We thank Reviewer #1 for his/her constructive comments. The main concern raised by the Reviewer was the lack of direct comparison of the results with observations. We will improve upon this to the extent allowed by the fact that comparing the results from the semi-idealized setup of UCLALES-SALSA with aircraft measurements, where conditions vary significantly even during a single flight leg, is not very straightforward. Below, we will list all of the Reviewer comments, followed by our response highlighted in *italics*
–

The changes to the manuscript regarding each comment are highlighted in italics.

1) P2, L10: To help put the present model developments in context, are you able to point out any previous LES models that have developed similar aerosol-cloud couplings? The text only mentions that such aerosol-cloud schemes are sparse.

Aerosol-cloud interactions have been available also in previously published models, but the approach we take to describe the evolution of the aerosol size distribution for both activated and non-activated particles in a bin model is rather unique. To account for this comment, we will add further discussion and references on aerosol-cloud modeling frameworks of similar level of sophistication (e.g. the LIMA scheme; Vié et al., 2016). We will also implement the suggestions by Reviewer #2 concerning the same topic.

We have added this and several other new references to page 2, lines 10-15 of the revised manuscript.

2) P3, L19: The sub-range indices 1a and 2a do not appear on Fig. 1 (only a and b are shown, not 1 and 2). Please check this on the figure and check the later references in this paragraph to 2a and 2b. The labels 1 and 2 are confusing because they do not appear on Fig. 1.

We will modify the figure according to the Reviewers suggestions.

The subrange indices are now indicated in Figure 1.

3) P4, L3-4: One goal of this work is stated as ‘to reproduce the evolution of the aerosol size distribution through cloud processing and wet scavenging by precipitation accurately’. Please consider whether the manuscript would be improved by showing aerosol size distributions. Figure 7 does show the time series of the number concentrations in each bin – would a size distribution figure for hours 0 and 8 be helpful to illustrate the changes? Also please consider showing observed size distributions to improve confidence in the simulations.

Figures showing the simulated size distributions as suggested by the Reviewer will be added. They show that the evolution of the dry as well as the total (activated+non-activated) aerosol size distributions remain consistent and robust with respect to the presented model processes. The initial aerosol conditions used in the model runs are based on the data given by Ackerman et al. 2009, which are somewhat idealized due to the high variability of the aerosol size distributions even within a flight leg.

A new figure (figure 7 in the revised manuscript) shows the aerosol size distributions as suggested by the Reviewer. This figure is discussed on page 12, lines 1-6.

4) P4, L8: ‘defined to be parallel’ – the meaning of this is not quite clear – please clarify.

The “parallel bins” in the text refer to setting the cloud droplet bin edges (according to the dry CCN size) identical to those in the corresponding non-activated aerosol bins. i.e. the bins are defined for same dry particle sizes. We have reiterated this in the manuscript.

The “parallel bins” part is explained more clearly on page 6, lines 10-13 in the revised manuscript.

5) P4, L9-11: ‘ This way, the properties of the aerosol size distribution are preserved upon cloud droplet activation, as well as evaporation of cloud droplet, though subject to the typical uncertainties inherent in the sectional approach’ – please consider rewording this sentence to clarify what is meant by ‘properties’.

We will reword “properties” as the shape of the distribution and number concentration of particles.

This is now included on page 6 lines 13-14.

6) Section 2: Could equations be added to describe the key microphysical processes?

We will add equations for the key processes and also provide some further details about their implementation.

More detailed description of the microphysical processes is found on pages 4-5 in the revised manuscript.

7) P5, L29: Please provide further details about the source for the coagulation kernels.

A reference for the source of the kernels will be added.

References have been added and the equations (3-5) are given on page 4 of the revised manuscript.

8) P5, L30-32: How is the dry size of the particle determined when the drizzle drop evaporates? Please clarify.

Upon evaporation of drizzle, the particle size is obtained by assuming that a single particle per droplet is released. The particle diameter is then the result from dividing the total aerosol mass with the bulk particle density. This will be stated more clearly in the text.

This is now explained in more detail on page 7, lines 31-33.

9) P8, L17: How do you define ‘deeper and more massive shallow convection elements’?

By the deeper elements we refer to the cumulus clouds occasionally arising from about 400 m height (about the decoupling inversion height), supported by the build-up of heat and moisture from the surface. A more detailed description will be given in the manuscript.

