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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.

Anonymous Referee #3

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Review of Marley et al.

This paper describes measurements of light scattering and absorption in the visible collected at the T0 and T1 sites during MILAGRO. This is an important topic with clear implications for local and regional air quality and climate. The paper mostly reports the measurement time series and describes them in limited detail. The paper has several important limitations, and in my opinion it needs to be significantly reworked before it would be acceptable for publication in ACP.

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Major issues

- The results presented here are compared to a very limited set of previous studies, many of which are one or two decades old and mostly by the authors. Very little cross-linking is made with the large number of publications arising from the MILAGRO campaign, from the MCMA-2003 field study, and other recent papers from Mexico City, including many papers on the same topics as this one. This could be appropriate for a technical report but it is definitely not appropriate for a paper in the peer-reviewed literature. Many suggested references are given below.

- Almost no use is made of the extensive gas-phase and particle-phase datasets collected by other instrumentation deployed at the T0 and T1 sites to aid on the interpretation. The paper reads as if the authors in isolation had decided to go to Mexico City to collect and interpret the measurements presented here, and as if very few other measurements had been reported at this location (which contrasts with Mexico City being now one of the most studied urban locations on Earth).

Critical Technical Issue

- Page 12632, line 5, how were the inlets specified to transmit that size range? It is actually hard to get rid of particles above 25 nm or so, did the authors specifically design their inlet so that diffusion losses reduced the concentration of particles in the 25-100 nm range. If so, how and for what purpose? This could actually create a significant negative bias for the absorption measurements as many diesel exhaust particles have aerodynamic and mobility diameters smaller than 100 nm (see for example Figure 8 of Park et al., 2003, Figures 8-9 of Kittelson et al., 1998). It seems that the authors should have rather fought to keep particles down to 25 nm or so, which is actually not that hard. Is the size cut actually 100 nm, or is this a poorly estimated lower limit? Have the data here been compared to other absorption measurements to evaluate this potential bias?

- Also, how was the upper size cut of 2 microns chosen, designed, and calculated?

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How certain is this size limit? Why 2.0 and not the more common 2.5 micron cut, which would allow more direct comparisons to other measurements?

Specific comments

- Abstract: the average absorption at T0 is 63% higher than that at T1, and I would not describe these as "similar". The range is indeed similar but the average is more representative of typical scattering at each site.
- Page 12267 line 17, the inhibition of dispersion due to the mountains around the city is limited, see for example de Foy et al. (2006) entitled "Rapid ventilation of the Mexico City basin... " and Fast et al. (2007).
- Page 12267 line 21, the boundary layer can reach up to 3-4 km above the surface, see Figure 4 in de Foy et al (2006).
- Page 12627 line 26, Barth et al. (1999, cited later in the paper) have reported an impact of Mexico City emissions at much longer distances.
- Page 12627 line 29, the Mexico City aerosol is 55% organic matter (OM), not organic carbon (OC). The OC fraction can be calculated with recently reported OM/OC ratios for this location (Aiken et al., 2008, average of about 1.7) and is about 32%. This should be corrected to avoid confusion.
- Page 12628, line 3: the emissions are of SO₂, not sulfate aerosols. Also the MCMA urban area is a relatively modest source of SO₂, the larger regional sources being the Popocatepetl volcano and the power plant and refinery in Tula (which is outside of the MCMA). See de Foy et al. (2007), DeCarlo et al. (2008), and Grutter et al. (2008) who report observations of SO₂ and sulfate for these various sources and regions. Also note that Barth et al. used the SO₂ emissions from a global inventory that did not accurately reflect the actual emissions. If the authors wish to make a quantitative comparison of the SO₂ emissions from Mexico City and the total for the world they should compare the numbers from de Foy et al. (2007) and Grutter et al. (2008) to an

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up-to-date global inventory.

- Page 12630, line 6, EC and BC are not the same, one is quantified by thermal methods and the other by absorption measurements with an assumed mass absorption efficiency. These definitions can result in significant differences so the authors should be careful here and potentially point to some of the relevant papers in the literature (Andreae and Gelencser, 2007, Saathoff et al. 2003).

- Page 12630 line 9, several studies have reached similar conclusions about a significant, but not dominant, impact from biomass burning sources as part of MCMA-2003 and MILAGRO, including Kleinman et al. (2008), Molina et al. (2007), Stone et al. (2008), Yokelson et al. (2007), DeCarlo et al. (2008), Salcedo et al. (2006) etc. Several of these studies used either more specific techniques such as levoglucosan (Stone et al.) or aircraft measurements (Kleinman, Yokelson, DeCarlo) and would reinforce the conclusion presented here.

- Page 12630 line 9, many millions of people live in Central Mexico outside of the MCMA, e.g. in Toluca, Cuernavaca, Puebla, Pachuca... and it is not surprising that their emissions affect the regional air. This should also be mentioned here.

