

We thank the reviewer for carefully reading our manuscript and for providing us with their helpful insight. We have adjusted our manuscript based on their recommendations, which we believe has strengthened the manuscript's overall impact. Our responses to specific comments are shown below in red.

Review of "A Two-Component Parameterization of Marine Ice Nucleating Particles Based on Seawater Biology and Sea Spray Aerosol Measurements in the Mediterranean Sea" by Trueblood et al.

#### General comment

This study investigated the ice-nucleating abilities of surface microlayer (SML), surface seawaters (SSW), and sea spray aerosol (SSA) particles collected/generated in the Mediterranean Sea. In parallel to the evaluation of the ice-nucleating abilities of the different samples, a large set of biogeochemical analysis were performed on the samples to understand the relationship between ocean biology and marine ice nucleating particles (INP). While the ice nucleation analysis of the SML and SSW samples was performed with a LED based Ice Nuclei Detection Apparatus (LINDA), the analysis for the SSA samples was performed using a Dynamic Filter Processing Chamber (DFPC). Taking into account the collected information, the authors developed a new two-component parametrization. Although the information collected/derived by the authors is very rich and valuable, in addition that they were collected in a poorly explored region on Earth, the manuscript is not easy to follow, it lacks important information, and the conclusions are not clearly supported by the provided data. The manuscript fits with the ACP scope, but the current version cannot be accepted for its publication.

Major Comment 1. Two different techniques were used to analyze the ice nucleating abilities of the samples, i.e., the LINDA and the DFPC. While the LINDA determines the INP concentrations via the immersion freezing mode, the DFPC does it via condensation freezing. Given that both data sets were used to develop the parametrization, I am wondering if the INP concentration delivered by both instruments are directly comparable. For example, I am wondering about the very low concentration of INPs reported for the SSA samples in comparison to literature data. It is a true number or is it an artifact related to the used method?

Only the DFPC IN data was used to develop the parameterization. The LINDA instrument was only used on seawater (SSW and SML) samples, and the results were used to help guide the selection of features for the parameterization of SSA INP based on their correlation with biological features. A more in-depth response regarding the process of using the DFPC and its comparison with immersion freezing methods is given in the response below.

2. I was unable to fully understand how the INP concentrations for the SSA samples were obtained as the DFPC was not properly described. Is this a new custom-made instrument? Is this the first data delivered/published by this instrument? If this is the case, a much deeper description needs to be provided. If this is not the case, how good is the agreement of the data delivered by the DFPC against other well-known ice nucleation instruments?

The method for the DFPC is as follows: aerosols are collected onto a filter which is then placed on a metallic support covered with a layer of paraffin. The paraffin is heated slightly (60-80°C) for ~10 seconds and then rapidly cooled to allow the paraffin to penetrate the filter pores. This process is done to modify the color of the filter from white to black for optical processing and to improve thermal conductivity. The filter is then placed inside the DFPC on top of a Peltier cooler plate. The filter is then continuously swept by humid and cool air. Upon activation, ice nuclei grow into ice crystals with size around 0.4 mm. The ice crystals are then recorded using a camera.

In this study, the DFPC was used at water saturation,  $S_w$ , of 1.02 and  $T = -22, -25, \text{ and } -18^\circ\text{C}$ . Under these conditions, we expect to see condensation freezing mode. It has been reported (Vali et al., 2015) that condensation freezing and immersion freezing are not distinguishable as unique.

The DFPC has been used in multiple previous studies and found to agree well with other INP monitoring instruments (DeMott et al., 2018; Hiranuma et al., 2019; McCluskey et al., 2018).

We have updated the methods section starting at line 167 of the main text to account for this added information.

3. I am surprised that the INPs concentrations of the SML, SSW, and SSA are not compared to literature data. Actually, a recent study by Gong et al. (2020) who also studied the SML, SSW, and SSA is not cited/discussed here. There are also other studies in marine environment performed at subtropical and tropical latitudes that deserved to be discussed in the context of the present study.

We have added Figure 2 and Figure 3 to the manuscript, along with discussion starting on line 222 to compare seawater and SSA INP concentrations to previous studies.

4. Given that the chemical composition of the SSA is linked to the size of the aerosol particles (e.g., O'Dowd et al. 2004; Prather et al. 2013), I am wondering how well the used apparatus to generate the SSA, reproduces the proper size distribution of the natural SSA. Also, the authors only provided the particle size distribution and the chemical characterization for particles smaller than 500 nm and 1000 nm, respectively. There is a big problem here because while the chemical analysis was performed for submicron particles only, but the INP concentration took into account total suspended particles. It has been shown that the aerosol particles with the highest potential to act as INP are those larger than 500 nm (DeMott et al. 2010), and especially the super-micron particles (Mason et al. 2015; Gong et al. 2020) ignored in the present study.

