Response to RC3 “Review of ‘Ice-Nucleating Particles Near Two Major Dust Source Regions’ by Beall et al.”

We thank reviewer 3 for their helpful comments and suggestions. Below we list their comments, our responses and subsequent changes to the text and figures. Section and line numbers reflect those in the revised manuscript.

Major Comments:

1. The sampling of subsurface seawater should be a separate section in parallel with aerosol measurement.

We have restructured the Methods sections as suggested. There are now two sections in the methods about INP measurements instead of one combined:

L267: 2.4 INP Measurements in Aerosol

L398: 2.5 INP Measurements in SSW.

2. Section 3.1 is way too long. The structure and logic flow could be further improved.

To improve the manuscript flow and readability, we have updated the Results section with additional subsections including:

- L429: Results 3.1 INP Concentrations in Aerosol
- L472: Results 3.2 Seawater Source Potential
- L515: Results 3.3 Ice-active Surface Site Densities in Aerosol
- L627: Results 3.4 Characterization of INPs in Aerosol
- L675: Results 3.5 Characterization of INPs in a Soil Dust Sample
- L701: 4 Discussion

We have also reordered the results section, placing the subsection on the SSW measurements earlier on to help build evidence toward the conclusion that marine INPs were not likely observed while avoiding repetition.

3. The soil dust IN results and connection with collected air-borne samples is vague and speculative. Further evidence on chemical or mineralogy links should be presented.

We agree that the results and discussion on INP sources could have been strengthened with single particle chemical composition and minerology. These measurements were unfortunately not available for this campaign. Lacking chemical composition/minerology, we do not have evidence that the soil dust sample is representative of the sources that influenced the INPs observed. However, we think it is meaningful to apply our methodology to a regional Saharan dust sample that was used to develop the N12 parameterization, and to consider the composition of one of the only soil dust samples available from a source region indicated by FLEXPART back trajectories.

We have added the following to clarify the limitations of this comparison:

Sect. 3.5, L678: FLEXPART back-trajectories indicate this source region for several samples (f006-10, f038), though it should be noted that dust sources cannot be confirmed in this study lacking aerosol and soil dust minerology.

Specific comments

L143: Please define SSA.

This has been corrected, thank you.

Sect. 1, L146: There are now two parameterizations available for the estimation of atmospheric concentrations of marine INPs emitted from the ocean surface: Wilson et al. (2015), which estimates cumulative INPs from total organic carbon (TOC) concentrations in simulated sea spray aerosol (SSA), and …

L155-156 : Can organic compounds be heat-liable as well?
Yes, the following clarification was added:

Sect. 1, L163: "Observed n_s are compared to dust and marine INP parameterizations in Sect. 3.3, followed by an analysis of thecontributions of heat-labile (e.g., proteinaceous) and heat-stable organic compounds to observed INP populations in aerosol (Sect. 3.4)."

L175-177: Please rephrase.
This line has been rephrased as follows:

Sect. 2.2, L193: A filter flag based on aerosol measurements was derived to identify and eliminate stack emissions and was applied to all aerosol data. The filter flag was based on short term variation in particle number concentration measured by a Condensation Particle Counter (CPC, TSI model 3787), black carbon concentrations (Aethalometer, Magee AE33), wind direction and speed.

L203: Please double-check the caption of Fig. S3.
Thank you for bringing this to our attention. The caption has been corrected for Fig. S3 (now Fig. S5 after revisions)

Caption Figure S5: Time series of sea salt concentrations in aerosol. Sea salt concentrations were estimated from soluble Na^+ concentrations measured in total suspended particles (Sect. 2.2) and were used as a proxy for SSA number concentrations. Red markers show where hourly composition data was linearly interpolated for four samples where data was partially missing (samples f020, f025, f036 and f037).

L209-226: Since MERRA-2 was detailed in Gelaro et al., 2017, this section contributes little to the major findings of this study. The authors may consider moving this section to supplement.

Thank you for this suggestion. We moved several details about MERRA-2 from the main text to the supplement as suggested so that Sect. 2.3 is brief and focused on the simulation of dust.

L211: Replace x and X with ×. Also applies to L253: "-fold". Please perform a consistency check on the usage of abbreviations and symbols throughout the manuscript.

Corrected as suggested. The rest of the text was checked and corrected for this inconsistency.

Sect. 2.5, L245: "Since dust concentrations were not measured during the campaign, hourly dust surface mass concentrations along the cruise track were obtained from the (0.5 × 0.625 °) Modern-Era Retrospective analysis for Research and Application, version 2 (MERRA-2; Gelaro et al., 2017) and were averaged over the region covered during each sampling period."

