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Dear Professor Szczytko

Thank you for inviting me to review “Modeling particle growth by condensation and deposition in turbulent mixed-phase cloud volumes” by Daniel G. Albuquerque.

The work explores if unresolved turbulent fluctuations interact with mixed-phase cloud microphysics by controlling supersaturation to produce more liquid phase than mean-field approaches. To achieve this a super-particle approach is taken where the local environmental properties of the particles (e.g. temperature and humidity) are prognosed and subject to mixing with the environmental mean. Some idealised cases are presented to demonstrate the differences.

Chapter 1 sets the scene and provides motivation, chapter 2 introduces an adiabatic homogeneous parcel cloud model. Chapter 3 outlines a highly simplified bulk model with temperature dependent thermodynamic phase fraction. Chapter 4 describes the Lagrangian microphysics that will act as the control against the modified version with the mixing included. While chapter 5 builds in the mixing terms for heat and moisture and explores the impact of these new equations in a variety of settings. The impact on mixed-phase evolution as well as droplet activation is described.

Chapters 1-4 demonstrate the candidates grasp of the fundamental theoretical components of the subject of the thesis covering the development of a parcel cloud model. The novel approach undertaken for this thesis is to augment the microphysical rate equations by a mixing term that replaces the mean field environmental variables with a time varying one. This work which forms the main demonstration of the new approach adopted here has satisfied peer review and already been published in the Quarterly Journal of the Royal Meteorological Society. This demonstrates that the candidate can pursue original scientific research. The extension of the microphysical equations to include turbulent mixing could be used in an LES setting as the basis of a more inexpensive parametrization to capture the effect of the turbulent fluctuations on mixed-phase cloud evolution. Improving the representation of this class of clouds is of importance for constraining climate prediction and improving NWP.

In conclusion, both criteria for the PhD have been met - the demonstration of theoretical knowledge and the original scientific research and therefore the degree of PhD can be awarded following a successful viva.

Although not mandatory, I have the following comments that the candidate may wish to consider.

It would have been very nice to see this approach implemented into an LES model. This would provide a way to demonstrate what the impacts of feedbacks from the cloud evolution were and potentially allow comparisons with observations to test the approach in a more realistic setting.

It was not quite clear to me how this approach could be scaled up for use in NWP and climate models. At its heart it is super particles that are not yet possible to include in climate simulations. Perhaps more could be discussed about this? A way forward might be to implement this in an LES as suggested above and then parameterise the LES behaviour with environmental variables that are readily available in climate models?

p.7 (end of first para). The parcel model has many simplifications (no sedimentation, riming etc). What do you think the impact of neglecting these additional processes is? A step before full implementation in LES might have been to introduce a simplified kinematic 1-D framework to probe the importance or not of these other processes.

p9 (beginning of 2nd para). I think these papers talked about the conditions for parcels moving under constant or oscillating vertical velocities - and so the criteria are valid for those situations. Later work did demonstrate that consideration of turbulence even when mean ascent velocities were zero would also be able to maintain liquid in the presence of ice (e.g. Field et al. QJRMS 2014. Tested against 10x10x5m les box output. This provides subgrid heterogeneity compared to 100m box.)

FitzRoy Road, Exeter
Devon, EX1 3PB
United Kingdom

enquiries@metoffice.gov.uk
www.metoffice.gov.uk

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p12 eq 2.4. was enthalpy, h, defined?

p20. Figure 3.1 seems to have different starting conditions to those in the text? To start at T=269K and ~80% RH would need a qv=23g/kg
Looks like theta~299K~T=299K and qv=15g/kg would give ~70% RH?

p22. Figure 3.2 - 80% liquid at -30C. That seems unusual? Must be for a highly specific case or regime?

p28. I could not see Req on fig 4.1?

p38. In bulk scheme... But in the bulk scheme the phase is determined totally by a temperature dependent function - and condensation is saturation adjustment so while the grid box mean is subsaturated you could view the droplet population as being at water saturation (the saturation adjustment) and the ice population being subsaturated with respect to liquid to give the grid box average.

