704R), was used as an impaction surface. The sampling period was from June 2013 to February 2014, and each sampling time was approximately 24 h. In each sample train, an additional PTFE filter (hereafter, the air-passed filter) was placed downstream of the sampling line. The sampling train was not grounded. We collected 73 sets of filter samples at the rooftop of a building of Keio University, located in a residential area (not a roadside) in

Yokohama, Japan.

2. 3 Faraday Cage Method

The amount of net electrostatic charge of the filter samples was measured using the Faraday cage method. A coulomb meter (Kasuga Denki, Inc., NK-1001) coupled with a Faraday cage (Kasuga Denki, Inc., KQ-1400), which consists of a double-layer metal cage, allowed us to measure the amount of net electrostatic charge of the filter samples. The PM$_{2.5}$ filters, air-passed filters, and unused (blank) filters were analyzed before and after each sampling. The precision of the replicated measurements was within 5% of the relative standard deviation (n＝5).

2. 4 Surface Potential Measurements

The surface potential of the filter samples was measured using a surface potential meter (Trek Inc., Model 323 non-contacting electrostatic voltmeter coupled with Model 6000B-7C vibrating capacitor probe). The probe head diameter was 11.2 mm. Details of the experimental procedure are described later. The PM$_{2.5}$ filters, air-passed filters, and unused (blank) filters were analyzed after each sampling.

2. 5 Elemental Analysis

Elemental analysis of the filter samples were performed using an energy-dispersive X-ray fluorescence spectrometer (EDXRF, Rigaku Inc., NEX CG, formerly known as EDXL300) without any pretreatments. Details of the analytical conditions of the EDXRF measurements have been described elsewhere (Okuda *et al.*, 2014, 2013a, b; Okuda and Hatoya, 2013). All EDXRF measurements were conducted after the measurements of the charging state.

3. RESULTS AND DISCUSSION

3. 1 Faraday Cage Method

3. 1. 1 Amount of Net Electrostatic Charge of PM$_{2.5}$ Collected on Filter Media

In this study, PM$_{2.5}$ particles were collected by passing air through filters. In this process, triboelectricity might be observed on the filters due to friction between the air and filter materials. Therefore, after PM$_{2.5}$ particles were collected on the first filter, an additional filter (the air-passed filter) was placed downstream of the sampling line. The net charge of the samples was calculated by subtracting the charge of the air-passed filter from that of the first (PM$_{2.5}$) filter. The charge of unused (blank) filters was also measured to determine the change in the charging state due to the handling of the samples. These results are presented in Table 1. The net charge amounts of the air-passed filters and the unused (blank) filters were almost 0 nC, clearly showing that the filters were not significantly charged by friction with air in the absence of PM. On the other hand, the net charge amounts of PM$_{2.5}$ collected on filters (-2.0 nC on average) were markedly different from those of the air-passed and blank filters. The data suggested that net charging state of PM$_{2.5}$ collected on the filters was likely to be negative.

3. 1. 2 Charge Amount of PM$_{2.5}$ after Removing Gaseous Ions

During the sampling, gaseous ions as well as particles contact with filter media. This may affect the charging state of the filter. Therefore, the effect of gaseous ions on the net charge amounts of sample filters was investigated. We prepared two sample trains; one of them was equipped with a gaseous ion denuder, which was a conductive tube (5 mm in diameter, 20 cm in length) that was grounded, at the air inlet, and the other was not equipped the gas denuder. For this experiment, the air flow rate was set at 5 L/min. The denuder was designed to remove more than 92% of electric charges carried by gaseous ions of which diffusion coefficient was 0.04 cm^2/s (Takahashi, 2003) by contacting of gaseous ions with grounded tube (Gormley and Kennedy, 1949). The results on the net charge amount of PM$_{2.5}$ collected on PTFE filters with/without gaseous ion denuder are shown in Table 2. The charge amount of filter with gas denuder seemed more negative than that without denuder, but this difference was not statistically significant (t-test, $p > 0.05$). Therefore, the effect of gaseous ions on the charge amount of the filter was not significant in this study.

