Changes in black carbon and PM_{2.5} in Tokyo in 2003–2017

By Tatsuhiro MORI,^{*1} Sho OHATA,^{*,*3} Yu MORINO,^{*4} Makoto KOIKE,^{*5}
Nobuhiro MOTOKI^{*5} and Yutaka KONDO^{*6,†}

(Edited by Atsuhiko NISHIDA, M.J.A.)

Abstract: Black carbon (BC) particles cause adverse health effects and contribute to the heating of the atmosphere by absorbing visible solar radiation. Efforts have been made to reduce BC emissions, especially in urban areas; however, long-term measurements of BC mass concentration (M_{BC}) are very limited in Japan. We report M_{BC} measurements conducted in Tokyo from 2003 to 2017, showing that M_{BC} decreased by a factor of 3 from 2003 to 2010 and was stable from 2010 to 2017. Fine particulate concentrations (PM_{2.5}) decreased by a much smaller factor during 2003–2010. The diurnal variations of BC size distributions suggest that the BC in Tokyo originates mainly from local sources, even after 2010. Our three-dimensional model calculations show that BC from the Asian continent contributes a small portion (about 20%) of the annual average M_{BC} in the Kanto region of Japan, which includes Tokyo. This indicates that continued reduction of BC emissions inside Japan should be effective in further decreasing M_{BC} .

Keywords: black carbon, PM_{2.5}, PAHs, Tokyo, vehicular emission, Asia

1. Introduction

Air pollution is a major health risk linked to respiratory and cardiovascular diseases. Particulate matter (PM) is one of the most important elements of air pollution and is responsible for adverse health effects.^{1)–3)} Current air quality standards for PM are based on the concentrations of PM_{2.5} or PM₁₀ (PM with effective diameters of less than 2.5 μm or less than 10 μm , respectively).¹⁾ Black carbon (BC) aerosols (or “soot”), a constituent of PM, are emitted

from the incomplete combustion of fossil fuels and biomass.^{4),5)} Incomplete combustion also produces polycyclic aromatic hydrocarbons (PAHs),⁶⁾ and the molecular precursors of BC are considered to be heavy PAHs with molecular weights of 500–1,000 dalton.^{7)–11)} Thus, PAHs are co-emitted with BC and exist in gas and particle phases in the atmosphere.^{12),13)} Moreover, nitropolycyclic aromatic hydrocarbons (nitro-PAHs) are formed by the reaction between PAHs and NO₂ in the atmosphere. Many PAHs and nitro-PAHs are carcinogenic or mutagenic.¹¹⁾ They thus contribute to the adverse health effects caused by BC particles, including allergic, respiratory, and cancer-like diseases.^{14)–18)} Therefore, the reduction of PM_{2.5} emissions, which contain BC particles, is beneficial to human health.^{19)–21)}

BC particles also contribute to atmospheric warming by absorbing solar radiation.^{4),5),22)–24)} They also act as cloud condensation nuclei and can influence cloud albedo, partially offsetting the atmospheric heating related to the direct radiative forcing of BC.⁴⁾ To mitigate the contribution of BC to global warming, measures aimed at reducing BC emissions are most effectively directed at BC sources, such as emissions from diesel engines.

Asia was the world’s largest source of BC aerosols in 2000,^{25),26)} and vehicular emissions generated the majority of BC aerosols in urban

^{*1} Department of Physics, Faculty of Science Division I, Tokyo University of Science, Tokyo, Japan.

^{*2} Institute for Space–Earth Environmental Research, Nagoya University, Nagoya, Aichi, Japan.

^{*3} Institute for Advanced Research, Nagoya University, Nagoya, Aichi, Japan.

^{*4} National Institute for Environmental Studies, Tsukuba, Ibaraki, Japan.

^{*5} Department of Earth and Planetary Science, Graduate School of Science, The University of Tokyo, Tokyo, Japan.

^{*6} National Institute of Polar Research, Tachikawa, Tokyo, Japan.

[†] Correspondence should be addressed: Y. Kondo, National Institute of Polar Research, 10-3, Midori-cho, Tachikawa, Tokyo 190-8518, Japan (e-mail: kondo.yutaka@nipr.ac.jp).

