

Mass concentrations of black carbon measured by four instruments in the middle of Central East China in June 2006

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Abstract. Mass concentrations of black carbon (BC) were determined in June 2006 at the top of Mount Tai (36.26° N, 117.11° E, 1534 m a.s.l.), located in the middle of Central East China, using four different instruments: a multi-angle absorption photometer (5012 MAAP, Thermo), a particle soot absorption photometer (PSAP, Radiance Research), an ECOC semi-continuous analyzer (Sunset Laboratory) and an Aethalometer (AE-21, Magee Scientific). High correlation coefficients ($R^2 > 0.88$) were obtained between the measurements of the BC mass concentrations made using the different instruments. From the range of the slopes of the linear least-square fittings, we concluded that BC concentrations regionally-representative of the area were measured in a range with a maximum-to-minimum ratio of 1.5 (an exception was that the BC (PM_{2.5}) concentrations derived from MAAP were ~2 times higher than the optical measurements (PM_{2.5}) derived from the ECOC analyzer). While this range is significant, it is still sufficiently narrow to better constrain the large and highly uncertain emission rate of BC from Central East China. In detail, two optical instruments (the MAAP and the PSAP equipped with a heated inlet 400°C) tended to give higher concentrations than the thermal EC concentrations observed by the ECOC analyzer. The ratios of optical BC to thermal EC showed a positive correlation with the OC/EC ratio reported by the ECOC analyzer, suggesting two explanations. One is that the optical instruments overestimated BC concentrations in spite of careful cancellation of the scattering effect in the MAAP instrument and the

expected evaporation of volatile species by heating the inlet of the PSAP instrument. The other is that the determined split points between OC and EC were too late when a large amount of OC underwent charring during the analysis, resulting in an underestimation of EC by the ECOC analyzer. High ratios of optical BC to thermal EC were recorded when the NO_x/NO_y ratio was low, implying the coating of the particles became thicker in an aged air mass and thus resulted in the optical instruments overestimating BC concentrations because of the lensing effect.

1 Introduction

Black carbon (BC) aerosol particles are an important component of the atmosphere contributing to global warming. A global average of the direct radiative forcing at the top of the atmosphere due to the present day increased atmospheric loading of BC has been estimated to be in the range of +0.2–0.6 W m⁻² (IPCC, 2007). The large uncertainty range partially stems from the uncertainties in the estimated BC emission rate and the absolute concentration in the atmosphere. In particular, the emission rate of BC from China is estimated to be large and having large uncertainty. Bond et al. (2004) gave a central value of the emission rate of BC from contained combustion (fossil and biofuel combustion excluding open burning) in China as 1365 Gg yr⁻¹ for 1996, nearly 30% of the global emission rate (4626 Gg yr⁻¹). Even when the contribution from open burning (124 Gg yr⁻¹) is added, the fraction of the emission rate from China (1489 Gg yr⁻¹) is still large (~19% of the global value, 7951 Gg yr⁻¹). The ratios of their highest estimation to their lowest estimation



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of the emission rates are 3.9 for both contained combustion and open burning in China. Streets et al. (2003) estimated the total emission rate from China to be 1049 Gg yr^{-1} in 2000 with an uncertainty of a factor of 5.8 (as the 95% confidence interval). The ratio between their highest and lowest estimates is as high as a factor of 34. In the vast geographical area that makes up China, Central East China (CEC) is a region with outstandingly high emission rates of atmospheric pollutants. Although measurements of BC have been undertaken at several urban sites in the region (Bergin et al., 2001; He et al., 2001; Dan et al., 2004; Yang, F. et al., 2005; Ye et al., 2003; Cao et al., 2007; Yang, H. et al., 2005), measurements at rural/remote locations with regional representativeness have only been made at Lin'an (Zhejiang Province, 53 km west of Hangzhou) in November 1999 and in February–April 2001 (Xu et al., 2002; Wang et al., 2004), at Xianghe (70 km southeast of Beijing) in March 2005 (Li et al., 2007), and at Shangdianzi (about 150 km northeast of Beijing) from 2003 to 2005 (Yan et al., 2008). In these studies, the observed BC concentrations have not been compared with model predictions. During the ACE-Asia and TRACE-P field campaigns performed in the downwind area of China in February–April 2001, modeled BC concentrations were compared to observed values, testing the emission rate of BC from China (e.g., Woo et al., 2003; Carmichael et al., 2003a, b; Uno et al., 2003). However, no solid conclusion was reached particularly for the validity of the Chinese emission rate. Therefore no stringent studies have been performed testing the validity of the emission rate through a comparison of the observed and modeled mass concentrations of BC in China.

Generally, two approaches are employed for measurements of BC concentrations. One is the optical method, in which measurements of optical transmittance, reflectance, or their combination are undertaken with respect to a filter on which the aerosol particles are deposited. The measured absorbance or the attenuation of light through the filter is related to the mass of BC. The other type is the thermal method, in which organic carbon (OC) and elemental carbon (EC) collected on a filter are successively evaporated under He and O_2/He atmospheres with thermal evolution and then the evolved carbon is quantified as CO_2 or CH_4 . During the evaporation, the optical transmittance (or reflectance) of the filter is monitored, which typically decreases first and then increases, due to the charring of OC during the analysis. The OC and EC are separated by defining a split point where the original optical transmittance (or reflectance) is recovered during the temperature-rising program, to correct for the charring. Two thermal protocols are often used; those of the Interagency Monitoring of Protected Visual Environments (IMPROVE) of the Desert Research Institute (Chow et al., 2001) and the National Institute for Occupational Safety and Health (NIOSH) (Birch and Cary, 1996). The EC concentrations determined by the thermal methods should agree with the optical BC measurements. However, a disagreement

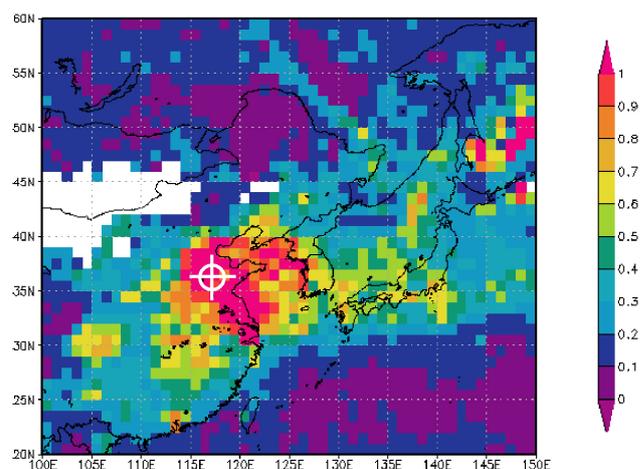


Fig. 1. Location of Mt. Tai (36.26° N , 117.11° E , 1534 m a.s.l.) shown together with aerosol optical depth at 550 nm measured by a MODIS sensor on board the Aqua satellite during the campaign period, June 2006, (<http://disc1.sci.gsfc.nasa.gov/G3/movas.shtml>).

by up to a factor of 4 has been reported (Park et al., 2006; Jeong et al., 2004; Bae et al., 2007; Hitznerberger et al., 2006; Reisinger et al., 2008), depending on the used techniques, sites, and aerosol properties. Although there has been much discussion about the selection of the temperature program and the mass absorption/attenuation coefficient assumed to calculate BC concentrations from the optical measurements, solid and systematic conclusions have not yet been reached. In this situation, one approach for the better quantification of BC concentrations would be to make measurements using several instruments and examine the differences. Such comprehensive measurements of BC aerosols have not been performed in China.

