

Response to the Reviewer #2:

We thank the reviewer very much for reading our paper carefully and giving us valuable comments.

Detailed responses to the comments are given below.

This paper discusses comprehensive shipborne O₃ and CO measurements covering a large oceanic region from the Arctic to the Southern Ocean over the period of 2012-2017. The dataset was thoroughly analysed and was compared to the simulation results from a tropospheric chemistry reanalysis model (TCR-2), demonstrating the usefulness of such dataset in critical model evaluation. The authors also carried out two focused analyses assessing the underlying processes causing models to underestimate Arctic O₃ and to overestimate O₃ in the western Pacific equatorial region, respectively, compared to observations. The paper is very well written with detailed and in-depth analyses. The dataset is a significant addition to the current surface O₃ database over remote oceanic regions and is valuable for model evaluations. I recommend the paper to be published after the authors have addressed some minor comments that are detailed below.

Specific comments:

- 1) Page 1, L31: "less efficient dry deposition" than assumed in the model sounds very speculative. Dry deposition coefficient is generally considered very slow over the ocean, and it is unlikely that there is much room for a significant impact when adjusting the dry deposition coefficient.

The discussion on dry deposition in the Arctic is revised.

The dry deposition velocity of TCR-2 (CHASER) on the Arctic ocean surface, $\sim 0.04 \text{ cm s}^{-1}$, is on the high side of $\sim 0.01\text{--}0.05 \text{ cm s}^{-1}$, a range adopted into global atmospheric chemistry models. This is now clarified in the revised manuscript (first in page 5, lines 16-20 in the model description section and then in page 12, lines 7-9 in the discussion section on the Arctic processes) as follows:

"The dry deposition velocity (v_d) of O₃ is computed as $(r_a + r_b + r_s)^{-1}$, where r_a , r_b , and r_s are the aerodynamic resistance, the surface canopy (quasi-laminar) layer resistance, and the surface resistance, respectively (Wesely, 1989). $1/r_s$ over ocean surface was assumed to be 0.075 cm s^{-1} globally, irrespective of regions (Sudo et al., 2002). As a result, v_d was $\sim 0.04 \text{ cm s}^{-1}$ over the Arctic open ocean in September, for instance. This will be a subject of discussion in Sect. 3.3.2."

"The v_d , $\sim 0.04 \text{ cm s}^{-1}$ over the Arctic open ocean in September for CHASER (TCR-2), is on the high side of $\sim 0.01\text{--}0.05 \text{ cm s}^{-1}$, a range adopted into global atmospheric chemistry models (see Fig. 4 of Hardacre et al., 2015)."

Then we cited Ganzeveld et al. (2009), having discussed potential impact of v_d change on the surface ozone concentrations in high-latitude regions (page 12, lines 9-10):

"Ganzeveld et al. (2009) discussed a sensitivity study shifting their standard v_d of 0.05 cm s^{-1} to 0.01 cm s^{-1} substantially increased surface ozone concentrations by up to 60% in high-latitude regions."

Indeed, with $v_d = 0.04 \text{ cm s}^{-1}$, assuming 500 m of boundary layer height, the lifetime of ozone due to dry deposition would be as low as about 14 days in a rough calculation. As the photochemical loss over the Arctic is weak, dry deposition can be an important loss process. In the future we will perform a sensitivity test with different assumption with v_d

On the basis of this discussion, in Abstract, the previous sentence is kept but is now clarified that the sentence is for CHASER (TCR-2) (page 1, line 33):

"For TCR-2 (CHASER), dry deposition on the Arctic ocean surface might also have been overestimated."

2) Page 1, L33: "the observed O3 level frequently decreased to . . .": could add "more" before "frequently"

The sentence is revised accordingly.

3) Page 3, L1-5: I am not sure CO2 observations are that relevant here.

We agree that CO₂ is not that relevant to the main topic of this manuscript, i.e., analysis of reactive species. However, we still think it important to learn the strategy how the network observations are maintained and how the data are widely collected. Thus, the previous sentences are preserved.

