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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

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

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Abstract

Carbon mass of aerosols and its division between EC and OC sources were continuously measured at hourly intervals from October 2009 to June 2010. During this 9-month period, we also measured the aerosol absorption coefficient at 7 wavelengths and obtained PM mass density data at 1-h resolution. The measurement was made at the Gosan ABC super site, which is an ideal location for monitoring long-range transported aerosols from China.

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 Absorption Ångström Exponent (AAE) of about 1.27, which we suggest represent the average carbonaceous aerosol (CA) AAE at Gosan. CA AAE is highest in winter, in which the monthly value is near 1.4.

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. Black carbon (BC) and organic aerosol (OA) absorption cross sections per carbon mass are found to be 5.1 (4.2–6.0) and 1.4 (1.1–1.8) $m^2\,g^{-1}$ at 520 nm respectively. From the estimated BC & OA MAC, we find that OA contributes about 45 % to CA absorption at 520 nm. OA AAE is found to be 1.7 (1.4–2.1). Compared with a previous estimate of OA MAC and AAE, 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

Black carbon (BC) aerosol and organic aerosol (OA) are emitted together by open biomass burning, indoor biomass combustion for cooking and heating, and fossil fuel combustion (IPCC-AR4-Chapter-2 et al., 2007; Lamarque et al., 2010; Lawrence and Lelieveld, 2010). While all the BC aerosols are emitted as primary particles, OA is emitted both as primary aerosols and in gaseous form which is subsequently converted

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to aerosols, referred to as secondary organic aerosol. OA, once emitted, is oxidized and grows in mass. We use OA or OC (organic carbon) aerosol to refer to organic aerosols while OC represents the carbon component of organic matter. We use "BC" to refer to BC aerosol, as commonly done.

Both BC and OA are carbon based, and are thus commonly referred to as carbonaceous aerosols (CA). Carbonaceous aerosols originate from incomplete combustion, since all the carbon would be emitted as CO_2 in complete combustion. How do BC and OA differ?

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 (or BC particles) consist of elemental carbon (EC) and other minor components. These minor components include OC (Watson and Valberg, 2001). 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.

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 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. 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.

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BC is known to be strongly light-absorbing aerosol. The light absorption characteristics of BC depend on a lot of factors. BC MAC (Mass Absorption Cross section), for example, was found to range from 8 to 14 $m^2\,g^{-1}$ at the mid-visible wavelength (see Table 3). BC MAC should vary, in view of various refractive indices of BC material (Bond and Bergstrom, 2006), different BC sizes, etc. Another reason for such a wide range of BC MAC is 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. When BC is coated by scattering material, all the absorption is attributed to BC. Some BC particles exist without coating, some with thin shells and the rest with thick shells (Adachi and Buseck, 2008; Moffet and Prather, 2009), and so coating should contribute significantly to the ambient BC MAC range.

Coating amplifies BC MAC. Theoretical studies (Gyawali et al., 2009; Chung et al., 2012) showed that coating also lowers BC Absorption Ångström Exponent (AAE). AAE is defined as: $\text{Absorption}(\lambda) = C_0 \times \lambda^{-\text{AAE}}$. AAE describes the spectral dependence of absorption, and is another important absorption characteristic. Although not highlighted, Schnaiter et al. (2005) showed in a laboratory study that coating enhances BC absorption more at 700 nm than at 450 nm (see Fig. 9). 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. Therefore, for coated BC, BC AAE should be lower than 1.0. In spite of this, we find it strange that recent studies (e.g. Khatri et al., 2010; Arola et al., 2011) still assume a fixed BC AAE value of 1.0. In the present study, we attempt to estimate BC AAE with ambient measurements rather than assume it. There are plenty of ambient measurement studies of estimating total aerosol AAE, but rarely is AAE for the BC component estimated.

OA was evaluated as slightly absorbing a decade ago (OPAC data; Hess et al., 1998). Recently, Andreae and Gelencsér (2006) explained that organics is decomposed into brown carbon (BrC) and colorless carbon, and only brown carbon absorbs.

