

We would like to thank the reviewers for their insightful and helpful comments, and encouragement for the need of vertical profiling of INPs. As a result of revision based on their feedback, the manuscript is much stronger. Below are the responses to reviewer comments in blue. We note that both reviewers were generally concerned with the hovering capability of the system in addition to presenting one successful flight out of three. As a result, we have significantly revised the tone and organization of the manuscript, in addition to providing the community with what did and did not work, and recommendations for those interested in developing similar systems in the future. More details and specific examples of where text was modified is provided in the responses below. We have attached a track changes version for reference of the specific revisions made.

Reviewer 1

General/Major Comments:

1) As mentioned above HOVERCAT's ability to hover at a constant altitude could not be reliably demonstrated (Figs. 2 and 7). Only 2 out of 6 samples show stable enough altitude conditions (one of which was collected close to the ground). Page 6, Line 3 states the original flight plan was a 5-step altitude profile with 500 m steps each. Yet, the actual flight profile looks nothing like that. Section 3.1 explains that the plan could not be fulfilled because of too windy conditions and a time delay between command and execution of buoyancy adjustments. The reader is then reassured that on a calmer day it might work. Yet, it leaves the reader wondering. How to achieve it in the future? Why didn't the authors do more flights until, the flight plan worked as planned? The rate of 1 out of 3 flights producing high enough sample volumes to be analyzed for INPs, seems also to be improvable. Have there been more test flights in the meantime?

We have softened the hovering aspect of the system throughout the manuscript to rectify the issue with the hovering flight plan. We also added, "However, this plan was ultimately not executed due to flight complications discussed herein." to section 2.4. The only places the word "hover" remains in the manuscript is in the HOVERCAT acronym. We also revised the definition of the BBFCS system such that the user can control the ascent and descent rates of the balloon instead of "controlling the altitude".

We realize we were not initially clear with what components were included in the HOVERCAT definition, which we intended to define as the aerosol components, since the honing on vertical cloud and aerosol properties is the responsibility of the aerosol instrumentation. We have now made this clear throughout the manuscript.

To address the issue with having only one successful flight, we note that the system *does* work, but with its caveats. Even the Ardon-Dryer et al. (2011) study that the reviewer mentions report only three filters from one day, indicating a substantial number of flights and samples is not required for publication of what (very) limited INP profiles exist. We emphasize that this is phase I and think it is important to discuss what *didn't* work as that is important to other researchers who may be interested in developing parallel systems in the future, such that they will not have to "reinvent the wheel" so to speak. This is the reasoning behind keeping the original flight plan, such that our idea of stepping is not as easy as one might think. However, we now modified section 3.4 to discuss our recommendations for those interested in controlled launched balloon measurements, including a list of what to do and not to do. This addition provides rationale for keeping in the flight plans, why we had one out of three successful flights, and provides suggestions for improvement. There is a certain stigma with publishing only partially successful results or deployments, but we aim to break that mold and show what didn't work to help those in the future. In addition, we originally intended to showcase the vertical profiling of both immersion and deposition mode INPs with the aerosol module, which is indeed novel, but have now emphasized that throughout the manuscript.

To provide some specific examples of where we modified the text:

- We revised the abstract to highlight the aerosol module and state that we provide recommendations for use of future launched platforms.
- We instead emphasize that the BBFCS has a benefit of being more cost effective and flexible (in terms of FAA regulations, launch locations, and personnel required) than traditional tethered platforms such that it does not require a winch and that we can slow the ascent rate in addition to providing a descent option which, combined, is not a feather attainable with traditional launched balloons. This information is provided in section 2.1.

- We completely revised and reorganized the methods section to clearly define HOVERCAT and describe that first, followed by describing but not showcasing the balloon platform.
- We revised section 3.4 to emphasize our scientific priorities (discussed in detail in our response to comment 2) and recommendations for future measurements.

In Section 3.4, where the future directions of HOVERCAT are listed, point 2) says: “operate successfully on a routine basis”, however, I feel this should be a prerequisite for the platform, before thinking about publishing. It feels like step two is done before step one. In the Conclusion the platform is stated to have “the capability to hover at desired altitudes, making it an ideal system to collect sufficient aerosol loadings at a range of altitudes up to 2.6 km AMSL”. The system may be capable of doing it (as sample 4 might suggest), yet I don’t feel like it has proven without a doubt.

This section was considerably revamped to instead discuss recommendations for future deployments of such systems in general. We agree that it has not been proven without a doubt that the system can hover—this concern was alleviated by toning down the hovering capability description throughout the entire manuscript.

2) On a more technical note: Why didn’t you sync the profiles with the sampling times? Wouldn’t it be better to have a flexible (e.g. command based) way to communicate with the sampling system to adjust for difficulties and only start the pump when the profile is stable? Alternately, why not include a buffer of some minutes between prescribed sampling periods to account for maneuvering to reach the desired altitude?

Certainly, it would be easier to enable ground-based bidirectional commands to change the sample spot when desired. However, the manuscript presents a pilot study focusing on specific priorities: (1) Can we recover the system? And (2) Can we control the ascent and descent? Both priorities fall under the umbrella of making a system that is relatively affordable, user friendly, and ensuring avoidance of complicated FAA COA paperwork. Adding complexity to the system for controlling altitude and sampling frequency adds cost, weight, and more variables that could fail. For clarity, we now emphasize that this was a pilot study throughout the manuscript (in addition what we already call “test” flights) and discuss our priorities in section 3.4.

More specifically, a bidirectional communication feature entails a significantly more advanced ground-based system and would add too much weight to HOVERCAT through additional processing hardware, an additional receiver, and batteries needed to control in such a manner. First, the BBFCS had bidirectional communication, but additional channels would need to be added to control the TRAPS as well, adding to the cost. And second, adding the necessary components to the TRAPS would add to both cost and weight, and given our 400 g buffer (i.e., calculated from the 2700 g limit and our HOVERCAT 2300 g weight), would likely push the system into a weight category that requires either restricted airspace or significant paperwork to obtain and FAA COA to fly outside of restricted airspace areas. Both of these issues make flying HOVERCAT more complicated and expensive, which fall outside of our vision to make an affordable and easy-to-use system.

3) In the Introduction the authors describe why more airborne INP measurements are needed and that there is a lack of alternatives to aircraft-based missions to gain vertically resolved information about INP. In this regard the paper by Schrod et al. (2017) should definitely be cited and summarized. Schrod et al. (2017) describe the first INP measurements made onboard of an unmanned aircraft system (UAS) and follow a similar train of thought. On a similar note, the paper by Ardon-Dryer et al. (2011) should be mentioned as well. In it INP samples were collected at the ground and from a tethered balloon in Antarctica.

Thank you for bringing these references to our attention, which are certainly important and noteworthy studies with regarding to the vertical distribution of atmospheric INPs. We have added both Schrod et al. (2017) and Ardon-Dryer et al. (2011) to the introduction, their advantages, and how our results differ from theirs. Schrod et al. (2017) measured INPs but only in the deposition mode. Ardon-Dryer et al. (2011) present results more relevant to ours, but only measured INPs below $-18\text{ }^{\circ}\text{C}$ in immersion mode on 3 samples and only up to 196 m a.g.l. The fact that only two studies before ours exist, and the limitations of those studies and ours, demonstrates the challenges associated with obtaining vertically profiled INPs, which is important to highlight and why we directly state this after describing their work in the introduction. We also removed the statement, “Additionally, to our knowledge, there have been no measurements of INPs via any balloon platform.”

4) Fig. 6: Why didn't you wait until all drops are frozen – is it due to instrument limitations? Also, Page 7 Line 28f says the measurements were continued, until all droplets were frozen. Which fraction is used for calculating the INP concentration by the Vali equation (Fig. 8)? The ones shown in Fig. 6 or the actual number of frozen droplets relative to the total number of droplets? If the frozen fraction as shown in Fig. 6 were used, you'd create a bias towards a more ice active sample. I would rather show the plots as done in Figs. 4 and 5 (with the frozen fraction not normalized to 100%) to avoid confusion about this matter.

We realize our description was confusing. At times, not all drops froze for the control experiments with UPW due to the system detection limit (i.e., it can only go down to approximately -32 to -33 °C). The fraction frozen was the number of frozen droplets divided by the number of droplets detected + unfrozen drops, which is why we did not always observe a frozen fraction of 1. However, all drops froze from the blanks with the bag, blanks with the filter, and the collected samples themselves. We more clearly define this in section 2.3.1.

5) Fig. 10: Does it show the onset conditions of the first observed ice nucleation? Or is it a constant activated fraction of X %? Is there any quantitative measure of this ice nucleation technique available at all? From what I understand from section 2.3.2 it is not (at least in this study). Therefore, I would advise to be careful in making too strong statements, when just the first ice activation was observed. Go through the paragraph with that in mind. E.g. rewrite the sentence on Page 12 Line 1ff to something like this: “Overall, ice activation onset conditions between the six samples were similar at all temperatures tested. However, at -40 °C Samples 3 and 4 showed first ice nucleation activity at saturation ice ratios X to X % earlier than the other samples and may be characterized as more efficient deposition INPs at that temperature as compared to the remaining samples. However, it should be emphasized that the presented results are not of a quantitative measure.”

Yes, Figure 10 shows the onset conditions of the first observed ice nucleation event. Because this is the first observed ice nucleation event, we can say that our percentage of ice is approximately 10^{-4} (1 out of 10^4 ; total number of particles on the sample disc) and thus our technique is semi-quantitative. We have changed this to read:

“Overall, ice activation onset conditions between the six samples were similar at all temperatures tested (Figure 10). However, at -40 °C, Samples 3 and 4 showed first ice nucleation activity at a saturation ice ratio of 1.12, which were lower than the other samples and may be characterized as more efficient deposition INPs at that temperature as compared to the remaining samples.”

6) My impression is that sections 3.2 and 3.3 are somewhat overanalyzed in terms of trying to explain the sources etc. After all, the result section of this manuscript depicts only six consecutive 30 minute samples of one day, most of which feature a considerable variance in sampling altitude, which should make it difficult to make general statements about the different samples. Also only 3 – 5 INPs were analyzed with Raman spectroscopy from each sampling spot, again making it difficult to allow general statements about each sample. I will give some examples: a) Page 11, Line 10ff: I think this statement is somewhat misleading. Sample 6 had only a short period of approx. 5 minutes where it was hovering close to the ground. The corresponding reasoning is rather speculative. b) Page 11, Line 17f: 1) and 2) are identical, but only phrased differently? c) Page 11, Line 18f: Although it is a logical statement, is this really supported by the data at this point? d) Page 12, Line 5f: It should be avoided to speak too generally here, since only 3-5 particles per sample were analyzed. How this few particles be representative of a whole sample? e) Page 12, Line 15f & Page 12, Line 17ff: This is again rather speculative.

Although Sample 6 was only near the ground for 5 minutes, those 5 minutes had OPC particle concentrations up to 248 cm^{-3} . Influence from short exposure to samples for drop freezing analysis can yield significant changes in the freezing temperatures and thus the INP concentrations, which were on a maximum of 0.04 cm^{-3} INPs for Sample 6. The ice nucleation community has this issue with even exposure of blank samples to laboratory air for a matter of seconds to minutes—even small levels of exposure to relatively “clean air” in the laboratory can introduce enough artefacts to increase the background level of even the purest water.

We revised the sentence summarizing the results in section 3.2 to, “Combined, the immersion INP, OPC, and BBFCS results indicate that: 1) total particle number concentrations and INP concentrations were highest when HOVERCAT sampled near the ground and 2) INPs of likely biological origin remained close to the surface, which is predominantly agricultural soils (Hill et al., 2016).” For point 2), this is indeed a logical statement given the freezing temperatures of

the samples—only biological particles have been shown to nucleate ice at temperatures above $-15\text{ }^{\circ}\text{C}$ in the atmosphere (see Murray et al. (2012) and Kanji et al. (2017) and references therein).

Murray, B. J., O'Sullivan, D., Atkinson, J. D., and Webb, M. E.: Ice nucleation by particles immersed in supercooled cloud droplets, *Chem Soc Rev*, 41, 6519-6554, 2012.

Kanji, Z. A., Ladino, L. A., Wex, H., Boose, Y., Burkert-Kohn, M., Cziczo, D. J., and Krämer, M.: Overview of Ice Nucleating Particles, *Meteorological Monographs*, 58, 1.1-1.33, 2017.

With regard to the Raman section, we dissolved the samples into HPLC grade water, shook and inverted them to ensure complete mixing, and then the sample was nebulized. From this method, we believe that most of the particles on the sample disc will be of similar composition. While we only analyzed 3 – 5 particles, they were always quite similar. We have added a sentence clarifying why we assume that most particles will be of similar composition: “Of the particles that nucleated ice, 3 – 5 particles were analysed for composition using Raman spectrometry for each sample. We assume that a majority of the particles are of similar concentration because the whole sample was dissolved in water, allowed to mix to a homogeneous solution, and nebulized onto the sample disc. Indeed, the particle composition was similar for each particle in any sample, while there was variation from sample to sample.”

Specific/Minor Comments:

1. The terming of HOVERCATS ability to measure “time-resolved” INP concentrations (Abstract, Page 13 Line 11) seems a little misleading, since it averages over broader time periods instead of measuring in real-time.

The purpose for defining our sampler as time-resolved was to distinguish it from previous balloon-borne measurements where a filter was collected for the entire flight. Time-resolved does necessarily indicate real-time, however, we removed “time-resolved” from most spots in the manuscript for to prevent such confusion. Since this is a part of the TRAPS acronym, we specifically define “time-resolved” when first discussing the sampler in section 2.2

2. Use meters above ground level throughout the manuscript (other than when discussion problems related to the flow of the pump). The Colorado plains has a rather high ground elevation, which may lead the reader to think the balloon was flying higher than it actually was.

Understanding the pressures in which the pump was functional at is an important aspect to the system. We wanted to provide the altitude above ground *and* sea level, to demonstrate that the pump had to work harder at the lower pressures aloft in Colorado as, say, compared to flights in Seattle in which 1000 m a.g.l. would have much higher ambient pressures than 1000 a.g.l. over Colorado. The balloon could certainly fly much higher a.g.l. in lower elevation locations than the 500 m a.g.l. in Colorado. However, for clarity we now ensure that anywhere we state a.m.s.l. that we provide a.g.l. when referring to the flights in Colorado and to be consistent with the a.g.l. in Figure 2, we changed Figure 7 to a.g.l. as well.

3. Page 2, Line 10: When listing the ice nucleation modes, a reference to Vali et al. (2015) should be considered. Also, the concept of pore condensation freezing as introduced by Marcolli et al. (2014) could be mentioned, when describing deposition freezing.

Vali et al (2015) was added, in addition to a mention of pore condensation freezing and the Marcolli et al. (2014) reference.

4. Describe in more detail how the system adjusts the altitude, how it lands and how it is retrieved. Is the balloon still intact after landing, can it be reused?

We added a description of how the balloon system adjusts to the altitude, how it lands, and how it is retrieved in section 2.2. We did not go into detail on how it adjusts to the altitude because we removed discussion on the hovering capability.

5. Why haven't you used a more powerful pump that can go above 1.2 lpm? I understand that payload restrictions are crucial for this approach, but I think the system would be much more flexible when the sample flow could go higher.

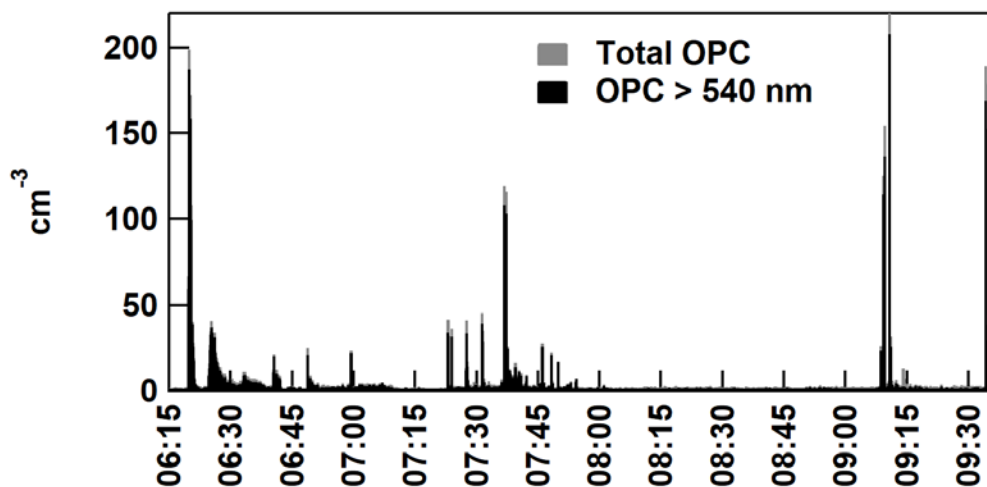
It is certainly true that higher flow rates would yield larger volumes of air and thus sufficient sample loading for ice nucleation measurements. However, as our manuscript and the reviewer note, putting a larger pump on the system would indeed exceed the FAA regulations on payload restriction. We are limited to 2.7 kg or less, and our aerosol module weighs 2.3 kg. We have researched a variety of stronger pumps, none of which weight less than 400 g.

6. Page 10, Line 28ff: I think it would be better to give median (or average) particle concentrations instead of the maximum, since near ground values showed episodic peaks. Also, a plot showing the particle concentration vs. altitude could be worthwhile.

We added the averages for when the system was close to the ground versus when it was aloft in section 3.1. We wanted to provide the maximum here to show how influential ground sources are to the particle counts. Figure 7 already demonstrates that when the system was flying near the ground, the OPC counts and mass concentrations clearly increased, thus, a correlation plot would be redundant. Additionally, as indicated by the spikes, the sources of high particle counts were localized at the ground, thus a correlation of such high time resolution data would not be observed. We added this to the discussion in the second paragraph of section 3.1.

