

Answers to RC1

We thank the reviewer for the careful reading and helpful commenting of our manuscript, giving very valuable feedback and suggestions. By addressing the comments we hope that our manuscript improved considerably.

The reviewer's comments are in blue with our answers in black. Extracts from the original manuscript are presented in *italic* and changes in *italic green*.

This study by Lacher et al. describes ice nucleating particle measurements made during 2 campaigns at the Storm Peak Laboratory (Rocky Mountains, CO, USA) during two different years. INP measurements were made with the online expansion chamber instrument PINE, with additional aerosol measurements of supermicron particles from an APS. Measurements ranged from fall to spring (2021-2022) and winter to spring (2025), with the lowest INP concentrations observed in winter, and the highest in spring in both years. Supplemental backtrajectory analyses of air mass source “footprints” was combined with an aridity dataset, and suggested elevated INP concentrations in spring were correlated with local/regional dust emissions. The sizes of INPs were investigated in a few different ways, including correlations with supermicron particle concentrations and direct size measurements of INP residuals. The indirect correlations suggested the importance of supermicron particles to INP concentrations, particularly in fall and spring. Unfortunately, the direct measurements were only performed for a short period in one winter campaign, but suggested both sub- and super- micron particles were important at Storm Peak Lab during the winter.

I found the article easy to read, the structure logical, and the figures well-labeled and clear. I have a few major comments about some additional analyses or text that would strengthen the sections on the impact of local/regional dust and the ice residual size measurements with the PCVI, which are included below.

Major Comments:

1. Cases with the highest INP concentrations during April 2022 and 2025 were additionally analyzed with backtrajectories to determine source footprints and compared to the aridity dataset. The determination of how important local/regional dust sources are to the SPL INP concentration would be stronger if some periods with lower INP concentrations were analyzed, both during the same month, and also in a different season, when INP concentrations were lower (ie winter). If the source region is different and/or the aridity is lower when the INP concentrations are low, it would help validate the role of local dust during the high INP concentration periods.

As the reviewer suggested, we performed additional backtrajectory analyses of two cases of generally lower INP concentrations in April, namely for April 23 00:00 UTC to April 24 00:00 UTC, when INP concentrations at -28 °C to -30 °C decreased by one order of magnitude, with values mostly below 10 INP stdL⁻¹, and for April 5 00:00 UTC to April 6

00:00 UTC, when INP concentrations in the same temperature range decreased from 80 – 100 to 10 – 50 stdL^{-1} . We show the footprints together with the VegDRI maps, and the back trajectories only from the STILT model for the discussed cases of high INP concentration in the manuscript together with the cases of low INP concentration in new Figures A3 and A4.

Interestingly, for these two cases of lower INP concentrations, the footprints from the STILT model are more widespread (Fig. A3; Fig. A4, panels e, f), as compared to the cases of high INP concentrations shown in the manuscript (Fig. 7, panels a, b, c, d; Fig. A4 panels a, b, c, d). There is no clearly defined upwind source region for the air parcels and the contour fields spread broadly and the isolines become more diffuse.

We interpret this finding that there is no distinct source of dust, especially when compared to our high INP case studies. When compared to the high INP case studies, the back trajectory footprints are much more concentrated over regions of known dust sources.

We add to the discussion in section 3.2 now:

“For comparison, we also performed back trajectory analyses in combination with surface maps of aridity for two cases of low INP concentration (Fig. A3). Interestingly, the footprints are more widespread as compared to the discussed cases of high INP concentration, and make an interpretation with regard to distinct local and regional dust sources difficult. The back trajectory calculations only (Fig. A4) show more clearly that the footprints for the low INP cases, in comparison to the high INP cases, are broader, indicating that during this time the station measured particles from more diverse source of aerosols and not only local dust sources.”

And included the two corresponding figures in the Appendix:

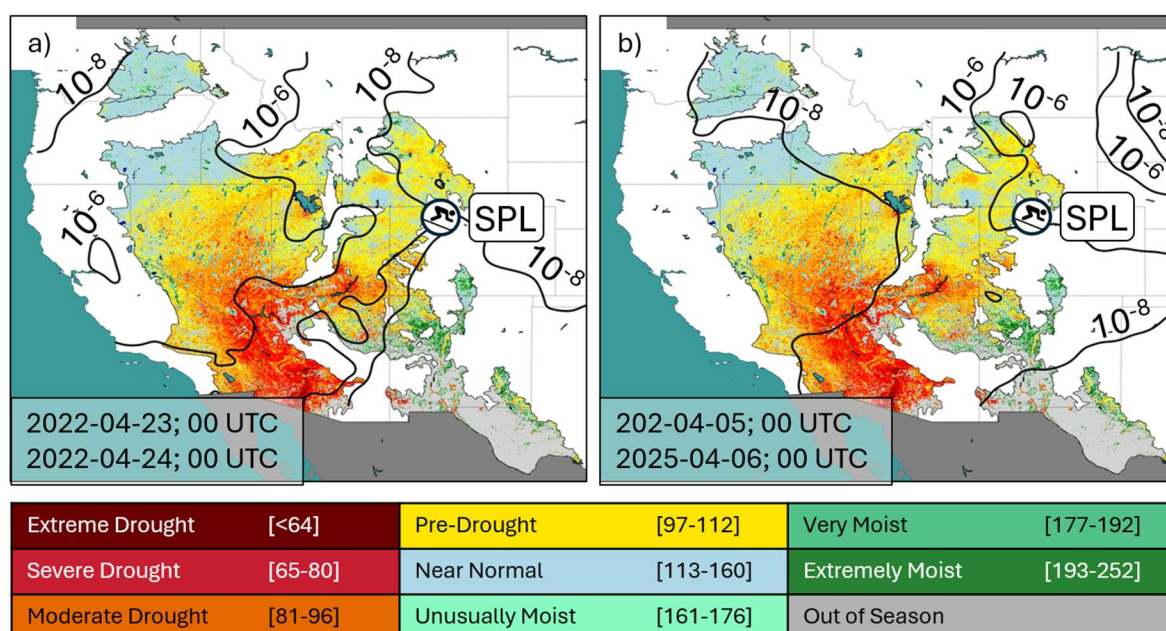
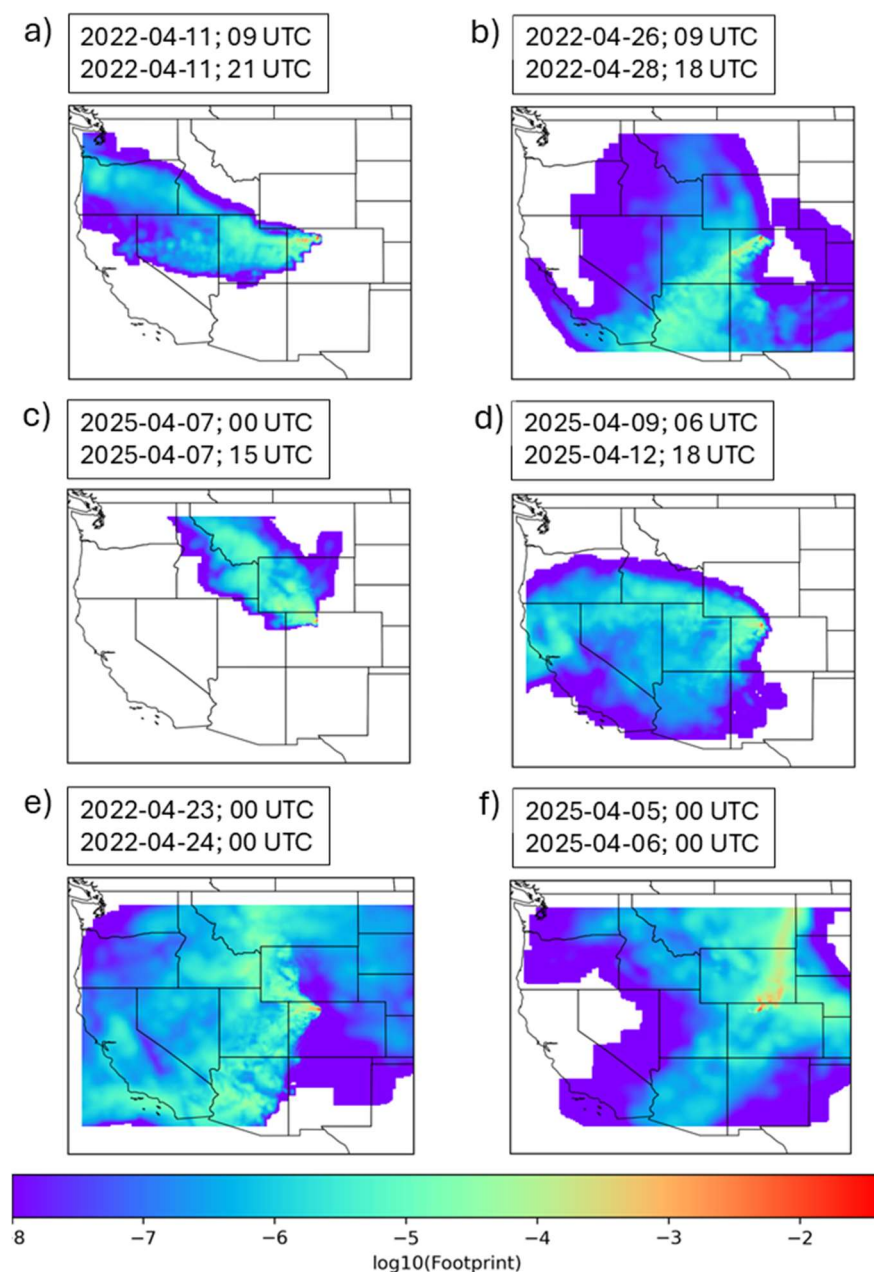


Figure A3: Footprints computed with the STILT model combined with surface maps of aridity using VegDRI of North American Desert ecoregions. Contours of the footprint are in units of $\text{PPM}/\mu\text{mole m}^{-2} \text{s}^{-1}$ (surface influence footprint; Fasoli et al., 2018). Events of low INP concentrations were analyzed on the 23rd April 2022 (panel a, INP concentration at -30°C mostly below 10 INP stdL^{-1}), on the 5th April 2022 (panel b, INP concentration at -30°C below 50 INP stdL^{-1}).



We hope that the new analyses strengthen our statement, but believe that more measurements on a long-term basis in combination with intensive model calculations of source regions would be required to ultimately answer the question of the importance of local and regional dust for the INP population in the Rocky Mountains. To make this point clear, we add to the summary:

„Thus, more and longer-term observations of INP properties should be conducted in combination with source emission sensitivity studies of dust sources.“

2. Additional information and/or calculations are needed regarding the PCVI measurements of ice residuals. These include:

2.1 Does the D50 depend on particle shape or density? Cloud droplets are generally spherical, while ice crystals are highly non-spherical, and cloud droplets, ice crystals, and aerosols all have different densities.

