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Atmospheric Chemistry and Physics Discussions

ACPD

8, S11893–S11901, 2009

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 Dr. Owen Cooper for the thoughtful and detailed comments on how the manuscript can be improved.

Major Comments 1: The authors need to make a clear statement about the purpose of the paper and what they hope to achieve. In the Introduction the authors state that "the response of local and regional ozone in Asia to choice and implementation of chemical mechanisms has not yet been evaluated ". This appears to be the main motivation of the paper but it needs to be more prominent. Also, in the Conclusions, the authors need to provide some recommendations for future model simulations and configurations.

Response to Major Comments 1:

In response to editor comments, the following statements about major contributions of this study have been added to the Introduction:

"However, the response of local and regional ozone in Asia to choice and implementation of chemical mechanisms has not been evaluated yet. Several studies have suggested the significant impacts of East Asia monsoon on ozone production over eastern China [He et al., 2008; Wang et al., 2008], but the evaluation on how well the meteorological models can reproduce the monsoon rainfall and its implication on ozone prediction is insufficient. "

This study employs the regional Community Multiscale Air Quality (CMAQ v 4.5.1) model [Byun and Ching, 1999; Byun and Schere, 2006] to examine seasonal and diurnal variations of boundary layer ozone over East Asia. The CMAQ model is driven by MM5 or WRF meteorological inputs and with boundary conditions from the global Model for Ozone and Related Tracers (MOZART v2.4) [Horowitz et al., 2003]. This is a contemporary model set-up and there are not many results presented yet for East Asia. 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 model results and observations, and make recommendations regarding optical regional-scale model configurations for future studies of East Asian ozone prediction."

Some recommendations are made in the Conclusion:

"Prediction of summertime ozone over central east China is found to be most sensitive to the chemical mechanism applied in CMAQ. Our findings suggest that future studies for this region should carefully examine the chemistry sensitivities especially in regulatory applications such as designing of emission control policies. Between CB4 and SAPRC99, the detailed SAPRC99 mechanism is sug-

ACPD

8, S11893–S11901, 2009

Interactive Comment

gested for future studies examining high ozone episodes, the impacts of large releases of reactants, and multi-day effects.

-The short-term simulation of WRF generally reproduces both the magnitude and spatial distribution of East Asian monsoon rainfall, which greatly improves the CMAQ prediction of ozone at mid-latitudes. Reinitializing meteorological models with high-quality reanalysis data every five to ten days is recommended to maximize the consistency with observed weather patterns.

-This study demonstrates clear benefits of using a high-resolution model in evaluating transport and chemical transformation of Asian pollutants. Boundary layer ozone over rural and urban areas exhibits a large vertical gradient reflecting the impacts of ground-level emissions and land-surface interactions. Depth of first model on the order of a few meters to twenty meters is recommended for future studies for Asia to better resolve surface inversion and land-surface interactions.

-We demonstrated that a simple measurement correction of boundary condition from the MOZART global model systematically improves the CMAQ performance near the domain boundaries. Correcting zonal flux using vertical profiles from ozonesonde stations worldwide is recommended for future studies. Regional modelers should choose a global chemistry model driven by assimilated meteorology. In the future, we would benefit optimal global chemistry model results with the assimilation of large-scale satellite observations of chemical species. Future development in regional models needs to strengthen the coupling between global and regional models, in particular, the treatment of upper boundary conditions."

Major Comments 2: I am concerned by the fact that the model is run for the year 2001, but only 12 of the 22 surface stations discussed in this study have data from 2001. I don't think the authors have done enough to address the influence of inter-annual variability when comparing a simulation of 2001 ozone to measurements from other years. The authors did point out that the transport patterns were slightly different in 2001 compared to the climatology, but what about the ozone values? How different was ozone in 2001 compared to other years? This comparison needs to made for the sites that have data from 2001 and other years, such as Beijing, Mondy, LinAn and Mt. Waliguan, as well as for several of the Japanese sites.

Response to Major Comments 2:

We have made great effort to compare model results with available observations in China to provide general insights into how the model can capture the seasonal cycle of surface ozone in Asia. Following the Editor's suggestion, we have obtained 2001 ozone measurements at LinAn from Dr. Tao Wang and at Mondy from Dr. Pakpong Pochanart. Only three months data is available in 2001 for LinAn. Although Ding et al. (2008) present a climatologically mean of Beijing ozone measurements during 1995-2005, most data were collected during 1997-1998 and around 2005. There are very limited (less than five) instant sample profiles in 2001 (Ding et al. 2008). Thus we still used the averaged values during 1995-2005 for Beijing.

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. The slightly different transport pattern found for August 2001 might contribute to ozone enhancements over central east China, but the impacts on ozone is likely lower than might 5-10 ppb.

In response to Reviewer 1, we have added TRACE-P measurements conducted in March 2001 to evaluate the predictions of ozone, PAN, OH, and H_2O_2 from the CB4 and SAPRC99 chemical mechanisms. From the analysis, we identified

ACPD

8, S11893–S11901, 2009

Interactive Comment

differences in these two mechanisms contributing to model discrepancies and made recommendations for future improvement.

Major Comments 3: A recent paper that needs to be referenced and compared to is Wang et al. [2008]. This paper describes the summertime ozone variation at Miyun, a rural site on the northern edge of the North China Plains and downwind of Beijing. These authors describe the influence of the summer monsoon on the decrease of ozone mixing ratios in July, even though CO mixing ratios increase. This paper is especially relevant to your discussion of ozone overproduction during summer on page 20251. Y. Wang, M. B. McElroy, J. W. Munger, J. Hao, H. Ma, C. P. Nielsen, and Y. Chen, Variations of O_3 and CO in summertime at a rural site near Beijing, Atmos. Chem. Phys., 8, 6355-6363, 2008

Response to Major Comments 3:

Wang Y. et al (2008) has been referenced and discussed in the revised manuscript.

