Changes in Fine PM Pollution Levels with Tightening of Regulations on Vehicle Emissions

Akihiro Iijima, and Kimiyo Kumagai

***Abstract*—**A long-term campaign for monitoring the concentration of atmospheric Particulate Matter (PM) was conducted at multiple sites located in the center and suburbs of the Tokyo Metropolitan Area in Japan. The concentration of fine PM has shown a declining trend over the last two decades. A positive matrix factorization model elucidated that the contribution of combustion sources was drastically reduced. In Japan, the regulations on vehicle exhaust emissions were phased in and gradually tightened over the last two decades, which has triggered a notable reduction in PM emissions from automobiles and has contributed to the mitigation of the problem of fine PM pollution.

***Keywords*—**Air pollution, Diesel-powered vehicle, Positive matrix factorization, Receptor modeling.

I. INTRODUCTION

RECENTLY, particulate matter (PM) has become one of the air pollution issues of greatest concern. In Japan, guidelines for the concentration of suspended particulate matter (SPM, representing PM less than or equal to 10 μm in aerodynamic diameter under the 100% cut-off condition) have been enforced since 1973. Owing to the tightening of regulations on several emission sources, the current SPM concentration has decreased notably [1]. Nonetheless, in the past several decades fine PM has been conclusively demonstrated to strongly affect human health [2]. In Japan, new guidelines for $PM_{2.5}$ (35 μg/m³ 24-h mean and 15 μg/m³ annual mean) were introduced in 2009 [3].

In 1989, local governments in the center and suburbs of the Tokyo Metropolitan Area started a joint research project that aims to clarify the major sources of fine PM ($PM_{2.1}$) pollution [4], with particular focus on the Tokyo Metropolitan Area, which is characterized by a large population, heavy traffic, and massive industrial complexes. In the last several decades, regulations on vehicle emissions have been phased in and gradually tightened, and regulations for the transport sector tend to become tighter as a global trend.

Prior to investigating the introduction of further regulations, it is important to evaluate the effects of previous policies. By using positive matrix factorization (PMF), this study discusses whether previously enforced regulations on vehicle emissions have contributed to alleviating the fine PM pollution problem in the Tokyo Metropolitan Area.

A. Iijima is with the Faculty of Regional Policy, Takasaki City University of Economics, Takasaki, Gunma 3700801 Japan (corresponding author, phone: +81-27-343-5417; fax: +81-27-343-4830; e-mail: a-iijima@tcue.ac.jp).

K. Kumagai is with the Gunma Prefectural Institute of Public Health and Environmental Sciences, Maebashi, Gunma 3710052 Japan (e-mail: k-kimiyo@pref.gunma.lg.jp).

II. EXPERIMENTAL

A. Sample Collection and Chemical Analysis

The Tokyo Metropolitan Area, covering a region with a radius of 70 km from the center of Tokyo, is characterized by a large population (more than 34 million people, corresponding to 30% of the total population of Japan) and heavy traffic (18 million vehicles corresponding to 20% of the total number of vehicles in Japan). Massive industrial complexes, including facilities operated by the steel and petrochemical industries, are located in Tokyo Bay.

A PM monitoring campaign was conducted between 1989 and 2007. Sample collection was carried out simultaneously at multiple sites (17 sites in 2007 (Fig. 1)) every summer and winter. PM samples categorized according to size ($PM_{2.1}$ and $PM_{2.1-11}$) were collected for 1 week by using an Andersen-type low-volume air sampler.
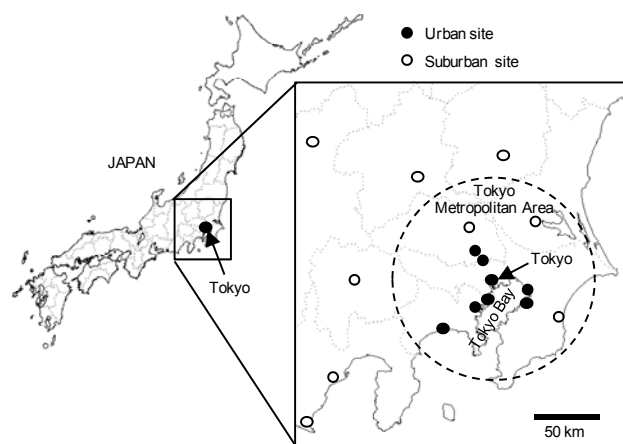