The reviewer is right, aerosol separation within all virtual impactors depends on aerodynamic diameter (line 166 in the new manuscript), which is a function of particle density and shape, which can have an influence in the separation in the PCVI and thus on the D50. In PINE, ice crystals are formed by immersion freezing of (spherical) cloud droplets, and we assume that the fast freezing process during expansion is likely to produce roughly spheroid particles. If highly non-spherical crystals were present their smaller aerodynamic size may reduce their PCVI transmission efficiency. However, this is likely to be a small contributor to the overall transmission efficiency.

Moreover, we considered it to be of importance to test our PCVI setup in the field under realistic sampling conditions, meaning that ice crystals of different shape and density can be present.

To make this point more clear, we updated the respective text in section 2.4:

“The setup was configured and tested during laboratory studies (see appendix for these experiments) and configured, tested and applied during the SPL campaign in winter 2022. The aim of the laboratory studies was to determine the D50 using ammonium sulphate particles and to define preliminary configuration settings. In the field, the setup underwent more thorough testing for the separation of aerosol particles, cloud droplets and ice crystals, which is more complex as the differences in particle shape, density and phase can have an impact (e.g., Kulkarni et al., 2011).“

2.2 Different concentration factors are listed for the PCVI throughout the manuscript- how much did it vary over time and with particle size, and how was it measured?

The PCVI concentration factor is simply the ratio of the measured total in flow (F_{inlet}) to total out flow (F_{out}). The range of factors is due to slight changes in the add flow throughout the study.

To make this more clear, we added a sentence to section 2.4, defining the concentration factor and to clarify the effect of changing flows to concentrating factor:

„The PCVI concentrates the ice particles (and thereby the residuals) by a factor of total inlet to total outlet flow.“

And modified the sentence:

„This initial flow reduction increased the PCVI counterflow slightly and increased the concentrating factor to as high as 38.6 but otherwise had little effect on PCVI operation.“

We also noted an error in the calculation of the concentration factor upper range and corrected the value in Tab. 1 to a range of 27 – 39.

2.3 Why was the calibration presented in the Appendix performed for an F_{inlet} of 4.2 lpm, when the campaign has an F_{inlet} of 3.3 lpm? Is the calibration presented here relevant to the conditions during ambient sampling? Does F_{inlet} impact the PCVI D50 or the transmission efficiency of particles of any size?

The PCVI characterization in Fig. A5 (formerly Fig. A3) was performed using the TSI APS instrument as mentioned, rather than the SPX instrument, since the APS measures aerodynamic diameter directly and has an extended size range that captures the D50 and beyond. The APS gives a PCVI out flow of 1 LPM. As the PCVI D50 depends mostly on in flow and counterflow (Kulkarni et al., 2011), we adjusted the in flow to higher values. The settings used in these laboratory experiments represent lower limits to those under SPL measurement conditions. Please also note that the D50 experiments mostly aimed at investigating the approximate size range of the transmitted particles and the more detailed experiments were performed in the field (presented in section 2.4 and in appendix).

For clarification, we updated the text:

„Then the original size distribution and the size distribution downstream of the PCVI was measured with an APS (3320, TSI Incorporated, Shoreview, Minnesota, USA), having a sample flow of 1 LPM, which equals F_{out} .“

„The D50 estimates in this flow configuration represent lower limits to those under SPL measurement conditions.“

Also, the caption of Fig. A5 (formerly Fig. A3) was updated to include the corresponding concentration factors:

„Fig. A5: Size distribution of ammonium sulfate particles *before* (black line) and behind the PCVI using different values for F_{add} . Transmission at sizes larger than $\sim 5 \mu\text{m}$ is artificially low due to enhanced impaction losses in the aerosol generation system when the PCVI was installed. Curves are corrected for dilution and the PCVI concentrating factor of 3.7 – 4.7.“

We also indicate in Tab. 1 now that the D50 is $> 4 \mu\text{m}$:

Table 1: PCVI configurations used when coupled with PINE.

F_{inlet} (= expansion flow)	3.3 LPM
F_{pump}	6.2 LPM
F_{add}	2.8 - 3 LPM
F_{out}	0.085 - 0.12 LPM
F_{eff} ($F_{add} - F_{out}$)	2.68 – 2.92 LPM
Concentration factor	27 - 39
D50	$> \sim 4 \mu\text{m}$

2.4 The conclusion from the droplet evaporation calculation would be strengthened in two ways. First, if these calculations are repeated for a warmer temperature ($> -28 \text{ }^{\circ}\text{C}$) where Fig. A4 suggests transmission of cloud droplets through the PCVI, do the calculations also suggest the cloud droplets are larger than the PCVI D50? And second, since the cloud droplets are being measured during expansions with the PINE OPC, are the sizes calculated in Fig. A5 consistent with the measurements?

We performed the cloud droplet growth calculation for $-25 \text{ }^{\circ}\text{C}$ (warmer than $-28 \text{ }^{\circ}\text{C}$, as the reviewer suggests), a temperature at which some droplets were transmitted through the PCVI based on the experiments presented in Fig. A7 (formerly Fig. A5). At this temperature, cloud droplets with diameters of $6.5 \mu\text{m}$ could form, which could be transmitted through the PCVI (Fig. 2).

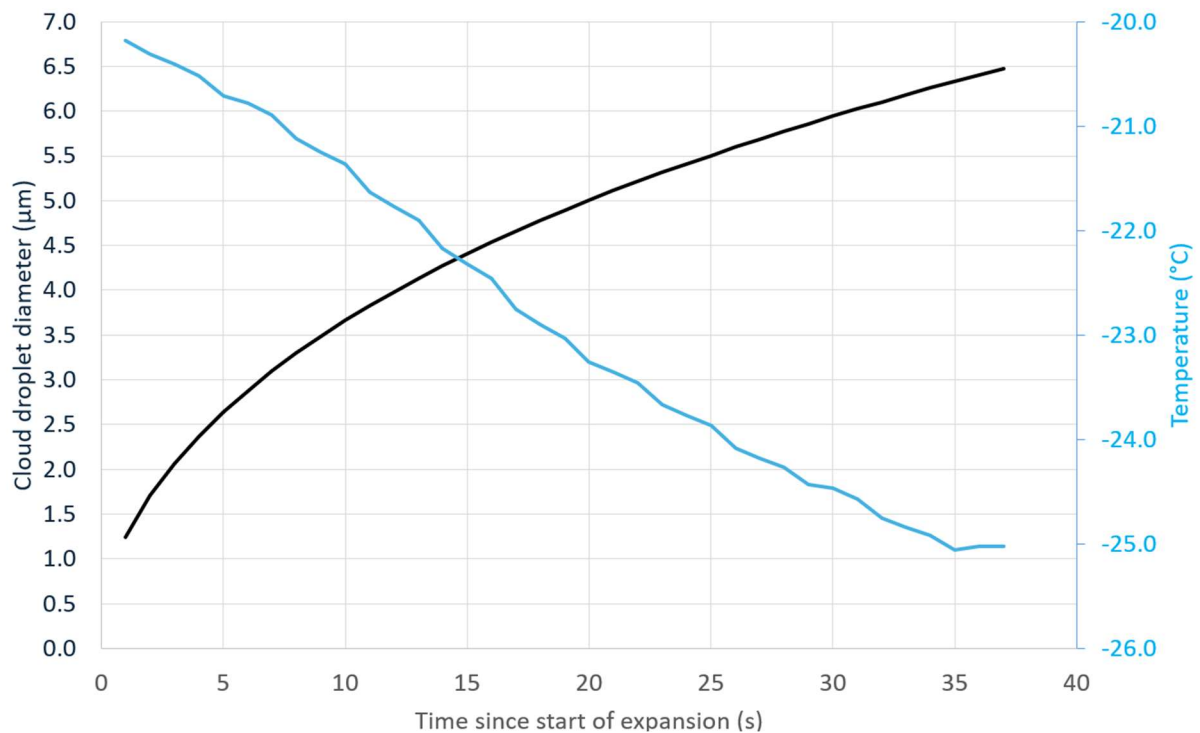


Figure 2: Calculated cloud droplet diameter during a PINE expansion with a start temperature of -20 °C, based on diffusional growth calculations.

We add the results of this calculations now in the corresponding section in the appendix:

“The formed cloud droplets during this experiment can reach sizes of 5.5 μm (Fig. A7). For comparison, the calculation for a temperature of -25 °C (referring to a temperature decrease during expansion from -20 °C to -25 °C) shows that cloud droplets can grow to 6.5 μm.”

And correct the following text:

“Cloud droplet sizes of 5.5 μm, which form at -29 °C, thus are completely evaporated before entering the PCVI body, and can thus be separated from ice crystals. Based on these calculations, cloud droplets of a size of 6.5 (formed at temperatures of -25 °C) μm are not completely evaporated, however, when entering the PCVI their size is still below the D50, and it can be assumed that the majority is still separated from ice crystals, but can lead to the observed increase in residuals during the experiments presented in Fig. A6.”

Moreover, cloud droplets are not sized accurately with the fidas-pine OPC, as it is tuned in a way to see only larger particles, meaning that a detailed comparison cannot be made unfortunately. In the future, a tandem setup using two in-line OPCs could be used, one for detecting single ice crystals with a high sensitivity, such as the fidas-pine OPC, and one for accurate sizing of cloud droplets, e.g., such as a welas-2500 sensor (Palas GmbH) which was used in PINE-1A in first field campaigns and can adjust to the rapid pressure decrease during expansion.

3. Some of the issues are potentially to do with pdf conversion, but many of the figures have blurry lines, text, or axes. The axes are generally very thin and faint, and the text is too small in many of the figures.

Yes, the conversion to pdf caused a lower quality of the figures. Moreover, we adjusted the text font and axes thickness in Figs. 4, 5, 8, 9, 11 and A5.

Minor Comments:

1. Line 24: Measurements of INP residuals downstream of related instruments (ie CFDC) have been performed previously, so be clear about what exactly is novel here (for example, Cornwell et al. (2019)). Is it just the coupling to PINE, specifically?

The reviewer is correct that the novelty here is that the coupling to PINE, an instrument that is capable to measure INP concentration autonomously and continuously, such that it can be operated in a monitoring fashion. By being able to couple PINE to a PCVI bears the great potential to investigate INP properties in a long-term operation mode.

To highlight this better we add to the abstract:

„Moreover, INP sizes were investigated by ranked correlation coefficient analysis of parallel measurements of super-micrometer particles, the application of a novel setup of a pumped-counterflow virtual impactor downstream of PINE to analyze the sizes of ice residuals, and alternated INP measurements at a 1 μm impactor. In addition, for the first time, PINE was coupled to a pumped counterflow virtual impactor to analyze the sizes of ice residuals.“

And to the summary and outlook section:

„This is of particular scientific interest, as PINE is emerging as a key instrument for the continuous global measurement of INPs, and being coupled to a PCVI would allow for determining INP properties such as size and chemistry on a longer-termed period.“

2. Lines 53-66, discussion of other methods/direct measurements of ice residuals: The addition of an impactor to CFDC instruments has enabled a similar set of chemical/size measurements on collected INP residuals in both mesocosm and field campaign settings. See, for example DeMott et al. (2023) and Twohy et al. (2021). See also Burrows et al. (2022) for an overview of other techniques that achieve single-INP chemical measurements (ie cold stage activation + microscopy). Some have been applied to field measurements. Or, if you are only discussing fully online techniques, clarify that.

