Author response file for manuscript amt-2020-307

Dear Editor,

Thank you very much for handling our manuscript. We meanwhile have prepared and uploaded author comments in reply to the four referee comments we received for our manuscript entitled "The portable ice nucleation experiment PINE: a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles".

Included in this file are the four comments and a the revised manuscript version with tracked changes.

With best regards,

Ottmar Möhler and co-authors

Author comment in response to the comments provided by Referee # 1

We thank referee # 1 for her/his effort in reading and commenting our manuscript. In the following, we report the referee's comments (in italics), give point-by-point answers, and suggest manuscript revisions based on the referee's comments and our answers. Respective reference will be given to the line numbers of manuscript version 1.

Referee comment: This manuscript details the design and performance of a new ice nucleation chamber. This instrument is based on an expansion principle, much like the AIDA chamber at KIT (location of several of the co-authors). In this regard the chamber is different than the continuous flow principle used on almost all current ice nucleation chambers. PINE therefore represents an important addition to the field. The design and performance is important and the use of a long term (in this case 45 days) makes this a solid paper and very appropriate for AMT. The paper is well written and only minor revisions are needed. There are a few points I'd like to ask the authors to consider: Starting in the Abstract but running through paper there are several unquantified terms : "... extensive ...", "... good ...", "... high time resolution ...". These are all subjective and need to be removed.

Answer: We scanned the manuscript for such unquantified terms and removed most of them (see lines 9, 15, 61, 297, 493) or replaced them with more quantitative statements (see lines 11, 371).

Referee comment: The Introduction, although highly comprehensive, is also very long for an instrumental paper (3 pages). It seems like it could be considerably shortened.

Answer: The third referee (Paul DeMott) also mentioned the introduction to be overly comprehensive. We agree and suggest removing or shortening the following parts:

Remove section about cirrus clouds (lines 37 to 43) which are not further subject of this paper. We had included this short paragraph in the first manuscript version because PINE is also capable of measuring INPs in the cirrus cloud temperature regime. This is subject of ongoing activities and the further development of the PINE instrument.

Reformulate lines 56 to 61: "Existing parameterizations are applied in models to calculate and predict primary ice formation in clouds, however, the atmospheric INP data that we can compare with global fields of model predicted INP concentrations are limited in spatial, ...

Remove lines 69 to 76 ("While high temperature INPs ... may dominate high temperature INPs in many regions (...)."

Modify line 77 to "Most of previous INP measurements were only sensitive to immersion freezing ...".

Modify lines 103/104 to "... in a wide temperature range. In this paper, PINE´s ability is demonstrated to measure in the mixed-phase cloud temperature regime from $-10\,^{\circ}\mathrm{C}$ to $-40\,^{\circ}\mathrm{C}$. PINE is also able to measure ice nucleation at cirrus cloud temperatures to about $-65\,^{\circ}\mathrm{C}$, which is the topic of ongoing studies."

Referee comment: The 'milestone' portion of the 2. Basic Principles section should be removed. It does not seem relevant to outline the timeline / dates (i.e. 20 years, first test 2016, etc.) since they don't impact the instrument performance. Please eliminate this part of the paper.

Answer: We believe that the experience in operating the AIDA cloud expansion chamber was indeed an important contribution to the PINE development. Referring to AIDA operation as a well-known and well-cited cloud simulation chamber also introduces the reader to the basic operating principles of PINE. Moreover, PINE was developed during relative short time, as e.g. compared to the development of continuous flow diffusion chambers. Thus, we believe that it is interesting to the reader to be informed about the development steps. Therefore, we like to keep this part, but have made the following modifications:

Change first sentence of section 2 (lines 111/112) to "The idea for PINE resulted from the experience in operating the AIDA facility for cloud experiments at simulated conditions of up-drafting atmospheric air parcels."

Modify the last two sentences of first paragraph of this section (lines 122 to 124) to: "Large aerosol particles, droplets and ice crystals are measured and counted with an optical particle counter (OPC). Placing the OPC in the vertically oriented pump tube below the cloud chamber was one of the critical development ideas for PINE (see patent applications DE 10 2018 210 643 A1 and US2020/0003671 A1). PINE can be operated both for ice nucleation research in the laboratory, and for INP measurements in field campaigns or long term monitoring activities.

Remove the sentence "This setup was operated in a cold room ... attached to the pump line (see patent applications ...)" (lines 127 to 130).

Referee comment: The dates of the SGP test (Oct 1 -) is found in Section 2 and then repeated 4 times in the paper; please state once.

Answer: Done as suggested (see lines 414, 466).

Referee comment: During HyICE, there are repeated references to CCN activation. Just as PINE is compared to AIDA, wasn't there a CCNC at HyICE? If so can the PINE droplet data be compared to those data? The topic of drop formation could be more fully developed in the paper and this would help.

Answer: We were using the term CCN activation because aerosol particles sampled into the PINE chamber first act as cloud condensation nuclei to form supercooled droplets which then eventually freeze when they include an ice-active aerosol particle (INP) at the given temperature. In other words, the same particle acts as CCN and INP. However, CCN activation happens in a fast cloud expansion process without independent control of the relative humidity or supersaturation. Therefore, an expansion cloud chamber like PINE or AIDA cannot control or quantify the CCN process as a function of relative humidity, just is then filled with the droplets resulting from the CCN activation and diffusional droplet growth processes, but without quantifying the parameters controlling those processes. The size distribution of the resulting droplet cloud can then well be measured with the OPC and compared for subsequent runs, as demonstrated by first field measurements with the PINE-1A prototype instrument during the HyICE field campaign. To clarify this, we suggest the following changes to the manuscript line 296: "This means that PINE is able to reproduce the formation of the supercooled droplet cloud in repeated runs at constant sampling and operation conditions, . . . "

Referee comment: Figures Please check f ice and others not in subscript ;

Answer: checked and corrected

Referee comment: Figure 8 : Does "in preparation of the HyICE field campaign" have an impact on the measurement? It seems highly extraneous.

Answer: Yes, agreed and removed.

Referee comment: Figure 11 : Is 'aerosol, right after the PINE-1A runs were finished.' the relevant point? Is 'using the same aerosol' correct?

Answer: Yes, correct, both chambers are using the same aerosol. Therefore, no modification needed here.

Referee comment: Figure 12 : Does not seem necessary to attribute funding to DOE here since this is typically done in the acknowledgements. Site location seems sufficient. Inset legend seems to mean '6 hour averaged data' (not daily)? And '45 day average'

Answer: Reference to DOE funding will be removed. The figure indeed shows temperature binned data for both 6 hour averaged data and average over all 45 days. We will change the legend and rephrase the figure caption as follows: "Temperature-binned concentrations data ($\Delta T = 1\,^{\circ}\mathrm{C}$) is shown for 6 hour time averaged data (black markers) and 45 days averaged data (green markers)."

For consistency with the updated Figure 12, we also suggest modifying lines 15/16 of the abstract as follows: "...with continuous temperature scans for INP measurements between $-10\,^{\circ}\mathrm{C}$ and $-30\,^{\circ}\mathrm{C}$."

Referee comment: Figure A1 : 'setup' can be removed, it is redundant after 'Schematic'

Answer: done

Referee comment: Figure A5 : Figure text appears to be of low quality and needs to be increased in resolution.

Answer: Figure with higher quality will be included in the revised version of the manuscript.

Referee comment: Figure A6 : 'foto' should be 'photograph'. Panel (b) appears redundant and can be removed.

Answer: "foto" changed to "photograph". Because the left shows a photograph of PINE-c at the SGP field site, and the right the 3-D construction of the commercial PINE version as a new instrument, we would like to keep the figure as is, but suggest to change the caption as follows: "Photograph of PINE-c (a) located at the ARM-SGP site in Oklahoma for 45 days of continuous INP measurements from October 1st to November 14th, 2019. Part (b) on the right shows a composite photograph of the same instrument before delivery."

Author comment in response to the comments provided by Referee #2

We thank referee #2 for her/his effort in reading and commenting our manuscript. In the following, we repeat the referee's comments (italics), give point-by-point answers, and suggest manuscript revisions based on the referee's comments and our answers. Respective reference will be given to the line numbers of manuscript version 1.

Referee comment: *A new laboratory instrument for INP measurement called "portable Ince Nucleation Experiment" (PINE) chamber is introduced in this manuscript. The design, working principles, and operational procedures of the PINE chamber are described in details, as well as preliminary results from the HyICE campaign, AIDA intercomparison and SGP-ExINP long-term measurements are provided as work cases in the paper. The development of the PINE chamber is a great contribution to the INP research field in specific and the Atmospheric science in general. It also provides long-term monitoring capability to operation-oriented organizations. The topic fits AMT scope perfectly. The paper is well organized and written. After the authors address my minor points, it should be in good shape for publication on AMT.*

Minor comments: *The font on many figures is too small.*

Answer: This was also mentioned by Referee #1. We will re-plot the figures with larger fonts.

Referee comment: *Line 175: remove "and"*

Answer: done

Referee comment: *Line 265: What is the aerosol size range for these concentrations?*

Answer: The majority of particles was smaller than $500\,\mathrm{nm}$ in diameter. Only a minor fraction ($< 0.1\,\mathrm{cm}^{-3}$) had diameters between $0.5\,\mu\mathrm{m}$ and $5\,\mu\mathrm{m}$. We will add the following to line 264: "..., with the majority of particles smaller than $0.5\,\mu\mathrm{m}$ in diameter, ..."

Referee comment: *Lines 271 to 275: Is the assumption of ice saturated condition at the beginning of expansion reasonable? The response of the OPC does agree with this assumption. But is it universally valid?*

Answer: Yes, we think this assumption is reasonable because the frost point temperature of the air sampled into the chamber was higher than the average wall temperature. The excess water vapor quickly condenses to the cold chamber walls, so that ice saturated conditions of the air inside the cloud chamber are reached. We re-phrase line 273 for including this information.

Referee comment: *Line 287: "larger than the dense"*

Answer: changed

Referee comment: *Figure 6: Can a turbulence be introduced to the chamber to mix the air so that the temperature is more uniform across the chamber?*

Answer: This is a good idea and suggestion we have already discussed among the PINE team members. Of course, such fan driven mixing is an important part for operating the large AIDA cloud chamber at homogeneous gas temperature conditions. Until now, we did neither test this option for the small $10\,\mathrm{L}$ PINE chamber nor did we discuss the technical solution. But it is an option for further developing and improving the PINE chamber.

Referee comment: *Line 446: replace "largest" with "highest".*

Answer: done

Referee comment: *Line 468: Based on Fig. 12, the minimum INP concentration is about 0.02 L-1, not 0.2 L-1.*

Answer: The referee is right, it will be corrected. Thanks for noting this.

Referee comment: *There are multiple places stating that details on HyICE results and SGP-ExINP results will be discussed in details in future papers. Can reduce the redundancy.*

Answer: Yes, we agree. We removed respective statements for the HyICE campaign because not of relevance here (e.g. line 377, 474), but kept it for the DOE SGP campaign section. We suggest reformulating the sentence in line 320 to "Ongoing activities for improving the operation and data analysis tools for PINE also focus on developing an automated procedure for setting this threshold."

Author comment in response to the comments provided by Referee #3

We thank Paul DeMott (referee #3) for his effort in reading and commenting our manuscript. In the following, we repeat the referee's comments (italics), give point-by-point answers, and report manuscript revisions based on the referee's comments and our answers. Respective reference will be given to the line numbers of manuscript version 1.

Referee comment: General Comments: *As a scientist in this research area who both continues strong collaborations with some of the group represented on this paper and has promoted use of continuous flow diffusion chambers (CFDCs) for ice nucleation measurements over many years, I figure it is important to self-identify in this review. In this manuscript, the PINE instruments are introduced, appearing to represent a great new tool for the community, and with promise for meeting certain INP monitoring and experimental study needs. The new results for unhindered immersion freezing are very encouraging, admirable in being achieved over a relatively short period of development. From the standpoint of a publication documenting a new method, there were a few things missing for me as a very interested reader. Hence, I list a number of specific comments/questions below, driven by my desire to understand the instrument clearly. In short summary, 1) there was not a full description of principles and device(s) in order to understand what challenges may be met in applying the method for the range of ice nucleation studies inferred to be possible (deposition and immersion freezing to* $-60\,^{\circ}\mathrm{C}$ *(i.e., only immersion freezing is thus far discussed to the point of homogeneous freezing conditions); 2) uncertainties were given relatively limited discussion (especially at the limit of detection); 3) there was no discussion of consistency of results with physical expectations that might be revealed from, for example, microphysical modeling considerations; 4) relatedly there seemed more cursory consideration given to defining the relevant temperature associated with a measurement (I did follow the arguments, although the confirmation was mostly by comparing to AIDA), the role of growth time and sedimentation if any, clear separation of water and ice given that the latter occurs usually a few orders of magnitude lower than the water drop concentrations; and 5) finally, the introduction of field data and field instrument was rather abbreviated considering the nature/nuances of that application and considerations that will impact operation across the stated T and supersaturation range of the device in the presence of varying atmospheric conditions and full aerosol distributions. The field data only serve the purpose of demonstrating a range of data collected during automated operation for a period, as there is no other discussion of the data provided. I expect that some of the lack of clarity that I sensed will be resolved in review here. I understand, of course, that full information on any new device is often revealed over some time, often in a number of separate publications. This is clearly underway as indicated by a paper in preparation (and other intercomparison studies I am aware of), but it suggests then that some of the statements herein may require a few caveats because supporting data are not fully shown. Hence, I might even suggest consideration of a title change to include something like "An introduction . . . " or "A first evaluation . . . " or "First description and results from . . . " or something to that effect. That is not an adamant request, simply a suggestion. The paper is otherwise well-written and an anticipated and welcome addition to the literature.*

Answer to General Comments:

We appreciate the comprehensive and meaningful comments from Paul DeMott, which we think not only contribute to improve the current manuscript, but are also very valuable and helpful for the further development of the new PINE instrument. We hope that our answers as well as the related revision of the manuscript will result in a sufficiently comprehensive first description of this new instrument which is clearly understandable to the interested reader.

We would also like to mention here that we purposely refrained from including everything we possibly could in this first paper, and instead address key aspects such as the operation in the field in separate papers where we can go into more detail. This first paper is intended to introduce this new instrument and to explain how it works, what it measures and how accurate the measurements are. The further development of the instrument itself and the data analysis systems are subject of ongoing activities, as the referee also noted, and will be discussed in more depth in upcoming publications, including more thorough uncertainty analysis and quality assurance measures.

Concerning general comment (1): Meanwhile, several measurements and test runs with PINE have been conducted in the cirrus cloud temperature regime. Thus, we indeed know that PINE is capable of measuring INPs of relevance for cirrus cloud formation, but we admit that this is not the subject in this paper (our initial focus is very much on mixed-phase clouds), and we will therefore modify the manuscript accordingly (see answers to specific comments below).

Concerning general comment (3): We think the referee is mainly referring here to the proof-of-concept runs of PINE for homogeneous freezing of poor water droplets and the comparison to AIDA results. We agree that freezing of water droplets

around $-35\,^{\circ}\mathrm{C}$ can also be expected from freezing rates derived from classical nucleation theory or reported in the literature. We therefore suggest to re-phrase lines 365-368 as follows:

"... and as such allowed the intercomparison of temperature-dependent freezing rates or INP concentrations. Homogeneous freezing of supercooled water droplets is known from classical nucleation theory and from literature results (Pruppacher and Klett, 2010; Koop and Murray, 2016) to occur at temperatures between about $-35\,^{\circ}\mathrm{C}$ and $-37\,^{\circ}\mathrm{C}$. Figure 7 shows the freezing of water droplets to be measured with PINE-1A in the expected temperature range. As in the experiments ...".

Setting up a microphysical model of the processes occurring within the chamber would be an interesting exercise and may become necessary in the future. However, for the study of INP relevant for mixed-phase clouds, our focus at the moment, we do not need a detailed microphysical model. The fact we create a liquid cloud defines saturation, we measure temperature directly and ice crystals are readily detected. Hence, we do not need a model to access any of the pertinent parameters. With or without a model we would place a great deal of emphasis on the comparison with AIDA, hence we focus on this comparison rather than theory.

Concerning general comment (2, 4): The referee's comments on the limited discussion of uncertainties and the comparison of PINE with AIDA refer to the more general question, whether it is possible to accurately and completely identify and specify all uncertainties of an instrument like PINE or a CFDC (Continuous Flow Diffusion Chamber), or whether a calibration to some standard or a direct comparison to a reference instrument is needed. We think the known sources of uncertainty of PINE are well mentioned and discussed in the manuscript. Up to this stage of development, we used the AIDA cloud chamber as a reference, and we think this is justified from previous intercomparison activities and results. This is our natural first order approach for an uncertainty estimate of PINE INP measurements, further systematic experiments and test are needed to quantify specific systematic uncertainties related e.g. to sampling efficiencies for aerosol and ice particles or to ice growth and size range issues. Concerning the nucleation temperature discussion, we agree that our argumentation is somehow "cursory", or to say it in other words, PINE is not capable of controlling the ice nucleation temperature as accurate as e.g. a CFDC is. This is one of the limitations of PINE, but when measuring immersion INP concentrations over a wide temperature range from about $-10\,^{\circ}\mathrm{C}$ to $-35\,^{\circ}\mathrm{C}$, an overall temperature uncertainty of $\pm 1\,^{\circ}\mathrm{C}$ according to current conservative estimates may be sufficient to quantify the temperature spectrum of INP concentrations.

Another important parameter for long-term observations, the PINE instrument is also developed for, is the precision for repeated measurements at the same sampling and operating conditions. Figure 1 shows a recent measurement with the new commercial PINE-04-01 when sampling a mixed aerosol (ammonium sulfate and natural dust) for more than 8 hours from the AIDA chamber. This figure well demonstrates the run-by-run stability and repeatability of PINE measurements. In this experiment we did not expect a constant but a steadily decreasing INP concentration (panel a), according to the steady decrease of the aerosol concentration (panel b) according to aerosol loss processes to the chamber walls. The ice-active particle number fraction (panel c) remained constant with a mean value of 1.8×10^{-4} and a standard deviation of 2.1×10^{-5}, or a relative uncertainty of about 12%, which demonstrates the precision of PINE INP measurements under these conditions. During this operation, an average number N_{ice} of about 90 ice crystals was measured during one run. Therefore, the relative uncertainty from counting statistics can be calculated as $\sqrt{N_{ice}}/N_{ice} = 10.5\%$, which is close to the observed standard deviation.

