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Atmospheric Chemistry and Physics Discussions

Interactive comment on "

Formation of semivolatile inorganic aerosols in the mexico city metropolitan area during the milagro campaign" *by* V. A. Karydis et al.

Anonymous Referee #2

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General comments:

In this study, the authors mainly used the measurements on March 2006 during the MILAGRO campaign to evaluate three dimensional CTM PMCAMx and then to use the model to test three different emission strategies (i.e., reducing 50% SO2, NH3 and NOx emissions, respectively). The subject of this study is suitable for interests of ACP. However, it seems the authors spend more paragraphs and figures to repeat evaluation of the CTM following the previous study (Karydis et al. 2010) against another set of

ground-level measurements during the MILAGRO campaign which is not novel while comparably fewer discussions are made on the most interesting and novel part of the current manuscript (i.e., to use the CTM to test three emission strategies is eventual goal of both MILAGRO field campaign and the chemical transport model as stated by the authors). Moreover, the difference of the model setup as well as emission inventories in the current work in contrast to the previous study (Karydis et al., 2010) is not clearly addressed, which brings a question: what's the advantage of this new version of PMCAMx? The abstract does not provides a concise and complete summary while the conclusions sound more like summary. The overall presentation of the manuscript needs to be significantly revised and following specific comments are needed to be clarified.

Specific comments:

1. In the abstract, the authors used lots of numbers for the predicted and measured inorganic aerosol concentrations that are overly loaded with details and are lacking in the important conclusions. For example, at page 21996, line 21-23, the authors listed the predicted and measured sulfate, nitrate, ammonium and chloride concentrations without any comments on the difference of the modeled vs observed values (e.g., the model underestimates nitrate, ammonium and chloride at the T0 site, etc), which makes the abstract tedious and verbose.

2. From the context, the authors evaluated model prediction against measurements at two sites (i.e., T0 and T1). However, only the results at T0 (model vs observation) is concluded in the abstract.

3. The brief conclusion from the sensitivity with respect to the hybrid method versus the equilibrium method is also missing in the abstract.

4. In the section of introduction, the authors used lots of vague statements when referring previous studies, for example, a. page 21998, line 22-23, "a general agreement although some differences were found"; b. page 21998, line 27-28, "an overall

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agreement was reported although some discrepancies were found at Long Beach"; c. page 21999, line 15, "general agreement was found"; d. page 21999, line 27-29, "...similar results in model predictions for total PM...although some differences over species concentrations and RH regimes are reported." The statements like "an overall agreement although some difference" are too general and uninformative when comparing the model predictions to the observed data. General agreement on what? Differences on what? It's unclear. The question is: how different or similar previous model studies are compared to the observations? What are advantages of your model compared to numerous previous model studies since the prediction of the partitioning of the semi-volatile inorganic aerosol components is one of the most challenging tasks? The authors need to clarify that.

5. Page 21998, line 25-27, if "the MILAGRO campaign was designed to follow the urban plume originated in Mexico City" as stated, it might be more interesting to compare the vertical profile from the model prediction of inorganic matter versus the measured ones by taking advantage of both 3D CTM and observed data.

6. Page 21999, line 2-3, "An analysis of model performance against measurements has been performed" is needed to be changed to "An analysis of model performance against measurements with respect to the particulate matter has been performed".

7. Page 22002, line 22-25, the authors stated that "The concentrations of the aerosol components at the boundaries of the domain were chosen based on results of the GISS-II global CTM for the month of March". Do you mean March 2006 or climatology March? I am curious how the model predictions on the inorganic PM are sensitive to these boundary conditions. For example, the predicted PM1 chloride seems sensitive to the south boundary conditions shown in Figure 2(d).

8. This study used meteorology fields outputted from the WRF model. What's the time interval of CTM as well as the meteorology field? Did you use the meteorology fields corresponding to March 2006? The model setup needs to be described in details. It's

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unclear how the model setup as well as the emission inventories used in the current work differs with that in the previous study (Karydis et al., 2010) although the authors stated the current work is based on previous efforts.

9. Page 22001, line 6-8, the authors stated the purpose of this study is to "evaluate our current understanding of the atmospheric processes responsible for the spatial, temporal and seasonal variability of fine inorganic PM over the Mexico City Metropolitan Area". This statement is not appropriate since only observations on March 2006 during the MILAGO campaign were used. I could not find any results related to seasonal variation with respect to either fine or coarse inorganic aerosols.