We performed an intensive field campaign on the summit of Mount Tai (36.26° N , 117.11° E , 1534 m a.s.l., Shandong Province of the People's Republic of China), located in the middle of CEC (Fig. 1), in June 2006 (The Mount Tai Experiment 2006, MTX2006) focusing on aerosol and ozone chemistry. The field site is free from local sources but is located at the center of the regionally polluted area over CEC (Fig. 1), providing a regionally-representative data set. One aim of the campaign was to determine BC concentration levels using multiple instruments to better constrain the emission rate from CEC that currently has large uncertainty. The instruments used were a multi-angle absorption photometer (5012 MAAP, Thermo) (Petzold and Schönlinner, 2004; Petzold et al., 2005), a particle soot absorption photometer (PSAP, Radiance Research) (Bond et al., 1999), and an ECOC semi-continuous analyzer (Sunset Laboratory) (Bae et al., 2004). An Aethalometer (AE-21, Magee Scientific) was also operated for several days during the first part of the campaign. In this paper, the observational results from the four instruments are compared. We found compact correlations but

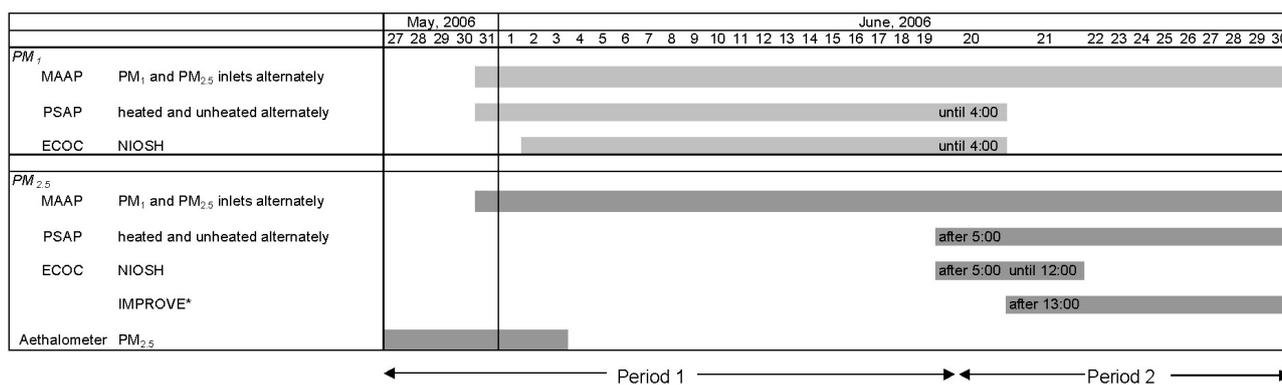


Fig. 2. Operation of the four instruments shown separately for PM_{10} and $PM_{2.5}$ measurements.

systematic differences between the mass concentrations derived from these instruments. From the range of the slopes of the linear least-square fittings, we conclude that the black carbon concentrations regionally representative over CEC were measured in a range with a maximum-to-minimum ratio of 1.5 (with one exception). This factor would be small enough to better constrain the emission rate of BC from CEC. We also investigate possible reasons for the optical measurements tending to give higher concentrations than the thermal method.

2 Experimental

Figure 2 shows the operation of the MAAP, PSAP, ECOC analyzer, and Aethalometer during the campaign. (The acronyms, full names, principles, operation modes, and detection limits of the instruments are summarized in Appendix A.) Most of the instruments were operated with PM_{10} (1- μm -diameter cutoff) cyclones during the early period (Period 1, before 20 June, 04:00 CST (Chinese Standard Time; UT+8 h)) and with $PM_{2.5}$ (2.5- μm -diameter cutoff) cyclones during the late period (Period 2, after 20 June, 05:00 CST). Thus the following comparisons are made for the two periods separately. For the MAAP instrument, an automated valve switched between the two cyclones (URG-2000-30EHB and URG-2000-30EH, URG Inc.) every 30 min such that sampling alternated between PM_{10} and $PM_{2.5}$. The MAAP instrument calculates absorbance from particles deposited on the filter using measurements of both transmittance and reflectance at two different angles. The absorbance is then converted to the mass concentration of BC using a fixed mass absorption coefficient at 670 nm of $6.6\text{ m}^2\text{ g}^{-1}$ as recommended by the manufacturer. The BC concentration derived from the MAAP instrument is hereafter referred to as MAAP_{BC}. The firmware used during the campaign was version 1.29. It is known that measurements with this firmware version show a leveling-off behavior at high concentrations (ca. $>7\text{ }\mu\text{g m}^{-3}$). Thus the raw data from the instrument were empirically corrected using a correction term provided

by the manufacturer. The correction was based on a comparison between the MAAP results (using version 1.29) and a reference method (DRI, Thermal-Optical-Reflectance correction, IMPROVE method) at two Japanese sites (Hasegawa et al., unpublished data, 2005). The version after the correction is referred to as version 1.29 corr. The uncertainty in the determined absorbance has been estimated to be 12% (Petzold and Schönlinner, 2004). The uncertainty in the BC mass concentration, including the uncertainty in the mass absorption coefficient, is estimated to be larger. The minimum detection limit is specified by the manufacturer to be $<0.1\text{ }\mu\text{g m}^{-3}$ with an averaging time of 2 min.

