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Brown carbon absorption linked to organic mass tracers in biomass burning particles

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Abstract

Traditional gas and particle phase chemical markers used to identify the presence of biomass burning (BB) emissions were measured for a large forest fire near Boulder, Colorado. Correlation of the mass spectroscopic marker of levoglucosan (m/z 60) with measured particle light absorption properties found no link at 532 nm, and a strong correlation at 404 nm. Non-black carbon (BC) absorption at 404 nm was well correlated to the ratio of the mass fractions of particulate organic matter (POM) that were m/z 60 (f_{60}) to m/z 44 (f_{44}). The f_{60} to f_{44} ratio did not fully explain the variability in non-BC absorption, due to contributions of brown carbon (BrC) absorption and absorption due to internal mixing of POM with BC. The absorption Ångström exponent ($\text{\AA}$) showed a good correlation to f_{60}/f_{44} ; however the best correlation resulted from the mass absorption efficiency (MAE) of BrC at 404 nm ($\text{MAE}_{\text{POM-404 nm}}$) and f_{60}/f_{44} . This result indicates that the absorption of POM at low visible and UV wavelengths is primarily driven by emissions of levoglucosan (and related compounds), although they do not contribute to 532 nm absorption in this fire. The linear relationship between $\text{MAE}_{\text{POM-404 nm}}$ and f_{60}/f_{44} suggests that the impact of BrC can be predicted by emissions of f_{60} -related organic matter.

1 Introduction

The identification of biomass burning (BB) plumes using chemical markers has immense value in identifying sources, transport and processing of emissions (e.g. Roberts et al., 2011; Stohl et al., 2007; Warneke et al., 2010; Hecobian et al., 2011; Simpson et al., 2011; Andreae, 1983) which can then inform discussions on the potential impacts and mitigation options of BB pollution near sensitive regions for human health or climate (Smith, 1987; Fullerton and Bruce, 2008; Innes, 2000; Ramanathan and Carmichael, 2008). The effectiveness of a chemical marker is related to its atmospheric reactivity and specificity to the BB source (Aiken et al., 2010; Andreae, 1983;

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Hennigan et al., 2010; Simoneit et al., 1999). Carbon monoxide (CO), acetonitrile, hydrogen cyanide potassium and levoglucosan for example help to distinguish BB plumes from other sources in the gas and particle phase (de Gouw et al., 2003; Simoneit et al., 1999; Andreae, 1983).

Levoglucosan is a common BB particle tracer. It is an organic molecule formed from the pyrolysis of cellulose that can form the backbone of a family of combustion-linked particulate organic matter (POM) having a characteristic atomic mass fragment of m/z 60. The fraction of m/z 60 (f_{60}), compared to the fraction of the “oxidized organic” mass fragment m/z 44 (f_{44}) provides a unique picture of BB POM aging and mixing (Cubison et al., 2011; Innes, 2000). f_{44} in a BB plume will increase, and f_{60} decrease, as the levoglucosan-related compounds oxidize (Adler et al., 2011; Cubison et al., 2011) while f_{60} , which has a small non-BB background, will increase as the absolute amount of BB increases (Aiken et al., 2008; Alfarra et al., 2007; Cubison et al., 2011).

The climate impacts of BB emissions are determined by the optical properties of the emitted particles, which contain mostly POM, black carbon (BC) and some inorganic material (Lack et al., 2012b; Kondo et al., 2011; Posfai et al., 2003, 2004). These components contribute to light scattering and, critically, to wavelength dependent absorption of BC, absorption enhancement through internal mixing of BC and POM, and absorption by POM itself (i.e. brown carbon or BrC) (Lack and Cappa, 2010; Lack et al., 2012b). Here we investigate the relationships between the aerosol mass spectrometer (AMS) measured POM chemical markers (m/z 60, m/z 44) and these optical properties.

