Responses to comments of the Referee #1.

Below we respond to the reviewer comments. The original comments are in the black color, our responses blue.

My understanding of the "scaled-DNS" presented in this manuscript is that one can simulate a large domain size by artificially increasing the kinetic viscosity of the airflow. The Reynolds number is kept unchanged in such simulations so that the computational cost is still feasible. Therefore, one can study how the supersaturation fluctuations can be affected by the large eddies. This is plausible as the small scales do not matter for the supersaturation fluctuations. The authors further tested the application of the superdroplet approach in such a setup to tackle the condensation process. I would recommend the publication of this manuscript after the authors carefully discuss the following comments.

The reviewer's summary of our methodology is correct.

The author addressed both in the abstract and in the conclusion that this is a "novel methodology". This statement should be treated carefully for the following reasons: 1. The method presented in this manuscript is DNS with large artificial kinetic viscosity, which is not a new method. 2. Mellado et al also used the same treatment (section 3 of Mellado et al).

We do not agree with this comment. Yes, Mellado et al. applied large domain in their simulations of the stratocumulus top. However, nowhere in Mellado et al. we find the formula showing how the viscosity needs to be increased to allow appropriate dissipation when the model grid length is far away from the dissipation length. This is not that important for the finite-difference model as long as proper dissipation is accomplished by the advection scheme as in the so-called implicit large eddy simulation. But the rescaling is critical for the spectral model that by design has no numerical dissipation. Our methodology targets traditional homogeneous isotropic turbulence simulation applying DNS that in its original form cannot consider droplet growth in large domains. We believe the methodology we propose and use is novel. That said, we added a reference to Mellado et al. and a few other papers (per comments by Rev.1 and 2) in the introduction.

Specific comments:

1. The paper by Mellado et al applied the same idea. Can the authors compare their work with the one of Mellado et al?

Mellado et al. applied similar technique to the cloud top simulation applying bulk condensation scheme. We are not sure what is there to compare.

2. Can the authors check Eq.3 again? If dR/dt=KrS/R, then Rˆ2/2=KrS, together with Eq.2, you will get a pre-factor of 2 instead of 4/3 in Eq.3, right?

No. Eq. 3 does not come from Eq. 2. Eq. 2 is the condensation rate applying the analytic droplet growth rate equation that is shown in the text below Eq. 2. (We corrected an error in Eq. 2 pointed out by Rev. 2). Eq. 3 is the condensation rate derived from the change of the droplet volume as predicted applying a finite time step. Eq. 3 ensures that the mass of water (water vapor plus droplets) is conserved because condensation rate as given by Eq. (3) is applied in the water vapor equation.

3. L.125: Should the intensity of turbulence determined by the single parameter, Reynolds number? The energy dissipate rate is a small-scale quantity which describes how fast energy dissipates in the dissipation range of turbulence. In other words, it characterizes how vigorous the small eddies of turbulence are. It is calculated from the trace of the strain tensor. This aspect is also discussed by the authors in the paragraph just below Eq.10. Did you mean the energy transfer rate here, which is the rate energy transfers from large to small eddies in 3-D turbulence?

Reynolds number describes the range of scales between the scale of energy containing eddies (about the size of the domain in DNS) and the dissipation scale, see Eq. 5 in the paper. The intensity of turbulence is determined by the eddy dissipation rate. Yes, the eddy dissipation rate is a small-scale quantity, but this is how intensity of turbulence is expressed in models and in observations. TKE depends on the eddy dissipation rate and on the characteristic eddy scale, Eq. 4 in the paper. This is why TKE increases with the domain size as shown in Fig. 2 (upper row), and the TKE dissipation rate is the same (lower row).

Perhaps a discussion in Grabowski and Abade (JAS 2017, p. 1485) can also help.

4. Can the authors normalize the energy spectrum in the same way as the one of Fig.1 of Li et al. (2019)? If the Reynolds number is the same, the normalized spectrum should collapse on top of each other.

This can be done. However, we feel the figure in its current form is more informative for a cloud physics reader that we target in our manuscript. The figure clearly shows the shift of spectrum with the increase of L. Also, the figure in the current form illustrates the TKE increases with the domain size for the same TKE dissipation rate. We prefer to leave the figure in its current format.

5. Why is there an initial spike in Fig.6? I don't understand why it is different for different domain sizes. The water mass loading is the same for all the simulations, right?

First, please note that the spike amplitude is very small compared to the standard deviation shown in Fig. 7. We mention that in the revised manuscript. Second, initial conditions are not in the equilibrium with droplets that are present locally, and establishing quasi-equilibrium takes some time. However, one may argue that this should not take as long as the figure shows. Another aspect is that "molecular diffusion" (in quotes as this is the scaled-up molecular diffusion), in addition to the local vertical velocity, impacts droplet growth. In fact, the duration of the spike (longer for larger domains) suggests that the diffusion is likely the key factor. This is where the absence of unresolved scales in the scaled-up DNS may play some role. Because those

transient conditions are just an artifact of the modeling setup, we do not think exploring this any further is warranted.

6. As the integral time scale is different for simulations with different domain size, could it be an idea that the authors normalize the time axis by the integral time scale?

We included two horizontal axes on bottom panels of figure 2 per reviewer request.

Technical corrections:

1. Fig.4, caption: standard deviation of supersaturation fluctuations?

2. L248: When “the” multiplicity. . .

Corrected.

Responses to comments of the Referee #2.

Below we respond to the reviewer comments. The original comments are in the black color and our responses are in blue color.

Overall, we feel the reviewer missed the key aspect of the paper: the largest turbulent eddies have the most significant impact on the diffusional droplet growth. Our desire to simulate the large-eddy end of the spectrum, and not scales close to the molecular dissipation, is the main motivation for the scaled-up DNS approach. The motivation has been discussed in the two paragraphs starting at line 64 of the introduction that sets the stage for the entire manuscript.

This study uses a modified DNS to investigate the effect of turbulence on the diffusional growth of cloud droplets in much larger domains (up to 64*64*64 mˆ3) compared with traditional DNS. The method is the combination of increasing air viscosity to allow DNS in a larger domain and using Lagrangian particle-based microphysics to lower the number of particles needed to be tracked in a larger domain. Results show that the variance of droplet radius increases with the square root of time, which is consistent with previous studies and the result from a stochastic model. Different domain sizes and multiplicities are also tested to check the convergence of the scaled-up DNS. The paper is interesting. However, I have some comments needed to be addressed before this manuscript can be accepted.

This is approximately correct summary of the paper. A small clarification: results from the stochastic model show that the supersaturation fluctuations in the domain are consistent with scaled-up DNS. The figure does not show the droplet radius variance.

Major comments:

1. Running DNS in a larger domain is not new, for example, Rotunno and Bryan (2018) recently applied direct numerical simulation to study lee vortices in a larger domain by setting viscosity as 1 m2/s. Although the combination of this technique and Lagrangian cloud model is interesting, it is not clear to me whether it is suitable to do that, or at least what we can learn from it, for the following reasons:

The lack of novelty comment echoes comments from the Reviewer 1. In general, running a fluid flow model with constant mixing coefficients, such as in Rotunno and Bryan (2018), goes long way back. See, for instance, Grabowski and Clark (JAS 1993, p. 555) for an earlier example. Another example is the so-called implicit large eddy simulation (ILES) approach where there is no explicit mixing and the computational stability is ensured by the numerical algorithm (i.e., the monotone finite-difference advection scheme). Such an approach is being used by many (see, for instance references at the beginning of section 3 in Grabowski JAS 2014, p. 4493). However, ILES would not work for a spectral model because proper dissipation at small scales is missing. In our study, we utilize a spectral DNS code with scaled-up viscosity and diffusivity, and use the code in simulations for domains that are impossible to consider with the traditional DNS. The basis of such an increased dissipation and its application for large domains is the key novelty of our study.

Because of this comment and a comment from the Reviewer 1, we added a couple sentences in the introduction of the revised manuscript.

1.1 When air viscosity is changed, it is unclear to me whether thermal diffusivity is also changed accordingly in this study. If not, the Prandtl number will be different compared with air. It means that this is a fluid that does not behave as air. If yes, this will be consistent with R&B (2018).

Thermal diffusivity is also changed in the same way as the viscosity. This is clearly stated in the abstract and we added a comment on that in section 3 below Eq. (8). The R&B study is irrelevant in this context, see above.

1.2 Changing air viscosity and thermal diffusivity will also slow down the condensational growth of cloud droplets following physical rules. But I guess this effect is ignored in this study. Therefore, the scaled-up DNS in this study simulates a turbulent cloud system that is not similar dimensionless to natural clouds. Therefore, it is unclear to me what we can learn from it. I hope the authors can comment on this.

This comment is incorrect. The molecular coefficients applied in the droplet growth equation are not scaled up and the typical droplet growth equation is used, see below Eq. (2) in the manuscript. The key for the condensational growth are the supersaturation fluctuations. Their magnitude increases with the turbulent eddy size and we aim at producing fluctuations typical for large eddies. The supersaturation fluctuations obtained from scaled-up DNS are consistent with the stochastic model as shown in figure 10. In short, the comment that we simulate a "cloud system that is not similar dimensionless to natural clouds" is simply incorrect.

2. For the same energy dissipation rate, if air viscosity increases, Kolmogorov's length scale increases and therefore a coarser resolution can be used. However, Komogrov's velocity scale also increases at the same time. Larger velocity fluctuation in a larger domain leads to larger supersaturation fluctuation (Figure 7), and thus faster broadening of droplet size distribution (Figure 9). The reason that the variances of droplet size distribution from DNS/scaled-up DNS are consistent with the stochastic model (Figure 10) is that they generate/use the same strength (e.g., PDF) of the velocity field. Therefore, their agreement and the consistency with the tˆ(1/2) scaling law is not a big surprise to me.

This comment provides an approximately correct summary of our results. However, the reviewer confuses increase of the Kolmogorov scales with the increase of the supersaturation fluctuations. Please see our response to the next comment.

The fact that the results are as expected supports validity of the proposed approach. Yes, it is expected that larger eddies feature larger and longer-lasting supersaturation fluctuations and thus have more impact on the spectral broadening. However, specific details (such as the spectral width increase with the domain size when following t^1/2 scaling, fig. 9) can only be obtained with the scaled-up DNS.

An important comment is that the approach developed in this paper is being used in ongoing studies with more realistic systems, such as a rising parcel with CCN activation. To be

relevant to a turbulent cloud, domains larger than a mere few meters are needed. This aspect is mentioned in the final paragraph of the paper.

However, in an idealized homogeneous isotropic turbulent cloud, the velocity (supersaturation) fluctuation should be independent of the volume we choose, meaning that in either a volume of 1 mˆ3 or 10 mˆ3, the energy dissipation rate, Komogrov's length scale and velocity scale should be the same. Therefore, it is unclear to me how the simulated cloud in a domain size associated with the increase of Komogrov's velocity scale, is related to the conceptual cloud with the same large domain with the original (smaller) Komogrov's velocity scale. Please comment on it.

The initial part of the comment is incorrect. The velocity and supersaturation fluctuations **increase** with the domain size when the eddy dissipation (and thus Kolmogorov scales) remain unchanged. This has been shown in past DNS studies that we refer to and is the key of the argument; see also Grabowski and Abade (2017). The largest supersaturation fluctuations come from the largest eddies, and these are related to the turbulent kinetic energy (TKE). This is because TKE represents energy of the largest eddies of a turbulent flow. TKE increases with the domain size and the correct scaling is given by Eq. 4 in the paper (line 129).

