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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Measurements of aerosol absorption and scattering in the Mexico City Metropolitan Area during the MILAGRO field campaign: a comparison of results from the T0 and T1 sites" *by* N. A. Marley et al.

N. A. Marley et al.

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The authors thank the reviewers for the detailed comments and suggestions. We have undertaken a major revision to the manuscript that has included changes in response to the reviewers comments and suggestions, as appropriate, and also includes the recalculation of all data sets and figures to 550 nm instead of the original 670 nm of the MAAP. This was done to offer a simpler and clearer comparison for all data sets presented. We feel that this revised manuscript has addressed all of the reviewer comments and should now be suitable for publication in ACP.

Specific responses to comments from reviewers follows:

Reviewer #1.

1. References to the wavelengths used have been clarified.

2. The size range of the aerosols sampled has been clarified as 0.1 to 2 micron aerodynamic diameter.

3. The rains on day 86 occurred very late (19:00) outside the daylight hours and after the MFRSR had stopped recording data. The SSA reported by Doran et al 2007 from the MFRSR are averaged over the daylight hours only as it is a radiometer.

4. A discussion of the problems associated with the filter based absorption measurements and how they may or may not impact this study has been added to the manuscript. We have also compared our aethalometer results to photoacoustic measurements made co-located at the T0 site (Paredes-Miranda et al., 2008)) and also with subsequent spectral analysis made in the laboratory by integrating reflectance spectroscopy (Kilaparty et al., 2008) and found excellent agreement. Sentence added and reference to Paredes-Miranda et al, 2008 added.

5. The SSAs reported are for fine aerosols (0.1-2 micron aerodynamic diameter) only. The calculated SSAs are not meant to be used to determine total radiative balance in the Mexico City area but are reported as a measure of the optical properties of the fine mode aerosols. Total aerosol SSAs used for the determination of total radiative forcing were measured by MFRSR and satellite during MILAGRO. However, the fine mode aerosols will remain in the atmosphere the longest and are responsible for local heating of the boundary layer effecting atmospheric circulation patterns (Ramanathan and Carmichael, 2008; Chung and Zhang, 2004). This difference is further clarified in the manuscript by noting fine aerosol SSA as appropriate, and a comparison has been added of total aerosol column SSA MFRSR results previously reported by Doran et al. with the fine mode aerosol SSA results from this work.

6. Submicron particle scattering peaks at 45 deg toward the forward direction, which is

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defined as 0 deg. This has been clarified in the manuscript.

Reviewer #2

1. The results of the INDOEX study and subsequent modeling efforts have shown that the effect of absorbing aerosols on the convective available potential energy (CAPE) of the boundary layer is strongly dependent on their vertical distribution. While the presence of absorbing aerosols aloft can result in a significant cooling of the underlying land surface decreasing the CAPE and stabilizing the boundary layer, the presence of absorbing aerosols near the surface has the opposite effect, increasing the CAPE and resulting in a large-scale rising over time (Chung and Zhang, 2004). The aerosols in this study were sampled at 10-15 m above ground level. A comparison with results from the G1 aircraft showed that the absorbing aerosols were not well mixed. Therefore the radiative effects of the aerosols sampled in this study would be at the surface. This has been clarified in the manuscript and references added.

2. The absorption and scattering measurements reported in this study were taken on a 5 min time resolution and averaged by using an hourly running average to reduce the effects of instrumental noise and short term excursions due to local sources. This method is similar to that used in a "Savitsky-Golay" data smoothing function. The SSAs were also smoothed according to the same method. The daily averages were made according to the method used by Doran et al. for direct comparison to their work.

3. The TSI wavelengths are 450, 550, and 700 nm. The typo on page 12632 has been corrected.

4. All absorbance data are reported for standard conditions as was the data reported by Doran et al. The data sets and figures have been reworked to report values at 550 nm instead of the original 670 nm. This was done as a simpler comparison of all data sets. Figures have been revised as well as clarified in the text.

5. We use the symbols "alpha"to designate the Angstrom coefficient for total extinction

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(absorbance + scattering) "alpha sub A" for the Angstrom coefficient for absorption and "alpha sub S" for the Angstrom coefficient for scattering. This has been further explained in the manuscript.

6. The typo on pg 12634 has been corrected.

7. Data have been normalized to the 550 nm wavelength of the PSAP instead of the 670 nm wavelength of the MAAP for a simpler comparison thus avoiding the need to correct the PSAP data. However, it is interesting to note that laboratory examination of fine aerosol filter samples by using integration reflectance spectroscopy have confirmed high Angstrom coefficients for absorption at site T1 (Kilaparty et al., 2008, Marley et al., 2008).

8. The TSI nephelometer was calibrated with CO2 according to the manufacturers suggested procedure. The Radiance Research nephelometer was calibrated by comparison to another nephelometer at the laboratory at UNAM. This has been clarified in the manuscript. Aerosol scattering also reported by a reciprocal nephelometer operated with a photoacoustic spectrometer agreed well with the results reported here for T1 (Paredes-Miranda et al., 2007; 2008).

9. The RB meters were not operated side by side during the study, however, they have been operated side by side in Chicago IL. The temperature stabilized RB meters have been used for long term monitoring of UVB in monitoring networks and have been found to be very stable over the lifetime of the RB meters. Measurement uncertainties are usually due to calibration uncertainties (Weatherhead, 1997). The Solar Light company offers NIST traceable calibrations to ensure a high level of accuracy. In addition, sensitivity differences would not appear as a function of zenith angle. This has been clarified in the manuscript and references added.

