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## On the attribution of black and brown carbon light

D. A. Lack and  
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# On the attribution of black and brown carbon light absorption using the Ångström exponent

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## Abstract

The absorption Ångström exponent ( $\hat{a}_{\text{Abs}}$ ) of black carbon (BC), or BC internally mixed with non-absorbing material ( $\text{BC}_{\text{Int}}$ ), is often used to differentiate the contribution of black carbon, dust and brown carbon to light absorption at low-visible wavelengths. This attribution method contains assumptions with uncertainties that have not been formally assessed. We show that the potential range of  $\hat{a}_{\text{Abs}}$  for BC (or  $\text{BC}_{\text{Int}}$ ) in the atmosphere can reasonably lead to +7 % to -22 % uncertainty in BC (or  $\text{BC}_{\text{Int}}$ ) absorption at 404 nm derived from measurements made at 658 nm. These uncertainties propagate to errors in the attributed absorption and mass absorption efficiency (MAE) of brown carbon (BrC). For data collected during a biomass-burning event, the mean uncertainty in MAE at 404 nm attributed to BrC using the  $\hat{a}_{\text{Abs}}$  method was found to be 34 %. In order to yield attributed BrC absorption uncertainties of  $\pm 33\%$ , 23 % to 41 % of total absorption must be sourced from BrC. In light of the potential for introducing significant and poorly constrained errors, we caution against the universal application of the  $\hat{a}_{\text{Abs}}$  attribution method.

## 1 Introduction

The Ångström exponent ( $\hat{a}$ ) was originally developed to describe the wavelength dependence of visible light extinction by atmospheric particles (Ångström, 1929). The  $\hat{a}$  for extinction is mostly influenced by particle size and has often been used to differentiate extinction by large particles (e.g. desert dust, sea salt) from fine mode particles (e.g. anthropogenic organics, sulfates and nitrates) in atmospheric samples (Bergstrom et al., 2007; Meloni et al., 2006). Equation (1) shows the simplest way in which  $\hat{a}$  can

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be calculated using a pair of observations at two different wavelengths:

$$\hat{a}(\lambda_1, \lambda_2) = \frac{\ln\left(\frac{b_{\text{Optical}}(\lambda_1)}{b_{\text{Optical}}(\lambda_2)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)} \quad (1)$$

where  $b_{\text{Optical}}$  is the optical coefficient of interest (e.g. light extinction, absorption or single scattering albedo) at wavelength  $\lambda$ , where  $\lambda_2 > \lambda_1$ .

The absorption Ångström exponent ( $\hat{a}_{\text{Abs}}$ ) for uncoated black carbon (BC) is predicted to be wavelength independent for small particle sizes (< 50 nm) taking a value  $\hat{a}_{\text{Abs}} = 1$  (Moosmüller et al., 2011; Moosmüller and Arnott, 2009; Bergstrom et al., 2002 and references therein). However, the  $\hat{a}_{\text{Abs}}$  for ambient particles (measured between a blue and a red wavelength) has often been observed to be larger than 1. There are a number of factors that could have contributed to these observations including the presence of organic/inorganic coatings on BC, and/or absorption by non-BC absorbers such as desert dust (Russell et al., 2010) and organics (i.e. brown carbon, BrC) (Kirchstetter et al., 2004). These mechanisms are treated in turn below.

BC absorption under dry atmospheric conditions can in theory be enhanced at all wavelengths by the presence of transparent organic/inorganic coatings (Bond et al., 2006; Jacobson, 2001; Bohren and Huffman, 1983; Fuller et al., 1999). The mechanism for this enhancement is one in which the coating acts as a lens to focus radiation into the absorbing BC core. Absorption enhancements of this nature have been observed in some laboratory and field studies (e.g. Cappa et al., 2012; Cross et al., 2010; Schnaiter et al., 2005; Lack et al., 2012b, 2009). Theoretical calculations have bounded the likely range of  $\hat{a}_{\text{Abs}}$  for BC internally mixed with non-absorbing material ( $\text{BC}_{\text{Int}}$ ). They have shown that the  $\hat{a}_{\text{Abs}}$  can vary from a baseline of 1 to an upper limit of  $\sim 1.7$ , depending on the size and optical properties of the core/coating and the wavelength pairs used to determine  $\hat{a}_{\text{Abs}}$  (Gyawali et al., 2009; Lack and Cappa, 2010).

Non-BC absorbers could potentially enhance  $\hat{a}_{\text{Abs}}$  more significantly, although it should be noted that the spectral variability of absorption for these components (i.e.