10. Page 22004, line 3, what's the MCMA 2006 official emission inventory? What's the frequency of the emissions of precursor species emitted into the CTM?

11. Page 22004, line 12-14, the authors stated that "There is also a little ammonium in the coarse mode because the coarse dust particles are alkaline". I do not understand this explanation since the formation of ammonium (i.e., NH4+, cation species, needs to be associated with anion.) favors the acidic condition (e.g., H2SO4, HNO3, or HCl). The authors further explained "The soluble crustal elements increase the PM water content and thus favor the ammonium nitrate formation". This is also confusing statement without supportive justification from neither the context nor figures. Actually the presence of crustal elements (Ca2+, Na+, K+, Mg2+) may compete with NH4+ for avaible HNO3 gas. What's the corresponding relative humidity near the dust region (Texcoco lake, where is it? Could you mark it in Figure 1)? How's aerosol water content predicted by the model?

12. In the section 5, the authors compared the inorganic particulate matter concentrations (i.e., sulfate, nitrate, ammonium, chloride and dust components) between model predictions and observations. In the entire section, the authors listed a lot of average values with respect to the inorganic PM when comparing to the observations. Comparing the mean values only is not very helpful to understand the discrepancy between the

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model predictions and observations. The inclusion of uncertainty analysis (e.g., standard deviation, relative difference, absolute difference, root mean square, etc) might be more informative than the mean value from the statistic point of view. It's easier for readers to follow with a neat table.

13. In Figure 4, the authors used PM2.5. What's your definition of PM2.5 here? How is it related to or deduced from PM1-10 in the model shown in Figure 3? Why do you use the PM1 for T0 site but use the PM2.5 for T1 site? This issue needs to be clarified.

14. Page 22005, line 21-29, the authors ascribed to the spike on March 18th predicted by the model not coincident with the observations to the same emission for SO2 every day as well as errors in meteorology. The authors did not explain the discrepancy of the sulfate concentration between the model predictions and observations occurred for the first 3 days at T0 as well as the spikes on March 14th at both T0 and T1. Clearly, the model predicted sulfate is much larger than the observed ones for the first 3 days at T0 site.

15. Page 22006, the authors tried to use the underestimated OH during the early morning to explain the underprediction of nitrate at T0 and T1. What about the temperature and relative humidity at T0 and T1 during this period of time? Note that the partitioning of semi-volatile species is also highly sensitive to these two parameters. Actually Figure 5d suggests that the underprediction of nitrate from the model occurred in the afternoon at T1 site while the overprediction happened in the early morning, which results in the slight overestimation from the model vs the observation if comparing the average values at T1 site. Do you have justification for this discrepancy in the afternoon that is different with what happened at T0 site?

16. Page 22007, the overestimation of dust components as stated by the authors may partially explain the overestimation of PM2.5 nitrate at T1 site shown in Figure 5c.

17. The authors conducted sensitivity test by comparing hybrid approach (HYB) versus equilibrium (EQ) approach. Which method (i.e., HYB vs EQ) is more close to the

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observations during the MILARGO campaign (e.g., at T0 and T1 site)?

18. In the section 7, the authors conducted the sensitivity test by arbitrarily reducing half of SO2, NH3 and NOx emissions to see the change of inorganic PM. What about the situation if there is 50% increase of SO2, NH3 and NOx emissions for the case we do not have emission control in futrue? What do you expect if there is 50% reduction of SO2 with 50% increase of NOx for the case we only control the sulfur emission? Although the authors claimed that these sensitivity tests "do not correspond to actual emission control strategies", more discussion about the indication from these sensitivity tests might be more insightful for the design of future emission control strategies since "a major component of the MILAGRO campaign was the use of the observed data to evaluate the performance of three dimensional chemical transport models and then used them for the design of emission control strategies" as stated by the authors in page 21998 line 3-6. I think this is also the major motivation of the current study.

19. In the section of conclusion, the conclusion by comparing the hybrid method versus the equilibrium method is missing. The section of "conclusions" needs to be revised by including more discussions of your findings replacing the summarized list given in the present manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 21995, 2011.