Atmos. Chem. Phys., 12, 6173–6184, 2012
www.atmos-chem-phys.net/12/6173/2012/
doi:10.5194/acp-12-6173-2012

Atmospheric Chemistry and Physics

Carbonaceous aerosol AAE inferred from in-situ aerosol measurements at the Gosan ABC super site, and the implications for brown carbon aerosol

C. E. Chung[1], **S.-W. Kim**[2], **M. Lee**[3], **S.-C. Yoon**[2], **and S. Lee**[2]

[1]Dept. Environmental Science & Engineering, Gwangju Institute of Science and Technology, Gwangju, Korea
[2]School of Earth & Environmental Sciences, Seoul National University, Seoul, Korea
[3]Dept. Earth & Environmental Sciences, Korea University, Seoul, Korea

Correspondence to: C. E. Chung (eddy@gist.ac.kr)

Received: 18 January 2012 – Published in Atmos. Chem. Phys. Discuss.: 8 February 2012
Revised: 8 June 2012 – Accepted: 26 June 2012 – Published: 17 July 2012

Abstract. The Mass Absorption Cross section (MAC) and Absorption Ångström Exponent (AAE) have been commonly estimated for ambient aerosols but rarely for black carbon (BC) or organic aerosol (OA) alone in the ambient conditions. Here, we provide estimates of BC (and OA) MAC and AAE in East Asian outflow, by analyzing field data collected at the Gosan ABC super site. At this site, EC (and OC) carbon mass, the aerosol absorption coefficient at 7 wavelengths and PM mass density were continuously measured from October 2009 to June 2010.

We remove the absorption data with significant dust influence using the mass ratio of PM_{10} to $PM_{2.5}$. The remaining data shows an AAE of about 1.27, which we suggest represent the average carbonaceous aerosol (CA) AAE at Gosan.

We find a positive correlation between the mass ratio of OC to EC and CA AAE, and successfully increase the correlation by filtering out data associated with weak absorption signal. After the filtering, absorption coefficient is regressed on OC and EC mass densities. BC and OA MACs are found to be 5.1 (3.8–6.1) and 1.4 (0.8–2.0) $m^2\,g^{-1}$ at 520 nm respectively. From the estimated BC and OA MAC, we find that OA contributes about 45 % to CA absorption at 520 nm. BC AAE is found to be 0.7–1.0, and is probably even lower considering the instrument bias. OA AAE is found to be 1.6–1.8. Compared with a previous estimate of OA MAC and AAE near biomass burning, our estimates at Gosan strongly suggest that the strongly-absorbing so-called brown carbon spheres are either unrelated to biomass burning or absent near the emission source.

1 Introduction

A number of field studies have been conducted to monitor the optical properties of ambient aerosols (Kim et al., 2004; Huebert et al., 2003; Jung et al., 2009; Nakajima et al., 2007). In these studies, commonly-retrieved optical properties include the single scattering albedo (SSA), and the absorption efficiency. Aerosols in the ambient atmosphere are normally a mixture of up to several aerosol species. Known aerosol species are black carbon (BC) aerosol, organic aerosol (OA), sulfate particle, dust, sea salt, nitrate particle, etc. Out of these aerosol species, BC aerosol (simply referred to as BC, as commonly done) and OA are emitted together by open biomass burning, indoor biomass combustion for cooking and heating, and fossil fuel combustion (Forster et al., 2007; Lamarque et al., 2010; Lawrence and Lelieveld, 2010). Because BC and OA tend to co-exist, rarely did past field studies retrieve the optical properties of BC (or OA) in BC-containing ambient aerosol samples. There is no guarantee that the optical properties of BC particles alone are the same as those of BC in ambient aerosols. BC (or OA) optical properties in the ambient atmosphere are very valuable, in that (a) they can be used to diagnose the BC (or OA) amount from ambient aerosol measurements, (b) they are needed to accurately compute the radiative forcing of BC (or OA) in a radiative transfer model.

In this study, we use field observations to quantify the Mass Absorption Cross section (MAC) and Absorption Ångström Exponent (AAE) for BC (and also OA). AAE describes the spectral dependence of MAC, and is defined

as: $MAC(\lambda) = C_0 \times \lambda^{-AAE}$, where λ denotes wavelength. BC MAC is used to infer BC amount in ambient aerosols (e.g. Park et al., 2005). BC (or OA) MAC is also used to determine solar radiation absorption by BC and BC SSA. Currently, the MAC and AAE of BC or OA in use are based on theoretical calculations, laboratory studies, and limited field studies. These field studies, though limited, are essential in validating the results from theoretical or laboratory studies. However, there is a need to revisit the findings from some of these field studies, in that they rely on untested assumptions. Furthermore, most of these field studies use unclear definitions of BC and OA. The present study was designed to give more robust estimates of BC (and OA) MAC and AAE in East Asian outflow. In the next, we discuss the issues of these limited field studies.

1.1 BC

The community generally uses the terms "soot", "soot carbon" and "black carbon" interchangeably (Andreae and Gelencsér, 2006), and so will we here. Soot quickly evolves into aggregates of fine particles (Katrinak et al., 1993). These fine particles are called spherules or monomers, and are commonly in the range of 20–60 nm in diameter in the ambient atmosphere (Alexander et al., 2008). Figure 1a visualizes a typical fresh BC particle. Aggregate soot particles (i.e. BC particles) consist of elemental carbon (EC) and other minor components. These minor components include organic carbon (OC) (Watson and Valberg, 2001). Here, we use "OC" to refer to organic material and not organic aerosol. OC included in aggregate soot particles was referred to as OC intrinsic to BC (Adler et al., 2010). BC thus describe particles made up largely of EC. In contrast, there is no EC in OA. Because BC contains OC, BC MAC should be clearly defined and was not so in many field studies.

While BC particles stay in the atmosphere, they become aged. The aging processes include size change due to coagulation, the oxidation of the intrinsic OC, and coating. When a BC particle is coated during the aging, the entire particle is surrounded by a coating shell (see Fig. 1b). Organics can be part of the coating shell (Moffet and Prather, 2009). We distinguish organics in the coating shell from BC-intrinsic organic. If a BC particle is coated by an organic shell, the particle should be referred to as an OA-BC internally mixed particle (or OA-BC combined particle), rather than as a BC particle. The reason is that both the coating by a two-particle collision (which implies mixing) and the coating by condensation would produce the same OA-BC particle. In summary, organics in the atmosphere exists as stand-alone organic aerosol, as internally mixed with other aerosols, as intrinsic to BC, and so on. We suspect that stand-alone OA is the dominant form of organics in the atmosphere, though to our knowledge there has not been any observational study of quantifying the portion.

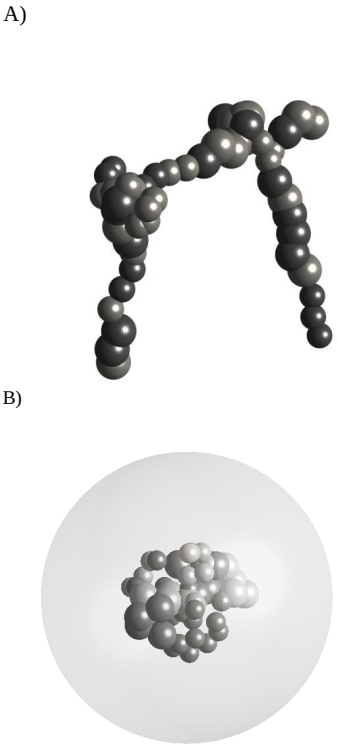

Fig. 1. (A) A typical fresh BC (Black Carbon) particle. A BC particle is made up of smaller spherical particles called spherules or monomers. Spherules in BC particles consist mainly of EC (Elemental Carbon) but also contains organics and inorganics (Watson and Valberg, 2001). Organics in BC is referred to as organics intrinsic to BC here, as in Adler et al. (2010). **(B)** An aged and coated BC particle. An entire BC particle is surrounded by a coating shell. The BC core becomes more compact when coated by shell (Schnaiter et al., 2005). The coating material was found to be sulfate, ammonium, organics, nitrate and water (Moffet and Prather, 2009).

