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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term change in the contributions of various source regions to surface ozone over Japan" *by* Tatsuya Nagashima et al.

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Response to the comment of Referee #2

The authors greatly appreciate your critical reading of our manuscript and highly valuable suggestions and comments. Our responses to your comments are listed below. (Pages and lines are those in the track-changed manuscript)

(RC): Referee Comment / (AR): Author Response

General comment 1:

(RC) This study used the NCEP renalysis data to drive the Chaser model. Please compared the meteorological parameters with observations (surface or satellite) if possible. For example, cloud information and temperature. This is important to ozone simulation.

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(AR) Thank you for the comment. Unfortunately, I didn't output any cloud parameters simulated in the model, but I could compare the surface temperature used in the model with those observed in Japan which were compiled by Japan Meteorological Agency (JMA). JMA selected 15 sites which undergo little urban influences to derive the average surface temperature over Japan. The modelled annual mean surface temperature averaged over whole Japan showed a significant warming during the simulation period, 0.44 $\pm$ 0.21 °C/decade, which well matched the observed warming of 0.45 $\pm$ 0.23 °C/decade. Add to this long-term trend, the inter-annual (year-to-year) variation was also well captured by the model, although there was a discrepancy that the modelled temperature was somewhat warmer than the observation in 2000s particularly in winter which might be related to the slight overestimation of winter surface O3 in the model depicted in the Fig.5. I added the following sentences to the manuscript. (P12, L520-526)

"The surface temperature over Japan in the model which was assimilated into NCEP/NCAR reanalysis data showed a warming of 0.44 $\pm$ 0.21 °C/decade in the annual mean during the simulation period which well corresponded to the observed warming of 0.45 $\pm$ 0.23 °C/decade (JMA, 2017). The IAV of the surface temperature was well captured by the model too, although the modelled temperature was somewhat warmer than the observation in 2000s particularly in winter which might be related to the slight overestimation of winter surface O3 in the model depicted in Fig.5."

General comment 2:

(RC) The author concluded that CHN contributed a lot to the trends of ozone in Japan. This can be expected because China's emissions are large and increases in last decades. I encourage the authors to analyze the contributing ability of each regions to JPN ozone. For example, how many is the contribution of China per NOx/VOCs emissions increase to JPN O3 trends in unit: ppbv/Gg NOx or VOCs.

(AR) Thank you for the comment, the concept of the contributing ability is quite in-

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teresting. However, because the sensitivity simulations to derive the contribution of emission trend in each region was done by varying all the O3 precursors emission simultaneously, it is not straightforward to estimate the contributing ability of a single O3 precursor individually. Nevertheless, I tried to estimate the contributing ability by conducting a multiple regression analysis with NOx and VOC emissions in a source region (e.g CHN) as explanatory variables and the contribution of that region on the surface O3 over Japan as the target variable, however, the analysis was failed because of the high correlation between NOx and VOC emission trends in the source regions such as CHN (p-value of the regression coefficient for NOx is 0.18) and KOR (that for VOC is 0.17). Therefore, deriving the contributing ability of different source regions should be addressed in the future study with a careful experimental design for that purpose.

Major comment:

(RC) The authors should gave a short discussion on the uncertainties of models and its impact on the conclusions. for example, the emission inventory. REAS should be different with other inventories (MEIC or EDGAR4). I wonders if the difference between inventories affected the conclusions.

(AR) I added a short discussion on the uncertainty of O3 precursor emission inventories and possible impacts on the conclusions in the last chapter as follows. (P14, L647-658)

“The results summarized above depended largely on the forcings of long-term simulation, particularly the long-term variation of the emissions of O3 precursors in Asia. Zhao et al. (2013) estimated the NOx emission in China for the period 1995âĂŤ2010 and compared it to the existing emission inventories including Hao et al. (2002), Zhan et al. (2007), and the version of REAS used in this study. They showed the log-term increasing trend in Chinese NOx emission in REAS was consistent with that in the other inventories, but the amount of emission was somewhat smaller in REAS than in the others. Therefore, the long-term increasing trend in the contribution of Chinese emission to the surface O3 over Japan showed in the preset study would be retained if the

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other emission inventories were used for the simulation but the specific values of the contributions could be affected. Further studies should address the impact these uncertainties in the different emission inventories on the trend of surface O3 over Japan."

Best regards,

Tatsuya Nagashima

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-1087/acp-2016-1087-AC2-supplement.pdf

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