For the PSAP instrument, an automated valve switched between inlet tubes with and without heating at a frequency of 30 min. Only a short portion of a stainless steel tube (o.d. 3/8", 21 cm long) was heated and kept at 400°C. Considering the sample flow rate of 1.0 L min^{-1} , the residence time of the sampled air in the heated portion of the inlet tubing was only $\sim 0.6\text{ s}$. With heating, volatile compounds of aerosols (nitrates, organics, sulfates) internally or externally mixed with BC would be evaporated and thus the BC particles without coating, if originally present, would be collected on a filter. Kondo et al. (2006) reported that such heating at 400°C removed about $97\pm 5\%$ of inorganic (SO_4^{2-} , NO_3^- , Cl^- , and NH_4^+) and organic components as measured by an Aerodyne aerosol mass spectrometer (AMS), while the charring of the organic species is kept to a minimum. It is assumed in this study that this holds for the PSAP instrument. Kondo et al. (2006) have also reported that such heating makes the mass absorption coefficient much more stable even when the degree of the mixing of BC is changing. A PM_{10} cyclone (URG-2000-30EHB) was used for the PSAP instrument before 20 June, 04:00 CST, but it was replaced by a $PM_{2.5}$ cyclone (URG-2000-30EH) on 20 June, 05:00 CST. For both heated and unheated measurements, a single mass absorption coefficient $10\text{ m}^2\text{ g}^{-1}$ at 565 nm was employed to convert the light attenuation to the BC mass concentration as recommended by the manufacturer. The filter was manually changed and only data with transmittance higher than

Table 1. Temperature Programs used for the ECO-C Analyzer during the Campaign.

Step	Atmosphere	NIOSH ^a		IMPROVE ^{*b}	
		Temperature (°C)	Hold times (s)	Temperature (°C)	Hold times (s)
1	He	–	30 [10] ^c	–	10
2	He	600	80	350	90
3	He	840	90 [70] ^c	550	120
4	He	Oven off	55	Oven off	0
5	He:O ₂	550	30	550	75
6	He:O ₂	850	110	700	75
7	He:O ₂	–	–	850	90
8	He:O ₂	Oven off	85 [0] ^c	Oven off	30
9	CH ₄	–	120 [100] ^c	–	120

^a NIOSH program was employed before 21 June, 12:00 CST.

^b IMPROVE^{*} program was employed after 21 June, 13:00 CST.

^c Analysis time was shortened between 18 June, 15:00 and 20 June, 05:00 CST.

0.5 were used for analysis. The uncertainty in the determined light attenuation is estimated to be $\pm 15\%$ in the heated mode, while the uncertainty in the BC mass concentration is estimated to be larger. A detection limit of $1.8 \times 10^{-6} \text{ m}^{-1}$ (signal-to-noise ratio of 2) with an averaging time of 1 min (Li et al., 2007) corresponded to a detection limit of heated PSAP_BC of $\sim 0.2 \mu\text{g m}^{-3}$, assuming a mass absorption coefficient of $10 \text{ m}^2 \text{ g}^{-1}$. In the following discussion the measurements made by the PSAP instrument are referred to as heated PSAP_BC and unheated PSAP_BC.

The ECO-C analyzer from Sunset Laboratory was operated with the NIOSH temperature program before 21 June, 12:00 CST and with a program similar to that proposed by the IMPROVE after 21 June, 13:00 CST (herein we refer to it as the “IMPROVE^{*}” program, where the temperature is raised only to 550°C under a He atmosphere similarly to the IMPROVE program but the analysis time was shorter than the original recommendation). The specific temperature-rise programs employed during the campaign are listed in Table 1. We employed transmittance for optical correction, as opposed to reflectance which is used in the normal IMPROVE method. The sampling line was equipped with a PM₁ cyclone before 20 June, 04:00 CST, and a PM_{2.5} cyclone after 20 June, 05:00 CST. Gas-phase organic carbon species were removed by a denuder (Sunset Laboratory). An hourly measurement cycle was employed; the sampling period (at a flow rate of $\sim 8 \text{ L min}^{-1}$) was 40 or 42 min, followed by an analysis period of ~ 15 min. The ECO-C analyzer was calibrated with a known amount of sucrose ($42.1 \mu\text{gC}$). As mentioned earlier, the ECO-C analyzer is equipped to measure the laser transmittance at 660 nm through the filter to determine the split point between OC and EC during the thermal analysis. The change in the transmittance during the sample collection was also used to

determine BC concentrations optically. The BC mass loading (in $\mu\text{g cm}^{-2}$) on the filter was empirically determined by $2.25 \times \text{ABS} + 0.75 \times (\text{ABS})^2$, where ABS (absorbance) is determined from the temporal decrease in the laser transmittance during sample collection. The mass loading was then converted to concentration using the filter area and sample volume. The thermal and optical measurements based on the ECO-C analyzer are hereafter referred to as EC and opt-EC, respectively. The uncertainty in the total carbon (TC=EC+OC) concentrations was estimated to be $\pm 25\%$, owing to the uncertainties in the sample flow rate and calibrations during the campaign. Unfortunately, the operation condition of the instrument during the campaign was not very good; the measurements of the laser transmittance and flow rate were frequently affected by electric noise. Please note that the uncertainty in the split point that would depend on the selection of the temperature program will produce additional uncertainty in EC and OC. The following analysis (Sect. 3.2) suggests a 31% uncertainty arising from the selection of the temperature program. The detection limit of EC with a sampling time period of 40 min was estimated to be $< 0.4 \mu\text{gC m}^{-3}$.

The Aethalometer measured light attenuation by aerosol particles deposited on a filter at two wavelengths, 880 nm and 370 nm. In this paper, measurements at 880 nm were used. A mass absorption cross section of $16.6 \text{ m}^2 \text{ g}^{-1}$ was used to convert the observed light attenuation to the mass concentration of BC. Aerosol particles were sampled through a 1/4" ID Tygon tube (2 m long) to the Aethalometer. A PM_{2.5} cyclone (BGI SCC 1.828–5 Lpm) was employed in the sampling line with a flow rate of 5 L min^{-1} . A typical noise level is $< 0.1 \mu\text{gC m}^{-3}$ on a 5-min basis.

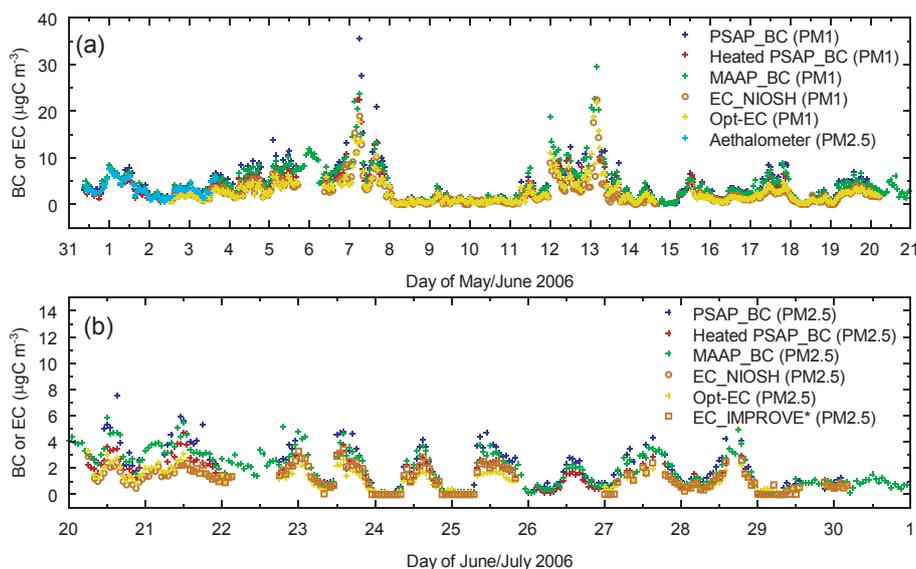


Fig. 3. Temporal variations in BC concentrations measured with four instruments. Panel (a): for the period between 31 May and 20 June (Period 1), when measurements with PM₁ cyclones are mainly performed, with an exception that Aethalometer data for PM_{2.5} are included. Panel (b): for the period between 20 and 30 June (Period 2), for which all plotted quantities are observed with PM_{2.5} cyclones.

