We thank the three referees for their constructive comments, which have led to considerable improvements in our manuscript. Below we provide our point-by-point responses (in blue color font) to each referee comment (in black color font). Revised text is highlighted in yellow in the updated manuscript.

Anonymous Referee #1

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.

Response: We agree that information about the interpretation of the MFE would improve the manuscript. Thank you for this constructive comment. The revised text states: "MFE ranges from 0 to +2. Following Boylan and Russell (2006), we treat a MFE value below 0.50 as indicating satisfactory model performance, with the MFE closest to zero indicating the best model performance among the simulation set." (page 16, line 1)

The model-measurement comparison of aerosol composition, especially OC, has many uncertainties that limit resulting conclusions. Omitting it (Section 3.1) would make the paper clearer and more concise.

Response: We agree that removal of Section 3.1 would make the paper clearer and more concise. The associated analysis had many uncertainties and was somewhat redundant with the analysis of the volume distributions. This section has been removed.

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.

Response: We revised the text to clarify how the emissions are varied regionally for the entire Arctic and near Arctic north of 50 °N, depending on the surface type and the location of the seabird colonies. Thank you for pointing out the need for clarification here.

The revised text states: "For simulations with Arctic seabird colony NH_3 emissions, these emissions are implemented following Riddick et al. (2012a) and Riddick et al (2012b) for the entire Arctic and near Arctic north of 50 °N, with modifications and spatial distribution of the colony-specific emissions, as described in Croft et al. (2016b) and Wentworth et al. (2016). The total summertime seabird-colony NH_3 emissions north of 50 °N of 36 Gg are spread uniformly in time between 1 May and 30 September and the point source emissions from the individual colonies are treated as well-mixed within the respective grid box on emission. Our simulations *also implement an NH_3 source from ice- and snow-free tundra for the entire Arctic, with a fixed emission rate of 2.2 ng m^{-2} s^{-1}. Due to knowledge gaps, these emissions are not temperature dependent." (page 11, line 12)*

and

"AMSOA-precursors are emitted in the entire Arctic and near Arctic north of 50° N over open seawater." (page 14, line 17)

and

"These simulations include particle precursor emissions for the entire Arctic as described in Sect. 2.2.2 and 2.2.5." (page 36, line 1)

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

Response: Our intention here was to indicate that there could be differences in the cycles and levels of oceanic biological activity for different marine regions. We removed the word 'types' and revised the text to state: "Due to the spatial and temporal variability, and diversity of organic precursor vapor sources and chemistry, the chemical character of AMSOA is not necessarily the same as MSOA arising from precursors originating in other marine regions. Other areas may have differing levels and cycles of marine biogenic activity (Facchini et al., 2008; Rinaldi et al., 2010) and/or different ship traffic emissions with differing VOCs than natural sources (Endresen et al., 2003)." (page 5, line 2)

Endresen, Ø., Sørgard, E., Sundet, J. K., Dalsøren, S. B., Isaksen, I. S. A., Berglen, T. F. and Gravir, G.: Emission from international sea transportation and environmental impact, J. Geophys. Res., 108(D17), 4560, doi:10.1029/2002JD002898, 2003.

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

Response: Thanks. The revised text states: "…verified for sizing on site using mono-disperse particles of polystyrene latex and of ammonium sulfate size selected with a Brechtel Manufacturing Incorporated Scanning Electrical Mobility Spectrometer." (page 7, line 5)

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?

Response: Lines 300-302 were meant to refer to a different particle size than the size discussed at lines 141-144. The revised text explicitly states the particle size as follows: "Implementation of the ternary scheme is supported by the findings of Giamarelou et al. (2016) that 12 nm-diameter particles in the summertime Arctic are predominantly ammoniated sulfates." (page 10, line 22)

and

"Limited observations indicate that growing Aitken-mode particles with diameters between 50 and 80 nm in the Canadian Arctic Archipelago are composed almost entirely of organics, suggesting a strong role for secondary organics (Tremblay et al., 2018)." (page 5, line 14)

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

Response: The revised text provides this comparison: "For the regions between 60 °W and 100 °W, with varying southward extent, the total implemented summertime tundra NH_3 emissions range from about 1.5- to 7-fold greater than the total summertime seabird-colony emissions, considering 72-90 °N and 50-90 °N, respectively." (page 11, line 29)

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

Response: We added a reference to Facchini et al. (2008) as support for the assumption that the sea spray organics are hydrophobic. However, as noted at the end of this paragraph, there are knowledge gaps about the hygroscopicity.

The updated text states: "This modification was introduced based on measurements indicating that sub-100 nm sea spray particles are composed mostly of hydrophobic organics, whereas larger particles have a progressively more dominant salt component (Facchini et al., 2008; Collins et al., 2013; Prather et al., 2013; Gantt and Meskhidze, 2013; Quinn et al., 2015). However, knowledge gaps remain related to the spatial distribution of sea spray composition and hygroscopicity (Collins et al., 2016)." (page 12, line 19)

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

Response: We added the following description of the conditions that promote activation of 20 nm particles: "In general, particles with diameters of 50 nm or larger activate in our simulations, although observations from the Canadian Arctic Archipelago indicate that particles as small as 20 nm could activate in clean summertime atmospheric layers above 200 m altitude when low concentrations of larger particles (diameters greater than 100 nm) enable relatively high supersaturations (Leaitch et al., 2016)." (page 13, line 20)

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

Response: Thank you pointing out the need for related citation here. We added citations as follows: "MSA that is produced by the DMS-OH-addition channel can contribute to condensational growth of existing particles (Chen et al., 2015; Hoffman et al., 2016; Willis et al., 2016; Hodshire et al., 2018a). In our simulations, MSA contributes to particle condensational growth, but not to particle nucleation." (page 13, line 24)

Chen, H., Ezell, M. J,. Arquero, K. D., Varner, M. E., Dawson, M. L., Gerber, R. B.: Finlayson-Pitts, B. J.: New Particle Formation and Growth from Methanesulfonic Acid, Trimethylamine and Water. Phys. Chem. Chem. Phys. 17, 13699–13709, 2015.