3. Figure 4 shows that results converge for different multiplications. As stated around line 257 "Note that for real DNS (Table 2 and Fig. 4), having a droplet in one of several dozens of grid volumes still results in supersaturation fluctuations in agreement with real droplets." However super droplet even with the multiplicity equals to 1 (real droplets) is one out of eight grid boxes, meaning that the density of droplet is low in the domain. Is it possible the evolution of the supersaturation fluctuation shown in Figure 4 is just the background even without droplet? I think a more careful test is to track more particles in real DNS, at least 10 per grid box, and then change the multiplicity but maintain the same number of particles in the domain.

The initial conditions for the simulations are as in Lanotte et al. DNS study, droplet concentration of 130 per cc and droplet radius of 13 microns (see line 119 of our ACPD paper). Indeed, clouds are fairly diluted systems. However, it is not true that the fluctuations for the case with droplets are the same as without droplets. The supersaturation variance in simulations without droplets for the two domain sizes shown in Fig. 7 are about 8×10^{-4} for the left panel and about 1.6×10^{-3} for the right panel, that is, larger than the mean of those shown in the figure. We do not think we need to discuss this in the paper.

The reviewer's suggestion at the end of the comment does not make sense. Droplet concentration (130 per cc) implies that it is impossible to have 10 droplets per grid box if the grid length is 1 mm. There are on average 0.13 droplets per grid box, or one droplet in about 8 grid boxes as the reviewer notices.

Minor comments:
1. Line 17: "mean droplet radius variance", should it be "droplet radius variance"?

2. Line 50: “however, see the comment on that paper by. . .” This sentence is not clear to me.
3. Line 101: “K is the molecular diffusion coefficient” should be “thermal diffusion coefficient”
4. Equation 2: I think \delta t should be removed to make sure the unit is correct. Please check.

These comments have been addressed in the revision.

Responses to comments of the Referee #3.

Below we respond to the reviewer's comments. The original comments are in black color and our responses are in blue. We do not agree with many suggestions as they seem to put our manuscript in a context that we feel is not appropriate for this work. Please see specific responses below.

This study applies the scaled-up DNS method to simulate supersaturation fluctuations and spectral broadening in an idealized framework of forced, isotropic turbulence. The supersaturation fluctuations are produced by the turbulent vertical motions. Scaled-up DNS is what I would consider to be the simplest possible form of large-eddy simulation. In scaled-up DNS, the molecular diffusivities are increased to maintain the same kinetic energy dissipation rate as an otherwise identical simulation with a smaller grid size. In the present application, the range of scales between the domain size and grid size was maintained. Droplet condensational growth was represented using the superdroplet method. The impact of the droplet multiplicity of the super-droplet method on the simulations was studied. The results (i.e., supersaturation standard deviations) of the scaled-up DNS were compared with those from a stochastic model, and exhibited very good agreement.

Specific Comments

1. lines 36-45: What is described here is not the only mechanism by which supersaturation fluctuations can be produced within a cloud, and probably not the most important. Entrainment and mixing also produce supersaturation fluctuations and are well-known to be an important source of spectral broadening, and should be mentioned here in order to place the focus of this study in proper perspective. As will be mentioned later, the set-up of the simulations actually implies an external forcing, which could be interpreted as a crude representation of entrainment.

Although we agree in general with the reviewer's comment, we do not think bringing detailed discussion of entrainment and mixing is needed. The paper discusses a very specific aspect of the homogeneous isotropic turbulence impact that was studied in the past applying DNS. Our approach extends those studies and targets DNS community. That said, we modified the introduction and brought several references to entrainment and mixing. We do not understand the last sentence – the simulations are forced in a way typical to traditional DNS.

2. lines 46-47: It would be appropriate to refer here to the even earlier study by Su et al. (1998) in which diffusional growth of cloud droplets in a turbulent environment was simulated using the Explicit Mixing Parcel Model, which is essentially a 1D kinematic DNS. lines 54-56: Droplet sedimentation was included in the EMPM simulations mentioned.
Su, C.-W., S. K. Krueger, P. A. McMurtry, and P. H. Austin, 1998: Linear eddy modeling of droplet spectral evolution during entrainment and mixing in cumulus clouds. Atmos. Res., 47–48, 41–58.

This part of the text has changed. We added several references to entrainment and mixing, including the one suggested. We do not want to single out EMPM as it is only marginally relevant to our study. Please see our response to 1 above.

3. line 57: Unclear. What has a “small impact on the droplet spectra"?

The supersaturation fluctuations. Text modified.

4. lines 56–64: State what mean supersaturation was used in these studies.

Vaillancourt et al. applied a rising adiabatic parcel setup. The mean supersaturation was never presented. Lanotte et al. and Li et al. applied no mean vertical motion (as in our study) and started with zero mean supersaturation. This is now mentioned in the modified text.

5. line 65: It is possible to do simulations with larger domains with the EMPM. It would appropriate to mention here that the EMPM simulations reported in Su et al. (1998) used a 20-m domain size, and EMPM domains up to 100-m domains were used in Tölle and Krueger (2014). Tölle, M. H., and S. K. Krueger, 2014: Effects of entrainment and mixing on the droplet size distributions in warm cumulus clouds. J. Adv. Model. Earth Syst., 6, 281–299, doi:10.1002/2012MS000209

As stated in our response to 1 and 2, we do not want to bring entrainment/mixing in this manuscript except in a brief comment in the final paragraph in the conclusion section.

6. lines 67–69: Be clear about the source of the supersaturation fluctuations that you are referring to, which vertical gradients of potential temperature, not entrainment and mixing. Noting this also makes the explanation of why large eddies produce larger fluctuations more obvious.

There are no mean temperature gradients in the traditional DNS and scaled-up DNS. In the framework we use, larger supersaturations come only from larger and longer-lasting fluctuations of the vertical velocity. We modified the discussion in this paragraph.

7. lines 69–72: It would also be appropriate to mention the EMPM approach here, because it certainly does perform “DNS-like simulations in computational domains comparable to the size of the LES grid box." Furthermore, the recent development of a linear-eddy based SGS model that is combined with the super droplet method by F. Hoffmann should be mentioned. Hoffmann, F. and G. Feingold, 2019: Entrainment and Mixing in Stratocumulus: Effects of a New Explicit Subgrid-Scale Scheme for Large-Eddy Simulations with Particle-Based Microphysics. J. Atmos. Sci., 76, 1955-1973, https://doi.org/10.1175/JAS-D-18-0318.1 Hoffmann, F., T. Yamaguchi, and G. Feingold, 2019: Inhomogeneous Mixing in Lagrangian Cloud Models: Effects on the Production of Precipitation Embryos. J. Atmos. Sci., 76, 113-133, https://doi.org/10.1175/JAS-D-18-0087.1

We do not agree that this aspect needs to be included in the manuscript. We see similarities between DNS and EMPM, but this is only tangentially related to the main thrust of our manuscript.

8. line 100, Eq. (1): In general, this equation should include a term $w\, dT/dz$. I suspect that this term is missing because $\overline{dT/dz} = 0$ is enforced due to the cyclic b.c. at the top and bottom boundaries. It this is the case, it should be mentioned. It should also be mentioned that forcing dT

/dz = 0 is equivalent to forcing a nonzero gradient of potential temperature, which acts as the source of temperature and supersaturation fluctuations.

The reviewer is correct. DNS by design cannot feature mean temperature gradients because of the triply-periodic boundary conditions. This is why Eq. (1) does not have the w dT/dz term. Eq. (1) is standard for the DNS of homogeneous isotropic turbulence (e.g., see Eq. 9 in Vaillancourt et al. JAS 2001). We prefer not to bring this aspect in the model description.

9. line 111. Eq. (3): State how changes in R3 are calculated if not by using (2).

We added a brief comment on that. The key is that droplet growth is calculated first, and then condensation rate follows from (3).

10. line 118: Please state the initial conditions, particular the initial temperature profile, as well as the initial supersaturation profile.

There are no profiles in the spectral DNS. The initial conditions are specified in the last paragraph of section 2 of the original submission. We added information about the assumed temperature and supersaturation.

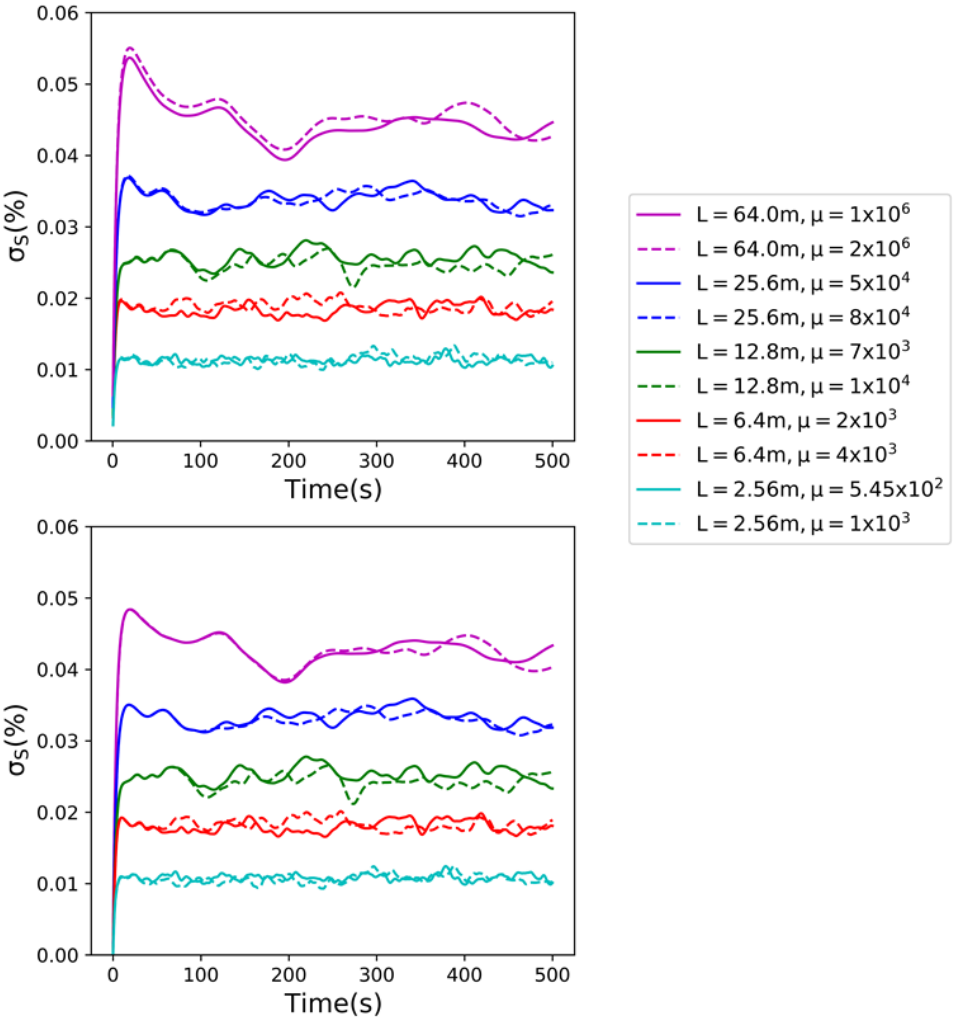
11. Figure 2: It would be enlightening to the readers to discuss how the TKE can be made non-dimensional in terms of flow parameters (a velocity scale specifically).

