Interactive comment on “The impact of biogenic carbon emissions on aerosol absorption in Mexico City” by N. A. Marley et al.

N. A. Marley et al.

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Response to Reviewers:

We wish to thank the reviewers for their effort, time, and kind comments on the quality of the paper. We also received private email that noted possible confusion with our original title for the paper between biogenic emissions of VOCs and biomass burning emissions. As our methods do not allow differentiation of the two sources, we are modifying the title to clarify that to “The Impact of Biogenic Carbon Sources on Aerosol Absorption in Mexico City.” In response to the reviewer’s comments we have revised the manuscript and are submitting revised paper and figures to ACP. Specific responses to the reviewer’s comments and suggestions follow:

D. Baumgardner 1. It was suggested that an analysis of the validity of AAEs derived
from the 7 wavelength aethalometer could be made by a comparison with AERONET data. The AAEs derived from the aethalometer were made on fine mode (< 2 micron) aerosols located at the surface while AERONET measurements represent total column aerosol loadings. As a total column measurement, AERONET Ångstrom exponents (AEs) are for total extinction measurements and therefore are a sum of the aerosol absorption Ångstrom exponent (AAE) and the aerosol scattering Ångstrom exponent (SAE). The value of the SAE is dependent on the particle size distribution with higher values (SAE > 1) typically observed for accumulation mode particles (0.1–2 micron) and lower values (SAE < 0) for coarse mode particles (>2 micron). In addition, the AEs calculated from AERONET data for the Mexico City area are for 440–870 nm whereas the aethalometer AAEs were calculated from 370–950 nm. The addition of this UVB wavelength in the aethalometer calculations would be expected to result in higher AAEs than those calculated from the AERONET data for similar aerosols. Therefore the measurement methods are sufficiently different that a comparison would not serve as a validation of the 7-channel aethalometer method.

Instead, we offer another comparison as a validation of the AAEs derived from the 7 wavelength aethalometer. We have obtained continuous spectra (280–850 nm) on the same samples used for carbon isotopic analysis in the laboratory by state of the art diffuse reflectance integration sphere spectroscopy. This method is not susceptible to the same interferences as the transmission technique used by the aethalometer. Reflectance intensities are not proportional to sample path length and therefore increases in path length arising from multiple reflections in the filter do not interfere with absorbance determinations as they do in transmission measurements. In addition, surface scattering resulting in intensity losses not associated with absorbance in the transmission technique are not important in the reflectance measurements due to the fact that all scattered light is collected in the instrument by the integration sphere. The AAEs calculated by using all 350 points of the diffuse reflectance spectra were compared to the AAEs obtained by the aethalometer averaged over the same time period. A detailed treatment of this comparison has been added to the manuscript.
2. Although the AAE is commonly used to describe the wavelength dependence of absorbing aerosols, we strongly agree that the assumption that this dependence is purely an exponential function may not be valid in all cases. We have seen evidence of narrow band absorptions in the aerosol diffuse reflectance spectra that are masked by the overuse of the AAE to describe the wavelength dependence of aerosol absorption. However, since an assumed aerosol AAE is very often used to convert absorption at one wavelength to that at another, we feel that a study of how they vary is important.

The estimation of the goodness of fit of the wavelength dependence to the exponential described by the AAE can be given by the correlation coefficient (r^2) of the least squares fit to Equation 3 for all calculations. This value varies from 0.9999 to 0.96 with a standard deviation of 0.004 in 2003 and from 0.9997 to 0.95 with a standard deviation of 0.006 in 2006. This discussion has been added to the manuscript.

Although in all cases the fit was not considered poor, the occurrence of an r^2 < 0.999 may serve as an indication of a narrow band absorption superimposed on the exponential background of the broadband absorbers. This is currently under investigation in our laboratory and will be the subject of a future publication.

3. We agree that a comparison of ground based measurements with total column measurements (e.g. AERONET) would be interesting. However, since total column measurements are of total extinction, any detailed comparison should be made with total extinction made at ground level. It is difficult to estimate the relative contributions of aerosol absorption and scattering to the total Ångström exponent (AE). Since total column measurements of AE are most often used to estimate particle size, it can only be concluded that the contribution of scattering is not insignificant. Therefore a comparison of ground based AAEs with AERONET AEs as an attempt to validate the AAEs determined at ground level may be misleading without also knowing the wavelength dependence of aerosol scattering or the aerosol size distribution in the total column.

4. As reported by Yokelson et al. (2007, 2009), springtime fires from deforestation
and crop clearing are common in the Yucatan as well as in the surrounding mountains of Mexico City. This most likely contributes to the overall elevated absorbing aerosol background in the area. However, spring of 2003 was a particularly intense period in the Yucatan. Both NOAA and NASA websites compare that period to the 1998 fires that impacted areas in the US as far north as Wisconsin. The study by Massie et al. indicated that heavy smoke plumes directly impacted Mexico City most during the last week of the 2003 study when both AAEs and modern carbon were highest. A more complete discussion of these impacts has been added to the manuscript.

The differences in impacts from forest fires (C-3 plants) and local grass fires (C-4 plants) in 2006 can be seen in the stable carbon isotopic content of the aerosols (Figure 7 & 8). Although there is most likely an elevated background level of aerosols in the entire area from continuing forest fires, the higher C13 seen at T1 is a direct indication of the aerosol input from burning C-4 plants (grasses).

5. Figure 4 (now Figure 5) has been changed from 2 panels to 3 panels for better clarity.

6. Typos in Figures 4, 6, and 7 (now Figures 5, 7, 8) have been corrected.

Referee 3.

1. Figures 2, 4-7 (now Figures 3, 5-8) have been altered in an attempt to improve clarity. Colors have been avoided whenever possible due to reproduction problems. Closed and open symbols along with solid and dotted lines are used instead. Figure 4 (now Figure 5) has been split into 3 panels.

2. Abscissa of Figure 3 (now Figure 4) has been changed DOY 55-90.

3. Figure captions of Figures 4, 6, and 7 (now Figures 5, 7, 8) have been corrected.

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