Fig. 1 Locations of the observation sites

Carbon components (organic carbon (OC) and total carbon (TC)) were analyzed by using a thermal carbon analyzer. OC was separated under at 600 °C in a helium atmosphere, and total carbon (TC) was separated at 950 °C in a 2% oxygen/98% helium atmosphere. Elemental carbon (EC) was defined as the amount of OC subtracted from the amount of TC.

Ion species (Na^+, Mg^{2+}, K^+, Ca^{2+}, NH_4^+, Cl^-, NO_3^-, and SO_4^{2-}) were analyzed by using ion chromatography. A portion of a sample-loaded filter was immersed into 20 mL of ultrapure water and sonicated for 15 min in an ultrasonic bath, and the resulting solution was filtered through a PTFE membrane filter.

The presence of various elements (V, Al, Mn, Cl, Na, Ca, Br, Ti, Mg, and Cu) was determined by instrumental neutron

World Academy of Science, Engineering and Technology
International Journal of Environmental and Ecological Engineering
Vol:6, No:12, 2012

activation analysis. A portion of a sample-loaded filter was placed in a polyethylene bag and irradiated with thermal neutrons for 40 s, after which the gamma-ray spectra of short-lived radioisotopes were analyzed.

The presence of polycyclic aromatic hydrocarbons (PAHs: B[k]F, B[a]P, and B[ghi]P) was determined by using high-performance liquid chromatography. A portion of a sample-loaded filter was immersed into 5 mL of dichloromethane and sonicated for 15 min in an ultrasonic bath. The resulting solution was filtered by using a PTFE membrane filter, and the filtrate was subsequently concentrated by blowing N_2 and dissolved in 1.5 mL of acetonitrile.

B. Principle of PMF Model

The PMF model can extract several factors from observational datasets according to similarities in their temporal or spatial characteristics [5]. The model essentially assumes that the number of factors is p, in other words, several sources impacting a receptor, and that the linear combination of the impacts from these pp factors gives rise to the observed concentrations of PM components [6]. The model can be expressed as in (1).

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \tag{1}$$

where $i = 1, \dots, n$ is the number of samples; $j = 1, \dots, m$ is the number of species; and $k = 1, \dots, p$ is the number of factors. Furethermore, x_{ij} is the observed concentration for the j th element in the i th sample, g_{ik} is the contribution of the k th factor to the observed concentration in the i th sample, f_{kj} is the chemical profile of the k th factor for the j th species, and e_{ij} is the residual for the j th species in the i th sample. In PMF, the objective is to estimate the contributions g_{ik} and chemical profiles f_{kj}. The requirement of PMF is to minimize the objective function Q, which is expressed as in (2).

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{u_{ij}} \right)^2 = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right)^2 \tag{2}$$

where u_{ij} is the uncertainty with respect to the j th species in the i th sample. The uncertainties for the respective observational data were calculated by using (3) in the case of $x_{ij} \leq MDL_j$ and (4) in the case of $x_{ij} > MDL_j$ [7].

$$u_{ij} = \frac{5}{6} \times MDL_j \tag{3}$$

$$u_{ij} = \sqrt{\left(Error\ Frac._j \times x_{ij} \right)^2 + MDL_j^2} \tag{4}$$

where MDL_j is the method detection limit in the j th species and $Error\ Frac._j$ is the percentage of uncertainty for the analytical value of the j th species. In this study, $Error\ Frac._j$ was set to 10%. Moreover, the uncertainties for each species were appropriately corrected according to the signal-to-noise (S/N) ratio, which is expressed as in (5) [8].

$$\left(\frac{S}{N} \right)_j = \sqrt{\frac{\sum_{i=1}^{n} \left(x_{ij} - u_{ij} \right)^2}{\sum_{i=1}^{n} u_{ij}^2}} \tag{5}$$

In order to minimize the impact on the PMF solution, the uncertainties for species with an S/N ratio of less than 2 were corrected to 3-fold of original values (set to "weak" in the model). The mathematical algorithm was processed with the EPA-PMF 3.0 software.