Indeed we aimed at discussing only online techniques, as the PINE-PCVI setup has the advantage (as other online techniques) to be operated in a more autonomous way. We update the text now:

*„A precise method to identify INPs **using online instruments** is the direct chemical and physical analysis of the INPs by using ice-selective inlets or cloud chambers coupled to pumped-counterflow virtual impactors (PCVI; Boulter et al., 2006; Hiranuma et al., 2016).“*

3. Lines 58-59: Coupled online INP + PCVI + single particle mass spectrometry has also been performed for ambient air at a coastal marine site, not just for cirrus clouds (Cornwell et al., 2019).

Thank you for referring to this study. We include now :

*„The currently existing coupled setups for ambient measurements **often** focus on cirrus formation, where only discrimination between ice crystals and aerosol particles is needed. A recent study by Cornwell et al. (2019) also applied a coupled system of a continuous-flow diffusion chamber with a PCVI at a coastal marine site at mixed-phase cloud conditions.“*

4. Line 70: Given the measurements presented here are from the US, I'm unsure why the focus here is on seasonal cycles in Europe. Similar annual cycles of INPs have also been seen in the high Arctic (Creamean et al., 2022; Tobo et al., 2024) and Greenland (Sze et al., 2023), among others. More relevant to this study are the 2 years of INP observations from SAIL, in Crested Butte, CO (Feldman et al., 2023; Zhou et al., 2025).

Thank you for pointing to these studies. We include them now:

*„Indeed, seasonal trends were identified at different measurement locations **in Europe**. In a boreal forest, Schneider et al. (2021) attributed the seasonal cycle in INP concentration with a maximum in summer to biogenic particles. Moreover, a seasonal cycle in the Arctic region was identified (Creamean et al., 2022; Tobo et al., 2024; Sze et al., 2023) related to different phenomena such as sea ice opening and melt and the transport from lower latitudes during spring.*

At different elevated sites in Europe and the US, seasonal cycles in the INP concentration were identified. A maximum in INP concentration in the warm season was also found at Alpine high-altitude sites (Brunner et al., 2022), caused by the impact of boundary layer air containing biogenic particles such as pollen and soil dust with peak INP concentration measured during Saharan dust events (i.e., mineral dust events). Also, at a mountain site in central France, Bras et al. (2024) identified a seasonal cycle with minimum INP concentrations in winter and increasing values in

spring. In a recent study in the Rocky Mountaings, Zhou et al. (2025) found lower INP concentrations in winter and maximum concentrations in summer. Such measurements at elevated sites are especially relevant as the INP population since ice crystal formation in clouds often occur at the same altitudes. Such measurements at elevated sites are especially relevant as the INP population can be directly important for ice crystal formation in clouds.“

5. Lines 119-120: What happens if the background experiments (sample air passed through a filter) have INP concentrations above zero?

If background measurements have non-zero INP concentrations after approximately 5 – 10 cycles of flush-expansion-refill, the instrument likely has a leak where ambient (humid) air and aerosol particles can enter the system. In such a case, the leak needs to be identified and fixed. During the campaigns in 2021/2022 and 2025, such an issue did not appear.

6. Line 124: The D50 of the PINE inlet is listed as 4 μm , as is the D50 of the PCVI later in the manuscript. This implies large aerosols near the 4 μm cutoff will be directly transferred to the PCVI, along with ice crystals or large cloud droplets. Why not use a larger D50 for the PCVI, to avoid this? What was the minimum size observed for ice crystals in PINE? Is there any overlap with the aerosol distribution?

The chosen D50 is based on the experiments performed first in the laboratory as presented in the appendix, and the experiments in the field. As we clarify now the D50 of the PCVI setup is > 4 μm (see answer to your major comment 2.3). While aerosol particles > 4 μm can pass the setup, the naturally very low ambient concentration of such particles would lead to a small bias of unactivated aerosol particles to be misclassified as ice crystals.

Moreover, based on our experiments, a larger D50 also leads to a decreased transmission efficiency (Fig. A5 (formerly Fig. A3)).

We add now to the description of Fig. 2 and related findings:

„As cloud droplets are not growing to sizes larger than the D50 of the setup (see appendix for more details), the increase above background is due to ice crystals passing the PCVI. Please note that ambient aerosol particles > 4 μm could be transmitted and misclassified as ice crystals, however, their naturally very low concentration makes such a misclassification unlikely.“

7. Line 126: Can you be more specific about what a “rime-free aerosol inlet” means? Is it heated?

The inlet is heated to 30 °C. We add now:

„PINE measurements were conducted at a rime-free aerosol inlet, which is heated to 30 °C.“

8. Line 127: What is the aerosol inlet D50 for realistic wind speeds sampled during the campaign?

At higher wind speeds of 10 ms⁻¹ and 15 ms⁻¹, the aerosol inlet has a D50 of approximately 7 µm and 5.5 µm (Petersen et al., 2019). The highest wind speeds occur in winter (December, January, February), with typical wind speeds below 12 ms⁻¹ (Fig. 1, similar to values shown by Hallar et al. (2025)). Therefore, transmission of particles that can enter PINE (D50 of 4 µm) is given under most wind conditions.

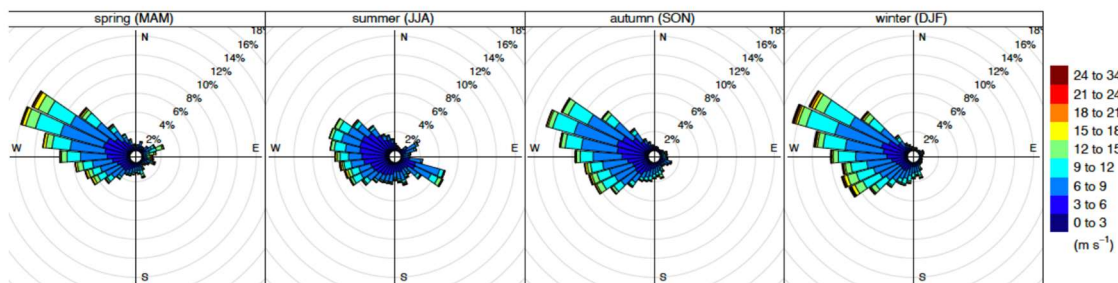


Figure 1: Seasonal wind speed and direction by proportional mean at SPL for the different seasons.

We also add now:

„At higher wind speeds of 10 m s⁻¹ and 15 m s⁻¹, the aerosol inlet has a D50 of approximately 7 µm and 5.5 µm, respectively (Petersen et al., 2019), allowing the transmission of particles that can enter PINE (D50 of 4 µm) as most of the time the wind speeds at SPL are below 10 ms⁻¹ (Hallar et al., 2025).“

9. Line 177: Are the size distributions mentioned (Fig. 2a) measured by the PINE OPC, or separate instruments (ie SMPS/APS)? If in PINE, were these distributions measured during the flush or expansion period- ie are there are cloud droplets and ice crystals at the larger sizes, or only unactivated aerosols?

The size distributions in Fig. 2 were determined with the SPX, not with the PINE OPC. We clarify this now:

„The size distribution was measured with the SPX and the flow settings of the final setup (Table 1) were used but with a slightly reduced Fadd of 2.5 LPM, and PINE was set to a temperature of -29 °C.“

10. Line 185: How was the ice transmission efficiency through the PCVI calculated? I'm unclear how to combine the INP concentration with the "small aerosol rejection factor" to give the transmission efficiency listed (16%).

The ice transmission efficiency is simply the ratio of the PCVI ice residual concentration to the ice crystals determined in PINE. The small aerosol rejection factor is independent of this transmission efficiency.

We update the sentence:

„During the expansions PINE measured an average INP concentration of 377 +/- 83 stdL-1, giving a transmission efficiency of ice crystals as compared to INP concentration through the PCVI in this configuration of 16%.“

11. Line 212: Are the footprints for each ensemble released (one per hour) averaged together to give an average footprint for each event? This is not clear from Sec. 3.2 and Fig. 7.

Yes, the footprints are calculated every hour and then averaged. We clarify this now:

„For our analysis, we selected four events of elevated INP concentrations, lasting between 12 hours and 3 days. For this, 1000 particle ensembles were calculated every hour from the receptor location (SPL) and tracked 72 hours back in time starting 4 hours before to 4 hours after each event. Then an average footprint for one event was calculated“

12. Line 220: Was the VegDRI dataset averaged separately over April 2022 and April 2025, or were both years averaged together? If the latter, why not leave them separate, since each event is being investigated separately?

The VegDRI dataset was averaged separately over April 2022 and April 2025, we clarify this now:

„The VegDRI was retrieved and averaged over April 2022 and April 2025 separately.“

13. Line 220-221: I'm unclear on what the "dry and more dry conditions" refers to. Are these specific definitions from the VegDRI dataset? What are these percent differences used for/where are they shown in the manuscript?

Thank you for this helpful input. We meant to say „moist“ instead of „more“, and corrected the sentence, and also move the sentence to the caption in Fig. A2, as we do not discuss this result in the main text.

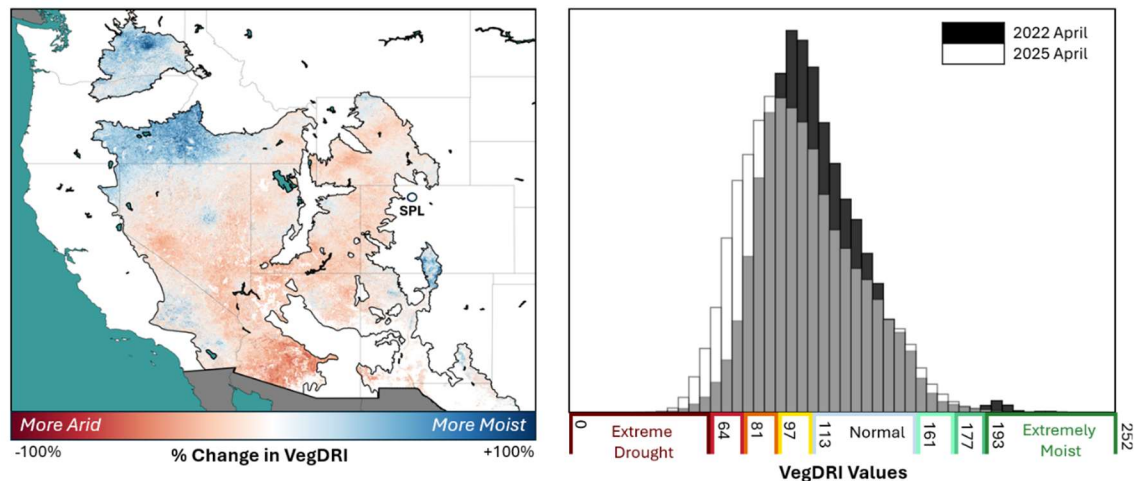


Figure A2: Surface map of percentage change in aridity of April 2022 as compared to 2025 of North American desert regions as indicated by the Vegetation Drought Response Index (VegDRI) (panel a) and distribution of extreme drought and extreme moist conditions during April 2022 and 2025 (panel b); VegDRI values presented in panel b correspond to aridity values in Figs. 7 and A3. The VegDRI was retrieved and averaged over April 2022 and April 2025 separately. Differences between dry and moist conditions between these two months were calculated as percentage differences: $100\% * (avg_2025 - avg_2022) / (avg_2022)$.