BC is known to be strongly light-absorbing aerosol. The mass absorption cross section is an appropriate measure of absorption efficiency by BC. Ambient BC MAC was found to range from 8 to 14 $m^2\,g^{-1}$ at the mid-visible wavelength (see Table 1). One reason for such a wide range of BC MAC would be that BC absorption is enhanced by a scattering shell. A number of studies (Schnaiter et al., 2005; Moffet and Prather, 2009; Chung et al., 2012) established the enhancement of BC absorption by a scattering coating shell. Some BC particles exist without coating, some with thin shells and the rest with thick shells (Adachi and Buseck, 2008; Moffet and Prather, 2009). Fresh BC particles are not coated and become coated over time (Moffet and Prather, 2009), and thus coating should contribute significantly to the ambient BC MAC variability.

Therefore coating amplifies BC MAC but does it also change BC AAE? Theoretical studies (Gyawali et al., 2009; Chung et al., 2012) showed that coating lowers BC AAE. Although not highlighted, Schnaiter et al. (2005) showed in a

Table 1. Reported ambient aerosol Mass Absorption Cross section (MAC) in units of $m^2\,g^{-1}$. The gram unit in MAC here is for carbon. For BC MAC, the carbon mass is for its EC component only. Some studies of estimating BC MAC (e.g. Clarke et al., 2004; Quinn et al., 2004) attributed all the CA absorption to BC absorption, and we exclude such studies here. For OC MAC here, the carbon mass includes BC-intrinsic OC, and the absorption is due to organic material in the aerosols.

MAC	λ in nm	Absorption instrument	Aerosol species	site	Reference
10.0±3.5	405	PASS-3	Denuded soot		Cross et al. (2010)
8.11±1.7	532	(Photoacostic)			
4.16±0.5	781				
2.1±0.1	405	PASS-3	OC	Gosan, Korea	Flowers et al. (2010)
$\sim 3.4 \pm 0.1$		(Photoacostic)			
14	550	Optical spectrometers and MULTI	BC	Urban samples	Kirchstetter et al. (2004)
0.6	550	Optical spectrometers and MULTI	OC	SAFARI	Kirchstetter et al. (2004)
10.0	520	PSAP (Filter-based)	BC	Xianghe, China	Yang et al. (2009)
8.8	590				
0.63	520	PSAP (Filter-based)	OC	Xianghe, China	Yang et al. (2009)
0.37	590				
0.71 ± 0.2	365	UV-Visible Spectorophotometer Cell	WSOC	Beijing, China	Cheng et al. (2011)
$\sim 1.79 \pm 0.24$		and Long-Path Absorption			

laboratory study that coating enhances BC absorption more at 700 nm than at 450 nm (see Fig. 9 of their study). Relatively more enhancement of absorption at longer wavelengths leads to a lowering of AAE. Kirchstetter et al. (2004) measured BC AAE near the BC emission sources, and found its value to be near 1.0. Given an AAE of 1.0 for fresh BC, the AAE of aged BC should be lower than 1.0 since BC is coated during the aging process. In spite of this, BC AAE of 1.0 continues to be used in field studies whether BC is fresh or aged (e.g. Gadhavi and Jayaraman, 2010), perhaps because there has not been any field study of estimating the AAE of BC downstream. In the present study, we attempt to estimate BC AAE in East Asian outflow, where BC is aged and presumably mostly coated.

1.2 OA

OA is emitted both as primary aerosols and in gaseous form which is subsequently converted to aerosols, referred to as secondary organic aerosol. Some of OA is natural, associated with biogenic or marine sources (Russell et al., 2011). OA, once emitted, is oxidized and grows in mass. OA was evaluated as slightly absorbing a decade ago (OPAC data; Hess et al., 1998). Recently, Andreae and Gelencsér (2006) explained that organic material is decomposed into brown carbon (BrC) and colorless carbon, and only brown carbon absorbs. The BrC optical characteristics however seem highly uncertain. Chakrabarty et al. (2010) generated BrC as primary particles and found BrC SSA to be much higher than 0.95 at 550 nm, consistent with the BrC particles analyzed by Hoffer et al. (2006). However, Alexander et al. (2008) identified different kind of BrC particles in outflows from China and showed that their SSA is 0.44 for the average BrC particle size. The difference between the two in absorption efficiency can be as large as a factor of 10 or more. Alexander et al. (2008) referred to these strongly-absorbing BrC particles as "brown carbon spheres". For clarity, we refer to the other BrC aerosols as the weakly absorbing BrC aerosols. Issues arise. Since Alexander et al. (2008) discovered the strongly-absorbing BrC particles in East Asia, does this mean that the strongly-absorbing BrC particles are abundant in East Asia and absent elsewhere? If the strongly-absorbing BrC aerosols have global abundance, is the ratio of the weakly absorbing BrC aerosols to the strongly absorbing BrC aerosols similar between biomass burning and fossil fuel combustion?

Can BrC aerosol coat BC in the ambient atmosphere? Alexander et al. (2008) found the strongly-absorbing BrC aerosol to always exist as stand-alone particles. Thus, it appears that BrC is either seldom or never part of the BC coating shell. On the other hand, organics intrinsic to diesel soot was found to be BrC (Adler et al., 2010). Here, we use BrC to refer to absorbing organic material (and not necessarily absorbing organic aerosol) while "BrC aerosol" refers to absorbing organic aerosol.

OA AAE poses another puzzle. Kirchstetter et al. (2004) extracted organics from biomass smoke aerosols, and found the AAE to be about 6.0. By extracting the organic component, they analyzed stand-alone OA, the organic shell and organics intrinsic to BC altogether. Conversely, we find that the strongly-absorbing BrC aerosol by Alexander et al. (2008) has an AAE of < 1.0 (see Sect. 6 for details). If the strongly-absorbing BrC aerosols were only present in East Asia, OA AAE would be very different between East Asia and elsewhere. How much does OA AAE change from region to region? In the present study, we will discuss this issue.

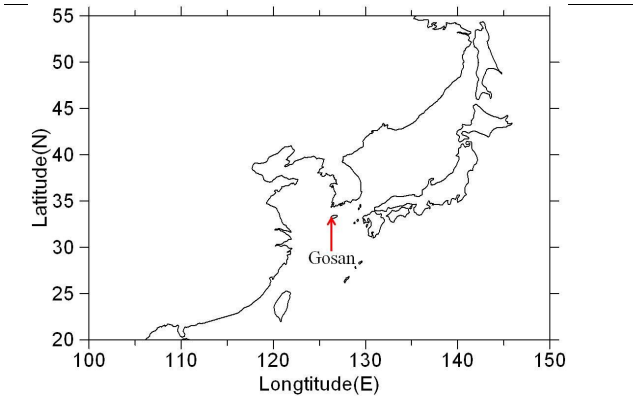

Fig. 2. Gosan is located at the western tip of Jeju Island, Korea. Gosan ABC super site is located at 33.28° N and 126.17° E.

2 Measurement

Gosan is a village located at the western tip of Jeju Island, and this island is south of the Korean Peninsula (Fig. 2). At the Gosan site, we measured EC mass, carbon mass in OC, and aerosol absorption all with a single inlet. The measurement was continuous at hourly intervals from October 2009 to May 2010, generating 6288 hourly data. Also, we collected PM (Particulate Matter) mass density and meteorology data, which were produced at the laboratory on the site operated by the National institute of Environmental Research (NIER) and Korea Meteorological Administration (KMA), respectively. The $PM_{2.5}$ and PM_{10} aerosol mass density were measured on an hourly basis using β-Ray Absorption Method. The $PM_{2.5}$ data on May 19th and 20th in 2010 were removed due to accuracy issues. We also removed the data with a warning tag. A warning tag was given when the instrument did not properly function (e.g. due to electronic power interruption) or the signal was below the detection limit.

In East Asia, air generally moves eastward. During the 9 month observation period, northwesterly winds blew at the 850 hPa height over Jeju Island overall (Fig. 3). This makes Gosan one of the ideal locations for monitoring long-range transported aerosols from China. Small-mode AOD (Aerosol Optical Depth), which effectively measures anthropogenic aerosol amounts, is largest in eastern China (Fig. 3). In view of this, the UNEP (United Nations Environment Programme) ABC (Atmospheric Brown Clouds) Project established one of the super sites at Gosan, and the Gosan site was named "Korea Climate Observatory-Gosan" (KCO-G). KCO-G collocates with an AERONET (AErosol RObotic NETwork) site.