L228-231: As mentioned by the other two reviewers, what is the impact of using different sampling inlets for aerosol and INP measurement?

We have added a figure to the supplement to illustrate the locations of the different sampling sites and inlets. We have also added the following clarification on the different sampling line and inlet set-ups and a comment on what the differences mean for our results.

Sect. 2.4, L268: Ambient aerosol sampling for offline measurement of INPs was conducted from 5 Jul – 31 Aug 2017 on the *Kommandor Iona's* wheelhouse top (platform above the bridge), ~15 m from the online aerosol measurements inlet and ~35 m from the ocean surface (Figs. S3-4).

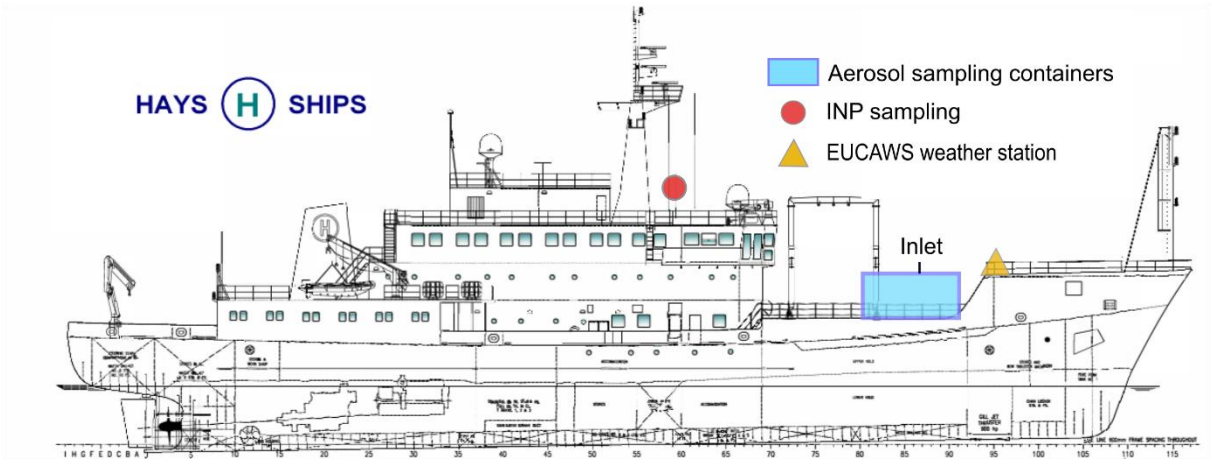
Sect. 2.4, L187: The inlet for the aerosol instrumentation was located at the top of a measurement container at a horizontal distance of about 15 m from the INP filter sampling unit **(Fig. S3)**.

(the details about the drying system in the sampling line for aerosol instrumentation remain in the following line)

Sect. 2.2, L235: "The MARGA sampling line was equipped with a PM_{10} cyclone but the sample was not dried as the instrument is not prone to condensation."

Sect. 2.4, L285: "Lacking a size-selective inlet for INP sampling, it is possible that aerosols > 10 µm were present in INP samples during dust events. Surface area may be underestimated for these samples due to the PM_{10} cutoff for aerosol sizing, but we do not expect this to affect our overall conclusions as increased aerosol surface area would further depress n_s (see Results Sect. 3.3 and Discussion Sect. 4)."

Supplementary Figure 3. Aerosol sampling, INP sampling and weather station locations.



L235 : Lpm -> LPM.
Corrected.

Sect. 2.4 L277: Aerosol sampling flow rates through the filter units were set to 10-13 L**PM** using…

L237-239: Is it possible that such an operation falsely omits the periods when terrestrial pollutants are transported to the sampling ship? The back trajectories in Fig. S9 and S10 suggest that most sampled air parcels passed through continents, which has also been stated by the authors between L419-26

It is possible that polluted air mass was occasionally omitted from sampling, but the omission of polluted air masses was unlikely to affect our results. Anthropogenic aerosols are not a known source of INPs in the temperature regime featured in this study (-6 - -25 °C) (Kanji et al., 2017). The stack emissions elimination strategy applied here may have been overly conservative because some degree of stack sampling may not have affected our results. Prior studies have demonstrated that terrestrial and marine INPs measured in a variety of environments were not influenced by pollutants (Schrod et al., 2020; Creamean et al., 2018).

The back-trajectory figures have been updated to exclude most of the free tropospheric parts of the back-trajectories, and they do not cover as much of the continents within the PBL as the previous figures showed.

L250: of collection -> since collection.
Sect. 2.4, L296: Samples were shipped in a dry shipper via Cryoport® High Vol Shipper at -180 °C and upon arrival at the laboratory were stored at -80 °C until processed, within 18 to 38 months **since** collection.

