Response to Reviewers, Simmons et al.

We appreciate the reviewer's close reading of the paper and have made changes to the manuscript accordingly. See detailed responses below. The comments from reviewers are in roman font and our response is immediately below in blue italics.

Review 1

Major comments:

1. It seems odd to me that the LWC should increase so much following the injection of NaCl and the injection of NaCl-Ti02. The baseline cloud in both cases had ~0.05 g/m3 LWC, but had spikes up to and exceeding 4 g/m3 after the test aerosol were injected. Is that much vapor available for uptake even if the RH in the chamber drops to the deliquescence value of ~80%? Also, why is there so much variability in the LWC values---I would not expect that. Is it possible that this is a result of poor sampling statistics of the larger droplets that are contain most of the mass of liquid?

At 20 C, the saturation vapor density is 17.3 g/m^3. When the powder deliquesces, RH drops from 100% RH to 75% in the local volume where it is dispersed. This equates to ~4g/m^3 of liquid water condensing.

There is, however, an intrinsic heterogeneity in the LWC after injection, particularly in the first 30 to 60s because of the local powder dispersal (the balloon burst) and because LWC is dominated by the larger droplets, which are not mixed uniformly through the chamber, especially in the time immediately after the dispersal of the powder. We are confident that the LWC does increase above the baseline value (for the reason outlined above) and we are showing the 1 Hz data so as to be transparent in the measurement that was actually made.

We have added a paragraph in the discussion of Figure 2 (also referencing Fig. 3) which outlines the uncertainties associated with the LWC derived in this way.

2. The results from the Arizona dust are also interesting, and seems counter-intuitive. The concentration increases significantly (albeit, mostly at small sizes, Figs 1 and B3)….this must result in a significant increase in LWC (not shown), but why? Why should the LWC increase after the injection of the dust?

Liquid water content does increase when we inject the dust, but not for the same reason as the hygroscopic powders. The dust overseeds the cloud, such that there is an enhanced concentration of small drops. Those small drops have a smaller fall speed and thus their residence time in the chamber is longer, which results in higher LWC. LWC content also increases when we inject hygroscopic burn in place flare smoke, for the same reason. (Those tests are the topic of another paper.)

Also note that the Welas is an optical particle counter. We can't differentiate between liquid water and dust particles using just that. The AZ dust is composed primarily of non *hygroscopic silica (69-77%) and aluminum oxide (8-14%), so all of the injected powder might not nucleate as water droplets as the NaCl and CSNT do. In the supersaturated cloud chamber, many of the dust particles would have been able to activate (this is evidenced by the shifting of the droplet mode in Figure 1 towards larger sizes in the minutes after injection), however the inability to discern liquid water from dry dust is the reason we have not computed liquid water content for the AZ dust injection.*

We have added a paragraph in the Methods section outlining the assumptions that the counters make and the uncertainty this may introduce in the size distribution.

3. For NaCl, following 600 s after injection the cloud returns to something that looks pretty similar to the baseline cloud (Fig B3), but this is less true for the NaCl-TiO2 and even less for the dust. Why?

As shown in Fig. A2, a significant portion of the injected NaCl powder remains in large particles. The resulting droplets formed on these large particles fall out of the chamber within the first two minutes, and the baseline cloud quickly recovers after this. In the case of the CSNT and AZ dust, a larger fraction of the injected mass is contained in particles smaller than 1 µm. These persist in the chamber for much longer, and in this time dominate the DSD (visible in Fig. 1) and delay the recovery of the baseline cloud.

The slow return of the baseline cloud is noted in the text (see first paragraph in Sec. 3.1). We have added a paragraph elaborating upon the temporal evolution of the LWC in the section where the dust tests are presented.

4. What are uncertainties associated with the measurements provided by the Welas and the CDMS-2? Are they identifying some of the dust and/or dry particles as liquid drops? If so, are they missizing them? While the results from the 2 instruments look similar (Fig 1 and B1), there are certainly differences. Are those differences significant when calculating bulk properties of the cloud? What does the CDMS-2 measure during the dust test?

The Welas and CDMS-2 both detect scattering of light by the measured particles. That signal is inverted to a particle size, given some assumptions about shape (sphericity) and refractive index. When those assumptions are not met, (for example when dry salt or dust is the scatterer), the reported size will be incorrect. As noted above, this one reason we chose not to report LWC for the AZ dust injection case. With the NaCl and CSNT injections, we assume that all of the material deliquesces, and this is supported by the drop in RH in the chamber.

The CDMS was not available at the time of the AZ dust injections.

We assume differences between the size distributions from the CDMS and Welas can primarily be attributed to differences in instrument positions within the Chamber, turbulent fluctuation in the chamber, and differences in measurement range and bin distributions. These do not impact the calculation of bulk cloud properties or qualitative analysis of the cloud DSD response to injection.

We have added a paragraph in the Methods section outlining the assumptions that the counters make and the uncertainty this may introduce in the size distribution.

