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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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

General Comments: This paper describes regional modeling studies of surface ozone over East Asia and focuses on how boundary conditions, chemistry schemes and model resolution influence comparisons with surface measurements. The study is competent and the results are useful as they highlight weaknesses in our current understanding of tropospheric photochemistry and in our ability to reproduce observed oxidant concentrations. A number of other papers have focused on ozone formation over East Asia (and these papers are acknowledged appropriately in the text), but this study provides further analysis and is therefore a useful addition to the literature. The focus on diurnal variability is one unique aspect of this study that provides a clearer assessment of the reasons for discrepancies between model results and observations.

Although valuable as a whole, the paper does not provide much additional new insight (in a quantitative sense) into how different processes control ozone over Asia, and I believe that this is a missed opportunity. The paper is strong on description, but weak on more detailed analysis and the conclusions are therefore less valuable than they could be. What do we learn from the differences between the chemical mechanisms? How important (quantitatively) is the Asian monsoon for suppressing surface ozone in summertime? How might PBL mixing be improved, and would this lead to closer agreement with observations? Additional analysis of any one of these aspects, along with some tightening of the abstract and conclusions, would strengthen the paper considerably. On balance, I believe that the paper is suitable for publication in ACP, but that it would benefit greatly from some revision, and I provide a number of suggestions for this below. In particular, the abstract and conclusions are vague (e.g., "complex interactions" are referred to but not identified) and these need to be rewritten to sharpen them up and to make it clear what the main contributions of this study are.

Response to General Comments: We have clarified in the revised manuscript that "The purpose of this paper is to provide a first step in evaluating the response of model simulations of local and regional ozone in Asia to the choice of chemical mechanisms, meteorological inputs, boundary conditions and model resolutions. From this analysis, we provide a detailed assessment of the reasons for discrepancies between models results and observations, and make recommendations regarding optical regional-scale CTM configurations for future studies of East Asian ozone production";

In response to Reviewer 1, we have changed the paper title to "Multi-scale model analysis of boundary layer ozone over East Asia"

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The Abstract and Conclusion has been rewritten to reflect the following detailed analysis added to the revised manuscript:

1) Evaluation with the TRACE-P measurements, which provides additional insights into how the predictions of O_3, PAN, OH, and H_2O_2 from the chemical mechanisms compare.

2) Evaluation of precipitation with the TRMM satellite measurements, which provides some insights on how the prediction of EA monsoon rainfall affects O_3 predictions.

3) A new alternative simulation of CMAQ driven by WRF and with higher vertical resolution. Through comparing the MM5-CMAQ and WRF-CMAQ predictions of ozone and NOx, we added the discussion about the impacts of PBL height on daytime ozone promotion and nighttime ozone destruction by NOx titration as well as dry deposition.

Specific Comments 1 Evaluation of CMAQ performance is covered very well in the paper, but it is not entirely clear how the results may be of use to others. The comparison of CBIV and SAPRC99 remains inconclusive; although there are sometimes large differences between the results, it is not clear why the more simplified CBIV scheme is "better". More concrete conclusions on this are required here.

Response to Specific Comments 1: We agree with the reviewer that the statement that "CB4 is better for regional ozone" is not defendable. Therefore, we have removed this statement and followed the suggestion by Review 1 to compare with TRACE-P measurements. The discussion about the chemical mechanisms has thus been rewritten in the revised manuscript. The overall conclusions are summarized below:

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 sensi-

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tive 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 HO2. SAPRC99 predicts 50

Specific Comments 2

The focus on diurnal variability would benefit from more quantitative analysis, so that future studies of these variations could be more clearly targeted. Sensitivity studies reducing the nighttime boundary layer mixing height would be particularly valuable here, as they would allow the contributions of mixing depth, titration and deposition to be more clearly distinguished.

Response to Specific Comments 2: In response to the reviewer's comments, we have rewritten the section on diurnal variations. The statistics of root mean square error, coefficient, and mean bias are calculated for the hourly mixing ratios of ozone (Table 3). The contributions of mixing depth, titration and deposition are analyzed in detail. A sensitivity test of WRF-CMAQ with the depth of first model layer setting to 17 m, in contrast to 73 m in earlier simulations, is conducted. The YSU boundary layer scheme from WRF predicts lower nighttime PBL height on some hours than the MRF scheme from MM5, which provides an opportunity to examine the impacts of reduced nighttime PBL height on NOx titration and deposition processes. Figure 14 in the revised manuscript shows that when nighttime PBL height substantially decreases lower than 250m, NOx increases can reach up to 20 ppb, which causes the rapid depletion of ozone at the ground level. The impacts of daytime PBL heights on ozone production are also discussed. For example, during July 26-27 the lower PBL height shown by WRF, as well as the weaker winds promotes up to 40 ppb of daytime O_3 mixing

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ratios.

