

## ***Interactive comment on “Influence of the variation in inflow to East Asia on surface ozone over Japan during 1996–2005” by S. Chatani and K. Sudo***

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I thank Anonymous Referee #1 for thorough review and variable comments. My replies are as follows.

(Anonymous Referee #1) A number of circumstances unfortunately limit the value of the study. Most bothersome is the poor correspondence between model results and observations. As long as the model can not accurately reproduce near-surface ozone over Japan (e.g. Figs. 4-6) it is difficult to trust the conclusions drawn from the model study.

(Reply) I agree that this is the weakest point of this study. However, other studies are also facing the common problem as referred in the manuscript. Several studies would

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be necessary to solve this problem. There were two points from which we judged that this study was applicable to investigate the influences of the inflow. The first point is the performance of the simulation at remote EANET sites (Ogasawara and Hedomisaki) located in southern islands. The simulation could capture the seasonal variation of surface ozone at these two sites. It means that the simulation has capabilities to reproduce background surface ozone which are mainly affected by the inflow while the simulation may have some difficulties to represent local situations. The second point is the relatively good performance of the simulation in reproducing the annual variation and the long-term trend of surface ozone as shown in Fig. 7 and Fig. 9, and as described in Sect. 4 and 5. Correlation coefficients and p values between observed and simulated surface ozone were calculated. They indicated that the variation of observed surface ozone was reproduced by the simulation well. The first paragraph of 3.3 will be slightly modified in the revised manuscript to clarify our idea on the first point described above. Correlation coefficients and p-values will be added to Fig. 7 of the original manuscript.

(Anonymous Referee #1) The poor performance of the model could partly be sought in the type data used for evaluation, i.e. measurements collected at >1000 “populated areas” in Japan. Given the resolution of the model (including the emission inventory) it would have been more appropriate to evaluate the model against data collected at regional monitoring stations, such as the EANET data, or similar. A brief attempt was made to compare the model results to individual time series at 10 Japanese EANET stations.

(Reply) I agree that it would be ideal to evaluate the model at regional monitoring stations such as the EANET data. However, there are two reasons why measurements collected at >1000 “populated areas” were chosen. The first reason is the temporal coverage of monitoring data in the populated area. They continued monitoring at least for ten years during 1996-2005. On the other hand, EANET sites started monitoring in around the year 2000. The temporal coverage of EANET monitoring is too short

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to evaluate the long-term trend of surface ozone. The second reason is indeed the motivation of this study described in the introduction. The purpose of this study is to investigate reasons why surface ozone is increasing while precursors are decreasing in Japan. This increasing trend has been reported by the Japanese government using monitoring data at >1000 “populated areas”. Therefore, we would like to investigate the influence of the inflow exactly at >1000 “populated areas”. The first paragraph of 2.3 describing monitoring at >1000 “populated areas” will be modified in the revised manuscript to clarify their importance.

(Anonymous Referee #1) It is remarkably that no evaluation (i.e. comparison with observations) of the model’s performance outside Japan is made, especially after speculating that the main reason for the model’s poor behaviour is excessive ozone production in NE China.

(Reply) The concentrations of observed surface ozone in China were discussed but any values were not shown in the original manuscript. The validation using observed surface ozone reported by Wang et al. (2009), Xu et al. (2008) and Lin et al. (2008) will be explicitly discussed in the revised manuscript. Descriptions of three monitoring sites will be added to 2.3 and Fig. 2 (as shown in Fig. 1 in this comment). After the first paragraph in 3.3, a new section “3.4 Monthly variation of surface ozone in China” will be added. The third paragraph of 3.1 containing brief explanations of the performance in China in the original manuscript will be moved to the new section 3.4, and some expressions will be changed to fit the new section. Monthly variations of concentrations of observed and simulated surface ozone in the BXX case averaged during 1996–2005 at three monitoring sites in China will be shown in the new figure (as shown in Fig. 2 in this comment). Dips in summer do not appear in simulated values at Linan and Shangdianzi. Especially, the concentration of surface ozone was significantly overestimated in summer at Shangdianzi which is located in a high concentration zone shown in Fig. 3. In addition, discussions in the second and following paragraphs in the original manuscript will be reorganized in a new section “3.5 Discussions on

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the performance of the simulation”. Some expressions will be modified to fit the new section.

