Aerosol-fog interaction and the transition to well-mixed radiation fog: Response to reviewer 1

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We thank the reviewer for their thorough review, and will update the manuscript accordingly. We have provided detailed responses to all points below, and would welcome any further comments the reviewer may have whilst the discussion phase is still open.

1. Demonstrate the usefulness of using 3 types of models (LES - NWP - climate model) : Previous studies on microphysical processes were done with NWP or 1D models or were the results of field experiments measurements. Please justify the use of LES to demonstrate the impact of aerosol on fog. What are the added values of LES study with respect to 1D model? This point is essential for the publication of this work.

The ultimate purpose of this work is to improve NWP or climate simulation of fog events. To do this, we have chosen two sources of information to aid our understanding - observations and LES. The main reason we have done this is because neither source alone can provide all of the information we need. The observations are obviously closest to the truth, but there are gaps in the observational dataset, both in terms of quantities measured, and spatial/temporal representivity of the data. Therefore, to assist with the interpretation and understanding of the observations, we have supplemented them with LES data.

The main information we require from the LES to support our conclusions is the downwelling longwave radiation from the period where the radiometer was frozen (Fig. 6). The partitioning between hydrated aerosol and activated cloud droplets comprising the size spectra in Fig. 9 is an additional piece of interesting information provided by the LES, for which we do not have observations. Finally, the mean profiles from the LES are closer to what we would expect an NWP or climate model to simulate. The observations are point samples and can be subject to considerable variability (e.g. the difference in consecutive tethered balloon profiles shown in Fig. 5). The ability to explore this variability in the LES allows us to determine whether changes seen in the observations are due to evolution of the fog layer or natural variability.

We feel that LES (as opposed to a 1D model) is the best tool for this job, because it is least reliant on parametrizations. The key focus of this paper is the coupling between aerosol processes and turbulence. The LES is able to resolve the important scales of turbulence, simulating the variability that exists within a fog layer forming over a spatially homogeneous terrain. We can therefore couple the microphysics directly to the turbulent flow, allowing for the activation in updrafts and supersaturated regions, without having to rely on sub-grid estimates of turbulent quantities from a parametrization. A 1D model would be providing these from a turbulence parametrization, which would itself, most likely have been derived from LES simulations, and therefore we are avoiding this intermediate step.

We will include this discussion and motivation in the revised paper.

The impact of cloud droplet number with NWP should be evaluated in a statistical way : How has this modification improved the fog forecast? This evaluation could be done with LANFEX data which provide many cases of "stably stratified fog" cases. Your conclusions are too speculative and need to be demonstrated.

We had not initially wanted to present a detailed analysis of the NWP model verification, as the key focus of the paper is a process modelling study which should be applicable to any NWP system. We merely wanted to illustrate with Figures 11 and 12 that lessons learned from the process modelling study do provide useful benefit in an NWP modelling system. We have included in Figure 1 and 2 the statistical results of the month-long trial of the full data assimilation and forecast system. These plots incorporate all data from UK land stations to verify the forecasts (4 per-day at 00, 06, 12 and 18 UTC) for their 36 hour forecast range. There are therefore many more forecasts and observations in this data-set than in, for example, the LANFEX archive.

Figure 1 shows categorical statistics for observed and forecast visibilities of 200 m and 1 km. Following Mason (2003), we define a 2×2 contingency table such that a is the number of hits, b is the number of false alarms, c is the number of misses and d is the number of correct negatives. We then determine the Equitable Threat Score (ETS) as $(a - X)/(a - X + b + c)$ where $X = (a + b)(a + c)/(a + b + c + d)$, Frequency Bias as $(a + b)/(a + c)$, Probability of Detection as $a/(a + c)$ and Probability of False Detection as $b/(b + d)$. Perfect scores are 1 for the ETS, Frequency Bias and Probability of Detection, and 0 for the Probability of False Detection. As shown, the "Control" model was over-forecasting low-visibility events, with a Frequency Bias > 1 and a Probability of False Detection > 0. Including the modified drop taper has clearly improved both of these metrics. Importantly, it has done this without significantly degrading the Probability of Detection, which remains largely unchanged, and therefore the ETS is improved. These results are consistent with the main results from LANFEX IOP1, and the case study from this trial period presented in Fig. 11. The model produces fog which is

