

## ***Interactive comment on “Spectral dependence of aerosol light absorption over the Amazon Basin” by L. V. Rizzo et al.***

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Comment 1: A 10 micron size cut for the Aethalometer was used for the Pasture/Dry 1999 experiment. Also, in general filter based absorption was used for all campaigns. Some filter-based methods have shown to have size-dependent biases (Lack et al. 2009; Nakayama et al. 2010). Please discuss these issues related to the assessment of absorption between campaigns. Response 1: To address this comment, we added the following paragraph to the section 2.3: “Recent works indicate that the multiple scattering correction depends on particle size, because it varies with the aerosol penetration depth into the filter. Nakayama et al. (2010) compared filter based absorption measurements of nigrosin dye generated particles to predictions from Mie theory, and found increased sensitivities with decreasing diameters, leading to overestimations as

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high as 100% for 100 nm aerosols in diameter. On the other hand, Lack et al. (2009) reported underestimations of 50% for absorbing polystyrene spheres of 500 nm, through the comparison of filter based to photoacoustic absorption measurements. As stated in section 2.1, in this study measurements were taken with a 10  $\mu\text{m}$  cut-off diameter. Fine mode particle mass (diameter < 2.5  $\mu\text{m}$ ) represented, respectively, 80% and 60% of PM10 at the pasture site (Artaxo et al., 2002) and at the forest site (Rizzo et al., 2010) during the dry season. Measurements of fine mode number size distribution during the SMOCC and LBA/ZF2-C14 experiments indicate that most particles had diameters between 10 and 400 nm, with mean geometric diameters around 100 nm (Rissler et al., 2006; Rizzo et al., 2010). With the available information, it is hard to say whether there will be an underestimation or overestimation of absorption coefficients due to aerosol size effects. Even though this effect is difficult to quantify, this is important to mention. To our knowledge a comprehensive investigation concerning the sensitivity of filter based measurements to ambient aerosol size is still missing.”

Comment 2: P11561 discusses Angstrom exponent decreases being due to a shift in size. This reads like the size of particles themselves are the cause of the absorption changes, when in reality it is likely that the particles are different and just happen to be different sizes. Response 2: We understand that the observed decrease in Ångström exponents from the dry to the wet season can be attributed either to the change in particle size, or to the change in the physicochemical properties of aerosols. To make this point clear, we rephrased the referred paragraph as follows: “The reason for reduced Ångström exponents during the wet season can be attributed either to the increase of particle sizes (Figure 4) or to a change in the physicochemical properties of the aerosol population. The variation of absorption spectral dependency as a function of aerosol bulk properties is still not well understood, especially for biogenic aerosols. Nevertheless, the effect of particle size changes on absorption Ångström exponents is relatively well known. For example, Moosmüller et al. (2011a) modeled the absorption spectra of homogeneous spherical brown carbon particles and found a decrease of Ångström exponents for larger particle diameters (from 0.1 to 10  $\mu\text{m}$ ). A similar result has been

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found by Gyawali et al. (2009) for uncoated black carbon spheres. In addition to the effect of aerosol size, another possible explanation would be the rain scavenging of soluble organic aerosols during the wet season, resulting in an enrichment of insoluble soot carbon particles in the remaining aerosol. This effect could also shift the Ångström exponents to lower values from dry to wet season.”

Comment 3: Please add a figure that shows the size distribution shifts between seasons (i.e related to P11561). Response 3: To address this suggestion we added the following figure:

Figure 4: Percent contribution of PM2.0 to PM10 aerosol mass concentration during the LBA/SMOCC experiment, based on gravimetric analysis of polycarbonate filters. The percentage of PM2.0 decreases from dry to wet season, in accordance with the shift from biomass burning to biogenic aerosols.

Comment 4: What do the results presented mean in terms of radiative effects? Presenting this as the final section would provide some needed context and significance of these results. A first order assessment based on the information presented should be achievable. Response 4: We agree that the assessment of the radiative effects of Ångström exponents for absorption provides context to our results. Therefore, we added the following text to section 3, as well as the Table 3: “We made a first order estimate of the influence of the observed absorption Ångström exponents on the aerosol radiative forcing. The 24-hour aerosol radiative forcing was calculated assuming an aerosol optical thickness (AOT) of 1.0 at 500 nm, typically observed in Amazonia during the dry season (Procopio et al., 2004), over a vegetated surface. The radiative fluxes were calculated between 450 and 880 nm by means of SBDART (Santa Barbara DISORT Atmospheric Radiative Transfer), using zero AOT as reference to calculate the aerosol forcings. The latitude of 5°S and the Julian day 258 were assumed in the calculations, representing the average latitude of the Amazon Region and the middle of the dry season, respectively. Aerosol optical properties at 500 nm were derived from a dynamic spectral aerosol model developed by Procopio et al. (2003) for

