Responses to reviews of "Technical Note: Equilibrium droplet size distributions in a turbulent cloud chamber with uniform supersaturation" by Krueger (acp-2019-932)

1 Reviewer Comments 1

The author thanks Reviewer 1 for his/her careful reading of the manuscript and helpful suggestions and comments. All were incorporated.

Specific Comments

Size distribution at $r = r_a$

From Eq. (9), it can be written as below

$$v(r_a) = \frac{r_a}{\xi}\int_{r_0}^{r_a} A(r)dr.$$

On the other hand, from the general solution Eq. (12),

$$v(r) = Dr\mathsf{exp}(-Cr^4/4).$$

Does this mean that Eq. (9) can be related to Eq. (12) by substituting $r = r_a$ in Eq. (12)? I think it might be informative for readers if the author adds an explanation on how Eq. (9) is connected to the general solution Eq. (12).

RESPONSE: Eq. (12) is now solved for D when $r = r_a$. The resulting expression is given by Eq. (13) which contains the boundary condition, $v(r_a)/r_a$ given by Eq. (9). The link between Eq. (12) and Eq. (9) should now be clear.

Nominal supersaturation in Fig. 6

In page 14, line 4, the author cites Rogers and Yau (1989) and explains that the critical radius for injected NaCl particles is about $r_* \sim 0.6\mu$m. I think the same textbook also gives an estimation of the critical supersaturation for those particles and I expect it to be about $S_* \sim 0.1$%. On the other hand, according to Figure 6, the inferred nominal mean supersaturations for the Pi-chamber experiments with two largest number densities of cloud droplets are smaller than 0.01% ($\overline{S}_{\mathsf{nominal}} < 0.01$%). This seems somehow strange, because aerosol particles cannot be activated to cloud droplets if the supersaturation $\overline{S}_{\mathsf{nominal}}$ is much smaller than the critical supersaturation S_*. Does the author have possible explanations for this apparent discrepancy? If so, providing those explanations in the manuscript might be helpful for readers.

RESPONSE: This comment motivated an important extension to the original study. The apparent discrepancy occurs because the analytic solution omits droplet curvature and solute effects, and therefore exhibits no dependence on aerosol properties, and does not have a critical supersaturation. In response, I modified the manuscript extensively in section 6.

On page 15, I gave a more complete characterization of the aerosol size distributions used in the cloud chamber and stated that "The potential impacts of both droplet curvature and solute effects on comparisons of analytic and measured DSDs will be discussed below, in section 6.2."

I added section 6.2 (pages 17–19) on inferred mean supersaturation and droplet activation. In this section, I noted that 99% of the injected aerosol particles have a dry diameter less than about 170 nm. The critical supersaturation for a NaCl particle with a dry diameter of 170 nm is 0.052%. I also noted that 6 DSDs in Figure 6 have inferred supersaturations less than 0.052%. The rest of section 6.2 discusses the implications of this situation. I discussed the following possibilities (excerpted from the revised manuscript):

1. Neglecting droplet curvature and solute effects in the analytic DSD governing equation produces significant underestimates of the inferred supersaturations. It could be that once curvature and solute effects are included in the droplet growth equation, the inferred mean supersaturations for all 11 measured DSDs will be large enough to activate at least the largest of the injected aerosols.

   To investigate this possibility, we used the droplet growth equation, both with and without the curvature and solute terms included, in the Monte Carlo model described in section 3 to calculate mean droplet radius versus supersaturation for 100 supersaturation values (Figure 8). Figure 8 shows that the mean droplet radius is smaller when these terms are included, for the same fixed supersaturation. This is due to the slower initial growth of the droplets. The differences in mean radius are largest for supersaturations slightly larger than the critical supersaturation.

How do the curvature and solute terms affect the inferred supersaturation? For a given droplet radius, the inferred supersaturation is larger with solute and curvature terms included. In our specific case, Figure 8 suggests that a measured DSD ($r > 2.5$ μm only) with a mean radius of about 4.4 μm or larger could have been activated and grown with a fixed supersaturation of 0.055%. Figure 6 shows that this requirement excludes the measured DSDs with the 5 smallest mean radii.

2. Even after including droplet curvature and solute effects, the inferred supersaturations of the 5 measured DSDs with the smallest mean radii are less than the critical supersaturation of the largest of the injected aerosols. In this case, we conclude that there must have been supersaturation fluctuations somewhere in the cloud chamber that exceeded the critical supersaturation for at least the larger injected aerosols. There are two possible situations:

   (a) Large supersaturation fluctuations occur only near the bottom and top boundaries of the cloud chamber, as is typical of Rayleigh-Bénard convection. In this case, it could be that activated droplets are transported away from the boundaries and then continue to grow consistent with inferred mean supersaturations calculated with droplet curvature and solute effects included. This scenario is analogous to droplets growing in a cumulus updraft: The droplets are activated by relatively large supersaturations just above cloud base, but then continue to grow in lower supersaturations at higher levels (Rogers and Yau 1989).