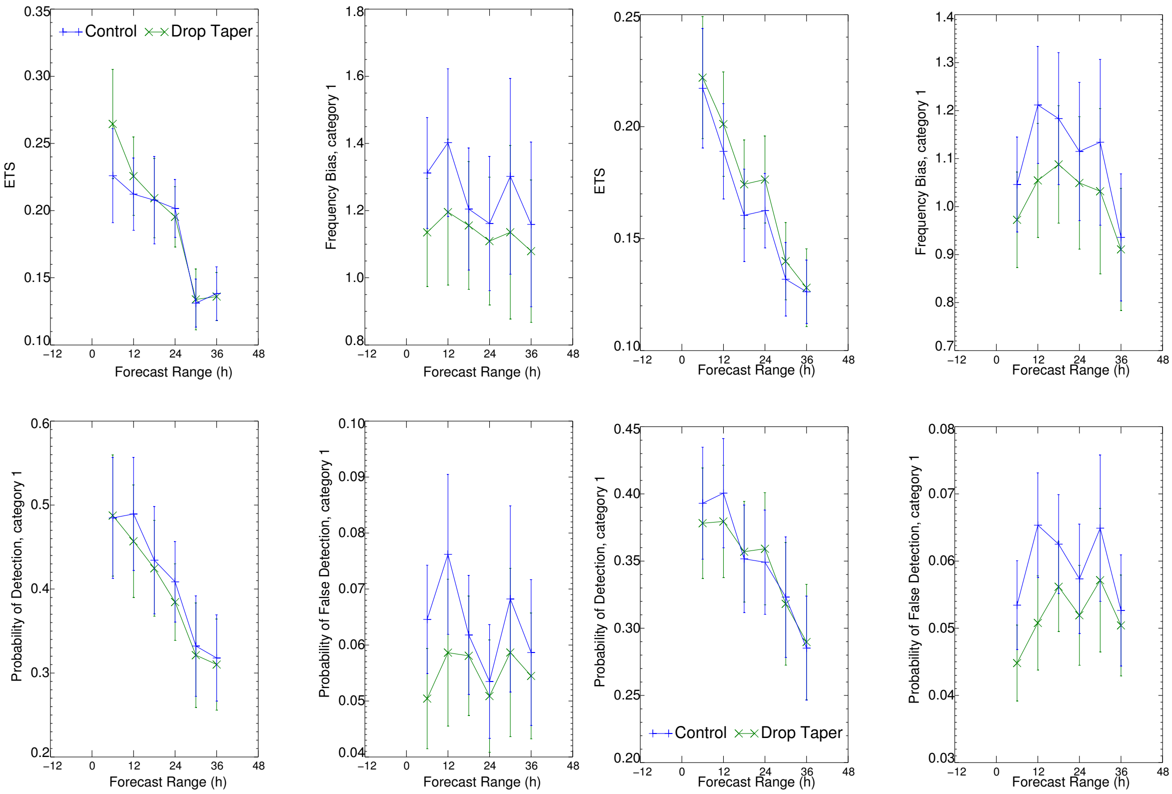

Figure 1: NWP model verification of visibility from 05/02/2015 to 05/03/2015 utilising all observations and forecasts (4 per-day) over UK land areas. Panels show the Equitable Threat Score (ETS), Frequency bias, Probability of Detection and Probability of False Detection for visibility thresholds of 200 m (left) and 1 km (right), for experiments "Control" and "Drop Taper".

too thick, too fast, and tends to persist for too long, i.e. it over-forecasts. By improving the droplet numbers, this behaviour is improved, and can be seen in the statistical analysis.

Fig 2 shows the mean error and root-mean-squared (RMS) error for visibility and screen-level temperature forecasts. The results show improved RMS errors when including the modified droplet taper, and a slightly degraded mean error, which is again consistent with the other results presented. The effect of the revised drop taper is to reduce low visibility events, which will lead to an increase in mean visibility. When the model already has a high bias in mean visibility, this will inevitably get worse. However, the high bias in mean visibility comes mainly from high visibility events (visibilities $>$ 10 km), and therefore by over-forecasting low visibility events we were compensating for this error in a mean sense. We have now removed one half of this compensating error.

The screen temperature shows improvements to both the mean and RMS errors. The improvement to the mean error tends to be removing a cold bias in the model, showing that the most significant effects on temperature verification are from allowing the fog to dissipate earlier during the morning period and allowing the model to warm up faster during the day.