2.1 Carbon mass density measurement

We measured the carbon mass of aerosols, referred to as TC (total carbon). TC is divided into EC (elemental carbon) and OC (organic carbon). EC and OC masses per unit volume were measured using a Model-4 Semi-Continuous OC-EC Field Analyzer (Sunset Laboratory Inc., USA). This instrument uses a thermal-optical transmittance (TOT) method with a laser-based pyrolysis correction and the National Institute for Occupational Safety and Health (NIOSH) 5040 protocol (Birch and Cary, 1996; Chow et al., 2005). The instrument collects samples on the filter for 45 min and then analyzes carbons, producing hourly data for EC and OC masses. The detection limits of EC and OC masses were determined to be 0.01 μgC m^{-3} and 0.24 μgC m^{-3}, respectively, based on three times the standard deviation of filter blanks. The sampling filter was changed when laser correction was lower than 0.88 to avoid a substantial decrease in the laser signal due to accumulation of refractory materials on the filter. This instrument has been widely used especially for long-term experiments and produced sturdy results (e.g. Polidori et al., 2006). Polidori et al. (2006) gives detailed methods and procedures.

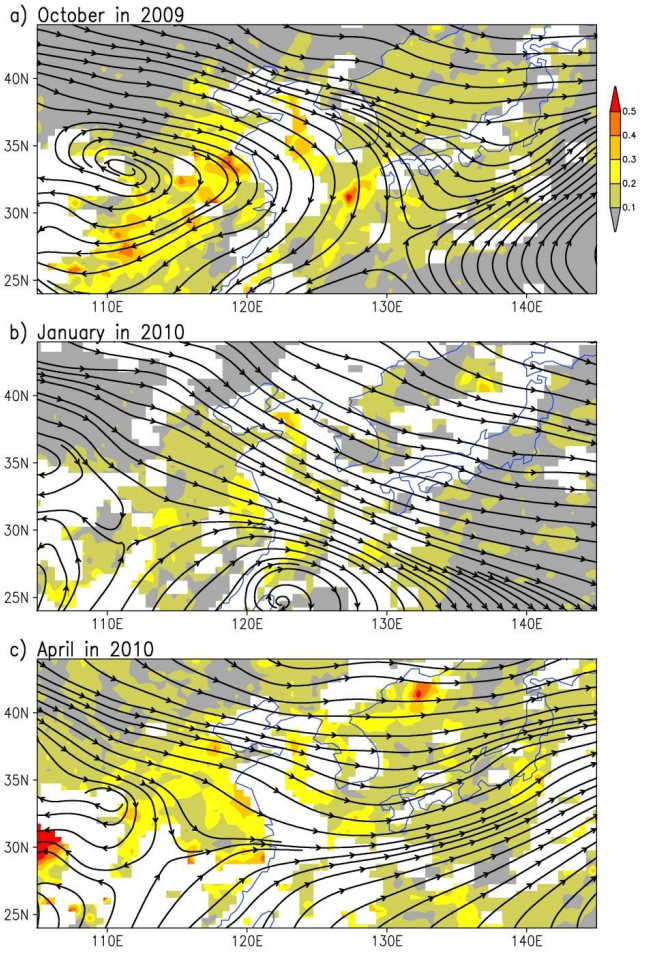

Fig. 3. 850 hPa wind stream from the ERA Interim Reanalysis (Dee et al., 2011) and MISR (Multi-angle Imaging Spectro-Radiometer) small-mode AOD at 555 nm.

2.2 Aerosol light absorption measurement

The aerosol light-absorption coefficient (σ_{ap}) was determined from the seven-channel aethalometer measurements of the attenuation of light passing through an aerosol-laden filter (Magee Scientific, AE31; Hansen, 2005). The aethalometer collects sample on a quartz fiber filter tape and performs a continuous optical analysis of filter transmission at seven wavelengths covering the ultra-violet to the near-infrared wavelength range (i.e. 370, 470, 520, 590, 660, 880 and 950 nm), with a time resolution of 5 min and 3.9 l min^{-1} of flow rate. Most of filter-based absorption instruments, such as aethalometer, suffer from various errors that needed to be corrected: (1) enhancement of light attenuation by multiple scattering by the fiber-filter substrates (multiple scattering correction), (2) enhanced attenuation due to scattering of aerosols embedded in the filter (scattering correction), (3) an increase of light attenuation with an accumulation of light absorbing particles in the filter (filter loading correction) (Collaud Coen et al., 2010). In this study, σ_{ap} was derived by the Arnott method (Arnott et al., 2005) which corrects for all of the above mentioned instrumental artifacts (Arnott et al., 2005; Collaud Coen et al., 2010).

We expect the aethalometer data to be particularly erroneous during precipitation events. Such data are manually removed in the present study. The Absorption Ångström Exponent (AAE) was determined by fitting the measured σ_{ap} at all seven wavelengths into: $\sigma_{ap}(\lambda) = C_0 \times \lambda^{-AAE}$.

3 CA AAE

Dust, BC and OA are known to be the dominant aerosol absorbers of the sunlight. Aerosols arriving at Gosan should have all the three absorbing aerosol species and scattering aerosols together in external and internal mixtures. So-called "internal mixture" delivers two different meanings to the modeling and measurement communities. We follow the modelers' definition, which refers to a type of mixing, whereby particles of different species stick together by complete mixture, coating or attachment. Even when a BC particle is coated by organics and the coating occurs through condensation, we regard this particle as a BC-OA internally mixed particle, since the outcome would be the same irrespective of whether the coating occurs through condensation or by collision between an OA and a BC particle. BC and OA are grouped as CA (Carbonaceous Aerosol).

To compute CA AAE, we assume that dust and CA contribute 100 % to solar radiation absorption by aerosols. Aerosol absorption at Gosan has dust influence at times but not always. During the observation period, there were about 11 days of Asian dust storms over Gosan. On some of the other days, dust is expected to have influenced aerosol absorption at Gosan. We attempt to compute CA AAE by first removing dust-influenced data and then computing AAE for the remaining hourly data. Our strategy of removing dust-influenced data is based on the mass density ratio of PM_{10} to $PM_{2.5}$. This ratio is referred to as $PM_{10}/PM_{2.5}$ for brevity here. Figure 4 shows the ratio from October 2009 to July 2010. The ratio is expectedly highest in spring season, when Asian dust storms commonly pass Korea. We thus use $PM_{10}/PM_{2.5}$ as an indicator of dust influence. Using $PM_{1.0}$ instead of $PM_{2.5}$ would be better but $PM_{1.0}$ measurements were only done sporadically.

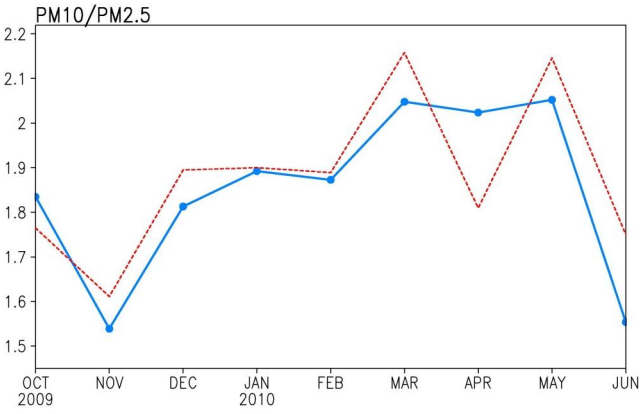

Fig. 4. Ratio of monthly PM_{10} mass density to monthly $PM_{2.5}$ mass density at Gosan. Monthly values are obtained from arithmetic mean (blue lines) and median (red lines).

We compute the average AAE over the observation period by averaging the absorption coefficient at each wavelength and then computing AAE. Without removing any data, the average AAE is found to be about 1.31. Figure 5 shows how the average AAE decreases as a result of removing data associated with high $PM_{10}/PM_{2.5}$ ratio. We interpret this as follows: Dust AAE is higher than CA AAE at Gosan. Some of the dust particles are polluted with BC attachment. By removing absorption data associated with high $PM_{10}/PM_{2.5}$ ratios, we remove both pure dust and polluted dust. Reported dust AAE values are 1.9–2.4 (Bergstrom et al., 2007; Eck et al., 2010; Russell et al., 2010). Some of the reported dust AAE values might pertain to polluted dust. Dust AAE at Gosan is expected to be lower than this range at times since some of the dust particles at Gosan travel from highly polluted areas over China. Figure 5 clearly demonstrates that dust AAE at Gosan is at least higher than 1.31.

