Author Response to RC #2

Answers to the comments are written in blue.

General comment:

The manuscript describes the PIA software for processing raw PINE chamber data into quality-controlled INP concentrations. The authors present the software architecture, an automated ice-threshold identification algorithm, and a set of quality control tests. The ice-threshold algorithm, the multi-instrument temperature sensor analysis, and the large-aerosol contamination characterisation are original contributions that deserve publication. However, the manuscript requires major revision before the paper can describe PIA as providing a rigorous processing workflow.

I suspect that the INP concentration calculation contains a systematic, unacknowledged bias that likely dominates all other stated uncertainties. As I understand, the entire ice crystal count from each expansion is divided by the total volume pumped through the OPC over the full ~40 second expansion window, yet most nucleation events occur near the reported lowest temperature, which is reached only in the final seconds of the expansion. The volume of air experiencing the minimal temperature is a fraction of the used denominator, leading to a systematic underprediction of the INP concentration. The stated 10% OPC uncertainty is explicitly an assumption rather than a calibration result, and accounting for the volume bias described above, for temperature uncertainty, for large aerosol contamination, or for ice-threshold uncertainty must be included in an uncertainty budget. As the software that produces INP concentrations, the PIA paper must present a self-contained and complete uncertainty budget analysis. Further, large sections of the manuscript reproduce content already available in Möhler et al. (2021) and on pia.readthedocs.io without adding scientific justification, and the paper's relationship to the GloPINE dataset (Herbert et al., 2026), which was produced using PIA, is never stated.

We would like to thank the referee for the detailed comments and will address the points raised in the general comment in the specific comments. However, we would like to point out that many of the points raised regarding the operation and the analysis of the PINE instrument are outside the scope of this work. This manuscript is intended to describe the PIA software and its functionalities. The method by which INP concentrations are retrieved from the PINE data is beyond the scope of this work.

Specific comments:

Introduction and Sec. 4.1: Add a statement linking the paper to the GloPINE article (Herbert et al., 2026) and how the PIA software is involved in the processing chain contributing to this dataset. Table 1 of the GloPINE paper lists the PIA software version used for different campaigns. Please explain differences in the previous PIA versions and version 3 and whether the quality-control remains identical or if not, how it affects the reported INP concentrations.

It was not possible to reference the GloPINE paper by Herbert et al. (2026) as it had not been submitted when this manuscript was submitted. We added a citation in the introduction listing the multiple field campaigns conducted with PINE. However, it is not relevant for this work how PIA was used for the GloPINE paper. If necessary, differences in the analysis should be discussed in the GloPINE paper itself.

Section 2.1 and Figure 1: Condense the instrument description to the essential operating principle and key specifications needed to establish physical context for the software and refer for all instrument detail to Möhler et al. (2021). Currently the text follows the same structure and uses nearly identical phrasing as Möhler et al. (2021). Remove any verbatim phrasing. Figure 1 b), c) could be replaced with an illustration of a typical run like the Figure 3 in Möhler et al. (2021) to visualize the chambers working principle.

Thank you for this input. Möhler et al. (2021) described the prototype PINE (PINE-1A) and the first-of-series PINE (PINE-c), and provided a detailed characterization of the setup and working principle. Here, we focus on the current version of PINE that is manufactured by Bilfinger Nuclear & Energy Transition GmbH, which we make clearer now:

„This section describes the setup and the working principles of the currently manufactured PINE instrument ~~as manufactured~~ by the company Bilfinger Nuclear & Energy Transition GmbH (Würzburg, Germany).“

The description of the PINE setup in Section 2.1 follows a similar structure to that in Möhler et al. (2021), as this is, in our opinion, the most straightforward way to explain the working principle of the instrument. This is common practice and we do not consider it to be plagiarism. Here too, the description is much shorter than in Möhler et al. (2021), and we focus on the information necessary for the reader to understand the general working principle of the instrument, the data generated, the general software analysis, and the quality control.