p45. Yes, for the inertial range we have some theory that allows us to describe the underlying distributions of humidity. For larger mesoscales outside of the inertial range we do not. But if we could produce a theory that could take account of mesoscale features such as fronts and MCS's then perhaps we could extend to larger scales?

p47. Eq 5.3 Are the <mean> values varying for the simulation or fixed?

p48. para 3. Tau_m - why is this not relatable to epsilon and the scale of the box (Delta) ?

p49. From Rodean 1997, Tau_decorrelation=2*sigma^2/(epsilon*C0). Here C0 is the lagrangian structure function. Typically has values of ~10 but is regime(flow) dependent. C_tau~2/C0 ~0.2? Tau_decorelation is how fast the turbulence forgets the velocity. And then the scalar-to-velocity factor is on top of this? But here have the two been combined? C_tau of 0.1-10 is used later so typical values are probably covered. There is a literature of estimates of C0 and scalar-to-velocity values that could be used for the sensitivity testing. The upper end of this range (10) may be unrealistic?

p50. Could envisage the reflection as representing the exchange of a similar distribution of particles at the upper and lower boundaries with identical parcels there?

p51. For the third group with pre-existing ice it would have been nice to demonstrate getting the same as published results (e.g. Korolev) for the same cases as a bench-mark of the model used here.

p52. Table 5.1. It would be good to add a column that indicates sigma for the turbulence cases so that we can gauge the relative strengths of the turbulence to the imposed mean velocity. It looks like 0.5m/s for the epsilon=1e-3 case. So the mean vertical velocity is similar or larger than the turbulent fluctuations velocity. Could also add a column for Tau_mix. Looks like for the range of sensitivities (C_tau=0.1,10, epsilon=1e-3, 1e-4) that Tau_mix will vary from 20s to ~1hour.

p52. I am probably missing something here. eq. 5.17 and 5.18 are conditional so it looks like they become undefined for purely liquid or purely ice cases. If so, then will it still be possible to use the <SI>=<SI>I assumption to get 5.19? It is clear that the mean supersaturation with respect to ice still has a value even if there is no ice present and vice versa for liquid.

p53. I may have missed it - what C_tau values was chosen for figure 5.4?

p60. Figure 5.7 - Is this just liquid only? Is ice also competing for vapour? If not, it would be interesting to see the impact of ice dampening the effect of the turbulence.Interesting that for these stochastic activation events even though the droplets are modifying the mean vapour field the first droplets are not quenching later ones. This has possible implications for geoengineering marine cloud brightening modelling?

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Devon, EX1 3PB
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enquiries@metoffice.gov.uk
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How do these results compare to standard widely used parameterisation such as Abdul-Razak and Ghan?

p63. Figure 5.8 (and figure 5.12) The C_tau values of 1 and 10 imply that the decorrelation timescale is very long. For C_tau=10 it is nearly as long as the oscillation timescale. Is this why the results seem insensitive to the choice of this value.
C_tau=1 and C_tau=10 will be very similar because of the reflection assumption that limits the vertical extent of turbulent fluctuations.

p68. There is a rich literature about homogeneous and inhomogeneous mixing and evaporation of droplets. This would be fruitful vein to explore with this setup.

p69 - And if you were to allow the ice to fall out (so that the sink of vapour to ice was able to reach a steady state) then you could keep generating new ice and sustaining liquid for as long as INP were made available.

p79. I think the statement about Furtado et al. is not quite right here. The grid box mean vertical velocity in climate models is ~0m/s. Any persistent supercooled liquid is supported only by the action of the subgrid turbulence.

Yours sincerely

Paul Field
Met Office Science Fellow and University of Leeds Professor of Climate.

FitzRoy Road, Exeter
Devon, EX1 3PB
United Kingdom

enquiries@metoffice.gov.uk
www.metoffice.gov.uk