Author's response to reviewer's comments

We thank the two reviewers for their constructive comments and suggestions, which have helped to improve the manuscript. Before responding to the specific individual comments from the reviewers, we note that, we have made substantial changes to the manuscript based on the reviewer comments. Specifically, we changed the focus from looking at the relationship between sea spray aerosol particle hygroscopicity and ATOFMS cluster-type fractions to one between hygroscopicity and organic matter volume fractions (ε_{org}). The OM volume fractions were estimated from the AMS organic matter/PM_1 mass fractions that were presented in the original manuscript. In the original manuscript, we did not use the ε_{org} quantitatively, as there are concerns regarding the detection efficiency of the AMS for these marine derived organics as particles containing a large fraction of sea salt have a higher susceptibility to particle bounce and organic matter contained in these particles may be inefficiently vaporized (as is suggested by the results presented by Frossard et al. (2014)). That said, in one of the references mentioned by Reviewer #2 (Ovadnevaite et al. (2012)), it was determined that sea salt aerosol had a collection efficiency (*CE*) in the AMS of 0.25. We have therefore now corrected the AMS organic matter/PM_1 mass fractions using a *CE* of 0.25, and the mass fractions were converted to ε_{org} assuming a density of 1 g/cm^3 for organic matter. The resulting ε_{org} are therefore relatively uncertain in terms of absolute magnitude, but the trends with time should be reasonably robust under the assumption that the *CE* did not change substantially across the measurement campaign. A thorough discussion of the uncertainties in ε_{org} as estimated from AMS organic matter/PM_1 mass fractions and the details for calculating ε_{org} has been added to section 2.2.1.

> "It is important to note that while the temporal trends of the AMS NR-OM/PM_1 fractions are likely reflective of the general behavior, the absolute values are more difficult to quantify because NR-OM associated with particles containing high sea salt fractions may not be vaporized efficiently by the AMS due to the refractory nature of sea salt (Frossard et al., 2014) and to the susceptibility of SSA particles to particle "bounce" in the AMS. Consequently, the SSA particles, including the NR-OM component, are detected with a collection efficiency (*CE*) lower than unity (Frossard et al., 2014). One previous study (Ovadnevaite et al., 2012) determined the *CE* value for organic-free sea salt sampled when RH < 70% is approximately 0.25. However, they also note that the *CE* is potentially instrument dependent, and further may not be applicable to the organic fraction in sea spray particles due to differences in ionization efficiency (which is a component of the overall *CE*) (Ovadnevaite et al., 2012). It is also possible that the *CE* differs between particles that have differing relative amounts of OM and sea salt. Despite such uncertainties in quantification of NR-OM by the AMS for sea spray particles, the NR-OM mass concentrations for the sampled SSA particles were determined in this study assuming *CE* = 0.25. The measured NR-OM mass concentrations were used to calculate NR-OM volume concentrations assuming a density (ρ) of 1.0 g/cm^3. A value of 1.0 g/cm^3 for ρ_{OM} is consistent with that of fatty acids ($\rho < 1$ g/cm^3), which are a significant fraction of marine-derived OM (Mochida et al., 2002; Cochran et al., 2016). However, this value serves as a lower bound for ρ_{OM} because OM with higher densities, such as sugars ($\rho \sim 1.7$ g/cm^3), have also been observed in SSA (Quinn et al., 2015). The NR-OM volume

> fractions of SSA (ε_{org}) were calculated as the ratio between the observed NR-OM volume concentrations and the integrated total particle volume concentrations from the size distribution measurements. Given the use of a lower-limit value for ρ_{OM} the ε_{org} are likely upper limits (not accounting for uncertainty in the assumed *CE*)."

The CE-corrected ε_{org} are now used as the primary compositional metric for understanding both the depression in *GF*(85%) values relative to inorganic sea salt and their temporal variability. Figures 3 and 5 have been updated to show the CE-corrected ε_{org} values. Discussion regarding the temporal variability in and absolute magnitude of the CE-corrected ε_{org} has been added.

