Responses to Reviewer#2

Review of 'Identifying the seeding signature in cloud particles from hydrometeor residuals' Konwar et al. AMT-2023-171

Summary: In this manuscript, the authors present airborne measurements from a mini-Aerosol Mass Spectrometer (mAMS) to identify regions in cloud that may have been influenced by cloud seeding. Following sampling using a counter flow virtual impactor (CVI) that removes hydrometeors and other aerosol with aerodynamic diameters smaller than a certain size (~7 um in this study), cloud droplets are evaporated and the residual aerosol are tested for the presence of chemical species, including potassium (K) and Chlorine (Cl) that are otherwise not present (or not detectable in natural form) within the natural cloud environment. The authors (correctly) state that unambiguously identifying cloudy regions in which seeding material is contained within hydrometeors remains elusive; and developing a method to identify such regions is a key step to evaluating hypotheses associated with how cloud seeding might impact cloud and precipitation evolution. In this manuscript, the authors present evidence that they are able to do just that, and thus I believe this work makes an important contribution and is therefore worthy of publication. However, there are numerous points (some major) that I feel the authors need to address before publication.

Response: Authors thank the reviewer for the helpful and constructive comments on the manuscript. We have revised the manuscript thoroughly for better readability and simplicity.

In the first case study (case I, on Aug 21); the authors obtain measurements upwind in a stratus cloud and follow that with a release of seeding material a bit downwind, which is again followed with additional passes downwind to measure the cloud after seeding. The data shown in Figure 5 (b and c) indicate that most of the measurements downwind of the seeding release show elevated K and Cl. That is somewhat counter to what I would expect. The seeding material should disperse downwind in a slantwise pattern…similar to that shown in Tessendorf et al (2019; BAMS) and other references coming out of SNOWIE. Thus, when flying the 3 passes downwind in this case, I would expect the aircraft to fly through a brief line (region) containing seeding material surround by most of those legs being outside of regions containing seeding material. The authors could develop a simple advection model utilizing the mean wind speed and the time of the release along the seeding leg to estimate how the seeding line advects downwind. That would then provide an estimate of where to (and where not to) expect the presence of seeding material in the cloud.

Response: Thank you for the interesting discussion. Figure 5 (b,c) did not earlier display the spatial distribution of K and Cl; they were plotted with respect to Nt and re. Since aircraft measurements are confined to a limited area, the present data is insufficient to fully explain the spatial distribution of the plumes. Nevertheless, as suggested, we utilized a simple advection model to understand the transportation of the seeding agents in the downwind direction. It's worth noting that we used reanalysis wind data at a 0.25 x

0.25°(https://cds.climate.copernicus.eu/) resolution at an altitude of ~4 km on August 21, 2019, which was further resampled to 0.125 x 0.125°.

As illustrated in the figure below, the research aircraft conducted measurements within the seeding plumes in the downwind direction, after the dispersion of seeding agents.

Now the Figure 5 (shown as Fig. 1B here) is revised as suggested by the reviewer for clarity and better presentation of data.

