Changes in black carbon and PM$_{2.5}$ in Tokyo in 2003–2017

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**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 PM$_{10}$ (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 NO$_2$ 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.$^{(4,5,22)}$ 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...
areas. 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.

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. 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 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 (MB) in Tokyo decreased by a factor of about 3 between 2003 and 2010. However, the changes in MB after 2010 have not been reported. It is also important to investigate the changes in MB relative to PM2.5 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/PM2.5 concentration ratios in addition to updating MB 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 PM2.5 with high accuracies on a long-term basis at key locations.

2. BC measurements

2.1. Observational sites. MB 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. BC and carbon monoxide were strongly correlated in measurements made in 2004 at Komaba. Carbon monoxide concentrations, measured at seven monitoring stations in Tokyo between 3 and 22 km from Komaba, were rather uniform and significantly correlated with MB.

PM2.5 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. 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. MB 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. The overall accuracy of MB 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.

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. The accuracy of MB measurements by COSMOS was estimated to be about 10% based on comparisons with measurements by a single particle soot photometer (SP2). The MB measurements by COSMOS and TOT instruments
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)

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

| Period          | Observation sites | Observation days | Measurement | Number of data | $M_{BC}$ (µg m$^{-3}$) |
|-----------------|-------------------|------------------|-------------|----------------|------------------------|
| May 2005–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)) |

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. 2a). 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 ($PM_{1}$) at Komaba in 2003. $PM_{1}$, 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).
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)\textsuperscript{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 study\textsuperscript{31} 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.\textsuperscript{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.\textsuperscript{44} The use of diesel particulate filters and low sulfur fuels beginning in 2003 has led to large reductions in EF$_{BC}$\textsuperscript{44} which accounted for the decrease in total EF$_{BC}$ from all sources during this period.\textsuperscript{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.\textsuperscript{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). 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 ($\Sigma$PAH) in winter decreased by a factor of about 3.6 from 1997 to 2005 and then remained steady from 2008 to 2014.\textsuperscript{46} The initial large decrease in $\Sigma$PAH was interpreted to result of a corresponding decrease in PAH emissions from vehicles.\textsuperscript{46} The similarity in the temporal changes in $\Sigma$PAH 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)\textsuperscript{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\textsuperscript{48,49} for anthropo-
genic sources in Japan (~1 and 10 km resolutions for vehicles and other sources, respectively), from the Regional Emission Inventory in Asia\(^\text{50}\) for anthropogenic sources of other Asian countries (0.25° resolution), and from the Global Fire Emission Database for biomass burning (0.5° resolution).\(^\text{51,52}\)

The effects of possible localized sources in Tokyo that were not included in these estimates should not
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 µg 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 distribution 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.

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