

Response to RC1

The authors would like to thank Anonymous Referee #1 for their constructive and insightful feedback. 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.

Reviewer comments in black, **author responses in red.**

Minor Comments:

P6L170-174: It appears that multiple inlets were used for this study. Do all inlets come with a similar particle transmission/loss rate? Was it accounted for the size distribution data (or neglected)? Please clarify in the text.

Thank you very much for pointing out the clarification needed. We expect the overall mass loss to be below ~5%, given that the maximum of the particle mass size distribution around 3-5 μm . The following was added to the text:

Sect. 2.2, L205: “Particle losses were estimated using the Particle Loss Calculator (von der Weiden et al., 2009). Losses were negligible (<1%) up to 3.5 μm and increased to 40% at 10 μm .”

Sect. 2.2, L226: “The overall uncertainty of derived particle surface area concentrations is estimated to be 30%, **including the uncertainty due to particle losses.”**

Sect. 2.2, L237: “Particle transmission losses to the MARGA were estimated using the PLC and found to be consistent with the aerosol sizing instruments described above.”

P7L198-204: MARGA measured TSP but it seems other measurements (i.e., FMPS and OPC) assessed particles up to 10 μm . Were there any >10 micron diameter particles measured during the campaign? Since the authors are adapting the MERRA-2 reanalysis data for the range of 0.1 – 10 micron (P2L49 & P8L220), further justification in the detected size consistency seems necessary here. Please elaborate.

The reviewer is correct in pointing out an inconsistency in the INP sampling set up and the aerosol sizing/TSP measurements. The MARGA sampling line was equipped with a PM₁₀ cyclone. The aerosol sizing sample inlet was not size-selective, however, as a consequence of selection of particle size range for the PM calculation, the PM results are PM₁₀. The INP sampling was performed using an open-face filter unit, with no inlet or size-cutoff applied. We added the following to clarify our assumptions in comparing the sizing/TSP data with the INP data. I also see that clarification on the MARGA size cut-off is needed:

Sect. 2.2, L228: “The water-soluble fraction of total suspended particles (TSPs) was monitored with hourly resolution using a Monitor for AeRosols and Gases in Ambient Air, MARGA (Metrohm Applikon model S2, Herisau, Switzerland).”

Sect. 2.4, L285: “Lacking a size-selective inlet for INP sampling, it is possible that aerosols $> 10 \mu\text{m}$ 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 (Sect. 2.2 and S3), but we do not expect this to affect our overall conclusions as increased aerosol surface area would further reduce n_s (see Results Sect. 3.3 and Discussion Sect. 4).”

P7L196-198: How did the authors estimate this overall uncertainty of 30%? The reviewer assumes this is some sort of systematic error. Please clarify in the text.

This estimate was derived from analysis of several factors that contribute to overall particle surface area concentration uncertainty, including : inlet and transport losses, re-binning uncertainty, FMPS scaling uncertainty and uncertainty due to counting statistics. These details have been added to the supplement Sect. S3.

What were the measurement time resolutions of OPC and FMPS? Were the data time-averaged afterward, and that is what is reported in Table 1 for aerosol surface area concentration?

FMPS and OPC measurements had a 1s and 6s time resolution, respectively. Measurements from both instruments were averaged over 1-minute intervals. For the “Aerosol Surface Area” column in the Table 1, the total surface area is calculated by summing up the 1-min surface area concentration data over the sampling period and dividing by the INP sampling volume, so the value in the table is a time averaged surface area over the INP sampling period.

The following was added to the text to clarify:

Sect. 2.2, L185: “The OPC measures particles in the size range $0.25 - 32 \mu\text{m}$, and the FMPS measures particles with sizes between 5.6 nm and 560 nm with a 6s and 1s time resolution, respectively.”

Sect. 2.2, L192: “OPC and FMPS data were averaged over 1-minute time intervals.”