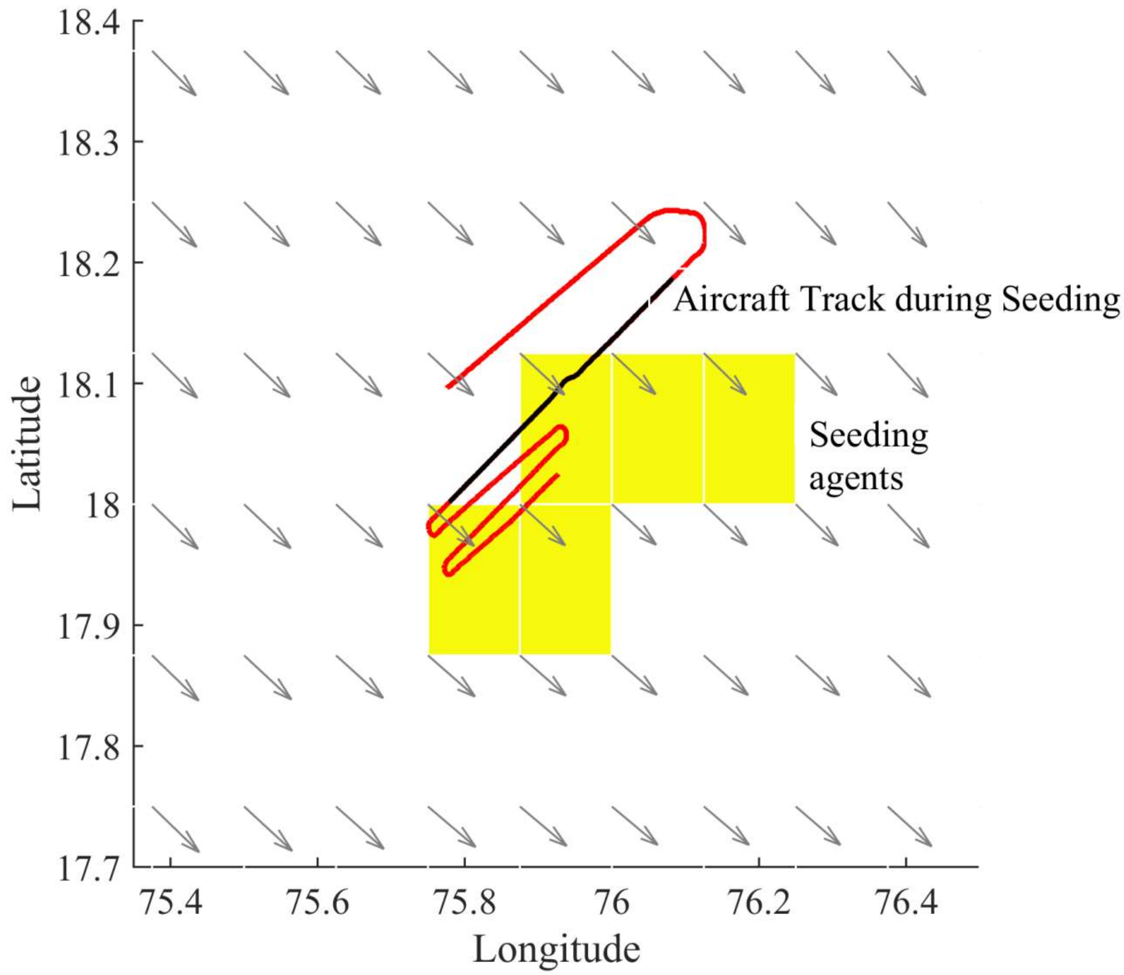

Fig 1A:The red solid line represents the aircraft's track, while the black line represents the moment when the seeding agents were released. Advections of the seeding agents are shown in the downwind direction approximately 3 minutes after the seeding event. Wind data at a resolution of 0.25° x 0.25° is obtained from https://cds.climate.copernicus.eu/ and is further resample to create data at a resolution of 0.125° x 0.125°.