4) Page 4, L27: please define "BC".

We mention "black carbon" in the revised manuscript.

5) Page 5, 1st paragraph: Can you briefly describe the chemical mechanism used in the TCR-2 framework? This will inform a later discussion of photochemical production.

In the revised manuscript, we will include the following sentences (page 5, lines 14-16):

"The base forward model CHASER V4.0 used for TCR-2 has been described by Sekiya et al. (2018). Briefly, 93 species and 263 reactions (including heterogeneous reactions) represent O_x-NO_x-HO_x-CH₄-CO photochemistry and oxidation of non-methane volatile organic compounds. Tropospheric halogen chemistry is not included."

6) Page 6, L10 & 11: Suggest naming the regions where these locations are.

We mention " the western Pacific equatorial region" in the revised manuscript.

7) Page 6, L22 & 23: “South of . . .” & “In equatorial regions. . .” seem overlap in what the authors try to convey; South of 15oN implies the equatorial region as well, if it doesn’t go beyond 15oS.

This part is revised as follows in the revised manuscript (page 7, lines 2-3):

"South of 15° N, even lower levels were dominant, i.e., <15 ppbv; particularly in equatorial regions, levels less than 10 ppbv of O₃ were frequently observed."

8) Page 7, L29: Could you elaborate a bit more on “overestimate of photochemical O₃ production”?

In the revised manuscript we will mention overestimation of photochemical O₃ production during long-range transport in TCR-2 (page 8, line 7).

9) Page 9, L27, Is “less efficient production of O₃” related to the chemical mechanism used in the model?

This part is describing a single case. Considering possibilities that J values and other important parameters are also not well represented in the model, it is difficult to relate the less efficient production of O₃ specifically to the chemical mechanism.

10) Page 10, L20: Could you elaborate “Processes other than daytime photochemistry. . .”?

We will mention "processes other than daytime photochemistry (e.g., nocturnal chemistry)" after revision.

11) Page 11, L6: the significance can be established by Student’s t test.

Welch’s t test, allowing unequal variances in the two group, was made. In the revised manuscript, we will mention that "the reanalysis significantly underestimated this (24.6 ppbv) based on Welch’s *t* test ($p < 0.001$)."

12) Page 11, L17: “unlike large increments obtained at low and. . .” – what does “large increments” refer to?

The sentence will be rewritten as follows:

"The reanalysis ozone over the Arctic Ocean can be similar to the model predictions, except when poleward transports are strong enough to propagate observational information from low and mid latitudes."

13) Page 11, L29-30: what is the dry deposition coefficient over this region used in the model? How does it compare with literature values for a similar land surface type? It is unlikely that dry deposition plays a significant role here, especially over the ocean; see above.

For the Arctic, the assumed dry deposition velocity over ocean was $\sim 0.04 \text{ cm s}^{-1}$ for CHASER (TCR-2), at the high side of a range often used in global atmospheric chemistry models ($0.01\text{-}0.05 \text{ cm s}^{-1}$). A rough calculation assuming 500 m of boundary layer height, the lifetime of ozone due to dry deposition over ocean is calculated to be as 14 days. For the air masses traveling over ocean for >3 days, which were typical for observations, dry deposition may have potential to reduce O_3 levels by $\sim 20\%$. Over the Arctic, as the photochemical loss is weak, dry deposition can be an important loss process. Indeed, Ganzeveld et al. (2009) discussed a sensitivity study shifting their standard v_d of 0.05 cm s^{-1} to 0.01 cm s^{-1} substantially increased surface ozone concentrations by up to 60% in high-latitude regions (page 12, lines 9-10). In the future we will perform a sensitivity test with different assumption with v_d .

14) Page 11, L31-32: Can you put a reference here?

McClure-Begley et al., 2014 is added.

15) Page 11, L33: what is “(AMAP 2015)”?