7. I wonder if you considered to calculate the correlation between OPC concentration larger $0.5 \mu\text{m}$ (as in DeMott et al., 2010, 2015) and the INP concentration at $T = -X \text{ }^\circ\text{C}$ averaged over each sampling period (although $N = 6$ isn't very good statistics), and/or add a scatter plot.

We did look at the OPC concentrations from the third bin to the last (i.e., $540 \text{ nm} - 17 \mu\text{m}$). However, (1) the lower limit can only be 540 nm and not 500 nm and (2) there was no strong correlation with the $\text{OPC} > 540 \text{ nm}$ concentrations and INP concentrations at $-10 \text{ }^\circ\text{C}$, $-15 \text{ }^\circ\text{C}$, $-20 \text{ }^\circ\text{C}$, $-25 \text{ }^\circ\text{C}$, nor the maximum INP concentration per sample. There is a correlation between the $\text{OPC} > 540 \text{ nm}$ and temperature in which a fraction frozen of 1 occurs, but as the reviewer notes, this is only for $N = 6$. Thus, we made no such changes in the manuscript. Here is a plot of total OPC concentrations versus concentrations of $> 540 \text{ nm}$ demonstrating a minor difference in the two:



8. Page 11, Line 12: Give the range of minutes (from X to X minutes) that HOVERCAT hovered near the ground. Following the reasoning on Page 11, Line 14ff it may be worthwhile to correlate the number of minutes close to the ground with the INP concentration of the corresponding sample.

Good idea. We have now added the % of time at the ground for the duration of the sample (i.e., $< 50 \text{ m a.g.l.}$) to the discussion in section 3.2. Sample 3 did have the highest percentage of time spent at the ground compared to the other samples (69%), followed by sample 1 (40%), sample 6 (19%), sample 2 (9%), sample 5 (2%), and sample 4 (0%).

9. In section 3.4 the authors should add a goal to improve on the ability to stay at a constant altitude with HOVERCAT.

Done.

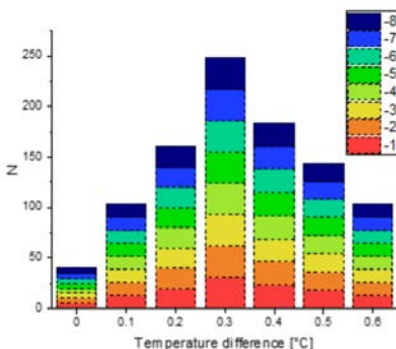
10. Add a Figure that shows a detailed schematic of the TRAPS. I see that a similar Figure can be found in Ogren et al. (2017). Yet, I feel it is essential to this manuscript as well and should be added therefore (also the design probably is different to what is shown in Ogren et al. (2017)). A few sentences describing the filter collection with TRAPS in more detail could also be added to the text.

Thank you for the suggestion. We have added a schematic of the TRAPS in Figure 1 and more details about the TRAPS filter collection in section 2.1.

11. Fig. 2: Indicate the exact sampling times (e.g. using a black sphere to mark the beginning of a new sample on the flight track, and a black line on the time color scale)

Sampling times are already shown in Figure 7.

12. Fig. 3: Y-axis and color scale give the same information, one of those seems redundant. A stacked (color-coded by cooling rate) histogram might give more information (or as additional Fig. b). Something like my Figure 1 attached to this review (not your data):



Great idea, we agree the histogram looks much better and have changed to that in Figure 3.

13. Fig. 7: Give the altitude in m AGL (starting from 0). Use more distinct colors for altitude and OPC (instead of light and dark grey). Coloring for Sample 2 is not the same orange as in the others figures. Maybe use logarithmic scale for the OPC concentration?

Done.

14. Fig. 10: Add information to the labeling describing what exactly is depicted on this figure. Is it the onset conditions of the first observed ice nucleation event (or of X % activated fraction)?

Thank you for the suggested clarification, we have changed the caption to read: “Depositional ice nucleation experiments on Samples 1 – 6 plotted by S_{ice} versus temperature. The values plotted here are of the onset conditions of depositional ice nucleation. For our experiments, this refers to the first particle to nucleate ice out of the 10^4 particles deposited on the disc in total, thus a percent activated fraction of 10^{-4} . Although temperatures measured were not exactly -25 °C, -40 °C, and -55 °C, these values are used for brevity for all samples within each grouping shown above.”

15. Fig. 11: What is meant by “most representative particle type per sample”? In section 2.3.2 it says that only 3 – 5 particles were analyzed with Raman spectrometry per sample. How do you know which particle type is representative for the whole sample when only 3 – 5 particles each were analyzed in total?

We have changed the wording here because we meant that the spectrum for each sample is representative of the entire sample. The particles on the disc had little to no variation in spectral features, which is expected from the sample preparation. The solutions that were nebulized were homogeneous. We added the following to clarify this: “Of the

particles that nucleated ice, 3 – 5 particles were analysed for composition using Raman spectrometry for each sample. We assume that a majority of the particles are of similar concentration because the whole sample was dissolved in water, allowed to mix to a homogeneous solution, and nebulized onto the sample disc. Indeed, the particle composition was similar for each particle in any sample, while there was variation from sample to sample.”

The caption for Figure 11 has been modified to read: “Raman spectra for a representative particle per sample. Characteristic vibrational frequencies for functional groups of organics (C-H; 2800 – 3000 cm^{-1}), carbonates (CO_3 ; 1070 – 1090 cm^{-1}), sulphates (SO_4 ; 972 – 1008 cm^{-1}), and nitrates (NO_3 ; 1032 – 1069 cm^{-1}) are noted for reference. Included are images of the particles that initiated depositional freezing for the Raman spectra shown. The length of the black line in each image represents a scale of 20 μm .”

Technical Comments:

Page 2, Line 17: Replace “adroit” with “efficient”

Done.

Page 5, Line 10: Remove “-“ in “1.2-L min⁻¹”

Done.

Page 5, Line 13: Add space between “12 V DC”

Done.

Page 10, Line 23ff: The term “profile” is a little confusing, since it does not correspond to the same samplings (e.g. profile 3 is sample 4). Maybe add a short sentence that the two terms are not the same.

We now define profile as “ascent followed by descent to ground” as provided in the first paragraph of section 3.1.

Page 12, Line 16: Typo in reference “Möhler et al., 2008”

Fixed.

Fig. 1 Caption: b) Picture of the aerosol module.

Fixed.

Fig. 8: Labeling of “UPW in breaker” is covering part of the data. The last sentence of the caption does not make sense for this plot.

Fixed labeling and removed last sentence of caption.

Reviewer 2

Page 1

Line 11. Precipitation processes “can” be modulated by aerosol cloud, not “are” since other dynamics also influence radiative and precipitation processes.

Fixed.

Line 15-20: Upon reading the manuscript and seeing how only 1 successful test has been presented herein, I would soften the ideas about “hovering” at as given altitude. It was clear that the authors struggled to hover at a given altitude because of vertical winds and the balloon could not respond fast enough to maintain altitude. I think one test flight is too soon to claim this.

The reviewer has a valid concern, similar to the first reviewer. As described at the beginning of this response, we have significantly revised the manuscript to soften the “hovering” aspect of the work, given the significant and unexpected challenges associated with such an idea situation, and instead highlight the utility of the aerosol module and evaluation of vertical distributions of INPs.

For realtime aerosol measurements, the vertical profile would still be achievable, but for INP analysis as the authors rightfully acknowledge more time is needed at each sampling altitude to get enough PM on their filter, and this is harder to achieve without fluctuations (sudden rises, or falls). Perhaps it would help if the authors stated a range xx +/- yy m hovering capability. So that instead of being sample to sample at say 500 M agl.. HOVERCAT can sample at 500 +/-100 m as an example.

We have softened the hovering aspect of the system throughout the manuscript to rectify the issue with the hovering flight plan. We also added, “However, this plan was ultimately not executed due to flight complications discussed herein.” to section 2.4. The only places the word “hover” remains in the manuscript is in the HOVERCAT acronym. We also revised the definition of the BBFCS system such that the user can control the ascent and descent of the balloon instead of “controlling the altitude”.

To provide some specific examples of where we modified the text:

- We revised the abstract to highlight the aerosol module and state that we provide recommendations for use of future launched platforms.
- We instead emphasize that the BBFCS has a benefit of being more cost effective and flexible (in terms of FAA regulations, launch locations, and personnel required) than traditional tethered platforms such that it does not require a winch and that we can slow the ascent rate in addition to providing a descent option which, combined, is not a feather attainable with traditional launched balloons. This information is provided in section 2.1.
- We completely revised and reorganized the methods section to clearly define HOVERCAT and describe that first, while describing but not showcasing the balloon platform.
- We revised section 3.4 to emphasize our scientific priorities (discussed in detail in our response to comment 2) and recommendations for future measurements.

Where we do discuss particular altitudes that were somewhat maintained (i.e., in section 3.1) we now provide a \pm range.

Page 2

line 10. An appropriate reference to add or even to replace Cziczo et al. (2017)(who talk about IN methods as a focus not the mechanisms) with would Vali et al. (2015) who have a nice discussion that included the IN community on the terminology and IN mechanisms.

We have added Vali et al. (2015).

Line 12: immersion freezing is the most relevant heterogeneous ice nucleation mechanism for MPCs but not for cloud ice formation. We don't know that, there is a fair bit of literature to suggest that majority of cloud ice may come from secondary ice multiplication. So I suggest re-phrase to make this more clear.

Good point. We have revised the sentence to, “Immersion freezing is the most relevant to primary ice formation in mixed-phase clouds...”

Page 3

Line 1: here the authors say 2 km AGL is a limitation but then the platform they present is only shown to go to 1.1 km AGL, this is contradictory. I suggest correct this or modify the way this is presented.

We assume the reviewer is talking about the top of page 4, but these are discussing limitations of tethered balloons. We do see the point that we only go up to 1.1 km a.g.l. in Colorado, but this limited by the low pressures associated

with this altitude. If, for example, we launched in Seattle, we could go much higher than 1.1 m a.g.l. However, to eliminate any confusion and focus more on the other caveats with tethered balloons, we now state, “In general, tethered balloons can handle much larger payloads than launched systems, but are limited to lower altitudes (i.e., up to approximately 2 km a.g.l. anywhere), have wind condition limitations, and involve more complicated logistics (e.g., use of a winch and personnel required to operate a winch) thus may not be ideal for sampling at various levels where clouds exist.”

Line 4 -5: there may have not been INP measurements on a balloon system, but a similar idea of collecting aerosol on filters/wafers on a UAV was already done couple years ago and published (Schrod et al., 2017). This study should be discussed and acknowledged here.

Reviewer 1 also brought to our attention Schrod et al. (2017) and Ardon-Dryer et al. (2011), which were actual INP measurements from balloon. We have added both Schrod et al. (2017) and Ardon-Dryer et al. (2011) to the introduction, their advantages, and how our results differ from theirs. Schrod et al. (2017) measured INPs by only in the deposition mode. Ardon-Dryer et al. (2011) present results more relevant to ours, but only measured INPs below -18°C in immersion mode on 3 samples and only up to 196 m a.g.l. The fact that only two studies before ours exist, and the limitations of those studies and ours, demonstrates the challenges associated with obtaining vertically profiled INPs, which is important to highlight and why we directly state this after describing their work in the introduction. We also removed the statement, “Additionally, to our knowledge, there have been no measurements of INPs via any balloon platform.”

Ardon-Dryer, K., Levin, Z., and Lawson, R. P.: Characteristics of immersion freezing nuclei at the South Pole station in Antarctica, *Atmos Chem Phys*, 11, 4015-4024, 2011.

Page 5

Line 8: Maybe state what density/shape factor relationships were used to convert OPC data into PM values

We added the density and refractive index values used (default of 1.65 g mL^{-1} and 1.5, respectively).

Line 10: typo should be 1.2 L not 1.2-L

Fixed.

Page 6

This is a nice description of the flight and the testing done. Indeed, it is exciting to read that such developments are taking place. My only concern about this is if two out of the three test flights were not successful and if the altitude of 2.5 km AMSL was determined to be the max altitude range of operation, why weren't more flights conducted for this study, once the operational parameters were clear? This should have been straightforward to do, no? Furthermore, if stronger pumps are being tested for higher altitude (lower pressure) sampling why not wait for those tests so that a test flight with higher altitude can also be conducted.

To address the issue with having only one successful flight, we note that the system *does* work, but with its caveats. Even Ardon-Dryer et al. (2011) that reviewer 1 mentions (i.e., the only other INP measurements by balloon in existence) present only three filters from one day in their paper, indicating a substantial number of flights and samples is not required for publication of what (very) limited INP profiles exist. We emphasize that this is phase I and think it is important to discuss what *didn't* work as that is important to other researchers who may be interested in developing a parallel system in the future, such that they will not have to “reinvent the wheel” so to speak. This is the reasoning behind keeping the original flight plan, such that our idea of stepping is not as easy as one might think. However, we now modified section 3.4 to discuss our recommendations for those interested in controlled launched balloon measurements, including a list of what to do and not to do. This addition provides rationale for keeping in the flight plans, why we had one out of three successful flights, and provides suggestions for improvement. There is a certain stigma with publishing only partially successful results or deployments, but we aim to break that mold and show what didn't work to help those in the future. In addition, we originally intended to showcase the vertical profiling of both

immersion and deposition mode INPs with the aerosol module, which is indeed novel, but have now emphasized that throughout the manuscript.

It is certainly true that higher flow rates would yield larger volumes of air and thus sufficient sample loading for ice nucleation measurements. However, as our manuscript, putting a larger pump on the system would indeed exceed the FAA regulations on payload restriction. We are limited to 2.7 kg or less, and our aerosol module weighs 2.3 kg. We have researched a variety of stronger pumps, none of which weight less than 400 g. We have modified the text in section 3.4 to explain that a larger pump could be used, but likely only on tethered balloon platforms or UAS that can hold a large payload.

Line 22-28: The data given in this paragraph is all for ground level or at a given height? I guess if the goal is vertical profiling, shouldn't the meteorological data for the different height be useful or may be needed to understand the aerosol properties (e.g. phase state) during sampling. Does it help to have all the ground based data when all the sampling is occurring above ground?

As noted, the data were obtained from the CDPHE at their Boulder Reservoir site, although we realize we did not directly state these data were from a ground site, which we now provide. To our knowledge, there are no meteorological data available above the ground near the locations we flew. The goal of describing the general conditions was to paint a picture of the overall weather that day.

Page 7

Line 1: make clear 47 mm is diameter.

Fixed.

Line 25: here the authors say control experiments were done with ultrapure water at various cooling rates. How was the water treated here. In order to have a true control, the water should also have been subjected to the same process as the samples i.e. “(i.e., samples) were successfully collected before the battery died. Each spot was placed in a 29-mL sterile Whirlpak® bag with 2 mL of UPW to resuspend particles deposited on the filter” The water should be treated as above to ensure there is no INP contamination coming from the whirlpak bag. Have such experiments been done? If not, I suggest these should be included in order to really be confident that the freezing is from the aerosol re-suspended into the sample.

The water was from a commercial ultrapure water system as described in the methods, the same UPW that was used for the samples. We did not conduct additional treatment to the UPW. Additionally, we did include spectra in Figure 8 of UPW alone in a beaker, UPW mixed in a WhirlPak® bag for 2 hours, and the filter used (EmFab®) mixed in UPW in a WhirlPak® bag for 2 hours. Thus, we did present results of blanks treated in the same manner as the samples. However, we made this clearer in the caption of Figure 8.

Page 8

Line 4: I understand how the temperature calibration experiment was done, but I tried hard but do not follow how the data are plotted in Figure 3. Which data are for the probe at the center of the copper plate and which for the drop on top of the stage? The vertical lines just mean that the temperature difference doesn't change with cooling rate, but then why are there so many vertical lines, and what determined the position of the data on the x-axis, this figure is difficult to follow. I also don't understand why there is a cooling rate on the y-axis and then the data are also colored by cooling rate?

We have completely revised Figure 3 to show a histogram and eliminate redundant information.

Line 27, why does it take more time to prepare drops with a pipette. Are you using a single tipped pipette, but you could use an 8-channel or 12 channel pipette for this too. It would speed things up.

It takes more time because it is a single pipette instead of a repeater (which we do not have access to). With the normal pipette, we have to draw and expel sample for each drop versus the syringe, which is filled with the whole 0.25 mL

then dropped on the plate. A multichannel pipette is a good idea in theory, but would not allow for the circular pattern of drops on the copper disc and would create a much smaller number of drops (we have already tried this and it did not work).

Page 9

Line 4. It was not clear to me why all 100 drops could not be recorded? If you place 100 drops on a cold stage what is the hindrance in recording data until all 100 drops are frozen?

The limitation of the DFCP is the hindrance: At times, not all drops froze for the control experiments with UPW due to the system detection limit (i.e., it can only go down to approximately -32 to -33 °C). Not all water freezes by these temperatures, so we had times where a few drops remained unfrozen. We more clearly define this in section 2.3.1.

Page 10

Line 10: based on your filter sampling, what was the efficiency of sampling particles larger than 10 μ m? From the deposition mode experiments the authors say they see particles between 1 – 50 μ m. This could be a strong indication of coagulation of the aerosol based on the suspension and nebulization methods used. Does it then make sense to assume that one particle lead to each ice crystal observed? And also try and analyses that particle? It is likely a particle that is composed of multiple particles. With more samples perhaps the authors could do a particle count by feeding the nebulized spray though an SMPS or even into a CPC to get a particle count. And then compare this to a particle count taken from image analysis of the deposition state to see if numbers are similar. This will provide some correction factor (if needed) for how much coagulation maybe taking place when the particles are being deposited onto the stage for deposition nucleation.

According to Ogren et al. (2017), there is an 80% collection efficiency of 10- μ m particles, but according to their Figure 2, the instrument can collect at larger sizes, just at a lower efficiency. However, it is important to note that the sampling efficiencies were calculated, and not actually measured. We have added this information to section 2.1, stating, “The TRAPS has the highest collection efficiency for particles in the 1 nm – 10 μ m aerodynamic diameter range—with particle losses of less than 10% for 5 nm – 7 μ m particles and less than 1% for 30 nm – 2.5 μ m particles at 1.0 L min⁻¹—but can collect particles with larger diameters (Ogren et al., 2017).”

