

Atmos. Chem. Phys. Discuss., referee comment RC2
<https://doi.org/10.5194/acp-2021-1101-RC2>, 2022
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Comment on acp-2021-1101

Anonymous Referee #2

Referee comment on "Ice-nucleating particles near two major dust source regions" by
Charlotte M. Beall et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2021-1101-RC2>, 2022

Review of Beall et al. 2022: Ice-Nucleating Particles Near Two Major Dust Source Regions

The study by Beall et al. 2022 evaluates INP concentrations in ambient air and sea water samples collected during the ship-borne AQABA campaign in the Red Sea, Gulf of Aden, Arabian Sea, Arabian Gulf, and part of the Mediterranean. The authors investigate changes in observed INP concentration with respect to origin and nature of the observed INPs. The authors put a specific emphasis on the discrimination of dust and marine contributions to the observed INPs in the air and underline the importance of organic and proteinaceous compounds on dust particles to their ice nucleation ability. The observations, analyses, and conclusions are of great interest to the scientific community and deserve publication in ACP. However, before publishing the study, the reviewer suggests addressing different general and specific comments, which hopefully help to improve the quality and impact of the manuscript.

LXXX refers to line XXX in the manuscript.

General comments:

Overall, the manuscript is a bit long-winded owed to the fact of an extensive analysis and many links to existing literature – what the reviewer highly appreciates. At some points the manuscript seemed repetitive (see specific comments below) and the readability/overview of the manuscript should be improved introducing more subsections (so far there are only two subsection titles for 17 pages of results) guiding the reader

through the manuscript. Furthermore, (to the reviewer) the climax of the manuscript is reached with the conclusion of different parameterizations needed for fresh and aged dust, respectively (L620-623). Afterwards, the results continue, however, with the analysis of the sea water samples, which are also used to assess the partitioning of INPs arising from desert and sea sources, a discussion that had previously been addressed using different proxies. The authors may want to cover the discussion of the sea samples earlier in the manuscript when discussing the source partitioning and in order to work towards the conclusion L620-623. This could potentially increase the readability of the manuscript, but the reviewer leaves the decision of changing the manuscript structure up to the authors.

The reviewer highly appreciates the effort of the authors to apply their methodology also to a Saharan dust sample of the previous study by Niemand et al. 2012. What would be valuable is to let the reader know how the results in this study compare to the initial measurements of Niemand et al. 2012. Was the same ice nucleation (IN) activity observed? Could storage effects (e.g., Beall et al. 2020, Stopelli et al. 2014) have altered the observed IN activity? From the reviewer's viewpoint this is essential information to assess how representative the reduction in IN activity due to the two treatments is, which is later used to support the conclusion for the need of different dust parameterizations. In addition, the sample preparation for analysis is not described in the manuscript and should be added in the methods section. Furthermore, the sample's association to the study by Niemand et al. 2012 should be indicated at all instances in the text and in the caption of Figure 5 for clarity. The authors may consider plotting the original INP concentration observed by Niemand et al. 2012 to Figure 5.

Generally, storage effects on both the obtained filter and seawater samples seem to not be addressed in the manuscript. Given the storage of the filters of up to more than three years (L251), had a change in IN activity been observed, e.g., a decrease in average IN activity with storage time? Could a potential decrease due to storage also explain the reduced INP concentrations in the samples compare to previous work? This should be added to the discussion and be potentially mentioned in the conclusions, where the possible explanations for the reduced observed INP concentrations are listed. Furthermore, how long have the sea water samples been stored before analysis?

It is not entirely clear to the reviewer why the MERRA re-analysis data was chosen to assess the dust mass concentration albeit aerosol in situ observations were available on the ship. It is understood that using MERRA source regions could potentially be identified and the aerosol classes allow for the discrimination of different sources. However, these capabilities seem to not be used (for the source apportionment FLEXPART is used, for discrimination the sources it appears an AMS and a MARGA was used). Furthermore, from Table 1 it seems that the re-analysis average dust concentration did not necessarily relate to the in situ aerosol surface area. If the re-analysis is representative, a potential relation should exist to PM_{10} derived from the in situ measurements assuming a representative dust density. To support the representativity of the re-analysis data, this relation should be explored and supported with e.g., a correlation coefficient. Would the usage of an in situ derived PM_{10} compare better to the INP concentration (cf. Figure S7)? If so, the usage of an in situ derived PM_{10} might be more appropriate. The authors may have addressed this approach in their analysis already. I would suggest elaborating briefly on their decision from where to take the dust concentration in the manuscript.

