

We would like to thank the reviewer for taking the time to review the presented manuscript and provide their appraisal of the research carried out. We will try to provide a satisfactory reply to all the questions raised in this document. The posed questions are in green and the replies are presented below them.

Before replying to the individual questions we would like to offer our apologies for the amount of typos found in the manuscript. A complete evaluation of the manuscript has been carried out again and both the issues presented by the reviewer and others found during this new evaluation have been addressed and we hope the new manuscript will present a clearer and more understandable text.

Major Questions

Question 1: After reading the manuscript several times, now have the impression that during the calibration campaign only runs with steady flow conditions have been performed. Is this true? If so, why did you not perform evacuations for cloud formation as well? How much data (e.g. in hours, or if evacuations how many) did you take during the calibration campaign. How many hours did you measure in flow conditions during the data campaign? I think this information would be useful and should be added to the manuscript. Please clarify! In general, more information about the experiments performed during both campaigns might be helpful, maybe summarise the experiments in a table?

The calibration campaign stated in the manuscript did indeed only consist of measurements taken during steady state conditions. No evacuations were performed. The reason for the calibration campaign was to create a calibration curve for the different permanent temperature strings that exist in CLOUD (the Pt100, TC and OS strings). In order to do this, several points at different operational temperatures in CLOUD are required. One temperature measurement is thus required to be acquired at stable conditions. Although expansions (evacuations) provide important information about the dynamics of the chamber, they do not provide information relevant to obtaining a calibration curve of the sensors. Each temperature point taken during the calibration campaign took 24 hours. During data campaigns the permanent sensor strings (Pt100, TC, OS) were constantly turned on and acquiring data. The relevant nucleation experiments used were taken during separate periods between 29th of September and 29th of October 2014. Each temperature point during data campaigns consists of a period no smaller than 3 hours. The manuscript has been altered to better relay this information and the focus of these experiments and a table containing the major relevant parameters of these nucleation experiments has been included.

Question 2: Why Pt100 sensors were chosen for the calibration strings? This is not motivated in the manuscript. However, as you state, they have a rather long response time (180 s), which seems insufficient for cloud experiments, where temperature drops much faster than this? How do you compare temperatures

from the fast response sensors to these slow response sensors? Please explain!

The manuscript was indeed confusing regarding this point. A distinction must be made between the permanent Pt100 string and the calibration PT100 strings (PTH and PTV). The description of the Pt100 string and more specifically the 180s response time was regarding the permanent Pt100 string. The calibration Pt100 strings use a completely different hardware and software construction, allowing for 1s time resolution measurements and smaller time response. A revised paragraph on the construction of the PT100 calibration strings was added, containing the following text distinguishing the Pt100 strings:

“These sensors, unlike the Pt100 string already present in the CLOUD chamber and despite also being four-wire sensors with National Instruments NI~9217 readout electronics, do not have the high mass of the first sensors, allowing for fast responses equal to the TC and OS sensors.”

Question 3: Why do you average the data over 15sec? Does it make sense in case of the Pt100 with time constants around 180sec? On the other hand, 15secs smooth out fluctuations in the fast sensors responses. Do you also smooth data during evacuations?

The data in the plots shown in figures 3 and 4 was altered using a median filter. This filter applies a median to a specified window of data (in this case 15 seconds) as opposed to the averaging mechanism suggested by the reviewer that applies an average instead. The reason for this data alteration is two-fold. First, to remove any outliers from the measurement of the distribution of temperature. Secondly to provide the reader with an easier to read figure, since without the filter, the reader would not be able to see the fluctuations of the data and the different signals shown. Indeed, temperature drops during cloud formation experiments (expansions) were at times much faster than 180 seconds. The temperature measurements shown in this manuscript for expansions, however, do not show measurements from the permanent Pt100 string, only the fast response OS and TC strings. The permanent Pt100 string only exists to measure nucleation experiments, the comparison of data from this string to the fast response sensors is thus valid, taking into account the total run time of nucleation experiments (several hours). The data for expansions was not altered since we were not measuring a constant temperature distribution but a time-changing one. In every instance where data was altered, it is explained to the reader in the manuscript. The following text was added to the caption of figure 3:

“The measurements are smoothed with a 15 s median window firstly to improve the measurement result by removing any outlier values of the measurement distribution and secondly to improve readability of the figure by the reader.”

Question 4: I would like to see a schematic of CLOUD chamber that shows more detail, e.g. the “serpentine” pipe (page 3, line 71), regulation and gate valves (line 51), sampling ports for cloud measurements. Particularly the valve positions in respect to the temperature sensors would be good to know!