Abbreviations: BC: black carbon; COSMOS: continuous soot monitoring system; EF_{BC}: emission flux of BC; M_{BC} : BC mass concentration; MMD: mass median diameter; PAHs: polycyclic aromatic hydrocarbons; PM: particulate matter; SP2: single particle soot photometer; TOT: thermal-optical transmittance.

areas.^{27)–29)} Emissions from the transport sector (on-road and off-road vehicles) constitute about 17% of the global BC burden; however, these estimates have large uncertainties.⁴⁾

Tokyo, with a population of about 14 million, represents a typical urban environment. Earlier studies have suggested that vehicular emissions cause the majority of the atmospheric aerosol loading in Tokyo.^{26),28),30),31)} PM in Tokyo is mainly emitted from diesel vehicles, and Japan started regulations for these emissions in 1994. Subsequent regulations have required new vehicles to reduce PM emissions,^{31),32)} and emission factors of PM from heavy-duty diesel vehicles have decreased greatly in the last 25 years.

An earlier study showed that ambient mass concentrations of BC (M_{BC}) in Tokyo decreased by a factor of about 3 between 2003 and 2010.³¹⁾ However, the changes in M_{BC} after 2010 have not been reported. It is also important to investigate the changes in M_{BC} relative to PM_{2.5} concentrations because the ratio of these two quantities is related to the single scattering albedo of aerosols; aerosols tend to cool the atmosphere as single scattering albedo increases. This study therefore estimated changes in BC/PM_{2.5} concentration ratios in addition to updating M_{BC} data for Tokyo.

In addition, the relative contribution of BC particles transported from the Asian continent may have changed because of recent regulations in different countries in East Asia. Therefore, it is important to investigate the effect of long-range transport when planning further reductions in BC emissions in Japan. In reporting updated data on Asian BC emissions for the period of 2010–2017 and integrating them with earlier results, this study also underlines the need to measure BC and PM_{2.5} with high accuracies on a long-term basis at key locations.

2. BC measurements

2.1. Observational sites. M_{BC} measurements were made at the Komaba II campus (the Research Center for Advanced Science and Technology campus) of the University of Tokyo (35.66°N, 139.68°E) between 2003 and 2010 and at the main Hongo campus of the University of Tokyo after 2014 (35.71°N, 139.76°E). These sites are within the area with the largest BC emissions in the Kanto region (Fig. 1) and are strongly influenced by vehicular emissions.^{28),30),31)} BC and carbon monoxide were strongly correlated in measurements made in 2004 at Komaba.²⁸⁾ Carbon monoxide concentrations, meas-

Fig. 1. BC sampling locations in Tokyo (35.5–35.9°N, 138.9–139.9°E). The gridded total BC emission rates in Kanto (35.0–37.0°N, 138.8–140.9°E), surrounded by dashed lines, for the year 2000 are also shown.³⁰⁾

ured at seven monitoring stations in Tokyo between 3 and 22 km from Komaba, were rather uniform and significantly correlated with M_{BC} .^{28),31)}

PM_{2.5} concentrations were measured at 40 roadside stations in central and suburban Tokyo and at 48 non-roadside stations in central and suburban Tokyo during 2000–2012.³²⁾ These stations were classified into four categories: roadside stations in central Tokyo, roadside stations in suburban Tokyo, non-roadside stations in central Tokyo, and non-roadside stations in suburban Tokyo. Komaba and Hongo are non-roadside stations in central Tokyo.

2.2. Measurement techniques. M_{BC} was measured at Komaba during 2003–2005 and 2006–2010 (Table 1). During 2003–2005, we used a semi-continuous elemental carbon-organic carbon analyzer (model RT3052, Sunset Lab., OR, U.S.A.) based on the thermal-optical transmittance (TOT) method with a time resolution of 1 h.^{28),33)} The overall accuracy of M_{BC} measurement using this method was estimated to be 22% based on the uncertainties in the sensitivity calibration, aerosol sampling, and temperature protocol used in the present study.^{28),34)}

From 2006 to 2010, we used the continuous soot monitoring system (COSMOS), which is based on a filter-based absorption photometer with a heated inlet.^{34)–37)} The accuracy of M_{BC} measurements by COSMOS was estimated to be about 10% based on comparisons with measurements by a single particle soot photometer (SP2) ($r^2 = 0.97$).^{34),36)–38)} The M_{BC} measurements by COSMOS and TOT instruments