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dust, BrC) may not be captured well by the  $\lambda$  model (e.g. Updyke et al., 2012). Measured values of  $\hat{\alpha}_{\text{Abs}}$  for desert dust have ranged from 2.5 to 6.0 (wavelength pairs: 467 nm, 660 nm) (e.g. Weinzierl et al., 2011), while  $\hat{\alpha}_{\text{Abs}}$  for BrC up to 9.5 have been observed (wavelength pairs: 400, 700 nm) (Andreae and Gelencser, 2006 and references therein; Rizzo et al., 2011; Flowers et al., 2010; Lewis et al., 2008; Corr et al., 2012).

## 2 Ångström attribution of BC absorption

The  $\hat{\alpha}_{\text{Abs}}$  has often been used in a simple method for attributing low-wavelength absorption to BC and non-BC sources. The attribution method utilizes a pair of measurements made at different wavelengths (e.g. 404 and 658 nm in Fig. 1a). Absorption at the longer wavelength is assumed to be due only to dry uncoated BC, or dry  $\text{BC}_{\text{Int}}$ . This absorption measurement is extrapolated to the lower wavelength using an assumed  $\hat{\alpha}_{\text{Abs}}$ , most commonly 1. The difference between the measured and extrapolated absorption at the short wavelength is then attributed to absorption by non-BC entities. Figure 1a shows a schematic of this method. The figure was constructed by arbitrarily setting the absorption level of BC to 1 at the longest wavelength (e.g. 658 nm) and assuming an  $\hat{\alpha}_{\text{Abs}}$  of 1 for uncoated BC (line [1]). Additional absorption at 658 nm was added to the BC absorption to illustrate internal mixing of non-absorbing material, also assuming an  $\hat{\alpha}_{\text{Abs}} = 1$  (line [2]). Additional absorption due to BrC was represented using an  $\hat{\alpha}_{\text{Abs}} = 2.5$  (line [3]). Table 1 summarizes the various absorption terms calculated using this method (referenced to Fig. 1a).

Other more complex multi-wavelength methods for absorption attribution have been reported (Tesche et al., 2011; Kaufman et al., 2002; Arola et al., 2011; Schuster et al., 2005; Bergstrom et al., 2004; Corr et al., 2012). However, we concentrate here only on the simple  $\hat{\alpha}_{\text{Abs}}$  approach on account of its widespread use in peer reviewed studies (Bahadur et al., 2012; Cazorla et al., 2013; Chung et al., 2012; Clarke et al., 2007; Esposito et al., 2012; Favez et al., 2009; Fialho et al., 2005; Gadhavi and Jayaraman,

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2010; Herich et al., 2011; McNaughton et al., 2011; Sandradewi et al., 2008a,b; Wang et al., 2013; Yang et al., 2009).

Uncertainties in the  $\hat{a}_{\text{Abs}}$  attribution method primarily arise from the choice of  $\hat{a}_{\text{Abs}}$  used to characterize BC or BC<sub>int</sub>. As stated previously,  $\hat{a}_{\text{Abs}}$  for uncoated BC is approximately 1 for particles < 50 nm in diameter, but can range from 0.7–1.2 for diameters > 50 nm (Gyawali et al., 2009). Larger BC particle sizes can exist in the atmosphere due to coagulation and collapse of smaller BC spherules (e.g. Cross et al., 2010; Zhang et al., 2008), or from direct formation during inefficient combustion (Schwarz et al., 2008). A recent study even suggested a lower limit of  $\hat{a}_{\text{Abs}} = 0.55$  for atmospheric “elemental carbon” (a term functionally similar to BC) (Bahadur et al., 2012). Despite this potential variability in  $\hat{a}_{\text{Abs}}$  for uncoated BC (represented in Fig. 1a by the vertical arrow linked to line [1]),  $\hat{a}_{\text{Abs}} = 1$  is a commonly used community standard.

