Answers to Reviewer2's comments

We thank reviewer 2 for his/her constructive comments that we feel we have well addressed. Reviewer's comments are in black while our answers are in blue.

In this manuscript, the authors conducted a study during a research cruise in the Southern Ocean using a sea spray simulation chamber to generate nascent sea spray aerosol. The authors varied the seawater temperature in the chamber while steaming across different water masses to investigate the impact of seawater temperature and the biogeochemical state of the ocean on the sea spray aerosol flux.

As the authors allude to in their introduction, this research question has received significant attention in the scientific community. Although it is increasingly evident that seawater temperature impacts sea spray aerosol flux, there are still differences in both the magnitude and direction of the relationship between seawater temperature and sea spray aerosol flux, which may be due to variations in experimental approaches and differences in water chemistry and biology across different studies.

Although the study did not resolve the longstanding issue of the differences in the relationship between seawater temperature and sea spray aerosol flux found in the literature, my major criticism of the work is not the lack of scientific significance. Rather, the quality of the manuscript is compromised due to a general lack of attention to detail. The methods section omits critical details, rendering it impossible to evaluate the authors' findings. Furthermore, the work is poorly presented, making it difficult to understand the authors' intended message. In addition, the study overlooks numerous uncertainties, making it impossible to verify some of the authors' claims. In summary, the study feels rushed and fails to do justice to the authors' effort in collecting the dataset.

Therefore, I strongly recommend that the authors revise the manuscript substantially and focus on presenting the methods and results more clearly. As a result, I am afraid that I can only recommend rejecting the article in its current form. Below I outline in more detail the major issues I have identified with the manuscript.

Methods were only briefly presented in the original manuscript because they were already extensively detailed in previous published works (Schwier et al. 2015, Schwier et al. 2017, Trueblood et al. 2021, Freney et al. 2021) and for N100 fluxes in Sellegri et al. 2021. The experiment is also described in Sellegri et al. 2023. This last reference is cited in the present paper and publicly available in free access to read (https://journals.ametsoc.org/view/journals/bams/aop/BAMS-D-21-0063.1/BAMS-D-21-0063.1.xml), in case the reviewer needed more details than provided in the present work.

1. Schwier, A. N., C. Rose, E. Asmi, A.M. Ebling, W.M. Landing, S. Marro, M.-L. Pedrotti, A. Sallon, F. Iuculano, S. Agusti, A. Tsiola, P. Pitta, J. Louis, C. Guieu, F. Gazeau, and **K. Sellegri**

Primary marine aerosol emissions from the Mediterranean Sea during pre-bloom and oligotrophic conditions: correlations to seawater chlorophyll-a from a mesocosm study, Atmo.Chem. Phys., 15, 7961-7976, doi:10.5194/acp-15-7961-2015, 2015

2. Schwier A.N. , **K. Sellegri**" S. Mas, B. Charrière, J. Pey, C. Rose, B. Temime-Roussel, D. Parin, J.-L. Jaffrezo, D. Picard, R. Sempéré, N. Marchand and B. D'Anna, "Primary marine aerosol physical and chemical emissions during a nutriment enrichment experiment in mesocosms of the Mediterranean Sea, Atmos. Chem. Phys., 17, 14645-14660, https://doi.org/10.5194/acp-17-14645-2017, 2017
3. **Sellegri Karine**, Alessia Nicosia, Evelyn Freney, Julia Uitz, Melilotus Thyssen, Gérald Grégori, Anja Engel, Birthe Zäncker, Nils Haëntjens, Sébastien Mas, David Picard, Alexia Saint-Macary, Maija Peltola, Clémence Rose, Jonathan Trueblood, Dominique Lefevre, Barbara D'Anna, Karine Desboeuf, Nicholas Meskhidze, Cécile Guieu and Cliff S. Law Surface ocean microbiota determine cloud precursors, *Sci Rep* **11,** 281 https://doi.org/10.1038/s41598-020-78097-5, 2021
4. Jonathan V. Trueblood, Alesia Nicosia, Anja Engel, Birthe Zäncker, Matteo Rinaldi, Evelyn Freney, Melilotus Thyssen, Ingrid Obernosterer, Julie Dinasquet, Franco Belosi, Antonio Tovar-Sánchez, Araceli Rodriguez-Romero, Gianni Santachiara, Cécile Guieu, and **Karine Sellegri**, A Two-Component Parameterization of Marine Ice Nucleating Particles Based on Seawater Biology and Sea Spray Aerosol Measurements in the Mediterranean Sea, Atmos. Chem. Phys., 21, 4659–4676, https://doi.org/10.5194/acp-21-4659-2021, 2021
5. Evelyn Freney, **Karine Sellegri**, Alessia Nicosia, Jonathan T. Trueblood, Matteo Rinaldi, Leah R. Williams, André S. H. Prévôt, Melilotus Thyssen, Gérald Grégori, Nils Haëntjens, Julie Dinasquet, Ingrid Obernosterer, France Van-Wambeke, Anja Engel, Birthe Zäncker, Karine Desboeufs, Eija Asmi, Hilka Timmonen, and Cécile Guieu, Mediterranean nascent sea spray organic aerosol and relationships with seawater biogeochemistry, Atmos. Chem. Phys., 21, 10625–10641, https://doi.org/10.5194/acp-21-10625-2021, 2021