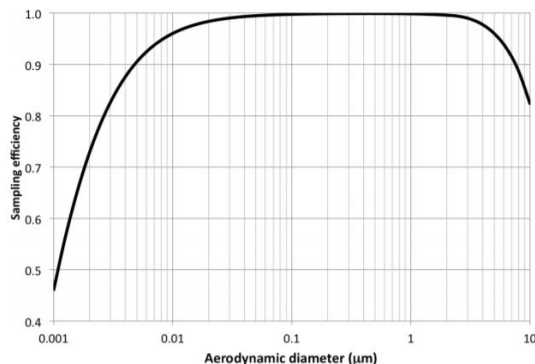


Figure 2. Calculated sampling efficiency of particles reaching the CLAP filter.

A more detailed experimental setup would be ideal, however, due to the limited amount of sample, we were not able to run extra experiments with size distribution measurements. We did add some text describing the caveats “Although the Raman spectral and ice nucleation analyses are helpful to observe the overall particle composition as temperature and relative humidity are changed, the experiment does not determine the size or mixing state of the particles as they were in the atmosphere. Further, the spectral resolution of 1 micrometre in our system does not allow smaller scales to be distinguished within the individual particles probed.”

Line 25-30: regarding sudden drops and hovering – see comment above that I made in abstract section. I think it is too soon to claim the ability to hover at a fixed altitude. Perhaps consider putting a range to that, given that the balloon

cannot respond fast enough to the updrafts experienced. I still think this method is super valuable even for example you say, each INP filter sample is conducted within a range of 100 m or so. This is a step in the right direction for vertical profiling

We completely agree with the reviewer that it is too soon to claim the ability to hover. We have significantly revised the manuscript to avoid this issue as discussed in our response to the reviewer's last comment on page 1.

Page 11

Section 3.2. Results are indeed interesting, but it would have been nice to see that each sample presented did not have overlapping altitudes. Is there no way to automatically shut off filter sampling when a certain threshold of a sudden rise or fall is crossed and then only resume sampling when the balloon is within this threshold again? Would this be too demanding, or is there not enough time in the flight for this. This way you ensure that your INP filter samples are restricted to sampling at a certain altitude and you can resolve better the vertical profiling of the INPS. For example in Figure 9, sample 2, if the sampling was shut off for the periods when the instrument was below 600 m AGL, that would allow sample 2 to be representative of only INPs above 600m AGL.

This is certainly a feature of interest, but extensive work would need to be executed to incorporate bidirectional communications yet maintain payload weight under the FAA regulations. It would be ideal to enable ground-based bidirectional commands to change the sample spot when desired. However, the manuscript presents a pilot study focusing on specific priorities: (1) Can we recover the system? And (2) Can we control the ascent and descent? Both priorities fall under the umbrella of making a system that is relatively affordable, user friendly, and ensuring avoidance of complicated FAA paperwork by obtaining a COA. Adding complexity to the system for controlling altitude and sampling frequency adds cost, weight, and more variables that could fail. For clarity, we now emphasize that this was a pilot study throughout the manuscript (in addition what we already call "test" flights) and discuss our priorities in section 3.4.

More specifically, a bidirectional communication feature entails a significantly more advanced ground-based system and would add too much weight to HOVERCAT through additional processing hardware, an additional receiver, and batteries needed to control in such a manner. First, the BBFCS had bidirectional communication, but additional channels would need to be added to control the TRAPS as well, adding to the cost. And second, adding the necessary components to the TRAPS would add to both cost and weight, and given our 400 g buffer (i.e., calculated from the 2700 g limit and our HOVERCAT 2300 g weight), would likely push the system into a weight category that requires either restricted airspace or significant paperwork to obtain and FAA COA to fly outside of restricted airspace areas. Both of these issues make flying HOVERCAT more complicated and expensive, which fall outside of our vision to make an affordable and easy-to-use system.

Section 3.3: should be deposition nucleation – not freezing, freezing implies liquid to solid transition, I think the authors' intent is to imply vapour to ice mechanism here, therefore I would take freezing out of the phrase.

Thank you for pointing this out. We have changed 'deposition freezing' to 'deposition nucleation' throughout the manuscript.

Page 12

Line 1: "...above homogenous freezing" are you referring to temperature or S_i ? Could clarify here already. Based on Fig. 10 it looks like you refer to both. However, some of your data points are quite close to water saturation (-25 for example), so it maybe a tough sell to claim those as be depositional ice nucleation.

Thanks for that catch. We mean the S_{ice} values are below that required for homogeneous freezing. For the lower temperatures, we are confident that we are observing depositional ice nucleation, but for the highest temperature it could be subsaturated immersion nucleation. This text was modified from the original version and we removed that statement.

Section 3.3: I like the discussion in this section. But here one must acknowledge that there could be artifacts of having your INPs as agglomerates of particles. So maybe the aerosol was externally mixed, but with the experimental method used, they could become or appear as internally mixed since you suspend and nebulize and then allow for evaporation to retain the residual particles on the cold stage. This should allow for some coagulation, and perhaps the Raman is investigating a single particle that is a result of multiple smaller coagulated particles.

We have added the following to address this: “Although the Raman spectral and ice nucleation analyses are helpful to observe the overall particle composition as temperature and relative humidity are changed, the experiment does not determine the size or mixing state of the particles as they were in the atmosphere. Further, the spectral resolution of 1 micrometre in our system does not allow smaller scales to be distinguished within the individual particles probed.”

Section 3.4: Again while reading this section, I can't help but feel that the paper is a little premature. Some of the plans and developments described herein could be already part of this manuscript, like the stronger pumps, a few more test flights, certainly more than 1, and the practice to control the balloon to stay at desired altitudes. i.e., the above should be part of Phase I. I understand regulatory approval work like compliance with FAA can be phase II as well as launching on other airborne platforms (UAS, reverse parachute). i.e. demonstrating their instruments are versatile enough for other platforms, can all be phase II.

We have completed overhauled this section to discuss possible improvements and recommendations for future flights as discussed in the responses above. We realize that there are improvements that could be made, but we aim to clearly highlight our phase I scientific objectives and questions, by starting simple and developing a system that can be deployed under limited resources. We hope by shifting the focus of section 3.4 to our recommendations that others can help contribute to improvement of the technology and contribute to INP measurements via balloon, which are extremely limited.

Page 13

Line 5. Certainly if the plans to operate at Jungfraujoch in Spring 2018 are on track (that is now), then stronger pumps have been implemented already. Since ground level at Jungfraujoch is already ~ 650 mb pressure.

We removed this part as deployment on Jungfraujoch was not executed.

Line 14: maybe put in also the altitude AGL to give context to your starting point.

Done.

Line 19-20: Soften the vertical profiling statement here because the authors showed overlapping vertical profiles, so it is not yet achievable in the strictest of sense.

Done.

Given that this is an instrument development paper, I would like to see a section on Benefits and limitations of the instrument. Where the authors state this clearly. I believe some of this is interspersed through the manuscript, but I think this should be brought together in one section to make it clear what are the benefits of HOVERCAT (and its current limitations).

Done. We have made this information more clear in section 3.4.

Figure 4. For clarity that the control experiments are done with pure water and also it would be good to indicate the volume of the drops in the caption.

Done.

Figure 5. I was a little confused – the “by hand” drops, are those not the same as the syringe drops? Or how were the 2.5 ul drops by hand produced? There must have been some sort of tool for these.

The 'by hand' is technically 'by syringe', which we now changed in the legend and caption for clarity.

Figure 7: Could you make it clear in the caption that the grey line plot is the one corresponding to the altitude and the scatter plot corresponds to the concentration from the OPC? Is it mentioned what densities or shape factors have been assumed to come up with the PM values from the OPC data?

We have revised this figure to better distinguish altitude and OPC number concentrations. We also now provide the density and refractive index used to estimate the PM concentrations.

Were the particles losses of 10% accounted for in the results? i.e. when calculating the concentrations in Fig. 8. Also, how were the control experiments for the ultra pure water in beaker and bag converted to INP concentrations? These fraction curves are not addressed sufficiently in the text I think.

We did not account for particle losses of 10%, because according to Ogren et al. (2017), losses vary depending on particle shape, which is difficult given we do not know the size distribution of the particles collected on our filter.

We now include discussion at the end of section 2.3.1 about how INP concentrations from the blanks were obtained. We assumed an average volume of air based on the samples collected and applied that to the Vali (1961) equation.

HOVERCAT: A novel aerial system for evaluation of aerosol-cloud interactions

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10 **Abstract.** Aerosols have a profound impact on cloud microphysics through their ability to serve as ice nucleating particles (INPs). As a result, cloud radiative properties and precipitation processes ~~are can be~~ modulated by such aerosol-cloud interactions. However, one of the largest uncertainties associated with atmospheric processes is the indirect effect of aerosols on clouds. The need for more advanced observations of INPs in the atmospheric vertical profile is apparent, yet most ice nucleation measurements are conducted at the ground or during infrequent and intensive airborne field campaigns. Here, we

15 describe a novel measurement platform that is less expensive and smaller (< 5 kg) when compared to traditional aircraft and tethered balloon platforms and that can be used for evaluating two modes of ice nucleation (i.e., immersion and deposition). HOVERCAT (Honing On VERTICAL Cloud and Aerosol properTies) flew during a pilot study a successful aerosol collection flight in Colorado, USA up to 2.6 km above mean sea level (1.1 km above ground level). ~~HOVERCAT and~~ consists of ~~the Boomerang Balloon Flight Control System that has the capability to “hover” at desired altitudes and~~ an aerosol module that

20 includes an optical particle counter for size distributions (0.38 – 17 μm in diameter) and a new time resolved sampler that collects up to ten filter samples for offline ice nucleation and aerosol analyses on a launched balloon platform. During the May 2017 test flight, total particle concentrations were highest closest to the ground (up to 50 cm^{-3} at < 50 m above ground level) and up to 2×10^2 particles were ice nucleation active in the immersion mode (at $-23\text{ }^\circ\text{C}$). The warmest temperature immersion and deposition mode INPs (observed up to $-6\text{ }^\circ\text{C}$ and $-40.4\text{ }^\circ\text{C}$, respectively) were observed closest to the ground, but overall

25 INP concentrations did not exhibit an inverse correlation with increasing altitude. HOVERCAT is a prototype that can be further modified for other airborne platforms, including tethered balloon and unmanned ~~aerial aircraft~~ systems. The versatility of HOVERCAT affords future opportunities to profile the atmospheric column for more ~~routine comprehensive~~ evaluations of aerosol-cloud interactions. Based on our test flight experiences, we provide a set of recommendations for future deployments of similar measurement systems and platforms.

1 Introduction

Some of the least understood atmospheric processes are aerosol-cloud interactions, and specifically, those with aerosols that serve as ice nucleating particles (INPs) (Boucher et al., 2013). Formation and microphysical modulation of cloud droplets and ice crystals is highly dependent upon the types and number of aerosols that serve as cloud condensation nuclei (CCN) and INPs. In the absence of CCN and INPs, clouds would in theory require $> 400\%$ humidity and $< -36\text{ }^\circ\text{C}$ to form droplets and ice crystals, respectively; conditions atypical of mixed-phase clouds (Pruppacher and Klett, 1997). Aerosol-induced microphysical modifications influence cloud lifetime and albedo (Morrison et al., 2005), as well as the production of more or less precipitation, particularly in mixed-phase cloud systems. INPs nucleate ice through pathways dependent upon temperature, saturation with respect to ice, and the INP type (Hoose and Möhler, 2012). The modes of heterogeneous ice nucleation include:

- 1) condensation freezing whereby ice is formed concurrently with the initial formation of liquid on CCN at supercooled temperatures,
- 2) immersion freezing whereby an INP is immersed in an aqueous solution or water droplet via activation of CCN during liquid cloud formation,
- 3) contact freezing whereby an INP approaches the air-water interface of a droplet (e.g., via a collision) and initiates freezing, ~~and~~
- 4) deposition ~~freezing-nucleation~~ whereby ice is ~~nucleated-formed~~ from supersaturated vapour with respect to ice ($RH_i > 100\%$) on an INP directly, ~~and~~
- 5) pore condensation and freezing whereby water vapour is condensed into voids and cavities followed by glaciation (Coluzza et al., 2017; Cziczo et al., 2017; Hoose and Möhler, 2012; Kanji et al., 2017; Marcolli, 2014; Vali et al., 2015).

Immersion freezing is the most relevant to primary ice formation in mixed-phase clouds ~~ice formation~~ and requires that INPs initially serve as, or in conjunction with, CCN, whereas deposition ~~freezing-nucleation~~ is prevalent in mixed-phase and dominant in cirrus cloud ice formation (Kanji et al., 2017). Aerosols such as mineral dust, soil dust, sea salt, volcanic ash, black carbon from wildfires, and primary biological aerosol particles (PBAPs) have been shown to serve as INPs (Conen et al., 2011; Cziczo et al., 2017; DeMott et al., 1999; Hoose and Möhler, 2012; McCluskey et al., 2014; Murray et al., 2012; Petters et al., 2009). Among these, dust and PBAPs are the most ~~adroit-efficient~~ INPs found in the atmosphere (Cziczo et al., 2017; Murray et al., 2012). Dust is the most atmospherically-abundant INP, forming ice as warm as $-10\text{ }^\circ\text{C}$, but primarily at temperatures $< -15\text{ }^\circ\text{C}$ (Hoose and Möhler, 2012; Murray et al., 2012). On the other hand, PBAPs are relatively rare in the atmosphere, but can form ice as warm as $-1\text{ }^\circ\text{C}$ (Despres et al., 2012; Schnell and Vali, 1976; Vali et al., 1976; Vali and Schnell, 1975). However, constraining aerosol-cloud impacts in models ranging from the cloud-resolving to climate scales, specifically when parameterizing INPs, remains a significant challenge due to limited observations (Coluzza et al., 2017; Cziczo et al., 2017; DeMott et al., 2010).

A number of previous ground-based field measurements dating back to the 1950s have provided noteworthy advancements in understanding the sources and efficiencies of INPs (e.g., Bigg, 2011; Durant et al., 2008; Garcia et al., 2012; Huffman et al., 2013; Jayaweera and Flanagan, 1982; Mason et al., 2015; McCluskey et al., 2014; Mossop, 1963; Murray et al., 2012; Petters et al., 2009; Prenni et al., 2009b; Prenni et al., 2013). Further, previous work has evaluated INP concentrations and at times

composition in detritus, soil, water from lakes and oceans, surface microlayers, and precipitation samples to assess INP sources (e.g., Conen et al., 2016; Creamean et al., 2014; DeMott et al., 2016; Hill et al., 2016; Irish et al., 2017; Moffett, 2016; O'Sullivan et al., 2014; Petters and Wright, 2015; Pietsch et al., 2017; Pouzet et al., 2017; Schnell, 1977; Schnell and Vali, 1972, 1973, 1975; Stopelli et al., 2015; Tobo et al., 2014). Analysis of INPs in precipitation samples take a step in the direction
5 of vertical profiling of INPs, making the assumption that the INPs in precipitation are what initiated ice formation in the clouds above; however, there are caveats associated with artefacts from scavenging during raindrop or snowflake descent, aerosolization methods, and redistribution of residue particles in collected liquid precipitation samples (Creamean et al., 2014; Hanlon et al., 2017; Petters and Wright, 2015).

Although observations at the ground afford detailed information regarding the characterization of INP sources, they may not
10 be representative of INPs in the atmospheric column, where they have the direct ability to impact cloud ice formation processes and may originate from a range of local to long-range transported sources. As a result, several INP quantification and characterization studies have been conducted in clouds at mountaintop atmospheric research facilities, such as Storm Peak Laboratory in the United States (Baustian et al., 2012; Cziczo et al., 2004; Richardson et al., 2007), Puy de Dôme in France (Joly et al., 2014; Joly et al., 2013), and Jungfraujoch in Switzerland (Chou et al., 2011; Conen et al., 2015; Stopelli et al.,
15 2017; Stopelli et al., 2016). Such studies provide routine or long-term measurements of INPs in clouds, yet one disadvantage is that profiling is not possible. Vertical profiling of INPs can serve as a connection between the ground and various altitudes below, in, and above cloud. Targeted aircraft campaigns have helped explain the role of INPs in cloud ice formation at all levels from below cloud, cloud base, in-cloud, and cloud top (e.g., Avramov et al., 2011; Creamean et al., 2013; Curry et al., 2000; DeMott et al., 2010; DeMott et al., 2003; Pratt et al., 2009; Prenni et al., 2009a; Rogers et al., 2001; Rogers et al., 1998;
20 Schnell, 1982). Although such campaigns yield results crucial for understanding the vertical distribution of INPs in cloudy environments, they are intensive with regard to personnel, cost, and time.

