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Interactive comment on "An investigation into the performance of three cloud droplet activation parameterisations" *by* E. Simpson et al.

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Response to Comments by Anonymous Referee 1

p. 1319, lines 13-20. Another relevant reference here is Ghan, S. J., G. Guzman, and H. Abdul-Razzak, 1998: Competition between sea-salt and sulfate particles as cloud condensation nuclei. J. Atmos. Sci., 55, 3340-3347.

This comment is referring to the competition between large CCN and small CCN. The suggested paper has now been included as a reference in the manuscript:

p. 1319, lines 16-17. 'which results in fewer ''smaller" particles activating (e.g. Ghan et al, 1998 and Sander, 1999).'

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p. 1324, lines 20-26. While it is true that assuming the aerosols have an infinite amount of time to activate is unrealistic for this case, is the large aerosol case realistic? Are there measure size distributions in which the number distribution is dominated by a 1.5 micron aerosol mode? I suspect not. If not, then a more realistic case should be chosen, or the conclusion here should be tempered. The point could certainly be made with a 1 micron aerosol mode, or perhaps even smaller.

The referee's point is valid that 1.5 microns is large for an aerosol mode. This was chosen to illustrate the point clearly. However, the effect still holds for 1 micron aerosols. Figure 1 has been redone with a 1 micron aerosol mode and a concentration of 500cm-3 (see below). We will include this figure in the revised manuscript and make the following change to the text:

p. 1324, line 5. 'and large aerosol median diameter (1000nm, the "large aerosol" case).'

p. 1328, lines 6-7. You've made the point about the infinite effective simulation time. Could you offer suggestions on how it might be overcome? I thought FN had a treatment of kinetic limitations.

Referee 2 also raised the point about better defining infinite effective simulation time, I will address this here.

The referee is correct in their assertion that FN includes the treatment of kinetic limitations. The infinite effective simulation time is a different concept as will now be described. The parameterisations calculate the maximum possible supersaturation given values for temperature, pressure, updraft velocity and aerosol characteristics regardless of the altitude that the parcel needs to rise to achieve this supersatuation. Using this maximum supersaturation the fraction of aerosol that can activate is calculated. The altitude of the maximum supersaturation may, in fact, occur above cloud top and therefore in reality no aerosol should activate as the parcel of air would have stopped

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rising before maximum supersaturation is reached. The parcel model takes this into account as a runtime is set, limiting the height to which the parcel can rise. In some of the cases, the runtime required to activate droplets resulted in the cloud base being deeper than the troposphere, which clearly is unrealistic.

Therefore to overcome this problem parameterisations need to include the altitude where cloud top occurs in addition to the altitude that the drops activate. If the altitude where, mathematically, drops activate is above cloud top there should be no activated drops. It is not immediately obvious how the current parameterisation could be modified to include this effect, but one approach could be to use empirical relationships from cloud parcel models to give the altitude that the drops activate.

Referee 2 also raised this point, but pointed to p1318, line 13 in the abstract. To clarify, we will alter the manuscript to read:

p. 1318, line 13. 'This problem arises in the parameterisations because it is assumed that a parcel of air rises to the altitude where maximum supersaturation occurs, regardless of whether this altitude is above the cloud-top. Such behaviour is problematic because, in some cases, large aerosol can completely suppress the activation of drops.'

Response to Comments by Anonymous Referee 2

Page 1318, Line 8. Ghan et al. (2011) performed systematic comparison of several parameterisations. Other works cited by the authors have done so as well, Thus the statement that this the "first systematic evaluation" is unsupported.

The referee is referring to our original statement in the abstract, where we said that this is the first systematic evaluation of these 3 parameterisations. Indeed Ghan et al. did evaluate several of these schemes over a smaller parameter space; hence we have change line 8 to read:

p. 1318, line 8. 'We present a detailed systematic evaluation of three schemes'.

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Page 1318, Line 12 and below. ”Large” and ”small” are too vague. Please be more specific on what specific values these statements refer too. Also in a sentence explain better what you mean by infinite time and how it affects the performance of the parameterisations.

The size ranges for ”large” and ”small” aerosol have now been defined in the text. We have changed p1318, line 11 to read:

p. 1318, line 11. ’when the aerosol particle ”median diameter” is large (between 250nm and 2000nm) ’

and p 1318, line 14 to read:

p. 1318, line 14. ’cases when the ”median diameter” is small (between 5nm and 250nm)’

A more detailed explanation for infinite effective simulation time has been included; please see Referee comment response p. 1318, lines 6-7.

Page 1318, Line 18. Since the parameterisations only differ in the way they approximate the maximum supersaturation, this statement is equivalent to say that this is due to difference in the parameterisations. Please be more specific.

We have changed the original statement in the abstract to read:

p1318, line 18: ‘methods used by the parameterisations to approximate the sink of water vapour’

Furthermore a detailed explanation of the differences between the parameterisations has been included in Section 2.1:

’ARG approximates the maximum supersaturation by assuming all particles start at their equilibrium size and then grow further depending on the supersaturation. FN splits the population of aerosol into two separate groups; those particles that are small and are therefore assumed to start at their equilibrium size and then grow further depending

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on the supersaturation, (similar to ARG) and those particles that are large and take time to grow to their equilibrium size before growing further. This method takes into account kinetic limitations to the growth of larger particles. ‘

Page 1320, Line 14. Correct ”performance”.

Correction made.

p. 1320, line 14. ’performance’

Page 1320, Line 24. Correct ”numerical”.