- General comment on the introduction: the recent review paper by Molina et al. (ACP 2007) which summarizes the results of the MCMA-2003 campaign touches on many of the issues described in the introduction and provides much more up-to-date information for many of the topics discussed here. We highly recommend that the authors read this paper and cite it (or its references) as appropriate here, which will provide the readers with more up-to-date links to the literature.

- Page 12631, lines 10-13, the annual climate parameters are given for T1, but the monthly climate parameters for the month of March, or the actual values during MILAGRO would be more informative.

- Page 12631, lines 22-23, again the climatological value of the maximum wind speed

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is of some interest, but it should be put in context of the wind speeds (range, average, median etc.) observed during this campaign.

- Page 12632 line 7, were there differences between the results at 530 nm and the other wavelengths? Either way this should be stated here.

- Page 12632 line 10, what is the wavelength used by the Meteorology Research nephelometers? Also 530 nm? Something else?

- Pages 12632-33. Literature references are available that describe, evaluate, and intercompare most of the instruments described here (3-wavelength nephelometer, PSAP, f(RH) measurement...). These should be given to allow the interested reader to more easily link the current observations to the literature.

- Page 12635 lines 1-2: the association of modern C with agricultural and forest fires during MILAGRO is less clear than implied here. Forest fires were very intense early in the campaign and then almost completely stopped for several days following the arrival of a clean air mass with a cold surge meteorological situation accompanied by rain, as described later in the paper. However the fraction of modern C in the aerosol only decreased slightly during this period. Together with the strong linking of a major fraction of the organic aerosol at the surface to urban primary and secondary sources by multiple researchers (Stone et al., 2008; Kleinman et al., 2008; Volkamer et al., 2006; Hennigan et al., 2008; Querol et al., 2008; Salcedo et al., 2006...) this strongly suggests that urban sources of modern C contribute a significant fraction of the modern C observed in Mexico City. These sources may include cooking, biofuel use, trash burning, SOA from modern precursors emitted by sources such as those, etc. A similarly simplistic interpretation of modern C data from the Northeast US would suggest that most of the organic aerosol in that region would arise from biogenic SOA. However many studies have pointed out a very strong association of the OC with anthropogenic sources and a weak one with biogenic VOCs and their reaction products (de Gouw et al., 2005, 2008; Weber et al., 2007; Sullivan et al., 2006). Thus only the excess modern C above that

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in the period with no fires can be clearly associated with non-urban biomass burning. The authors should avoid simplistic interpretations of the modern C fraction which do not take into account all the other pieces of evidence about organic aerosol sources in Mexico City.

- Page 12635 line 6, the average fractional impact of biomass burning at the T0 site is 32% according to Stone et al. (2008). The impact of this source alone cannot explain the trend described here since this source is not dominant. The parameters appropriate for urban primary and secondary OA should also be discussed here.

- Page 12635 line 10, the dependence of aerosol absorption on wavelength in Mexico City between the wavelengths used here has been reported by Barnard et al. (2008, Fig. 1) and follows an Angstrom exponent of 1 and not 2.1. Stone et al. (2008) report similar relative source influences at the T0 and T1 sites, and thus the empirical correction reported here may be incorrect. The value of 2.1 could be valid during specific days with very high fractional impact of biomass burning, but it most likely does not hold at all times during the campaign (given for example the very variable impact of biomass burning).

- Page 12637, line 10: here the results of this study are compared only with those from a single study in Santiago, Chile, for which most of the data were measured in 1988 (2 decades ago) and using a relatively rudimentary technique (integrating plate). The sources in both Santiago and Mexico City have changed a lot since 1988. Many more recent references exist, including measurements in Mexico City (Barnard et al., 2007, Barnard et al., 2008; Baumgardner et al., 2007; Johnson et al., 2005; Jiang et al., 2005; Raga et al., 2001, Marley et al., 2007) etc.) and other developing countries in recent years. Besides Santiago, it would be of interest to compare the results here with recent reports from rapidly developing countries such as China and India.

- Page 12637, lines 21-25, aerosol composition measurements were carried out at both T0 and T1 by several groups (Stone et al., 2008; Querol et al., 2008; Hennigan et al.,

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2008; Aiken et al., 2008...) and should also be linked to this discussion.

- Page 12637, line 28: the authors attribute the scattering diurnal profile solely to the formation of ammonium nitrate, which they had described earlier (page 12628, line 1) as comprising only 10% of the fine PM mass in Mexico City. The glaring omission here (and also at various other locations in the paper) is the contribution from the rapid formation of secondary organic aerosol in polluted urban air, which has been documented by many studies in Mexico City and is at least as large as ammonium nitrate (Volkamer et al., 2006, 2007; Kleinman et al., 2006; Hennigan et al., 2008; Stone et al., 2008; Salcedo et al., 2006; Aiken et al., 2008; DeCarlo et al., 2008 etc.) and elsewhere, for example the studies from New England cited above, also Crosier et al., (2007) for the Po Valley, Cubison et al. (2006), Johnson et al. (2006) in the UK, Miyakawa et al., 2008 in Tokyo, Docherty et al. (2008) for the Los Angeles area, etc.

