



Research Article

Spatiotemporal Variations of Fine Particulate Organic and Elemental Carbons in Greater Tokyo

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Received: 19 March 2019

Revised: 24 May 2019

Accepted: 11 June 2019

ABSTRACT Hourly concentrations of fine particulate OC and EC are monitored for more than one year by optical monitors at three sites in and out of Tokyo, Japan. Distances between the sites are 20 to 50 km. SOC concentrations are estimated from the OC and EC concentrations by an EC tracer method. Site A in an industrial site shows higher EC concentrations than site B in a residential area, but differences between the sites are much reduced for OC. Site C in a rural area, where measurements are available in colder months, shows pronounced seasonal variations with high peaks in October and December and characteristic diurnal variations in OC with the highest in the evening in October, due probably to open field burning. Photochemical formation of SOC is suggested but does not rise up so much OC concentrations in summer, which are quite lower than other seasons. A comparison between relative concentrations of EC among the sites and local emissions amounts indicates missing or considerable underestimation of EC emissions near the rural site C. Time lag correlations suggest transport of OC produced by open field burning around site C to site A. These findings would be useful for improving model performances in OC/EC predictions.

KEY WORDS PM_{2.5}, OC, EC, APC-710, Hourly measurements

1. INTRODUCTION

Fine particulate matter (PM_{2.5}) in the atmosphere is highly concerned worldwide because of its adverse effects on human health. In Japan, air quality standards for PM_{2.5} were set in September 2009; 15 $\mu\text{g m}^{-3}$ for annual averages and 35 $\mu\text{g m}^{-3}$ for daily averages. Monitoring of PM_{2.5} mass concentrations begun at general ambient and roadside stations in whole Japan since April 2010, the beginning of Japanese fiscal year (FY) 2010. Before FY2010, PM_{2.5} concentrations had been measured by research institutes and national and local governmental initiatives. According to monitoring results in the 2000s, annual-mean concentrations of PM_{2.5} tended to gradually decrease, and a national-wide annual mean averaged over all general ambient air monitoring stations came down to 15.1 $\mu\text{g m}^{-3}$ in FY2010 (Itahashi and Hayami, 2018). The PM_{2.5} concentration after FY2010 moderately varied around 15 $\mu\text{g m}^{-3}$ but considerably decreased in a recent couple of years. In FY2016, the annual-mean PM_{2.5} concentration was 11.9 $\mu\text{g m}^{-3}$, and the air quality standards for daily- and annual-mean concentrations of PM_{2.5} were met at about

88% of all 1,008 monitoring stations (Ministry of the Environment, 2018). The recent decrease in $PM_{2.5}$ is mainly associated with decrease in trans-boundary air pollution from the Asian Continent resulted from emissions reductions of sulfur dioxide and nitrogen oxides in China (Uno *et al.*, 2017). $PM_{2.5}$ concentrations in Japan are often influenced by both trans-boundary transport and locally-produced pollution from sources inside Japan (Shimadera *et al.*, 2013). The reduction in the trans-boundary air pollution would relatively enlarge domestic contributions and lead to much more discussions about emission controls on domestic sources.

Air quality models are useful for understanding behaviors of pollutants, analyzing source apportionment, and predicting future air quality. However, current air quality models have difficulties in predicting $PM_{2.5}$ concentrations; overestimation of nitrate and underestimation of organic carbon (OC) and wintertime sulfate (Shimadera *et al.*, 2018). Among them, the present paper will focus on OC, which is very important because it dominates $PM_{2.5}$ mass by 23% in Japan (Ministry of the Environment, 2018) and by 30% (as organic matter) in world megacities (Cheng *et al.*, 2016). Toward the underestimation of OC, many efforts have been devoted through emission modeling, chemical modeling, laboratory experiments, and field observations (e.g., Sato *et al.*, 2018; Morino *et al.*, 2015; Kumagai *et al.*, 2009). However, hourly measurements of OC are very limited, which are necessary for model validation, even though models output hourly concentrations. Until now, automatic monitors that can measure OC concentrations hourly have been commercially available, including Sunset Laboratory field OC/EC analyzers and Rupprecht and

Patashnick (R&P) series 5400 ambient carbon particulate monitors (Venkatachari *et al.*, 2006). However, annual datasets of hourly OC concentrations are still limited (Höller *et al.*, 2002).

This paper presents hourly OC and EC concentrations measured for more than one year at three sites in the Greater Tokyo Area. Our monitoring mainly aims at providing hourly measurements of OC/EC for validation of air quality models in a model intercomparison project named J-STREAM (Japan's STudy for REference Air quality Modeling) (Chatani *et al.*, 2018). Three monitoring sites are located in a coastal, industrialized area, a residential area and a rural area. Distances between the sites are 20 to 50 km, which corresponds to few to ten of horizontal grid cells (~ 5 km) used in typical urban-scale modeling. In the present paper, OC/EC measurements are mainly analyzed for general spatiotemporal patterns to encourage model evaluations for average behaviors. High-concentration episodes will be analyzed elsewhere.