Sect. 2.2, L185: “Particle surface area concentrations were derived from the 1-min time-averaged FMPS and OPC measurements as follows.”

Were the statistical relative deviations of surface area concentrations within 30% over individual sampling times?

Yes, the 30% overall uncertainty is inclusive of statistical relative deviations from counting statistics. This is explained in the details that have been added to the supplement Sect. S3 (see the end of Sect. 3.5).

“Therefore, we estimate that the overall counting statistics-related uncertainty of the particle surface concentration is in the order of 5-7% for 1-minute averages.

Considering all the components to the overall particle surface concentration uncertainty described above, we conservatively estimate the uncertainty of the particle surface concentration to be 30%.”

Table 1: Each sampling interval covers several hours. Air masses came from a similar source throughout individual sampling periods? The authors may consider showing the min-max ranges of surface area for each timestamp or some sort of data deviation range. They may help clarify to the readers if air masses were consistent over each sampling interval (or not).

Thank you for this idea. Minimum and maximum aerosol surface area were added to Table 1. The range was fairly consistent, $<320 \mu\text{m}^2 \text{cm}^{-3}$ for all samples except for f024, for which the range was $>600 \mu\text{m}^2 \text{cm}^{-3}$.

The following was added to the text:

Sect. 3.1, L455: The ranges of aerosol surface area concentrations for all sampling periods were $<320 \mu\text{m}^2 \text{cm}^{-3}$, with the exception of f024, for which aerosol surface area range was $>600 \mu\text{m}^2 \text{cm}^{-3}$ (Table 1).

Have the authors analyzed the correlation between INP concentration and ambient meteorological conditions? Were there any precipitations during the campaign, where rain may have washed out the dust and fresh particles (thus, showing low INP conc.?)?

Since there were relatively few data points during this cruise campaign (26 unique datapoints for INPs at a given freezing temperature), we did not attempt an analysis of correlations between meteorological conditions. A prior study that undertook such an analysis featured 2 years' worth of measurements at a single site (Stopelli et al., 2016). There were no precipitation events during the campaign along the cruise transect or FLEXPART back trajectories.

Sect. 2.3. What is the uncertainty involved in the estimation of reported dust & sea salt mass concentrations? The MERRA-2 spatial scale seems big, and the reviewer wonders if such large-scale data can represent the pin-point dust concentration at the sampling point during the authors' campaign. Is the MERRA-2 data sensitive and representative of the surface dust concentration at the sampling height that the authors employed?

Thank you for raising this point. There isn't yet a specific uncertainty estimate to apply to MERRA-2 dust mass concentrations. Deriving bias and uncertainty from evaluations of model products could be a subfield of the modeling domain, though this analysis is being done for a select few MERRA-2 products (Liu and Margulis, 2019). To further support the use of MERRA-2 for dust mass concentrations, we added the following text and figures to Sect. 2.3 and the supplement.:

Sect. 2.2, L248: "Buchard et al. (2017) showed a high degree of correlation between MERRA-2 and surface dust concentration observations ($r \geq 0.69$), particularly during dust storms ($r \geq 0.92$). MERRA-2 surface dust mass concentrations also correlated well with PM_{10} observed during AQABA ($r \geq 0.71$) (Fig. S6)."

Supplementary Figure 6. Comparison of MERRA-2 surface dust mass concentrations and observed PM₁₀ during INP sampling periods.