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The BrC optical characteristics however seem highly variable Chakrabarty et al. (2010) generated BrC as primary particles and found BrC SSA (Single Scattering Albedo) to be much higher than 0.95 at 550 nm, consistent with weakly absorbing BrC particle analyzed by Hoffer et al. (2006). However, Alexander et al. (2008) identified different kind of BrC particles in East Asia 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". It is not certain yet if BrC coats 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 while "BrC aerosol" is used as such.

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 below 1.0 (see Sect. 6 for details). Alexander et al. (2008) discovered the BrC particles in outflows from China. Does this mean that the strongly-absorbing BrC particles are abundant in East Asia and absent in biomass burning aerosols? How much does OA AAE change from region to region? Here, we attempt to answer these questions.

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 from October 2009 to May 2010, generating 6288 hourly data. Also, we collected PM

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(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 19 and 20 May in 2010 were removed due to accuracy issues.

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). CA (Carbonaceous Aerosol) should contribute predominantly to TC. TC is divided between EC (elemental carbon) and OC (organic carbon). EC and OC masses per unit volume are measured using a Model-4 Semi-Continuous OC-EC Field Analyzer (Sunset Laboratory Inc.). This instrument uses a thermal-optical transmittance (TOT) method with a laser-based pyrolysis correction with the National Institute for Occupational Safety and Health (NIOSH) 5040 protocol (Birch and Cary, 1996) method. 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 $\mu gC\,m^{-3}$ and 0.24 $\mu gC\,m^{-3}$, respectively, based on three times the standard deviation of filter blank filtered air measurement. The sampling filter was changed when laser correction was lower than 0.88 to avoid a substantial decrease in the laser signal

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due to accumulation of refractory materials on the filter. The reported uncertainty of this system was 5 %.

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) (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; 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 dominant absorbers of the sunlight. Aerosols arriving at Gosan should have all the three absorbing aerosol species and scattering aerosols

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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 the particle as a BC-OA internally mixed 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. Aerosol absorption at Gosan has dust influence at times but not always. 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 ideal but $PM_{1.0}$ measurements were only done sporadically.

We compute AAE averaged 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 probably lower than this range since the dust particles often travel through highly polluted areas before making it to Gosan. Figure 5 clearly demonstrates that dust AAE at Gosan is at least higher than 1.31.

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Figure 5 shows that AAE approaches 1.27 as the dust influence weakens. We propose that CA AAE at Gosan is about 1.27. One might argue that even with 1.27 there might still be significant dust influence. When we remove data with $PM_{10}/PM_{2.5} > 1.6$, we are removing more than 70 % of the total data volume. It is difficult to imagine that the remaining data have significant dust influence. Given the percentage of filtered data, 1.27 is likely to represent CA AAE accurately.

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 almost equally strong, when we remove dust-influenced data (Fig. 6b). This indicates that at Gosan (a) CA AAE dominates total aerosol AAE or (b) dust AAE is not much higher than CA AAE. 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 BC coating by sulfate and organics. 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 4 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. 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.

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.

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First, we collocated temporarily all the hourly data for the analysis. Then, the data associated with reported Asian dust events were eliminated to better analyze the characteristics of CA. 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. 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, and others.

Figure 7 shows the correlation, as we remove 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 $10 \times 10^{-6}\,m^{-1}$. In view of this, our technique of minimizing the data error is to eliminate the data associated with 10 or less $10^{-6}\,m^{-1}$ 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 error will show a particular bias. On the other hand, these errors can impact the relationship between OC/TC and AAE.

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

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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 Fig. 7 and Fig. 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.

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 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 absorption coefficients at different wavelengths, as measured by aethalometer, and λ denotes wavelength. EC (or OC) is measured carbon mass by the Sunset analyzer, and MAC refers to Mass Absorption Cross section. In solving the Eq. (1), MAC_BC represents BC absorption cross section per EC mass, and MAC_OA OA absorption cross section per OC mass.

