

Response to Referee Comment 1, point by point

egusphere-2026-3543

Companion to the revised manuscript (v11) and its Supplement (v7)

How to read this

The report is not numbered; it is written as prose under three headings (*Data removal*, *Including zero counts* and *Correction factors*) after a general assessment. We have labelled each question A1–A6, B1, C1–C4 and D1–D2 in the order of the report, and quote the referee’s text in full for each.

The preprint is not amended, on the handling editor’s instruction; Author Comment AC1 is the correction of record. Corrections the referee did not raise are listed in **AC1.Add.pdf** and are cross-referenced here where relevant. Section, table and figure numbers under “Where” and “Carried into” are those of the revised manuscript and Supplement, which will be submitted to the editor after this discussion and differ in places from the preprint.

Summary

	Referee’s point	Verdict	Carried into
A1	Is the LOD calculation correct?	Accepted; premise corrected	Sect. 2.6; Table 2
A2	Why applied to Figs. 3 and 4?	Premise corrected; cause identified	Sect. 4.1; Table 2 caption
A3	Why not applied to transit and Jeju?	Premise corrected	Sect. 2.6; Table 2
A4	Data removed below 3 L ⁻¹ biases high	Accepted in full	Table 2; Abstract; Sects. 3.1, 3.2, 4.1
A5	Confirm the Jeju 0.15 L ⁻¹ limit	Two causes found, neither a volume	Fig. 4a; Fig. 3; Table 2 footnote
A6	Add Jeju Level 1 to Fig. 4	Added	Fig. 4(c–e)
B1	Resample including zero counts	Accepted in full	Sect. 2.6; Table 2
C1	CF methodology flawed; does not belong	Accepted without qualification	Sect. 4.4; the correction-factor appendix of the discussion paper withdrawn
C2	Read Robinson et al. (2026)	Read; adopted	Sect. 4.4

	Referee’s point	Verdict	Carried into
C3	Size-range claim needs evidence	Withdrawn <i>and reversed</i>	Sect. 4.4
C4	Revisit whether CFs are needed at all	No; they are not	Sect. 4.4; Supp. Fig. S5
D1	Occasionally verbose	Repetition removed; still longer	Abstract; Figs. 3–5; Sects. 2.2, 2.5.2, 3.4, 4.4
D2	Not publishable in current form	Understood	—

A. Data removal

A1 Is the LOD calculation correct?

“Is the LOD calculated by the authors correct? Even if only a single expansion was included in their LOD equation (Eq. 1) this should equate to $3/2 = 1.5 \text{ L}^{-1}$.”

The arithmetic is right: a single 2 L expansion gives 1.5 L^{-1} . Our limit is computed over the pooled period rather than one expansion, so it is much smaller; on the revised daily pooling it ranges from 0.020 to 0.062 L^{-1} for ARA15A. However, the Sect. 2.6 limit was never applied to Figs. 3 or 4 (see A2), so it is not what produced the floor. Every concentration in Table 2 now carries its own limit.

Where: Sect. 2.6 (*Data analysis and statistical methods*); Table 2, bracketed values in every cell.

A2 Why was the limit applied to Figs. 3 and 4?

“Why was this applied to the data presented in Figure 4 (and Figure 3) when it is stated to only be used for the climatological trend analysis?”

It was not, but the question led us to the cause of the floor. Two effects combined. First, the plotting code kept only bins with `cn_ice > 0`; in the quality-controlled ARA15A Level-2 record, 52,745 of 59,867 temperature bins (88.1 %) contain no ice crystal, and 90.3–97.4 % between -25 and -19°C . Second, the remaining values are quantised: a 0.5°C bin samples 0.15–0.23 stdL, so one crystal gives $1/V_{\text{bin}}$, with an ARA15A median of 4.42 L^{-1} .

There is no hard cut at 3 L^{-1} . The smallest non-zero Level-2 value is 1.526 L^{-1} and 65 values are below 3; the 1st and 5th percentiles, 3.13 and 3.33 L^{-1} , form the floor the referee saw. As a result the published ARA15A cells (3.58 , 3.68 , 3.67 and 3.66 L^{-1} at -19 , -20 , -22 and -25°C) equal the one-crystal quantum to two decimal places. Their near-constancy across 6°C , which we had read as a physical flatness, comes from the estimator and not from the air.