To avoid similar phrasings, we made the following changes:

„This section describes the setup and the working principles of the currently manufactured PINE instrument ~~as manufactured~~ by the company Bilfinger Nuclear & Energy Transition GmbH (Würzburg, Germany). For a more detailed description see Möhler et al. (2021). The abbreviations for the instrument sensors used by the control and the analysis software are shown in brackets. The PINE instrument ~~consists~~ is composed of several main components, including an inlet system, a temperature-controlled cloud chamber ~~with a cooling system~~, an optical particle counter (OPC) ~~a particle detection system,~~ and a control system. Upon entering PINE via the aerosol inlet, the sampled air is guided through two parallel Nafion™ membrane diffusion dryers (Perma Pure, MD-700-24S-1; Fig. 1(a)). By controlling the water vapour pressure gradient across the membrane, the humidity of the sampled air is reduced to the required measurement conditions. The humidity of the sampled air is ~~measured~~ monitored with a dew point sensor (Vaisala, DRYCAP® DMT143) ~~at ambient temperature in front of the chamber~~ located downstream of the dryers and is expressed as dew point temperature (DP). Although the sensor reports the dew point above 0°C and the frost point below, this study will always use the term dew point temperature or DP for simplicity. For proper operation, the ~~The~~ DP has to exceed the chamber temperature ~~of the sampled air needs to be higher than the temperature inside the chamber~~ during expansion ~~in order to allow~~ to enable the formation of cloud droplets, while still being sufficiently low to prevent increased ~~, but at the same time, DP needs to be low enough to avoid~~ frost formation that eventually can cause an ice background in the chamber. The conditioned sampled air enters the chamber via the main valve. The cloud chamber has a volume of 10 L and is located within a vacuum chamber and cooled by a Stirling system. Five thermocouples (Thermoelement Typ K IKT 10/070 D2.5x15/2KK06/09 IEC 583-3 class 1, ~~unknown manufacturer~~ ES Electronic Sensor GmbH) located inside the cloud chamber (Ti1 – Ti5) measure the gas temperature, and three pt-100 temperature sensors (Pt100 RS 1/5 class B, Sensorshop 24; Tw1 – Tw3) measure the temperature of the chamber wall. A pressure

sensor attached to the inlet tubes upstream of the cloud chamber monitors the chamber pressure (Pch). ~~An optical particle counter~~ The OPC ~~(OPC;~~ (fidas-pine, Palas GmbH) ~~located directly below the cloud chamber~~ positioned directly below the cloud chamber determines the number and size of the particles within the sample flow.

Operation of the PINE instrument follows a cyclic sequence consisting ~~is operated in cycles~~ of flushing the aerosol sample through the chamber (flush mode; Fig. 1(b)), expanding the air to create cloud droplets and ice crystals (expansion mode; Fig. 1(c)), and refilling the chamber with filtered and dried air to balance the pressure to ambient conditions (refill mode). During the flush mode, the dried sample air is guided through the cooled cloud chamber at a constant mass flow rate ~~in order~~ to exchange the air inside the chamber. The photomultiplier (PM) voltage of the OPC is ~~tuned~~ configured to detect relatively large particles such as ~~to the size range of~~ large droplets and ice crystals. Therefore, it cannot detect smaller aerosol particles. The expansion mode is started by closing the main valve upstream of the cloud chamber and the~~. The~~ expansion flow (Fe) is ~~set~~ regulated to a constant volume flow rate~~, which is regulated~~ by a mass flow controller (MFC; ELFLOW Select F-201CV 10 l, Bronkhorst Instruments GmbH; reference conditions are $T = 273.15$ K and $p = 101\,325$ Pa) according to the current temperature and pressure conditions inside the chamber while the air is expanding. The INP concentration is therefore reported in number per standard litre (stdL-1). The cooling resulting from the decrease in pressure induces an increase in relative humidity ~~As soon as the saturation ratio in regard to water is exceeded~~ that exceeds water saturation, allowing cloud droplets and ice crystals to form in the presence of cloud condensation nuclei and INPs, respectively. As aspherical ice crystals are optically larger than spherical cloud ~~droplets for the OPC used in PINE~~ when measured with the OPC in PINE, ice crystals can later be identified on the basis of their optical size (see Sect. 4.2). During expansion, the wall temperatures remain approximately constant and therefore cause an increasing heat flux from the wall to the gas. Due to this heat source, which leads to the evaporation of the cloud droplets inside the chamber, the cloud formation and ice crystal detection time is limited to approximately 40 seconds, depending on the expansion flow rate. ~~The~~ To maintain a stable inlet flow ~~along the inlet system is kept constant~~ during the expansion ~~by an additional~~ a bypass flow is applied (see 1(c)). In the refill mode, ambient pressure conditions are established by refilling the chamber with filtered and dried air. One cycle of these three modes is called a run and takes about 5 to 6 minutes, depending on the duration of the flush, the expansion and refill flow, the minimum pressure, as well as ambient conditions.”

