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Optical Method for Measuring Deposition Amount of Black Carbon Particles on Foliar Surface

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ABSTRACT

To perform quick measurements of black carbon (BC) particles deposited on foliar surfaces of forest tree species, we investigated an optical method for measuring the amount of BC extracted from foliar surfaces and collected on quartz fiber filters. The seedlings of *Fagus crenata, Castanopsis sieboldii, Larix kaempferi* and *Cryptomeria japonica* were exposed to submicron BC particles for one growing season (1 June to 7 December 2009). At the end of the growing season, the leaves or needles of the seedlings were harvested and washed with deionized water followed by washing with chloroform to extract the BC particles deposited on the foliar surfaces. The extracted BC particles were collected on a quartz fiber filter. The absorption spectrum of the filters was measured by spectrophotometer with an integrating sphere. To obtain the relationship between the absorbance of the filter and the amount of BC particles on the filter, the amount of BC particles on the filter was determined as that of elemental carbon (EC) measured by a thermal optical method. At wavelengths below 450 nm, the absorption spectrum of the filter showed absorption by biological substances, such as epicuticular wax, resulting in the low coefficient of determination (R^2) in the relationship between the amount of EC on the filter (M_{EC}, μg C cm^{-2} filter area) and the absorbance of the filter. The intercept of the regression line between M_{EC} and the absorbance of the filter at 580 nm (A_{580}) was closest to 0. There was a significant linear relationship between the A_{580} and M_{EC} (R^2 =0.917, $p<0.001$), suggesting that the amount of BC particles collected on the filter can be predicted from the absorbance. This optical method might serve as a simple, fast and cost-effective technique for measuring the amount of BC on foliar surfaces.

Key words: Black carbon particles, Integrating sphere spectrophotometer, Thermal optical method, Forest tree species, Foliar surface

1. INTRODUCTION

Black carbon (BC) is emitted from the combustion of fossil fuels and biomass (Ramanathan and Carmichael, 2008). BC produces a warming effect in the atmosphere by absorbing solar radiation and has a negative impact on human health (WHO, 2012; Forster *et al.*, 2007). The major sources of BC are developing nations in the tropics and East Asia (Ramanathan and Carmichael, 2008). It is projected that BC emissions in Asia will continue to increase (Ohara *et al.*, 2007). East Asian countries are facing the problem of transboundary air pollution, including BC and other aerosol particles, and its effects on vegetation (Izuta and Funada, 2010). Therefore, it is necessary to clarify the effects of BC particles on the growth and physiological functions of East Asian forest tree species.

Although there is little information on the effects of BC on the growth and physiological function of plants, it has been suggested that the BC deposited on the foliar surface at >400 mg C m^{-2} projected leaf area increases leaf temperature and intercepts the irradiation light, which results in a reduction in net photosynthesis (Hirano *et al.*, 1995, 1991). Although many re-

searchers have reported deposition amounts of insoluble particulate matter and their elemental composition on foliar surfaces (e.g. Sæbø *et al*., 2012; Dzierzanowski *et al*., 2011; Takamatsu *et al*., 2001, 2000; Beckett *et al*., 2000; Turunen *et al*., 1997), there is no information on the specific amount of BC particles deposited on foliar surface. To clarify the effects of BC particles on growth and leaf or needle gas exchange rates of forest tree species, it is necessary to make field estimates for the amount of BC particles deposited on foliar surfaces (e.g. Matsuda *et al*., 2012). Furthermore, it is important to validate (using actual measurement) the estimated amount of BC deposited on the forest canopy. However, a large number of sample measurements are required for validation because of the huge and seasonal variation in the amount of BC deposition (Matsuda *et al*., 2012). Therefore, it is necessary to establish a simple, fast and cost-effective technique for measuring amount of BC deposited on the foliar surface of forest trees.