Table 1. Mean and median values of BC mass concentrations (M_{BC})

Period	Observation sites	Observation days	Measurement	Number of data	M_{BC} ($\mu\text{g m}^{-3}$)	
					Mean ($\pm 1\sigma$)	Median (25%, 75%)
May 2003–Aug 2005	Komaba	308	TOT	7,386 (hourly data)	2.30 ± 0.45	2.29 (2.05, 2.56)
Dec 2006–Dec 2010	Komaba	540	COSMOS	12,965 (hourly data)	1.12 ± 0.48	1.00 (0.88, 1.23)
Feb 2014	Hongo	16	SP2	21,365 (1-min data)	0.71 ± 1.04	0.35 (0.18, 0.69)
Jul–Aug 2014	Hongo	20	SP2	26,775 (1-min data)	0.41 ± 0.31	0.31 (0.20, 0.56)
May 2016	Hongo	7	COSMOS	8,765 (1-min data)	0.94 ± 0.86	0.61 (0.28, 1.53)
Sep 2016	Hongo	8	COSMOS	11,380 (1-min data)	1.10 ± 1.41	0.79 (0.37, 1.27)
May 2017	Hongo	24	COSMOS	30,290 (1-min data)	0.70 ± 0.83	0.54 (0.33, 0.81)

agreed to within about 10% ($r^2 = 0.92$) at various sites in Asia.³⁴⁾

After 2014, we measured M_{BC} at Hongo via SP2 and COSMOS during intensive measurement campaigns that lasted 1–4 weeks (Table 1). Because M_{BC} measurements by TOT, COSMOS, and SP2 agree to within $\sim 10\%$, in this paper we use the M_{BC} data without reference to the technique used.

PM_{2.5} concentrations were based on the measured mass of aerosols collected using filters.³²⁾

3. Temporal variations

3.1. BC and PM_{2.5} during 2003–2017. The annual average PM_{2.5} concentrations at the stations in the four categories agreed to within about 10% (Fig. 2b). From this, we conclude that PM_{2.5} data in central Tokyo are representative of the whole Tokyo metropolitan area, consistent with the uniform distribution of carbon monoxide concentration and its strong correlation with M_{BC} (section 2.1). It is therefore likely that all aerosol measurements at Komaba and Hongo are representative of the Tokyo metropolitan area.

Measurements at Komaba show that M_{BC} decreased by a factor of 3 during 2003–2010, with the rate of change apparently slowing in 2009 (Fig. 2a). Similarly, the annual average PM_{2.5} concentrations for each of the four station categories decreased by about 35% from 2003 to 2010 (Fig. 2b). Our data from Hongo during 2014–2017 were rather stable and similar to M_{BC} observations at Komaba in 2010. We infer that M_{BC} did not decrease significantly after 2010. These reductions in BC and PM_{2.5} are probably due mainly to the regulation of emissions from diesel vehicles.³²⁾

3.2. Diurnal BC variations. Diurnal BC variations are useful for analyzing the effects of local BC emissions on M_{BC} . Figures 3 and 4 show the

diurnal variations of M_{BC} and the size distribution of BC measured by SP2 in July and August 2014. M_{BC} increased in the morning, which is likely due to emissions from diesel vehicles.^{28),31)} The average number and mass size distributions shifted to smaller diameters with this increase in M_{BC} (Fig. 4). The mass median diameter (MMD) reached its minimum when M_{BC} reached its maximum in the morning (Fig. 3). Sizes of freshly emitted BC particles in urban areas are generally smaller than those of aged BC particles because they are less influenced by coagulation. Similarly, BC particles observed downwind from the Asian continent are considerably larger than those emitted from Japan.^{39),40)} It is thus likely that locally emitted BC influenced M_{BC} in Tokyo strongly even in 2014, when M_{BC} had decreased by a factor of 3 since 2003.

In 2014, M_{BC} reached its daily maximum value around 10:00 local time (Fig. 3a), whereas the maximum occurred around 07:00 local time during 2003–2005.²⁸⁾ These diurnal trends are further evidence that BC emissions from diesel vehicles had decreased as a proportion of the total BC emissions.