Major issues

The authors must provide a clear description of the sea spray simulation system they used in the methods section, as it is increasingly evident from the literature that the scale of laboratory systems used to generate nascent sea spray impacts the relationship between the number and size of aerosols generated as seawater temperature changes.

If the reviewer has the knowledge of a study intercomparing plunging jet systems and concluding their size is a main factor influencing the shape of the SSA size distribution as a function of temperature, we would like to read and cite this. We believe that the main factors are the jet orifice size, the jet flowrate, and the distance to the seawater surface (these impact the amount of air entrained in the seawater) rather than size. Operating the system in a continuous or disruptive manner, and the distance between two jets is probably also influencing the way bubbles are prematurely broken or not. In our system bubbles did not have any contact with the tank's walls, and provide SSA size distributions very similar to those observed in the natural clean marine sector of the region investigated (Sellegri et al. 2023).

In the current version of the manuscript, the authors have only referred to another paper (Sellegri et al. 2022) which is not included in the reference list. Given this, I assume that the system used is the same as that described in Schwier et al. (2015), …

The paper Sellegri et al. 2022 took a very long time to be processed - it was supposed to be published and accessible before the present manuscript – however, it is now available as Sellegri et al. 2023. It is clearly stated in the original text that: "Sea spray was continuously generated with a plunging jet system, as described in detail in Sellegri et al. (2022) and previously used in Schwier et al. 2015 and 2017, Trueblood et al. 2021, Freney et al. 2021 and Sellegri et al. 2021.", with the last 4 references accessible in ACP. Sellegri et al. 2023 is in now accessible and included in the reference list.

… which is relatively small compared to other systems used for simulating sea spray aerosol generation in the laboratory.

Although our system is several times smaller than systems such as those described in Dall'Osto et al. (2022), this reduces the seawater residence time, so limiting changes in biology or sedimentation of large phytoplankton species that occur in larger chambers (Dall'Osto et al. 2022). In addition, a smaller system also eliminates potential gas-phase reactions with air that may occur in other larger systems. We now clarify these advantages in the Method section:

"Given the jet flowrates of 1.2 LPM, the relatively small seawater volume results in low residence time (4 min), so preventing changes in biology or sedimentation of large species that occur in larger chambers (Dall'Osto et al. 2022). The small dimensions of our system also correspond to a short residence time of air in the headspace (12s), also preventing potential gas-phase reactions with lab air."

The water depth in the system used by Schwier et al. (2015) was only 10 cm deep, which may have resulted in increased interaction between bubbles and the chamber walls.

Our plunging jets are equally spaced along the chamber diagonal and penetrate the seawater volume at a depth of 7 cm, and therefore do not interact with the chamber walls. Now specified in the method section.

Moreover, the sea spray chamber used by Schwier et al. (2015) had multiple plunging jets, and there may be differences between this setup and those that use single plunging jets. However, since the details of the chamber used are not clear, it is currently impossible to determine this.

This was stated in the previous literature that we cited, but it is now specified that we use 8 jets. There are indeed likely differences with the multiple jet system used in different studies (including the MART), with a one jet system.

Regarding the air entrainment rates described by the authors, how they were obtained is unclear. As the authors normalized their fluxes to this parameter, a clear description of the methodology used to determine the air entrainment rates is necessary in the manuscript.