14. Line 246: The definition of spring used in this manuscript (March/April/May) should be moved up here, with the definitions of the other seasons. Also, February seems to be sometimes included with winter and sometimes excluded entirely, so please be clear in these definitions and consistent throughout the discussion.

We include the definition of spring here now, and also correct the statement that winter refers to December, January and February:

„In general, a seasonal variability is observed with a slight decrease in the median INP concentration from autumn (October/November) to winter (December/January/February) (Fig. 4; Tab. A1), and an increase in spring (March/April/May).“

15. Line 247: The statement that both years have similar values in the same month is clear from Fig. 4b, except for January. Could shading or some other uncertainty estimate be added to the figure to help with this assessment?

For clarity, we would like to keep Fig. 4 as it is and not add more information than the temperature dependent median INP concentration. Also, we show the interquartile-range in Fig. 6, which is giving more information about the data.

16. Line 276-277: The way this sentence is written: "...strong increase...in the median and inter-quartile range...", it sounds like sounds like the median increased and the range also got larger/wider during spring, which doesn't appear to be the case based on Fig. 5. Did you just mean that all the concentrations increased?

We meant that in general the INP concentration increased, which is visible by the median INP concentration and the inter-quartile range. We correct the sentence now:

„An interesting feature of the data is the strong increase in the INP concentration of approximately one order of magnitude in spring, with the median value and inter-quartile range mostly exceeding 10 INP stdL⁻¹ within the respective temperature range.“

17. 3.2: Some of the INP concentrations quoted are for -28°C, some for -30°C, and some don't have a temperature listed. Please list temperatures for all the measurements and consider standardizing the concentrations in this section to one temperature.

We would not like to standardize to one temperature, as we refer to specific features of the data. However, we corrected statements where it is not clear to which temperature we refer to:

„During both April 2022 and 2025, the highest INP concentrations were measured with values above 1000 INP stdL⁻¹ at -30 °C. For a better understanding of potential source regions during events of peak INP concentrations in the observed temperature range -22 to -32 °C, so-called footprints were calculated and combined with surface maps of aridity of arid ecoregions, namely North American Desert regions (see section 2.5). The median INP concentration and IQR these months are similar in the observed temperature range (see section 3.1), and interestingly, also the occurrence of extreme drought and drought conditions in April 2022 and 2025 are similar (Fig. A2).“

„Two case studies in April 2022 and 2025, when events of peak INP concentrations occurred were analyzed in more detail using back-trajectories in combination with aridity information (Fig. 7). An increase in INP concentration was observed across the entire measurement range (-22 to -32 °C). For clarity, the INP concentrations within the narrower range of -28 to -30 °C are presented below.“

18. Line 299: When, exactly, are the footprints plotted in Fig. 8 from? Are they some average over part of each event? The footprint coinciding with just the peak INP concentrations?

We assume the reviewer refers to Fig. 7. The footprints refer to the complete duration of the event, we add now to the corresponding sentence:

„The footprints of the back-trajectories during these events...”

19. Line 300: The two events in April 2022 have quite different footprints, with April 11 having sources directly west in a narrow latitude band, while the April 26 footprint covers almost the entire US desert ecoregion, with the strongest source coming from the southwest, not the northwest. If you want to talk about them together, perhaps just say "...indicate arid regions west of SPL contributed to the measurements, where pre-drought...".

Agreed and changed:

„The footprints of the back-trajectories during these events indicate arid regions west of SPL contribute to the air mass, where pre-drought to severe drought conditions were observed.“

20. Line 303: Suggest clarifying that “only pre-drought conditions occurred in the source region” for the April 7 2025 case because the footprint for this case is northwest of SPL, where the aridity is lower, whereas other cases are more west or southwest.

Thank you for this suggestion, we change the sentence now accordingly:

„However, in contrast to the other cases, only pre-drought conditions occurred in the source region. Also, in contrast to the other cases, the source region here is more north-west of SPL, which might explain the less strong increase in INP concentration.“

21. Lines 335-337: The final sentence in this paragraph feels out of place, which has no other mention of instrumentation, and also unbalanced/missing nuance. 2.5µm captures a significant portion of the supermicron aerosol number distribution in most cases. Although it is certainly true some portion of the distribution is missed by CFDCs, the same is true for PINE, with a maximum size of 4µm. Was a significant portion of the supermicron aerosol seen between 2.5-4µm in this campaign?

We agree that the sentence is out of place in this section and delete it.

The reviewer is right that no significant portion of particles was in the range of 2.5 - 4 µm during the campaign.

22. Line 346: Instead of just saying these correlations are “relatively high” compared to other studies, could the range of correlation coefficients be listed for one or a few examples, for comparison?

We agree and added:

„This is a relative high correlation coefficient compared to other studies. E.g., DeMott et al. (2010) investigated the relation between INP number concentration from global

measurements and particle concentration > 0.5 μm and found values between 0.6 and 0.8 for temperatures between -11.5 and -33.5 $^{\circ}\text{C}$. At a high-altitude station, correlation coefficients with values between 0.4 and 0.6 were found for INP concentration at -31 $^{\circ}\text{C}$ and particle concentration > 0.5 μm (Lacher et al., 2018b). Again, this might be an indication that larger aerosol particles such as dust contributes considerably to the INP population in general and at SPL during the respective months.“

23. Line 348: Was the correlation with smaller aerosols also tested? Showing that the correlation with submicron aerosols was smaller than the correlation with supermicron aerosols would strengthen this point.

Unfortunately, information on aerosol particle size distribution at smaller sizes was not continuously available throughout the campaign due to instrument maintenance; therefore, we did not assess the correlation between INP concentration and submicron particles.

24. Line 363-364: What is the rationale for separating the ice residual distribution at a size of 0.45 μm ? The focus throughout the rest of the paper, including Sec. 3.3.1, is on submicron vs supermicron particles, so this seems out of place.

This size was chosen as a break between small and large ice residuals based on the shape of the SPX size distribution of the flush (background), where a sudden change in particle concentration is observed at this size; we cannot exclude for certain that the small ice residuals are impacted by non-ice residuals, which we discuss in section 3.3.2:

„However, it is notable that smaller ice residuals were more abundant than large residuals, and their size distribution strongly resembles the background during flush (Fig. 10, panel a). It is possible that processes within the PCVI such as transmission of unactivated particles in the wake of ice crystals (Pekour and Cziczo, 2011) contributed to small residuals during expansions. However, based on the experiments by Pekour and Cziczo (2011) at much higher concentrations, only 1% of residuals might be artifacts from wake capture. Certainly, a so-far unknown mechanism in the PCVI being coupled to PINE could contribute to aerosol particles or small droplets being inadvertently transmitted. In future studies, such phenomena should be investigated further.“

To make the reasoning for a break at 0.45 μm more clear, we add

„At a size of 0.45 μm a clear reduction of particles in the flush (background) is observed. The ice residual concentrations during expansions of 31.9 and 29.7 stdL^{-1} (small particles below 0.45 μm , large particles above 0.45 μm) were higher than the

background concentrations during the flush mode of 0.18 and 0 stdL-1 by a factor of 180 and more than 2560, respectively (lower limit).“

25. Line 372: Is the larger enhancement for larger ice residuals due to a size-dependent enhancement in the PCVI? Different concentration factors are listed for the PCVI throughout the manuscript- how much did it vary over time and with particle size, and how was it measured?

The enhancement factor is certainly size dependent, however, here we meant simply that the larger particles ($> 0.45 \mu\text{m}$) are more abundant during the cloud formation process in PINE (flush mode) than during the background measurement when no cloud forms (flush mode). We update the respective sentence:

„The larger particles ($> 0.45 \mu\text{m}$) have a greater enhancement relative to the background, which is in agreement to the general finding in this study of the importance of super-micrometer sized INP.“

We did not determine the size-dependent concentration factor specifically. Please also see our answer to your major comment 2.2 regarding the different concentration factors listed.

26. Line 372: Please clarify the scientific reason for comparing the INP residual distribution to the SPX/PCVI background measurement? Shouldn't the residual size distribution be compared to the PINE OPC size distribution for the flush prior or immediately after the expansion of interest?

Unfortunately, the PINE OPC does not determine the true size distribution, as it is tuned to only measure larger particles (cloud droplets and ice crystals).

The background measurements refer to the ambient size distribution, and by comparing the size distribution during the background/flush to the expansion, we determine the contribution of the ambient particles to the ice residual size distribution. We clarify this now:

„The larger particles have a greater enhancement during the expansion mode, when ambient particles, cloud droplets and ice crystals can be present, relative to the size distribution in the flush mode, which is the size distribution of the ambient particles only. Thus, super-micrometer sized particles appear to contribute more to the INP population.“

27. Paragraph starting with line 381: This section would flow better if this paragraph (“Moreover...”) was moved up above the previous paragraph (“However, it is notable...”).

We would like to keep the ordering, as we first want to present the results and then discuss the uncertainties of them.

28. Line 393: What is meant by “PSL diameter”?

PSL refers to the particles used for calibration, Polystyrene Latex particles, we correct this now:

*„Sizes are reported as **Polystyrene Latex particles diameter used for calibration.**“*

29. Line 429: Same as Minor Comment #16. The change in inter-quartile range is likely to be interpreted as getting wider.

The inter-quartile range in the spring months is indeed increasing, with values being ~one order of magnitude higher than in the other months. Thus, we would like to keep the sentence as it is.

30. Line 433: What is meant by “INP concentration **is solid**”?

We meant here that the observation of increasing INP concentration in spring is solid/robust as we observe this in two years. We replace the word now:

*„We believe this observation of increasing springtime INP concentration is **robust**,...“*

31. Lines 436-437: The last sentence of the paragraph doesn't fit together. Both statements are true, but they don't go together. Changes in desert emissions may impact cloud properties, which will be important to measure. And improving model representations of ice processes are also important. But simply making more measurements at SPL or other sites will not improve modeled microphysics in a broad way. For that, measurements in a variety of environments, with differing aerosol, dynamics, and cloud properties are needed.

In our opinion an improvement in the representation of cloud microphysical processes, such as ice nucleation and growth processes of ice crystals, can be improved by comprehensive set of observations at a single site, as such longer term measurements help to put different items together (e.g., seasonal patterns, longer-term trends). Also, a single site such as SPL, or others such as the DOE ARM sites (e.g.,

Creamean et al., 2025) is facing different conditions regarding aerosol and cloud properties.

DeMott et al. (2010) clearly demonstrates that the “strong sensitivity of climate forcing to INP suggests that long-range import of INP from dust storms, boreal biomass burning, and pollution could lead to feedbacks on mixed-phase clouds, impacting precipitation”. In order to achieve major advances in representing INPs and their impacts on clouds and precipitation in regional and global cloud models, particle surface area (including supermicron particles), and size-resolved aerosol composition are needed, which is often provided by measurement stations, and which is useful to parameterize INP in models, as commonly used INP parameterizations often depend on both of these measurements. A major advantage of continuous long-term measurements at a stationary site is being able to track seasonal and diurnal changes. This data will provide a deeper understanding, which will place intensive field studies in context.

