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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of meteorological variability on interannual variations of the springtime boundary layer ozone over Japan during 1981–2005" *by* J. Kurokawa et al.

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The authors would like to thank the Editor, Dr. Cooper, for taking the time to review our manuscript and giving very constructive and informative comments. These comments were very helpful and enabled us to improve the quality and clarity of the manuscript. We revised our manuscript based on them. Our detailed responses to the comments are below.

Reply to general comments:

> Comment:. . . In this study the authors found that their regional model over predicted Japanese ozone due to problems with too much ozone entering the model at the boundaries (Dr. Lin can provide the revised version of the paper: mlin26@wisc.edu). I wonder if your model might also have a similar problem because ozone in your study is too high for years with strong westerly flow (transport from Asia) and also for years with strong southerly flow (transport from the Pacific).

Reply:

We agree that the boundary conditions are one reason for the discrepancy between simulated and observed results. We added a new section in which we evaluated the general model performance for O3 over East Asia and compared the springtime averaged O3 simulated by EyyMyy with EANET observation data from remote sites in Japan. However, we found that the simulated results were not systematically larger than the observations. Therefore, it is not likely that the boundary conditions are the major reason for systematic over-prediction of O3 over Japan at the air quality monitoring stations managed and operated by the Ministry of the Environment of Japan and by local governments. We plan to conduct long-term simulations with lateral and upper boundary conditions that include interannual variations in a future study; the results will be analyzed in detail and compared with those of this study.

> Comment: Regarding your hypotheses that the problem is due to the model not resolving ozone titration, a simple test would be to compare the model output to measured Ox in three groups: rural, suburban and urban. The ozone titration problem should be more evident for the urban sites, while the rural sites should have better agreement with the model. Finally, examining time series of Ox broken down by rural, suburban and urban locations will be one final check that upwind ozone has a major impact on Japan. If upwind ozone and interannual variability affect all regions of west central Japan, then similar ozone trends and IAV should be seen for the three types of measurement sites. Adding a figure showing the trends for these three categories would be helpful for understanding your analysis and will also be of interest to

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researchers who keep track of ozone trends.

Reply:

We agree that in the previous manuscript, we did not discuss the large overestimation of simulated O3 compared with observations of air quality monitoring stations over Japan with sufficient detail. Unfortunately, the stations used in this study do not provide information about site characteristics. Thus, we classified the sites of the stations as urban, suburban, or rural on the basis of the 21-year average (during 1985–2005) of springtime NOx concentrations observed at each station. As expected, observed Ox values were smallest over urban areas and largest over rural areas. However, simulated O3 values over WCJ were still about 5–10 ppbv larger than observed Ox over rural areas, which suggests that even sites defined as rural might be influenced by neighboring urban or suburban areas. In order to further investigate the cause of the differences between observed and simulated results, we compared concentrations of Ox' (the sum of simulated O3 or observed Ox and NO2 generated secondarily by the oxidation of NO in the atmosphere) and found that the simulated Ox' values and the interannual variability of them agreed well with observations. From these results, we inferred that differences between observed Ox and simulated O3 over WCJ were mainly caused by the dilution of NOx emissions in the coarse model grid of this study. In addition, all observation types, namely Ox at urban, suburban, and rural areas and Ox' over WCJ, showed very similar trends, which suggests that the interannual variation of O3 over WCJ is influenced by large-scale factors rather than by local ones. We added these results and discussion to the revised manuscript.

Reply to additional comments:

> Comment: Some additional explanation of the Ox measurement methodology is required. If I understand you correctly, the Japanese government does not directly measure O3 but instead measures Ox. Please list the major species that contribute to Ox.

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Reply:

The definition of Ox has been added, and the major species of Ox listed.

> Comment: Mixing ratios of Ox must be greater than mixing ratios of O3 and there must be some sites in Japan where both are measured simultaneously. Please give an indication of the difference between O3 and Ox, with the understanding that this will vary between sites, especially between rural and urban sites.

