Investigation of Variations in Suspended Particulate Matter with Enforcement of Regulations on Diesel Vehicle Exhaust in Suburban Japan*

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In the summers of 2003 and 2004, size-separated suspended particulate matter (SPM) samples were collected with a high-volume Andersen air sampler at a site adjacent to Saitama Prefectural Route 57 in Saitama City. This sampling site is in an atmospherically polluted area that is also one of the "Specified Areas concerning Special Measures for Total Emission Reduction of Nitrogen Oxides and Particulate Matter from Automobiles" established in October 2002. We investigated carbonaceous compounds in the SPM before and after the Regulation on Diesel Vehicle Exhaust came into effect in October 2003 in Tokyo Metropolis and Saitama, Chiba, and Kanagawa prefectures. At the sampling site, elemental carbon (EC) in the fine particles (< 2 μm) was derived mainly from diesel vehicle exhaust emissions, and crustal metals such as Al, Fe, and Mg in the coarse particles (> 2 μm) were generated as road dust by vehicular traffic and wind. Correlations among chemical components generated by heavy-duty diesel vehicles suggest that the air quality is improving at the sampling site as a result of the enforcement of the Regulations on Diesel Vehicle Exhaust.

Key Words: Suspended Particulate Matter, Fine Particles, Coarse Particles, Road-Dust, Regulations on Diesel Vehicle Exhaust

1. Introduction

Suspended particulate matter (SPM), one of the major air pollutants in urban areas[1], is generated by many sources, including by automobile exhaust and industrial combustion and mechanical processes, and secondarily from gaseous pollutants[2]. Recently, ambient fine particles have been reported to have serious health effects; their current levels in ambient air are associated with increased cardiovascular and respiratory morbidity and mortality[3),(4)]. However, it is not yet known whether these health effects are caused by high concentrations (high numbers) of particles or by toxic components in the fine particles. To characterize recent SPM pollution, it is necessary to study the mass concentrations, size distributions, and origins of major chemical components in SPM in the urban and suburban atmosphere.

Atmospheric pollution in the Kanto District has become a serious problem[5)-(8)]. In October 2002, new vehicle-category regulations were implemented under the "Law concerning Special Measures for Total Emission Reduction of Nitrogen Oxides and Particulate Matter from Automobiles in Specified Areas" (hereinafter referred to as the "Automobile NO_x/PM Law") to address emissions from vehicles already in use[9]. Furthermore, in October 2003, Tokyo Metropolis, and Chiba, Saitama, and Kanagawa prefectures began to enforce the Regulation on Diesel Vehicle Exhaust Gas, based on the Ordinance on Environmental Preservation to Secure the Health of Citizens. This ordinance regulates the operation of heavy-duty vehicles (trucks and buses) that fail to comply with specified PM emission standards in the respective regulated area. Therefore, vehicles not in compliance with this regulation must be replaced with low-emission vehicles or equipped with PM reduction systems.

In Japan, many studies on the sources and chemical composition of atmospheric particles in urban and suburban areas have been carried out[5),(8),(10),(11)]. However,

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few studies on SPM characteristics before and after the Regulation on Diesel Vehicle Exhaust came into force in October 2003 have been conducted in the greater Tokyo Metropolitan Area; thus, more information is needed.

In this study, we collected size-separated atmospheric particles alongside Saitama Prefectural Route 57 during 2003 – 2004 in a relatively polluted area, Saitama City. We investigated the characteristic variations in SPM before and after the Regulation on Diesel Vehicle Exhaust Gas began to be enforced in October 2003, and inferred sources of SPM on the basis of correlations among the measured chemical components.

2. Experimental

2.1 Site description

The Tokyo Metropolis and Chiba, Saitama, and Kanagawa Prefectures (i.e., the capital city and the three adjacent prefectures) in the southern part of Kanto District (South Kanto) constitute the Tokyo Metropolitan Area. Saitama Prefecture is located in central Kanto District, and Saitama City (with a population of more than one million, in the southeastern part of the prefecture) is the seat of the Prefectural Government. Thus, Saitama City is in the center of South Kanto.