2 Experimental

2.1 Instrumentation

Experimental details for these measurements can be found in Lack et al. (2012b). Briefly, the measurements reported here were taken 24 h after ignition of a large

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Ponderosa Pine forest fire near Boulder, Colorado. 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-denuded absorption (sample heated to 200 °C) at 532 nm and 404 nm ($b_{\text{Abs-532-BC}}$, $b_{\text{Abs-404-BC}}$) (Lack et al., 2012a). Total non-refractory particle mass (POM and ammonium nitrate) was measured using a compact time-of-flight aerosol mass spectrometer (AMS, Bahreini et al., 2009). For the signal levels measured and time resolution reported, measurement uncertainties are estimated as follows; AMS – $\pm 38\%$, PAS 532 nm and 658 nm – $\pm 5\%$, PAS 404 nm – $\pm 10\%$. From these measurements the following absorption-related properties were calculated; absorption enhancements at 532 nm and 404 nm ($E_{\text{Abs-532}}$, $E_{\text{Abs-404}}$) using Eqs. (1) and (2); absorption Ångstrom exponent ($\text{\AA}$) using Eq. (3); and mass absorption efficiency of POM at 404 nm using Eq. (4):

$$E_{\text{Abs-532}} = \frac{b_{\text{Abs-532}}}{b_{\text{Abs-532-BC}}} \quad (1)$$

$$E_{\text{Abs-404}} = \frac{b_{\text{Abs-404}}}{b_{\text{Abs-404-BC}}} \quad (2)$$

$$\text{\AA} = \frac{\ln\left(\frac{b_{\text{Abs}}(\lambda_1)}{b_{\text{Abs}}(\lambda_2)}\right)}{\ln\left(\frac{\lambda_1}{\lambda_2}\right)} \quad (3)$$

$$\text{MAE} = \frac{b_{\text{Abs-404-POM}}}{m_{\text{POM}}} \quad (4)$$

The BB plume sampled was the same from the study of Lack et al. (2012b). Within this work, there were distinctly different periods of emissions that were classified according to the apparent influence of BB. Some figures in this work are clarified using these data periods. Two periods were almost devoid of BB influence (colored black and yellow in Figs. 2 and 3) while there were two periods heavily influenced by BB emissions (colored brown and orange in Figs. 2, 3) that were found to differ in their 404 nm

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optical properties by an additional absorption contribution from internal mixing. Lack et al. (2012b) removed this additional absorption term using a modeling step to, constrained by measurements of BC core size and total particle size, and particle mass, produce a MAE for BrC at 404 nm, which is used in this study.

An in-situ, dual channel, gas chromatograph–mass spectrometer (GC-MS) measured acetonitrile (and propene, 1,3-butadiene, and benzene used in the Supplement). Two 350 ml gas-phase samples were simultaneously collected via cryogenic trapping. The samples were then analyzed using an Al₂O₃ porous layer open tubular column (Channel 1) or a DB-624 capillary column (Channel 2) coupled to a quadrupole mass spectrometer (Roberts et al., 2011). The entire sample acquisition (5 min) and analysis (25 min) scheme repeated every 30 min. Measurement precision is ±5 % and accuracy is ±20 %. Carbon monoxide (CO) was measured using a UV fluorescence method (AeroLaser Model AL-5002) (Williams et al., 2009).

2.2 Positive-matrix-factorization analysis of AMS POM

Positive-matrix-factorization (PMF) has been used in several studies in order to identify the contribution of different factors, representing different sources or atmospheric processes, to the organic aerosol (OA) mass and spectra (Ulbrich et al., 2009; Zhang et al., 2011). In several studies, in addition to hydrocarbon-like OA (HOA) factor representing urban-combustion related primary OA, and an oxygenated OA (OOA) factor representing oxidized, secondary OA, a BB factor (BBOA) has been identified (Aiken et al., 2008; Lanz et al., 2010). The BBOA correlates well with gas phase biomass markers such as HCN and acetonitrile as well as CO (Aiken et al., 2010; Lanz et al., 2010). Mass spectra of the BBOA factor has a strong ion signature of levoglucosan from BB at *m/z* 60 (Aiken et al., 2010; Alfarra et al., 2007; Cubison et al., 2011; Lanz et al., 2010), but it may also have a hydrocarbon signature similar to HOA, making complete separation of BBOA and HOA challenging with PMF (He et al., 2011). Based on an extensive PMF analysis (see Supplement) separation of HOA and BBOA emissions in this study is not well achieved. This is not surprising considering limitations of the

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PMF technique to separate factors that co-vary in time (for example in this study the BB plume mixed with urban emissions for part of the time and were then transported together to the sampling site). In this regard, it is more straightforward to consider variability of the optical properties with the variability in the contribution of m/z 60 (f_{60}) since it is a dominant marker for BBOA.