We agree that replicating the production mechanism of SSA is certainly an important and difficult task. The plunging apparatus used in this study has been well characterized in previous studies (Schwier et al., 2015; Schwier et al., 2017). Furthermore, the characteristics of the setup (water flow rates, plunging water depth, etc.) were selected to mimic Fuentes et al. (2010), which have been shown to mimic quite well the production of nascent SSA. To this end, we are confident that the SSA analyzed in this study are an accurate representation of natural SSA.

It is not yet clear whether INP from sea spray are mainly from super or submicron sizes. While Gong et al. (2020) reported high concentrations of INP for supermicron particles, they also note these likely came from dust particles rather than marine aerosols. As studies have shown that the IN active components of SSA are typically of biogenic origin (DeMott et al., 2016), and that the organic mass fraction of SSA is greatest for submicron particles (Gantt and Meskhidze, 2013), it would seem likely that the INP characteristics of SSA can be well described in terms of the submicron regime. Indeed, comparison of INP concentrations in PM1 and PM10 filter samples of both clean marine air and continental land-mass air at Mace Head in the North Atlantic have recently provided evidence that the majority of SSA INP are in the submicron regime and that terrestrial aerosol INP are in the supermicron regime (McCluskey et al., 2018). Field campaigns in the equatorial Pacific and Gulf of Mexico have also found the marine INPs are most abundant for particle diameters less than 500 nm (Rosinski et al., 1986, 1987; Rosinski et al., 1988). However, we want to stress here that our INP measurements are performed over the full range of SSA sizes, and we do not ignore its supermicronic fraction.

For a response to the concern of particle size distributions, please see our response to comment number 5 below.

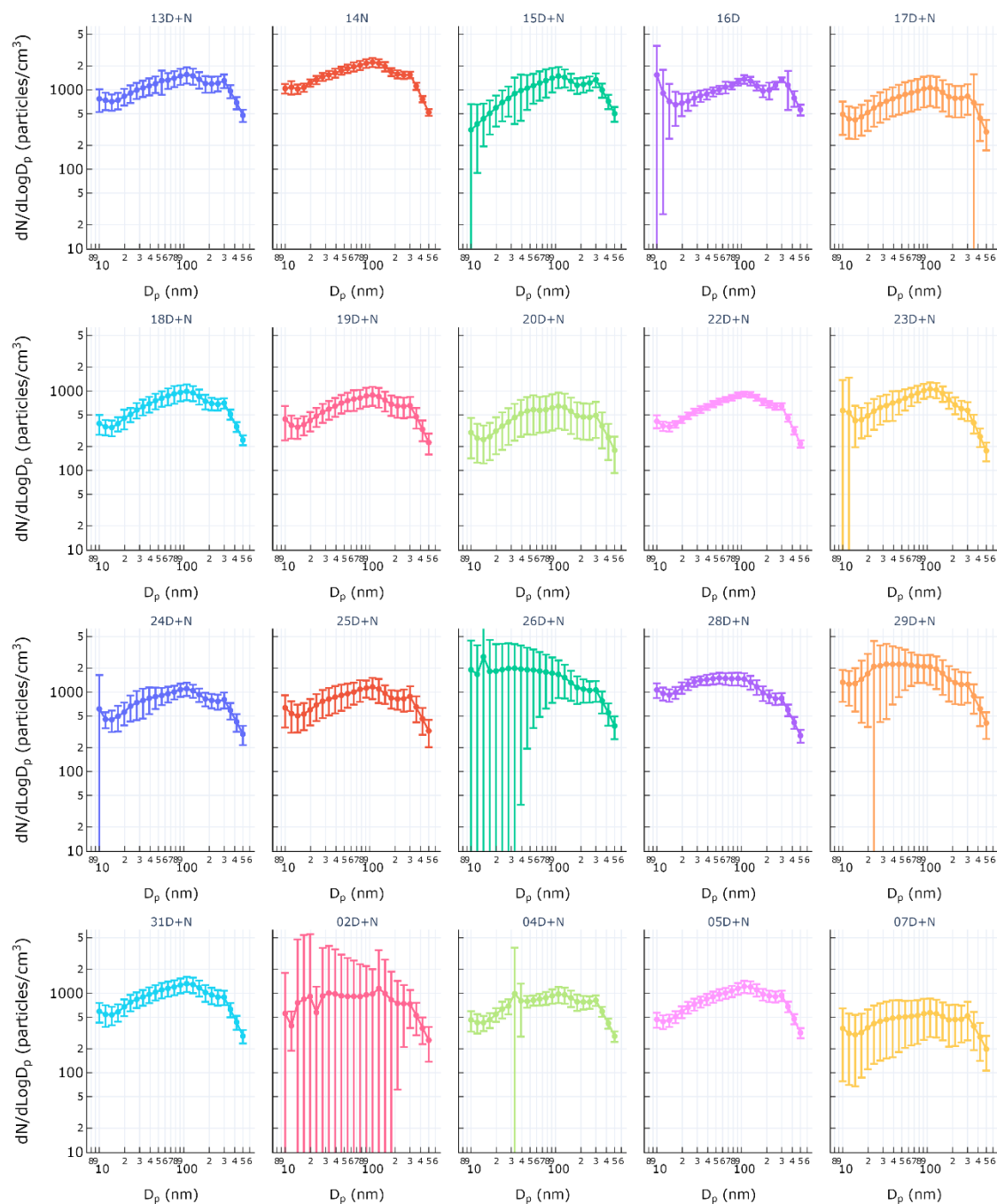
Concerning the chemical analysis of SSA, it is correct that only the submicron concentration of SSA has been measured. However, previous analysis of the chemical composition of marine aerosol have shown that most of the organic matter is found in the submicron fraction (O'Dowd et al. 2004, Gantt and Meskhidze 2013 for a review). Therefore, we base our analysis that the WIOC, WSOC and OC concentrations provided in this work are representative of the whole size range of SSA. This is now mentioned line 698-700 of the manuscript.

