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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 #1

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The paper uses GEOS-Chem-TOMAS to interpret measurements of aerosol composition and size distributions measured at Alert, Eureka, and on a cruise in the Canadian Archipelago. The comparisons of simulated and measured size distributions provide useful information on the potential sources and mechanisms affecting Canadian Arctic Archipelago aerosol. That said, agreement between measured and modeled parameters, which is the basis for the entire paper, is defined by the mean fraction error (MFE) but a metric for a "goodness of agreement" based on MFE is not provided. This omission makes it difficult to assess comparisons for the many simulation types and measured data sets. The model-measurement comparison of aerosol composition, especially OC, has many uncertainties that limit resulting conclusions. Omitting it (Sec-

tion 3.1) would make the paper clearer and more concise. Finally, it is not clear how the simulations developed for the Canadian archipelago are extrapolated to the entire Arctic for the direct and indirect radiative effect calculations. Are emissions varied regionally by extent of open water and location of sea bird colonies? It should be made clearer that the local effects of seabird emissions are accounted for in the pan-Arctic calculations. Additional comments are provided below.

Line 133: What would be included in "differing types of marine biogenic activity"?

Lines 207 – 208: The BMI SEMS is size selecting aerosol, not generating aerosol.

Lines 300 – 302: Yet it is pointed out above (Lines 141 – 144) that growing Aitken mode particles in the Canadian Arctic Archipelago are composed almost entirely of organics. Is this a difference in particle size for the ammonium sulfate vs. organic content of the Aitken mode? Or is this due to regional variability in the composition of Arctic aerosol?

Lines 319 – 322: How does the emission rate of 2.2. ng/m2/s of NH3 from tundra compare to seabird colony emissions?

Lines 342 – 345: What is the assumption that the sub-100 nm sea spray aerosol organics are hydrophobic based on?

Line 378: Please detail the conditions that promote activation of 20 nm particles.

Lines 380 – 381: Please provide older, published references that provide evidence for the contribution of MSA to condensational growth of existing particles.

Lines 390 – 393: Does the "gas-phase precursor" oxidize and lead to new particle formation? While the "SOA tracer" condenses onto existing particles leading to particle growth?

Lines 395 – 396: What potential impacts might result from the exclusion of aqueous phase production of SOA?

Lines 440 – 441: So black carbon is assumed to be externally mixed in the radiative

transfer calculations? How does this assumption impact the results? What about in the determination of the hygroscopicity parameter (Lines 448 – 449)?

Lines 474 – 479: NH3 emissions from sea birds are given in Gg while emissions from tundra are given in ng/m2/s. Is it possible to provide them in the same units so the reader has a sense of the magnitude of the difference in emitted NH3 for these two sources?

Equation 2: Please provide a sense of a "good" MFE, i.e., what values would indicated agreement between measured and simulated values?

Section 3.1: As the authors note, the value of this comparison is limited given the uncertainties in both the measured and simulated OC concentrations. The OC concentrations could be artificially high due to the absorption of gas phase organics on the sampling substrates. The many assumptions that go into the simulated OC concentrations (assumed MSOA precursor vapor source flux, efficiency of MSOA formation, formation rate of newly formed particles, MSOA assumed volatility, etc.) make those concentrations even more uncertain. Furthermore, the time periods of the measurements and the simulations do not overlap. Comparisons for inorganic ions are more constrained as only sources of NH3 are varied. It is shown that the agreement between measured and simulated NH3 concentrations improves when seabird colony and tundra emissions are included. This is not a surprising result. Given all of the knobs to turn, and the uncertainty in both measured and simulated OC concentrations, it is not clear that the "mass-based comparisons offer confidence that the simulations which include Arctic MSOA are reasonable". The authors reinforce this point when they say that "several uncertainties affect the interpretation of the model-measurement 862 comparisons for the quartz filter OC mass concentrations" (Lines 861 – 862). Omitting this section from the paper would make a long paper shorter.

Lines 658 – 660: It is not clear that number concentrations at Alert and on the ship are both elevated at the same time on Aug. 10, 11, and 16. X-axis gridlines might

help clarify. Also – no information is provided (back trajectories, etc.) to explain these results.

Lines 697 – 699: Please put these MFE values (and all others reported in the paper) into some kind of context. For example, Boyland and Russell (2006) state that "a model performance goal has been met when both the mean fractional error (MFE) and the mean fractional bias (MFB) are less than or equal to +50% and $\pm$30%, respectively". Are similar criteria applicable here?

Lines 937 – 941: Please point out that Giamarelou used volatility analysis to determine the composition of the sub-12 nm particles.

Lines 984 – 986: The existence of sub-100 nm organics is due to the choice of a seasalt source function that emphasizes the flux of sub-100 nm particles. This parameterization is in conflict with the canonical sea salt size distribution reported by Lewis and Schwartz (2004) based on number size distributions measured in the marine boundary layer and with sea spray aerosol size distributions generated in a wave channel (Prather et al., 2013). It is not clear why a parameterization would be chosen that produces unrealistic sea spray aerosol size distribution regardless of the motive.

Lines 997 – 1000: What is the mechanism that transports organics but not sulfate from lower latitudes? Figure 7 includes non-marine sources of organics but not sulfate.

Section 3.6: How are the emissions of ammonia from seabirds and tundra and MSOA precursor gases from open water varied regionally in these calculations? The impacts of ammonia emissions from sea birds on particle formation and growth are local in nature. Is this reflected in the calculations?

References:

Lewis, E. R. & Schwartz, S. E. Sea Salt Aerosol Production: Mechanisms, Methods, Measurements, and Models - A Critical Review (American Geophysical Union, 2004).

Prather, K. A. et al. Bringing the ocean into the laboratory to probe the chemical com-

plexity of sea spray aerosol. Proc. Natl Acad. Sci. USA 110, 7550–7555 (2013).

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