III. RESULTS AND DISCUSSION

A. Dataset for PMF Modeling

To create the dataset for PMF modeling, we selected 16 species (OC, EC, Na^+, Mg^{2+}, K^+, Ca^{2+}, NH_4^+, Cl^-, NO_3^-, SO_4^{2-}, Al, V, Mn, B[k]F, B[a]P, B[ghi]P), whose concentrations were continuously monitored during the 19-year period of the monitoring campaign. The dataset (x_{ij} matrix) includes approximately 20,000 entries of observation data. Table I summarizes the statistics of the dataset for PMF modeling.

TABLE I
METHOD DETECTION LIMIT (MDL), MEAN, MAXIMUM AND MINIMUM
VALUES OF THE ANALYZED SPECIES, AND NUMBER OF SAMPLES (N=1230)
WITH CONCENTRATION BELOW THE MDL

Species	Unit	MDL	Concentration			# Samples
			Mean	Max.	Min.	<MDL
OC	$\mu g/m^3$	0.10	3.93	22.3	0.12	0
EC	$\mu g/m^3$	0.05	2.62	14.8	0.07	6
Na^+	$\mu g/m^3$	0.01	0.44	7.2	0.02	5
Mg^{2+}	$\mu g/m^3$	0.005	0.06	0.83	0.01	49
K^+	$\mu g/m^3$	0.005	0.14	0.99	0.01	3
Ca^{2+}	$\mu g/m^3$	0.01	0.33	2.87	0.02	22
NH_4^+	$\mu g/m^3$	0.005	1.23	9.11	0.01	26
Cl^-	$\mu g/m^3$	0.005	0.96	13.6	0.01	9
NO_3^-	$\mu g/m^3$	0.01	1.37	10.6	0.02	3
SO_4^{2-}	$\mu g/m^3$	0.03	2.54	25.8	0.06	3
Al	$\mu g/m^3$	0.005	0.28	2.84	0.01	12
V	ng/m^3	0.5	3.53	35.0	1.0	84
Mn	ng/m^3	5.0	18.3	160	10.0	269
B[k]F	ng/m^3	0.005	0.28	4.18	0.01	81
B[a]P	ng/m^3	0.005	0.34	5.37	0.01	64
B[ghi]P	ng/m^3	0.005	0.65	13.2	0.01	106

B. Temporal Trends for $PM_{2.1}$ and EC

Fig. 2 shows the temporal trends in averaged mass concentrations of $PM_{2.1}$ and EC for all sites in the winter campaign. The concentrations of $PM_{2.1}$ and EC show clear declining trends during the last two decades, and a similar temporal trend can be seen in the cases of OC, NO_3^-, Cl^-, and PAHs. On the other hand, there are no notable changes in the concentrations of the other species. This study focuses on the reasons for this remarkable reduction in $PM_{2.1}$ concentration in winter.
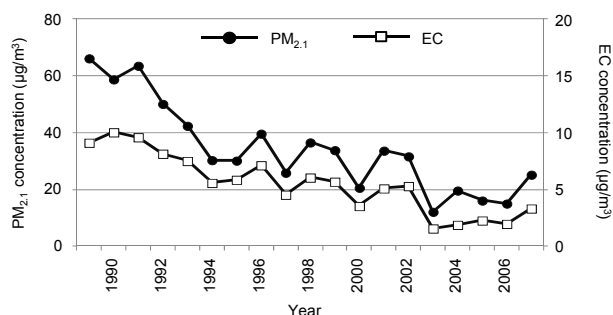
World Academy of Science, Engineering and Technology
International Journal of Environmental and Ecological Engineering
Vol:6, No:12, 2012

Fig. 2 Temporal trends in mass concentrations of $PM_{2.1}$ and EC in winter campaign

C. Identification of $PM_{2.1}$ Sources and Trends in the Contribution from Automotive Exhaust Products

Preliminary cluster analysis has suggested that between 5 and 7 factors might be extractable from the dataset. We extracted 6 base factors on the basis of the following criteria.