We measured the air entrainment in our system using an approach similar to Salter et al. 2014. The procedure to use this air entrainment flux to derive an equivalent wind speed is described in the text:

Calibration experiments performed following the procedure of Salter et al. (2014), enabled to established that the air entrainment flowrate in our system is 4.5 Lair min-1 under the jet operational condition (seawater flowrate of 1.25 L min^{-1}, orifices' diameters, jet distance to seawater surface). According to Long et al. (2011), the flux of air entrained (Fent) during wave breaking can be related to a wind speed at 10 m (U10) following:

$$F_{ent} = 2 * 10^{-8} U_{10}^{3,74} \quad (3)$$

Given that, we calculate that our plunging jet system simulated a bubble volume distribution equivalent to that produced at a wind speed of 9 m s^{-1}.

The authors need to provide a more detailed description of how they controlled the temperature of the sea spray chamber. While they mention the use of a 50 L temperature-controlled reservoir, it is unclear how this was connected to the sea spray chamber. Specifically, it is not clear whether the sea spray chamber was immersed in the reservoir or connected to it via some other means. A schematic of the experimental setup would be useful in clarifying this point. Further, how were the temperature experiments conducted?

A schematic of the experimental setup is provided in Sellegri et al. 2023, and is now also presented in Figure 1.

The authors mention that they applied temperature gradients ranging from 2°C to 15°C to the seawater over approximately 1 hour, but the exact form of these experiments is unclear. Given that these experiments are crucial to the study, it would be helpful for readers to see a typical experiment as a figure in either the main manuscript or the supplementary materials. This would provide more context and enable better understanding of the results.

All temperature experiments are reported Figure 4a (given that fluxes vary as SSA concentrations as stated equation 1).

In addition, these are fast temperature ramps which leads to the question of how repeatable the measurements were. Were any experiments conducted over a longer time period at constant temperature to determine the impact of quickly ramping the temperatures on the fluxes versus holding the system at a steady temperature?

Fast changes were applied so the seawater biology did not have the time to react to temperature changes as our goal was to investigate the physical dependance of fluxes to instant biogeochemistry. Holding the system for longer time periods would, in our opinion, lead to unrealistic changes in biology or chemistry and also our reservoir had a limited

volume that prevented these tests, so we did not conduct experiments at constant temperature. Now specified in the text.

There are some important details missing about the aerosol measurements. It is unclear how the instruments were connected to the sea spray chamber. Were all instruments connected through a single connection, and was the sampling conducted isokinetically?

The instrumental set-up is now shown in Figure 1

The type of differential mobility particle sizer used is also not specified. Was it purchased or custom-built in-house? Additionally, it is unclear whether an impactor was used to prevent particles larger than 500 nm or some other cutoff from entering the instrument. It is also unclear whether the aerosols were dried before sampling or whether they were measured under ambient conditions in the sea spray chamber, and if so, what was the relative humidity of the sample. Again, some of these details could be better explained with the inclusion of a schematic of the setup.

All of these details are given in Sellegri et al. 2023, now reported here again.

" For submicron particles, SSA were taken through a ¼ inch stainless steel line to a 1-m long silica gel diffusion drier followed by an impactor with PM1 diameter cutoff. Particle size distributions were monitored by a differential mobility particle sizer system (DMPS) at 1 LPM, with a separate line to a condensation particle counter (MAGIC CPC, flowrate 0.3 LPM) connected in parallel for validation. The DMPS system was preceded by a soft X-ray aerosol neutralizer (TSI Model 3088) and consisted of a TSI-type custom-built differential mobility analyzer (length 44 cm) operated at a sheath flow rate of 5.0 L/min for selecting particle sizee range of 10-500 nm across 26 size bins during a 13 min 40s scan and a TSI CPC model 3010. Relative humidity at the inlet was monitored, and kept below 35% at all times. Another short, smooth curvature antistatic Teflon ½ inch line brought the generated SSA to a Waveband Integrated Bioaerosol Sensor (WIBS) for diameters ranging from 500 nm up to 4500 nm"

In their study, the authors use the flux of particles larger than 100 nm as a proxy for cloud condensation nuclei (CCN). However, they should provide a detailed explanation of how they obtained this flux. Although they mention using the flush air flow and water surface of the tank, they fail to clearly explain the process. To help the reader understand, the authors should provide a mathematical explanation of the process and how they normalized it to wind speed. Moreover, the authors mention using air entrainment, but the details of how this was done are unclear, and it should be explained to the reader. To make it easier to understand, the authors should describe the process used mathematically.