In the (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, so the independence is warranted. The (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 ratio of BC-intrinsic OC to EC for ambient aerosols is not known. Thus, with an implicit assumption of no BC-intrinsic OC, we solve the Eq. (1) (see Table 1). At 520 nm, BC MAC is found to be 5.1 and OC MAC 1.4.

We conduct the sensitivity of the results to the presence of BC-intrinsic OC in Table 2. OC mass measured by the Sunset analyzer includes this BC intrinsic component. Considering this component, our regression method should be revised to:

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$$\mathrm{Abs}(\lambda)=\mathrm{MAC\_BC}(\lambda)\times(\mathrm{EC+BC-intrinsic\ OC})+\mathrm{MAC\_OA}(\lambda)\times(\mathrm{OC\text{-}BC\text{-}intrinsic\ OC}). \quad (2)$$

When we assume that BC-intrinsic OC mass is 20 % of EC mass, for example, we find that BC MAC decreases slightly while OA MAC changes insignificantly. OA MAC changes very little, perhaps because OA mass dominates TC mass.

Overall BC MAC appears to be about 5.0 $m^2\,g^{-1}$ at 520 nm. This value is somewhat lower than the reported range in Table 3. 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. Additional piece of evidence in support of the damages of coated BC particles is our inferred BC AAE of 0.9–0.95 as shown in Table 2. BC AAE is computed using BC MAC values. Kirchstetter et al. (2004) measured BC AAE near the BC emission source, and found its value to be near 1.0. We believe that a BC AAE of 1.0 represents fresh (thus uncoated) BC. Our inferred BC AAE is very close to 1.0, indicating that the derived absorption coefficients from aethalometer measurements neglect the enhanced absorption by coating to a great degree. We propose that BC AAE at Gosan is likely lower than 0.9–0.95.

6 OA MAC and AAE, and implications for BrC

We find that OA MAC and AAE are insensitive to the presence of BC-intrinsic OC (Table 2). In addition, absorbing OA, i.e. BrC particles, appear to exist as stand-alone particles (i.e. without coating), and so our estimates are likely immune to the aforementioned aethalometer errors with regard to coating destruction. Our estimated OA

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MAC is about 1.4 $m^2\,g^{-1}$ at 520 nm. 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 OC contributes about 45 % to CA absorption while BC contributes the rest 55 %.

How does our estimated OA MAC value compare with previous estimates? Table 3 lists the reported values for OC MAC. In this table, we use a term "OC MAC", since the previous estimates included BC-intrinsic OC. We showed in Table 2 that OA MAC & AAE are insensitive to the BC intrinsic OC component at Gosan, and so we assume that OC MAC & AAE can generally be regarded as OA MAC & AAE. 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, as mentioned earlier. 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 are 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 estimated OA AAE is 1.7 (1.4–2.1) using OA MAC over 370–950 nm. If we interpolate 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.

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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 in a log normal size distribution was assumed. A standard Mie scattering code is 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 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.

Why is the Gosan OA AAE higher than the strongly absorbing BrC AAE and lower than biomass smoke OC AAE? 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.

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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. 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 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:

1. ambient BC AAE and MAC and how the aging process affects them;
2. ambient OA AAE and MAC in biomass burning areas versus fossil fuel combustion areas; and
3. ambient OA AAE and MAC near an emission source and downstream.

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

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Table 1. Estimated Mass Absorption Cross section (MAC) in units of $m^2\,g^{-1}$. The gram unit in MAC here is for carbon. The results here assume 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)

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Table 2. Sensitivity of estimated MAC to a mass ratio of EC to BC-intrinsic OC. Here, BC MAC refers to BC absorption cross section per BC carbon mass in units of $m^2\,g^{-1}$. OA MAC refers to OA (stand-alone OA + organic shell) absorption cross section per OA carbon mass, where absorbing OA (i.e. BrC particle) is assumed to be externally mixed with BC.