We will include this figure and the respective text to the appendix C (new Figure A6). We also suggest to modify and extend the last sentence of section 5 (lines 407 to 410):

"This also underlines the assumption, that the ice formation in PINE is mainly controlled by the coldest temperature in the bottom part of the chamber and that the number concentration of ice crystals, and by that the number concentration of ice-active aerosol particles in laboratory experiments and of INPs during field measurements can be calculated with Eqs. 5 and 6 within the above given uncertainty estimates for the number concentration and the nucleation temperature. These estimates are justified by the comparison of PINE with AIDA results. Further systematic uncertainties like the loss of large ice crystals between the PINE cloud chamber and OPC, size range overlap of small ice crystals with large aerosol particles not activated to droplets, or the sampling efficiency of large aerosol particles into the cloud chamber may have to be considered for calculating the overall accuracy of INP measurements.

A more comprehensive uncertainty assessment for PINE may result from recent intercomparison studies with other methods and instruments and ongoing long-term operation in field campaigns. For long-term measurements, another important parameter is the precision for repeated measurements at the same sampling and operating conditions. In a recent test experiment at the AIDA cloud chamber, the new commercial PINE-04-01instrument sampled a mixed aerosol (ammonium sulfate and natural

dust for more than 8 hours from the AIDA chamber (Figure A6). During this experiment, a mean ice-active particle number fraction of 1.8×10^{-4} was measured with a standard deviation of 2.1×10^{-5}, which corresponds to a relative uncertainty of about 12%. During this operation, an average number Nice of about 90 ice crystals was measured during one run. Therefore, the relative uncertainty from counting statistics can be calculated as $\sqrt{N_{ice}}/N_{ice} = 10.5\%$, which is close to the relative standard deviation of the run by run data from the mean value. For measurements with a much lower number of ice crystals detected in one run or a consecutive number of runs, the measurements uncertainty from counting statistics can be much larger. Next versions of the PINE analysis software tools will also include uncertainty analysis for low counting cases close to the PINE detection limit."

We also noted that there is an error in line 401. The temperature uncertainty during an AIDA cloud expansion chamber is noted as $\pm 1\,^{\circ}\mathrm{C}$. This should be corrected to $\pm 0.3\,^{\circ}\mathrm{C}$.

Concerning general comment (5): The description of the field instrument was brief because it's operating principles are identical. Also the discussion of the data was brief because we are planning a succession of papers focused on the field data (as the referee notes).

Concerning the title: We feel our existing title is accurate, and the phrase "a new" implies that this is the first description.

Further suggested changes to the manuscript will be included below along with our answers to the specific referee comments.

Specific Comments

Introduction:

Referee comment:

1) The introduction was comprehensive, perhaps overly so for a paper describing a new instrument. It was long, and not so much related to the development itself other than attempting to meet motivations.

Answer:

Referee 1 had a similar comment. See answers given there. Further revision is mentioned below in response to more specific comments.

Referee comment:

2) Lines 87-89: It seems clear that both low and high time resolution are desirable things for different scenarios. High time resolution is arguably not useful if one is attempting to document the most special INPs, the ones that even the PINE instrument may struggle to measure in all but INP-rich environments such as shown later in this paper. I see higher time resolution measurements as highly useful, but not sufficient, unless their resolution can match higher volume collections. Some of the studies referenced to preface this statement were made with instruments capable of even higher time resolution than the PINE, but the issue I am speaking of is resolving low INP concentrations in some environments. Those other methods have been developed for automation as well, a point I will raise next.

Answer:

We agree the important point are missing here, and suggest to modify lines 87-89 as follows:

"Depending on the specific campaign goals and objectives, different instruments and methods were used like CFDCs with higher time resolution to e.g. characterize changing air masses (e.g. Boose et al., 2016a; Lacher et al., 2018), or aerosol filter based offline methods to achieve high sensitivity for characterizing INPs at higher temperatures or in clean environments (e.g. Wex et al., 2019), or a combination of both (e.g. Welti et al., 2018). What is missing so far are long-term monitoring of INPs with high time resolution and over a wide temperature range."

Referee comment:

3) Lines 97-100: I would say to be fair that these statements need modification or qualification for other studies in the recent literature. I think that the continuous flow chamber developments reflected in Bi et al. (2019) and Brunner et al. (2020) meet the criteria of operating more than "periodically" and of saving "intensive man-power and time for operation or offline analysis." Such developments are advancing rapidly across the discipline. Those instruments also appear to be capable of higher time resolution than is demonstrated for the PINE instrument in this paper. It is also implied here that time resolution of minutes will somehow solve the INP size and chemistry resolution issue, although due to statistics (sample volume and particle numbers) it is hard to imagine this as yet being achievable for single INPs except in high loading situations. Rather, this would occur by correlation to independent high resolution composition measurements for all aerosols, which sometimes does not work for comparing directly to specialized INPs. Hence, I see high resolution INP capabilities as one piece needed in the course of a full development.

Answer:

Thanks for this very valuable comment, and thanks for referring to Bi et al. (2019) and Brunner and Kanji (2020). In fact, we have not cited Bi et al. (2019), and this error has been rectified. Brunner and Kanji (2020) was available as a discussion paper when writing, and it will also be cited in the revised paper. We also agree that other statements in this section are somehow imbalanced and suggest to re-phrase lines 94 to 100 as follows:

"Most of the INP methods showed reasonable agreement with each other, but many of them are time and operator intensive. A general feature is, that offline methods based on aerosol filter samples have poor time resolution depending on required aerosol sampling time of hours to days, and require intensive man-power and time for both operation and offline analysis. Most online instruments can only be operated periodically, and also require operator time during the measurements, but can be operated for INP measurements at higher time resolution in particular at low temperature or in polluted environments where concentrations are higher. Only recently, newly developed INP instruments with a higher degree of automation became available (Bi et al., 2019; Brunner and Kanji, 2020). The automated CFDC instrument used by Bi et al. (2019) performed INP measurements during a period of one month in 2018 at temperatures between $-20\,^{\circ}\mathrm{C}$ and $-30\,^{\circ}\mathrm{C}$. The CFDC instrument called HINC-Auto (Horizontal Ice Nucleation Chamber) used by Brunner and Kanji (2020) autonomously measured immersion freezing INP for 90 consecutive days, but only at one temperature of $-30\,^{\circ}\mathrm{C}$. A combination of both, high time resolution and wide temperature range for long-term INP measurements, together with a comprehensive set of high resolution aerosol analytics, would challenge the comparison to potential driving factors for atmospheric ice nucleation."

Referee comment:

4) Line 103: The stated temperature range is what the instrument is designed for, but no exploration of capabilities to make useful measurements to as low as $-60\,^{\circ}\mathrm{C}$ are given in this manuscript. It appears as a potential capability, only in that the temperature can be achieved in PINE-c. One can imagine that challenges in operating and interpreting data to that lower limit could be significant (e.g., control on final T and RH of expansion, low water vapor pressure and slow ice crystal growth rates), and not simply depend on the capacity of the cooling system (line 121). I suggest to stick to what is demonstrated in this paper, as far as confirmed operational capabilities and to clearly identify capabilities that remain to be defined.

Answer:

We agree and suggest to modify lines 103/104 to "... in a wide temperature range. This paper demonstrates the instrument's ability to measure in the mixed-phase cloud temperature regime from $-10\,^{\circ}\mathrm{C}$ to $-40\,^{\circ}\mathrm{C}$. PINE is also able to measure ice nucleation at cirrus cloud temperatures to about $-65\,^{\circ}\mathrm{C}$, which is the topic of ongoing studies." (see also answer to comments from referee #1).

Basic principles and milestones of the PINE development:

Referee comment:

1) Line 121 repeats the assertion that likely requires "potential" as a caveat. No low temperature data are shown excepting the homogeneous freezing onset for grown droplets.

Answer:

We suggest to remove "and thereby the temperature range of ice formation and INP detection"

PINE instrument setup:

Referee comment:

1) Line 169 or thereafter: Have particle losses been characterized through the nafion dryer system? For that matter, I realize in reviewing these comments that particle transfer versus size into the PINE systems has not been discussed.

Answer:

We characterized the particle loss through the dryers at the NAUA aerosol chamber, using a natural dust sample from Marocco. An APS was used to measure the aerosol size distribution before and after the dryers, which were mounted in a way to represent their orientation at PINE, with vertical orientation and the sampled air flowing in upward direction, then bending by 180 degrees for straight downward flow into the PINE cloud chamber. Figure 2 shows both the aerosol particle size distribution with and without the dryers, demonstrating that the loss of particles in the size range of up to about $2\,\mu\mathrm{m}$ is minor. Only particles larger than approximately $4\,\mu\mathrm{m}$ experience a major loss of more than 50%. As such we have confidence that a large fraction of atmospherically relevant particles will enter PINE. In setups were the sample flow can be taken in strictly vertical downward orientation, the particle loss can be expected to be much smaller. More systematic experiments of this kind will be performed in the future, where the particle loss in the dryers and in the PINE chamber will be characterized.

We suggest adding the following paragraph at line 180:

"In the commercial version, the standard location of the dryers is next to the cloud chamber with vertical orientation, so that the sampled air flows in upward direction through the dyers, then passes a $90°$ bend, a horizontal distance of $50\,\mathrm{cm}$ and another $90°$ bend to then flow downward into the PINE cloud chamber. The aerosol particle loss for this setup was measured to be less than 20% for particles smaller than $2\,\mu\mathrm{m}$ diameter. It decreased to about 50% for particles with an aerodynamic diameter of about $4\,\mu\mathrm{m}$. The dryers can also be mounted above the PINE chamber for a strictly vertical sample flow, for which a further reduced particle loss can be expected. More systematic sampling efficiency measurements for different configurations and operations will be performed in the future."

Referee comment:

2) Lines 172-173: Perhaps this is irrelevant since an aircraft system is not yet described, but I wondered about the use of the nafion system on aircraft where the pressure drop will be limited at higher altitudes. Will the system work over the needed ranges in this scenario?

Answer:

We know from first estimates and test series that a dryer will not be needed when sampling dry air in the middle/upper troposphere. We may even need a humidifier instead, depending on air temperatures and relative humidity. Nevertheless, the nafion system can still be operated at reduced absolute pressure, but what makes it inefficient for application in the free troposphere is not the reduced pressure drop over the membrane (you could even think of operating the dryer with dry synthetic air) but the drying efficiency of the membrane itself which seems to be limited to an absolute frost point temperature of about $-20\,°\mathrm{C}$.

Referee comment:

3) Lines 177-180: I am curious about the later tests shown for background, simply because I did not understand the implications of no background particles found after five runs. Why does it take five runs to decrease, and does it mean that any background is then absent from thence forward in time? Have you explored this systematically, and/or after hours of operation? My personal understanding from an overlapping study in time with the one at the SGP site, is that the dewpoint was $-10\,°\mathrm{C}$ in that case, and that background counts at some level were always detected, if minimal. Hence, the basic question is if it is understood what dewpoint is sufficient for frost-free operation at any given T?

Answer:

We have to distinguish here between two sorts of background, one coming from frost build-up on the cold chamber walls during longer time operation at temperatures lower than the frost point temperature of sampled air, and the other coming from large aerosol particles or liquid droplets overlapping in size with ice crystal detection. Here we only argue about the absence of the background from frost artifacts when operating PINE with filtered, particle free air. It takes up to five runs to completely flush the chamber with filtered air and to achieve particle free conditions. Only then we can be sure that any remaining ice counts would come from frost at the walls. In longer term operations of PINE, we do such frost background checks not only once but usually repeat them every day, so we already tested the long term behavior for frost artefacts. To make this more clear we suggest to rephrase line 178 to

"…resulting in zero particle counts in the detection range for ice crystals after about 5 consecutive runs …', and to add in line 180 "Such frost background tests are usually repeated once every day in long term operation of PINE."

Referee comment:

4) Lines 183-184: It was not clear to me what actually constitutes the cooling system? Is it a plenum around the chamber and this is fed by the large chiller reservoir?

Answer:

Good point. The PINE-1A cloud chamber is actually cooled by circulating ethanol from the bath chiller through special thermo-conductive plastic tubes wounded around the cloud chamber. To make this clear, we suggest adding the following sentence in line 185:

"This is achieved by circulating the chilled ethanol from the bath chiller through thermo-conductive EPDM (ethylene propylene diene monomer rubber) tubes wounded around the chamber."

We also suggest removing the word "precisely" in line 183.

Referee comment:

5) Line 191: I expected that the minimum air temperature achieved would be colder than the minimum cooling temperature? Why are they the same?

Answer:

Thanks for this comment. The minimum gas temperature reached at a wall temperature of $-33\,^{\circ}\mathrm{C}$ is about $-40\,^{\circ}\mathrm{C}$. We will correct this error. Related to this, we noted that a wrong lower limit for the PINE measurement range is given in line 7 of the abstract. This will be changed from $-38\,^{\circ}\mathrm{C}$ to $-40\,^{\circ}\mathrm{C}$.

Referee comment:

6) Line 195: Can you explain the Stirling cooler method of cooling the wall of PINE-c for those of us unfamiliar with the exact cooling mechanism? E.g., fluid versus expansion cooling or whatever it is. The details on cooling systems in general does not match the later attention to detail of the OPCs.

Answer:

We suggest to add the following text at line xy of the manuscript:

"A dual opposed pistons compressor driven by linear motors with moving magnet flexure bearing design drives a Stirling-type pulse tube. As a consequence, there is only little vibration introduced to the cloud chamber in direct thermal contact to the pulse tube. The compressor of the cryocooler is force-flow air-cooled. Therefore, no cooling liquids are required and the cooling system is maintenance-free."

We also suggest to add the following reference:

D.L. Johnson, I.M. McKinley, J.I. Rodriguez, H. Tseng, and B.A. Carroll, Characterization testing of the Thales LPT9310 pulse tube cooler, in Cryocoolers 18 (S.D. Miller and R.G. Ross, Jr., eds.), pp. 125–133, Plenum Press, 2014.

Referee comment:

7) Line 198-200: Again, the cooling is understood, but the utility for performing low temperature ice nucleation experiments, especially where this will presumably involve more special control over the expansion conditions to meet some final peak relative humidity, is not yet discussed or demonstrated herein.

Answer:

OK, but we still would like to mention here the technical capabilities for future work with PINE and therefore suggest to modify lines 198-200 as follows: "PINE-c can also be cooled to a lower wall temperature of $-60\,^{\circ}\mathrm{C}$ and can therefore be operated at cirrus cloud temperatures in upcoming studies."

Referee comment:

8) Line 222: Does this more limited volume used to define ODV explain the higher value of lowest detection limit concentration listed in Table? Perhaps worth noting here, since It only comes up again at the end of section 4.

Answer:

Yes, this is indeed the reason for the different detection limits. At the end of the same paragraph we already mentioned the detection limit to depend on the volume flow through the OPC. Therefore, we do not see a need for change or extension here.

PINE operating principle:

Referee comment:

1) Line 250: To this point, the definition of ice crystals versus drops has not been made. Perhaps add a short note about this, "... as discussed later in this section"? Otherwise, this raised a number of questions immediately.

Answer:

Good point. We suggest to change line 249 to "...are then activated to form liquid cloud droplets and/or ice crystals, depending on ...", and to re-phrase the sentence in line 250: "Both droplets and ice crystals are measured with an OPC downstream of the chamber. Ice crystals are distinguished from droplets by their larger optical size, as discussed later in this section."

Referee comment:

2) Lines 269-270: Figure 3 is an important figure, and it raises a number of questions that were mostly answered in time over this section. However, I will list a number of them here. Immediately I wondered why the lowest temperature measured was used. As an aside, this point (lowest T used) should also be stated in the figure caption, for clarity. What differences are seen in these temperatures, and what uncertainty does this create? Are concentrations referenced to the entire integrated time interval and volume of expansion (and will this be the case also for the PINE-c), and do they represent the lowest temperature achieved (e.g., there is a $4\,^{\circ}\mathrm{C}$ cooling shown in the figure over the time of the expansion)? Hence, is it one measurement or

many, and how are the sub-intervals defined? A range of apparent ice crystal sizes are shown in Fig. 3, up to 100 microns. Are these ice sizes consistent with expectations of grown sizes for the conditions and growth times? The PINE chamber is quite small compared to the AIDA chamber where volumetric concentrations are assessed in situ. Is there sedimentation that could impact inferred concentrations and their reference temperature for the smaller geometry of the PINE? Have any such calculations been made at this time, or are they planned?

Answer:

We agree that this is an important figure, but more in the sense of explaining the basic measurement principle of PINE and the three different modes of what we call a run. The temperature chosen for this plot is only of minor importance here, but we agree to add this information to caption of Figure 3. The relation of increasing number of ice with decreasing temperatures in the course of an expansion is discussed in lines 330 to 345. In runs with a larger number of ice crystals, one may obtain a number of INP data points in certain temperature subintervals. We analyzed the PINE data in this way in a number of field and laboratory based operations, and may come back to this approach on future publications. For now, we decided to report the cumulative number of INPs that corresponds to the minimum nucleation temperature in a PINE run. The analysis program sums up all ice crystals detected during one run, and calculates the number density by dividing this number by the total volume that passed the OPC. According to the ice nucleation active surface site density concept, this cumulative number of ice is well defined and independent of the start temperature for droplet and ice formation during the expansion mode.

Concerning the temperature uncertainty see our answer to the general comments above. The reason and justification for using the lowest temperature measured as the "nucleation temperature" is mentioned in lines 343 to 345, and also the results of comparing PINE with AIDA results (see Figures 7 to 11).

