Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2020-228-AC1, 2020

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.

Hajime Akimoto et al.

akimoto.hajime@nies.go.jp

Received and published: 8 June 2020

Answer to Referee #1

(General Comments) It is not clear to me, why only choose those periods because it should be based on the overall general or a year-round period to secure the generality of characterization. In this sense, this conclusion needs to be very carefully characterized and the re-analysised according to the separate specific events and their differing conditions. There are several choices how to select models and seasons for the dis-

cussion of discrepancies between models and observation in the marine O3 under the framework of MICS-Asia III project. As for the selection of the month, according to Figure 3 of the overview paper for ozone in the MICS-Asia III (Li, J. et al., 2019), the substantial overestimate of surface O3 in EA3 region covering the Northwest Pacific and the Sea of Japan, can be seen only in summer and early september by most of the models. This is the reason we selected only July as a representative month of summer for the discussion in this paper. The relative importance of long-range transport, in-situ photochemistry and dry deposition to ocean surface contributing to the overestimate would be different by months during the season, quantitative discussion of the cause of discrepancies among the models is out of the scope of this paper. A short notes relevant to the selection of only July have been added in Line 103-107.

(Major Comments) (1) Under- or over-estimation by model is presumably caused by very complicated factors in the regional CTM models. Authors employed two models with different versions of CMAQ. I thought it may make sense to compare it to a completely different and diverse models, such as the CAMx or WRF-Chem or GEOS-Chem model. This is because, as we do not know the "true" values of dry deposition velocities, and thus authors should open to the different possibilities of other controlling factors. For example, as depicted in Fig. 8, NAQM showed considerably lower ozone mixing rations over the whole domain than CMAQ, which in turn could derive the relatively lower ozone than CMAQ over Hedo and Ogasawara as well (it would be quite natural to me), rather than pointing out relatively higher dry deposition velocities than CMAQ. As authors are aware, models employ their different dry deposition parameterizations, mostly generating different values. We agree that inclusion of other models would be more persuasive and useful for the discussion of this paper. However, the reason we selected only the two CMAQ and NAQM is as follows. As seen in Figure 7 of the overview paper for ozone (Li, J. et al., 2019), there are large variability of surface O3 spatial distribution in the targeted marine region in summer among the 13 models submitted to MICS-Asia III. Even among the six WRF-CMAQ, and the three WRF-Chem models, the variability is substantial and it is not possible to select

representative WRF-CMAQ or representative WRF-Chem. Since the purpose of our papers in this special issue is to elucidate the main element causing the discrepancies, rather than to make quantitative discussion of the cause of whole variability among the models, we selected only limited number of models for discussion. More practical reason that we selected two CMAQs and NAQM is that we could use the hourly data of concentrations of O3 and NOx, and process analysis of net chemical O3 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. Although NAQM gives relatively lower O3 values than CMAQ over the whole region in East Asia as shown in Fig. 7, we think the cause of lower O3 is different between over land with high photochemical O3 production and over ocean where such activity is minimal. The cause of lower O3 in the heavily polluted region giving better agreement with the observation have already been discussed by our previous paper (Akimoto et al., 2019) in this special issue. We think the lower O3 in the oceanic region is caused by quite different reason as we discussed in this paper.

(2) As authors described in page 7 (Long range transport of O3), Oki is influenced by O3 inland area (and thus mostly urban O3). Probably CMAQ which usually simulate "higher-than observation" ozone mixing ratios over inland, and CMAQ model improvement over "land" area may automatically remove ozone biases in "marine" area where authors claim that dry deposition velocity should be higher. Yes, we think overestimate of O3 over land by CMAQ definitely contribute to the overestimate of the transported peak on July 6 and 23. The reduction of the overestimate would remove the substantial part of the overestimate on these days. A sentence has been added in Line 234-236.

(3) (Line 355-375) It is also confusing that, among three models, NAQM reproduce ozone well over Hedo and Ogasawara primarily due to the higher dry deposition velocities than those of CMAQ. However, in the case that dry deposition velocities of Bohai Bay and Yellow sea have to be raised, then Oki will be expected to be also down, but ozone will be also down simultaneously in Hedo and Ogasawara where originally there

were no biases of ozone mixing ratios. I guess increasing dry deposition velocities in model NAQM over Bohai Bay and Yellow sea will not satisfy both. We think the dry deposition velocity over Bohai Bay/Yellow Sea region is much different from the open Pacific south of Japan, since the former is expected to be much larger than the latter due to the presence of much higher amount of organic materials deposited from the polluted atmosphere to cover the ocean surface. Underestimate of dry deposition velocity of NAQM is expected only over Bohai Bay/Yellow Sea region. Therefore, region-specific dry deposition velocity should be given to the model simulation although the necessary quantitative data is not available at the moment. Therefore, in the Section of Future Research Recommendations, we recommended the measurement of deposition velocity of O3 in the Northwestern part of the Pacific Ocean particularly over Bohai Bay and Yellow Sea.

(4) Finally I recommend to secure some more simulation cases. Authors used at least global model for initial condition such as GEOS-chem and CHASER: dry deposition velocities from two global models vs. over- (or under-) estimation of ozone mixing ratios can be also useful to justify authors' conclusion from multiple cases. Or in some cases, authors may reach a different conclusion (i.e., model internal errors instead of different dry velocities). The dry deposition velocity used in the GEOS-Chem and CHASER has been given in the text, and a comment has been added in Line 364-369.

(Specific comments) 1. Specify the dry deposition parameterizations (with references) for all two (or three) models. Description on the parameterizations of Wesely (1989) and M3DRY (Pleim et al., 2001) used in the NAQM and CMAQ was added in Line 131-132.

2. During July 23-26 in Fig. 3(c), observation of ozone in Ogasawara showed strong diurnal variations with big differences between max. in daytime and min. in night time. Should there be a photo-chemical reactions? because Ogasawara is thought to be a real background site: it has nothing to do with both local photo-chemical formation and transport of NOx from other areas I guess. Although Ogasawara is generally

thought to be a real background site based on its remote location over the open Pacific ocean, the surroundings of the monitoring station is actually surrounded by trees and the data may be affected by the very local emissions of biogenic VOCs and soil NOx, which may possibly cause some photochemical activity to form in-situ O3 under certain meteorological conditions. Such a note has been added in Line 207-213.

3. Line 230. — three models reproduce observation reasonably well.. It is confusing because, in the previous sentences, only NAQM's results matches well with observations in Ogasawara. Here the "increment" of transported O3 is reproduced well in all the three models. Overestimate of by the CMAQ is mostly caused by the overestimate of "background" O3 and resulted the overall better matching of NAQM at Ogasawara (Line 248).

4. Please indicate the locations of Bohai Bay and Yellow sea areas. The location of Bohai Bay and Yellow Sea has been added in Fig.1.

5. is it possible to analyze the case different periods like May or June where there are high ozone mixing ratios with (or without Long-range transport processes) over East Asia?, because just one single month test might have a possibility of sometimes misleading the conclusions. As noted in the answer to the general comments at the top of this document, substantial overestimate of surface O3 in EA3 region covering the Northwest Pacific and the Sea of Japan, can be seen only from July to September. In May there is excellent agreement of monthly averaged mixing ratio of O3 and there is much less overestimate by the model in June than July. We agree that analysis of these months may yield different contribution of the transport, in-situ photochemistry and dry deposition to each site, we wish to limit the purpose of this paper to identify these three elements as possible cause of discrepancies between the model simulations and observation.

Over