	BC MAC		OA MAC	
	No BC-intrinsic OC	BC-intrinsic OC being 20 % of EC	No BC-intrinsic OC	BC-intrinsic OC being 20 % of EC
370 nm	6.0	5.4	2.7	2.7
470 nm	5.6	5.0	1.7	1.7
520 nm	5.1	4.5	1.4	1.4
590 nm	4.6	4.0	1.2	1.2
660 nm	4.2	3.7	1.0	1.0
880 nm	3.0	2.6	0.6	0.6
950 nm	2.6	2.3	0.5	0.5
AAE	0.9	0.95	1.72	1.72

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Table 3. Reported ambient aerosol 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, the carbon mass includes BC-intrinsic OC. OC absorption means absorption due to organic matter.

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

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Table 4. 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)

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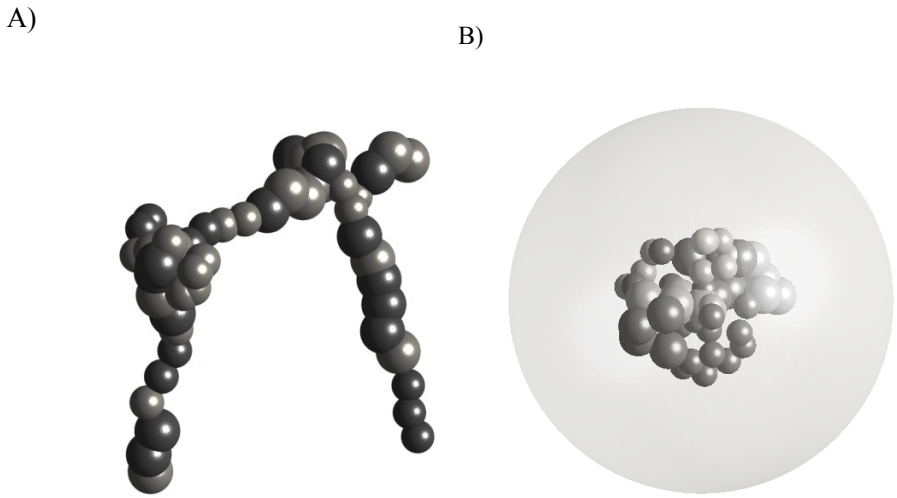

Fig. 1. **(A)** A typical fresh BC (Black Carbon) particle. A BC particle is made up of smaller spherical particles called spherules or monomers. Spherules in BC particles consist mainly of EC (Elemental Carbon) but also contains organics and inorganics (Watson and Valberg, 2001). Organics in BC is referred 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).

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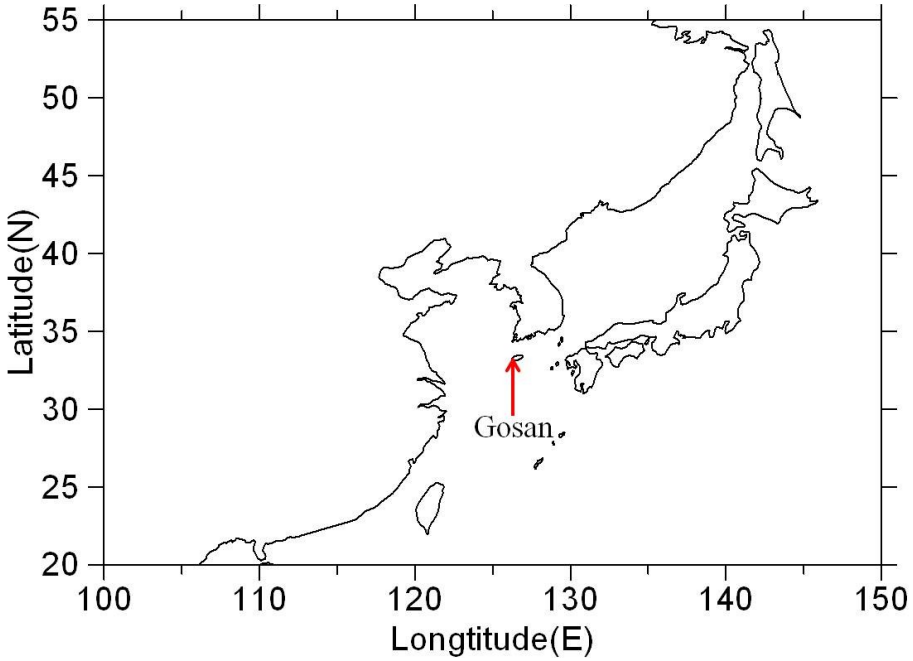