Where: Sect. 4.1; caption of Table 2, which now states what produced the superseded values; Fig. 3, where the quantum is drawn on the frequency panels.

A3 Why was it not applied to the transit and Jeju data?

“Why was this not applied to the ‘Transit data’ and Jeju data?”

The zero-exclusion filter was applied to Jeju too, in the same line of code. Its effect there is small because Jeju concentrations lie well above the quantum: the Level-2 bin median is 19.6 L^{-1} and

51.5% of bins are zero, against 88.1% in the Arctic. The same code was harmless for one dataset and distorting for the other. All datasets now receive the same daily-pooled treatment.

The transit runs never entered the run-level product, because the contamination screen of Sect. 2.5.2 drops runs without a wind record and the 2024 transit (two operations running 11–28 July from 33.8 to 59.6° N) has none. Sect. 3.3 had therefore used a separate file that had not been screened and from which 47.9% of the expansions had been discarded. It has been recomputed from the run-level product with zeros kept. The bimodal profile remains, the minimum moves from 45–55 to 55–60° N, and the 2025 transit is withdrawn because all its runs fall in the excluded inlet-malfunction period (item 3.6 of AC1_Add.pdf).

Where: Sect. 2.6; Table 2, where every campaign is on one estimator.

A4 The removal biases the dataset high and may mask the true temperature dependence

“Removing the data below 3 L^{-1} strongly biases the dataset towards higher values — and may result in the largely temperature-independent INP spectrum used in all subsequent analysis in the study. The absence of strong temperature-dependence is routinely seen as evidence of biogenic influence, yet if the data is biased high then this may be masking the true temperature dependence.”

Accepted in full. All Arctic concentrations are recomputed by daily pooling, with zero-crystal expansions kept in the denominator and nothing imputed for a zero. The median change across the nine Arctic cells of Table 2 is a factor of 0.027, about one part in 37; across all 18 cells, Jeju included, it is 0.35. At -20°C ARA15A goes from 3.68 to 0.099 L^{-1} .

The temperature dependence the referee expected is there. Over the cold regime (-31 to -24°C), where the concentrations are 11 to 56 times their own detection limits, ARA15A gives $\beta = -0.166 \text{ K}^{-1}$ ($R^2 = 0.97$). After the ARA16A wind file was rebuilt (items 3.7 and 3.51 of AC1_Add.pdf), that campaign has 1,162 runs in 164 daily periods instead of 200 runs in 18, and gives $\beta = -0.129 \text{ K}^{-1}$ ($R^2 = 0.93$). Both campaigns are therefore steep in the cold regime. Over the warm regime no slope is resolved ($R^2 = 0.01$ for ARA15A), and we do not give a value for it. We no longer compare these slopes with the -0.07 to -0.09 K^{-1} band of the discussion paper, which we could not trace to any source. Slopes derived from the deposited spectra of four Arctic filter records range from -0.117 to -0.165 K^{-1} ; ARA16A falls inside this range and ARA15A 0.001 K^{-1} beyond it (items 3.15 and 3.16 of AC1_Add.pdf).

The three arguments in Sect. 4.1 against an instrumental artefact are withdrawn, because a saturated statistic is flat within any subset and cannot be tested that way. The campaign-wide medians (8.02 , 6.83 and 6.57 L^{-1}) recompute to 12.1 , 0.524 and 0.265 L^{-1} and have since been removed at the second referee’s request (RC2QnA.pdf, R2-2); taken temperature by temperature, the Jeju-to-Arctic ratio runs from 2.5 to 158 . The “spectral crossover” described in the Abstract does not exist.

Where: Table 2; Abstract; Sect. 3.1 (Overview); Sect. 3.2 (Temperature spectra); Sect. 4.1 (Source characteristics); Sect. 4.4; Supp. Table S6 and Fig. S6; Table 1 note ^c for the ARA16A rebuild.