Actual measurement of atmospheric BC has been performed by thermal elemental carbon (EC) analysis (e.g. Schmid *et al*., 2001). It is not, however, realistic to carry out the large number of required measurements because the thermal optical method takes too long. On the other hand, an optical method using an integrating sphere was established and has been used for the measurement of BC (Hitzenberger *et al*., 2006, 1996). This method allows the measurement of spectral absorbance in a short time, and thus, allows the measurement of a large number of samples. It has been reported that light-absorbing compounds, such as brown carbon, affects the accuracy of the optical method for measuring BC amount (Reisinger *et al*., 2008). However, accuracy could be improved by using an appropriate wavelength based on the absorbance spectrum and by correction using a calibration curve with actual measurements using the thermal optical method. In the present study, therefore, we investigated the optical method for measuring the amount of BC extracted from the foliar surface of forest tree species and collected on a quartz fiber filter to analyze a large number of samples within a short time.

2. MATERIALS AND METHODS

2.1 Plant Materials

From 1 June 2009 to 7 December 2009, 3-year-old *Fagus crenata* seedlings (deciduous broad-leaved tree), 2-year-old *Castanopsis sieboldii* seedlings (evergreen broad-leaved tree), 1-year-old *Larix kaempferi* seedlings (deciduous conifer) and 1-year-old *Cryptomeria japonica* seedlings (evergreen conifer) were planted in 2 L pots and grown in 6 phytotron chambers (Koito Industries Co., Ltd., Japan) at the Tokyo University of Agriculture and Technology (Fuchu, Tokyo, Japan). During the growing season, air temperature and relative air humidity in the chambers were maintained at $25.0 \pm 1.0/18.0 \pm 1.0$°C (6:00-18:00/18:00-6:00) and 70 ± 5%, respectively. All seedlings were fertilized at two-week intervals with 200 mL liquid fertilizer diluted 2,000 times (HYPONeX, N : P : K=6 : 10 : 5, Hyponex Japan Co. Ltd., Japan).

2.2 Exposure to Black Carbon Particles

The seedlings were exposed to submicron BC particles generated by an aerosol generator system centered in the chamber. The system is based on an electrostatic-spray (Lenggoro *et al*., 2002) and an ultrasonic nebulizer (Wang *et al*., 2008). The run time for electrostatic-spray and ultrasonic nebulizer, using suspension of BC particles, was 10 and 5 min, respectively. The source of BC was a powder sample (TokaBlack, Tokai Carbon Co. Ltd., Tokyo) with primary particle size around 30 nm. The nanopowders were prepared by a combustion process of hydrocarbon with oxygen containing mixture gases (Harada and Akabane, 2011). The powder samples were mainly consisting of elemental carbon (EC), after a measurement using a DRI OC (Organic carbon)/EC carbon analyzer (Model 2001A) with the thermal optical reflectance (TOR) method. The size distributions of generated and suspended aerosols in the dry condition (i.e. the solid BC particles) were measured by a real-time technique based on a differential mobility analyzer (DMA) method (Wang *et al*., 2008; Lenggoro *et al*., 2002). The measured mean size of BC particles was between 100-300 nm. This result indicates that the generated (dry) aerosols were formed by the aggregation of BC particles having primary particle size of around 30 nm. From 13 June 2009 to 7 December 2009, seedlings assigned to three chambers were exposed to BC particles every two days between 6:00-9:00 (BC treatment), and seedlings assigned to the remaining three chambers were not exposed to the particles (control treatment). The seedlings assigned to the BC treatment were placed around the aerosol generator system and exposed to BC particles in the chamber.

2.3 Extraction of BC Particles Deposited on the Foliar Surface

The treated and control seedlings were harvested and separated into the plant organs on 8-12 December 2009. Dry weights of harvested leaves or needles ranged from 0.31 to 1.51 g (i.e. from 45 to 198 cm^2 projected leaf area) for *F. crenata*, from 1.28 to 5.19 g (i.e. from 82 to 374 cm^2 projected leaf area) for *C. sieboldii*,

