Response to Reviewer 2.

We greatly appreciate the careful review and constructive comments from the reviewer. We agree with the reviewer on most comments and tried very hard to address all the concerns in the responses and in the revised manuscript. More detailed descriptions were added about the equations used to calculate the diffusional growth. We clarified the description about the impact of seeding on the evolution of the water drops formed on background aerosol particles. The meaning of Ostwald ripening effect used in this study was also explained.

In this document, the original comments are in blue font and our responses are in italic type and black, furthermore the suggested modifications in the manuscript are in normal type and black.

In this manuscript, the authors used a cloud parcel model to investigate the characteristics of cloud droplet spectral evolution by condensation and collision and coalescence for various background CCN distributions and different updraft conditions. Then the impact of hygroscopic seeding material on cloud droplet spectral broadness was examined. The model they used seemed very appropriate for calculating cloud droplet growth processes in an adiabatic cloud parcel, limiting numerical diffusion by adopting moving bin boundaries for calculating condensation processes. The limitation was that this model did not take into account the entrainment and mixing processes, which certainly affect cloud droplet growth processes and droplet distributions in real clouds. However, for examining cloud droplet spectral broadening at earlier stages of cloud development, such limitation may be tolerable. The described impact of hygroscopic seeding material seems somewhat expected. Certainly seeding effect would be pronounced when seeding particles are big and such effect would be diminished when background CCN include many big particles. The scientific contribution of this manuscript mainly comes from the development of hybrid bin scheme that can be used for calculating condensational growth process without numerical diffusion. I think that this manuscript deserves publication in ACP after minor revision, addressing the comments I made below.

Major comments:

It is good to see that the Ostwald-ripening (OR) effect on droplet spectral broadening can also be significant under non-oscillating vertical velocity conditions. In a strict sense, however, what was presented in this manuscript was not exactly the same as the OR effect described in Yang et al. (2018), where the spectral broadening occurred since larger droplets grew but smaller droplets shrank. Such phenomenon can occur easily under oscillating vertical velocity condition: during updraft all droplets can grow but during downdraft larger droplets can still grow but smaller droplets may evaporate as they may become deactivated. In this manuscript, vertical velocity was always positive (updraft), although the value itself varied. So all activated droplets grew throughout the ascent regardless of their sizes but the important point was that the radius growth rate of larger droplets could be higher than that of smaller droplets near cloud base altitudes especially under low updraft conditions, resulting in broadening of the cloud droplet distribution. Such spectral broadening can also be called the OR effect but the subtle difference from Yang et al. (2018) should be noted.

A sentence is added to explain the difference between our interpretation of the OR and that of Yang et al. (2018) at line 198 in the original version of manuscript and at line 216 in the revised manuscript:

"In this study, the Ostwald-ripening effect is subtly different from Yang et al. (2018). In Yang et al. (2018), the spectral broadening is a result of the shrinking of the smaller drops and growth of larger drops in an oscillating vertical velocity condition. In our study, the vertical velocity is always positive and all droplets larger than the critical size can growth throughout the ascent, but the larger drops grow faster than the smaller drops, resulting in broadening of the size distribution."

In fact, the characteristics of spectral broadness of droplets that are grown by condensation under different CCN and updraft conditions were extensively examined by Yum and Hudson (Atmospheric Research, 2005), which clearly explained with cloud parcel model calculation and theoretical assessment that it was the differences between the ambient (cloud) supersaturation and the equilibrium supersaturations of different size droplets that determine spectral broadness of condensationally grown droplets: at lower ambient supersaturation, the differences between the ambient supersaturation and the equilibrium supersaturations of different size droplets are relatively larger than those at higher ambient supersaturation, and therefore broader spectra. Yum and Hudson (2005) should be cited when discussing the dependence of spectral broadening on supersaturation.

Thank you for drawing our attention to this paper. It is fully relevant.

We cite the paper in section 4.1 (line 218) and another paper published by these authors is also cited in section 5 (line 335).