Hoffmann, E.H. Tilgner,A. Schrödner,R. Braüer,P. Wolke, R.; Herrmann, H.: An Advanced Modeling Study on the Impacts and Atmospheric Implications of Multiphase Dimethyl Sulfide Chemistry. Proc. Natl. Acad. Sci. U. S. A., 113, 11776−11781, 2016.

Hodshire, A. L., Campuzano-Jost, P., Kodros, J. K., Croft, B., Nault, B. A., Schroder, J. C., Jimenez, J. L., and Pierce, J. R.: The potential role of methanesulfonic acid (MSA) in aerosol formation and growth and the associated radiative forcings, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-1022, in review, 2018a.

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?

Response: No, the oxidized gas-phase precursor does not contribute to new particle formation in our simulations, but does contribute to growth as it forms the SOA tracer. We revised the text for clarification as follows: "Our simulations include growth of particles by condensation of the oxidized gas-phase SOA precursor, as well as by condensation of gas-phase H_2SO_4 and MSA, but do not allow initial formation of nascent particles by clusters of organic vapors arising from the oxidation of the gas-phase SOA precursor." (page 16, line 23)

and

"The SOA scheme introduces two additional tracers, a gas-phase SOA precursor, and a SOA tracer that immediately condenses on the pre-existing particles. The gas-phase SOA precursor oxidizes to form the immediately condensed SOA tracer on a fixed timescale of 1-day." (page 14, line 7)

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

Response: Exclusion of aqueous phase production of SOA could result in missing pathways of SOA formation in our simulations. It would also change the impact of SOA on the size distribution as it would predominately be added to already CCN-sized particles. This is now explicitly acknowledged in the following revised text: "The model employed for this study does not include explicit aqueous-phase production of SOA, which could further increase the SOA production and change the shape of the particle size distribution." (page 14, line 13)

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)?

Response: In the summertime Arctic changes in transport from lower latitudes sources, more vigorous wet scavenging and few black carbon sources in the region limit the impact of black carbon on the DRE and AIE relative to other seasons. We added the following discussion: "Kodros et al. (2018) found that the Arctic springtime DRE for all aerosol is less negative than the external mixing-state assumption by 0.05 W m^{-2} when constraining by coating thickness of the mixed particles and by 0.19 W m^{-2} when constraining by BC-containing particle number fraction. The radiative-effect sensitivity to the assumed black carbon mixing state is expected to be less for the

Arctic summer than in springtime since changes transport and wet removal, along with low regional sources limit the summertime black carbon concentrations (Xu et al., 2017)." (page 18, line 5)

Kodros, J. K., Hanna, S. J., Bertram, A. K., Leaitch, W. R., Schulz, H., Herber, A. B., Zanatta, M., Burkart, J., Willis, M. D., Abbatt, J. P. D., and Pierce, J. R.: Size-resolved mixing state of black carbon in the Canadian high Arctic and implications for simulated direct radiative effect, Atmos. Chem. Phys., 18, 11345-11361, https://doi.org/10.5194/acp-18-11345-2018, 2018.

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?

Response: We added the following text to give a sense of the magnitude of the difference for these two sources: "For the regions between 60 °W and 100 °W, with varying southward extent, the total implemented summertime tundra NH_3 emissions range from about 1.5- to 7-fold greater than the total summertime seabird-colony emissions, considering 72-90 °N and 50-90 °N, respectively." (page 11, line 29)

As well, we moved the description of the total implemented seabird emissions out of Section 2.3 and into Sect. 2.2.2, preceding the above statement.

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

Response: Thank you for noting this omission. We added the following text to state the MFE values that indicate acceptable agreement between the measurements and simulations: "Following Boylan and Russell (2006), we treat a MFE value below 0.50 as indicating satisfactory model performance, with the MFE closest to zero indicating the best model performance among the simulation set." (page 16, line 2)

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.

Response: We agree that Section 3.1 was of limited value given the uncertainties in the comparison and was also somewhat redundant with the evaluation of the volume distributions. We have removed this section and renumbered the remaining sections.

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.

Response: We have removed these lines because this additional discussion was distracting from the main point of our analysis in this section. Our focus here is intended to be on the evaluation of the total amount for the N4 and N10, as opposed to a focus on the exact timing of the spikes. We chose to present the figure to indicate that the simulation does exhibit bursts of particle formation and to help understanding of our comparison of the time-averaged number concentration. We do not expect that the model will always get the timing right for these events, particularly in the Arctic where the assimilated meteorology may not be as good as at lower latitudes and the model's grid-box mean may not be perfectly representative of the measurement site.