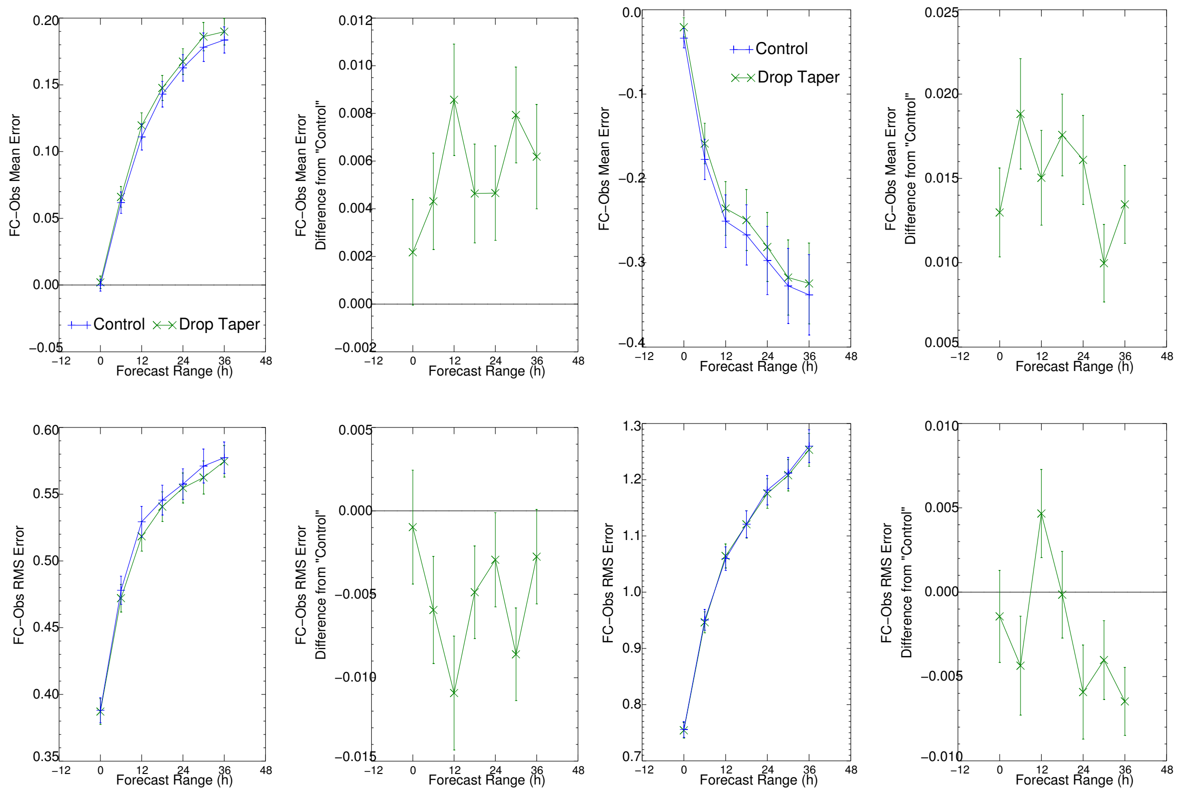

Figure 2: NWP model verification of visibility (left) and screen-level temperature (right) from 05/02/2015 to 05/03/2015 utilising all observations and forecasts (4 per-day) over UK land areas. Panels show Mean Error and Root-Mean-Squared Error for experiments "Control" and "Drop Taper", and the difference of Drop Taper from Control.

In the revised manuscript, we will replace Figure 12 with some of this statistical evidence, to add further support to our results. The manuscript will then contain a detailed process modelling study, an independent example case, and statistical evidence from many cases, all showing the same behaviour and model improvements.

2. Validate the microphysical parameterization : Your work is based on the use of a specific microphysical parametrization. In my opinion, the main characteristics of this parametrization should be detailed in the revised manuscript. Particularly, the dependence of activation parametrization with respect to radiative cooling and turbulent processes should be explained. This microphysical parametrization should also be validated for fog cases, with comparison with observations from LANFEX for example.