Overall, a key gap in ice nucleation research is routine vertical profiling of INP abundance, efficiency, and chemical and physical characterization (Coluzza et al., 2017). Tropospheric measurements via balloon-based systems have been a desirable means of measuring aerosol properties on an inexpensive and thus, more frequent basis. However, such measurements can be
25 limited in terms of time, measurements made, or location. For example, long-term records of tropospheric aerosol particle size distributions have been reported in Wyoming, United States (i.e., 20 years) (Hofmann, 1993). The same launched balloon system was deployed in Antarctica, demonstrating the utility of this platform in multiple environments (Hofmann et al., 1989). Particle size distributions have also been measured via launched balloons in several locations in China using optical particle counters (Iwasaka et al., 2003; Kim et al., 2003; Tobo et al., 2007). One major caveat with these studies is that it is not clear
30 if the balloon systems were retrievable, given their maximum flight ceilings were located well into the stratosphere. In addition, the launched balloon platforms provide information on 1 – 2 aerosol profiles (i.e., ascent and sometimes descent) and are limited by payload weight. Particle spectrometers have also been deployed and retrieved on tethered balloon systems (de Boer

et al., 2018; Greenberg et al., 2009; Maletto et al., 2003; Renard et al., 2016; Siebert et al., 2004; Wehner et al., 2007), affording information on aerosol layer locations and evolution by means of multiple profiles. A few studies have deployed miniature aerosol filter samplers on launched or tethered balloon systems, yielding information on aerosol chemistry (Hara et al., 2011; Rankin and Wolff, 2002); ~~however, such samplers contained one filter per flight, thus providing information on aerosol properties at only one altitude (i.e., not a profile).~~ ~~A noteworthy study by Ardon-Dryer et al. (2011) consisted of measurements of immersion mode INP concentrations from a tethered balloon flight in Antarctica, although only at temperatures below -18°C from three filters collected below 200 m above ground level (a.g.l.).~~ ~~however, such samplers contain one filter per flight, thus providing information on aerosol properties at only one altitude (i.e., not a profile).~~ In general, tethered balloons can handle much larger payloads than launched systems, but are limited to lower altitudes (i.e., up to approximately 2 km ~~above ground level (AGL a.g.l. anywhere)~~), and have wind condition limitations, ~~and involve more complicated logistics (e.g., use of a winch and personnel required to operate a winch)~~ thus may not be ideal for sampling INPs in all conditions ~~at various levels where clouds exist.~~ Schrod et al. (2017) present INP measurements from several flights using unmanned aircraft systems (UASs) over the Eastern Mediterranean, but only in the deposition nucleation mode. To our knowledge, the results from Ardon-Dryer et al. (2011) and Schrod et al. (2017) are the only reported vertical INP measurements using smaller, unmanned systems. ~~The fact that only two published studies exist, in addition to the limitations of such studies (and our limitations as discussed in more detail herein) demonstrate the challenges associated with obtaining INP measurements aloft.~~

Overall, both launched and tethered balloon platforms, ~~and UASs,~~ have their advantages and disadvantages in terms of flight ceiling, profiling, retrieveability, ~~cost, operational logistics,~~ and payload restrictions. ~~Additionally, to our knowledge, there have been no measurements of INPs via any balloon platform.~~ A solution to reduce the limitations of these methods is a launched balloon system that can be controlled in terms of ~~altitude ascent and descent,~~ ~~to afford~~s multiple profiling and payload retrieval capabilities, and ~~a system that~~ collects aerosol loadings sufficient for altitude-resolved offline ice nucleation measurements. Here, we present ~~a measurement system called HOVERCAT (Honing On VERTICAL Cloud and Aerosol properTies) deployed on an an~~ experimental launched balloon system, ~~called HOVERCAT (Honing On VERTICAL Cloud and Aerosol properTies), that possesses such capabilities that possesses such capabilities.~~

25 2 Methods

The first prototype of HOVERCAT was recently built and tested in Colorado, United States, consisting of ~~an aerosol module for measuring real-time particle size distributions and a miniaturized filter sampler for aerosol collection for offline ice nucleation analyses.~~ ~~The novel~~ balloon platform, ~~capable of ‘hovering’ at desired altitudes~~ called the ~~Boomerang Balloon Flight Control System (BBFCS)~~ the Boomerang Balloon Flight Control System (BBFCS), was used to fly HOVERCAT. ~~Additionally, HOVERCAT contains an aerosol module for measuring real-time particle size distributions and a time-resolved filter sampler for aerosol collection for offline ice nucleation analyses.~~ The current version of HOVERCAT is experimental,

thus we consider it as in Phase I of its development, and is described herein. As discussed later, we provide future directions for modification and improvement of HOVERCAT and recommendations for non-tethered balloon systems in general for future deployments.

2.1 HOVERCAT: The aerosol instrumentation package

5 The aerosol module package contains: 1) an optical particle counter (Alphasense OPC-N2) for particle size distributions (16 size bins for 0.38 – 17 μm in diameter) and estimated particle mass concentrations with optical diameters $< 1, 2.5,$ and $10 \mu\text{m}$ ($\text{PM}_{1,}$ $\text{PM}_{2.5,}$ and $\text{PM}_{10,}$ respectively) and 2) the National Oceanic and Atmospheric Administration (NOAA)-built miniaturized Time-Resolved Aerosol Particle Sampler (TRAPS) for collection of up to 10 samples. The time resolution can be set at the desired rate, but was set for 30 minutes in the current study. The OPC-N2 operates at 175 mA in operation mode and weighs 105 g. Flow rates are adjusted based on ambient pressure to maintain a 1.2 L min^{-1} flow using a patented ‘pump-less’ design. Data are stored on a microprocessor within the OPC during collection. A default density of 1.65 g mL^{-1} and refractive index of 1.5 were used to estimate particle mass concentrations. The TRAPS design is based on the filter components of the NOAA Continuous Light Absorption Photometer (CLAP), without the optical components and measurements (Ogren et al., 2017) (Figure 1a). It is connected to a small 12 V DC vacuum pump (Brailsford & Co., Inc. TD-4X2N), which nominally enables a flow rate of approximately $1.2 \pm 0.1 \text{ L min}^{-1}$ through the TRAPS when a 47-mm diameter filter with $0.2 \mu\text{m}$ pore size is in place. A Honeywell AWM43600V mass flow meter measures sample flow rate. Ten miniature solenoid valves select the active sample spot and are controlled by an on-board microprocessor preselected for the desired time resolution, which was 30 minutes per sample spot for the HOVERCAT test flights. The TRAPS flow rate at 30 minutes provides approximately 40 total litres of air through each spot, which is ideal for measuring more realistic INP concentrations (Mossop and Thorndike, 1966). Sample loaded spots average to a coverage area of 19.9 mm^2 (equates to a spot diameter of approximately 4.46 mm). The TRAPS has the highest collection efficiency for particles in the 1 nm – 10 μm aerodynamic diameter range—with particle losses of less than 10% for 5 nm – 7 μm particles and less than 1% for 30 nm – 2.5 μm particles at 1.0 L min^{-1} —but can collect particles with larger diameters (Ogren et al., 2017).

25 The TRAPS, micropump, and OPC are all operated by battery: the TRAPS and micropump run off a battery pack containing three 18650 rechargeable Li-ion batteries (Panasonic NCR18650B, 12 V output, 3400 mAh) and the OPC runs off one rechargeable battery (Anker PowerCore 5000, 5 V output, 5000 mAh). The OPC can operate for several days on its portable battery, while the TRAPS and pump can operate for up to 5 hours on its battery pack. Both the TRAPS and OPC are connected to inlets composed of an 8-inch segment of 1/4-inch ID black conductive tubing connected to a stainless-steel funnel (5 cm in diameter) with the opening covered with stainless steel mesh. All components are seated in a foam enclosure with removable lid and inlets extending out of the bottom (Figure 1b).

2.12 Balloon platform BBFCS: The balloon platform

~~BBFCS-The BBFCS~~ is a real-time, remote device that allows the user to control the ~~altitude-ascent and descent~~ of standard latex weather balloons (Figure 1**bc**~~a~~). The primary features are a lift-gas vent valve in the control module that permits negative buoyancy adjustments and a sand ballaster (i.e., ballast module) that permits positive buoyancy adjustments. Buoyancy adjustments as small as 5 g of lift are possible. For example, if a faster or slower ascent is desired, ballast can be dropped or venting can be done, respectively. If descent is desired, a longer and faster venting is applied. Due to the ability to slow down the fall speed by a combination of the appropriate amount of venting and dropping ballast, if needed, landing the system is relatively gentle and did not result in instrumental damage during the test flights. It is possible the balloon itself can be reused (i.e., we used the same balloon for two flights).

Two-way communication is achieved through a 70-cm line-of-sight LoRa radio link. The system features a 1/4W transceiver that uses a low baud rate and a slow 4-second time-division multiple access (TDMA) cycle to achieve ranges in excess of 300 km. The system also features redundant termination methods, anti-collision strobes, positioning, and flight sensors. A recovery parachute is included for emergency termination and faster fall speeds than slow balloon deflation. The BBFCS was manually controlled for this project. We utilized a software interface on a ground-based computer to analyse the real-time flight conditions and send the necessary buoyancy control commands to achieve the desired flight profile. We drove in the approximate trajectory of the balloon in order to stay within the 300-km communications range, thus were able to physically retrieve it when it ultimately landed. Early morning launches were conducted to maximize the calm low-troposphere atmospheric conditions as flight control is much easier in such conditions. Because this project entailed low-altitude flights that did not exceed 9.6 km above mean sea level (~~AMSLa.m.s.l.~~) or approximately 8.1 km a.g.l., 300 g latex balloons were used. These relatively small balloons, for a 3.9-kg payload, ensured that the envelope was always under tension and would expel lift-gas whenever the vent valve was opened, while ensuring that the burst altitude was above the expected operational altitude. Burst altitude was calculated to be 13 – 14 km ~~AMSLa.m.s.l. (11.5 – 12.5 km a.g.l.)~~ depending on how much lift gas had been vented. The BBFCS is designed to allow Federal Aviation Administration (FAA) part 101 exempt flights, even when carrying a reasonably-sized payload (i.e., total payload weight of less than 5.5 kg and no one module greater than 2.3-~~7~~ kg).

~~2.2 Aerosol module~~

~~The aerosol module package contains: 1) an optical particle counter (Alphasense OPC N2) for particle size distributions (16 size bins for 0.38 – 17 µm in diameter) and estimated particle mass concentrations with optical diameters ≤ 1, 2.5, and 10 µm (PM₁, PM_{2.5}, and PM₁₀, respectively) and 2) the National Oceanic and Atmospheric Administration (NOAA) built miniaturized Time Resolved Aerosol Particle Sampler (TRAPS) for collection of up to 10 samples. The OPC N2 operates at 175 mA in operation mode and weighs 105 g. Flow rates are adjusted based on ambient pressure to maintain a 1.2 L min⁻¹ flow using a patented ‘pump less’ design. Data are stored on a microprocessor within the OPC during collection. The TRAPS design is based on the filter components of the NOAA Continuous Light Absorption Photometer (CLAP), without the optical components and measurements (Ogren et al., 2017). It is connected to a small 12 VDC vacuum pump (Brailsford & Co., Inc.~~

TD-4X2N), which nominally enables a flow rate of approximately $1.2 \pm 0.1 \text{ L min}^{-1}$ through the TRAPS when a 47-mm filter is in place. A Honeywell AWM43600V mass flow meter measures sample flow rate. Ten miniature solenoid valves select the active sample spot and are controlled by an on-board microprocessor preselected for the desired time resolution, which was 30 minutes per sample spot for the HOVERCAT test flights. The TRAPS flow rate at 30 minutes provides approximately 40 total litres of air through each spot, which is ideal for measuring more realistic INP concentrations (Mossop and Thorndike, 1966). Sample loaded spots average to a coverage area of 19.9 mm^2 (equates to a spot diameter of approximately 4.46 mm). The TRAPS collects particles in the 1 nm – 10 μm aerodynamic diameter range, with particle losses of less than 10% for 5 nm – 7 μm particles and less than 1% for 30 nm – 2.5 μm particles at 1.0 L min^{-1} (Ogren et al., 2017).

The TRAPS, micropump, and OPC are all operated by battery: the TRAPS and micropump run off a battery pack containing three 18650 rechargeable Li ion batteries (Panasonic NCR18650B, 12 V output, 3400 mAh) and the OPC runs off one rechargeable battery (Anker PowerCore 5000, 5 V output, 5000 mAh). The OPC can operate for several days on its portable battery, while the TRAPS and pump can operate for up to 5 hours on its battery pack. Both the TRAPS and OPC are connected to inlets composed of an 8 inch segment of 1/4 inch ID black conductive tubing connected to a stainless steel funnel (5 cm in diameter) with the opening covered with stainless steel mesh. All components are seated in a foam enclosure with removable lid and inlets extending out of the bottom (Figure 1b).

2.4 Test flight details

The overall launch mass of HOVERCAT was 4250 g with 450 g of free lift to achieve an initial 3 m s^{-1} ascent rate. System masses were: 350 g for the balloon and connection spindle, 900 g for the BBFCS control module and parachute, 2300 g for the aerosol module HOVERCAT, and 700 g of ballast. Initial flight planning called for a 5-step flight profile with 500-m altitude steps. This allocated 100 g of ballast per step, 1.5 m s^{-1} anticipated ascent rate between steps, with a 200-g reserve for the flight to help maintain the desired altitude. However, this plan was ultimately not executed due to flight complications discussed herein. The flight train for this project consisted, from top to bottom: latex balloon, valve and flight computer modules, 500 mm of line, aerosol module, 500 mm of line, and ballast module (Figure 1e1d). The recovery parachute was attached to the bottom of the flight computer module and hung off to the side. The parachute's apex was attached to the termination clamp and was released by this clamp during termination or by aerodynamic drag if the balloon had prematurely burst. The OPC was started during balloon inflation and the TRAPS and micropump were started via Bluetooth just prior to take off. ~~HOVERCAT-Two~~ also had two miniature cameras (Mobius Basic ActionCam with wide angle lens) were mounted to and facing the BBFCS valve module and aerosol module HOVERCAT for time lapse photos during take-off, flight, and landing.

Three test flights were conducted in central Colorado during 24 – 26 May 2017. Two of the three flights had instrument operational issues (i.e., 24 and 26 May), so only data from the 25 May flight is presented herein. Briefly, communications

were lost during the 24 May flight and as a result, controlling the valve and ballast modules was not possible. HOVERCAT The system reached 9.68.1 km AMSL a.g.l. and ambient pressure was too low for the TRAPS pump to operate. The 26 May flight reached > 2 km AMSL a.m.s.l. (> 500 m a.g.l.), in which the TRAPS pump also did not operate correctly. For both the 24 and 26 May flights, the total volume of air pulled through the filters was 1 – 12 L above 2.5 km AMSL a.m.s.l. (1.1 km AGL), equating to loadings too low for offline analyses (i.e., calculated INP concentrations were below detection limits). Based on the successful 25 May flight and unsuccessful flights on 24 and 26 May, we have concluded that in its current configuration, HOVERCAT can operate below 2.5 km AMSL a.m.s.l., otherwise at the low pressures, the current micropump cannot generate sufficient flow. New, higher volume pumps are being tested.

The 3-dimensional flight path for 25 May is shown in Figure 2. The horizontal distance between launch and landing was 16.8 km, bird's eye view. Conditions were partly cloudy with surface air temperatures ranging from 16 – 21 °C, relative humidity from 35 – 47%, and wind speeds from 2 – 3 m s⁻¹ from the north and south (hourly meteorological data during flight times obtained from the Colorado Department of Public Health and Environment (CDPHE) at the Boulder Reservoir ground site; 40.07°N, 105.22°W; <https://www.colorado.gov/pacific/cdphe/data>). HOVERCAT did not fly through the clouds present that day, but remained below cloud base, based on visual identification of the system while tracking in real-time (i.e., HOVERCAT the system was always in line-of-sight).

2.3 Offline ice nucleation analyses

2.3.1 Drop freezing assay for immersion mode ice nucleation

For the 25 May flight, aerosol samples were collected on 47 mm filters (Pallflex® EmFab™). Pre-treatment of the filters by means of a 6 N nitric acid bath (Certified ACS Plus, Fisher Scientific), 3 times rinse with ultrapure water (UPW; Barnstead™ Smart2Pure™ 6 UV/UF), and baking at 150 °C for 30 minutes, was conducted to remove possible filter INP artefacts. Out of the filters tested, EmFab™ possessed the lowest contribution from artefacts compared to cellulose nitrate and polytetrafluoroethylene and survived the pre-treatment process.

Immersion mode freezing was tested using a drop freezing assay (DFA) cold plate apparatus. This cold plate technique was based on previous but slightly modified apparatuses (Hill et al., 2016; Stopelli et al., 2014; Tobo, 2016; Wright and Petters, 2013). For brevity, we call this system the NOAA Drop Freezing Cold Plate (DFCP). Following collection and prior to analysis, sample filters were stored frozen for approximately six months. After removing from the freezer, each sample spot was carefully cut and separated from the 25 May filter; ~~only~~ six spots (i.e., samples) were successfully collected before the battery died. Each spot was placed in a 29-mL sterile Whirlpak® bag with 2 mL of UPW to resuspend particles deposited on the filter. The bags were sealed and shaken at 500 rpm for two hours (Bowers et al., 2009). Copper discs (76 mm in diameter, 3.2 mm thick) were prepared by cleaning with isopropanol (99.5% ACS Grade, LabChem. Inc.), then coated with a thin layer of

petrolatum (100%, Vaseline®) (Bowers et al., 2009; Tobo, 2016). Three of the spots on the filter had visible aerosol deposits that were successfully transferred to the UPW (i.e., based on visual identification).

Following sample preparation, a sterile, single-use syringe was used to draw 0.25 mL of the suspension and 100 drops were pipetted onto the petrolatum-coated copper disc, creating an array of ~2.5- μ L aliquots. Drops were visually inspected for size; however, it is possible not all drops were the same exact volume, which could lead to a small level of indeterminable uncertainty. However, previous studies have elucidated that drops need to be orders of magnitude different in volume to significantly perturb the freezing temperature from drop size, alone (Bigg, 1953; Hader et al., 2014; Langham and Mason, 1958). The copper disc was then placed on a thermoelectric cold plate (Aldrich®) and covered with a transparent plastic dome. Small holes in the side of the dome and copper disc permitted placement of up to four temperature probes using an Omega™ thermometer/data logger (RDXL4SD). The Omega™ meter has a 0.1 °C resolution and accuracy of \pm (0.4% + 0.5 °C) for the K sensor types used. During the test, the cold plate was cooled at 1 – 10 °C min⁻¹ from room temperature until all drops on the plate were frozen or until the DFCP detection limit –of approximately –32 to –33 °C. Control experiments with ~~ultrapure water~~UPW at various cooling rates within this range show no discernible dependency of drop freezing on cooling rate, akin to previous works (Vali and Stansbury, 1966; Wright and Petters, 2013). Frozen drops were detected visually, but recorded through software written in-house, providing the freezing temperature and cooling rate of each drop frozen. For the control experiments with UPW, some experiments resulted in unfrozen drops at the DFCP lower temperature limit, thus, the fraction frozen was calculated from the number of drops detected, including the unfrozen remaining, which is the reason why not all fractions frozen = 1. However, all drops froze for tests with blanks for the sample handling and the samples themselves. The test continued until all 100 drops were frozen, typically occurring around –30 °C. Each sample was tested three times with 100 new drops for each test. From each test, the fraction frozen and percentage of detected frozen drops were calculated. The results from the triplicate tests are then binned every 0.5 °C to produce one spectrum per sample.