We are not sure what the reviewer has in mind here. The velocity scale comes from TKE, see section 5. Perhaps the discussion in Grabowski and Abade (JAS 2017) can help.

12. Figures 6–8: Please state whether the supersaturation statistics plotted are at the droplets or everywhere in the flow. They are most useful for understanding the DSD properties if they are at the droplets, as those for the stochastic model are.

This is a good point. For DNS, the statistics are for the flow and we mention this in the revision. At some point we compared the statistics for the flow and at the droplet positions. The differences between the two methods are small as shown in the figure below, so we decided to use the flow statistics as a much simpler to calculate. We feel this is because gradients at the grid scale are small due to molecular (or scaled-up molecular) transport coefficients and droplets (or super-droplets) present only in the small fraction of grid volumes.

The upper panel in the figure below shows the statistics derived using the flow data. The figure includes some data shown in the manuscript's Fig. 7. The bottom panel shows statistics calculated by interpolating the supersaturation to the droplet position. There are some small differences, but the mean values are close. We mention that in the footnote of the revised section 4.



13. Figure 7; It would be enlightening to the readers to discuss how σS can be made non-dimensional in terms of flow parameters.

We do not think this is possible. There are several parameters that likely play the role, and only some (like the domain size) vary in our study. Perhaps Fig. 10 provides something along the lines suggested by the reviewer.

14. Figure 9: It would be enlightening to the readers to discuss how σR2 can be made non-dimensional in terms of flow parameters.

Please see our reply to 13 above.

A general comment to 13 and 14 above: the problem has several parameters. From the fluid flow, the eddy dissipation rate and the domain size are the key (see Grabowski and Abade JAS 2017). From the cloud droplet perspective, the droplet concentration and initial size are relevant. We doubt our limited set of simulations, used primarily to demonstrate the approach strength, allows

to draw specific conclusions in terms of nondimensional parameters. This has to be left for future follow-up studies.

15. lines 272-73: Clarify this sentence: “The fluctuating in space supersaturation in the dynamic simulations (i.e., real DNS or scaled-up DNS) is modelled as independent realizations of the fluctuating in time supersaturation." I am not sure what you mean. Modelled by what? The stochastic model? Also clarify that the stochastic model predicts supersaturation fluctuations at the droplets. (See comment 12.)

The text has been modified to better explain how this is done in the stochastic model. The fact that the stochastic model predicts supersaturation in the droplet vicinity is mentioned in the footnote in section 4.

16. line 283 and Figure 10: Clarify that the stochastic model predicts supersaturation fluctuations at the droplets.

See response to 15.

17. line 317: Please explain why small-scale motions would reduce σS.

Just simply because of the heuristic argument that missing scales of motions would provide additional smoothing of supersaturation gradients. See also our response to 21 below.

18. Section 6: It would probably be helpful to many readers to make clear how scaledup DNS differs from LES. In my view, it is really LES but with a constant eddy viscosity, which is not a good SGS model. Explain why you chose this approach rather than using a better SGS model? I would also suggest that you discuss the advantages and the disadvantages of the scaled-up DNS approach. For the problem addressed, it seems that the stochastic model captures the important physics, and that the scaled-up DNS does not add any additional insights.

We do not agree with this comment. The paper shows how to expand DNS to larger domains at the same Reynolds number and thus to include larger eddies featuring larger supersaturation fluctuations. We think a better analogy is the so-called implicit LES, that is, an approach used in finite-difference models where no SGS scheme is used. Unfortunately, such a technique cannot be used with a spectral model and this is where the scaled-up DNS fits. We mention this in the revised introduction.

19. lines 332-33: I agree with this statement. You may want to mention again other possible methods that have been developed (such as those mentioned in comments 2, 5, and 7).

Again, we specifically target homogeneous isotropic turbulence simulation methodology. We do not want to bring other methods as not relevant to what we present.

20. lines 365-6: This might be too general of a statement. The large eddies dominate for this mode of supersaturation fluctuation because they span a larger potential temperature difference for the same mean vertical gradient. For other modes of supersaturation fluctuation generation

such as entrainment, large eddies also dominate, but for a different reason (their greater mixing time scale).

This comment is incorrect. Larger eddies feature larger and longer-lasting vertical velocity fluctuations because of the way TKE scales with L for the same eddy dissipation rate. As explained above, spectral DNS has no mean vertical gradients.

21. lines 381-82: "Finally, one can also consider applying scaled-up DNS in simulations of the turbulent entrainment and mixing similar to those discussed in Kumar et al. (2018)." There is a significant drawback for this application of scaledup DNS due to the importance of the small-scale supersaturation fluctuations in determining DSDs. Such near-Kolmogorov-scale variability is not present in scaled-up DNS.

It remains to be seen if the small-scale supersaturation fluctuations are important compared to large-scale fluctuations. The reviewer seems to believe so. We are not sure. Grabowski (JAS 2020) makes that point while discussing ILES simulations of the Pi chamber. He states that only true DNS can answer this question through the comparison with LES or ILES. We added a reference to Paoli and Shariff (2009) that is yet another method to include the impact of entrainment/mixing into DNS and scaled-up DNS in addition to Kumar et al. (2018).

Technical Comments

1. line 139: It would be helpful to define L1 and L2.

Text modified.

Diffusional growth of cloud droplets in homogeneous isotropic turbulence: DNS, scaled-up DNS, and stochastic model

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Abstract. This paper presents a novel methodology to use the Direct Numerical Simulation (DNS) to study the impact of isotropic homogeneous turbulence on the condensational growth of cloud droplets. As shown by previous DNS studies, the impact of turbulence increases with the computational domain size, that is, with the Reynolds number, because larger eddies generate higher and longer-lasting supersaturation fluctuations that affect growth of individual cloud droplets. The traditional DNS can only simulate a limited range of scales because of the excessive computational cost that comes from resolving all scales involved, that is, from large scales at which the turbulent kinetic energy (TKE) is introduced down to the Kolmogorov microscale, and from following every single droplet. The novel approach is referred to as the 'scaled-up DNS'. The scaling-up is done in two parts, first by increasing both the computational domain and the Kolmogorov microscale, and second by using super-droplets instead of real droplets. To ensure proper dissipation of TKE and scalar variance at small scales, molecular transport coefficients are appropriately scaled-up with the grid length. For the scaled-up domains, say, meters and tens of meters, one needs to follow billions of real droplets. This is not computationally feasible, and so-called super-droplets are applied in scaled-up DNS simulations. Each super-droplet represents an ensemble of identical real droplets, and the number of real droplets represented by a super-droplet is referred to as the multiplicity attribute. After simple tests showing validity of the methodology, scaled-up DNS simulations are conducted for five domains, the largest of 64^3 m^3 volume using a DNS of 256^3 grid points and various multiplicities. All simulations are carried out with vanishing mean vertical velocity and with no mean supersaturation, similarly to past DNS studies. As expected, the supersaturation fluctuations as well as the spread in droplet size distribution increase with the domain size, with the ~~mean~~ droplet radius variance increasing in time t as $t^{1/2}$ as identified in previous DNS studies. Scaled-up simulations with different multiplicities document numerical convergence of the scaled-up solutions. Finally, we compare the scaled-up DNS results with a simple stochastic model that calculates supersaturation fluctuations based on the vertical velocity fluctuations updated using the Langevin equation. Overall, the results document similar scaling as in previous small-domain DNS simulations and support the notion that the stochastic subgrid-scale model is a valuable tool for the multi-scale simulation of droplet spectral evolution applying large-eddy simulation model.

1 Introduction

The impact of turbulence on the growth of cloud droplets is an important and still poorly understood aspect of cloud physics. This is because of the wide range of spatial scales that affect droplet growth, from the Kolmogorov microscale (about a millimeter for typical atmospheric turbulence levels) to the scale of the entire cloud or cloud system. Cloud droplets grow by the diffusion of water vapor and by gravitational collision/coalescence, with the former dominating growth until droplets are large enough so the collisional growth can be initiated and eventually led to drizzle and rain formation. For the gravitational collision/coalescence, the frequency of droplet collisions depends on the droplet spectrum width. It follows that understanding processes leading to the observed droplet spectra is important for the understanding of the rain onset. Observations of natural droplet spectra go back to the early days of aircraft cloud studies (e.g., Warner, 1969) and continue in numerous subsequent investigations (e.g., Jensen et al., 1985; Brenguier and Chaumat, 2001; Pawlowska et al., 2006; Prabha et al., 2012, among many others; see also references in Grabowski and Wang, 2013). Those observations typically show that observed droplet spectra are wider than predicted by simple models of cloud dynamics and microphysics. ~~Reasons for~~ In many instances, such a discrepancy ~~have occupied cloud physics community for decades, with some of those mechanisms involving cloud turbulence.~~ can be explained by cloud entrainment (e.g., Warner, 1973; Paluch and Knight, 1984; Su et al., 1998; Lasher-Trapp et al., 2005, among many othe . However, presence of a significant spectral broadening in undiluted and weakly diluted cloudy volumes is more difficult to explain. One can wonder if the presence of small-scale turbulence can lead to appreciable widening of the droplet spectra during diffusional growth within otherwise uniform cloudy volumes.

~~Srivastava (1989) was first to argue that the traditional approach to the growth of a cloud droplet population by the diffusion of water vapor, the macroscopic approach, is not appropriate, and it should be replaced by an approach where each droplet grows in response to the supersaturation in its immediate environment, the microscopic approach. Such an argument is consistent with a study by Cooper (1989) who conjectures that droplets observed at a given location within a turbulent cloud arrive there following different trajectories and experiencing different growth histories, and that this process results in a significant broadening of the droplet spectrum. This mechanism is referred to as the eddy hopping in Grabowski and Wang (2013) , see also Grabowski and Abade (2017) and Abade et al. (2018). Lasher-Trapp et al. (2005) show that eddy-hopping can indeed result in a significant spectral broadening and can simulate droplet spectra in a better agreement with in-situ observations. Cooper et al. (2013) argue that spectral broadening resulting from eddy hopping can significantly accelerate drizzle and rain formation in cumulus clouds.~~

~~Motivated by Srivastava (1989),~~ Vaillancourt et al. (2001, 2002) were first to apply direct numerical simulation (DNS) approach to study diffusional growth of cloud droplets in ~~a turbulent environment~~homogeneous isotropic turbulence applying a rising adiabatic parcel setup. DNS has been initially applied to turbulent particle-laden flows to study the so-called preferential concentration (or clustering) of inertial particles in turbulence (e.g., Eaton and Fessler, 1994, see also references in Shaw, 2003). ~~Arguably, the clustering can also affect diffusional growth of cloud droplets as argued in Shaw et al. (1998); however, see the comment on that paper by Grabowski and Vaillancourt (1999).~~ For the DNS of cloud droplets growing by the diffusion of water vapor, ~~the~~ droplets respond to the supersaturation fluctuations in their immediate environment as sug-

gested by Srivastava (1989). Limited by the computational resources, Vaillancourt et al. (2002) were only able to consider small volumes of a turbulent cloud, around 1 liter. Three sets of simulations were performed with turbulent kinetic energy dissipation rates relevant to cloud conditions and including droplet sedimentation. Earlier studies of particle-laden turbulent flows typically exclude sedimentation (see Eaton and Fessler, 1994) but this is inappropriate for weak to moderate turbulence intensities typical for natural clouds (Grabowski and Vaillancourt, 1999). Vaillancourt et al. (2002) simulations show a small impact of local supersaturation fluctuations on the droplet spectra: the standard deviation of the initially monodisperse droplet distribution increases very slowly with time, of the order of 0.01 μm per minute. Similar simulations reported in Lanotte et al. (2009) applying larger domains and no mean ascent clearly show that the impact, although still relatively small (a few tenths of 1 μm), does increase with the domain size (see Fig. 3 therein). In similar DNS simulations, Li et al. (2019) demonstrate the increase of spectral broadening with the increase of the domain size (i.e., the ~~increasing~~ Reynolds number) and the increase of the length of the simulations, see Figs. 3 and 4 therein. For the largest domain of 512^3 and about a minute of the simulation time, the initially monodisperse 10 µm droplets evolve into a spectrum with about 1 µm width.