The revised text focuses on the evaluation of the time-averaged total number concentration and states: "These episodic bursts in number concentration are indicative of particle formation and growth events. Figure 2 also shows the time series of coincidently sampled simulated number concentrations for five of the simulations described in Table 1 and Sect. 2.3. The simulations have episodic bursts in total number concentration similar to the observations. However, the simulated grid-box mean total number concentration may not always well represent the measurement site such that simulating the exact timing of the bursts is a greater challenge than simulating the time-averaged magnitude of the number concentration. The simulations may perform better for large-scale (few hundred km) growth events in the Canadian Arctic Archipelago, such as those shown by Tremblay et al. (2018). As an evaluation of the magnitude of the simulated total particle number, we calculated the model-to-measurement fractional bias (FB) using the period-averaged number concentrations for the first 22 days of August (Eq. 5, N=1 and removing absolute value in numerator). The BASE simulation is associated with the greatest FB values for the ship track (-1.93) and Alert (-1.86). The simulations better capture the total particle number when including NH_3 sources from seabird colonies and tundra, with FB values of +0.12 (ship track) and +0.34 (Alert) similar to the findings of Croft et al. (2016b)." (page 21, line 18)

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 ±30%, respectively". Are similar criteria applicable here?

Response: We agree that information was needed to interpret the MFE values. We added this information at page 16, line 2, as quoted above

and

"We consider a MFB between -0.3 and +0.3 indicates satisfactory model performance." (page 16, line 9)

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

Response: We revised the text to state: "For example, Giamarelou et al. (2016) found using volatility analysis that 12 nm-diameter particles in the Svalbard region were primarily ammoniated sulfates, pointing to the importance of particle formation by ternary nucleation of gas-phase NH_3*,* H_2SO_4 *and water and initial growth by low volatility sulfur-containing vapors." (page 32, line 18)*

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.

Response: Our focus here was to demonstrate that the Mårtensson parameterization, considered as being extremely favorable to ultrafine sea spray production, was not anywhere close to producing the number of ultrafine particles needed to match the observations. We revised the text to acknowledge that the parameterization exceeds the canonical distribution of Lewis and Schwartz (2004) and to explain that we use this parameterization to support our analysis that the missing aerosol mass is not likely to arise from sea spray.

The revised text states: "We use the Mårtensson et al. (2003) parameterization, which in comparison with other parameterizations yields among the largest sub-100 nm diameter sea spray particle production fluxes for temperatures near 273 K (de Leeuw et al. 2011, Fig. 9). As well, for particle diameters from 100 nm to 500 nm, the Mårtensson et al. (2003) parameterization exceeds the uncertainty ranges identified by Lewis and Schwartz (2004), thus the role of primary marine emissions is likely over estimated by this parameterization for this size range. There is evidence that primary organics could contribute 10-20% of the mass of particles with diameters less than 500 nm (de Leeuw et al., 2011). Thus, a portion of the mass fraction labeled as sea salt on Fig. 8 for sizes 100 to 500 nm could be organics that are misrepresented as sea salt. However as the sea-spray fraction in Fig. 8 indicates, this potential primary-organic contribution is considerably smaller than the AMSOA mass fraction. As a result any missing POA for 100 nm to 500 nm diameter particles is likely not sufficient to yield a match for the volume distributions shown in Figs. 3-5." (page 34, line 13)

and

"The simulated contribution of primary organics of sea-spray origin to sub-100 nm particle mass fractions was largest for the ship track simulation in the marine boundary layer, with mass fractions approaching 20% for particles with diameters around 10 nm to 20 nm, and was likely over estimated by the sea spray parameterization." (page 39, line 31)

Lewis, E. R., and Schwartz, S. E.: Sea Salt Aerosol Production: Mechanisms, Methods, *Measurements and Models—A Critical Review, Geophys. Monogr. Ser., vol. 152, 413 pp., AGU, Washington, D. C., 2004.*

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

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.

Response: We did not intend to indicate that there is no transport of sulphate from lower latitudes. The sulphate transport from lower latitudes is included in the 'anthropogenic and biomass burning sulfate' category on Fig. 8. We added the following text to clarify: "Sulfate transported from lower latitudes is included in the anthropogenic and biomass-burning category (shown in orange shading on Fig. 8)". (page 35, line 1)

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

Response: Yes, to a certain extent the regional variation and local nature of the emissions is included in the calculations, but future work is needed to refine these source functions.

The revised text related to seabird-colony and tundra NH_3 emissions states: "For simulations with Arctic seabird colony NH_3 emissions, these emissions are implemented following Riddick et al. (2012a) and Riddick et al (2012b) for the entire Arctic and near Arctic north of 50 °N, with modifications and spatial distribution of the colony-specific emissions, as described in Croft et al. (2016b) and Wentworth et al. (2016). The total summertime seabird-colony NH_3 emissions north of 50 °N of 36 Gg are spread uniformly in time between 1 May and 30 September and the point source emissions from the individual colonies are treated as well-mixed within the respective grid box on emission. Our simulations also implement an NH_3 source from ice- and snow-free tundra for the entire Arctic, with a fixed emission rate of 2.2 ng m^{-2} s^{-1}. Due to knowledge gaps, these emissions are not temperature dependent." (page 11, line 12)

and as related to AMSOA:

"AMSOA-precursors are emitted in the entire Arctic and near Arctic north of 50° N over open seawater. Like other biogenic SOA sources, these vapors are emitted with a 50/50 split between the gas-phase precursor and a vapour that is immediately condensed. Given knowledge gaps,

these AMSOA precursor emissions are not dependent on other parameters such as temperature or marine biologic activity." (page 14, line 17)

and as related to our radiative calculations:

"We caution that several uncertainties are associated with our quantification of the DRE and AIE. The sources for AMSOA precursor vapors, and also for the seabird-colony and tundra ammonia are uncertain. As well, there are uncertainties in the DRE and AIE due to the simulated cloud fields, surface albedo and particle size distributions in the absence of AMSOA. Future work is needed to improve the emissions parameterizations for Arctic particle precursors. Our simulations include AMSOA and tundra NH_3 emissions that vary spatially with land type, but additional factors such as temperature and biological activity could also control these emissions and could be investigated in future studies. Further work is also needed to better understand the source and nature of AMSOA-precursor vapors. Additionally, work to examine the impact of a sub-grid plume processing parameterization for the seabird colony emissions could be beneficial. These effects could change the spatial distribution and magnitudes of the radiative effects attributed to AMSOA, and reduce associated uncertainty. As a result of these uncertainties and knowledge gaps, we consider the presented values for the DRE and AIE as an indication of the order of magnitude that AMSOA may contribute to the DRE and AIE. However, we view these calculations as identification that the impact of condensational growth by AMSOA is expected to be relevant for the Arctic climate." (page 37, line 13)

Anonymous Referee #2

GEOS-Chem-TOMAS chemical transport model with size-resolved aerosol micro- physics 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?

Response: Thank you for pointing out the need for clarification about the methodology for determining the VOC flux. We agree that a clearer presentation of these details would strengthen the manuscript.

The revised text states: "The top-down estimate of the flux (500 µg m^{-2} d^{-1}; north of 50° N) for our simulations is adopted by tuning the VOC flux in a simulation set (with the seabird-colony and tundra NH_3 emissions) until a MFE below 0.5 was achieved for the three measurement platforms. Further details on the related results are presented in Sect. 3. To put the implemented flux in context, this value exceeds either the estimated isoprene flux from a north temperate deep lake (Steinke et al., 2018) or tundra VOC emissions (Lindwall et al., 2016) by a factor of about 5-10. As this flux was tuned specifically to yield model-measurement agreement for our study, it should not be over-interpreted as being fully representative of Arctic marine VOC emissions. Future measurements of marine VOC concentration, fluxes, and volatility are needed for a bottom-up estimate of the marine SOA-precursor source flux." (page 16, line 12)

As well, the paragraphs preceding this text in Sect. 2.2.5 now provide details about the equations used for the calculations and the parameterizations related to AMSOA.

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.

Response: The text was revised as follows to explain why the spatial variation depends only on surface type in our simulations: "AMSOA-precursors are emitted in the entire Arctic and near Arctic north of 50° N over open seawater. Like other biogenic SOA sources, these vapors are emitted with a 50/50 split between the gas-phase precursor and a vapour that is immediately condensed. Given knowledge gaps, these AMSOA precursor emissions are not dependent on other parameters such as temperature or marine biologic activity." (page 14, line 17)

and

"Future measurements of marine VOC concentration, fluxes, and volatility are needed for a bottom-up estimate of the marine SOA-precursor source flux." (page 16, line 20)

This additional future work is needed for development of parameterizations with greater temporal and spatial variation than the one we have implemented in this study.

"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.

Response: Thank you for this constructive suggestion. The text was revised to clarify our methodology as quoted in the preceding author comments and additionally the revised text states: 'This fixed flux of 500 µg m^{-2} d^{-1} of AMSOA-precursor vapors (with a yield of unity) emitted from open seawater in the Arctic and near Arctic (north of 50° N) was determined by tuning the *simulated flux to achieve model-measurement agreement for the first four moments of the aerosol size distributions at the three measurements platforms in the Canadian Arctic Archipelago." (page 38, line 31)*

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.

Response: We agree that presentation of these definitions in the methods section would clarify the discussion. We moved the definitions to Sect 2.2.5 of the methods. (page 15, line 7)

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.

Response: In response to this comment, we added text to clarify that: "We include the four moments to yield a more complete evaluation that gives equal weighting to aerosol number, integrated diameter, surface area and volume." (page 16, line 4)

As well, we added the following discussion of each the four moments in the context of the new Fig. 6, which shows fractional bias for each of the four moments separately and replaces Table 4: "Considering each moment separately, Fig. 6 shows the model-measurement FB (defined in Sect. 2.2.5) for the first four moments of the size distributions, for the three measurement platforms and all simulations. Among the moments, the 0^{th} moment (number) is most sensitive to the addition of the seabird-colony and tundra NH_3 emissions, whereas the 3^{rd} moment (volume) shows the least sensitivity. The 1^{st} and 2^{nd} moments show an intermediate sensitivity to the NH_3 source. The volume distribution shows the highest sensitivity to the AMSOA source with relatively less sensitivity towards the lower moments. Figures 3-5 show that AMSOA contributes more than half of the simulated total volume distribution. Figure 6 shows that the combination of NH_3, nucleation scaling, and mixed volatility AMSOA is required to simultaneously bring all four moments within the range of satisfactory model performance at all three measurement platforms (simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv/sv), excepting a small exceedance for Eureka's 2^{nd} moment. The volume moment provides a year-matched constraint on the total aerosol mass concentrations in our simulations. Simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv/sv has the lowest volume distribution FB for both Alert (+0.07) and the ship track (+0.01), while for Eureka two simulations had the lowest FB, BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv (-0.06) and BASE+TUNDRA+BIRDS+100xnuc+2xAMSOAnv/sv (+0.06). For all three sites, implementation of AMSOA reduced the volume fractional bias within the bounds of satisfactory model performance relative to an otherwise similar simulation without AMSOA. These general improvements of the simulations with the addition of AMSOA offers support for a key role of marine biogenic emissions in shaping the Arctic size distributions." (page 29, line 22)

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.