For BC<sub>int</sub> the situation is somewhat different. As described previously, the theoretical  $\hat{a}_{\text{Abs}}$  for BC<sub>int</sub> can range from 1 to ~ 1.7 (Gyawali et al., 2009; Lack and Cappa, 2010). In contrast, Bahadur et al. (2012) assumed that internal mixtures did not affect  $\hat{a}_{\text{Abs}}$  and used an  $\hat{a}_{\text{Abs}}$  for BC<sub>int</sub> = 0.55. Analysis of a range of atmospheric measurements of  $\hat{a}_{\text{Abs}}$  for aerosol sourced from fresh fossil fuel, and regional urban pollution (where the dominant absorber was BC) show an average  $\hat{a}_{\text{Abs}}$  (467 nm and 660 nm) of  $1.1 \pm 0.2$  ( $1\sigma$ ) (Lack et al., 2008; Clarke et al., 2007; Virkkula et al., 2005; Rosen et al., 1978; Bergstrom et al., 2007, 2002; Kirchstetter et al., 2004). We acknowledge that for these studies the extent to which internal mixing, or even BrC, impacted  $\hat{a}_{\text{Abs}}$  was unknown. However these studies have been used previously to support the use of an  $\hat{a}_{\text{Abs}} = 1$  for BC (Bond et al., 2013). At a minimum we suggest that these studies provide a better indication of the potential range of  $\hat{a}_{\text{Abs}}$  for BC<sub>int</sub> compared to the wider range of values determined theoretically (this range is represented on Fig. 1a by the vertical arrow on line [2]).

In this study we will focus on extrapolation of absorption to 404 nm from 658 nm for BC<sub>int</sub> rather than bare BC. If an  $\hat{a}_{\text{Abs}}$  of 1 is used for 404 nm BC<sub>int</sub> absorption attribution, when another  $\hat{a}_{\text{Abs}}$  in the range 0.55 to 1.7 would have been more appropriate, then

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the extrapolated absorption will contain an uncertainty. Figure 1b (grey line) shows that this uncertainty ranges from +20 % to −40 %. An experimentally based range of  $\hat{a}_{\text{Abs}}$  for  $\text{BC}_{\text{Int}}$  derived from the studies referenced above is also shown in Fig. 1b (red markers). These data span an  $\hat{a}_{\text{Abs}}$  range of 0.8 to 1.4 and suggest that a more likely range in extrapolated  $\text{BC}_{\text{Int}}$  absorption uncertainty is +7 % to −22 %. This uncertainty will contribute to uncertainty in attributed BrC absorption, the magnitude of which will be dependent on the total level of absorption.

### 3 Ångström attribution of BrC absorption

Following determination of the  $\text{BC}_{\text{Int}}$  absorption at short wavelength using the Ångström interpolation method (assuming no other non-BC absorbers), BrC absorption can be attributed. The confidence in the BrC absorption attribution can be assessed by combining the uncertainty introduced from the  $\hat{a}_{\text{Abs}}$  used for attribution of  $\text{BC}_{\text{Int}}$  absorption with the uncertainty in the absorption measurements themselves. This confidence level depends on the fractional contribution of BrC to the total aerosol absorption.

First we consider uncertainties in the  $\hat{a}_{\text{Abs}}$  attribution method. In Sect. 2 we showed that the uncertainty in short-wavelength absorption by  $\text{BC}_{\text{Int}}$  determined by extrapolation using an  $\hat{a}_{\text{Abs}} = 1$  ranged from +7 % to −22 %. To simplify this section, rather than report asymmetric uncertainties, we report high (low) uncertainty estimates. For example, the uncertainty in attributed  $\text{BC}_{\text{Int}}$  absorption is represented as  $\pm 22\%$  ( $\pm 7\%$ ).

Second we consider experimental uncertainties. For this analysis we assume measurements were made using the PAS technique, which offers the lowest uncertainty absorption measurement of currently available techniques of  $\pm 5\%$  (Lack et al., 2012a, 2009). We investigate a system where we assume that the PAS was used to measure  $\text{BC}_{\text{Int}}$  absorption at 658 nm ( $\pm 5\%$ ), which was then used to attribute  $\text{BC}_{\text{Int}}$  absorption at 404 nm. The  $\hat{a}_{\text{Abs}}$  and PAS measurement uncertainties are treated independently and propagated in quadrature yielding an uncertainty range in attributed  $\text{BC}_{\text{Int}}$  absorption at 404 nm of  $\pm 22.6\%$  ( $\pm 8.6\%$ ). To this estimate, we add the uncertainty in PAS-measured

total absorption ( $BC_{\text{Int}} + \text{BrC}$  absorption) at 404 nm ( $\pm 5\%$ ), which yields a total uncertainty range for attributed BrC absorption at 404 nm of  $\pm 23\%$  ( $\pm 10\%$ ). It is clear that the most significant contributor to the total uncertainty arises from the choice of  $\hat{a}_{\text{Abs}}$  used.

This analysis indicates that in order to achieve an uncertainty in attributed BrC absorption at 404 nm of  $\leq \pm 100\%$  the absolute amount of BrC absorption must be at least 23% (10%) of  $BC_{\text{Int}}$  absorption. To achieve a reasonable uncertainty in attributed BrC absorption of e.g.  $\pm 33\%$ , BrC absorption must be at least 69% (30%) of  $BC_{\text{Int}}$  absorption, or alternatively, BrC must contribute at least 41% (23%) of total absorption.