An additional Figure has been added to Section 2.1 showing a cycle of flush, expansion and refill.

Line 59ff.: Clarify the relevance of the ambient temperature for the measurement of the dew point temperature. Does the Vaisala sensor report dew point or frost point below 273K? Quantify what dew point is required by the measurement and considered low enough. Below the frost point? On line 411 it is implied that frost formation on the chamber walls is not a severe problem, even helpful to achieve supersaturation.

The wording here is imprecise. The sensor reports the dew point temperature above 273.15 K and the frost point temperature below it. This has been adapted in this section, with the reference to ambient temperature removed (see answer before).

While frost formation on the chamber walls does not necessarily have a negative impact on the measurements, it can lead to ice particles falling down from the walls and causing inaccurate measurements. Too high humidity can also lead to an icing of the chamber inlet. To identify such erroneous measurements different quality tests are implemented: Particles during refill, large aerosol particles during flush mode, and icing of chamber inlet. It is the responsibility of the PINE operator to find the proper range of the dew point temperature, which cannot generally be quantified.

Line 70, and 86: Figure 2 c) in Möhler et al. (2021) indicates that the refill air is usually not filtered. Please clarify.

The refill air is filtered for all PINE versions being analysed with the PIA software.

Line 73: Provide a reference explaining the OPC tuning protocol.

This lies within the responsibilities of the PINE operator and is not part of the PIA software. The boundary conditions for setting the photomultiplier voltage of the OPC are described here, so that the ice threshold finder can function efficiently.

Line 87-90: The INP concentration is computed as the number of ice crystal counts divided by the total volume pumped through the OPC during the entire expansion. The implicit assumption, that all air volume in the denominator was at the minimum temperature, is physically incorrect. The chamber cools adiabatically by 4-8 K over the expansion. Most ice crystals form near the end, as can be seen for example in Figure 3 in Möhler et al. (2021), when only a fraction of the total air volume was at or near minimum temperature. For a 40 second expansion with most nucleation in the final ~10 seconds, the volume of air close to the minimum temperature is less than 25% of the total. The denominator is therefore roughly 4 times too large, systematically underpredicting INP concentration at the assigned temperature by the same factor. The authors should acknowledge that the expansion method gives an integral INP concentration activating from the start to the end temperature and supersaturation of the expansion. If they want to report a single activation temperature, this bias from the expansion parameters (flow rate, total expansion duration, and the estimated duration of the cold tail) must be quantified. It could be assessed whether it is feasible using the high-resolution chamber condition records and OPC particle timestamps to segment the data into concentrations at different temperature intervals during an expansion. However, the uncertainty budget should be updated accordingly, considering either the sampled volume at the reported, minimum temperature or the change in temperature during an expansion.

As mentioned before, this work does not cover the analysis method of PINE. However, laboratory comparisons with the AIDA cloud chamber (Möhler et al., 2021) as well as other INP instruments (Vogel, 2022) and field campaigns (e.g., Lacher et al., 2024) demonstrate a high level of agreement with alternative measurement techniques. These results confirm that the established PINE analysis method is accurate within the uncertainty range and does not underestimate the INP concentration by a factor of 4.

Line 92-95: The entire uncertainty budget is outsourced to Böhmländer et al. (2025), which itself is incomplete and states that the 10% OPC uncertainty is a conservative assumption, not a calibration result. The PIA paper, as the software producing INP concentrations, must provide a complete, self-contained uncertainty budget, combining the volumetric bias described above, OPC uncertainty with proper justification, Poisson counting statistics, temperature labelling uncertainty (see comment on Section 5.1 below), ice-threshold uncertainty (Section 4.2.3), and large-aerosol contamination (Section 5.3). It is likely that these uncertainties together far exceed the stated 10% from the OPC, meaning the current stated uncertainty is not the dominant term. The uncertainty budget in Böhmländer et al. (2025), which omits several of these terms entirely, should be updated accordingly.