Response: We agree that our analysis would be more complete with a consideration of the fractional error for the separate moments. We added the new Fig. 6, which shows the fraction bias for the separate moments and added related discussion as quoted above. We chose to present fractional bias here as opposed to fractional error to indicate whether the individual moments under or over predict the measured values.

The boundaries of the summation on equation 2 should be from i=0 to i=N-1

Response: Corrected (now renumbered as Eq. 5). Thank you. (page 15, line 26)

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.

Response: Corrected in Figs. 3, 4 and 5 – thank you noticing these typos.

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.

Response: We agree that the word coastal is vague here. We removed the word throughout and the revised text states: 'Our simulations suggest that condensation of secondary organic aerosol (SOA) from precursor vapors emitted in the Arctic and near Arctic marine (ice-free seawater) regions plays a key role in particle growth events that shape the aerosol size distributions observed at Alert (82.5° N, 62.3° W), Eureka (80.1° N, 86.4° W), and along a NETCARE ship track within the Archipelago." *(page 2, line 8)*

and

"In this study, the terminology AMSOA indicates SOA formed from any organic precursor vapors emitted from ice-free seawater north of 50° N, excluding methane sulfonic acid (MSA), which we treat as a separate aerosol component, for consistency with most filter-based aerosol species mass measurements." (page 4, line 28)

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

Response: We added the following text: "Terrestrial volatile organic compounds (VOCs) from tundra and lakes are an additional biogenic influence (Potosnak et al., 2013; Lindwall et al., 2016; Steinke et al., 2018)." (page 4, line 16)

and

“To put the implemented flux in context, this value exceeds either the estimated isoprene flux from a north temperate deep lake (Steinke et al., 2018) or tundra VOC emissions (Lindwall et al., 2016) by a factor of about 5-10.” (page 16, line 15)

Potosnak, M. J., Baker, B. M., LeStourgeon, L., Disher, S. M., Griffin, K. L., Bret-Harte, M. S. and Starr, G.: Isoprene emissions from a tundra ecosystem, Biogeosci., doi:10.5194/bg-10-871-2013, 2013.

Lindwall, F., Schollert, M., Michelsen, A., Blok, D., and Rinnan, R.: Fourfold higher tundra volatile emissions due to arctic summer warming, J. Geophys. Res. Biogeosci., 121, 895–902, doi:10.1002/ 2015JG003295, 2016.

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.

Response: We revised the text to clarify as follows: “Due to the spatial and temporal variability, and diversity of organic precursor vapor sources and chemistry, the chemical character of AMSOA is not necessarily the same as MSOA arising from precursors originating in other marine regions. Other areas may have differing levels and cycles of marine biogenic activity (Facchini et al., 2008; Rinaldi et al., 2010) and/or different ship traffic emissions with differing VOCs than natural sources (Endresen et al., 2003).“ (page 5, line 2)

Endresen, Ø., Sørgard, E., Sundet, J. K., Dalsøren, S. B., Isaksen, I. S. A., Berglen, T. F. and Gravir, G.: Emission from international sea transportation and environmental impact, J. Geophys. Res., 108(D17), 4560, doi:10.1029/2002JD002898, 2003.

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

Response: We added the following clarification: “Standalone CPC measurements were not available at Eureka.” (page 21, line 16)

5. Line 658: "Interestingly, at several times the elevated number concentrations occur at both Alert and at the ship, such as on August3,8,9,10,11,15and16. "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.

Response: We removed these lines as our focus here is on the evaluation of the total number concentration as opposed to exact timing.

The following revised discussion of Fig. 2 draws our focus to total number: “The measurement time series shows episodic bursts of particle number concentration greater than 500 cm^{-3}*. These episodic bursts in number concentration are indicative of particle formation and growth events. Figure 2 also shows the time series of coincidently sampled simulated number concentrations for five of the simulations described in Table 1 and Sect. 2.3. The simulations have episodic bursts in* *total number concentration similar to the observations. However, the simulated grid-box mean total number concentration may not always well represent the measurement site such that simulating the exact timing of the bursts is a greater challenge than simulating the time-averaged magnitude of the number concentration. The simulations may perform better for large-scale (few hundred km) growth events in the Canadian Arctic Archipelago, such as those shown by Tremblay et al. (2018). As an evaluation of the magnitude of the simulated total particle number, we calculated the model-to-measurement fractional bias (FB) using the period-averaged number concentrations for the first 22 days of August (Eq. 5, N=1 and removing absolute value in numerator). The BASE simulation is associated with the greatest FB values for the ship track (-1.93) and Alert (-1.86). The simulations better capture the total particle number when including NH_3 sources from seabird colonies and tundra, with FB values of +0.12 (ship track) and +0.34 (Alert) similar to the findings of Croft et al. (2016b)." (page 21, line 17)*

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.

