

ACP Review of 'A Two-Component Parameterization of Marine Ice Nucleating 2 Particles Based on Seawater Biology and Sea Spray Aerosol 3 Measurements in the Mediterranean Sea'.

General Comments:

The paper Trueblood et al. 2020 is a nice study which considers INP data from oligotrophic/Mediterranean waters and shows that eutrophic parameterisations (W15 and MC18) result in over-prediction. The occurrence of a dust deposition event over the measurement periods, in conjunction with measurements from the SML, SSW, and SSA, makes for very interesting reading, although it is a shame that the dataset ends before INP_{SSA} concentrations reached a clear maximum. However, this brings up the question can the two-component temperature dependent parameterisation from this study be relevant to much larger bodies of water? I am happy that the authors themselves addressed this in the need for future work relating INP_{SSA} to POC and NCBL measurements in the Southern Ocean. However, the difficulty in choosing POC and NCBL in relationship to INP is that all variables must be directly measured. The authors give no indication how to apply this parameterisation in a global model. The largest problem with the current study is that no uncertainties or error in the INP measurements (or biological measurements for that matter) are shown or discussed. This paper should not be published without the addition or evaluation of the inherent errors and uncertainties in the measurements themselves and the application of the measurements to creating a parameterisation. Also, the authors have not convincingly shown that the temperature dependent parameterisations are necessary to model INP concentrations, although they have shown that oligotrophic waters may need different parameterisations to eutrophic waters. A question also arises of whether INP_{SSA} increases after the dust event are really to do with the dust event or not? INP_{SSA} did not seem to be very connected with SML conditions (which surprised the authors and may therefore necessitate more attention). Lastly, throughout the text, Figure and Table descriptions are kept too short and often do not fully describe what is shown.

Specific Comments:

Temperature nomenclature (TM) varies throughout the text, sometimes for example as -15C or -15 C. Please keep consistency and it is suggested to use the proper format of e.g. -15°C.

All figures appear blurry, this should be corrected.

Line 102 – SSW properties were obtained from two depths 20 cm and 5 m, why this is done at two depths is never explained. It is important as POC is measured at 5 m depth while SML and 20 m depth SSW samples were measured simultaneously and both calculated NCBL.

Line 110 – Why is there a specific empirical relationship for PEACETIME? Will this affect other estimations of POC used for the parameterization?

Line 126 – methodology should be described in brief, or else simply cited if it is the only established measurement practice.

Line 140 – calculation should be described in brief. What are the associated errors/uncertainties of this methodology using LINDA?

Line 153 – You talk about bin size or 100-500 nm, but what is this? Is it the dry particle (electrical?) mobility diameter? This must be stated explicitly.

Line 161-164. Confusing description of how measurement of WSOC was measured vs how TOC was measured. Then how was WIOC measured?

Line 166-175. It seems that no measurements of ambient INP were taken. This seems concerning as often tank and ambient measurements do not always compare well to one another. Do you have evidence that the plunging jet SSA measurements were similar to that of the ambient SSA over the Mediterranean?

Line 166-175. What are the associated errors/uncertainties of this methodology using DFPC?

Line 178 – 183. How are INP from SSW measured (I assume it is LINDA – but this is not included in your methodology)? Which SSW measurement is tested for INP?

Line 191 – 192. The use of the term 'peak' here is a bit confusing in two ways. Purely graphically it is true that $INP_{SSA-25C}$ peaked on May 12, however, the implication that it is truly peaking is false as this is the first measurement it could have been higher before measurements commenced. Has contamination of the plunger tank system been ruled out as it is by far the greatest disparity between different temperatures for INP_{SSA} ?

Line 196. Again the use of the word peak is a bit misleading as measurements ended before the true peak could be observed. In this case can you really comment on the time difference between one peak in SML and SSA?

Line 204. Were there any differences in cell counts between SSW at 5 m or 20 cm depth?

Line 206-207. Are these the ranges associated with Pujo-Pay et al. 2011, or the ranges for this study? If the latter then perhaps give the expected range as well.

Line 209. How are you calculating enrichment factor? It is good to state as sometimes confusion arises.

Line 236-240. This paragraph feels like it is out of place as a discussion paragraph crammed between the synopsis of the results in the same Figure. It does not add much to the discussion. What do these two studies mean for your results? If anything they imply that you must compare INP_{SSA} to SSA bacterial abundance.

