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Comment

Interactive comment on “Megacity impacts on regional ozone formation: observations and WRF-Chem modeling for the MIRAGE-Shanghai field campaign” by X. Tie et al.

Anonymous Referee #1

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In this manuscript, Tie et al. provides an overview of the field experiment conducted in the Shanghai megacity during September 2009. Ozone and related trace gas as well as PM_{2.5} were measured at several ground sites during the campaign, and the data obtained from the two sites (Pudong (PD) and Dongtan (DT)) are used for this study. A regional chemical/dynamical model, WRF-Chem, is also applied to simulate the distributions and variations of ozone and related trace gases in the Shanghai region, and results are compared with measurements at the two sites. Model sensitivity tests are performed to evaluate the effect of emission ratio of NO_x/VOC on ozone formation in the region. The manuscript addresses the scientific questions that are within the scope of ACP and it is certainly well suitable for this special issue. The measurement items

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in this study are relatively complete (though short-lived radicals are missing), providing a valuable dataset for the first time for the study of ozone chemistry in the Shanghai megacity. My major concerns and questions arise from the model analysis of this work. I think the manuscript should/could be improved by increasing the links between the more practical or sensible source emissions and model simulations as well as between the measurement data and model analysis.

Specific comments

1) The source emission distributions of primary pollutants in the Shanghai region are not clearly presented in the manuscript. I would suggest that the authors provide an additional figure showing the regional emission distributions of primary pollutants (NO_x, VOCs, CO) and perhaps biogenic VOCs in the inner domain (right panel of Fig.1). What emission inventory used for the model simulations of this study is not clearly stated in the manuscript. The authors only refer to their previous work of Tie et al. (2009b) (should be Tie et al. (2009a)), where the TRACE-P inventory of gaseous pollutants developed by Streets et al. (2003) for the year 2000 had been applied. The question is: are there any great changes in the source emissions from 2000 to 2009 for the Shanghai region? Note that an emission inventory for the Yangtze Delta region has been developed recently (Huang, et al. ACP, 4105-4120, 2011).

2) Where is the center point of urban Shanghai in the right panel of Fig.1, BS, XJH, PD or other place? It may not sound to describe the cost site DT as “remote” site. How many distances are the PD and DT sites from the city center? It would be nice if a km scale is given in the right panel of Fig.1. While the main focus of this study is the megacity impacts on the regional ozone formation, no observations at a suburban or rural site downwind of Shanghai were performed or presented. As demonstrated in Fig.13, maximum ozone is simulated to occur at about a distance of about 100 km from the city center. Could the measurement dataset of this study be applied to access such model-simulated plume evolution process?

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3) There have been numerous studies on regional ozone in the Yangtze Delta region including both measurements and model simulations, e.g. the results from the CHINA-MAP project. These previous studies should have been recognized and reviewed in the manuscript. Certainly, the authors might argue that in contrast to previous studies, their focus is on the Shanghai megacity where their super sites are located. If so, I would suggest that “urban” (instead of “regional”) ozone formation be emphasized in the title as well as the body of the text.

4) While most species are generally well simulated with comparison to the mean values of measurements, the variability of some important species such as O₃ is not well predicted. Scatter plots of measured and simulated mixing ratios of important species would be helpful to see the performance of the model and at least they can be given in the Supplement. Scatter plots showing the correlations of some species with CO (Figs.8-10) are not necessary to appear in the formal manuscript since no much more information can be learned by this investigation than from emission inventories.

5) In this study, the observational data are used merely for evaluating the model performance with respect to the simulation of the species' mixing ratios. In-depth analysis with measurement data can be done to look at the chemical character of the city plume as revealed by observations. Otherwise, it may not be entirely sufficient to be a campaign overview paper.

6) The mean value of simulated HNO₂ is nearly an order of magnitude smaller than its measured mean value over the experimental period. The effects of such HNO₂ biases on simulated ozone should be quantified. How about the diurnal cycles of simulated HNO₂ and other reactive nitrogen species with comparison to the measurements?

7) How was PM_{2.5} sampled and analyzed for this study? While the PM_{2.5} concentrations are simulated, does the model include the effect of aerosols on ozone chemistry? Note that measured PM_{2.5} could reach as high as 100 $\mu\text{g m}^{-3}$ at the PD site, which might have considerable impacts on ozone formation through heterogeneous reactions

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and photolysis rate changes. There are few analysis and discussion about the simulation of aerosol formation in the manuscript. If no feedback on ozone chemistry from aerosols is included in the model, the authors may consider skipping over the parts about PM_{2.5} (both measurement and modeling results) in the manuscript.