SPM samples were collected with a high-volume Andersen air sampler (HVA; Model AH-600F, Shibata Scientific Technology Ltd., Tokyo, Japan) alongside Prefectural Route 57 near the Waste Liquid Treatment Institute of Saitama University in Saitama City (Fig. 1). The HVA was installed in close proximity to the highway (ca. 1.5 m from the drive-way's edge on the north side of the road). SPM was sampled in summer (July and August 2003 and August 2004).

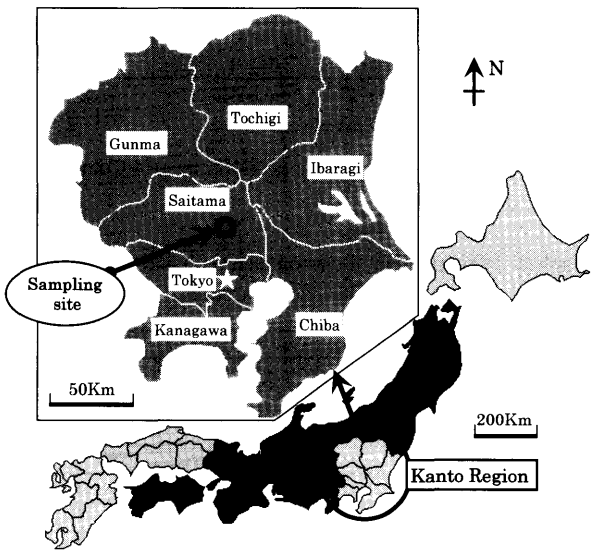

Fig. 1 Sampling site location in Saitama City, Saitama Prefecture

2.2 Description of the measurement equipment

The size-separated SPM samples were collected using an HVA with a standard five-stage cascade impactor. SPM were collected on four quartz fiber filters and on a quartz fiber back-up filter (2500QAST, Pallflex; Shibata Scientific Technology Ltd.) on the impactor stages, separating the particles according to their diameters into five size fractions from coarse to fine (equivalent aerodynamic diameters, < 1.1, $1.1-2.0$, $2.0-3.3$, $3.3-7.0$, and $> 7.0\,\mu m$). The sampler was operated at a flow rate of $566\,L min^{-1}$.

The sampling was continuous from 00:00 to 23:30 JST each day during the sampling periods (30 July to 7 August 2003 and 14 to 21 August 2004). After the sampling, loaded filters were stored for 24 h or more in an apparatus that controlled the temperature at 25°C and the relative humidity (RH) at 50%, and then weighed. Subsequently, they were stored in a freezer at about −40°C to prevent evaporation of volatile components.

Ambient temperature, RH, wind speed, and wind direction were monitored during the sampling periods by an EDO MET-9800 system (Shibata Scientific Technology Ltd.). Traffic volumes along Route 57 during the sampling periods were recorded on videotapes for later manual counting. The total volume of vehicles in both directions was counted during a 10-min period each hour. The average total traffic volume during the sampling period was 20 500 vehicles (about 5 000 heavy-duty diesel-fueled trucks and buses and about 15 500 light-duty gasoline-fueled automobiles) per day on weekdays and 15 000 vehicles (about 2 700 heavy-duty diesel-fueled trucks and buses and about 12 400 light-duty gasoline-fueled automobiles) per day on Saturdays and Sundays (Fig. 2).

2.3 Chemical analyses

2.3.1 Carbonaceous aerosol analysis We analyzed all loaded filters for organic carbon (OC) and elemental carbon (EC) using a thermal optical carbon analyzer (DRI model 2001, Shibata Scientific Technology Ltd.). The IMPROVE thermal/optical reflectance (TOR) protocol[12] was used for the carbon analysis. In the protocol, a 0.503-cm^2 punch aliquot of a sample quartz fil-