> "The NR-OM volume fractions of SSA varied from 0.29 to 0.50 throughout the course of the indoor MART microcosm experiment (Figure 3). The observation of such large ε_{org} values is consistent with the substantial depressions in the *GF*(85%) values relative to pure, inorganic sea salt (2.1). The temporal variation in the ε_{org} was generally similar to that of the *GF*(85%) values, with smaller *GF*(85%) values corresponding to larger ε_{org} values, although the peak in ε_{org} is somewhat sharper than the dip in the *GF*(85%). The inverse relationship between the *GF*(85%) and ε_{org} is consistent with organic compounds being less hygroscopic than sea salt."

The original Figure 4, which showed the relationship between the *GF*(85%) values and the ATOFMS non-sea salt cluster fractions, has been replaced. The new Fig. 4 now shows the relationship between *GF*(85%) and the CE-corrected ε_{org}. We now use the Zdanovskii-Stokes-Robinson (ZSR) mixing rules to estimate a *GF*(85%) value for the organic matter component of the SSA particles specifically. The fitting procedure is described on Page 16 Lines 12-20 in the updated manuscript.

There has also been an evolution in our understanding of the ATOFMS clusters determined for nascent sea spray since the manuscript was originally submitted. A portion of the non-sea salt (sodium-depleted) clusters can be explained by the incomplete ionization of sea salt particles (Sultana et al., In Prep.). This change in our understanding of ATOFMS cluster types further supports our decision to use the CE-corrected ε_{org} from the AMS in place of ATOFMS cluster types to understand the dependence of the *GF*(85%) on variations in particle composition. A brief discussion of this new understanding regarding the ATOFMS clusters for nascent sea spray has been added to the manuscript in the methods section.

> "It is important to note, however, that dried SSA particles sampled by the ATOFMS can be spatially chemically heterogeneous, with shells depleted in Na and rich in Mg, K, and Ca (Ault et al., 2013). Thus, some fraction of the particles identified as having Mg or SSOC type spectra may be partially explained by the incomplete ionization of sea salt particles (Sultana et al., In Prep.). However, variations in the thickness of this Na-depleted shell likely reflect variations in the total particle organic content. Therefore, increases in the fraction of SSOC or Mg type mass spectra generated suggest a net increase in SSA particle organic content."

We made other changes, where deemed appropriate and added additional figures and tables as supplemental materials. Our point-to-point response to both reviewers follows below.

Key: Black = Reviewer, Blue = Response

Response to Reviewer #1

The paper by Forestieri et al. presents results from two microcosm experiments on the properties of sea spray aerosols, focusing on their hygroscopic and optical properties, as a function of the seawater composition. The seawater composition was artificially modified in the microcosms by the addition of nutrients. The authors infer an average hygroscopic growth factor (HGF) for the whole sea spray aerosol population, from the measurement of the aerosol extinction enhancement due to the uptake of water vapour at 85% humidity. Results show a decrease of the HGF by 10 to 19% relative to pure inorganic sea salt. The authors then infer an average chemical composition from the HGF with the hypothesis that the organic fraction is hydrophobic. No linear link between the increase of Chl-a levels and the change in aerosol chemical composition (organic content, mainly) was observed. The study of the impact of the presence of organic matter in primary sea spray on its optical properties through the effect of a decreased water uptake has never been investigated in the past to my knowledge. Whether this has an important impact or not is important. In this view this is a very valuable study. However, the measurement methodology relay on several hypothesis and approximations that could be better justified (see detailed comments), and the article is more focusing on inferring the organic fraction of primary organic aerosol than on evaluating this impact, which could be more emphasised (the impact on scattering is only mentioned in the conclusion as a range from 10 to 35% for 85% humidities). I would have expected a time series of the extinction (wet and dry) in order to directly evaluate the impact of a phytoplanktonic bloom on the optical properties of sea salt aerosol. I recommend publication after major revisions.

