Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC1
https://doi.org/10.5194/acp-2022-21-RC1, 2022

Comment on acp-2022-21

Anonymous Referee #1

Referee comment on "Predicting atmospheric background number concentration of ice nucleating particles in the Arctic" by Guangyu Li et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-21-RC1, 2022

Review of “Predicting atmospheric background number concentration of ice nucleating particles in the Arctic” by Li et al., submitted to ACPD

In this study, data from two measurement periods in Ny-Alesund (Svalbard) are used. Collected data concern mainly INP concentrations (both in-situ and off-line analysis), as well as some aerosol properties (size distributions for both seasons, fluorescence signals for one season). It is argued that INP concentrations cannot be described well based on the aerosol properties, and a new INP parameterization is derived from the data, given as a line, similar to the Fletcher or the Cooper curve, together with a 95% confidence interval (covering roughly one order of magnitude).

The work as such is sound. However, I have a number of issues with both content and formulations, which I elaborate on below. Besides for three main concerns, there is a number of general comments, followed by editorial comments. The three main concerns all center around how broadly applicable the new parameterization really is, due to the time periods during which measurements were done, due to the fact that parameterizations based on aerosol properties were not done (it was merely compared with older parameterizations for other locations, which is not enough to reject these kinds of parameterizations) and about some data which may have been overlooked in an analysis connecting the new parameterization to literature data (SPIN data from Hartmann et al., 2021, a data source of which, however, off-line data was used). Also, overall, tuning down some of the statements made in the text would make the work more scientifically sound.

Overall, this is an interesting study which will add to the community’s understanding of Arctic INP, and it certainly can be revised such that it will merit publication. However, (rather major) revisions are needed.

Three major issues:

1)

One of my main concerns is about the time periods during which you measured. You happen to have measured during times which were described as transition periods from low to high (spring) or high to low (fall) INP concentrations before (e.g., Creamean et al. 2018; Wex et al. 2019). I am aware that Schrod et al. (2020a) did not find such an annual variation, but they only examined temperatures < -20°C. The pronounced changes shown in Creamean et al. (2018) and Wex et al. (2019) occurred at higher temperatures, for which you report the bulk of your data. Also, there is a new publication which you may not know yet (Porter et al., 2021), in which INP concentrations in the North Pole region were measured in summer. The there reported INP concentrations are MUCH higher than yours (by roughly close to 3 orders of magnitude), which means that while you overestimate concentrations in winter, compared to these new data, you will underestimate them in summer. (And while Porter et al. (2021) is similar to a discussion paper in ACPD, I am sure it will be published in peer reviewed version soon, and then your statements about a general applicability of your fit will be outdated already from its start.) Also, Tobo et al. (2019) and Sanchez-Marroquin et al. (2020) suggested that Arctic mineral dust sources contribute to atmospheric INP. It is unknown but possible that average land based INP concentrations may differ from averages of data taken over the ocean. This should be discussed more.

2)

The second of my main concerns is your suggestion that a single line (together with a confidence interval) will represent INP concentrations better than a parameterization based on aerosol properties. With this, I do not mean to say that such a parameterization based on aerosol properties will work out. But judging from what I see from your data in Fig. 4, A3 and A4, doing fits for n>0.5mm, S or fluorescent conc. for your dataset would cause a similar uncertainty (or confidence interval) of one order of magnitude as your temperature dependent parameterization. And as such, aerosol parameters may represent the data just as well as the parameterization you present. (While, admittedly, additional measurements are needed to retrieve them, so I get the nice part of your approach.)

Instead for examining also these other types of parameterizations in detail, you are just showing parameterizations from totally different environments and then turn them down on false claims. Obviously also the line fits from other environments (Fletcher, Cooper, Meyers) similarly do not fit. So you need to change your arguments for not using an aerosol parameter based parameterization.

Therefore, it needs to be discussed why you think that such a temperature dependent parameterization would outperform parameterizations related to aerosol parameters, which, as said, aren't even introduced and much less examined in the current study. Also, while your parameterization gives a general temperature dependent trend, it does not reproduce the variability as such (besides for giving a range for it, which covers, overall, 2 orders of magnitude). All of this needs to be stated much clearer.