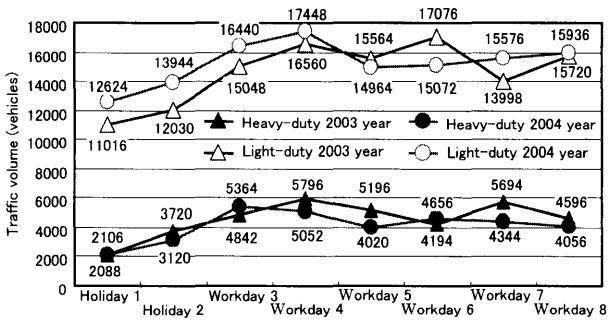

Fig. 2 Traffic volume at the sampling site alongside Saitama Prefectural Route No. 57

ter is heated at temperatures of 120 (OC1), 250 (OC2), 450 (OC3), and 550 (OC4)°C in a non-oxidizing helium (He) atmosphere, and at 550 (EC1), 700 (EC2), and 800 (EC3)°C in an oxidizing atmosphere of 2% oxygen and 98% helium. The repeatability, determined from replicate analyses, was better than 5% for total carbon (TC) and 10% for OC and EC[13].

2.3.2 Analysis and pretreatment method of metallic elements We analyzed SPM samples for aluminum (Al), iron (Fe), and magnesium (Mg). The elemental composition of the samples was quantified using inductively coupled plasma with atomic emission spectroscopy (ICP-AES, JICP-PS1000UV, Japan Electron Optics Laboratory Co., Ltd., Tokyo, Japan) and atomic absorption spectrophotometry (AA-6300, Shimadzu Corporation, Kyoto, Japan) at the Molecular Analysis and Life Science Center of Saitama University. Pre- and post-processing of the samples were performed in a clean laboratory. All plastic vessels were nitric acid-cleaned prior to use. Three-eighth parts of filters were digested in a solution of 4 mL of 46% HF, 6 mL of 61% HNO_3, and 2 mL of 46% $HClO_4$, on a hot plate at 110°C (±3°C) for 6 h. After acid digestion, the samples were evaporated to dryness at 60°C, redissolved in 1% nitric acid, and filtered before ICP-AES analysis. Field blanks were also analyzed, and the resulting values were subtracted from the analytical results for the respective samples.

3. Results and Discussion

3.1 Average concentrations of carbonaceous species

Carbonaceous species, including OC and EC, are major components of SPM, especially fine particles, in Japan[10),(11)]. EC is graphitic carbon formed during incomplete combustion of carbon-based fuels[14]. OC is derived from both primary and secondary generated carbon and includes polycyclic aromatic hydrocarbons and other components with possible mutagenic and carcinogenic effects. Soot carbon, which is predominantly EC and a major component of $PM_{2.5}$, has been linked to a significantly increased risk of death from lung cancer and other severe respiratory ailments[15].

The size-separated concentrations of EC and OC collected alongside Route 57 are shown in Fig. 3, and, for reference, the carbon data with and without laser correction of the size separate are shown in Table 1. The average concentration of EC in the < 1.1 μm fraction in July and August 2003 was clearly higher than that in August 2004, but the average concentrations of EC in the other size fractions in 2003 were almost the same as those in 2004. As a general trend, the OC concentration was higher than the EC concentration in each size fraction, except in the < 1.1 μm fraction. Furthermore, most EC and OC was found in the < 1.1 μm fraction, which suggest that they were derived from anthropogenic sources such as combustion processes or diesel vehicle exhaust emissions[16]. Moreover, $OC_{>7\,\mu m}$ also occurred in relatively high concentrations.

3.2 Average concentrations of major metal components

Mass concentrations of the size-separated particles increased from the smaller to larger particle-size fractions, and relatively high concentrations of Al and Fe were observed (Fig. 4). These elements, along with Mg, are crustral elements. In general, diesel-vehicle exhaust is believed to be the strongest local contributor to fine particle mass alongside roads[17]. Moreover, it is well known that high concentrations of coarse particles alongside roads are mainly due to resuspension of the road dust by traffic and wind[18], and that, in addition to road dust, coarse particles such as tire dust are strongly related to the volume of vehicular traffic[19),(20)].