3. 1. 3 Charge Stability

The charge amount of the filter samples may change

Table 1. Net charge amount of PM$_{2.5}$ collected on PTFE filters, filters that air without particulate matter passed through, and unused (blank) filters.

	Net charge amount [nC]		
	Mean	SD	n
PM$_{2.5}$ filter	-2.0	2.2	73
Air-passed filter	-0.1	1.0	72
Unused (blank) filter	0.2	0.5	34

Table 2. Net charge amount of PM$_{2.5}$ collected on PTFE filters with/without gaseous ion denuder.

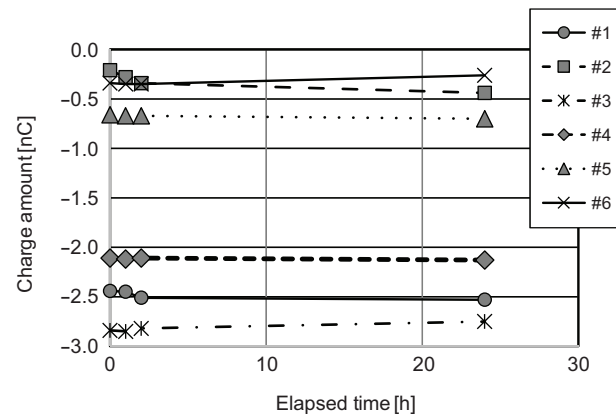|	Net charge amount [nC]		
	Mean	SD	n
Without gas denuder	0.0	1.9	4
With gas denuder	-0.8	1.5	4

Fig. 2. Change in the amount of electrostatic charge of PM$_{2.5}$ collected on PTFE filters during a 24-h storage. Six individual samples were tested. The air temperature and RH during the experiment were $23 \pm 2°C$ and $24 \pm 9\%$, respectively.

over time. Therefore, the time-trend of the charging state was observed by measuring the charge amount of the filter at 0, 1, 2, and 24 h after sample collections (Fig. 2). The charge amount of the filter samples did not change significantly within 24 h after the sampling. The charge amounts were re-measured after two-months of storage. Each sample filter was stored in a plastic petri slide, and then the slides were put in a shelf desiccator that was not grounded. The temperature and relative humidity (RH) in where the samples were stored were 20-25°C and 50%, respectively. During the storage, no changes were observed (Fig. 3); i.e., neither charge addition nor leakage was observed.
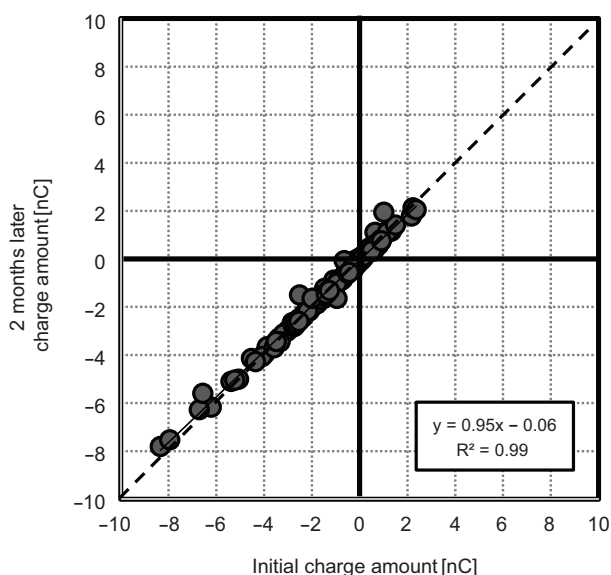
Fig. 3. Amount of electrostatic charge of the filter samples, measured in November 2013 and January 2014 (n = 148).
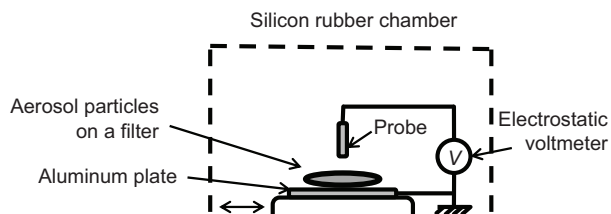

Fig. 4. Schematic of the system used to measure the surface potential of $PM_{2.5}$ collected on a filter medium.