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smoke aerosols in Amazonia. The spectral dependency of aerosol optical properties was extrapolated using four extinction Ångström exponents ( $\alpha_{ext}$ ), calculated using the following equation taken from Moosmüller et al. (2011b): , (14) The single scattering albedo and the scattering Ångström exponent were assumed to be 0.92 and 2.0, respectively (Chand et al., 2006), and independent of wavelength in the spectral range under consideration. This is a reasonable assumption, as we estimate a weak spectral dependency for  $\omega_0$ , ranging from 0.92 at 450 nm to 0.91 at 880 nm. Four values of  $\alpha_{abs}$  were considered for the 24-hour aerosol forcing calculations: 1.0 (usual absorption spectral dependency assumed for soot carbon); 1.4 (corresponding to the average of  $\alpha_{abs}$  for the Pasture Dry 2002 experiment minus one standard deviation – refer to Table 2); 1.8 (average of  $\alpha_{abs}$  for the Pasture Dry 2002 experiment); and 2.2 (average of  $\alpha_{abs}$  for the Pasture Dry 2002 experiment plus one standard deviation). The value of  $\alpha_{abs}$  was also assumed to be wavelength independent, in accordance with our results (Table 2). Aerosols were assumed to be distributed homogeneously in an atmospheric layer of 1.6 km of altitude. Table 3 shows a first order assessment of the variability of the 24-hour aerosol radiative forcing as a function of absorption Ångström exponents. The mean forcing found with this parameterization for AOT = 1 (500 nm) was -32 W m<sup>-2</sup> at the surface and -8 W m<sup>-2</sup> at the top of atmosphere (TOA), with 24 W m<sup>-2</sup> being absorbed in the atmosphere. This result is comparable to other studies conducted in the Amazonian region, e.g., Procopio et al. (2004). Changing  $\alpha_{abs}$  from 1.0 to 2.2 causes a reduction of less than 1% in the absolute values of instantaneous aerosol forcing at the surface and in the atmosphere. At TOA, the instantaneous forcing calculated assuming  $\alpha_{abs} = 2.2$  can be up to 4.5% greater in absolute values, as compared to the forcing calculated assuming the absorption spectral dependency of soot aerosol ( $\alpha_{abs} = 1.0$ ). For the 24-hour aerosol forcing at TOA, the corresponding increase is about 0.9%. Overall, the results indicate a small impact of absorption Ångström exponents on 24-hour aerosol forcings, at least in the spectral region of 450-880 nm. Further studies should be taken to assess the corresponding impact in the UV spectral range.”

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Table 3: First-order estimate of the 24 hour radiative forcing between 450 and 880 nm at the surface, top of atmosphere (TOA), and in the atmosphere, considering AOT = 1.0 and different spectral dependencies for the absorption coefficient.

Comment 5: P11562: and Figure 6: Add shading which shows sunrise and sunset times. How will diurnal variability that you discuss affect radiative forcing? Will this 20% difference have an impact? Response 5: Following the referee's suggestion, we added shading on (former) Figure 6 to represent nighttime periods. The 20% decrease of absorption Ångström exponents at daytime will not have a significant impact on the aerosol radiative forcing, as discussed above.

Comment 6: P11563 L10: The absorption variability may also be due to chemical composition of the primary organic as it is emitted. Your statements assume that the primary emission has a constant optical character. Response 6: We agree that biomass burning emissions may not have a constant optical character. To address this point, we rephrased the sentence as follows: "The decreased absorption Ångström exponents observed at daytime might be either an outcome of increased particle sizes (Moosmüller et al., 2011a), or a consequence of changes in particle chemical composition due to combustion conditions or biomass fuel type, or even a consequence of the condensation of absorbing species with a weaker spectral dependence to the surface of aerosols."

Comment 7: P11564 L18: Many other studies indicate caution also. I think this work should be discussed with this in mind. Response 7: We agree, and we think that the data shown in our work reinforce the point that absorption measurements might not be extrapolated to all regions of the solar spectrum without care.