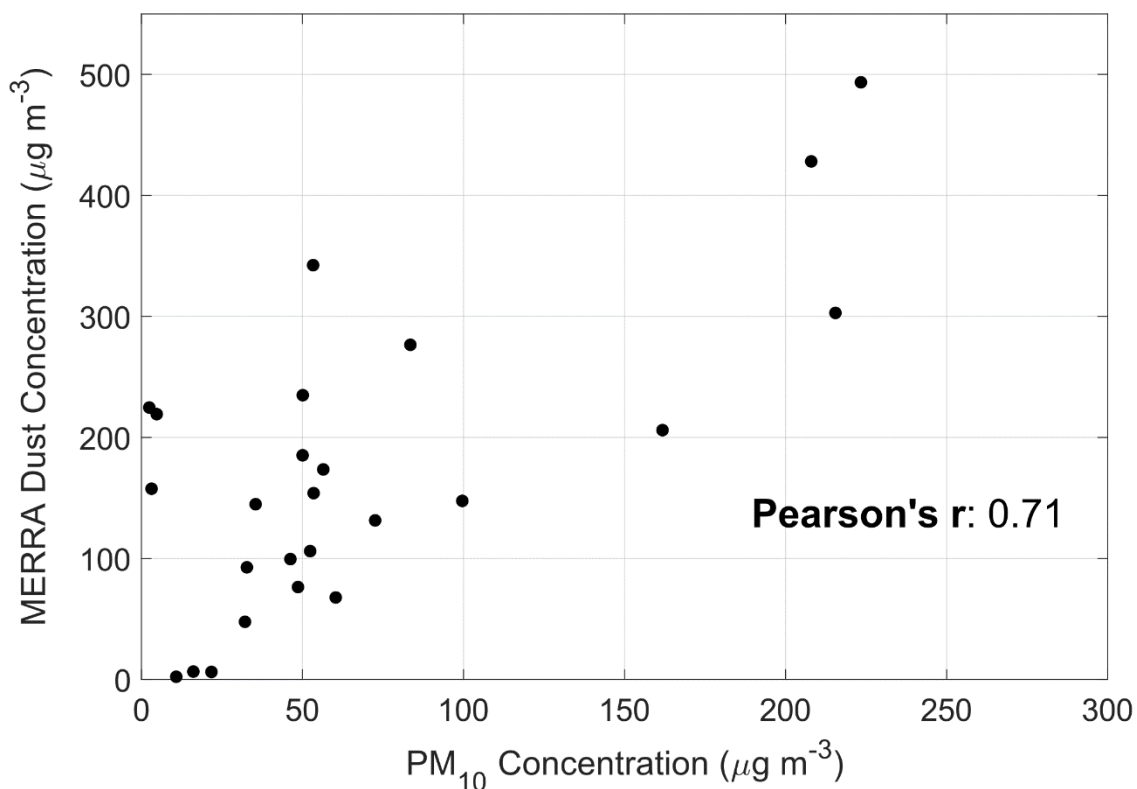


Fig. S5: There seem some gaps between x1 and diluted results beyond the CI95% (e.g. f035, f042). Which n_{INP} (upper or lower bound) would be representative for the overlapping T region (i.e., ~ -10 to -15 dC)?

For the freezing temperature ranges where there is overlap between the diluted and undiluted spectra, most of the 95% CI overlap, but yes, there are one or two data points in f035, f042, f044 where the confidence intervals do not overlap. We consider both values from the diluted sample and the undiluted sample, and these are shown in Figs. 2-3. In the case of Fig. 1, the value for INPs at -15 °C as measured in the undiluted sample is shown, and thus could be considered conservative for f035 and f042. Since -15 °C was beyond the limit of detection in the undiluted f044 sample, the diluted sample value for INP concentration was used.

We have made the following updates to the text.

Figure 3 caption: “For the 8 samples on which a dilution was performed (Fig. S8), n_s for both the raw undiluted and diluted sample are shown.”

Figure 4 caption: “For the 8 samples on which a dilution was performed (Fig. S8), n_s for both the raw undiluted and diluted sample are shown.”

P11L319-P12L325: How was this H₂O₂ treatment protocol developed? The reviewer thinks that it will be informative for the reader to know how the 1.6:0.8 mL ratio and this particular concentration of H₂O₂ were selected to be used in this study. It would be nice to have a reference here if the same procedure has been used in a previous study. Otherwise, please explain.