Regarding the reviewers comment as to presentation of a time-series of extinction (wet and dry), we note that for this study the absolute values of extinction are not nearly as important as the relative values between the humidified and dried extinction (i.e. f(RH) = wet extinction/dry extinction). Although MARTs produce particles with size distributions that are similar to particles produced from breaking waves in the ocean, the absolute particle concentrations (and therefore the absolute scattering) is different than what would be observed in the ambient marine atmosphere, as the absolute values depend on the sample flow rates, MART size, plunging frequency, etc. (Stokes et al., 2013). Thus, a focus on the extensive properties (e.g. dry and wet scattering) is, in our opinion, not as important as a focus on the intensive properties (e.g. f(RH)). In the original manuscript, we presented a time-series only growth factor values that were derived from the f(RH) measurements and the size distribution measurements, not the f(RH) measurements themselves. However, to address the reviewers concern, we have now added to the supplemental material a time-series of measured wet and dry extinction measurements and of the associated f(RH).

Regarding discussion of the limited evaluation of the impact of organic material on scattering, we intentionally kept this discussion "simple". The reason for this is that the ultimate impacts depend on not only variability in the organic fraction, but also in real variations in relative humidity fields. Thus, a comprehensive evaluation would likely require assessment within a climate model, which is outside the scope of this work. To address the reviewers suggest that we emphasize the potential impact to a greater extent, we have added the following sentences to the conclusions:

> "Regardless, the results presented here suggest that OM in SSA particles may have a non-negligible, yet variable impact on the light scattering by SSA particles in the ambient atmosphere. Most likely, the simulated cooling effect of SSA particles due to aerosol-radiation interactions (i.e. the "direct effect") would be decreased relative to the assumption that all SSA behaves as sea salt."

Therefore, f(RH) (= wet scattering/dry scattering), which is an intensive parameter, is used to assess radiative impacts. A figure comparing the f(RH) of particles observed in this study to the f(RH) expected for pure, inorganic sea salt calculated using average size distributions for both microcosm experiments has been added to the supplementary material (Figure S8).

Page 4, lines 6-7 : "SSA particles sampled from the MARTs are primary, since the average residence time in the MARTs is much shorter than the time scale required for secondary processing of SSA particles (e.g. heterogeneous gas-phase reactions) (Lee et al., 2015)." What is the residence time in the microcosm headspace, what are briefly the results from Lee et al. 2015 to support this hypothesis ? How can the absence of any photochemical reactions producing condensing organic matter be excluded?

Oxidants which accompany secondary processes, such sulfates and nitrates, were not present in the ATOFMS spectra. This is as expected as zero air (particle, ozone, and volatile organic species free) was used to feed the MART headspace, so secondary processes should be highly minimized even over extended periods. Hydroxyl radicals should not be generated by the lamps used here, as the higher-energy UV radiation needed to photolyze water would be filtered by the acrylic chamber walls. The residence time in the headspace is particle size dependent (as discussed in Stokes et al., 2013). At the flow rate used here, for particles in the size range 400-600 nm the e-folding lifetime was 11 minutes and in the size range 1-2 microns was 8.5 minutes.

Page 5 lines 13_18 : "The same seawater as used in the indoor MART was added to a separate MART and sampled immediately after collection and before nutrient addition. However, the resulting particle size distribution from this MART differed substantially from those measured from the indoor MART, with a much greater contribution of large particles. Thus, the measurements from this separate MART are not directly comparable to the measurements from the indoor MART and are not considered further" Is there any explanation for this ? Could it be that the same difference in original size distribution (before enrichement) was observed in the outdoor experiment ?

The large supermicron mode observed during in the pre-nutrient period may be due to differences in MART conditions, such as water level or sampling tube length, but we cannot entirely rule out that it is some difference due to the nutrient addition. Unlike the indoor MART, the outdoor MART was not sampled prior to enrichment, and thus we cannot directly address the reviewers' second question. In separate (unpublished) MART experiments, strong differences in the size distributions before/after nutrient addition are not observed, suggesting that in this particular case the size distribution difference was driven by external factors (which resulted from the somewhat more complex experimental setup during the IMPACTS study due to the large number of instruments involved).

Page 6, line 10 : "Group 1 sampled for 1.5 h, group 2 sampled for 2 h, and Group 3 for sampled 1 h each day that sampling was conducted" Was sampling always performed in this order ? Can there be a bias due to the position of the sampling period during the day ? Has this been tested ?