Given that we have taken steps to accurately mimic the SSA production mechanism, extended our SSA surface calculation to include particle with diameters smaller than 10  $\mu\text{m}$ , and clearly mention the hypothesis that organic carbon in submicron SSA is representative of the OC in the totality of the SSA, we are confident that our approach is sufficient to characterize total SSA INP concentrations. However, future studies are needed to better constraint the characteristics of SSA in sub and supermicron regimes.

5. INPSSA was normalized by the particle surface area. However, the NPSSA were derived from samples with total suspended particles, but the particle surface area was derived from particles ranging between 10 and 500 nm only. There is a big mismatch here that can hide important information or can even conduct the authors to deliver wrong conclusion. That is why the following was found: "no statistically significant correlations were seen between total submicron particle counts or total SSA surface area and INPSSA at all three temperatures". Actually, it would have been more appropriate to use the size distribution of super-micron particles to calculate the particles surface area.

Please see below our response to reviewer 1 who had the same concern:

First, we calculated adjusted daily mean particle size distribution based on sampling time intervals from the differential mobility particle sizer (DMPS) that aligned better with when the filters later analyzed by the Dynamic filter processing chamber (DFPC) were collecting particles. In our original manuscript, daily means of DMPS data were calculated on a 24-hour time interval beginning and ending at midnight. As DFPC filter samples were not collected at these exact times, there existed a small misalignment between DMPS and DFPC sampling intervals. We therefore re-calculated the DMPS daily mean across each DFPC sampling period. We also did the same adjustment for daily means of underway data when comparing underway data to the DFPC INP concentrations. We added error bars to represent the standard deviation throughout each sampling period to the resulting size distributions, produced from the bubbling system during each DFPC sampling period, shown in Figure R1. This figure has been added to the supporting information as Figure S3 on line 18.



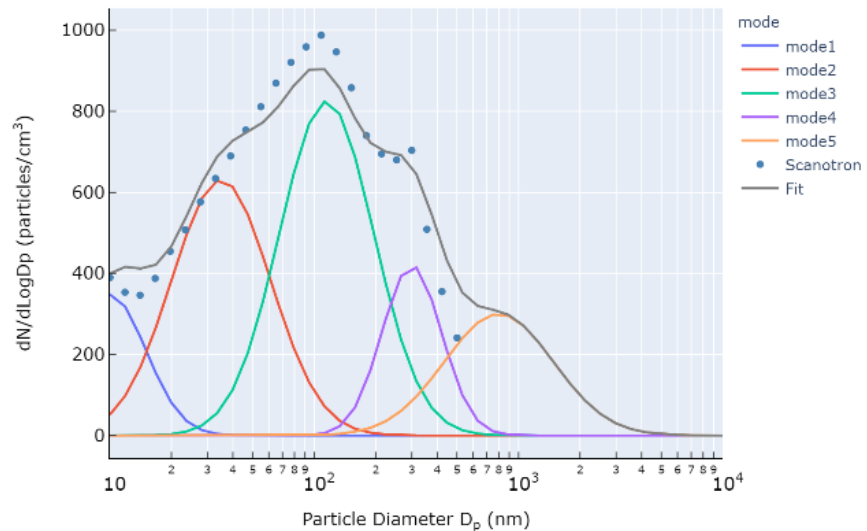
**Figure R1.** Average size distributions of SSA produced by the plunging apparatus as observed by DMPS across each DFPC sampling period. Error bars represent standard deviation.