3)

My last main concern is about your comparison with literature data. In Fig. 7, you include some data from Hartmann et al. (2021), a ship-based summer campaign. More specifically, you include the off-line data in Fig. 7, but not the SPIN data. Later in the text you argue that the off-line data are lower than yours, and you mention as possible reasons a) local differences and b) that the off-line samples may have degraded during transport and storage, based on an extrapolation of these data towards SPIN data shown in Hartmann et al. (2021). Specifically, your argument b) makes me think that you feel that the SPIN data is more trustworthy than the off-line data. But this opens up the question why SPIN data is not included in Fig. 7.

Then you say that without including the Hartmann et al. (2021) data, "approximately 97 % of the data falling within the confidence interval for the summer." I assume that for your analysis comparing with literature data, you only included the off-line data from Hartmann et al. (2021) shown in Fig. 7. Or were the SPIN data ever included in your comparison to previous Arctic INP field observations? I guess not, and this is disturbing. I assume you overlooked to include these data and will do so in the revised version. Or maybe there is a good reason for excluding these data. In this case, it needs to be explicitly stated in the text that you exclude them and why.

Summarizing these major concerns, claims you make about the wide applicability of your new parameterization and its outperformance of others seem to be exaggerated and need to be tuned down. I refer to this issue below explicitly where needed.

General comments:

First a general comment on having an appendix: The way the text is structured, it is necessary to go back and forth in the file to get all information during reading. Also, the appendix is quite long. I suggest to prepare supplemental information as a separate file. If I refer to the appendix below, this also always alternatively stands for supplemental information.

Line 8-9: It would be informative to already know from the abstract, during which months you measured.

Line 53: The method to fit INP concentration distributions at one temperature with a frequency distribution was first suggested by Welti et al. (2018), who was, to my knowledge, the first in the INP community, who referred to the paper by Ott et al. (1990), which you cite here. You cite Welti et al. (2018) later, so you should know the content of this work. I was waiting for Welti et al. (2018) to be cited already above, where you describe different types of fitting. Refer to Welti et al. (2020) and the fact that he fitted frequency distributions to INP data either above or related to this sentence here.

Line 76-77: "A flow splitter (custom-built), a 0.5 m long 90°-bend and a three-way ball valve (…) connected a blower (…) and a high flow-rate impinger (…), both operating at 300 L min−1, downstream to the inlet." Please revise this sentence, as it is difficult to understand. Imagining the equipment, I guess I know what you mean, but was the bend in one of the lines after the flow splitter, or before? What was the diameter of the tubing? Maybe an additional sketch would be good as well.

Line 89-90: It remains totally unclear, how INP measurements were done. Liquid was sampled by the impinger, and then? I assume measurements were done right after 1 h sample collection? Or were the samples stored frozen? The measurements you did with DRINCZ, and basics of how DRINCZ works need to be explained at least shortly. This location here is the one where this information fits best.

I was also wondering, in Fig. 1 (b), where DRINCZ really is – I assume it is the thermostat. If so, move the word "DRINCZ" to the left, and also move "Coriolis impinger" to where this really is.

Line 90: The parameter N_INP is not officially introduced, therefore, behind "INP concentration" add "(N_INP)".

Equ. 1: You explain how to derive INP concentrations from impinger samples/DRINCZ measurements. But how about data from HINC?

Line 99: You refer to a background correction. Collecting samples in an impinger (with added water to keep its performance) will enrich impurities in the water in the samples.

For a background correction, you likely collected particle free air into pure water in the impinger for the typical sampling time of 1 hour? Or did you determine this otherwise? In any case, it is needed to explain how you determined the background. Also add at least one plot (best in the appendix) with raw measured INP data (frozen fractions) from samples and from background, as suggested as good practice by Polen et al. (2018).

Line 104: Did HINC use a pre-impactor to reduce false signals from large aerosol particles, as it is typically done for these in-situ instruments? Please state this in the text.

