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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.

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Answer to Referee #3

(General Remarks) 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. It is true that the amount of long-range transport is affected by the mixing ratios of both in the boundary layer and free troposphere. It is generally analyzed by using three-dimensional trajectory analysis (Akimoto et al., 1996; Pochanart et al., 2002). However, the assessment of quantitative contribution of each is beyond the scope of this study, and no aircraft campaign data of vertical distribution of O3 is available in the targeted period of summer in 2010 in this area. A note has been added in the text in Line 384-389.

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. In the model, net photochemical production of O3 was calculated by the difference between model-calculated O3 formation terms and O3 loss terms by chemical reactions. As for the NO2* peak on July 2 at Oki appeared in the observation, it is apparently due to accidental local perturbation near the monitoring site or due to malfunction of the instrument, since the peak appeared only in the one-hour averaged data at 7 am (Japan Standard Time). Such a note has been added in the figure caption.

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. NAQM gave substantially lower O3 than CMAQ agreeing reasonably well with observation in the polluted source region over land according to our previous paper (Akimoto et al., 2019). We think, however, the lower continental O3 does not contribute much to the lower O3 at Hedo and Okinawa in July, since the frequency of long-range transport of continental airmass to these sites is rare in this month. Meanwhile, as noted just below, inclusion of the gas-phase halogen reactions in the model would decrease surface O3, and NAQM would underestimate the mixing ratio of monthly averaged O3 in this region (see Lines 392-409).

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. We added the discussion of the possible reduction of marine O3 by inclusion of the gas-phase chemistry of halogens into the models (Lines 392-409).

(Detailed Remarks/Suggestions for Revision) Line 82: Please define 'NOz' here Here, 'NOz' has been changed to 'NOy' (Line 71).

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. Some explanation about WRF has been added in Line 128-131.

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. We changed the 'transport amplitude' to 'amount of O3 increase due to the transport' (Line 206).

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. We added the similar figures for CMAQ5.0.2 and NAQM as Figs 4(b) and (c).

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. We confirmed that since O3 does not accumulate in the upper MBL during nighttime being different from over land where near surface O3 is depleted by NO titration and high deposition to ground during nighttime, the downward mixing after sunrise doesn't cause O3 buildup in the morning. Therefore, we think the observed diurnal variation of O3 is mainly due to in-situ photochemistry rather than vertical mixing.

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. As stated in the answer to the second comments of General Remarks, we think the observational NO2* peak in the morning of July 2 at Oki, is apparently due to artifact, either by accidental local perturbation near the monitoring site or malfunction of the instrument. Such a note has been added in the figure caption.

Line 270: How these hourly net chemical ozone production rates are calculated? As answered to the second comments in the General Remarks, the net photochemical production of O3 was calculated in the model by the difference between model-calculated O3 formation terms and O3 loss terms by chemical reactions.

Line 276: Should be 'in-situ photochemical ozone production in the CTMs, which contributes to the overestimate : : :' The sentence has been changed as suggested (Line

298-300).

Line 290: Better to show similar figures such as Fig 5 for Ogasawara site in the supplementary material to support this statement. We added the similar figures for Ogasawara in Fig. 5S-2 in the supplementary material.

Line 298: I am surprised that NAQS predicted much lower ozone concentrations compared 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. We selected NAQM not as a model reproducing well the observational data, but from the practical reason that we could use the submitted hourly data of concentrations of O3 and NOx, and process analysis of net chemical O3 production along with two CMAQ 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. Although we agree that comparison including WRF-Chem and other models would strengthen our discussion, we limited ourselves within the framework of MICS-Asia III. It is true that Fig. 8 shows NAQM gives lower O3 in most of the region in East Asia, However, as shown in our previous paper (Akimoto et al., 2019), NAQM reproduced well the O3 and NOx in megacity areas in Beijing and Tokyo in contrast to the overestimate of CMAQ 4.7.1 and 5.0.2. This paper has suggested that the lower mixing ratios of marine O3 in Northwestern Pacific would be due to the higher dry deposition velocity of O3 over oceanic water. Thus, the cause of lower O3 of NAQM would be different by region. It would be worthwhile to elucidate the cause of lower O3 by NAQM in PRD and YRD region, but it is beyond the scope of this paper.

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. As responded to the forth comments in the General Remarks and also to the comment of Reviewer #2, 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 392-409.

Line 368: Any observations support that the statement that the ozone concentrations in the Bohai Bay and Yellow Sea are overestimated? Unfortunately, as far as we know, there is no reported data of the mixing ratios of O3 over Bohai Bay and Yellow Sea. We recommended such observation in Future Research Recommendation (Line 421, 429-430)

Line 370: Which one? Do NAQS and CMAQ have similar sensitivity to water surface resistance? Here we described general statement not for the specific model. We omitted the sentence and replaced by a new the statement (Line 389-391).

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. Since we didn't make any quantitative analysis of the contribution of marine boundary layer and free tropospheric O3 to the transported O3 in the marine region of Northeast Asia, the statement has been modified taking into account the reviewer's comment. (Line 384-389; 459-462)

Figures Figure 4: Consider using different shapes to represent these 3 sites, for the readers who are not familiar with the names. We changed the symbols of the three sites and gave legend in the figure caption.

Over

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