Atmos. Chem. Phys. Discuss., 11, C6731–C6737, 2011
www.atmos-chem-phys-discuss.net/11/C6731/2011/

Atmospheric Chemistry and Physics Discussions

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

L. V. Rizzo et al.

luvarizzo@gmail.com

Received and published: 21 July 2011

Comment 1: The authors suggest to measure the spectral absorption from the UV to near IR (c.f. page 11564, lines 21 to 32). The Aethalometer AE30 measures from 450 to 950 nm. Other types (e.g. AE-31) measure from 370 nm to 950 nm. Is there any indication that the extension of the spectral range down to wavelength of 370 nm improves the information content of absorption measurements? Response 1: Our results did not clearly indicate an improved definition of the absorption spectra by extending measurements from 450 to 370 nm. However, we had absorption measurements at 370 nm only during one of the three field experiments studied here (Forest Dry). As it is expected that the absorption cross section of non-soot carbon may increase sharply with decreasing wavelength (Andreae and Gelencsér, 2006), there is a possibility that

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

the absorption Ångström exponent would increase in the spectral region below 450 nm at the Pasture site. We wish we had absorption measurements at 370 nm from all the three field experiments to objectively address this question. Anyway, we recognize that we do not have enough evidence to recommend absorption measurements in the UV spectral range. Therefore, we rephrased the sentence as follows: "Especially in remote areas, it is advisable to measure aerosol absorption at several wavelengths to accurately assess the impact of non-soot aerosols on climate and photochemical atmospheric processes."

Comment 2: Page 11548, line 20 and page 11564, lines 13 to 15: Low absorption coefficients are correlated with low Ångström exponents (<1). The authors's explanation is that biogenic aerosols from the Amazonian have a weak spectral dependence. The authors should support their thesis by an error analysis for these cases. How high are absorption coefficients compared to the detection limit? Please give values for the uncertainty of the Ångström exponent. Response 2: This comment contains a number of different questions. Let's start with the issue about the uncertainty of the Ångström exponent. According to equations 7, 9 and 10, the calculation of absorption coefficients from Aethalometer attenuation coefficients depends on the previous knowledge of the spectral dependency of the single scattering albedo (ω0), among other parameters. The calculation of corrected absorption coefficients at several wavelengths is necessary to obtain the Ångström exponents for absorption. As we did not have simultaneous measurements of either single scattering albedo or scattering coefficients, the best we could do was to use average values for the albedo at a reference wavelength, and for the scattering Ångström exponent, as explained in the section 2.3. Errors associated with the use of averages instead of specific values were evaluated through sensitivity tests, discussed in section 2.4. A range of values of ω0 (0.88 to 0.96) and åscat (1.2 to 2.8) was tested, and we verified that different combinations of ω0 and åscat result in a variation of absorption coefficients and Ångström exponents. In our understanding, this is not possible to obtain a precise value for the uncertainty on absorption coefficients or Ångström exponents, but it is possible to predict a range of uncertainties. Through

ACPD

11, C6731–C6737, 2011

sensitivity tests, we verified that the main source of error to σabs and åabs is the choice of the Ångström exponent for scattering (åscat). In the worst case, assuming the average value åscat = 2.0 may cause a maximum deviation of $\pm$ 10% on σabs and $\pm$ 40% on åabs, depending on the different possible combinations of åscat and ω0 values. In the best case, the respective deviations would be $\pm$ 1% on σabs and $\pm$ 2% on åabs. That is the range of uncertainties predicted to σabs and åabs. To make this issue clear, we rephrased a couple of sentences on pages 11558 and 11559 as follows: "The sensitivity tests indicate that the main source of error to σabs and åabs is the choice of the scattering Ångström exponent (åscat). In the worst case, assuming the average value åscat = 2.0 may cause a maximum deviation of $\pm$ 10% on σabs and $\pm$ 40% on åabs, depending on the different possible combinations of åscat and ω0 values. In the best case, the corresponding deviations would be $\pm$ 1% on σabs and $\pm$ 2% on åabs. All values reported in this work should be considered under these uncertainty ranges. However, neither worst nor best case uncertainties might be applicable for every point of the dataset analyzed here. In our view, the typical uncertainties are $\pm$ 5% on σabs and $\pm$ 20% on åabs.". Concerning the issue of low absorption coefficients being correlated with low Ångström exponents, we added error bars to Figure 4, considering the mentioned typical uncertainties. Answering the question about detection limits, (former) Figure 4 includes exclusively absorption coefficients inferred from attenuation coefficients that satisfied the filtering criteria stated in Section 2.5: I) The adjusted R2 of the quadratic fit should be greater than 0.85 II) The measured attenuation should be above the detection limit of the instrument, considering its sampling time and flow. Even with the inclusion of the error bars in (former) Figure 4, we see a clear association between low absorption coefficients and low Ångström exponents. In our understanding, this is an indication that Amazonian biogenic particles have a spectral dependency weaker than Amazonian biomass burning particles. We agree with the referee in the sense that we should not mention a value for the Ångström exponent (<1) without mentioning the uncertainties. Therefore, we rephrased and included a couple of sentences as follows: Page 11561, line 14: "Also, Fig. 4 shows that in the SMOCC experiment