Line 250. DOC EF is positively correlated with $INP_{SML-15C}$, and you state this is due to the dust event, and in the next statement say that the fraction of DOC enriched in the SML during the dust event has specific IN properties. It seems possible that the DOC came from non-marine originating bacteria and that the deposition event also deposited terrestrial DOC which is the origin of increased IN ability. Or more so, could the correlation be coincidental with another

correlating factors from the dust event (i.e. Fe)? No indication is given of why the authors believe it to be 'likely connected to the CSP abundance, albeit not to the TEP', which if given may add value to the statement.

Line 291-293. What are the total particle counts referred to in Line 292? How are they measured and how do they match well with SSA counts in the range? In terms of SSA surface area: (1) how was SSA calculated from D_p ? (2) SSA have two noticeable modes larger than 500 nm, one is a submicron mode and the other is the jet-drop mode which are found to have mean dry mobility diameters near at 0.83 (Ovadnevaite et al. 2014) and $\sim 2 \mu\text{m}$ (Wang et al. 2017, Lewis & Schwartz 2004), respectively. According to Figure S3, most of the surface area distributions have already peaked by $0.5 \mu\text{m}$ particle diameter (with the possible exception of 2017-05-17), yet a significant portion of surface area for particles with $D_p > 0.5 \mu\text{m}$ seems to be lost. It seems an overstatement to say 'most of the surface area of sea spray is comprised between this size range'.

Ovadnevaite, J., Manders, A., de Leeuw, G., Ceburnis, D., Monahan, C., Partanen, A. I., Korhonen, H., and O'Dowd, C. D.: A sea spray aerosol flux parameterization encapsulating wave state, *Atmos. Chem. Phys.*, 14, 1837-1852, 10.5194/acp-14-1837-2014, 2014.

Wang, X., Deane, G. B., Moore, K. A., Ryder, O. S., Stokes, M. D., Beall, C. M., Collins, D. B., Santander, M. V., Burrows, S. M., Sultana, C. M., and Prather, K. A.: The role of jet and film drops in controlling the mixing state of submicron sea spray aerosol particles, *Proceedings of the National Academy of Sciences*, 114, 6978-6983, 10.1073/pnas.1702420114, 2017.

Lewis, E. R., and Schwartz, S. E.: *Sea Salt Aerosol Production: Mechanisms, Methods, Measurements and Models-A Critical Review*, American Geophysical Union, 2004.

Line 313. What is the difference between SSA OC and TOC here? How is OC calculated from the SSA?

Line 326/327. It would be good to state the relevant conclusions of Freney et al. 2020.

Line 368. How are you calculating OMSS? Why is this in agreement with Cochran et al. 2017?

Line 413. It is stated that '...the INP concentrations measured in the SSW are in line with the INP measured in the SML...'. There is only one comparison of INP shown of the two (figure 2a) and only one temperature is shown for the SSW. Is there further evidence to back this statement? Indicate what evidence is referred to in the text.

Table 1 – Description of table needs to state what p , $R(R^2)$ and n are. Is the p value of NCBL EF 0.78? This looks like a typo. Review the rest of the table to double check for other typographical issues. Why does it say $\text{CSP}_{\text{abundance}}$, when in there is no explanation of the difference between CSP and $\text{CSP}_{\text{abundance}}$?

Table 2. Description of table needs to state what p , $R(R^2)$ and n are. The table is stretched over a page break. This should be corrected to be on one page. Change POC to POC_{SSW} .

Table 3. Description of table needs to state what p , $R(R^2)$ and n are.

Figure 1. The image is blurry. The points indicated on the map are names with abbreviations that are never explained nor referred to in the text. If these refer to the dates mentioned in other graphs, this should be made clear. If not, then why are they there?

Figure 2. Why is there no uncertainty associated with each measurement? INP measurements have some of the largest uncertainties in aerosol science, this can't be neglected. How do you explain why $INP_{SSA,-25C}$ and $INP_{SSA,-22C}$ are sometimes anti-correlated and sometimes not? Some other minor corrections are needed. This graph is blurry and should be higher resolution. It would be nice to have different keys for a) and b). The y-axis in a) should be written scientifically – i.e. either 10,000 or 1×10^4 . It is difficult to differentiate the colours, effort should be taken to use different markers. The bottom axis should probably be the 'Date' not 'Day Number' (see same issue in other graphs).

Figure 3. This figure is also blurry with no error/uncertainty on the measurements shown.