Text has been modified:

"The flux of SSA was calculated from the SSA total number concentration, as follows:

$$\boldsymbol{F_{tot}\ (\#\ m^{-2}s^{-1}) = \frac{CN_{tot} * Q_{flush}}{S_{tank}}} \quad (1)$$

where CN_{tot} is the concentration of SSA measured from the MAGIC CPC, Q_{flush} is the flushing air flowrate inside the tank's headspace, and S_{tank} is the surface of seawater inside the tank. In Sellegri et al. (2021), hereafter referred to as SELL21, the concentration of > 100 nm particles was used as a proxy for CCN concentration. For comparison to SELL21 we also calculated fluxes of SSA larger than 100 nm. The flux of CN100 (F_{CN100}) was calculated in a similar manner to Equation (1):

$$\boldsymbol{F_{CN100}\ (\#\ m^{-2}s^{-1}) = \frac{CN_{100} * Q_{flush}}{S_{tank}}} \quad (2)$$

where CN_{100} is the concentration of SSA with a diameter larger than 100 nm. Calibration experiments performed following the procedure of Salter et al. (2014), enabled to established that the air entrainment flowrate in our system is 4.5 Lair min-1 under the jet operational condition (seawater flowrate of 1.25 L min-1, orifices' diameters, jet distance to seawater surface). According to Long et al. (2011), the flux of air entrained (Fent) during wave breaking can be related to a wind speed at 10 m (U10) following:

$$F_{ent} = 2 * 10^{-8} U_{10}^{3,74} \quad (3)$$

Given that, we calculate that our plunging jet system simulated a bubble volume distribution equivalent to that produced at a wind speed of 9 m s-1. For the data acquired with a seawater flowrate that deviated from 1.25 L min-1, fluxes were normalized to the 9 m s-1 equivalent windspeed with the following relationship:

$$F_{normalized} = F_{original} * \frac{1.25^{2.4}}{Q_{SW}^{2.4}} \quad (4)$$

Where Q_{SW} is the seawater flowrate. Equation (4) was obtained by varying QSW over a short period (less than an hour) and fitting the flux dependence to Q_{SW}. Normalization resulted in less than 30% change in the fluxes for 80% of the data."

The authors conducted an experiment to measure the surface tension of seawater at different temperatures. They froze the water samples and then allowed them to warm up to 15°C over approximately one hour. It would be helpful to include a typical experiment's data plotted in the supplement or manuscript (this maybe figure 4 but it is not completely clear).

Yes, this was Figure 4 (which is now Figure 4b). We make this very clear now in the method section.

"Results of surface tension measurements as a function of sample temperature during unfreezing are shown in Figure 4b"

The authors suggest that the short time period of the experiment reduced the impact of changing biogeochemistry on their measurements. However, they do not discuss the potential impact of freezing on the surface tension. For instance, freezing could rupture phytoplankton cells present in the sample, releasing organic matter into the water, which could impact the surface tension. Did the authors conduct an experiment where they

measured the surface tension of a fresh sample at ambient temperature, froze it, and then returned it to the same temperature? If so, the measurements should be similar if freezing had limited impact, and this potential issue should be discussed.

Yes, freezing may have impacted surface tension measurements, as it does for many offline analysis when samples need to be taken back from ships to the laboratory. We did not conduct the tests suggested by the reviewer, and now warn the reader that a bias in the surface tension may have occur due to the impact of freezing. This effect should be investigated separately, as we anticipate that the impact is dependent on biology and would be a nice subject of investigation.

"A bias may exist in the surface tension measured here after samples have been frozen, compared to the surface tension of a sample that would have not experienced freezing, due to the impact of freezing on, for example, the rupture of phytoplankton cells releasing organic matter. Future studies should investigate how freezing may impact surface tension."

Furthermore, to rule out contamination from the sample tubes, were any measurements of pure water taken in the Falcon tubes? Including this information would enhance the clarity of the authors' findings.

Blank measurements were regularly performed using milliQ water, and we expect minimal contamination of Falcon tubes by surfactants.