3. 2 Surface Potential of $PM_{2.5}$ Collected on Filters

The following experiments were conducted to measure the surface potential of $PM_{2.5}$ collected on the filters.

(1) Experimental setup and measurement procedure
(2) Heterogeneity of the surface potential over the filter samples
(3) Distance between the probe and filter surface
(4) Surface potential measurements of the filter samples
(5) Stability of the surface potential of the filter samples
(6) Changes in the surface potential caused by ionization treatment and XRF measurement

3. 2. 1 Experimental Setup and Measurement Procedure

The schematic of the experimental setup is shown in Fig. 4. The measurement system was placed inside a simple chamber constructed using silicon rubber sheets that were not grounded. Air temperature and RH inside the chamber were monitored. The sample stage was a grounded aluminum plate. A zero check was performed by measuring the plate surface prior to each measurement. The electric potential difference between the probe and sample surface was measured using an electrostatic voltmeter.

Representative raw data of the surface potential measurements are shown in Fig. 5. The measurement procedure was as follows. First, zero setting was performed by measuring the plate surface where the distance between the measuring point and the edge of the
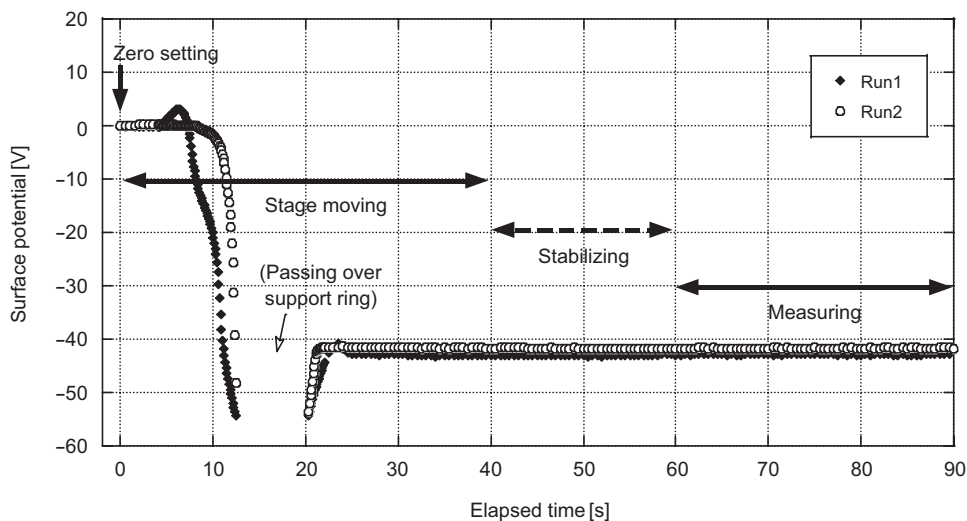

Fig. 5. Representative raw data of the surface potential *vs*. elapsed time along with the moving sample stage. The measured sample was $PM_{2.5}$ collected on a PTFE filter (PP-ring supported).

sample filter was 50 mm, so that the reading of the electrostatic voltmeter showed zero at the beginning of the measurement. Second, the aluminum plate stage, on which a sample filter was placed, was moved until the probe pointed to the center of the filter. The stage was moved for a total of 40 sec in each measurement. When the probe passed over the polypropylene support ring, the voltmeter reading started to display negative values, and then went off scale. This "off-scale" reading was observed in each measurement. This may be caused by negatively charged polypropylene support ring. After passing over the support ring, the reading returned and a particular value was displayed. Third, after 20 sec stabilization time, the reading averaged over 40 sec was considered as the surface potential of the filter sample (-43 V in the example case).