3.3. BC/PM_{2.5} ratio. BC and PM_{2.5} concentrations at non-roadside stations in central Tokyo decreased by 70% and 35%, respectively, during 2003–2010. This resulted in a decrease in the BC/PM_{2.5} ratio by a factor of 2.3 (Fig. 2c). Kondo *et al.* (2010)⁴¹⁾ showed that light-scattering particles, which include organic and inorganic aerosols, constituted about 80–90% of aerosol mass concentrations for PM with diameters less than 1 μm (PM₁) at Komaba in 2003. PM₁, in turn, is the dominant contributor to scattering coefficients for visible radiation in urban atmospheres.⁴²⁾ In this study, we used the BC/PM_{2.5} ratio as a qualitative measure of aerosol optical properties. BC/(PM_{2.5} – BC) ratios were similar to the BC/PM_{2.5} ratios (Fig. 2c).

Fig. 2. Temporal variations of (a) BC mass concentration (M_{BC}) observed at Komaba (black circles) and Hongo (black squares), (b) PM_{2.5} concentrations for four station categories (roadside and non-roadside stations in central Tokyo and suburbs of Tokyo), reported by Wakamatsu *et al.* (2013),³²⁾ and (c) BC/PM_{2.5} and BC/(PM_{2.5} - BC) ratios at non-roadside stations in central Tokyo. The error bars in (a) represent the standard deviation of the annual means at Komaba and the monthly means at Hongo.

Reducing the BC/PM_{2.5} ratio leads to a corresponding increase in single scattering albedo, which contributes to net radiative cooling by aerosols. Quantitative estimates of these effects require radiative transfer calculations, which are beyond the scope of this study.

Murphy *et al.* (2011)⁴³⁾ reported reductions of more than 25% in BC and PM_{2.5} at stations in national parks and other remote regions in the United States during 1990–2004. Despite the decrease in BC, these changes were estimated to have

caused warming in the United States because the rate of decrease in BC was comparable to that for light-scattering particles during the period.

4. Emission of BC in Tokyo and Japan

A previous study³¹⁾ estimated the emission flux of BC (EF_{BC}) in Tokyo for the period 2000–2011 on the basis of EF_{BC} determined in the Kanto region for the year 2000.³⁰⁾ The annual EF_{BC} was derived from the annual number of vehicles for each category (cars, buses, medium- and heavy-duty diesel trucks, and light-duty diesel trucks) for which statistical data were reported by the Ministry of Environment, Japan.⁴⁴⁾ The use of diesel particulate filters and low sulfur fuels beginning in 2003 has led to large reductions in EF_{BC} ,⁴⁴⁾ which accounted for the decrease in total EF_{BC} from all sources during this period.³¹⁾ The estimated decrease in total EF_{BC} is consistent with the observed changes in M_{BC} reported here (Fig. 2a); however, the study found that EF_{BC} from sources other than on-road vehicles did not substantially change in 2000–2011.³¹⁾

Estimated EF_{BC} values for all of Japan and the Kanto region decreased by factors of 3.0 and 2.8, respectively, from 2003 to 2015 (Fig. 5).⁴⁵⁾ The change in estimated EF_{BC} in Kanto agrees well with the change in observed M_{BC} in Tokyo. Although the EF_{BC} from transport in Kanto (including diesel vehicles) decreased greatly during 2003–2015, the EF_{BC} from industry slightly increased after 2011, which slowed the rate of decrease in total BC emissions after 2011.

In a study that measured concentrations of nine PAHs in the total PM in Tokyo, and in Kanagawa Prefecture (in the Kanto region), from 1997 to 2014, the total concentration of all nine PAHs (ΣPAH) in winter decreased by a factor of about 3.6 from 1997 to 2005 and then remained steady from 2008 to 2014.⁴⁶⁾ The initial large decrease in ΣPAH was interpreted to result of a corresponding decrease in PAH emissions from vehicles.⁴⁶⁾ The similarity in the temporal changes in ΣPAH and M_{BC} in Tokyo is consistent with the similarities of their sources, as discussed in section 1.

5. Transport of BC from the Asian continent

Morino *et al.* (2017)⁴⁷⁾ calculated the distribution of aerosols, including BC, in East Asia for the year 2012 using the three-dimensional chemistry and transport model of the Models-3 Community Multi-scale Air Quality modeling system. They used EF_{BC} data from the Auto-Oil Program^{48),49)} for anthropo-

Fig. 3. Average diurnal variations of (a) M_{BC} and (b) mass median diameter (MMD) measured by SP2 at Hongo in July and August 2014.