Response: We added the following details to describe the calculation, and we now present fractional bias as opposed to fractional error to indicate whether the simulations over or under predict the measurements: "As an evaluation of the magnitude of the simulated total particle number, we calculated the model-to-measurement fractional bias (FB) using the period-averaged number concentrations for the first 22 days of August (Eq. 5, N=1 and removing absolute value in numerator)." (page 21, line 27)

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

Response: We agree that these details were redundant with earlier discussion in the methodology. We removed the associated sentences.

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.

Response: The model does capture many of the nucleation events. Times when the total particle number exceeds about 500 cm^{-3} (as shown on Fig. 2) are generally times of nucleation and growth events. We added a sentence to identify that: "These episodic bursts in number concentration are indicative of particle formation and growth events." (page 21, line 18)

Figure 2 shows that the model does capture many of the events, and a result we do not consider that the simulation of the event shown in Fig. 7 was a coincidence. However, there are events that are missed or incorrectly timed by the simulations. The model is likely to perform better when the event are of a greater regional extent. Tremblay et al. (2018) presented the possibility for wide-scale regional events in the Canadian Arctic Archipelago, as quoted in the revised text above.

Following your suggestion, we have revised the horizontal scale on Fig. 7 (now renumbered from 6) to show hours of growth event. Due to the 10 nm cut-off for the SMPS, we are unable to present time after nucleation. However, the update to hours of growth event removes the focus from the exact date and time, unlike the previous label. Thank you for this suggestion. (page 32, line 1)

Editorial comments:

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

Response: We have changed terminology to AMSOA throughout the manuscript.

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

Response: We have divided the sentence into shorter sentences. Updated text reads: "Particle growth due to the condensable organic vapor flux contributes strongly (30-50%) to the simulated summertime-mean number of particles with diameters larger than 20 nm in the study region. This growth couples with ternary particle nucleation (sulfuric acid, ammonia, and water vapor) and biogenic sulfate condensation to account for more than 90% of this simulated particle number, a strong biogenic influence." (page 2, line 19)

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

Response: We retained the abbreviation Sect. 3 because our understanding of the guidelines was that this is the ACP convention for use of this word in mid-sentence. Please advise us if our understanding of the guidelines is not correct here.

4. Line 189: "Figure1"

Response: Space inserted before the 1. (page 6, line 30)

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

Response: Abbreviation is removed.

6. Line 608: Delete the extra return.

Response: This section (original Sect. 3.1) has been removed, following the suggestion of referee 1.

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

Response: This table was removed with the removal of the original Sect. 3.1.

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

Response: Repetitive acronym description for MSA removed. Now defined only at page 4, line 29.

9. Line 898: Please fix the format.

Response: Early line break removed.

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

Response: X-axis tick marks are moved to outside for visibility (renumbered as Fig. 8) (page 35, line 3)

Anonymous Referee #3

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.

Response: The revised text includes the following details about how the VOC flux magnitude was estimated: "The top-down estimate of the flux (500 µg m^{-2} d^{-1}; north of 50° N) for our simulations is adopted by tuning the VOC flux in a simulation set (with the seabird-colony and tundra NH_3 emissions) until a MFE below 0.5 was achieved for the three measurement platforms. Further details on the related results are presented in Sect. 3. To put the implemented flux in context, this value exceeds either the estimated isoprene flux from a north temperate deep lake (Steinke et al., 2018) or tundra VOC emissions (Lindwall et al., 2016) by a factor of about 5-10. As this flux was tuned specifically to yield model-measurement agreement for our study, it should not be over-interpreted as being fully representative of Arctic marine VOC emissions. Future measurements of marine VOC concentration, fluxes, and volatility are needed for a bottom-up estimate of the marine SOA-precursor source flux." (page 16, line 12)

As well, further details about the calculations and AMSOA-related parameterizations are now located in the paragraphs preceding the above-quoted text.

We removed the original Sect. 3.1, which was redundant with the analysis of the volume distributions. This change has reduced the length of the manuscript. In place of that discussion, we added the following: "We also conducted comparisons of mass concentrations with filter measurements at Alert (not shown) and all simulations with seabird and tundra NH_3 *matched the sulfate+ammonium+MSA mass within 20% (and contributions of other measured species, e.g. nitrate, were minor) so organic aerosol mass was likely the most uncertain species." (page 26, line 14).*

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.

Response: Thank you pointing out this omission in our methods section. We have added a detailed description of our methodology in Sect. 2.2.5. The revised text explicitly states that the flux was estimated by tuning as opposed to being bounded by VOC flux observations, as quoted above and as follows: "Given this uncertainty and the lack of a marine SOA source in our standard simulations, we introduced and tuned a simulated fixed AMSOA-precursor vapor source flux (AMSOA formed with a mass yield of unity) from the ice-free seawater in the Arctic and near Arctic (north of 50° N) for simulations with seabird and tundra NH_3*. We tuned to a satisfactory model-measurement for the first four moments of the aerosol size distributions for Alert, Eureka and the ship track." (page 14, line 31)*

We also include the definitions for the four moments in the revised Sect. 2.2.5 and state: "Following Boylan and Russell (2006), we consider a MFE value below 0.50 indicates satisfactory model performance, with the MFE closest to zero indicating the best model performance among the simulation set. We include the four moments to yield a complete evaluation that gives equal weighting to aerosol number, integrated diameter, surface area and volume. The absolute value in the MFE numerator prevents cancellations of over predictions and under predictions between the moments." (page 16, line 2)

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.