Throughout the manuscript, the authors need to perform a delicate balance between the analysis of individual samples and the implications to the overall big picture. At some locations in the manuscript very specific and detailed information of individual samples are given without putting it into context with other samples or the big picture (e.g., for L661-663, while the relation to the study of McCluskey et al. 2018 is very interesting to an INP researcher, indication in only one observed sample does not fully allow general conclusions). The authors may consider reading critically through their manuscript, removing side thoughts to streamline the manuscript. The reviewer acknowledges that the importance of such small connecting statements is subjective and leaves it up to the authors to assess their importance.

The presented supplementary information is appropriately chosen to support the understanding of the manuscript while not interfering with the main storyline of the manuscript. Despite being supplementary, the material should be presented in a more structured way, i.e., all figures should feature a corresponding caption beneath, and – in the reviewer's opinion – a brief explanatory description as text should be given. Additionally, the reviewer encourages the authors to revise some of the figures in terms of readability, consistency, and quality. Specific comments follow below.

Moreover, the reviewer sees generally the need for more consistency throughout the manuscript in both the text and figures. Specific comments follow below.

Specific comments:

Abstract

No finding related to the SSW samples is mentioned in the abstract. The authors may consider adding an obtained insight from these samples.

L49: It should be annotated that the dust concentration was obtained from re-analysis data in contrast to the in situ measured INP samples.

Introduction

L120: The reviewer enjoyed the rather extensive but nicely written introduction to INP parameterizations.

Methods

L174: Different instruments were connected to different inlets. How do the different inlets compare, were the sampling lines similar per instrument? Furthermore, it would be very beneficial to have an overview picture/schematic of the different sampling sites to see their relative locations, e.g., are there any obstacles between the different inlets?

L176: The authors may want to consider adding the range of typical RHs during the campaign.

L177-L182: It would be beneficial to briefly explain the used thresholds/logical arguments of the filter flag.

L187-L188: Is there a reference for the mass ratio assumption? The authors may consider adding the values of used refractive indices and shapes, including a reference if possible.

L191: Is there a reference for the AMS data obtained during AQABA?

L198: How was the uncertainty of 30% derived? Please elaborate.

L208: Here again, an overview graphic (in the main text, supplementary, or referenced from a different publication) would strongly help to understand the relative locations.

L211: What is meant by "area-averaged"? Please elaborate.

L230: Are the ~25 m displacement vertically or horizontally?

L233: What determined the sampling length? What determined the sampling frequency? Please elaborate.

L238-L242: Just to ensure: The relative wind direction towards the sampling unit has not been used to switch off the sampling pump?

L253: How was the shaking realized?

Eq. (1) + E1. (2): Consider adding a reference for the equations.

L287-L289: Given the spread of two orders of magnitude of the background INP concentration, the reviewer does not fully agree with the used background correction methodology. The degree of contamination of the field blanks when inserting them in the sampling unit and removing them could likely be a function of the ambient aerosol concentration while taking the field blank. Thus, subtracting the timely closest background from a sample might be more meaningful. However, the reviewer acknowledges that this change would not substantially affect the background corrected INP concentrations and does not need to be implemented.

L293: When overlapping, were the INP concentrations from diluted samples combined with the INP concentrations from the undiluted samples? If so, please specify.

Eq. (3) + Eq (4): While Eq. 3 and Eq. 4 are presented in Kanji et al. 2017, a reference like e.g., Hiranuma et al. 2015 explaining the approximation to A_{tot} might be more helpful to the reader.

L315: What was the motivation/criteria for the selection of the specific 12 samples? Please elaborate.

L315-L338: Has this procedure/protocol of heating and hydrogen peroxide treatment been previously described? If yes, the authors may want to shorten the description and cite the existing literature.

L345-L346: Also here, how were the 5 samples selected? Please elaborate.

Line 349ff: How frequent were back trajectories released, how many were investigated per filter sample, and which time (e.g., start or end time of sampling) was considered for choosing the displayed trajectories? Is there a reason for choosing 72 hours? Furthermore, the authors may consider indicating the trajectory locations in plots S8 and S9 where the trajectories were below a certain threshold height indicating potential air mass uptake.

Results

L415: It could be insightful to add a flag to Table 1 to indicate which samples are thought to be sampled during dust events. Otherwise consider changing "(Table 1)" to "(dust concentration > 50 $\mu\text{g m}^{-3}$, see Table 1)"

L415-L416: How was a high probability of dominant dust contributions determined? To the reviewer's understanding this assessment is based on increased PM_{10} loadings of the re-analysis data. However, it seems that the reanalysis data does not necessarily correlate with the in situ observed particle surface area concentration (Figure S7). Has the relation between re-analysis data and in situ observations been investigated to support the assessment of dust contributions solely on re-analysis data? If no relation prevails the reviewer would suggest tempering the statement from high probability to likely given a potential absence of in situ increased concentrations.