Figure 4. Y-axis scale is difficult to interpret, should be written for example 10^8 not 10_8 . On the x-axis the authors might consider writing Temperature ($^{\circ}C$) rather than (C). Again error/uncertainties should be shown, or else noted that the error bars are not larger than the data points. The description of Figure 4 is on a different page than the figure, this should be corrected. It is difficult to tell day=2017-05-24 from day=2017-06-06. The authors could probably omit the 'day=' in the key and make the text larger.

Figure 5. Description does not mention INP normalised to SSA. Why use $/cm^3$ rather than $/nm^2$ which is what the surface area is shown in in Figure S3? When you normalise INP to SSA, should it not still be in term of $(/m^3 \text{ of air} \cdot SSA \text{ cm}^2)$? Top left panel, should read ' 3×10^{-4} ' not ' $3 \times 10_{-4}$ '.

Figure 6. Description should be below figure, and should include some more details of the graph. The figure is blurry, and need to be corrected. OMSS not explained.

Figure 7. Description should mention only significant correlations shown. Text should not state that these panels are a matrix. The scatter plots are blurry and should be corrected to higher resolution. The authors may choose to add r-values to each panel to make it easier for readers to study the results.

Figure 8. Graph should be made larger and enhanced to be less blurry. Y-axis scale is difficult to interpret, should be written for example 10^1 not 10_1 . It is difficult to read the axes. Your 3 panel axes seem to be in different units, some per L and some per m^3 . These are all SSA INP so they should be terms of their atmospheric concentration. This should be explained in the description.

Additionally, it seems clear from the graphs that while both the W15 and MC18 models over predict INP concentrations the over prediction is not really temperature dependent. The graph seems to show more of the difference between oligotrophic waters and eutrophic waters. How much does the authors' own parameterisations differ if only the colder (eq. 2) or warmer (eq. 1) parameterisation is applied to all the results? Are there any data of eutrophic waters which suggest a temperature dependence might improve the agreement?

Supplementary Info – consider adding a schematic of measurements taken from the tank.

Table S1. Usually tables come before Figures. Description of table needs to state what p , $R(R^2)$ and n are. Place a '0' before all values in column p .

Figure S1. Where possible, missing data should be deleted rather than shown as a line jumping from the last measured point to the next. There should be graph panel specific keys as each factor is not shown on every graph. It would be nice if more detail could be given in the description of where/how these measurements were taken. A description of what POC or biovolume covers here could also be useful.

Figure S2. Grey outline squares around a) and b) are somewhat off centre and cut-off the a) and b). Fe axis should be shown on the same scale in a) and b).

It would be nice to see INP_{SSA} measurement overlaid in time with those SML and SSW conditions considered to be contributing most prominently to INP_{SSA} concentrations.

Figure S3. It is nearly impossible to tell some of these 'variable' apart as the same color is used for multiple days. Please graph in such a way that the surface area spectrums can be identified for each variable. If they are daily averages than the stdev should also be graphed. Y-axis, change from ' $(nm^2/(cm^3))$ ' to ' (nm^2/cm^3) '. The authors could probably omit the 'variable=' in the key. Also, it is low resolution. What is a scanotron? Were these not measured by the DMPS as stated in the methodology?

Figure S4. Color of 'variables' again overlap for multiple days. Please graph in such a way that the number size distribution spectrums can be identified for each variable. If they are daily averages than the stdev should also be graphed. The y-axis shows $dN/d\log D_p$ in ' $(particles/(cm^3 nm))$ ' the extra nm is likely a typo? It should be ' $(/cm^3)$ '. The authors could probably omit the 'variable=' in the key. Also, the graph resolution is low. What is a scanotron? Were these not measured by the DMPS as stated in the methodology?

Technical corrections:

Line 22 – delete the 's' after INP, as INP is defined plural earlier.

Line 29 - delete the 's' after INP. This occurs many more times so check throughout the text.

Line 33 – delete extra space '...to SSW parameters (POC_{SSW} ...'. Add an 'and' or a ',' between ' $(POC_{SSW} INP_{SSW,-16C})$ '.

Line 56 – delete ')(' between references and replace with ';'. Delete '-' after SSA.

Line 62/63 – refer to study simply as 'Wilson et al. 2015 identified a temperature-dependent...'. Either delete the 's' from the end of the word entities or from concentrations.

Line 68 – TM (see specific comments).

