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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Assessing the regional impacts of Mexico City emissions on air quality and chemistry" *by* M. Mena-Carrasco et al.

M. Mena-Carrasco et al.

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Question: I have only two major criticisms that should be addressed before publication, both of which overlap: “discernible impact&”would be a much more citable result if it were quantified, in the text, conclusions, and abstract. Is “discernible” 10%? More/less? ii) The paper has a good clear discussion of the method and uncertainties, but little effort is made to quantify either the uncertainty or the variability of the conclusion. Is the “discernible”; impact always frequently, or occasionally present? How much does it exceed the error?

Answer: We thank the reviewers for their time in providing valuable insight. Indeed, the concept of “discernible impact” is broad, and we should support this statement with quantitative estimations. We add the quantification of “discernible mexico city influence” more explicitly in the abstract, discussion and

conclusions. We also quantify the frequency of distinct outflow patterns vs. more disperse outflow patterns based on the 31 modeled days on the surface and the 3km level, to occur 9 times, i.e. classified as “occasionally present”. We also add discussion on why we chose this case study vs other cases. We also add the phrase “This flight was chosen because while other flights do show similar results in concentration enhancements attributable to the MCMA closer to the city, this was the only day in which strong predicted outflow combined with a sampling strategy to capture the outflow over the Gulf of Mexico.”

Second reviewer

Question: Section 2.1.2: emission inventories and boundary conditions a) There are several sources of emission inventories used to construct the baseline emission inventory for simulating the MILAGRO conditions. i) Were those emissions adjusted to a common year or not?

Answer: The emissions were not adjusted to a common year. The MCMA emissions inventory used was the latest available at the time of the project, and for regions surrounding Mexico City global emissions inventories were used. We did not adjust to a common year, as growth of emissions on a national scale based on EDGAR would largely be marked by growth of emissions from MCMA, possible introducing unnecessary uncertainty in the modeling. Question: ii) Why was BEIS2 used for biogenic emissions? BEIS 2 seems outdated and has been superseded for models such as MEGAN. Please justify.

Answer: BEIS2 was used since model comparison to observations of isoprene in the C-130 was appropriate (R=0.11). Also, while these emissions are uncertain, they do not contribute significantly to VOC reactivity in the region in comparison to MCMA. Question: b) There is no Table with the aggregated emissions for the whole MILAGRO domain, this ought to be included for comparison with other studies. Answer: We have included a table that summarizes the inventory for MCMA used in this study. We do

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not include a summary of total MILAGRO domain emissions since the heterogeneous nature of this merged inventory (which considers different resolution, compounds, sectors, methodology, and base year). Question: c) I would encourage the authors to make their emission inventory available, so that the scientific community may benefit from such info. Answer: The emissions inventory is available freely through UNAM. The addition of emissions surrounding MCMA based on EDGAR and BRAVO is of little value to modelers outside this project, but is freely available upon contacting the University of Iowa. Also our gridded emissions of the Mexico National Emissions Inventory, which was used in another publication in this journal, is and has been freely available online since it was done in 2007.

Question: Section 3. Results and discussion The first paragraph needs editing. It does not convince me that the base model run hasbeen obtained from a systematic model improvement. In fact the model overestimates ozone and CO, and NOx (NO and NO2) are underestimated. Correlation coefficients in Table 1 are OK but they do not tell the whole picture; also NOy is overestimated by the model, which is certainly consistent with the ozone overestimation. Thus there are biases in the emissions of different species, in the boundary conditions of some of them (figure 5) and on photolysis rates (figure 8); these pitfalls have to be taken into account when photochemistry processing is analyzed. As discussed further below, the manuscript needs editing to achieve clarity.

Answer: The correlation coefficients are lower than those obtained while modeling surface sites. Surface sites benefit from being subject to strong diurnal patterns in temperature and PBL heights. Aircraft based modeling performance evaluation is more complex, as meteorological uncertainties grow, and the conditions sampled present strong gradients, both vertical and horizontal. In comparison to studies during ICARTT and TRACE-P modeling performance indeed is better, presenting less mean standardized bias, and higher correlation coefficients, for all species. Longer lived species such as ozone, CO, NOy tend to present better correlation coefficients, and highly reactive

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species such as NOx present lower coefficients in all campaigns. Large improvements are documented from the first run to the latest run. Further improvements would require adjoint analysis, and would be the subject of a completely different publication.

The model is also sensitive to boundary conditions. Previous work during ICARTT shows that in the upper troposphere performance is dependent on these. A specific sensitivity analysis for selecting a model run was carried out using RAQMS and MOZART, and upper tropospheric bias in ozone and CO was improved by solely changing the boundary conditions.

Question: Section 3.2.2. Time series of modeled and observed values along flight track Figure 5 indicates that CO and ozone boundary conditions may be overestimated, at least for that episode of March 19. Was this behavior persistent for the whole March simulation? This BC issue needs to be addressed.