Both reviewers expressed concern that the DMPS data used to calculate surface area of SSA did not include particles above 500 nm in diameter. The reviewers correctly pointed out that an additional mode at 800 nm exists, which contains a large portion of SSA surface area. Ovadneveite et al. (2014) developed a sea spray aerosol source function consisting of 5 log-normal modes based on in-situ particle number concentration measurements at Mace Head and open-ocean eddy correlation flux measurements from the Eastern Atlantic. Comparison of parameters from their fit with those from the fit of our number-size distribution revealed good agreement between the two. The parameters are shown in Table R1 below. This table has been added to the SI on line 1 as Table S1.

**Table R1.** Lognormal parameters for a sea spray source function parameterization from Ovadnevaite et al. (2013) and for the fit of observed particle counts during the PEACETIME cruise. For each mode ( $i$ ), a geometric standard deviation ( $\sigma_i$ ), count-median diameter ( $CMD_i$ ), and total number flux ( $F_i$ ) or amplitude is shown. For the fit from the literature (Ovadnevaite et al., 2014),  $F_i$  is a function of Reynolds number  $Re_{HW}$  which we selected as  $3.1 \times 10^6$  based on the air flow across the surface of the water in our bubbling apparatus.

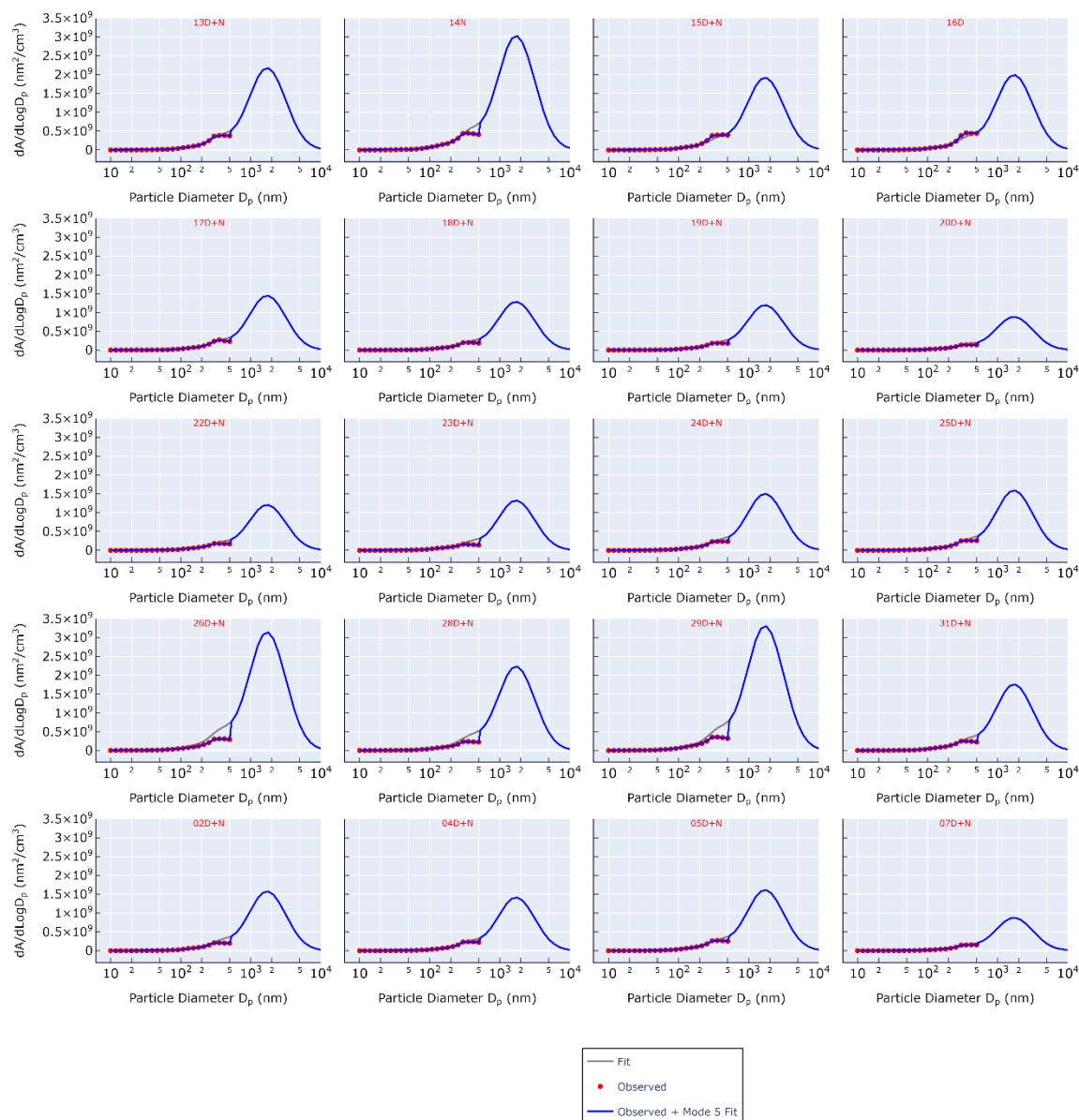
| $i$                              | $\sigma_i$ | $CMD_i$ | $F_i/Amplitude$                          |
|----------------------------------|------------|---------|--|
| <b>Ovadnevaite et al. (2013)</b> |            |         |  |
| 1                                | 1.37       | 0.018   | $104.5(Re_{HW} - 1 \times 10^5)^{0.556}$ |
| 2                                | 1.5        | 0.041   | $0.0442(Re_{HW} - 1 \times 10^5)^{1.08}$ |
| 3                                | 1.42       | 0.09    | $149.6(Re_{HW} - 1 \times 10^5)^{0.545}$ |
| 4                                | 1.53       | 0.23    | $2.96(Re_{HW} - 1 \times 10^5)^{0.79}$   |
| 5                                | 1.85       | 0.83    | $0.51(Re_{HW} - 1 \times 10^5)^{0.87}$   |
| <b>PEACETIME Cruise</b>          |            |         |  |
| 1                                | 1.5        | 0.01    | 0.01                                     |
| 2                                | 1.75       | 0.035   | 0.025                                    |
| 3                                | 1.7        | 0.115   | 0.031                                    |
| 4                                | 1.4        | 0.300   | 0.01                                     |