Concerning the ice crystal size, please note that these are optical sizes. We know from experience with welas measurements at AIDA, and from scattering phase function calculations, that the sideward scattering geometry of both the welas and fidas sensors detect a-spherical particles with a much larger scattering intensity than spherical particles of the same volume and refractive index. Järvinen et al. (2014) determined an average oversizing factor of 2.2 for a welas sensor. For individual ice crystals, this factor can be much larger depending on their size, shape and orientation in the OPC detection volume. Therefore, the geometric size of ice crystals is much smaller than shown in Figure 3. We suggest to add the following text at the end of line 306:

"The use of a simple size threshold to distinguish between ice crystals and droplets is supported by the fact that the sideward scattering geometry of both the welas and fidas sensors detect a-spherical particles with a much larger scattering intensity than spherical particles of the same volume and refractive index. Järvinen et al. (2014) determined an average oversizing factor of 2.2 for the welas sensor. For individual ice crystals, this factor can be much larger depending on their size, shape and orientation in the OPC detection volume."

In this paper, we suggest to stay with demonstrating and documenting the quality and accuracy of PINE measurements and data analysis procedures by comparison to AIDA results (see also our answers to the general comments above).

Additional reference:

E. Järvinen, P. Vochezer, O. Möhler, and M. Schnaiter, "Laboratory study of microphysical and scattering properties of corona-producing cirrus clouds," Appl. Opt. 53, 7566-7575 (2014).

Referee comment:

3) Lines 273-275: Regarding the starting vapor saturation ratio for expansion, you assumed this or you set that partial pressure based on a room temperature RH measurement? Why would it be ice saturated if there is no ice on the walls? Or is it close enough as determined on some other basis? This would seem important for future use toward other measurements than immersion freezing.

Answer:

We did not say that there will be no ice at all at the wall. We only stated that no frost fragments are observed even after any deposits have eventually accumulated over longer operation periods while sampling slightly ice supersaturated air. When the frost point temperature of air added to the cloud chamber is higher than the wall temperature we assume that the excess water vapor still deposits to the wall so that ice saturated conditions are reached or at least approached at the beginning of the expansion mode. Part of this wall ice deposit may be removed again when refilling the cloud chamber which causes an adiabatic warming of the gas inside the cloud chamber. But the referee is raising an important point here which we will have in mind for further test series, in particular at lower temperatures of PINE operation.

Referee comment:

4) Line 288: Here an important distinction may arise, but perhaps the authors can correct any misconception I have. While described as purely immersion freezing, the temperature is already cold at the point of expansion, and so does the measurement also not integrate some proportion of INPs from any/all INP mechanisms, other than contact freezing, that ensue as the air rapidly cools and ultimately exits the chamber through the OPC? That is, somewhat similar to CFDCs when they are operated for bringing air to a final RH that is well above water saturation?

Answer:

Yes, the referee is right, other modes of nucleation are possible. In the case of very high aerosol concentrations in an AIDA cloud expansion experiment a clear development of a supercooled droplet cloud does not necessarily occur in the course of an expansion. We did observe this case in recent laboratory tests and calibration runs with high INP number concentrations. In all field operations so far we always saw a clear development of a supercooled droplet cloud and only a minor number fraction of ice crystals, as in the case mentioned in line 288. In this case we believe that most, if not all ice observed can only be formed by immersion freezing INPs.

Referee comment:

5) Line 317-318: Concerning addressing the size threshold setting for ice crystals, I struggled a bit to reconcile Figures A5 and 5. In A5, the scale is frequency, and it spans about three orders of magnitude out to 10 microns. Is there an issue in the fact that if cloud droplet concentrations range up to 1000 per cubic centimeter, and activated INP concentrations could range down to 1 per liter, then assessment of cloud drop frequency would have to be made over a greatly extended period of time to capture the tail of the distribution? Or is it simply the case that repeated experiments like the one in Fig. A5 never indicated a drop even in the size range greater than 10 microns? It might help to add the time and/or volume of assessment represented in Fig. A5. Clearly, Fig. 5 shows particle numbers appearing in these larger size ranges at 4-5 orders of magnitude below cloud droplet concentrations at least. This is an issue that perhaps deserved more attention in the paper, but if I understand, sensitivities of the ice cut size threshold will be more extensively covered in Adams et al. It would be good to add a reference for that paper, if it is now in submission.

Answer:

Figure A5 is just one example of a droplet size distribution without the presence of INPs. Of importance is here the sharp edge of the size distribution, which we also observed in many other cases. Given that the expansion is rapid, there is little opportunity for some droplets to grow more than others and a tail of the droplet size distribution towards larger diameters was not observed so far. This is also expected given the diameter growth rate of a spherical droplet in the continuum regime to be inversely proportional to its diameter. Systematic uncertainties related to the size threshold may more result from a potential overlap of the ice crystal size distribution with the droplet size distribution, less to a tail of the droplet size distribution towards ice crystal sizes. We have selected a safe threshold size to be sure we never catch the high size end of the droplet distribution, at the expense of eventually undercounting the ice crystal concentration.

This will indeed be investigated in more detail in upcoming publications. The one by Adams et al., however, is not yet submitted. We will therefore remove reference to this paper or replace by statements like “will be discussed in more detail in upcoming publications”.

Referee comment:

6) Lines 348-349 and lines 359-360: Note that the first statement repeats from earlier in the manuscript. One example is provided in Fig. A3. Perhaps repeating myself also, is this the very start of operations, or a period during the midst of operations? Why does it take 5 cycles at all, and does the background then stay that low in all cases? What does this depend on? The question arises again in the later sentence where long time operating detection limits are listed. Do not these very low detection limits listed for long operation imply the need for validating backgrounds being below such levels over such long times?

Answer:

As already stated above, it takes several cycles with filtered air to remove all or at least most of the aerosol particles and by that also the INPs from the cloud chamber and by that then prove that frost background is indeed zero. Such background operations are then repeated at least once a day to check for frost to accumulate or not. When not frost is accumulated, then we consider the chamber walls to stay free of accumulated frost formation and by that free or background frost artefacts. We agree to the referee that such background test have to be done also over longer operation times and already started to do so.

Laboratory tests of the prototype version PINE-1A:

Referee comment:

1) Lines 374-375: In Fig. 6, there looks to be up to 1C temperature uncertainty in defining the lowest temperature attributed to ice nucleation. Since T is not spatially uniform in the chamber, do you anticipate a bias in sampling only part of the flow as in PINE-1A versus all of the flow in PINE-c? Also, Figure 6 and its caption could use a little attention to description. At present the data are described as "all single ice crystals measured." Should it say something like "Data points indicate all single ice crystal event temperatures . . . "

Answer:

In a number of test runs with PINE-1A using either welas or fidas we did not observe a difference in the freezing temperature measured for pure water droplets. Such a difference or bias is also not expected because it is the same portion of air pumped through the OPC, just a different fraction analyzed. We agree to modify the caption of Figure 6 to

"The data points show event temperatures of all ice crystals measured with PINE-1A . . . in Figs. 4 and 5. The events are plotted as a function of the relative run time they were detected and the gas temperatures measured at the same time with three sensors . . . "

Referee comment:

2) Line 391: Just a note that there seems an inconsistency between the statement of a minimum pressure reduction every 5th cycle versus what is shown in Fig. 8 (and stated in that caption). It looks like 4 cycles. It is 5 cycles in Fig. 10.

Answer:

Thanks for noting this. It is even every third run in the example shown in Fig.8. We correct the text body and the figure caption for this.

Referee comment:

3) Lines 401 to end of section: The basic agreement shown between AIDA and PINE in Figures 7, 9 and 11 (over a more limited range) is excellent. I again wonder here about the percentage uncertainties being constant over the entire dynamic range of ice concentrations. For example, at the LOD, the true uncertainties must be larger, no? That statistical uncertainty does not appear to be captured in defining uncertainties based on the OPC ODV alone. I guess I expected based on statistical count considerations that the uncertainties should be larger for lower INP concentrations. Additionally, given that ice concentrations are integrated over the range of temperatures present throughout the volume, and if some of the crystals grow in that time to 50-100 microns (would be good to state the typical mode size), does sedimentation assuredly not impact/skew the results attributed to one temperature? There could be differences as to how this is measured temporally in situ in AIDA versus drawing the entire tank flow from the PINE, and there is some room for not discerning that in the comparisons shown. Nevertheless, a minor point overall.

Answer:

See our answers given to the general comment above, and the new Figure A6.

Field measurements with PINE-c:

Referee comment:

1) Lines 416-417: With an expansion mode time of 60 *to* 90 s*, a question arises as to the applicability of the discussion of temperature attribution and method for calculating INP concentrations with the PINE-c versus PINE-1A. Were they exactly the same (lowest T used, etc) for these presented analyses?*

Answer:

Yes, for both instruments, we analyzed and plotted the INP number concentrations for the lowest temperature reading, and used the same equations as discussed in the manuscript.

Referee comment:

2) Fig. 12: This is a nice compilation of results, if leaving a lot of room for discussion of their meaning still (i.e., variability of 2 orders of magnitude temporally at any T,) and raising all of the questions listed in the last sentence of this section. It is a minor concern for showing them in this manner, simply as a demonstration that the data were collected more or less autonomously over this period (maintenance or other attention needed were not discussed). Let me ask one thing though. The flattening of the INP concentrations toward the higher temperature limit of detection is interesting, but raises a question regarding the confidence in these results. The uncertainties are based on relative standard errors. The percentage errors are

quite small and I wonder how these can be the same at the LOD as they are at any other conditions. This is the same question raised for PINE-1A.

Answer:

We intend to introduce these compiled quasi-raw PINE data in a snapshot single figure to demonstrate the PINE's capability for fairly long-term continuous operation in a simple manner. Nevertheless, we certainly understand the reviewer's concerns - there is ample room for further discussion on many details. To mitigate the reviewer's and reader's misgivings, we have revised our sentence in L425-427 to address remaining items to be investigated in the future (please see our response below). The nice uncertainties in this figure are based on relative standard errors of time-averaged data, which appear to be small - equivalent to or smaller than the "systematic" error of OPC ($\pm 20\%$). We have started to run detailed statistical error analysis with an inclusion of estimated backgrounds for a subset of our PINE-c field data, and confirmed that the nice uncertainty near the LOD at relatively high temperatures propagates and becomes apparent, as a relative importance of background contribution becomes prominent in such temperatures. Some of the authors of this paper will carefully characterize the data in this region and address our findings along with other detailed topics (i.e., L425-427) in our future paper. We suggest to add the following sentence at the end of the Figure 12 caption: "Statistical errors from low counting signals are not considered here and will be the subject of further analysis."

Referee comment:

3) Lines 423-424: I am not sure what is meant by "warranted" here. Possible? Also, can the point regarding the dewpoint temperature be clarified? Dewpoint is not controlled somehow? It would be much higher in summer and much lower in winter. How might this affect the operational range, background etc, or does this remain to be investigated?

Answer:

Yes, "possible" is a better and more appropriate word choice here. Furthermore, thanks to the referee's comment, we noted that we should refer here to the dew point temperature of the sample air after passing the dryer, and not to the dew point of the ambient air. We therefore suggest to rephrase lines 423-424 as follows:

"This temperature range represents the PINE-c condition, where ice nucleation through immersion freezing was possible below the frost point temperature of the sample air, which passes the membrane diffusion dryers operated at maximum drying efficiency. For measurements at higher temperature, the drying efficiency has to be reduced, in order to increase the dew point of the sampled air and to exceed water saturation during the expansion mode at higher temperature. Next versions of the PINE control program will include this option for operation at higher temperature."

Referee comment:

4) Lines 425-427: What exactly is meant by deconvolution of nucleation modes? Meaning different operation of the PINE than discussed in this paper, which is immersion freezing? Or meaning resolving what I mentioned earlier in this review, the temporal evaluation of data during single expansions? This is a point that should be clarified, as it is important to state which potential aspects of PINE measurement capabilities are demonstrated in this first publication and which remain.

Answer:

It was meant for separating/estimating ice crystals formed through immersion freezing from other ice nucleation paths, however not just from PINE measurement but in combination with all other instruments data and measurements. PINE alone will only be able to measure immersion freezing in the temperature range of interest in this paper. We admit the sentence in lines 425-427 is confusing. For clarity, we suggest to revise the sentence as:

"Any further scientific discussions regarding PINE-c operations and observations, in combination with other INP and aerosol measurements during the ExINP-SGP campaign, are beyond the scope of our current study, and will be followed up in future publications."

Other editorial comments:

Line 96: typo - based

Corrected

Line 266: Suggest "one of" after "An example of . . . "

Added

Line 282: "so" not needed before "calculated"

Removed

Line 287: Suggest "than" for "as"

Changed

Figure 3 caption: Suggest to add "Calculated" at start of sentence starting "Liquid water . . . "

Changed

References:

Bi, K., G. R. McMeeking, D. Ding, E. J. T. Levin, P. J. DeMott, D. Zhao, F. Wang, Q. Liu, P. Tian, X. Ma, Y. Chen, M. Huang, H. Zhang, T. Gordon, and P. Chen, 2019: Measurements of ice nucleating particles in Beijing, China. Journal of Geophysical Research: Atmospheres, 124, 8065–8075. https:// doi.org/10.1029/2019JD030609

Brunner, C. and Kanji, Z. A.: Continuous online-monitoring of Ice Nucleating Particles: development of the automated Horizontal Ice Nucleation Chamber (HINC-Auto), Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-306, in review, 2020.

Author comment in response to the comments provided by Referee #4

We thank referee #4 for her/his effort in reading and commenting our manuscript. In the following, we repeat the referee's comments (italics), give point-by-point answers, and suggest manuscript revisions based on the referee's comments and our answers. Respective reference will be given to the line numbers of manuscript version 1.

Referee comment:

The knowledge of ice nucleating particles and their impacts on clouds was restricted by the development of measurement techniques and instruments. This manuscript presents a new instrument based on expansion chamber for both laboratory studies and field observations to measure ice nucleating particles. The authors successfully demonstrate the applicability of their new instrument to be compared with AIDA and deployed in a field campaign. Different from the commonly used Continuous Flow Diffusion Chamber (CFDC), PINE is truly the first commercial instrument capable of automated long-term continuous observation, and its development provides an excellent complement to enrich measurement methods. This research aligns well with the scope of AMT. The manuscript is well written and easy to follow, thus should be acceptable for publication after considering following minor comments:

Referee comment:

P2 Line28: The abbreviation "(INPs)" should not be linked directly after "...atmospheric aerosol particles". Moreover, two sets of parentheses are used instead of one. For example, "...atmospheric aerosol particles (INPs) (Vali et al., 2015)".

Answer:

Thaks for noting this. We suggest to reformulate lines 27/28 as follows:

"In the absence of homogeneous freezing, the cloud ice phase is initiated in various ways by ice nucleating particles (INPs), a very small fraction of atmospheric aerosol particles (Vali et al., 2015)."

Referee comment:

P2-3: The need for promoting INP monitoring was put forward until P3 Line 63, and the new methods and instruments for INP measurements were discussed from P3 Line 90. Please consider adding more descriptions and comparisons of existing instruments, especially the most commonly used CFDC, and simplifying the context on INP.

Answer:

Referees #1 and #3 have also suggested similar revisions to the introduction. We have removed some parts and in particular revised lines 84 to 100 (see our answers to referees #1 and #3). We have in particular added reference to recent developments of by Bi et al. (2019) and Brunner and Kanji (2020), and for other instruments like CFDCs we referred the reader to the paper by DeMott et al. (2018).

Referee comment:

P9 Line275: A parenthesis is missed.

Answer:

Yes, added.

Referee comment:

P12-14: PINE-c performed the field measurements to demonstrate its capability, however, the comparisons with AIDA and performance tests were conducted by the prototype version PINE-1A. PINE-c is a further developed version with major upgrades in chamber type, cooling system, controlled temperature range, particle detector, and so on. So direct characterizations and tests of the performance of PINE-c would be helpful.

Answer:

This is a good point. Unfortunately, there was time for only a few test runs of PINE-c at the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud chamber facility, before the instrument had to be delivered for participation in the ARM-SGP field campaign. Meanwhile, three more instruments of type PINE-c have been built, and two of them are currently extensively tested and operated at the AIDA facility (see for instance the new Figure A6 in the appendix) and also next to the PINE-1A version. A more comprehensive comparison of PINE-c with other methods and instruments will be teh subject of upcoming paublications.

Referee comment:

Figure 7: Please notice the superscript of the unit, "...5 l min-1".

Answer:

Yes, corrected.

References:

Bi, K., G. R. McMeeking, D. Ding, E. J. T. Levin, P. J. DeMott, D. Zhao, F. Wang, Q. Liu, P. Tian, X. Ma, Y. Chen, M. Huang, H. Zhang, T. Gordon, and P. Chen, 2019: Measurements of ice nucleating particles in Beijing, China. Journal of Geophysical Research: Atmospheres, 124, 8065–8075. https:// doi.org/10.1029/2019JD030609

Brunner, C. and Kanji, Z. A.: Continuous online-monitoring of Ice Nucleating Particles: development of the automated Horizontal Ice Nucleation Chamber (HINC-Auto), Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2020-306, in review, 2020.

DeMott, P. J., Möhler, O., Cziczo, D. J., et al.: The Fifth International Workshop on Ice Nucleation phase 2 (FIN-02): laboratory intercomparison of ice nucleation measurements, Atmos. Meas. Tech., 11, 6231–6257, https://doi.org/10.5194/amt-11-6231-2018, 2018.

The portable ice nucleation experiment PINE: a new online instrument for laboratory studies and automated long-term field observations of ice-nucleating particles

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Abstract. Atmospheric ice-nucleating particles (INP) play an important role in determining the phase of clouds, which affects their albedo and lifetime. A lack of data on the spatial and temporal variation of INPs around the globe limits our predictive capacity and understanding of clouds containing ice. Automated instrumentation that can robustly measure INP concentrations across the full range of tropospheric temperatures is needed in order to address this knowledge gap. In this study, we demonstrate the functionality and capacity of the new Portable Ice Nucleation Experiment (PINE) to study ice nucleation processes and to measure INP concentrations under conditions pertinent for mixed-phase clouds, with temperatures from about $-10\,^{\circ}\mathrm{C}$ to about ~~$-38\,^{\circ}\mathrm{C}$~~ $-40\,^{\circ}\mathrm{C}$. PINE is a cloud expansion chamber which avoids frost formation on the cold walls, and thereby omits frost fragmentation and related background ice signals during the operation. The development, working principle, and treatment of data for the PINE instrument is discussed in detail. We present ~~extensive~~ laboratory based tests where PINE measurements were compared with those from the established AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud chamber. ~~The results show good agreement of PINE~~ Within experimental uncertainties, PINE agreed with AIDA for homogeneous freezing of pure water droplets and the immersion freezing activity of mineral aerosols. Results from a first field campaign conducted at the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) observatory in Oklahoma, USA, from October 1 to November 14, 2019 with the latest PINE design (a commercially available PINE chamber) are also shown, demonstrating PINE's ability to make automated field measurements of INP concentrations at ~~high~~ a time resolution of about 8 minutes with continuous ~~wall temperature scans between -5 and $-35\,^{\circ}\mathrm{C}$~~ temperature scans for INP measurements between $-10\,^{\circ}\mathrm{C}$ and $-30\,^{\circ}\mathrm{C}$. During this field campaign, PINE was continuously operated for 45 days in a fully automated and semi-autonomous way, demonstrating the capability of this new instrument to be also used for longer term field measurements and INP monitoring activities in observatories.