Figure 5 shows that AAE approaches 1.27 from 1.31 as the dust influence weakens. 1.27 is likely to represent the AAE value with very little or no dust influence. We thus propose that CA AAE at Gosan is about 1.27. When only the first 4 wavelengths (i.e. 370, 470, 520, 590 nm) are used, we find the CA AAE to be about 1.2, while the CA AAE is 1.35 with the last 4 wavelengths (590, 660, 880 and 950 nm). That the total AAE and the CA AAE differ only by 0.04 (i.e. 1.31–1.27) implies that at Gosan (a) dust AAE is not much higher than CA AAE (which points to

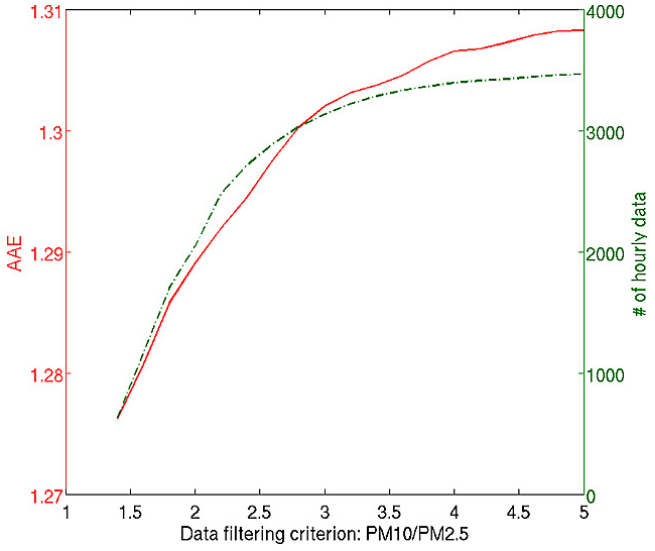

Fig. 5. AAE (Absorption Ångström Exponent), as obtained from averaged aethalometerabsorption coefficients at 370, 470, 520, 590, 660, 880 and 950 nm. Absorption data are averaged after removing the values corresponding to $PM_{10}/PM_{2.5}$ being greater than a threshold (i.e. data filtering criterion). $PM_{10}/PM_{2.5}$ refers to the mass density ratio of PM_{10} to $PM_{2.5}$, and is used as an indicator of dust influence.

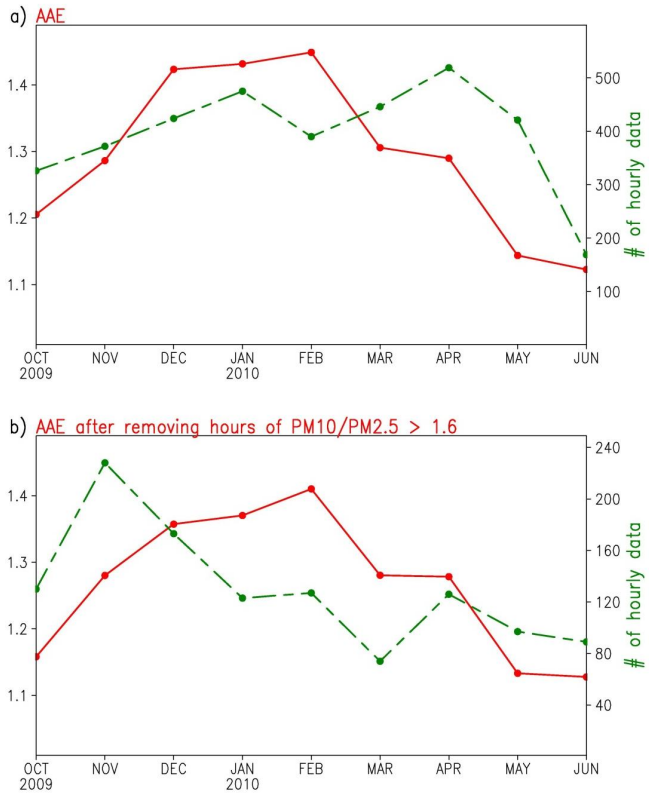

Fig. 6. Monthly-mean AAE at Gosan.

highly-polluted dust) or (b) dust contributes very little to total aerosol absorption. We also propose to filter out the data associated with $PM_{10}/PM_{2.5} > 1.6$ in order to retain the data with very little dust influence. At this filtering criterion, the AAE is very close to 1.27. After removing the data with $PM_{10}/PM_{2.5} > 1.6$, we end up with less than 30 % of the total data volume. It is very unlikely that this less-than-30 % data have significant dust influence. Using a filtering criterion of less than 1.6, though it might weaken the dust influence further, will leave us too few data. Using the criterion of greater than 1.6 increases AAE and would increase dust influence. Data with little dust influence are needed to infer the optical properties of BC and OA in our methodology. We will conduct the sensitivity of the BC and OA optical property results to the dust filtering criterion.

Figure 6a shows monthly averaged AAE. AAE is highest in winter, and is to our surprise relatively low in spring in which Asian dust storms pass commonly. This "low-AAE in spring" feature is also very strong, when we remove dust-influenced data (Fig. 6b). This explains Fig. 6a features. CA AAE is lowest in May and June during the observation period. Low CA AAE in June does not appear related to relatively higher amounts of BC over OA given the Sunset EC / OC mass data. One possible explanation for the low CA AAE in May and June is more (or thicker) BC coating by sulfate. Air is more humid in these two months, and higher humidity facilitates hygroscopic growth of sulfate aerosols. As discussed in Sect. 1, coating lowers BC AAE. Another possible explanation is that OA AAE is particularly low in these two months. As Table 2 shows, the AAE of OA rich aerosols varies depending on the source. We will also show in Sect. 6 that strongly-absorbing BrC aerosols by Alexander et al. (2008) have very low AAE values compared to other brown carbon aerosols. Strongly-absorbing BrC aerosols might abound particularly in May and June. We find it very difficult to ascertain the hypotheses, since the volume of filtered data on a monthly scale is too low to give credible answers. More studies are needed to answer these questions.

4 Filtering out collocated data errors

Absorption data from aethalometer generally have higher error-to-signal ratios when the signal is weak. The carbon mass data from the Gosan Sunset Analyzer are probably subject to the same issue. We have developed a statistical technique of reducing these data errors, to be explained next.

First, we collocated temporarily all the hourly data for the analysis. Then, the data associated with reported Asian dust storm events were eliminated to reduce dust effects. We then calculated the correlation between AAE and the mass ratio of OC to TC. We expect the correlation to be positive in view of Kirchstetter et al.'s (2004) finding that OC AAE $\gg$ BC AAE.

Table 2. Reported ambient aerosol AAE.

AAE	λ range (nm)	Site	Aerosol species	Reference
0–2.0	470–660	N. America	Pollution	Clarke et al. (2007)
1.2–2.5	470–660	N. America	Biomass burning aerosol	Clarke et al. (2007)
1.7–4.7	470–660	N. America	Mixture of dust and pollution	Clarke et al. (2007)
1.42–2.07	405–870	N. California	Biomass smoke by wildfire	Gyawali et al. (2009)
1.0	325–1000	N. America	Pollution	Russell et al. (2010)
1.45	325–1000	S. Africa	Biomass burning aerosols	Russell et al. (2010)
2.3	325–1000		Dust dominated aerosols	Russell et al. (2010)
1.3 ± 0.1	470–660	Gosan, Korea	OC rich pollution	Lee et al. (2012)
1.1 ± 0.1	470–660	Gosan, Korea	Sulfate rich pollution	Lee et al. (2012)
1.5 ± 0.1	470–660	Gosan, Korea	Polluted dust	Lee et al. (2012)
1–1.4	405–870		Burning of juniper and flowering shrubs	Lewis et al. (2008)
1.5–2.0	405–870		Burning of pines	Lewis et al. (2008)
2.5–3.5	405–870		Burning of duffs	Lewis et al. (2008)

Kirchstetter et al. (2004) analyzed biomass burning aerosols. Russell et al. (2010) supported Kirchstetter et al.'s (2004) by showing that biomass burning AAE is higher than fossil-fuel combustion aerosol AAE. Biomass burning is known to emit more primary OA over BC than fossil fuel combustion (Lamarque et al., 2010). Despite all these, we find the correlation to be only 0.06. Our hypothesis is that this weak correlation is due at least to (a) remaining dust influence, (b) data errors, (c) some of the OA particles as non-absorbing ones and others.