3 Results

3.1 Traditional markers

The traditional gas-phase markers of acetonitrile and carbon monoxide (CO) show the largest enhancements during periods of elevated f_{60} . This indicates that BB gas phase markers are directly linked to the levoglucosan particle phase species which shows consistent identification of a BB influenced air mass (Fig. 1). The f_{44} and f_{60} relationship is also consistent with Cubison et al. (2011) on aging phenomenon of POM in BB particles and/or mixing with more oxidized POM (Fig. 1). The data presented here perhaps contains a lower overall contribution of f_{44} than observed by Cubison et al. (2011). One period of sampling shows an average f_{60} of 0.042, which is described by Cubison et al. (2011) as an upper limit to the background level of f_{60} , and indicates very little BB influence (shown as the black data in Figs. 2 and 3). Although the relationship between f_{44} and f_{60} in Fig. 1 cannot explicitly separate aging or mixing, the relationship between reactive VOC markers for BB (e.g. benzene, propene, and 1,3-butadiene) and acetonitrile indicate that the two biomass burning plumes were of similar photochemical ages (see Supplement and Fig. S1).

3.2 AMS f_{44} , f_{60} and absorption enhancement (E_{Abs})

E_{Abs} is the amount of absorption that is due to non-BC material through radiation lensing of coatings on BC cores and/or absorption by BrC (note that we assume there was no absorbing dust present). At 404 nm E_{Abs} increases as f_{60} increases, although the

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relationships are dependent of the period of sampling (Fig. 2a). The two periods of elevated $E_{\text{Abs-404}}$ observed in Fig. 2a (brown and orange data) diverge at elevated levels of f_{60} . These periods correspond to the heavily BB influenced periods identified in Lack et al. (2012b). The black and yellow data correspond to background air masses with little or no BB influence. These data periods were separated based on the observed levels of PM mass, and absorption enhancements (Lack et al., 2012b). The difference in $E_{\text{Abs-404}}$ between these periods was shown to be due to the influence of internal mixing of non-absorbing POM with BC, possibly from anthropogenic sources or secondary production of POM (Hennigan et al., 2011; Grieshop et al., 2009). Despite obvious correlations, f_{60}/f_{44} does not fully explain the behavior of $E_{\text{Abs-404}}$. In addition, there is no evidence of a relationship between $E_{\text{Abs-532}}$ and f_{60}/f_{44} (Fig. 2b), which indicates there is no contribution of f_{60} to 532 nm absorption.

3.3 Brown carbon-linked optical properties and AMS f_{60} , f_{44}

3.3.1 Absorption Ångstrom exponent ($\text{\AA}$)

Absorption Ångstrom exponent is often interrogated for indications of BB influenced particles, where it is assumed that an $\text{\AA} > 1$ is indicative of absorption by more than just BC, such as dust and BrC (Gyawali et al., 2009; Lewis et al., 2008; Corr et al., 2012; Yang et al., 2009; Favez et al., 2009). There is a direct correlation between wavelength dependency of absorption for this heavily BB influenced plume ($\text{\AA}$) and f_{60}/f_{44} (Fig. 3). This is strong evidence that the $\text{\AA}$ and absorption at 404 nm for BB emissions is directly correlated to the levoglucosan-based products of BB combustion. There is some scatter in this relationship however, particularly between the three sampling periods, which may be due to the influence of particle size or coating effects on the $\text{\AA}$ (Lack and Cappa, 2010; Moosmuller et al., 2011)

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3.3.2 Mass absorption efficiency (MAE) of brown carbon

Of the optical parameters investigated, the MAE of the POM at 404 nm ($MAE_{POM-404\text{ nm}}$) shows the most robust linear relationship with f_{60}/f_{44} (Fig. 4). The ratio of f_{60}/f_{44} appears to be a good proxy for BrC absorption (for this fire). The MAE reported does not contain any absorption contribution from internal mixing (removed via a modeling step, Lack et al., 2012b) and the subsequent relationship does not show any differentiation between the two different emissions periods observed in the $E_{Abs-404}$ (Fig. 2a). This difference indicates that there was considerable absorption by internal mixing during one period of the fire not due to the f_{60} -linked combustion products. The emissions sampled here were close to the emissions source, and gas phase markers indicate that there was minimal atmospheric processing (shown in the Supplement). The large range in f_{60} and f_{44} therefore indicates extensive mixing of air masses with minimal atmospheric processing. Therefore, the observed link between MAE and f_{60}/f_{44} shows that the strength of BrC absorption is linked to the emissions of the f_{60} -related organics. Some studies hint at the possibility of POM absorption efficiency increasing as it is oxidized (Corr et al., 2012; Marley et al., 2009; Adler et al., 2011; Bones et al., 2010), presumably from processing of emitted POM or secondary POM formed from VOC emissions, to produce low-visible and UV-active chromophores. Our data does not provide an opportunity to assess this link between atmospheric processing of f_{60} related organics and BrC optics.

