

Response to Dr. Hongyu Liu's comments:

We thank Dr. Hongyu Liu for his valuable and thoughtful comments. Our responses to the comments are provided below, with Dr. Hongyu Liu's comments italicized and our responses in plain and bold fonts.

This paper presents a GEOS-Chem model analysis of surface ozone and its precursors (NO_x, PAN, C₂H₆, C₃H₈, CO) observed at Summit, Greenland during the period of July 2008 - June 2010, with a focus on their concentrations and seasonal variations. Modeling tropospheric ozone in the Arctic has been challenging, and it is very interesting to use a state-of-the-art chemical transport model to test and improve our understanding of its sources and variability. The authors identified the discrepancies between the GEOS-Chem simulations and observations, which were then examined using various model perturbation experiments. The results are original, and the paper is concise and very well written. I recommend its publication on ACP with some minor modifications, as itemized below.

1). Title - Using "tropospheric ozone" in the title is a bit misleading. Although this study also compared the model vertical profiles of ozone and specific humidity with ozonesonde observations, the main scope of this paper is "surface ozone".

Response: We agree with the reviewer's comment. We therefore change the title as "Surface ozone and its precursors at Summit, Greenland: comparison between observations and model simulations".

2). Section 2: It is not clear which version of the GEOS-5 meteorological data archive was used. Is it GEOS-5.1.0 or GEOS-5.2.0? See e.g., http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-5_met_field_reprocessing and "http://wiki.seas.harvard.edu/geos-chem/index.php/List_of_GEOS-5_met_fields".

Response: Thanks for pointing this out. It is GEOS-5.2.0. We have added this in Section 2 as "The GEOS-Chem model has fully coupled O₃-NO_x-VOC-Aerosol chemistry mechanism and is driven by assimilated meteorological data from the Goddard Earth Observing

System version 5.2.0 (GEOS-5.2.0) of the NASA Global Modeling Assimilation Office.”
(lines 105-107)

3). Section 2, 2nd paragraph: "Time series data were archived with 3-hr temporal resolution at the Summit grid box" — I think you meant "grid column". Moreover, it is not clear how the model output was sampled in the vertical. The elevation of Summit is 3212m asl. Did you sample the model bottom layer, or the model vertical layer that is about 3212m above the sea level? The latter may very well be different than the former because the topography is not well resolved at coarse resolution. Would the results about model overestimates or underestimates found in this paper be different if the alternative way of model sampling is used (e.g., lines 206-207)?

Response: Good points. We archived the time series data with 3-hr temporal resolution at Summit grid box for each model vertical level, including the model bottom layer. For comparison with surface observations at Summit, Greenland, we sampled the data for the model bottom layer. Indeed, the topography is not very well resolved at coarse model resolution, and we believe diagnostics for the model bottom layer would work better than those for the 3212 m level for comparison to the surface measurements.

Minor editorial comments:

Line 43: change the "and" before "volatile organic compounds" to comma.

Response: Changed.

Line 56: the ARCTAS mission

Response: Corrected.

Line 66: What do you mean "O₃ mixing ratios below the boundary layer"? Within the boundary layer?

Response: Yes, it is within the boundary layer. We have therefore corrected the sentence as “Wespes et al. (2012) also revealed that model simulated O₃ mixing ratios within the

boundary layer were significantly underestimated during spring-summer, compared with ARCTAS measurements.” (lines 68-70)

Line 77: "....used to be the global default anthropogenic C₂H₆ emission inventory" - Do you mean "default" in GEOS-Chem or any other models?

Response: Yes, it is the GEOS-Chem default anthropogenic C₂H₆ emission inventory.

Line 228: add "over Greenland" after "PAN".

Response: Added.

Line2 268-270: "relative to NEI11_MIX" – isn't this relative to NEI11? Remove it?

Response: Well, it is relative to NEI11_MIX. In terms of emissions, you are right because we don't change the MIX emissions in this sensitivity simulation. Therefore, in order to avoid confusion, we give a simulation name for this sensitivity run and change this sentence as “We therefore run a sensitivity simulation by increasing the NEI11 C₂H₆ emissions by 40% and keeping other model configuration identical to NEI11_MIX (hereafter referred to as NEI11_40_MIX). We find this update leads to an increase in the model simulated annual mean surface C₂H₆ mixing ratios over Summit by only 6% during the period of 07/2008-06/2010 (figure not shown), still not able to explain the high model bias.” (lines 283-287)

Line2 279-281: The annual mean agrees quite well with observations, but the simulation is worse in summer.

Response: Points are well taken. We have changed this sentence to “We find that the simulated annual mean surface C₂H₆ mixing ratios at Summit from NEI11_MIX20 agree quite well with observations (within 1%). Similarly, better agreement between model and observations are found for monthly average values for October - January. However, the new simulation is not able to reproduce the seasonal cycle of C₂H₆ - the model significantly underestimates in February – April but overestimates in June – September (Fig. 5).” (lines 296-301)

Lines 283-288: Good point, but this long sentence needs a break.

Response: Thanks for pointing this out. We have divided the long sentence to “Note that this standard version of GEOS-Chem does not account for the sink of C₂H₆ from the reaction with chlorine, which could reduce the global annual mean surface C₂H₆ mixing ratio by 0-30%, and the global burden of C₂H₆ by about 20% (Sherwen et al., 2016). However, this may introduce additional uncertainty for our measurement-model comparison, together with the uncertainty in the seasonality of C₂H₆ chemistry.” (lines 304-308)

Line 325: "Unfortunately, ..." – "However, ..."

Response: Corrected.

Lines 339-340: "..., which implies that GEOS-Chem possibly underestimates STE for O₃ over Summit" – This is interesting and appears consistent with Choi et al., ACP 2017 (<https://www.atmos-chem-phys.net/17/8429/2017/>), see their Fig. 6), where the GMI CTM driven by MERRA (GEOS-5.2.0) underestimates ozonesonde-observed ozone in the Northern Hemisphere high-latitude upper troposphere.

Response: Thank you for providing us a reference source, which also attributed the model low bias to STE. We have included a discussion about this study in the text as “This is consistent with the study by Choi et al. (2017), which found low bias with model simulated O₃ mixing ratios over high-latitude upper troposphere of the Northern Hemisphere, compared with ozonesonde data, and attributed the low bias to weak STE in the model.” (lines 359-361)

Lines 358-359: Summit, Greenland; surface ozone

Response: Good suggestion. We have changed the whole sentence to “We combine model simulations with two-year (July 2008 - June 2010) ground based measurements at Summit, Greenland, to investigate the abundance and seasonal variations of surface O₃ and related species in the Arctic.” (lines 370-372)

Figures 2, 3,5, S1: In the caption, state briefly what the perturbation simulations are and refer the reader to the text for details.

Response: We have added additional descriptions in the captions for Figures 2, 3, 5, and S1. Please refer to our revised manuscripts for details.