Figure 7 shows the correlation, as we remove the data associated with high $PM_{10}/PM_{2.5}$ ratio and low absorption at 520 nm. The correlation improves when dust influence weakens. When data associated with weak absorption at 520 nm is removed, the correlation improves as well. The correlation keeps improving until we filter out the data associated with 10 or less $10^{-6}\,m^{-1}$ in 520 nm absorption coefficient. After this threshold, the correlation fluctuates. This strongly suggests that for our collocated data the noise-to-signal ratio decreases with signal till $10 \times 10^{-6}\,m^{-1}$ in 520 nm absorption coefficient and the ratio is stable after $10 \times 10^{-6}\,m^{-1}$. When the first 4 (last 4) wavelengths are used to compute AAE instead, the ratio is stable after $8 \times 10^{-6}\,m^{-1}$ ($12 \times 10^{-6}\,m^{-1}$). In view of this, our technique of minimizing the data error is to eliminate the data associated with less than $10 \times 10^{-6}\,m^{-1}$ ($8–12 \times 10^{-6}\,m^{-1}$) in 520 nm absorption coefficient. 520 nm absorption coefficient of $10 \times 10^{-6}\,m^{-1}$ is fairly large. The threshold for the Sunset carbon mass data alone might be lower. We are not certain whether this large threshold is due mainly to aethalometer absorption data or to both the absorption data and carbon mass data. Note that we computed CA AAE in Sect. 3 without removing weak absorption data. This is because there is no reason to believe that the weak-absorption data error will show a bias in monthly-mean AAE. On the other hand, these errors can impact the relationship between OC / TC and AAE.

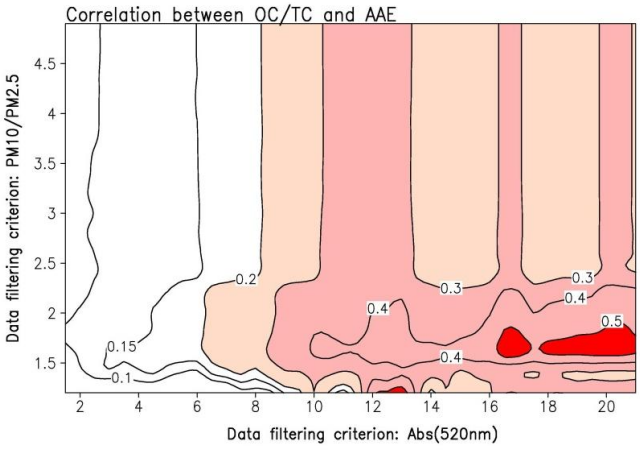

Fig. 7. Correlation between OC / TC and AAE. TC refers to total carbon (i.e. EC + OC). OC / TC refers to the mass ratio of OC to TC. Correlation is calculated in the following: Mean OC / TC and mean AAE are computed with mean OC, mean TC and mean absorption at each wavelength instead of the mean of OC / TC and the mean of AAE. Data filtering criterion based on $PM_{10}/PM_{2.5}$ is as in Fig. 5, and the data filtering criterion based on 520 nm absorption means that absorption < threshold is removed. Absorption coefficient at 520 nm is in units of $10^{-6}\,m^{-1}$.

Figure 7 gives an impression that the correlation is highest when the data with $PM_{10}/PM_{2.5} > 1.6$ are filtered out. Actually, the number of surviving data is fairly small after the data associated with $PM_{10}/PM_{2.5} > 1.6$ are removed. Figure 8 shows that the correlation rather fluctuates than decreases when the filtering criterion decreases from 1.6. In light of Figs. 5, 7 and 8, we delete the data associated with the ratio $PM_{10}/PM_{2.5}$ of 1.6 or higher, in order to minimize the dust influence on AAE.

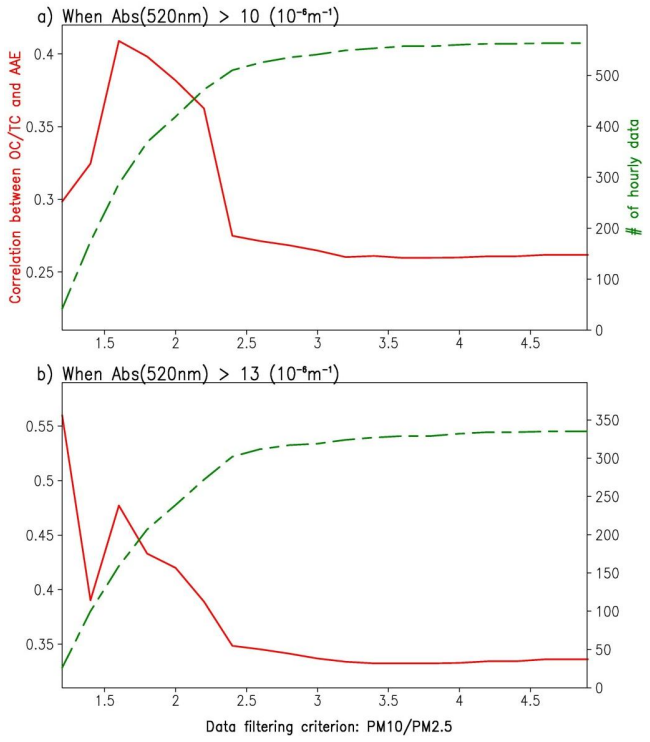

Fig. 8. Correlation between OC / TC and AAE, as a function of the data filtering criterion based on PM_{10} / $PM_{2.5}$. See Fig. 5 for the discussion of the filtering criterion. Also shown is the number of hourly data corresponding to data filtering criterion.

5 BC MAC and AAE

The temporally-collocated hourly data are removed if associated with PM_{10} / $PM_{2.5}$ of 1.6 or higher and also if the absorption coefficient is lower than $10 \times 10^{-6}\,m^{-1}$ at 520 nm. The surviving data are considered to have very little dust influence and minimized measurement errors. We fit the surviving data into the following equation:

$$\mathrm{Abs}(\lambda) = \mathrm{MAC\_BC}(\lambda) \times \mathrm{EC} + \mathrm{MAC\_OA}(\lambda) \times \mathrm{OC} \quad (1)$$

Abs(λ) refers to the absorption coefficients at different wavelengths, as measured by aethalometer, and λ denotes wavelength. EC (or OC) is measured carbon mass density by the Sunset analyzer, and MAC refers to Mass Absorption Cross section. The Eq. (1) is solved for MAC_BC(λ) and MAC_OA(λ) by regressing the absorption coefficient on EC mass density and OC mass density. The regression is done for each wavelength.

In the Eq. (1) regression method, absorption is related to EC mass and OC mass. For this method to work well, absorption related to EC should be independent of that related to OC. There has not been any empirical evidence yet that BrC coats BC, and so the independence is nearly warranted. The Eq. (1) regression method also assumes that there is no OC in BC, which however violates the results by laboratory studies (Watson and Valberg, 2001; Adler et al., 2010). The carbon mass ratio of BC-intrinsic OC to EC depends greatly on the BC emission source (Watson and Valberg, 2001), and this ratio is unknown for East Asian outflow aerosols. Thus, with an implicit assumption of no BC-intrinsic OC, we solve the Eq. (1) (see Table 3). At 520 nm, BC MAC is found to be $5.1\,m^2\,g^{-1}$ and OA MAC 1.4.

Table 3. Our standard estimates of the MAC for BC (black carbon) and OA (organic aerosol) in units of $m^2\,g^{-1}$. The gram unit in MAC here is for carbon. In this estimate, we only used the data associated with PM_{10} / $PM_{2.5} \leq 1.6$ and also absorption coefficient $\geq 10 \times 10^{-6}\,m^{-1}$ at 520 nm. It is also assumed that there is no OC in BC. The range in brackets denotes the range of obtained MAC at the 95 % confidence level.

λ in nm	BC MAC	OA MAC
370	6.0 (4.7–7.3)	2.7 (2.3–3.2)
470	5.6 (4.6–6.6)	1.7 (1.4–2.1)
520	5.1 (4.2–6.0)	1.4 (1.1–1.8)
590	4.6 (3.8–5.4)	1.2 (0.9–1.5)
660	4.2 (3.5–4.9)	1.0 (0.7–1.2)
880	3.0 (2.4–3.5)	0.6 (0.4–0.8)
950	2.6 (2.2–3.1)	0.5 (0.3–0.7)

We conduct the sensitivity of the estimated BC MAC to assumptions and chosen data filtering parameters. Table 4 lists the conducted sensitivity tests. In the first test, the presence of BC-intrinsic OC is assessed. OC mass measured by the Sunset analyzer includes this BC intrinsic component. Considering this component, our regression method should be revised to:

$$\mathrm{Abs}(\lambda) = \mathrm{MAC\_BC}(\lambda) \times (\mathrm{EC} + \mathrm{BOC}) + \mathrm{MAC\_OA}(\lambda) \times (\mathrm{OC} - \mathrm{BOC}), \quad (2)$$

where BOC refers to BC-intrinsic OC. When we assume that BC-intrinsic OC carbon mass is 20 % of EC mass, for example, we find that BC MAC decreases slightly. We also change the dust filtering criterion and the criterion of deleting weak absorption data within the uncertainty range. Taken together with statistical significance, BC MAC is estimated to be between 3.8 and $6.1\,m^2\,g^{-1}$ at 520 nm.