Answer:

We do not consider there is bias due to boundary conditions. Bias in boundary conditions usually manifests in the upper troposphere. Observations were within mid troposphere and were not near the borders of the domain. Bias over the domain would not manifest only on the high values, but would probably have an overall effect. In fact mean modeled ozone is larger than mean observed ozone. Similarly the same applies for NOy. Question: Section 3.2.3 Impact of MCMA emissions on ozone formation and regional photochemistry In the first paragraph figure 6 is introduced and discussed. Both panels depict the same data with different labels and show two clusters of data points: a large one that extends to high NOz values, and a small one that extends up to 7 ppb of NOz, within 100 km of MCMA and that has a distinctive slope higher that the one defined by thelarge cluster; both clusters have similar high values of ozone. Thus I wonder what is the slope value reported in the figure. Is that slope computed for all data points or not? The slope is calculated separating by those observations with at distances less than 100km, and those beyond that. Observations showed similar

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ozone concentrations, but lower NOz values indicate higher efficiency. We did not add the fit to the plot for clarity to the observations.

Question: The second paragraph states that aerosols reduce NO2 photolysis (modeling results) and so they reduce ozone production and concentrations (figure 7). Given that the model overestimates ozone (see Table 1) this also shows that there is a problem with the boundary conditions and emission inventories of ozone precursors.

Answer: While photolysis rates are reduced, our modeled estimation of the impact of aerosol composition and concentration shows that this is maximum 5%, and in an effect that is more long range than local. This does not vary our previous description that ozone is overestimated due to a bias in emissions inventories of NOx on a regional scale (NOx limited). Also, as we have also discussed that the BC problem is constrained to the upper troposphere, and that improvements have been made in obtaining boundary conditions that enhance model performance in the upper troposphere.

Question: This is an appropriate place to critically examine model runs. One way to sort things out is to acknowledge that the baseline scenario needs further improvements, which will be addressed in another publication. And so the claims on first paragraph in Section 3 would be toned down.

Answer: We add “Indeed, it is apparent that there is room for further improvement. However, for the specific analyses illustrated in this paper, model performance was substantially better (R$>$0.8) providing better support for our findings. “ Question: Third paragraph. The authors recognize that photolysis rates are over predicted 10-50 per cent near MCMA. Thus we find another source of model error that needs to be addressed in this section. Answer: Essentially this is because of an underestimation of aerosol concentration and composition in the basin. However it is difficult to evaluate since the nature of the emissions inventory in Mexico City, which does not speciate PM in terms of absorbing or reflecting aerosol. Also there are large uncertainties in the estimation of Aeolian dust, which are incorporated in the model,

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but not specifically adjusted for the region. Also a large portion of Mexico City aerosol is organic in nature, of which an important is secondary. Unfortunately STEM does not consider secondary organic aerosol formation, and models that do incorporate this are still underdevelopment. Even though there are some point bias estimations overall model performance showed a correlation coefficient over 0.8. Question: That third paragraph then goes back to discussing aerosol effects upon photochemistry, and this discussion would better be in another paragraph.

Answer: We reorganized section and edited for clarity.

Question: 4. Conclusions I agree with most of the conclusions presented here, for they are supported by the analyses carried out. I particularly agree with the need of combining model simulations and observations to estimate a megacity air pollution footprint, as reported extensively in this manuscript for MCMA and its regional scale surroundings. However, some caveats about model uncertainties ought to be summarized here.

Answer: We add “While there are inherent uncertainties in models in terms of boundary conditions, emissions inventories, and transport, these have been evaluated and support insight provided by these.” We also add “which captured the majority of observations with little bias and high correlation coefficient to the sentence on the March 19th, 2008, Question:

Detailed comments Abstract. a)Include the period analyzed. b)Please clarify sentence running from lines 17 to 18, write explicitly whether you are referring to observed or modeled quantities. Current syntax is ambiguous to me Answer: We added “modeled” to the sentence.

Question: Introduction. Last paragraph. Has the term air quality footprint been used before? If so please cite accordingly

Answer: The term has only been used loosely in sites related to carbon footprint calculations. It has not been defined in scientific literature

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Question: 3 Results and discussion. First paragraph. Since the paper cited therein is not available to me, I would appreciate a better explanation of the process of adjusting emission inventories, a non-trivial task in modeling studies.

Answer: We change the reference to a doctoral dissertation available online, in addition to the previously cited manuscript.

Question: Page 20294. The sentence running from lines 6 to 7 needs some editing. Answer:Sentence was rewritten Question: Page 20296. The sentence running from lines 18 to 20 needs editing: the NOx levels are high because of the MCMA emissions, not because of aerosol-reduced photolysis rates (this can only be a second order effect)

Answer: We added “which extend NOx lifetime within the city”. Questions: Figure 1. First two columns have the labels swapped. Figure 4. Check caption, a number (5) is mistyped. Answer: Fixed

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 20283, 2008.

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