2. EXPERIMENTS

Hourly OC and EC concentrations were monitored at three sites in Greater Tokyo; Koto (site A), Komae (site B) and Kazo (site C) (Fig. 1). Site A is on the roof (23.5 m AGL) of the building of the Tokyo Metropolitan Research Institute for Environmental Protection, which is very close to the coast of the Tokyo Bay with many large-point sources, heavy-traffic roads, airports and ports. Site B is on the ground in a residential area of Tokyo with no large factories and heavy traffic. Site C is on the roof of a one-story building (~ 4 m AGL) in an inland, rural area in Sai-

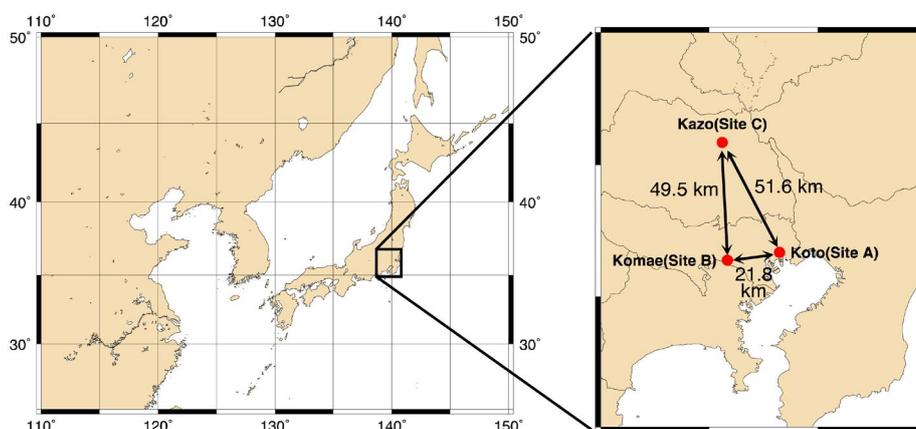


Fig. 1. Monitoring sites in this study.

tama Prefecture. Distances are 21.8 km between site A and site B, 51.6 km between site A and site C, and 49.5 km between site B and site C. Monitoring periods are from March 2016 to June 2017 at site A, from March 2016 to March 2017 at site B, and from October 2016 to June 2017 at site C. Measurements during a common period of April 2016 to March 2017, which is a Japanese fiscal year of 2016, are mainly analyzed in this paper.

An automatic continuous particulate carbon monitor, APC-710 (Kimoto Electric Co., Ltd.), was used at each of the sites. The APC-710 collects PM_{2.5} on a PTFE tape-filter that rolls up once an hour and detects optical transmittance and reflectance at wavelengths of 375 nm and 890 nm from each sampled spot on the filter. Measurements of the APC-710 were assured through comparisons with offline filter-base samples analyzed by IMPROVE-A thermal-optical protocols (Li *et al.*, 2017). However, our outputs of the APC-710s were multiplied by 0.75 and 2.63 to obtain OC and EC concentrations, respectively, based on a comparison study conducted at site A (Saito *et al.*, 2018).

It is important for both scientific modeling and policy making to separate OC into primary OC (POC) and secondary OC (SOC). Some methods have been proposed to estimate SOC from a measurement set of OC and EC by using EC as a tracer of POC (Turpin and Huntzicker, 1995). Recently, Wu and Yu (2016) presented a new EC tracer method to estimate OC/EC ratios at primary sources, $(OC/EC)_{pri}$. They consider that $(OC/EC)_{pri}$ should

give a minimum correlation between EC and SOC, if variations in EC and SOC are independent in a certain period. Once $(OC/EC)_{pri}$ is given, SOC is estimated by the following equations (Turpin and Huntzicker, 1995).

$$POC = (OC/EC)_{pri} EC \quad (1)$$

$$SOC = OC - POC \quad (2)$$

In the present study, $(OC/EC)_{pri}$ was estimated for each month and site by the method of Wu and Yu (2016), who.

3. RESULTS

3.1 Semiannual Statistics

Six-months mean and 98-percentile (hereinafter, 98%) concentrations of measured OC and EC and estimated SOC are summarized in Table 1. The monitor at site A worked almost all hours of the whole year. The monitor at site B worked for only 35% in November due to an operational error but almost fully in the other months. The monitoring at site C begun in late September 2016.

