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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Mechanisms controlling surface ozone over East Asia: a multiscale study coupling regional and global chemical transport models" *by* M. Lin et al.

M. Lin et al.

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Response to Referee 1

We thank the reviewer for the thoughtful comments on the manuscript.

General Comments:

This paper addresses the application of CMAQ to simulate ozone in East Asia. The paper title suggests that the focus is on the mechanisms controlling surface ozone in East Asia. However, as written the paper is focused on the sensitivity of CMAQ predictions to the to various model settings. These sensitivities provide valuable information about ozone prediction in East Asia, but the results as presented and discussed do not

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provide insights or new information about mechanisms of ozone production. I suggest that the paper title reflect the prediction aspects. With such a focus I think it is a good contribution. If the focus is to remain on mechanisms, then I feel much more work is needed before publications.

In this paper the CMAQ model is driven by MM5 and with boundary conditions from the global model MOZART. This is a contemporary model set-up, and there are not many results presented yet for East Asia. The sensitivities studied include: model resolution; chemical module, and treatment of boundary conditions. These are important tissues, as regional scale air quality modeling of ozone is of growing importance in East Asia, and many groups are beginning to build programs. They are making decisions about what models, what resolution and what research needs to be conducted to improve model performance for East Asia applications. The sensitivity results presented will be of interest to many and helpful in designing research plans.

The results presented are well documented and discussed. They include many of the most important components. Another important component not discussed in sufficient detail is dry deposition. There have been some studies of this in the East Asia context in the MICS-Asia papers.

Response to General Comments:

We appreciate the reviewer for recognizing the important contributions of this paper. We agree with the reviewer that the initial paper title did not reflect the prediction aspects of the results as presented and discussed in the manuscript. Therefore, we changed the paper title to "Multi-scale model analysis of boundary layer ozone over East Asia".

We haven't done any sensitivity tests yet with different parameterizations of dry deposition processes. However, we have added some discussion on how the dry deposition processes can affect the diurnal variation of ground-level ozone at a rural site near Nagoya, and at a mountain site in central Japan. Impact of

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boundary layer height on dry deposition is also described. We referenced the MICS-Asia paper and discussed the uncertainties in estimating dry deposition velocities in regional model studies for Asia.

"Dry deposition flux of ozone differs considerably between MM5-CMAQ and WRF-CMAQ (Fig. 14). Higher PBL height reflects stronger turbulence and thus might increases dry deposition velocity. For example, the YSU scheme in WRF predicts slightly higher PBL height at nighttime on July 25–26 than the MRF scheme in MM5, which in part contributes to the increasing dry deposition flux of ozone. Wang et al. [2008] suggested that dry deposition velocity is one of major discrepancies in model simulations for Asia. The parameterization schemes used in the current models have been developed and tested using experimental data obtained in North America and Europe, while the deposition processes depend largely on local conditions. Measurements of dry deposition velocities for major air pollutants in Asia would be extremely valuable in the future to improve the parameterizations in model application for Asia. It would be also valuable to use dynamic land use data from satellite measurements instead of static data."

"On July 7 and 10, highest observed ozone mixing ratios are found near midnight at mountain site Happo, and are not reproduced by the model. Such behavior is characteristics of mountain stations located above the nocturnal inversion, which isolates the site from the effects of surface deposition. The 27-km horizontal scale used in this study is still too coarse to represent the mountain topography. The model due to unrealistic removal of ozone through dry deposition generally underestimates nighttime. It would be valuable in the future to examine if high spatial (1km) resolution model can reproduce the ozone diurnal behavior at high altitude sites."

Specific Comments:

1) Focus on 2001 and comparison with 2001 data is good. The inclusion of data from

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Interactive Comment

other years is generally not relevant and does not add much to the discussion.

Response to Specific Comments 1:

The purpose of comparing model results with observations in other years is to provide general insights into how the model can capture the observed seasonal cycle of surface ozone that does not change significantly over years. We have made great effort to obtain 2001 ozone measurements at a few sites such as Lin'An and Mondy. In response to editor comments, we have compared ozone measurements in 2001 and the average values during 2001-2007 at the Japanese sites and at the Mondy site in Siberia to examine the influence of inter-annual variability. The comparison suggests that the general pattern of ozone seasonal cycle at the Japanese sites did not change in 2001. Our analysis suggests that the slightly different transport pattern found in August 2001 might contribute to ozone enhancement over central east China, but the impacts on ozone is likely lower than 5-10 ppb.

2) The discussion around the chemical mechanism results is generally good, and includes the issue related to the emissions inventory. The discussion also includes PAN. The generalization to CBiv is better for this problem (regional ozone) and SAPRAC for another, is not defendable. We need to know that there are differences, but we ultimately need to know which one is right or wrong for the right reasons. Since the simulation is 2001, they can compare their results to TRACE-P aircraft data, which should provide some insights into how the predictions of PAN and other relevant species (OH, HO_2, formaldehyde) compare.

Response to Specific Comments 2:

We agree with the reviewer that it is not defendable to state that CB4 is better for regional ozone and SAPRC99 for local ozone. We have specifically followed the reviewer's suggestion to compare O_3, OH, H_2O_2 and PAN with the TRACE-P data (See Figures 10, 11 and 12). The discussion about the chemical mechanisms has

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thus been rewritten in the revised manuscript. The concluding remarks are:

Central eastern China is found to be the most sensitive region to the choices of chemical mechanisms. Central eastern China appears to be the most sensitive region in our study to the choice of chemical mechanisms. Evaluation with TRACE-P aircraft measurements reveals that neither the CB4 nor the SAPRC99 mechanisms consistently capture observed behavior of key photochemical oxidants in springtime. However, our analysis finds that SAPRC99 performs somewhat better in simulating mixing ratios of H_2O_2 and PAN at flight altitudes below 1km. The CB4 mechanism overpredicts H_2O_2 by a factor of two, which is caused by higher self-reaction rates of HO_2. SAPRC99 predicts 50% of higher PAN because more PAN products and lower decomposition rate of PAN are considered in SAPRC99 than in CB4. The overall higher load of VOC, explicit organic peroxy radicals, and inclusion of radical recycling processes explain why SAPRC99 predicts 10-20 ppb higher O_3 during summertime for central eastern China and downwind areas. Our findings suggest that future model studies over this region should carefully examine chemistry sensitivities, especially in regulatory applications such as designing of emission control policies. Between CB4 and SAPRC99, the more detailed SAPRC99 mechanism is suggested for future studies examining high O_3 episodes, the impacts of large releases of reactants, and multi-day effects. But results need to be interpreted with care.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 20239, 2008.