It is a report from AMAP, Arctic Monitoring and Assessment Programme, Arctic Council, as shown in the reference list. Now the reference is given correctly as (AMAP, 2015).

16) Page 12, P14: It looks like the comparison between observations and ACCMIP models depends on the frequency ranges, and it is too general to claim the comparisons are poorer for December. Maybe you could elaborate on the seasonal difference in model performance? Is there any systematic model bias that are season dependent?

As found in the subsection title, our focus here is to compare occurrence of $< 10 \text{ ppbv}$ in the model and observational datasets. And as the observations are basically limited to the two months, it is difficult to discuss the seasonal difference in detail. Thus we will just simply add " for any of the percentiles" to explain the poor comparison for December in the revised manuscript (page 12, lines 28-30):

"For December (Fig. 12b), the performances of the models were poorer; although CESM-CAM-super fast and GISS-E2-R again captured the observed distributions in low ranges, all others, including the ACCMIP ensemble median and TCR-2, overestimated the mixing ratios for any of the percentiles."

17) Page 12, L16-17: “The large variations among the model results could be the result of different assumptions regarding the dry deposition velocity of O_3 ” – Do you have any reference to back this up? It surprises me that differences in dry deposition among the models over the ocean would result in such a large model spread. I’d rather think that differences amongst the models in the efficiency of transport of mid-latitude polluted air to the Arctic, coupled to maybe differences in mid-latitude ozone production, is

likely the driving factor. The large model spread in the middle ranges in March might reflect the impact of large variations in transport.

For this part, discussing low O₃ levels at low latitudes, we will delete a previous speculative sentence regarding possible variability in the dry deposition velocity among the ACCMIP models in the revised manuscript, as the reviewer pointed out. Instead, we added that "the large variations among the model results may reflect the impact of large variations in transport, particularly in March", as suggested by the reviewer (from page 12, lines 30-31).

18) Page14, L22: again, what is the dry deposition coefficient over the ocean used in here? See my previous comments regarding the unlikely impact from dry deposition over the ocean. Many of the ACCMIP models use "off-line" dry deposition schemes characterized by prescribed, fixed dry deposition velocities over open water and ice which are documented in the literature. Hence an assertion that variations in these assumed dry deposition velocities drive the differences in the simulation of O₃ needs to be backed up by a discussion of this literature and does not need to be the subject of speculation.

As discussed above, we still think that the relatively fast dry deposition assumed in the CHASER (TCR-2) would at least partially explain the lower-than-observed O₃ levels over the Arctic. The sentence in Sect. 4 is retained but is now clarified that this sentence is for TCR-2.

19) Page14, L29: I would replace "later inter-comparisons" with "future intermodal comparisons".

We will revise accordingly.

20) Technical comments:

Page 6, L5 & L9: may replace "namely" with "i.e." Page 12, L5: Do you mean "studied region"? Figure 1: swap positions of (a) and (b), domain 3 is not clearly visible Figures 6 and 7: "CO" missing from the captions – should be "surface CO and O₃" Figure 13: add "mixing ratios" after "maximum"

We will revise accordingly.

Finally, we added a co-author Takashi Sekiya, who contributed significantly to development of data assimilation system and TCR-2.

We again thank the reviewer for the important suggestions.

References

McClure-Begley, A., Petropavlovskikh, I., and Oltmans, S.: NOAA Global Monitoring Surface Ozone Network. 1973-2014. National Oceanic and Atmospheric Administration, Earth Systems Research Laboratory Global Monitoring Division. Boulder, CO. <http://dx.doi.org/10.7289/V57P8WBF>, 2014.

Sudo, K., Takahashi, M., J.-I. Kurokawa, and Akimoto, H.: CHASER: A global chemical model of the troposphere 1. Model description, *J. Geophys. Res.*, 107(D17), 4339, doi:10.1029/2001JD001113, 2002.

Wesely, M. L.: Parameterization of surface resistance to gaseous dry deposition in regional-scale numerical models, *Atmos. Environ.*, 23, 1293-1304, 1989.