The semiannual-mean concentrations of EC and OC at site B are quite low, which are comparable to annual-mean concentrations at a rural background site in Central Europe (EC of 0.65 ± 0.53 and OC of 2.85 ± 1.91 $\mu\text{gC m}^{-3}$) (Mbengue *et al.*, 2018). The mean and 98% concentrations of EC at site A are obviously higher than

Table 1. Six-months statistics of hourly concentrations of EC, OC and SOC at the three sites.

	Site A (Koto)	Site B (Komae)	Site A (Koto)	Site B (Komae)	Site C (Kazo)
Period	Warmer months (Apr. 2016 to Sep. 2016)		Colder months (Oct. 2016 to Mar. 2017)		
n [hr] (completeness) ¹	4,392 (100%)	4,209 (95.8%)	4,309 (98.6%)	3,899 (89.3%)	4,350 (99.6%)
EC [$\mu\text{gC m}^{-3}$]					
98%	3.3	2.3	5.5	3.6	6.7
avg. \pm s.d.	1.03 ± 0.83	0.51 ± 0.79	1.33 ± 1.31	0.80 ± 1.17	1.59 ± 1.49
R to Site A	–	0.63	–	0.69	0.56
OC [$\mu\text{gC m}^{-3}$]					
98%	5.4	5.1	8.9	8.2	13.5
avg. \pm s.d.	2.48 ± 1.25	2.30 ± 1.16	3.17 ± 1.97	3.14 ± 1.88	3.58 ± 2.74
R to Site A	–	0.66	–	0.82	0.62
SOC [$\mu\text{gC m}^{-3}$]					
98%	2.9	3.4	4.2	4.9	6.4
avg. \pm s.d.	1.32 ± 0.79	1.64 ± 0.87	1.46 ± 1.18	1.89 ± 1.29	1.43 ± 1.47
R to Site A	–	0.71	–	0.71	0.38
SOC/OC	53%	71%	46%	60%	40%

¹(Measured hours)/(Total hours in each period)

those at site B. However, the mean and 98% concentrations of OC at site A are slightly higher than those at site B. Moreover, the mean and 98% concentrations of SOC at site A are lower than those at site B. As a result, the mean SOC/OC ratios at site A are lower than those at site B. From these findings, it seems that site A is more influenced by primary sources of EC and OC than site B. All the concentrations at sites A and B in the colder months (October to March) are higher than those in the warmer months (April to September). This is due probably to higher emissions and more stable atmospheric conditions in the colder months. SOC/OC ratios in the warmer months are higher than those in the colder months, which implies more active secondary formation in the warmer months. Correlations between sites A and B for EC, OC and SOC are relatively high, and higher in the colder months than in the warmer months. It could be considered that both sites A and B are basically under the same atmospheric conditions on an hourly basis.

For the colder months, measurements at site C are available for discussion. The mean and 98% concentrations of EC and OC at site C are remarkably higher than those at sites A and B. For SOC, however, the mean concentration is lower than site B, and the 98% concentration is higher than site B. The mean SOC/OC ratio at site C is smaller than those at sites A and B. It seems that site C is much influenced by primary emissions of EC and OC, compared to sites A and B.

3.2 Seasonal and Diurnal Variations

Monthly-mean concentrations of EC, OC and SOC are shown in Fig. 2. The EC concentration at all the sites shows similar seasonal variations with the highest in December. This tendency is more pronounced at site C than sites A and B. For OC, variations in the concentration are surprisingly harmonized among the three sites except for October at site C. The estimated SOC concentration shows similar seasonal tendencies among the sites, low in summer and winter and high in spring and autumn. However, the seasonal variation in the SOC concentration is less pronounced compared to those in EC and OC. This means that the pronounced seasonality in OC is mainly contributed by primary OC.

Fig. 3 shows the estimated $(OC/EC)_{pri}$ for each month and site. The $(OC/EC)_{pri}$ mostly falls between 1 and 2, with the highest for each site found in January. Basically, a tendency is found that the ratio at site B is larger than sites A and C. Miyazaki *et al.* (2006) estimated $[OC/EC]_{pri}$ ratios from OC and EC concentrations measured in ano-