This is noted on page 10, lines 26-31 in the revised manuscript.

10) P8, L32-34: In comparing the LEV3 and LEV4 simulations, it would be helpful to have a clearer description of the parameterization of drizzle formation/loss in LEV3 (the default UCLALES configuration). Perhaps this could be added earlier on in the model description.

We will add a more detailed description of these processes for the default UCLALES.

Description of the drizzle formation in the default UCLALES is given on page 8, lines 10-15 of the revised manuscript.

11) Fig 4: Where is LEV3 on panel 4b?

The LEV3 (as in the default UCLALES) does not contain a description for aerosols (apart from the prescribed CCN concentration used to yield the number of cloud droplets). Therefore this cannot be added. The panel 4b serves the purpose of illustrating the abilities of UCLALES-SALSA. We will make a better note of this in the manuscript to avoid confusion.

It is now explicitly stated on page 8, lines 8-9 that the UCLALES does not contain a description for aerosol and thus for scavenging. Moreover, it now says "Figure 4 shows the surface precipitation rate in LEV3 and LEV4 simulations as well as the rate of removal of sulphate aerosol embedded inside precipitating droplets in UCLALES-SALSA, … " on page 12, lines 8-10.

12) P9, L33-35: How is scavenging treated in the below-cloud layers? Please consider adding this information.

Collision and collection processes are treated between all different particle and droplet classes using the coagulation equations. Upon collision between an aerosol particle and drizzle drop, the mass of the aerosol particle is moved to the drizzle bin in question. This will be elaborated on in the manuscript.

Scavenging by precipitation below-cloud is explained on page 7, lines 28-29 of the revised manuscript.

13) P10 L6: 'lack of representation for aerosol scavenging' – How is aerosol scavenging represented in LEV3? Consider adding this information earlier on in the text to help the reader in understanding these comparisons between the LEV3 and LEV4 simulations.

As stated in the response to comment No 11, LEV3 does not contain a representation for aerosols, and therefore the wet scavenging process is not represented by LEV3. This will be stated more clearly in the manuscript.

This is stated on page 8, lines 8-10.

14) P10, L11-12: 'LWP and rain water path show quite similar features as those obtained with a cloud system resolving model with interactive aerosols' – Please state these 'similar features' more explicitly.

The results in the low aerosol cases of this paper showed a similar depletion of cloud water caused by aerosol scavenging and drizzle. We will explain this more clearly.

It now says "Interestingly, the reduced cloud water and increased drizzle caused by depletion of aerosol, as shown by Figure 3 for LEV4, resemble the corresponding effects shown by (Yamaguchi and Feingold, 2015) … " on page 12, lines 30-32.

15) Section 3.2: This section includes a detailed discussion of the simulation results for the case DYCOMS-II flight RF02, which was a marine stratocumulus case that took place off the coast of California. Would there be observations available that could be explicitly compared to the simulation output presented here?

We will add flight-mean estimates of LWP and surface precipitation to Figures 3 and 4, respectively. It is shown that the model results fit the observed values quite well, given the assumptions used in the model runs. A more detailed discussion will be added to the manuscript.

The observed estimates for LWP and surface precipitation retrieved from literature are now show in fiigures 3 and 4 and commented on page 11, lines 1-3 and lines 11-13.

16) P11, L22: Why was the drizzle formation switched off for this fog case?

Even though the drizzle formation was not explicitly used in these simulations, the fog droplets can grow freely, given the ambient conditions, and reach the size range when they begin to be removed by sedimentation. However, as stated also by Porson et al. (2011), the liquid water content remains relatively small, so explicit drizzle parameterization is not needed. We follow this notion to conform with their model setup.

This is noted on page 14, lines 23-25 in the revised manuscript.

17) P12, L19: Consider adding a table to describe the simulations A200, A400, A800 A400W.

We will add a table with the details of the simulation setups.

Table 2 has been added with the requested details and noted on page 15, line 10.

18) P13, L4-5: 'These findings illustrate the ability of the UCLALES-SALSA to provide a realistic description of not only the thermodynamic and microphysical properties. . ..' – Please consider if this statement would be better supported by explicitly showing model-observation comparisons in the manuscript.

Here, we refer to the results presented in Porson et al. (2011) and Price (2011). More elaborate discussion about comparing our model results with the afore mentioned data will be added to the manuscript.