32. Line 447: Same as Minor Comment #1, be specific about what is “novel” about this setup.

We add now:

„Finally, a novel setup selecting ice residuals activated in PINE and separated in a PCVI was tested. This is the first time that PINE was coupled to a PCVI, which is different to existing setups, as the cloud formation process is based on a rapid reduction of pressure in PINE (expansion cooling). The setup was applied for continuous measurements in January and February 2022, where ice residuals sizes were analyzed with a SPX size distribution instrument.“

33. Line 664: Does the PCVI D50 depend on particle shape or density? Cloud droplets are generally spherical, while ice crystals are highly non-spherical, and cloud droplets, ice crystals, and aerosols all have different densities.

See our answer to your major comment 2.1.

34. Line 715: Are there any measurements to verify the assumption that $S < 1.01$? Does the cloud droplet maximum size meaningfully change if $S = 1.02$ or 1.03 , for example?

The supersaturation in PINE during expansion is not yet completely characterized, therefore this value is only an assumption.

However, the cloud droplet maximum size would only change slightly based on the calculations shown in Fig. A5. For $S = 1.01$, a maximum size of $5.41 \mu\text{m}$ is reached, while for $S = 1.02$ ($S = 1.03$), a maximum size of $5.42 \mu\text{m}$ ($5.6 \mu\text{m}$) is reached.

35. Line 716: What is SPL01?

We used an internal naming of the campaign (Storm Peak Lab 01), we change this now:

„For the calculation of cloud droplet size, the temperature and duration of a typical expansion during *the campaign*...”

36. Line 716: Would the expansion described, with a temperature decrease from -24 to -29°C, be considered a measurement at -29°C?

The reviewer is right that such a measurement is considered as a measurement at -29 °C, as this is the minimum temperature reached during expansion. We clarify this now:

„...the temperature decreased from -24 to -29 °C during 40 seconds (referred to an INP concentration measurement at -29 °C).“

37. Line 725: Citation missing for “Hinds”.

Corrected.

38. Grammatical suggestions are included in the attached pdf.

Thank you for reading and correcting the manuscript thoroughly, we included most suggestions (see track-change manuscript).

Figure/Table Notes:

1. Figure 1: The caption says “...only the larger ice crystals are of sufficient size...”. Does that mean there is a potential bias in the size or number of the ice crystal residuals if only a portion of the ice crystal distribution is sampled and measured by the SPX? Or do you just mean the ice crystals are larger than the cloud droplets and aerosols?

1. Also, a label for F_{out} is missing from the diagram.

We only meant that ice crystals are large enough (larger than cloud droplets and ice crystals) to be transmitted. We update Fig. 1 for F_{out} and correct the caption, and also considered the comments from your attachment:

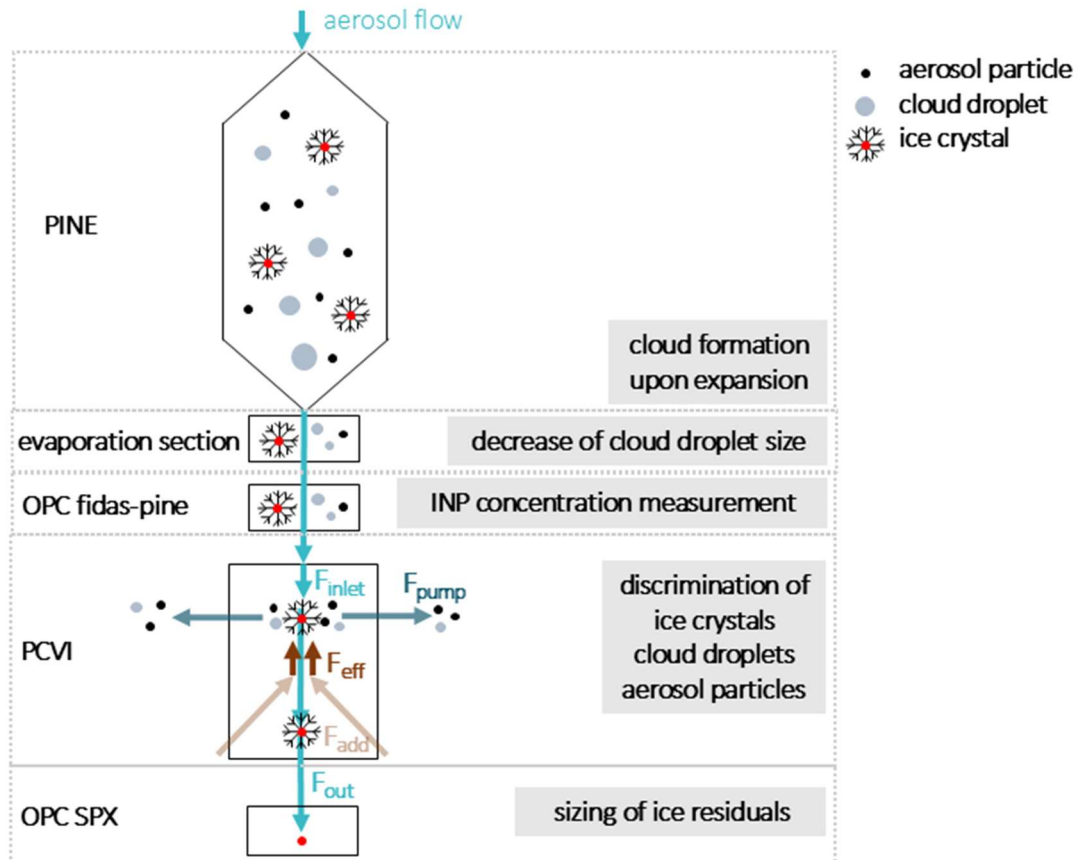


Figure 1: Setup of PINE coupled to a PCVI. The cloud droplets, ice crystals, and unactivated aerosol particles that form in PINE during expansion pass an evaporation section and the fidas-pine OPC. Upon entering the PCVI body, only the larger ice crystals are of sufficient size, *as compared to smaller cloud droplets and aerosol particles*, to be selected and are then, after sublimation of ice, further analyzed for their size. The discrimination between these particles in the inlet flow (F_{inlet}) is achieved by a combination of a pump flow (F_{pump}) and a counterflow (F_{eff}). *Ice residuals are transmitted in the output flow (F_{out})*.

2. Table 1/Figure 2a: The D50 for the PCVI is the same as was listed for the PINE inlet cutoff D50 (line 124), implying the largest aerosol particles/cloud droplets (if not evaporated enough) may pass through the PCVI and be counted as ice crystal residuals. From the PINE OPC, what was the typical ice crystal size distribution? Was there any overlap between ice, cloud droplets, and large aerosols $\sim 4\mu\text{m}$? If Fig. 2a shows PINE OPC size distributions during an expansion, is the ice crystal peak at $\sim 3.5\mu\text{m}$?

Indeed, some larger aerosol particles, which are not activated as cloud droplets and ice crystals, as well as cloud droplets which are not evaporated completely, might be able to pass the PCVI, however, based on the experiments and calculations presented here (Fig. 2; Fig. 10 and related discussion; appendix section „Validation experiments

PINE-PCVI“ and „Calculation of cloud droplet size and evaporation“), we believe that the contribution is little, also given that large aerosol particles are preferably activated as cloud droplets.

The PINE OPC is not determining the true size of the ice crystals, thus we unfortunately cannot make such a comparison.

Fig. 2 shows measurements from the SPX, which we clarify now in the caption:

„Figure 2: Size distributions measured during a PCVI diagnostic experiment at SPL at a temperature of -29 °C determined with the SPX...“

3. Figure 2: The PCVI concentration factor mentioned in the caption should be discussed in the methods section 2.4.

We discuss the concentration factor now in section 2.4 (see also our answer to your major comment 2.2):

„The PCVI concentrates the ice particles (and thereby the residuals) by a factor of total inlet to total outlet flow. One experimental challenge for a coupling a PCVI to PINE is that the ice residual characterization instrument(s) must adapt to the rapidly changing system pressure during a PINE expansion experiment. The SPX inlet flow, which is F_{out} , was set to control at a constant volumetric flow of 0.12 LPM. During the start of expansions the flow dropped to as low as 0.085 LPM before the nominal flow was re-established. This initial flow reduction increased the PCVI counterflow slightly and increased the concentrating factor to as high as 38.6 but otherwise had little effect on PCVI operation.“

4. Table 2: Could the min values listed as 0.1 instead be listed as <LOD, for clarity? Or make it clear in the caption that values of 0.1 represent concentrations below the LOD.

We add to the caption:

„Table 2: INP concentration statistics at each measurement temperature (± 1 °C) for the whole campaign. INP concentrations below the LOD depicted as values of 0.1 $stdL^{-1}$.“

1. Would it also be possible to list the 2022 and 2025 values here to see if there are any inter-annual differences? Something like "overall value (2022 value / 2025 value)" for each temperature? Table A1 in the appendix provides this information, but since it is also separated by month, the reader would have to calculate all these statistics themselves to get the annual values.

We would not like to include this information in Table 2, as this might be confusing the reader. We believe that the information about the individual months in Appendix Table A1 is sufficient.

5. Figure 4: The February line (light blue) and all line (light gray) are quite difficult to see. This is especially true for the dashed line in panel b. Consider making these colors darker, make the lines thicker, or otherwise make them more visible.

1. Does the "all" line in panel a include 2025, or just 2021/2022 data?

Thank you for the comment. We changed the line thickness to make it more visible, and added the information that the whole campaign (all) includes both campaigns in 2021/2022 and 2025.

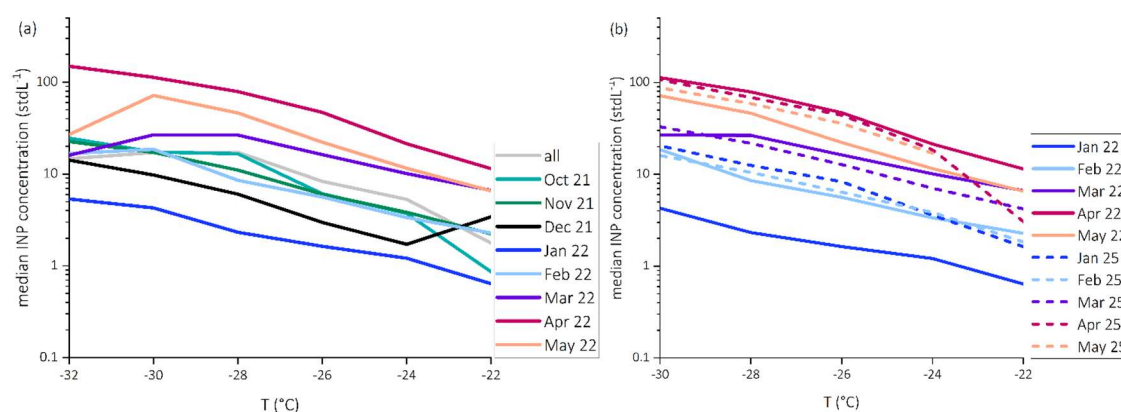


Figure 4: Median INP concentrations as a function of temperature (± 1 °C) for the whole campaign (2021/2022 and 2025) and the individual months (October 2021 – May 2022, panel a) and for the months January to May from the two field campaigns in 2022 and 2025 (panel b). Values below the LOD included as 0.1 INP stdL⁻¹.