The authors have presented time series of different parameters in Figure 1, but there are some significant issues with the data presentation. Firstly, the linear axis appears to have the same distance between the values of 0.00E+00 and 1.00E+06 and the values of 1.00E+06 and 2.00E+06.
Additionally, all the subscripts are missing from the axis labels and legend entries, which may seem minor, but it suggests carelessness on the part of the authors.
Moreover, the authors argue for a trend in this data, which is impossible to determine considering there are no error bars on the data points. Without an idea of the uncertainty on each data point, it is difficult to find a trend in the data when it is so noisy. Furthermore, it is not clear what the authors have plotted in Figure 1. Have they plotted the integrated number flux of all particles following normalization, or is it the integrated number flux of all particles larger than 100 nm? The authors should clarify this point to help readers better understand their results.

We modified Figure 1 (now Figure 2) and accounted for all the reviewers observations

The authors initially mentioned fitting "single lognormal modes" to their measurements of aerosol size distribution. However, in the following sentence, they reported finding "four modes in the submicron range and two modes in the supermicron range". This description is unclear and adds to the manuscript's overall confusion. Upon examining Figure 6, it becomes evident that the authors actually fitted a series of lognormal modes, not "single lognormal modes".

Now clarified in text:
"These were averaged over two different temperatures ranges, 2-3 °C and 7-9°C, and fitted with a combination of single lognormal modes (Figure 5)."

However, they did not provide any information about their fitting procedure or the quality of the fits. It is generally recommended to present data and its associated uncertainty (e.g., mean/median and standard deviation/error) before comparing contrasting datasets. If the authors wish to include fits, they could be presented in another panel or the supplement. Comparing the actual data across different temperatures is crucial to determining whether differences exist between the presented temperature regimes. Without the data, it is impossible to determine whether such differences exist.

A mode fitting procedure was applied for each experiment to the average size distribution measured in the coldest temperature range and also the highest temperature range, in order to visualize the change in the mean diameter of each mode, as reported Table 1 (and now commented in text). We now show the quality of the fitting procedure in Figures S1 and S2 for the respective temperature ranges. Also, standard deviations were added to Figure 6 (now figure 5) highlighting how significantly the two average size distributions were.