The assumed mass absorption cross sections for the optical instruments are the ones empirically determined for particles under certain conditions. The conditions are not necessarily identical for all instruments. Therefore, it is not obvious that they are directly applicable for measurement of the black carbon particles at the top of Mt. Tai. Cross sections, depending on their size, shape, and the mixing state of the particles theoretically, should have uncertainties of a factor of ~ 2 . In this study, we first use the cross sections recommended by the manufacturers for simplicity and then discuss systematic differences in the BC concentrations derived from the instruments.

3 Results and discussions

The hourly data from the MAAP, PSAP, ECOC, and Aethalometer instruments are plotted as a time series in Fig. 3a and b for Periods 1 and 2, respectively. The hourly BC concentration (PM_{2.5}) as measured by the MAAP instrument (shown only for Period 2) ranged from -0.1 to $+40.8 \mu\text{gC m}^{-3}$ with an average of $3.7 \mu\text{gC m}^{-3}$. Two episodes with high BC concentrations are easily recognized for 5–7 June and 11–13 June. The high concentrations likely stem from significant open biomass burning of crop residues (winter wheat) over the North China Plain as discussed elsewhere (Li et al., 2008; Akimoto et al., 2008). It should be noted in the following discussion that the high concentration episodes occurred during Period 1, when the NIOSH program was employed for the ECOC analyzer. We found a regular diurnal variation pattern with a daytime maximum and nighttime minimum in Period 2 (Fig. 3b). The pattern is due

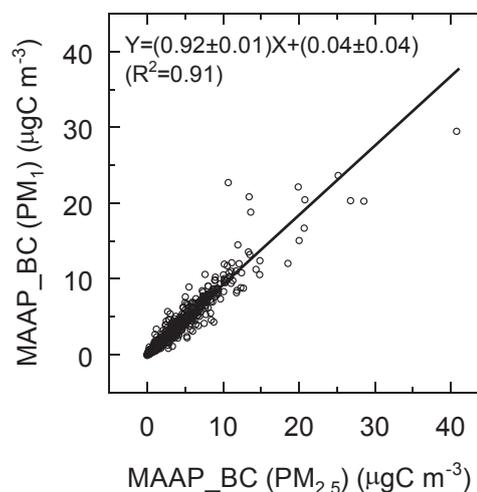


Fig. 4. Comparison between BC measurements made by a MAAP instrument employing PM₁ and PM_{2.5} cyclones. The sequential PM_{2.5} and PM₁ measurements within an hour (30 min for each) were regarded as a pair of hourly data.

to polluted air mass being transported to the mountain top during the daytime, which is associated with the buildup of the planetary boundary layer. During the night, on the other hand, the observed air mass is isolated from the sources. A similar diurnal pattern has been previously observed for CO and ozone at the site (Gao et al., 2005). In the following subsections, we separately discuss various aspects of the comparisons.

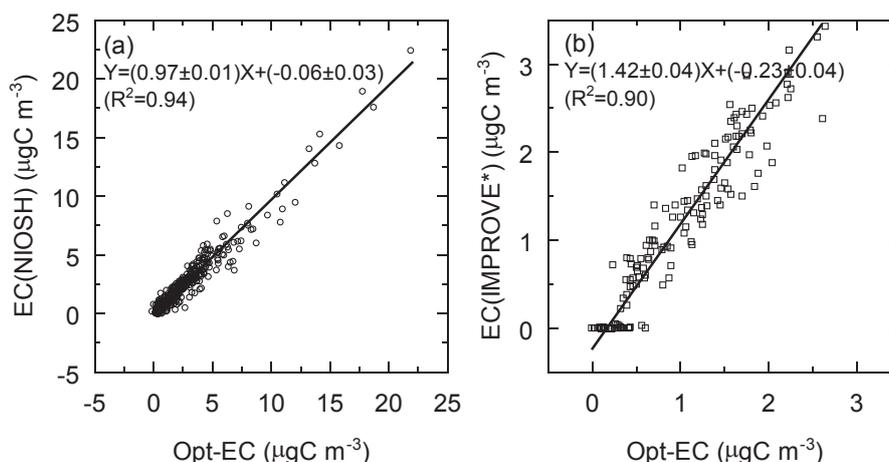


Fig. 5. Relationship between opt-EC with thermal EC measurements employing (a) NIOSH and (b) IMPROVE* temperature programs.

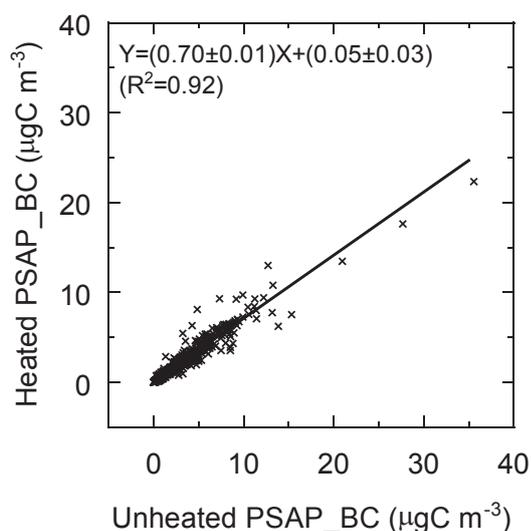


Fig. 6. Comparison of BC concentrations measured by a PSAP instrument with heated and unheated inlet tubes.

3.1 PM₁ vs. PM_{2.5} (MAAP)

Figure 4 shows a comparison between BC concentrations measured by a MAAP instrument employing PM₁ and PM_{2.5} cyclones. The sequential PM_{2.5} and PM₁ measurements within an hour (30 min for each) were regarded as a pair of hourly data. Although the measurement time periods did not exactly match, a tight correlation was found between them. This is because the temporal variation in the BC concentration was dominated by low-frequency components. The slope was 0.92 ± 0.01 , suggesting that 92% of BC is present in the submicron fraction of aerosol particles.