Fig. 4. Hourly mean size distributions of BC by (a) number and (b) mass measured by SP2 at Hongo in July and August 2014.

Fig. 5. Temporal variations of total BC emissions (dots) and BC emissions from different sectors in (a) Japan and (b) Kanto during 2003–2015. Data from Kurokawa *et al.* (2018).⁴⁵⁾

genic sources in Japan (~ 1 and 10 km resolutions for vehicles and other sources, respectively), from the Regional Emission Inventory in Asia⁵⁰⁾ for anthropogenic sources of other Asian countries (0.25°

resolution), and from the Global Fire Emission Database for biomass burning (0.5° resolution).^{51),52)} The effects of possible localized sources in Tokyo that were not included in these estimates should not

Fig. 6. Contributions of BC emitted in Japan and Asia to M_{BC} for (a) all of Japan and (b) Kanto in winter, spring, and summer 2012. Data from Morino *et al.* (2017).⁴⁷⁾

influence the present results considering the spatial uniformity of observed PM_{2.5} concentrations in Tokyo. Their calculated average M_{BC} values for winter, spring, and summer for all of Japan and for Kanto are shown in Fig. 6, along with the contributions from emission sources in Asia and Japan.

The M_{BC} value of about $0.4 \mu\text{g m}^{-3}$ for Kanto in 2012 (Fig. 6) is about half that observed at Komaba and Hongo during 2010–2017 (Fig. 2a). BC emitted within Japan contributed about 60% of the average M_{BC} in Japan and about 80% of the average M_{BC} in Kanto, which has large local as well as internal sources of BC (Fig. 6). This analysis shows that M_{BC} in Kanto can be further decreased by continuing to reduce BC emissions in Japan as well as Kanto as long as BC emissions in East Asia do not increase enough to offset those reductions. To further reduce domestic BC emissions, it will be necessary to reduce emissions from other sources besides on-road vehicles. To improve estimates of these contributions, further analyses using the latest emission datasets with higher spatial resolutions are needed.

6. Conclusions

Our measurements near the urban center of Tokyo show that M_{BC} decreased by a factor of 3 during 2003–2010 and remained steady during 2011–2017, whereas annual average PM_{2.5} concentrations in Tokyo decreased by only 35% during 2003–2010. As a result, BC/PM_{2.5} ratios decreased by a factor of 2.3, leading to corresponding increases in single scattering albedo and the cooling effect of aerosols. The diurnal variations of M_{BC} and the size distribu-

tion of BC in 2014 suggest the significant contribution from BC emitted within Tokyo. The stability of M_{BC} during 2010–2017 indicates that EF_{BC} from sources other than vehicular emissions in Tokyo did not decrease substantially. The pattern of the temporal changes in M_{BC} was similar to that of ΣPAH in Tokyo, consistent with the similarities of their sources. Three-dimensional modeling of M_{BC} values in Kanto, the region that includes Tokyo, showed that the contribution of domestic BC emissions to M_{BC} was about 80% in Kanto as compared with about 60% for all of Japan. These results indicate that M_{BC} in Tokyo can be further lowered by reducing emissions of BC inside Japan as well as in Kanto.

Acknowledgements

We thank Jun-ichi Kurokawa of the Asia Center for Air Pollution Research for providing the BC emission rate data from different sectors in Japan. We also thank Kazuichi Hayakawa of Kanazawa University for providing the PAHs data for Kanto. This work was supported by the Japanese Ministry of Education, Culture, Sports, Science, and Technology (MEXT), the Environment Research and Technology Development Fund (2-1403 and 2-1703) of the Environmental Restoration and Conservation Agency of Japan, Japan Society for the Promotion of Science KAKENHI Grant (JP16H01770), and the Arctic Challenge for Sustainability project. The data used in this study are available at URL (<https://ads.nipr.ac.jp/dataset/A20191212-002>).