We insert the following sentences at line 218 in the original version of manuscript and at line 246 in the revised manuscript:

"Similar conclusions were reported by Yum and Hudson (2005). They numerically simulated the diffusional growth of water drops formed on aerosol particles with homogenous chemical composition (salt), and they also found that the curvature and solution terms could significantly impact the broadening of the DSD near the cloud base if the updraft velocity was small. "

and the following text is inserted at line 335 in the original version of manuscript and at line 368 in the revised manuscript:

"If the ratio of the Q_{seed}/Q_{ctrl} is larger than 1 the LWC is superadiabatic, because in the control cases, the LWC is close to the adiabatic value at 100 m above the cloud base. The uptake of vapor by highly hygroscopic particles under the cloud base, especially in the case with a weak updraft, increases the LWC to above the adiabatic value. The amount of the surplus is small, so the effect is significant only when the adiabatic LWC is small (e.g., near the cloud base) and becomes negligible at a higher level above the cloud base. We hypothesize that the efficient vapor uptake by the coarse-mode hygroscopic particles can partly explain the observed superadiabatic LWC near the cloud base with weak updrafts in some field campaigns (Blyth and Latham, 1985, Yum and Hudson, 2001)."

The description of Eq (1) is a little confusing. The indices i and k appear together for m and D. Does it mean that there exist multiple k values for each droplet size bin boundary, i? According to Table 1, a specific kappa value is associated with a specific mode of aerosol particles. So I guess that a specific k value is associated only with a certain range of i values.

This should be clearly stated.

The mass variables for the aerosol particles are given by one dimensional array, that means we use the same bin intervals for the different types of the aerosol particles. The size variables for the mass of drops and the diameter of water drops are described by two-dimensional arrays in this model. The first dimension is the size, and second dimension means the different (distinct) types of the aerosol particles the drops formed upon. So, we can simulate the diffusional growth of water drops separately even if the aerosol particles inside the drop have the same size but different hygroscopicities. In this study, the hygroscopicity is given in non-overlapping, separate size categories of the aerosol particles. This is the reason that we deleted the k index associated with aerosol mass in Eq. 1. However, even in this case, we need two-dimensional variables for the drops. Although at the start of the simulation there is no size overlapping among the aerosol particles at different hygroscopicities, size overlapping can occur at a later time as the drops formed on aerosol particles with smaller size but larger hygroscopicity can grow faster than the drops formed on aerosol particles with larger size and smaller hygroscopicity (e.g. in BGQNC_1 case).

Texts at line 84 is modified in the original version of the manuscript and at same line in the revised manuscript:

"where $m_{d,i,k}$ and $D_{d,i,k}$ are two-dimensional variables of the mass and the diameter of the water drop, respectively. The index i represents the bin boundary for the mass or size of the particles. The index k represents the type of the aerosol particles on which drops formed. The size distributions of the different types of aerosol particles are given with the same mass bin intervals, the $m_{ap,i}$ is the mass of the aerosol particles at the i^{th} bin boundary."

The sentence at the line 95 in the original version of manuscript and at line 102 in the revised manuscript is modified:

"At 100 m above the cloud base, the size of drops is large enough to neglect both the curvature and the solution terms, so the aerosol mass inside of the drops is not tracked, and Eq. 1 can be simplified to Eq. 2:"

Line 173: Are the temperature and vertical velocity profiles different for different aerosol conditions or are they given as initial conditions? Temperature in the cloud parcel may become slightly different for different initial aerosol conditions since latent heat release can be slightly different. But the vertical velocity profile should have been prescribed. This sentence can misleadingly indicate that vertical velocity profile can be affected by the given initial aerosol distribution. This may be so but I doubt that the model took that into account.