from 0.94 to 1.96 g for *L. kaempferi*, and from 1.05 to 4.07 g for *C. japonica* seedlings. The harvested leaves or needles were first washed with 20 mL of deionized water. Washed leaves were then dried in an oven (DX 402, Yamato Scientific Co., Ltd., Japan) at 40°C. Subsequently, dried leaves or needles were washed with 20 mL of chloroform for 20 seconds in an ultrasonic bath (AU-25C, Aiwa Medical Industry Co., Ltd., Japan). It has been reported that the particulate matter was trapped in waxes (Dzierzanowski *et al*., 2011). Because the plant cuticle, which coats the foliar surface and its outer layer is epicuticular wax, is hydrophobic (Samuels *et al*., 2008; Bargel *et al*., 2006), hydrophobic particles such as BC would adhere to the epicuticular wax. Although a partial amount of BC particles deposited on foliar surface can be extracted by water, it was necessary to dissolve the epicuticular wax to achieve complete extraction of the particles. Because chloroform has been used to dissolve and extract epicuticular wax quickly since the 1980s (e.g. Sæbø *et al*., 2012; Dzierzanowski *et al*., 2011; Takamatsu *et al*., 2001; Turunen *et al*., 1997; Cape *et al*., 1989), we also used chloroform to wash the leaves or needles to completely extract the BC particles deposited on the foliar surface.

2. 4 Measurement of BC Amount

The particles suspended in the deionized water and chloroform were individually collected on quartz fiber filters (QR-100, Advantec MFS, Inc., Japan) by gravitational filtration. Immediately before the filtration, the quartz fiber filters were submerged in deionized water or chloroform to completely remove air from the filter, and they were kept wet until filtration. After filtration, the filters were dried and used for the measurements. The absorbance of the filters used for the collection of BC particles from the foliar surface was measured by a spectrophotometer with an integrating sphere (U-4100, Hitachi High Technologies Corp., Japan). The amount of BC collected on the quartz fiber filters was determined as the amount of EC measured by the TOR method using a DRI OC/EC carbon analyzer (Model 2001A). The EC was determined as follows. The OC was evolved under a stream of He while heating the sample in four temperature steps (120, 250, 450 and 550°C). To evolve the EC and pyrolyzed OC, the sample was then heated under a mixture of 2% O_2 +98% He in three temperature steps (550, 700 and 800°C). Correction for the pyrolysis contribution to EC from OC was achieved by monitoring the reflectance of a laser beam by the filter. The detection limit of total EC measured by DRI OC/EC carbon analyzer is 0.19 μg C cm^{-2} (MOE, 2007), indicating that the limit of quantification is about 0.32 μg C cm^{-2}. The reproducibility of random error in the measurement of total EC is typically 5% for uniformly distributed sample (MOE, 2007). In this study, the measurement errors of absorbance were not considered, since the errors are considered to be much lower than the prediction errors described below.

2. 5 Collection Efficiency of BC Particles by Filtration Using Quartz Fiber Filters

Quartz fiber filters are not completely efficient collectors of submicrometer BC particles suspended in solution. However, the quartz fiber filter has been used for the filtration of BC particles in rainwater and snow (Matsuda *et al*., 2012; Cerqueria *et al*., 2010; Doherty *et al*., 2010; Clarke and Noone, 1985; Ogren and Charlson, 1983). In a study by Matsuda *et al*. (2012), the filtration procedure was the same as that mentioned above, and they reported that the collection efficiency of BC particles suspended in water was considered to be greater than 80%. However, the collection efficiency of BC particles suspended in chloroform by filtration using quartz fiber filters has not been reported. To confirm the collection efficiency, the commercial BC particles described above were prepared by weighing several hundred micrograms using a microbalance (MC5, Sartorius, Germany) and suspended in chloroform (20-100 mL) using an ultrasonic bath. The BC particles suspended in chloroform were collected on a quartz fiber filter by gravitational filtration. The amount of BC collected on the quartz fiber filters was determined as the amount of EC measured by the TOR method.

3. RESULTS AND DISCUSSION

The relationship between the amount of BC in chloroform applied to a quartz fiber filter and the amount of BC collected on the filter, determined as that of EC, is shown in Fig. 1. There was significant correlation between the amount of BC applied to the filter and the amount of EC on the filter. The slope of the regression line was 0.917. This result indicates that the collection efficiency of BC particles suspended in chloroform was greater than 90%.