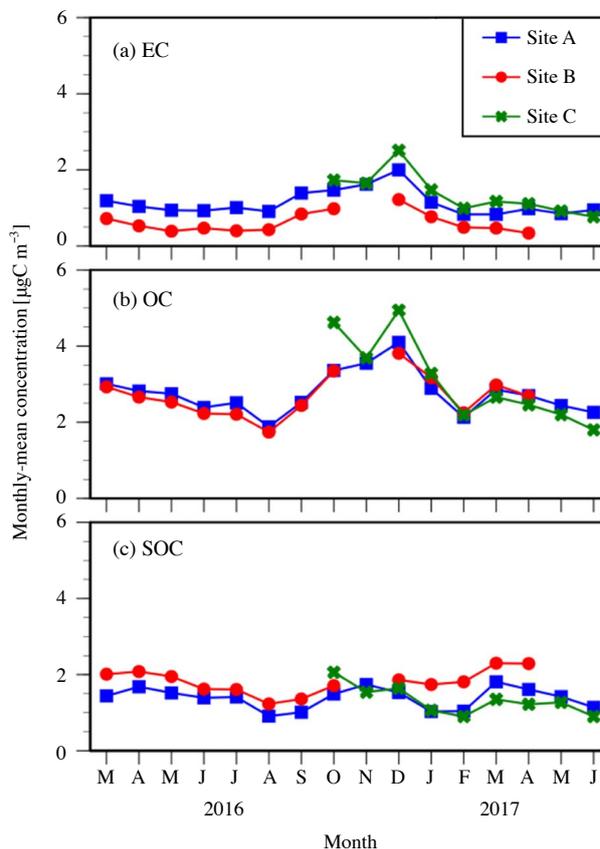


Fig. 2. Monthly-mean concentrations of EC, OC and SOC.

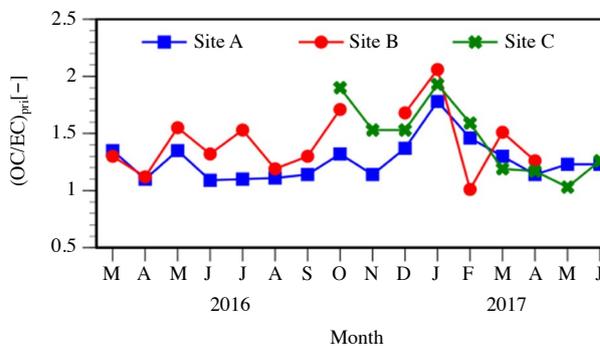


Fig. 3. Primary OC/EC ratios.

ther residential area of Tokyo in 2004 with a semi-continuous analyzer (Sunset Laboratory Inc.). The analyzer was operated for about two weeks in each season at an hourly cycle (45 min sampling and 15 min analysis). Therefore, their temporal resolution is very close to, but the number of operational days is much shorter than ours (1 hour and more than 1 year). Their estimates of $[OC/EC]_{pri}$ were 1.46 in winter, 1.36 in summer, and 1.33 in fall. These