20247 l.8: It is clear that adjusting the boundary conditions improves model agreement in a consistent way. It would be helpful to suggest a reason for this overprediction in MOZART.

A reason for the overprediction MOZART has been suggested in the revised manuscript. "The summertime overprediction is likely due to coarse horizontal scale implemented by MOZART, which has limited ability in simulating cloud activities and thus enhances ozone production on clear sky."

20248 l.11: The distributions in Fig 4 are not very informative as they are geographically very similar. It would be more helpful to quantify the burden of O_3 and PAN over the region, and identify whether these differences arise because of the differing production of each species or differing lifetimes. You've started to do this on the next page (20249 l.12), but further detail would provide more insight into the differences between the chemical mechanisms. The differences between the schemes here is worrying: does SAPRC99 overestimate the PAN yield, or does CBIV underestimate it? If you could identify which aspect of the schemes lead to the difference it would go a long way towards resolving the problem.

We have changed the correspondent figure to show the absolute differences of ozone and PAN between these two mechanisms (see Figure 9 in the revised manuscript). The discussion about the impacts of chemical schemes is rewritten (see section 4 for details). Major conclusions are summarized in the "Response to Specific Comment 1"

20249 l.8: I assume that CBIV has a treatment of isoprene oxidation, and if so then the results should be equally sensitive to the estimated emissions? I believe that isoprene treatment has been upgraded in the more recent CB05 mechanism, might this explain some of the differences?

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CBIV does have a treatment of isoprene oxidation, but the isoprene chemistry in CB4 is much coarser than that in SAPRC99. Another major difference between CB4 and SAPRC99 in the treatment of biogenic emissions is that SAPRC99 includes both isoprene and terpene but CB4 includes isoprene only.

20249 l.17: Glatthor reference: it would be more appropriate to cite an earlier paper for this finding, e.g., Moxim et al. [1996, JGR101, p12621]

Changed

20251 l.16: Cloud activity and convective mixing are referred to here without any explanation. How might they contribute to the bias? Monsoon flow (and perhaps biases in photochemistry) is more likely explanations for the summertime overprediction.

Impacts of cloudiness associated with the monsoon rainfall on ozone production are explained in the revised manuscript. Clouds have significant radiative effects on the photochemistry of O_3 production near surface [Lefer et al., 2003; Liu et al., 2006]. Transmission of solar radiation is reduced below cloud level, thus photochemical production of O_3 is suppressed in the presence of increased cloudiness associated with the monsoon rainfall.

20251 l.25: The Wang et al. 2006 measurements focus on outflow from Beijing and direct comparison with MOZAIC data is therefore not appropriate as they are representative of different flow regimes and regions (as outlined in Ding et al. 2008). The following discussion of meteorological differences in August is more relevant.

We agree with the reviewer that it is not appropriate to compare with the Wang et al. (2006) measurements. Thus the correspondent discussion and comparison have been removed from the revised manuscript.

20253 l.16: The difference in nighttime ozone is also affected by the intensity of NOx emissions (direct removal of ozone) and by deposition processes. These are both influenced by PBL height and mixing, but should be acknowledged here as contributing to the nighttime differences.

The influence of PBL height and mixing on nighttime ozone and deposition has been added to the manuscript. See Response to Specific Comments 2.

20254: The effects of agricultural burning in June are described in more detail in Fu et al. [2007, JGR, D06312], and citation of this paper might replace some of the discussion here.

Fu et al. (2007) was referenced in the revised manuscript. We also added discussion about possible underestimates of VOC emissions for China and its impact on ozone prediction.

20255 l.19: It would be useful to make a more quantitative assessment of the effects of resolution here. How much does the RMSE depend on resolution?

The statistics of root mean square error, coefficient, and mean bias are calculated for the hourly mixing ratios of ozone (Table 3). The 27-km simulation gives better statistical scores at most sites than the 81-km simulation. The RMSE of ozone by the 27-km prediction decreases up to 12 ppb at some sites, for instances, Sado-seki and Oki.

20258 l.11: What mixing height would be required to simulate the ozone measurements correctly? This information would be useful to help identify the errors in the current PBL treatment in CMAQ.

See response to Specific Comments 2

20269, Fig 2a: Note in the caption that these figures are for 81 km simulations, and that the adjustments involve reduction in the boundary conditions (i.e., are actually negative).

Done

Minor Points and Typos 20244 l.4: Fig 2a -> Fig 1

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Corrected

20244 l.21: mostly -> almost the (or equivalent)

Corrected

20245 l.11: 2008b -> 2008a

20248 l.1: remove "while"

Corrected

20251 l.9: decreasing trend of -> decrease in

Corrected

20256 l.28: left and right are reversed here.

Corrected

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

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