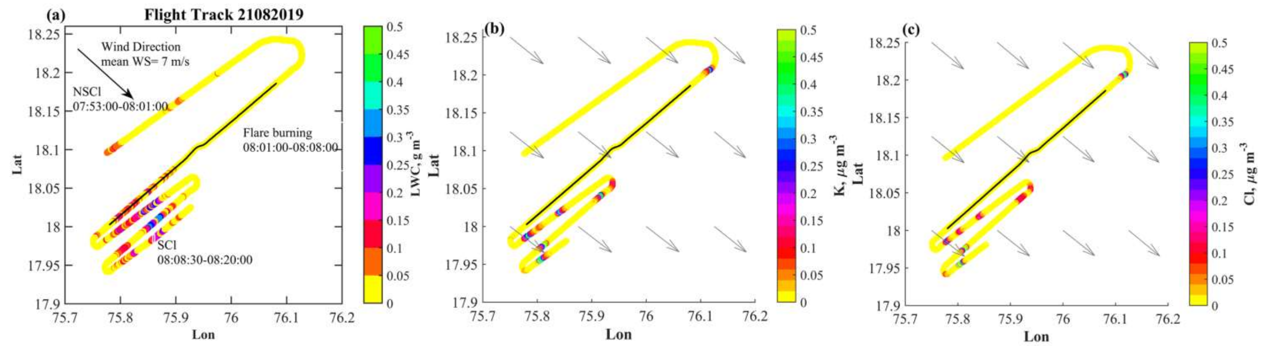

Fig. 1B: (a) Flight path during the seeding experiment on 21 August 2019. Liquid water content (LWC, gm^{-3}) at 1 Hz resolution is indicated. Periods during which cloud measurements were made for non-seeded cloud (NSCl) and seeded cloud (SCl) are indicated. Mass concentrations ($\mu g\ m^{-3}$) obtained from the mAMS of (b) K and (c) Cl during the seeding experiment are shown along the flight track. The spatial background winds shown are obtained from https://cds.climate.copernicus.eu/ (0.25ºX0.25º), which are reampled to 0.125º X 0.125º.

That the authors are able to detect the presence of K and Cl is exciting, however, I think the onus is on them to prove that it is related specifically to the region that contains seeding material. My gut instinct is that it indeed is identifying those regions, but I am still a bit concerned based on the measurements provided in figure 5 as I describe in the above paragraph, that the method ‘over-identifies’ regions containing seeding material. I also wonder, since the same aircraft is used for releasing seeing material and making measurements; is there any potential for contamination? Were any experiments conducted where NSCl was resampled upwind following the release of seeding material?

Response: We could sample the seeding clouds downwind, after the dispersal of seeding agents in the cloud.

Since the stratus cloud usually covers a large area, assuming spatial uniformity in the cloud properties, the measurement of the seeded clouds appears un-contaminated due to the transect made by the aircraft.

Major Comments

1. Introduction—I think the introduction could be stronger. For example, in the second and third paragraphs the authors discuss the use of tracer materials to identify how researcher might understand the transport of seeding material through clouds. They describe two technologies previously used (chaff and chemical tracer such as SF6); while describing downsides of both, I think they miss the main point that neither of those methods would allow investigators to determine if seeding material actually makes it into cloud hydrometeors. This is touched on around line 85 ('can changes is cloud microphysical processing be linked to seeding materials'); in order for this to happen, the seeding material must actually interact with hydrometeors. The method developed in this manuscript does exactly that, by sampling residuals from cloud droplets. The author's should highlight this

Response: Thank you for the valuable suggestion. Now a few sentences are added emphasizing the limitation of past techniques.

2. Lines 131 – 139; and Table 1: Measurements of Cloud properties. The authors need some additional discussion on uncertainties in their measurements. These should come from published literature and simply referencing the manufacturer website is not sufficient. For example, the CDP provides a measurement/estimate of the size distribution, but there are uncertainties associated both with counting (though likely small) and sizing (which could be significant in some size bins)…these errors/uncertainties can lead to further (and much larger) uncertainties in integrated quantities such as liquid water content. The authors use LWC as a threshold in this study to identify regions less likely to be impacted by entrainment/mixing; thus having a discussion of potential errors in this measurement is important.

Response: The uncertainties related to measurements with single particle optical spectrometers, particularly from the CDP and CIP have been quantified and documented in multiple refereed publications. We now state these uncertainties in the modified text along with the appropriate references as written below.

"The uncertainties associated with the CDP, and single particle light scattering instruments like the CDP, have been well characterized and documented (Baumgardner et al., 1983, 2001, 2016; Lance et al., 2010). In water droplets the sizing uncertainty is ±20% and counting accuracy ±16%, which propagates into a LWC uncertainty of ±38%."

Limitation of selecting adiabatic fraction in the seeded cloud is now discussed in the revised manuscript. Please see section 2.1.

3. It appears that in the three cases presented, the authors knew well the cloud base conditions. Lines 144-5; the authors describe a threshold method to identify parcels that are either not or only slightly diluted. Why use LWC_max rather than LWC_ad? A threshold of 0.75 * LWC_max could be a highly diluted if the maximum LWC during that penetration is significantly less than the adiabatic value. Using adiabatic can be a direct estimate of 'how diluted' a parcel may be.