Line 85 - delete the 's' after INP. Delete the 'the' before title of study. Here is it the title of the cruise or study? I suggest replace the word 'cruise' with 'study' and delete 'study' from the end.

Line 87 – add space, 'May 10 – June 10, 2017'.

Line 88 – delete 'were'.

Line 92 – what is 'R/V'? Here 'Pourquoi Pas?' is written differently than later. Keep consistency.

Line 94 – replace 'fashion from 35° to 42°' to 'fashion between 35° to 42°'.

Line 109 – HPLC acronym not explained.

Line 113 – FWS and SWS acronym not needed as never used again.

Line 124 – ICP-MS acronym not explained. Replace with full title as acronym not needed.

Line 130 – MQ acronym not explained. Replace with full title as acronym not needed.

Line 131 – add space between '0.5L'

Line 132 – HCL acronym not explained, although it is well known as Hydrochloric acid. Authors may choose to spell it out as it is not repeated.

Line 135 - add space, 'May 22 - June 7'.

Line 137 – TM (see specific comments).

Line 146 – change meter to 'm'

Line 147 – ACSM acronym not described. DMPS and CPC acronym used before description.

Line 153 – correct to '10-500 nm'.

Line 159 – MSA acronym not explained. Replace with full title as acronym not needed.

Line 160 – KOH acronym not explained. Replace with full title as acronym not needed.

Line 161 – WSOC acronym used for first time and is not defined.

Line 166 – '24h' change to '24-hour' to keep consistency.

Line 167 – delete the 's' after INP, as INP is defined plural earlier.

Line 169 – change to '47 mm' with space.

Line 173 – TM (see specific comments). Add '...(for air temperatures of ... -22.3 C, respectively)'.

Line 175 – add 'INP/volume of air'

Line 181/182 – TM (see specific comments). June 4 not 4th.

Line 192 – use scientific notation for INP/m³ (i.e. 1.47x10⁻² not 14.7x10⁻³).

Line 196 – the peak in INP_{SSA} occurred three days after INP_{SML} peaked, not one day. Unless the authors meant to suggest that INP_{SSA} only saw an increase begin a full day after the INP_{SML} peak?

Line 200 – Delete '(SI)'.

Line 204/205 – keep same scientific notation for describing cells/mL.

Line 209 – add 'Enrichment factors (EF)...'

Line 214 – delete 'next' (optional).

Line 220 – consider adding in '...positive or negative correlations...'

Line 254 – uppercase L for litre, such that 'TOC $\mu\text{gC/L}$ '. Replace 'particulate organic carbon' with POC.

Line 255 – Replace 'dissolved organic carbon' with DOC.

Line 256 – Should be '(INP per gram of TOC)' not 'OC'. Is this cumulative INP as in W15, or is this INP/mL?

Line 282 – Do you mean '...between seawater OC' or 'TOC'?

Line 291 – add space between '500' and 'nm'.

Line 294 – Only normalised size distribution shown in Figure S4, not number concentration. Perhaps add it in the graph key? Replace 'dependence of' with 'dependence on'.

Line 298 – add space between '500' and 'nm'.

Line 300 – add 'in' ahead of 'Table 2'.

Line 307 – Give correlation stats for $INP_{SSW,-16C}$

Line 351 – replace 'the' with 'that'.

Line 353 – Replace 'At this temperature, INP_{SSA} ' with just ' $INP_{SSA,-25C}$ '.

Line 361 – some overlap issue with graph and line numbering.

Line 362 – change 'R=.84' to 'R=0.84'. Check for other numbering mistakes throughout the text.

Line 380/381 – TM (see specific comments).

Line 392 & equations – Warm INP defined as $\geq -24C$, but in eq. (1) says -22. Also, in eq. (1) 'POC' should be rewritten ' POC_{SSW} ' to keep clarity (unless authors want any POC to be used in which case more explanation should be given).

Line 393 – this entire line should come before eq. (1) and (2).

Line 425 –INPSWL? Change 'INPSWL and INPSML' to ' INP_{SSW} and INP_{SML} '.

Line 430 – Is INPSSA measured at -16C or it -18C? Leave and 'and' or ',' between POC and INP.

Line 436 – '...seawater POC and SSW microbial abundance' seems redundant or repetitive.

Line 446 – it is written here 'RV' but elsewhere 'R/V'. '*Pourquoi Pas ?*' is also written differently elsewhere.