Fig. 2 shows typical absorbance spectrums of the quartz fiber filter used for the collection of BC particles from the foliar surfaces of *F. crenata, C. sieboldii, L. kaempferi* and *C. japonica* seedlings. In all the tree species, the absorbance of the filter increased when the wavelength at which the absorbance was measured decreased. At a wavelength below 450 nm, the increase in the absorbance was greater than that above 450 nm. This tendency was observed in absorbance spectrums of all filters used for filtration of chloroform washings

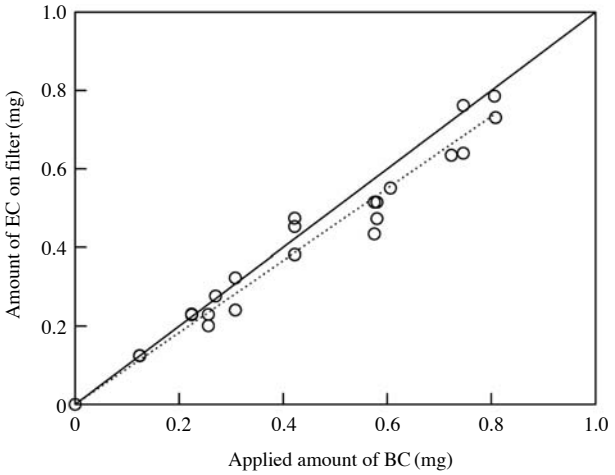

Fig. 1. The relationship between the amount of BC in chloroform applied to quartz fiber filter and the amount of BC collected on the filter determined as that of EC. Solid and dashed lines indicate a 1 : 1 line and the regression line of the relationship, respectively. The regression line was obtained from the linear regression analysis with the intercept forced through zero. The coefficient of determination (R^2) and slope (±standard error) of the regression line were 0.991 ($p < 0.001$) and 0.917 (±0.018), respectively.

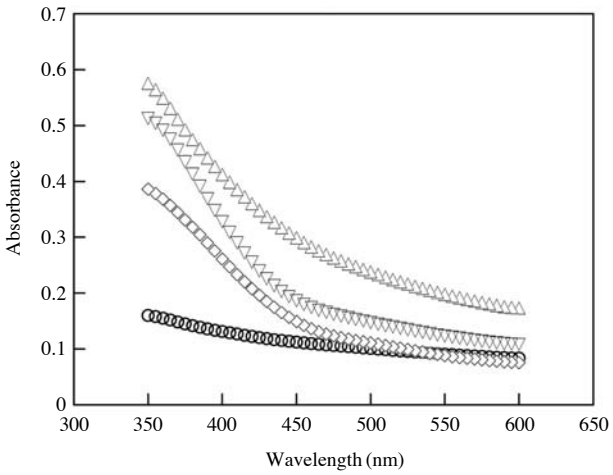

Fig. 2. Typical absorbance spectrums of quartz fiber filter used for the collection of BC particles extracted from the foliar surface of *F. crenata* (○), *C. sieboldii* (△), *L. kaempferi* (◇) and *C. japonica* (▽) seedlings.

but was not observed for that of deionized water washings. More than 95% of the ultraviolet radiation (UV, wavelength below 400 nm) is absorbed by cuticular, suberized outer layers of the epidermis and pigments, such as flavonoids, in plant leaf (Larcher, 2003). Because the chloroform used for the extraction of BC particles dissolves the epicuticular wax of the leaves and needles (Cape *et al.*, 1989), the absorption at a wavelength below 450 nm might be due to the epicuticular wax. In addition, in the seedlings of *C. sieboldii, L. kaempferi* and *C. japonica*, the increase in the absorbance below 450 nm tended to be greater than that in the *F. crenata* seedlings (Fig. 2). The species difference in the wavelength-dependent absorbance was observed in all filters used for filtration of chloroform washings. It has been suggested that the amount and composition of epicuticular wax changes the UV absorptivity of wax (Sase *et al.*, 1998). Therefore, the species difference in the degree of the increase in the absorbance at a wavelength below 450 nm might be due to a difference in the amount and/or composition of the epicuticular wax among the species. It is important to identify the light-absorbing compounds and their optical absorption property for further application of the present method to field measurement practices. However, the aim of the present study can be achieved by selecting an appropriate wavelength to avoid the influence of absorption by light-absorbing compounds on the accuracy of the present optical method. Therefore, absorbance at a wavelength above 450 nm was used for the quantification of the amount of BC collected on the filter.