Response: We agree that our simulations could be missing primary sea spray organics for the diameters between 100 nm and 500 nm. We added the following text to estimate the implications of POA mass at these larger particle diameters (100-500 nm): "There is evidence *that primary organics could contribute 10-20% of the mass of particles with diameters less than 500 nm (de Leeuw et al., 2011). Thus, a portion of the mass fraction labeled as sea salt on Fig. 8 for sizes 100 to 500 nm could be organics that are misrepresented as sea salt. However as the sea-spray fraction in Fig. 8 indicates, this potential primary-organic contribution is considerably smaller than the AMSOA mass fraction. As a result any missing POA for 100 nm to 500 nm diameter particles is likely not sufficient to yield a match for the volume distributions shown in Figs. 3-5." (page 34, line 20)*

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

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.

Response: There are many dimensions to the DRE uncertainties. We added the following discussion of the uncertainties to help in the interpretation of the presented direct and indirect radiative: "We caution that several uncertainties are associated with our quantification of the DRE and AIE. The sources for AMSOA precursor vapors, and also for the seabird-colony and tundra ammonia are uncertain. As well, there are uncertainties in the DRE and AIE due to the simulated cloud fields, surface albedo and particle size distributions in the absence of AMSOA. Future work is needed to improve the emissions parameterizations for Arctic particle precursors. Our simulations include AMSOA and tundra NH_3 emissions that vary spatially with land type, but additional factors such as temperature and biological activity could also control these emissions and could be investigated in future studies. Further work is also needed to better understand the source and nature of AMSOA-precursor vapors. Additionally, work to examine the impact of a sub-grid plume processing parameterization for the seabird colony emissions could be beneficial. These effects could change the spatial distribution and magnitudes of the radiative effects attributed to AMSOA, and reduce associated uncertainty. As a result of these uncertainties and knowledge gaps, we consider the presented values for the DRE and AIE as an indication of the order of magnitude that AMSOA may contribute to the DRE and AIE. However, we view these calculations as identification that the impact of condensational growth by AMSOA is expected to be relevant for the Arctic climate." (page 37, line 13)

We also added the following clarification in the abstract: "AMSOA accounts for about half of the simulated particle surface area and volume distributions in the summertime Canadian Arctic Archipelago, with climate-relevant simulated summertime pan-Arctic-mean top-of-the-atmosphere aerosol direct (-0.04 W m^{-2}) and cloud-albedo indirect (-0.4 W m^{-2}) radiative effects, which due to uncertainties are viewed as an order of magnitude estimate." (page 2, line 31)

Specific comments:

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

Response: While there is a consensus in the literature on the sign of the effect, the magnitude is not yet well quantified. We revised the text to state: "In this study, we did not include additional *chemistry related to production of dimethylsulfoxide (DMSO), which could increases the yield of MSA and reduce sulfate concentrations (Breider et al., 2014; Hoffman et al., 2016). Future studies are needed to quantify the impact of multi-phase DMS chemistry." (page 13, line 28)*

Hoffmann, E.H., Tilgner,A., Schrödner,R., Bräuer,P., Wolke, R., Herrmann, H.: An Advanced Modeling Study on the Impacts and Atmospheric Implications of Multiphase Dimethyl Sulfide Chemistry. Proc. Natl. Acad. Sci. U. S. A., 113, 11776−11781, 2016.

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.

Response: We agree that the DRE is sensitive to this common assumption of BC external mixing. We added the following text to indicate the potential impact of this assumption: "Kodros et al. (2018) found that the Arctic springtime DRE for all aerosol is less negative than the external mixing-state assumption by 0.05 W m^{-2} when constraining by coating thickness of the mixed particles and by 0.19 W m^{-2} when constraining by BC-containing particle number fraction. The radiative-effect sensitivity to the assumed black carbon mixing state is expected to be less for the Arctic summer than in springtime since changes transport and wet removal, along with low regional sources limit the summertime black carbon concentrations (Xu et al., 2017)." (page 18, line 5)

Kodros, J. K., Hanna, S. J., Bertram, A. K., Leaitch, W. R., Schulz, H., Herber, A. B., Zanatta, M., Burkart, J., Willis, M. D., Abbatt, J. P. D., and Pierce, J. R.: Size-resolved mixing state of black carbon in the Canadian high Arctic and implications for simulated direct radiative effect, Atmos. Chem. Phys., 18, 11345-11361, https://doi.org/10.5194/acp-18-11345-2018, 2018.

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.

Response: Yes, the AMSOA precursor is tuned in the model to best represented the aerosol measurements. The revised text now explicitly states: "Given this uncertainty and the lack of a marine SOA source in our standard simulations, we introduced and tuned a simulated fixed AMSOA-precursor vapor source flux (AMSOA formed with a mass yield of unity) from the ice-free seawater in the Arctic and near Arctic (north of 50° N) for simulations with seabird and tundra NH_3. We tuned to a satisfactory model-measurement for the first four moments of the aerosol size distributions for Alert, Eureka and the ship track." (page 14, line 31)

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.

Response: To avoid confusion we use only the value of 500 µg m^{-2} d^{-1} throughout the revised text. This value is the implemented flux rounded to one significant figure. The difference between the two values has a negligible impact on our results. The revised text states: "The top-down estimate of the flux (500 µg m^{-2} d^{-1}; north of 50° N) for our simulations is adopted by the tuning *the VOC flux in a simulation set (with the seabird-colony and tundra NH_3 emissions) until a MFE below 0.5 was achieved for all three measurement platforms." (page 16, line 12)*

Line 510 Missing opening parenthesis.