4 Conclusions

The expected link between gas and particle phase chemical markers for biomass burning emissions was observed for a forest fire near Boulder, Colorado. The absorption of BrC in the particle emissions of this fire is correlated to the mass ratio of the fragments at m/z 60 and m/z 44. The m/z 60 fragment is sourced from levoglucosan and levoglucosan-related organics; products of incomplete combustion of cellulose. Mea-

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surements of the enhanced absorption due to internal mixing of POM and BC and BrC show a strong link to the m/z 60 fragment, however divergence of $E_{\text{Abs-404}}$ for two sampling periods heavily influenced by BB emissions and non-linear behavior between $E_{\text{Abs-404}}$ and f_{60}/f_{44} indicate that there are additional contributors to non-BC absorption at 404 nm (other than levoglucosan-related organics). A somewhat scattered relationship between wavelength dependence to absorption ($\hat{a}$) and f_{60}/f_{44} indicate that the 404 nm absorption is linked to levoglucosan, or related organics. The most robust linear relationship was found between the mass absorption efficiency of the organic matter at 404 nm ($\text{MAE}_{\text{POM-404 nm}}$) and f_{60}/f_{44} . Given that the photochemical age of the sample plume did not vary, this link shows that the evolution of BrC absorption observed is linked to just the absolute amount of levoglucosan-related organics. Future work will need to assess how the BrC absorption is related to atmospheric processing. This observed relationship may provide a pathway to predicting the contribution of BrC absorption from biomass burning emissions using the chemical composition of the organic particles produced from biomass combustion.

Supplementary material related to this article is available online at:
**[http://www.atmos-chem-phys-discuss.net/12/29129/2012/
acpd-12-29129-2012-supplement.pdf](http://www.atmos-chem-phys-discuss.net/12/29129/2012/acpd-12-29129-2012-supplement.pdf)**

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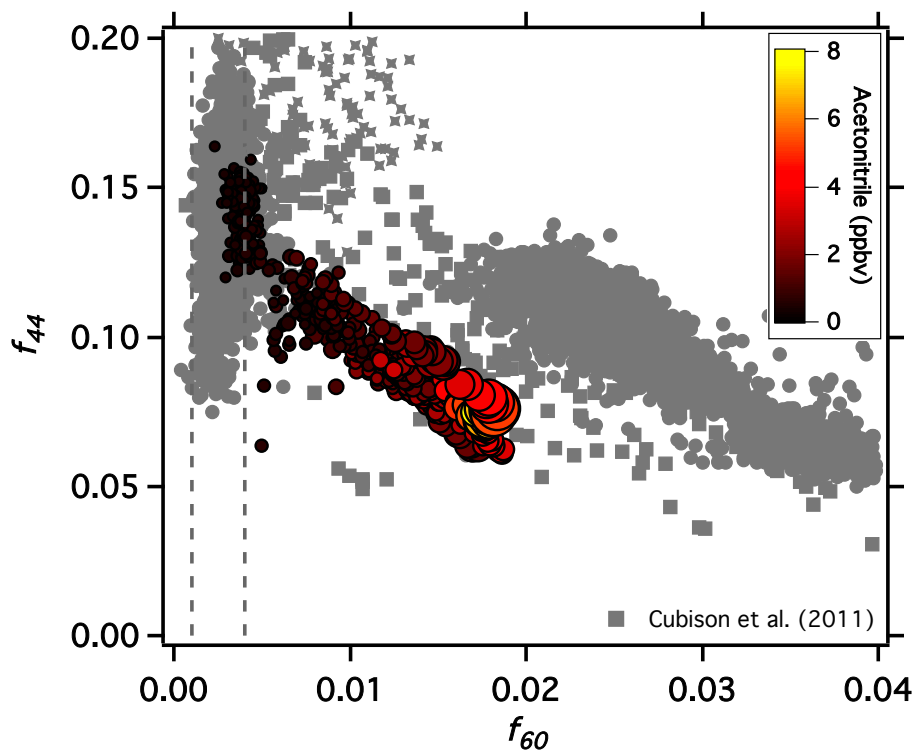