This comment is considered in the added discussion on page 16, lines 10-30 in the revised manuscript.

19) P 13, L8-9 'growth rate is considerably lower than the observed'. . .'see figure 5 in Porson et al., 2011' – are there observations that could be explicitly shown here to help the reader understand these comparisons?

We will use the data on fog layer growth presented in Porson et al. (2011) based on tethered balloon measurements. The data points are added to our Figure 9.

Observed data is now shown in Figure 10 (note the changed number!) in the revised manuscript.

20) P13, L21: 'These results point towards the importance of detailed representation of the microphysical processes.' This sentence does not appear to be finished – do you mean in cases of fog?

Yes, in this context we refer to the fog case. We will reword the sentence.

It now says "The results point towards the importance of detailed representation of the microphysical processes in cases of fog formation." on page 16, line 21.

21) P13, L22: 'UCLALES-SALSA does well' – Are you able to quantify what is meant by 'does well'?

This refers to the occurrence of the peak droplet number concentration mentioned in the next sentence. We will adjust the wording.

It now says "In particular, the size resolving microphysics in UCLALES-SALSA results in a peak number concentration in the fog droplet size distribution at approximately 25 µm in terms of the wet diameter, which agrees with the observed range between 20 µm and 25 µm based on the measurements presented in Price (2011)." on page 16, lines 22-24 in the revised manuscript

22) P13, L26 'UCLALES-SLASA also agrees well with observations' – again please quantify what is meant by 'agrees well' and consider showing model-observation comparisons in the manuscript.

Again we refer to Porson et al. (2011) where it is shown that droplet concentrations between 20 and 60 cm-3 were measured for this forg case. The droplet concentrations in the experiment A400 fit to this range, except when the fog layer eventually transforms into a shallow cloud later in the morning. We will discuss this in more detail in the manuscript.

Some new discussion is added on page 16, lines 26-31.

23) P13, L30: 'a more detailed land surface scheme is needed' – did you test any limiting cases?

No, we did not. The surface heat capacity was tuned to match the observed surface temperature, and the surface was assumed to be wet.

24) P14, L29-30 'very similar to the observations'. . ..'even more resembles the observed properties' – Please consider showing these comparisons in the manuscript, likewise showing some model-observations comparisons would be helpful for understanding the model performance for the stratocumulus case.

Measured data is added to Figure 9 of the revised manuscript for fog layer growth (comment #19). Moreover, observed estimates of LWP and surface precipitation are now shown in Figures 3 and 4 (comment #15).

Changes are covered by earlier reponses.

25) P14, L29: If a realistic wind profile improved the model-measurement agreement – why was the case with winds not used as a default? Did you test A200W and A800W?

We wouldn't consider the no-wind simulations as the "default". Instead, they were considered first, because such a simple setup allows us to demonstrate the effect of aerosols specifically, which we are most interested in. With a realistic wind profile, the simulations were performed also with other aerosol concentrations. However, the mixing caused by wind shear dominates the growth rate of the fog layer over the initial aerosol concentration. Moreover, supersaturation inside the fog is also strongly affected by mixing, which makes the differences in fog droplet concentrations less clearly defined between the different aerosol concentrations.

Technical corrections:

1) P2,L10: Do you mean 'of' instead of 'off'? *- Corrected*
C42) P5, L20: 'Evolution of the drizzle droplet population' – should this read drizzle/rain since the upper diameter limit is 2mm? *- Done.*
3) Fig. 3a: Should HI be removed from the legend? *- Done.*
4) P12, L27: Do you mean Fig 8 as opposed to Fig. 9? *- No, Fig 9 is correct.*
5) P12, L31: Consider starting a new paragraph with the start of the Fig. 11 discussion. *- Done.*
6) P13, L13: Do you mean Fig. 9 as opposed to Fig. 10? There is no dashed line on Fig. 9. *- Corrected.*
7) Fig. 1: What is the meaning of the light blue arrows on the dark blue for the drizzle rain bins? What is the size range for the cloud droplets? *– They were to represent the rain drop growth, but indeed might be misleading. They are removed. Size range is added.*
8) Fig 2: Could g kgˆ-1 be placed beside the color bar? *- Done.*
9) Fig 3: Could drizzle be added to the title of panel b? Also, please check legend for error in simulation names. *- Done.*
10) Fig 7: Please check units on the legend – did you mean m? *- Yes, corrected.*

All the corrections above have been implemented.