3.3 Correlations between traffic volume and chemical components

We examined the relationships between traffic volume and concentrations of the chemical components (Table 2). A high positive correlation indicates that the concentrations of that chemical component vary similarly over time with the variation in the traffic volume and sug-

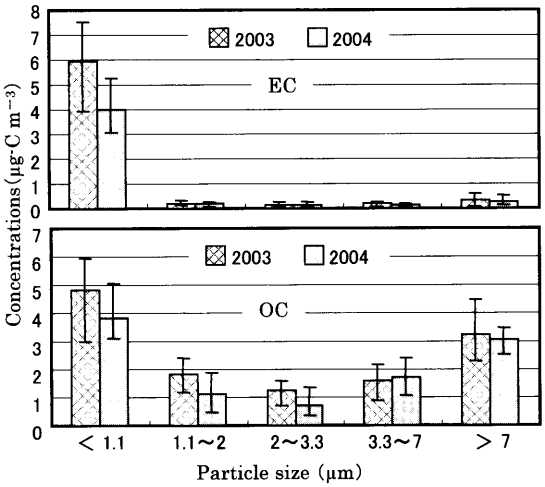

Fig. 3 Variation of average concentrations of EC and OC at sampling site

Table 1 Comparison of carbon data with and without laser correction of the size separations

Component		Year	< 1.1 μm	1.1~2 μm	2~3.3 μm	3.3~7 μm	> 7 μm
With laser correction	EC	2003	5.94	0.19	0.16	0.18	0.34
		2004	3.99	0.19	0.12	0.14	0.28
	OC	2003	4.80	1.81	1.25	1.61	3.21
		2004	3.83	1.12	0.73	1.70	3.07
Without laser correction	EC	2003	7.71	0.82	0.49	0.64	1.35
		2004	5.03	0.44	0.33	0.52	1.16
	OC	2003	3.03	1.17	0.91	1.15	2.19
		2004	2.79	0.87	0.52	1.32	2.18

Note: Unit of concentration (μ g-C m^{-3})

gests that the chemical component may originate from vehicle exhaust or traffic. Their relationships were represented with correlation coefficient (r) and significance level (p).

3.3.1 Correlation between traffic volume and chemical components in resuspended dust Correlations between traffic volume and crustal metal elements such as $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$ and $Mg_{>2\,\mu m}$ in the resuspended dust alongside the road are shown in Table 2. We found significant correlations between heavy-duty traffic volume and concentrations of all metal elements in the resuspended dust both in 2003 and 2004 (r, 0.64 – 0.89). However, only $Al_{>2\,\mu m}$ in 2003 and $Mg_{>2\,\mu m}$ both years were moderately correlated with light-duty traffic volume. We found fairly strong linear correlations in data of 2003 and 2004 only between the heavy-duty traffic volume and the concentrations of $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$, or $Mg_{>2\,\mu m}$ component (Table 2). These facts suggest that the resuspended dust mass was strongly dependent on heavy-duty traffic volume. Metal elements such as Al, Fe, and Mg found mainly in coarse particles (> 2 μm) were abundant constituents of all road dust and soil particles. Therefore, these results clearly suggest that these mineral aerosols are likely generated by the resuspension of road dust and soil (resuspended dust)[21]. Moreover, increase in the number of heavy-duty vehicles on the road probably increases the proportion of the road-dust contribution to SPM mass.