A5 Confirm the Jeju lower limit of 0.15 L^{-1}

“[I]nspecting Figures 3e to 3i and the box plots in Figure 4a suggest a lower limit of around 0.15 L^{-1} . If this is from the Level 1 data then this implies a much larger volume was sampled (around 6 L). Can the authors check and confirm the data is correct?”

Checked. There are two causes, and neither is a sample volume. First, a small correction to the arithmetic: the Jeju protocol samples 1.94 L , not 1.5 L , so the minimum non-zero concentration is 0.516 L^{-1} rather than 0.66 ; for Araon it is 2.00 L and 0.500 L^{-1} . The two campaigns are therefore even closer than the referee estimated.

In Figs. 3e–3i the Jeju panels were built from PIA operation-mean files, which average up to 40 expansions per bin and so reach 0.025 L^{-1} , while the Arctic panels used single-expansion data. Both

are now rebuilt from per-expansion Level-2 spectra. In Fig. 4a the 0.15 L^{-1} was the axis clip constant of the plotting routine, not a concentration; for Jeju Clean the 10th and 25th percentiles and the median across the 27 temperature bins are all zero. The revised panel marks zero quantiles at the axis floor and shows the one-crystal Jeju limit of 0.516 L^{-1} . Five published Table 2 cells have no run-level counterpart and are left blank with a footnote.

Where: Fig. 4a and its caption; Fig. 3 (2 of 2); Table 2 footnote *a*; Sect. 2.2.1 for the category dates.

A6 Can the Level 1 Jeju data be included on Figure 4 as a third panel?

“And can the level 1 Jeju data be included on Figure 4 as a third panel?”

Added, as a third row (c–e) below the two original latitudinal panels (a) and (b). The lowest stripe in each campaign is the one-crystal quantum:

Panel	Runs	Sampled volume	Lowest stripe	Counted no crystal
(c) ARA15A	5,596	2.00 L	0.500 L^{-1}	54.2 %
(d) ARA16A	1,162	1.99 L	0.503 L^{-1}	59.0 %
(e) Jeju 2024	14,345	1.94 L	0.516 L^{-1}	8.4 %

No Level-1 Jeju value reaches 0.15 L^{-1} , and the three floors agree within 3 %. What separates the datasets is the share of runs with no crystal, 54 % in the Arctic against 8.4 % at Jeju, which is why the same processing distorted one dataset and not the other. A further panel (f), on a second page, shows the latitudinal transect recomputed on the daily-pooled basis.

Where: Fig. 4(c–e) and its caption; Fig. 4(f) on the second float page.

B. Including zero counts

B1 Resample the timeseries including zero-INP expansions

“[O]ther PINE users have resampled the timeseries and taken means of the INP concentrations, including the zero INP expansions. Figure 2 from Herbert et al. 2026 demonstrates this. I suggest the authors consider performing this extra step before presenting their data as it will help remove some of the temperature independence that is seen in the dataset.”

Accepted in full. Expansions are pooled by day within $\pm 1^\circ\text{C}$ of each reported temperature. The pooled concentration is total crystals over total sampled volume, so expansions with no crystal count in the denominator, and no value is imputed for a zero. The detection limit is $3.0/V_{\text{total}}$ (Sect. 2.6). Computing per expansion instead gives almost the same result (0.107 against 0.099 L^{-1} at -20°C for ARA15A).

Pooling does not lift every temperature clear of the limit:

ARA15A	−28	−25	−22	−20	−19
Above own limit	55.6×	11.2×	2.2×	5.0×	1.5×
Periods above limit	25/25	24/25	21/23	23/23	16/21
ARA16A	−28	−25	−22	−20	−19
Above own limit	4.4×	1.4×	1.0×	0.9×	—
Periods above limit	11/14	8/13	11/16	9/16	—

Cells close to or below their limit, such as ARA15A at -19°C and ARA16A at -20°C , are reported with their limits and nothing is built on them.

Herbert et al. (2026), the GloPINE dataset paper, uses the same convention: expansions below the limit are kept, and expansions are combined so that the sampled volume grows and the limit falls. The only difference is the averaging interval (6 h and 2 K in GloPINE, one day and $\pm 1^{\circ}\text{C}$ here), which changes the result by a few per cent.