Response: We thank the reviewer for the valuable suggestion. Here, the main goal was to select the DSDs of higher LWC, which in general located in the core region of the cloud transect. The adiabatic LWC values are always greater than the maximum LWC measured in a cloud due to entrainment and mixing processes. In the present study, the adiabatic fraction i.e.LWC_{max}/LWC_{ad} varies between 13% to 96%, which provides some idea of the degree of dilution. This means even in the cloud core entrainment of dry air take place lowering the Maximum LWC value. However, it may be noted that after seeding the distribution of LWC profile changes significantly compared to the non-seeded cloud. Parcel model simulations (without consideration of entrainment mixing process) of seeded and non-seeded cloud suggest changes in the distribution of LWC profiles (please see Figure 7 of Konwar et al., 2023). It is due to the reason that the supersaturation (SS) values at the lower altitude decreases in a seeded cloud (please see Figure 5 of Konwar et al., 2023). It is due to the reason that the coarse-mode aerosols nucleate and cloud droplets grow bigger consuming the available water vapor lowering the SS values. This means that the LWC profile in a seeded cloud would change due to the activation of coarse mode aerosols at the lower altitude. This would result changes in the adiabatic fraction of a seeded cloud. Therefore adiabatic fraction may not be the only parameter to understand the rate of dilution in a seeded cloud. This requires further detail model study with observational data. Limitations of using selecting the DSDs in the slightly diluted region are now discussed in section 2.1.

4. Specific for Seed Case i: Fig 5, Table 3, and paragraph contained in lines 329-343 – Several questions come up when I look at the presentation of this case: How are the incloud times selected for which dots are represented in figs 5b and 5c? I don't think I'd call these 'profiles' in fig5 b & c because all thee downwind (SCl) passes are at one level and the theNSCl pass is at a slightly higher level. I understand wanting a consistency in figure style between this case and the other two; but presenting this case in this style doesn't make sense to me. The data in table 3 indicates that one of the NSCl cases in this case had a LWC of 0.003 +/- 0.003 – this seems marginal as a cloud to me; less than 0.01 LWC is below what most would consider in cloud. It

seems odd to include data such as this as a comparison of cloud? LWCs are generally higher after seeding (although the measurements are lower in cloud…why? Seeding should not result in increased condensed water, but rather just changes in how that condensed water is distributed across hydrometeor distributions. On line 340 the authors suggest that increase in mid- sized droplet concentration may be due to collection, but this does not make sense…these droplets are <20 µm collection is extremely inefficient at these.

Response: Thank you for the valuable suggestions.

We agree the cloud pass had very small LWC values. We now have selected a new NSCl pass of nearly 30 sec with higher LWC than the previous case. As suggested the word 'profile' is removed now for this cloud.

As suggested, the presentation for this seeding case is now changed for clarity.

5. For Seed Case ii: In Fig 6 b and c – Need a better way to differentiate between which dots represent NSCl and which represent SCl – perhaps shape? One could be square— the other circle? Line 392 – the authors claim that after seeding Nt increased at lower altitudes—I disagree. The lowest altitude above cloud base sampled before seeding was 0.8 km and after seeding was 0.3 km – these two cannot be compared---the so seed is more than twice the distance above cloud base. Further, in the after seeding case, a pass was made at 0.96 km, and that contained lower concentrations than the no seed at 0.8 km. The authors need more rigor in their data analysis before making such claims. On line 398 the authors claim that large standard deviations in re are likely the result of entrainment/mixing with dry air….I agree; but then they need to be more rigorous in trying to eliminate regions which are influenced by mixing in their analysis. This is a possible indicator that their threshold of 0.75*LWC_max is not sufficient to do accomplish this. On line 412 the authors suggest that because drizzle is encountered after seeding and not before that the production of drizzle can be attributed to seeding…while this is one possibility, there are many other possibilities that could account for that. Different stages of lifecycle, pre-conditioning of the environement, etc are all possibilities. Studies of cumulus continually show that drizzle may form at the same levels in a cumulus cluster that indicated no drizzle development several minutes earlier.

Response: As suggested the symbols are improved and now distinguishable for SCl and NSCl cases.