Thank you for pointing out this omission. A concentration of 10% was chosen to provide a generous excess of oxidant. A recent study found that occasionally the organic material requires longer than 20 minutes to further decompose and suggested that this may have been due to the protection of organic matter within aggregates, their adsorption onto mineral surfaces, and/or the inability of the H₂O₂ to oxidize organo-mineral complexes and chemically resistant compounds such as black carbon and aliphatic hydrocarbons (Testa et al., 2021). Thus, the methods applied in the present study may occasionally result in an underestimate of the contribution of organic INPs.

The following has been added to the text:

Sect. 2.4, L364: “Heat and peroxide treatments were applied to a subset of samples (12 of 26) to test for heat-labile biological (e.g., proteinaceous) and organic INP composition, respectively, following the procedure described in McCluskey et al. (2018b) and Suski et al. (2018).”

P12L323-325: Does the catalase-added H₂O₂ suspension blank have a similar background freezing spectrum as compared to the field blanks? There is no suppression of background INP in pure water by these catalysts, correct? Perhaps, the authors can add the spectrum of the H₂O₂-treatment blank on Fig. S6 or may consider showing it elsewhere in this manuscript.

Thank you for this suggestion. Yes, the H₂O₂ + catalase suspension has a similar background freezing spectrum. The following has been added to the main text and supplement:

Sect. 2.4, L379: “Figure S7 shows the estimated n_{INP} in a heat and H₂O₂-treated blank sample.”

Supplementary Figure 7. INP concentrations measured in 26 aerosol samples collected during AQABA, in simulated INP concentrations from 7 blank filter samples, and simulated INP concentrations in a heat-treated and H₂O₂-treated blank filter sample.