10. Figure 6A, the mass absorption coefficient for total carbon has been deleted. Although the assumption of 30% BC is an approximation, we justify the choice by referencing past work in Mexico City and other urban areas (Chow et al., 2002; Tanner

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et al., 1982; Gaffney et al., 1984). In addition, the values in Figure 4 were averaged over the daylight hours so variations in % BC over the period (possibly higher in the morning and lower in the afternoon) would be averaged out. This is reported to allow comparison to previous results and back trajectories reported by Doran et al.

11. We have looked at the ozone data and the concentrations at site T0 and T1 were very similar at both sites for the days used in the UVB comparisons. If anything, T1 was slightly lower on the average than T0. This observation has been added to the manuscript.

12. Equation 5 has been consolidated into the explanation for total Angstrom coefficient (Equations 1-3). The Angstrom coefficient for scattering reported is for the fine mode aerosols only and is used to estimate the fine mode particle size and the scattering asymmetry factor for the fine mode aerosols.

13. The direction of light scattered by particles is dependent on the size and shape of the particle and not the particle composition. Mie scattering theory predicts that particles of the same size as that of the wavelength of the incoming radiation will scatter the radiation most favorably 45 deg to the forward direction. The dominant particle size of the fine mode particles is determined by the Angstrom coefficient for scattering to be 0.3 micron or 300 nm, which is of a similar size as the UVB radiation. Therefore the UVB spectral range will be scattered toward this forward direction by these fine mode particles. This will not necessarily be the case for radiation of longer wavelengths. This is clarified in the manuscript.

14. The data in Figures 10 and 11 were not averaged. They are on the original five minute time resolution on which the original data were obtained. This is clarified in the text.

Reviewer #3

General Comments: All appropriate references suggested by the reviewer have been

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added to the manuscript. All typos have been corrected.

The primary focus of this manuscript is to describe the fine aerosol optical properties at sites T1 and T0 for other investigators involved in aerosol radiative effects related to climate. As the reviewer points out, there are many excellent publications on aerosol composition from the MILAGRO and MCMA2003 studies. These references have been added to the manuscript for easy access to the reader. However, a detailed comparison of the aerosol optical properties with composition studies is beyond the scope of this work and is best left for a future publication.

Specific Comments:

1. Although it is true that hot diesel exhaust at the exit of the tailpipe contains particles smaller than 100nm, these ultrafine particles very rapidly diffuse to surfaces and become incorporated into the accumulation (fine) mode (Williams et al., 2002). It is not the purpose of this study to measure the optical properties of all particles in the total atmospheric column, but to focus on particles that most efficiently effect radiation and climate on regional scales. These are the particles that both most efficiently scatter and absorb radiation and have lifetimes in the atmosphere sufficiently long to influence regional climate i.e. the fine mode or accumulation mode aerosols.

2. The major mechanisms for particle loss in sampling inlets are impaction on external surfaces. These losses can easily be calculated by using the particle Stokes number and the inlet bend ratio. This has been reported for an inlet similar to ours with two 45 deg bends and a flow rate of 10 L/min and the particle size cut off has been calculated to be 2 micron (Hermann et al., 2001). This reference has been added to the manuscript for clarification. This size cut off was confirmed during the MCMA2003 study by comparison of aerosol scattering results to a LASAIR particle size counter (Salcedo et al., 2006).

3. The overall averages have been removed from the abstract. We still believe that the similarity of the absorption ranges and diurnal profiles at both sites T0 and T1 are

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much more similar than that for the scattering at the same sites.

4. The values for BC and OC have been reported from Chow et al. (2002) as 32% and 15%. It is very interesting to note that the OM data reported in 2003 also gives values for OC of 32% and a BC of 11%. This is a very strong indication that the atmospheric aerosol carbon content has not changed significantly since 1997. This has been clarified in the manuscript.

5. The discussion of sulfate aerosol emissions from the MCMA basin is deleted since it is only a minor point in the introduction.

6. Most references to BC or EC have been avoided as Doran et al., reported EC by thermal evolution and our total carbon measurements were also made by thermal evolution. Therefore the measurements are operationally comparable.

7. The contributions of the sources outside the megacity to the region is not the focus of this manuscript. Moreover, the section in question is only a summary of observations made by Doran et al. 2007.

8. Meteorological parameters for both sites for the month of March have been added.

9. The difference in aerosol scattering at the three different wavelengths of the nephelometer is dependent on the particle size, which determines the Angstrom exponent for scattering. These results are used later in the manuscript to determine fine mode aerosol size distribution.

10. The MRI nephelometers operated at 550 nm.

11. It is stated in the manuscript that C14 analysis confirms a large portion of aerosol carbon arises from biomass sources. It is admitted that these sources can include forest and grass fires as well as biogenic SOA and urban burning. However, it is beyond the scope of this manuscript to provide an in depth analysis of the sources. This is the focus of another manuscript which will shortly appear in ACPD (Marley et al, 2008).

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12. The discussion of aerosol Angstrom absorption exponents and their possible sources at site T1 has been removed due to the change in wavelength correction of the data from 670 nm to 550 nm. Focus is now on T0. An in depth discussion of the aerosol absorption Angstrom exponents at T1 is left to another manuscript.

13. A review of the rapid morning formation of secondary organic aerosols in Mexico City has been added.

14. The numbers for day of year have been given for easy comparison to back trajectories reported by Doran et al. In all cases the corresponding dates are now also given.

15. The section on diffuse radiation field has been expanded.

16. All units in the manuscript are SI units.

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