References

- 1) European Environment Agency (2018) Air quality in Europe–2018 report. EEA Rept. No. 12/2018. <https://www.eea.europa.eu/publications/air-quality-in-europe-2018>.
- 2) Lelieveld, J., Klingmüller, K., Pozzer, A., Burnett, R.T., Haines, A. and Ramanathan, V. (2019) Effects of fossil fuel and total anthropogenic emission removal on public health and climate. *Proc. Natl. Acad. Sci. U.S.A.* **116**, 7192–7197.
- 3) Lelieveld, J., Klingmüller, K., Pozzer, A., Pöschl, U., Fnais, M., Daiber, A. *et al.* (2019) Cardiovascular disease burden from ambient air pollution in Europe reassessed using novel hazard ratio functions. *Eur. Heart J.* **40**, 1590–1596.
- 4) Bond, T.C., Doherty, S.J., Fahey, D.W., Forster, P.M., Berntsen, T., DeAngelo, B.J. *et al.* (2013) Bounding the role of black carbon in the climate system: A scientific assessment. *J. Geophys. Res. Atmos.* **118**, 1–173.
- 5) Kondo, Y. (2015) Effects of black carbon on climate: Advances in measurement and modeling. *Monogr. Environ. Earth Planets* **3**, 1–85.
- 6) Khalili, N., Scheff, P.A. and Holsen, T.M. (1995) PAH source fingerprints for coke ovens, diesel and gasoline engines, highway tunnels, and wood combustion emissions. *Atmos. Environ.* **29**, 533–542.
- 7) Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R. and Simoneit, B.R.T. (1993) Sources of fine organic aerosol. 2. Nuncatalyst and catalyst-equipped automobiles and heavy-duty diesel trucks. *Environ. Sci. Technol.* **27**, 636–651.
- 8) Mansurov, Z.A. (2005) Soot formation in combustion processes (review). *Combust. Explos. Shock Waves* **41**, 727–744.
- 9) Xi, J. and Zhong, B.-J. (2006) Soot in diesel combustion systems. *Chem. Eng. Technol.* **29**, 665–673.
- 10) Fizpatrick, E.M., Jones, J.M., Pourkashanian, M., Ross, A.B., Williams, A. and Bartle, K.D. (2008) Mechanistic aspects of soot formation from the combustion of pine wood. *Energy Fuels* **22**, 3771–3778.
- 11) Hayakawa, K., Tang, N., Nagato, E.G., Toriba, A., Sakai, S., Kano, F. *et al.* (2018) Long term trends in atmospheric concentrations of polycyclic aromatic hydrocarbons and nitropolycyclic aromatic hydrocarbons: A study of Japanese cities from 1997 to 2014. *Environ. Pollut.* **233**, 474–482.
- 12) Chen, Y., Sheng, G., Bi, X., Feng, Y., Mai, B. and Fu, J. (2005) Emission factors for carbonaceous particles and polycyclic aromatic hydrocarbons from residential coal combustion in China. *Environ. Sci. Technol.* **39**, 1861–1867.
- 13) Araki, Y., Tang, N., Ohno, M., Kameda, T., Toriba, A. and Hayakawa, K. (2009) Analysis of atmospheric polycyclic aromatic hydrocarbons and nitropolycyclic aromatic hydrocarbons in gas/particle phases separately collected by a high-volume air sampler equipped with a column packed with XAD-4 resin. *J. Health Sci.* **55**, 77–85.
- 14) Samet, J.M., DeMarini, D.M. and Mallng, H.V. (2004) Do airborne particles induce heritable mutations? *Science* **304**, 971–972.
- 15) Somers, C.M., McCarry, B.E., Malek, F. and Quinn, J.S. (2004) Reduction of particulate air pollution lowers the risk of heritable mutations in mice. *Science* **304**, 1008–1010.
- 16) Jansen, K.L., Larson, T.V., Koenig, J.Q., Mar, T.F., Fields, C., Stewart, J. *et al.* (2005) Associations between health effects and particulate matter and black carbon in subjects with respiratory disease. *Environ. Health Perspect.* **113**, 1741–1746.
- 17) Nel, A. (2005) Air pollution–related illness: Effects of particles. *Science* **308**, 804–806.
- 18) Alessandrini, F., Schulz, H., Takenaka, S., Lentner, B., Karg, E., Behrendt, H. *et al.* (2006) Effects of ultrafine carbon particle inhalation on allergic inflammation of the lung. *J. Allergy Clin. Immunol.* **117**, 824–830.
- 19) Janssen, N.A.H., Hoek, G., Simic-Lawson, M., Fisher, P., van Bree, L., ten Brink, H. *et al.* (2011) Black carbon as an additional indicator of adverse health effects of airborne particles compared with PM₁₀ and PM_{2.5}. *Environ. Health Perspect.* **119**, 1691–1699.
- 20) Yorifuji, T., Kawachi, I., Kaneda, M., Takao, S., Kashima, S. and Doi, H. (2011) Diesel vehicle emission and death rates in Tokyo, Japan: A natural experiment. *Sci. Total Environ.* **409**, 3620–3627.
- 21) International Agency for Research on Cancer, World Health Organization (2013) IARC: Outdoor air pollution a leading environmental cause of cancer deaths. Press Release No. 221. https://www.iarc.fr/en/media-centre/pr/2013/pdfs/pr221_E.pdf (last accessed on April 26, 2017).
- 22) Ramanathan, V. and Carmichael, G. (2008) Global and regional climate changes due to black carbon. *Nat. Geosci.* **1**, 221–227.
- 23) Jacobson, M.Z. (2010) Short-term effects of controlling fossil-fuel soot, biofuel soot and gases, and methane on climate, Arctic ice, and air pollution health. *J. Geophys. Res. Atmos.* **115**, D14209.
- 24) Intergovernmental Panel on Climate Change (2013) The physical science basis, contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, U.K.
- 25) Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S.D., Fu, Q., He, D. *et al.* (2003) An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geophys. Res. Atmos.* **108**, 8809.
- 26) Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X. *et al.* (2007) An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. *Atmos. Chem. Phys.* **7**, 4419–4444.
- 27) Jiang, M., Marr, L.C., Dunlea, E.J., Herndon, S.C., Jayne, J.T., Kolb, C.E. *et al.* (2005) Vehicle fleet