3. 2. 2 Heterogeneity of the Surface Potential over the Filter Samples

The surface potential of certain areas on the filter samples may vary because the PM deposition on the filter is essentially heterogeneous. Therefore, heterogeneity of the surface potential over the filter samples was examined by determining the surface potential at five measurement positions. The schematic of this experiment is shown in Fig. 6 and the results are presented in Table 3. The distance between the probe and sample surface was fixed at 3.5 mm. The results indicate that deviations in the surface potential values due to sample heterogeneity were smaller than inter-sample variation. The values of positions #2-5, which were not located at the center of the filter samples, may be affected by the adjacent support ring. Hence, the position #1 value (the center of the filter) was
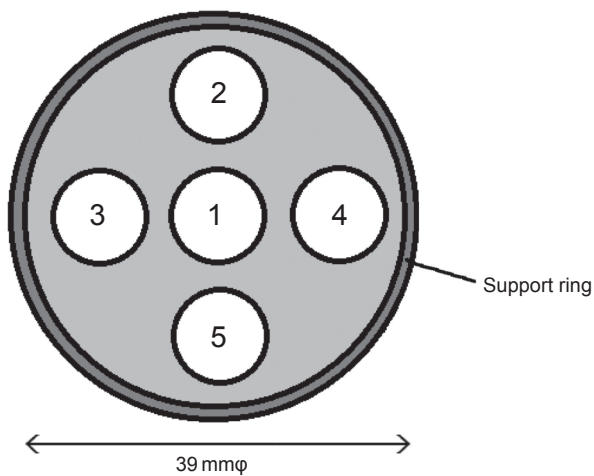
adopted as the measurement value.

3. 2. 3 Distance between the Probe and Filter Surface

The voltmeter readings may depend on the distance between the probe and filter surface. The shorter the distance, the smaller the surface area the probe can be affected by. It is important to optimize the distance between the probe and filter surface so that voltmeter readings represent the entire particle deposition area on a filter. Note that the optimum probe distance should have a certain maximum limit because factors other than particles (such as the support ring of the filter) affect the probe. In this experiment, the voltmeter readings were recorded as the distance between the probe and filter was changed. The results for two representative samples are shown in Fig. 7. As the distance increased from 3 to 7 mm, the voltmeter readings became more negative in sample I but less negative in sample II. At a distance greater than 7 mm, the voltme-

Table 3. Variations in surface potential readings measured in different positions on the filter surface.

	Surface potential [V]		
	Sample a	Sample b	Sample c
1 (center)	5.0	-43.1	-200.0
2 (upper)	5.1	-37.4	-158.2
3 (left)	5.3	-33.6	-193.5
4 (right)	4.7	-6.3	-142.6
5 (bottom)	4.6	-36.0	-131.7
Mean	4.9	-31.3	-165.2
SD	0.3	14.4	30.4

Fig. 6. Each measuring point of the surface potential on a filter sample. The distance between the probe and filter surface was fixed at 3.5 mm.
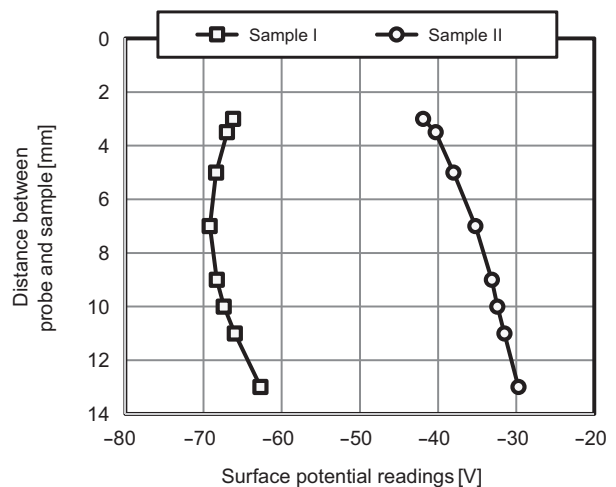

Fig. 7. Changes in the reading of surface potential by changing the distance between probe and sample.