Although the methodology behind DFA is well established, control experiments were conducted with UPW for full system characterization of the DFCP. First, temperature differences were measured within the range of cooling rates using UPW on petrolatum-coated copper discs between the centre of the disc (thermocouple inserted in a small diameter hole in the side of the disk) and a drop on top of the plate with a thermocouple inserted into the drop (Figure 3). As expected based on previous work (Vali and Stansbury, 1966; Wright and Petters, 2013), there was no dependence of the temperature difference on cooling rate, but on average, the drop temperature was 0.33 \pm 0.15 °C warmer than the centre of the plate. Thus, a +0.33 °C correction factor was added to any temperature herein and an uncertainty of 0.15 °C was added to the probe accuracy uncertainty.

Second, various hydrophobic coatings with UPW were tested for the best combination of materials to use with the least influence from artefacts (Figure 4). Materials tested were chosen based on those used in previous work and included: 1) direct petrolatum (Tobo, 2016), 2) 15% w/v petrolatum in xylenes (Certified ACS Reagent Grade, Ricca Chemical) (Bowers et al., 2009), 3) silicone fluid (710 fluid, Dow Corning®) (Polen et al., 2016), and 4) squalene (\geq 98%, Sigma-Aldrich®) (Hader et

al., 2014; Wright and Petters, 2013; Wright et al., 2013). The silicone fluid was difficult to use for cold plate experimentation because droplets would coalesce during the experiment and freezing detection by eye was difficult due to the glare of the substance. Squalene was less viscous than the silicone fluid, inducing more drop coalescence but freezing detection was easier than the silicone fluid. Both materials remained in the fluid state, thus are not ideal for direct cold plate use, but have been
5 proven suitable for cold stages that use covered sample dishes or trays and smaller drop sizes (Hader et al., 2014; Polen et al., 2016; Wright and Petters, 2013; Wright et al., 2013). The petrolatum and xylenes solution creates a thin layer of petrolatum after drying to evaporate the xylenes and alleviate the coalescence problem; however, as evidenced by the freezing spectra in Figure 4, is not the best option in terms of limiting artefacts. To summarize, a hydrophobic coating is needed on the copper plate and the option with the least influence from contaminants is direct petrolatum smeared onto the plate using UPW.

10 Last, the effect of drop size was tested using UPW and petrolatum-coated copper plates (Figure 5). Normally, 2.5- μ L drops are created by hand using a sterile syringe. Because such drops are created without the use of a pipette, possible small variations in drop volume may occur. The same volume drops were created with a pipette and sterile tips and tested against syringe drops. Additionally, tests with 1.5- μ L and 5.0- μ L drops were conducted to evaluate the effects of larger changes in volume. One major caveat with the pipette technique is that it takes substantially more time to create the arrays of 100 drops (approximately
15 five times slower than the syringe method). Overall, the best method in terms of onset freezing temperatures and fraction frozen was the 2.5- μ L drops created via syringe. This test was comparable in terms of fraction frozen to the 1.5- μ L drops colder than -21 °C. One possible explanation for the higher onset temperature and higher concentrations of impurities in the 2.5- μ L pipetted drops as compared to the 2.5- μ L syringed drops is contamination from the pipette tips. The 5.0- μ L test demonstrated that drops of this size are too large such that they induce freezing at warmer temperatures and are subject to large
20 variability—in theory, the larger the drop volume, the larger the abundance of impurities within a single drop that may facilitate ice formation (Bigg, 1953). Overall, our drop size tests demonstrate the efficiency and reliability of 2.5- μ L drops created via syringe.

Out of the 100 drops for each test, $95 \pm 5\%$ on average (ranging between 84 – 100%) were detected as frozen and recorded from all tests (Figure 6). Some of the tests within the same sample were reproducible within error, demonstrating the reliability of
25 the method (e.g., samples 1 and 3). However, variability from test-to-test within the same sample could occur due to: 1) detection of rarer INPs at specific temperatures during 1 – 2 of the tests or 2) uncertainties arising from instrumental artefacts, such as contamination between tests. These results demonstrate the importance of running triplicate (or more) tests for DFA techniques—to capture some of the rarer INPs that may exist in the samples or account for test-to-test variabilities. Such rarer INPs may be missed or over accounted for if only one test is conducted. The cooling rate was variable during each test, but
30 maintained within the $1 - 10$ °C min^{-1} range and the fraction frozen did not show a noticeable dependence on the cooling rate, as discussed above.

From the fraction of drops frozen and the known total volume of air per sample, we calculated the estimated INP concentration (L^{-1} of air) with the universally applied equation by Vali (1971):

$$[INP](L^{-1}) = \frac{\ln(1 - f)}{V_{drop}} \times \frac{V_{suspension}}{V_{air}}$$

where f is the proportion of droplets frozen, V_{drop} is the volume of each drop, $V_{suspension}$ is the volume of the suspension (i.e., 2.5 mL for the sample tests), and V_{air} is the volume of air per sample. We averaged the total volume of air from the six field samples collected and applied that to the equation to calculate INP concentrations for the blanks, in order to conduct a direct comparison and evaluate the INP concentrations in the samples relative to the blanks.

2.3.2 Raman microscopy for deposition mode ice nucleation

Depositional ice nucleation was measured using a Nicolet Omega XR Dispersive Raman Spectrometer outfitted with a Linkham THMS600 environmental cell and a Buck Research CR-1A chilled-mirror hygrometer. The Raman spectrometer was coupled with an Olympus BX51 research-grade optical microscope with 10x, 20x, and 50x magnification abilities. The environmental cell and CR-1A hygrometer allow for temperature control and dew/frost point measurements to back calculate saturation ice ratios, S_{ice} . The environmental cell was connected to two UHP grade N_2 tanks, one is humidified and the other is a “dry” tank that is not humidified. These two were then mixed, fed through the environmental cell, and lastly the CR-1A measures the dew/frost point. In these experiments, the water vapour was kept constant while the temperature was decreased, which resulted in an increase in S_{ice} . This experimental set up, calibration, and calculation is explained in more in detail in Baustian et al. (2010), Schill and Tolbert (2013), and Primm et al. (2017).

An aliquot of the solutions from the previous immersion mode experiments were used for deposition mode ice nucleation experiments (i.e., untested sample solution). The solution derived from each spot on the collected filter sample was nebulized onto a fused silica disc, which was then placed into the environmental cell at ~0% RH to allow for evaporation of water from the particles. The temperature was then decreased at a rate of 0.1 K min^{-1} , while water vapour was held constant. Temperature and dew point were recorded during the entire experiment. S_{ice} was determined from the temperature and dew point where ice was first visually identified. The different S_{ice} values at different temperatures were determined by performing the same procedure, but changing the starting water vapour pressure. This difference in water vapour pressure changes the S_{ice} value at different temperatures. Temperatures which were analysed for depositional ice nucleation were chosen to cover a wide range of those previously reported and relevant for several cloud regimes (Hoose and Möhler, 2012). Nebulization onto the disc resulted in 5000 – 10000 particles, with a range of $1 \mu\text{m}$ to $50 \mu\text{m}$ in diameter, deposited on the surface depending on the spot from the filter paper. Of the particles that nucleated ice, 3 – 5 particles were analysed for composition using Raman spectrometry for each sample. Because the purpose of the analysis was to prove that particles could be analysed for depositional

ice nucleation using samples collected by HOVERCAT, only the first few particles that formed ice at each temperature regime were recorded. A more statistical approach (i.e., analysing more particles) to characterize the depositional INP population during the flight is outside the scope of this manuscript.

3 Results and discussion

5 3.1 Operation of HOVERCAT instruments during test flight

Although the ability to ~~hover at an exact altitude control the exact altitude of the system~~ was difficult due to vertical winds—which was determined by abrupt ascent or descent and horizontal transport ~~of HOVERCAT~~ while tracking in real-time—the ~~system was able to profile~~ ~~we were able to control gas venting and dropping ballast to slow down ascent and descent~~, and sample at altitudes from the ground level up to 2543 m AMSL a.m.s.l. (approximately 1053 m AGL a.g.l.) for 3 hours (Figure 7). The ability to control the BBFCS to execute the step-wise flight plan was difficult given the winds and the several-second delay in time when venting or dropping ballast to decrease or increase in altitude, respectively. Minor fluctuations in BBFCS control to maintain altitude was not possible during 25 May conditions, but may be on a calmer day aloft. Because of such issues, the first two profiles (i.e., ascent followed by descent to ground) during the first hour of flight (up to 2316 and 2543 m AMSL a.m.s.l.) were abrupt and parking at desired altitudes was not achieved. We were able to ~~hover maintain altitude~~ at 1771±80 ~1800 m AMSL a.m.s.l. (281 m a.g.l.) during the third profile (08:00 – 09:00), with a short drop in altitude around 08:50. Starting at 09:07:40, we were able to ~~hover maintain altitude~~ just above the ground at ~1536±2000 m AMSL a.m.s.l. (46 m a.g.l.) until 9:15, with a final profile up to 2098 m AMSL a.m.s.l. (608 m a.g.l.) at 09:25. Ultimately, the balloon deflated and ended the flight at 09:36.

While controlling the exact altitude of the BBFCS was difficult, the aerosol measurements were fruitful. The OPC measured particle concentrations up to 250 cm⁻³ while at the ground (average of 6 cm⁻³), with the lowest concentrations occurring at the highest altitudes (< 1 to 2 cm⁻³; average of 1 cm⁻³). However, episodic spikes in number occurred when stable at the ground, indicating localized sources of high concentrations of particles. PM concentrations followed a similar inverse relationship with altitude (Figure 7). The total flow through the filter in TRAPS was fairly consistent throughout the flight, starting at 40 L for Sample 1 and decreasing to 32 L for Sample 6. The slight decrease possibly resulted from: 1) inconsistent power supply by the battery pack to the micropump or 2) strain on the micropump with altitude, although the latter is less likely given the variability in altitude throughout the flight.

3.2 Immersion freezing ice nucleation

From the six filter sample spots that were collected, aerosol loading was sufficient to conduct INP measurements using the DFCP system. Cumulative INP spectra show relatively low concentrations (i.e., 10⁻² – 10⁻¹ L⁻¹) of warm temperature INPs (>

-10 °C, likely of biological origin (Murray et al., 2012)) for all samples, while reaching up to 10^1 L^{-1} at temperatures below -20 °C (Figure 8). Such concentrations are within range of those previously reported in Colorado: Prenni et al. (2013) reported $1 - 10^2 \text{ L}^{-1}$ at -25 °C. The highest INP concentrations were observed from Sample 3, which corresponded to the time where HOVERCAT was closest to the ground (i.e., 69% of sample time was < 50 m a.g.l.), on average (Figure 9a). Sample 6 had the highest concentrations of INPs active between -8 and -12.5 °C, which also corresponds to when HOVERCAT hovered just above ground level (19% of the time; Figure 77). It is important to note that all samples aside from Sample 4 hovered near the ground: Samples 1, 2, and 5 were close to the ground 40%, 9%, and 2% of the time, respectively. Thus, altitude-dependent results could be skewed by collection nearest to the local source of aerosol. It is important to note that the samples that spend little to no time at the ground corresponded to the lowest INP concentrations (i.e., Samples 4 and 5). However, based on OPC number concentrations, there was not always a clear decrease of aerosol concentrations with altitude (e.g., Sample 5). Additionally, concentrations were calculated and based on total volume of air, indicating that the altitude in which the sample was collected at for the most amount of time is representative of the overall sample INP population. Combined, the immersion INP, OPC, and BBFCS results indicate that: 1) total particle number concentrations and INP concentrations were highest ~~closest when HOVERCAT sampled near the to the ground and, 2) collection at higher altitudes resulted in lower particle concentrations, and 3) INPs of likely biological origin remained close to the surface, which is predominantly agricultural soils in this region~~ (Hill et al., 2016). The relative abundance of INPs to total particles is also consistent with previously reported values (DeMott et al., 2010): INPs represented 1 in every 10^2 to 10^5 number of particles detected by the OPC, although the OPC does not measure below 380 nm so the fractions might in reality be even lower (Figure 9b). However, INPs are thought to be relatively large (i.e., > 200 nm in diameter) based on previous work (DeMott et al., 2010; Fridlind et al., 2012; Kanji et al., 2017; Mertes et al., 2007; Niedermeier et al., 2015), so the OPC may be relevant for supporting INP measurements. Although these results may not be surprising (e.g., total particle, INP concentrations within range of previous work and generally highest near the ground, and biological INPs sourced from an agricultural region) and yield results consistent with previous work (DeMott et al., 2010; Hill et al., 2016; Murray et al., 2012; Prenni et al., 2013), they demonstrate the utility and reliability of the collection and analytical methods of HOVERCAT and the DFCP systems.

25 3.3 Deposition ~~freezing~~ ice nucleation

Depositional ice nucleation analysis of the six filter samples was conducted using the extra volume of resuspension left from the immersion freezing analysis (i.e., the portion of the 2 mL that was not used on the DFCP). Of the particles that nucleated ice, 3 – 5 particles were analysed for composition using Raman spectrometry for each sample. We assume that a majority of the particles are of similar concentration because the whole sample was dissolved in water, allowed to mix to a homogeneous solution, and nebulized onto the sample disc. Indeed, the particle composition was similar for each particle in any sample, while there was variation from sample to sample. Although the Raman spectral and ice nucleation analyses are helpful to observe the overall particle composition as temperature and relative humidity are changed, the experiment does not determine

the size or mixing state of the particles as they were in the atmosphere. Further, the spectral resolution of 1 micrometre in our system does not allow smaller scales to be distinguished within the individual particles probed.

5 Overall, ice activation onset conditions between the six samples were similar at all temperatures tested (Figure 10). However, at -40 °C, Samples 3 and 4 showed first ice nucleation activity at a saturation ice ratio of 1.12, which were lower than the other samples and may be characterized as more efficient deposition INPs at that temperature as compared to the remaining samples. Overall, the samples contained deposition INPs that enabled ice formation above homogeneous freezing. Saturation ice ratios of all six samples were similar at the temperatures tested, except for Samples 3 and 4 at approximately -40 °C (Figure 10), indicating these samples contained more efficient deposition INPs at that temperature as compared to the remaining samples. These samples contained slightly more efficient INPs at -25 °C, but similar efficiencies to the remaining samples at 10 -55 °C. Raman spectrometry demonstrates that most of the samples were compositionally disparate from each other (Figure 11). The first three samples show a very intense fluorescence signal (i.e., the curve-like characteristic of the baseline), which is consistent with either biological or organic materials (Baustian et al., 2012). Additionally, Sample 2 contained a peak for carbonate, which is indicative of a mineral dust signature (Baustian et al., 2012). The sample collection time periods for these samples occurred directly over a dense agricultural region in the Colorado plains, supporting the observation of highly 15 fluorescent particles (Figures 2 and 7). Interestingly, Sample 3 contained efficient immersion mode INPs as well, that were likely of biological origin due to the relatively higher INP concentrations at temperatures greater than -10 °C (Figure 8). Samples 4, 5, and 6 show a C-H stretch peak, and occasionally sulphate (SO₄²⁻) and nitrate (NO₃⁻) peaks, which is consistent with the composition of typical anthropogenic aerosols in the atmosphere (Zhang et al., 2007). Sample 5 had the most intense anthropogenic peaks while yielding the least efficient immersion mode and deposition mode (i.e., at the two highest 20 temperatures measured) INPs. It is possible any INPs present in this sample were affected by sulphate or nitrate coatings, which have been shown to inhibit the ice nucleating abilities of aerosols (e.g., Cziczo et al., 2009; Möhler et al., 2008; Reitz et al., 2011; Sullivan et al., 2010). Collection of Samples 5 and 6 coincided with when HOVERCAT flew close to the ground near I-25, where vehicular traffic and industry lining the multilane interstate likely contributed to the larger signal from anthropogenic functional groups and less efficient INPs. However, the Raman spectrum for Sample 6 also has a weak 25 fluorescent signature, indicating a possible biological contribution. HOVERCAT flew from over I-25 to the west over more agricultural lands. Sample 6 also contained high concentrations of INPs at -10 °C, indicating the sample also contained biological INPs. Combined, these results from Sample 6 suggest a mixture of biological and anthropogenic sources.

3.4 Future directions for HOVERCAT Recommendations for future airborne INP measurements on small platforms

30 As indicated earlier, Phase I of the BBFCS and HOVERCAT combination exists in its current prototype state. The priorities of Phase I were to develop a system that is cost effective, user-friendly, versatile, and in compliance with FAA regulations without the need for special approvals or restricted airspace. Under these priorities, our objectives were to address if we could develop such a system that was: (1) recoverable and (2) controllable. Recoverability was a requirement as we needed to obtain

the filter samples for the offline INP analysis, while controllability was an added benefit to have altitude-resolved INP measurements. We successfully achieved the first objective by recovering the system after it landed, and controlling the BBFCS such that the landing was not damaging to the instrumentation. The second objective, however, is still in need of improvement as discussed here. The benefits of the system as a whole are that it is cost effective and easy to operate relative to traditional airborne measurements of INPs, and did not require special FAA approvals, providing flexibility to fly anywhere at any time. HOVERCAT alone has the benefit of having time-resolved filter sampling capabilities that, if able to control altitude, would yield vertically-resolved INP measurements. However, as discussed throughout, both the BBFCS and HOVERCAT have their limitations. Here, we discuss these limitations and provide recommendations not only for a Phase II system for HOVERCAT, but also recommendations generally applicable towards INP measurements on small airborne platforms.