Fig. 2. Gosan is located at the western tip of Jeju Island, Korea. Gosan ABC super site is located at 33.28° N and 126.17° E.

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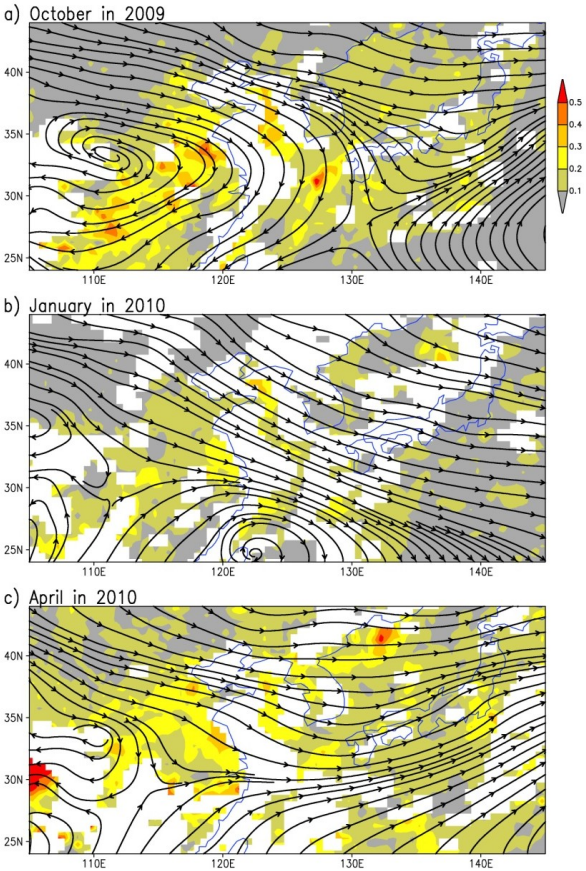

Fig. 3. 850 hPa wind stream from the ERA Interim Reanalysis (Dee et al., 2011) and MISR (Multi-angle Imaging Spectro-Radiometer) small-mode AOD at 555 nm.

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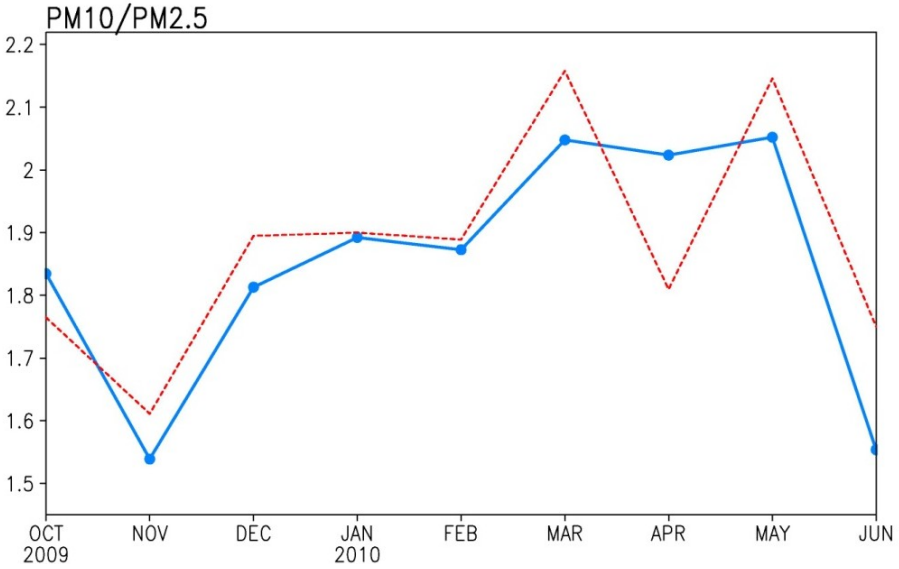