We assume here you are referring to the microphysical parametrization in the LES model. In UCLALES-SALSA the aerosol size distribution is represented with a sectional method using 10 size classes (bins), and water condensation on particles in different bins is calculated by numerically solving the condensation equation at every time step in every grid point. Thus we are able to explicitly simulate how the radiative cooling (which increases water saturation ratio S) and turbulence (updrafts enhance S and downdrafts decrease S) affect the water saturation ratio and how this affects the size of aerosol particles. As soon as the first particles grow above their critical size given by Kohler theory, they will be counted as cloud/fog droplets and technically transferred into separate cloud droplet size distribution. Water condensation on droplets is also solved through the condensation equation, and condensation on both aerosol particles and droplets is affecting the water amount in the gas phase. Thus we do not employ any such parameterization for droplet activation which are traditionally used in large eddy or other atmospheric modelling applications, but instead we simulate the actual supersaturation and growth of aerosol particles into droplets. We have tested with a more detailed cloud parcel model (e.g. Kokkola *et al.*, 2008) that the growth of particles into cloud droplets is solved with a good accuracy in SALSA for the case of adiabatic air parcels with different updraft velocities.

In LANFEX we are missing the aerosol information to validate the microphysical scheme, but supersaturation simulated by UCLALES-SALSA agrees well with measured aerosol activation in the existing field campaigns already cited in the manuscript. We have already provided qualitative comparison of model results against similar observations to LANFEX in Tonttila *et al.* (2017), and detailed analysis of model simulations of radiation fog in Maalick *et al.* (2016). The model was found to capture the main characteristics of radiation fog formation, and to provide similar results to other LES models. Finally, it's worth noting that the performance of the LES against observations for this case (as presented in Figs. 2-10) is very good, and has been achieved without any specific tuning of the LES.

We will make this more clear in the manuscript and add references into the recent literature where the lack of radiative cooling is found problematic for activation parametrizations, and explain how this is avoided in our case.

3. Validate the numerical model used and particularly the frost-dew deposition : The dew and frost deposition play a key role during the formation phase of fog and particularly in the transition to well-mixed radiation fog (eg Guedalia and Bergot 1994). Frost deposition could prevent the formation of dense fog despite radiative cooling. It is necessary to demonstrate that this process is correctly simulated. Otherwise, your modification may simply compensate the errors in the estimate of deposition by the model.

We again assume here you are referring to the LES model. Dew deposition is clearly an important process, and something we plan to study more as part of the LANFEX project, because this is likely to be an area in which all models are deficient. During LANFEX, we have measurements of dew deposition available from the instruments described in Price and Clark (2014), in addition to typical near surface humidity measurements.

Figure 3 shows observations of dew deposition from IOP1, and equivalent simulated quan-

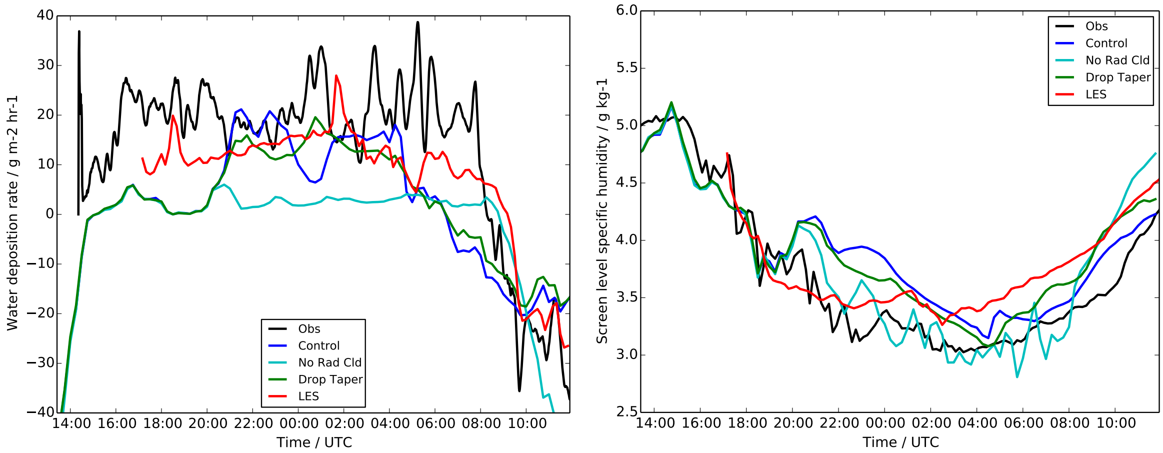

Figure 3: Time series of surface water deposition rate (i.e. dew, left) and screen level (1.5 m) specific humidity (right) from observations (black), LES (red) and UKV experiments: control (blue), radiatively inactive cloud (cyan) and modified droplet number (green)

tities from all the models analysed. During the main period analysed (20 UTC to 04 UTC) the LES (and NWP models) are in reasonable agreement with the observations – they are at the lower end of what the observations are showing, but certainly within the uncertainty of the instrument. During the early stages (before 20 UTC) the models appear to be under-estimating the deposition, and this is likely to be due to hygroscopic absorption, a process not accounted for in the model's surface latent heat flux parametrization, as discussed in Price and Clark (2014). During the latter stages (after 04 UTC), the models again appear to be under-estimating the deposition, and this is due to a compensation between the fog droplet sedimentation, which is still in good agreement with the observations, and the latent heat flux, which is now positive causing the surface to lose water.