~~DNS~~ Homogeneous isotropic turbulence simulations of Vaillancourt et al. (2002), Lanotte et al. (2009) and Li et al. (2019) are limited by the computational domain size. As a result, simulations featuring domains larger than a fraction of a cubic ~~meters~~ meter are simply not yet possible. At the same time, as argued in Grabowski and Wang (2013) and documented in Grabowski and Abade (2017; see Fig. 4 therein) and Li et al. (2019; see Fig. 4 therein), the impact of ~~turbulent supersaturation fluctuations~~ supersaturation fluctuations in homogeneous isotropic turbulence on the spectral width increases with the domain size. A simple argument is that this is because the largest turbulent eddies ~~impact most strongly the local~~ feature the largest vertical velocity perturbations that result in the largest and longest-lasting supersaturation fluctuations and thus ~~the spread of the~~ have the largest impact on the spread of droplet growth histories. From the point of view of realistic cloud modelling, developing and validating robust subgrid-scale schemes for contemporary large eddy simulation (LES) models (i.e., featuring grid lengths of a few tens of meters) requires performing DNS-like simulations in computational domains comparable to the size of the LES grid box. Such an attempt was made in Kumar et al. (2018) who conduct DNS simulations applying up to $(2\ \mathrm{m})^3$ domain size and report systematic analysis of the dependence of mixing processes on the DNS domain size.

To this end, we propose to use what we refer to as the "scaled-up DNS" approach. Since the largest eddies are the key for the condensational growth, one would like to apply the DNS technique in simulations with domains much larger than currently possible. For instance, taking a 128^3 DNS simulation with 10 cm grid length gives computational domain of 12.8^3 cubic meters, that is, comparable to the grid volume of an LES cloud simulation. To ensure a proper dissipation of the turbulent kinetic energy (TKE) at the smallest scales, one needs to scale up the molecular viscosity with the increase of the model grid length. The increase of the small-scale dissipation is critical for traditional DNS models applying spectral techniques to simulate homogeneous isotropic turbulence as applied in this study. This is different from past turbulence-related studies applying finite-difference models with large domains and spatially-uniform diffusion coefficients (e.g., Grabowski and Clark, 1993; Mellado et al., 2 or no explicit diffusion at all as in the so-called implicit large eddy simulation (e.g., Margolin et al., 2006; Grinstein et al., 2007) . The increased number of droplets in the large domain can be accounted for by the so-called ~~"~~super-droplet ~~"~~ technique in

which each super-droplet represents an appropriately scaled-up number of natural droplets (referred to as the multiplicity factor, Shima et al., 2009) as already applied in the appendix of Lanotte et al. (2009) and in Li et al. (2019).

The paper is organized as follows. The next section presents the model and modelling setup. Section 3 presents a general methodology of the scaled-up DNS and discusses numerical tests of this approach. Cloud droplets are added to scaled-up DNS simulations in section 4 applying the super-droplet method. Section 5 compares DNS and scaled-up DNS supersaturation fluctuations with those obtained from a simple stochastic model. Concluding discussion is the focus of section 6.

2 The model and modelling set up

The numerical code used here is that of Kumar et al. (2012, 2014). It solves evolution equations for the three velocity components (u, v, w), the temperature T and the water vapor mixing ratio q_v. Cloud droplets are represented as point particles followed in space and they grow or evaporate as dictated by their local environment. Droplet collisions are not considered. The Navier-Stokes equations are solved by a pseudo-spectral method over a cubic volume with periodic lateral boundary conditions in three directions using the fast Fourier transforms. Time stepping is performed using a second order predictor-corrector method. The code is parallelized in two dimensions and the cubic domain is decomposed into so-called pencils. The same procedures as in Kumar et al. (2012) are followed for the initial turbulent state preparation and the turbulence maintenance. See Kumar et al. (2012, 2014) for more details.

Two modifications have been made to the code to carry out the present study. First, we included an additional source/sink term in the temperature equation that was missing in the original code. The term describes evolution of temperature fluctuations affected by the vertical velocity. This effect is incorporated in the DNS through the source/sink term $-gw/C_p$, where g is the gravitational acceleration, w is the local vertical velocity, and C_p is the specific heat capacity of air at constant pressure. The complete equation for the evolution of temperature fluctuations is:

$$\frac{\partial T'}{\partial t} + \mathbf{u}.\nabla T' = K\nabla^2 T' + \frac{L}{C_p}\frac{L_v}{C_p}C_d - \frac{g}{C_p}w' \ , \tag{1}$$

where K is the ~~molecular~~ thermal diffusion coefficient, ~~L~~ L_v is the latent heat of vaporization, and C_d is the condensation rate. Without the last term, the vertical velocity simulated by the DNS has no impact on the supersaturation fluctuations. Since the emphasis in Kumar et al. (2014, 2018) was on the mixing between cloudy and clear air, this omission has a negligible impact on results presented there. However, this term is critical for the current study.

Second, we modified the way condensation rate is calculated for a single droplet. The analytic formulation applied originally has the form:

$$C_d = \frac{4\pi K_r \rho_w}{\rho_0 \, \Delta V} SR \, \Delta t, \tag{2}$$

where S is the supersaturation, R is the droplet radius, ρ_0 is the air density, $\rho_w = 10^3$ kgm^{-3} is the liquid water density, $K_r = 5.00 \times 10^{-11}$ m^{2}s^{-1} is the condensational growth constant (i.e., $dR/dt = K_r S/R$), $\Delta V = dx \times dy \times dz$ is the grid box

volume~~, and Δt is the time step~~. To ensure mass conservation, (2) is modified to:

$$C_d \Delta t = \frac{4\pi\rho_w}{3\rho_0\,\Delta V}[R^3(t) - R^3(t-\Delta t)]\,, \tag{3}$$

where Δt is the time step, $R(t)$ and ~~$R(t-\Delta t)$~~ $R(t-\Delta t)$ are droplet radii at time t and ~~$t-\Delta t$~~$t-\Delta t$, respectively. This means that droplet growth is calculated first, and then (3) is used to derive the condensation rate.

The coupling of the Eulerian fields and the droplets is done using trilinear interpolation. The condensation rate is calculated for each droplet by interpolating the values of T and q_v from the grid points enclosing the droplet. The condensation rate is calculated at the droplet position and then redistributed to the nearest eight grid points through a reverse procedure. The condensation rate provides a feedback on the temperature and water vapor evolutions. Inertial effects and gravitational settling are included in the droplet motion. More details can be found in Kumar et al. (2012).

The modeling setup follows one of the simulations discussed in Lanotte et al. (2009). We consider an initial mono-disperse droplet distribution of 13 μm radius and the concentration of 130 cm^{-3}. The liquid water content (LWC) is 1.19 gm^{-3}. The initial conditions include uniform temperature of 283 K and zero supersaturation. The later gives the water vapor mixing ratio of 7.65 gkg^{-1}. Since the mean velocity inside the DNS domain is zero, the total cloud water does not change with time, but the initial monodisperse droplet size distribution broadens because the supersaturation fluctuates in time and space affecting the distribution (cf. Li et al., 2019; Saito et al., 2019). The two specific aspects are discussed in the next two section that allow extending the DNS methodology ~~into~~ to apply large spatial domains.

3 Scaling-up DNS simulation

The intensity of turbulence is typically expressed by the turbulent kinetic energy (TKE) dissipation rate ε. Increasing the domain size L for the same ε increases kinetic energy of turbulent motions. The TKE determines velocity fluctuations and controls the supersaturation variations that play the key role in the condensational growth of cloud droplets. The TKE dependence on ε and L is typically expressed as (e.g., Pope, 2000):

$$E \sim (L\varepsilon)^{2/3}\,. \tag{4}$$

In the classical DNS, the grid length has to be close to the Kolmogorov microscale η to allow proper TKE dissipation at the smallest scales. Increasing the domain size L without changing the number of grid points implies that the grid length increases as well. We will refer such simulations as "scaled-up DNS". With the increased grid length, one needs to increase molecular transport coefficients to maintain proper TKE dissipation as well as the removal of scalar fluctuations. Assuming that the domain size L represents appropriate scale of energy-containing eddies, the L/η ratio represents the flow Reynolds number Re:

$$\frac{\eta}{L} \sim Re^{-3/4} \tag{5}$$

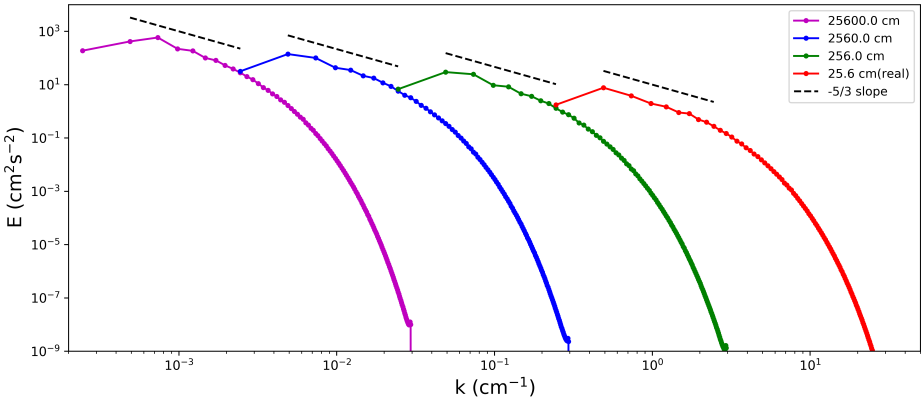

Figure 1. Comparison of energy spectra for real and scaled-up DNS

(e.g., Pope, 2000; Grabowski and Clark, 1993). Keeping the Reynolds number the same for the actual and scaled-up DNS implies that

$$\frac{L_1}{\eta_1} = \frac{L_2}{\eta_2}, \tag{6}$$

where subscripts 1 and 2 refer to different domain sizes and corresponding Kolmogorov microscales. The Kolmogorov microscale is given by $\eta = (\frac{\nu^3}{\varepsilon})^{1/4}$ where ν is the viscosity. Applying η with the same TKE dissipation rate ε for both ν_1 and ν_2 leads to

$$\frac{L_1}{\nu_1^{3/4}} = \frac{L_2}{\nu_2^{3/4}}. \tag{7}$$

Eq. (7) implies that when the TKE dissipation rate is supposed to be the same in DNS and scaled-up DNS, the molecular viscosity needs to be scaled up as:

$$\nu_2 = \nu_1 \left(\frac{L_2}{L_1}\right)^{4/3}, \tag{8}$$

where ν_1 is the viscosity in the real DNS, ν_2 is the scaled-up viscosity, L_1 is the real-DNS domain size, and L_2 is the scaled-up domain size. Thermal ~~diffusivity is~~ and water vapor diffusivities are also changed in the same way as the viscosity.