(Anonymous Referee #1) From Figs. 4-5 it appears that the regional model merely shifts the level of ozone by a constant value, i.e. the seasonal cycle of WRF/Chem is more similar to CHASER than to the measurements. The statistical analysis does not convincingly show that the regional model is better than the global model (except for the overall bias). I suggest a more rigorous evaluation of WRF/Chem, focussing on background stations in the whole domain rather than urban stations in Japan only.

(Reply) I agree that the regional model is effective to reduce the overall bias. Descriptions in the first paragraph of 3.2 meant only this advantage. An evaluation in China will be added in the revised manuscript as described in my reply above.

(Anonymous Referee #1) The discussion about possible reasons for the overestimation of ozone in Northeast China on pp. 30833-30834 is a bit speculative and far-fetched, in my opinion. The authors discuss possible deviations of emissions during certain months in certain areas (e.g. “Beijing region”) or refer to studies in the Pearl River delta which is quite a distance from Beijing and Japan. It is suggested that erroneous emissions may be the cause for the overestimated ozone concentrations in Japanese populated areas from summer through early winter. Would it not be worthwhile to test the impact of neglecting temporal (seasonal and diurnal) variations of emissions as well as neglecting biomass burning in the present set-up. Emissions from biomass burning often contribute to a very large fraction of ozone precursors.

(Reply) In summer, the concentration of simulated surface ozone is high over Northern China, and it appears that Japan is affected by the transport by westerly winds from a high concentration zone in Fig. 3 of the original manuscript. The concentration in a high concentration zone is significantly overestimated as discussed above. Therefore, possible reasons of the overestimation in Northeast China are discussed. I agree that emissions in Japan may also cause the overestimation. Brief descriptions of one

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study (Chatani et al., 2011) will be added as a reference in the new paragraph in the end of 3.5. It indicated that the much finer resolution and seasonal and diurnal variations of emissions could reduce the overestimation. If biomass burning emissions are included in the simulation, the concentration of simulated surface ozone may become even higher. The influence of biomass burning emissions is remaining issues as mentioned in my reply to Anonymous Referee #2.

(Anonymous Referee #1) On page 30833 it is also stated that “. . .transport of ozone from a high concentration zone to Japan may be overestimated in summer.” Could it not be that the transport from the clean Pacific during summer is underestimated? The model performance at the remote island stations Ogasawara and Hedomisaki seem to be good throughout the year. The problem with the model performance could therefore be sought in the description of the wind speed and direction (both in CHASER and in WRF/Chem). Maybe you could evaluate this important meteorological parameter?

(Reply) Although any figures and descriptions were not included in the manuscript, no problem which may cause the overestimation was found in the meteorological field. What I meant here is not about the meteorological field. The major wind direction is southerly in summer, but the wind direction is not constant for all days. Ozone would be transported from the west for some days with westerly winds. Average concentrations are calculated by weighting the ozone concentration transported from each direction and its frequency. Therefore, the contribution of the transport from each direction to the average concentration is determined by not only the frequency but also the ozone concentration. Problems in the meteorological field would mainly influence the frequency, but the amount of ozone transported from each direction is also important factor on the average concentration. The current expressions in the original manuscript seem to be confusing, and will be replaced by a following sentence. “the contribution of the transport of ozone from a high concentration zone to the average concentration of surface ozone over Japan may be overestimated in summer.”

(Anonymous Referee #1) On page 30827 almost a full paragraph is spent discussing

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CMAQ. While I appreciate all references to previous work on the subject, I don't think it's necessary to go into details of e.g. deficiencies in the vertical transport of CMAQ.

(Reply) I hope to retain these descriptions because this is the important motivation to choose WRF/chem in this study. I am also extensively using CMAQ in other studies, but I could not solve this problem yet. I would like to share this experience with other readers and stimulate studies which aim at solving this issue.

(Anonymous Referee #1) Page 30829, line 22: Is it really the “Japanese government” that has noticed that ozone represent the major fraction of all photochemical oxidants. It would be good, if you could support this statement with a reference.

(Reply) It is true while no relevant reference is available. However, expressions may be inappropriate in this context. This sentence will be deleted and expressions in preceding sentences will be modified in the revised manuscript as follows. “instruments which detect only ozone have been officially approved by the Japanese government because differences in concentrations of photochemical oxidants and ozone are expected to be small. They have been used at some of the monitoring stations. Therefore, differences in concentrations between ozone and photochemical oxidants were ignored in this study.” It must be noted that NO<sub>2</sub> is not included in photochemical oxidants in the Japanese definition.