Response: Revised text has opening parentheses. (page 16, line 12)

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

Response: We removed this statement.

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.

Response: We added the following text as justification of the volatility assumption: "We also conducted simulations with the assumption that 100% of the AMSOA behaved as semi-volatile compounds and found excessively suppressed growth of the sub-40 nm particles relative to observed size distributions. Thus for our simulations, we settled on 70% as a reasonable intermediate between 50% and 100% (the range from Riipinen et al., 2011) of the AMSOA being semi-volatile." (page 20, line 29)

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.

Response: We chose this metric because our focus was on evaluation of the overall magnitude rather than the exact timing of the events. The following revised text clarifies that our focus is on the evaluation of magnitude: "As an evaluation of the magnitude of the simulated total particle number, we calculated the model-to-measurement fractional bias (FB) using the period-averaged number concentrations for the first 22 days of August (Eq. 5, N=1 and removing absolute value in numerator)." (page 21, line 27)

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.

Response: The definitions for MFE and MFB are now provided in Sect. 2.2.5 and the revised text clarifies that relative to MFE: "Mean fractional bias (MFB) is similarly defined, but without the absolute value in the numerator and ranges from -2 to +2. We consider a MFB between -0.3 and +0.3 indicates satisfactory model performance." (page 16, line 7)

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.

Response: We have removed these organic mass comparisons as being redundant with the analysis of the volume distributions. The original Sect. 3.1 was removed. This also makes the overall presentation more concise.

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?

Response There is a gap in the measurements on Aug 17, 2016, so we are not able to determine if there is a strong over prediction. However, there does appear to be a factor-of-3 over prediction on Aug 15, 2016. This could be related to sub-grid scale effects such that the simulated grid box mean was not a close match to the conditions exactly at the measurement site. This is particularly an issue when looking at a time series as opposed to monthly mean values.

We revised the text to provide the following related discussion: "The simulations have episodic bursts in total number concentration similar to the observations. However, the simulated grid-box mean total number concentration may not always well represent the measurement site such that simulating the exact timing of the bursts is a greater challenge than simulating the time-averaged magnitude of the number concentration. The simulations may perform better for large-scale (few hundred km) growth events in the Canadian Arctic Archipelago, such as those shown by Tremblay et al. (2018)." (page 21, line 21)

As well, this section was revised to focus on evaluation of the period-mean number concentration as opposed to the exact timing, as quoted in the above responses.

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.

Response: R2 values would bring the focus more towards the timing of the events. As a result, we chose to use the fractional bias since our focus here is on the evaluation of the magnitude of the total number concentration over the time period as opposed to the exact timing.

Units in Figures 3-5 wrong.

Response: Units on the vertical axis of the panels in Figs. 3-5 are now revised. Thank you for noting the need for correction here. As well, we have changed the color font on these figures to avoid confusion between the two shades of green used in the original version.

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.

Response: In response to this comment, we added the 20th and 80th percentiles for simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOnv/sv in Figs. 3-5. We also revised the text to state "As *shown on Figs. 3-5, this simulation also has a range of variability between the 20th and 80th percentiles that is similar to that of the measurements for all four moments." (page 29, line 8)*

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?

Response: We agree and revised the sentence to state: "This pattern is consistent with the hypothesis of an important role for open water in building summertime Arctic size distributions (Heintzenberg et al., 2015; Willis et al., 2017; Dall´Osto et al., 2018a), along *with the contribution of the more prominent continental influence at lower latitudes." (page 24, line 15)*

Lines 833-838 Ambiguous sentence. Please reformulate.

Response: We reformulated this sentence to read: "This inter-site difference in the AMSOA precursor source flux magnitude that yields a MFE of 0.1 suggests development of a parameterization for the precursors' volatility-dependent spatial distribution could be of benefit. Such a parameterization could also help to better capture the increase in the magnitude of the mode for the number, diameter, area and volume distributions between Alert and Eureka. However, our current parameterizations do capture the larger magnitude of the mode value for all four moments for the ship track relative to those for Alert and Eureka (simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv/sv)." (page 29, line 1)

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).

Response: We removed this sentence as being redundant with the following quantitative discussion of the growth rate in the updated Sect. 3.4: "Collins et al. (2017) and Burkart et al. (2017a) also report growth rates of about 2-4 nm h^{-1} for similar size aerosols during other growth events observed from the CCGS Amundsen during the 2016 cruise."

and

"The top right panel shows that without the source of AMSOA (simulation BASE+TUNDRA+BIRDS+100xnuc), the nascent particles do not exhibit sufficient growth beyond about 15 nm by condensation of H_2SO_4 and MSA alone. The bottom left panel shows that with the source of non-volatile AMSOA for simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv, there is growth from about 10 nm to about 40 nm over 8 hours, a growth rate that is slightly faster than observed for this event and faster than reported by Burkart et al. (2017a). The bottom right panel of Fig. 7 shows for simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv/sv, particles grow from about 10 nm to 20 nm over about 8 hours, which is slightly slower than the

observed rate and slower than the simulation BASE+TUNDRA+BIRDS+100xnuc+AMSOAnv, which assumed non-volatile AMSOA." (page 31, line 11)