Copyright statement. TEXT

1 Introduction

Atmospheric ice-nucleating particles ~~(INP)~~ induce ice formation in atmospheric clouds, and by that are important for initiating precipitation in mixed-phase clouds and determining the phase of clouds, their albedo, lifetime and other important properties (DeMott et al., 2010). However, the details of these aerosol-cloud-climate interactions remains highly uncertain (Boucher et al., 2013; Fan et al., 2017; Lohmann, 2017). This is partly due to the fact that such clouds are rather complex systems, and that the knowledge on the formation, the concentration and the fate of ice crystals is still uncertain (Heymsfield et al., 2017; Korolev et al., 2017).

In the absence of homogeneous freezing, the cloud ice phase is initiated in various ways by ice nucleating particles (INPs), a very small fraction of atmospheric aerosol particles ~~(INPs)~~ (Vali et al., 2015). In mixed-phase clouds, immersion freezing is thought to be the dominating freezing mechanism (de Boer et al., 2011; Hande and Hoose, 2017; Hoose et al., 2010). Vergara-Temprado et al. (2018) showed INPs to have a strong control of cloud reflectivity over the Southern Ocean. Mülmenstädt et al. (2015) and Field and Heymsfield (2015) showed the ice or snow phase to exist in a large fraction of precipitating clouds, in particular over the continents. This underlines the importance of INPs for cloud radiative properties and precipitation formation, but it should be noted here that the cloud ice phase not only depends on the primary ice formation by INPs, but is also largely influenced by a cascade of secondary ice formation and interaction processes, in particular at temperatures above $-15\,^{\circ}\mathrm{C}$ (Field et al., 2016). Increased ice crystal concentrations can e.g. lead to rapid cloud glaciation and associated dissipation (Campbell and Shiobara, 2008; Paukert and Hoose, 2014), as also observed recently in a laboratory cloud chamber experiment (Desai et al., 2019) .

~~At higher altitudes with temperatures below about $-35\,^{\circ}\mathrm{C}$, cirrus cloud ice crystals can either be formed by homogeneous freezing of aqueous aerosol particles at relatively high ice supersaturations (Koop et al., 2000; Kärcher and Lohmann, 2002), or by heterogeneous ice nucleation processes at lower ice supersaturations (Hoose and Möhler, 2012; Kärcher and Lohmann, 2003; Krämer et . As in the mixed-phase cloud regimes, the heterogeneous pathways of cirrus ice crystal formation are limited and controlled by the abundance of INPs in the upper troposphere, in addition to other factors like dynamic, thermodynamic or kinetic processes (Heymsfield et al., 2017).~~

Throughout the troposphere, INPs are difficult to identify and to quantify due to their low and largely variable number fraction to the total aerosol concentration (DeMott et al., 2010; Kanji et al., 2017). This fraction strongly depends not only on temperature and relative humidity conditions, but also on the particle type, size, and surface properties (Pruppacher and Klett, 2010; Holden et al., 2019). Nevertheless, cloud, weather and climate models need to formulate and quantify primary ice formation as accurately as possible (Vergara-Temprado et al., 2018; Waliser et al., 2009). This is achieved by calculating the abundance of INPs with parameterizations based on either laboratory ice-nucleation experiments (Hoose and Möhler, 2012; Murray et al., 2012; Sesartic et al., 2013; Spracklen and Heald, 2014; Vergara-Temprado et al., 2018) or field measurements (DeMott et al., 2010; McCluskey et al., 2018; Tobo et al., 2013; Wilson et al., 2015). A number of different parameterizations

for the various pathways of atmospheric ice nucleation in supercooled liquid and cirrus clouds have been developed under different assumptions, based on either temperature and time dependent ice formation rates according to classical nucleation rate formulations (Barahona and Nenes, 2009; Kärcher and Lohmann, 2002, 2003), the number concentration of larger aerosol particles (DeMott et al., 2010, 2015), or the temperature-dependent ice nucleation active site (INAS) density on the surface of aerosol particles (Connolly et al., 2009; Harrison et al., 2019; Niemand et al., 2012; Ullrich et al., 2017).

~~The proper use of aerosol particle specific INP parameterizations, however, requires aerosol type specific knowledge of parameters like number concentration and size distribution, needed as input to the calculation and prediction of INP concentrations. The application of these ice nucleation parameterizations can be challenging, because of limitations in aerosol characterization in field campaigns and modelling studies. In particular, information on the types, chemical nature, and mixing state of aerosol particles is often missing, but may have a strong impact on the ice nucleation activity or INP abundance (Möhler et al., 2008) . At present, the~~ Existing parameterisations are applied in models to calculate and predict primary ice formation in clouds, however, the atmospheric INP data that we can compare with global fields of model predicted INP concentrations are ~~extremely~~ limited in spatial, temporal and concentration ranges (Burrows et al., 2013; Vergara-Temprado et al., 2017). Hence, there is an urgent need for more INP observation and monitoring, not only for constraining INP predictions by models and representing a fuller range of INP sources in those models, but also to extend the data base for a better understanding of temperature dependent INP concentrations throughout the atmosphere and the year.

Existing measurements of ambient INP concentrations at mixed-phase cloud temperatures (Kanji et al., 2017) show a great variability not only across the temperature range from about $-5\,^{\circ}\mathrm{C}$ to $-35\,^{\circ}\mathrm{C}$ (10 orders of magnitudes), but also at a single temperature (~ 4 orders of magnitude). Different aerosol types were found to dominate the INP population at specific temperatures. ~~While high temperature INPs are typically associated with biological particles (e.g., DeMott et al., 2010; Creamean et al., 2013; Prenn , their atmospheric implication remains uncertain (Després et al., 2012; Hummel et al., 2018). Marine aerosol particles were identified to be ice active at $T > -30\,^{\circ}\mathrm{C}$ (Alpert et al., 2011; Brier and Kline, 1959; DeMott et al., 2015; Mason et al., 2015a, b; McCluske . They might be an important source for INPs in the absence of more ice active aerosol particles (Burrows et al., 2013; Vergara-Temprado et . Mineral dust particles are very efficient INPs at $T < -20\,^{\circ}\mathrm{C}$ (Boose et al., 2016c; Harrison et al., 2019; Ullrich et al., 2017) and may dominate the INP number concentrations in many locations (Atkinson et al., 2013; Sanchez-Marroquin et al., 2020; Tobo et al., 20 .~~

Most of ~~these~~ previous INP measurements were only sensitive ~~for~~ to immersion freezing INPs in the temperature range of mixed-phase clouds, and were carried out at boundary layer field sites which were considered to be predominantly influenced by different aerosol types. Measurements in the free troposphere were either performed at high altitude mountain stations (Boose et al., 2016a, b; DeMott et al., 2003a; Conen et al., 2015; Lacher et al., 2018a, b) or with aircraft-based measurements (Rogers et al., 2001; DeMott et al., 2003b; Prenni et al., 2009; Pratt et al., 2010; Eidhammer et al., 2010; Field et al., 2012), but most of them were also limited to measure immersion freezing INPs at higher temperatures. DeMott et al. (2003b) also measured the concentration of INPs active in the deposition mode at temperatures below $-40\,^{\circ}\mathrm{C}$.

The identification of INP types in ambient air remains challenging. Most ambient studies focus on sampling INPs in campaigns over a limited time period and focused on specific air masses like Saharan dust events (Boose et al., 2016b), biogenic

source regions (O'Sullivan et al., 2018) or marine environments ~~(Mason et al., 2015a)~~(Mason et al., 2015a; McCluskey et al., 2018), or use back trajectories to identify source regions (e.g., Lacher et al., 2017; Wex et al., 2019). ~~Such approaches are not only in need of high-time resolution INP measurements to~~ Depending on the specific campaign goals and objectives, different instruments and methods were used like CFDCs with higher time resolution to e.g. characterize changing air masses ~~, but also~~ (Boose et al., 2016a; Lacher et al., 2018b), or aerosol filter based offline methods to achieve high sensitivity for characterizing INPs at higher temperatures or in clean environments (Wex et al., 2019), or a combination of both (Welti et al., 2018). What is missing so far are long-term monitoring of INPs ~~to capture the bigger picture and not only short-term periods of the atmosphere~~with high time resolution and over a wide temperature range.

An increasing number of new methods and instruments for INP measurements have been developed and compared to each other during the previous years (DeMott et al., 2011; Hiranuma et al., 2015; Wex et al., 2015; DeMott et al., 2018). The most recent and comprehensive INP instrument and method intercomparison study was the Fifth International Workshop on Ice Nucleation Research (FIN-2), and many of the latest developments for atmospheric INP measurements are included and described with respective references in the overview paper by DeMott et al. (2018). Most of the INP methods showed reasonable agreement with each other, but ~~most~~ many of them are time and operator intensive. A general feature is~~that available online instruments can only be operated periodically, and offline methods base on aerosol~~ , that offline methods based on aerosol filter samples have poor time resolution depending on required aerosol sampling time of hours to days~~. All existing methods~~ , and require intensive man-power and time for ~~operation or~~ both operation and offline analysis. ~~The low time resolution of offline techniques challenges~~ Most online instruments can only be operated periodically, and also require operator time during the measurements, but can be operated for INP measurements at higher time resolution in particular at low temperature or in polluted environments where concentrations are higher. Only recently, newly developed INP instruments with a higher degree of automation became available (Bi et al., 2019; Brunner and Kanji, 2020). The automated CFDC instrument used by Bi et al. (2019) performed INP measurements during a period of one month in 2018 at temperatures between $-20\,^{\circ}\mathrm{C}$ and $-30\,^{\circ}\mathrm{C}$. The CFDC instrument called HINC-Auto (Horizontal Ice Nucleation Chamber) used by Brunner and Kanji (2020) autonomously measured immersion freezing INPs for 90 consecutive days, but only at one temperature of $-30\,^{\circ}\mathrm{C}$. A combination of both, high time resolution and wide temperature range for long-term INP measurements, together with a comprehensive set of high resolution aerosol analytics, would challenge the comparison to potential driving factors for ~~ice nucleation, as e. g. the size and chemistry of the aerosol population. For such measurements, online INP instruments are desirable, having a high-time resolution of minutes.~~ atmospheric ice nucleation.

This paper presents the development, technical description, working principle, as well as first laboratory and field applications of the new Portable Ice Nucleation Experiment PINE. PINE is the first fully automated instrument for laboratory ice nucleation studies and long-term field observations of INPs in a wide temperature range~~from $-10\,^{\circ}\mathrm{C}$ to about $-60\,^{\circ}\mathrm{C}$, including~~ . This paper demonstrates the instrument's ability to measure in the mixed-phase cloud ~~and cirrus cloud regimes and related primary ice formation processes~~temperature regime from from $-10\,^{\circ}\mathrm{C}$ to $-40\,^{\circ}\mathrm{C}$. PINE is also able to measure ice nucleation at cirrus cloud temperatures to about $-65\,^{\circ}\mathrm{C}$, which is the topic of ongoing studies. Similar to the AIDA (Aerosol Interaction and Dynamics in the Atmosphere) cloud simulation chamber, PINE is based on a pumped expansion principle to

induce ice and water supersaturated conditions for aerosol particles sampled either from laboratory setups or natural environments. The instrument is operated in repeated cycles of sampling the aerosol into a pre-cooled cloud chamber, activating the aerosol particles as supercooled droplets and ice crystals by expanding the air inside the cloud chamber, and refilling the cloud chamber with fresh aerosol for the next cycle (see section 4).

2 Basic principles and milestones of the PINE development

The idea for PINE resulted from ~~almost 20 years of experience~~ the experience in operating the AIDA facility for cloud experiments at simulated conditions of up-drafting atmospheric air parcels. Cloud formation in the rigid but large AIDA chamber with a volume of $84\,\mathrm{m}^3$ is induced in a controlled way by lowering the pressure at different rates, starting from well controlled thermodynamic conditions (Möhler et al., 2003, 2005). With a volume of only about $10\,\mathrm{L}$, the PINE cloud simulation chamber is much smaller, transportable, and operated in a fully automated sequence. Similar to the AIDA cloud chamber, PINE also uses the principle of pressure reduction by controlled pumping of air out of the cloud chamber. By that, the temperature in the chamber decreases due to expansion cooling, while the relative humidity increases. This causes the aerosol particles, which are present in the chamber prior to the expansion, to act as Cloud Condensation Nuclei (CCN) and/or INPs to form liquid cloud droplets and ice crystals, depending on the temperature, ice supersaturation and the type of aerosol. The starting temperature of each expansion run ~~, and thereby the temperature range of ice formation and INP detection,~~ can be set in a wide range from about $-10\,^{\circ}\mathrm{C}$ to $-60\,^{\circ}\mathrm{C}$, depending on the capacity of the cooling system. Large aerosol particles, droplets and ice crystals are measured and counted with an optical particle counter (OPC)~~mounted directly to the~~ . Placing the OPC in the vertically oriented pump tube below the cloud chamber was one of the critical development ideas for PINE (see patent applications DE 10 2018 210 643 A1 and US2020/0003671 A1). PINE can be operated both for ice nucleation research in the laboratory, and for INP measurements in field campaigns ~~or~~ includig long term monitoring activities.

The first version of PINE was successfully tested in January 2016. It consisted of a simple perplex chamber of $10\,\mathrm{L}$ volume with manual valve and flow control, and a welas 2300 single particle optical detector from Palas GmbH, Karlsruhe, Germany. ~~This setup was operated in a cold room at temperatures around $-10\,^{\circ}\mathrm{C}$ and sampled Snomax© aerosol particles for first proof-of-concept studies of immersion freezing in the small cloud expansion chamber. The critical development idea for PINE was to pass the total pump flow during the cloud expansion cycle through the optical particle counter directly attached to the pump line (see patent applications DE 10 2018 210 643 A1 and US2020/0003671 A1).~~ Another prototype chamber of about $7\,\mathrm{L}$ volume was then built of stainless steel and also operated in a cold room for further proof-of-concept experiments.

Based on the development and operational experience with the prototype versions, we developed the first mobile versions PINE-1A and PINE-1B with their own cooling systems and a control system for semi-autonomous operation during laboratory ice nucleation measurements and field INP observations. Because both systems are almost identical, we only refer to PINE-1A in the following sections, for simplicity. PINE-1A can be operated in a temperature range from $-10\,^{\circ}\mathrm{C}$ to about $-40\,^{\circ}\mathrm{C}$, was characterized in a series of laboratory experiments, and was used in a first field campaign~~(Adams et al. , in prep.).~~ . As a next step, the version PINE-c was developed, which is now commercially available from Bilfinger Noell GmbH in Germany (see

https://www.noell.bilfinger.com/pine/#c167514). PINE-c is operated in the same way as PINE-1A, but received a few new components and features making it more compact and autonomous for operation in both field and lab studies. This will be detailed in Sect. 3, together with a general technical description of the new PINE instrument. The typical working principle and operation of PINE will be explained in Sect. 4. In Sect. 5 we summarize and discuss some first results from laboratory test and characterization experiments of PINE-1A in comparison with the AIDA cloud chamber. Finally, in Sect. 6, we will present and discuss some results from a first field application of PINE-c, which continuously measured during all 45 days of a field campaign at the DOE SGP site in Oklahoma from October 1st to November 14th, 2019.

3 PINE instrument setup

As illustrated in Fig. 1, PINE consists of 5 major parts, (I) an inlet system, (II) a cloud chamber, (III) a cooling system, (IV) a particle detection system, and (V) a control and data acquisition system. Figure 2 shows a simplified schematics of the PINE setup in the different operational flow configurations that will be discussed in Sect. 3.

The inlet system (Fig. 1, part I) is composed of an inlet or sampling tube, a diffusion dryer, a humidity sensor and a bypass flow section with aerosol particle filter for background measurements. The relative humidity, measured with a dew point sensor (Vaisala DRYCAP® DMT143) has to be high enough to allow cloud droplet formation upon expansion cooling, and at the same time low enough to avoid frost formation on the chamber walls (see Sects. 4 and 5). Both the prototype version PINE-1A and the commercial version PINE-c (see Tab.1 and Sect.6), are equipped with two nafion membrane diffusion dryers (Permapure, MD-700-24S-1, length 62 cm) in parallel, in order to reduce the flow through one single dryer and by that enhance the drying efficiency.

Figure A1 shows the PINE sample flow dryer setup with two nafion diffusion dryers and other major components. The sample flow passes the straight nafion tube of $1.7\,\mathrm{cm}$ diameter and $62\,\mathrm{cm}$ length from top to bottom. The nafion tube is located inside an airtight stainless steel tube of $2.5\,\mathrm{cm}$ diameter. A second air flow is passing the annular gap between the coaxial nafion and stainless steel tubes from bottom to top (counter flow arrangement). For simplicity, the PINE system uses ambient air for this counter flow, but at reduced absolute pressure. The absolute pressure reduction also reduces the water vapour partial pressure compared the sample flow inside the nafion tube at ambient pressure. This water vapour partial pressure difference across the nafion membrane, which is permeable for water molecules, drives a diffusional flow of water molecules from the sample flow to the counter flow. The molar flux of water molecules increases with the pressure difference across the membrane and the residence time of the sample air inside the nafion tube. As seen in Fig. A2, the drying efficiency increases with pressure reduction. The pressure of the counter flow air is controlled with a pressure controller (Wagner-MSR type P-702), located between the dryer and the vacuum pump, and the volumetric flow rate of the counter flow air is controlled with a critical orifice at the inlet side. In comparison to conventionally used diffusion dryers with water adsorption material, the membrane dryers used in the PINE setup have the great advantage that they can be operated for long-term without decreasing their drying efficiency.