As discussed in Guedalia and Bergot (1994), the key effect dew deposition has on the fog lifecycle comes through its modification of the near-surface specific humidity, which is also shown in Figure 3 and profiles at various times are shown in Figure 4. The observations show a reduction in specific humidity from 5 $\mathrm{gkg^{-1}}$ to 3 $\mathrm{gkg^{-1}}$ during the development stages of the fog event, and this is reasonably well simulated by the models. The profiles presented in Fig. 4 show how this reduction in water vapour grows through the lower levels of the atmosphere, up to 80 m by 03.30 UTC. Again, this reduction is well simulated by all of the models, except for "No Rad Cld" which it not a particularly realistic simulation. This reduction in specific humidity is consistent with water transfer into the condensed state (the fog), for which observations show that the water contents are well simulated by the models (Figs 4 and 5a in the paper), and deposition onto the surface (Fig. 3).

Therefore, we are confident that this process is being correctly simulated in the model, and not leading to erroneous fog development. However, this is also clearly an uncertain process requiring further work. Indeed it will interact with the aerosol effects discussed in this paper.

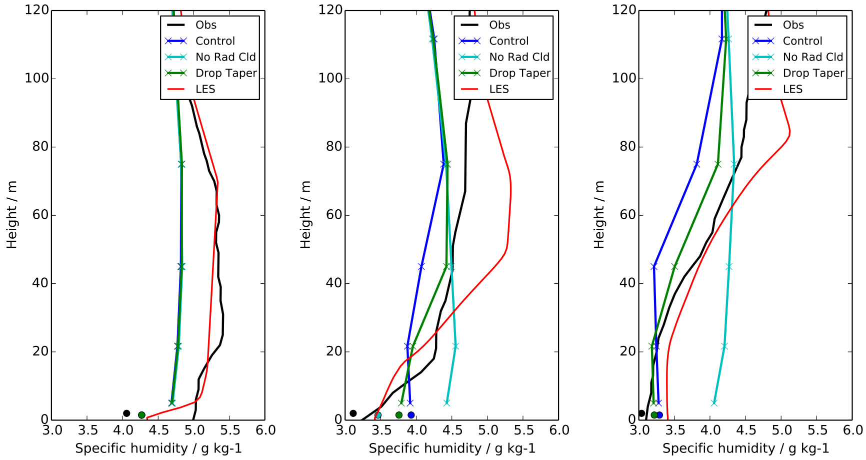

Figure 4: Profiles of specific humidity at 17.30 UTC (left), 22.30 UTC (middle) and 03.30 UTC (right), showing observations (mast: black circles, radiosonde: black line), LES (red) and UKV experiments: control (blue), radiatively inactive cloud (cyan) and modified droplet number (green). Model profiles show model-level data (crosses) and diagnosed screen level data (filled circles).

The droplet sedimentation flux is inversely proportional to the number of fog droplets, and therefore deposition will be high in situations with low fog droplet number, such as IOP1. In situations with higher droplet number, the deposition will be much lower. This is another mechanism (in addition to the radiative mechanism discussed in the paper) which will lead to strong sensitivity of fog processes to aerosol.

We shall add some of this discussion to the revised manuscript, to demonstrate that the dew deposition is realistic in the models, but is another area of uncertainty which requires further study.

It is also absolutely necessary to validate the model used for turbulent processes, radiation and soil-atmosphere exchanges.

As mentioned in response to point 2, Maalick *et al.* (2016) and Tonttila *et al.* (2017) have analysed and validated the LES for radiation fog cases similar to those presented here. The LES model used also has an extensive pedigree for modelling turbulence and the interaction of radiation with other types of clouds (e.g. Stevens *et al.*, 1999, 2005), where it has been shown to be of similar quality to any other commonly used LES code. Finally, the comparison of LES results to turbulence and radiation observations for the case presented (e.g. Figs. 2, 3, 7) is very good. As the surface temperature is specified throughout the simulation, there are no soil-atmosphere exchanges or land surface feedbacks within the LES. This allows us to avoid the large uncertainty in simulation of land surface processes, and choice of initial conditions for soil heat and moisture content, something which was pointed out by Maronga and Bosveld (2017) as one of the main uncertainties in the formation time of fog.