We note that if filter based methods for measuring absorption are used (e.g. Aethelometer or particle soot absorption photometer), which have uncertainties of the order of 25% (Bond et al., 2013), the minimum contribution of BrC to total absorption must exceed 50% for the  $\hat{a}_{\text{Abs}}$  attribution method to provide quantitative results (this analysis neglects the influence of additional suspected biases that may add uncertainty to filter-based absorption measurements, Lack et al., 2008; Subramanian et al., 2007).

## 4 Experimental method for BrC attribution

In Sect. 5 we assess uncertainties associated with the  $\hat{a}_{\text{Abs}}$  attribution method using an experimental dataset in which absorption by BC and BrC was measured directly at multiple wavelengths. Full experimental procedures can be found in Lack et al. (2012b) and are briefly summarized in this section.

Dense biomass burning emissions were sampled 24 h after the start of a Ponderosa Pine forest fire near Boulder, Colorado during September 2010. Particle emissions were dominated by BC and organics. Total non-refractory particle mass ( $m_{\text{NR-PM}}$ , encompassing organic and ammonium nitrate) was measured using an aerosol mass spectrometer (AMS, Bahreini et al., 2009). Organic material was consistently 90% of  $m_{\text{NR-PM}}$ . A photo-acoustic aerosol absorption spectrometer (PAS) measured dry total absorption at 658 nm, 532 nm and 404 nm ( $b_{\text{Abs-658}}$ ,  $b_{\text{Abs-532}}$ ,  $b_{\text{Abs-404}}$ ) and thermal-

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denuded absorption at 532 nm and 404 nm ( $b_{\text{Abs-532-BC}}$ ,  $b_{\text{Abs-404-BC}}$ ) (Lack et al., 2012a). The thermal-denuder heated the sample flow to 200 °C and removed the majority of semi-volatile coatings. The bulk MAE (404 nm) of the NR-PM was determined using Eq. (2). The actual MAE of the NR-PM with the effects of BC internal mixing removed,  $\text{MAE}_{\text{BB-BrC}}$ , was calculated independently of the  $\hat{a}_{\text{Abs}}$  method using Mie theory constrained by measurements of BC size and coating characteristics (Lack et al., 2012b). The contribution of dust to total absorption in the biomass plume was assumed to be minimal. This was consistent with observations of low background levels of aerosol extinction and absorption measured under conditions not strongly influenced by the biomass plume, and sample mass size distributions that peaked in the accumulation mode at approximately 300 nm.

$$\text{MAE}_{\text{Obs}} = \frac{b_{\text{Abs-404}} - b_{\text{Abs-404-BC}}}{m_{\text{NR-PM}}} = \frac{b_{\text{Abs-404-non-BC}}}{m_{\text{NR-PM}}} \quad (2)$$

The  $\hat{a}_{\text{Abs}}$  attribution method was used to calculate MAE assuming four different  $\hat{a}_{\text{Abs}}$  values for  $\text{BC}_{\text{int}}$ : 0.55 (lower limit; Bahadur et al., 2012), 1.0 (community standard), 1.7 (upper limit; Gyawali et al. (2009) and Lack and Cappa, 2010), and 1.2. This last value represents a calculated average  $\hat{a}_{\text{Abs}}$  for BC coated in non-absorbing material from the fire sampled by Lack et al. (2012b). This value was determined using measured BC core size distributions, measured total particle size distributions, and Mie theory. All  $\hat{a}_{\text{Abs}}$  used were for the wavelength pair 404 nm and 658 nm.

## 5 Experimental vs. Ångström attributed BrC absorption

The 404 nm absorption data measured for the forest fire described in Sect. 4 showed a significant contribution from internal mixing of BC and absorption by BrC (Fig. 2 and Lack et al., 2012b). These data were used to calculate two MAE values independently of the  $\hat{a}_{\text{Abs}}$  method. The first MAE value ( $\text{MAE}_{\text{Obs}}$ ) was calculated using



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Eq. (2) and includes contributions from both internal mixing effects and BrC absorption. A histogram of  $MAE_{Obs}$  at 404 nm is shown in Fig. 3a, and has a mean  $\pm 1\sigma$  value of  $1.58 \pm 0.60 \text{ m}^2 \text{ g}^{-1}$ . A second MAE was calculated for BrC only,  $MAE_{BB-BrC}$ , and is shown in Fig. 3b. Removal of the internal mixing lensing effect (performed in Lack et al., 2012b) reduced the MAE to  $0.98 \pm 0.45 \text{ m}^2 \text{ g}^{-1}$ . These two MAE values were not derived using the  $\hat{a}_{Abs}$  method and are therefore available to compare to  $\hat{a}_{Abs}$ -attributed MAEs.

