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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.

J.-i. Kurokawa

kurokawa.junichi@nies.go.jp

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The authors would like to thank Anonymous Referee #2 for taking his or her time to review our manuscript and for giving very constructive and informative comments. These comments helped us improve the quality and clarity of the manuscript. We revised our manuscript based on them. Below are our detailed responses to the comments.

Reply to major comments:

> Comment: *** B7558-L24: Why do you focus on the "springtime BL O3" ? Here,

the authors should show clearly their motivation for concentrating on springtime O3, neither summertime nor annual O3. And, why do you use BL concentration instead of surface one in this study ? Discussion using the modeled surface ozone would be more straightforward, I feel. The authors could compare their results with observations more directly as well.

Reply:

As mentioned in the previous manuscript, we expect that transport of polluted air masses from continental Asia will continue to increase. The trans-boundary O3 seems to have a large impact on the maximum O3 concentration and exceedances of air quality standards in Japan, especially in the springtime. Therefore, in this work we focused on the influences of interannual variability of continental-Asian outflow on the springtime O3 over Japan. We have added these descriptions to the revised manuscript. However, we agree that analysis of the interannual variation for other seasons and the annual average is also very important. We plan to conduct this analysis next.

With respect to the reason for using BL O3, our focus is on the influence of large-scale meteorological variability such as continental-Asian outflow and inflow from the Pacific Ocean. Therefore, we consider that the analysis using BL O3 is preferable because surface O3 is more sensitive to the effects of local emissions, meteorology, and topography. Moreover, as we mention in the reply to "Comment *** B7562-L1:", the effects of the difference between BL and surface O3 on the results of this study are likely small.

> Comment: *** B7560-L1-L2: The authors state that their chemistry and aerosol schemes are not applicable to the stratosphere. Does it mean that your model does not consider chemistry in the stratosphere ?

Reply:

Yes. In this study, intrusions of O3 from the upper layers across the lateral boundaries

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determine the stratospheric and upper tropospheric O3 concentrations. We assumed that the influence of stratospheric O3 on the interannual variations of O3 over WCJ was relatively small because Terao et al. (2008) indicated that 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 regions likely play a dominant role. However, upper boundary conditions could cause the discrepancy between modeled and observed O3 over Japan. The influences of upper boundary conditions on interannual variation of O3 over Japan will be examined in a future study.

> Comment: *** B7560-L15-: How about reproducibility of your RAMS-CMAQ simulation for meteorological fields like temperature, wind, precipitation, water vapor ?

Reply:

Meteorological parameters such as temperature, wind speed and direction, relative humidity, and precipitation, simulated by RAMS with the same configurations of this study have been validated by Yoshida et al. (2006) and Uno et al. (2003, 2005).

> Comment: *** B7561-L1: I didn't get the point. what do you mean by "inflow concentrations" ? I presume it is a parameter separate from boundary condition in CMAQ. Could you explain more about this in the text ?

Reply:

The description of the inflow of stratospheric O3 in the model (the upper boundary conditions) was not adequate and has been modified in the revised manuscript.

> Comment: *** B7562-L1: This section discusses the modeled BL ozone. Is it possible for you to mention the difference between BL and surface (model 1st layer) ozone in you model ? and, how does it affect your discussions in this section?

Reply:

We compared the simulated springtime surface and BL O3 and found that their anoma-

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lies over WCJ were almost the same as between them. In addition, BL O3 showed a very similar horizontal distribution with surface O3, although absolute concentrations of BL O3 over WCJ were larger than surface O3 by about 2.5 ppbv on average. We therefore consider any effects of the difference between BL and surface O3 on the results of this study to be small.

> Comment: *** B7563-L5: "photochemical oxidants(Ox)": Presumably, this Ox includes oxidant species like PAN as well as O3 itself. If this is correct, the authors should use modeled Ox field instead of O3. In highly polluted area, O3 accounts for only 70 or 80% of Ox in some cases.

Reply:

First, we added the definition of Ox to the revised manuscript and listed the major species of Ox. With respect to the evaluation of influences of Ox other than O3, comparisons of the observed results between the KI and UV methods at the same time and place are required. However, we did not have such data sets. Thus, we selected 10 pairs of neighboring stations and examined the relations 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 done at exactly the same place, the difference between Ox values observed by the KI method and those observed by the UV method seems to be small. In addition, Maeda et al. (1997) reported that concentrations of peroxyacetyl nitrate determined 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 that the use of different observation methods is not a critical problem for the analysis of interannual variation of springtime O3 over Japan. We have added these results and discussion to the revised manuscript.