Correction made.

p. 1320, line 24. ’numerical’

Page 1320, Line 26. Better say ”benchmark for comparison” instead of ”ground truth”.

Correction made.

p. 1320, line 26. ’taken as the benchmark for comparison’

Page 1322, Lines 3-5. Say that ARG is written in terms of dimensionless parameters.

Change made.

p. 1322, line 3-4. ’ ARG is written in terms of dimensionless parameters to account for the errors made by simplifying the droplet growth rate’

Page 1322, Line 19. define dP/dt.

A definition of dP/dt has been added.

p. 1322, line 19. ’the rate of change of pressure with respect to time, dP/dt=-[P/(TRa)]gw,’

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Page 1324, Line 18. Correct ”artifact”.

Correction made.

p. 1324, Line 18. ’artifact’

Page 1325, Section 3.2. This description seems somehow superficial. The differences may be understood in terms of the differences in the assumptions behind each parameterisations. It also looks like the parameterisations fail most of the time and except for some FN GCCN cases, none is able to reproduce the parcel model for d > 100nm. Is this contrary to what is presented in other works?

Here, we are merely trying to highlight what the difference between the parameterisations and the parcel model are. The differences between the parameterisations are certainly due to the differences in the assumptions made by each parameterisation. These differences in approach are now described more thoroughly in Section 2.1 (see reply to comment above).

The results presented in our paper cover a much wider range of aerosol size distributions than those used in previous evaluations of the parameterisations including those presented by ARG, FN and Barahona et al, 2010; however, they are consistent with those previous results.

Page 1326, Line 8. Figure S8 provides valuable information and maybe should not be supplemental. Similarly for Figure S6 later on.

Thanks for pointing this out, we agree. Both figures, S8 and S6, are now included in the manuscript.

Page 1326, Line 10-15. Can this be understood in terms of the approximations made for each parameterisation?

Here the referee is referring to our description of the results of the dual-mode experiments where ARG and FN GCCN underestimate the fraction of activated drops and

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FN highly overestimates the fraction of activated drops.

As explained above in response to the point in the abstract, the ARG parameterisation assumes all aerosol particles start at their equilibrium size, which is not true for the larger particles. Hence, this formulation would tend to over predict the sink of water vapour, and hence underestimate the activated fraction – this is consistent with our findings.

The FN parameterisation calculates the maximum supersaturation by splitting the population of aerosol into two groups; those that are free from kinetic limitations to growth and those where kinetic limitations dominate. Spart is estimated as the division in an aerosol population between these two groups. For particles that are free from kinetic limitations their contribution to water vapour depletion is calculated assuming they start at their equilibrium size, similar to ARG. For particles where kinetic limitations dominate their contribution to water vapour depletion is calculated including the water vapour they uptake in order to reach their equilibrium size. In theory this should improve the comparison with the parcel model; however, Spart is estimated using an empirical function derived from parcel model simulations over a limited range of aerosol characteristics and it would appear that this function is not appropriate for all aerosol distribution tested here.

Page 26, Line 16. Why does the correction in FN GCCN work well in this case but it does not in the single-mode experiment?

It is unknown at this point why the correction in FN GCCN works well in the bimodal case but not in the monomal case. Here we have evaluated and reported on the results from both a bimodal and a monomodal case. It should be noted that an evaluation on the performance of FN GCCN on a monomodal case has not been seen in the literature previously, and was not presented in Barahona et al, 2010.

Page 1327, Section 3.4. It does not seem completely random that all the monomodal cases result in overestimation whereas all the bimodal cases in underestimation. What

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is the origin of these systematic differences?

Thanks for pointing this out. Indeed it is not random. There are two effects: (i) where relatively large aerosol particles (d> 250nm) take longer to grow to activation sizes than smaller particles, and almost activate but not quite; and (ii) where very large aerosol particles are very far from the activation size and do not activate.

The poor agreement in the monomodal cases is where the median aerosol particle diameter is > 250nm and you are correct that the parameterisations tend to overestimate here. This is because scenario (i) occurs: the growth of the large aerosols is inertially limited and they take too long to grow to sizes where they can activate as cloud drops. The effect is captured well in the parcel model but not in the parameterisations: hence they overestimate the activated fraction.

In the bimodal cases, scenario (ii) occurs: ARG underestimates the activated fraction because it hugely overestimates the vapour sink due to assuming all aerosol particles start at their equilibrium size. Since the bimodal cases contain a mode with a very large median diameter this vapour sink has a significant effect on the results.

In the monodal cases where the median aerosol diameters are >250nm the parameterisations tend to overestimate the fraction of activated drops. This is because the growth of these aerosol particles should be inertially limited. FN GCCN also overestimates the fraction of activated drops in the monodal cases for sizes >250nm as it does not work well in monodal cases. In the bimodal cases ARG underestimates the fraction of activated drops as it overestimates the vapour sink by assuming all particles start at their equilibrium size.

Page 1327, Section 3.5. Since the FN GCCN parameterisation performs better, the authors should recommend including the effects of unactivated large particles in cloud modes.

Thanks for pointing this out. This recommendation has now been made in the Conclu-

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sions section of the manuscript.

p. 1328, line 9. 'Due to the substantial improvement that the Barahona et al, 2010 amendment makes to the FN parameterisation, we recommend that the effects of large unactivated particles on the maximum supersaturation be included in cloud models.'

Interactive comment on Geosci. Model Dev. Discuss., 7, 1317, 2014.