Using the  $\hat{a}_{Abs}$ -attributed method, BrC MAEs were calculated using  $BC_{int} \hat{a}_{Abs} = 0.55, 1.0, 1.2$  and  $1.7$  respectively (Fig. 3c–f). A number of features distinguish the BrC MAE values calculated using the  $\hat{a}_{Abs}$  attribution method. Firstly,  $MAE_{Obs}$  and  $MAE_{BB-BrC}$  do not show any values below zero. In contrast, the attributed  $MAE_{\hat{a}_{Abs}-1.0}$ ,  $MAE_{\hat{a}_{Abs}-1.2}$ , and  $MAE_{\hat{a}_{Abs}-1.7}$  all show values below zero, with the fraction of MAE values  $< 0$  increasing with the assumed  $\hat{a}_{Abs}$  used. Uncertainty in the absorption measurements ( $\pm 5\%$ ) used to derive the attributed MAE suggest that some negative values may be expected. However, observation of a progressively larger number of negative MAE values as the assumed  $\hat{a}_{Abs}$  increases is likely an indication of a fundamental methodological problem with the  $\hat{a}_{Abs}$  approach.

The MAE derived using  $\hat{a}_{Abs} = 0.55$  is similar to  $MAE_{Obs}$  ( $1.54 \pm 0.63 \text{ m}^2 \text{ g}^{-1}$  vs.  $1.58 \pm 0.60 \text{ m}^2 \text{ g}^{-1}$ , Fig. 3). However, it is important to recognize that that the  $MAE_{Obs}$  includes absorption by internal mixing whereas the attributed  $MAE_{\hat{a}_{Abs}-0.55}$  should not. Given that the primary interest is in determining the MAE of BrC, it is more appropriate to compare values to  $MAE_{BB-BrC}$ . The best agreement between the  $\hat{a}_{Abs}$  attribution method and  $MAE_{BB-BrC}$  is obtained using  $\hat{a}_{Abs} = 1$  ( $MAE = 0.99 \pm 0.75 \text{ m}^2 \text{ g}^{-1}$  vs.  $0.98 \pm 0.44 \text{ m}^2 \text{ g}^{-1}$ , respectively).  $MAE_{\hat{a}_{Abs}-1.2}$  and  $MAE_{\hat{a}_{Abs}-1.7}$  produce lower MAE values with wider distributions ( $MAE_{\hat{a}_{Abs}-1.2} = 0.66 \pm 0.83 \text{ m}^2 \text{ g}^{-1}$ ; and  $MAE_{\hat{a}_{Abs}-1.7} = -0.05 \pm 1.04 \text{ m}^2 \text{ g}^{-1}$ ) and a substantially larger number of negative values.

In order to make a quantitative assessment of the differences between these methods we generated histograms of the percentage difference between the  $MAE_{BB-BrC}$  time

series and those for  $MAE_{\hat{a}_{Abs}=0.55}$ ,  $MAE_{\hat{a}_{Abs}=1.0}$ ,  $MAE_{\hat{a}_{Abs}=1.2}$  and  $MAE_{\hat{a}_{Abs}=1.7}$  ( $\Delta MAE\%$ , Fig. 4a–d respectively). For each histogram, the mean difference was also calculated. A negative  $\Delta MAE\%$  indicates that, on average, the attributed MAE was less than  $MAE_{BB-BrC}$ . We recognize that the  $\Delta MAE\%$  distributions for each  $MAE_{\hat{a}_{Abs}}$  trial had different characteristic shapes and therefore the mean does not provide a truly representative statistic for comparing trials. However, despite this limitation it still serves as a useful general metric. Figure 4a shows that on average there was a 56 % over-estimation of the BrC MAE when derived using  $\hat{a}_{Abs} = 0.55$ . Figure 4b suggests that there was no significant difference between  $MAE_{\hat{a}_{Abs}=1.1}$  and  $MAE_{BB-BrC}$ . This result would appear to support the use of the  $\hat{a}_{Abs}$  absorption attribution method with  $\hat{a}_{Abs} = 1$ . However, evaluation of the true  $\hat{a}_{Abs}$  based upon the observations suggests that the likely  $\hat{a}_{Abs}$  for the  $BC_{Int}$  was in fact  $\sim 1.2$ . As such, it appears that the good agreement between  $MAE_{\hat{a}_{Abs}=1.1}$  and  $MAE_{BB-BrC}$  was fortuitous. Using the more likely  $\hat{a}_{Abs}$  of 1.2, or the upper bound  $\hat{a}_{Abs}$  of 1.7, the average MAE was 34 %, or 104 %, lower than  $MAE_{BB-BrC}$ . The inset in Fig. 4 shows the relationship between assumed  $\hat{a}_{Abs}$  and  $\Delta MAE\%$ . Uncertainties in MAE of +20 % to –60 % are possible depending on the assumption made for  $BC_{Int}$   $\hat{a}_{Abs}$ . It is important to note that these uncertainties are calculated for a best-case scenario where BrC was a significant contributor to the total absorption. As BrC absorption contribution drops compared to  $BC_{Int}$  absorption, these uncertainties will increase.