> Comment: *** B7564-L21: "...are generally about 20 ppbv larger than observed..." The authors should take this discrepancy more seriously in fact. This can break the

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validity of the discussions on O3 anomaly in this manuscript. Underestimation of the NOx titration effect in urban area can be a candidate for the reason of this discrepancy as the authors suggest. This can be easily confirmed by comparing your model results only with the data selected for non-urban observation sites in the WCJ region. The authors use BL O3 for their discussions. I presume that BL O3 is systematically larger than surface O3 in the model because of higher contributions from the stratospheric O3 transport at 1km altitude. At least, I think, the authors should evaluate their model results further using any other observational data.

Reply:

We agree that our discussion of the large overestimation of simulated O3 compared with the observations of air quality monitoring stations over Japan was not adequate in the previous manuscript. First, we compared the simulated springtime O3 with EANET observation data at remote Japanese sites. We found that the simulated results were not systematically larger than the observations. With respect to the air quality monitoring stations managed and operated by the Ministry of the Environment of Japan and by local governments, there is unfortunately no information about site characteristics. Thus, we classified the sites as urban, suburban, or rural using the 21-year average (during 1985–2005) of springtime NOx concentrations observed at each station. As expected, observed Ox 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 values over rural areas, which suggests that even stations defined as rural might be affected by neighboring urban or suburban areas. In order to further investigate the cause of 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 simulated Ox' values and the interannual variation of them agreed well with observations. From these results, we inferred that differences of observed Ox and simulated O3 over WCJ were caused mainly by the dilution of NOx emissions in the coarse model grid of this study.

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We have added this discussion to the revised manuscript. With respect to the influence of the difference between surface and BL O3, please see our answers to “Comment *** B7558-L24” and “Comment *** B7562-L1”.

> Comment: *** B7565 This section discusses the ozone anomaly in the context of pressure anomaly fields. Their focus seems to be only on transport processes like outflow from China. However, the authors should have described also anomalies in chemical processes involving O3 production intensity in the source regions (mainly China) and O3 loss (lifetime) in the downwind regions (around Japan and western Pacific). Please describe more about changes in chemical fields to clarify the roles of chemistry in their simulated ozone anomalies.

Reply:

In order to evaluate the influence of chemical processes on the IAV of O3 over WCJ, we investigated the chemical production, chemical loss, and net chemical production of simulated O3 over East Asia. However, we could not find any obvious influences of chemical processes on the IAV of springtime BL O3 over WCJ. Anomalies of O3 and its net chemical production over WCJ were not related. Anomalies of net chemical production of O3 over CEC were positively correlated with those of O3 over WCJ, but both the slope of the regression line and the correlation coefficient were relatively small. Therefore, in this study, we did not focus on the role of chemistry in the springtime BL O3 anomalies over WCJ. We present these results in detail in the supplementary material of revised manuscript.

> Comment: *** B7569-L23: "...by the hypothesis described in Sect. 3.3 (1) and (2)" I don’t recognize well what this represents. Please be more specific here.

Reply:

The corresponding sentence has been rewritten and made more specific.

> Comment: *** B7570-L8: "ENSO and their influences on tropospheric O3

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(...:Koumoutsaris et al.,200 8)" Koumoutsaris et al. 2008 showed significant increases in O3 exported from Asia to the western Pacific including Japan in an ENSO phase (1998). This appears to be in the opposite direction to your results. Please describe this point and you're your interpretation.

Reply:

We examined the O3 anomalies in March 1998 simulated by E00Myy and found similar positive O3 anomalies over the western Pacific. In our results, O3 anomalies over the central part of Japan were also positive, although those over the western part were negative. However, positive O3 anomalies over the western Pacific in our model domain almost disappeared in April and May. Thus, influences of El Niño in 1998 on BL O3 over Japan might be different between March and the subsequent springtime months of April and May. We have added this information to the revised manuscript.

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.

Uno, I., Carmichael, G. R., Streets, D. G., Tang, Y., Yienger, J. J., Satake, S., Wang, Z., Woo, J.-H., Guttikunda, S., Uematsu, M., Matsumoto, K., Tanimoto, H., Yoshioka, K., and Iida, T.: Regional chemical weather forecasting system CFORS: Model descriptions and analysis of surface observations at Japanese island stations during the ACE-Asia experiment, J. Geophys. Res., 108(D23), 8668, doi:10.1029/2002JD002845, 2003.

Uno, I., Ohara, T., Sugata, S., Kurokawa, J., Furuhashi, N., Yamaji, K., Tanimoto, N., Yu-

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9, C2661–C2668, 2009

mimoto, K., and Uematsu, M.: Development of the RAMS/CMAQ Asian Scale Chemical Transport Modeling System (in Japanese), J. Jpn. Soc. Atmos. Environ., 40, 148-164, 2005.

Yoshida, Y., Yumimoto, K., and Uno, I.: Evaluation of meteorological parameters over East Asia simulated by the Regional Meteorological Model, RAMS, (in Japanese), J. Jpn. Soc. Atmos. Environ., 41, 235-248, 2006.

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