Overall, our estimated BC MAC is about $5.0\,m^2\,g^{-1}$ at 520 nm at Gosan. This value is much lower than the reported range in Table 1. It is beyond the scope of the present study to fully investigate the reasons for the difference between our BC MAC and previous estimates. Previous studies of estimating BC AAE were conducted with different locations, and different methodologies. Another difference between the present study and previous studies is measurement instrument. Aethalometer is a filter based device. When coated BC particles reach the filter, the coating shell might get distorted or destroyed. In view of this, BC MAC at Gosan is likely higher than indicated by aethalometer data.

Table 4. Sensitivity of estimated AAE. In a sensitivity experiment where BC contains OC, OA MAC refers to OA absorption cross section divided by the carbon mass in OC (i.e. OC in OA + BC-intrinsic OC). AAE is computed using MAC at all of the 7 wavelengths.

	BC AAE	OA AAE
Standard (PM_{10} / $PM_{2.5} \leq 1.6$ and absorption coefficient $\geq 10 \times 10^{-6}\,m^{-1}$ at 520 nm; no BC-intrinsic OC)	0.9	1.72
Standard except that BC-intrinsic OC = 20 % of EC	0.95	1.72
Standard except $PM_{10}/PM_{2.5} \leq 2.0$	1.0	1.82
Standard except absorption coefficient $\geq 8 \times 10^{-6}\,m^{-1}$	1.02	1.59
Standard except absorption coefficient $\geq 12 \times 10^{-6}\,m^{-1}$	0.68	1.68
Data not used in standard	1.23	1.38

BC AAE is estimated to be about 0.9 by solving the Eq. (1) with the standard data filtering parameters, and is estimated to be from 0.68 to 1.02 by applying various sensitivity tests (Table 4). BC AAE is computed from BC MAC values at 7 wavelengths. Kirchstetter et al. (2004) estimated BC AAE near the BC emission source, and found its value to be near 1.0 (0.7–1.3). Our inferred BC AAE of about 0.7–1.0 tends to be somewhat lower than estimated by Kirchstetter et al. (2004), though the difference is not very significant. Kirchstetter et al. (2004) used different measurement devices, and furthermore identified BC absorption by extracting OC (thus also extracting OC within BC). Therefore, a direct comparison with the study by Kirchstetter et al. (2004) should be made carefully. Given the tendency of aethalometer to destroy some of the BC coatings, we propose that the true BC AAE is likely lower than 0.7–1.0 at Gosan. Thus, our estimates of BC AAE demonstrate that aged BC has less than 1.0 for AAE.

6 OA MAC and AAE, and implications for BrC

OA MAC is estimated to be 1.4 (0.8–2.0) m^2 g at 520 nm, by applying the standard data filtering parameters, performing various sensitivity tests as in Table 4, and considering statistical significance. Together with our estimated BC MAC, we can then partition CA absorption between OA and BC at Gosan. The OC mass and EC mass densities averaged over the 9 month observation period are 2.15 and 0.73 $\mu g\,m^{-3}$ respectively. Thus, we find that at 520 nm OA contributes about 45 % to CA absorption while BC contributes the rest 55 %. OA absorption is zero or miniscule compared to BC absorption in climate models, and our results ask for significant changes in OA absorption treatment in the models.

How does our OA MAC estimate compare with previous estimates? Table 1 lists the reported values for OC MAC. In this table, we use a term "OC MAC", since the previous estimates included BC-intrinsic OC. It is not certain how much difference there is between so-called OC MAC and OA MAC. Since BC-intrinsic OC is part of BC, it is more reasonable to exclude BC-intrinsic OC and estimate OA MAC. In our methodology, we find that estimated OA MAC is very insensitive to the BC intrinsic OC component (from Eq. 2). The OA MAC estimate by Flowers et al. (2010) was derived by assuming a BC MAC value. The OA MAC estimate by Yang et al. (2009) was derived by assuming that there is no OA absorption at long wavelengths. The only OA MAC estimate without such assumptions is by Kirchstetter et al. (2004) who analyzed biomass burning aerosols in Africa. We emphasize that the samples they analyzed were collected near the sources.

Our OA MAC value is much larger than that by Kirchstetter et al. (2004). A much higher absorption efficiency for OA in our estimate indicates relatively more strongly-absorbing BrC aerosols at Gosan than near biomass burning sources. Alexander et al. (2008) named these strongly-absorbing BrC particles "brown carbon spheres", and noticed that brown carbon spheres were distinct from previously-noted brown carbon particle such as tar ball. The source of brown carbon spheres has yet to be known. Alexander et al. (2008) discovered the brown carbon spheres over the Yellow Sea – a sea between China and Korea.

Our OA AAE estimate is 1.7 (1.6–1.8) using OA MAC over 370–950 nm (Table 4). If we interpolate the OA MAC values onto 440, 675 and 870 nm, OA AAE changes insignificantly. 440, 675 and 870 nm are the wavelengths for AERONET AAOD (Absorption AOD). Our estimated OA AAE of about 1.7 at Gosan is significantly lower than the OC AAE by Kirchstetter et al. (2004). In Fig. 9, the OC AAE by Kirchstetter et al. (2004) is compared with the Gosan OA AAE. In our methodology, estimated OA AAE is very insensitive to the BC intrinsic OC component (Table 4). It is not certain how much difference there would be between OC AAE and OA AAE in the OC extraction methodology used by Kirchstetter et al. (2004), but the difference between our OA AAE and Kirchstetter et al.'s (2004) OC AAE is significant enough to justify the comparison in Fig. 9.

To better understand the Gosan OA AAE, we also show in Fig. 9 the AAE of the brown carbon spheres. Using the BrC refractive index and CMD (Count Median Diameter) of 230 nm from Alexander et al. (2008), we computed absorption cross section at different wavelengths and AAE. In the computation, GSD (Geometric Standard Deviation) of 1.5

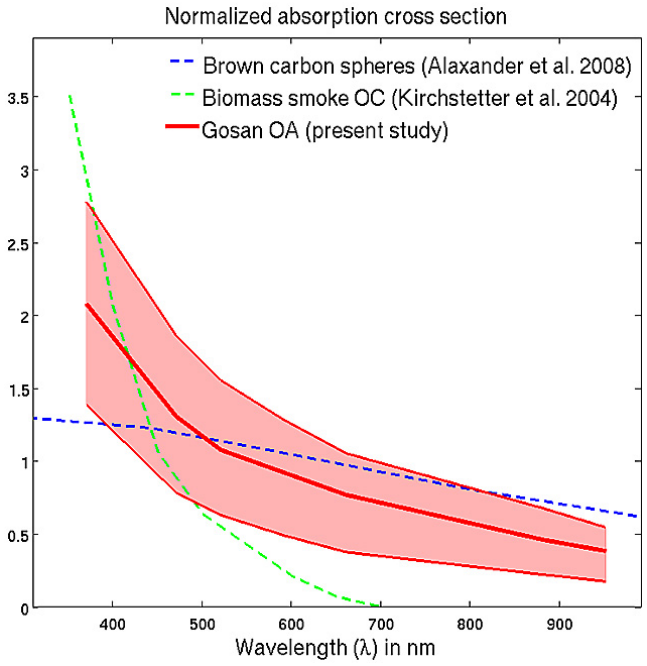

Fig. 9. Absorption cross section divided by its average w.r.t. wavelength. Biomass smoke OC refers to organic material (and not organic aerosol) in biomass burning aerosols. In this case, OC absorption was estimated by removing the OC component in the aerosol samples and measuring the absorption for the remaining aerosols. Gosan OA is represented by a thick red line (corresponding to our standard estimate) and shading. This shading shows the uncertainty of our estimate due to data shortage and uncertainties in the chosen parameters for data processing.

in a log normal size distribution was assumed. A standard Mie scattering code was used for absorption calculation. As Fig. 9 shows, the strongly-absorbing BrC aerosols have an even lower AAE than the Gosan OA AAE. At 800 nm or higher, the strong-absorbing BrC aerosols still show strong absorption, and the Gosan OA has sizable absorption, while the OC analyzed by Kirchstetter et al. (2004) shows no absorption. Figure 9 further supports our view that the strong-absorbing BrC particles are abundant at Gosan and rare near biomass burning sources.

