Reviewer #2 Comments

(Authors Response in italics, excerpts from text in bold)

This document intends to provide information about sea salt aerosol sourced from the southern hemisphere. Justification for the study is given as a lack of southern hemisphere measurements and underestimation of low-level cloud cover. The measurements come from a 23-day ship voyage off the coast of New Zealand. Several chemical speciation measurements were taken along with VH-TDMA (water) and UFO-TDMA (ethanol) measurements. The author uses statistical analysis of the many variables to survey for correlations. Some of those correlations do not have legitimate causation. The document ends by trying to resolve the issues using OCEANFILMS (vs ZSR). The amount of work is significant and clearly represents measurements from the southern hemisphere. Some changes should be made prior to full publication.

The authors thank the reviewer for their detailed review, helpful comments/suggestions and broadly positive review.

The authors have addressed concerns surrounding causation between lognormal modes and composition, for example the apportionment of organics to lognormal modes has since been removed in response to the reviewers comments.

Another change to the manuscript worth noting is the removal of the NaSO4 hydrate component from the computation of fio, which has a subsequent impact on the OVF computed from volatility measurements. This component was removed because it has been shown that the contribution to volatility from NaSO4 hydrates very small (Rasmussen et al. 2017).

Page 3: Line 16: Does the sea salt samples (from manufactured sources) not create salt hydrates?

Yes the laboratory sea salt and the inorganic sea salt component of sea spray both contain hydrates and this needs to be accounted for.

Page 3: Line 18: OVF not defined.

Defined earlier "Exceptions include Quinn et al. (2014), who observed an organic volume fraction (OVF) of up to 0.8 using CCN measurements"

Page 4: Line 14: HGF not defined

The text has been changed and the first use of HGF is now defined.

Page 5: Line 8: beta 660 backscatter, pCO2, and DMSsw not previously defined.

The text has been simplified.

> Section 2.1 SOAP voyage
>
> **"seawater parameters (Chl-a, dimethyl sulfide, and carbon dioxide concentrations)"**

Page 5: Line 10: DMS not previously defined

The text has been changed and the first use of DMS is now defined.

Page 7: Line 4: "Subsequent to heating the SSA was exposed to 90% RH and the hygroscopic growth factor was measured." Please insert a comma or adjust to better display the subordinate clause.

The authors agree and the text has been simplified.

"After heating the SSA hygroscopic growth factor at 90% RH was measured."

Page 7 Line 4: RH not previously defined.

The authors agree and the first use of RH has been defined.

Page8: DOC not previously defined

The authors agree and the first use of DOC has been defined.

Page10: Were the inverted volatility scans used as inputs to the Gysel inversion routine to calculate growth factor as insinuated by equation 4? After performing volatility, the particles shrink some. How is this shrink, prior to hygroscopic growth, represented using the Gysel inversion?

Volatility measurements and hygroscopicity measurements were inverted separately. A single VGF mode was used to correct HGF for volatility. A comment on this has been added

2.3 Data analysis.

"Volatility measurements and hygroscopicity measurements were inverted separately and a single VGF mode was used to correct HGF for volatility."

Page 10: Line 14 and 15: recompose sentence to read that sulfate mass was calculated from S, not all inorganics from S measurements.

The authors agree and the sentence has been changed.

Page 11, Line 8
"The inorganic mass (IM) was computed as the sum of Na, Mg, SO4, Cl, K, Ca, Zn, Br and Sr. The measured S mass was used to calculate the SO4 mass, all S was assumed to be in the form of SO4."

Page 11: Why would a salt hydrate have a growth factor of 1?

The hydrate, water component has a GF of 1. The anhydrous salt has a higher HGF than the hydrated salt. This was a way of explicitly including the hydrate volume fraction (which varies between samples) in the ZSR assumption.

Page 13: I am assuming the number of size distribution modes correlates with the four sintered glass filters. Is this true? If not, please dispel the misconception.

No not necessarily – for example Fuentes et al. 2010 used a single porosity sintered glass frit and fitted 4 lognormal modes. This has been pointed out in the text.

