

## ***Interactive comment on “Discrepancies between MICS-Asia III Simulation and Observation for Surface Ozone in the Marine Atmosphere over the Northwestern Pacific Asian Rim Region” by Hajime Akimoto et al.***

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Answer to Referee #2

(Major Comment) 1. What're the major differences between CMAQ 5.02 and 4.7.1? If they're very similar in lots of aspects, what's the point to compare these “two” models? It would be much more interesting to compare it with another “independent” model. As a result, the difference between CMAQ 4.7.1 and 5.0.2 was not very much for the targeted Marine region although substantial differences were found in the simulated

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NO<sub>x</sub> mixing ratios and net O<sub>3</sub> production at Oki and Hedo as shown in Figs. 5 and 6. Since we found that the different version of two CMAQ models gave substantially different results for heavily polluted land area over Beijing and Tokyo (Akimoto et al., 2019 in this special issue), we are interested in how much difference can be seen between the two different versions of CMAQ over oceanic region. Practical reason that we selected only NAQM and two CMAQ models in this paper is that we could use the submitted hourly data of concentrations of O<sub>3</sub> and NO<sub>x</sub>, and process analysis of net chemical O<sub>3</sub> production only for these models. Since the submission of hourly data and process data were not specifically requested by the MICS-Asia III project, such data were not submitted from other models for comparison.

2. I would suggest a detailed analysis of O<sub>3</sub> budgets, including chemical production/loss terms and physical removal terms of O<sub>3</sub>, at the sites, which might be also helpful for understanding the model overestimates. For example, underestimates of chemical loss of O<sub>3</sub> due to the halogen chemistry in the models would also contribute to the overestimates of O<sub>3</sub> at these oceanic sites. Unfortunately, the detailed terms consisting of net chemical production, production and loss terms, were not available under this project, and the direct comparison between the chemical and physical loss terms could not be made. We agree that the additional discussion of the contribution of the gas-phase halogen chemistry is worthwhile to be included in the paper. A description was added at the end of 3.4 in Lines 410-412.

3. One major conclusion in this paper is that the overestimate in O<sub>3</sub> over the study region in CMAQ is due to too small O<sub>3</sub> dry depositions whereas the better agreement between NAQM and observation might have resulted from a relatively larger O<sub>3</sub> dry dep. It would be interesting to conduct a few sensitivity model simulations, e.g., applying higher O<sub>3</sub> dry deposition rates in CMAQ or implementing the dry deposition that used in NAQM into CMAQ, to support the authors' major conclusions. Although we agree that such sensitivity analysis is very useful to strengthen our conclusion, it was beyond the activity of MICS-Asia III project and we could not perform this activity (Line

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407-409).

(Minor comments) 1. Line 69: I don't think the present paper aims to "solve" the discrepancies between models and observations. It may provide some useful information on advancing the current understandings of discrepancies between the two models and between modeled and observed O<sub>3</sub> at the sites. We agreed and modified the sentence in Line 56-58.

2. Line 102-105: It would be good to elaborate a little bit here to illustrate how important the role of air-sea exchange plays instead of just citing the papers. According to the suggestion, a paragraph was revised as in Line 93-102.

3. Line 106: It's not clear to me that a "more detailed comparison" is compared to which study? "more detailed" has been deleted.

4. Does JST mean Japanese local time? Need to clarify. "Japan Standard Time" has been added.

5. unit of O<sub>3</sub> needs to be consistent: in some places, it's ppb but others as ppbv. All have been unified to "ppbv"

6. Fig 1: Overlapping monthly winds in the figure may help readers better understand the transport patterns of O<sub>3</sub> in the region. We added the wind vectors to Fig.1.

7. Line 197: I would suggest the authors to avoid stating the model/models "excellently" capture observations. It seems that NAQM still overestimates observed O<sub>3</sub> by ~30-40% during nighttime. "excellently" has been deleted.

8. Line 229: " $\frac{1}{4}$  along with the edge of the Pacific High": it would be good to add isopiestic lines or wind patterns in Fig 4. It would be better to add the spatial pattern of O<sub>3</sub> on this day from NAQM as well. "along with the edge of the Pacific High" has been deleted. Spatial distribution of surface O<sub>3</sub> by NAQM has been added in Fig. 4 as 4(c).

9. Line 281: I'm wondering if this soil NO<sub>x</sub> from sugar cane fields had been considering

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any of these models? MIX emission inventory used for the modeling considers NO<sub>x</sub> emission from agricultural sources without specifying crop species. The sentence has been added in Line 288-290.

10. Line 282: It's not clear to me what the term "diurnal O<sub>3</sub> formation" means. "diurnal O<sub>3</sub> formation" has been changed to "daytime O<sub>3</sub> peaks".

11. Fig 5: I would suggest the authors plot the comparisons of observed and simulated NO<sub>2</sub> in one plot and the comparisons of NO in the other. That would be more consistent with the analyses in other similar figures and clearer for NO values. Figures for NO<sub>2</sub> and NO have been separated in Fig. 5.

12. Fig 6: I would suggest plotting the comparisons of modeled chemical production between three models in the same figure. Fig 6 has been modified according to the suggestion.

13. Fig 7: What're the simulated values from the other two models like at these two sites? Fig. 7 has been modified including the data for CMAQ 5.0.2 and NAQM according to the suggestion and combined to Fig. 6 as Fig. 6(b) and 6(c).

14. Line 419: " : : identified as the cause of overestimate": I would say the three factors are possible causes of models overestimates in O<sub>3</sub> since there could be other possible reasons. "cause" has been changed to "possible cause".

15. Line 422: May need to clarify the regions or sites that were suffered from overestimates in transported O<sub>3</sub> from continents. I'm also curious about whether the transport patterns of O<sub>3</sub> over the study region are significantly different in June or August from that in July. "at Oki in July" has been specified in the sentence (Line 456). From our former experience of study (Pochanart et al., Atmos. Environ, 36, 4235-4250, 2002), frequency of the transport of continental outflow is higher in June, followed by August and July is the least at Oki and Ogasawara during summer.

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