ACPD

11, C6731–C6737, 2011

low absorption coefficients were associated with Ångström exponents below 1.0, with a typical uncertainty of ± 20% (refer to Section 2.4)." Page 11562, line 3: "The fact that 90% of the observed Ångström exponents are below 1.5 (with a typical uncertainty of ± 20%) reinforces the point that Amazonian biogenic particles may have a weak spectral dependence for absorption." Page 11563, line 13: "The inference of aerosol absorption coefficients from attenuation coefficients requires a previous knowledge of the spectral dependency of the single scattering albedo, among other parameters. In the absence of the necessary supporting measurements, average values were used to obtain corrected absorption coefficients from Aethalometer observations. Errors associated with the use of averages instead of specific values of single scattering albedo were evaluated through sensitivity tests. A precise value for the uncertainty on absorption coefficients or Ångström exponents is not achievable, but it is possible to predict a range of uncertainties. We estimate the typical uncertainties as ± 5% on σabs and ± 20% on åabs. All values reported in this work should be considered under these uncertainties." Page 11548, line 20: "Also, results indicate that low absorption coefficients were associated with low Ångström exponents. This finding suggests that biogenic aerosols from Amazonia have a weaker spectral dependence for absorption in comparison to biomass burning aerosols, contradicting our expectations of biogenic particles behaving as brown carbon."

Comment 3: Page 11554, line 1: should be "ΔATN"? Response 3: Yes, it should be "ΔATN", and we corrected it in the text.

Comment 4: Page 11554, line 13: there might by a typo in "Eqs. (4) to (2.5)" Response 4: The correct form is "Eqs. (4) to (6)". This mistake may have happened during the conversion to Latex, as the right form figures in our original .doc file.

Comment 5: Page 11555, line 12: Was the value of 20% calculated from Eqn. 8? Which ATN corresponds to this value? Response 5: The filter loading correction (R) for the Amazonian aerosol was obtained by Schmid et al. (2006) through a comparison between Aethalometer attenuation coefficients and photoacoustic spectrometer (PAS)

ACPD

11, C6731–C6737, 2011

absorption coefficients used as the reference measurement. Each measurement cycle of the Aethalometer begins with an acclimatization phase during which a pristine spot of the filter tape is put into place and the measured ATN is defined as 0. As absorbing aerosols deposit onto the filter spot, the filter gets darker and the light attenuation ATN increases. According to their set-up, at the predefined ATN value of 75 the filter tape was automatically advanced to expose a new pristine filter spot, and the cycle starts again. They verified that when ATN reached 75, the maximum loading correction observed was 0.8 (Figure 7 from Schmid et al., 2006). This is how they arrived at the value of 20%. We decided not to include this discussion in the article, because we understand that the reader might refer to Schmid et al. (2006) for further details.

Comment 6: Page 11555, line 23: Is it valid to use values forms and C from Arnott et al. (2005)? To my knowledge in Arnott et al. (2005) no biomass burning or biological particles were investigated. Can the authors comment on that? Response 6: Arnott et al. (2005) compared Aethalometer measurements with those from a PAS (photoacoustic spectrometer), supported by aerosol scattering measurements. Their values of ms (the fraction of the aerosol scattering coefficient that is erroneously interpreted as absorption) and C* (the multiple scattering correction factor that includes the effects of aerosol and filter matrix scattering) were obtained for ammonium sulfate aerosol, i.e., non-absorbing particles. Therefore, Arnott's parameters might represent the upper limit of absorption overestimation due to aerosol scattering effects. Unfortunately, during the experiments analyzed here, there were no simultaneous scattering measurements. If there had been, we could use a procedure similar to that used by Arnott et al., (2005) to derive site-specific values for ms and C* for the SMOCC experiment. Thus, for lack of better choices, we followed the procedure from Schmid et al. (2006), who also used Arnott's parameters in their calibration. Other than Schmid et al. (2006), we are not aware of other published articles investigating the calibration of Aethalometer with regard to Amazonian biomass burning or biogenic aerosols that could provide us with better estimates for ms and C*. We added the following sentence to Page 11555: "The parameters ms and C* are provided by Arnott et al. (2005) for

ACPD
11, C6731–C6737, 2011

ammonium sulfate particles. We are not aware of other published works investigating the performance of Aethalometers specifically with regard to Amazonian aerosols that could provide us with better estimates for ms and C*."

Comment 7: Page 11557, lines 16 to 18: The authors derived factors for the conversion of attenuation to absorption coefficients. I think the authors should compare their conversion factors to values given in the literature, e.g. Collaud Coen et al. (2010). Response 7: We included the following sentences to page 11557: "As a matter of comparison, Collaud Coen et al. (2010) found multiple scattering correction constants (C) ranging between 2.8 and 7.8 at 660 nm for several sites, measurement conditions and correction procedures. In this work, C averaged 5.72 $\pm$ 0.14 at 660 nm."

Comment 8: Page 11559 line 3: The worst case error is given to be 25 %. Does this worst case error explain the contradicting expectation discussed in the abstract (page 11548, line 22) and on page 11561, line 14. Response 8: Please refer to Response #2.

Comment 9: Page 11560 line 2: "dmËĘ3" is that a typo? Response 9: Curiously, in the version published at the ACPD website, "dm3" figures, and not "dmËĘ3". It may be that this referee had another version of the manuscript.

Comment 10: Page 11560 line 5: Can the authors explain how the detection limit was determined? Response 10: The detection limit was determined based on equation 13. To make the text more clear, we rephrased the sentence as follows: "Equation 13 was used to calculate the Aethalometer detection limit for the different field studies considered here, as can be seen in Table 1."

Comment 11: Figures 2 and 4: Typos in the vertical axis title "Mm-1" Response 11: The typos were corrected.

Comment 12: Figure 5,4,6,3: Typos in the horizontal axis title "angstrom" Response 12: The typos were corrected.

ACPD

11, C6731–C6737, 2011

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 11547, 2011.

ACPD

11, C6731–C6737, 2011

Interactive Comment