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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term INP measurements from four stations across the globe" *by* Jann Schrod et al.

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General Comments

This paper is excellent as a large compilation of INP data that has been processed in a consistent manner. The effort is to be commended for that reason alone. It is also a very well written manuscript, and with most of the details one would wish for, and the abstract highlights several key points: well mixed populations that do not vary greatly overall between northern and southern continental and marine sites, short-term variability dominating at all sites, certain site specific aerosol drivers of INPs, but no universal driving aerosol property driver, and no indication of anthropogenic influences. Nevertheless, as I read the paper as it is currently organized, I struggled in

knowing how to relate the method and results from the standard FRIDGE method to drop freezing assays (or the immersion mode method sometimes applied using the FRIDGE device), which are possibly the most widely used present method. It seems to me that two things are required to assist readers in understanding the nature of the results, and potentially how to consider them in relation to immersion freezing data. First, the title should explicitly describe the basis for INP measurements. In other words, "Long-term deposition/condensation freezing INP measurements. . ." or something to that effect. When one sees the INP versus ice supersaturation data in this manuscript, there is no discontinuity that occurs at water saturation (as the authors readily note), and so it seems apparent that immersion mode freezing is indeed not represented at all. The authors provide a discussion of the dominant mechanisms at play in the data and the likely underestimate in comparison to immersion freezing mode operation of the FRIDGE only very late in the paper. This is critically important in understanding if the findings can be ascribed only to deposition and condensation-freezing mode INPs, or if the same is expected for immersion freezing populations. I suggest in the specific comments that the methods used may indeed limit assessment of strong local/regional impacts, at least for biomass burning. Of course, it will not be possible to make a conclusion about what was not measured, but it should be highlighted as a question for future inspection. This should all be made crystal clear. Hence, the second recommended change is to bring a discussion forward of what types of INPs the data describe, and what types the generalized results may not describe. It will not detract from the great effort the authors have made to collect large quantities of ice nucleation data from multiple sites and discern answers to some of the key and enduring questions related to INP sources. However, I believe that it will better frame future needs.

Specific Comments

1) Introduction

Page 2, Lines 8-9: Is there a reason to separate primary biological aerosol and marine biological aerosols? They are both primary biological aerosols, no? If referring to

secondary marine aerosols, you might require evidence that those play any role as INPs.

Page 2, Lines 30-32: I find this statement quickly becoming untrue, with many laboratories now involved in long-term measurements of immersion freezing (e.g., Schneider et al., 2020), some with agency support, and multiple online instruments are in development (or are already there) for automated or semi-automated operation.

Page 3, line 30 to end of paragraph: With regard to anthropogenic influences, I do think that there is some literature on this topic. Some is recent, e.g., Levin et al. (2019) found no apparent influence of urban pollution on INPs in studies in CA, USA. Chen et al. (2019) and Bi et al. (2019) discuss urban pollution impacts in Beijing.

2) Methods

Page 4, lines 28-29: Have larger particle losses been quantified? This is important, as it is a weakness compared to an open-faced filter for example, and it is not clear as an advantage over the in situ instruments mentioned in the last sentence of the paragraph. For example, Schrod et al. (2016) report collection efficiencies only to 3 microns, which is not measurably much different that impactors used on some in situ devices. And larger particles might be imagined as the most efficient deposition nuclei. While collection of and a role for larger INPs is evident ultimately in Fig. 9 for the AZ site, one wonders if the drop off of INPs at sizes above 2 microns reflects the true contributions in these size classes or is influenced at all by collection efficiencies.

Page 5, line 13 paragraph: This description of the aerosol samples had me already wondering about sampler inlets and placement. You might state that this will be covered for each specific site. I do question the statement that 100 L samples provide for "well-resolved ice crystal numbers for a broad spectrum of temperatures..." INP concentrations can span several orders of magnitude from -5 to -35°C . This paper covers a 10C range for data presentation. Finally, is the statement on storage effects necessarily assured for biological INPs that might be exposed to dessicated and higher

temperature conditions? This was qualified in Schrod et al. (2016).