(Anonymous Referee #1) On page 30831 you mention that the present study only deals with “surface ozone”, contrary to other studies, e.g. Kurokawa et al (2009a). What do you mean with “surface ozone”. Is it from the lowest layer of your model, or is it scaled to a particular height over the surface? Please clarify.

(Reply) Ozone in the lowest layer is defined as “surface ozone” in this study. This explanation will be added in the second paragraph of 2.1.

(Anonymous Referee #1) Page 30835: When discussing interannual variability of ozone over Japan you may also want to refer to: Calori, G., Carmichael, G.R., Streets,

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D., Thongboonchoo, N., Guttikunda, S.K., 2001. Inter-annual variability in sulphur deposition in Asia. Journal of Global and Environmental Engineering 7, 1–16.

(Reply) It was difficult to link the description on ozone in the original manuscript to the inter-annual variability of sulfur deposition described in the proposed paper. Therefore, this paper will not be included in the revised manuscript and the references.

(Anonymous Referee #1) The “wind-arrows” in Figs. 3, 8, 11 are too small. They need to be larger.

(Reply) Wind-arrows will be slightly larger in the revised manuscript as shown in Fig. 3 of this comment.

(Anonymous Referee #1) Use identical colour-legend for the seasonal and annual values in Fig. 3.

(Reply) Identical colour-legend will be used in the revised manuscript as shown in Fig. 3 of this comment.

(Anonymous Referee #1) Exchange “Whole months” to Annual Mean (or similar) in Fig. 3 onwards.

(Reply) “Whole months” will be replaced by “All months” in the revised manuscript as shown in Fig. 3 of this comment. Other figures will be also fixed.

(Anonymous Referee #1) Page 30824, line 1: “Air quality simulation . . .” -> “Air quality simulations . . .”

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30824, 20: “carbons” -> “CO<sub>2</sub>” or “carbon”

(Reply) This part will be corrected to “CO<sub>2</sub>” in the revised manuscript.

(Anonymous Referee #1) 30825&30826: Consider rephrasing some of the instances of “influence” since it is used in great numbers on these pages.

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(Reply) Some of the instances of “influence” will be replaced by similar expressions in the revised manuscript.

(Anonymous Referee #1) 30825, 27-28: “for the past years” Unnecessary, delete?

(Reply) This part will be deleted in the revised manuscript.

(Anonymous Referee #1) 30826, 2: “its” -> change to “the regional CTM’s” ?

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30826, 19: “both” Unnecessary, delete?

(Reply) “both temporal variations” will be corrected to “daily and annual variations” in the revised manuscript.

(Anonymous Referee #1) 30826, 25: “in other seasons as well as spring” Unnecessary, delete?

(Reply) This part will be deleted in the revised manuscript.

(Anonymous Referee #1) 30827, 21: “major” -> change to “a large” ?

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30827 & 30828: use “x” instead of a dot to describe the grid-size and number of gridcells?

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30827, 25: “sigma-P” -> change to “sigma-p” ?

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30828, 15: “simulation results” Unnecessary, delete?

(Reply) This part will be deleted in the revised manuscript.

(Anonymous Referee #1) 30829, 3: Start a new paragraph after “. . . lower strato-

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sphere.” ?

(Reply) A new paragraph will be started after “. . . lower stratosphere.” in the revised manuscript.

(Anonymous Referee #1) 30829, 3&26: Don't use point when writing the number of monitoring stations. Write 1045 (or 1 045).

(Reply) This part will be corrected to 1045 in the revised manuscript.

(Anonymous Referee #1) 30830 onwards: “whole months” is confusing. Use “all months” or “annual mean” or “yearly average” etc. to label this category.

(Reply) “whole months” will be replaced by “all months” in the revised manuscript.

(Anonymous Referee #1) 30830, 21: “southern ocean” ? For me southern ocean is the part of the Atlantic, the Pacific and Indian Ocean that is immediately north of Antarctica. Is this what you mean or is southern ocean a Japanese name for the part of the Pacific north of the equator?

(Reply) “southern ocean” will be replaced by “Pacific Ocean” in the revised manuscript.

(Anonymous Referee #1) 30831, 13: “. . . that band of a high ozone” -> “. . . that a band of high ozone” ?

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30833, 3 onwards: “The simulation-reproduced . . .” The whole sentence needs rephrasing.