Major Comments 4: Please elaborate on the vertical collapsing to 8 layers. Does this mean that CMAQ only calculates transport and chemistry on 8 levels? How many levels are in the lowest 2 km? Wouldn't coarse vertical resolution explain why O_3 is over predicted in Beijing at night because you are not developing a strong surface inversion? Similarly in Section 4.2.1 you discuss the effect of model resolution on boundary layer depth. You state that the finer scale MM5 run has a better resolved nocturnal boundary layer depth, but I don't see why this would matter if the MM5 data for both the 27 km and 81 km runs is degraded to only 8 levels. Shouldn't both CMAQ runs have nocturnal boundary layers that are equally resolved by this coarse vertical resolution of only 8 levels?

Response to Major Comments 4:

The initial purpose of vertical collapsing to 8 layers is to reduce the comput-

ing cost for annual simulations. Depth of first CMAQ layer is set to 75 m and there are 5 layers in the lowest 2 km to simulate transport and chemistry in the boundary layer. We agree with the editor that strong surface inversion might not be adequately developed in CMAQ with such a coarse vertical resolution, thus we have done a new CMAQ simulation driven by WRF with 29 vertical layers and depth of first model layer setting to 17 m. The new WRF-CMAQ simulation conducted for March and July is compared with the initial MM5-CMAQ simulation, and discussion about the impact of nocturnal boundary layer on nighttime ozone destruction is added to the revised manuscript. The results show that the new WRF-CMAQ simulation significantly improves nighttime ozone prediction at Ijira, a rural site near the Nagoya city in central Japan. In response to Reviewer 2, we have added the discussion about the impacts of boundary layer height on daytime ozone build-up and nighttime ozone destruction by NO_x titration and deposition processes.

For Beijing, the discussion is for daytime ozone with little effects from surface inversion. In light of the significant impacts of East Asian monsoon on ozone seasonality over this region, we have evaluated both MM5 and WRF simulated precipitation with the TRMM satellite measurements. The evaluation suggests that the overprediction of daytime ozone at Beijing in July and August relates primarily to the underprediction of the monsoon rainfall and associated cloud activities over Beijing.

Major Comments 5: The standard of English is quite good, but there are many instances of grammatical errors or awkward phrasing that need to be corrected. Please proof read the paper for these types of errors.

Response to Major Comments 5:

We have proof read the revised the manuscript and corrected grammatical errors.

Minor comments:

ACPD

8, S11893–S11901, 2009

Interactive Comment

Abstract, line 7 change inflows to inflow

Done

Abstract, line8 shouldn't be over the northern domain?

Corrected

Page 20244 line 14 What do you mean by physical parameterization? Do you mean parameterization of deep convection, and/or shallow convection?

The correspondent sentence was removed. The physical options applied for MM5 and WRF simulations are clarified with a Table (Table 1 in the revised manuscript).

Page 20250, line 6 change response to respond

Done

Page 20251 lines 22-24 You note that ozone at a site north of Beijing in 2005 is greater than ozone in Beijing over 1995-2005, and imply this is due to inter-annual variability. But couldn't this just be spatial variability, with the site north of Beijing sampling the aged urban plume that has more time to produce ozone?

We agreed with the editor that this might be due to spatial variability, thus the discussion about this site has been removed.

Page 20252, line 4-5 I don't follow why you need to state that the 2004 data are not shown, when the model was not run for 2004.

The reason we mentioned the 2004 data is that measurements at three mountain top sites were for 2004. Nevertheless, the correspondent sentence has been removed from the revised manuscript.

Page 20252 line 21-24 Here you state that the typical flow pattern of air to central China during the monsoon is from the southwest, but to me it looks like it's from the south or

ACPD

8, S11893–S11901, 2009

Interactive Comment

southeast.

Corrected

Page 20258, line 27 wouldn't high nighttime ozone at Mt. Happo be explained by the fact that this is a high altitude site that becomes decoupled from the surface and dry deposition processes at night?

In response to this comment, we have revised the discussion about diurnal variations of ozone at Mt. Happo.

"Although a similar diurnal pattern of boundary layer heights is shown at Ijira and Happo, ozone at Mt. Happo shows little diurnal variation. On July 7 and 10, highest observed ozone mixing ratios are found near midnight, 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 ozone at Happo. 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. "

Throughout the paper, when discussing data in units of ppbv, refer to the data as mixing ratios and not concentrations.

Corrected

Figure 6 Here the back trajectories from Mt Tai are initialized at 1000 m, but Mt Tai is at 1500 meters. Why the discrepancy? The back trajectories need to be initialized at 1500 m. In the text you need to give more explanation of the clustering method. Did you specify that only 4 clusters would be produced for each year? What does the color of each trajectory signify?

ACPD

8, S11893–S11901, 2009

We have decided to remove the correspondent figure from the manuscript since it did not add much to the discussion and the inclusion of TRACE-P and TRMM measurements data has increased the length of the manuscript.

Figure 2a The caption mentions whiskers but I don't see any in the figure. Please be specific about which years you are showing data from, for the model and the observations.

Corrected (Measurements in 2001 and other years are distinguished with black and gray lines).

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

ACPD

8, S11893–S11901, 2009

Interactive Comment