8) The authors use the ratios of CH₂O/NO_y to identify whether the ozone formation is under the VOC-limited or NO_x-limited conditions in the Shanghai region. Based on what chemical principle or for environmental conditions did Sillman (1995) propose this method? Can the CH₂O/NO_y ratio threshold value (0.28) derived by Sillman (1995) be applied to the Shanghai region since the chemical character of Shanghai can be quite different? The authors also give a discussion about the effect of the concentration and emission ratios of NO_x/VOC on ozone formation in Shanghai compared to Mexico City. Are there any quantitative connections of these NO_x/VOC ratios to the concentration ratios of CH₂O/NO_y for the Shanghai region? CH₂O is an oxidation product of VOCs, and NO_y are the oxidation products of NO_x or both NO_x and VOCs (e.g. PAN). Can the NO_x/VOC ratios be used to evaluate the ozone formation regime since CH₂O was not measured in this study? Nevertheless, an in-depth analysis should have been performed to address these issues.

9) The authors performed 4 different model runs to study the effect of the NO_x/VOC emission ratio on regional ozone formation in the Shanghai region. It is not clear how they changed the NO_x/VOC emission ratios for model simulations (maybe I missed something in the manuscript). Are the NO_x emission changed with a constant VOC emission, or the VOC emission changed with a constant NO_x emission? It is quite difficult to understand that Run-R3 (also Run-R4) can represent either a reducing NO_x emission case or an increasing VOC emission case unless two simulations were performed for Run-R3. Are the emissions in the entire model domain changed? Are the biogenic VOC emissions changed? How about the situations if NO_x and/or VOC emissions are reduced, e.g. by 50%? Nevertheless, I do not think such simple model sensitivity tests without practical emission scenarios could provide highly valuable in-

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formation for the understanding of ozone formation in this region, not mention to the planning of ozone control strategies.

10) In the Summary section, the conclusions based on wind directions in Highlights 1 and 4 are limited to the case of this study (summertime in Shanghai). These results are too local to be referred by other studies. Highlight 3 may not be necessary to be a conclusion, as mentioned above. There is neither measurement nor model simulation to support the conclusion about the contribution of biogenic isoprene emissions to ozone formation as stated in Highlight 5. Of course, Fig.16 provides some highlights, but it would be more convincing if a model sensitivity study had been performed.

Technical corrections

1) Abstract and Introduction: The full name of an abbreviation should be given when it is firstly used, e.g. NO_x and VOC.

2) P1676, L21: Tie et al. (2009) should be Tie et al. (2009a)?

3) P1677, L29: participants?

4) P1678, L22: the Institute of Chinese Meteorological Science? Not found in the author's affiliations.

5) Fig.1: There are three large red points, instead of two, all of them referring to the super sites? Where are the white points? A description of observational stations in the panel is needed. A distance scale in km should be given.

6) Fig.2: This plot may be not necessary. Otherwise, the distributions of major pollutants (CO, NO_x, NO_y and O₃) as well as RH at the two sites (PD and DT) can be shown and discussed in the text.

7) P1680, L21 and L 24: should be Tie et al. (2009a).

8) P1682, L6, “polluted period” and “clean” period”: it may not be suitable to saying so, merely according to the observations at one site.

9) P1683, L28–P1684, L1, “It shows that.....during the experiment”. What do you mean? One should know if PAN and MPAN are measured by the instrument performance itself.

10) P1684, L1-4: The NO_x/NO_y ratios of 0.92 and 0.99 are mean values. How about the variations and the minima?

11) Fig. 11: it would be better to show both measured and calculated ages for PD with one panel and calculated ages for DT with another panel. There appear larger differences between measured and calculated ages at PD, indicating that the plume feature is not well captured by the model.

12) P1691, L20: should be Tie et al. (2009a).

13) P1691 and Fig. 16: What is OH reactivity and how it was calculated? The unit should be given in the figure. Note that OH reactivity is different from the ozone formation potential.

14) P1692, L12, “In addition to.....”: should be rewritten.

15) Figs. 12, 14-17, the same model domain should have been used, the latitudes and longitudes should have been labeled, and the PD and DT sites marked out.

16) It might not be necessary to put all the original version of figures into the Supplement.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 1673, 2013.

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