We agree that we need to clarify the description of Fig. 1. The velocity profiles are prescribed, and they are not impacted by the latent heat release of condensation. However, the temperature profiles are calculated. We mentioned the types of the seeding materials in the text and the types of the background aerosol particles in the figure caption relevant for Figs. 1a and 1b. To better clarify this point, we add more information on the initial aerosol conditions in the caption of Fig. 1.

We modify the figure caption:

"Figure 1: The simulated temperature profiles and the prescribed updraft profiles in the numerical simulation for (a) the SPEC UAE cases and (b) the QCSRP cases. The background aerosol particles are BGUAE_1 and BGQNC_1, seeding materials are ICE70_2 and ICE70_4 in the figure (a) and (b), respectively. The plotted temperature profiles are calculated at the updraft profile of w_3 for each panel. The horizontal blue lines denote the altitude of the cloud base. The three horizontal black lines from bottom to top denote the level where the calculation of the diffusional growth starts (RH = 70%), the levels 100 m and 1000 m above the cloud base."

Line 211: What the model calculates is the adiabatic LWC in the sense that the model does not allow heat exchange and mixing of the outside air. However, this adiabatic LWC can be different for different updraft conditions because different supersaturation (indicating the amount of excess vapor remaining without being condensed) can be generated for different updraft conditions, as demonstrated in this manuscript. What the authors indicate in this sentence is the maximum adiabatic LWC that can be obtained in the pseudo-adiabatic process which assumes that all excess water vapor is condensed and just saturation is maintained during the ascent. Make it clear.

We agree that we did not compose the sentence correctly. The liquid water content cannot be exactly equal to the adiabatic LWC, because the supersaturation is never equal to zero in these simulated cases even in the case of weak updraft. We modified the text to clarify this at line 210 in the original version of manuscript and at line 235 in the revised manuscript:

"In the case of weak updrafts, the LWC approaches the adiabatic LWC immediately above the cloud base. (The LWC profile simulated with the weakest vertical velocity is closest to the adiabatic LWC profile. This difference is not discernible in Fig. 4b.) In the strong updraft (updraft profile of w_1) the vapor surplus due to the updraft exceeds the vapor depletion due to the condensation. Therefore, LWC approaches the adiabatic value only at a higher elevation, at about 100 m above the cloud base. Adiabatic LWC is defined as equal to zero before the parcel reaches saturation, and the supersaturation is zero above the cloud base."

Line 261: It is stated that seeding has no significant effect on the growth rate of the drops formed on background aerosol particles. I would guess that adding seeding material would increase total droplet concentration and decrease the supersaturation, leading to broader spectra even only for the droplets formed on background aerosols. What were the change or difference of total droplet concentration and supersaturation caused by seeding?

The total amount of the condensed water is hardly changed by seeding, comparing to the control cases. Seeding particles grow by depleting water vapor, and at the same time, suppressing the growth of the background small droplets (Ostwald-ripening effect). When nanoparticles are used as seeding material, the maximum supersaturation was not impacted by seeding. When the flare particles are used for seeding (e.g., ICE70), the supersaturation is reduced. This reduction is consistent with the significant increase of the CCN due to seeding. However, even in this case, the total amount of condensed water vapor was the same in the control and seeded cases. Seeding results in a significant decrease of the water vapor uptake by the background aerosol particles (see line 308 in the manuscript). Because the average size of the flare particles is nearly the same as that of the background fine particles, they do not have the advantage of competing for more water vapor, so the size distribution remains narrow. To demonstrate this effect, the maximum supersaturation and the mass of the condensed water vapor in the control and seeded cases are added for each plotted case in Fig. 6. The impact of the seeding on the concentration of the activated background aerosol *particles in the case of the nanoparticles is not clearly described, so one sentence clarifying this is added to the text. Although the impact of the flare particles on the activation of background aerosols is mentioned in the original version of the manuscript (see lines 379 – 383 in the current version of the manuscript), sentences are inserted to make this effect clearer.*

The following sentences are inserted in the manuscript at line 263 in the original version of manuscript and at line 293 in the revised manuscript:

"Seeding by nanoparticles, due to their small concentration, hardly impacts the supersaturation above the cloud base and the amount of water vapor uptake by the background aerosol particles (see $s_{max,ctrl}$ and $s_{max,seed}$ as well as $Q_{bg,ctrl}$ and $Q_{bg,seed}$ in Fig. 6). In line with the small impact of the seeding on the supersaturation and the LWC the results of the numerical simulation show that the number concentration of the drops formed on background CCN does not change due to the seeding."

and at line 308 in the original version of manuscript and at line 343 in the revised manuscript:

"The doubling of the aerosol concentration due to seeding reduces the maximum supersaturation by 20% and halves the amount of the condensed water on the background aerosol particles (see $s_{max,ctrl}$ and $s_{max,seed}$ as well as $Q_{bg,ctrl}$ and $Q_{bg,seed}$ in Fig. 6e). The flare particles suppress the growth of the drops formed on the background aerosols, they mainly suppress the growth of drops formed on the fine-mode background aerosol particles and only slightly suppress the growth of the drops formed on the coarse-mode aerosols. However, their growth rate does not exceed significantly the growth rate of the background aerosol particles having the same size."

Caption for Fig. 6 is also modified:

"Figure 6: The vertical profiles of the growth rate of drops formed on background aerosol particles with different sizes and hygroscopicities (first column) and that formed on different types of seeding materials with different sizes and hygroscopicities (second column). In the left panels, the solid lines denote control cases, and the dashed lines denote seeded cases. Blue and black vertical lines denote the location of the cloud base and that of the maximum supersaturation. The $s_{max,ctrl}$ and $s_{max,seed}$ mean the calculated maximum supersaturation belong to the unseeded and seeded cases, respectively. The $Q_{bg,ctrl}$ and $Q_{bg,seed}$ mean the amount of the condensed water on the background aerosol particles in the control and seeded cases at 100 m above the cloud base, respectively. "

Line 339: Background CCN concentration does not decrease. The number of activated cloud drops from background CCN may decrease. Rewrite the sentence.
In all size distribution plots, y-axis label is written as N/dlog(r), not dN/dlog(r). Are you sure? Then what does N mean here?

We assumed that the potential cloud condensation nuclei are called condensation nuclei (CN), and the activated condensation nuclei (subset of CN) are called CCN. However, we realize that this distinction is not generally applied, and we modify the sentence in line 339 in the original version of manuscript and at line 383 in the revised manuscript:

”A notable decrease in the number concentration of activated drops from background CCN was only found in the case of a weak updraft (w = 1.0 m s^{-1}).”

Sorry that the label text is wrong both in Figs. 4 and 5 (and Figs. 2 and 7 too). The correct label is dN/dlog(r). We modify the labels in these figures.

Figure 3: What do closed circles mean? No explanation is given in caption or in the text.

The solid circles represent all of the six other background cases (BGUEA_1, BGUAE_3, BGUAE_4, BGUAE_5, BGUAE_6 and BGQNC_2 with three, corresponding updraft profiles). Because we changed to the logarithm scale in the case of 'x' axis the dots belong to the case BGUAE_4 cannot be plotted ($H_c = 0$ in this case). A sentence is added to the figure caption and one sentence is inserted in the text to clarify this.

The figure caption is modified:
"Figure 3: Reflectivity calculated from the modelled DSD (a) 100 m and (b) 1000 m above the cloud base for non-seeded control cases. Symbols with different colours represent different updraft velocities at the cloud base as indicated by the key. Three different background aerosol size distributions are presented by solid triangles, squares, and open circles, respectively (see the legend in panel a.). The solid circles represent all of the six other background conditions (BGUEA_1, BGUAE_3, BGUAE_5, BGUAE_6 and BGQNC_2 with three corresponding updraft profiles, except BGUAE_4 case where H_c is zero)."