Because the current PINE instrument versions are typically operated with a sample flow rate of up to $4\,\mathrm{L\,min^{-1}}$ (see Sect. 4), two nafion dryers are used in parallel operation, in order to limit the sample flow through each dryer to $2\,\mathrm{L\,min^{-1}}$. If needed, the dryers can then be operated with a maximum pressure difference of about $800\,\mathrm{hPa}$ to achieve a high drying efficiency with a drop in the sample flow dew point temperature of at least $10\,^{\circ}\mathrm{C}$. As mentioned above, the frost point temperature of the sample air should be close to the wall temperature of the PINE cloud chamber. If the sampled air is too humid, frost may form at the coldest wall sections, potentially causing ~~and~~ an increasing ice background due to frost artefacts. However, this was not the case when operating PINE-1A during a first field application ~~(Adams et al., in prep.)~~ for several weeks at temperatures below $-25\,^{\circ}\mathrm{C}$ and sample air frost point temperatures around $-15\,^{\circ}\mathrm{C}$. This was tested by passing the sample flow through the particle filter (see Fig. 2) resulting in zero particle counts in the detection range for ice crystals after about 5 consecutive runs (Fig. A3~~; see also Adams et al., in prep.~~). This means that when the sample air is passing through the bypass particle filter, the system detects neither aerosol particles, nor activated droplets nor ice crystals. In the commercial version, the standard location of the dryers is next to the cloud chamber with vertical orientation, so that the sampled air flows in upward direction through the dyers, then passes a $90°$ bend, a horizontal distance of $50\,\mathrm{cm}$ and another $90°$ bend to then flow downward into the PINE cloud chamber. The aerosol particle loss for this setup was measured to be less than $20\,\%$ for particles smaller than $2\,\mu\mathrm{m}$ diameter. It decreased to about $50\,\%$ for particles with an aerodynamic diameter of about $4\,\mu\mathrm{m}$. The dryers can also be mounted above the PINE chamber for a strictly vertical sample flow, for which a further reduced particle loss can be expected. More systematic sampling efficiency measurements for different configurations and operations will be performed in the future.

The heart of a PINE instrument is a temperature controlled cloud chamber (Part II in Fig. 1). The PINE-1A cloud chamber has a volume of about $7\,\mathrm{L}$ and is made of stainless steel, with a central cylindrical part and two cones at the top and the bottom (Tab. 1). With the cooling system (part III in Fig. 1), the wall temperature of the cloud chamber can either be ~~precisely~~ controlled at a constant value, or changed at constant cooling or heating rates of up to $0.3\,^{\circ}\mathrm{C\,min^{-1}}$. The PINE-1A cloud chamber is temperature-controlled with an ethanol bath chiller (Lauda RP 855; Lauda-Königshofen, Germany). This is achieved by circulating the chilled ethanol from the bath chiller through thermo-conductive EPDM (ethylene propylene diene monomer rubber) tubes wounded around the chamber. The wall temperature of the chamber is measured with three thermocouples attached to the outer chamber walls at three different locations. The gas temperature inside the cloud chamber is also measured with three thermocouples located in the bottom, middle and upper section of the chamber about 5 cm off the wall (see Fig. A4). All thermocouples have been calibrated to a reference sensor (Lake Shore, Model PT-103, Sensor Typ Platinum Resistor) with an accuracy of $\pm 0.1\,^{\circ}\mathrm{C}$. A minimum wall temperature of about $-33\,^{\circ}\mathrm{C}$ can be reached with PINE-1A. With additional expansion cooling of the chamber volume (see Sect. 4), a minimum gas temperature of about ~~$-33\,^{\circ}\mathrm{C}$~~ $-40\,^{\circ}\mathrm{C}$ is then reached for ice activation of the aerosol particles.

PINE-c is equipped with a thin-walled aluminium cloud chamber with a slightly larger volume of $10\,\mathrm{L}$ as compared to PINE-1A (see Table 1). Mainly for thermal insulation, the cloud chamber is located in an evacuated stainless steel container and is cooled with a Stirling cooler (Thales, LPT9310~~). The~~ , Johnson et al. (2014)). A dual opposed pistons compressor driven by linear motors with moving magnet flexure bearing design drives a Stirling-type pulse tube. As a consequence, there is only little vibration introduced to the cloud chamber in direct thermal contact to the pulse tube. The compressor of the cryocooler is

force-flow air-cooled. Therefore, no cooling liquids are required and the cooling system is maintenance-free. The combination of the low mass and heat capacity of the thin-walled cloud chamber and the high cooling power of the stirling cooler allows to cool the PINE-c cloud chamber at a rate of up to approximately $0.6\,^{\circ}\mathrm{C\,min^{-1}}$ without any notable effects of measurement disturbance (see Sect. 6). The heating rate of the chamber can also automatically be set to a value up to $0.6\,^{\circ}\mathrm{C\,min^{-1}}$. By that, faster temperature scans than with PINE-1A can be achieved for temperature-dependent ice nucleation and INP measurements. PINE-c can also be cooled to a lower wall temperature of -60°C and can therefore be operated ~~for ice nucleation experiments and INP measurements~~ at cirrus cloud temperatures in upcoming studies.

The PINE particle detection system (part IV in Fig. 1) consists of an OPC connected to the control and data acquisition system (part V in Fig. 1). Depending on the OPC type, aerosol particles, activated cloud droplets and ice crystals are detected during the different run modes as described in Sect. 4. The OPC is mounted to the pump tube, with a minimum distance to the cloud chamber in order to minimize warming of the cold air flow from the cloud chamber and by that avoid evaporation of supercooled cloud droplets and sublimation or melting of ice crystals. PINE-1A is equipped with a welas 2500 sensor connected to a Promo© 2000 system (Palas GmbH, Karlsruhe, Germany). The same sensor has been operated for many years at the AIDA cloud chamber for cloud droplet and ice crystal detection (Möhler et al., 2006; Wagner and Möhler, 2013). It measures aerosol particles, water droplets and ice crystals with optical sizes between 0.7 and $220\,\mu\mathrm{m}$ diameter, depending on the sensitivity setting of the photomultiplier measuring the intensity of light scattered by single particles when passing the optical detection volume (ODV). The welas sensor records for each detected particle the time of detection, the transit time through the ODV, and the intensity of light scattered into a range of scattering angles around $90°$ (sideward scattering). This optical arrangement is favourable for the selective detection of a-spherical ice crystals, which are measured at a larger optical size than spherical droplets of the same volume and can therefore more easily be distinguished from droplets by setting a simple threshold for the optical size (see Sect. 4).

The welas 2500 sensor has a well confined ODV with a sample flow cross section area $A_w = 0.24\,\mathrm{mm^2}$ and a length $l_w = 0.31\,\mathrm{mm}$. Because the transect time τ_w of particles through the ODV is also measured, the sample flow rate through the ODV can be calculated as

$$F_w = \frac{A_w l_w}{\tau_w}. \tag{1}$$

With the count rate c_p of detected particles, one can then calculate the particle number concentration

$$n_p = \frac{c_p}{F_w}. \tag{2}$$

On average, the ratio of the volume flow through the ODV to the total volume flow through the welas 2500 sensor is about 0.105. This means that the sensor detects only about $10\,\%$ of the particles sampled from the cloud chamber. The PINE-c version uses a new OPC called fidas-pine (Palas GmbH, Karlsruhe, Germany). This new OPC was developed especially for the PINE-c instrument and analyses the full sample flow of up to $5\,\mathrm{L\,min^{-1}}$ for particles in a size range similar to the welas 2500 sensor. For PINE-c, the particle number concentration can still be calculated with Eq.2, just by replacing the flow rate through the ODV of the welas 2500 sensor by the total sample flow rate F_{em} during the expanion mode (see Sect. 4). Therefore, fidas-pine has a 10 times higher detection rate of particles, and by that a 10 times lower INP concentration detection threshold than PINE-1A.

PINE is controlled via a bespoke LabVIEW program, which sets the respective measurement condition, displays the parameters such as particle size, temperature, pressure, and flows, and saves the data internally. Metadata describing the experiment are saved automatically using LabVIEW, such as date and time, type of OPC used and its configuration, temperature and pressure conditions.

4 PINE operating principle

The PINE instrument can either be used in an individual operator controlled mode for laboratory ice nucleation experiments and measurements, or in a fully automated mode for long-term field measurements and observations of INPs. The instrument's settings during a laboratory or field campaign and the data storage systems of PINE are organized in a well-defined sequence of operations and runs. All data and metadata are saved automatically using the LabVIEW program.

An operation is defined as a specific series of runs, which can be, for example, performed at one temperature, and during a specific sampling time. Each run is composed of a sequence of three modes called "flush", "expansion", and "refill". The flow settings of PINE in these three run modes are illustrated in Fig. 2. In the flush mode (Fig. 2a), the sample flow is passed through the cloud chamber to fill it with the aerosol under investigation. This can either be ambient air at a field station where PINE measures INP concentrations, or an aerosol generated in a laboratory for ice nucleation studies. For PINE-1A and PINE-c, the sample flow rate is limited to about $4\,\mathrm{L\,min^{-1}}$ (see Sect. 4). In the flush mode, the sample flow can also be passed through a aerosol filter for background, particle-free measurements.

In the expansion mode (Fig. 2b), the sample flow is kept constant, but switched to a bypass line around the cloud chamber. At the same time, a valve at the chamber inlet is closed and the OPC flow rate is set to a value between 2 and $5\,\mathrm{L\,min^{-1}}$, such that the pressure in the cloud chamber is lowered at a constant rate and to a pre-defined minimum pressure. This forced gas expansion in the cloud chamber causes an adiabatic cooling and thereby an increase of the relative humidity. When the relative humidity exceeds ice or water saturation, the aerosol particles in the cloud chamber, which were sampled during the flush mode, are then activated to form ~~ice crystals~~ liwuid cloud droplets and/or ~~liquid cloud droplets~~ice crystals, depending on the temperature and the type of aerosols. ~~The number of ice crystals is measured with the~~ Both droplets and ice crystals are measured with an OPC downstream of the chamber~~, and equals the number of INPs in the same sampling volume~~. Ice crystals are distinguished from droplets by their larger optical size, as discussed later in this section. The expansion mode flow rate F_{em} is limited to 2 and $5\,\mathrm{L\,min^{-1}}$, because both the welas 2500 and fidas-pine OPCs can only be operated to a maximum sample flow rate of $5\,\mathrm{L\,min^{-1}}$. Smaller flow rates can cause cloud droplet evaporation or ice crystal sublimation in the tube connection between the cloud chamber and the OPC. The end pressure is typically 200 to $300\,\mathrm{hPa}$ lower than the start pressure that is given by the pressure of the aerosol sampled during the flush mode.

The refill mode (Fig. 2c) is the final run mode and is carried out to safely re-pressurize the PINE chamber to the start pressure. Once this pressure is reached, the sample flow is immediately switched back to pass the cloud chamber, starting the next run with the same series of flush, expansion and refill modes. A full run takes about 4 to 6 minutes, depending on the flush time, the pump flow rate during the expansion mode and the end pressure. The higher the sample flow rate, the faster the air

in the cloud chamber is replenished and renewed for the next run, and the shorter the flush time can be chosen. The lower the minimum pressure during expansion, the longer the refill mode time.

Figures 3 to 6 show results from a PINE-1A operation on March 25, 2018 during the HyICE field campaign, which includes a series of 59 identical runs. Each run took about 6 minutes, so the whole operation lasted almost 6 hours. During this time, the ambient total aerosol concentration varied between about 900 and 2300 cm^{-3}, with the majority of particles smaller than 0.5 µm in diameter, and PINE-1A sampled ambient air at a flow rate of 3 $\mathrm{L\,min}^{-1}$. The flush time was set to 4 minutes. Each expansion was started at a wall temperature of about $-26\,^{\circ}\mathrm{C}$ with pump flow rate of 4 $\mathrm{L\,min}^{-1}$, and took about 40 seconds. An example of one these 59 runs is depicted in Fig. 3, which shows the end of the flush mode, the expansion mode and the refill mode. The data time series are plotted as a function of the time in seconds relative to the start of the expansion mode. A temperature and pressure decrease of about $6\,^{\circ}\mathrm{C}$ and 300 hPa, respectively, is observed during the expansion (Fig. 3a). Here, only the data from the lowest of the three gas temperature sensors (see Fig. A4) is plotted, which reaches a minimum value of about $-32\,^{\circ}\mathrm{C}$ at the end of the expansion after about 40 seconds.

The relative humidity is not directly measured in the PINE cloud chamber, but can be calculated from the change of the temperature dependent water saturation pressure, assuming ice saturated conditions at the start of the expansion and omitting water vapour sources and sinks during the expansion. ~~As mentioned above, we assumed ice saturated conditions~~ This assumption is reasonable, because the frost point temperature of the air sampled into the chamber was slightly higher than the average wall temperature. The excess water vapor quickly condenses to the cold chamber walls, so that the water vapor partial pressure at the start of expansion equals the ice saturation pressure calculated as function of the wall temperature at start of expansion (~~$p_{w,0} = p_{sat,ice}(T_{g,0})$~~ $p_{w,0} = p_{sat,ice}(T_{g,0})$), and the corresponding saturation ~~ration~~ ratio with respect to liquid water is $S_w = 0.79$ at the same start temperature $T_{g,0} = -26\,^{\circ}\mathrm{C}$. During the expansion mode, the liquid water saturation ratio was then calculated as

$$S_w = \frac{p_w}{p_{sat,liq}(T_g)} \tag{3}$$

with

$$p_w = p_{w,0}\frac{p}{p_0} \tag{4}$$

where p_0 is the pressure at start of expansion and p the pressure during the expansion. It can be seen that after about 10 seconds, the ~~so~~ calculated S_w exceeds 1 (Fig. 3b). Note that S will in reality be limited by the growth of cloud droplets, but that conditions of $S > 1$ indicate conditions where a liquid cloud could form. This roughly corresponds with the start of cloud droplet activation as shown in panel (c) of Fig. 3, shown by the sudden occurrence of a large number of particles with diameters up to 10 µm. This panel shows each single particle detected by the OPC plotted as a single blue dot at the time of occurrence and with its measured optical diameter. With ongoing pressure reduction and related cooling, a small number of particles is detected at larger optical size, with diameters larger ~~as~~ than the dense "cloud" of liquid droplets (Fig. 3c). Those particles are identified as ice crystals formed by immersion freezing of only a minor droplet fraction. The expansion mode stops after about 40 seconds and the chamber is refilled to ambient pressure within about 1 minute. The next run is started with the flush mode,

filling the cloud chamber again with ambient aerosol particles for the next expansion run. Refilling causes compression of the chamber air and related warming (see upper panel of Fig. 3). This also leads to the evaporation of the droplets and ice crystals after some time, however, the abrupt stop of particle recording is related to the fact that the pump flow rate through the OPC is stopped at the end of expansion, so that only a few particles are moving through the OPC detection volume during the refill mode.

For the same PINE-1A operation during the HyICE field campaign, there is little run-to-run variation for the total OPC counts per second of run time (Fig. 4). This means that PINE is able to reproduce ~~aerosol CCN activation and super-cooled droplet formation~~ the formation of the supercooled droplet cloud in repeated runs at constant sampling and operation conditions, which provides a good basis for conducting series of immersion mode INP and freezing measurements at high precision. The small grey dots in this figure show the OPC count rates of individual runs, the bigger black circle the mean over all 59 runs of this operation. The variation can partly be explained by the natural variability of the ambient aerosol concentration which also causes a variation of the droplet number concentration. As mentioned above, the aerosol number concentration varied by about a factor of two between 900 and 2300 cm^{-3}.

Not only cloud droplets but also ice crystals were detected during the same operation as shown by the occurrence of particles larger than $\sim 10\,\mu\mathrm{m}$ (Fig. 3c). The whole size distribution of both cloud droplet and ice crystals (Fig. 5) reveals that there is only little variation from run to run, at least for the droplet mode with maximum diameters of $\sim 10\,\mu\mathrm{m}$. Larger particles are identified as ice crystals, and can be distinguished from the droplets by setting a size threshold close to the end (the "right edge") of the sharp droplet mode. The use of a simple size threshold to distinguish between ice crystals and droplets is supported by the fact that the sideward scattering geometry of both the welas and fidas sensors detect a-spherical particles with a much larger scattering intensity than spherical particles of the same volume and refractive index. Järvinen et al. (2014) determined an average oversizing factor of 2.2 for the welas sensor. For individual ice crystals, this factor can be much larger depending on their size, shape and orientation in the OPC detection volume.

Based on Eq. 1, the immersion mode INP number concentration measured in one run of the PINE-1A system can then be calculated by dividing the total number ΔN_{ice} of ice crystals detected by the total volume ΔV_w of air passing the ODV of the welas OPC during the expansion mode after the formation of the supercooled liquid cloud

$$n_{INP,w} = \frac{\Delta N_{ice}}{\Delta V_w} = \frac{\Delta N_{ice}}{F_w \Delta t_{em}} \tag{5}$$

where F_w is the volumetric flow rate through the optical detection volume of the welas sensor, and Δt_{em} the duration of the expansion mode from the start of liquid cloud formation (see also Sect. 3 and Eq. 2). For the welas 2500 sensor, ΔV_w is about 10 % of the total volume ΔV_{em} passing the OPC during the same time. For the PINE-c system equipped with a fidas-pine (fp) sensor analysing the total pump flow $F_{em} = \Delta V_{em}/\Delta t_{em}$ for particles (see Sect. 3), the INP number concentration results from

$$n_{INP,fp} = \frac{\Delta N_{ice}}{\Delta V_{em}} = \frac{\Delta N_{ice}}{F_{em} \Delta t_{em}} \tag{6}$$

Examples from PINE-1A at higher temperatures without ice crystal formation prove that this "right edge" of the activated droplet size distribution is indeed rather sharp in typical expansion runs (Fig. A5). For data interpretation, the size threshold

to distinguish between droplets and ice crystals can be set manually, however it varies with operation temperature and droplet number concentration. ~~Therefore, Adams et al. (in prep.) developed~~ Ongoing activities for improving the operation and data analysis tools for PINE also focus on developing an automated procedure for setting this threshold. ~~Setting this size threshold and counting all larger particles~~ Counting all particles larger than this threshold as ice crystals is a simple straightforward procedure, but neglects smaller ice particles which may also be present in the overlapping size range with the droplets and may cause an underestimation of the ice crystal number concentration. Therefore, the PINE instrument was also operated next to the AIDA cloud chamber for homogeneous droplet freezing and immersion freezing experiments in order to identify and quantify potential systematic uncertainties and biases (see Sect. 5).