The PIA software does not currently calculate or provide any uncertainties. Therefore, this paper is not the place to introduce any further uncertainties. As Böhmländer et al. (2025) and this work state, for low INP concentrations, Poisson counting statistics become more relevant. The data producer is expected to consider this when interpreting the results. Regarding large-aerosol contamination, it is stated in Section 5.3, that it is unclear whether large aerosols influence the INP concentration at all, but they should be considered in the uncertainties if they exceed the statistical uncertainties.

Section 4.2.3: The validation of the ice-threshold algorithm all uses the fidas-pine OPC, with no sensitivity analysis converting threshold uncertainty to INP concentration uncertainty. Discuss the physical reasons why the threshold varies between repetitions in the same environment as shown in Figure 4. Böhmländer et al. (2025) describe a manual fallback procedure, whose frequency and magnitude are not discussed in the PIA paper. The validation should be extended to include welas OPC data, a test with synthetic data where the true threshold is known, a quantification of INP concentration sensitivity to a up to 5 bin threshold shift, statistics on the frequency of manual fallback across campaigns, and an extended discussion of conditions where the algorithm may fail (e.g., very low ice counts, high background, large aerosol presence).

The PINE mostly measures at atmospheric conditions, as can be seen in the example in Figure 4 (now Figure 5). Naturally, no two samples will be identical. This results in the differences observed in the particle size distribution shown in Figure 4 (now Figure 5). As the ice threshold is determined individually for each run and sample, the ice threshold finder detects different minima in the size distributions. As described in step 4 of the ice threshold finder algorithm, the ice threshold is set to the first bin with a non-negative second gradient after the minimum. This means that the ice threshold is set below the smallest detected ice particle. Therefore, the ice threshold can vary over a few bins above the liquid cloud without influencing the INP concentration.

The manual fallback procedure described in Böhmländer et al. (2025) is not part of the PIA software, but was part of the manual revision of the data done by Böhmländer et al. (2025). The software used for the analysis was PIA v2.0.2, which did not include the test for variability of the ice threshold, yet. This test now enables users to manually inspect and correct suspicious ice thresholds if necessary.

As Figure 4 (now Figure 5) shows, changes in atmospheric conditions and the aerosol population lead to changes in the ice threshold and how well it is determined. Using synthetic data for validation would not provide any useful information about how the ice threshold finder works with real observational data.

For low INP concentration, close to the detection limit, the uncertainty is governed by Poisson statistics, which encompass the uncertainty introduced by the ice threshold finder. For higher INP concentrations, a variable ice threshold will impact the INP concentration less in relative terms.

Figure 5: The last sentence in the caption mentions that zero concentrations are displayed as a ratio of one. Where on the x-axis are these data?

Thank you for pointing this out. Due to the logarithmic scale, we overlooked the values with a concentration of 0 in this plot. The plot has been adjusted to display these values too.

Section 4.3 and Table A2: The QC thresholds are stated to derive from empirical observations but are not scientifically justified. Test conditions could be included in Table A2. For each threshold the paper should state its physical basis, the fraction of expansions it flags, and provide a sensitivity analysis of mean INP concentration if a threshold is tightened or relaxed by 10–20 %. This will give confidence that the QC choices are not arbitrary. The bulk of this section and Table A2 reproduce content already on pia.readthedocs.io. The manuscript should present the scientific justification for the QC design, not a re-statement of the implementation.

The Section on quality control has been restructured to avoid duplicating straightforward tests such as range tests. Table A2 has been expanded to include the test parameters and information on whether the test was performed on a single data point or the entire run. Tests that can be sufficiently described by the table have been removed from Section 4.3.

The test conditions are either based on instrument specifications, have been defined after analysis of many datasets and haven been adapted over time in an iterative process to find stable settings, or are based on established statistical methods such as standard deviation and outlier detection algorithms. Lines 323f have been adapted to clarify this:

“Unless stated otherwise, the test conditions are based on empirical observations and have been adapted in an iterative process to get stable test results or are based on instrument specifications.”

