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Atmospheric Chemistry and Physics Discussions

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.

Anonymous Referee #3

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This manuscript investigated the CTMs performance of simulating ozone mixing ratios over marine regions in East Asia. This study suggested that the overestimates of three CTMs simulations could be caused by: 1) the overestimate of long-range transport (LRT) from the continent, 2) the overestimated in-situ photochemical production, and 3) the underestimated dry deposition of ozone over the ocean. The authors pointed that the underestimate of ozone dry deposition could be important. The text is concisely written and well documented. The topic is applicable for the Atmospheric Chemistry & Physics journal. However, the current manuscript lacked discussion and necessary

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analysis (please see the remarks below). The authors proposed couple of hypotheses, but did not show solid evidence to support them. First, the current manuscript used the surface observations (O3, NO2*, etc.) and modeled surface air pollutants and dry deposition velocity for LRT effects. The LRT is usually dominated by the transport in the free troposphere, and the PBL dynamics (the buildup of PBL in the early morning and the downward mixing of ozone and its precursors to the surface). So it is hard to make this conclusion only using the surface data and model results. The revised manuscript may investigate the PBL simulations if the authors can find aircraft observations or sounding data. Second, the paper did not state how the net in-site photochemical production rate was calculated. From the observation at Oki, the peak NO2 or NO2* on July 2 is not associated with high ozone levels, which did not support the hypothesis of local photochemical production. Third, the NAQS model did show good results simulating the ozone concentrations at these marine observation sites. However, NAQS tends to have consistent low bias as compared with two CMAQ simulations. If NAQS has significant underestimation of ozone levels in the source regions, we cannot conclude that NAQS has better model performance in simulating marine ozone concentrations. Lastly, the CMAQ model was developed by EPA to regulate the air pollution mainly over the land. So for the two versions used in this study, complex air-sea interactions and halogen chemistry are not included. So it is not surprised to see CMAQ has poor performance here. In summary, the current manuscript shows some results but lacks further discussion or analysis. Major revisions as indicated in the comments and remarks below are needed before consideration of publication in ACP.

Detailed Remarks/Suggestions for Revision

Line 82: Please define ‘NOz’ here

Line 125: This paragraph discussed the set-up of these 3 CTMs, and WRF was used to generate meteorological fields. But there is no information about the WRF simulations, such as the physical options and if observation/analysis nudging was used, which are

important for ozone transportation and deposition. I also cannot find these details in Akimoto et al., 2019 ACP paper. Different configuration of WRF could influence the regional CTMs simulations. The authors need to add explanations in the revised manuscript.

Line 195: I believe the 'transport amplitude' means the ozone enhancements due to the LRT. If yes, please revise this sentence to make it clear.

Line 211-214: As raised above, did these WRF runs generate consistent circulation patterns for these two episodes? Figure 4 shows the results from CMAQ v4.7.1, how about the ozone contours in CMAQ v5.0.2 and NAQS? Please include figures similar as Figure 4 in the supplementary material.

Line 239: This statement needs further analysis to support it. I agree that if the in-situ photochemical production is important at Oki, the observations should show a similar diurnal cycle which did not exist. Another possible explanation is that the LRT occurred usually in the free troposphere, and the downward mixing due to the PBL build-up can cause the same diurnal cycle I suggest the authors to examine the vertical profiles of ozone from CTMs over Oki to rule out this possibility.

Line 258-260: The observations in Fig 5a shows high NO2* concentrations around 07/02. However, I didn't see significant enhancement in ozone at Oki in Fig. 3a on the same day. The net photochemical production of ozone should be anticipated if the NO2 levels are higher. Need some explanation or discussion here.

Line 270: How these hourly net chemical ozone production rates are calculated?

Line 276: Should be 'in-situ photochemical ozone production in the CTMs, which contributes to the overestimate . . .'

Line 290: Better to show similar figures such as Fig 5 for Ogasawara site in the supplementary material to support this statement.

Line 298: I am surprised that NAQS predicted much lower ozone concentrations com-

pared with two CMAQ simulations. Especially for the Pearl River Delta (PRD) region, NAQS predicted extremely low monthly mean ozone, as low as 10-20 ppbv, for July. Same phenomenons are found in the Yangtz River Delta (YRD), Wuhan, Seol, and Tokyo. So the good performance of NAQS simulating marine ozone at these 3 marine sites could not be that the model successfully captured the nature, but NAQS has a systematic low bias for surface ozone. If that is the case, why select this model run? The Akimoto et al., 2019 ACP paper listed 12 regional model simulations for the MICS-Asia III project, and some WRF-Chem simulations should be introduced here.

Line 362-364: Not sure about NAQS. CMAQ models did not include halogen chemistry until version 5.2. So I am not surprised that the halogen chemistry did not impact the dry deposition of ozone over Bohai Bay and the Yellow Sea.

Line 368: Any observations support that the statement that the ozone concentrations in the Bohai Bay and Yelloe Sea are overestimated?

Line 370: Which one? Do NAQS and CMAQ have similar sensitivity to water surface resistance?

Line 426: I disagree with this argument. The LRT of ozone in the free troposphere should be more important than the transport near the water body. The KORUS-AQ campaign results in 2016 support this hypothesis. In my opinion, the underestimate of ozone deposition could only impact the surface ozone levels.

Figures Figure 4: Consider using different shapes to represent these 3 sites, for the readers who are not familiar with the names.

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