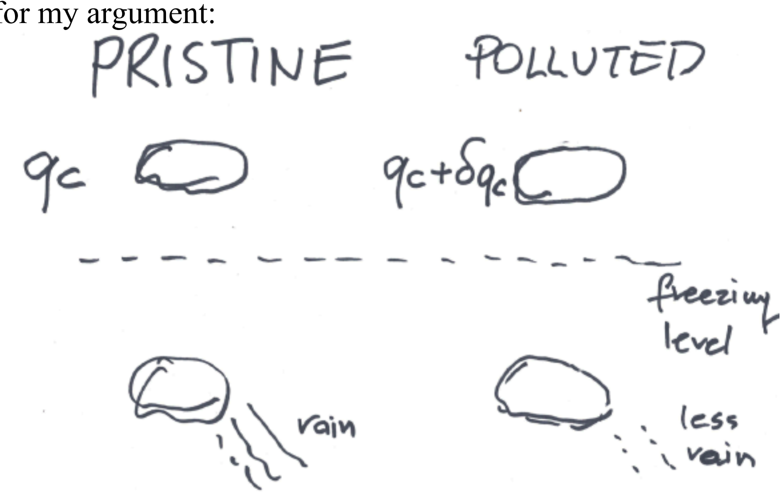
Responses to the reviewer' questions about the cold-phase and warm-phase invigoration by aerosols.

The reviewer's comments:
It presents the authors view that is not supported by simple arguments and by other studies. For instance, the "cold-phase invigoration" as described in lines 42-45 is simply not possible because the latent heat released by freezing the cloud water carried across the melting level in the polluted case only balances the weight of the water carried upwards. So where does the invigoration come from? The explanation of the "warm-phase invigoration" is simply incorrect and it repeats the incorrect argument used in papers the authors cite. The latent heating does not depend on the droplet concentration and droplet radius as long as the updraft velocity does not change. This is strictly true when the in-cloud supersaturation is equal to the quasi-equilibrium supersaturation. The validity of the quasi-equilibrium supersaturation approximation has been argued in many studies, at least in the absence of ice (e.g., Politovich and Cooper JAS 1988). Such an incorrect interpretation is repeated in lines 334-337. I suggest the authors consult the recently accepted manuscript by Grabowski that provides a thorough discussion of the two invigorations, see section 2 there. The manuscript is available on EOR in JAS (https://journals.ametsoc.org/doi/abs/10.1175/JAS-D-20-0012.1). I also suggest the authors consult and cite a paper by Varble (JAS 2018) for a less biased discussion of the invigoration problem.

Here are the follow-on comments from the reviewer on the cold-phase and warm-phase invigoration.

I appreciate the authors' responses to my initial comments. However, the response has fundamental flaws and thus I cannot consider the issues settled. I think the authors need to reconsider my comments about the invigoration and correct obvious flaws in their arguments.

About the cold-phase invigoration:
The authors clearly misunderstood my argument. The figure below illustrates the starting point for my argument:



The left part of the panel shows a cloudy parcel that rises through the melting (freezing) level in pristine conditions. The total liquid condensate above the freezing level is q_c. The right part of the figure shows situation when a similar parcel rises in polluted conditions. Because of the less efficient warm rain processes, the parcel above the freezing level carries more liquid condensate, $q_c + \delta q_c$. Freezing of δq_c in the authors' opinion (and in many other papers) is the reason for the invigoration. However, to carry δq_c across the freezing level requires extra buoyancy when compared to the left panel. As shown in section 2a of Grabowski and Morrison (2020), the two effects approximately balance each other. It follows the original sentences in the manuscript under review that say: "… a well-known theory is that increasing aerosol concentrations can suppress warm rain as a result of increased droplet number but reduced droplet size. This allows more cloud water to be lifted to a higher altitude wherein the freezing of this larger amount of cloud water induces larger latent heating associated with stronger ice microphysical processes, thereby invigorating convective updrafts…" are simply incorrect and require additional explanations. For instance, the invigoration would be possible if the frozen condensate was removed through precipitation processes.

That said, I really do not think referring to invigoration is needed for this manuscript. If the authors insist, then the introduction and references to the invigoration in the text should provide a less biased discussion, for instance, as in Grabowski and Morrison JAS papers and as in Varble (JAS 2018).

