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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Arctic marine secondary organic aerosol contributes significantly to summertime particle size distributions in the Canadian Arctic Archipelago" *by* Betty Croft et al.

Anonymous Referee #2

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GEOS-Chem-TOMAS chemical transport model with size-resolved aerosol microphysics is used in this manuscript to interpret measurements conducted during the summertime of 2016 in the Canadian Arctic Archipelago. Arctic marine secondary organic aerosol (AMSOA) is introduced to the simulation and this implementation significantly reduced the discrepancy between measured and modeled aerosol size distribution. This discrepancy is further decreased by shifting the volatility of organic vapor precursors of AMSOA. The simulated size-resolved composition shows that the highest AMSOA contribution was on ultrafine particles larger than 10 nm. Implications of AMSOA were also examined in the manuscript by estimating pan-Arctic direct and indirect radiation force causing by AMSOA. This work represents a potentially substantial

contribution to arctic aerosol formation and growth, which is well within the scope of the journal and the quality of presentation is good in general. However, I do have several concerns mostly related to the scientific approaches. I will support publication of this manuscript if the authors can properly address the following comments.

1. Line 491 to 515: The top-down estimation of the "fixed VOCs flux of 500 μg m-2 d-1" is an essential part of the simulation but it is presented underwhelmingly here. First, it is unclear to me how this quantity is "adopted to best represents observations as shown in the following sections within the context of our simulations". What are the specific observations and setup of simulations for the estimation? At line 1105-1106, the authors doubled this flux to get the lowest MFE in Eureka so this quantity definitely varies. Fixed VOCs flux is used in this model for simplicity. However, the authors stated several places in the manuscript that spatial variability of this flux is causing part of the uncertainties, for example at line 834. Since the VOCs are indicated to be biogenic, their variation should be significant, especially when estimating pan-arctic radiation effects. It is unclear to me why temporal and spatial variation of this flux has not been explored in this simulation. "To put the implemented flux in context, this is within an order of magnitude of the isoprene flux estimated from a north temperate deep lake (Steinke et al., 2018). Future work should include a bottom-up estimate of the SOA-precursor source flux." I do think the authors should do include a short summary of measurement of VOC concentration, fluxes, and volatility. The VOCs measurements were mentioned in introduction and conclusion but quantitative evidence supporting this VOCs flux is lacking in the manuscript.

2. Line 557: MFE is defined as the average of fractional error of size distributions with zeroth to third moment weight of diameters. Definitions of the moments of size distributions are in Section 3.3 at line 712. The authors should consider moving them to the method section. Technically, one of these moments, for example, number distributions in (a) of figures 3, 4 and 5 have all information for the rest of these figures. Integrated diameter (1st moment) and surface area (2nd moment) were not discussed

in texts. Please justify the necessity of all these moments in this manuscript. I do not oppose to include all four moments but they should be discussed more properly. MFE is oddly defined as the average of the four moments. It would be interesting to show the fractional error of each of the moment (perhaps in a figure instead of Table 3). This comment also applies to fractional bias.

The boundaries of the summation on equation 2 should be from i=0 to i=N-1 and from i=0 to i=3 in equation 6 in Hodshire et al. (2018). The units on the y-axes in figures 3, 4 and 5 are not correct. DlogDp has no unit (dlogDp=logDp2-logDp1=log(Dp2/Dp1)) so dN/dlogDp should always have the same unit as N for example. I believe that these are just typos and no numbers need to be updated but mistakes like this reflect poorly on the completeness of this paper. More proofreading is necessary in the revised manuscript.

Minor Comments:

1. Line 43: "Open water and coastal". Coastal is a vague word here. Since VOCs are from "ice-free seawater" (Line 502), I would remove "coastal" here and hereafter.

2. Line 105-122: Terrestrial emissions of VOCs from lakes are tundra should also be included here for comparison. See more above.

3. Line 131-133: "... as other types of MSOA arising from precursors... more strongly influenced by shipping and differing types of marine biogenic activity." The authors seem to consider marine SOA from natural sources in the rest of the manuscript. Clarifications are needed here how other types of MSOA are influenced by shipping.

4. Line 654: The reason for only show time series of Alert and Ship track but not Eureka is not clear to me.

5. Line 658: "Interestingly, at several times the elevated number concentrations occur at both Alert and at the ship, such as on August 3, 8, 9, 10, 11, 15 and 16. " It is not obvious in Figure 2. The authors might suggest that there was a regional aerosol

concentration pattern but this should not be left to readers to speculate.

6. Line 673: " error (MFE) (Eq. (2)) for the simulations of total number concentration shown on Fig. 2." Equation 2 is not the correct equation for total number concentration. I assume that MFE here is fractional error averaged with time. See more above.

7. Line 671 to 710: These four paragraphs contain some repeated information, for example, “This scaling acts as a surrogate for the parameterization of particle nucleation by materials in addition to the simulated gas-phase NH3, H2SO4, and water, as described in Sect. 2.3.” and “This simulation treats the Arctic MSOA as a 30/70 mix of non- and semi-volatile species.” The paragraphs should be shortened for

8. Line 920: As pointed out by the authors, the timing of this event in observation and model does not match very well. The start of nucleation event is off by about 5 hours based on Figure 6. I wonder how well the model simulates other nucleation and condensation growth events. Can the model capture most of them only with timing off? Does the model miss some of them or predict events that were not observed? Since it is challenging for global models for simulating these events at the correct time, is it possible that Figure 6 was just a coincidence? I agree with the key role for semi-volatile Arctic MSOA during the frequent summertime growth events. Time after nucleation instead of specific date and time can be shown if the modeled nucleation and condensation growth events were not very consistent with observations.

Editorial Comments:

1. Line 46: "Arctic marine SOA (Arctic MSOA)". Can the authors change it to AMSOA as the editor suggest? It was abbreviated as AMSOA later in the manuscript, for example, captions for figure 7 and figure 8.

2. Line 52-56: I would avoid long sentences to reduce potential confusion.

3. Line 180: " Sect. 3 ". Please change to Section 3 for consistency.

4. Line 189: "Figure1".

5. Line 295: "ACDC". No need to abbreviate if not using later.

6. Line 608: Delete the extra return.

7. Line 616: Align the texts in Table 2.

8. Line 379: MSA has been introduced before at Line 196

9. Line 898: Please fix the format.

10. Line 952: Ticks are needed on x-axes in Figure 7.

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