Where: Sect. 2.6; Table 2, bracketed limits and period counts; footnote *b* of Table 2 for the -19°C caution.

C. Correction factors

C1 The methodology is flawed and the analysis does not belong in this paper

“I don’t believe there is sufficient justification for combining INP datasets by including correction factors to bring them in line — to then look at something as fundamental as linking retreating sea ice with INP concentrations in the Arctic. The methodology is flawed and the current analysis does not belong in this paper.”

Accepted without qualification. The fixed correction factor and the harmonisation of historical datasets onto the PINE scale are removed. The correction-factor appendix of the discussion paper is withdrawn with its table and figures, as are Supplement Figs. S3 and S4, which did not plot measured data (item 2.1 of AC1_Add.pdf); the comparison is now made on measured spectra in Fig. 7.

The sea-ice result does not depend on the factor. With published filter values used unscaled, $\rho = -0.57$ ($p = 5.2 \times 10^{-6}$, $n = 55$), and the Newey–West HAC slope is -0.245 $[-0.41, -0.08]$, against -0.22 $[-0.36, -0.09]$ on harmonised values.

Where: Sect. 4.4; Sects. 1, 2.6, 2.7 and 5; Fig. 6; Supp. Sects. S4, S14 and S17, Table S9, Fig. S5.

C2 Read Robinson et al. (2026)

“I recommend the authors read Robinson et al. (2026) [...] Their results suggest there is a strong element of temperature-dependence to the differences, as well as from the INP species / material. If this is the case, then a systematic CF, as applied in this study, will not be able to account for these differences.”

Read. Robinson et al. (2026) state that without knowledge of composition and mixing state a universal correction factor cannot be applied, which is why we withdrew the factor rather than revising it.

The offset also changes sign between aerosol regimes. In continental air PINE reads low because its inlet excludes coarse particles (see C3); in remote marine air the filter reads low because free ice-nucleating macromolecules are not fully recovered from the substrate. Wilbourn et al. (2024) ran the same PINE-03 chamber at two sites: at continental SGP the online and offline values agree, 2.33 ± 0.50 against $1.90 \pm 0.21 \text{ L}^{-1}$ at -20°C , while at remote-marine ENA they differ by one to two orders of magnitude, 0.02 against $0.40 \pm 0.03 \text{ L}^{-1}$. A constant factor cannot represent this. An earlier draft gave ranges of 3.3–79.7 at ENA and 0.5–1.2 at SGP; these are not in Wilbourn et al. (2024) and are withdrawn. The ratios now shown in Fig. 7 and Supplement Sect. S9 (3.3–80 at ENA, 0.47–1.18 at SGP) are recomputed from the PANGAEA deposit and labelled as such; what is withdrawn is their attribution to the text of Wilbourn et al. (2024). Wilbourn et al. leave the cause open, so the mechanism above is our interpretation.

Following Robinson et al., published filter concentrations are now treated as a lower bound rather than rescaled. Their wash-off curves are also flatter above -22°C , so spectral flatness can itself be a filter artefact, which is a further reason not to read flatness as a sign of biogenic influence.

Where: Sect. 4.4.

C3 What size range does PINE measure, and what evidence supports the claim?

“With regards to the justification in Section 4.4. ‘First, the PINE chamber samples a broader size and freezing-mode population than immersion-only offline filter assays’. What size ranges does PINE measure? [...] PINE has significant losses of particles around 5 micron (see Mohler et al., 2021 and Knopf et al., 2021 SI) — is this better than the filter-based methods? [...] This kind of statement needs to be supported by evidence.”

The claim does not hold; it was wrong in direction. Möhler et al. (2021) report inlet losses below 20 % under $2\text{ }\mu\text{m}$ and about 50 % at $4\text{ }\mu\text{m}$, and do not report the $5\text{ }\mu\text{m}$ loss attributed to them. Knopf et al. (2021) describe an upper size cut-off rather than a loss: PINE-c and the CSU-CFDC sample up to 5 and $2.5\text{ }\mu\text{m}$ aerodynamic diameter respectively. For the $2.6\text{--}5$ and $5\text{--}12\text{ }\mu\text{m}$ fractions roughly $0.04\text{--}0.5\text{ INP L}^{-1}$ were detected at -20°C , and filter sampling has no comparable cut-off. PINE therefore misses part of the coarse-mode INP that filters collect. The statement is withdrawn and replaced by its converse.