Fig. 1. Relationship between AMS f_{44} and f_{60} , colored by acetonitrile and sized by CO concentration. Range of background f_{60} levels shown as the vertical dashed lines (Cubison et al., 2011). Grey data is all data from Cubison et al. (2011). Acetonitrile determined from interpolation of the discrete GC-MS samples.

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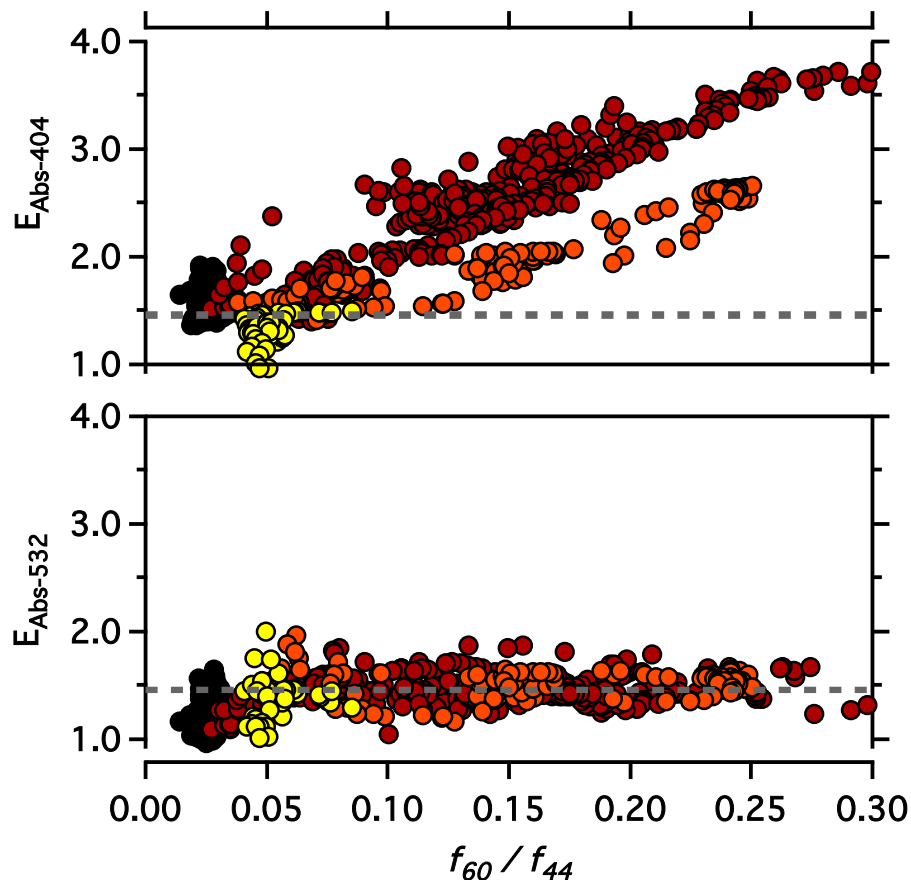


Fig. 2. Absorption enhancement relationship with f_{60}/f_{44} at **(a)** 404 nm and **(b)** 532 nm. The color-coding shows the different sampling periods of the BB plume. Horizontal dashed line indicates reasonable E_{Abs} (1.5) expected from theory from internal mixing only (Bond et al., 2006).