· Ensuring conformance of Q to the theoretical value.
· Maintaining the ion balance in the extracted factors.
· Reproducing the concentrations of 16 species.

Significant positive correlation ($r = 0.98$, $p < 0.01$) was found between the observations and calculated values for 6 base factors. A bootstrap run confirmed the stability of the base run solutions. Moreover, no significant rotation freedom was found in the base run solutions. Consequently, we adopted these 6 base factors as final solutions.

Fig. 3 shows the factor profiles as a distribution of chemical species across these 6 factors. By focusing on the indicative species, the potential sources of the respective factors were interpreted as follows.

Factor 1 includes NH_4^+ and SO_4^{2-}, representing the impact of secondary sulfates (ammonium sulfate). In addition, OC, EC, K^+, and V, which are mainly emitted from combustion sources, can also be associated with this factor. Consequently, we interpret Factor 1 as secondary sulfates, including the impact of combustion sources as the origin of precursors of secondary sulfates.

Factor 2 includes OC, EC, and PAHs. These are species indicative of combustion sources.

Factor 3 includes NH_4^+, Cl^-, and NO_3^-, representing the impacts of secondary chlorides (ammonium chloride) and secondary nitrates (ammonium nitrate).

Factor 4 includes Ca^{2+} and Al, which are emitted mainly as a result of weathering of geological media. In this study, we interpret Factor 4 as road dust.

Factor 5 includes Na^+, Mg^{2+}, and NO_3^- as indicative species. Sodium nitrate or magnesium nitrate can be formed by a heterogeneous reaction between sea salt particles and HNO_3 gas. Therefore, Factor 5 represents aged sea salt.

Factor 6 includes Na^+, Mg^{2+}, and Cl^-, representing the impact of fresh sea salt.

Factors 1, 2, and 3 contribute strongly to the $PM_{2.1}$ fraction, suggesting that these factors originate mainly from anthropogenic sources, whereas Factors 4, 5, and 6 are typically associated with the $PM_{2.1-11}$ fraction, indicating that they

originate from natural sources. These features are consistent with the above interpretations.

Fig. 4 shows the variation in averaged factor contribution to the $PM_{2.1}$ concentration in the winter campaign. As mentioned above, the averaged mass concentration of $PM_{2.1}$ has shown a trend toward decrease over the last two decades, and Factor 2 contributes notably to this trend. As shown in Fig. 3, Factor 2 includes OC, EC, and PAHs as indicative species, which are mainly emitted from combustion sources such as internal combustion engines, biomass burning, and oil burning. There is a fair positive correlation ($r = 0.90$, $p < 0.01$) between the EC concentration as calculated from Factor 2 and the observed EC concentration. Therefore, the reduction in EC concentration can be attributed to the reduction of PM emissions from combustion sources.
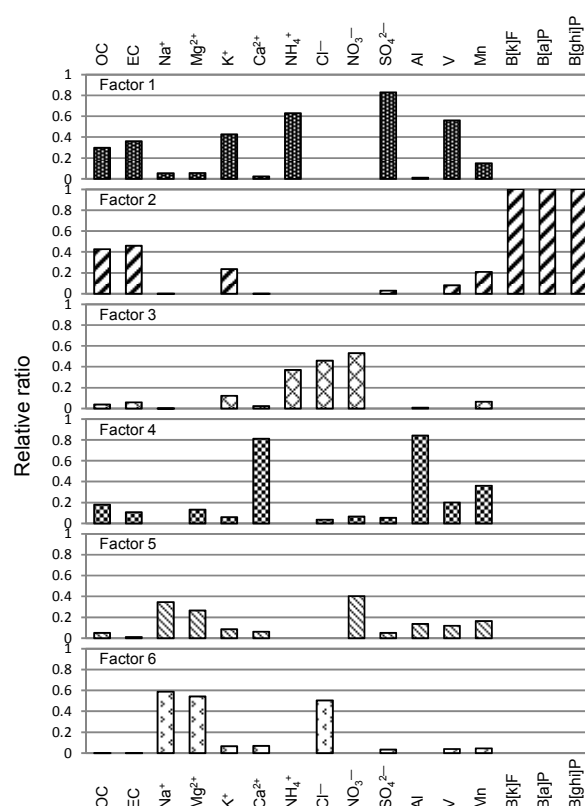