Section 2.2: It is worth carefully explaining the valid activation modes for this work (should be deposition and condensation "freezing" mode on line 22), perhaps by reiterating a few points from Schrod et al. (2016). This first paragraph appears to be the clear place to expound on what is known about the potential underestimations compared to immersion freezing mode INP data as well. Instead, there is only a sentence, "In this context. . .", which is awkward and defensive considering that the FRIDGE instrument pre-dated many of the droplet freezing assays. The instrument is clearly a tool within the wider array of ice nucleation instrument types, and to my knowledge one of the few well-characterized and documented ones that allows for exploring the full temperature and ice relative humidity space (in the mixed-phase cloud regime) for single samples, in the same manner that droplet freezing assays allow for full temperature spectra. All of the advantages of the technique compared to more labor intensive diffusion chambers and drop freezing assays are well acknowledged. What is missing for this assessment of long-term records at multiple sites is a clear indication of the relation of the modes assessed to immersion freezing. What is known and what remains for future exploration, if the method could be meshed with additional immersion freezing measures?

Page 5, line 24: The word meaningful seems unnecessary.

Page 7, line 15: An additional question here is if there are any considered additional particle losses in the inlet entry to the sampling system. That is, is sampling from the main inlet isokinetic (or sub- or super-isokinetic) and are any additional large particle losses characterized for that last step in collection? Similarly on page 8, line 23, it says that the sampler and the OPS instruments were connected to a 2 m stainless steel line at OVSM. Were particle transmission efficiencies characterized/expected to be the same at this site? Given the outsized role of larger particles as INPs at some surface sites (e.g., Mason et al., 2016), it seems important to know if the relative collection efficiencies were the same, and what the upper limit might be. I also note no mention

of sampling inlet protocol for either TO or SB sites.

Page 7, line 29: A minor note here that it would be interesting to know the vegetative differences in these sites. Images of the sampling sites could also be interesting, for supplemental information.

Page 9, line 20: What is meant by “direct influence of sea salt aerosol”? Is the Zeppelin site not within the boundary layer? This is important to know with regard to what influences are being measured there. Sea spray particles would seem as one key source

3) Results and Discussion

Page 10, lines 10-11: Considering the discussion above about INP mechanisms, this statement about deposition being considered relatively unimportant for mixed phase clouds is confusing. Is this not what is measured by the FRIDGE instrument? If the traces of INP versus ice supersaturation are continuous, how to know the difference between deposition and condensation freezing? Is not the highest RH value of processing used here so that the highest INP concentrations assessable are accessed? This is the only way to understand the following statement that “incomplete” condensation freezing is assessed. Again, this may be material to consolidate in the Methods section, where it can be pointed out that an emphasis will be placed on the highest RH values for inter-comparison of site data.

Page 10, lines 31-32: It is great that the authors qualify the results regarding timing of the sampling, storage impacts, etc. However, I am not sure what this statement means about long-term trends being better captured by different sampling strategies. Can you expound? Does it mean spreading the sampling periods out across daily periods? Larger volume samples collected over longer time periods? Additional use of immersion freezing methods, as in that study, to investigate if that mode of ice nucleation also shows a lack of long-term trends at sites. Also, please note that the full publication on the noted results is now in press and under review in ACP (Schneider et al., 2020).

That study does show trends linked to a regional source. One can imagine that regions close to mineral dust sources also show impacts of a strong regional source, where much higher INP concentrations are noted (e.g., Price et al., 2018). Likewise, higher latitude and polar regions, especially from ship campaigns in the Southern Hemisphere (McCluskey et al., 2018; Welti et al., 2020), appear to represent extraordinarily pristine INP environments. It is simply the case that for the sites selected for this paper and the methods applied, strong cycles are not noted and short-term variability dominated. The extent to which this can be generalized for tropical and mid-latitude regions remains to be seen.

Page 11, lines 15-16 and elsewhere: I have a suggestion to consider for demonstrating the spectral differences between sites, and where they are distinguished for given sites. Currently, a temperature spectral plot is not included in the paper, with too much emphasis on ice supersaturation in my opinion. Figure 5 could be made differently or augmented with an additional panel. While sometimes a linear scale is preferable, in this case if you alternately (or additionally) put these data on the same log scale, one could see the temperature differences more clearly. For example, if the y-axis scaled from 0.01 to 10 on a log scale, the temperature spectra becomes evident for conditions near water saturation, which are arguably the most important for clouds.

Page 12, lines 18-19: This comment harps back a little bit to the statement in Methods regarding the large dynamic range of measurements. While 100 L samples are more useful than the smaller sample volumes used in online instruments, the lack of resolution in the -20°C and warmer regime means that there is little or no access to the temperature range where one might expect most sites to be distinguished, considering for example the results shown in Petters and Wright (2015). This is also an important point to remember in the discussion here regarding whether any of the sites are distinguished by apparent biological particle influences. The measurements are just touching the regime of interest.