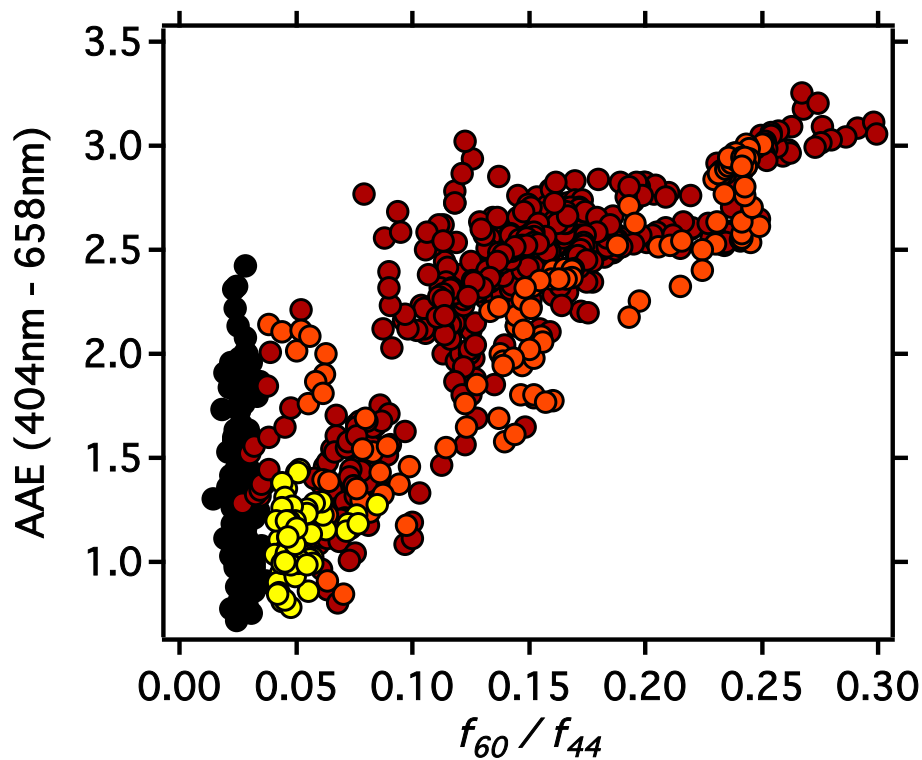


Fig. 3. Absorption Ångström Exponent ($\text{\AA}$) versus f_{60}/f_{44} . The color-coding shows the BB plumes sampled (Lack et al., 2012b).

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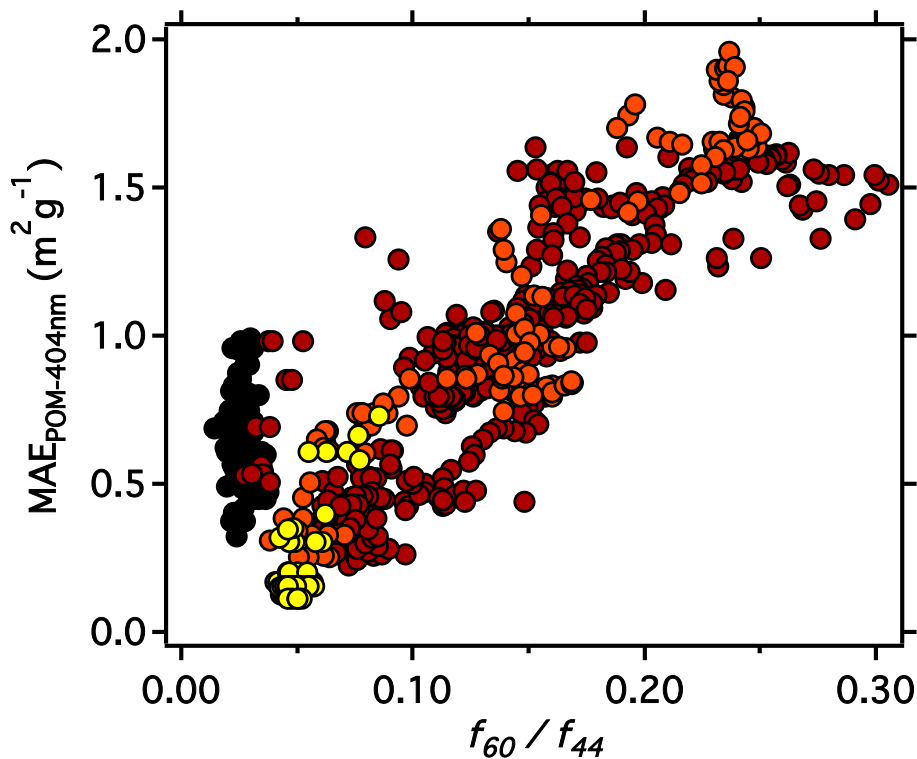


Fig. 4. Mass absorption efficiency (MAE, at 404 nm) versus f_{60}/f_{44} . The color-coding shows the BB plumes sampled (Lack et al., 2012b).