L276-278 Please elaborate the sampling setup of field blanks, e.g. how long was the sampling time instead of “momentarily”?

These details have been added to the text.

Sect. 2.4, L325: "Background n_{INP} were estimated using measured n_{INP} in aerosol sample field blanks, which had been placed in the sampling apparatus ~ 5s (without actuating the pump) before removal and unloading and storage of the filter."

L281 and L290: Please keep consistent notation throughout the paper.

The following line was corrected. The rest of the text was also checked and corrected for this inconsistency.

Sect. 2.4, L340: For this study, the detection limit was 0.68 $\boldsymbol{n_{INP}}$ mL^{-1} liquid or 0.001-0.0024 $\boldsymbol{n_{INP}}$ L^{-1} air for…

L303-305: The readers would be happy to read such discussion. Can the authors add a comment after the discussion, such as the impact of including all ambient aerosol types that are not IN active on *n*S calculation?

This discussion is included in Sect. 3.3, but we agree that it could be better linked to the n_s approximation comment in the Methods.

We have updated the following text:

Sect. 3.3, L580: Heterogeneous aerosol composition in the sampled air masses likely contributed to some of the low n_s spectra observed due to the contribution of non-INPs to the measured aerosol surface area **(see description of n_s approximation in Sect. 2.4)**. However, the difference between n_s observed during the most extreme dust events, i.e., when the aerosol population was likely approaching homogeneity in composition, and the n_s predicted from N12 and U17 was still greater than 2 orders of magnitude.

This point was also mentioned in Sect. 3.3, L549:

As noted in Gong et al. (2020), some deviations could be expected due to the difference between approximated n_s based on total particle surface area in ambient measurements and true n_s based on surface area of a homogeneous aerosol population (see Methods Sec**t**. 2.4).

L318: Do the authors explicitly state that heating reduced the IN activity of the samples in Methods?

We have rephrased this line to be neutral for the Methods section.

Sect. 2.4, L368: For each heat-treated sample, a 2 mL aliquot of the original ultrapure water suspension was heated to 95 °C for 20 min in a water bath and re-tested for changes in INP concentrations.

L322: Please rephrase.
This line was rephrased.

Sect. 2.4, L374

To remove residual H_2O_2 and prevent freezing point depression, the solution was cooled and catalase (Cat. number IC10042910, MP Biomedicals, Santa Ana, California, USA) was added.

L324: The reviewer is no expert in biology, but catalase is supposed to consume instead of being "decomposed by" H2O2? Besides, please keep consistent usage of H2O2 (and molecular representations for other species) or peroxide.

This line has been removed and replaced with references in which the procedure was described.

L348-354: Consider moving to supplement.

The FLEXPART section has been moved to the supplement as suggested.

L382-385 was left blank. Was it intentionally?

No, these spaces were placed here for the purpose of getting Table 1 on one page. The spaces have been removed.

L391: Please define "M18".

The definition has been added.

Sect. 3.3, L519: "Overall, observations nearly bridge the full regime between the parameterization for marine INPs (**hereafter "M18";** McCluskey et al., 2018c),…"

L436-437: Please rephrase. IN is more relevant to aerosol number concentration rather than mass concentration.

This line has been rephrased as follows:

Sect. 3.3, L569: "However, considering that SSA is associated with 1000 times fewer IN sites per unit surface area than dust (i.e. 1000× lower n_s) (McCluskey et al., 2018c), the characteristically low IN activity of untreated SSW (even in light of the modest changes expected from storage, Sect. 3.2), and the frequency of dust events during AQABA, our findings suggest it is unlikely that the observed INPs originated from SSA."

L453: compare well with Price et al. (2018)?

This clarification has been added.

Sect. 3.3, L593: Yet **overall**, the aerosol surface area concentrations compare very well **with those observed by (Price et al., 2018)**, indicative of comparable dustiness in the two studies.

L451-L456: Can the authors infer the mixing state of dust and marine particles based on the data?
Unfortunately, this is not possibly due to the lack of single particle chemical composition measurements, as mentioned above.

L467-468: Is dust responsible for the observed IN? It might be better to present the IN results of processed samples before discussion. E.g. L494-495.

Thank you for bringing this to our attention. This paragraph has been moved to the Discussion section after the treatment results section (Sect. 4).

L472: Please define PDF.

The definition was added.
Sect. 4, L807: "…and proposed a probability density function **(**PDF**)**-based approach to predicting INPs at a given freezing temperature."

L660: Incorrect parenthesis.
This line was corrected.

Sect. 3.2, L503: "These results are indicative of the particulate organic carbon (POC) type of marine INP defined in McCluskey et al. (2018a),..."