(Reply) This sentence will be simply corrected in the revised manuscript as follows. “The simulation reproduced dips in summer at Ogasawara and Hedomisaki,”

(Anonymous Referee #1) 30835, 8: “picking up” -> “selecting” ?

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30835, 16: “southwest ocean” Not a familiar name of an oceanic regions for me.

(Reply) “over the southwest ocean” will be replaced by “in the southwest of Japanese islands” in the revised manuscript.

(Anonymous Referee #1) 30835, 17-18: “but they are not obvious in ‘Low’ years.” Language needs improvement.

(Reply) This sentence will be deleted in the revised manuscript because it is not necessary.

(Anonymous Referee #1) 30836, 19: “. . . variation in inflow.” -> variation in modelled inflow.” ?

(Reply) This part will be corrected to “variation in modeled inflow” in the revised manuscript.

(Anonymous Referee #1) 30837, 29: “. . . were calculated . . .” -> “. . . were also calculated . . .”

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30838, 18: “. . . increasing rate of ozone” Reformulate? 30838&30839: Consider replacing a few of the instances of “increasing rate”.

(Reply) Alternative good expressions have not been found yet.

(Anonymous Referee #1) 30839 11-12: “. . . a regional CTM with a global CTM were coupled” -> “. . . a regional CTM were coupled with a global CTM”

(Reply) This part will be corrected in the revised manuscript.

(Anonymous Referee #1) 30840, 111: “Multiple simulations studies . . .” -> “Several modelling studies . . .”

(Reply) This part will be corrected to “Several modeling studies . . .” in the revised manuscript.

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## Reference

Chatani, S., Morikawa, T., Nakatsuka, S., Matsunaga, S. and Minoura H.: Development of a framework for a high-resolution, three-dimensional regional air quality simulation and its application to predicting future air quality over Japan, *Atmos. Environ.*, 45, 1383-1393, 2011.

Lin, W., Xu, X., Zhang, X., and Tang, J.: Contributions of pollutants from North China Plain to surface ozone at the Shangdianzi GAW Station, *Atmos. Chem. Phys.*, 8, 5889–5898, doi:10.5194/acp-8-5889-2008, 2008.

Wang, T., Wei, X. L., Ding, A. J., Poon, C. N., Lam, K. S., Li, Y. S., Chan, L. Y., and Anson, M.: Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994-2007, *Atmos. Chem. Phys.*, 9, 6217-6227, doi:10.5194/acp-9-6217-2009, 2009.

Xu, X., Lin, W., Wang, T., Yan, P., Tang, J., Meng, Z., and Wang, Y.: Long-term trend of surface ozone at a regional background station in Eastern China 1991-2006: enhanced variability, *Atmos. Chem. Phys.*, 8, 2595–2607, doi:10.5194/acp-8-2595-2008, 2008.