6. Figure 5: Perhaps this is just the pdf conversion, but the legends in this figure are very hard to read. The boxes with colors are partially blanked out and vary in size, so it is unclear which color is which month/season/year. The same is true of the individual measurements in each panel- some are partially blanked out, some have outlines of a different color.

We improved Figure 5 regarding the mentioned drawbacks, caused by a too low resolution. We also changed the colors to match better the color scheme in Fig.6 and changed the label naming to make it clear that we refers to January and February (see also comment from RC2):

„In spring (March/April/May), an increase in the INP concentration is detected, with highest values in April when the median INP concentration increased to values above 10 stdL⁻¹ across the observed temperature range. As compared to January and February, when only 42% of the measurements are above 10 INP stdL⁻¹ (45% in January

and February 2022 and 33% in January and February 2025), 86% of the measurements in April are above this value (84% in April 2022 and 87% in April 2025; Fig. 5).“

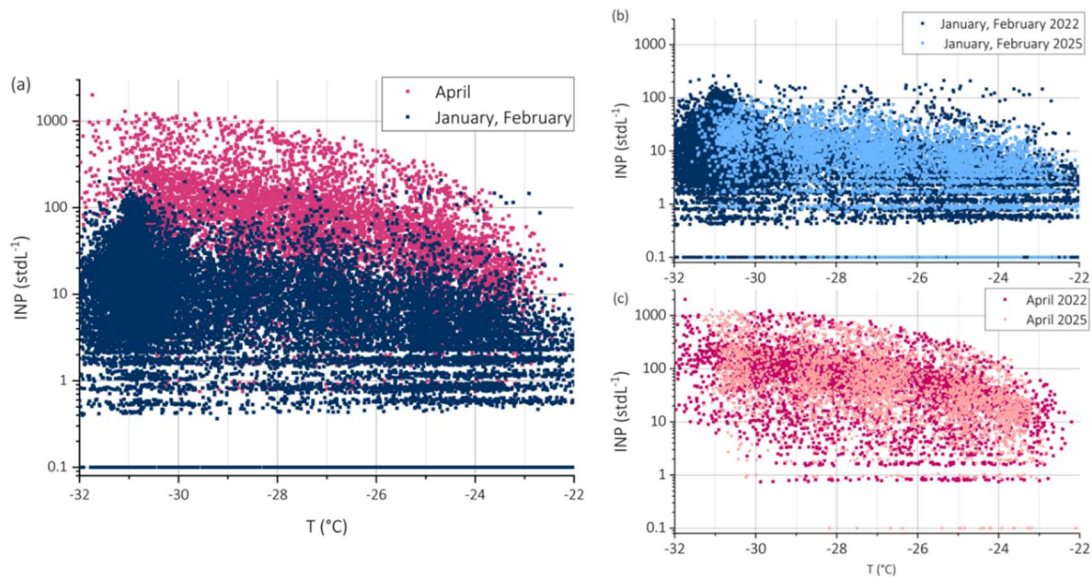


Figure 5: INP concentration as a function of temperature for the measurements in April and January, February from both measurement campaigns in 2021/2022 and 2025 (panel a), for the measurements in January, February 2022 and 2025 (panel b) and for the measurements in April 2022 and 2025 (panel c). Values below the LOD are shown as 0.1 INP stdL⁻¹.

7. Figure 6: Could the "all" range be added to all the panels, so it is easier to compare?

1. Since the different years are separated in this figure, please use the same colors for the same months. The lighter colors for winter 2025, and particularly February 2025, are hard to see.

For clarity of each panel, we did not include the all range, as each panel includes already up to three overlapping ranges. We updated the figure regarding the colors to match the seasons.

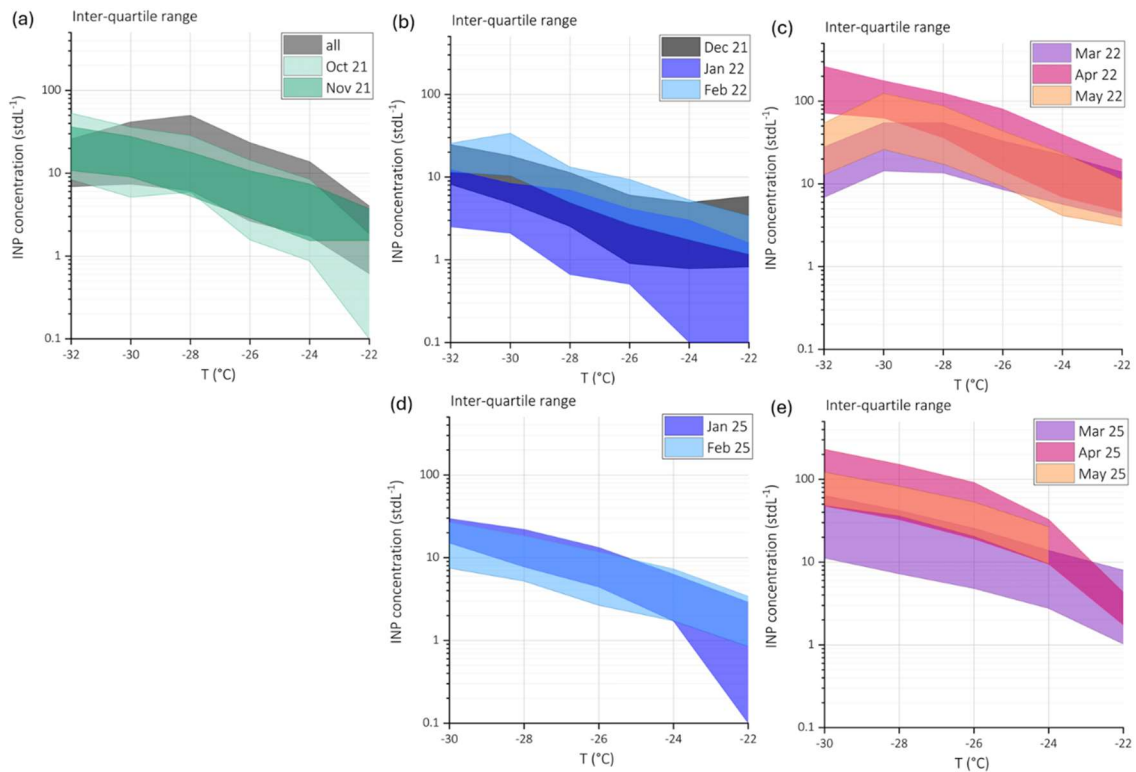


Figure 6: Inter-quartile range (25% to 75% of the data) of INP concentrations as a function of temperature ($\pm 1^\circ\text{C}$) for all the campaign and autumn 2021 (panel a), winter 2021/2022 (panel b), spring 2022 (panel c), winter 2025 (panel d), and spring 2025 (panel e). Values below the LOD are included as values of $0.1 \text{ INP stdL}^{-1}$.

8. Figure 7: Please list the years with each date, for clarity.

1. The figure caption appears to have been cut off in the middle; there is no description for panels b-d, the legend for the VegDRI colorbar, or the icon (I assume) depicting the location of SPL.

We added the years in the figure caption and also corrected the error of the cut caption text. Moreover, we also include the legend for the VegDRI colorbar and the icon for SPL.

We also found an error in the labeling of the contour line in panel d); the 10^{-6} on the west coast was originally labeled as 10^{-8} , which is wrong and corrected now:

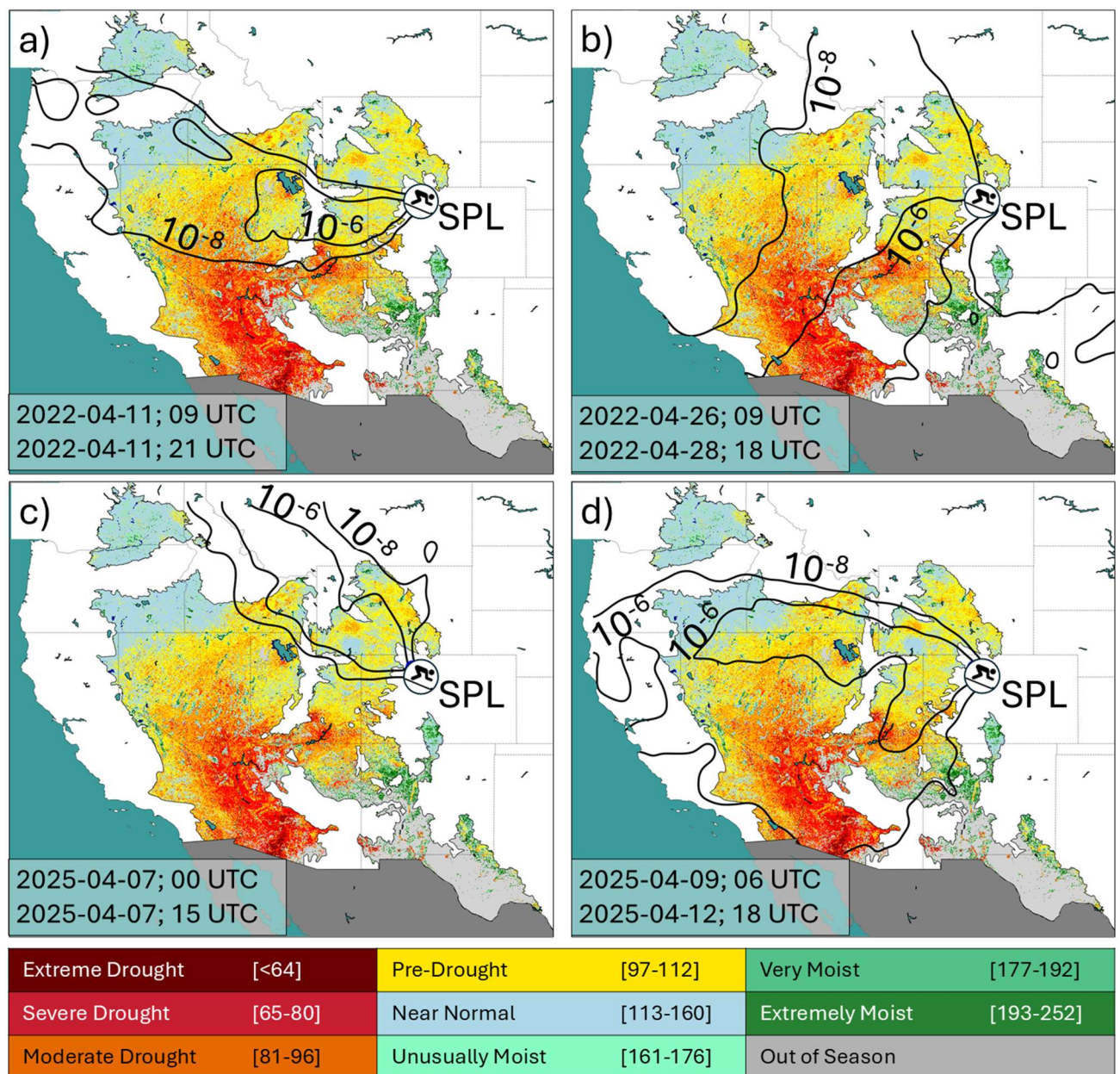


Figure 7: Footprints computed with the STILT model combined with surface maps of aridity using VegDRI of North American Desert ecoregions. Contours of the footprint are in units of $\text{PPM}/\mu\text{mole m}^{-2} \text{s}^{-1}$ (surface influence footprint; Fasoli et al., 2018). Events of elevated INP concentrations were analyzed on the 11th April 2022 (panel a, INP concentration at -30°C up to $1000 \text{ INP stdL}^{-1}$), on the 26th to 28th April 2022 (panel b, INP concentration at -30°C up to $1000 \text{ INP stdL}^{-1}$), on the 7th April 2025 (panel c, INP concentration at -30°C up to $500 \text{ INP stdL}^{-1}$), and on the 9th to 12th April 2025 (panel d, INP concentration at -30°C up to $1000 \text{ INP stdL}^{-1}$).