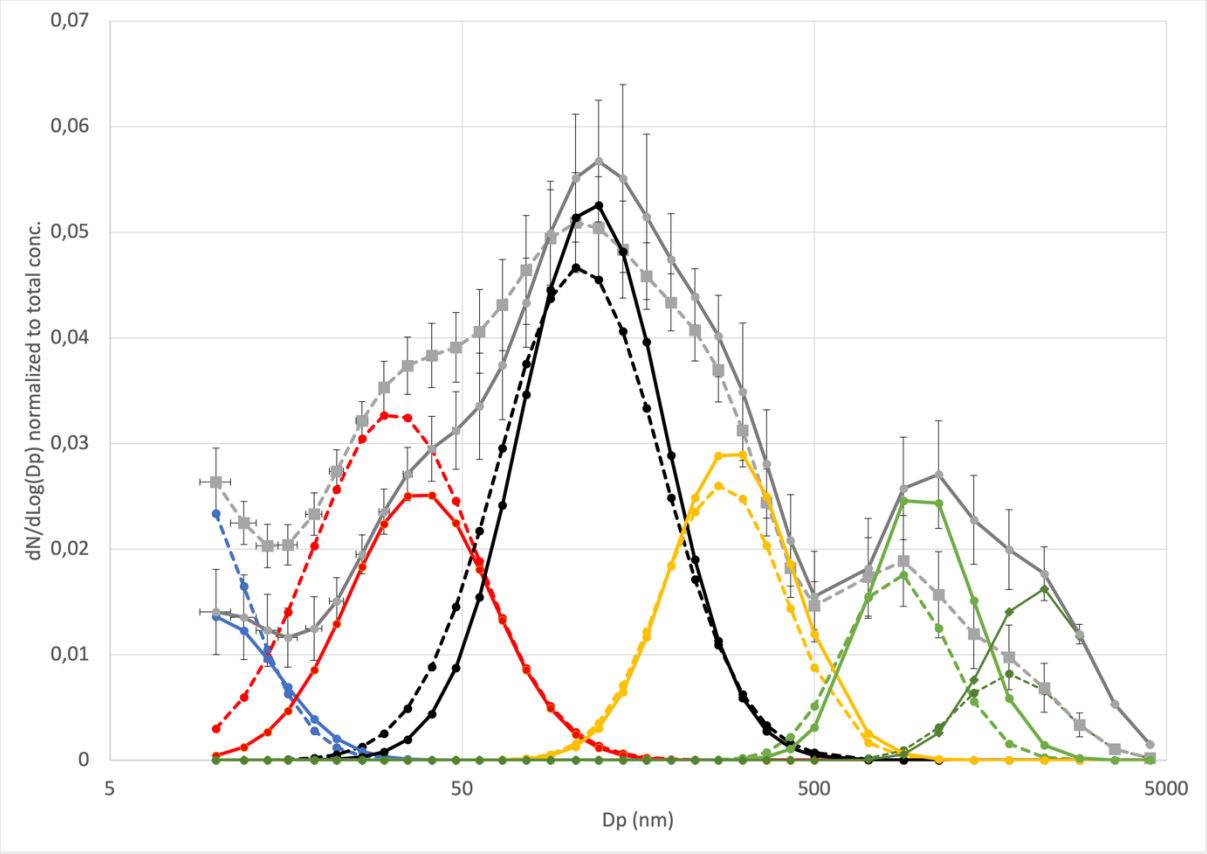

Figure 5: Average sea-spray size distributions normalized to the total sea-spray concentrations for the temperature range 7 - 9 °C (plain lines) and 2-3 °C (dash lines) reconstructed from their decomposition in a combination of single log-normal (nucleation mode (blue), Aitken mode (red), first accumulation mode (black), second accumulation mode (yellow) and the two coarse modes (light and dark green). Error bars are from standard deviation on the averaged measured size distributions.