Fig. 2: Will it be helpful to distinguish dust- and marine-dominant aerosol population in this figure? Even though the authors declare that there is no common standard to classify dust events.

We tried multiple ways of including the dust concentration in this figure because we agree it would be nice to see, but in the end decided that including the marker color for dust concentration resulted in a figure that was too busy and difficult to understand. This was exacerbated by the fact that there was so much overlap between the high dust n_s and the low dust n_s. Although not as convenient as seeing all the features at once, we think plotting the dust concentration in Fig. 4 (as it is now numbered in the revised manuscript) improved the figure clarity. Fig. 4b and Fig. 3 can be directly compared.

Fig. 4: - L508-509: The linear gradient filling in legend marker is misleading. The readers might recognize it as a semifilled symbol.

Thank you for bringing this to our attention. The legend has been moved to the side of the figure so that the marker's coloring won't confuse the reader.

- L509: The markers for heat-treated samples are not filled.
Yes, this was intentional as noted in the caption for Fig. 5, as numbered in the revised manuscript. "Markers of heat-treated and H_2O_2-treated samples are filled to indicate significant INP concentration difference from untreated samples according to Fisher's Exact Test ($p < 0.05$)."

The unfilled markers indicate either insignificant difference according to Fisher's or the lack of data in the untreated sample (due to e.g., concentrations below or exceeding the measurement detection limit) at the given temperature.

Fig. S3: - The y label is "Sea Salt Mass Concentration" while the caption indicates "dust concentrations".

Thank you for bringing this to our attention. The caption has been corrected.

Figure S5. Time series of sea salt concentrations in aerosol. Sea salt concentrations were estimated from soluble Na^+ concentrations measured in total suspended particles (Sect. 2.2) and were used as a proxy for SSA number concentrations.

Other changes:

A line was added to better connect the point about elevation to the discussion.

Sect. 4, L779: "The differences between Price et al. (2018) and the two surface-based studies draws attention to the need for vertical profiles of n_s > -25 °C in dust-laden air masses."

A member of the INP community who saw the pre-print kindly sent us a reference reporting INP measurements from the Red Sea and Indian Ocean made in 1979. We included the reference in Sect. 3.1

Sect. 3.1, L435: "This range agrees within an order of magnitude with that of Prodi et al. (1983) who measured n_{INP} in the Mediterranean, Red Sea, Gulf of Aden and Indian Ocean nearly 4 decades prior to the present study (4×10^{-2} to 2 L^{-1} **at -16 °C**)."

References

Creamean, J M, Kirpes, R M, Pratt, K A, Spada, N J, Maahn, M, de Boer, G, Schnell, R C, and China, S: Marine and terrestrial influences on ice nucleating particles during continuous springtime measurements in an Arctic oilfield location, Atmos. Chem. Phys., 18, 18023–18042, https://doi.org/10.5194/acp-18-18023-2018, 2018.

Gelaro, Ronald, McCarty, Will, Suárez, Max J, Todling, Ricardo, Molod, Andrea, Takacs, Lawrence, Randles, Cynthia A, Darmenov, Anton, Bosilovich, Michael G, Reichle, Rolf, Wargan, Krzysztof, Coy, Lawrence, Cullather, Richard, Draper, Clara, Akella, Santha, Buchard, Virginie, Conaty, Austin, da Silva, Arlindo M, Gu, Wei, Kim, Gi-Kong, Koster, Randal, Lucchesi, Robert, Merkova, Dagmar, Nielsen, Jon Eric, Partyka, Gary, Pawson, Steven, Putman, William, Rienecker, Michele, Schubert, Siegfried D, Sienkiewicz, Meta, and Zhao, Bin: The Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2), J. Clim., 30, 5419–5454, https://doi.org/10.1175/JCLI-D-16-0758.1, 2017.

Kanji, Zamin A., Ladino, Luis A., Wex, Heike, Boose, Yvonne, Burkert-Kohn, Monika, Cziczo, Daniel J., and Krämer, Martina: Overview of Ice Nucleating Particles, Meteorol. Monogr., 58, 1.1-1.33, https://doi.org/10.1175/amsmonographs-d-16-0006.1, 2017.

Schrod, J, Thomson, E S, Weber, D, Kossmann, J, Pöhlker, C, Saturno, J, Ditas, F, Artaxo, P, Clouard, V, Saurel, J.-M., Ebert, M, Curtius, J, and Bingemer, H G: Long-term deposition and condensation ice-nucleating particle measurements from four stations across the globe, Atmos. Chem. Phys., 20, 15983–16006, https://doi.org/10.5194/acp-20-15983-2020, 2020.