First, HOVERCAT could only operate in its current design up to 2.5 m a.m.s.l. (1.1 m a.g.l.). Although this is an improvement over previously reported tethered measurements of INPs (e.g., Ardon-Dryer et al. (2011) reached 196 m a.g.l.), achieving higher altitudes is desired to capture the profile of INPs in and above clouds using a launched platform that affords the flexibility to essentially fly anywhere. To improve operation for higher altitudes, modifications should be made to incorporate a stronger micropump that would yield higher flows and operation at lower pressures. The main issue is that to fly at free will (i.e., under FAA compliance), payload weight must be maintained under 2.7 kg for any single module (i.e., HOVERCAT). Thus, stronger pumps, which are by nature heavier, may not be realistic for HOVERCAT on a launched balloon system. Implementing a stronger pump would require either: (1) a FAA Certificates of Waiver or Authorization (COA), (2) flights in restricted airspace, or (3) flights on a tethered balloon system, all of which do not align with the priorities to maintain simplicity and versatility. However, we generally recommend future parallel measurements be made with a better pump. One option could be to reduce weight of the other components (e.g., replace the metal protective enclosure of the TRAPS with lightweight foam). This may not afford enough margin to incorporate the weight of a better pump, but is a possible alternative that needs to be tested.

Second, the hovering capability needs improvement, either by further testing with the BBFCS or modification to a traditional launched balloon system. We were able to control the altitude ± 80 m, but executing the step-wise flight plan proved to be more difficult than anticipated. The venting and ballasting functioned properly, but room for improvement could be focused on accounting for natural conditions (i.e., updrafts and downdrafts) that affect the altitude and truly enable the BBFCS to hover at desired altitudes. As another option, HOVERCAT could be deployed on a traditional launched balloon with a slow rise rate and less helium, or a reverse parachute (i.e., less buoyancy and more drag) to afford a steady vertical profile, although this eliminates the hovering capability of the system unless the free lift is adjusted such that the system may hover near inversions. For instance, an ascent rate of 0.5 m s^{-1} would provide a 90450 m vertical resolution (at 30 minutes per sample). If such a system were successful, the need for bidirectional communication to control TRAPS sampling intervals would not be required.

and would eliminate the need for additional hardware, receivers, batteries, and other data processing components in HOVERCAT and for the ground station. In general, we recommend implementing advanced controllability features into traditional launched balloons to not necessarily hover, but afford a consistent and slow rise for sample collection, and components to terminate the flight at the desired altitude such that the package is still recoverable.

- 5 Third, the Phase I pilot study involved sampling in clear air to conceptually prove HOVERCAT could perform as desired. Ideally, operation of such a system would be in clouds and harsher conditions such as the Arctic. To function in harsher environments, testing the modules in humidified, pressure-controlled, and temperature-controlled conditions is required at temperatures down to -40°C . Ardon-Dryer et al. (2011) measured INPs successfully using a filter sampler in the Antarctic, but did not collect samples in cloud. Schrod et al. (2017) deployed their sampler on a small unmanned aircraft system up to
- 10 2.5 km a.g.l., but did not fly in cloud or ambient temperatures below approximately 15°C . Combined, even though our system and these previous systems are subject to limitations, they are a significant advancement towards a more flexible and versatile manner in which INPs above ground level can be measured. In general, additional research is needed to continue to improve such systems with regard to cost, performance, and enhanced spatial and temporal coverage to improve understanding of INP impacts on clouds.
- 15 ~~As indicated earlier, Phase I of HOVERCAT exists in its current prototype state. Plans for a Phase II to develop a more operational measurement platform are underway to: 1) improve operation at higher altitudes, 2) operate successfully on a routine basis, 3) function suitably in more extreme environments, such as in cloud and the Arctic, 4) improve the aerosol module's versatility to adapt to other platforms such as tethered balloon systems or unmanned aerial systems (UASs), and 5) transmit data to a ground station in real time. To improve operation for higher altitudes, modifications will be made to the~~
- 20 ~~micropump. The main issue is that to fly at free will (i.e., under FAA regulations), payload weight must be maintained under 2.7 kg for the aerosol module. However, more efficient yet lightweight micropumps can be used and tested in a pressurized chamber prior to deployment to improve operation at lower pressures and thus higher altitudes. Routine operations require more frequent flights and practice controlling the BBFCS to maintain a desired altitude, even in conditions with influential vertical winds. Additionally, the aerosol module could be deployed on a launched balloon with a slow rise rate and less helium,~~
- 25 ~~or a reverse parachute (i.e., less buoyancy and more drag) to afford a steady vertical profile, although this eliminates the hovering capability of the system unless the free lift is adjusted such that the system may hover near inversions (i.e., $< 0.5\text{ m s}^{-1}$). To function in harsher environments, testing the modules in a humidified and temperature controlled chamber is planned at temperatures down to -40°C . Operational testing of the aerosol module in cloud (i.e., under supersaturated and likely riming conditions) will occur at the high alpine research station in Jungfraujoeh, Switzerland during spring 2018. The aerosol module~~
- 30 ~~may easily adapt to other platforms through modification of the foam enclosure for tethered balloons or adding a smaller, protective case to fit on small UASs. Transmitting data during flight will require an on board computer and communications~~

~~between the modules and a ground station, as is done with launched balloons. In general, several modifications are planned to improve performance, utility, and versatility of HOVERCAT for future deployments.~~

4 Conclusions

5 Here, we present a novel airborne ~~aerosol and ice nucleation platform-measurement system~~ called HOVERCAT ~~that was tested~~
during a pilot study on the BBFC platform. HOVERCAT ~~that measures-measured~~ time ~~and altitude~~-resolved particle number
and INP concentrations ~~a range of altitudes up to 2.6 km a.m.s.l. (1.1 km a.g.l. at a ground elevation of 1.5 km a.m.s.l.).~~ -
~~Although controlling the ascent and descent of the balloon platform was difficult, we provide recommendations for future~~
~~platforms and measurements using similar non-tethered balloon systems. The platform has the capability to hover at desired~~
~~altitudes, making it an ideal system to collect sufficient aerosol loadings at a range of altitudes up to 2.6 km AMSL.~~ Unlike
10 similar ~~platforms~~systems, HOVERCAT can vertically resolve particle number concentrations in addition to both immersion
and deposition mode INPs. To our knowledge, this is the first platform to perform such measurements in tandem. Phase I of
HOVERCAT has been ~~successfully deployed~~tested, while ongoing efforts for improvement and modification are ~~underway~~
~~desired~~ for Phase II to enable HOVERCAT to fly higher and in more inclement conditions.

15 The ability to evaluate vertical distributions of INP concentrations and glaciation temperatures is of crucial importance in order
to inform and constrain process level models to improve understanding of aerosol-cloud interactions. Additionally, more
routine measurements of INP properties are needed to understand the evolving nature of aerosol-cloud interactions under a
wide range of cloud regimes, locations, and time of year.

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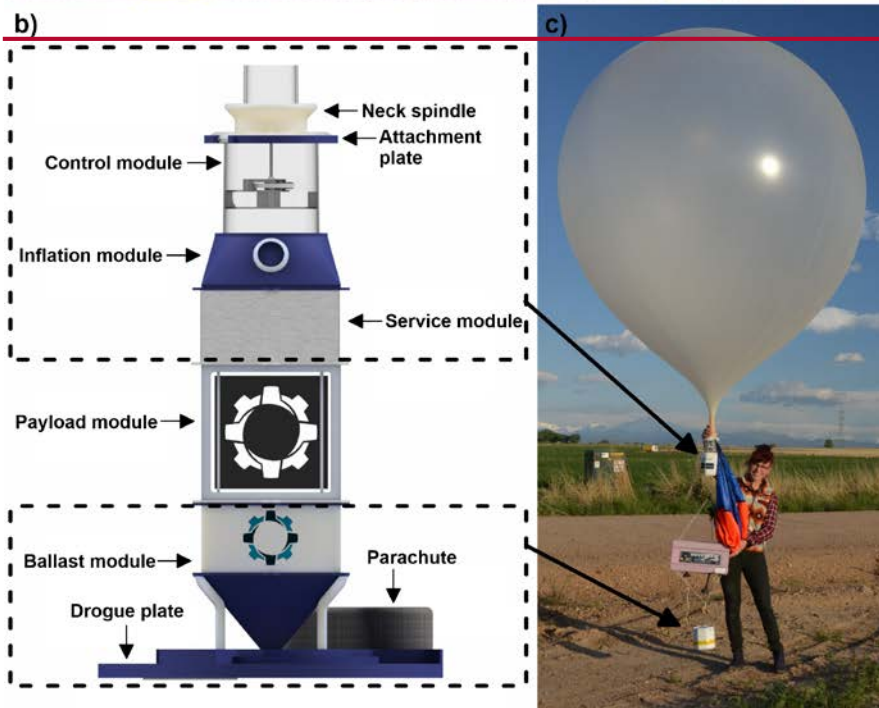
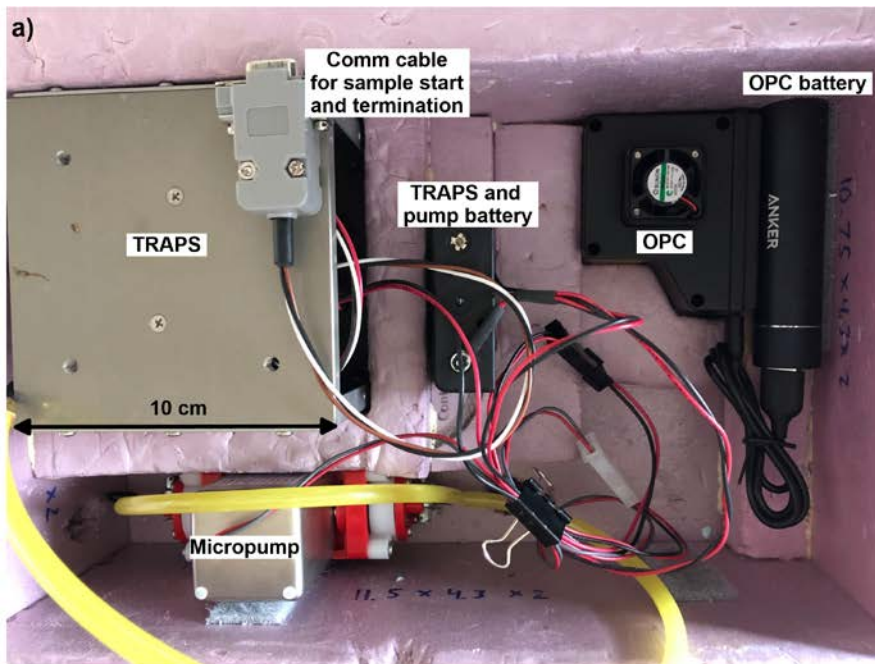
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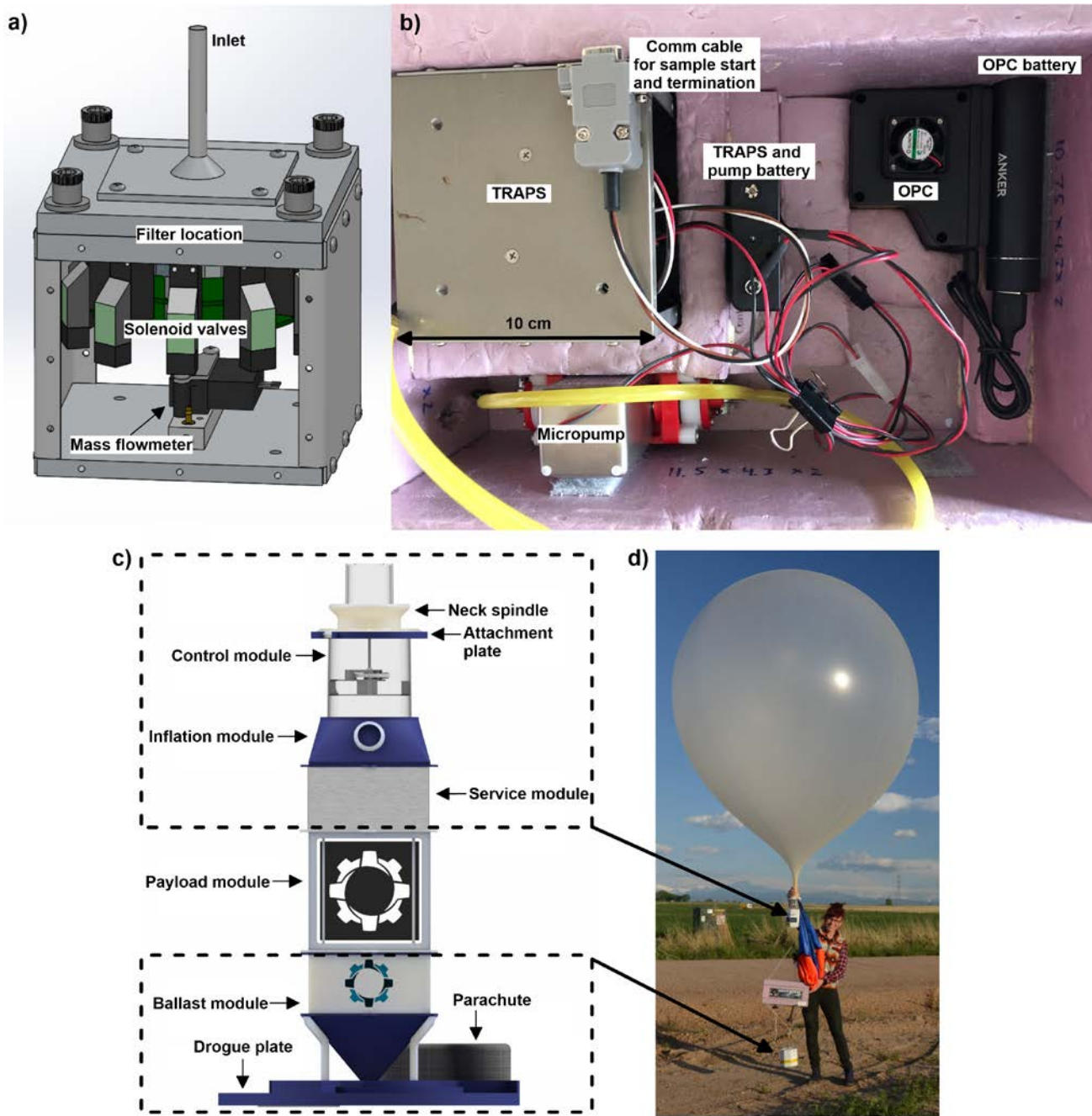


Figure 1: **Components of the complete flight system, including a) schematic of the TRAPS, b) a) Schematic of the BBFCS, b) Picture of inside the aerosol module, c) schematic of the BBFCS, and d) flight train for test flights.** Note that the service module on the BBFCS was separated approximately 1 m from the ballast module with the aerosol module (i.e., payload) in between. The ballast module was controlled by the on-board computer in the control module via an extended cable that ran down the tether string. The separated BBFCS modules were housed in foam for flights.

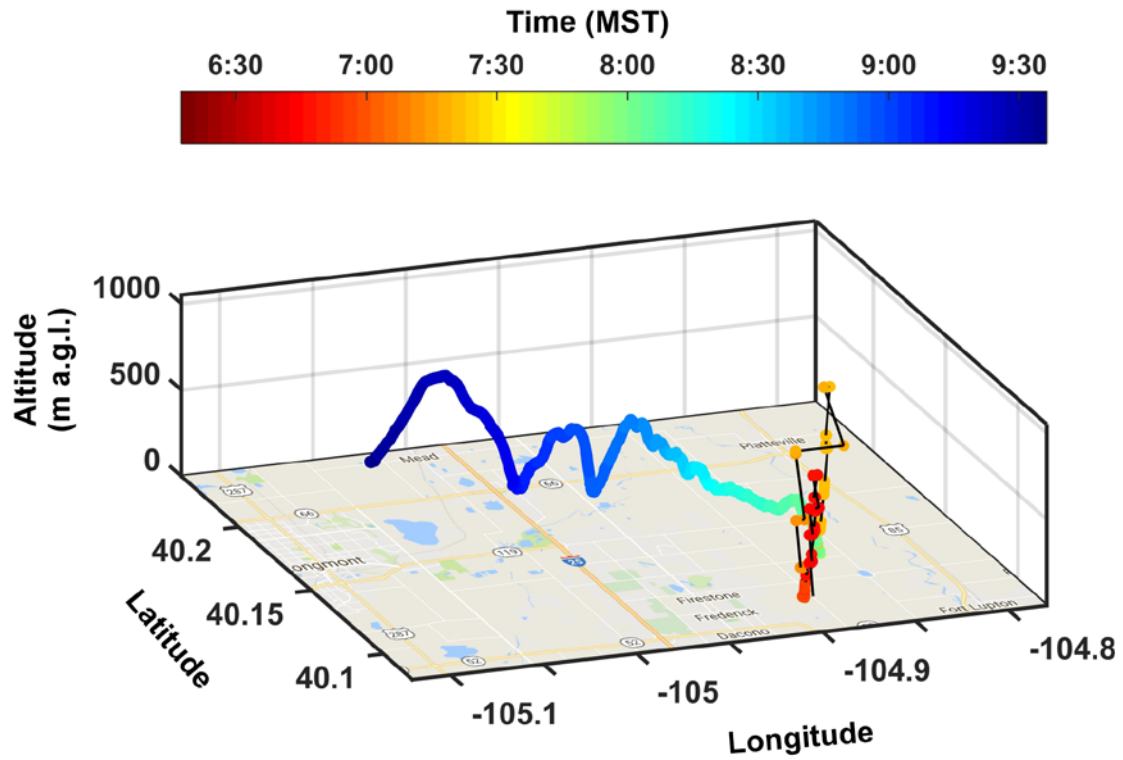


Figure 2: Four-dimensional flight path of HOVERCAT during the 25 May 2017 test flight, coloured by time in Mountain Daylight Time (MDT). Black lines between data points indicate missing GPS data, which occurred between 7:01 – 7:07 and 7:23 – 7:51. Meters AGL a.g.l. was calculated by subtracting 1490 from m AMSL a.m.s.l. to roughly show the altitude above ground.