We used a DNS with 256^3 grid points to study scaling-up simulations without droplets. Real DNS was run for $L = 0.256$ m and scaled-up DNS was run with domains of sizes $L = 2.56$ m, $L = 25.6$ m and $L = 256$ m. According to (8), the viscosity (taken as $\nu = 0.15$ cm^{2}s^{-1} for the real DNS) has to be scaled-up by 21.54, 464.16 and 10,000 times for $L = 2.56$ m, $L = 25.6$ m and $L = 256$ m, respectively. All simulations are forced as described in Kumar et al. (2012) applying TKE dissipation rate ε of 10 cm^{2}s^{-3} as in Lanotte et al. (2009).

Fig. 1 shows energy spectra for the real 25.6 cm DNS and the three scaled-up DNS. The black dashed lines represents the -5/3 slope expected in the inertial range. The spectral peak shifts to the left and its value increases as the domain size increases (i.e., the wavenumber k decreases). The slope remains approximately similar for the four simulations.

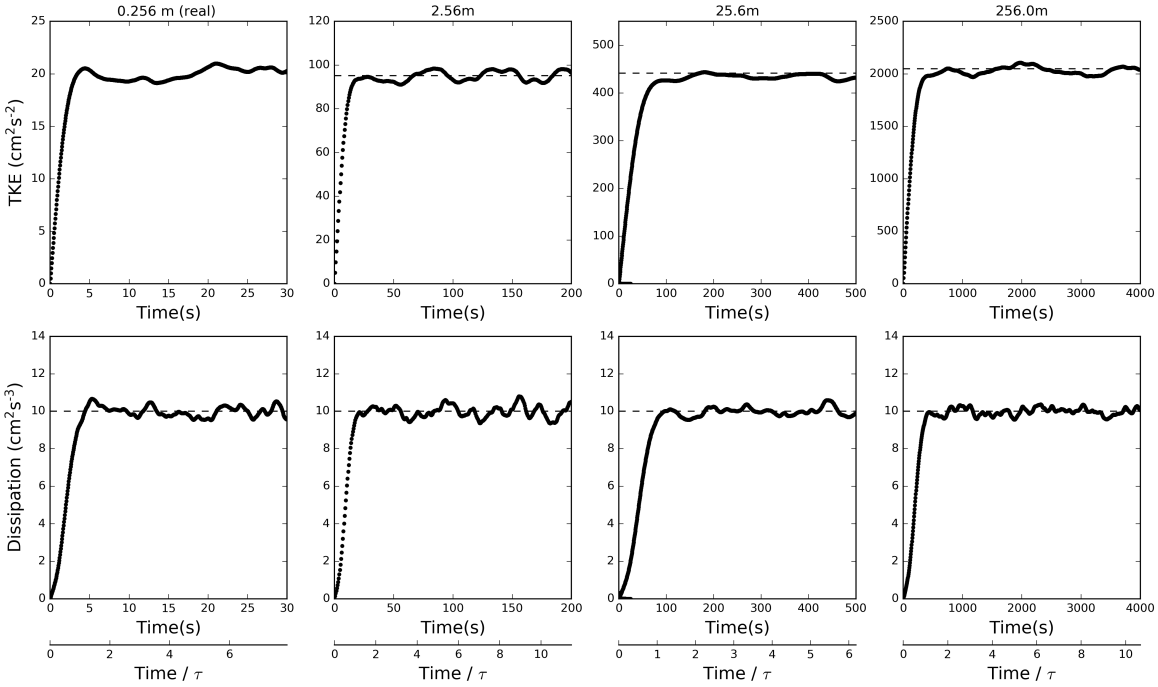

Figure 2. Evolution of TKE (upper panels) and TKE dissipation rate (lower panels) for four simulations mentioned in text. The dashed lines are theoritical values. Bottom panels show nondimensional time using eddy turnover time, see (13) in section 5.

TKE in the scaled-up simulations for the same TKE dissipation rate ε should increase following the scaling originating from (4), that is,

$$E_1 = E_2 \left(L_1/L_2\right)^{2/3}, \tag{9}$$

where E_1 and L_1 are for the scaled-up DNS and E_2 and L_2 are for real DNS (e.g., $L_2 = 0.256$ m and $E_2 = 20$ cm^{2}s^{-2}). Fig. 2 shows the evolution of TKE and TKE dissipation rate for the four simulations in Fig. 1. For the TKE evolution, dashed lines show the expected scaling based on (9). TKEs from the scaled-up DNS simulations agree with the theoretical scaled-up TKE values. To show that the DNS and scaled-up DNS feature the same TKE dissipation rate, we also show the dissipation rate calculated using the simulated enstrophy as typically done in DNS studies. The plots show that the forcing is approximately correct in the scaled-up simulations. The scaled-up simulations need to be run for longer times, with the time scale following the $L/E^{1/2}$ scaling of the large-eddy turnover time. The simulations show that the scaled-up DNS with viscosity modified according to (8) produces expected TKE.

The simulations shown in Fig. 1 and Fig. 2 feature the same dynamic range, that is, the same Reynolds number and the L/η ratio. However, one may also consider scaled-up DNS simulations where the dynamic range is changed. For instance, one may compare simulations with the same ε and L, and different numbers of grid points N covering L. For such simulations, the change of the Kolmogorov microscale $\eta = L/N$ suggests the required rescaling of the dissipation coefficients. Since $\eta =$

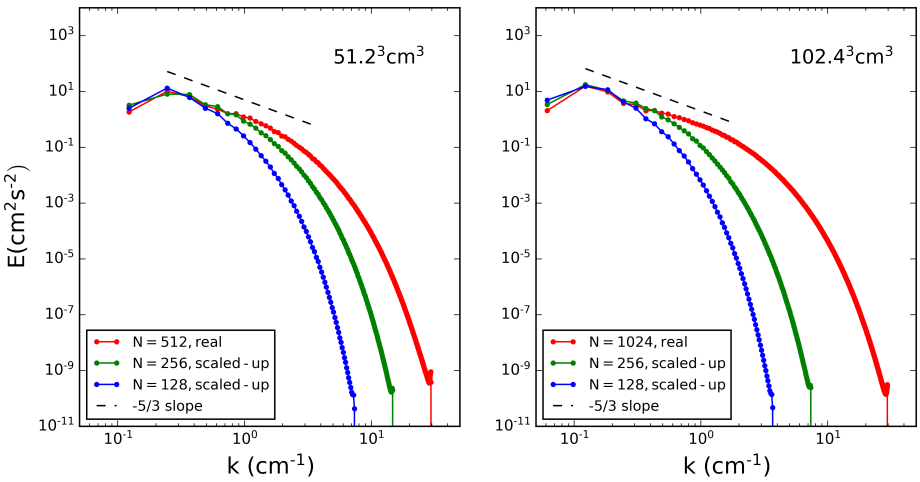

Figure 3. Energy spectrum comparison of real DNS and scaled-up DNS. Left/right panels correspond to $512^3/1024^3$ real DNS.

$\left(\frac{\nu^3}{\varepsilon}\right)^{1/4}$, assuming ε = const gives the scaling similar to (8):

$$\nu_2 = \nu_1 \left(\frac{N_1}{N_2}\right)^{4/3} \quad (10)$$

that is, with the number of grid points rather than the domain size providing the scaling.

The scaling (10) is illustrated in Fig. 3 that shows the spectra in simulations with the domain size of either 0.512 m or 1.024 m and applying either DNS or scaled-up DNS. The spectra are obtained at final simulation times. The red lines represent spectra for the real DNS, and green and blue lines show spectra for scaled-up DNS. Scaling-up accurately predicts the energy at the largest scales, but some energy at smaller scales, still far from the dissipation, is lost. This means that the total TKE for a scaled-up DNS is slightly lower than the real DNS within the same volume. For the simulations shown in Fig. 3, TKE for L of 0.512 m is 34.2, 32.0, and 26.0 $\mathrm{cm^2 s^{-2}}$ for real DNS (N=512), and scaled-up DNS with N=256 and 128, respectively. For ~~L~~ L of 1.024 m, TKE is 55.0, 50.0, and 41.0 $\mathrm{cm^2 s^{-2}}$ for real DNS (N=1024), and scaled-up DNS with N=256 and 128, respectively. Because for the condensational growth the interest is on the largest scales as discussed in the introduction, the energy loss at smaller scales can be considered less important. However, this aspect is relevant for the comparison between scaled-up DNS and the stochastic model as discussed in section 5.

4 Applying superdroplets for the scaled-up DNS

For a scaled-up DNS, one needs to follow significantly larger number of droplets when compared to DNS. For instance, for the droplet concentration of 130 $\mathrm{cm^{-3}}$ one needs to follow 1.3×10^{11} droplets for a domain of $L = 10$ m. This is not computationally feasible. To overcome this problem, one can use the so-called super-droplets (Shima et al., 2009) instead of real droplets, where each super-droplet represents an ensemble of real droplets with the same radius. Position and velocity of

Table 1. Number of super-droplets and their multiplicity for real DNS domains of volume 6.4^3 cm^3 and 12.8^3 cm^3.

~~L~~ L = 6.4 cm	~~L~~ L = 12.8 cm
$N_s = 34078; \mu = 1$	$Ns = 272630; \mu = 1$
$N_s = 17039; \mu = 2$	$Ns = 54526; \mu = 5$
$N_s = 6815; \mu = 5$	$Ns = 27263; \mu = 10$

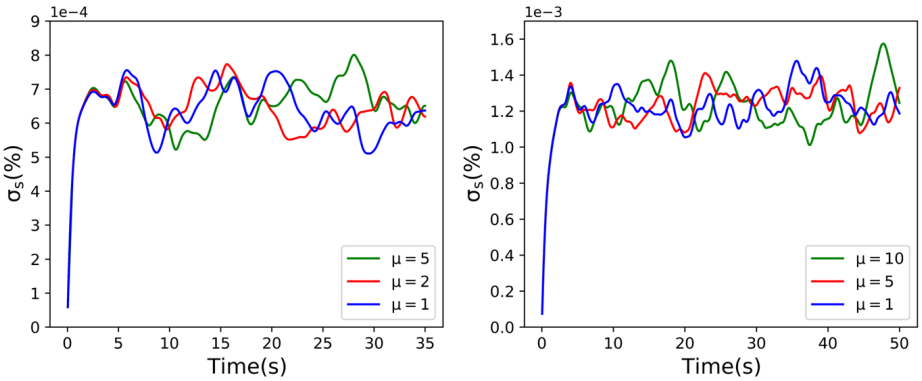

Figure 4. Standard deviation of supersaturation fluctuations for 6.4^3 cm^3 real DNS (left panel) and 12.8^3 cm^3 real DNS (right panel). Colors represent different multiplicity as marked inside each panel.

each super-droplet is predicted in the same way as for the real droplet. The number of real droplets represented by a single super-droplet is referred to as the multiplicity attribute μ (Shima et al., 2009).

205 At the onset of simulations, super-droplets are inserted into the computational domain in the same way as regular droplets, that is, they are randomly positioned inside the domain and subsequently followed in space and time as regular droplets. The condensation rate for a super-droplet is calculated as in (~~2~~3) except for an additional multiplicity factor μ. The evolution of the temperature and water vapor mixing ratio fluctuations is affected by the condensation rate of super-droplets within a grid box in the same way as regular droplets.