About the warm-phase invigoration:

The phase relaxation time scale and the quasi equilibrium supersaturation estimates in the authors' response above are simply wrong. Below I include a table from Politovich and Cooper (JAS 1988) that shows phase relaxation time scale for different combinations of droplet concentrations and radii. These values are much smaller than those shown in the authors' response.

TABLE 1. Time constant characterizing supersaturation. (Values of $\tau = 1/(a_2 I)$ s for p = 771 mb, T = 4.3°C)

Radius (μm)	Droplet concentration (cm^{-3}) 100	300	500	1000
2	14.1	4.7	2.8	1.4
3	8.7	2.9	1.7	0.87
5	4.9	1.6	0.98	0.49
10	2.3	0.77	0.46	0.23

The key question is why?

The explanation is relatively simple. The authors say that they use the formulas from Pinsky et al., eq. (4) therein. However, Pinsky et al. apply a simplified (and in my view incorrect) droplet growth equation that is different from the comprehensive formula used in Politovich and Cooper (JAS 1988). The key point is that one has to apply exactly the same droplet growth equation in the phase relaxation time scale calculation (and thus in the quasi-equilibrium supersaturation) as used in the numerical model. I expect Khain's bin microphysics applies a correct droplet growth formulation that is close to the one used in Politovich and Cooper (JAS 1988), and not the simplified droplet growth equation applied in Pinsky et al. The supersaturation simulated by the model can be compared to the diagnosed quasi-equilibrium supersaturation only if exactly the same droplet growth equations are used in both. This was the case for the relatively good agreement shown in Grabowski and Morrison (JAS 2017), at least below the freezing level, see Fig. 15 therein. In summary, the values shown in the authors' response above have to be

Corrected.
The above discussion requires the authors to modify their responses and revise their paper accordingly. Note that the second part of my rebuttal impacts some of my other original comments. I strongly object publication of the manuscript unless those comments are appropriately addressed

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As we noted earlier, the two sides of arguments have been existing for a while, and it should not be the role of this paper to debate and resolve this issue. Here we only provided our key points, the detailed review and comment paper was submitted to J. Atmos. Sci., which would allow both sides to discuss and debate there. The bulk of the above comments are chiefly the expression of the reviewer's view on the aerosol invigoration effect, rather any substantial objection to the scientific importance and the findings of this study.

For the "cold-phase invigoration", the reviewer's argument "the increase in the buoyancy by freezing is completely offset by the buoyancy for carrying the extra cloud water across the freezing level" has several issues:
(1) Droplet ascending and then freezing are subsequent at different locations; also, the two processes can take at very different time scales (freezing is instant but ascending could take much longer time). How do they compensate each other at different time scale and locations? Responses of a complex non-linear dynamical system in deep convective clouds strongly depend on duration and location of the forcing.
(2) In the process of ascending in updrafts, droplets will grow through condensation, and the changes in latent heating and condensate loading from this are not considered in this argument.
(3) The argument neglected the subsequent enhanced riming and deposition resulting from more ice particles formed from enhanced droplet freezing. This leads to (a) a further increase in latent heating and (b) a reduction in condensate loading because more graupel and hail form due to increased supercooled liquid content and precipitate. Rosenfeld et al. (2008) indeed considered the possible compensation between the extra condensate loading and the extra latent heat of freezing. They showed (in line d of their Fig. 3) that the total buoyancy excesses and invigoration occurs after the ice hydrometeors are unloaded (i.e., precipitated) from the cloud parcel. The unloading is quite efficient in the case of rich supercooled liquid water content where large particles like graupel and hail can form. Many modeling studies have showed that the latent heat released from deposition and riming is much larger (at least an order of magnitude) than freezing. The increase in latent heating by aerosols is mainly from the increase in deposition and riming at the high-levels. Overall, the buoyancy increase via latent heat release exceeds the buoyancy decrease resulting from the increase in condensate loading, leading to a positive net buoyancy (e.g., Fan et al. 2012a, 2018; Tao and Li, 2016; Lebo et al. 2012; Chen et al. 2020).

For the "warm-phase invigoration", our interpretation of the mechanism is the enhanced condensation by larger droplet nucleation in the polluted conditions releases more latent heat, enhance buoyancy this updraft intensity. This is consistent with many literature studies (e.g., Khain et al. 2012; Igel et al. 2015; Sheffield et al. 2015; Chen et al. 2017; Fan et al. 2018; Lebo 2018). The reviewer argued that condensation rates only depend on updraft velocity with the quasi-steady assumption (i.e., true supersaturation is approximated with equilibrium supersaturation), therefore they interpreted that it is the lower equilibrium supersaturation in polluted conditions that lead to a larger buoyancy, thus enhanced updraft speeds and condensation.