L428+L429: In Table 1 the seasalt concentration is given. If this is used as proxy for SSA, the authors should specify this and explain their reasoning for choosing this proxy.

L437-L439: The higher relative abundance of dust over sea salt concentration seems to be the essential part of the last two paragraphs. If the authors see a possibility, they could abridge their argumentation in L415-L443 to reach this statement.

L450: What is meant by efficiencies? Please elaborate.

L455: The exclusion of the sample by Price et al. 2018 seems a little bit arbitrary. Could the authors add the observed surface area of the excluded samples and the maximal observed surface area during the present study to relate the two observational ranges?

L459: Is there also a possibility for regional difference affecting the aerosol/INPs differently (Price et al. 2018 sampled at the western end of the Sahara)?

L492+: In the following, is sensitivity referred to as degradation? The authors may specify that.

L543-L546: Is there a plot supporting this argument?

L550-L560: This passage has more introductory character. The authors may consider moving it to the introduction

L585: Cite the referred studies.

L626: Does Figure 5 refer to the soil sample used previously in the Niemand et al. 2012 study? What about the ambient aerosol observations during the research cruise? The authors may consider including the results presented in Figure 4 to the discussion here.

L644: Add a reference for the SSW samples collected at the Scripps Memorial Pier. If the data has not been published yet, the authors should add a description of the sampling and analysis in the methods section.

L657: Why were not all samples processed? Please elaborate here or in the methods section.

L661-L663: The reviewer is not fully sure what this result adds to the discussion. The authors should consider elaborating on this and highlight how the observation in an individual sample relates to a bigger implication.

L670-671: The authors may consider adding the total number of observed samples.

Conclusions

L678: The authors should add a quantitative statement on what is meant by "showed agreement" for the different data they refer to.

L685: The authors may consider stating explicitly the temperature ranges they refer to in the conclusions.

L685-L689: To the reviewer this statement has rather introductory than concluding character. The authors may consider either moving the sentence to the introduction or shortening the statement and directly combining it with observation of the study.

L689-L691: The authors may add that the analyzed sample originates from the analyzed samples in Niemand et al. 2012.

L698: Also here, what is considered a well agreement? Agreement within an order of magnitude? Please support with a quantitative statement.

L700-L707: In the reviewer's opinion, the possibility of sample degradation and the potential impact of sampling close to the ground (as discussed in the results) should be added.

L709: The authors may want to add a brief explanation of the working principle/data basis of the methods by Gong et al. 2020 to the sentence.

Editorial notes:

L37: The authors may consider changing "reflectivity, and precipitation efficiency" to "radiative properties, and precipitation initiation efficiency", to avoid using reflectivity, which is often connotated to radar observations, and be more specific about the role of INPs towards precipitation. This comment also applies to other occurrences in the manuscript.

L47: Increase consistency: sometimes e.g., "1-3 orders of magnitude" is used, sometimes "one to three orders of magnitude".

L50: Despite standard in our community, the authors may want to specify the usage of "hydrogen peroxide". In the manuscript, different versions (peroxide, hydrogen peroxide, H₂O₂) – the authors may use one version for consistency.

L51: Consistency: here \geq is used, whereas in L56 and L58 $>$.

L57: Consistency: modest (three instances) or moderate?

L67: Consider adding "primary" to "ice formation".

L73: The long-term INP observations of Brunner et al. 2021 may be an additional suitable

reference for this statement.

L76: Consider adding "average" to "global dust loading".

L79: emissions \square emission.

L89: Consider adding "by number" or "by mass" after "dominant".

L111: Consider introducing A13 here as well.

L112+L114: Add 'hereafter,' before introducing the abbreviations of N12 and D15.

L121: Consider adding "in situ" before "INP measurements".

L126: Consider adding "west" before "of the Sahara".

L144: "SSA" not defined yet.

L149-L159: If further subsections are added to the manuscript, the different sections could be added here after the individual analysis steps to allow easy finding of specific parts of the manuscript.

L152: Consider adding "ambient" before "aerosol sample".

L156: Leave out " ' "

L190: d_{opt} , d_{geo} , and PM_1 are not defined yet.

L198: Consider adding "geometric diameter" after " μm ".

L254: At different instances “-fold”, “x”, or “-times” are used. Consider using one version throughout the manuscript.