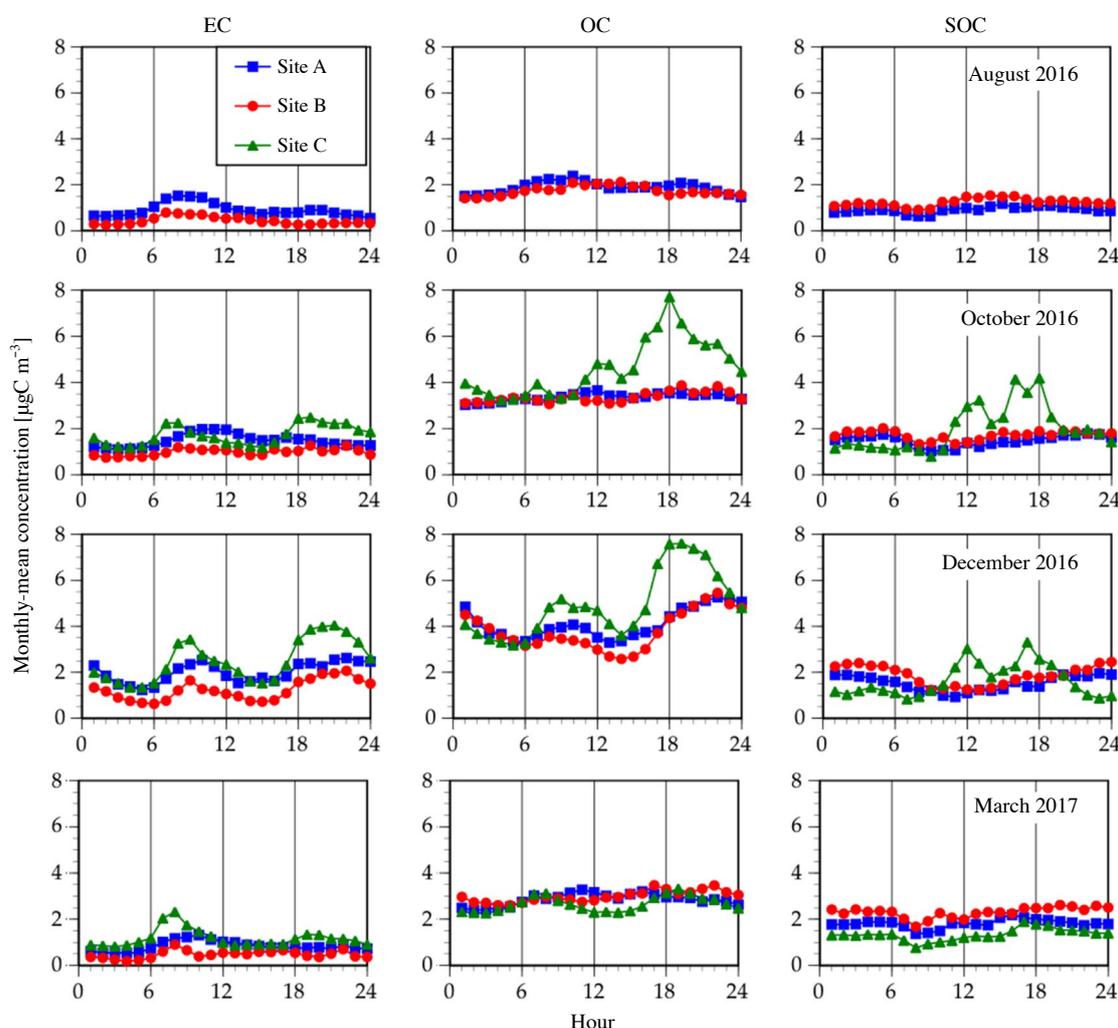


Fig. 4. Monthly-mean diurnal variations of EC, OC and SOC in August, October and December 2016 and March 2017 (from top to bottom).

results are generally consistent with the present study showing the highest (1.78) in January at all the sites.

Fig. 4 shows monthly-mean diurnal variations of EC, OC and SOC concentrations in several months. Each hour indicates time at the end of sampling; e.g., an hourly-mean concentration from 9 to 10 is plotted at 10. In August when the monthly-mean concentrations of EC, OC and SOC are very low as shown in Fig. 2, mean diurnal variations are also very small. For EC at sites A, a peak in the morning is marginally recognized. The SOC concentration slightly increases in the afternoon. Photochemical formation of SOC will be discussed later in section 4.4.

In October, concentrations of OC at site C become considerably high in the evening, although those in the morning are in equal to those at sites A and B, where almost no diurnal variations are shown. Variations in SOC at site

C is also different from those at sites A and B. Diurnal variations in the EC concentration are very small and inconsistent among the sites.

In December when the monthly-mean concentrations are the highest, the EC concentration shows two distinct peaks in the morning and the evening. The evening peak is higher than the morning peak. The same feature was found in other urban areas including Athens (Grivas *et al.*, 2012) and Beijing (Jia *et al.*, 2016). According to Mbenque *et al.* (2018), the evening peak is related to the evening rush hour combined with worsening of atmospheric dilution. However, this explanation seems insufficient for our case, as discussed later. The diurnal variation in OC is similar to but more pronounced than that in EC. SOC concentrations at sites A and B are higher in nighttime than daytime, and those at site C have two peaks around