The gas input pipes are located below the lower mixing fan, which is then responsible for pulling the inputted gas and mixing it with the air inside the chamber. They insert the gases at a height of a couple of centimeters above the bottom of the chamber ($-150 \text{ cm} < z < -140 \text{ cm}$ using figure1) Figure 1 has been altered to include a representation of the gas input valves along with a representation of the axis used to calculate the sensor positions in the chamber. The reader can now estimate the position of the pipes in relation to the temperature sensors (using figure 1 and table 1).

Question 5: A large part of the introduction (i.e. from line 28) reads more like a potential chapter 2 “chamber operation” (or similar). I would expect more introduction about cloud chambers and the importance of temperature measurements, temperature stability e.g. what motivates your manuscript.

The manuscript has been changed to reflect the expressed views. Section 2 is now named chamber operation and part of the offending text in the introduction was moved there. The following text was included to stress the relevance of temperature characterization of chamber measurements:

“Maintaining temperature stability and uniformity in these chamber measurements ensures that the chemical reaction rates in the chamber do not fluctuate either in time or in space [2]. Accurate measurement of temperature is also necessary to measure the onset of ice formation in chamber experiments [3, 1].” (citations properly introduced in manuscript)

Calibration Runs

Question 1: Are calibration and data runs performed at the same relative humidity? (Think of cloud formation, latent heat release...)

No dedicated measurement of relative humidity was taken during the calibration runs. However, the proximity of the calibration sensors to the TC and OS sensors as well as the sheer length of measurements during the calibration campaign drastically reduces the uncertainty caused by varied values of relative humidity. The campaign data was also taken at various relative humidity values. The nucleation experiments were a part of scheduled experiments of one of the many institutes that are a part of the CLOUD consortium and were not subject to any change. There were no dedicated experiments during the data campaign for temperature measurements (either nucleation or expansion experiments). The manuscript has been altered to reflect this concern.

Question 2: Are there calibration runs that were performed at data run like flow rate? You could simply let the instruments suck as well – higher flow rate might increase the temperature instability. Thus, it would be necessary to show. How are clouds formed in a calibration run? If clouds are only formed in expansions, what is the meaning of calibration runs for cloud studies in the chamber?

The instruments referred to in the manuscript and this question were not yet present at the time in CLOUD. The instruments present in CLOUD data

campaigns are the property of several institutes in the CLOUD consortium (and outside it as well). These instruments are taking measurements at several sites around the world during the year. Only at specific times are they present at CLOUD. At the time of the calibration campaign, no instrument had yet arrived. The very high cleanliness standards of CLOUD also prevent us from keeping instrument ports open for large periods of time. Thus it was simply impossible to mimic the flow conditions of CLOUD data campaigns without inadvertently increasing the pressure in the chamber. No clouds were formed in the calibration run. As stated in the first question, the goal of the calibration campaign was to provide a calibration curve for the temperature sensors. No other instruments were calibrated during this campaign. The manuscript was altered to clarify the definition of the calibration campaign throughout the text.

Question 3: You only show examples of the measurements in the figures for the calibration campaign. Is there a way of showing all data in one plot? Are there any expansions made during the calibration campaign? As the calibration strings were only installed during the calibration campaign, this would need to mimic conditions as they would be during a measurement campaign. If not, what is the aim of the calibration campaign?

Figure 3a, 6a, 6b, 7 and 10 show data taken during CLOUD data campaigns. If the reviewer is referring to the measurement distributions provided in figure 4, this can be remedied. It would however, require the reviewers clarification. No expansions were made during the calibration campaign as explained in the first and previous question. See previous question for reason of calibration campaign

Question 4: You state e.g. in line 148 that around 300 expansions have been performed in the latter campaign. I would expect something like scatterplots showing all data and median/average values (if necessary grouped into classes by speed of expansions to show all data. “Various experimental conditions” are mentioned in the abstract, not mentioned any further later on! What are these various conditions? They could be used to group data for plots. Where is the statistical analysis? What about significances?

Over 300 expansions were indeed performed in the CLOUD campaign. An indirect representation of the distribution of expansions is presented in figure 9 accompanied with statistical analysis. The temperature change in an expansion is indirectly related to the time of the expansion. The variation of experimental parameters is too large to provide an easy visualization of the parameters. Presenting the different expansions also goes outside the scope of the manuscript, as only the relevant dynamic behavior of expansions in the CLOUD chamber is studied in this manuscript. A statistical analysis is provided with the use of the expansion reheating parameters in figures 9 and 8. The reheating parameter in our opinion provides a quantitative parameter to analyze individual expansions.

