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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 #3

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The manuscript presents evidence that arctic MSOA plays an important role in determining aerosol size distributions and growth in the Canadian Archipelago. GEOS-Chem-TOMAS is used to model measurements conducted at two fixed locations (Alert and Eureka) and on board the CCGS Amundsen. The authors argue that a secondary biogenic marine organic aerosol source is needed to close the gap with filter OC measurements. It is clear from their analysis that adding a constant VOC flux (that subsequently oxidizes and partitions to the particle phase) closes the gap with filter OC measurements. Moreover, the modeled moments of the aerosol size distributions are closer to measurements when adding a constant marine VOC flux. The manuscript is mostly well written, but a rigorous discussion of how the VOC flux magnitude was

estimated is missing. Another drawback of the manuscript is its length. The authors have done a good job at summarizing literature results. The topic of the manuscript is relevant and within the scope of the journal. I recommend publication if the following concerns are properly addressed.

1. The discussion of the constant VOC flux is vague. How was that flux estimated? Was it tuned in the model to get the best agreement with measurements? If so, the authors should explicitly state that this flux is only useful in the context of this study and should not be used as representative of arctic marine VOC emissions given the numerous assumptions made to transform the VOC flux into an SOA mass. Or was this VOC flux bounded by observations? The authors need to clearly articulate these important details in the method section.

2. The authors present evidence that an organic carbon source is missing from their base simulation to accurately model the summer average OC measurements. However, I am not yet entirely convinced that the missing OC mass originates from the oxidation of secondary vapors emitted from the arctic sea only. Primary organic matter contributes significantly to sub-0.1 μm aerosol mass (as included in their analysis). But there is also evidence that primary organics can make 10-20% of the mass of particles with diameter < 0.5 μm (De Leeuw et al. 2011), potentially contributing significantly to total OC mass. The authors need to estimate the implications of primary organic mass at larger particle diameters to strengthen their conclusions.

3. There is no discussion of the uncertainty in reported direct and indirect radiative impacts from including AMSOA. The authors only present values of -0.04 W/m2 for DRE and -0.4 W/m2 for AIE. The error associated with these reported numbers should be included for interpretation of their results.

Specific comments:

Line 383 Please quantify "slightly underpredicted" and "slightly overpredicted", 5%, 10%?

Lines 440-441 The authors need to provide convincing justification for treating BC as externally mixed. This has significant consequences on estimated direct radiative effects.

Line 501 “We identified a fixed arctic MSOA-precursor vapor source flux (Arctic MSOA formed with a mass yield of unity)”. This goes back to my earlier comment of a vague AMSOA discussion. Is the AMSOA precursor VOC flux tuned in the model to best represent measurements? In that case, assuming a mass yield of unity is understandable. Or was this VOC flux estimated from observations? In that case assuming a yield on unity is questionable.

Line 509 Not clear what the difference between the value of 500 μg.m-2.d-1 and the value of 468 μg.m-2.d-1. Please clarify.

Line 510 Missing opening parenthesis.

Line 512 “which are an upper limit on primary organic aerosol contribution”. Not sure how the authors reached that conclusion.

Line 543 “whereas 70% of the arctic MSOA behaves as idealized semi-volatile compounds. . .”. The authors provide adequate justification for using a fraction of semi-volatile organic material greater than 0.5, however, their used value of 0.7 is unfounded. The authors should thoroughly justify this assumption.

Line 554 Is there a reason the authors chose the mean fraction error (MFE) as opposed to cosine similarity? Cosine similarity an intuitive way of comparing size distributions.

Line 631 Mean fractional error (MFE) and not mean fractional bias (MFB) is discussed in section 2.3. Are these the same? If so, please adopt a single notation.

Line 647 “These mass-based comparisons offer confidence that the simulations which include arctic MSOA are reasonable”. Not sure this sentence logically follows from the previous. Please clarify.

Figure 2: Why is it that the model (BASE+TUNDRA+BIRD+100xnuc+AMSOAnv/sv) grossly over predicts measured N10 number concentrations from measurements on Aug 15 and Aug 17, 2016 at Alert?

Figure 2: I believe the comparison of model and measurements of aerosol number concentrations would be easier to interpret if the authors use r2 values instead of MFE values.

Units in Figures 3-5 wrong.

Figures 3-5. The authors should include the one-standard deviation on top of median distributions for at least their BASE+TUNDRA+BIRD+100xnuc+AMSOAns/sv simulation, for all four moments. It is clear from Figures 3-5 that the model captures median measured moments of the size distribution, but the reader has no sense of how well the model performs in predicting variability. The authors should also include a discussion of this in the text.

Line 774 “This pattern is consistent with the hypothesis of an important role for open water in building summertime aerosol size distributions” Is it possible that this is due to a more prominent continental influence with decreased latitudes?

Lines 833-838 Ambiguous sentence. Please reformulate.

Lines 924-926 Please quantify the better agreement with measurements when including AMSOA.

References:

De Leeuw, G., Andreas, E. L., Anguelova, M. D., Fairall, C. W., Lewis, E. R., O’Dowd, C., ... & Schwartz, S. E. (2011). Production flux of sea spray aerosol. Reviews of Geophysics, 49(2).

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