Page 14 Line 2

"The measured size distributions were broken up into four log-normal modes characterised by geometric mean diameters ranging from 33 to 320 nm, as seen in Fig. 4. This is consistent with the number of <u>lognormal</u> modes fitted by Fuentes et al. 2010 and is not a direct result of the use of multiple glass filters in this study."

Page 14: Although this may be a little over critical, the natural sea water normalized concentration is missing 1% in Table 1.

A rounding issue, an extra decimal point has been included for clarity.

Page 14: Line 17 and 18 and Figure 5: how do we know that the non-volatiles (OVFNV) are organic? If you have a proxy for total organic mass and a proxy for semi-volatile mass, wouldn't the involatile be the difference between the two using assumptions for density?

The linear model provides a calculated total organic volume fraction and semi-volatile organic volume fraction (for preselected 50 nm SSA). The non-volatility/low volatility component is the difference between these two values.

Page 15: The hygroscopic growth measurements are based on number population (as described in Section 3.4). The volume fraction (used in volatility) is based on both number and diameter. (unless everything is singly charged, the two numbers do not correlate). FYI, 80% of the population is singly charged for this situation. The averaged sampled population from mode 3 is 17% by number and 27% by volume. See table below. These calculations are based on three items: the non-diffusing DMA transfer function (

Stolzenburg and McMurry 2008) and your reported DMA 1 settings, the charging fraction as defined by (Wiedensohler 1988), and the reported size distributions in Table 1. In the numbers below, I have multiplied the normalized population numbers in Table 1 by 100,000 for clarity.

Page 16: Feel free to use the numbers above to try to resolve any issues in error in volume fraction. I should note that the numbers above are based on your published average settings and will not be representative of an individual scan.

Thank you kindly for your helpful information.

This apportionment of the organic fraction (based on volatility) to the lognormal modes has since been removed in the absence of any size resolved composition measurements (particularly around the accumulation mode), in response to questions from reviewer #1.

Page 18-Figure 7 caption: "Stars in bottom right plot represent the mean EF from TEM-EDS measurements of SSA generated from laboratory seawater, dotted error bars show standard deviation in the mean." – I do not see any stars in the panel.

"Stars" changed to "Triangles" to reflect the figure.

Page 19: OM not previously defined.

First use of OM now defined in the text

Page 19 line 5: tot should be to

Text changed to fix typo

Page 19 bottom paragraph: I noticed that the number fractions in the growth factor distribution roughly correlate with charges: the first charge constitutes 80% of the population. How do you know that the lower growth mode isn't the singly charged particles?

The number fraction of the first HGF mode varies between the seawater samples, ranging from 0.47 to 1, which isn't consistent with a (stable) charge fraction.

The doubly charged particles would have a diameter of approximately 75 nm. The HGF difference between 50nm and 75 nm SSA should be reasonably small.

Page 20 line 1 through 5: This could be true (using the above tables), but it is likely more complicated. The first size distribution mode could also create the higher growth mode, by theory. I understand that there was statistical correlation, but I find no causal relationship for size distribution 3 being the only size distribution mode related to the second growth factor mode.

The authors agree that the relationship between the lognormal modes and HGF modes/composition is not certain. The text has been amended to reference the possibility of different lognormal mode compositions, but acknowledging the uncertainty/limitations in this study.

> *Page 21, Line 7*
>
> **"The fraction of the second HGF mode at 50 nm correlated with the proportion of lognormal mode 3 (R2 of 0.39, p-value < 0.01, and slope of 0:87_0:3). This suggests that the lognormal modes may have different composition and/or morphology, which has previously been observed for nascent SSA (Collins et al., 2013), however in the absence of size resolved compositional measurements further conclusions are not possible."**

Page 20 Figure 8: Is it possible to keep the ordinate of panels (a) and (b) the same to show the increase in HGF due to heating?

Yes, the authors agree that this would be clearer and the figure has been changed to make the y-axis scales aligned.

Page 28 Line 22 and 23: I do not see any evidence in this work that shows a discrepancy between modeled CCN in the atmosphere and actual CCN measurements during the study. Use of the word "improve" seems inappropriate given the lack of evidence. A verb similar to "change" or "alter" seems more appropriate.

This text has subsequently been changed in response to comments from reviewer #1 and no longer uses this wording.