L256: SIO not defined.

L265: In the reviewer’s perception, the ‘INP’ subscript should not be in italics, i.e. n_{INP} . This applies throughout the manuscript, including the figures. Further, the authors should consistently use n_{INP} or INP concentration in the text.

L267: Consider adding “drop” before “volume”

L273-L290: Is n_{INP} in bold font intended?

L282: When comparing to Figure S6 the concentrations seem different at -20 °C.

L285: The linear regression seems to have been done on the log-transformed data. If so, please specify.

L314: °K □ K

L328-L331: Maybe add that the results of each test are indicated in the corresponding figures.

L362: The referred abbreviation PM_{10} does not appear in Table 1.

Table 1: Remove “unt” from sample ID to stay consistent with text. Consider adding (°N) and (°E) to latitude and longitude. At what sampling time are the presented latitudes and longitudes (start or stop datetime?). To enrich the information of the table, the authors may consider adding the INP concentration at an exemplary temperature.

L392: Has M18 already been introduced?

L396: Did the authors mean to refer to Yang et al. 2020?

L397+L398+L401: The authors may consider adding the sample IDs of the mentioned samples/spectra.

L403: It may not be entirely clear to which study "same region" refers to.

L433: DMSO₂ and VOCs remain undefined.

L452: Figure S7 should be number S9.

L467: DeMott et al. 2015a □ DeMott et al. 2015

L533+L595: Consistency: change "dusty" to "dust-laden".

L534: "N." □ "North"

L565+L566: Consider rephrasing the citation text ("e.g.") as it is not directly evident what statement the citation supports.

L569: The authors consider adding exemplary references for the mixed and contradictory results already here.

L581: Both ">" and "below" is used. For consistency the authors may want to use </> or below/above consistently throughout the manuscript.

L611: Remove second "x".

L612: Remove whitespace.

L616: Missing unit after "(±65,000)".

L620: For Kanji et al. 2017 being a summary, consider putting it last in line and add "and references therein".

L629: What are the temperature ranges considered "high" or "moderate"? The authors should consider adding the temperature ranges here, and/or explicitly introduce the referred temperature ranges earlier in the manuscript.

L660: "POV" was already introduced.

L663: "DOC" was already introduced.

L670: Consider adding "Observed" before "INP concentration".

L675: The authors may consider adding that the PM₁₀ observations were obtained from re-analysis.

Editorial notes on the figures:

Generally, many of the figures (e.g., Figure 3, 4, and 6) feature strange gray lines not belonging to the plot itself. Further the indication of units is inconsistent, e.g., "°C" after "Temperature" is never set in brackets. Legends that apply to different subplots could be put aside of the subplots and not within one subplot. The reviewer kindly asks the authors to check all their figures (including supplement) to enhance their quality.

Figure 1: Datapoints at 25°N/35°E are overlaying and barely visible. Change order or add some alpha to scatters. "µm" in colorbar.

Figure 2: The readability of the plot may be increased when using generally open symbols in combination with some alpha.

Figure 3: Mention the errorbars in caption. Maybe mark sample f020 in plot (a) as reference. Consider stretching the y-axis limits in (b).

Figure 4: The labels of the subfigures need to be increased. "INPs" to "INP concentrations" in caption.

Figure 6: Shouldn't the y-label read $n_{\text{INP,L}}$? "INP spectrum" to "INP spectra". Also name the number of samples from the other studies in the caption.

Figure S1: It seems that f006-f009 and f018+f019 overlap. Were several sampling units running at the same time? If so, this should be mentioned in the methods section.

Figure S3: The authors may consider reducing the dot size for readability.

Figure S5: In consistency to Figure 5, the dilutions could be indicated with a different marker than undiluted samples.

Figure S7: The labels are rather small and could be increased in size. What correlation coefficient was used? To the reviewer's perception Spearman's correlation coefficient should be used. The reviewer is not fully sure about the expressiveness of the figure. Maybe a table with Spearman correlation coefficients between the INP concentration and both surface area and dust concentration at all temperatures could help for a more complete picture.

Figures S8+S9: The authors may consider indicating only locations of the trajectories below a certain height threshold.

Figure S9: To add information, the cross of the missing sample could be colored according to the observed dust concentration.

Figure S11: There seems to be data gaps, which are indicated in black as is the land contour. Color either the land or data gaps in a different color. Remove gray lines around colorbar. Add shown quantity to the colorbar.

Figure S12: Shouldn't the y-label read " $n_{\text{INP,L}} (\text{mL}^{-1})$ "?

References

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