210 The super-droplet approach is first tested in the real DNS. Fig. 4 shows evolutions of the standard deviation of the supersaturation spatial fluctuations[1] for real DNS of $L = 0.064$ m (64^3 grid points) and $L = 0.128$ m (128^3 grid points) with different multiplicity parameter. Number of superdroplets used and their multiplicity are listed in Table 1. For randomly distributed real droplets, the two simulations have droplets in one out of about eight grid boxes. For super-droplets in 64^3 simulations, this number changes to one out of about 15 grid boxes for μ = 2 and about 38 grid boxes for μ = 5. For the 128^3 simulations, super-

[1] Supersaturation statistics in DNS and scaled-up DNS are calculated using fluid flow grid data and not the supersaturation interpolated to droplet positions. Limited tests suggests that the differences between the two methods are small (not shown). Supersaturation statistics for the stochastic model in Section 5 are for the vicinity of a droplet. Discussion in the Appendix A of Vaillancourt et al. (2001) is pertinent to this issue.

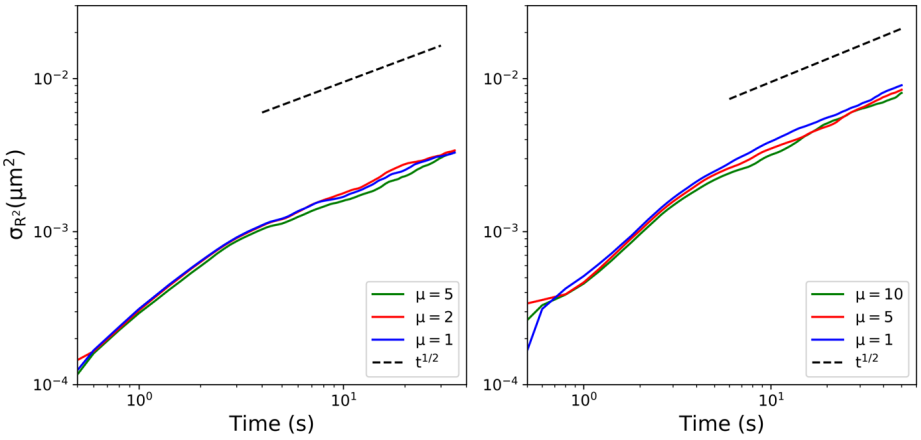

Figure 5. Evolutions of the radius squared standard deviation (σ_{R^2}) for real DNS with different multiplicity parameters. Left/right panels corresponds to $64^3/128^3$ real DNS ($L = 6.4/12.8$ cm). Colors represent different multiplicity as marked inside each panel.

droplets are on average in one out of about 43 and 87 grid boxes for μ = 5 and 10, respectively. The mean supersaturation is close to zero as expected (not shown). Supersaturation standard deviations fluctuate similarly in all simulations with the mean values close among all multiplicities. The mean value of the standard deviation is larger for the larger domain in agreement with simulations discussed in Lanotte et al. (2009).

In general, the multiplicity value should be decided carefully because too large multiplicity results in too many grid boxes without droplets when compared to real droplets and this may cause undesirable effects in the mean supersaturation and its spatial variability. In the two DNS cases, slight deviations in the mean supersaturation are present, although the simulations are not long enough to document the impact with confidence. For the scaled-up DNS, the number of droplets is in billions and we have to select higher multiplicity values to make computations feasible. The evolution of the radius squared (R^2) standard deviation (σ_{R^2}) from the above simulations with droplets and super-droplets is shown in Fig. 5. Initially (i.e., at t = 0), the distributions are monodisperse (i.e., σ_{R^2} = 0). Supersaturation fluctuations in response to local vertical velocity fluctuations lead to the increase of σ_{R^2} in time. After some initial adjustment, the increase approximately follows the $t^{1/2}$ scaling with t being the time from the start of the simulation. This agrees with the study by Sardina et al. (2015) who applied a stochastic model and DNS. Similar result is also shown in Li et al. (2019) and Saito et al. (2019). As expected, the σ_{R^2} values are larger for the larger domain, in agreement with Fig. 3 in Lanotte et al. (2009) and Li et al. (2019).

After the super-droplet technique is tested in DNS, the same method is used in scaled-up DNS. In general, one may expect that if the multiplicity is increased beyond a certain value, the results will start deviate from those with a low multiplicity featuring a larger number of super-droplets. However, high multiplicity is desirable to reduce the number of super-droplets that need to be followed. For the DNS, the number of super-droplets was shown to be relatively low to maintain similarity between real droplet and super-droplet solutions (see Fig. 4 and Fig. 5; as low as one super-droplet in a few dozen of grid volumes).

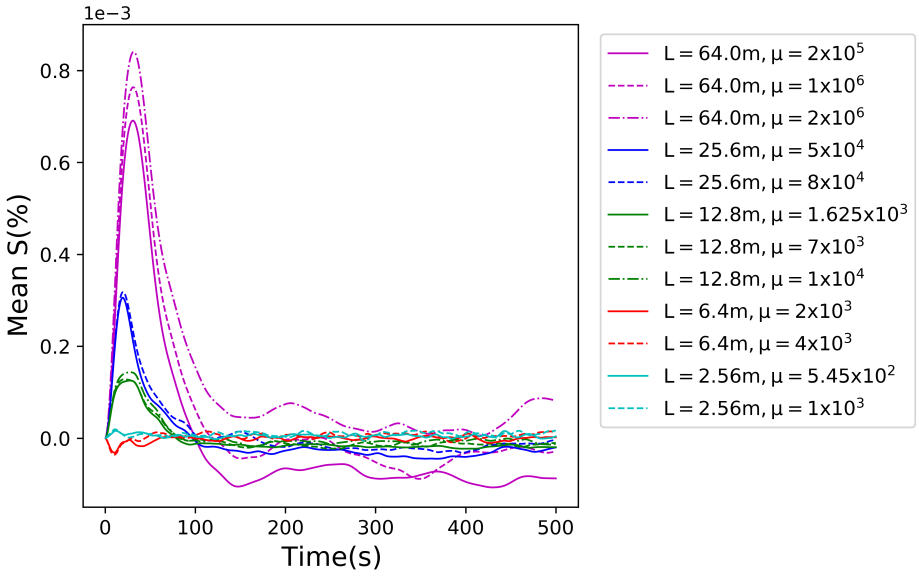

Figure 6. Evolution of the mean supersaturation for various scaled-up domains. Colors represent different domain size; different line styles correspond to different multiplicities. The additional simulation of 10 super-droplets per grid volume is only shown for 12.8^3 m^3 and 64.0^3 m^3 volumes.

With scaled-up DNS, one might expect a different requirement because of a stronger local forcing of the supersaturation due to higher TKE and thus larger vertical velocities.

For the scaled-up DNS study, we apply a 256^3 domain to represent volumes with characteristic lengths of several meters. The TKE is scaled as explained in section 3 with relevant parameters listed in table 2. As the table shows, scaled-up DNS simulations typically have a relatively small number of super-droplets per grid box, similarly to DNS. This is because of computational efficiency considerations. However, one may question such an approach because scaled-up DNS include a large number of real droplet (e.g., $\sim 10^9$ for scaled-up DNS with 1 cm grid length and much larger numbers for scaled-up DNS with larger grid lengths). To show that the standard deviation of the supersaturation spatial distribution is not affected by the small number of super-droplets considered in the scaled-up DNS simulations, we included additional simulations (shown in bold in the Table 2) that include about 10 super-droplets per grid volume and follow about 160 millions of super-droplets. Although arguably still a small number, 10 super-droplets per grid box is the number considered in one of the sensitivity simulations in Grabowski (2020). By comparing results of simulations with various numbers of super-droplet per grid volume, Grabowski (2020) shows that the number as small as 10 is sufficient to reasonably represent condensational growth of natural droplets in idealized simulations of laboratory chamber experiments. Fig. 6 and Fig. 7 present evolutions of the mean supersaturation and standard deviation of its spatial distribution for the scaled-up simulations from Table 2. The five scaled-up domains shown in the table and figures correspond to the domain size L of 2.56, 6.4, 12.8, 25.6 and 64 m. Note that the simulations extend to times of several minutes, that is, a significant fraction of a small convective cloud life cycle. All ~~simulations except 12.8 and 64~~

Table 2. Details of DNS and scaled-up DNS. From left to right: domain length L, grid length l, viscosity ν, Turbulent kinetic energy E, number of superdroplets in the domain N_s, multiplicity μ, number of superdroplets per grid volume N_s/N^3

	$L(\mathrm{cm})$	$l(\mathrm{cm})$	$\nu(\mathrm{cm^2 s^{-1}})$	$E(\mathrm{cm^2 s^{-2}})$	N_s	μ	N_s/N^3
Real DNS	25.6	0.1	0.15	20.0	$\sim 2.2 \times 10^6$	1.0	0.13
Scaled-up DNS	256.0	1.0	3.231	94.0	$\mathbf{\sim 1.7 \times 10^8}$	**13.0**	**10.0**
					$\sim 4.0 \times 10^6$	5.45×10^2	0.24
					$\sim 2.2 \times 10^6$	1.0×10^3	0.13
	640.0	2.5	10.965	171.0	$\mathbf{\sim 1.7 \times 10^8}$	**203.125**	**10.0**
					$\sim 1.7 \times 10^7$	2.0×10^3	1.01
					$\sim 8.5 \times 10^6$	4.0×10^3	0.51
	1280.0	5.0	27.630	270.0	$\mathbf{\sim 1.7 \times 10^8}$	$\mathbf{1.63 \times 10^3}$	**10.0**
					$\sim 3.9 \times 10^7$	7.0×10^3	2.32
					$\sim 2.7 \times 10^7$	1.0×10^4	1.61
	2560.0	10.0	69.624	420.0	$\mathbf{\sim 1.7 \times 10^8}$	$\mathbf{1.3 \times 10^4}$	**10.0**
					$\sim 4.4 \times 10^7$	5.0×10^4	2.62
					$\sim 2.7 \times 10^7$	8.0×10^4	1.61
	6400.0	25.0	236.235	750.0	$\mathbf{\sim 1.7 \times 10^8}$	$\mathbf{2.03 \times 10^5}$	**10.0**
					$\sim 3.4 \times 10^7$	1.0×10^6	2.03
					$\sim 1.7 \times 10^7$	2.0×10^6	1.01

~~m~~ scaled-up simulations are run with ~~two~~ three different multiplicities for super-droplets as listed in Table 2. As Fig. 6 shows, the mean supersaturation for all five scaled-up cases is close to zero after the initial spike. The spike magnitude, about hundred times smaller than the standard deviations shown in Fig. 7, increases as the domain size increases, and it is slightly larger for the higher multiplicity. Higher multiplicity also causes larger fluctuations after the initial spike, but the mean does not seem to be significantly affected. The standard deviation shown in Fig. 7 increases with the domain size as expected. For all domains, standard deviations are similar for various multiplicities. In particular, based on 12.8 m and 64 m simulations, the low number of super-droplets per grid volume (desirable for computational efficiency) seem to insignificantly impact the supersaturation statistics.