Table 4. Surface potential values of $PM_{2.5}$ collected on PTFE filters, filters that air without particulate matter passed though, and unused filters.

	Surface potential [V]		
	Mean	SD	n
$PM_{2.5}$ filter	-115.5	90.0	73
Air-passed filter	-21.6	50.6	73
Unused (blank) filter	2.0	0.5	3

ter readings became less negative for both samples. Therefore, the optimal distance to set was assumed as between 3 and 7 mm. According to the manufacturer's information, the measurement area of the sample surface should be 25 mm in diameter when the distance between the probe and surface is 3.5 mm (Trek Inc., 2014). We considered 25 mm in diameter to be large enough to represent the total particles deposited on a filter. Consequently, a 3.5 mm distance between the probe and filter surface was adopted in this study.

### 3. 2. 4 Surface Potential Measurements of the Filter Samples

The surface potentials of $PM_{2.5}$ filters, air-passed filters, and unused (blank) filters were measured (Table 4). The surface potential of $PM_{2.5}$ collected on filters (-115.5 ± 90.0 V) were considerably different from those of the air-passed (-21.6 ± 50.6 V) and blank filters (2.0 ± 0.5 V). Therefore, net charging state of $PM_{2.5}$ collected on the filters was likely to be negative, a phenomenon that is consistent with the results of the charge amount measurements.

3. 2. 5 Stability of the Surface Potential of the Filter Samples

The surface potential of the filter samples might change over time. Therefore, the time-trend of the surface potential was monitored. The surface potential was re-measured after two months (Fig. 8). No obvious changes in the surface potential were observed during the two-month period as with the case of charge amount. Additionally, temperature and RH at the day of measurement did not affect the surface potential of the filter samples significantly.

3. 2. 6 Changes in the Surface Potential Caused by Ionization Treatment and XRF Measurements

Weighing and chemical analysis are often carried out using filter media. It is common to neutralize the electrostatic charge of filter media using an ionizer installed in an electronic balance during weighing. This procedure could alter the surface potential of filter samples. Similarly, elemental composition of filter
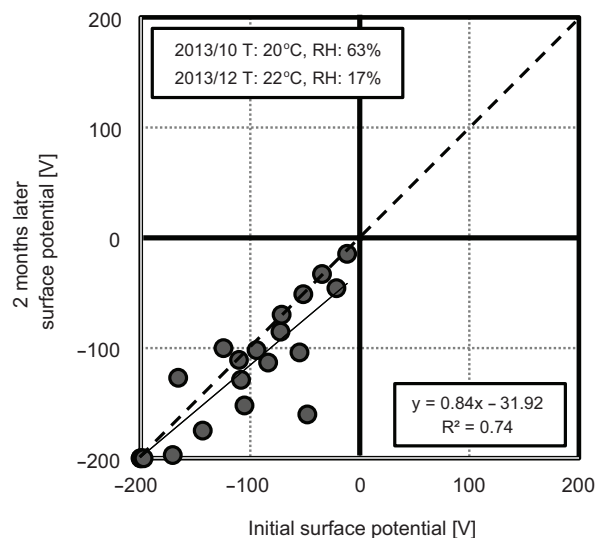
Fig. 8. Surface potential values of $PM_{2.5}$ collected on PTFE filters measured in October and December 2013 (n = 20).

Table 5. Changes in surface potential values of $PM_{2.5}$ collected on PTFE filters, before and after ionization treatment or EDXRF measurement.