- emissions of black carbon, polycyclic aromatic hydrocarbons, and other pollutants measured by a mobile laboratory in Mexico City. *Atmos. Chem. Phys.* **5**, 3377–3387.
- 28) Kondo, Y., Komazaki, Y., Miyazaki, Y., Moteki, N., Takegawa, N., Kodama, D. *et al.* (2006) Temporal variations of elemental carbon in Tokyo. *J. Geophys. Res. Atmos.* **111**, D12205.
 - 29) Han, S., Kondo, Y., Oshima, N., Takegawa, N., Miyazaki, Y., Hu, M. *et al.* (2009) Temporal variations of elemental carbon in Beijing. *J. Geophys. Res. Atmos.* **114**, D23202.
 - 30) Kannari, A., Tonooka, Y., Baba, T. and Murano, K. (2007) Development of multiple-species resolution hourly basis emissions inventory for Japan. *Atmos. Environ.* **41**, 3428–3439.
 - 31) Kondo, Y., Ram, K., Takegawa, N., Sahu, L., Morino, Y., Liu, X. *et al.* (2012) Reduction of black carbon aerosols in Tokyo: Comparison of real-time observations with emission estimates. *Atmos. Environ.* **54**, 242–249.
 - 32) Wakamatsu, S., Morikawa, T. and Ito, A. (2013) Air pollution trends in Japan between 1970 and 2012 and impact of urban air pollution countermeasures. *Asian J. Atmos.* **7**, 177–190.
 - 33) Birch, M.E. and Cary, R.A. (1996) Elemental carbon-based method for occupational monitoring of particulate diesel exhaust: Methodology and exposure issues. *Analyst* **121**, 1183–1190.
 - 34) Kondo, Y., Sahu, L., Moteki, N., Khan, F., Takegawa, N., Liu, X. *et al.* (2011) Consistency and traceability of black carbon measurements made by laser-induced incandescence, thermal-optical transmittance, and filter-based photo-absorption techniques. *Aerosol Sci. Technol.* **45**, 295–312.
 - 35) Miyazaki, Y., Kondo, Y., Sahu, L.K., Imaru, J., Fukushima, N. and Kano, M. (2008) Performance of a newly designed continuous soot monitoring system (COSMOS). *J. Environ. Monit.* **10**, 1109–1240.
 - 36) Kondo, Y., Sahu, L., Kuwata, M., Miyazaki, Y., Takegawa, N., Moteki, N. *et al.* (2009) Stabilization of the mass absorption cross section of black carbon for filter-based absorption photometry by the use of a heated inlet. *Aerosol Sci. Technol.* **43**, 741–756.
 - 37) Ohata, S., Kondo, Y., Moteki, N., Mori, T., Yoshida, A., Sinha, P.R. *et al.* (2019) Accuracy of black carbon measurements by a filter-based absorption photometer with a heated inlet. *Aerosol Sci. Technol.* **53**, 1079–1091.
 - 38) Moteki, N. and Kondo, Y. (2007) Effects of mixing state on black carbon measurements by laser-induced incandescence. *Aerosol Sci. Technol.* **41**, 398–417.
 - 39) Shiraiwa, M., Kondo, Y., Moteki, N., Takegawa, N., Sahu, L.K., Takami, A. *et al.* (2008) Radiative impact of mixing state of black carbon aerosol in Asian outflow. *J. Geophys. Res. Atmos.* **113**, D24210.