Fig. 4. Ratio of monthly PM_{10} mass density to monthly $PM_{2.5}$ mass density at Gosan. Monthly values are obtained from arithmetic mean (blue lines) and median (red lines).

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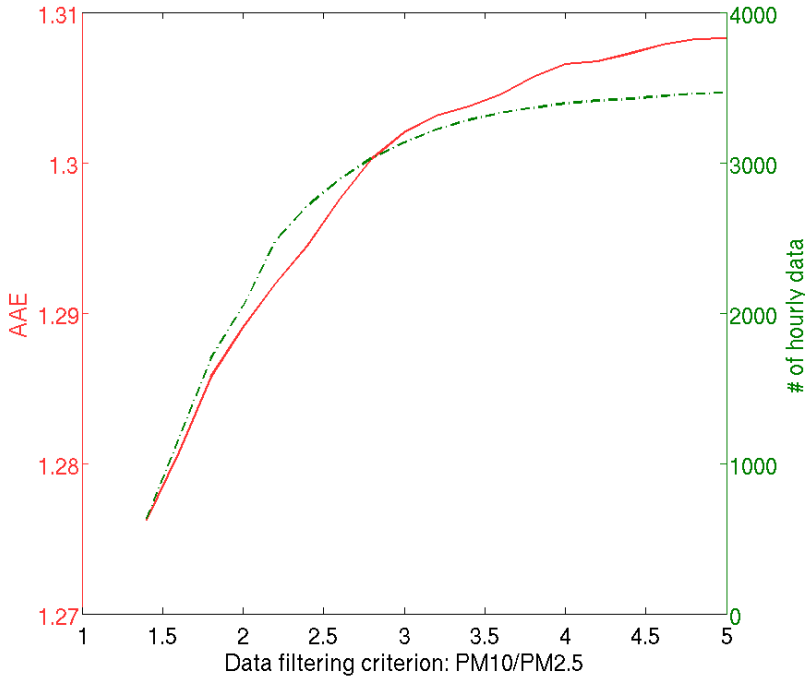

Fig. 5. AAE (Absorption Ångström Exponent), as obtained from averaged aethalometer absorption coefficients at 370, 470, 520, 590, 660, 880 and 950 nm. Absorption data are averaged after removing the values corresponding to $PM_{10}/PM_{2.5}$ being greater than a threshold (i.e. data filtering criterion).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