Line 363: Clarify if the pump is turned off and not just the flow redirected.

Thank you for pointing this out, the pump is indeed not turned off during refill, but the MFC between OPC and pump is regulated to zero and the flow redirected. This was corrected in Line 340f:

“The first two time bins of the refill mode are ignored, as it is possible that there are still particles in the line after the flow through the OPC is turned off.”

Line 385: Are ice crystal counts corrected for the large aerosol BG measured during flushing?

No, as stated in Section 5.3 the large aerosols should be considered in the uncertainties. A correction for the large aerosols during flush mode would possibly lead to a low bias of the INP concentration, as these larger particles will exhibit a smaller optical diameter, when activated as CCN.

Line 403: Clarify why -35°C is essential for the range test.

This has been clarified by the additional statement (now Lines 363ff):

“Below -35 °C PINEair can measure homogeneous freezing by conducting a rapid expansion. In this case 9 sec are enough time to detect the ice crystals with the OPC. For heterogeneous freezing it takes a longer time for the ice crystals to form and to be guided through the OPC.”

Line 407ff: Specify if supersaturation refers to ice or water, and DP to dew point or frost point.

As written in Section 2.1 supersaturation refers to water. DP refers to the value reported by the dew point sensor, which can be dew point or frost point depending on the temperature range. This is clarified in this section as follows (now Lines 366f):

"If water supersaturated conditions are not given inside the chamber, a liquid cloud cannot form and the ice threshold finder would not function efficiently."

Equation 4: Dubble check the units of the equation. It should be seconds, but in its current form the right-hand side units cancel out. Also explain where the 60 s come from.

The flow is reported in L/min, to get the resulting time in seconds it needs to be divided by 60. Therefore the "s" must be removed from the formula and the 60 moved behind the fraction. This has been corrected and specified with the following sentence:

"As the expansion flow is given in L min^{-1}, the whole term needs to be multiplied by 60 to get the result in seconds."

Section 5.1: The finding that PINE-05-02 had Ti5 mispositioned near the chamber wall causing it to report temperatures higher than Ti4 and biasing the assigned minimum temperature warm is an important result but its consequence for INP concentrations in affected measurements is not quantified. To quantify this type of error the Temperature Error Factor framework of Schrod and Bingemer (2025) could be applied. For typical ambient INP spectra Schrod and Bingemer (2025) show that even a 1 K warm bias in the assigned temperature translates to a factor of up to 5 underestimations of INP concentration at the reported temperature. The authors should estimate what the magnitude of the temperature offset was before sensor repositioning, and what the resulting concentration bias is for affected campaigns (LIFE-FROSTDEFEND in the GloPINE table 1). Please calculate the sensitivity of INP concentration to temperature uncertainty for the typical PINE operating range and report the resulting contribution to the combined uncertainty.

The results from this section show that mispositioning or malfunction of a temperature sensor can easily be detected by making this comparison. As a result of this test, the faulty sensors can be fixed and no concentration bias needs to be applied. The LIFE-FROSTDEFEND campaign that you are referring to was conducted after the temperature sensors in PINE-05-02 were placed correctly. The uncertainties are not part of the PIA software.

Figure 7: The large particle BG seems not to depend on the measured INP concentration. This could be mentioned and the y-axis changed to INP concentration flush instead of the ratio.

Yes, the large particle background concentration is only slightly dependent on the INP concentration determined during the expansions. We have – as suggested – modified the y-axis accordingly and also added the values for INP concentrations of 0. We also calculated the Pearson correlation coefficient resulting in a value of $r = 0.273$ with $p = 0$. We have adjusted the description and the text in the paper accordingly (Line 452ff):

"Figure 8 shows the concentration of ice-sized aerosols during flush as a function of the INP concentration during expansion. There is only a slight dependency (r = 0.273) between large

aerosols during flush and the INP concentration during expansion, therefore the relative influence of large aerosols becomes relevant for concentrations below 10 stdL−1."