Lab Calibration

Question 1: You mention that the WIKA reference thermometer is calibrated in the temperature range 0-100°C. How do you use it at temperatures below freezing? What confidence do you have in its performance there?

The WIKA thermometer was only used for the positive temperature values, including the 0°C point. The appendix has been edited to clarify this point.

Question 2: You mention liquid nitrogen as calibration point for cold temperatures. Is it valid to assume linear calibration between -196.21°C and 0°C, why? How did you get calibration points at temperatures between -70°C and 0°C, i.e. at temperatures that would potentially be used for experiments in the chamber? (E.g. you could add one point by using salt/ice mix and one point by using dry ice in a Dewar flask).

As expressed in the manuscript, the Pt100 sensors were calibrated using the Callendar-van Dusen equation. This equation provides a curve for resistance of Pt100 sensors in the range of temperature of [-200 °C, 600 °C]. This equation is a quadratic equation for temperatures above 0°C and a third degree equation for temperatures lower than 0 °C. This relationship is based on the properties of platinum that are part of the sensors. There were no laboratory temperature points in the range of CLOUD's negative temperatures especially because the reference thermometer was not absolutely calibrated below 0°C. The Callendar-van Dusen equation along with the estimated parameters will provide a relationship between each sensor's resistance and temperature in the whole range of CLOUD operating temperatures. The uncertainty of the measurements of the Pt100 sensors was estimated on a worst case scenario using the Monte Carlo study for the ranges of temperatures in CLOUD (including the negative temperatures). The manuscript was altered to clarify first the use of the Callendar-van Dusen equation and secondly the importance of the Monte Carlo study in the calculation of the temperature uncertainty for points without laboratory measurements.

The suggested salt-water mixture suggested by the author was considered but the use of said mixture in the laboratory would go against the strict cleanliness standards of the CLOUD experiment, running the risk of contaminating further campaigns after placing the strings in the chamber. The following text was added to the appendix:

“In order to comply with CLOUD's cleanliness standards, only pure water and liquid nitrogen were allowed for the laboratory calibration of the Pt100 sensors.”

Question 3: How did you calibrate at 0 °C, this is not mentioned in the text? Figure A2 is showing a very different behaviour of the sensors at 0°C compared to the water bath calibration points (which, as you state, start at 2°C). So, how trustworthy is the point at 0°C?

The calibration at 0°C was accomplished by using a mixture of milipore water and ice. During this phase transition it is guaranteed that the temperature is at 0°C. There are, however some concerns to take into account, mostly

related to the volume of the Huber unit used. The most important of which is the possible existence of convection currents that increase the uncertainty of the measurement. The following text has been added describing the 0° C point:

“A 0° C point was obtained using a millipore water bath containing ice, ensuring that the temperature was at 0° C while ice was present in the bath.”

Question 4: Why are the OS, TC and Pt strings not calibrated directly in the lab as well? How exactly is linear interpolation performed for the Pt string (page 8, line 237/238)? Elaborate!

The reason for the creation of these specialized strings was to make an in-situ calibration of the permanent temperature sensors. It was noticed that when the sensors of the permanent strings were disconnected from their readouts, their calibrations changed. This makes it impossible to disconnect the strings from the chamber and make a proper calibration only to reconnect the readouts and find out that the calibration had changed. These specialized strings were made in such a way that the string could be unmounted and mounted in the chamber while keeping the individual sensors connected to their respective readouts, allowing for a laboratory calibration and subsequent in-situ chamber calibration. The manuscript now stresses this point throughout with passages such as the following which has been added to the end of section 2 to clarify this point:

“Removing the sensor strings to calibrate requires that the sensors be disconnected from their respective electronics, this results in a shift of the previous calibration. It is thus impossible to remove the strings for calibration and place them back in the chamber using the same calibration.”

In response to the concern as to how the interpolation of the sensor was made. This was a spatial linear interpolation of the temperature between the position of PTH sensors and the permanent Pt100 sensors. The following text was added to the manuscript as an effort to explain how the interpolation was made for the also non working PTH2 (see question 4 in next section), which also applies for the permanent Pt100 string and this has been stressed in the manuscript:

“PTH2 was non responsive after being placed in the chamber. Thus it’s measurement was replaced by a spatial linear fit between the measurements of PT1H and PT3H defined by:

$$T_{PTH2}^* = T_{PTH1} + (r_{PTH2} - r_{PTH1}) \frac{T_{PTH3} - T_{PTH1}}{r_{PTH3} - r_{PTH1}} \quad (1)$$

where r_{PTY} is the radial position of sensor PTHY and T_{PTY} is the temperature measured by sensor PTHY (see table 1 for details).”