To further study the impact of the multiplicity, additional scaled-up DNS simulations are run with 256^3 grid points for a domain of size 12.8^3 m^3. All simulations are listed in Table 3, with some already considered in Table 2 and Fig. 6 and

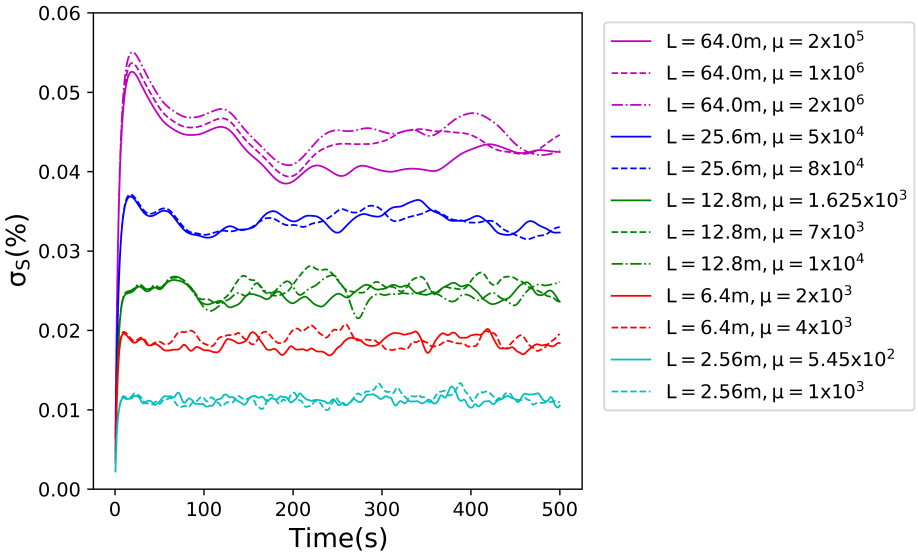

Figure 7. Evolution of standard deviation of supersaturation fluctuations for different domain sizes. Colors represent different domain size; different line styles correspond to different multiplicities. The additional simulation of 10 super-droplets per grid volume is only shown for 12.8^3 m^3 and 64.0^3 m^3 volumes.

Fig. 7. Total number of real droplets for the 12.8^3 m^3 domain with droplet concentration of 130 cm^{-3} is about 2.7×10^{11}. The grid volume of the 256^3 grid points and 12.83 m^3 simulation is 125 cm^3. When the multiplicity is 1625, the number of super-droplets is close to 170 millions and there are on average 10 super-droplets per grid volume. When μ is 7×10^3, the number of super-droplets is close to 40 millions and there are on average about 2.3 super-droplets per grid volume. When μ is further increased to 1×10^4, the number of super-droplets per grid volume decreases to about 1.6. For $\mu = 5 \times 10^4$, the number further decreases to about 0.32 (i.e., a super-droplet in about 3 grid volumes). Finally, for $\mu = 8.192 \times 10^5$, a super-droplet is approximately in one out of 50 grid volumes.

Results obtained from these simulations are shown in Fig. 8 with some results already shown in Fig. 7. As the figure shows, only the largest multiplicity, with a super-droplet in one out of 50 grid volumes differs significantly from other simulations. The highest multiplicity simulation also results in the non-zero mean supersaturation (not shown). Note that for real DNS (Table 2 and Fig. 4), having a droplet in one of several dozens of grid volumes still results in supersaturation fluctuations in agreement with real droplets. This suggests that the maximum multiplicity that can be used in scaled-up DNS depends on the domain size. This perhaps should not be surprising because the magnitude of the vertical velocity perturbation and thus the supersaturation forcing increases with the domain size. Results for the largest domain considered in the current study (64^3 m^3) suggest that the multiplicities selected for the scaled-up DNS provide robust (i.e., independent of the multiplicity) outcomes.

As shown in Fig. 9, evolutions of the radius squared standard deviation σ_{R^2} for scaled-up DNS domains follows the same trend as in the real DNS, that is, the standard deviations increase in time t as $t^{1/2}$. The results are shown for the five scaled-up cases mentioned above. Scaled-up DNS for each domain was run for three different multiplicity values, one of them being

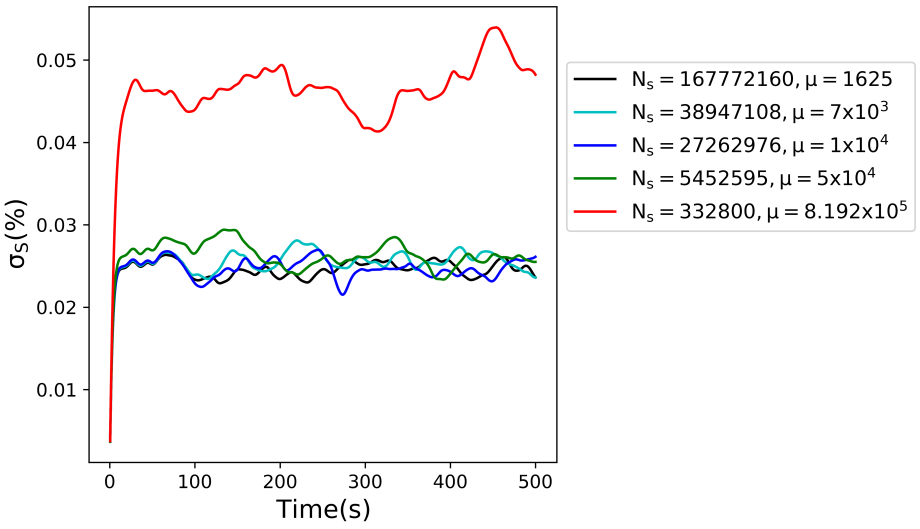

Figure 8. Standard deviation of supersaturation fluctuations for 12.8^3 m^3 scaled up domain. Colors indicate different multiplicities.

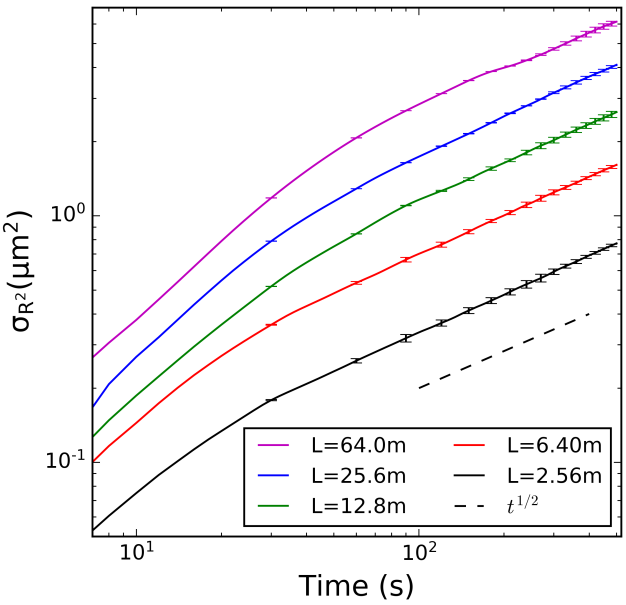

Figure 9. Evolutions of the radius squared standard deviation σ_{R^2} for different domain sizes in the scaled-up DNS simulations. Horizontal bars along each line show variability resulting from different multiplicity used for each domain size.

Table 3. Number of super-droplets and multiplicity for different 12.8^3 m^3 scaled-up domain simulations.

Number of superdroplets(N_s)	Multiplicity (μ)
$\sim 1.6 \times 10^7$	1625
$\sim 3.9 \times 10^7$	$\sim 7 \times 10^3$
$\sim 2.7 \times 10^7$	$\sim 1 \times 10^4$
$\sim 5.4 \times 10^6$	$\sim 5 \times 10^4$
$\sim 3.3 \times 10^5$	$\sim 8 \times 10^5$

10 super-droplets per grid volume. The error bars correspond to the standard deviation among realizations with different multiplicities. Overall, the scatter resulting from different multiplicities is relatively small. The key result in Fig. 9 is that the spectral width increases with the domain size. For domain sizes of a few tens of meters, the spectral width after a few minutes reaches values of 1-2 μm that is comparable to those observed in near-adiabatic cores of small cumuli (e.g., Jensen et al., 1985) or subtropical stratocumulus (e.g., Pawlowska et al., 2006).

5 Stochastic model

We apply the stochastic model similar to that in Grabowski and Abade (2017) to simulate fluctuating supersaturation and compare results to the real and scaled-up DNS. The fluctuating in space supersaturation in the dynamic simulations (i.e., real DNS or scaled-up DNS) is modelled in the stochastic model as independent realizations of the fluctuating in time supersaturation as described below. For each realization, the supersaturation fluctuations are driven by the vertical velocity fluctuations as given by the Ornstein-Uhlenbeck process (e.g., Pope, 1994). In its finite difference implementation, the velocity perturbations are updated as in Grabowski and Abade (2017):

$$w'(t+\delta t) = w'(t) e^{-\delta t/\tau} + \sqrt{1 - e^{-\frac{2\delta t}{\tau}}}\, \sigma_{w'} \psi \,, \tag{11}$$

where δt is the model timestep, $\sigma^2_{w'}$ is the vertical velocity variance obtained from TKE as

$$\sigma^2_{w'} = \frac{2}{3} E \,, \tag{12}$$

ψ is a Gaussian random number with zero mean and unit variance generated at every time step, τ is the eddy turnover time calculated as

$$\tau = \frac{L}{(2\pi)^{1/3}} \left(\frac{C_\tau}{E} \right)^{1/2} \,, \tag{13}$$

where C_τ is a constant equal to 1.5 as in Lasher-Trapp et al. (2005). Supersaturation fluctuations evolve according to the equation

$$\frac{dS'}{dt} = a_1 w' - \frac{S'}{\tau_{relax}} \,, \tag{14}$$

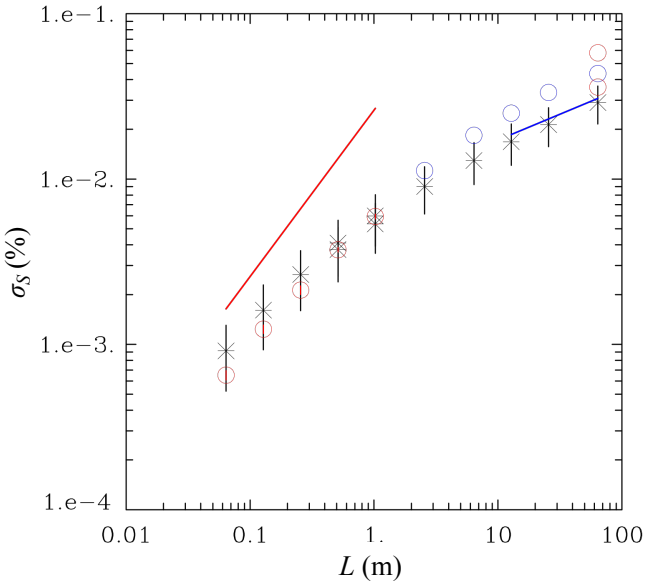

Figure 10. Standard deviation of supersaturation fluctuations in DNS and scaled-up DNS (colour circles) and the stochastic model (black stars). Vertical lines for the stochastic model represent variability among individual realizations. For DNS and scaled-up DNS, the variability comes from different multiplicity for super-droplets; it is not shown as it is smaller than the symbol size. Red circles in the left half are for DNS with three different multiplicities as in Fig. 4. Blue circles are for 256^3 scaled-up DNS simulations. Red circles for $L = 64$ m are for 128^3 (lower symbol) and 512^3 (upper symbol) scaled-up DNS simulations. The two data points for the stochastic model with $L = 0.512$ m and $L = 1.024$ m come from applying TKE from either DNS or scaled-up DNS. The red and blue lines show results from the stochastic model without droplets and with the quasi-equilibrium supersaturation, respectively. See text for details.

where w' is the vertical velocity perturbation evolving as in (11), a_1 is a temperature-dependent numerical coefficient and τ_{relax} is the phase relaxation time that depends on the temperature and pressure as well as on the droplet concentration and mean radius. For the conditions considered in this study, $a_1 = 4.753 \times 10^{-4}$ m^{-1} and $\tau_{relax} = 3.513$ s.