In addition to detecting the accurate number of ice crystals, the quality of ice nucleation and INP measurements also depends on measuring the precise temperature, at which the ice crystals are actually nucleated, either homogeneously or at the surface of an INP. The variability of the gas temperature in the PINE cloud chamber during 59 expansions is illustrated in Fig. 6. Here, all ice crystals detected during the 59 expansions are plotted for the relative time after start of the run in which they were measured, and the respective gas temperature measured with three sensors located in the lower (blue), the middle (green) and the upper (red) part of the chamber (see Fig. A4). First of all, one can see that the number of ice crystals, and thereby also the number of immersion freezing INPs that caused the ice formation in these expansions, increases with decreasing temperature, which reflects the temperature dependent INP number concentration in ambient air. For individual sensors, the temperature variability from run to run is less than about $0.5\,^{\circ}\mathrm{C}$, clearly underlining that PINE is able to detect the temperature dependent ice crystal formation from run to run at well controlled conditions. However, there is an increasing deviation of the temperature readings at the different locations in the PINE cloud chamber, with the lowest temperature measured at the bottom and the largest at the top. This inhomogeneity of the temperature across the chamber arises from the fact that there is an increasing temperature difference between the expanding gas and the almost constant wall temperature. This causes an increasing heat flux into the chamber volume and by that an increasing temperature distortion and deviation from the adiabatic temperature profile. The hereby formed warm air tends to be collected in the top part of the chamber. The related temperature variability inside the cloud chamber impacts the temperature uncertainty for the INP and ice nucleation detection. However, it can be assumed that most of the ice crystals detected in the PINE expansion mode are formed at the lowest temperature in the bottom part of the chamber, where all the air flowing to the OPC passes through. Since ice nucleation is a strong function of temperature, it is a good first order approximation to assume the coldest temperature in the chamber to guide the ice nucleation. This assumption will be solidified by the results of experiments presented and discussed in the following section.

An important part of PINE operations are the background runs, during which the sampled air is guided through a filter, while the operation runs are ongoing. After typically 5 to 10 runs, the chamber becomes particle free, as such any remaining particle counts indicate the presence of frost formation on the walls or a leak in the chamber or pipework. A typical background measurement, where almost no particles are present after 5 cycles, is presented in Fig. A3. Regular background run series are performed with PINE at least during longer measurement phases at low temperatures. A frost-free chamber is a prerequisit for operating PINE with highest sensitivity. In case of zero background conditions, the detection limit for INP number concentrations can be calculated by dividing the minimum number of ice crystals detected in a certain volume of air. In one expansion

with PINE-1A and PINE-c analysing about 0.2 and 2 liters of air per run, respectively, the resulting one count detection threshold is $5\,\mathrm{L}^{-1}$ and $0.5\,\mathrm{L}^{-1}$, respectively (see also Tab. 1). Note that the detection limit of PINE-1A is a factor of 10 lower because only about 10 % of the pump flow during the expansion is analysed, whereas the OPC of PINE-c detects all ice crystals in the pump flow. If 10 consecutive runs are conducted and summed up in one hour, assuming the total run time is set to 6 min, about 10 times more volume of air is analysed, and all ice crystals detected can be summed-up, so that the INP detection limits are reduced by a factor of 10 to $0.5\,\mathrm{L}^{-1}$ and $0.05\,\mathrm{L}^{-1}$ for PINE-1A and PINE-c, respectively, with a time resolution of one hour. When summing-up over a whole day of subsequent runs, the detection limits are further reduced to $0.02\,\mathrm{L}^{-1}$ and $0.002\,\mathrm{L}^{-1}$, respectively.

5 Laboratory tests of the prototype version PINE-1A

During several test series, immersion freezing and cloud droplet freezing measurements with PINE-1A were compared to the AIDA cloud camber results. For these intercomparison studies, PINE-1A sampled aerosols directly from the AIDA chamber and was operated at similar wall temperatures as the AIDA cloud chamber. By that, the cloud expansion runs covered a similar temperature range, and as such allowed the intercomparison of temperature-dependent freezing rates or INP concentrations. Homogeneous freezing of supercooled water droplets is known from classical nucleation theory and from literature results (Pruppacher and Klett, 2010; Koop and Murray, 2016) to occur at temperatures between about −35 °C and −37 °C. Figure 7 shows the ~~results for homogeneous freezing of supercooled water droplets , which are known to start freezing in a typical AIDA cloud expansion run at about −36 °C, in good agreement with other experimental results and formulations for classical nucleation theory (Benz et al., 2005)~~freezing of water droplets to be measured with PINE-1A in the expected temperature range. As in the experiments by Benz et al. (2005), aqueous sulphuric acid particles were first added to the AIDA chamber. Then, the aerosol particles with a diameter around 0.8 µm and a number concentration of about $200\,\mathrm{cm}^{-3}$ were sampled into the PINE-1A chamber for its homogeneous freezing experiments, followed by an AIDA cloud expansion experiment with the same aerosol. Figure 7 shows ~~good agreement for~~ the onset temperature of the homogeneous freezing in PINE-1A ~~and the~~ to agree with the results of the AIDA cloud expansion experiment. The PINE-1A data is plotted as a function of the temperature measured with the bottom temperature sensor, which always measures the lowest temperature during a run (see Fig. 6). This result underlines the assumption, that the ice formation measured with PINE is mainly controlled by the minimum temperature in the cloud chamber.

PINE-1A was also operated next to the AIDA cloud chamber during the EXTRA18 campaign in February 2018. This campaign was mainly organized to test and calibrate the newly constructed PINE-1A in preparation to ~~the~~ a first field campaign~~, which will be described in more detail in a follow-up paper~~. During this campaign, PINE-1A sampled aerosol particles directly from the AIDA chamber again, and measured their ice nucleation activity in the same temperature range covered by AIDA cloud expansion runs with the same aerosols. Arizona test dust (ATD) and illite NX aerosols where used during this campaign. These aerosols are well studied for their ice nucleation activities and were also used in previous intercomparison experiments for INP instruments (DeMott et al., 2011, 2018; Hiranuma et al., 2015). We used the same aerosol sources as Steinke et al.

(2015) for ATD and Hiranuma et al. (2015) for illite NX, and the methods for generating and characterizing these aerosols are described in these papers.

The supercooling or minimum temperature reached in a PINE expansion can be controlled by two parameters, the pump flow rate and the end pressure. This allows for a quick scan through a certain temperature range of ice nucleation activity. Both higher pump flow rates and lower end pressure cause a larger supercooling of the air in the cloud chamber, means a lower minimum temperature at the end of expansion. An example is shown in Fig. 8. In this operation, PINE-1A sampled ATD aerosol directly from the AIDA chamber and measured the number fraction f_{ice} of ice-active ATD particles in a series of runs starting from a temperature of about $-18\,^{\circ}\mathrm{C}$. The expansion flow rate was $5\,\mathrm{L\,min^{-1}}$ in all runs, but the end pressure was stepwise reduced every ~~5~~ 3 runs from about $800\,\mathrm{hPa}$ at the beginning to about $500\,\mathrm{hPa}$ at the end of this operation (see panel (a) of Fig. 8). This caused a stepwise decrease of the minimum gas temperature in the cloud chamber, as also shown in panel (a). The welas 2500 single particle data (Fig. 8, panel (b)) indicates an increasing amount of ice formation with decreasing minimum temperature. This stepwise increase in the number concentration of ice crystals or INPs is shown in panel (c) of Fig. 8, which depicts the time series of the ice crystal number concentrations measured at the end of each expansion.

Figure 9 depicts the ice crystal number fraction calculated with Eq. 5 devided by the aerosol number concentration for each individual run as function of gas temperature measured with the sensor in the bottom of the PINE-1A cloud chamber. The measured number concentration of ice crystals equals the number concentration of ice-active ATD particles measured in an AIDA cloud chamber experiment with the same aerosol (Fig. 9). For the PINE measurements, we estimate a temperature uncertainty of $\pm 1\,^{\circ}\mathrm{C}$, mainly caused by the inhomogeneous temperature distribution in the PINE cloud chamber during the expansion run (see Fig. 6). The temperature uncertainty during AIDA cloud expansion experiments is ~~$\pm 1\,^{\circ}\mathrm{C}$~~ $\pm 0.3\,^{\circ}\mathrm{C}$. The estimated uncertainty for the ice number concentration is $\pm 20\,\%$ for both PINE and AIDA, mainly due to the uncertainty in the dimension of the ODV of the welas sensor and the measured transect time of particles passing the ODV (see Eq. 1).

The same measurements as for ATD were also performed with illite NX aerosol (Figs. 10 and 11), but with both AIDA and PINE-1A starting their cloud expansions at a slightly lower temperature of about $-22\,^{\circ}\mathrm{C}$ because of the somewhat lower ice nucleation activity of illite NX compared to ATD. Within the given uncertainty ranges, the PINE-1A data is in excellent agreement with the AIDA data for both ATD (Figs. 8 and 9) and illite NX (Figs. 10 and 11). This also underlines the assumption, that the ice formation in PINE is mainly controlled by the coldest temperature in the bottom part of the chamber and that the number concentration of ice crystals, and by that the number concentration of ice-active aerosol particles in laboratory experiments and of INPs during field measurements can ~~correctly~~ be calculated with Eqs. 5 and 6 ~~.~~ within the above given uncertainty estimates for the number concentration and the nucleation temperature. We should note here, that these uncertainty limits are so far only justified by comparison of PINE with AIDA results. Further systematic uncertainties like the loss of large ice crystals between the PINE cloud chamber and OPC, size range overlap of small ice crystals with large aerosol particles not activated to droplets, or the sampling efficiency of large aerosol particles into the cloud chamber may have to be considered for calculating the overall accuracy of INP measurements.

A more comprehensive uncertainty assessment for PINE may result from recent intercomparison studies with other methods and instruments and ongoing long-term operation in field campaigns. For long-term measurements, another important parameter

is the precision for repeated measurements at the same sampling and operating conditions. In a recent test experiment at the AIDA cloud chamber, the new commercial PINE-04-01instrument sampled a mixed aerosol (ammonium sulfate and natural dust) for more than 8 hours from the AIDA chamber (Figure A6). During this experiment, a mean ice-active particle number fraction of 1.8×10^{-4} was measured with a standard deviation of 2.1×10^{-5}, which corresponds to a relative uncertainty of about $12\,\%$. During this operation, an average number Nice of about 90 ice crystals was measured during one run. Therefore, the relative uncertainty from counting statistics can be calculated as $\sqrt{N_{ice}}/N_{ice} = 10.5\,\%$, which is close to the relative standard deviation of the run by run data from the mean value. For measurements with a much lower number of ice crystals detected in one run or a consecutive number of runs, the measurement uncertainty from counting statistics can be much larger. Next versions of the PINE analysis software tools will also include uncertainty analysis for low counting cases close to the PINE detection limit.

6 Field measurements with PINE-c

We performed ground-based INP measurements with PINE-c at the SGP observatory in Oklahoma, where long-term measurements provide statistical context (www.arm.gov/capabilities/observatories/sgp). ~~The ARM SGP field campaign, the so-called~~ During the ExINP-SGP (www.arm.gov/research/campaigns/sgp2019exinp), ~~was held from October 1 to November 14, 2019. Briefly,~~ we have successfully operated PINE-c ~~at the SGP site~~ (Fig. A7) via remote control for INP concentration measurements on a continuous basis for 45 consecutive days. During the ~~ExINP-SGP~~ entire campaign, PINE-c was operated with an expansion mode time of 60 to 90 seconds, resulting in an averaged sampled gas volume of $3.7 \pm 0.6\,\mathrm{L}$. This resulted in the minimum detectable INP concentration of about 0.2 to $0.3\,\mathrm{L}^{-1}$ for a single run of approximately 8 minutes duration. PINE-c was set to automated wall temperature control with ramping back and forth between $-5\,^{\circ}\mathrm{C}$ and $-35\,^{\circ}\mathrm{C}$ every 90 minutes, without any substantial technical issues during the whole campaign period.

Shown in Fig. 12 is the overall summary of compiled $n_{ice}(T)$ spectra measured during the ExINP-SGP campaign. Individual data points (black dots) represent 6 hours time-averaged data with a temperature interval of $1\,^{\circ}\mathrm{C}$. Here, we display the PINE-c n_{ice} data for the temperature segment of $-10\,^{\circ}\mathrm{C} \geq T \geq -30\,^{\circ}\mathrm{C}$. This temperature range ~~virtually~~ represents the PINE-c condition, where ice nucleation through immersion freezing was ~~warranted below local ambient dew point temperature.~~ possible below the frost point temperature of the sample air after passing the membrane diffusion dryers operated at maximum drying efficiency. For measurements at higher temperature, the drying efficiency has to be reduced, in order to increase the dew point of the sampled air and to exceed water saturation during the expansion mode at higher temperature. Next versions of the PINE control program will include this option for operation at higher temperature. Any further scientific discussions regarding PINE-c operations and observations, in combination with other INP and aerosol measurements during the ExINP-SGP campaign~~(e.g., deconvolution of nucleation modes, relationship between measured microphysics and local dynamics/thermodynamics, potential artefacts etc.)~~ , are beyond the scope of our current study, and will be followed up in future publications.

7 Summary and conclusions

We present a new instrument called PINE (Portable Ice Nucleation Experiment) for laboratory studies of ice nucleation and field measurement of ice-nucleating particles (INPs). Inspired by the large AIDA cloud chamber (Möhler et al., 2003, 2005), the PINE instrument also uses the principle of expansion to expose aerosols from different sources to cloud-relevant conditions. By that, the sampled aerosol particles are activated to form both supercooled water droplets and ice crystals, which are detected with an optical particle counter (OPC). However, with a volume of only about $10\,\mathrm{L}$, PINE is much smaller than the AIDA cloud chamber. The instrument is sensitive to detect ice formation and INPs in the immersion freezing, pore condensation freezing and deposition nucleation modes in a wide temperture range from $-10\,^{\circ}\mathrm{C}$ to $-65\,^{\circ}\mathrm{C}$. Equipped with a LabVIEW control system, PINE can be operated autonomously over longer time periods and is therefore also suitable for INP monitoring at atmospheric field sites and observatories.

The operation of PINE is organized in a well defined sequence of runs. Each run is composed of three modes called "flush", "expansion", and "refill". During the flush mode, the aerosol under investigation is sampled into the pre-cooled cloud chamber. The sampled aerosol particles are activated as supercooled cloud droplets and ice crystals during the expansion mode, depending on the pre-set wall temperature, the expansion rate and the minimum pressure reached at the end of the expansion mode. Droplets and ice crystals are detected with the OPC, and the fraction of ice-active aerosol particles or the number concentration of INPs in the sample can be calculated from the total number of ice crystals detected during the expansion mode and the volume of air that has passed the detection volume of the OPC. During the refill mode, the cloud chamber is just refilled to the ambient pressure to immediately start the next run. In the current PINE versions, one run takes about 4 to 6 minutes, which defines the ~~largest~~ highest time resolution that can be achieved with PINE when e.g. measuring time series of atmospheric INP concentration.

Here we presented and discussed the construction and performance of both the prototype version of the new instrument, called PINE-1A, and the more advanced and commercially available version PINE-c (Bilfinger Noell GmbH). PINE-1A has a stainless steel cloud chamber of $7\,\mathrm{L}$ volume that is cooled with a chiller to measure immersion freezing INPs at temperatures between about $-10\,^{\circ}\mathrm{C}$ to $-40\,^{\circ}\mathrm{C}$. This instrument was tested and characterized in a series of laboratory measurements in comparison with the benchmarked AIDA chamber. PINE-1A results for both homogeneous freezing of cloud water droplets and immersion freezing of ATD and illite NX aerosols were in excellent agreement with AIDA results. The first operation of PINE-1A during the HyICE field campaign in Hyytiälä, Finland, also demonstrated that there is only little variability of the measured droplet and ice size distribution from run to run.The INP concentration is measured with a high precision and repeatability. The temperature uncertainty is estimated to be about $\pm 1\,^{\circ}\mathrm{C}$, mainly influenced by an increasing temperature inhomogeneity during the expansion mode. The field operation also showed that the welas 2000 OPC can well distinguish between ice crystals and droplets by setting an optical size threshold, and that PINE-1A was operated over longer time periods at almost zero background conditions without any detectable frost formation on the cold cloud chamber walls. A follow-up stuy will present more results from the HyICE field activity and will discuss in more detail the performance of PINE-1A during long-term field operation.

The advanced instrument version PINE-c has a somewhat larger cloud chamber of 10 L volume which is made of thin-walled aluminium and locted in an evacuated chamber for thermal insulation. The cooling system is based on a Stirling cooler and allows cooling the chamber to temperatures as low as −60 °C. PINE-c was successfully operated for the first time during a field campaign conducted at the Atmospheric Radiation Measurement (ARM) Southern Great Plains (SGP) observatory in Oklahoma, USA~~, from October 1st to November 14th, 2019.~~ During this field campaign, PINE was continuously operated for 45 days in a fully automated and semi-autonomous way at ~~high~~ time resolution of about 8 minutes with continuous wall temperature scans between −5 and −35 °C. The overall INP concentrations ranged from about ~~0.2 L^{-1}~~ 0.02 L^{-1} at −10 °C to about 200 L^{-1} at −30 °C. More results from this field activity will be presented and discussed in a follow-up study.