We next took the ratio of mode 5 to mode 3 from the Ovadnevaite (2014) fit and applied it to our fit to calculate a fifth mode accounting for particles ranging in size between 500 nm and 10  $\mu$ m. Figure R2 shows an example of the result of this process using daily mean data from March 18. The total fit is shown in gray, which consists of modes 1-4 as calculated from our DMPS data, as well as mode 5 calculated as described above. Blue circles represent observed values.



**Figure R2.** Example of resulting size distribution fit based on comparison of fit from observed PEACETIME particle counts with a 5 lognormal-mode fit from the literature (Ovadnevaite et al., 2014). Blue markers denote particle counts by the DMPS instrument (named Scanotron). Modes 1-4 are fit based on observed data. Mode 5 is calculated by taking the ratio of Mode 5/3 from the Ovadnevaite et al. (2014) fit and applying it to our observed mode 3.

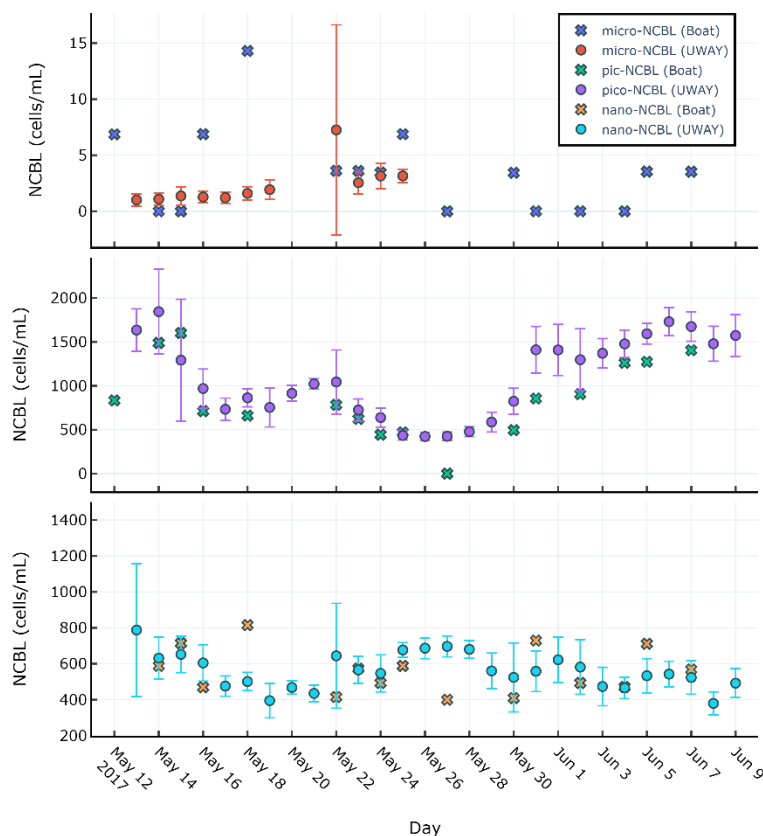
We applied this calculation to the mean data from the DMPS for each DFPC sampling period. From the resulting fits, we calculated aerosol surface area distribution, shown in Figure R3 (also found on line 22 of the Supporting Information as Figure S4). Finally, we used this adjusted surface area value to re-calculate surface area normalized INP concentrations. We have added description of this calculation to the main text on line 172.