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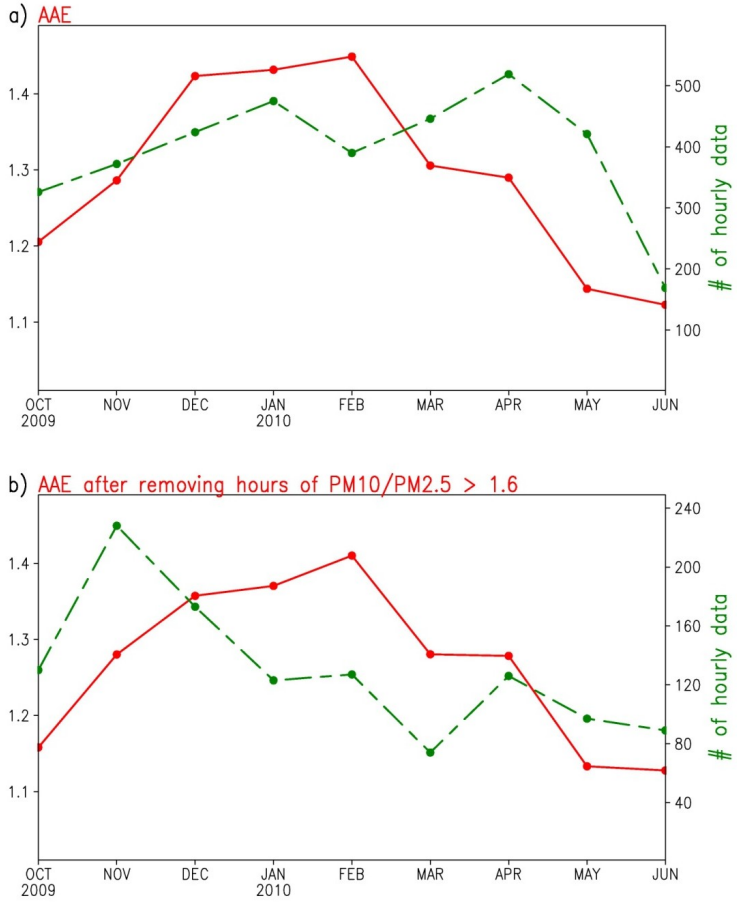

Fig. 6. Monthly-mean AAE at Gosan.

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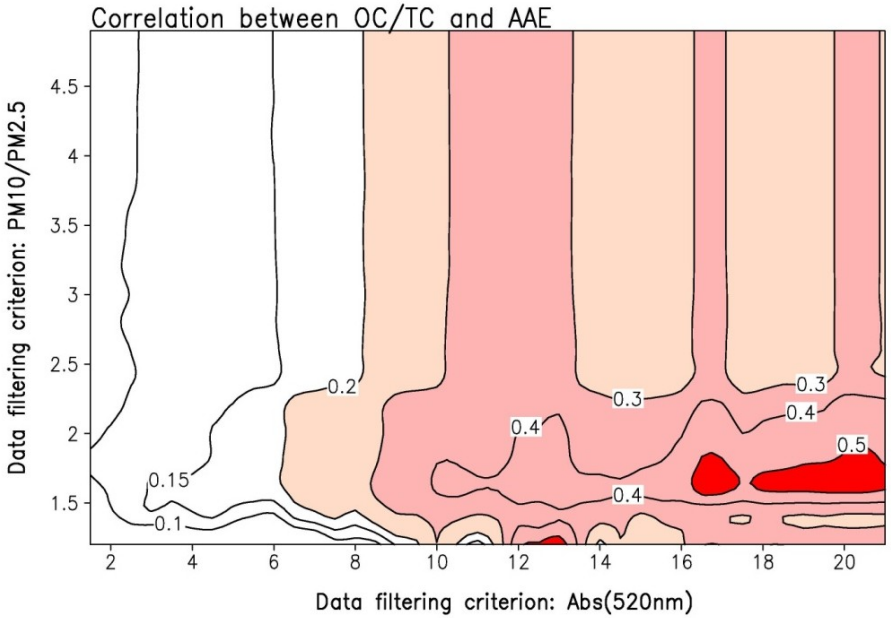

Fig. 7. Correlation between OC/TC and AAE. TC refers to total carbon (i.e. EC + OC). OC/TC refers to the mass ratio of OC to TC. Correlation is calculated in the following: mean OC/TC and mean AAE are computed with mean OC, mean TC and mean absorption at each wavelength instead of the mean of OC/TC and 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}.