One of the unique features of PINE, in contrast to flow diffusion or mixing devices, is its operation under dry and frost-free wall conditions. Therefore, long-term continuous operation over days and weeks can be performed without the occurrence of increasing background from frost artefacts. This is achieved by drying the sampled aerosol to a frost point temperature close to the minimum wall temperature. This was proven in a series of measurements during a field campaign in Hyytiälä, Finland. The ~~PINE-1A results form this campaign will be discussed in more detail in a follow-up publication. The~~ sampled air needs to be humidified when its frost point temperature is clearly below the lowest chamber wall temperature. This may only be the case when sampling from extremely cold or dry environments, like polar regions or desert areas, or when sampling laboratory aerosols generated in extremely dry air. In most surface-based atmospheric sampling locations, the sample includes sufficient humidity and needs to be dried before entering the PINE chamber. Future versions of PINE may therefore also include an optional air humidification system in addition to the diffusion dryers. In addition, the newest version PINE-c is operated with a novel and liquid-free cooling system, which makes it suitable to be even operated autonomously at remote measurement sites.

Given the dearth of atmospheric INP measurements with which to challenge and inform our aerosol, cloud and climate models, an instrument, such as PINE, capable of making measurements on a routine and autonomous basis is needed. The development of PINE is timely, since INP control the radiative properties of clouds around the globe and are first order for defining cloud feedbacks (Vergara-Temprado 2018; Tan 2016). We anticipate that PINE will become a standard autonomous instrument at atmospheric observatories around the globe as well as a versatile laboratory and research tool.

Appendix A: Membrane diffusion dryer

The PINE instruments are equipped with a dual membrane dryer system (Fig. A1) to reduce the humidity of the aerosol sampled into the cold cloud chamber and by that to avoid frost formation on the cold cloud chamber walls. The drying efficiency of the nafion tube was measured as a function of the pressure difference Δp between the sample flow and the counter flow and also as a function of the volumetric sample flow rate. The drying efficiency is plotted in Fig. A2 as the difference ΔT_d of the sample air dew point temperatures measured with a chilled mirror dew point sensor (MBW type 393) before and after the dryer. The measurements shown in Fig. A2 were conducted with the dew point temperature of the sample air ranging from about 6 to 7 °C. The drying efficiency is increasing with the pressure difference and decreasing with the sample flow rate. ~~High~~ A drying

efficiency with a drop in dew point temperature of more than $10\,°C$ is achieved when operating the dryers with a sample flow rate below 2 to $3\,L\,min^{-1}$ and at the maximum pressure difference of about $800\,hPa$ across the membrane.

Appendix B: Backgroud measurements

Operating PINE with high sensitivity for INP detection requires low or even zero background conditions. Therefore, the control system allows for regular background checks, where the instrument is set to flush mode and passing the sample flow through the bypass line with particle filter (via dashed line in Fig. 2a). A typical background run sequence (operation) from the HyICE field measurements with PINE-1A (Fig. A3) shows that the particle counts approach or drop to zero after about 4 to 5 runs. More details about background behaviour of PINE will be presented and discussed in a follow-up paper.

Appendix C: PINE construction and operation

Figure A4 shows the construction of the PINE-1A cloud chamber with the location of the three gas temperature sensors. For PINE measurements, a size threshold is used in order to distinguish larger ice crystals from smaller liquid water droplets in the OPC single particle data (see discussion in Sects. 3 and 4). In the absense of INPs, the droplet size distribution measured with the OPC has a sharp edge to larger particle diameters (Fig. A5), which is favorable for setting the size threshold. Fig. A6 shows a recent measurement with the new commercial PINE-04-01 when sampling a mixed aerosol (ammonium sulfate and natural dust) for more than 8 hours from the AIDA chamber. This figure well demonstrates the run by run stability and repeatability of PINE measurements. In this experiment we did not expect a constant but a steadily decreasing INP concentration (panel a), because of to the steady decrease of the aerosol concentration (panel b) according to aerosol loss processes to the chamber walls. The ice-active particle number fraction (panel c) remained constant with a mean value of 1.8×10^{-4} and a standard deviation of 2.1×10^{-5}, which demonstrates the precision of PINE INP measurements under these conditions. Figure A7 shows the first version of the PINE-c instrument in operation at the ARM SGP field campaign ExINP-SGP (www.arm.gov/research/campaigns/sgp2019exinp).

Code availability. TEXT

Data availability. Data shown in 3-12, A2, A3, A5, and A5 is available in KITOpenData under https://dx.doi.org/10.5445/IR/1000122157.

Code and data availability. TEXT

Sample availability. TEXT

Video supplement. TEXT

Author contributions. O.M. wrote this manuscript; O.M. and B.J.M lead the PINE development and coordinated the laboratory and field activities; M.A., L.L., F.V. conducted the laboratory experiments and field activities during the HyICE field campaign in Hyytiälä, Finland, and analysed the respective measurements; J.N. and R.F. developed the control and data analysis software for PINE and contributed to the analysis and interpretation of the measurements; C.B., T.P., A.H., and M.W. contributed to the engineering and construction of the PINE instrument; H.S.K.V and N.H. conducted the measurements with PINE-c and analysed the data; all co-authors participated in the data evaluation and interpretation and contributed in drafting this manuscript

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. Configuration and operational parameters of PINE prototype version 1A as well as the currently available commercial version PINE-c.

	PINE-1A	PINE-c
Chamber type	Stainless steel, single walled	Aluminium, thin-walled
Thermal insulation	2 cm thick armaflex layer	Vacuum chamber
Chamber length	75 cm	57 cm
Chamber diameter	15 cm	18 cm
Chamber volume	7 L	10 L
Cooling system	Chiller Lauda (RP855)	Stirling (Thales, LPT9310)
Wall temperature range	0 °C to −33 °C	0 °C to −60 °C
Measurement temperature range	−10 °C to −40 °C	−10 °C to −65 °C
Temperature uncertainty	±1 °C	±1 °C
Wall cooling rates	0.3 °C min^{-1}	0.6 °C min^{-1}
Wall heating rates	0.3 °C min^{-1}	0.6 °C min^{-1}
Particle detector	welas 2500	fidas-pine
Inlet dryer	Permapure, MD-700-24S-1	Permapure, MD-700-24S-1
Detection limit at 6 minute time resolution (single run)	5 L^{-1}	0.5 L^{-1}
Detection limit at 1 hour time resolution (10 runs)	0.5 L^{-1}	0.05 L^{-1}
Detection limit at 24 hour time resolution (240 runs)	0.02 L^{-1}	0.002 L^{-1}

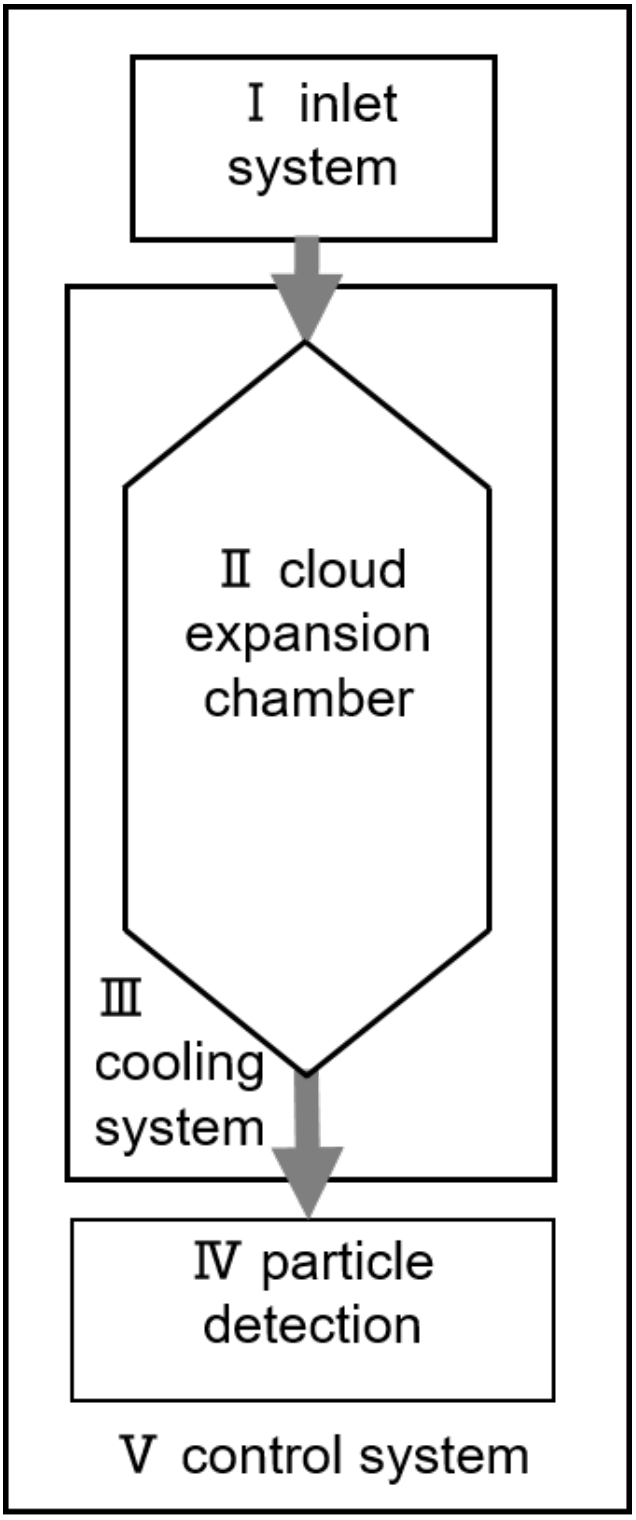

Figure 1. Scheme of a PINE instrument with its five basic components.

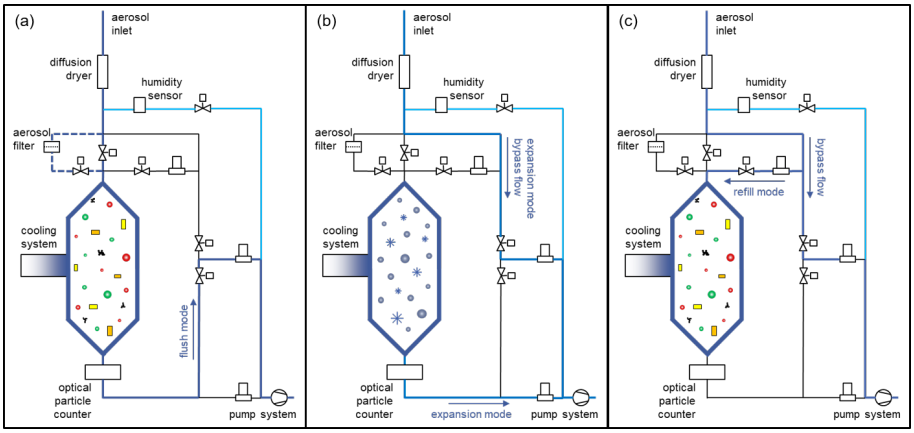

Figure 2. Schematic setup of the PINE-1A. The three figures show the same instrument, but in the different run modes (a) flush, (b) expansion, and (c) refill. The thick blue lines indicate which parts of the flow setup are active in the respective modes. The sampling gas flow through the humidity sensor (light blue line) is active all the time in a bypass line to the sampling pump. A background measurement can be done by passing the sample flow over an aerosol filter (dashed line, panel a). In the flush mode (a), aerosol particles are sampled (coloured various symbols), and activate into cloud droplets and ice crystals during the expansion mode (panel b, blue circles and stars, respectively). During the refill mode, aerosol particles are entering the chamber again (panel c, coloured various symbols).

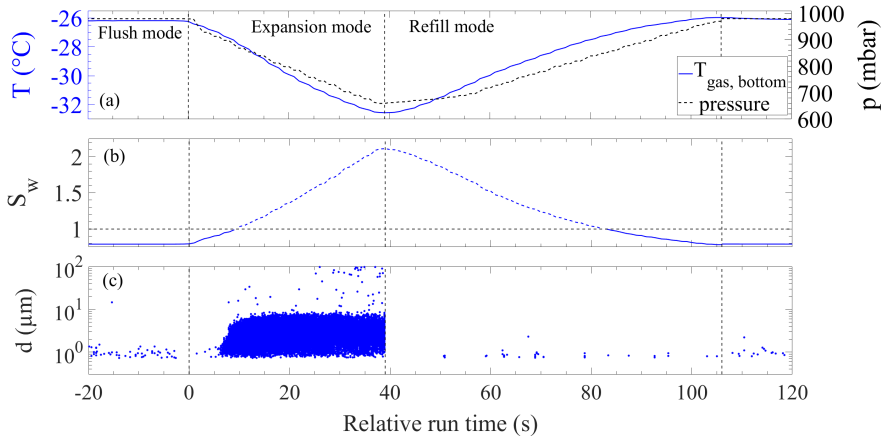

Figure 3. A typical run of PINE-1A showing both cloud droplet formation and ice formation during the cloud expansion mode. Upper panel: ~~Temperature~~ Lowest gas temperature (T; blue line) and pressure (p; black line) measured in the chamber. Middle panel: ~~Liquid~~ Calculated liquid water saturation ratio (~~Sw~~S_w). Lower panel: Optical particle diameter (d) detected in the OPC. This panel shows each single particle detected by the OPC plotted as a single blue dot at the time of occurrence and with its measured optical diameter.

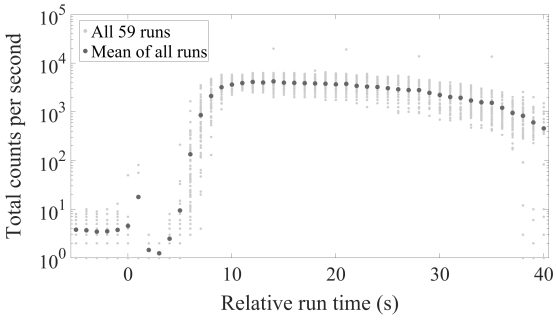

Figure 4. Total number counts measured with PINE-1A in 1 sec time intervals of 59 consecutive runs during the HyICE field campaign (operation 64 on 25th March 2018). The measured count rates are plotted as a function of time relative to the start of expansion. The small grey dots in this figure show the OPC count rates of individual runs, the bigger black circle the mean over all 59 runs of this operation. Before start of expansion, only larger aerosol particles are measured. The sharp increase after about 6 s of expansion is due to CCN activation of the aerosol particles in the chamber and the growth of droplets.

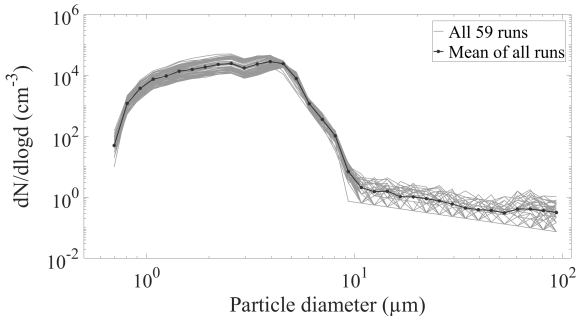

Figure 5. Particle size distribution for the same series of runs shown in Fig. 4.

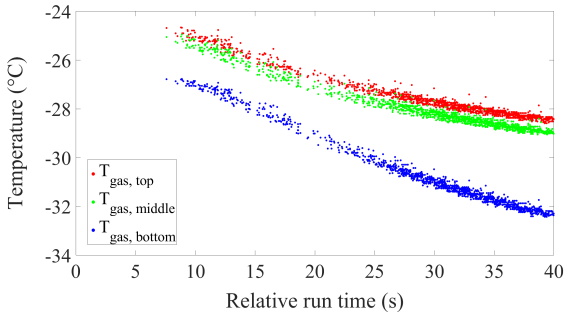

Figure 6. ~~All single~~ The data points show event temperatures of all ice crystals measured with PINE-1A during the same operation of 59 runs shown in Figs. 4 and 5. The ~~ice crystals~~ events are plotted ~~for the relative time after start~~ as a function of the relative run time they were ~~measured,~~ detected and the ~~respective~~ gas ~~temperature~~ temperatures measured at the same time with three sensors located in the lower (blue), the middle (green) and the upper (red) part of the chamber.

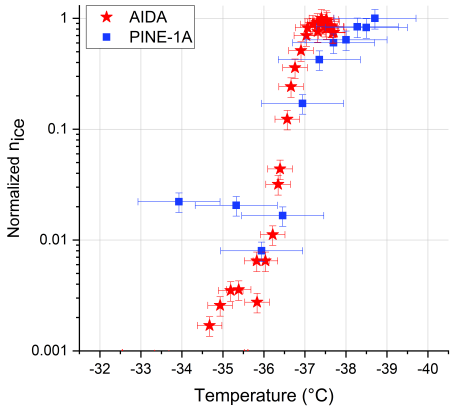

Figure 7. Homogeneous freezing of supercooled water droplets measured with PINE-1A and with AIDA during a PINE characterisation campaign in December 2018. For this measurement, the PINE-1A was equipped with a welas 2500 OPC and sampled sulphuric acid aerosol directly from the AIDA chamber. PINE-1A was operated at a wall temperature of about $-32.5\,^{\circ}\mathrm{C}$, the expansion run was done with a flow rate of ~~5 l min-1~~ $5\,\mathrm{l\,min^{-1}}$, and reached a minimum gas temperature of $-39\,^{\circ}\mathrm{C}$. The AIDA expansion was started at a temperature of about ~~-31°C~~ $-31\,^{\circ}\mathrm{C}$ and reached a minimum temperature of about $-38\,^{\circ}\mathrm{C}$.

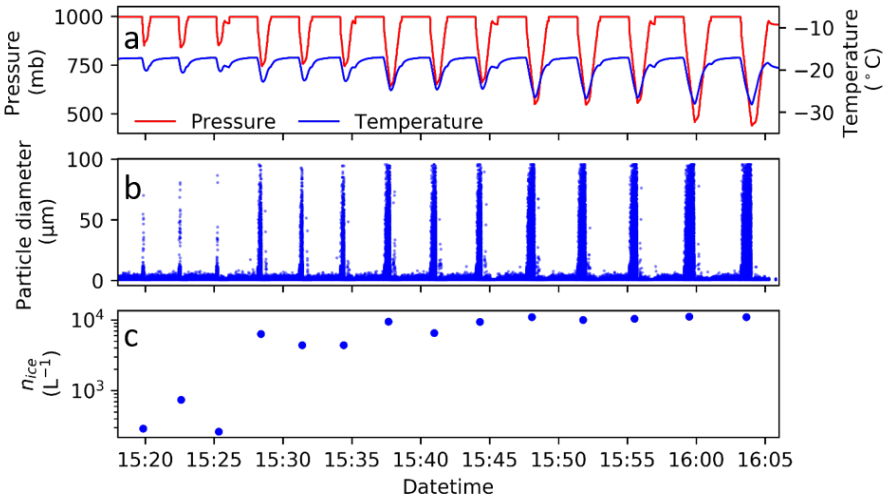

Figure 8. Repeated runs of PINE-1A sampling ATD aerosol from the AIDA cloud chamber during the EXTRA18 ~~laboratory test~~ campaign~~in preparation of the HyICE field campaign~~. The runs were started at the same temperature of about $-18\,°C$ (blue line), but the minimum expansion pressure (red line) and by that also the minimum gas temperature in the PINE cloud chamber was stepwise changed every ~~5th~~ 3^{rd} run (upper panel). Therefore, the number of ice crystals formed by immersion freezing also stepwise increased, as shown in the single particle plot from the welas 2500 OPC data (middle panel) and the ice crystal concentration measured at the end of each expansion (lower panel).