Line 123: This whole chapter 2.2 is difficult to follow and has a quite technical content. It may be better to add a more describing chapter here, in which you roughly explain what you did, and move this rather technical chapter to the appendix.

But no matter if you move most of the following content to the appendix or not, the text here needs to start with one or a few explanatory sentence(s) about what will be introduced in the following.

Line 168: "indistinguishable" may be the wrong word, here. Also, at the end of the sentence add "for the months during which we did our measurements." This is important (as elaborated on repeatedly in this review), as there is a seasonal variation in the Arctic (also observed at Ny-Alesund, see Wex et al., 2019), and that is exactly one of the reasons why your parameterization may not be as broadly applicable as you suggest in your text, performing worse when comparing to literature data taken in winter and summer.

Figure 4: Do I get it right, that D15 was shown as solid line only in the "applicable temperature range" at -20°C and -30°C, and else is given at dotted line? Then add the dotted line with a description to the legend. (The same holds for Fig. A3.)

Also, maybe use brighter blue and green colors for the text in the plots (concerning rho and p). (This also applies for other respective plots.)

Line 190: "For a few decades" – if this is true, then why do you cite two papers from 2021 on this issue? And what do you mean by "temperature dependence"? It seems to imply that knowing the slope of the curve is correct. But I guess that is not what is meant? Revise the test to be more specific on that.

Line 203: "To our knowledge, it is the first attempt to predict INP concentrations in the Arctic utilizing in-situ measurements." Collecting a sample for 1 hour and then examining the resulting bulk liquid (which is, I think, you did with DRINCZ) is not an in-situ measurement. Also, Schrod et al. (2020a), which you cite a number of times, gave an INP concentration parameterization, albeit based on relative humidity. Nevertheless, your statement is incorrect. Revise this sentence or remove it completely.

Line 206: Here you compare to the Fletcher, Cooper and Meyers parameterizations and state that your parameterization is lower than these. It may be good to add that these older parameterizations were done for different environments (i.e., not the transition months in the Arctic).

Figure 7: This relates to my main concern #3. Either include SPIN data from Hartmann et al. (2021) in both figure and analysis, or give a good reason for omitting it.

Line 216ff: This section about how well the parameterization fits, regardless of season and in general needs to be revised, also maybe in light of the addition of SPIN data, which may influence overall performance of your parameterization, or at least its performance during summer.

Line 233ff: A possible regional dependence of your results, which you suggest here, could be of interest. From your big data-set, did you try to compare terrestrially collected data with other data (from ships or airborne)?

Line 251: State clearly, following "95% confidence interval", that this confidence interval covers two orders of magnitude.

Line 254-256: What do you mean by well-mixed INP air masses? Also: the reasoning of the sentence here is not logical. INP are so rare, that most "overall" aerosol properties will not be useful to parameterize them. This means that a good aerosol parameter which can predict INP has not yet been found (and we may not find one), but it says nothing about the mixing state of air masses wrt. INP. At most, it may indicate that the majority of aerosol particles and the majority of INP may have different sources. This sentence needs a thorough revision.

Line 259-261: Given the limitations I listed in this review, this sentence needs to be tuned down. It sounds like you are trying to sell something. Particularly, I don't see how you can make any inference about a vertical INP profile!

Line 256-257: "new INP parameterization can be used as a proxy to estimate the pre-industrial or pristine INP level" This was not discussed in the text at all, and is highly questionable. Remove this text. Besides for this not being a topic in your study at all, here some more reasoning: You do not know how "pristine" your data is, given that it was collected on land, and then close to a settlement in the Arctic (Ny-Alesund). As for pre-industrial times, while it has been argued that Arctic haze does not add INP to Arctic air masses (at least in the temperature range which also you are looking at), it is not clear, to my knowledge, if overall INP concentrations have or have not changed. I know of two studies examining Arctic ice cores (Hartmann et al., 2019 and Schrod et al., 2020b – not intended for inclusion in your work, I only need them for my argument here), which however only examined ice cores dating up to ~ 1990, which is before Arctic amplification clearly started to show. These two studies also come to differing conclusions, the former saying that up to this time, no change was observed, while the latter said that in the modern-day period for temperatures < -22°C, there were higher and more variable INP concentrations. So overall, nothing is known about the performance of your parameterization to describe pre-industrial times.