3.2 Opt-EC vs. EC: estimating the difference between NIOSH and IMPROVE* temperature programs

The EC measurements with different temperature protocols, NIOSH and IMPROVE*, are plotted against opt-EC in Fig. 5a and b. Tight correlations are found for both cases, implying that the ECOC analyzer measured the same component of aerosol particles by the two different techniques (i.e., optical and thermal techniques). The slopes of the bivariate linear regression analyses were 0.97 ± 0.01 and 1.42 ± 0.04 , respectively. It should be noted the concentration ranges for the two figures are different: Fig. 5a includes PM₁ data before 20 June, 04:00 CST (with high values during biomass burning episodes) and PM_{2.5} data between 20 June, 05:00 CST and 21 June, 12:00 CST, while Fig. 5b contains measurements of PM_{2.5} after 21 June 13:00 CST (with lower values). When opt-EC data lower than $2.64 \mu\text{gC m}^{-3}$ were selected, the slope of the regression line between opt-EC and NIOSH-EC was 1.08 ± 0.03 . This analysis suggests IMPROVE*-EC is systematically higher than NIOSH-EC by 31% on average, although we cannot eliminate the possibility that the difference arose from the different aerosol properties in the two periods. The magnitude of the difference is slightly larger than that reported for Tokyo of 21% (Kondo et al., 2006). Because the NIOSH protocol utilizes higher temperatures under a He atmosphere, EC might have started combustion and been counted as OC under the NIOSH program. Conversely, OC that can be evaporated only at higher temperature might have been included as EC under the IMPROVE* program.

3.3 Heated and unheated PSAP

Figure 6 shows a comparison between BC concentrations measured by a PSAP instrument with heated and unheated inlet tubes. Similar to the case above (Sect. 3.1), the two

measurements were only made alternately by switching a 3-way valve every 30 min. Both the data for Periods 1 and 2 are included in Fig. 6. The slope of a regression line is 0.70 ± 0.01 ($R^2=0.92$), suggesting that using the heated inlet tube resulted in BC concentration readings 30% lower than for the case using the unheated tube when an identical mass absorption coefficient ($10 \text{ m}^2 \text{ g}^{-1}$) was used. This indicates the mass absorption (attenuation) coefficient for the unheated tube is 30% higher than that for the heated tube, on average. Scattering by externally mixed particles deposited on the filter might have been counted as absorption in the unheated mode. In addition, internal mixing of transparent components might have changed the absorbance. As discussed by Kanaya et al. (2008), the NO_x/NO_y ratio was only 0.18 ± 0.10 on average during the campaign, suggesting the observed air mass is normally aged. The air mass age for a NO_x/NO_y ratio of 0.18 was roughly estimated to be 35 or 20 h, using model-derived OH concentrations of 1.8×10^6 or 3.1×10^6 radicals cm^{-3} as 24-h or daytime 12-h averages for this campaign period (Kanaya et al., 2008), respectively. In this calculation, we neglected conversion between NO_x and peroxy acetyl nitrate (PAN), continuous input of NO_x during the aging process, and removal of NO_y during transport. During the aging process, BC particles might have been coated by transparent components (e.g., OC), which could enhance absorption by the lensing effect. Bond et al. (2006) suggested that absorption by aged particles can be 1.5 times greater than that of fresh particles due to the coating.

3.4 Comparisons across instruments

Data from the Aethalometer ($\text{PM}_{2.5}$) can be compared only with the data from the MAAP operated with a $\text{PM}_{2.5}$ inlet in the same period. Figure 7 shows a tight correlation between them, with a slope of the regression line of 0.88 ± 0.03 ($R^2=0.92$). A good agreement was obtained, although coincident observations were made by the two instruments only for 4 days.

Correlations and least-square fittings for each pair among EC (NIOSH), opt-EC, heated_PSAP, and MAAP for Period 1 are shown in Fig. 8a. We found strong correlations ($R^2 > 0.88$) for all the pairs. The slopes and intercept values of regression lines ranged from 1.03 to 1.54 and from -0.08 to $+0.51 \mu\text{gC m}^{-3}$, respectively, suggesting general agreement among the three instruments. From the slope values, we can infer the general magnitude relationship as $\text{EC (PM}_1, \text{NIOSH)} \sim \text{opt-EC (PM}_1) < \text{heated PSAP_BC (PM}_1) \sim \text{MAAP_BC (PM}_1)$. The largest discrepancy was found between EC (PM_1 , NIOSH) and MAAP_BC (PM_1), with a slope of 1.54.

Similar analyses for Period 2 (Fig. 8b), using the EC data with the IMPROVE* temperature rise protocol, yielded similar ranges of the slope (1.07–1.46) and intercept (-0.23 – $+0.10 \mu\text{gC m}^{-3}$) with high R^2 values (0.88–0.94), except that MAAP_BC was significantly higher

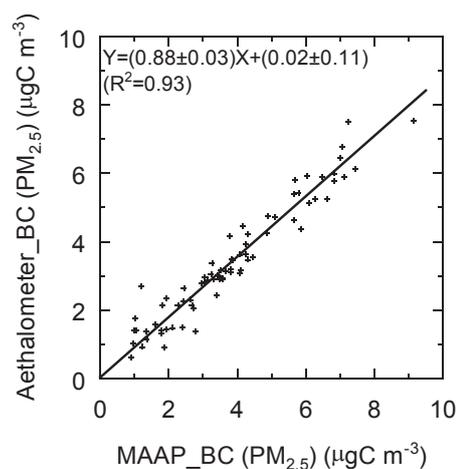


Fig. 7. Comparison of BC concentrations from the Aethalometer ($\text{PM}_{2.5}$) and MAAP ($\text{PM}_{2.5}$).

than opt-BC ($\text{MAAP_BC (PM}_{2.5}) = 1.95 \times (\text{opt-EC PM}_{2.5}) - 0.16 \mu\text{gC m}^{-3}$, with $R^2=0.91$). A similar scatter plot between the MAAP_BC and opt-EC for Period 1 for a limited data set satisfying $[\text{opt-EC}] < 2.64 \mu\text{gC m}^{-3}$ (similar to the opt-EC range during Period 2) also yielded a high slope value (2.13), suggesting opt-EC tends to give low values in the low concentration range. The MAAP vs. EC (IMPROVE*) plot for Period 2 yielded a slope of 1.45 ± 0.04 , slightly lower than 1.54 ± 0.03 for the MAAP vs. EC (NIOSH) plot for Period 1. As studied in Sect. 3.2, the IMPROVE* temperature protocol tends to give higher concentrations than does NIOSH, showing a slightly better agreement with the MAAP data.

