

*Referees comments are in Helvetica italics.*  
My replies are in Times standard.

### **Anonymous Referee #1**

*This paper describes an evaluation of a global chemical transport model using the extensive trace gas and particulate measurements collected during the MILAGRO field campaign. The model is then used to determine the sources of pollution, the age of pollution, and ozone production over Mexico. The paper is well organized, details of how the analyses are performed are described adequately, and the findings are articulated clearly; however, my primary concern is that the motivation for the particular model analyses needs to be described better. The introductory material should be revised to describe why the topics investigated in this paper are important. The conclusions should also reflect on whether the present results regarding this megacity applicable to other megacities around the world.*

Some text has been added to the Introduction regarding the importance of the topics covered in this paper. A paragraph has been added to the Conclusions discussing the uniqueness of Mexico City in relation to other megacities.

*The following is a list of other specific comments I have:*

1) *Page 3461, line 21: Suggest changing “Z” to “UTC” for time.*  
done

2) *Page 3462, first paragraph: Please include whether estimates of volcanic emissions of sulfur dioxide are included or not in the model run. Are dust emissions included in the model run? Although not central to the paper, the effect of NO<sub>y</sub> uptake on dust is mentioned on page 3471.*

This paragraph did mention that volcanic SO<sub>2</sub> emissions are included in MOZART-4, but additional discussion has been added, including mentioning the Popocatepetl emissions in the model are significantly lower than those observed by Grutter et al. (ACP). A description of how dust is included in the model (climatological 3-d fields) has also been added.

3) *Page 3463, line 16: Include the number of flights used in this analysis for both the C-130 and D-8.*

The number of flights used in this analysis for both the C-130 (12) and DC-8 (6) has been added.

4) *Page 3463, first paragraph in Section 3: It would be useful to state why measurements from the G-1 aircraft are not included in the analysis. Presumably, the flight paths occur largely in one or a few MOZART grid boxes so the model would show as much variability as observed.*

The reasons for not using the G-1 data (due to the city-scale coverage of the flights) have been added.

5) *Page 3464, line 14: The authors refer to uncertainties in volcanic emissions, but do not state earlier whether they are included or not in the simulation. Previous papers*

*suggest that sulfur dioxide emissions from Popocatepetl could be quite large compared to other sources in the region.*

See response to comment 2.

*6) Page 3464, line 16: It is true that the simulated sulfate could be improved by adding emissions of sulfate, but it would be better to either add more realistic volcanic emissions or evaluate why the particulate mechanism in the model is producing too little sulfate.*

I removed comment about including direct sulfate emissions, since the amount (a few percent) sometimes included in model simulations would not significantly improve the simulation.

*7) Page 3464, line 20: Suggest changing the text to indicate the model over-predicts the average OA concentration somewhat, but there are very large over-predictions at times.*  
The discussion of the model performance for OA has been re-written to more simply state that the model significantly under-predicts the observations at times.

*8) Page 3465, line 17: Different regimes are mentioned. Perhaps it would be useful to change Figs. 1 and 2 to use flight paths within the same latitude/longitude box so that the results are more comparable. Or perhaps breaking the analysis in Figs. 1 and 2 into flight paths over central Mexico and those transects further downwind. This*

It is a very good suggestion to separate the measurements near and far from Mexico City, so we have separated Figures 1 and 2 each into these two categories. A figure of the flight track locations and altitudes has been added.

*9) Page 3465, line 24: As far as the underestimation in photolysis rates, could this be due to uncertainties in simulated clouds? How are cloud fields included in MOZART? This is probably described in another paper, but seems relevant here.*

A brief description of the formation of clouds in MOZART-4 has been added, with acknowledgement that their misrepresentation could be cause of errors in the photolysis rates.

*10) Page 3465, line 28: OVOCs has not been defined yet. Here and elsewhere, the authors should check that acronyms are defined before being used.*

Done.

*11) Page 3467, lines 14-16: This sentence does not represent what is in the figure, and needs clarification. I agree that other large cities in central Mexico contribute to regional pollutants, but the figure that is being referred to does not really distinguish those cities from Mexico City. All that is seen is a single source in central Mexico.*

The discussion of the additional cities surrounding Mexico City has been moved to the discussion of Figure 5 (Fig.6 in the revised paper), which shows the emissions used in the models.