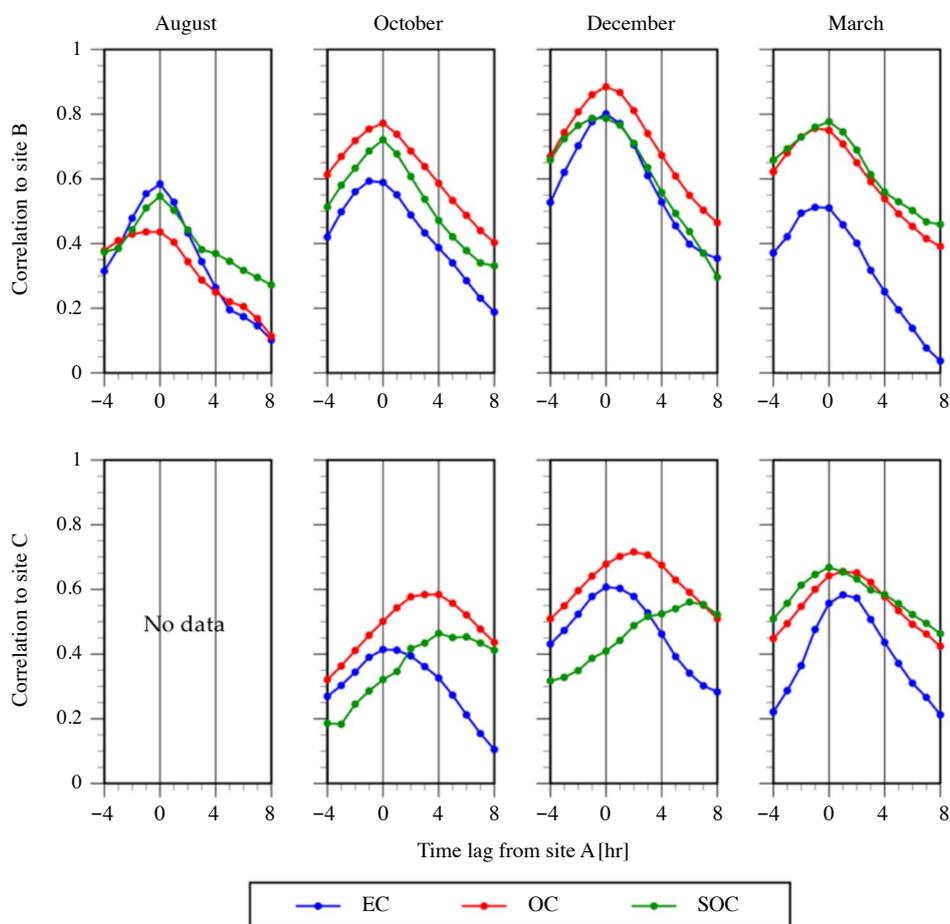


Fig. 5. Time-lagged correlations of EC, OC and SOC at site A to sites B and C.

the noon and in the evening.

In March 2017 with the highest monthly-mean concentration of SOC at sites A and B, diurnal variations in EC, OC and SOC are very small with some exceptions including relatively high EC concentrations at site C in the morning and relatively low OC concentrations at site C in the afternoon. SOC concentrations are higher at site $B > A > C$, in contrast to EC.

4. DISCUSSION

4.1 Time-Lagged Correlations

Site-by-site correlations given in Table 1 were computed for pairs of simultaneous measurements. Here in this section, time-lagged correlations are calculated in order to see spatial relationships among the sites. Fig. 5 shows time-lagged correlations of EC, OC and SOC at site A to sites B and C in characteristic months of August, October

and December 2016 and March 2017. In Fig. 5, positive time lags mean that concentrations at site A behind change after those at site B or C.

Overall, the highest correlations of EC are found within ± 1 hour of lag time, which means that EC concentrations almost simultaneously vary among the three sites. As mentioned in 3. 1, however, concentration levels of EC are different among the sites. It is considered that EC concentrations at each site are influenced by local sources with similar diurnal patterns but different intensities in emission. Correlations of EC are higher in December than in other months. This is likely because all the sites are covered by the same atmospheric conditions with thinner and more stable layers in winter than other seasons. Correlations of OC are higher than those of EC except in August. OC is partly composed of SOC, which is originated from non-local emissions from various kinds of sources. So, concentrations of OC are more uniformed

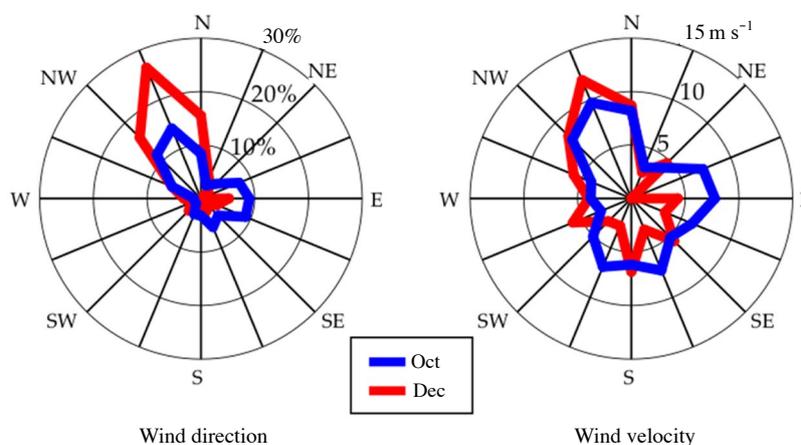


Fig. 6. Wind roses at the Saitama observatory in October and December, 2016.

in space than those of EC, which increase time-lag correlations.

Between sites A and B in March, correlations of SOC well correspond with those of OC and much higher than those of EC. This suggests that the behavior in OC in March at both sites is mainly controlled by SOC and much less influenced by local sources.