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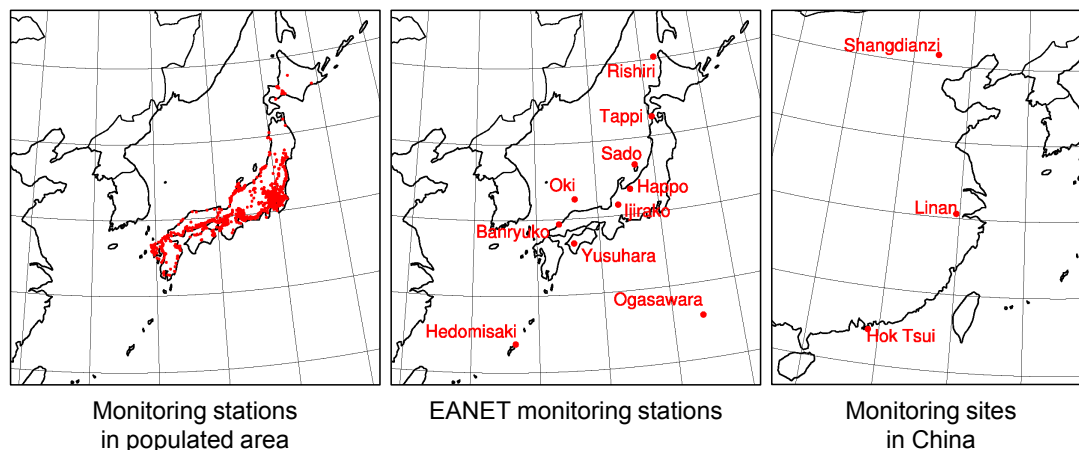
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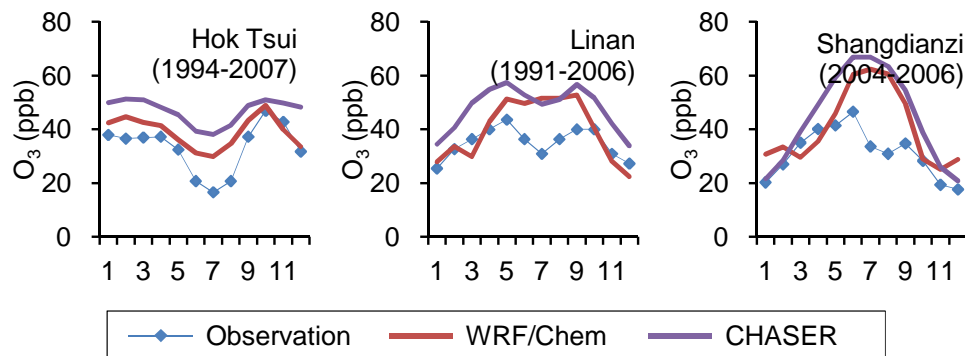
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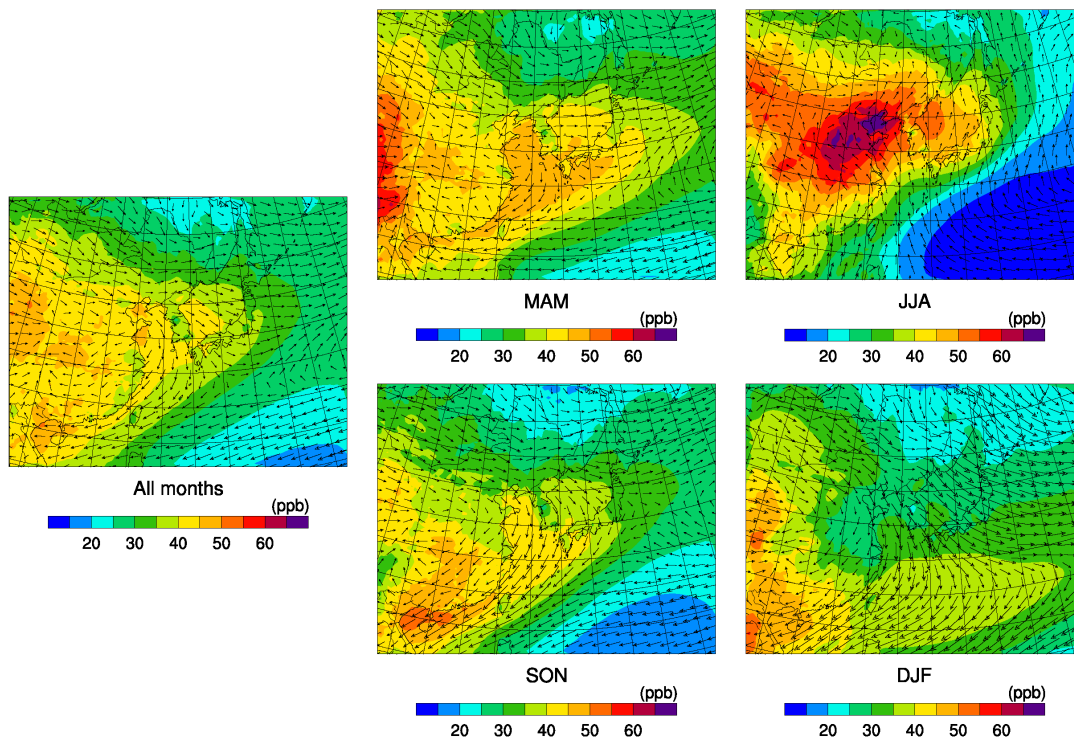
**Fig. 1.** Location maps of monitoring stations in populated area, EANET monitoring stations in Japan, and monitoring sites in China which were used in this study. (to be replaced with Fig. 2)

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**Fig. 2.** Monthly variations of observed and simulated concentrations of surface ozone in the BXX case averaged during 2000–2005 at each of three monitoring sites in China.

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**Fig. 3.** (to be replaced with Fig. 3 of the original manuscript)

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