Where: Sect. 4.4.

C4 With the data corrected, are correction factors needed at all?

“[I]f the data below $3L^{-1}$ has been erroneously removed from the Araon datasets [...] then it might be that the PINE data falls closer to the filter-based data. I recommend the authors look into their data to ensure it is correct, then revisit this analysis to see whether CFs are even needed.”

The referee’s expectation was right, and the gap largely closes. At -20°C the revised ARA15A value is 0.099 L^{-1} , below the 0.44 L^{-1} summer mean of the MOSAiC filter record reported by Barry et al. (2024). Recomputed from the archived MOSAiC files, the same record’s summer median is 0.080 L^{-1} , so on a median basis the ARAON value is the higher; the comparison will be restated on one estimator (RC2QnA.pdf, closing note). On either basis the difference is far smaller than the factor the correction was meant to close. The comparison is season-matched (the autumn MOSAiC value is 0.016 L^{-1}), and the revised values fall within the factor of five reported by Lacher et al. (2024) for most methods.

The recomputed sensitivity analysis shows that dropping the factor costs nothing: from $\times 1$ to $\times 47$ the Spearman correlation stays between -0.38 and -0.57 , and only the linear slope changes. The $\times 1$ case is the only one that needs no assumption about composition or mixing state.

Where: Sect. 4.4; Supp. Sect. S14, Table S9 and Fig. S5.

D. General

D1 The paper is a little verbose at times

“The paper is very well written — though perhaps a little verbose at times.”

Addressed in two passes, although the revision is still longer than the discussion paper. Withdrawing the correction-factor appendix removed about three pages, and two Supplement figures went with it. We then removed repetition: the abstract, which had reached 504 words in an intermediate draft, was cut to about 370 words (the discussion paper’s abstract had about 280), repeated exclusion counts and cycle times were cut, a 178-word sentence listing the contributions became a list, long paragraphs in Sects. 2.5.2, 3.4 and 4.4 were tightened, seventeen numerical substitutions moved to Supplement Table S11, and results were moved out of figure captions. The intercomparison evidence behind Sect. 4.4 moved to Supplement Sect. S19, and the account of the repaired data deposit moved

to **AC1_Add.pdf**. In paragraphs of 250 words or more the text fell from 8,115 to 6,703 words, and the number of paragraphs over 400 words from nine to four.

Typeset identically, the revision is 44 pages against 41, mainly because every value now carries its own detection limit and the corrections themselves take space. Sect. 3.3 was left at its length on purpose, because it carries the detection-limit argument behind A2 and A4.

Where: Abstract; Figs. 3, 4 and 5 captions; Sects. 2.2, 2.5.2, 3.4, 4.4 and 5; Supplement Table S11.

D2 Not publishable in its current form

“I have several major concerns with this study — to a degree that I will not provide a full review until my primary concerns have been addressed. [...] For these reasons, I do not recommend publication in its current form, but I look forward to the author’s responses in the hope that this can be published in the near future.”

Both primary concerns are accepted. The Arctic dataset was biased high, and the published values were the counting quantum rather than concentrations. The correction-factor framework is withdrawn in full. We would welcome comment on the reversal of the Sect. 2.5.1 operation-level rejection, on the warm-regime behaviour, and on the unresolved questions listed in **AC1_Add.pdf**.

Where: The manuscript as a whole; the two primary concerns are answered in A1–A6, B1 and C1–C4.

Open items bearing on these answers

1. The route by which the published Jeju 0.134 L^{-1} arose is still not established. This is the one part of A5 we cannot close.
2. Whether ship-contamination criterion (3) was applied to the published Table 2 cannot be determined from the archive.
3. The 991 published ARA16A runs cannot be reproduced from the archive.