From these analyses that yielded the ratios of the highest to lowest concentrations of ~ 2 at worst (generally ≤ 1.5), we conclude that while the differences among the BC concentrations from the four different instruments were significant, the range was still sufficiently narrow to better constrain the BC emission rate from CEC (with uncertainties similar to those of all China, a factor of 3.9 (Bond et al., 2004) and a factor of 34 (Streets et al., 2003) as noted in the Introduction), even when the model results are associated with an uncertainty of a factor of 1.5. Yamaji et al. (2008) will simulate BC concentrations using a regional-scale chemical transport model and compare them with the observations, testing the BC emission rate from CEC.

The deviations from unity of the slope values in Figs. 7 and 8 could be at least partially attributable to wrongly assumed absorption cross sections for the optical instruments. But it is not possible to re-determine the cross sections because none of the instruments can be regarded as standard. Nonetheless, it should be noted that there is a common tendency that PSAP and MAAP need to have larger cross sections than originally assumed to reduce the BC values down to the levels of EC (or that the EC values are consistently underestimated for some reasons). This tendency is studied in more detail in the next section.

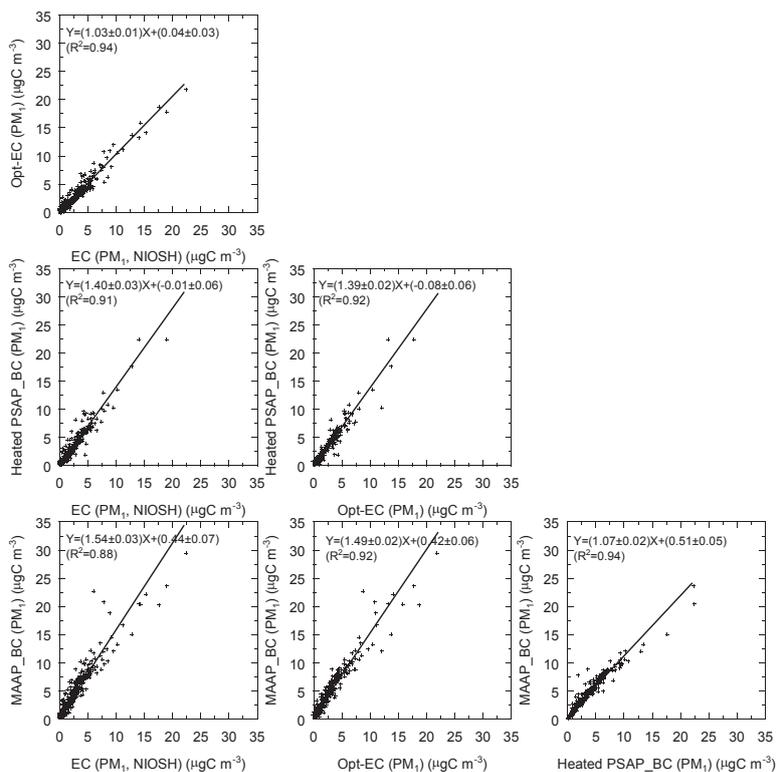


Fig. 8a. Correlations and least-square fittings for each pair among EC (NIOSH), opt-EC, heated PSAP_BC, and MAAP_BC for Period 1.

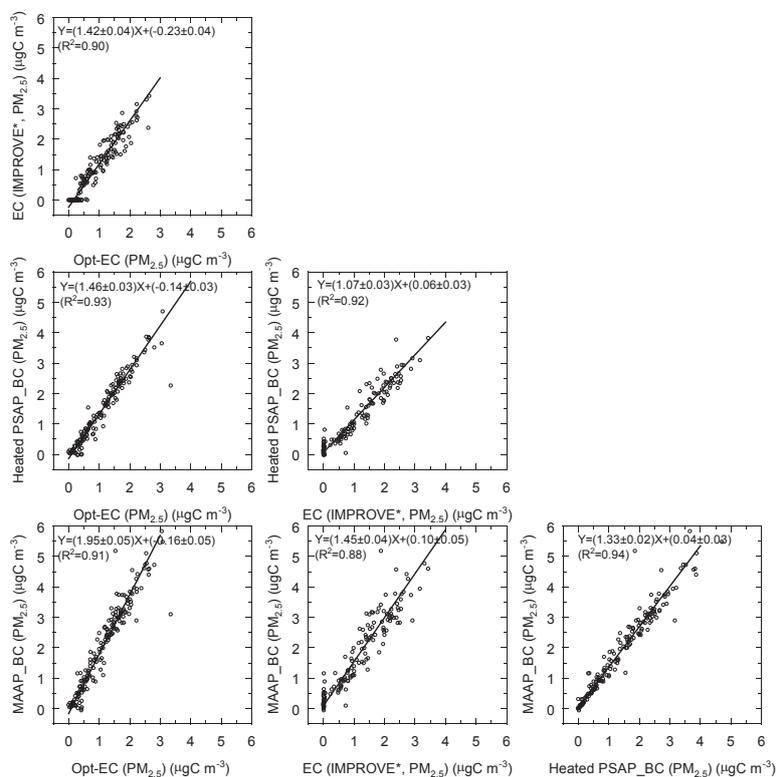


Fig. 8b. Correlations and least-square fittings for each pair among EC (IMPROVE*), opt-EC, heated PSAP_BC, and MAAP_BC for Period 2.

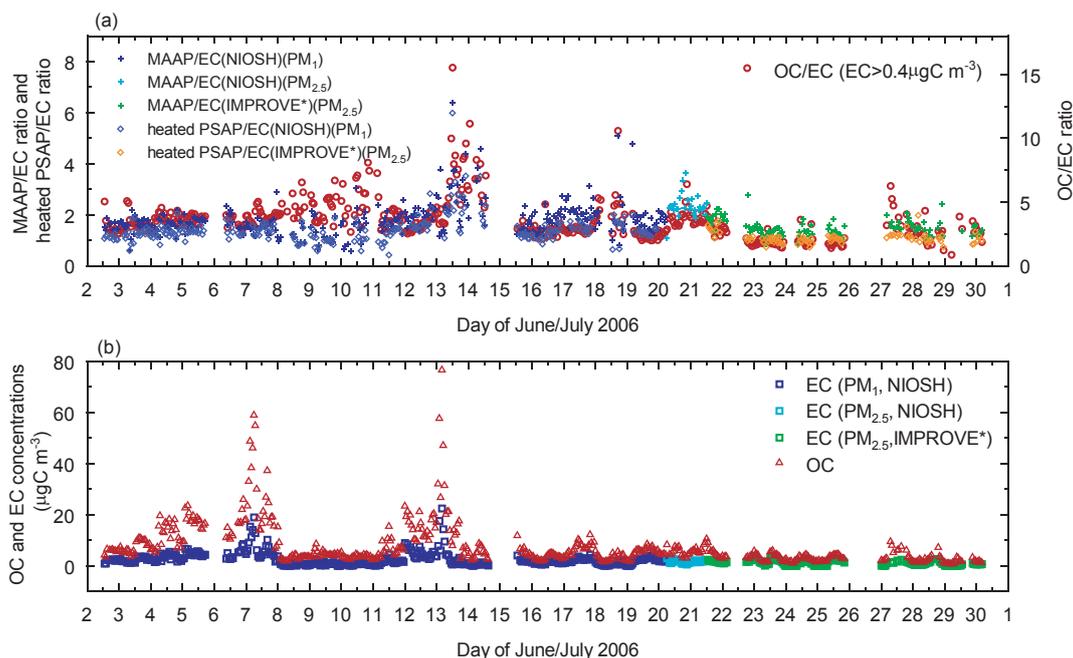


Fig. 9. (a) Temporal variations in the MAAP_{BC}/EC ratio and heated PSAP_{BC}/EC ratio (left axis) compared with temporal variations in the OC/EC ratio (red, right axis). (b) Time series of OC and EC mass concentrations observed by the ECO analyzer.