We have revised the discussion keeping in mind the limited data and a statement is included on the small data volume. We already had acknowledged the possible role of natural variability on drizzle production.

6. For Seed Case iii: Why is the LWC 3-5 times higher after seeding than before, when observations were made at the same level above cloud base? One possibility is that these difference are due solely to difference in amount of dry air mixed into parcels that were sampled, and any differences in microphysics may be attributed to that rather than to seeding?

Response: Thank you for pointing out the important aspect. Now we have done additional analysis to indicate the role of strong updrafts in these clouds. A discussion is now added to the revised manuscript: "Another aspect to consider here is the effect of strong updraft of 8 m s^{-1}. Using the Twomey (1959) equation the maximum droplet concentration formed in an updraft (W) can be expressed in terms of W and CCN-SS spectra, i.e. N_{CCN}=C SS^k i.e. (Roger and Yau, 1989),

$$N \approx 0.88\, C^{2/(k+2)} \left[7\, X\, 10^{-2}\, W^{3/2}\right]^{k/(k+2)} \qquad (4)$$

Here, W is in cm s^{-1}, N_{CCN}= 799 $SS^{0.43}$, which is obtained from the CCN counter (Roberts and Nenes, 2005; Nenes et al., 2001 and reference therein) operated in the research aircraft. During the cloud passes, maximum updrafts of W= 2.89 m s^{-1}, 1.00 ms^{-1} and 1.91 m s^{-1} were obtained. These values suggest that droplets formed in these updrafts could be 593 cm^{-3}, 448 cm^{-3} and 531 cm^{-3}, respectively. If we use the maximum updraft speed of 8 m s^{-1} measured below cloud base, the droplet concentrations formed in this updraft could be as high as 777 cm^{-3}. Therefore, the presence of strong updrafts that yield high SS could be one reason for the increasing N_t in the seeded clouds; while dry air mixing in the NSCl cases could be another reason for the smaller concentration of N_t. "

Specific Comments

1. Abstract, Line 31 – ….attempts to link precipitation enhancement….remained inclusive? Not sure what is meant by this, could it be the authors mean that attempts remained elusive?

Response: Yes now the word ‘elusive’ is used.

2. Abstract, Line 37 -- ….cloud droplets underwent a drying process, … I think it would be more accurate to state that droplets were evaportated.

Response: We agree, now it is revised.

3. Line 67 – ‘…then these traces are tried to measure higher in the cloud.’ Wording here is incorrect.

Response: OK, now the sentence is revised.

4. Line 91 – ‘…to identify seeding material in the residual cloud droplets.’ The seeding material is identified within ‘cloud droplet residuals’; i.e. the aerosol that remains after evaporation of the cloud droplets.

Response: OK, now the sentenced is revised.

5. Line 92 – ‘….hypothesis relies on a chain of microphysical mechanisms.’ The wording here is not correct; there is no such thing as a chain of mechanisms. I think the authors are referring to a chain of events with each event being a specific microphysical process.

Response: We referred to the chain of microphysical processes. Now the sentence is revised.

6. Line 99 – ‘CCN do..’ (not does, CCN is plural)

Response: OK, now the sentence is corrected.

7. Line 107 – replace ‘seed’ with ‘seeding agent’ and delete the word ‘the’ before broadening.

Response: OK, now corrected.

8. Sentence on lines 111-113 – I read this sentence in relation to the evolution of microphysical processes resulting from hygroscopic seeding (previous sentence); however, this has been accomplished in glaciogenic seeding experiments as the authors reference French et al. (2018). It seems to me that the processes are more dynamic and complex in cold clouds, yet that linkage has been made.

Response: This reference was included to emphasize understanding of physical processes of cloud seeding experiment, in general. The reviewer is correct, now this sentence is precluded from the revised manuscript.

9. Lines 127-129 – isn’t all the data presented in this study from ‘warm’ cloud seeding? Why reference flare racks for ‘cold’ cloud seeding. And, by ‘warm’ and ‘cold’ are the authors referring to hygroscopic vs glaciogenic seeding? If so, they should be referred to as such.

Response: Two relevant references are cited in the revised manuscript.