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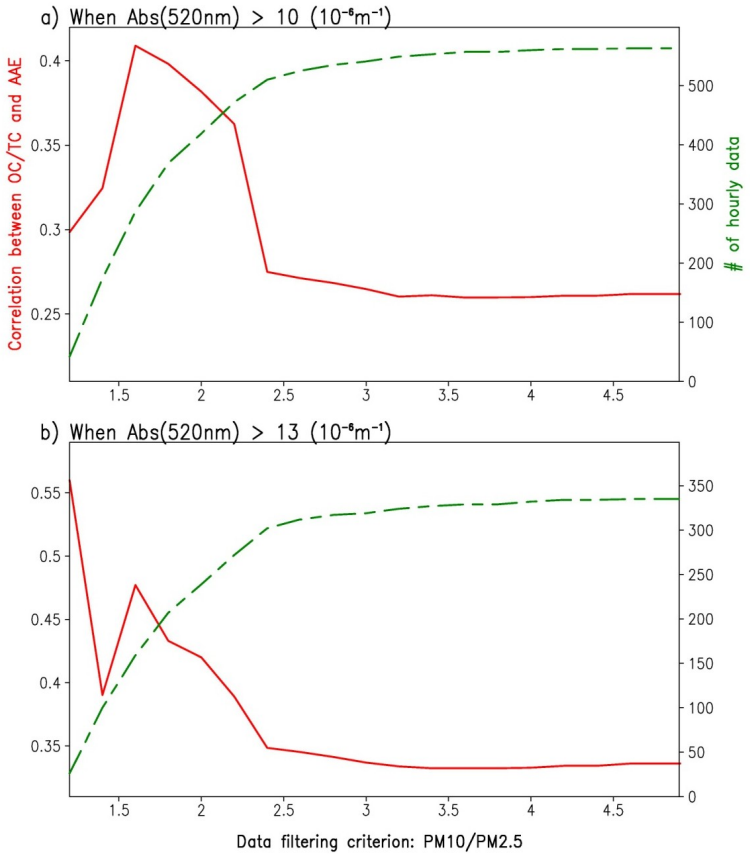

Fig. 8. Correlation between OC/TC and AAE, as a function of the data filtering criterion based on $PM_{10}/PM_{2.5}$. See Fig. 5 for the discussion of the filtering criterion. Also shown is the number of hourly data corresponding to data filtering criterion.

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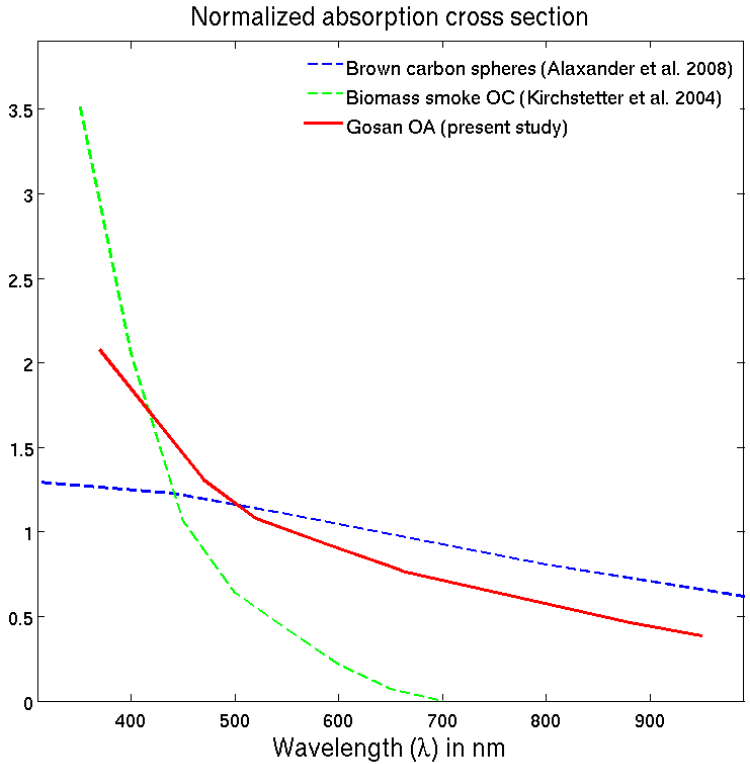

Fig. 9. Absorption cross section divided by its average w.r.t. wavelength. Biomass smoke OC refers to organic material in biomass burning aerosols.

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