We shall clarify these points in the revised manuscript.

4. Contribution of this study with respect to bibliography : Another shortcomings of the current manuscript is that it does not cite available literature. For example papers from Bott (1991) or Rangognio et al. (2009) have studied in detail the effect of aerosol on radiation fog. It needs to be shown what new results does this work provide compared to those already published. What are the differences in the model, observation and methods used? What are the differences in results found?

We agree that not citing the Bott (1991) paper was an over-site on our part, and this shall be corrected. The Bott (1991) study is indeed very complimentary to our own, demonstrating the effect of aerosol concentrations on the development of fog in a 1D model. Our work has reproduced and analysed in detail the mechanisms for this effect in two independent models (LES and NWP), utilising different parametrizations of microphysics, radiation and turbulence. Furthermore, we have provided observational evidence for this effect in a real case study, and perhaps most importantly have provided a simple method of incorporating this effect in an operational NWP system, demonstrating the benefits of including it on forecast quality. This latter point is key – it is 26 years since the study of Bott (1991) and yet very few operational NWP models include the interaction of aerosols with fog in their systems, despite the obvious importance which is noted by Bott (1991) – "in the morning, the urban fog is not dissipated by the solar radiation". This discussion shall be included in the revised manuscript.

Whilst it is an interesting paper, we are reluctant to cite the study of Rangognio *et al.* (2009) because it was never published, due to errors discovered in the numerical model. It is therefore difficult to fully assess any similarities or differences with our work.

You say in conclusions that "key factors affecting the development of well-mixed fog include :" "(i) the amount of time available for development before sunrise" : the point is well-known and has also be demonstrated by numerous studies. Please cite the current literature and please evaluate your contribution with respect to existing work. "(ii) the speed with which the fog layer can deepen , strongly governed by humidity profile" : please evaluate your contribution for this well-known result.

We don't believe we have claimed to make any contribution to these well known results, we were merely trying to synthesise for the reader all the key processes governing the development of well-mixed fog. We shall clarify this in the revised text.

"(iii) the amount of accumulation and coarse mode aerosol for activation" : this point is in contradiction with the LES study of Maronga and Bosveld (2017) on

a Cabauw case which say in abstract that "the choice of droplet number concentration ... has a high impact on the liquid water content within the fog layer but a rather small effect on its life cycle". Please elaborate.

The comparison with Maronga and Bosveld (2017) is interesting, and certainly something we should discuss in the manuscript. The case presented by Maronga and Bosveld (2017) transitions to well-mixed fog almost instantly in the model - this can be seen in their Fig. 2 where the visibility reduces to 100 m at all vertical levels (2, 10 and 20 m) within 30 minutes, and Fig. 3 where the downwelling longwave radiation rapidly increases to match the upwelling longwave radiation at the time of modelled fog onset. Therefore, their simulation is very similar to our "Control" run. Their sensitivity studies to fog droplet number encompass the range 100-200 cm^{-3}, which is approximately the vertical variability in droplet number shown by our control run (Fig. 5). Our observations and LES show drop numbers much lower than this range (< 50 cm^{-3}), and it is only when we reduce the drop numbers to this value that we achieve a simulation with a slow transition to well-mixed fog. It's also worth noting that the case investigated by Maronga and Bosveld (2017) probably also has a slow transition to well-mixed fog in reality, with the visibility falling to 100 m, at 2 m height, 2 hours before it does at 10 m, and 3 hours before it does at 20 m, in the observations. This is in stark contrast to the model which achieves this vertical development in 30 minutes. It would be interesting to re-run the simulations of Maronga and Bosveld (2017) with a lower droplet concentration, to see if this transition can be improved.

Another point, which we should make clearer, is that the feedback between aerosol and fog development is dependant on longwave scattering in the radiation parametrization, a process which is often ignored in NWP models (Edwards and Slingo, 1996). And indeed if we turn off this process in our NWP model, we do not see such strong sensitivity of the fog development to the aerosol concentration. It may be that non-inclusion of longwave scattering is a reason for reduced sensitivity seen in other studies.

We will add discussion and clarification of these points to the revised manuscript.

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