10. Use of abbreviations and nomenclature for height of measurements in cloud— introduced on Lines 146 & 7 – The authors use the nomenclature 'cloud depth' (abbreviated CD) to describe the heights (above cloud base) for measurements throughout the manuscript. I find this confusing as 'cloud depth' normally refers to a measurement of the total depth (or thickness) of a cloud. I suggest the authors should change all instances in text, figures, and tables from 'cloud depth' to 'height above cloud base'.

Response: Ok, as suggested we now used the term D* (km) for vertical distance above the cloud base.

11. Line 161 – Delete 'As mentioned earlier'

Response: OK.

12. Line 165 & 6 "…the CVI inlet segregates and samples cloud elements." Replace that with: 'the CVI inlet cloud hydrometeors with aerodynamic diameters larger that a certain size depending the rate of the counter flow.

Response: OK.

12. Line 165 & 6 "…the CVI inlet segregates and samples cloud elements." Replace that with: 'the CVI inlet cloud hydrometeors with aerodynamic diameters larger that a certain size depending the rate of the counter flow.

Response: OK.

13. Line 168 – 'larger than >7 μm' ' how was this verified for the setup in this experiment? The cutoff size is dependent not only on flow rate, but also aircraft speed, possibly mounting location on the aircraft, altitude, etc. Verifying this (somehow) seems to be a critical thing in this experiment.

Response: The flow rates of the CVI were verified using a Gilibrator flow meter as the standard before the project started. The CVI adjusted flow rates with its internal software based on true air speed (TAS) provided to the CVI computer from the AIMMS probe. The AIMMS measured true air speed and the M300 *acquisition system* sent this out to the CVI once per second and flow rates were adjusted accordingly. Flow rates were monitored and verified that they were being kept at the set points. Brechtel Engineering, the provider of the CVI, has characterized the cut-size as a function of air pressure and air speed. The angle of the CVI was determined from the average angle of attack. The reviewer is correct that a number of factors determine this cut size and the text has been modified to state that this cut-size is known to approximately +/- 1 um. This has little impact on the results and conclusions.

In addition to the above point we want to state that we have now corrected the mAMS data for enhancement factor as the ambient aerosol concentration concentrated in the CVI tip (Shingler et al., 2012). A sentence on this aspect is now added page 15.

14. Line 172 – I cannot access the Golderger et al. (2020) reference.

Response: Thank you for pointing this out. The following link is now provided in the reference list: https://www.arm.gov/publications/tech_reports/handbooks/doe-sc-arm-tr-254.pdf

15. Lines 230 & 231 and Figure 3—Is 'start' and 'stop' the times of 'Pass Begin' and 'Pass End' shown in the figure? If so, be consistent in labeling. Label 'maximum total mass time' in figure 3.

Response: OK, now corrected.

16. Lines 253 through 260 – The authors repeat items that are already discussed earlier and restate the same things multiple times in lines in this paragraph. I think the authors should rewrite this paragraph to make it more concise and less repetitive.

Response: This paragraph is now revised for clarity.

17. Line 285 and 286 reword to: …'with three cloud passes of the same cloud before and three passes after seeding are shown in Fig 4.'

Response: **OK.**

18. Line 288 – change 'above' to 'compared to'

Response: **OK.**

19. Line 290 – the wording seem awkward here…maybe change 'consist of' to 'containing'

Response: **OK.**

20. Line 291 & 2 – I do not see increased SO4 concentrations in the seeded clouds (perhaps at mid level, but certainly not at the highest level…

Response: Now this sentence is revised, mentioning mid level.

21. Line 307 – 'several cloud passes….were made….before dispersal of seeding material.' From Figure 5a, it looks like one pass was made.

Response: Now the sentence is revised.

22. Line 311-314 – The AIMMS 20 does not measure whether in cloud, I assume the authors use some threshold on the CDP measurements? Possibly a threshold on the LWC derived from the integrated size distribution?

Response: Now it is mentioned, Nt>10 cm^{-3} and passes greater or equal than 5 sec was the criteria.

23. Line 326 – sentence that begins ‘Discussions on cloud probes…’ ---delete this.

Response: OK.