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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An overview of the MILAGRO 2006 campaign: Mexico City emissions and their transport and transformation" *by* L. T. Molina et al.

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Response to Reviewer #1:

We would like to thank the reviewer for careful reading of this manuscript and for the thoughtful comments and constructive suggestions, which help to improve the quality of this manuscript. Our response follows:

General comments: This paper provides an overview of the spring 2006 MILAGRO field campaign. A well-crafted overview paper is important for such a field study, as it serves not only as a guide to the goals, operation and results of the program, but also ties the results together in a coherent manner. This paper largely achieves these

objectives. The paper is generally very well written (with a few exceptions noted below), and follows a clear and logical thread, introducing the entire MILAGRO program including its historical context, describing the measurement platforms and instruments, and then focusing on an extensive summary and synthesis of the results.

I have one primary reservation about this paper: it is extremely long (165 pages). The authors would be well advised to eliminate simple recitation of quantitative results and focus on a) the synthesis of results where possible and b) qualitative description of the work done with reference to the published papers. As much as possible recitation of quantitative results from published work should be eliminated. In these regard, I think that Section 8 on PM can be taken as a guide. I recommend that the paper be published once the following specific points have been addressed.

Reply: We agree that the article is very long; however, as indicated in the Abstract, the objective of this overview article is to provide a roadmap for the scientific community interested in understanding the emissions from a megacity such as the MCMA and their impacts. We have reviewed over 120 papers and have subdivided the measurements and the results under different topics (meteorology, emissions, gases and PM, photochemistry, aerosol radiative properties, and transport). It is written such that each subsection can be considered a stand-alone document.

We are trying to target both readers that are interested in the comprehensive study and readers that are only interested in subsections. The current form serves both of these needs. We are concerned that if we cut out the small overlaps in the introduction to each section as well as the technical material, the subsections will not be readable to the reader who is only interested in one or a few sections. Therefore we would like to keep the manuscript largely in its current form.

However, following the suggestion of the reviewer, we have reviewed carefully the entire manuscript and have removed redundancies in the revised manuscript.

Specific comments:

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1) The caption for Fig. 2 needs improvement with regard to ozone. It is clearly not simple annual averages that are plotted.

Reply: We have revised the caption, replacing "Annual Average" with "Estimated as the average of the 5th annual maximum from all stations with valid data for a given year."

2) In Fig. 10, the geographic location is not clear. Some specific features on the ground (e.g. the outline of MCMA from Fig. 1) should be clearly shown to orient the reader. Or is that the Gulf Coast of the U.S. shown? If it is a much clearer presentation is required.

Reply: We have revised Figure 10 and added the legend, showing clearly T0 and T1 supersites on the ground.

3) In the discussion of Fig. 10 the OH reactivity of background CO should be indicated. Only a fraction of the CO reactivity at the furthest extent is due to MCMA outflow. It should be contrasted with the reactivity of background CO, which is something like 130-140 ppbv in March at northern mid-latitudes.

Reply: As shown in Figure 10, the background CO is approximately 70-80 ppbv and the CO measured in the plume outflow is around 120ppbv, which agrees well with MOZART. We have added the following sentence at the end of the first paragraph in Sect. 6.5:

"At the C-130 interception point, approximately 60% of the CO reactivity is from the background CO."

4) Pg. 7866 – I do believe that the conclusion of the following statement is necessarily accurate: "However, afternoon ozone concentrations showed minimal changes over the weekend with occasional increases, providing direct empirical evidence that ozone production is VOC-limited." It may be that simple titration of ozone by the higher NOx emissions on weekdays accounts for the weekend-weekday differences. This requires a more complete discussion.

Reply: We agree that the simple titration of ozone by the higher NOx emissions on

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weekdays can contribute to the observed weekend-weekday differences. We have revised the text to more accurately reflect this effect:

"However, afternoon ozone concentrations showed minimal changes over the weekend with occasional increases, providing direct empirical evidence that ozone production is VOC-limited and NOx-inhibited during workdays."

A more complete discussion can be found in Stephens et al., 2008, as referenced in the manuscript.

5) Pg. 7871 – The following statement is not clear: "An important scientific and air quality management question in many of these megacities is quantifying the relative contributions of different sources such as mobile, industrial, biogenic, biomass burning, etc., as well as separating the contributions from primary emissions vs. secondary processes, both of which were an important objective of the MILAGRO Campaign." Contributions to what is not specified. Total PM, organic component of PM2.5?