- Figure 8: The right y-axis color is much lighter than the corresponding symbols for aerosol concentration. Could they be made to match?

1. Could the size be explicitly listed on the right y-axis label? ie "particle concentration > 1 μ m".
2. Could the particle concentration <1 μ m also be shown, or was that not measured?

The color of the y-axis was adjusted to the color of the marker of the aerosol particle concentration > 1 μ m, and also the size is now explicitly named in the y-axis label. However, the particle concentration < 1 μ m was not measured.

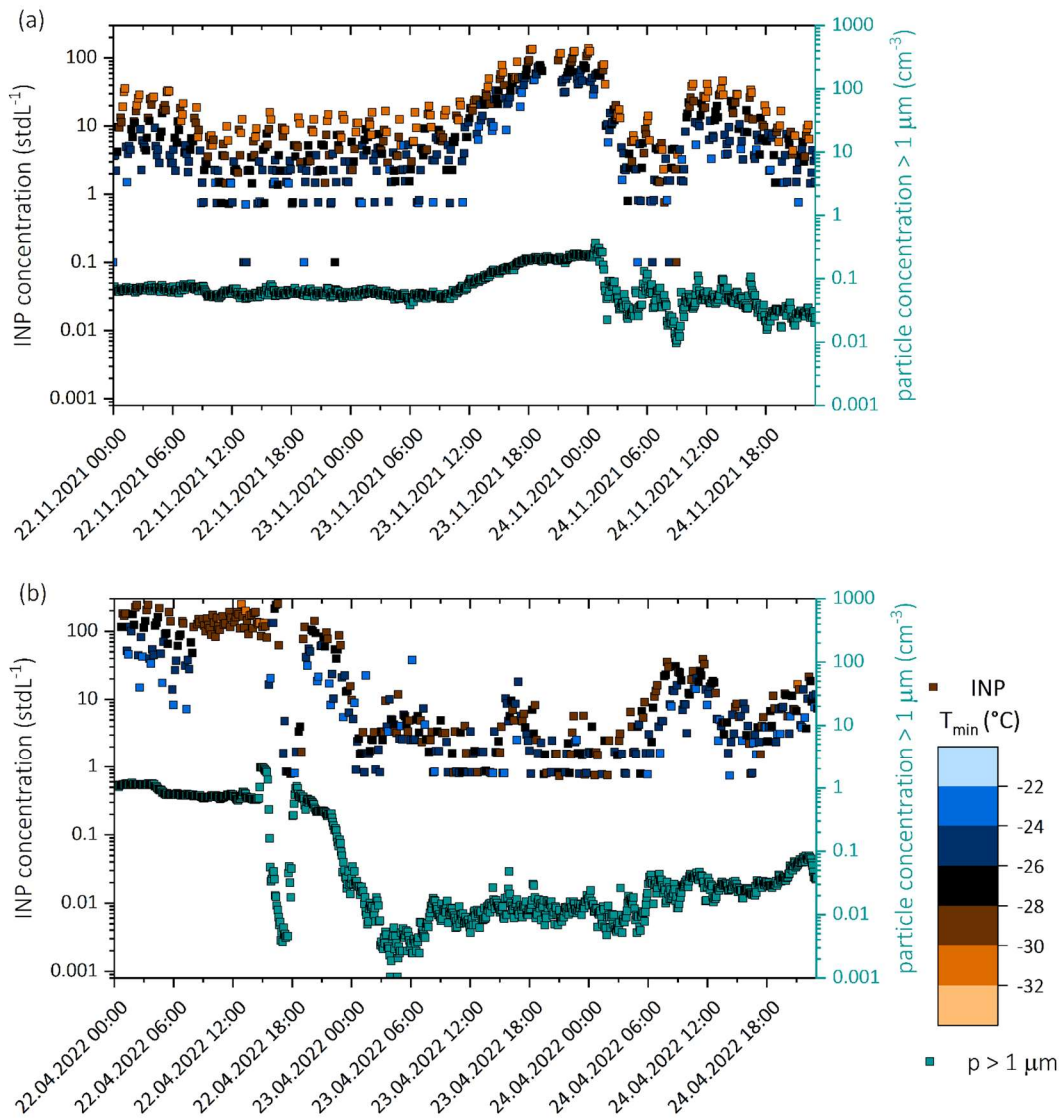


Figure 8: Timeseries of the INP concentration and aerosol particle concentration larger than 1 μm from November 22nd to 24th 2021 (panel a) and from April 22nd to 24th 2022 (panel b). Blue and brown colors present the nucleation temperature (± 1 °C), and green aerosol particle concentration larger than 1 μm . INP concentrations below the LOD are presented as values of 0.1 stdL^{-1} .

10. Figure 9: The text is very small on this figure, could it be increased to improve readability?

1. Could the correlation coefficient be listed on each panel, for comparison?

Thank you for these suggestions, we changed the text size and listed the correlation coefficients in the panels.

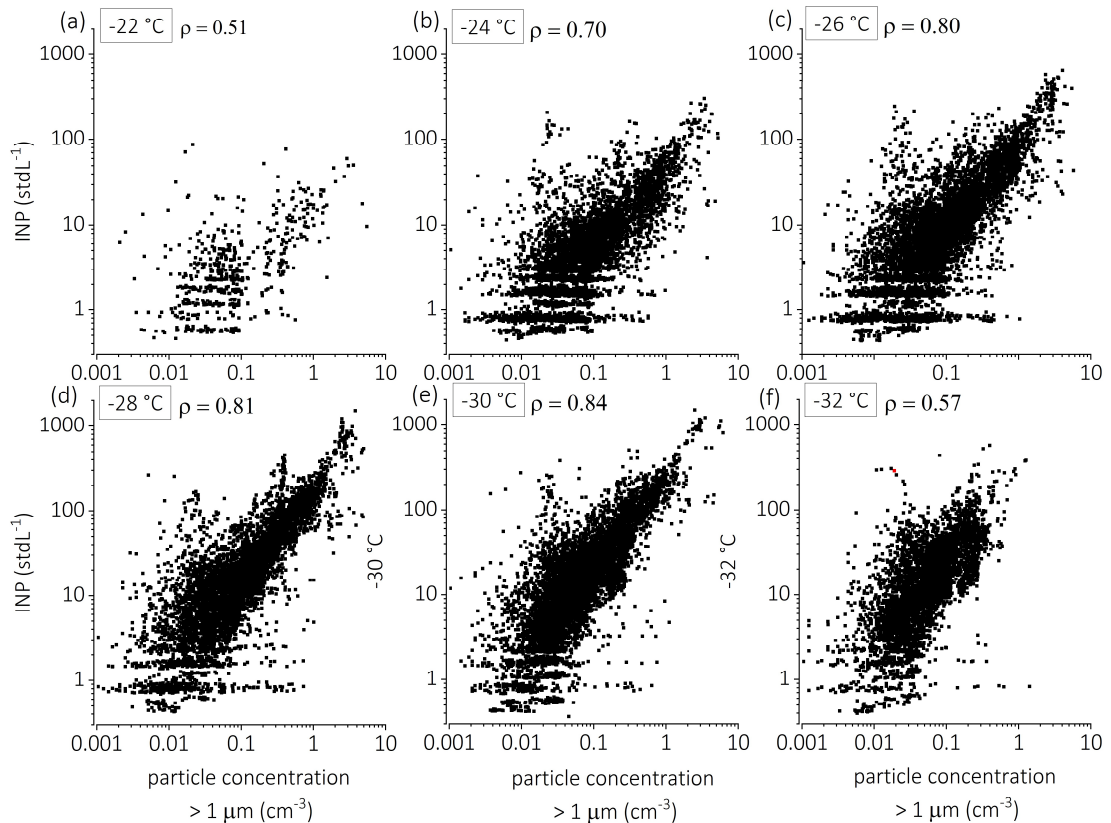


Figure 9: Relation between the INP concentration and particle concentration larger than 1 μm for the whole campaign at temperatures between -22 and -32 °C (panels a – f). *Spearman's correlation coefficient ρ is given for each temperature.*

11. Figure 11: The lines, axes, and text on this figure are very light/faded.

We made the lines, axes and text thicker.