On all days, except for 7/9, the instruments sampled at the same time each day. It is possible that the composition differed when the cavity-ringdown (CRD) sampled versus when the ATOFMS sampled, but this has not been systematically tested. In fact, some differences are to be expected given that the observations suggest changes in composition and f(RH) from one day to the next. However, the correlation between the ATOFMS organic markers and the CRD growth factors indicates that compositional changes were gradual and the composition of particles sampled by both instruments was consistent on a given day, even though the ATOFMS sampled 9 hours after the CRD.

Page 6, line 25 : "The SEMS and APS distributions were merged using the SEMS distribution up to 1 µm and the (dp,m equivalent) APS distribution at larger diameters." How did the two instrument compare on their common size range ? Why was the APS preferred over the APS on the 1-1.9 micron size range ?

A comparison between the SEMS and APS suggests that the SEMS under-counted particles with mobility diameters > 1 micron. This was characterized during separate experiments in which substantial fractions of supermicron particles were sampled. At just above 1100 nm, the SEMS undercounted by ~20% and at 1.5 µm, the SEMS undercounted by ~90%. We suspect that this difference resulted from the SEMS not being optimized in these experiments to transmit larger particles, and thus internal losses increased as size increased. It is for this reason that we chose to use the merged distribution, as opposed to the SEMS distribution by itself. However, in the size region that is most relevant to the experiments considered here, namely below ~ 1 micron, there was no substantial difference between the SEMS and merged SEMS + APS distributions. Since the fraction of supermicron particles was very small for these MART studies the impact of SEMS vs. APS differences at larger sizes is negligible.

Page 7, line 2 : "Light absorption by the SSA particles was negligible, and thus extinction is equal to scattering, i.e. bext = bsca." Was this assumption validated ? Can Brown carbon contribute the SSA absorption ?

The SSA absorption was measured by a UC Davis photoacoustic spectrometer (Lack et al., 2011; Cappa et al., 2012) and the observed absorption was 0 Mm^{-1} (within uncertainty), whereas the measured extinction was ~250 Mm^{-1}.

Page 9, lines 16-17: "Unlike f(RH), GF values are independent of the dry particle size, and thus only depend on composition" This is only true for larger particles, the smaller the particles the highest the kelvin effect is. Maybe it is useful to argue that this hypothesis is true for the sizes of particles relevant here.

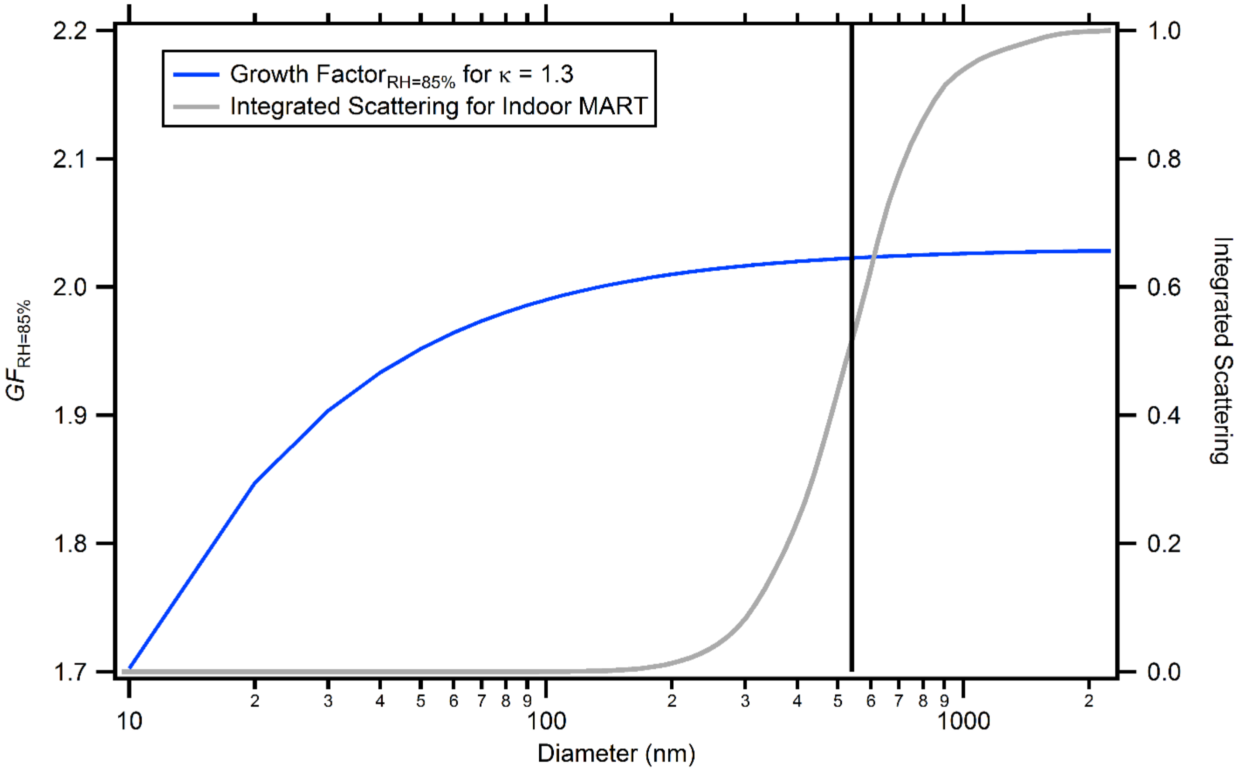
The reviewer raises an important point. We have therefore calculated theoretical growth factors as a function of particle diameter using equation 11 from Petters and Kreidenweis (2007), assuming 85% and assuming κ = 1.3. The relevant equation is:

$$\frac{RH}{\exp\left(\frac{A}{D_{dry}} \cdot GF\right)} = \frac{GF^3 - 1}{GF^3 - (1 - \kappa)}$$

The Kelvin effect is inherent in the *A* term in the above equation since:

$$A = \frac{4 \cdot \sigma \cdot MW_{H2O}}{RT \cdot \rho_{H2O}}$$

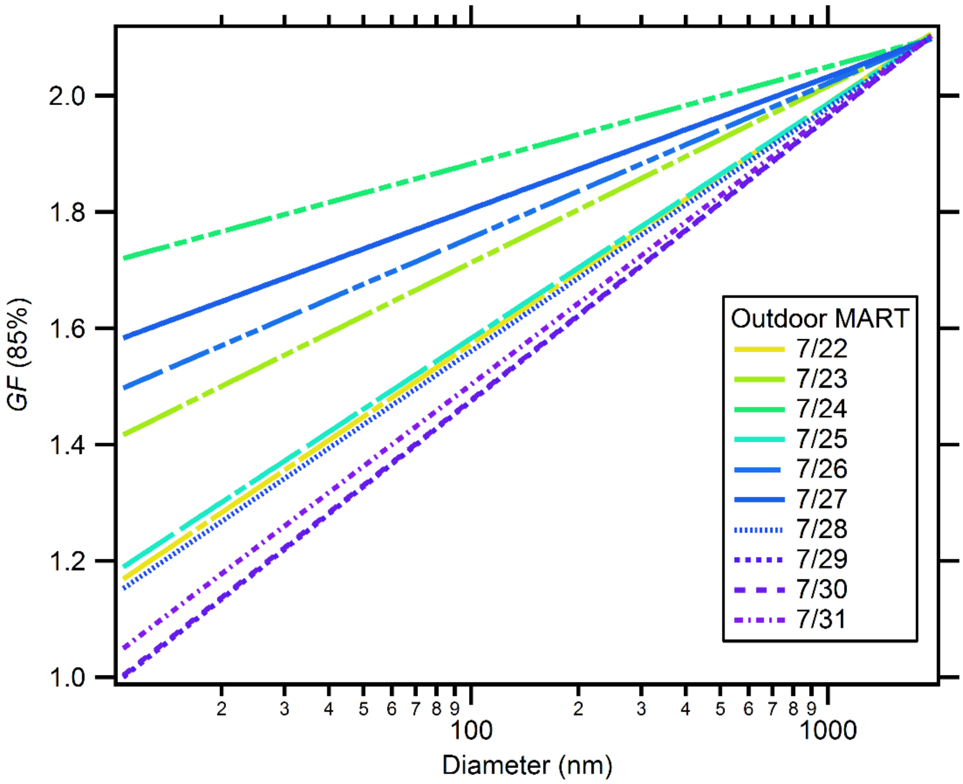
where σ = surface tension, MW_{H2O} is the molecular weight of water and ρ_{H2O} is the density of water. Results of these calculations are shown in the figure below. The black line indicates where the median (50%) integrated scattering occurred in our experiments. The *GF*(85%) values change by only ~1% over the range of sizes that contributed substantially to the observed scattering and thus changes in the measured GF values should only depend on composition. (To the extent that composition depends on size, size will play a role. But the Kelvin effect can be ignored for these experiments.)