## 6 Summary

The attribution of light absorption to black and brown carbon has commonly been achieved using the absorption Ångström exponent ( $\hat{a}_{Abs}$ ) relationship, assuming an  $\hat{a}_{Abs}$  for BC absorption of unity, and assuming that  $BC_{Int}$  does not alter  $\hat{a}_{Abs}$ . Theoretical investigations (Fig. 1b) suggest that this assumption can lead to modest uncertainty in attributed BC (or  $BC_{Int}$ ) absorption at 404 nm of +7 % to –22 %. This uncertainty will contribute to uncertainties in attributed BrC absorption. To achieve reasonable uncer-

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tainty in attributed BrC absorption of  $\pm 33\%$ , 23% to 41% of total absorption must be from BrC (assuming an absorption measurement uncertainty of  $\pm 5\%$ ). In the fire data presented, which is a best-case scenario for the occurrence of strongly absorbing BrC, 60% to 80% of absorption was due to BrC, making the  $\hat{a}_{\text{Abs}}$  attribution method viable.

For a test case examining absorption in a biomass burning plume, we found that the  $\hat{a}_{\text{Abs}}$  attribution method produced differences in assigned BrC absorption compared to direct analyses that did not use the  $\hat{a}_{\text{Abs}}$  approach. When using an  $\hat{a}_{\text{Abs}}$  of 1, as is often done, the average difference between the  $\text{MAE}_{\text{BrC}}$  calculated using the  $\hat{a}_{\text{Abs}}$  and  $\hat{a}_{\text{Abs}}$ -independent method was close to zero. However, the use of  $\hat{a}_{\text{Abs}} = 1$  for this system was not justified, given that the system contained BC coated in significant amounts of organic matter which would have yielded an  $\hat{a}_{\text{Abs}}$  (without the inclusion of brown carbon absorption) of at least 1.2. When this  $\hat{a}_{\text{Abs}}$  was used to attribute absorption, an underestimation in BrC MAE of  $-34\%$  was found. In addition to these uncertainties, the  $\hat{a}_{\text{Abs}}$  method often produces negative MAE values, which are (generally) physically unreasonable results.

It is almost certain that absorbing particles emitted from the same source will display a range of  $\hat{a}_{\text{Abs}}$  based upon size, coatings and composition. This  $\hat{a}_{\text{Abs}}$  is also very likely to change as atmospheric transport and processing occurs. Evolution of these  $\hat{a}_{\text{Abs}}$  distributions will lead to a range in attributed BrC absorption determined using the  $\hat{a}_{\text{Abs}}$  method. We therefore caution the general application of the  $\hat{a}_{\text{Abs}}$  method for absorption attribution suggesting that significant uncertainties can result. In order to minimize uncertainties, the technique should only be applied when a significant fraction of the total absorption is sourced from BrC.

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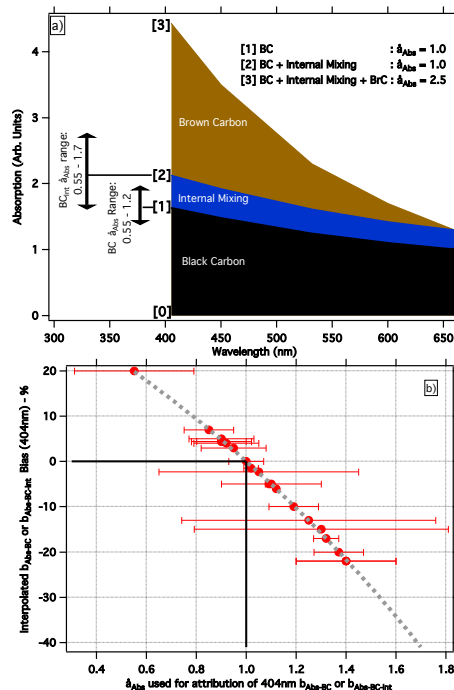
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**Table 1.** Summary of Idealized  $\hat{a}_{\text{Abs}}$  Absorption Attribution Products (refer to Fig. 1a).