- Page 12638, line 13: it would be better to refer here to the papers (de Foy, Fast) that focus on the meteorology instead of those by Doran et al.

- Page 12638 line 21: there is no reason to cite a non-peer reviewed source for the presence of photochemically aged aerosols in the afternoon, there is overwhelming evidence from published studies as described above, including at (and aloft over) the T1 site.

- Page 12639 line 1, the SSA of freshly emitted BC is not 0. These particles still scatter some light. For example Ban-Weiss et al. (2008) report SSA of 0.31 for gasoline vehicles and 0.2 for diesel vehicles.

- Page 12639 line 8: a study published in 1989 reporting data collected in 1986 is very far from "recent". Please make an effort, here and throughout the paper, to also link with more up-to-date literature.

- Page 12639 line 16: here, and also later in the paper and in the figures, the authors use day numbers. This is hard to follow and makes the comparison with other studies

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difficult, since everyone else uses the date ("March 21" etc.). Please replace the day numbers with dates throughout the paper. A table with the correspondence between the dates and the day numbers should be included in the Supplementary Information if the authors think that the day numbers can be useful to someone.

- Page 12640 line 11: here the first 2 days of the study are referred to as "the first part of the study" while and the other 18 days as "the second part of the study." Please rephrase to more precisely describe the data.

- Page 12640, lines 28-29: the MAE reported here are compared with measurements from Mexico City in 1997 (Chow) and 25-year old measurements in the other papers (Tanner, Gaffney). The sources of black carbon such as diesel engines have undergone major changes in the last 25 years. Again, please make an effort to link with the more up-to-date literature, including that from Mexico City (Barnard et al., 2007, Barnard et al. 2008; Baumgardner et al., 2007; Johnson et al., 2005; Jiang et al., 2005; Raga et al., 2001, etc.). Even a previous publication from the authors in the same topic, Marley et al. (2007) is not cited or compared with.

- Page 12641: what is the correlation between the time series of the measurements at T0 and T1?

- Page 12642 line 2: the previous mistake of associating scattering aerosols only with inorganic species is repeated here, the contribution of POA and SOA should also be noted.

- Page 12642 lines 10-14: here and elsewhere where similar comparisons are made: could a t-test be used to evaluate whether the differences are statistically significant? If some properties were relatively constant across times / locations within statistical uncertainty, this would be useful information for modelers.

- Page 12643 line 1: again compare with more recent measurements in Mexico City and not only with those from 1997.

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- Page 12643, line 23: similar results have been reported for Mexico City (Castro et al., 2001; Barnard et al., 2008) and they should also be discussed here. Note that the significant reduction in UV radiation in Mexico City has been reported before the start of the biomass burning season by Castro et al. (2001), so as expected it is also due to urban aerosols and not only biomass burning.

- Page 12644 line 4: the finding about the diffuse radiation field is important, but it is not explained in enough detail or related to the rest of the paper. Please expand this discussion.

- Page 12645 line 7: the fraction of secondary aerosols at T1 was at least as large as that at T0 if not larger (Hennigan et al., 2008; Stone et al., 2008; Kleinman et al., 2008).

- Page 12646 line 2: rephrase to be more accurate, as this study only reports on one megacity, not several.

- Figures 1 and 2: these figures as a time series, while not completely useless, are very hard to read. They should be moved to the supplementary information (with vertical gridlines to make it easier to interpret) and replaced in the main paper by the diurnal cycles of the properties measured, that will be much more informative for the reader.

- Similarly Figure 9 & 10 would be much more useful as diurnal cycles, while the current versions should be moved to the supplementary information.

Technical, grammar etc.

- Page 12626 line 7, English units are not appropriate for an international journal, please use SI units throughout the paper (km instead of miles). English units could be kept in parentheses after the SI units.

- Page 12626 line 13: "hose" seems to be typo

- Page 12627 line 16, the mountains rise much more than 1000 m above the basin floor of 2200 m. Popocatepetl reaches 5426 m, or 3200 m above the basin.

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- Page 12627 line 22 and page 12628 line 22, the same idea is repeated in these 2 locations.

- Page 12269 line 6, "that" should be "those"

- Page 12632 line 17, "decrease" should be "increase"

- Page 12634 line 2, "low" should be "high"

- Page 12634 line 25, "site T1" should be "the T1 site"

- Page 12636, line 19, replace scfm with SI units

- Page 12639 line 8: the author of this study is Clarke not Clark.

- Page 12639, lines 12 & 14: Doran et al. (2007) is repeated in this sentence.

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