 - 40) Moteki, N., Mori, T., Matsui, H. and Ohata, S. (2019) Observational constraint of in-cloud supersaturation for simulations of aerosol rainout in atmospheric models. *npj Clim. Atmos. Sci.* **2**, 6.
 - 41) Kondo, Y., Takegawa, N., Matsui, H., Miyakawa, T., Koike, M., Miyazaki, Y. *et al.* (2010) Formation and transport of aerosols in Tokyo in relation to their physical and chemical properties: A review. *J. Meteorol. Soc. Jpn.* **88**, 597–624.
 - 42) Seinfeld, J.H. and Pandis, S.N. (2010) *Atmospheric Chemistry and Physics*. John Wiley & Sons, New York.
 - 43) Murphy, D.M., Chow, J.C., Leibensperger, E.M., Malm, W.C., Pitchford, M., Schichtel, B.A. *et al.* (2011) Decreases in elemental carbon and fine particle mass in the United States. *Atmos. Chem. Phys.* **11**, 4679–4686.
 - 44) Ministry of Environment (2008) Investigation on Intensity and Total Amount of Emission from Motor-Vehicle Exhaust. Research Report (207939), Tokyo (in Japanese).
 - 45) Kurokawa, J., Yumimoto, K., Itahashi, S., Nagashima, T., Maki, T. and Ohara, T. (2018) Historical trends of air pollutant emissions in Asia: Development of regional emission inventory in Asia (REAS) version 3. Abstract ID No. 1.071. 14th iCACGP Quadrennial Symposium/15th IGAC Science Conference, Takamatsu, Japan.
 - 46) Hayakawa, K. (2016) Environmental behaviors and toxicities of polycyclic aromatic hydrocarbons and nitropolycyclic aromatic hydrocarbons. *Chem. Pharm. Bull. (Tokyo)* **64**, 83–94.
 - 47) Morino, Y., Ueda, K., Takami, A., Nagashima, T., Tanabe, K., Sato, K. *et al.* (2017) Sensitivities of simulated source contributions and health impacts of PM_{2.5} to aerosol models. *Environ. Sci. Technol.* **51**, 14273–14282.
 - 48) Japan Auto-Oil Program (2012) Emission estimates from motor vehicles. JPEC-2011AQ-02-06, Tokyo, Japan (in Japanese).
 - 49) Japan Auto-Oil Program (2012) Emission estimates from sources other than motor vehicles. JPEC-2011AQ-02-07, Tokyo, Japan (in Japanese).
 - 50) Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T. *et al.* (2013) Emissions of air pollutants and greenhouse gases over Asian regions during 2000–2008: Regional emission inventory in Asia (REAS) version 2. *Atmos. Chem. Phys.* **13**, 11019–11058.
 - 51) Van der Werf, G.R., Randerson, J.T., Giglio, L., Collatz, G.J., Kasibhatla, P.S. and Arellano, A.F. (2006) Interannual variability in global biomass burning emissions from 1997 to 2004. *Atmos. Chem. Phys.* **6**, 3423–3441.
 - 52) Morino, Y., Nagashima, T., Sugata, S., Sato, K., Tanabe, K., Noguchi, T. *et al.* (2015) Verification of chemical transport models for PM_{2.5} chemical composition using simultaneous measurement data over Japan. *Aerosol Air Qual. Res.* **15**, 2009–2023.

(Received Sep. 22, 2019; accepted Jan. 16, 2020)