Fig. A1: This figure isn't referred to in the appendix, only in the main text, which I find confusing. At the same time, I found it confusing that the appendix starts with referencing to Fig. A2. I suggest to add a paragraph on the fitting as A1, which would bring all that in order.

Editorial comments:

Line 115: Mention in section 2.1.2 explicitily, that the WIBS instrument was only used during fall. Right now, one has to get down to A2 to know about that, which is too late.

Line 130: Delete “the” before “heteroscedastic”.

Line 158-159: “The overview of the measurement site and the experimental setup are given in the Section 2.1 and Fig. 1a.” - This doesn't need to be repeated here and is rather interrupting the flow of the text -> remove this sentence.

Line 161: In the parenthesis, “further” should not be capitalized.

Line 165: What do you mean by “virtually log-linear pattern”? Maybe delete “virtually”???

Line 165: Delete “the” before “decreasing”.

Caption of Fig. 4: “as a function of the particle larger than 0.5 μm (volume equivalent diameter) number concentration (n>0.5μm)” should better be something like “as a function of n>0.5um (the number concentration of particles larger than 0.5um volume equivalent diameter)".

Line 166-167: “we conducted unpaired t-tests with a significance level of 5 % for observed INP concentrations at each temperature” I know what you did, here, but maybe this could be formulated a bit better.

Line 186 ff: “relations were first fitted between … relations were further linked with temperatures” – this is not the correct description. Typically, a parameter like the surface site density n_s was calculated based on measured (temperature dependent) INP concentrations together with the surface area, and then the obtained (temperature dependent) calculated values were fitted- so these two sentences here are off and need rewording.

Line 189: Add “area” after “surface”.

Line 191: First word should be in plural: “parameterizations”.

Line 191-192: “We present a methodology to optimally fit the temperature dependence of INP concentration from frequency distributions.” Sounds somewhat strange, think about rewording. Part of my bad feeling about this may be that you don’t fit a temperature dependence, but the frequency distributions.

Line 198: Delete “the” before “trimmed”.

Line 206: Add “than these parameterizations” at the end of the sentence.

Line 207: Replace “overestimated” with “"overestimating our values".

Caption of Fig. 5: “inidcated” needs to be corrected. AND add an “are” after “parameterizations”.

Figure 5 versus Figure 7: Check which line is which, and which ones are correct – Fletcher, Cooper, and Meyers lines should be the same in these two plots, but they are different, at a first glance. (It could be that Fletcher is different?)

Figure 7: In the legend, Ny-A..lesund (at Schrod 2020) needs to be corrected.

Figure 7: "The temperature in parenthesis of the Schneider et al. (2021) parameterization represent the average ambient temperature observed during the autumn (-4.1 °C) and spring (-13.1 °C) campaigns." - I don't see any temperature in parenthesis for the Schneider et al. (2021) parameterizations. Please check the figure or change this sentence.

Line 238: To describe SPIN, change the text in the parenthesis with "(an online instrument based on the same measurement principle as HINC)".

Line 249: In the text you typically use the expression of a 95% confidence interval, here you now refer to 2sigma. Might be better to have this consistent throughout the text.

Line 271: You did not measure the surface (that could be done with an epiphaniometer), but calculated it. Therefore, delete "and surface", or mention that you derived it.

Line 271: Sentence starting with "Generally" misses a verb ("was observed" or so).

Line 274: "concentration of fluorescent particle concentration" - delete one "concentration".

Page 15, caption for Tab. A2: As the abbreviation was only used here, give the explanation "confidence interval" for "CI".

Figure A5: Concerning this figure, it is only mentioned in the text that it exists. Add a paragraph or so (in the appendix), at least explaining what it shows.

Literature:

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