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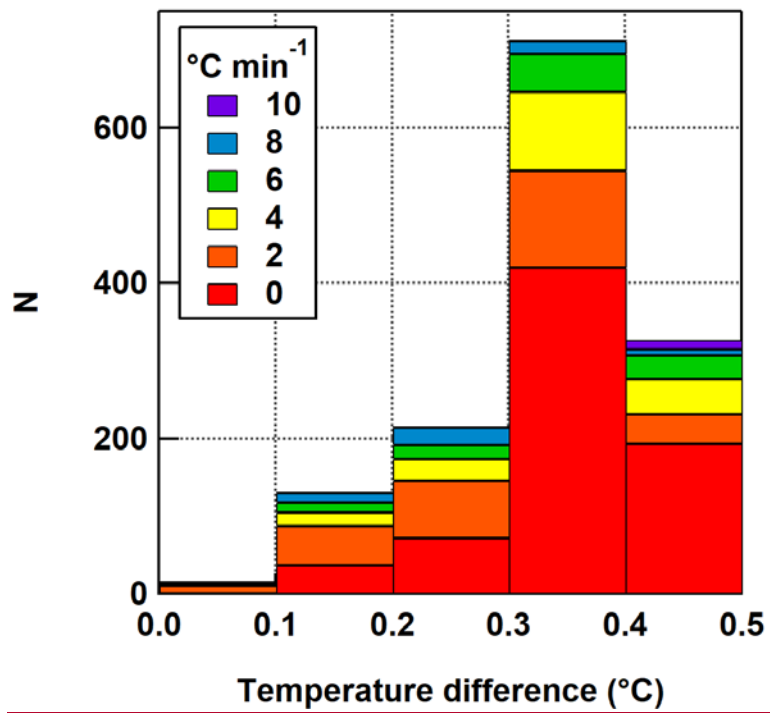
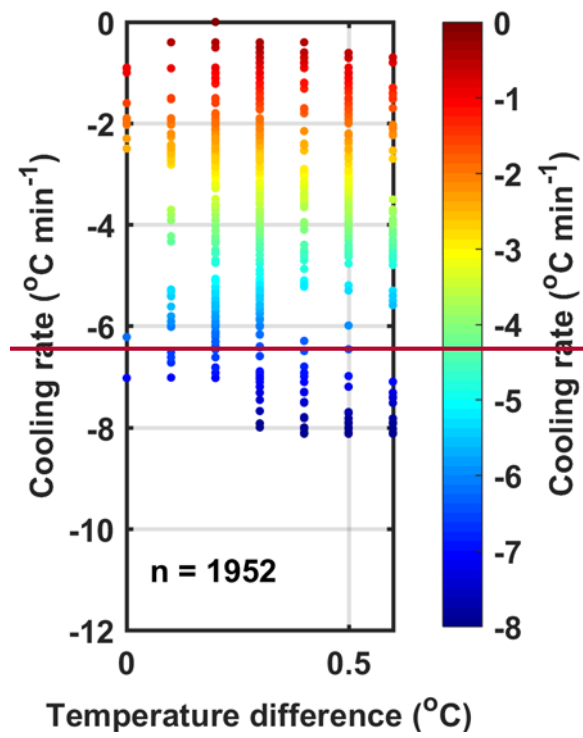
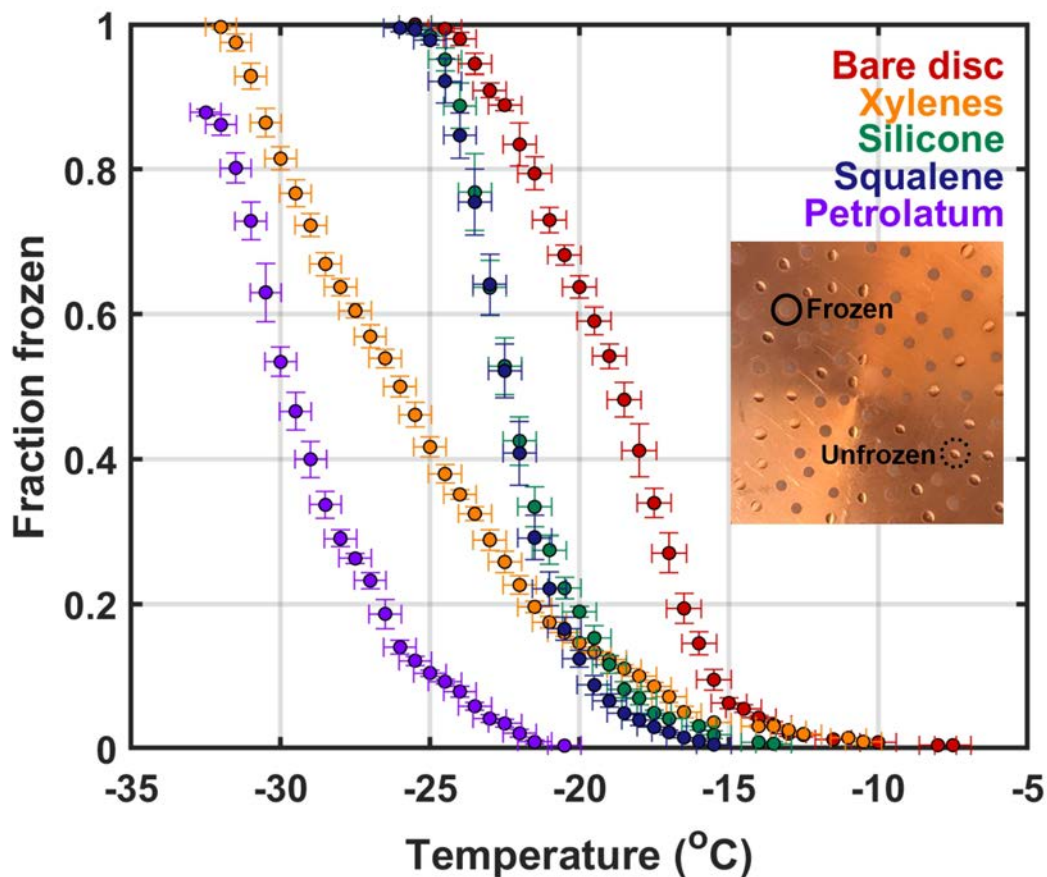
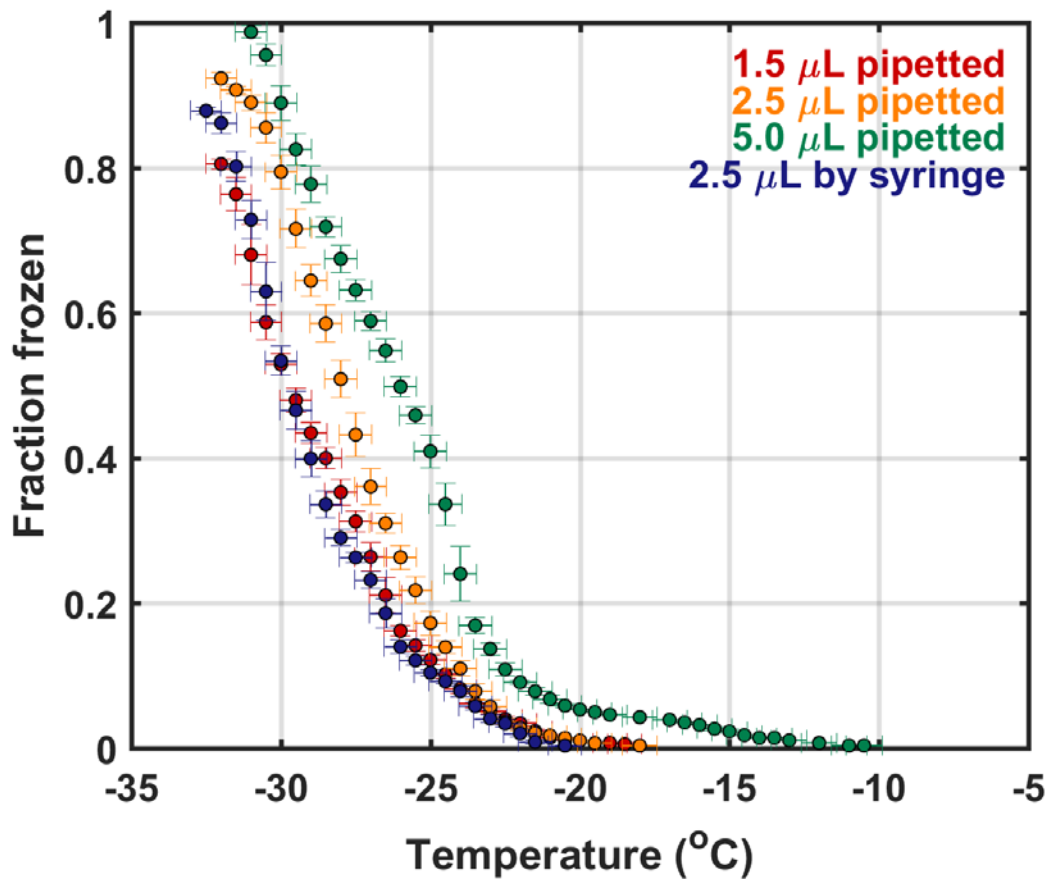


Figure 3: Histogram of ΔT temperature differences between measurements from a probe at the centre of the copper plate and drop on top of the plate coated with petrolatum coloured by cooling rate. The 1-second data are from three different tests and are coloured by cooling rate. The average difference used for the temperature correction was 0.33 ± 0.15 °C.



5 Figure 4: Freezing spectra for the control experiments conducted to characterize the DFCP system. Results included here are tests evaluating the most proficient hydrophobic coating with blank UPW drops. Error bars for the y and x axes correspond to standard deviation per 0.5 °C bin and temperature probe/plate versus drop variability standard deviation, respectively. Spectra that do not reach a frozen fraction of 1 indicates not all drops froze at the lower limit of the DFCP. The inset shows an example of the appearance of frozen versus unfrozen 2.5-µL drops on the copper disc.

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5 Figure 5: Freezing spectra for the control experiments conducted to characterize the drop size chosen for DFCP analysis. Results included here are tests evaluating pipetted versus ~~hand~~syringe-aliquoted drops and at different volumes. Error bars for the y and x axes correspond to standard deviation per 0.5 °C bin and temperature probe/plate versus drop variability standard deviation, respectively. Spectra that do not reach a frozen fraction of 1 indicates not all drops froze at the lower limit of the DFCP.

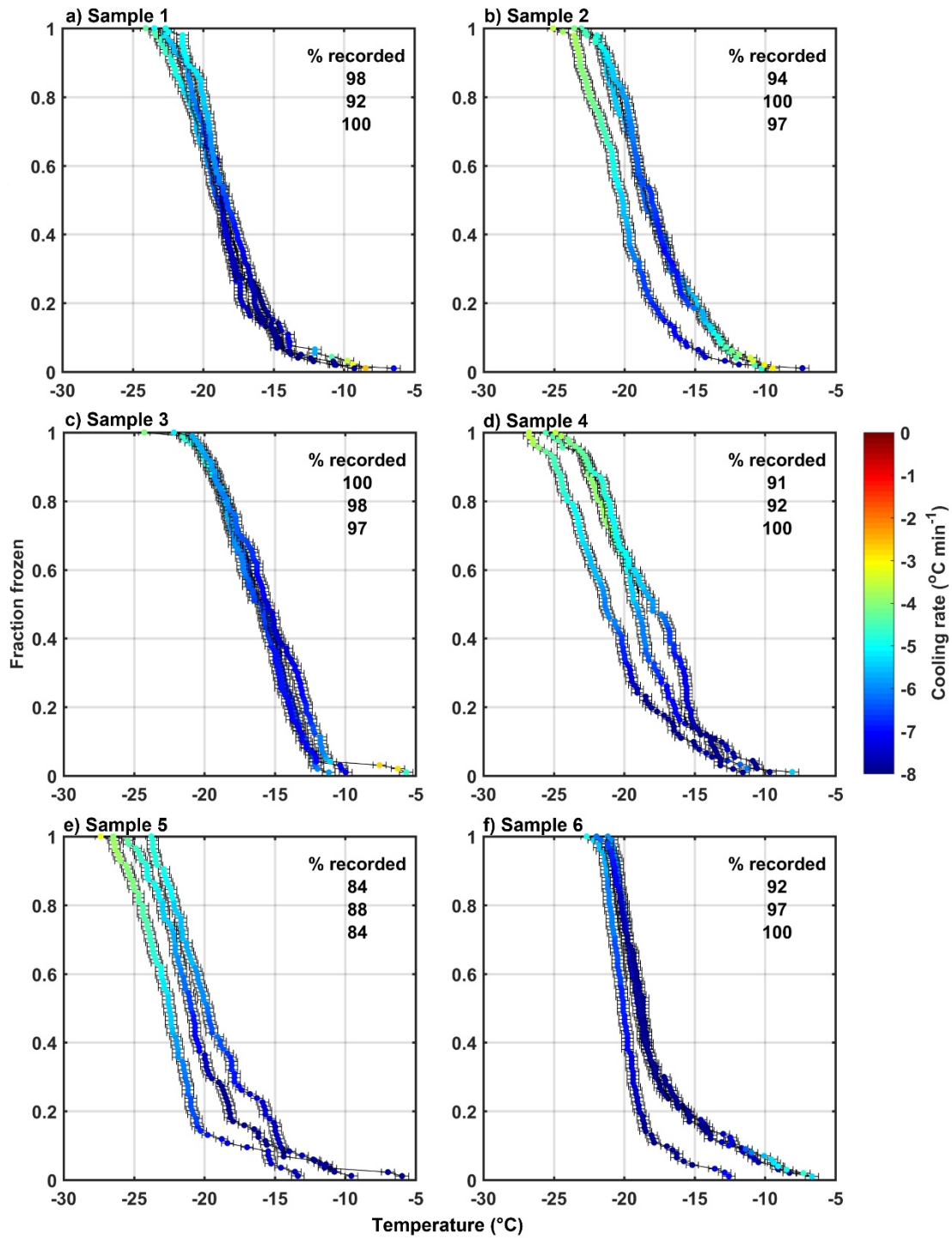
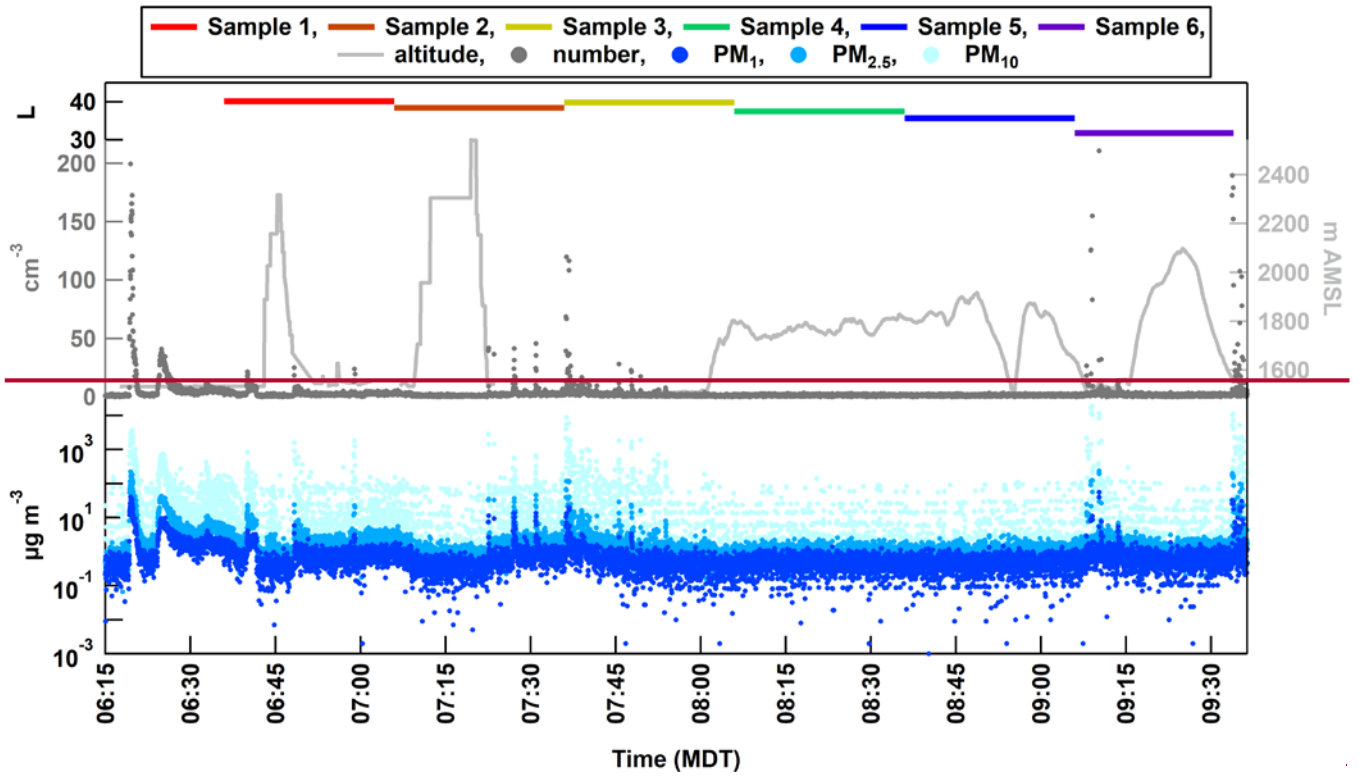


Figure 6: Freezing spectra for the three tests of each of the samples collected from HOVERCAT during the 25 May 2017 test flight. Each data point is coloured by cooling rate and has error bars associated with Omega temperature probe uncertainty. The percentage of recorded frozen drops is provided for each sample.