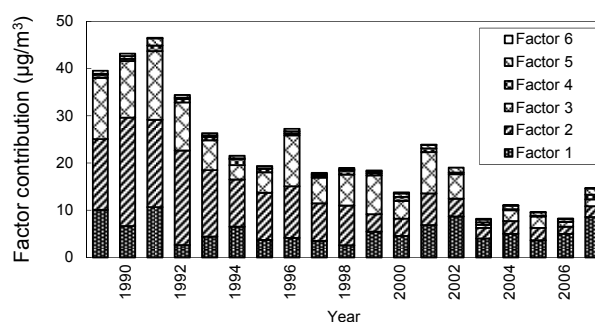Fig. 3 Distribution of chemical species across 6 factors

Fig. 4 Temporal trends in averaged factor contributions to $PM_{2.1}$ in winter campaign

World Academy of Science, Engineering and Technology
International Journal of Environmental and Ecological Engineering
Vol:6, No:12, 2012

Focusing on the recently implemented regulations on vehicle emissions, as shown in Fig. 5, regulations on vehicle exhaust emissions have been phased in and gradually tightened over the last two decades. With respect to the regulations on new cars, the Japanese government introduced new regulations on emission standards for NO_x and PM in 1994, 1998, 2003, and 2005 (corresponding to (A), (B), (D), and (E) in Fig. 5, respectively). These regulations resulted in the reduction of the emission of air pollutants from vehicles, particularly from diesel-powered vehicles. This drastic change was most likely due to the Law Concerning Special Measures for Total Emission Reduction of Nitrogen Oxides and Particulate Matter (corresponding to (C) in Fig. 5), which was enforced in July 2001. In accordance with national legislation aimed at curbing NO_x and PM emissions in major metropolitan areas, inspection compliance cannot be issued for vehicles that fail to meet the legal standards at inspection time. The enforcement of these regulations means that restrictions are imposed on diesel-powered vehicle circulation in the areas concerned.
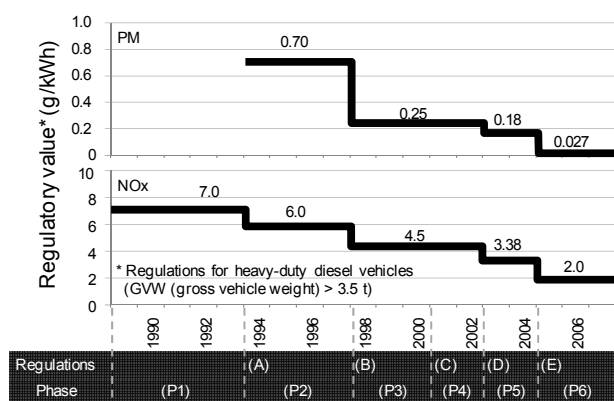

Fig. 6 Contribution of Factor 2 to $PM_{2.1}$ concentration in different regulation phases

IV. Conclusion

Regulations on vehicle emission have contributed to a steady reduction in fine PM pollution over the last two decades in Japan. Nevertheless, the current fine PM concentration remains at level which is higher than that defined in the Japanese air quality standards for $PM_{2.5}$. Therefore, in addition to primary sources of PM, close attention must be paid to secondary PM sources as well.

Fig. 5 History of impact of the tightening of regulatory values on NO_x and PM concentrations

The following regulations were introduced: (A) Short-term automobile exhaust gas regulations (1994), (B) Long-term automobile exhaust gas regulations (1998), (C) Law Concerning Special Measure for Total Emission Reduction of Nitrogen Oxides and Particulate Matter (2001), (D) New short-term automobile exhaust gas regulations (2003), and (E) New long-term automobile exhaust gas regulations (2005). The monitoring campaign was divided into 6 intervals ((P1)–(P6)) associated with the tightening of the regulations.