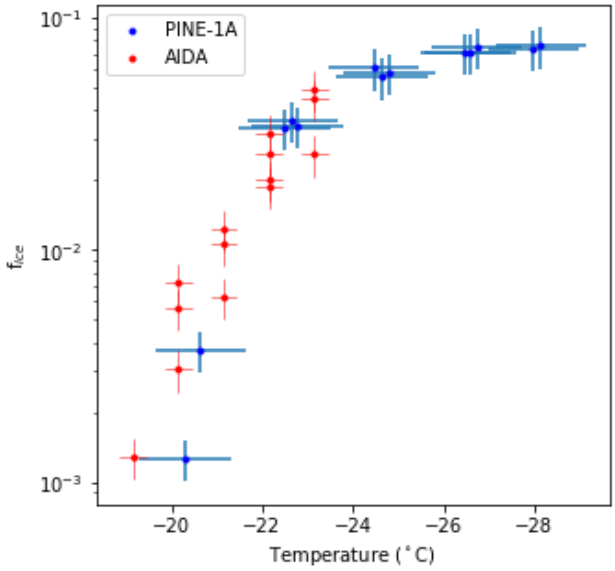

Figure 9. Ice-active particle fraction ~~fice~~ f_{ice} measured with PINE-1A for ATD as a function of temperature (see also Fig. 8), in comparison to ~~fice~~ f_{ice} measured in an AIDA cloud expansion experiment with the same aerosol, right after the PINE-1A runs were finished.

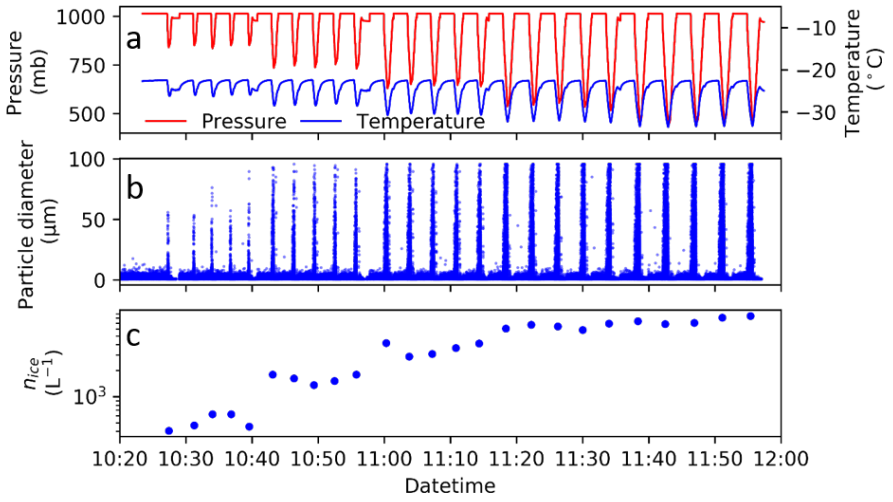

Figure 10. Same plot as shown in Fig. 8, but with PINE-1A sampling illite NX aerosol from the AIDA cloud chamber, and with a lower start temperature of about -22°C (see upper panel, blue line). As for ATD runs, the minimum expansion pressure (red line) and by that also the minimum gas temperature in the PINE cloud chamber was stepwise changed every 5th run (upper panel). Therefore, the number of ice crystals formed by immersion freezing also stepwise increased, as shown in the single particle plot from the welas OPC data (middle panel) and the ice crystal concentration measured at the end of each expansion (lower panel).

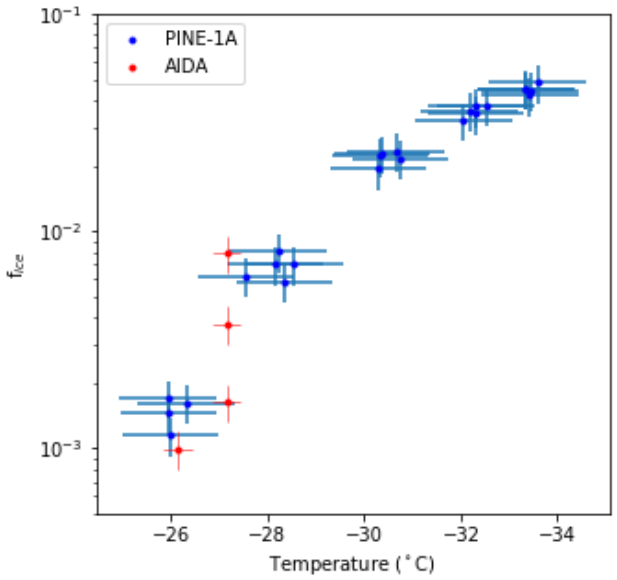

Figure 11. Ice-active particle fraction ~~fice~~ f_{ice} measured with PINE-1A (blue dots) for illite NX as a function of temperature (see also Fig. 10), in comparison to ~~fice~~ f_{ice} measured in an AIDA (red dots) cloud expansion experiment with the same aerosol, right after the PINE-1A runs were finished.

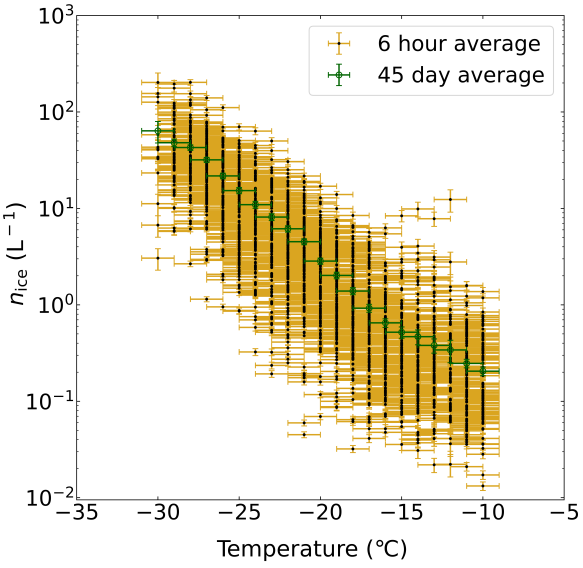

Figure 12. PINE-c INP concentration (n_{ice}) measured as function of the minimum gas temperature during ~~a U.S. Department of Energy funded~~ the ExINP-SGP campaign at the ARM-SGP site in Oklahoma. PINE-c measured continuously for 45 days from October 1st to November 14th, 2019. ~~Individual 6 hour time-averaged~~ Temperature-binned concentrations data ~~and overall temperature-binned data~~ (~~$\Delta T = 1°C$~~ $\Delta T = 1\,°\mathrm{C}$) ~~are~~ is shown ~~in~~ for 6 hour time averaged data (black markers) and 45 days averaged data (green markers). Note the temperature uncertainty of ~~$\pm 1.5°C$~~ $\pm 1\,°\mathrm{C}$ based on the homogeneous freezing temperature calibration with ammonium sulfate aerosol particles. The n_{ice} uncertainties represent relative standard errors of 6-hour averaged measurements at given temperatures. Statistical errors from low counting signals are not considered here and will be the subject of further analysis.

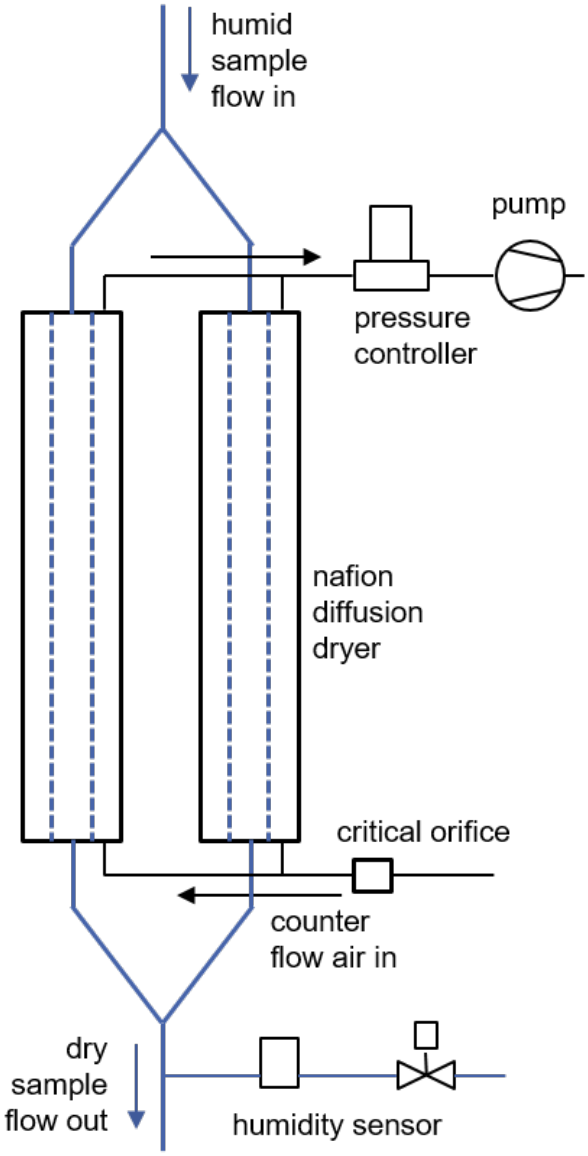

Figure A1. Schematic setup of the dual nafion dryer ~~setup~~ as part of the PINE inlet system.

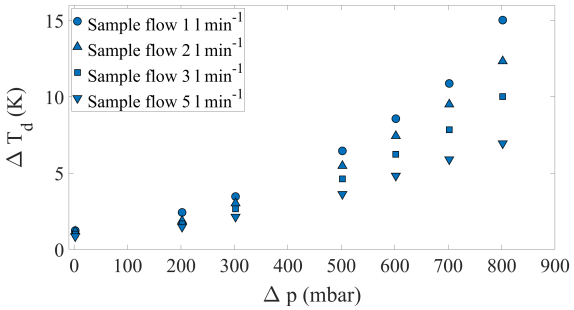

Figure A2. Drying efficiency of one nafion diffusion dryer, plotted as the difference ΔT_d of the dew point temperatures measured in the sample air before and after the nafion tube. The drying efficiency is increasing with the pressure difference Δp between the sample air and the counter flow air, and decreasing with the sample flow.

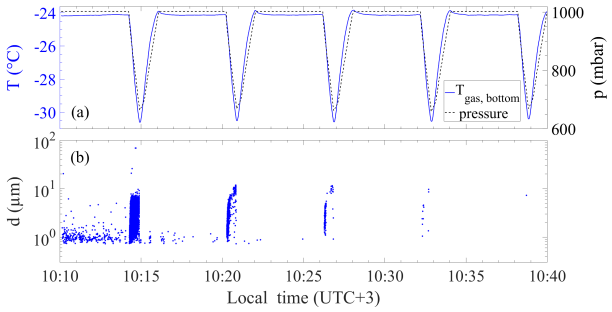

Figure A3. Background test run showing that after 4 consecutive expansion runs the total particle count is almost zero (only one droplet count detected in expansion no. 5).

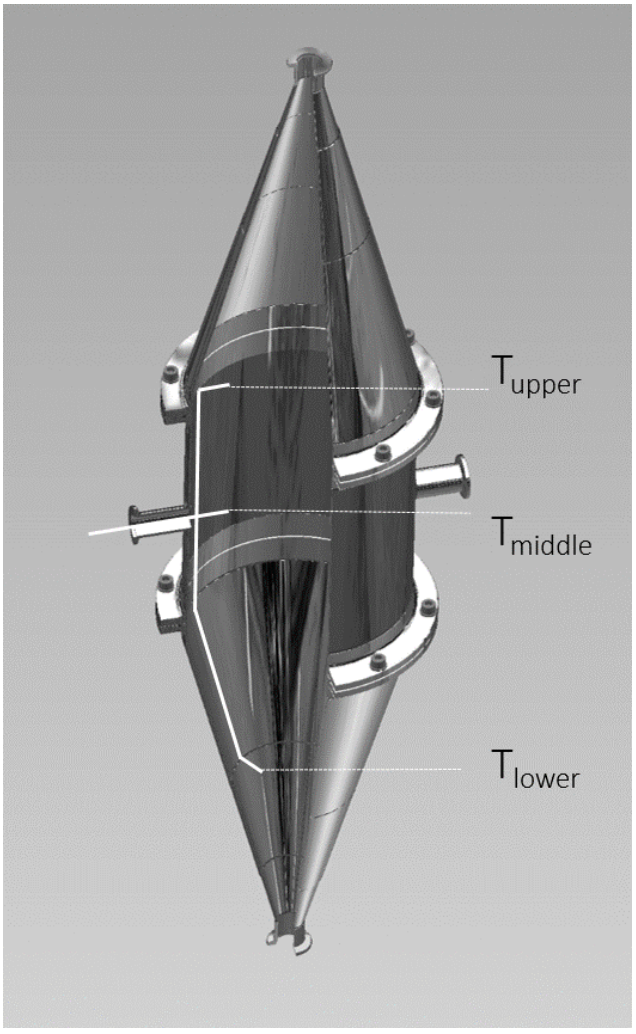

Figure A4. Construction of the PINE-1A stainless steel cloud chamber, without cooling and thermal insulation. The white lines indicate the location of the three thermocouples measuring the gas temperature inside the cloud chamber.

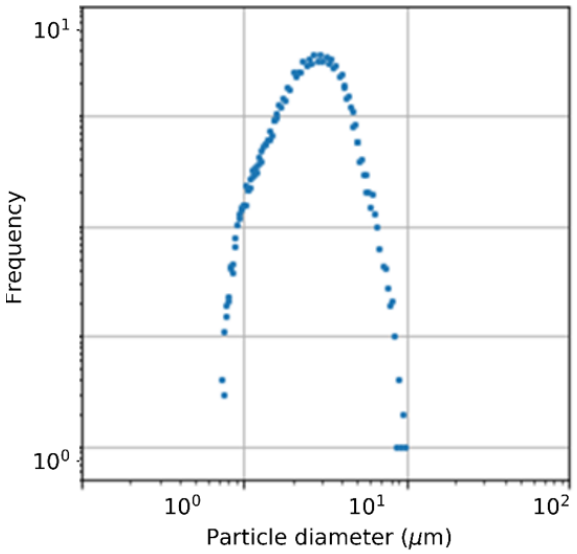

Figure A5. Size distribution of activated droplets measured with PINE-1A at high temperature conditions where no active INPs were present.

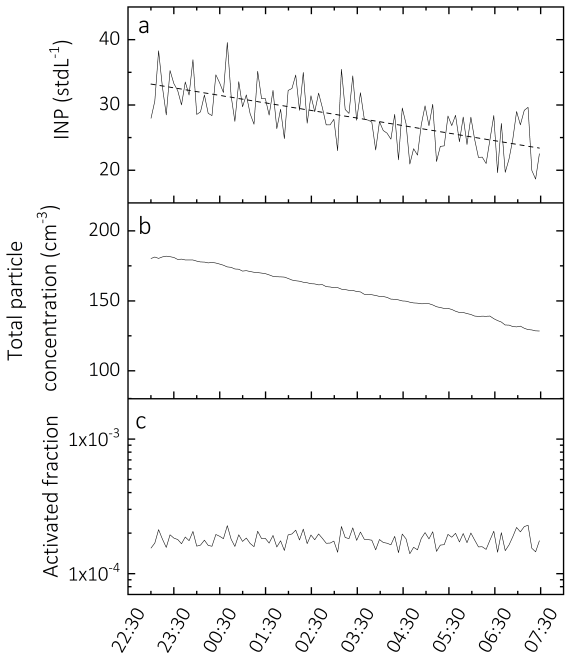

Figure A6. ~~PINE-c operated at~~ Time series from an experiment with the ~~SGP site~~ new commercial PINE-04-01 instrument sampling aerosol from the AIDA cloud chamber, with INP concentrations measured at 247.2 K (panel a)~~for continuous INP measurements for 45 days from October 1st to November 14th~~, ~~2019. The foto on the right side~~ total aerosol particle number concentration measured with a condensation particle counter (panel b)~~shows~~ , and the ~~same instrument with all~~ ice-active particle fraction fice calculated from the ~~side plates in place~~ratio of the INP number concentration to the aerosol number concentration, with a mean value of 1.8×10^{-4} and a standard deviation of 2.1×10^{-5}.

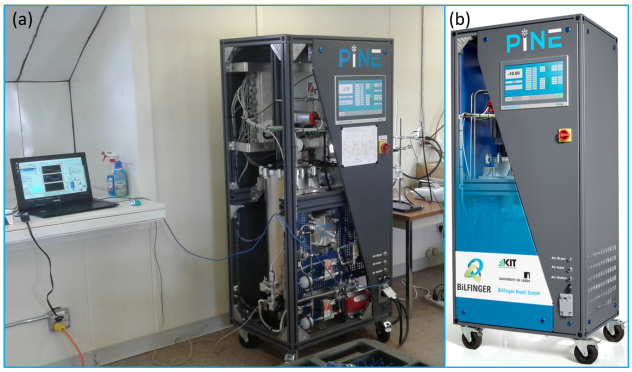

Figure A7. Photograph of PINE-c (a) located at the ARM-SGP site in Oklahoma for continuous INP measurements for 45 days from October 1st to November 14th, 2019. Part (b) on the right shows a composite photograph of the same instrument before delivery.