**Figure R3.** Daily average of adjusted SSA surface area distributions. Sampling time is indicated in red text at the top of each plot, where numbers indicate the day of the month and D/N indicates whether sampling was conducted at day/night, respectively. The gray line shows the combined fit of modes 1-4 from observed data with the additional contribution of mode 5 as calculated using the Ovadnevaite et al. (2013) fit . Red circles represent observed values and blue line represents the surface area from observed values through 500nm plus theoretical contribution from mode 5 from the gray fit. The small difference between blue and gray lines indicates the goodness of the fit.

6. It is unclear if the SSW samples are really superficial waters (as defined by the authors), bulk waters, or deep waters. I could not find the depth at which those samples were collected. Also, the SSA was generated from waters collected at 5 m depth. Would not have been more relevant to use superficial waters instead? How comparable are the ice-nucleating abilities of the SSA particles (from “deep” waters, 5 m) with those from the superficial SML and SSW samples?

We do mention the sampling depth of discrete SSW samples and underway continuous seawater (respectively 20 cm and 5 m). As the 5 m waters showed similar properties to SSW collected from 20



cm (see Figure R4), we are confident that they can be considered similar in nature.

**Figure R4.** Daily average of continuous NCBL measurements from the underway (UWAY) system, where error bars represent standard deviation compared with discrete daily samples from the workboat at 20 cm.

7. Section 4, the most important, is extremely short and too general without the required information to follow it. Two parametrizations were developed, one for temperatures above  $-22^{\circ}\text{C}$  and one for temperatures below  $-22^{\circ}\text{C}$ . Therefore, this means that the INPSSA were included in both parametrizations as the INP concentration for temperatures between  $-18^{\circ}\text{C}$  and  $-25^{\circ}\text{C}$  were obtained for the SSA samples. However, as mentioned above the chemical analysis for the SSA samples was performed for submicron particles only. C3 Therefore, this parametrizations may be valid for submicron SSA particles only and are not representative for marine aerosol particles.

Concerning the chemical analysis of SSA, see our answer at point 4.

Reviewer 1 rose similar concerns regarding Section 4. Please see our response below:

To ensure selection of the model that best fits the data, we formulated various parameterizations consisting of different time periods, features, and number of components for temperature ranges. Predictor features were chosen based upon their correlation with INP concentrations as described in the previous section. Single component parameterizations in which INP across all three temperatures were linked with the same features were compared with two-component parameterizations in which INP were split into warm and cold categories, each having their own predictor features. Finally, we developed and compared altered versions of the W15 and MC18 models to account for the oligotrophic seawater of the Mediterranean Sea, as the existing models were formulated from observations of eutrophic waters. Each parameterization was recalculated using data across all days of the cruise as well as for only days before the dust deposition event in order to



determine the impact of the dust event on the ability to predict INP. The complete set of parameterizations and their associated fit metrics ( $R^2$  and  $R_{adj}^2$ ) are given in Table R2.