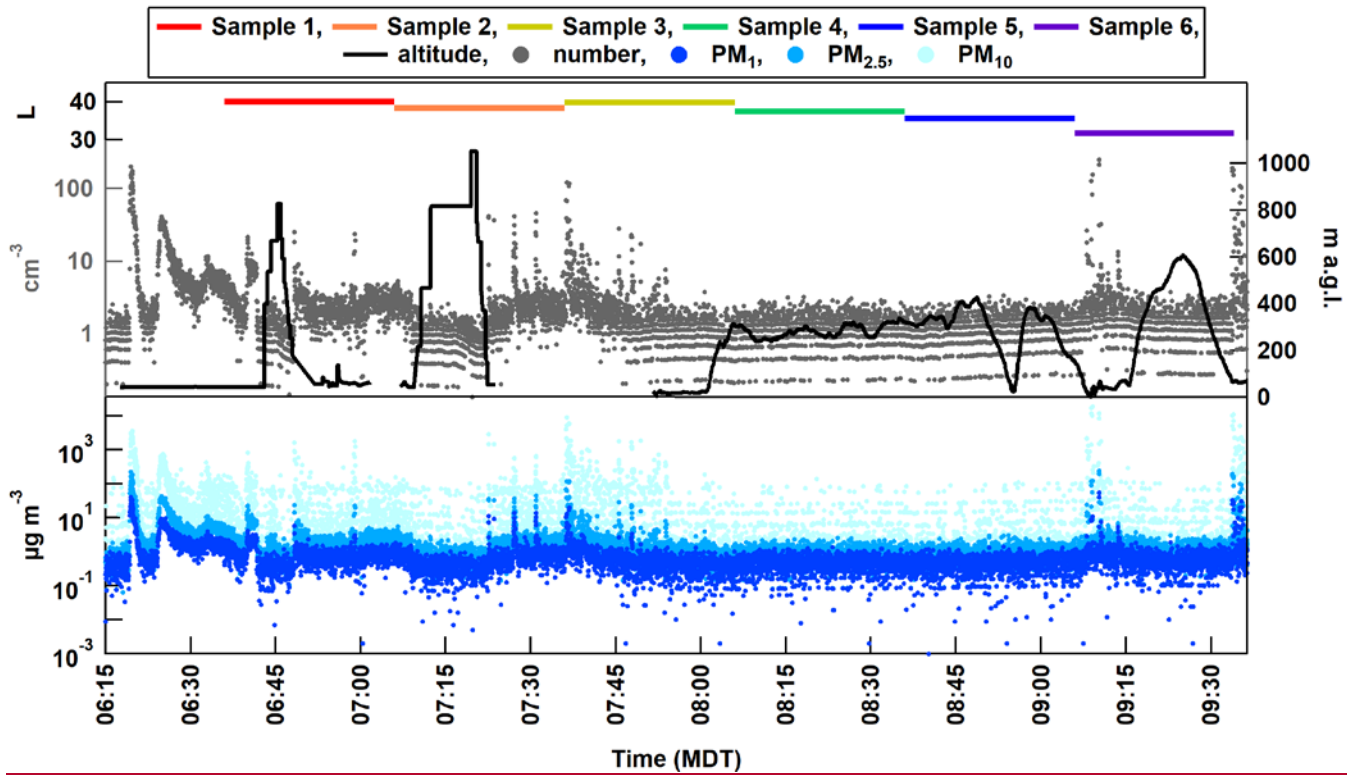


Figure 7: Time series of TRAPS total volume per sample (L; of air), OPC number concentrations (cm^{-3}), ~~HOVERCAT~~ altitude (m ~~AMSL a.g.l.~~), and estimated particulate mass (PM) concentrations from the OPC ($\mu\text{g m}^{-3}$). The width of the TRAPS total volumes corresponds to the collection time per sample (i.e., 30 minutes).

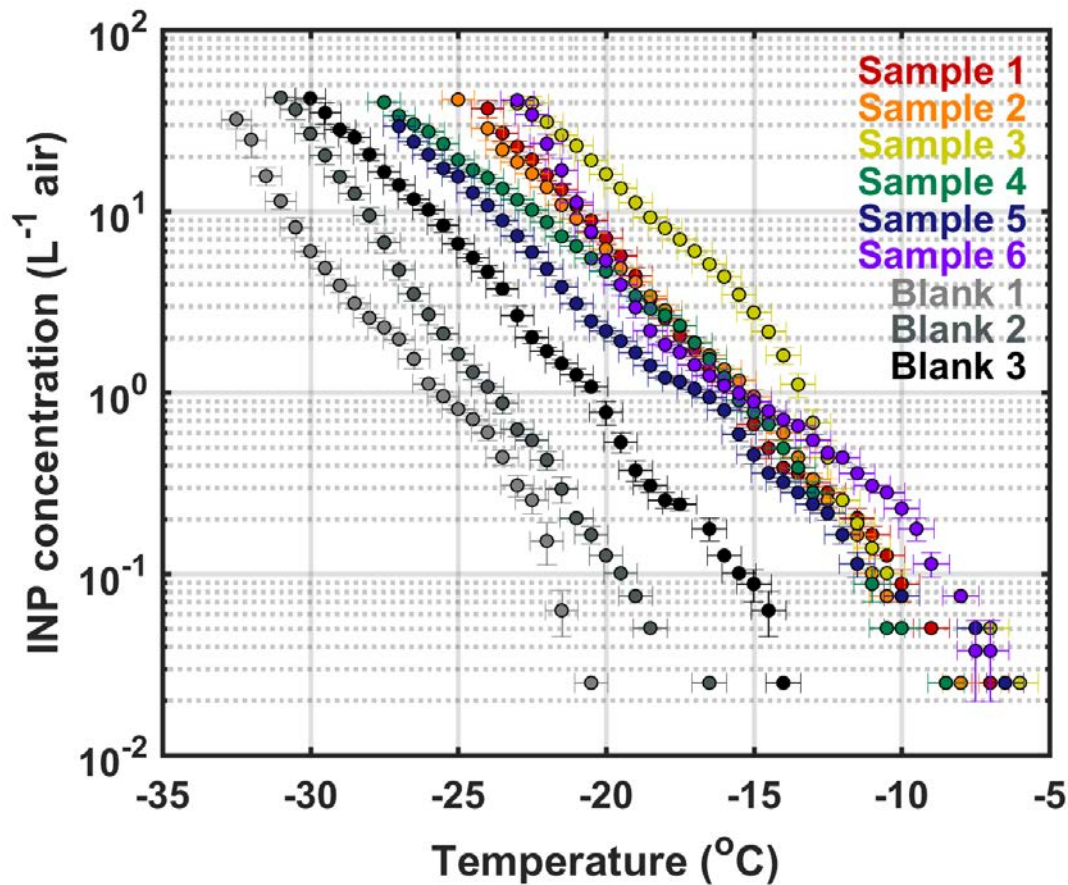


Figure 8: Cumulative INP spectra from the samples collected during the 25 May 2017 HOVERCAT test flight. Triplicate tests are binned every 0.1 °C. The blanks indicate a triplicate test from: a blank-UPW test mixed alone in a beaker for 2 hours (Blank 1), UPW mixed in a WhirlPak® bag for 2 hours (Blank 2), and an EmFab® filter mixed in UPW in a WhirlPak® bag for 2 hours (Blank 3). The latter is closest to how the samples were prepared, and a test with EmFab™ filter prepared in UPW in the same manner as the samples. Error bars for the y and x axes correspond to standard deviation per 0.5 °C bin and temperature probe/plate versus drop variability standard deviation, respectively. Spectra that do not reach a frozen fraction of 1 indicates not all drops froze at the lower limit of the DFCP.

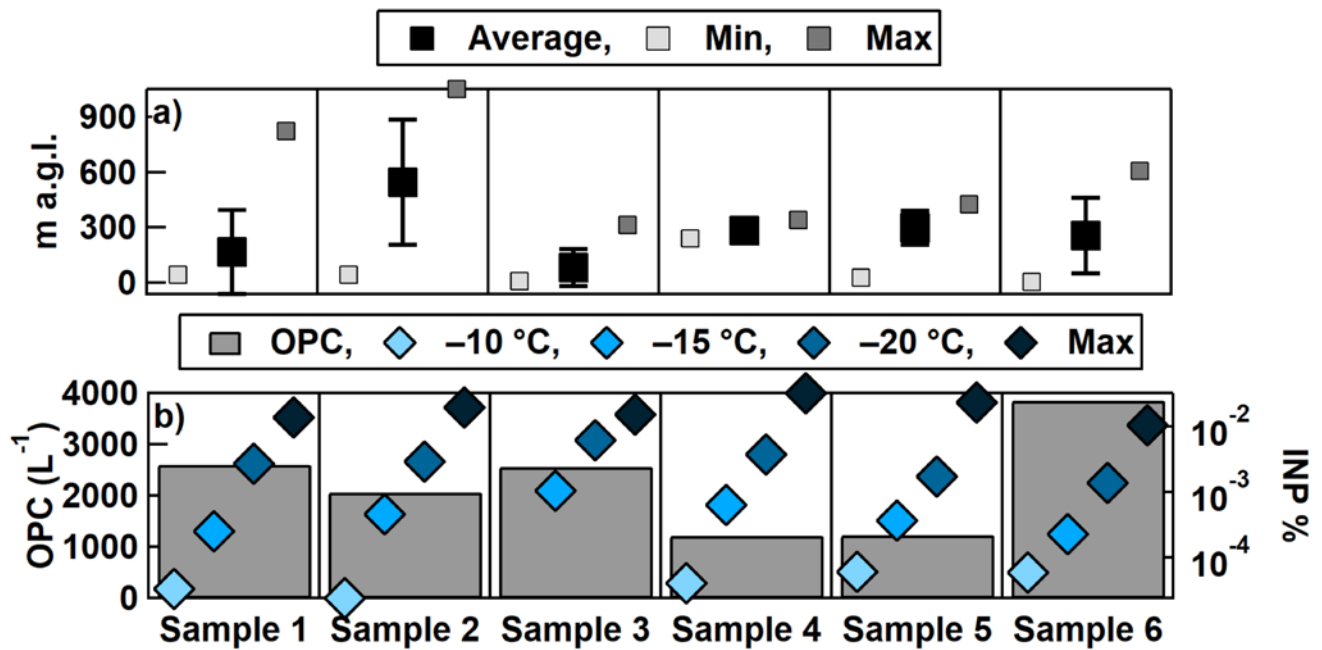


Figure 9: a) Average, minimum, and maximum altitudes HOVERCAT flew during each sample collection time period. Error bars represent one standard deviation. b) The average number concentrations of total particles from 380 nm – 17 μm in diameter measured by the OPC (left axis) and fraction of INPs out of total OPC number at -10 °C, -15 °C, -20 °C, and the maximum INP concentration measured at the temperature in which the last drop froze (right axis).

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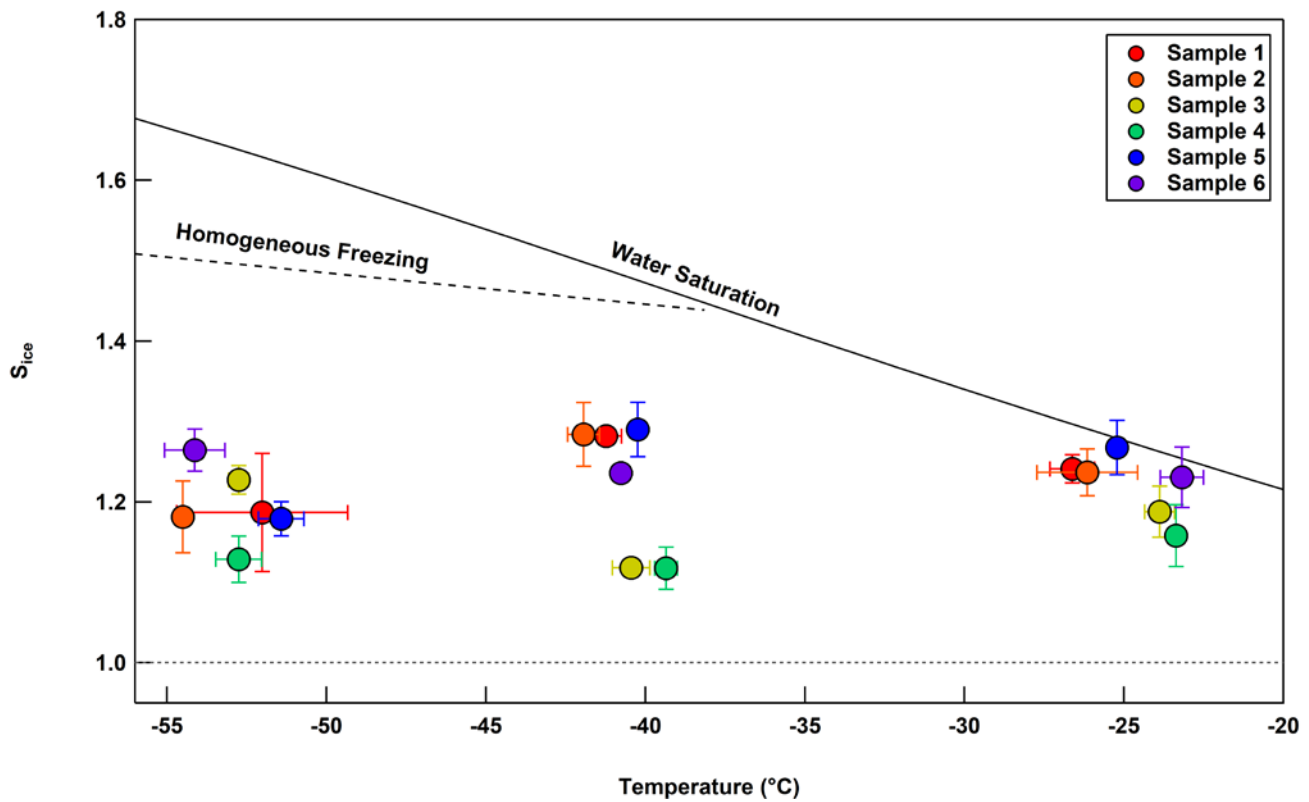


Figure 10: Depositional ice nucleation experiments on Samples 1 – 6 plotted by S_{ice} versus temperature. The values plotted here are of the onset conditions of depositional ice nucleation. For our experiments, this refers to the first particle to nucleate ice out of the 10^4 particles deposited on the disc in total, thus a percent activated fraction of 10^{-4} . Although temperatures measured were not exactly -25 °C, -40 °C, and -55 °C, these values are used for brevity for all samples within each grouping shown above. Depositional ice nucleation experiments on Samples 1 – 6 plotted by S_{ice} versus temperature. Although temperatures measured were not exactly -25 °C, -40 °C, and -55 °C, these values are used for brevity for all samples within each grouping shown above.

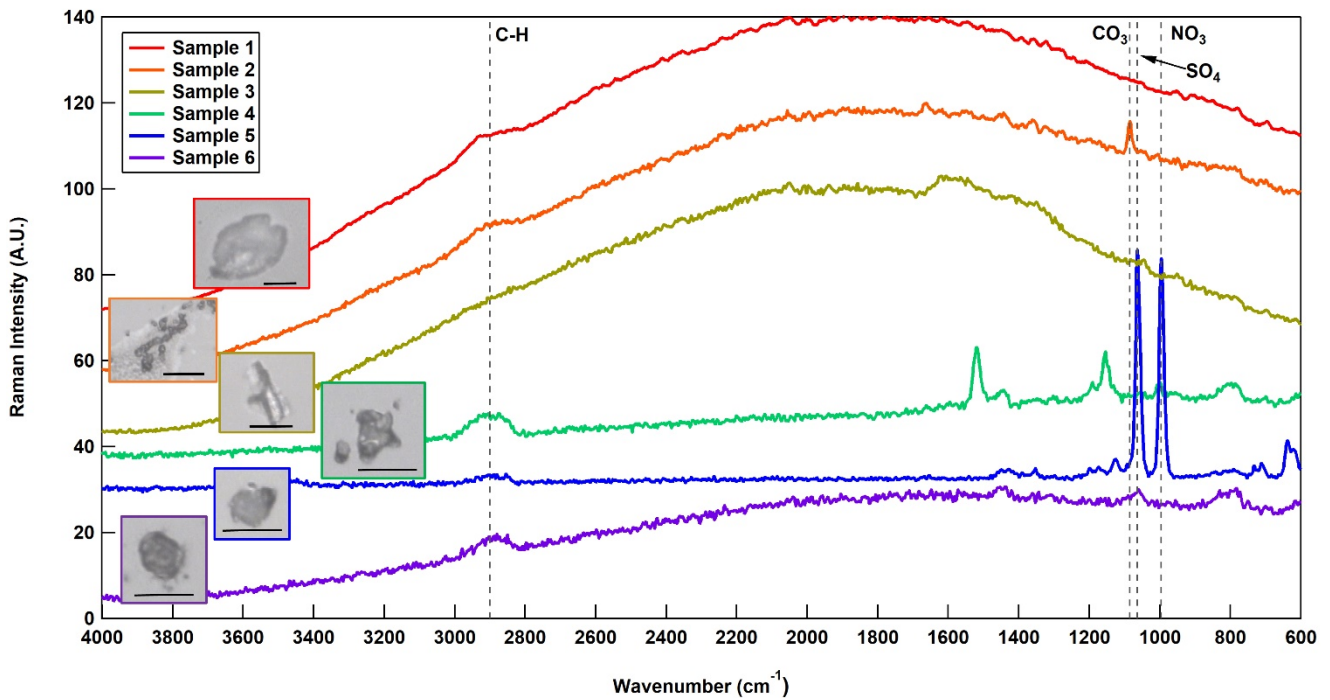


Figure 11: Raman spectra for a representative particle per sample. Characteristic vibrational frequencies for functional groups of organics (C-H; 2800 – 3000 cm^{-1}), carbonates (CO_3 ; 1070 – 1090 cm^{-1}), sulphates (SO_4 ; 972 – 1008 cm^{-1}), and nitrates (NO_3 ; 1032 – 1069 cm^{-1}) are noted for reference. Included are images of the particles that initiated depositional freezing for the Raman spectra shown. The length of the black line in each image represents a scale of 20 μm .