	Surface potential [V]	
	Initial value	After treatment
Ionization at weighing		
Sample A	-127.0	-77.0
Sample B	-150.7	-162.5
Sample C	-111.3	-23.7
Blank D	-1.6	1.8
EDXRF measurement		
Sample E	-96.8	-3.6
Sample F	-66.4	-1.7
Blank G	-2.0	0.9
Blank H	-3.6	2.1

samples can be measured using X-ray fluorescence (XRF); however, X-ray irradiation can likewise alter the charging state of samples. In this study, changes in the surface potential of the filter samples during the ionization process and XRF measurements were examined (Table 5).

Obvious changes in the surface potential were observed during the ionization treatments and EDXRF analysis, with maximum values of 87.6 V and 93.2 V, respectively. Consequently, measurements of the charging state of filter samples should be carried out prior to weighing and XRF analysis.
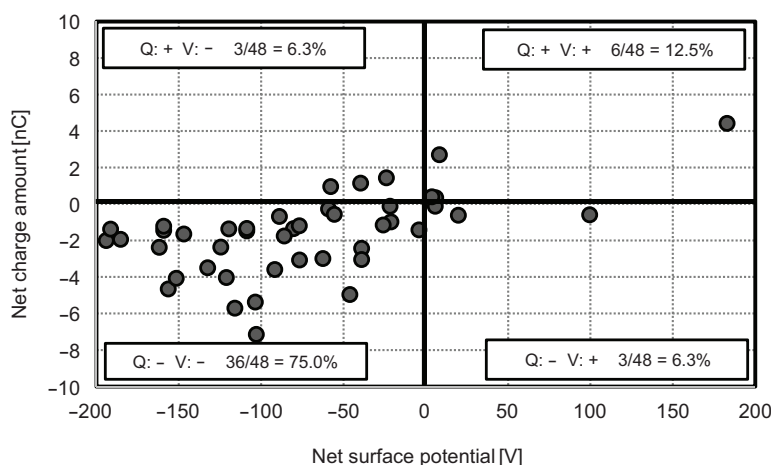
Fig. 9. Net charge amount and surface potential values of PM$_{2.5}$ collected on PTFE filters (n = 48).

3. 3 Relationship between Charge Amount and Surface Potential of the Filter Samples

Relationship between the net charge amount and surface potential measurements were investigated (Fig. 9). The polarity of the net charging state measured using these methods was in agreement for 42 out of 48 samples (87.5%), and that 36 samples (75.0%) were negatively charged. Therefore, net charging state of PM$_{2.5}$ collected on the filters was considered to be negative in most cases, based on the assumption that the charging state of the aerosols is not altered by the process of PM collection by filter itself.

Although the polarity of the charging state measured using these methods agreed well, individual plots in Fig. 9 did not lie on a particular regression line but were somewhat scattered. The specific reason for this is not clear; however, several possible reasons can be suggested. First, the Faraday cage method measures the entire charge amount of the filter whereas the surface potential meter measures the surface potential of part of the sample surface (see Table 3). Second, the Faraday cage method showed considerably better repeatability of measurements than the surface potential meter. Further studies are needed to resolve the cause of the observed scattering and the reason behind the superior repeatability of the Faraday cage method.

3. 4 Relationships between the Charging State of PM$_{2.5}$ and Other Parameters Such as Meteorological Conditions and Chemical Composition

The relationships between the net charging state of PM$_{2.5}$ and other parameters such as meteorological
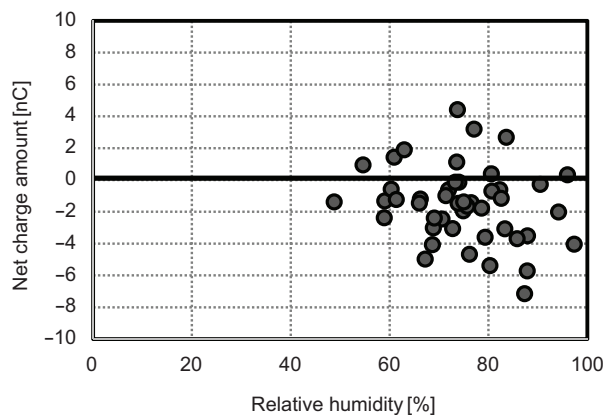