Page 13, discussion of Fig. 7: Figure 7 is a remarkable figure, and I find it astonishing

that local sources do not come into play for either TO or AZ. I wonder if the authors might comment on whether INP removal is also a factor to consider, not only dilution/mixing out from strong sources, as is inferred in the comment about “background” air masses?

Page 13, section 3.2.1: First, can you please clarify the timing of the “dry season” at AZ? It becomes obvious in Fig. 8, but it would be nice to see it stated in the discussion. And then one has to go back to figures to note the lack of an apparent influence of smoke. The reduction is AF is not really unexpected, right, in consideration of previous results regarding biomass burning INPs? Considering laboratory studies of surrogate and real combustion particles (Petters et al., 2009; Levin et al., 2016; Kanji et al., 2020) and field studies (Prenni et al, 2012; McCluskey et al., 2014; Schill et al., 2020)? Hence, the discussion could be clarified here, including the most recent references. One might even support that for realistic combustion particles, and not only black carbon isolated (Kanji et al., 2020) or contained in real biomass burning particles (Schill et al., 2020), water supersaturations and immersion freezing are required to see the influence of biomass burning on INP concentrations (e.g., Petters et al., 2009; Schill et al., 2020). That is, there are clear impacts of biomass burning on regional INP concentrations already demonstrated in the literature for other regions. I think this discussion needs more specifics than referencing a review paper and a single laboratory study on black carbon surrogates. Activity within the deposition and condensation freezing regime up to water saturation may be quite limited, so this may represent a case where the methods applied in this paper cannot resolve real influences on INPs, or it may indicate that fires are not sources at AZ. I think it is unresolved still.

Page 14, lines 17-18: Following in the same line of comment, in fact the INP concentration results herein seem to be a factor of several lower compared to Prenni et al. (2009). It would be good to quantify what is stated presently as “on the low end”.

Page 15, lines 25-26: It is unclear if the conclusion here is that marine contributions to the INPs at MQ are represented in the lower range of values observed?

Page 15, lines 32-33: Is this correlation with PM10 at TO shown anywhere? Can you at least state the r^2 and p values?

Page 16, SB section: As I read this section, I wondered about the issues brought forward at the end of the section with regard to signal to noise ratio, and how this influenced the lack of a seasonal cycle. For example, Hartmann et al. (2020) should also be referenced here. They also report winter values consistent with Tobo et al. (2019) and Wex et al. (2019). Hence, one wonders why no seasonal cycle is present in the data here. Is it just noise, or is the baseline potentially somehow even higher than you have estimated from blank data?

4) Conclusions

Page 17, lines 11-13: I find alluding to the Welti et al. paper results to not be a great comparison. In fact differences in the most remote locations were striking compared to mid-latitude and tropical locations in that paper and in other recent ship campaigns (McCluskey et al., 2018).

Page 17, line 20: I think you should add “at all sites” when referring to the inability of single parameters to describe results. This is important, as influences were noted at some sites.

Page 18, lines 3-6: This is the discussion point that needs to be introduced earlier in the paper, as I mentioned previously. One even wonders if the processing conditions emphasize certain INP types that are more well mixed in the atmosphere and contain few hygroscopic materials that would limit ice nucleation until strong condensation occurs at most of the temperatures investigated.

5) Outlook

Page 18, lines 18-19: One wonders about varying sampling times over daily schedules to represent diurnal cycles. However, here, I wonder if it is necessarily true that longer sampling times would reduce short term variability? Would several hour samples reflect

less differences than the short sample times used in this study? How do you know?

Page 18, lines 21-22: Again I find myself disagreeing with this conclusion that automated and higher frequency sampling methods are too much of a technological challenge. It simply needs impetus and being made a priority, and I would judge that the time has already arrived.

Page 18, lines 30-32: A reason that immersion freezing is considered so important is because clouds, and how they form, in many cases determine this result. Could immersion freezing measurements become an integral part of sampling and processing protocol for a device like the FRIDGE? Then all mechanisms except contact freezing would be assessed.

Page 19, lines 9-10: I find the calling out of a single device to be inappropriate here, from a conference paper no less. Fortunately for this reference, the prime publication on the PINE came out the same day as this review (Möhler et al., 2020). However, automated CFDC instruments are already being built for surface sites (Bi et al., 2019) and under development for aircraft use. I do not understand the statement about a "vital intermediate step".

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