The following sentence is inserted at the line 193 in the original version of manuscript and at line 210 in the revised manuscript:
"(all the other background cases are not distinguished from each other by using different symbols, and they are denoted by closed circle)"

Figure 4: Integration of droplet size distribution would produce total droplet concentration. If I do that for the two droplet size distribution for two different updraft shown in Fig. 4a, I would find that the total droplet concentration is higher for the lower updraft. The y-axis is in log scale. So the actual difference of the concentrations might not be as dramatic as shown in the plot but it should still be true that the concentration is higher for the lower updraft. I do not understand this.

Unfortunately, the resolution of the plots is not good enough to distinguish the two curves at the peak.

This table shows the values of the functions belong to the two updraft profiles near the peak of the curves.

radius (μm)	*dN/dlog(r) (m^{-3}) at w_3*	*dN/dlog(r) (m^{-3}) at w_1*
2.48	*8.32×10^8*	*1.45×10^8*
3.13	*7.41×10^9*	*9.73×10^9*
3.94	*8.78×10^8*	*2.40×10^8*

4.96	*5.29×10⁷*	*2.37×10⁷*

The data in the table shows that dN/dlog(r) at w_1 (strong updraft) is larger than that at w_3 only at the radius of 3.13 µm. This positive deviation at the peak is compensated by the negative deviation in other radii. We tried to zoom in Fig. 4a by starting the vertical scale at 10^4. It helps a little bit.

Figure 8: E is not clearly defined in the caption. Make it clear.

We modified the figure caption:

"Figure 8: (a) The impact of the change of the H_c parameter on the vapor uptake. The vertical coordinate represents the ratio of liquid water contents belong to the seeded and associated control cases. (b) This panel shows how the broader size distribution generated by the Ostwald-ripening effect close to the cloud base (E_{cb+100}) can result in broadening of the size distribution comparing to the control case 1000 m above the cloud base ($E_{cb+1000}$). E_{cb+100} and $E_{cb+1000}$ mean the calculated change of reflectivity due to seeding (Eq. 4) at 100 and 1000 m above the cloud base, respectively."

Table 4: What is NCM? No explanation in caption or in the text.

To our knowledge there is no publication about the NCM seeding materials, sentence at lines 171-172 in the original version of manuscript and at lines 187 – 189 in the revised manuscript is modified to add more information about the NCM particles:

"The NCM seeding material (Table 4 and Fig. 2c) is released from hygroscopic flare recently developed by the National Center of Meteorology at UAE. Comparing to the ICE70 particles, the size distributions of the NCM particles are narrower, and it consists of only fine particles."

Minor comments:
L178: rewrite w1.
The text is modified.

L212: vapor flux → vapor surplus
The text is modified:

L238: Move "(Wehbe et al., 2021)" to the end of the previous sentence.
The text is modified.

L251: remove 'of' in front of *bg*.
The text is modified.

The following references are added to the manuscript:

Blyth, A.M., Latham, J., 1985. An airborne study of vertical structure and microphysical variability within a small cumulus. Q. J. R. Meteorol. Soc. 111, 773–792.

Yum, S.S., Hudson, J.G., 2001. Cloud microphysical relationships in warm clouds. Atmos. Res. 57, 81– 104.

Yum, S.S., Hudson, J.G., 2005: Adiabatic predictions and observations of cloud droplet spectral broadness. Atmos. Res. 73, 203– 223.

Bermeo, M., Hadri, N. E., Ravaux, F., Zaki, A., Zou, L., Jouiad M., 2020: Adsorption Capacities of Hygroscopic Materials Based on NaCl-TiO 2 and NaCl-SiO 2 Core/Shell Particles. Journal of Nanotechnology **2020,** *2020* , 1-16. https://doi.org/10.1155/2020/3683629

Lompar, M., Ćurić, M., Romanic, D., Zou, L., Liang, H., 2018: Precipitation enhancement by cloud seeding using the shell structured TiO2/NaCl aerosol as revealed by new model for cloud seeding experiments. Atmos. Res. 212, 202-212