3.5 Analysis of differences between optical BC and thermal EC concentrations

Figure 9a illustrates that the MAAP_{BC}/EC and heated PSAP_{BC}/EC ratios show temporal variations similar to that of the OC/EC ratio determined by the ECO analyzer. During Period 1, the correlation coefficients were 0.59 ($n=330$) and 0.68 ($n=253$), respectively. On the afternoon of 13 June for example, the MAAP_{BC}/EC and heated PSAP_{BC}/EC ratios had elevated values together with high OC/EC ratios. It should be noted this case does not correspond to the intense biomass burning event with high OC and EC concentrations; these maxima in the BC/EC ratios occurred several hours after the event (see Fig. 9b).

There are two possible explanations. First, the optical methods might have overestimated the BC concentrations when the OC is more abundant. The OC might have coated BC particles and enhanced absorption by the lensing effect. The MAAP instrument, in principle, measures the reflected light at two different angles to cancel the scattering effect of the deposited aerosols. However, the lensing effect is not likely to be taken into account in principle. For the PSAP instrument, the heating temperature (400°C) might not have been high enough for all the OC to be volatilized. Secondly, the ECO analyzer might have underestimated EC concentrations when OC was present at relatively high concentrations. It is possible that the determined split point between OC and EC was too late, resulting in an underestimation of EC and overestimation of OC.

For the enhanced BC/EC event on the afternoon of 13 June, the raw thermogram from the ECO analyzer shows the presence of an OC fraction volatilized only at a high temperature of more than 400°C (Fig. 10a, between ca. 100 and 250 s). For this case, the BC concentrations from both the MAAP and heated PSAP instruments were 2.5 times the EC concentration and the OC/EC ratio was 7.1 ([OC]=12.9, [EC]=1.8 μgC m⁻³). This fraction of OC might not have been vaporized but deposited on the filter even in the heated mode of the PSAP measurement, leading to an overestimation of the BC concentration by enhancing the absorption, although heating was done in air (containing oxygen) for the PSAP instrument as opposed to pure He for the ECO analyzer. On the other hand, if OC and EC were split at an earlier point, the agreement between the optical BCs and EC should have been improved. When a fraction of OC vaporized only at high temperature is present, the EC and OC concentrations might be sensitive to the determination of the split point. It is also possible that the pyrolyzed fraction of OC becomes more important when the OC/EC ratio is high and subsequently the uncertain determination of the split point between the pyrolyzed carbon and the original EC might have affected the BC/EC ratios.

A contrasting case is that of 16:00 CST on 16 June, when OC mainly consisted of species volatilized at lower temperatures (Fig. 10b). The heated PSAP_{BC} was in good agreement with EC (heated PSAP_{BC}/EC ratio=1.02) and the MAAP_{BC} was higher than EC only by a factor of 1.24. The OC/EC ratio was 2.4 ([OC]=4.9, [EC]=2.0 μgC m⁻³). We were unable to conduct a more detailed analysis of the

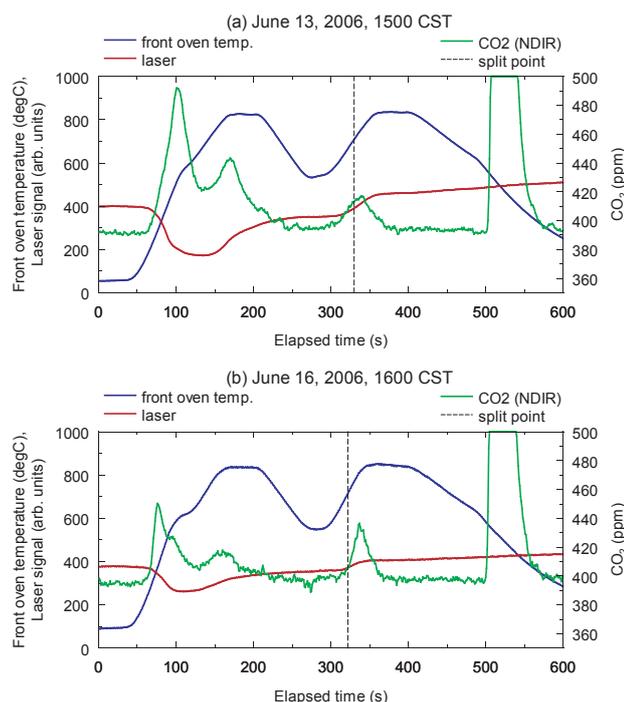


Fig. 10. Thermograms of the ECOC analysis on (a) 13 June 2006, 15:00 CST and (b) 16 June 2006, 16:00 CST. The analysis was made using the NIOSH protocol, under a He atmosphere before 255 s and an O₂/He atmosphere after that. The large peak after 500 s is for calibration.

dependence of the BC/EC ratios on the volatility of OC during the campaign, because the time period of thermal analysis was too short to subcategorize OC by volatility and quantify the subcategories individually.

In Fig. 9a, we see the MAAP_BC/EC ratio had a similar co-variation with the OC/EC ratio in Period 2 (21–30 June), for which the IMPROVE* temperature rise program was employed. The qualitative relationship between the MAAP_BC/EC ratio and the OC/EC ratio was basically unaltered by the selection of the temperature program. This may support the first possibility that the optical instruments overestimated the BC concentrations rather than the second possibility that the thermal EC measurement was underestimated. However, we cannot eliminate the second possibility because using the transmittance (not reflectance) in determining the split point could be more important than the selection of the temperature program for the late split point. The MAAP_BC/EC ratio also showed weak positive correlations ($R=0.54$, 0.49 , and 0.55) with nitrate (PM₁)/EC (PM₁, NIOSH), sulfate (PM₁)/EC (PM₁, NIOSH) and ammonium (PM₁)/EC (PM₁, NIOSH) ratios, suggesting that these species might also have enhanced absorption measured by the MAAP instrument owing to their coatings. However, the analysis was less reliable because of the limited number of aerosol filter samples ($n=21$).