Fig. 1.a Line Labels	Absorption Attribution
[1]–[0] =	BC Absorption: $\hat{a}_{\text{Abs}}$ of BC = 1.0: Can reasonably vary from 0.80–1.2
[2]–[1] =	Internal Mixing Absorption: $\hat{a}_{\text{Abs}}$ of internally mixed particles = 1.0 Can reasonably vary from 0.8–1.7
[3]–[2] =	BrC Absorption: $\hat{a}_{\text{Abs}}$ of internally mixed particles = 1.0
[3]–[1] =	Non-BC Absorption: $\hat{a}_{\text{Abs}}$ of BC = 1.0

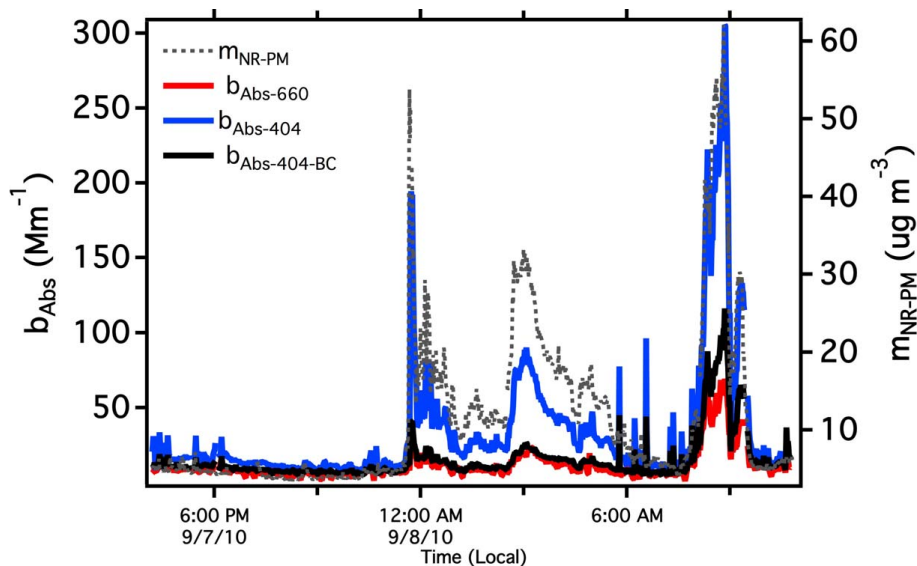
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**Fig. 1.** (a) Schematic of the attribution of absorption using the absorption Ångström exponent. We assume this system does not contain any dust. The absorption by BC is assumed to have an  $\hat{a}_{Abs} = 1$ , line [1], and can range from 0.55 to 1.2 (uncertainty arrow on line 1). Absorption by BC internally mixed with non-absorbing material ( $BC_{Int}$ ) is often assumed to have an  $\hat{a}_{Abs} = 1$ , line [2]. When in the atmosphere this  $\hat{a}_{Abs}$  may range from 0.55 to 1.7, uncertainty arrow on line [2]. In this example the  $\hat{a}_{Abs}$  of all ambient particles, including brown carbon is 2.5, line [3]. (b) Theoretical uncertainties in attributed BC (or  $BC_{Int}$ ) 404 nm absorption when  $\hat{a}_{Abs} = 1$ , rather than another value within the possible range suggested for BC or  $BC_{Int}$ . Red data shows average  $\hat{a}_{Abs}$  measured for particles assumed to be mostly BC (y-axis for red data forced to match black line for clarity).