*12) Page 3468, lines 8-11: Why are the biomass-burning emissions seemingly centered over Mexico City? Were the largest fires located in that region during the month? I would have expected that fires would be distributed throughout the region. Perhaps*

*adding a panel on the locations of the biomass-burning source would be helpful to the reader.*

A plot of the monthly average CO fire emissions has been added to Fig. 5 (Fig.6 in the revised paper), replacing the NO emissions plot. There were many fires in the hillsides surrounding Mexico City, in addition to the agricultural fires throughout the region. On the scale of the model there is significant overlap with the location of the anthropogenic emissions.

*13) Page 3468, line 21: The winds did not shift to northerly until March 23 (the third Norte described in Fast et al.) and Fig. 6 shows the increase of US+Canada sources on this day and not March 20.*

March 20 changed to March 23.

*14) Page 3468, line 22: Change “increases” to “minor increases”?*

Done.

*15) Page 3470: line 5: I am not sure this is a “large “region. Suggest changing “A large region over the Gulf of Mexico, north of the Yucatan peninsula, contains“ to “Just north of the Yucatan peninsula is a region containing”.*

Done.

*16) Page 3470, line 9: “Long ages” seems awkward.*

Changed to “large ages”.

*17) Page 3470, line 16: The second part of the sentence starting at “the majority of the CO” is redundant.*

I disagree. No change made.

*18) Page 3471, line 14: Is there a reference for the box model that can be included? Does the box model contain the same mechanism as MOZART? If not, would this not affect the comparisons in Fig. 9 where the unconstrained MOZART model results are compared to the constrained box model? Please explain.*

There is a reference on line 17. It has been moved to directly after ‘box model’. No, the chemical mechanisms are different. This could certainly play a role in the comparison. The assumption is that the box model will more accurately reproduce the truth through its constraint to the observations and more detailed chemistry. This has been clarified in the text.

*19) Page 3471, line 21: I do not understand the sentence starting “However, the MOZART-4 . . .”*

The sentence has been re-written to clarify that MOZART-4 is being compared to the box model diurnal averages.

*20) Page 3472, line 1: Can the authors speculate why MOZART had greater difficulty in simulating the high concentrations in this region. Is it primarily the spatial resolution?*

Yes, the coarse model resolution limits the maximum values in the simulation. This is added to this sentence.

21) Page 3472, line 26: *At the end of this paragraph, I got the impression that it may be important to compute OPE from MOZART if it is expected to be lower than reality (as a result of resolution). Perhaps, the discussion needs some refinement.*

I don't understand what the reviewer is suggesting.

22) Page 3474, line 23: *Here and elsewhere, the authors suggest that coarse resolution is a reason for some of the uncertainties. I agree that it is certainly a likely factor. But given other higher resolution trace gas simulations that have been performed for MILAGRO, can the authors provide some evidence that the time series (or stats) from their study differ from other higher resolution model studies?*

Not sure this will really add much, so have not done this.

23) Page 3475, line 2: *What is the implication of this statement and why is this relevant.*

This sentence has been expanded to explain: "The age spectra show that the atmospheric composition around Mexico City is generally composed of air with a range of ages, therefore the mean photochemical age may not be an appropriate representation of a given air parcel."

24) Page 3475, line 11: *Similarly to my comment on line 2, what is the implication of OVOCs?*

Sentence added at end of this paragraph: "These results highlight the need for understanding the chemical evolution of pollution outflow to properly assess the impact of megacities such as Mexico City on the regional atmospheric composition."

25) Page 3475: *Somewhere in the conclusions (and perhaps earlier in the text too) there should be some discussion on how the relative contribution of anthropogenic and biomass burning sources from this study should be compared to other studies of fossil vs modern carbon (via C14 measurements). The C14 studies suggest a much higher fraction of modern carbon. The biomass-burning inventory likely misses many small fires and other sources of burning may be neglected by current emission inventories.*

The use of C14 measurements to identify anthro and fire contributions seems to be quite complex. A comment about this has been added referring to Vay et al., 2009 and Aiken et al., 2009.

26) Figs. 1 and 2: *Include how many flights were included in the analysis in the caption.*  
Done.

27) Fig. 3: *Add "(MZ4)" after "MOZART". Change "C130" to "C-130" in the figure labels.*  
Done.

28) Fig. 8: *Change "Dashed line" to "Black dashed line". Include what the white dashed line means.*

Done.

29) Fig 9: *Definition of the red lines and dashed blue lines is not given.*

Added that they are the quartile ranges.

30) Fig. 11: *Add "(MZ4)" after "MOZART". Would it be useful to have a similar plot for those days with strong transport only (e.g. March 19)?*

This is shown in Apel et al. (2010) so a reference to that has been made in the text.