Line 543: The manuscript describes version 3.0.0, but the Zenodo DOI cited in the code availability section (zenodo.15592883) is labelled Büttner and Fösig, 2024, while Herbert et al. (2026) and Böhmländer et al. (2025) both cite zenodo.15592431 as PIA Software (v3.1.0), Büttner and Fösig, 2025. It is unclear whether these are distinct archived releases. The manuscript could provide a brief changelog noting any algorithm changes between v2.x, v3.0.0, and the latest version, for tracking reproducibility of datasets processed under earlier versions.

The DOI cited in this manuscript belongs to the PIA version 3.0.0. Herbert et al. (2026) and Böhmländer et al. (2025) cite the concept DOI of the PIA software. This DOI always resolves to the latest PIA version. Böhmländer et al. (2025) additionally cites version 2.0.2, which was used to analyse their data.

As a software is a living product that changes over time, the aim of this paper is to describe the state of the software at version 3.0.0, which was the most current version during the writing process. Any changes between different versions can be found in the changelog accompanying the software code.

Figure A4: Indicate flush, expansion, refill. Mention that this is a temperature ramp experiment.

The Figure and the description have been adapted accordingly.

Figure A5: Clarify what causes the pressure drop at the end of refill. A run without inlet icing could be added for comparison.

The pressure drop at the end of refill is caused by the same effect as it is at the beginning of the expansion. Switching the valve causes the gas inside and in front of the chamber to equilibrate, leading to a pressure drop due to inlet icing and the resulting lower pressure inside the chamber. This was added to the caption of Figure A5:

"Example of detected inlet icing. At the start of the expansion and the end of refill a steep drop can be seen in the pressure. This is due to the reduced pressure inside the chamber resulting from the iced inlet. The pressure drop results from the equilibrating gas once the valve switches. The plot shows operation 198, runs 10 to 12 from the CORONA campaign."

Technical corrections:

Line 93: "uncertainty budged" should be "uncertainty budget."

This was fixed.

Line 175: the default Level 2 temperature bin of 0.5 K is finer than the stated ±1 K instrument temperature uncertainty and implies precision not supported by the instrument. The default should be widened to ≥1 K or a clear caveat added.

The default temperature binning of 0.5 K is a common binning size for other instruments measuring INP concentrations (e.g., INSEKT (Schneider et al., 2021)). Therefore, this binning size was chosen to allow for straightforward comparison. If a coarser binning is desired, the results can be averaged accordingly.

Line 332: the upper dew point QC range limit of +8°C should be verified. It would cause immediate frost formation, suggesting a sign error.

The range test for the dew point values is used for any technical problems with the dew point sensor and therefore spans a wider range than typically used for measurements. Any dew point issues relevant for operating the PINE will be caught by other QC tests.

The expansion flow is given as 3 L min^{-1} while the flush flow is 2–3 stdL min^{-1}. Use consistent standardised units throughout.

The mass flow controllers report the data in stdL/min, which we are directly referring to when talking about the flush flow. During expansion the mass flow is constantly adapted to ensure a constant volume flow, which has the unit L/min and is required to keep the velocity of the particles constant through the OPC.

Language that could be interpreted as commercial endorsement should be avoided. Instead of describing PINE's potential as a "key instrument" or "reference instrument", stick to neutral, factual statements.

We agree and adapted the following sentences (Lines 35ff):

"~~PINE is commercially available, and d~~Due to its automated operation and minimal requirements for user input, ~~it~~PINE ~~has a great potential to become a key instrument~~ is well suited for INP monitoring. For example, PINE is ~~a reference instrument within~~ one of the instruments used by the ACTRIS Aerosol, Clouds and Trace Gases Research Infrastructure; Laj et al., 2024) Topical Centre for Cloud In Situ Measurements (CIS)."

Competing Interests statement: The declaration of no conflict of interest seems inconsistent with acknowledging KIT Technology Transfer project N059 PINE funding, the project that produced the commercially available PINE instrument. This standard academic-commercial relationship should be declared transparently.

Thank you for pointing this out. We added the following statement:

"OM, LL, and BM were involved in a prior technology transfer project between KIT, University of Leeds and Bilfinger Nuclear & Energy Transition GmbH (Würzburg, Germany) related to the development and commercialization of the underlying PINE instrument. The software described in this work is independent of this project, open source and freely available. The authors declare no competing financial interests."

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