

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-319

Anonymous Referee #2

Referee comment on "Arctic tropospheric ozone: assessment of current knowledge and model performance" by Cynthia H. Whaley et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2022-319-RC2>, 2022

In this study, Whaley and coauthors perform an ambitious comparison between simulations and observation of tropospheric ozone in the Arctic. The model output result from simulations for the Arctic Monitoring and Assessment Programme (AMAP) performed with a number of models with different characteristics. The comparison also includes ozone precursors like NO_x and CO. The results for the different compounds are evaluated with surface, aircraft, and satellite measurements. The major conclusion is that the performance of the models concerning the correct simulation of tropospheric ozone has not significantly increased in the last 10 to 20 years. Overall, this is an interesting paper, which merits publication in ACP. However, similar to referee #1 I recommend major revisions before publication.

The manuscript appears like an offspring of a previously published paper on a model evaluation of short-lived climate forcers in the Arctic (Whaley et al., 2022). This paper also included model results for ozone in the Arctic including a comparison to observations. This manuscript here describes the results and comparisons for ozone in more detail. However, as already mentioned by referee #1 a number of relevant processes and studies related to arctic ozone are mentioned, but are not analyzed in sufficient detail. This concerns for example the springtime depletion of ozone and the related halogen chemistry, for which a number of requirements have been postulated in the literature (temperature thresholds, presence of first-year ice, second-year ice, or frost flowers, etc.) or the emission of NO_x from the snowpack as local source. Moreover, previous reviews and modeling studies as well as observations beyond the coastal stations are missing. Some examples are given in the comments below.

Further comments

The authors may want to add a point on iodine chemistry, which was very recently pointed out as important for the destruction of Arctic ozone (Benavent, N., et al., Substantial

contribution of iodine to Arctic ozone destruction, *Nature Geoscience*, <https://doi.org/10.1038/s41561-022-01018-w>, 2022).

Fig. 1: I'm somewhat confused by the representation of the Arctic. The Arctic is actually a large ocean covered permanently or seasonally by sea ice with continents around. While there are some local emissions due to shipping or other marine activities in the Arctic Ocean, the majority of the emissions are on the continents around the Arctic Ocean. I think this could be better represented in the figure. The figure also gives the impression that the stratosphere-troposphere exchange happens in the Arctic (and only in the Arctic). Isn't there also a fraction of Arctic ozone with a stratospheric origin that is mixed down in lower latitudes and then it is transported to the Arctic in the free troposphere. Is this correct or is this negligible? If this plays a role, I think this could be added to the figure, too. Finally, in L. 104f it is stated "...which show marked seasonal and inter-annual variations (Figure 1)." It is not clear to me, how these variations are represented in the figure.

L.148: "...shipping would become the main surface O₃ precursor source."?

L. 178f: "These phenomena are most commonly observed at Arctic coastal locations in March/April..." I'm missing here the reference to Helmig et al., who did the first review on Arctic ozone observations (Helmig, D., et al., A review of surface ozone in the polar regions, *Atmos. Environ.* 41, 5138-5161, 2007.) Moreover, it would be important to mention here that the few available springtime O₃ measurements over the Arctic Ocean actually show an even more pronounced depletion (Bottenheim, J.W., et al., Ozone in the boundary layer air over the Arctic Ocean: Measurements during the TARA transpolar drift 2006-2008, *Atmos.Chem.Phys.* 9, 4545-4557, 2009; Jacobi, H.-W., et al., Observation of widespread depletion of ozone in the springtime boundary layer of the Central Arctic linked to mesoscale synoptic conditions, *J.Geophys.Res.* 115, D17302, doi: 10.1029/2010JD013940, 2010 and the more recent observations during MOSAiC also in Benavent, N., et al., Substantial contribution of iodine to Arctic ozone destruction, *Nature Geosci.*, <https://doi.org/10.1038/s41561-022-01018-w>, 2022)

L. 181ff: "Interestingly, Yang et al. (2020) and Huang et al. (2020) were able to explain major depletion events in a model study by introducing the wind-induced release of bromine from the snowpack. However, the models could not explain the depletion events observed at low wind speeds. Swanson et al. (2022) used the GEOS-Chem model to show that both blowing snow and the snowpack are important sources of bromine during the spring." It is unclear why these three studies are selected here, while many more modeling attempts can be found in the literature, e.g. (without being complete) Toyota, K., et al., Analysis of reactive bromine production and ozone depletion in the Arctic boundary layer using 3-D simulation with the GEM-AQ: Inference from synoptic-scale patterns, *Atmos.Chem.Phys.* 11, 3949-3979, 2011; Yang, X., et al., Snow-sourced bromine and its implications for polar tropospheric ozone, *Atmos. Chem. Phys.*, 10, 7763-7773, <https://doi.org/10.5194/acp-10-7763-2010>, 2010.

L. 192f: "Their findings suggest a dark wintertime source of reactive bromine (halogens) that could feed halogen photochemistry at lower latitudes as the sun returns." A direct impact of a dark mechanism on ozone (and mercury) has been observed over sea ice in Antarctica according to Nerentorp Mastromonaco, M., et al., Antarctic winter mercury and ozone depletion events over sea ice, *Atmos. Environ.* 129, 125-132, doi: 10.1016/j.atmosenv.2016.01.023, 2016.

Ch. 3: For the comparison of the model output with the observations the authors use a multi-model median using the model grid boxes containing the measurement site. However, the models have different spatial resolution. Is this considered? Does this impact the calculated medians?

L. 254f: "...but negative IASI biases were found compared with aircraft data in the lower troposphere, due to low thermal contrast in the Arctic boundary layer." Does that mean that there is a bias in the retrieval of O₃ from IASI data in the boundary layer or do the authors refer to a boundary layer process? Please clarify.

Ch. 5: The authors refer here for the first time to the "high Arctic". It would be good to define earlier in the manuscript what the authors mean by this region and probably also if they refer to the Arctic itself.

L. 303: Figure 5a appears in the text before Figure 4.

L. 317: "Seasonal cycles at Arctic stations are not extensively discussed in the literature,..." See Helmig et al., 2007 (see above).

L. 369ff: "Therefore, while none of the Arctic sites currently exhibit summertime surface maxima due to photochemical production, as often observed in polluted locations further south, this may change in the future with increasing local anthropogenic emissions (e.g. Marelle et al.2018)." This is highly speculative since at the moment summertime minima are still observed. Maybe the authors could give a more quantitative estimate here, i.e. by how much would the different O₃ precursors need to grow to actually turn the Arctic into an area with summertime photochemical production of ozone?

L. 595f: "...but surface bromine/halogen chemistry needs to be included to simulate springtime surface O₃ at coastal Arctic locations ..." I think it would be safe to say that halogen chemistry needs to be include to simulate springtime O₃ in the entire Arctic and not only at coastal stations, where observations exist to compare the model output.

Finally, there are some sensitive agreements between the current manuscript and the already mentioned paper by Whaley et al., 2022 (Atmos. Chem. Phys., 22, 5775–5828, 2022) when it comes to the model descriptions (see pages 6, 7 and 9 of the iThenticate.com Similarity Report). I leave it to the editor to decide if these matches need a revision.