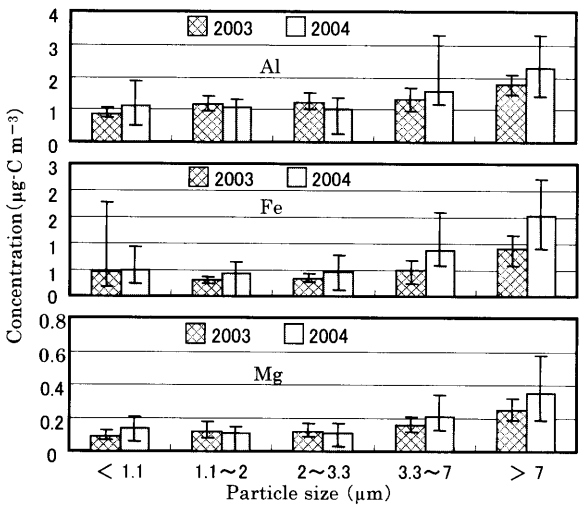

Fig. 4 Average mass concentrations of metals at the sampling site

3.3.2 Correlation of traffic volumes and diesel exhaust emission Although traffic volume was not correlated with $OC_{<2\,\mu m}$, we observed fairly strong correlations between heavy-duty traffic volume and the concentration of $EC_{<2\,\mu m}$ (r, 0.61 and 0.81) (Table 2). Correlations between light-duty traffic volume and $EC_{<2\,\mu m}$ were very low (r, 0.33 and 0.35). These results suggest that vehicle exhaust emission was strongly dependent on the volume of heavy-duty diesel vehicle traffic. Many studies have demonstrated that the EC content in fine particles is an indicator of diesel exhaust particles because EC constitutes a large fraction of the mass of diesel particles[22]. However, the slope (slope = Δconcentration/Δtraffic volume) of the correlation equation was 0.001 6 in 2003 and 0.001 1 in 2004, a decrease of about 31%, suggesting that enforcement of the Regulation on Diesel Vehicle Exhaust Gas may have reduced the amount of elemental carbon in diesel vehicle exhaust gas.

3.4 Correlations between major crustal components and EC

We also investigated the relationship between the crustal elements $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$, and $Mg_{>2\,\mu m}$ and the main component of heavy-duty diesel vehicle exhaust, that is, $EC_{<2\,\mu m}$ (Fig. 5). All of these elements were highly correlated with the volume of heavy-duty diesel vehicle traffic, as shown previously, and strong correlations of $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$, and $Mg_{>2\,\mu m}$ with $EC_{<2\,\mu m}$ were also found. Therefore, heavy-duty diesel vehicles strongly influence roadside atmospheric pollution in both fine and coarse particles.

A comparison of the slopes (slope = $\Delta EC_{<2\,\mu m}$ concentration/$\Delta Al_{>2\,\mu m}$ or $\Delta Fe_{>2\,\mu m}$ or $\Delta Mg_{>2\,\mu m}$ concentration) of the linear regression equations between concentrations of the coarse crustal elements and fine EC showed a clear decrease in each slope from 2003 to 2004 ($Al_{>2\,\mu m}$, from 3.73 to 1.48, −60.3%; $Fe_{>2\,\mu m}$, from 6.99 to 2.60, −62.8%; and $Mg_{>2\,\mu m}$, from 19.61 to 7.41, −62.2%). This

Table 2 Correlation between traffic volume and chemical components during the sampling periods

	Slope × 1000				r				p			
	Heavy-duty		Light-duty		Heavy-duty		Light-duty		Heavy-duty		Light-duty	
Component	2003	2004	2003	2004	2003	2004	2003	2004	2003	2004	2003	2004
Al > 2 μm	0.30	0.70	0.17	0.07	0.79	0.77	0.77	0.11	0.019	0.027	0.025	0.788
Fe > 2 μm	0.20	0.42	0.05	0.16	0.69	0.89	0.31	0.49	0.058	0.003	0.449	0.209
Mg > 2 μm	0.05	0.10	0.02	0.07	0.71	0.64	0.64	0.67	0.048	0.085	0.088	0.068
EC < 2 μm	1.60	1.07	0.48	0.39	0.61	0.81	0.35	0.33	0.108	0.011	0.390	0.419
OC < 2 μm	0.95	0.49	−0.26	0.29	0.29	0.22	0.18	0.18	0.490	0.600	0.660	0.671

Note: $n = 8$
X axis: Traffic volume
Y axis: Concentration of chemical components
r: Correlation coefficient
p: Significance level