   (b) Droplet growth in the chamber for these DSDs is primarily or entirely due to supersaturation fluctuations throughout the cloud chamber. In this case, the analytic DSD solution, which assumes that there are no supersaturation fluctuations, is not valid. Chandrakar et al. (2020b) found that analytic solutions for DSDs when mean supersaturation is absent (but fluctuations are present) have nearly the same shape as DSDs for no supersaturation fluctuations. As a result, it is difficult to distinguish the two cases based only on the consistency of the moments.

I also revised the conclusions by adding the following text to pages 25–26:

> We found that neglecting the curvature and solute terms in the droplet growth rate equation can sometimes affect the inferred supersaturations. For a given droplet radius, the inferred supersaturation is larger with solute and curvature terms included. Calculations with a Monte Carlo model with solute and curvature terms included suggest that for the aerosols injected into the cloud chamber, a measured DSD ($r > 2.5$ μm only) with a mean radius of about 4.4 μm or larger could have been activated and grown with a fixed supersaturation of 0.055%. This excludes the DSDs with the 5 smallest mean radii. To produce these DSDs, there must have been supersaturation fluctuations somewhere in

the cloud chamber that exceeded the critical supersaturation for at least the larger injected aerosols.

Technical Corrections

1.	p. 3, Sec 2.3, line 1	:	$u/h\Delta t = k_1 r^2/h\Delta t$	$\longrightarrow$	$(u/h)\Delta t = (k_1 r^2/h)\Delta t$
2.	p. 4, line 6	:	$k_1 r^2/h\Delta t$	$\longrightarrow$	$(k_1 r^2/h)\Delta t$
3.	p. 9, Figs. 3 & 4, y-axis	:	$\mathsf{pdf}(\mu\mathsf{m})^{-1})$	$\longrightarrow$	$\mathsf{pdf}((\mu\mathsf{m})^{-1})$ or $\mathsf{pdf}(\mu\mathsf{m}^{-1})$

All done.

2 Reviewer Comments 2

The author thanks Reviewer 2 for his/her careful reading of the manuscript and helpful suggestions and comments. Essentially all were incorporated.

Minor Comments

Sec. 2.4 I believe that Srivastava (1991) also requires some recognition in this subsection. He investigated, also analytically, the mean, standard deviation, and dispersion of droplet spectra, including the effects of droplet surface tension.

Srivastava (1991) is now cited in section 2.1 (p. 3, line 9).

P. 4, ll. 28 ff. A supersaturation of 10 % is relatively high for a typical cloud. For plotting the analytical solutions, a more realistic value of 0.1 % is used. I suggest to also use this lower supersaturation in the Monte-Carlo calculations of section 3. However, this will not change any conclusions.

Done. (Figures 1 and 2 replaced.)

P. 5, ll. 11–12 I would emphasize that the "stochastic nature of the droplet fallout process" includes the assumed stochastic rearrangements of the droplets along the z-axis, i.e., turbulent motions, since the sedimentation process itself is deterministic.

Done. (P. 5, last sentence.)

Sec. 4.2 It is possible to speed up the derivation of $w(s)$ using $v(r)$...

Replaced original derivation with this one (Sec. 4.2, pp. 7–8).

Eqs. (12) and (17) Because Eqs. (12) and (17) clearly violate the assumptions $v(r_0) = w(s_0) = 0$, it might be helpful to state—again—that the analytical solutions are only defined for $r > r_a > r_0 > 0$ and $s > s_a > s_0 > 0$.

Done. (P. 8. line 12).

P. 7, l. 10 The first term of the equation contains one minus sign too much.

Eq. no longer included because of comment on Sec 4.2.

Eq. (26) Also this deviation can be shortened: ...

Done. (Eq. (22) on p. 10.)

P. 21, l. 20 The usual citation for the phase relaxation timescale is Squires (1952).

Replaced. (P. 25, line 10.)

Technical Comments

P. 2, l. 26: Throughout the paper, the author uses plural personal pronouns ("we" or "us"). Thus, this single "I" feels odd.

P. 3, l. 27; p. 4, l. 6; p. 4, l. 26: For clarity, add parentheses to the equations for the fall out probability: $(u/h)\Delta$t and $(k_1 r^2/h)\Delta$t instead of $u/h\Delta$t and $k_1 r^2/h\Delta$t, respectively.

P. 4, l. 31; p. 5, l. 9: Since the analytical solution will be introduced further below, I suggest adding a "to-be-determined" in front of "analytical solution".

P. 6, l. 1: $A(r)$ has been previously introduced as the "production of (activated) droplets from the injected aerosol" (p. 3, ll. 8 – 9). Here, it is called the "production of droplets by activation". Although these processes are identical in the described framework, I suggest homogenizing the terminology.

Eq. (18): I would add a comma (",") to the end of the equation.

Figs. 3 and 4: An opening parenthesis ("(") is missing in the ordinate title.

P. 10, l. 14: I would add a comma (",") to the end of the equation.

P. 11, ll. 20 – 22: This comment feels too technical. I would omit it or state this information in a footnote.

All of these were done except for the last one. The ACP Author's Guide states that footnotes should be avoided, as they tend to disrupt the flow of the text. In addition, there was not a good place to add a footnote.