Reply:

We agree that comparisons of results observed by the KI and UV methods at the same time and place are very important. However, we did not have such data sets. Thus, we selected 10 pairs of neighboring stations and examined the relationships between Ox concentrations observed by the KI method at one station and those observed by the UV method at the other station of each pair. Although this comparison was not of observations made at exactly the same place, the difference between Ox values observed by the KI method and those observed by the UV method seemed to be small. In addition, Maeda et al. (1997) reported that concentrations of peroxyacetyl nitrate measured by the KI method are about one-fifteenth the actual concentrations; thus, the sensitivity of this method to Ox other than O3 is relatively small. Accordingly, we consider the difference in observation methods not to be a critical problem for the analysis of interannual variability of springtime O3 over Japan. We have added these results and discussion to the revised manuscript.

> Comment: page 7563 line 7-8 Do you mean to say: The majority of Ox instruments were calibrated with the KI method.

Reply:

Yes. The corresponding sentence has been changed for clarity. Furthermore, we noticed that the percentage given in the sentence (70%) was wrong and corrected it (60%).

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> Comment: page 7563 line 16 I don't understand what is meant by: Also KI scale for Ox showed about 9% larger sensitivity than O3 scale. Please elaborate and/or re-phrase. And please define what you mean by "scale".

Reply:

The corresponding sentence has been rewritten for clarity, and the word "scale" is not used now.

> Comment: page 7563 line 19 What percentage of the 136 stations is rural, suburban and urban?

Reply:

As mentioned in the reply to the second comment, we classified the station sites as urban, suburban, or rural on the basis of NOx observations at each station. By this means, we determined that 17%, 65%, and 18% of the 136 stations are urban, suburban, and rural, respectively.

Reply to minor comments:

> Comment: page 7561 line 1 You state that the inflow concentrations of O3 from the stratosphere are set to zero, but what about ozone in the stratosphere within the model domain Does it contain ozone so that any fresh stratospheric intrusions in the model domain will impact tropospheric ozone?

Reply:

The description about the inflow of stratospheric O3 in the model (the upper boundary condition) was not appropriate and has been modified in the revised manuscript. We assumed that the influence of stratospheric O3 on the interannual variation of O3 over WCJ was likely to be relatively small because according to Terao et al. (2008), in regions other than Canada and Europe, the stratospheric influence on the variability of tropospheric O3 is small, and local sources or variability in transport from other source

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regions apparently play a dominant role. However, the upper boundary conditions as well as the lateral boundary conditions could be the cause of the discrepancy between modeled and observed O3 over Japan. In addition to the effects of the lateral boundary conditions on interannual variation of O3 over Japan, those of the upper boundary conditions will be examined in a future study.

> Comment: page 7562 line 4 When calculating BL ozone do you include data from all times of day in the average?

Reply:

Yes. We have added a brief sentence to the revised manuscript to specify this.

> Comment: page 7570 line 13 and elsewhere When describing the NINO3 index please specify that it corresponds to the Niño 3 region which is commonly used in studies of El Niño

Reply:

The "NINO3" index used in this study is the same as the Niño3 index that is commonly used in the studies of El Niño. We have specified this in the definition of the Niño3 index and have added the abbreviation "NINO3" to Table 1.

> Comment: Figure 4 The panels are too small. Please increase by at least 50 %. Also, please change the units of e) and f) to hPa.

Reply:

We have increased the size of the panels in Fig. 4 (Fig. 6 in the revised manuscript) by about 50%. The units of Fig. 4e and 4f have been changed to hPa.

> Comment: Figure 5 and 6 Modify the legend to read: blue dot: ASPA > 1 or ASPA <-1 green dot: -1 < ASPA < 1

Reply:

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The legends of Figs. 5 and 6 (Figs. 7 and 8 in the revised manuscript) have been modified as the Editor suggested.

> Comment: In the comments below, if no explanation is given, please insert (or substitute) the text provided into the appropriate place in the manuscript.

Reply:

Thank you very much for correcting the English of our manuscript. All indicated problems have been corrected.

References

Maeda, J., Liang, C., Bandow, H., Maeda, Y., and Mizoguchi, T.: Response of KI oxidant analyzer for peroxyacetyl nitrate (in Japanese), J. Jpn. Soc. Atmos. Environ., 32, 425-430, 1997.

Terao, Y., Logan, J. A., Douglass, A. R., and Stolarski, R. S.: Contribution of stratospheric ozone to the interannual variability of tropospheric ozone in the northern extratropics, J. Geophys. Res., 113, D18309, doi:10.1029/2008JD009854, 2008.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 7555, 2009.