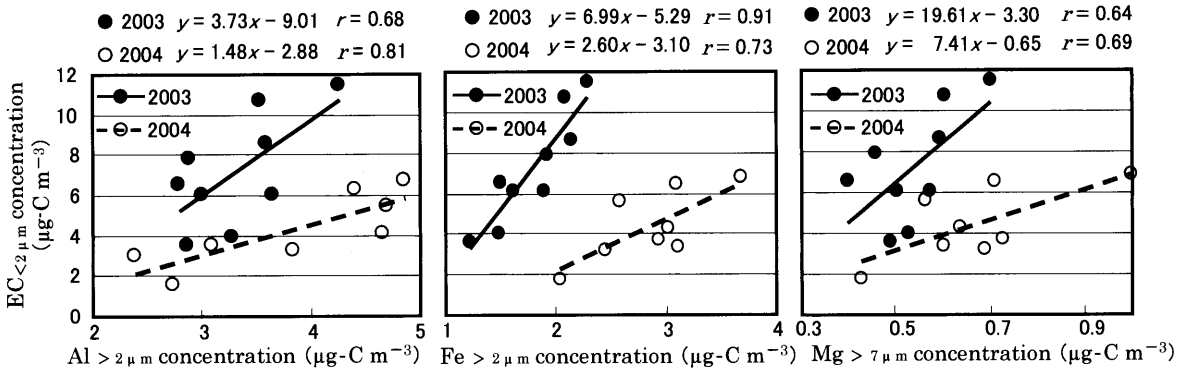

Fig. 5 Correlations between $EC_{<2\,\mu m}$ and suspended road-dust components at the sampling site

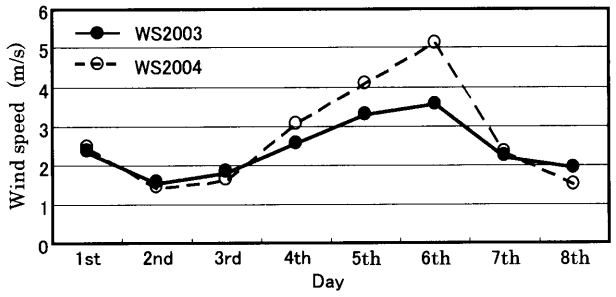

Fig. 6 Comparison of wind speed between 2003 and 2004 during the sampling period

slope decrease can be explained by an increase in the concentrations of crustal elements due to road dust generated by total vehicular traffic or by a decrease of EC in vehicle exhaust gases.

Concentrations of various air pollutants depend strongly on the meteorological conditions, especially wind speed during the sampling period. The most frequent wind direction was south–southwest and the mean wind speed was 2.47 and 2.68 m/s, respectively, during the sampling periods in 2003 and 2004 (Fig. 6). In general, because concentrations of fine particles decrease with an increase in wind speed[23], it may be presumed that the $EC_{<2\,\mu m}$ concentration in 2004 decreased by about 10% because of the change in wind speed. Thus, we estimated a decrease of about 20% in the $EC_{<2\,\mu m}$ concentration from 2003 to 2004, after excluding the dilution due to the average wind speed during the SPM sampling. Although we attributed most of the decrease in the EC concentration to the Automobile NO_x/PM Law and the Regulation on Diesel Vehicle Exhaust Gas, multi-year sampling and a more detailed data analysis are needed to clarify the influence of these regulations on the atmospheric SPM concentration.

4. Summary and Conclusions

Size-separated SPM samples were collected by a high-volume Andersen air sampler alongside Prefectural Route 57 in Saitama City. The crustal elements, Al, Fe, and Mg, occurred mainly in coarse particles (> 2 μm) and EC mainly in fine particles (< 2 μm). Average mass concentrations of carbonaceous species in five size-separated particle fractions were generally higher in 2003 than in 2004. From the correlations between the chemical components and the heavy-duty traffic volume, we inferred that $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$, and $Mg_{>2\,\mu m}$ were generated by the high-duty diesel vehicle traffic and that $EC_{<2\,\mu m}$ was mainly emitted as diesel automobile exhaust. Moreover, we found strong correlations between $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$, and $Mg_{>2\,\mu m}$ and $EC_{<2\,\mu m}$, which is an indicator of diesel vehicle exhaust.

Slopes of the linear regression equations between of the $Al_{>2\,\mu m}$, $Fe_{>2\,\mu m}$, or $Mg_{>2\,\mu m}$ concentration and the $EC_{<2\,\mu m}$ concentration decreased clearly from 2003 to 2004. Moreover, we found a similar decrease in slope between the $EC_{<2\,\mu m}$ concentration and the volume of heavy-duty diesel vehicle traffic. These results suggest that most of the decrease in the EC concentration can be attributed to the Automobile NO_x/PM Law and Diesel Vehicle Exhaust Gas Regulation. A detailed analysis of multi-year variation in the $EC_{<2\,\mu m}$ concentration at the other roadside site is now in progress.

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