This quasi-steady assumption is neither physically justified for the strong updrafts of deep convective clouds nor is it suitable for studying aerosol effects on deep convective clouds which requires the exact solution of supersaturation. Previous studies (e.g., Politovich and Cooper 1988; Korolev and Mazin 2003, Pinsky et al. 2013) showed that the quasi-steady assumption is invalidated in conditions of (a) low droplet concentrations (pristine condition) and (b) intense condensation and evaporation (e.g., cloud base and strong updrafts) due to long relaxation time (larger than a few seconds). Note that both Politovich and Cooper (1988) and Korolev and Mazin (2003) evaluated the phase relaxation time under the assumption of the constant drop radius, which is not as accurate as Pinsky et al. (2013) that used the accurate equation for supersaturation. However, the reviewer mistakenly thought that Politovich and Cooper (1988) used an accurate droplet growth equation but Pinsky et al. (2013) used a simplified one.
So we followed Eq. 3 and Eq. 4 in Pinsky et al. (2013) to calculate S_{eq} and phase relaxation time τ, respectively. Fig. r4 shows the calculated phase relaxation time as a function of droplet number and radius from SBM_anth. The values we got are quite consistent with the Table 1 of Politovich and Cooper (1988) for droplet number concentrations (Nc) of 100, 300, and 500 cm-3, which proves that our calculation has no problem. The reviewer said our values "are simply wrong" in his follow-on comment which were from the same calculation except we showed the mean value for the updrafts with a velocity greater than 2 m s−1 (Fig. r5). In these relatively strong updrafts, the phase relaxation time is long (Fig. r5c) because of low Nc (Fig. r5d) due to fast conversion of droplets to rain. Fig. r4 showed that most of the updrafts have Nc of 5-20 cm-3. The averaged τ for the updrafts greater than 2 m s−1 are around 10-15 s (with large values exceed 60 s). Above the cloud base, the quasi-equilibrium supersaturation is much higher than the true supersaturation (Fig. r5a) where low droplet number concentration (Fig. r5c) and strong updrafts (Fig. r5b) are seen. Therefore, it is clear that the short phase relaxation time (a few seconds) is only true near cloud base with large droplet number concentrations (~ hundreds cm-3) and weak updrafts. However, in relatively strong updrafts, the droplet number above the cloud base is much reduced (~ tens cm-3 in this case) due to fast conversion of droplets to rain, thus the phase relaxation time is much longer (> 10 s and even over 60 s) and the assumption of $S=S_{eq}$ is not valid any more. This is particularly true for the pristine case (SBM_noanth), we can see the S_{eq} can be much higher than the true supersaturation (Fig. r2a), so assuming $S=S_{eq}$ would lead to a large bias in condensation and evaporation in the pristine case.

Therefore, appropriately simulating aerosol effects on deep convective clouds requires an exact supersaturation calculation (Eq. 6 in Pinsky et al. 2013), in which the condensation depends on droplet number and size, and more droplets in the polluted clouds increase condensation and decrease supersaturation, which clearly showed our interpretation is physically solid. In addition, as shown in our responses to the comment #6, we see the polluted case (SBM_anth) has larger condensation rates for the same updrafts than the pristine case (SBM_noanth) (Fig. r3, a vs c) above cloud base where the increase of cloud droplet number is significant (Fig. 15a). This clearly shows that higher droplet number leads to larger condensation rate for the same vertical velocity, which rebuts the reviewer' argument that condensate rates are similar under the same vertical velocity.