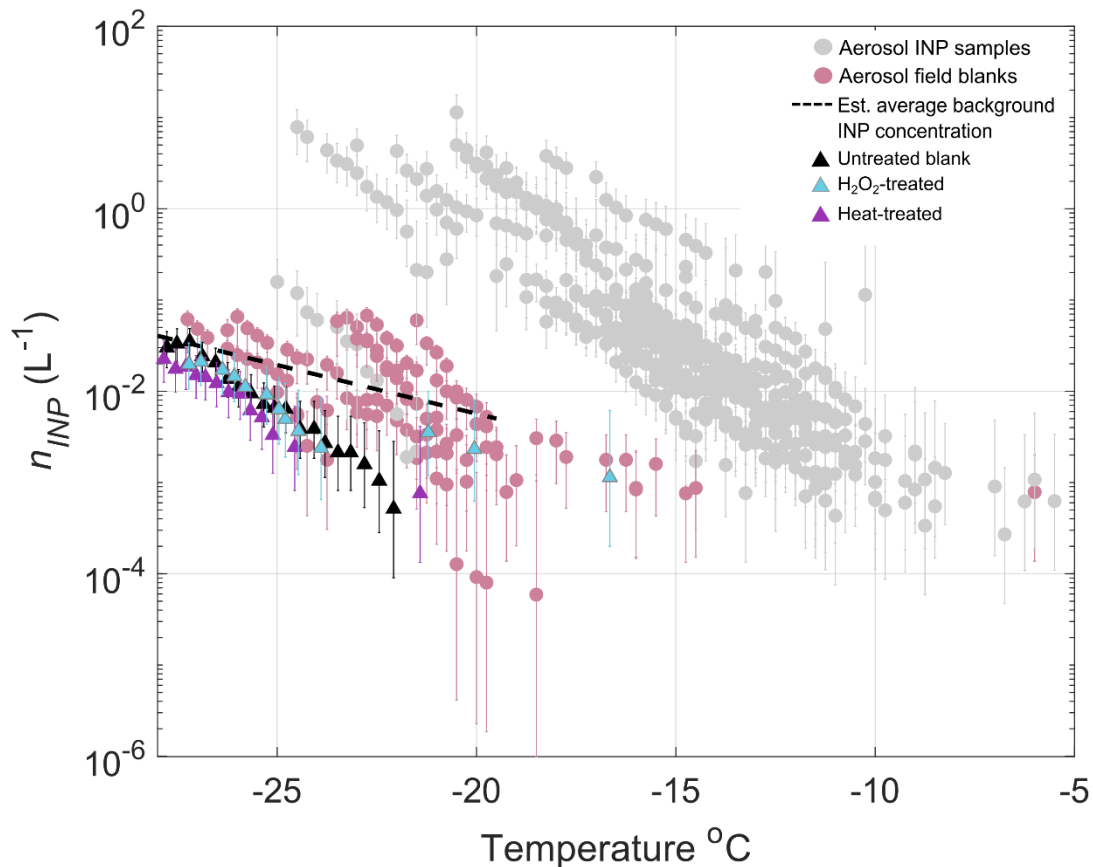


Fig. 2: This reviewer encourages the authors to provide the error bars for the observation data (at least to some representative data points at higher and lower ends of T). They do not need to be statistical uncertainties. Can be systematic uncertainties instead.

Fig. 3 has been updated with error bars.

Fig. 3 caption: “Error bars represent 95% binomial sampling confidence intervals (Agresti and Coull, 1998).”

P20L482-483: How were these 12 samples selected? The author may provide a brief explanation here or in Sect. 2.4 (~L315).

The following was added to Sect. 2.4:

Sect. 2.4, L366: “The 12 samples were selected based on sampling location with the aim of getting a representative measurement from each region.”

P21L501-504: Interesting. This release/exposure of INA core upon an application of heat can seemingly be a good future study topic in the IN research community. The authors may consider mentioning this somewhere as one of the outlook study topics.

Thank you for this idea! We agree.

The following has been added to Sect. 3.4, L653:

“However, increases in IN activity after heat treatment have been reported previously for airborne Saharan desert dust and aerosol collected during Saharan dust intrusions (Boose et al., 2019; Conen et al., 2022) as well as SSA and precipitation (Martin et al., 2019; McCluskey et al., 2018a) and should be further investigated in future studies.”

P31L718: The authors may consider adding “potentially” in front of enabling. The proposed tagging would not warrant to link INP and aerosol properties as aerosol composition is not necessarily identical to INP composition. Perhaps, more reasonable properties to link in this context would be the relationship between ice crystal residual composition, n_s , and n_{INP} . Knowing that air masses are typically influenced by dust and maritime source (P5L149-150), other physicochemical properties of particles (e.g., mixing state) may play a substantial role over aerosol composition.

We agree. Pulling the INP signal out of the total aerosol composition data is a tremendous challenge, so winnowing down the data with ICR composition is a good strategy. To avoid defining ICR so late in the text, we have simply added “potentially”.

Sect. 5, L932: “This information could be used to potentially “tag” different classes of organics and biological aerosols, enabling investigations of relationships between n_s , n_{INP} and organic signatures in, e.g., mineral dusts and agricultural soil dusts.”

Fig. S12: The authors may include the explanation of the difference between open and solid symbols in the figure caption. Please clarify why the solid purple symbol data are not available for s007. Also, why does the highest T data point in s001 at $T > -15$ dC has a higher c_{INP} than the next data point at T of < -15 dC?

The following was added to the S13 caption:

“Markers of heat-treated, filtered and H_2O_2 -treated samples are filled to indicate significant INP concentration difference from untreated samples according to Fisher’s Exact Test ($p < 0.05$).”

With regard to s007, you can see in this figure the two open purple data points near -25C. In this sample, all of the INPs measured between -12 and -21 in the untreated sample were eliminated after heating, an example of high heat-sensitivity. The reason the markers are not filled are that we cannot apply Fisher’s Exact Test where data for the sample to be compared (in this case the untreated sample) are unavailable, usually at freezing temperatures above or below detection limit.