Comment 8: Figures 3 – 5: Uncertainty should be added to these figures. Response 8: We added error bars to Fig. 5, considering the typical uncertainties of  $\pm 10\%$  on  $\sigma_{\text{abs}}$  and  $\pm 20\%$  on  $\hat{\alpha}_{\text{abs}}$ , and included the following sentence in the legend: "Error bars represent typical uncertainties:  $\pm 5\%$  on  $\sigma_{\text{abs}}$  and  $\pm 20\%$  on  $\hat{\alpha}_{\text{abs}}$ ". Concern-

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ing Figures 3 and 5, we added the following sentence to their legends: "The typical uncertainty on  $\hat{\alpha}_{\text{abs}}$  is  $\pm 20\%$  (refer to Section 2.4). "

Comment 9: P11550 L13: 'Soot' Angstrom is size dependent, but likely to be  $1 \pm 0.1$  around sized of interest. Response 9: We agree, and rephrased the sentence as follows: "Due to its morphology and relatively constant refractive index, the absorption spectrum of soot carbon is expected to exhibit Ångström exponents of about  $1.0 \pm 0.1$  for particles with diameters in the range 10-100 nm (Sun et al., 2007; Gyawali et al., 2009)."

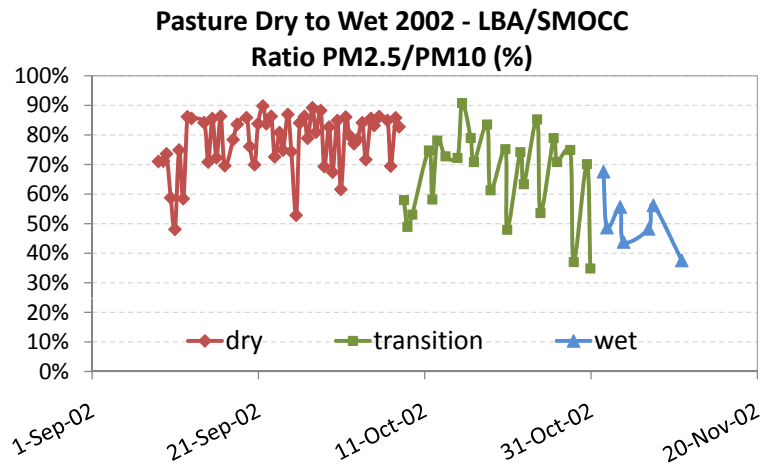
Comment 10: P11551 L19: What does biogenic origin include? Primary bio-aerosol, secondary organic, primary particles from biomass combustion? Response 10: We use the word "biogenic" following the terminology suggested by Fuzzi et al., 2006. The term "biogenic" comprises primary aerosols of biological origin, including whole organisms (e.g., bacteria), reproductive material (e.g., pollen, spores), fragments (e.g., plant waxes), decaying biomass, as well as secondary aerosols produced from gaseous chemical species of biological origin. Biomass burning aerosols do not fit in this definition. To make that clear in the manuscript, we rephrased the sentence as follows: "It provides a general description of the wavelength dependence of absorption both during the Amazonian wet season, when the aerosol population is dominated by biogenic particles, comprising primary and secondary aerosols of biological origin (Fuzzi et al., 2006), and during the dry season, when there is a strong influence of biomass burning emissions."

Comment 11: P11554 L 12: Is this the exact reverse procedure used by the instrument? Response 11: Yes, this is the exact reverse procedure, according to the manufacturer's manual and also to, e.g., Müller et al., 2011.

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**Fig. 1.** Fig.4: Percent contribution of PM2.0 to PM10 aerosol mass concentration during the LBA/SMOCC experiment, based on gravimetric analysis of polycarbonate filters.

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Table 3: First-order estimate of the 24 hour radiative forcing between 450 and 880 nm at the surface, top of atmosphere (TOA), and in the atmosphere, considering AOT = 1.0 and different spectral dependencies for the absorption coefficient.

Absorption Ångström exponent	Extinction Ångström exponent	Forcing at surface [W m <sup>-2</sup> ]	Forcing at TOA [W m <sup>-2</sup> ]	Forcing in atmosphere [W m <sup>-2</sup> ]
1.0	1.92	-32.07	-8.03	24.04
1.4	1.95	-32.03	-8.05	23.99
1.8	1.98	-32.00	-8.07	23.93
2.2	2.02	-31.95	-8.10	23.86

**Fig. 2.** Table 3: First-order estimate of the 24 hour radiative forcing between 450 and 880 nm at the surface, top of atmosphere (TOA), and in the atmosphere.

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