Fig. 10. Net charge amount of PM$_{2.5}$ collected on PTFE filters *vs*. RH during each sampling period (n = 46).

conditions and chemical composition were investigated to elucidate the factors controlling the charging state of PM$_{2.5}$ particles. The relationship between the net charge amount of the filter samples and RH during the sampling period (Japan Meteorological Agency, 2014) was investigated since the charging state of particles would have been affected by ambient RH (Gouveia and Galembeck, 2009). The results are shown in Fig. 10. No obvious relationship was observed between the two parameters. Similarly, no clear relationships were observed between net charging state and other meteorological parameters such as temperature, wind speed, rainfall amount, or sunshine duration. Based on these results, it does not seem that the net charging state of aerosols is related to any meteorological parameters.

Chemical composition of aerosol particles varies depending on sources and/or transport processes; there-
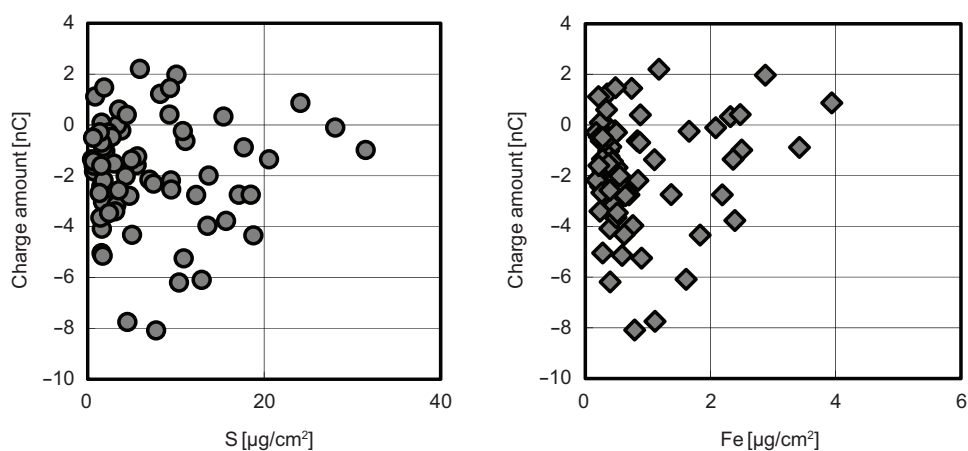
Fig. 11. Net charge amount of $PM_{2.5}$ collected on PTFE filters *vs.* amounts of selected elements on the filters (n = 71).

fore, it may affect charging state of the particles (Takahashi, 2003). The relationship between net charge amount and chemical composition was investigated, and representative results are shown in Fig. 11. No obvious relationships were observed between the net charge amount and concentrations of sulfur, iron, or other elements. It is possible, however, that net charge amount did not vary much from zero, even as the concentrations of these elements increased. We obtained similar results when surface potential was used as dependent variable instead of charge amount. The reason why net charging state of $PM_{2.5}$ collected on the filters was negative is still open for discussion. Further studies regarding this issue are critically needed.

4. CONCLUSIONS

Net charging state of $PM_{2.5}$ collected on the filters was concluded to be negative in most cases, based on the assumption that the charging state of the aerosols is not altered by the actual process of PM collection using filter. The filters were not significantly charged by friction with air alone. No changes in the charge amounts of the filter samples were observed during a two-month period after PM collection. Moreover, it does not appear that the net charging state of aerosols is related to any meteorological parameters or to their chemical composition. In addition, measurements of the charging state of filter samples should be carried out prior to weighing and XRF analysis because these processes can cause significant changes in the charging state. The reason why net charging state of $PM_{2.5}$ collected on the filters was negative is still open for discussion. Further studies regarding this issue are critically needed.

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