The decrease in n_{inp} between the two data points the reviewer points out here is a result of the correction applied for the instrumental blank. At the lower temperature, 1 or more of the aliquots of ultrapure water used as a control were frozen whereas at the higher temperature, none had yet been frozen. L321 from 2.4 may be helpful:

“Prior to calculating n_{INP} , the fraction of unfrozen wells (f) was adjusted for contamination in the water used for suspension by subtracting the number of frozen ultrapure water wells per temperature interval from both the total number of unfrozen wells and total wells of the sample.”

Technical comments

P1L1 vs. P2L35: ice-nucleating particles vs. ice nucleating particles – the reviewer suggests the authors be consistent in this terminology.

Thank you. P2L35 has been corrected to ice-nucleating particles.

P2L47 vs. P11L295: ice nucleation site densities vs. ice-active surface site density – the authors may consider using consistent terminology.

L295 has been corrected to “ice-active surface site density.”

P2L50 vs. P3L72: ice nucleation (IN) vs. ice-nucleating (IN) – please be sure the abbreviation is consistent throughout the manuscript.

This instance has been corrected to ice-nucleating. The rest of the text was checked and corrected in a few other instances of “IN properties”.

P2L55-57: The reviewer is a bit confused here – the point that the authors want to make is that the ice nucleation active (INA) organics are limited in terms of quantity as compared to INA minerals, but it dominates the ice nucleation at T above -15 dC; therefore, they are important, correct? Currently, it sounds like organics are less important as INA component of aerosol particles than minerals by reading this part alone. The authors may rephrase this sentence accordingly.

Good point – I see how this could be confusing. Since the important point about the dominance of organics at T > -15 dC is made a few lines above, this sentence was deleted.

P6L154: `

Removed, thank you.

P9L256: SIO – abbreviation

Corrected.

L303: INP concentrations were measured using the Scripps Institution of Oceanography Automated Ice Spectrometer (SIO-AIS), an immersion freezing droplet assay instrument that is described in detail in Beall et al. (2017).

P11L306: ice nucleating à ice nucleation active

Corrected.

L353: “The difference between the n_s approximation (Eq. 4) and n_s (Eq. 3) is that many particle types are typically included in the n_s approximation, and in an ambient aerosol measurement most of these are not IN-active”

P19L452: S7 appears after S8 & S9 (P13L372). Please fix the figure number sequence.

All figures have been renumbered.

P27L611: ??? what does (6-20X)X mean?

Typo fixed and clarification added.

Sect. 4, L811: Prior studies of aerosolized dust demonstrated that it is frequently enriched in organic matter (6-20×) **compared to soil dust** and that wind erosion selectively removes the chemically-enriched, fine portion of the soil higher in plant nutrients, organic matter and metals (Aryal et al., 2012; Delany and Zenchelsky, 1976; Van Pelt and Zobeck, 2007).

P27L615-616: 155,000 in what physical unit?

Thanks for catching this.

Sect. 4, L815: “Furthermore, a recent study that measured airborne concentrations of prokaryotic cells over the Red Sea characterized the region as a “global hot spot” with average concentrations of 155,000 ($\pm 65,000$) **cells m⁻³**, 19× higher than that over the subtropical and tropical open oceans (Mayol et al., 2014; Yahya et al., 2019).”

P27L620-623: The reviewer likes this statement. This is one of the well-summarized take-home messages.

Thank you!

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

Testa, Baptiste, Hill, Thomas C J, Marsden, Nicholas A, Barry, Kevin R, Hume, Carson C, Bian, Qijing, Uetake, Jun, Hare, Hannah, Perkins, Russell J, Möhler, Ottmar, Kreidenweis, Sonia M, and DeMott, Paul J: Ice Nucleating Particle Connections to Regional Argentinian Land Surface Emissions and Weather During the Cloud, Aerosol, and Complex Terrain Interactions Experiment, *J. Geophys. Res. Atmos.*, 126, e2021JD035186, <https://doi.org/https://doi.org/10.1029/2021JD035186>, 2021.