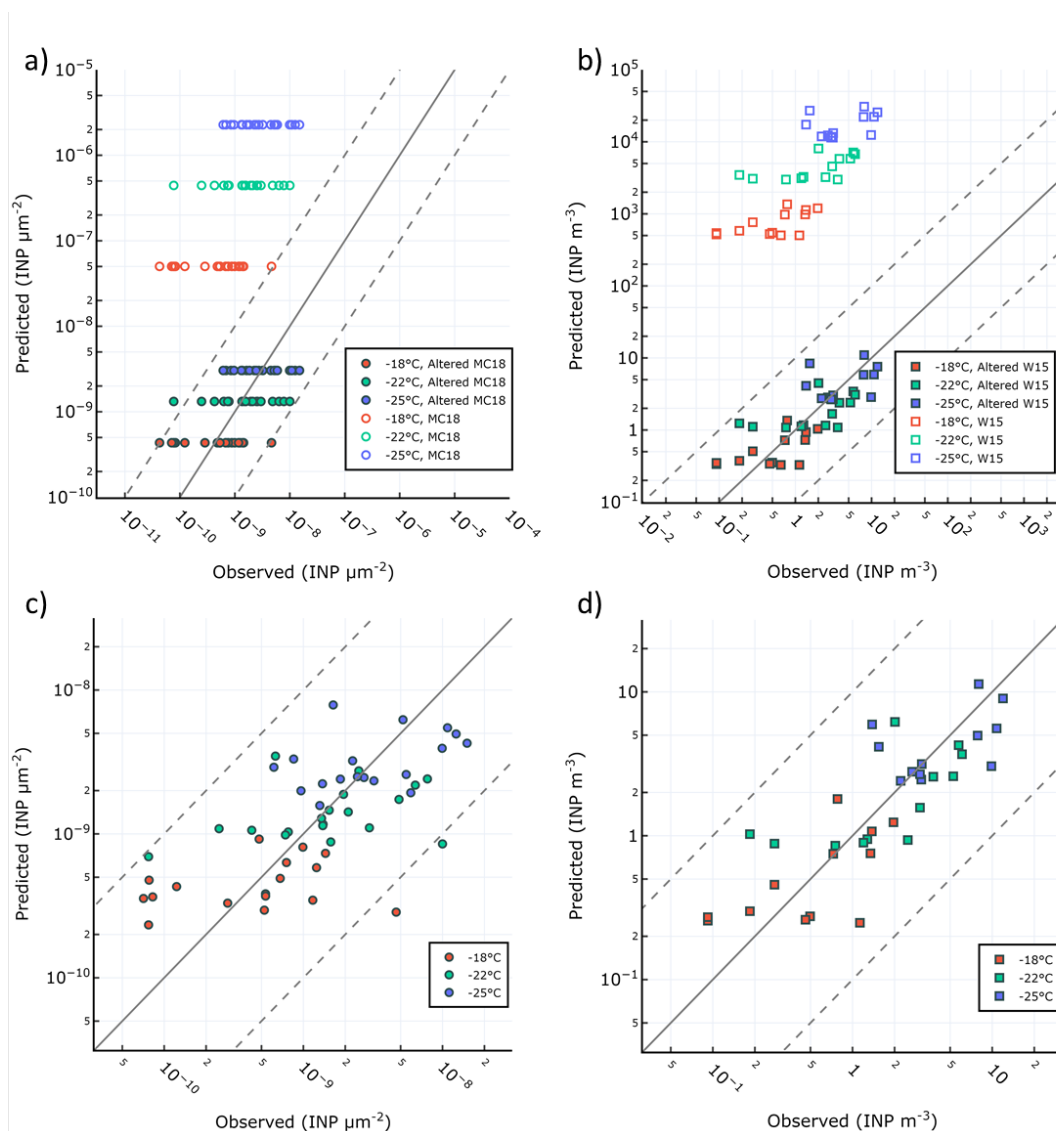
Figure R5a shows observed vs predicted  $INP_{SSA}$  for the W15 model, while Figure R5b shows the same but using the MC18 parameterization. Similar to our results for seawater INP, a large overprediction is found relative to our observations when using W15. Figure 5b shows that while MC18 is a slight improvement over the W15 approach, it still overpredicts INP by two orders of magnitude. We also present re-calculated best-fit-lines to data using the same features as in W15 and MC18 (i.e., OC and SSA surface area) in order to account for possible changes due to the oligotrophic nature of the Mediterranean Sea. We term these two parameterizations the altered Wilson fit for oligotrophy, which is given by:

$$\frac{INP}{m^3} = \exp(-7.332 - (0.2989 * T) + (0.3792 * OC_{SSA}))$$

and the altered McCluskey fit for oligotrophy, given as:

$$\frac{INP}{\mu m^2} = \exp(-26.57 - (0.2782 * T))$$

The results for these fits are shown in Figure R5a,b alongside the results of the original W15 and MC18 parameterizations. Both altered models offer improvements over the original parameterizations. The adjusted  $R^2$  of the altered Wilson fit for oligotrophy on log-transformed INP abundance was  $R_{adj}^2=0.59$  and was  $R_{adj}^2=0.32$  for the altered McCluskey fit for oligotrophy.





Interestingly, the adjusted Wilson fit for oligotrophy performs better than the adjust McCluskey fit for oligotrophy, which is the opposite of what was found when comparing the original models.

**Figure R5. Different parameterizations for prediction of INP in SSA. a) W15 and refit of same method using PEACETIME observations b) MK18 and refit of same method using PEACETIME observations c) single-component parameterization for INP/ $\mu\text{m}^2$  SSA surface area where INP at all temperatures are related to  $\text{POC}_{\text{SSW}}$  d) two-component parameterization for INP/ $\text{m}^3$  where  $\text{INP}_{\geq -22^\circ\text{C}}$  are related to OC and  $\text{INP}_{< -22^\circ\text{C}}$  are related to WIOC.**

We also tried a range of novel parameterizations based on the observed correlations between  $\text{INP}_{\text{SSA}}$  with seawater and SSA properties. Below we describe two parameterizations which offered good fits to the data. The single-component parameterization assumes the abundance of INP per unit surface area of total SSA at each temperature can be predicted from  $\text{POC}_{\text{SSW}}$  concentrations:

$$\frac{\text{INP}}{\mu\text{m}^2} = \exp(-28.5324 - (0.2729 * T) + (0.0361 * \text{POC}_{\text{SSW}}))$$