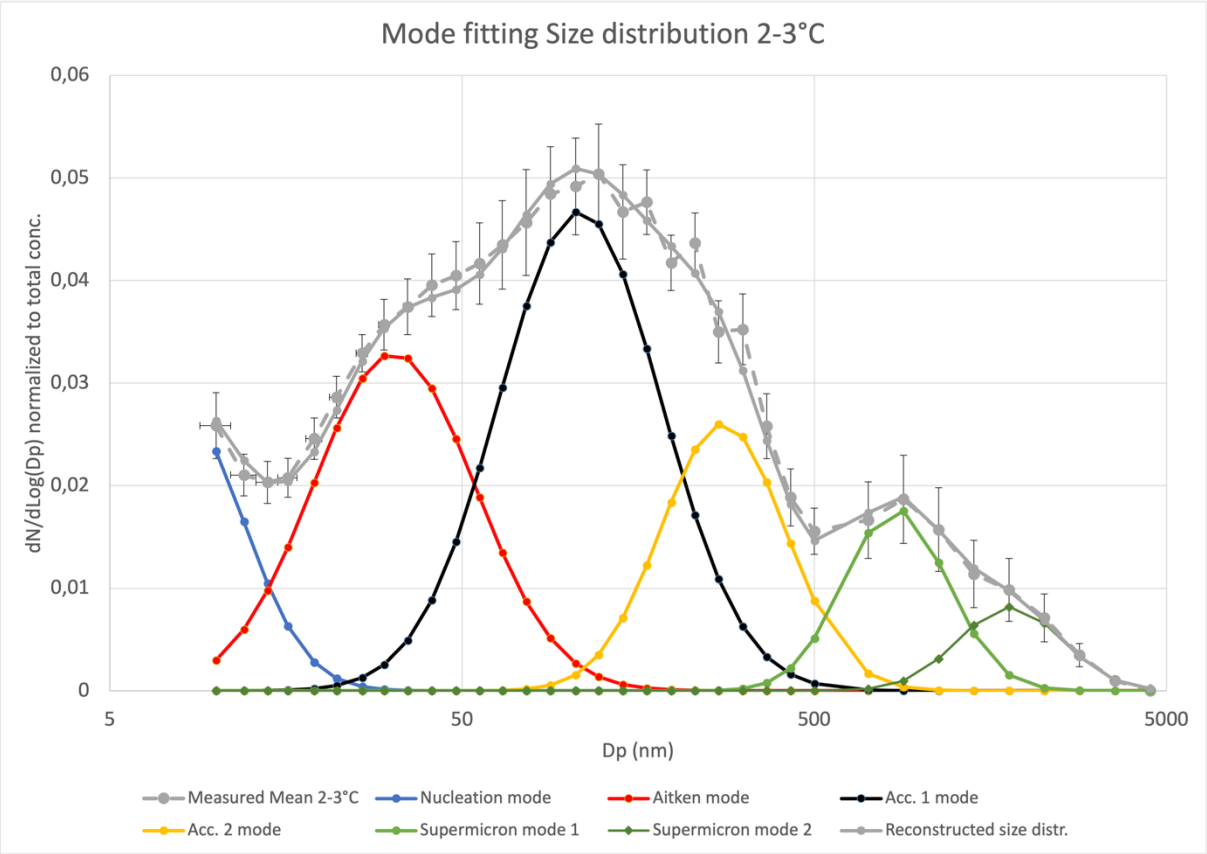

Figure S1: Measured sea-spray size distributions from merged SMPS and WIBS data, normalized to the total sea-spray concentrations and averaged for the temperature range 2-3 °C (dash grey line). Decomposition in a combination of single log-normal modes show the nucleation mode (blue), Aitken mode (red), first accumulation mode (black), second accumulation mode (yellow) and the two coarse modes (light and dark green) and reconstructed size distribution from the addition of individual modes (plain grey line). Error bars are from standard deviation on the averaged measured size distributions.

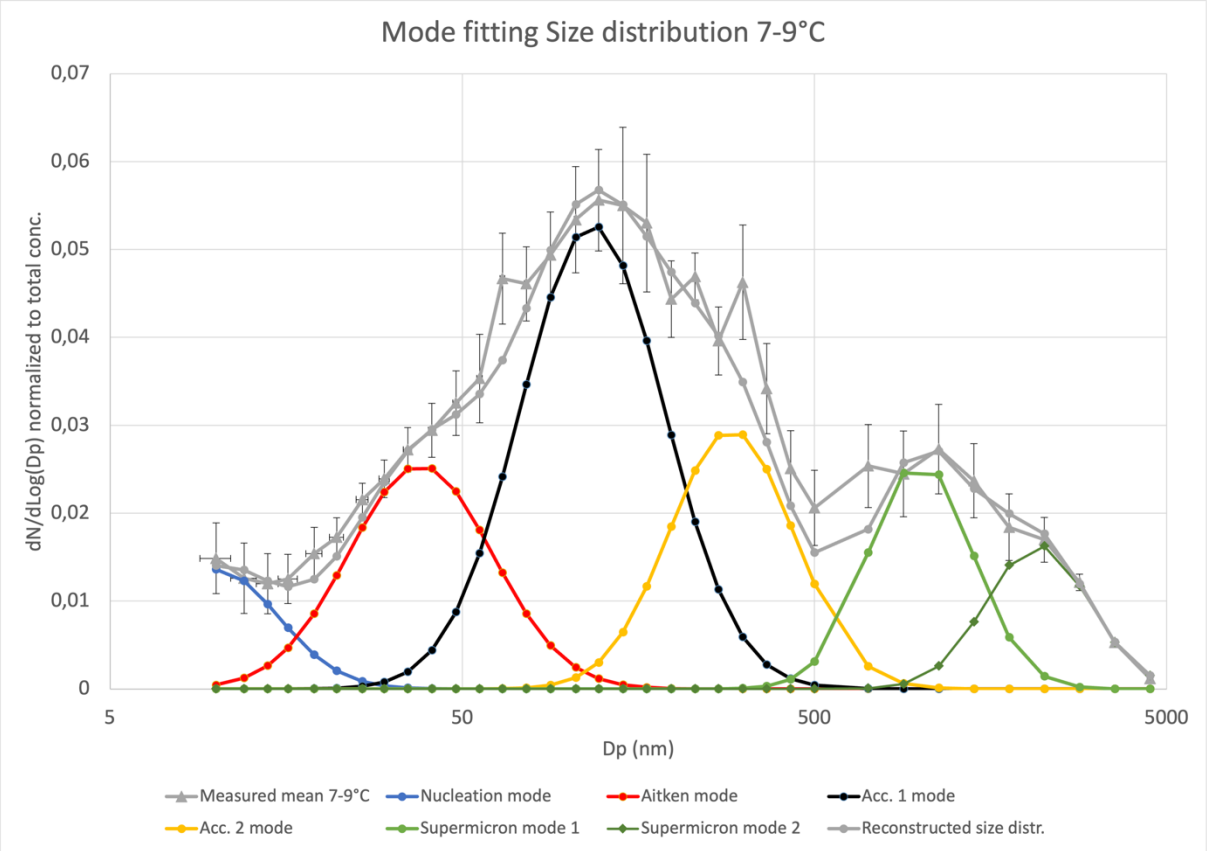

Figure S2: Same as S1 for the temperature range 7-9 °C.