Pages 9 and 10 : calculation of GF(RH) : the underlying hypothesis for such a iterative calculation is that the chemical composition of the aerosol is homogeneous over the whole range of sizes (independent of the particle diameter). Figure S6 does not show this. How does this impact the results ?

This simplification has no major impact on our results. The iterative approach used in our analysis can be modified to allow GF values to vary with size. If a functional form is assumed then one can allow the parameters describing this relationship to vary, as opposed to a single average value, to match the observed f(RH) values. We have actually made this model modification, assuming that GF values are a linear function of $\log(d_p)$, increasing as d_p increases. Specifically, we assumed that $GF(d_p) = a + b*\log(d_p)$ with the added constraint that GF(1800 nm) = 2.1, i.e. that these particles are pure sodium chloride and that all values must be >= 1. It was fully possible to adjust the a and b coefficients for each day to match the calculated and observed f(RH) values. One then finds that the lines (and a and b coefficients) vary day-to-day in a manner consistent with the derived variations in the optically-weighted GF values. The results of this approach are shown in the figure below for the Outdoor MART. The reason that this approach was not adopted as the default approach is that we did not want to introduce another assumption regarding the form of the *GF* vs. diameter relationship, and thus opted for the simpler (albeit, potentially less physically realistic) approach. We now discuss this further in the manuscript in Section 2.2.2 and have added the following text.

> "It is assumed that the growth factors are size independent, namely that $GF_x = GF$ for all d_p. Thus, this method retrieves an effective, optically-weighted *GF* value that explains the observed influence of water uptake on light scattering for the sampled size distribution. An alternative approach was considered in which the GF_x were assumed to vary with size, specifically as $GF_x = 2.1 - b\ (\log(1.8\ \mu m) - \log(d_{p,m}))$, and where the value of b was allowed to vary during the optical closure, with the condition that $GF_x \geq 1$. (This expression assumes that particles with $d_{p,m} = 1.8$ have a $GF_x = 2.1$, i.e. that of NaCl. The GF_x decrease as size decreases.) The derived *b* values exhibit a similar temporal dependence as the derived optically-weighted *GF* values. The general conclusions reached in this study are therefore independent of the assumptions made regarding the size-dependent behavior of GF_x. Thus, rather than introducing an uncertain functional form, the simpler assumption (namely, size-independent GF_x values) is used here."



Page 10, lines 1ç-21 : "The measured GF(85%) for NaCl was 2.09 +/- 0.03 and for (NH4)2SO4 was 1.59 +/- 0.05, which compare very well with literature values of ~2.1 for NaCl (Cruz and Pandis, 2000; Laskina et al., 2015; Hansson et al., 1998) and ~1.55 for ammonium sulfate (Laskina et al., 2015; Wise et al., 2003)." The literature values should be reported for a given aerosol size (or size range).

The aerosol size ranges have been added to the manuscript.

Page 12 , lines 13_14 : "Uncertainty in the assumed RI value for the dry particles may explain a small fraction (<5%) of the difference." How was this assessed ? Has the chemical analysis of the aerosol been used to estimate the real RI? All hypothesis for possible discrepancies addressed in this paragraph should be detailed in the methodology section (or at least in the supplementary material). An overall uncertainty on the HGF retrievals procedure should be calculated and compared to the measured HGF variability and consequent Org frac variability, so the reader can be convinced that the measured time variations are real. The uncertainty on the calculation method should be less than the 10 to 19% decrease in HGF for the results of the paper to be significant.