The second parameterization separates INP into warm and cold classes, where warm INP ( $\geq -22^\circ\text{C}$ ) are related to SSA OC and cold INP ( $< -22^\circ\text{C}$ ) are related to the concentration of SSA WIOC. This two-component parameterization predicts the concentration of INP/ $\text{m}^3$  through the following

$$\frac{\text{INP}_{T \geq -22^\circ\text{C}}}{\text{m}^3} = \exp(-7.9857 - (0.3178 * T) + (0.4643 * \text{OC}_{\text{SSA}}))$$

$$\frac{\text{INP}_{T < -22^\circ\text{C}}}{\text{m}^3} = \exp(-6.6606 - (0.2712 * T) + (0.5755 * \text{WIOC}_{\text{SSA}}))$$

equations:

Figure R5c,d shows the results of our single-component model using  $\text{POC}_{\text{SSW}}$  and the two-part model which uses SSA WIOC and OC and considers the separate temperature classes of INP. The adjusted  $R^2$  for each model on the log-transformed INP abundance were  $R_{\text{adj}}^2=0.404$  for the single component model using  $\text{POC}_{\text{SSW}}$  and  $R_{\text{adj}}^2=0.60$  for the two-component model using OC and WIOC. This result reveals that they both fit the observations better than the altered McCluskey parameterization for oligotrophy, while the two-component method performs as well as the altered Wilson parameterization. Each parameterization's fit to the data is improved when considering pre-dust days only ( $R_{\text{adj}}^2=0.63$  for the two-component parameterization and  $R_{\text{adj}}^2=0.57$  for the single-component parameterization). The improvement is more pronounced for the single-component parameterization using  $\text{POC}_{\text{SSW}}$ , further pointing to the fact that such dust deposition events can alter the INP properties of surface waters and the subsequent SSA, either through

**Table R2.** Summary of tested parameterizations to the PEACETIME dataset.

| Model Name          | INP Units           | Days     | # Cat. | Features           | Warm Features      | Cold Features     | R <sup>2</sup> | R <sub>adj</sub> <sup>2</sup> |
|---------------------|---------------------|----------|--------|--------------------|--------------------|-------------------|----------------|-------------------------------|
| PD-2TC_OC_WIOC      | INP/m <sup>3</sup>  | Pre-Dust | 2      |                    | OC <sub>SSA</sub>  | WIOC              | 0.66           | 0.63                          |
| PD-1TC_OC           | INP/m <sup>3</sup>  | Pre-Dust | 1      | OC <sub>SSA</sub>  |                    |                   | 0.63           | 0.61                          |
| PD-1TC_WSOC_WIOC    | INP/m <sup>3</sup>  | Pre-Dust | 1      | WSOC, WIOC         |                    |                   | 0.64           | 0.60                          |
| AD-2TC_OC_WIOC      | INP/m <sup>3</sup>  | All Days | 2      |                    | OC <sub>SSA</sub>  | WIOC              | 0.63           | 0.60                          |
| AD-1TC_OC           | INP/m <sup>3</sup>  | All Days | 1      | OC <sub>SSA</sub>  |                    |                   | 0.61           | 0.59                          |
| PD-2TC_POC_PHYTO-L  | INP/μm <sup>2</sup> | Pre-Dust | 2      |                    | POC <sub>SSW</sub> | Micro-NCBL        | 0.62           | 0.59                          |
| AD-1TC_WSOC_WIOC    | INP/m <sup>3</sup>  | All Days | 1      | WSOC, WIOC         |                    |                   | 0.62           | 0.58                          |
| PD-1TC_POC          | INP/μm <sup>2</sup> | Pre-Dust | 1      | POC                |                    |                   | 0.59           | 0.57                          |
| PD-1TC_POC_PHYTO-L  | INP/μm <sup>2</sup> | Pre-Dust | 1      | POC, Micro-NCBL    |                    |                   | 0.58           | 0.53                          |
| PD-2TC_WSOC_WIOC    | INP/m <sup>3</sup>  | Pre-Dust | 2      |                    | WSOC               | WIOC              | 0.53           | 0.49                          |
| AD-2TC_WSOC_WIOC    | INP/m <sup>3</sup>  | All Days | 2      |                    | WSOC               | WIOC              | 0.45           | 0.41                          |
| AD-1TC_POC          | INP/μm <sup>2</sup> | All Days | 1      | POC <sub>SSW</sub> |                    |                   | 0.43           | 0.40                          |
| AD-2TC_POC_PHYTO-L  | INP/μm <sup>2</sup> | All Days | 2      |                    | POC <sub>SSW</sub> | Micro-NCBL        | 0.43           | 0.39                          |
| AD-2TC_POC_PHYTO-LM | INP/μm <sup>2</sup> | All Days | 2      |                    | POC <sub>SSW</sub> | Micro-, Nano-NCBL | 0.43           | 0.38                          |
| AD-1TC_T            | INP/μm <sup>2</sup> | All Days | 1      | Temperature        |                    |                   | 0.33           | 0.32                          |

We have added this discussion to line 411 of the manuscript.

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