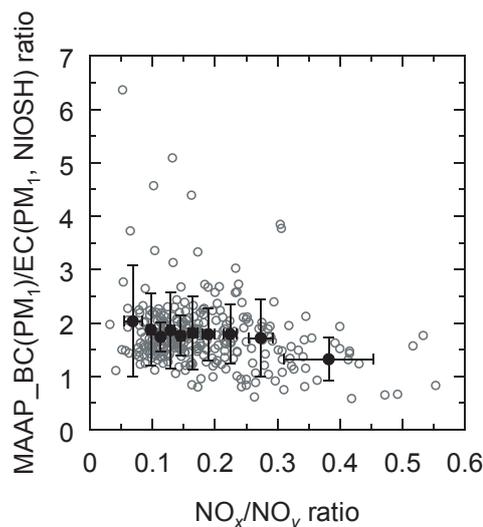


Fig. 11. Relationship between the MAAP_BC to EC ratio and the NO_x/NO_y ratio. Gray circles: hourly data; solid circles: data binned by NO_x/NO_y ratio.

Figure 11 shows the enhanced MAAP_BC/EC ratios occurred only for low NO_x/NO_y ratios, implying that aging is related to large MAAP_BC/EC ratios. Given that the tendency for underestimation of EC is not straightforwardly explained in terms of aging, it is more reasonable to conclude that the BC particles are coated by transparent materials after aging and that the high MAAP_BC/EC ratios are attributable to the overestimation by the optical instruments owing to the lensing effect induced by the coating. Moteki et al. (2007) and Shiraiwa et al. (2007) illustrated that the fraction of thickly coated black carbon particles increased on a time scale of 12 h in urban plumes from Japan. In the New England Air Quality Study field campaign in summer 2002, Kleinman et al. (2007) showed that the ratio of BC (optically measured with a PSAP instrument) to CO concentrations increased by a factor of 2.4 in the air masses in which the NO_x/NO_y ratio has decreased to 0.1, and attributed it to the increase in the mass absorption efficiency with age that could be explained by the deposition of nonabsorbing secondary aerosol and its lensing effect. Slowik et al. (2007) suggested from their laboratory studies that the reading of the MAAP instrument can be increased by 20% by a 60 nm anthracene coating.

Jeong et al. (2004) indicated that the Aethalometer overestimated BC by factors of 3.3 and 2.7 in comparison to EC concentrations (PM_{2.5}) measured with a semi-continuous Sunset Laboratory ECOC analyzer in Rochester and Philadelphia in the United States. The tendency that the optical measurements resulted in higher BC concentrations is in accordance with the results of this study. However, they observed a lower factor (0.35) during a Canadian

Table A1. Acronyms, full names, principles, operation modes, and detection limits of the instruments used in this study.

Acronym	Instrument full name	Model	Principle	Operation modes	Detection limit
MAAP	Multi-Angle Absorption Photometer	5012MAAP, Thermo	Transmittance and reflectance at two different angles	PM ₁ and PM _{2.5}	<0.1 μg m ⁻³ (2 min)
PSAP	Particle Soot Absorption Photometer	PSAP, Radiance Research	Transmittance (attenuation)	Unheated and heated inlet	~0.2 μg m ⁻³ (1 min)
ECOC	Elemental Carbon/Organic Carbon	Semi-continuous, Sunset Laboratory	Thermal evolution – optical transmittance correction Laser transmittance (attenuation) for opt-EC	NIOSH and IMPROVE* temperature programs	<0.4 μg m ⁻³ (EC, 40-min sampling)
Aethalometer	Aethalometer	AE-21, Magee Scientific	Transmittance (attenuation)	–	<0.1 μg m ⁻³ (5 min)

forest fire event in Philadelphia when OC was relatively abundant. The tendency that the optical instrument underestimated BC concentrations when OC is relatively abundant contradicts our results. Bae et al. (2007) also showed a similar trend in Gosan, Korea; an Aethalometer gave lower BC values in comparison to the thermal EC measured by a semi-continuous ECOC analyzer, when OC was relatively abundant (their case 2). Although commonly based on an optical method, the property of the Aethalometer might be different from the properties of the MAAP and PSAP. In a comprehensive comparison in Fresno, the United States, Park et al. (2006) showed the ratio of thermal EC (from a Sunset ECOC analyzer) to MAAP_BC was in the range of 0.58–0.73, consistent with the magnitude relationship in our study. Reisinger et al. (2008) implied that a MAAP instrument overestimated BC concentrations when the aerosol contained appreciable amounts of brown carbon in winter in Vienna, Austria. This seems consistent with our finding that the MAAP instrument yielded higher values than thermal EC values, especially when an OC fraction that only volatilized at a high temperature was relatively abundant.

4 Summary

We conducted optical and thermal observations of BC using four instruments at the top of Mount Tai, Shandong Province, China, in June 2006. The concentrations measured by the instruments were highly correlated and the slopes of the regression lines normally ranged between 1.0 and 1.5, with an exception of ~2 for the comparison of MAAP_BC (PM_{2.5}) and opt-EC (PM_{2.5}). The general agreement enables (1) model-to-observation comparisons of BC concentrations over this important area to narrow the emission rate, which currently has high uncertainty, (2) accurate analysis of the emission ratio (for example, the BC/CO emission ratio), and (3) a more reliable long-term observation using the MAAP instrument at Mount Tai.

In a more detailed analysis, it is inferred that the EC concentrations determined with the IMPROVE* temperature rise program for the ECOC analyzer were higher than those with the NIOSH program by 31% and showed slightly better agreement with the MAAP_BC values. The MAAP_BC/EC and heated PSAP_BC/EC ratios increased when high OC/EC ratios were reported from the ECOC analyzer. This suggested either that the optical instruments (MAAP and PSAP) overestimated BC concentrations or that the thermal instrument underestimated EC. Possible explanations for the overestimation of BC by optical instruments are that the lensing effects influenced the MAAP instrument or that less-volatile OC particles were not removed despite heating of the inlet tube of the PSAP instrument and subsequently increased the absorption by the particles deposited on the filter. The underestimation of EC by the ECOC analyzer would be explained by the possibility that some EC has evolved before the split time between OC and EC. The fact that the large MAAP_BC/EC ratios are associated with low NO_x/NO_y ratios suggests that BC particles might have been coated by transparent materials after aging and that the optical instruments have overestimated BC concentrations owing to the lensing effect. In the future, a systematic analysis studying the discrepancy as a function of air mass age should be valuable. Information on size and morphology of BC particles is also necessary because they are known to affect the mass absorption cross section of BC particles in the atmosphere (Bond and Bergstrom, 2006).

Appendix A

Summary of instruments used in this study

The acronyms, full names, principles, operation modes, and detection limits of the instruments used in this study are summarized in Table A1.

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