The uncertainties in the measured f(RH), relative humidity, refractive index, and diameter all contribute to uncertainty in GF retrieval. A fuller discussion of the contributions of uncertainty in each term to the uncertainty in the retrieved GF values, and how this was determined, has been added to the supplementary material. The precision of the GF values for each experimental day ranged from 1.7 to 2.2% (determined as the standard deviation of the individual measurements over each sampling period), which is far less than the 5 to 15% (in the updated manuscript the GF(85%) for pure sea salt is assumed to be 2.1 instead of 2.2) decrease in GF relative to pure, inorganic sea salt.

Page 12, lines 21-24 : "We have tested the sensitivity of the retrieval method to an 8% increase in the particle diameters. The retrieved GF values are increased by a marginal amount (0.015-0.03) when the diameters are increased, and thus such potential sizing uncertainty does not affect the main conclusions presented here" Does this mean that a particle diameter increase of 8% was actually applied to the data set ?

This statement refers only to the sensitivity tests. In the sensitivity test, the particle diameters were all increased by 8%, and then the retrieval was performed. No adjustment was applied to the "data set" beyond this sensitivity test.

Page 17, lines 10-12 : "By using a campaign-average scaling factor, it is implicitly assumed that the actual variations in fSS,0.75µm are captured by fSS,avg, which seems reasonable given the general constancy of the size distributions over the course of each of the microcosm experiments, c.f. Fig. 2." Why would the relative stability of the size distribution shown on fig 2 insure that the non-sea salt content of the aerosol (shown to increase in the course of the experiment) evolves uniformly with size ?

The ATOFMS sea salt fractions are no longer used quantitatively, so we no longer adjust the sea salt fraction to a vacuum aerodynamic diameter 0.75 µm.

Page 17, lines 19-24 : ". . .organic volume fraction (εi) of 0.56 – 0.88 for these particle types if it is assumed that volume mixing rules apply (i.e. the Zdanovskii-StokesRobinson mixing rules (Stokes and Robinson, 1966)). Since the non-SS values range from 0.53 – 0.74, if it is assumed that the SS-type and non-SS particle types have similar size distributions, then the implied ensemble average εi would be about 0.33 – 0.52." I understand that the first time that εi is used it refers to the fraction of hydrophobic material in the non-SS fraction, while the second time it is used it refers to the fraction of hydrophobic material in the overall aerosol. If this is right, the same terminology should not be used for both.

We now have an estimate for the organic volume fraction (ε_{org}) of the SSA particles and no longer estimate the non-SS organic fraction. Thus, there is no longer a need to change the terminology.

Page 18, lines 9-12: "Both DOC and heterotrophic bacteria concentrations increased as the bloom progressed until they stabilized around the point when Chl-a concentrations had returned approximately to their pre-bloom levels, with DOC concentrations ranging from 200 to 300 µM C and heterotrophic bacteria concentrations from 1 x 106 to a peak of 1.7 x 107 mL-1 (Figure S5B)." Are those values realistic for natural seawaters ?

As noted on Page 13 Lines 5-6 in the original manuscript, the peak DOC range is somewhat larger than values typically observed for blooms in the ocean, which are only ~130 - 250 µM C (Kirchman et al., 1991; Norrman et al., 1995). Regarding heterotrophic bacteria, the range of heterotrophic bacteria concentrations in surface ocean waters range from around 1 to 5x10^6 cells per mL (Li, 1998), which is comparable to the bacteria concentrations observed in the MART, although the peak MART concentrations exceed those in the ocean. This has been added to the manuscript.

Technical comments
Page 5, line 8 : mesocosm or microcosm ?

”Mesocosm” has been replaced with “microcosm” to keep terminology consistent.

Figure 3 (B) : description of Org not in the figure text. "the reported uncertainties for all properties is 1 sigma: : : "should be "the reported standard deviations for all properties is 1 sigma: : :" as those are not uncertainties on the measurements

This has been updated in the manuscript.

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