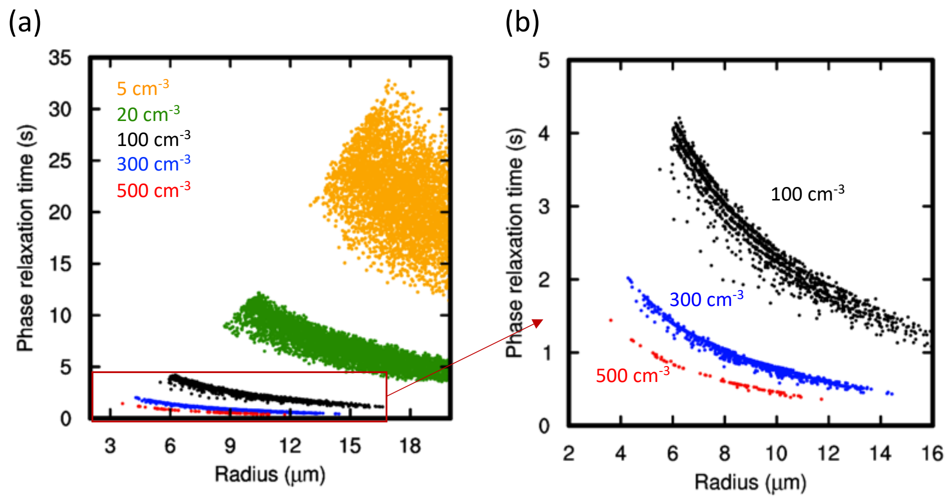

Figure r4 (a) Relationship between phase relaxation time and droplet radius for different droplet number concentrations from the simulations SBM_anth. (b) is the same as (a), except zooming in for droplet number concentrations of 100 cm^{-3}(black), 300 cm^{-3}(blue), 500 cm^{-3}(red).

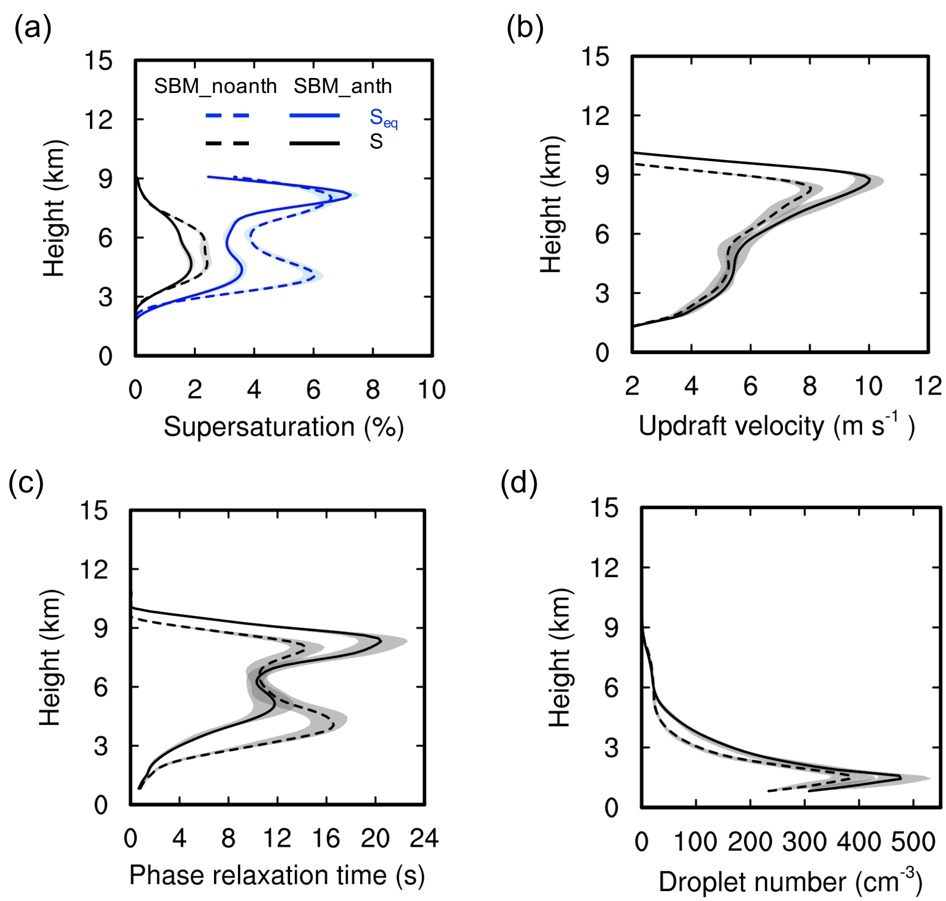

Figure r5 Vertical profiles of (a) supersaturation and quasi-equilibrium supersaturation, (b) updraft velocity, (c) phase relaxation time and (d) droplet number concentration averaged over the updrafts with value greater than 2 m s^{-1} from the simulations SBM_anth and SBM_noanth, over the analysis domain as shown in the red box in Figure 7 during the strong convection period (2000 – 2300 UTC, 19 Jun 2013). The shaded areas mark the spread of ensemble runs.