Reply: To clarify the text for the reader, the sentence has been reworded to read:

"An important scientific and air quality management question in many of these megacities is quantifying the relative contributions to concentrations of PM from different sources such as mobile, industrial, biogenic, biomass burning, etc., as well as separating the contributions from primary emissions vs. secondary processes, both of which were an important objective of the MILAGRO Campaign."

6) Pg. 7872 - The following statement is not clear: "Scanning Transmission X-Ray Microscopy (STXM) spectra of collected particles (Moffet et al., 2010), and 14C content of organic carbon, elemental carbon, Water-Soluble OC (WSOC), and Water-Insoluble OC (WIOC) (Aiken et al., 2009b) were measured in the MCMA." Was 14C content of all four (OC, EC, WSOC and WIOC) really measured separately?)

Reply: Yes, 14C context was really determined for all four fractions. To clarify this, the text has been changed to read:

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"14C content of carbon fractions (organic carbon, elemental carbon, water-soluble OC (WSOC), and water-insoluble OC (WIOC))."

7) Pg. 7886 - The following statement is not clear: "This study found that organics were causing a surface depression of 10–15%." What was depressed?

Reply: The word "tension" is missing after "surface." The corrected sentence now reads:

"This study found that organics were causing a surface tension depression of 10–15%."

Technical Corrections:

1) Pg. 7840, line 19 – Replace "data is" with "data are".

Reply: The suggested correction has been made.

2) Pg. 7844 – There is no need to reiterate percentages in the text that are already clearly displayed in Fig. 6.

Reply: Although the percentages are displayed in Fig. 6, we have decided to leave the text as is to highlight the importance of the contributions from gasoline and diesel vehicle fleets as well as other important emission sources.

3) Pg. 7846, line 8 – Eliminate the term "research grade, real-time", as it is ill defined. Perhaps "sensitive, fast response" would be more informative. The same comment applies to pg. 7909, line 17.

Reply: We have revised the term as suggested.

4) In contrast to most of the paper, Section 5.3 has several typos and grammatical errors, and the writing is not clear in places. It should be carefully edited.

Reply: We agree with the suggestion; this section has been carefully copy-edited.

5) In Fig. 10, the geographic location is not clear. Some specific features on the ground (e.g. the outline of MCMA from Fig. 1) should be clearly shown to orient the reader. Or

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is that the Gulf Coast of the U.S. shown? If it is the latter, a much clearer presentation is required.

Reply: Figure 10 has been revised with legend showing T0 and T1 supersites on the ground.

6) Pg. 7865, line 28 – The phrase "that emission controls would depend on location and meteorology" is incorrect. What would be correct is ":that the effectiveness of particular emission control strategies would depend on location and meteorology"

Reply: We agree; the text has been revised as follows:

"Furthermore, meteorological conditions led to large variations in regime for the relatively low-NOx emitting area, implying that that the effectiveness of particular emission control strategies would depend on location and meteorology [Song et al., 2010]."

7) Pg. 7905, line 5 – The conclusion "– Many hydrocarbon emissions show greater enhancement ratios in the MCMA than the US." is not a clear statement in isolation. It should be clarified.

Reply: We agree; the sentence has been modified as follows:

"Many hydrocarbon emissions show greater enhancement ratios to CO in the MCMA than the US due to the widespread use of LPG and higher industrial and evaporative emissions of aromatics in Mexico City."

8) Pg. 7905, line 7 – The conclusion – Total OH reactivity due to VOCs in the MCMA remains largely unchanged from the 2003 study; however the speciated attribution is quite different; the present study found that formaldehyde and acetaldehyde were the two most important measured VOC species for OH reactivity - is not clear. Has the VOC speciation in the ambient atmosphere changed, or is the MILAGRO result an improvement over an earlier, erroneous result? This is important to clarify.

Reply: We cannot quantitatively compare these results due to different techniques used

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and different locations; we have modified the sentence as follows:

“Formaldehyde and acetaldehyde were the two most important measured VOC species in terms of OH reactivity in the MCMA.”

We have also modified the text on page 7857, line 21, as follows:

“The OH reactivity determined at T0 is broadly consistent with the results from the MCMA-2003 study at the CENICA supersite located approximately 7 km SE of downtown Mexico City. During MCMA-2003 Shirley et al. [2006], using the Total OH Loss Measurement instrument (TOHLM), reported an average daily OH reactivity of 33 s-1, and estimated that 72% of it (∼24 s-1) was due to VOCs.”

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C5857/2010/acpd-10-C5857-2010-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 10, 7819, 2010.