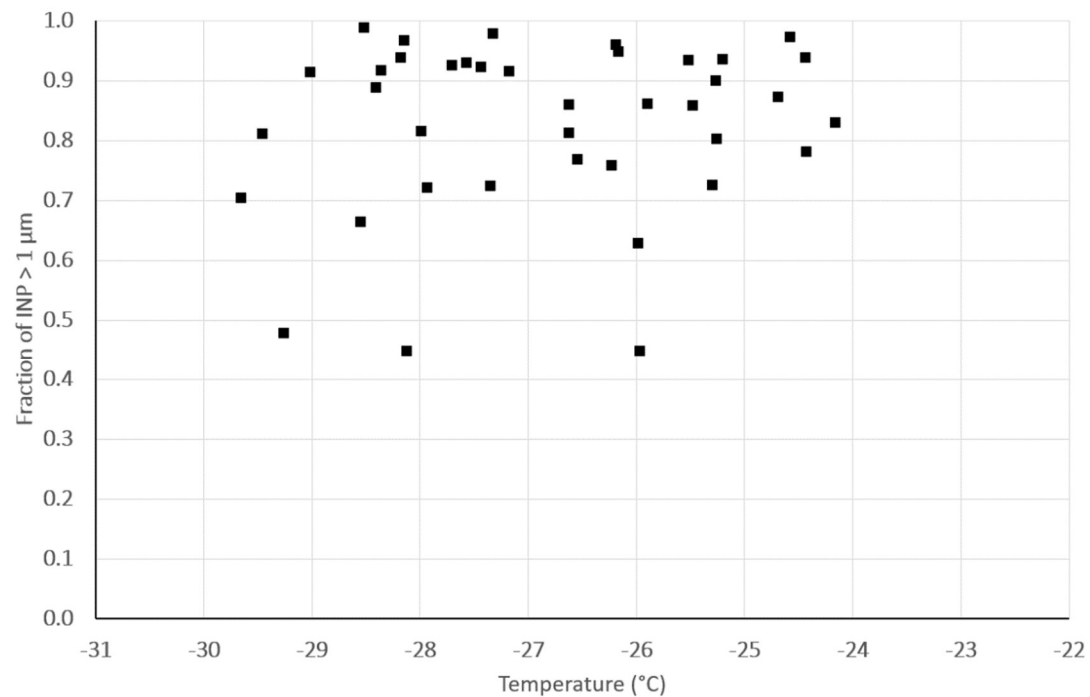
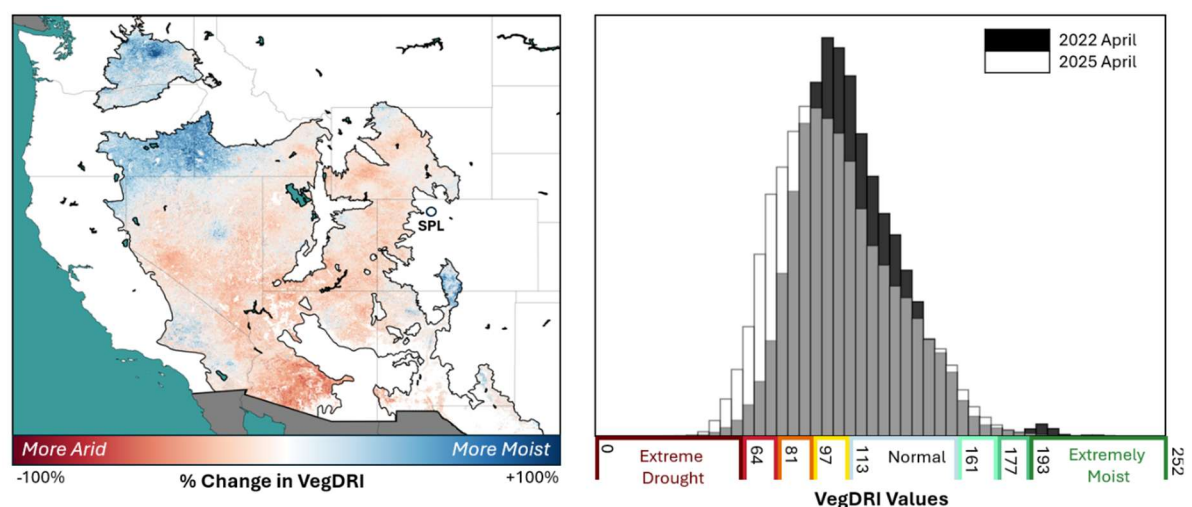


Figure 11: Fraction of INP larger than $1\ \mu\text{m}$ as a function of temperature. Fractions were calculated using consecutive measurements on and off the concentrator in May 2022.

12. Figure A2: What is the “percentage change in aridity relative to? The previous month? Some climatology for April?

The plotted aridity change in panel a is the percent change in average DRI index between April 2022 and 2025. The red represents where the DRI index decreased in 2025 relative to 2022 (more arid), and the blue represents where the DRI index increased in 2025 relative to 2022 (more moist). The percentage change in aridity is calculated as $100\% \times (\text{avg_2025} - \text{avg_2022}) / (\text{avg_2022})$ (see new text in figure caption). We update the legend.



„Figure A2: Surface map of percentage change in aridity of April 2022 as compared to 2025 of North American desert regions as indicated by the Vegetation Drought

Response Index (VegDRI) (panel a) and distribution of extreme drought and extreme moist conditions during April 2022 and 2025 (panel b); VegDRI values presented in panel b correspond to aridity values in Figs. 7 and A3. The VegDRI was retrieved and averaged over April 2022 and April 2025 separately. Differences between dry and moist conditions between these two months were calculated as percentage differences: $100\% * (\text{avg_2025} - \text{avg_2022}) / (\text{avg_2022})$."

1. The scale/colorbar for panel b needs more information. What do values in the middle mean? Where is "normal"? Are they numerical, or only categorical values?

Agreed and changed (see above).

2. Can April 2022 and April 2025 be shown separately instead of averaged together? The main text notes they are similar, but it is not possible for the reader to see that when they have been averaged together.

We corrected the legend to make it clear that it is not showing an average, see also our answer to comment 12.

13. Figure A4: Should the y-axis label on panel b just be "SPX IR (stdL-1)"? If both sets of markers in panel b are really small IR, a different legend is needed for panel b.

Thank you for the suggestions, we updated the figure accordingly:

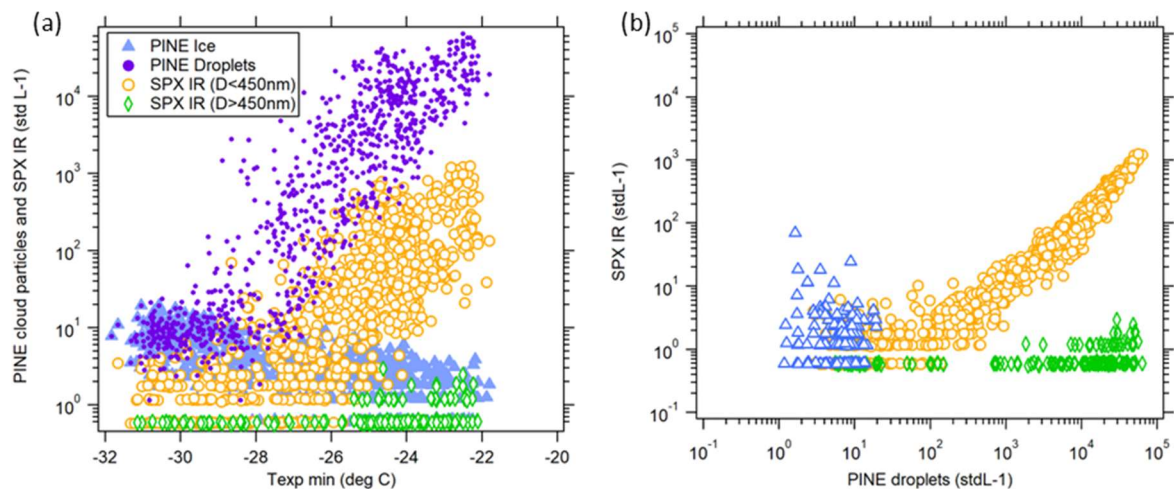


Figure A5: Comparison of PINE residual concentrations (PINE Ice) with PINE cloud particle concentrations (PINE Droplets) during a series of 909 expansions on Jan 15-16, 2022 where the minimum temperature was varied from -22 to -32°C. Panel a) Both small residuals (SPX IR (D<450 nm) and PINE droplet concentrations rise with increasing temperature (panel a). Panel b) A strong correlation is observed between small residuals and droplet concentration (at larger than 100 droplets stdL-1), corresponding to temperatures above -28 °C, indicating that the PCVI is transmitting droplets as well as ice. At temperatures below -28 °C, small residual concentrations

become constant and represent the combination of small INP and interstitial aerosol breakthrough in the PCVI. Residual concentrations are corrected by the PCVI concentrating factor.

1. Could a correlation between PINE INP and SPX IR be shown in Fig A4 as well? That would support the claim that only ice crystals are being transmitted. Since the PINE droplet concentration is fairly constant below -28°C, the SPX IR also being constant below that temperature is not conclusive by itself.

Unfortunately for this set of experiments under clean conditions, the PINE Ice concentrations (at $T < -28^{\circ}\text{C}$) had low variability, and the broad scatter of PCVI ice residual data near detection limit, and thus no significant correlation was found.

14. Figure A5: The axes and text on this figure are very faint and small.

Corrected. We also noted a minor mistake in the data, which is corrected now.

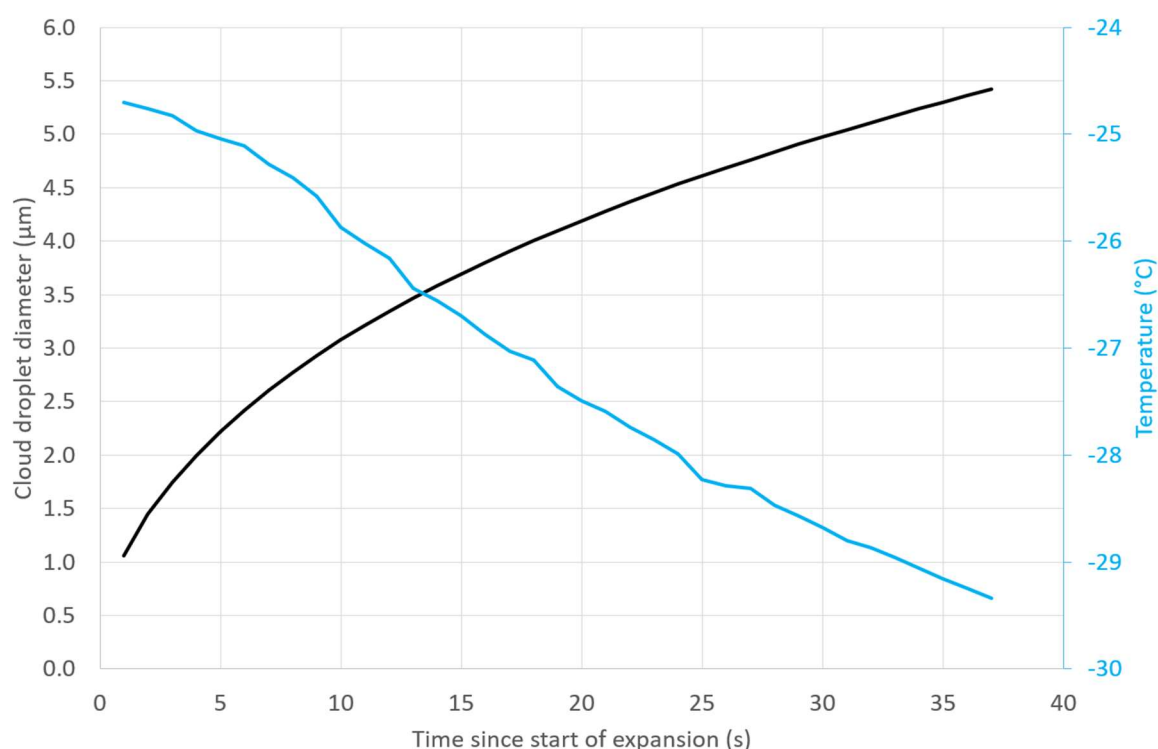


Figure A5: Calculated cloud droplet diameter during a PINE expansion with a start temperature of -25°C , based on diffusional growth calculations. Temperature measurements were taken from a typical expansion with an expansion flow of 3 LPM. Initial droplet sizes entering the PCVI are smaller than this calculation due to the usage of an evaporation section.

15. Table A2: What do “percentage completions” above 100% indicate?

This indicates that the cloud droplet is completely evaporated, and simply is the value based on the formula used.

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