To estimate the effects of these regulations, we divided the 19-year period of the monitoring campaign into 6 intervals associated with the tightening of regulations (corresponding to (P1)–(P6) in Fig. 5), and Fig. 6 shows the contribution of Factor 2 to $PM_{2.1}$ concentration in the respective intervals. During the first decade of the campaign, the contribution of Factor 2 to $PM_{2.1}$ concentration was greater than 30%; however, phased tightening of the regulations on vehicle emissions has clearly triggered a notable reduction in the contribution from Factor 2, which is currently approximately 10%. In conclusion, the tightening of the regulations on vehicle emissions has decidedly contributed to the mitigation of the fine PM pollution in the center and suburbs of the Tokyo Metropolitan Area.
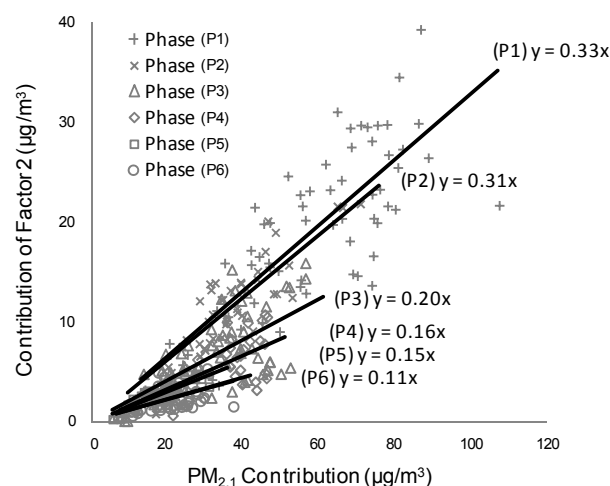
References

[1] Ministry of the Environment, Japan, *Annual Report on the Environment, the Sound Material-Cycle Society and the Biodiversity in Japan 2012*, Chiyoda-ku, Tokyo: Ministry of the Environment, Japan, 2012.

[2] D. W. Dockery, C. A. Pope, X. Xu, J. D. Spengler, J. H. Ware, M. E. Fay, G. G. Ferris, and F. E. Speizer, "An association between air pollution and mortality in six U.S. cities," *N. Engl. J. Med.*, vol. 329, pp. 1753–1759, 1993.

[3] S. Wakamatsu, "Overseas trends of $PM_{2.5}$ and future challenges in measurement, monitoring and componential analysis," *J. Jpn. Soc. Atmos. Environ.*, vol. 46, pp. 77–83.

[4] A. Iijima, G. Shimizu, and K. Kozawa, "Characterization of atmospheric aerosols in Kanto air basin," in *Proc. 50th Annu. Meeting of Jpn. Soc. Atmos. Environ.*, Yokohama, 2009, p. 466.

[5] A. Iijima, H. Tago, K. Kumagai, M. Kato, K. Kozawa, K. Sato, and N. Furuta, "Regional and seasonal characteristics of emission sources of fine airborne particulate matter collected in the center and suburbs of Tokyo, Japan as determined by multielement analysis and source receptor models," *J. Environ. Monit.*, vol. 10, pp. 1025–1032, 2008.

[6] P. Paatero and U. Tapper, "Positive matrix factorization: a non-negative factor model with optimal utilization of error estimates of data values," *Environmetrics*, vol. 5, pp. 111–126, 1994.

[7] A. V. Polissar, P. K. Hopke, P. Paatero, W. C. Malm, and J. F. Sisler, "Atmospheric aerosol over Alaska 2: Elemental composition and sources," *J. Geophys. Res.*, vol. 103, pp. 19045–19057.

[8] P. Paatero and P. H. Hopke, "Discarding of downweighting high-noise variables in factor analytic models," *Anal. Chim. Acta*, vol. 490, pp. 227–289.