Fig. 3 shows the relationship between wavelengths, at which the absorbance of quartz fiber filters was measured, and (a) the coefficient of determination (R^2) and (b) the intercept of the regression line between the amount of EC on the filter (M_{EC}, μg C cm^{-2} filter area) and the absorbance of the filter. The linear regression analyses were performed using the collective data of four tree species and two sample types (water and chloroform extract) due to the small number of data per species and sample type. The R^2 value increased drastically up to a wavelength of 450 nm and subsequently increased gradually. This tendency might be because of the absorption by biological substances such as epicuticular wax (Fig. 3). This result suggests that biological substances soluble in chloroform could hamper the relationship between M_{EC} and absorbance of the filter. Therefore, for more accurate estimation, it is necessary to confirm the absorption spectrum of the filter for each tree species to clarify whether there is an absorption maximum originating from these biological substances.

All the R^2 values were statistically significant (Fig. 3a). Relatively high R^2 values (above 0.9) were observed at wavelengths above 435 nm, and the highest value was observed at a wavelength of 600 nm (Fig. 3a). While increasing the wavelength, the intercept of the regression line drastically decreased up to a wavelength of 450 nm and subsequently decreased gradually (Fig.

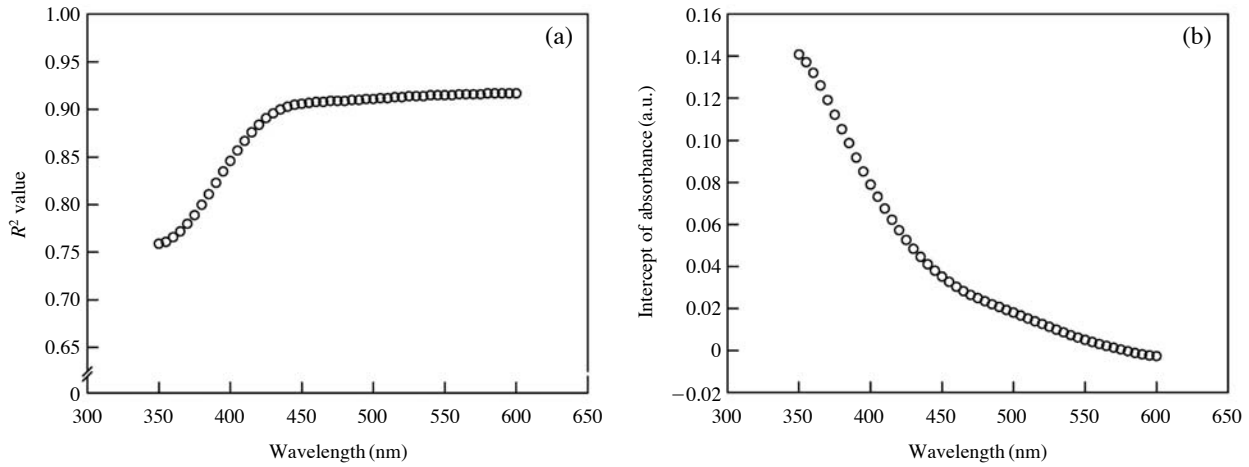

Fig. 3. The relationship between the wavelength, at which the absorbance of quartz fiber filters was measured, and (a) coefficient of determination (R^2) and (b) intercept of the regression line between the amounts of EC on the quartz fiber filter used for the collection of BC particles extracted from the foliar surface and the absorbance of the filter.