Other questions - which were not answered in other sections

Question 1: “experimental hall temperature” – wall temperature?

This indeed refers to the hall temperature at CERN where the CLOUD chamber is placed. Since it is a large hall shared by many other experiments, not to mention provides access to all scientists in and out of CLOUD to (verify instrumentation, etc..), it is simply impossible to provide temperature control for this hall at CLOUD operational temperatures). The feed-through gas pipes travel a short path (a few meters) through this hall before going either through the temperature controlled serpentine cable or being directly sent into CLOUD and there is an inevitable alteration of temperature during this path.

Question 2: Appendix: This describes the calibration in detail. Isn't the temperature calibration a main point in this study?

This question was also considered by the authors. The decision was to provide an appendix to the paper due to the fact that the manuscript is related to the analysis of the temperature stability, uniformity of the chamber. While the calibration of the sensors and how it was done is indeed important to show, it should not overshadow the end goal of the manuscript.

Question 3: Figure 4: How did you choose which sensors you show here? Motivate your choice. You could also show all other sensors in a supplement.

The sensors were chosen as they are the sensors at the middle of each respective string. A supplement has been prepared with the measurements of all other sensors in the string at the respective temperature, containing the requested plots, which are shown at the end of this document. The following text was added to figure 4:

“OS3, TC3 and PT3 were chosen due to being the central sensors of each respective string. The temperatures chosen represent common temperatures used during CLOUD campaigns.”

Question 4: Figure A4: You mention a malfunction of the PTH2 sensor, was this true for the whole campaign?

This was indeed true during the whole calibration campaign. This was resolved in data analysis by creating a virtual sensor via a spatial linear interpolation of the temperature between the radial position of PT1H (145 cm) and PT3H (90 cm) much as the same used for calibrating the permanent Pt100 string. An explanation of the interpolation mechanism can be found in question 4 of the previous section and as that question states, the manuscript has been updated.

Question 5: Were the OS4 and OS5 sensors replaced after the calibration campaign (as obviously they showed an unusual behaviour)?

The sensors were not replaced, as doing so would involve removing all other sensors from their respective readouts. This has been clarified in the manuscript.

Question 6: Table 1: From this table one could think that the TC and PTH / OS and PTV sensors have the same position. It would be better to indicate

the offset in the table, eg. by saying “0 (-20)” in the Height column (TC and PTH), and accordingly for the radius column for the OS and PTV sensors.

The sensors were placed in their strings and installed in the CLOUD chamber in an effort to place them in the closest possible position to each sensor to be calibrated. No measurement was made to determine the offset, visual inspection concluded that the sensors were offset by no more than 1 cm. As suggested the values $PT \pm 1$ cm were added to the PTH and PTV sensor positions in table 1 to indicate that their positions did not exactly match.

Question 7: Table A4: Why are PT4 and PT6 missing?

The permanent Pt100 string is only a 5 sensor string. PT4 was not functioning at the time of the calibration campaign and data campaign. This malfunction occurred during one of many efforts to calibrate said string in lab. One of such efforts must have damaged the sensing tip. During these efforts we noticed the shift in calibration when reconnecting the electronics which led us to make the calibration process described in the manuscript. The manuscript has been altered to address this concern.

We again would like to thank the reviewer for the time taken to appraise this manuscript. We hope that these replies along with the provided supplemented material and changes to the manuscript can provide a more favorable recommendation.

References

- [1] PJ Connolly, C Emersic, and PR Field. A laboratory investigation into the aggregation efficiency of small ice crystals. *Atmospheric Chemistry and Physics*, 12(4):2055–2076, 2012.
- [2] Howard G. Maahs. Kinetics and mechanism of the oxidation of s(IV) by ozone in aqueous solution with particular reference to SO₂ conversion in nonurban tropospheric clouds. *Journal of Geophysical Research*, 88(C15):10721, 1983.
- [3] Birte Riechers, Frank Wittbracht, Andreas Hütten, and Thomas Koop. The homogeneous ice nucleation rate of water droplets produced in a microfluidic device and the role of temperature uncertainty. *Physical Chemistry Chemical Physics*, 15(16):5873, 2013.