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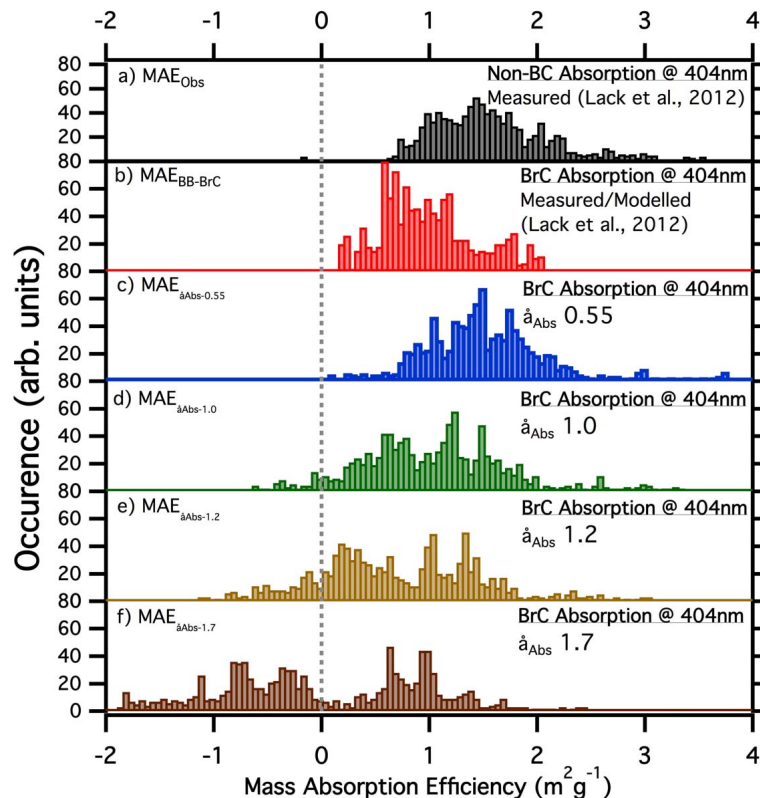


**Fig. 2.** Time series of measured 404 nm and 658 nm absorption of ambient particles (blue and red lines), 404 nm thermo-denuded absorption (black) and non-refractory aerosol mass (dashed grey line).

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**Fig. 3.** Histograms of measured and calculated mass absorption efficiency of NR-PM. **(a)** Measured MAE including internal mixing and BrC, **(b)** MAE of just BrC from Lack et al. (2012), and the MAE of BrC using the  $\hat{A}_{\text{Abs}}$  attribution method using **(c)**  $\hat{A}_{\text{Abs}} = 0.55$ , **(d)**  $\hat{A}_{\text{Abs}} = 1.0$ , **(e)**  $\hat{A}_{\text{Abs}} = 1.2$ , and **(f)**  $\hat{A}_{\text{Abs}} = 1.7$ .

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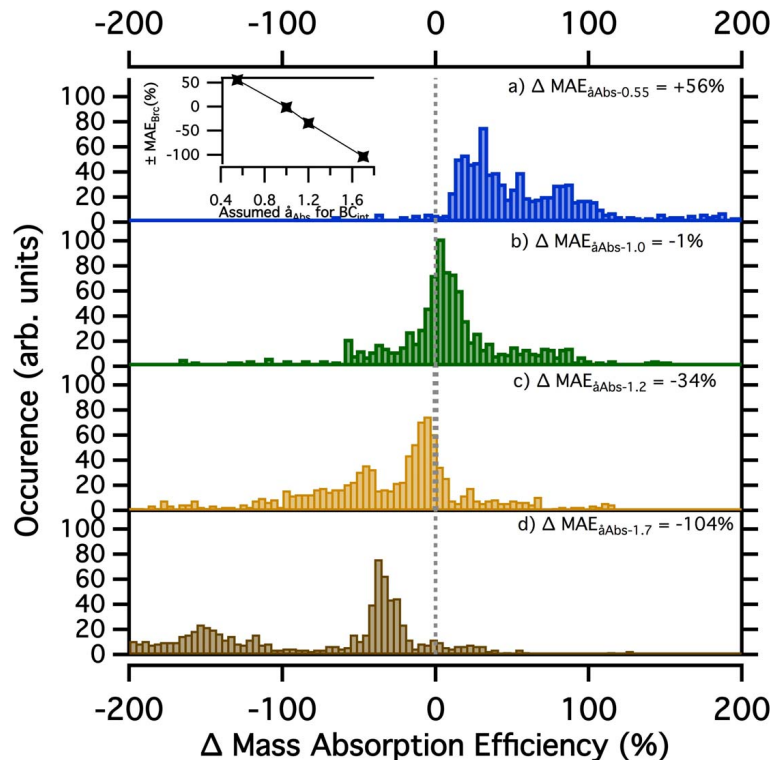
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**Fig. 4.** Histograms of the percent difference in time series of  $MAE_{BB-BrC}$  with (a)  $MAE_{\bar{a}_{Abs-0.55}}$ , (b)  $MAE_{\bar{a}_{Abs-1.0}}$ , (c)  $MAE_{\bar{a}_{Abs-1.2}}$  and (d)  $MAE_{\bar{a}_{Abs-1.7}}$ . Inset graph shows averaged relationship between assumed  $\bar{a}_{Abs}$  for internally mixed BC and the uncertainty in attributed MAE of BrC compared to the measured/modeled  $MAE_{BB-BrC}$ .

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