3b). Because the intercepts were not significantly different from 0 above 425 nm (data not shown), the absorbance of the filter at wavelength above 425 nm was statistically applicable to the estimation of the amount of BC particles collected on the filter. However, for more accurate estimation, it is desirable to use the absorbance at a wavelength above 450 nm because of absorption by epicuticular wax (Fig. 2). Furthermore, the intercept of the regression line between M_{EC} and the absorbance of the filter at 580 nm was closest to 0 (Fig. 3b). Ideally, the intercept of the regression line should be 0. Therefore, we assumed that the absorbance at 580 nm was suitable for estimating the amount of BC particles collected on the filter.

The relationship between M_{EC} and the absorbance at 580 nm (A_{580}) is shown in Fig. 4; a significant linear relationship (R^2=0.917, $p<0.001$) is obtained with a coefficient of variation for the slope of 6% as $A_{580}=(0.052\pm0.003)\ M_{EC}-(0.000\pm0.017)$. From the standard error (σ=0.017) of the interception of the relation, the detection limit (3σ) and limit of quantification (10 σ) of M_{EC} were calculated as about 1.0 and 3.3 μg C cm^{-2}, respectively. The 95% confidential intervals of the regression line were about ±0.4 μg C cm^{-2} around 4 μg C cm^{-2}, ±0.5 μg C cm^{-2} around 7 μg C cm^{-2} and ±1.0 μg C cm^{-2} around 10 μg C cm^{-2}. These results indicate that the method can predict M_{EC} with an error of about 10% for moderately loaded filter samples ($0.2<A_{580}<0.5$), and filter loading of 4 to 10 μg C cm^{-2} is the useful range of the prediction. The amount of light-absorbing compounds will vary depending on the amount of foliar samples, composition of biological substances and amount of contaminant deposited on the foliar surface. Therefore, it is necessary to confirm the appropriate wavelength and an applicable range of the relationship for each sample type, as determined by tree species and harvest region. This method, however, may serve as a simple, fast and cost-effective technique to measure amount of BC particles on foliar surface.

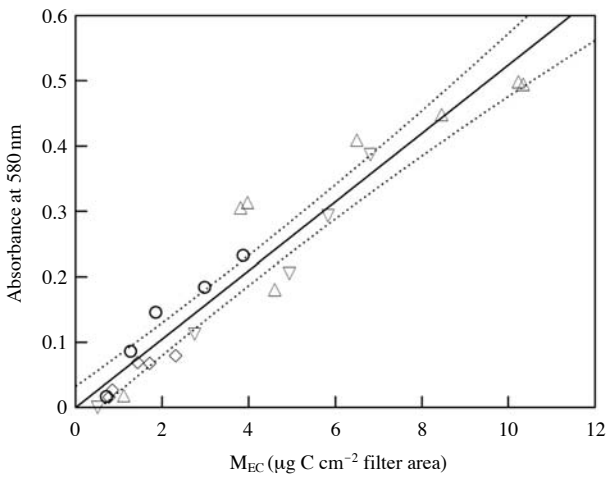

Fig. 4. The relationship between the amount of EC on the quartz fiber filter (M_{EC}, μg C cm^{-2} filter area) used for the collection of BC particles extracted from the foliar surface and the absorbance at 580 nm of the filter. The relationship was obtained by linear regression analysis using the collective data of the four tree species. The dashed lines indicate the 95% confidential intervals of the regression line. The coefficient of determination (R^2) was 0.917 ($p<0.001$) and slope and intercept (±standard error) of the regression line were 0.052 (±0.003) and −0.000 (±0.017), respectively. ○: *F. crenata*, △: *C. sieboldii*, ◇: *L. kaempferi*, ▽: *C. japonica*.

4. CONCLUSIONS

The results of the present study demonstrated that the amount of BC particles extracted from foliar surfaces of forest tree species and collected on a quartz fiber filter can be estimated from the absorbance of the filter. Part of the results were obtained using artificial BC particles, therefore, it is unclear whether the collection efficiency and calibration curve reported are directly applicable to the leaves or needles of forest trees in the field. However, this optical method can serve as a simple, fast and cost-effective technique for field measurements of amount of BC deposited on the foliar surface of forest trees.

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