The stochastic model used here applies 1000 realizations, each starting from a random velocity perturbation [i.e., $\sigma_{w'}\psi$ as in (11)] and zero supersaturation, and run for 6 eddy turnover times. The time step in (11) and (14) is taken as one thousandth of the eddy turnover time. The number of realizations is sufficient to give results that change insignificantly when the number is further increased. Subsequently, the standard deviation of the supersaturation temporal evolution for each realization is derived. Its mean value averaged over all realizations together with the standard deviation among realizations is used in the comparison with the DNS and scaled-up DNS simulations. Fig. 10 shows the standard deviation of supersaturation fluctuations (σ_S) derived from the stochastic model as explained above for different domain sizes together with similar results from the DNS and scaled-up DNS dynamic simulations. The first five points (L = 0.064, 0.128, 0.256, 0.512, 1.024 ~~m~~m) correspond to the real DNS, whereas the last five points (L = 2.56, 6.4, 12.8, 25.6, 64.0 ~~m~~m) correspond to scaled-up DNS. The stochastic model uses TKE simulated by either DNS or scaled-up DNS, and for L = 0.512 and ~~1.02 4m~~ 1.024 m by both. The vertical lines for the stochastic model show twice the standard deviation among the realizations. The red circles in the left part of the figure are from

DNS simulations. Standard deviations from different multiplicities as shown in Fig. 4 are smaller than the circle radius and thus they are not shown. The blue symbols are for scaled-up ~~256^3~~ 256^3 DNS simulations; standard deviations from different multiplicities are again smaller than the radius. ~~Finally, the~~ The red circles for ~~$L = 64$ m~~ $L = 64$ m show σ_S in scaled-up DNS simulations with grids of 128^3 and 512^3. The red line shows results from the stochastic model without droplets, that is, with no last term on the rhs of (14). Finally, the blue line is for the stochastic model assuming quasi-equilibrium supersaturation S'_{eq}, that is, with the lhs of (14) equal to zero and thus $S' = S'_{eq} = a_1 w' \tau_{relax}$.

Overall, the stochastic model seems to reasonably represent the scale dependence of the supersaturation fluctuations. At small scales (i.e., L = 0.064 and 0.128 ~~m),~~ m), presence of droplets has a small impact on supersaturation fluctuations and the scale dependence is approximately as for the case without droplets as shown by the red line. At those scales, DNS seems to underestimate supersaturation fluctuations. Arguably, this is because of the small Reynolds number and thus a poor separation between forcing and dissipation scales. For scaled-up DNS, the stochastic model underestimates supersaturation fluctuations and the spread between the scaled-up DNS and stochastic model increases with the increase of the spatial scale. At the largest scales considered (i.e., L = 12.8, 25.6 and 64 m), the quasi-equilibrium supersaturation provides a good estimate of the supersaturation fluctuations as shown by the blue line. This agrees with the argument put forward in Grabowski and Wang (2013, see discussion in the last paragraph of section 3.4).

There are a few reasons for the discrepancy between the stochastic model and scaled-up DNS. First, stochastic model uses TKE obtained from the scaled-up DNS. However, scaled-up DNS features reduced TKE when compared to the real DNS as documented in section 3. Allowing more TKE on input for the stochastic model would shift the stochastic model results upwards, that is, closer to the scaled-up DNS. But increasing the Reynolds number in the scaled-up DNS increases σ_S as well. This is illustrated by three data points for L = 64m scaled-up DNS with 128^3, 256^3, and 512^3 simulations. Second, scaled-up DNS excludes scales of motion that are smaller than the scaled-up Kolmogorov microscale. For instance, for L = 25.6m and 256^3 simulation, the scaled-up Kolmogorov microscale is 0.1 m. Hence, scales of motion between 10 cm and 1 mm are excluded when compared to the real DNS. Arguably, these small-scale motions in real DNS can affect supersaturation fluctuations and reduce σ_S. Such an argument seems to be contradicted by the results with L = 64m because σ_S increases, not decreases, between 128^3, 256^3, and 512^3 scaled-up DNS. ~~.~~However, it is unclear if the increase of σ_S with the further increase of the Reynolds number (i.e., the number of grid points) continues once real DNS is approached with further increase of the simulation towards the 64000^3 real DNS limit. Finally, one might argue that assuming a Gaussian vertical velocity distribution in (11) is an increasingly poor assumption with the increase of the domain size. Higher frequency of large vertical velocity perturbations (i.e., above the Gaussian distribution) should result in larger supersaturation fluctuations.

6 Discussion and Conclusions

This study presents a novel modelling methodology that extends the traditional technique to simulate homogeneous isotropic turbulence, the direct numerical simulation (DNS). DNS is typically used for small-scale simulations applying grid lengths of the order of the Kolmogorov microscale, that is, about a millimeter for typical levels of atmospheric turbulence. Such a choice

allows proper dissipation of the turbulent kinetic energy (TKE) that cascades through the inertial range from large scales where TKE is introduced. To reach domain sizes of about 1 cubic meter and beyond with a grid length of about 1 mm requires tremendous computation resources, with simulations featuring spatial scales of tens of meters and beyond (i.e., volumes of 1,000s of cubic meters and larger) impossible for a foreseeable future. At the same time, one should expect that the largest turbulent eddies affect the diffusional growth of cloud droplets most significantly because such eddies feature the largest and the longest-lasting vertical velocity and supersaturation perturbations. It is thus desirable to have a modelling approach similar to the traditional DNS, but capable of reaching significantly larger spatial scales, say, tens and hundreds of meters.

This paper presents such an approach. The key idea is simple: rather than assuming that the dynamic model grid length is the Kolmogorov microscale η, we start with the DNS domain size ~~L~~ L and adjust the Kolmogorov microscale given the computational resources. For instance, for ~~L~~ L = 100 m and 512^3 simulation, $\eta \simeq 0.2$ m. However, to use a traditional DNS code one needs to allow proper TKE dissipation (as well as the scalar variance removal) at the smallest scales. It follows that the molecular transport coefficients from the traditional DNS need to be properly increased. The Reynolds number similarity is applied to develop a proper scaling, see (8) and (10). The modified modelling approach is referred to as the scaled-up DNS. Section 2 presents numerical simulations applying the spectral DNS code that document robustness of the scaled-up DNS technique. We show that DNS and scaled-up DNS simulations with the same Reynolds number (i.e., the same L/η ratio) forced to maintain the same TKE dissipation rate feature the expected TKE scaling (4). However, when real DNSs are replaced by scaled-up DNSs with a reduced Reynolds number (i.e., keeping ~~L~~ L the same and increasing η), a small fraction of the TKE is lost. As one might expect, the closer scaled-up DNS L/η ratio is to the real DNS ratio, the closer are TKEs between the two simulations (cf. Fig. 3 and its discussion).

For simulations targeting growth of cloud droplets in homogeneous isotropic turbulence, the scaled-up DNS faces the problem of a large number of droplets that need to be followed inside the computational domain. For instance, a cube volume with $L = 100$ m and droplet concentration of $100\ \mathrm{cm}^{-3}$ contains about 10^{14} droplets. Following all of them is computationally not possible. We apply a method already used in Lanotte et al. (2009) and in Li et al. (2019) and referred to as the super-droplet method in Shima et al. (2009). A super-droplet represents an ensemble of real droplets with the same radius; position and velocity of each super-droplet is predicted in the same way as for the real droplet. The number of real droplets represented by a single super-droplet is referred to as the multiplicity attribute (Shima et al., 2009). The multiplicity attribute is included in the condensation rate calculations. The super-droplet method ~~was~~ is first tested in real DNS and then implemented in the scaled-up DNS. Real DNS with $L = 0.064$ m (64^3 grid points) and $L = 0.128$ m (128^3 grid points) and with different multiplicity parameters give consistent results even if the multiplicity parameter results in a super-droplet present in one out of a few dozen grid boxes. For scaled-up DNS (and likely for the real DNS as well), there is an upper limit for the multiplicity parameter before supersaturation fluctuations start deviating from the value obtained with lower multiplicities. Scaled-up DNSs presented here suggest that there should be at least a few super-droplets per grid box for approximately converged solutions. Such an estimate agrees with the result of idealized laboratory cloud chamber simulations reported in section 4 of Grabowski (2020).

The scaled-up DNSs starting from unimodal droplet distribution with no mean ascent (i.e., as in Lanotte et al., 2009; Li et al., 2019) extend the validity of the scaling relationship obtained previously in either DNS simulations (e.g., Li et al., 2019;

Saito et al., 2019) or in stochastic model simulations (Sardina et al., 2015). The scaling implies that the standard deviation of the droplet radius squared increases in time t as $t^{1/2}$. DNS results of Li et al. (2019) show that the evolution of the droplet distribution spread depends on the Reynolds number (i.e., the DNS domain) and is insensitive to the TKE dissipation rate. The Reynolds number dependence is consistent with the eddy hopping argument (see section 3.5 in Grabowski and Wang, 2013) and the dominating impact of the largest eddies for the spread of the droplet size distribution. The standard deviation of the droplet radius squared increases in our simulations as $t^{1/2}$ as well, with systematically larger values for larger scaled-up DNS domains as shown in Fig. 9.

Finally, we also consider supersaturation fluctuations in a simple stochastic model of a droplet ensemble (Grabowski and Abade, 2017) and compare the fluctuations to those simulated in DNS and scaled-up DNS. The key advantage of the stochastic model is that its computational cost is just a tiny fraction of a DNS simulation. The simulation time of the stochastic model is typically a mere few seconds on a laptop computer comparing to hours of wall clock time of high performance massively parallel computer applied in DNS and scaled-up DNS simulations. As argued in Grabowski and Abade (2017), the stochastic model provides a simple and physically appealing approach to multiscale large-eddy simulation of a cloud applying Lagrangian particle-based microphysics (see Grabowski et al., 2019, for a discussion of the Lagrangian microphysics).

The scaled-up DNS methodology presented here was developed with diffusional growth of cloud droplets in mind. The next step can be to apply this approach in a rising parcel simulations as in Grabowski and Abade (2017) to understand the impact of turbulence on the cloud condensation nuclei activation/de-activation near the cloud base (see discussion in Abade et al., 2018). One can argue that scale-dependent supersaturation fluctuations can induce significant droplet concentration heterogeneities at the cloud base that arguably affect droplet growth aloft. One may also consider applying the scaled-up DNS to the problem of droplet collisions. However, since collisions between cloud droplets take place at sub-Kolmogorov scales, applying scaled-up DNS for turbulent collisions is not straightforward. Finally, one can also consider applying scaled-up DNS in simulations of the turbulent entrainment and mixing similar to those discussed in Kumar et al. (2018) and Paoli and Shariff (2009). Such simulations would extend the still relatively small-domain DNS simulations into domain sizes comparable to the large entraining eddies in natural cumuli as discussed in Grabowski and Clark (1993). We hope to explore some of these research directions in the future.

Data availability. Data supporting the study is available at https://www.tropmet.res.in/~majfiles/Lois-Thomas/ACPfiles.zip and can be accessed upon request.

Author contributions. LT ran simulations and performed data analysis under the supervision of GW and BK. GW and LT developed the idea of scaled-up DNS. All three authors were involved in preparing the manuscript. BK helped in accessing and using HPC system of IITM that was used to run the DNS.

Competing interests. The authors declare that they have no conflict of interest.

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