That the Gosan OA AAE differs significantly from the strongly-absorbing BrC AAE compels that organic aerosols at Gosan include the strongly-absorbing BrC and other brown carbon particles both. The other brown carbon particles should collectively have very high AAE values so as to explain an OC AAE of 1.7 at Gosan.

Furthermore, we can propose two scenarios to justify the OA MAC and AAE at Gosan:

1. The strongly-absorbing BrC aerosols abound in East Asia and they seldom exist in biomass burning aerosols. Biomass burning produces the type of brown carbon that collectively has high AAE and low absorption efficiency. Kirchstetter et al. (2004) analyzed this particular brown carbon.

2. The strongly-absorbing BrC aerosols are abundant in both East Asia and biomass burning, but not near the source. The aerosol samples at Gosan represent the East Asian outflow, while Kirchstetter et al. (2004) obtained the samples near the source.

We are not certain which scenario is true. Alexander et al. (2008) speculated that their BrC particles belong to secondary organic aerosols. If the strongly-absorbing BrC aerosol is indeed secondary organic aerosol, the scenario 2 is more likely. Since the strongly-absorbing BrC particles have higher absorption efficiency and lower AAE than other brown carbon, the scenario 2 also implies that OA absorption efficiency increases and OC AAE decreases during the transport. More studies are needed to confirm it.

7 Suggestions

While conducting this study, we realized that very few observational studies had been done to reveal the optical properties of OA and BC separately in the ambient conditions. Most observational studies focus on characterizing overall aerosols. Some studies (e.g. Yang et al., 2009; Flowers et al., 2010) attempted to derive OA AAE but only with an assumption on the optical properties of BC or OA. In the present study, we attempted to derive the AAE and MAC of BC and OA without making any assumption on the optical properties of BC and OA. Kirchstetter et al. (2004)'s study is the only previous study we know of that is comparable to our study.

In light of this, we hope that observational studies will be launched to investigate the following issues.

a. BC, once emitted, is aged over time. Aged BC differs from fresh BC in size distribution, whether coated or not, etc. How do we describe BC AAE and MAC as functions of BC age in the ambient atmosphere?

b. How do ambient OA AAE and MAC in biomass burning areas differ from those in fossil fuel combustion areas overall?

c. How do ambient OA AAE and MAC change from near the emission source to the downstream areas?

d. We find that OA MAC at Gosan is larger than previously estimated near biomass burning. Does this mean that OA SSA would also be smaller at Gosan than near biomass burning? What is the global distribution of OA SSA? OA SSA can make significant impacts on the OA top-of-the-atmosphere radiative forcing (including the sign of the forcing).

Definite answers to the above issues will lead to drastic improvement of aerosol-chemistry models and aerosol climate forcing estimates.

Acknowledgements. The authors are thankful to H. Cha for his technical help and J. Lee of Chosun Univ. for her expertise on carbon measurement. This work was supported by the Korea Meteorological Administration Research and Development Program. Specifically, C. E. Chung's participation was supported by CATER 2012-7100, M. Lee's participation by CATER 2012-7102, and S.-C. Yoon's participation by CATER 2012-3020. S.-C. Yoon's participation was also supported by the BK21 program of the School of Earth and Environmental Sciences at Seoul National University.

Edited by: T. Kirchstetter

References

Adachi, K. and Buseck, P. R.: Internally mixed soot, sulfates, and organic matter in aerosol particles from Mexico City, Atmos. Chem. Phys., 8, 6469–6481, doi:10.5194/acp-8-6469-2008, 2008.

Adler, G., Riziq, A. A., Erlick, C., and Rudich, Y.: Effect of intrinsic organic carbon on the optical properties of fresh diesel soot, P. Natl. Acad. Sci. USA, 107, 6699–6704, 2010.

Alexander, D. T. L., Crozier, P. A., and Anderson, J. R.: Brown carbon spheres in East Asian outflow and their optical properties, Science, 321, 833–836, 2008.

Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131–3148, doi:10.5194/acp-6-3131-2006, 2006.

Arnott, W. P., Hamasha, K., Moosmüller, H., Sheridan, P. J., and Ogren, J. A.: Towards aerosol light-absorption measurements with a 7-wavelength aethalometer: Evaluation with a photoacoustic instrument and 3-wavelength nephelometer, Aerosol Sci. Tech., 39, 17–29, 2005.

Bergstrom, R. W., Pilewskie, P., Russell, P. B., Redemann, J., Bond, T. C., Quinn, P. K., and Sierau, B.: Spectral absorption properties of atmospheric aerosols, Atmos. Chem. Phys., 7, 5937–5943, doi:10.5194/acp-7-5937-2007, 2007.

Birch, M. E. and Cary, R. A.: Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust, Aerosol Sci. Tech., 25, 221–241, 1996.

Chakrabarty, R. K., Moosmüller, H., Chen, L.-W. A., Lewis, K., Arnott, W. P., Mazzoleni, C., Dubey, M. K., Wold, C. E., Hao, W. M., and Kreidenweis, S. M.: Brown carbon in tar balls from smoldering biomass combustion, Atmos. Chem. Phys., 10, 6363–6370, doi:10.5194/acp-10-6363-2010, 2010.

Cheng, Y., He, K.-B., Zheng, M., Duan, F.-K., Du, Z.-Y., Ma, Y.-L., Tan, J.-H., Yang, F.-M., Liu, J.-M., Zhang, X.-L., Weber, R. J., Bergin, M. H., and Russell, A. G.: Mass absorption efficiency of elemental carbon and water-soluble organic carbon in Beijing, China, Atmos. Chem. Phys., 11, 11497–11510, doi:10.5194/acp-11-11497-2011, 2011.

Chow, J. C., Watson, J. G., Louie, P. K. K., Chen, L. W. A., and Sin, D.: Comparison of $PM_{2.5}$ carbon measurement methods in Hong Kong, China, Environ. Pollut., 137, 334–344, 2005.

Chung, C. E., Lee, K., and Müller, D.: Effect of internal mixture on black carbon radiative forcing, Tellus B, 64, 1–13, doi:10.3402/tellusb.v64i0.10925, 2012.

Clarke, A. D., Shinozuka, Y., Kapustin, V. N., Howell, S., Huebert, B., Doherty, S., Anderson, T., Covert, D., Anderson, J., Hua, X., Moore, K. G., II, McNaughton, C., Carmichael, G., and Weber, R.: Size distributions and mixtures of dust and black carbon aerosol in Asian outflow: Physiochemistry and optical properties, J. Geophys. Res., 109, D15S09, doi:10.1029/2003jd004378, 2004.

Clarke, A., McNaughton, C., Kapustin, V., Shinozuka, Y., Howell, S., Dibb, J., Zhou, J., Anderson, B., Brekhovskikh, V., Turner, H., and Pinkerton, M.: Biomass burning and pollution aerosol over North America: Organic components and their influence on spectral optical properties and humidification response, J. Geophys. Res.-Atmos., 112, D12S18, doi:10.1029/2006JD007777, 2007

Collaud Coen, M., Weingartner, E., Apituley, A., Ceburnis, D., Fierz-Schmidhauser, R., Flentje, H., Henzing, J. S., Jennings, S. G., Moerman, M., Petzold, A., Schmid, O., and Baltensperger, U.: Minimizing light absorption measurement artifacts of the Aethalometer: evaluation of five correction algorithms, Atmos. Meas. Tech., 3, 457–474, doi:10.5194/amt-3-457-2010, 2010.

Cross, E. S., Onasch, T. B., Ahern, A., Wrobel, W., Slowik, J. G., Olfert, J., Lack, D. A., Massoli, P., Cappa, C. D., Schwarz, J. P., Spackman, J. R., Fahey, D. W., Sedlacek, A., Trimborn, A., Jayne, J. T., Freedman, A., Williams, L. R., Ng, N. L., Mazzoleni, C., Dubey, M., Brem, B., Kok, G., Subramanian, R., Freitag, S., Clarke, A., Thornhill, D., Marr, L. C., Kolb, C. E., Worsnop, D. R., and Davidovits, P.: Soot Particle Studies – Instrument Inter-Comparison – Project Overview, Aerosol Sci. Tech., 44, 592–611, doi:10.1080/02786826.2010.482113, 2010.

Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M. A., Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A. C. M., van de Berg, L., Bidlot, J., Bormann, N., Delsol, C., Dragani, R., Fuentes, M., Geer, A. J., Haimberger, L., Healy, S. B., Hersbach, H., Hólm, E. V., Isaksen, L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A. P., Monge-Sanz, B. M., Morcrette, J. J., Park, B. K., Peubey, C., de Rosnay, P., Tavolato, C., Thépaut, J. N., and Vitart, F.: The ERA-Interim reanalysis: configuration and performance of the data assimilation system, Q. J. Roy. Meteor. Soc., 137, 553–597, 2011.

Eck, T. F., Holben, B. N., Sinyuk, A., Pinker, R. T., Goloub, P., Chen, H., Chatenet, B., Li, Z., Singh, R. P., Tripathi, S. N., Reid, J. S., Giles, D. M., Dubovik, O., O'Neill, N. T., Smirnov, A., Wang, P., and Xia, X.: Climatological aspects of the optical properties of fine/coarse mode aerosol mixtures, J. Geophys. Res., 115, D19205, doi:10.1029/2010jd014002, 2010.

Flowers, B. A., Dubey, M. K., Mazzoleni, C., Stone, E. A., Schauer, J. J., Kim, S.-W., and Yoon, S. C.: Optical-chemical-microphysical relationships and closure studies for mixed carbonaceous aerosols observed at Jeju Island; 3-laser photoacoustic spectrometer, particle sizing, and filter analysis, Atmos. Chem. Phys., 10, 10387–10398, doi:10.5194/acp-10-10387-2010, 2010.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schulz, M., and Van Dorland, R.: Changes in Atmospheric Constituents and in Radiative Forcing, in: Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Av-

eryt, K. B., Tignor, M., and Miller, H. L.. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

Gadhavi, H. and Jayaraman, A.: Absorbing aerosols: contribution of biomass burning and implications for radiative forcing, Ann. Geophys., 28, 103–111, doi:10.5194/angeo-28-103-2010, 2010.

Gyawali, M., Arnott, W. P., Lewis, K., and Moosmüller, H.: In situ aerosol optics in Reno, NV, USA during and after the summer 2008 California wildfires and the influence of absorbing and non-absorbing organic coatings on spectral light absorption, Atmos. Chem. Phys., 9, 8007–8015, doi:10.5194/acp-9-8007-2009, 2009.

Hansen, A. D.: The Aethalometer – User manual, 2005.

Hess, M., Koepke, P., and Schult, I.: Optical Properties of Aerosols and Clouds: The Software Package OPAC, B. Am. Meteorol. Soc., 79, 831–844, 1998.

Hoffer, A., Gelencsér, A., Guyon, P., Kiss, G., Schmid, O., Frank, G. P., Artaxo, P., and Andreae, M. O.: Optical properties of humic-like substances (HULIS) in biomass-burning aerosols, Atmos. Chem. Phys., 6, 3563–3570, doi:10.5194/acp-6-3563-2006, 2006.

Huebert, B. J., Bates, T., Russell, P. B., Shi, G., Kim, Y. J., Kawamura, K., Carmichael, G., and Nakajima, T.: An overview of ACE-Asia: Strategies for quantifying the relationships between Asian aerosols and their climatic impacts, J. Geophys. Res., 108, 8633, doi:10.1029/2003jd003550, 2003.

Jung, J., Lee, H., Kim, Y. J., Liu, X., Zhang, Y., Hu, M., and Sugimoto, N.: Optical properties of atmospheric aerosols obtained by in situ and remote measurements during 2006 Campaign of Air Quality Research in Beijing (CAREBeijing-2006), J. Geophys. Res., 114, D00G02, doi:10.1029/2008jd010337, 2009.

Katrinak, K. A., Rez, P., Perkes, P. R., and Buseck, P. R.: Fractal geometry of carbonaceous aggregates from an urban aerosol, Environ. Sci. Tech., 27, 539–547, 1993.

Kim, K. W., He, Z., and Kim, Y. J.: Physicochemical characteristics and radiative properties of Asian dust particles observed at Kwangju, Korea, during the 2001 ACE-Asia intensive observation period, J. Geophys. Res., 109, D19S02, doi:10.1029/2003jd003693, 2004.

Kirchstetter, T. W., Novakov, T., and Hobbs, P. V.: Evidence that the spectral dependence of light absorption by aerosols is affected by organic carbon, J. Geophys. Res., 109, D21208, doi:10.1029/2004jd004999, 2004.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K., and van Vuuren, D. P.: Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application, Atmos. Chem. Phys., 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.

Lawrence, M. G. and Lelieveld, J.: Atmospheric pollutant outflow from southern Asia: a review, Atmos. Chem. Phys., 10, 11017–11096, doi:10.5194/acp-10-11017-2010, 2010.

Lee, S., Yoon, S. C., Kim, S. W., Kim, Y. P., Ghim, Y. S., Kim, J. H., Kang, C. H., Kim, Y. J., Chang, L. S., and Lee, S. J.: Spectral dependency of light scattering/absorption and hygroscopicity of pollution and dust aerosols in Northeast Asia, Atmos. Environ., 50, 246–254, doi:10.1016/j.atmosenv.2011.12.026, 2012.

Lewis, K., Arnott, W. P., Moosmüller, H., and Wold, C. E.: Strong spectral variation of biomass smoke light absorption and single scattering albedo observed with a novel dual-wavelength photoacoustic instrument, J. Geophys. Res., 113, D16203, doi:10.1029/2007jd009699, 2008.

Moffet, R. C. and Prather, K. A.: In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates, P. Natl. Acad. Sci., 106, 11872, doi:10.1073/pnas.0900040106, 2009.

Nakajima, T., Yoon, S.-C., Ramanathan, V., Shi, G.-Y., Takemura, T., Higurashi, A., Takamura, T., Aoki, K., Sohn, B.-J., Kim, S.-W., Tsuruta, H., Sugimoto, N., Shimizu, A., Tanimoto, H., Sawa, Y., Lin, N.-H., Lee, C.-T., Goto, D., and Schutgens, N.: Overview of the Atmospheric Brown Cloud East Asian Regional Experiment 2005 and a study of the aerosol direct radiative forcing in east Asia, J. Geophys. Res., 112, D24S91, doi:10.1029/2007jd009009, 2007.

Park, R. J., Jacob, D. J., Palmer, P. I., Clarke, A. D., Weber, R. J., Zondlo, M. A., Eisele, F. L., Bandy, A. R., Thornton, D. C., Sachse, G. W., and Bond, T. C.: Export efficiency of black carbon aerosol in continental outflow: Global implications, J. Geophys. Res.-Atmos., 110, D11205, doi:10.1029/2004JD005432, 2005.

Polidori, A., Turpin, B., Lim, H. J., Cabada, J., Subramanian, R., Pandis, S., and Robinson, A.: Local and regional secondary organic aerosol: Insights from a year of semi-continuous carbon measurements at Pittsburgh, Aerosol Sci. Tech., 40, 861–872, 2006.

Quinn, P. K., Coffman, D. J., Bates, T. S., Welton, E. J., Covert, D. S., Miller, T. L., Johnson, J. E., Maria, S., Russell, L., Arimoto, R., Carrico, C. M., Rood, M. J., and Anderson, J.: Aerosol optical properties measured on board the Ronald H. Brown during ACE-Asia as a function of aerosol chemical composition and source region, J. Geophys. Res.-Atmos., 109, D19S01, doi:10.1029/2003JD004010, 2004.

Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O., and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, Atmos. Chem. Phys., 10, 1155–1169, doi:10.5194/acp-10-1155-2010, 2010.

Russell, L. M., Bahadur, R., and Ziemann, P. J.: Identifying organic aerosol sources by comparing functional group composition in chamber and atmospheric particles, P. Natl. Acad. Sci. USA, 108, 3516–3521, 2011.

Schnaiter, M., Linke, C., Möhler, O., Naumann, K. H., Saathoff, H., Wagner, R., Schurath, U., and Wehner, B.: Absorption amplification of black carbon internally mixed with secondary organic aerosol, J. Geophys. Res., 110, D19204, doi:10.1029/2005jd006046, 2005.

Watson, A. Y. and Valberg, P. A.: Carbon Black and Soot: Two Different Substances, AIHAJ – American Industrial Hygiene Association, 62, 218–228, doi:10.1080/15298660108984625, 2001.

Yang, M., Howell, S. G., Zhuang, J., and Huebert, B. J.: Attribution of aerosol light absorption to black carbon, brown carbon, and dust in China – interpretations of atmospheric measurements during EAST-AIRE, Atmos. Chem. Phys., 9, 2035–2050, doi:10.5194/acp-9-2035-2009, 2009.