Between sites A and C, the highest correlations of OC are found at +3 to +4 ($r=0.58$) and +2 hours ($r=0.72$) for October and December, respectively, although the highest correlations of EC ($r=0.41$ and 0.61 , respectively) are at 0 hour (no time lag). This suggests that OC is transported from site C to site A with a typical speed of 13 km hr^{-1} ($= 51.6 \text{ km}/4 \text{ hr}$) in October and 26 km hr^{-1} ($= 51.6 \text{ km}/2 \text{ hr}$) in December. Fig. 6 shows wind roses in the evening to early night (17 to 21) of every day in October and December 2016 at the Saitama meteorological station, which is located just in the middle between sites A and C. Those hours are when OC concentrations at site C become high (Fig. 3). In October, mean wind velocities are 7.8 , 9.7 and 8.2 km hr^{-1} from dominant wind directions of NW, NNW and N, respectively. Winds in December are more remarkably from NNW and stronger than those in October; 8.3 , 12.0 and 8.7 km hr^{-1} from dominant wind directions of NW, NNW and N, respectively. It is reasonable that December is faster than October for both the transport speed and wind velocity, and that dominant air flows correspond to the direction from site C to site A in both months. However, transport speeds of OC between the sites estimated above are much faster than mean wind velocities in both months. This is probably because open field burning is carried out across a wide

area between sites A and C (Fig. 1).

4.2 EC Concentrations and Nearby Emissions

As shown in Table 1, mean concentrations of EC in descending order are site $A > C > B$. According to the Japan Auto-Oil Program (JATOP) emission inventory database (JEI-DB) (Chatani *et al.*, 2018), annual emissions of EC in 2010 are estimated as 247, 111, and 31 tons from $15 \text{ km} \times 15 \text{ km}$ grid squares with sites A, B, and C, respectively. It is reasonable that relative concentrations of EC at site B to site A ($0.51/1.03 = 0.50$ in the warmer months and $0.80/1.33 = 0.60$ in the colder months from Table 1) are very close to a relative annual emission of EC in the site B grid to the site A grid ($111/247 = 0.45$). However, a relative concentration ratio of EC at site C to site A ($1.59/1.33 = 1.20$ in the colder months) is nearly nine-times larger than a relative annual emission of EC in the site C grid to the site A grid ($31/247 = 0.13$). This discrepancy is too large to be explained by seasonal variations. A plausible cause is local emissions. Site C is surrounded by agricultural fields and rice paddies, where open field burning emissions could be still underestimated.

4.3 High OC Concentrations at Site C

As shown in Fig. 4, OC concentrations at site C become remarkably high in the evening in October and in the morning and evening in December, compared with sites A and B. Fig. 7 shows hourly concentrations of OC at each site in October.

OC concentrations at site C in October are mostly shown in late half of the month. Except for the steeply high concentrations, OC concentrations at site C are at

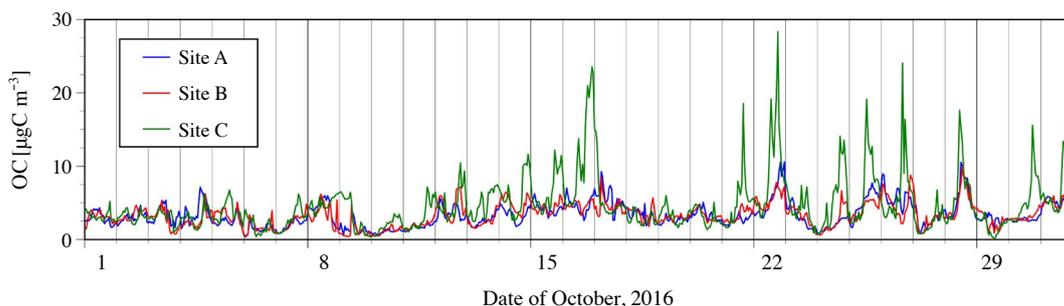


Fig. 7. Hourly concentrations of OC in October, 2016.