Minor comments:

1. Many of the figures are plotted as a function of time and demonstrate changes from before to after the injection of the test aerosol. However, the authors change the reference times used in many of the figures (Fig 1 for instance has the injection at shortly after 600 s, Figs 2 & 3 have them shortly after 900 s, and fig B1 have time in minutes). All of these figures should use the same time scale…and I further suggest that time t=0s can be set as the injection time, such that pre-injection, time is negative and post injection, time is positive.

Done.

Review 2

Major comments

- Cloud collapse: In Line 89, the definition of the cloud collapse is not properly done in my opinion. Only after presenting all results, a discussion and hypothesis on the process chain is done. I believe the understanding of this process chain is crucial and comes here too short. It is fine to have the discussion of the cloud collapse after the first result presentation, but then I would avoid having this very short definition here early on in the text. It rather raises questions than it helps the reader's understanding.

We have removed mention of "cloud collapse". We now refer to dissipation of the cloud, which should remove the ambiguity in phrasing.

- Changes in LWC: I a missing some uncertainty parameters on the measurements. Looking at Figure 3a, there is a single data point (is it one measurement or an average?) at 10 g/m3 right after the injection seeding material. Is this trustworthy? How confident are the authors on the single data points? Would a running mean help here?

There is an intrinsic heterogeneity in the LWC after injection, particularly in the first 30 to 60s because of the local powder dispersal and because LWC is dominated by the larger droplets, which are not mixed uniformly through the chamber, especially in the time immediately after the dispersal of the powder. We prefer to show the 1 Hz data so as to be transparent in the measurement that was actually made. We are confident that the single data points are accurate representations of the size distribution measured in that time period for that point in the chamber. We prefer to show the "noisy" 1 Hz data because it better captures intrinsic heterogeneity in the system.

We have added a paragraph in the discussion of Figure 2 (also referencing Fig. 3) which outlines the uncertainties associated with the LWC derived in this way.

- Different responses across the seeding agents: I would be curious to hear the authors thoughts on the underlying mechanisms in the seeding responses of all three agents. What is the mechanism for the dust sample? If it does not grow hygroscopically, what is happening? Why is the change in Ts higher for NaCl than for CSNT (Figure 4)? Why is the cloud collapse longer-lived in CSNT than for NaCl?

The AZ dust will still adsorb water and nucleate water droplets in supersaturated environments. This can be seen in the appearance of the larger particles when dust is injected, but also the fact that small mode in cloud droplets is enhanced upon dust injection. The greater change in Ts for NaCl indicates that a higher mass of water vapor is condensing onto droplets. The larger mass of the NaCl dispersed into the chamber will result in more water condensing out, even if a substantial portion of that mass is removed very quickly (i.e. the large clumps) through gravitational settling. We are unsure why the cloud dissipation appears to last longer for the CSNT test.

Minor comments

- "microns": in several instances throughout the manuscript microns is used, I would prefer the proper unit in µm.

References to "microns" have been changed to µm or micrometer

- Line 9: I would specify the size of the background cloud population to highlight the change in size more clearly

We have removed the reference to the background cloud at this point in the manuscript, preferring to focus on the the increase in LWC and appearance of large droplets.

- Line 17: I would just call them ice nucleating particles, and avoid having after the first sentence already a bracketed information.

We have removed the parenthetical statement, but we prefer to leave sublimation nuclei to preserve the sense of the original papers.

- Line 27: Here caking is used without explanation, while in Line 31 caking is added after dispersal. A short explanation about that would help to make the manuscript more accessible to wider audience.

We have added a brief explanation of caking in the Introduction after the paper by Biswas and Dennis is mentioned.

- Line 39: "KCl" was not introduced and is not used later, so I would spell it out.

Updated to "potassium chloride"

- Line 88: I do not expect any appendix on the tests of dry air entrainment, but would like to have some more quantifying statement than "minimal impact".

We have added additional context. "No change in the DSD or droplet concentration as measured by the Welas was observed after the bursting of empty balloons".

- Line 97: "drop" to "droplet" to be consistent with the wording in the manuscript

Changed all instances of water "drop" to "droplet".

- Figure 2: why is here the injection time at 900 s and not 600 s (same goes for Figure 3)? What do the data points represent? Are these averages or the 1 Hz recorded data points?

Figs 1, 2, and 3 have been replotted with the moment of injection labeled as t = 0. Previously, the time series in Figure 1 were cropped and plotted using relative timestamps so that the injection times all lined up at exactly 600 s, whereas Figs 2 and 3 show the actual injection times within the measurement duration.

The DSDs in Fig 1 show a running average over 5 s increments because the resulting color contours are less noisy and therefore easier to read. The LWCs in Figs 2 and 3 are the raw 1Hz data, which better illustrates the fluctuations immediately after injection as well as the cloud response on shorter timescales.

- Line 138: second "and" after CSNT is too much

Corrected.

- Figure 4: I recommend to replot this figure by avoiding having same line widths for the injection time and the time series. It is hard to distinguish. Changing the line width for the injection time as well as the color for CSNT will solve this issue.

The colors of the time series have been changed so that it's easier to differentiate between the CSNT, NaCl and injection markers. The time series have also been shifted so that injection occurs at t = 0 s

- Appendix B: the additional figures should at least be referenced in the main text.

The additional figures are now referenced in the caption to Fig. 1.