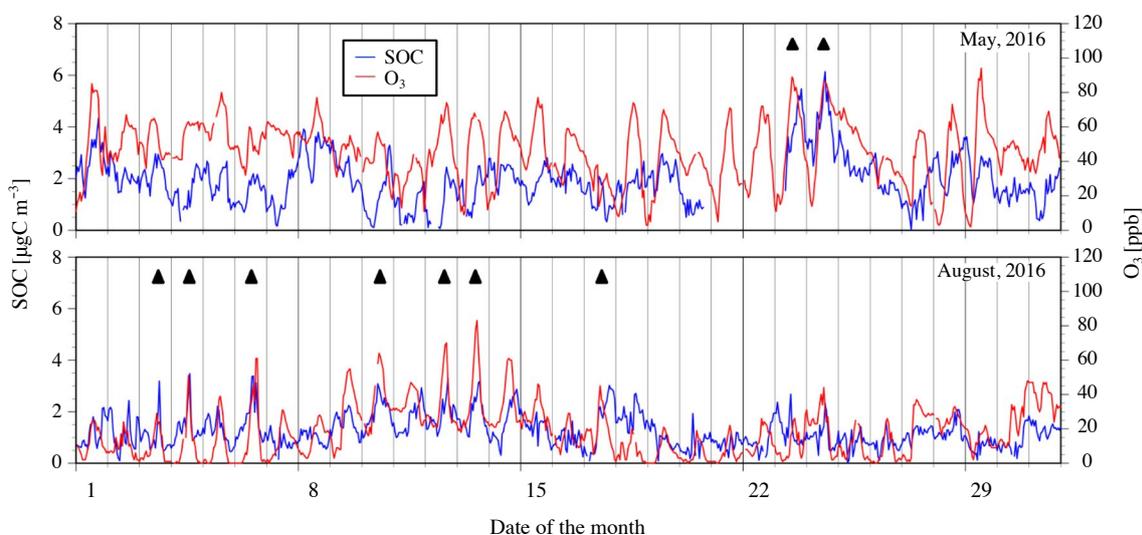


Fig. 8. Hourly concentrations of SOC and O₃ at site B in May and August 2016. Triangles indicate days with the top-ten hourly concentrations of SOC (two days in May and seven days in August).

the same level as sites A and B. Therefore, it is suggested that the high concentrations measured only at site C are influenced by local sources. Site C is surrounded by agricultural fields and rice paddies, and late October is during the highest season of open field burning (Hasegawa, 2017). Ichikawa and Naito (2017) conducted field experiments for chemical composition of PM_{2.5} in biomass burning smokes in nine farmlands in Chiba, Japan, and obtained an overall mean OC/EC ratio of 6.5 and site averages from 2.7 to 18.8. At site C, OC concentrations exceeded 15 µgC m⁻³ for 20 hours in October. At such high concentrations, the OC/EC ratios ranged 2.2 to 14.5, which is nearly correspond to those in open burning smokes. It is, therefore, suggested that the high concentration of OC in October at site C is contributed by open field burning. Compared to October, OC/EC ratios in December at site C are small, from 1.5 to 4.1 in

15 hours with high concentrations of OC over 15 µgC m⁻³. This means that EC concentrations in December are also higher than those in October, when OC concentrations exceeded 15 µgC m⁻³. OC/EC ratios of biomass burning smokes considerably vary with kinds of biomass (Ichikawa and Naito, 2017). It is suggested that kinds of biomass in December are different from those in October and/or site C is influenced by both open field burning and other EC sources.

4.4 Relationships between SOC and Oxidant

As shown in Figs. 2 and 4, monthly-mean SOC concentrations in August are not high, although SOC is formed through photochemical reactions. Takahashi *et al.* (2011) analyzed PM_{2.5} chemical compositions collected six-hourly in summer in Greater Tokyo and showed that the SOC concentration was well related with the Oxidant

concentration ($n = 26$, $r = 0.604$, $p < 0.005$), where Oxidant = $O_3 + NO_2 - 0.1(NO + NO_2)$. It was sustained that the formation of SOC was activated under photochemical conditions. We examined this relationship between SOC and Oxidant for site B, where SOC concentrations were mostly the highest among the sites, by each month and obtained relatively high correlations in August ($r = 0.67$), July ($r = 0.47$) and May ($r = 0.44$), compared with other months. These high correlations also suggest the photochemical formation of SOC. In addition, the top ten hourly concentrations of SOC in each month appear on just two straight days in May (23rd to 24th), but on non-sequential 7 days in July (1st, 2nd, 4th, 11th, 12th, 17th and 18th) and 7 days in August (3rd, 4th, 6th, 10th, 12th, 13th and 17th) (Fig. 8). This implies that both SOC and Oxidant increase in local-scale, short-term events in July and August and in large-scale, long-term events in May. The latter case suggests trans-boundary transport of polluted air masses, which is often observed in the spring season.

4. CONCLUSIONS

Hourly concentrations of fine particulate OC and EC were monitored for more than one year by optical monitors at three sites in and out of Tokyo, Japan. SOC concentrations were estimated from the OC and EC concentrations by the EC tracer method (Wu and Yu, 2016). Seasonal and diurnal variations in OC, EC and SOC in Greater Tokyo were revealed from the year-long hourly measurements. These findings made clear what to be expressed in air quality modeling of particulate carbons. For EC, it is important to precisely estimate spatiotemporal variations in emissions and to predict atmospheric conditions. It would be required to reexamine sources in agricultural areas. For OC, precise emissions variations like EC would not be required, because spatiotemporal variations in OC are moderate, except in colder months when open field burning influences much. Photochemical formation of SOC was suggested but would not have large impacts on OC, because the OC concentration was low in the summer of 2016.

ACKNOWLEDGEMENT

This research was funded by the Environment Research and Technology Development Fund (S-1601 and S-

1604) of the Environmental Restoration and Conservation Agency. The authors are grateful to the Kimoto Electric Co., Ltd. for technical support on their manufactured APC-710s.

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