We thank Reviewer #2 for his/her comments and feedback. The largest changes in the manuscript in response to these comments will be the improved presentation of technical details in addition to the other more specific suggestions given by the Reviewer which will also be implemented in the revised manuscript. Below, we will list all of the Reviewer comments, followed by our response highlighted in *italics.*

–

The corrections to the manuscript regarding each comment are highlighted in italics.

Paper content and a general impression

The submitted manuscript describes a newly-developed software package for research on aerosol-cloud-precipitation interactions. The presented framework is composed of a modified (adapted) version of the free and open-source Large Eddy Simulation (LES) system UCLALES, coupled with a modified (extended) version of the SALSA aerosol process modelling package. The paper consists of brief description of both pre-existing software packages and of how they were adapted, extended and coupled to result in the UCLALES-SALSA system. Moreover, the capabilities of the developed tool, in particular its applicability to capture aerosol-cloud interactions, are exemplified for two different simulation set-ups.

The topic of the paper matches well with the current interests of the cloud-modelling community – the study fits into an active stream of development of modelling techniques to study aerosol-cloud-precipitation interactions in LES-type frameworks. These concurrent endeavours are not referenced comprehensively in the paper, though.

My main major concern is the misalignment of the paper content with the scope of GMD. The model description itself amounts to ca. 3 pages out of 15 (not counting figures or references), while the rest of the paper deals with the case studies. Of course, this is not the page-count that matters and the case studies do provide valuable examples and validation of the model capabilities. Yet, the model formulation and implementation – in my understanding the key elements of the GMD scope – are clearly described in not sufficient detail. There is not a single equation used in describing the new extensions to the UCLALES and SALSA, even though the authors admit that "coupling the extended SALSA module into UCLALES yields extensive changes in the thermodynamic core of the model". A key component newly introduced into the model formulation, representation of cloud droplet collisions and coalescence, is commented with just a single sentence without detailing the numerical method or its implementation. The reader is left without any information about software engineering aspects of the project (without studying the references, the reader would not even learn about the programming language in which the code is written; more importantly such aspects as parallelisation techniques, required environment and tools to use and extend the model are not mentioned and these cannot be guessed). There is no information on how the modified UCLALES and the extensions to SALSA are planned to be disseminated within the community, how existing UCLALES and SALSA users can benefit

from the described developments. If the authors would rather prefer to keep the paper focused on the case studies, and not the model formulation and implementation, I suggest submission of a revised version of the text to ACP or similarly scoped journal. The model code is also not publicly available as of now. It not only makes the paper not compliant with the GMD guidelines, but it also prevents me to fulfil the reviewer's duties – the GMD board clearly states that all papers "must be accompanied by the code, or means of accessing the code, for the purpose of peer-review", moreover the journal guidelines

”strongly encourage referees to compile the code, and run test cases supplied by the authors”1. The authors do not detail how the paper readers may reproduce the discussed results.

For the reasons listed above, I am requesting a second round of the review to follow. In my opinion, the manuscript requires substantial changes to reach a good level of readability and to match contemporary standards of research reproducibility pioneered by GMD. Nevertheless, let me repeat that the described research and the developed tool are of prime interest to the community. In particular, the described system is capable of simulating aerosol processing by clouds through activation-collision-deactivation cycles as well as resolving aerosol sources – features not widely available in other LES-type systems.

As a general statement, we will improve on the description of the model technical details in the manuscript. In the current version, it is stated that the model will be available through Github, and upon request before its release. We are currently still in the process of cleaning up the code for a public release and expect this to be finished shortly.

Lots of new details have been added especially to Section 2.1. Moreover, new Section 2.3 gives further information about the technical details.

General remarks

Few references to other LES aerosol-cloud-precipitation interaction studies
For the purpose of giving a comprehensive background, as well as of highlighting the unique features of UCLALES-SALSA, I strongly suggest supplementing the list of referenced works with some seminal
and/or recent papers on aerosol-cloud-precipitation interaction modelling with LES-type tools. In the list below, I suggest some that might be worth checking. The list is certainly not exhaustive, though:
• Lebo and Seinfeld, 2011: 10.5194/acp-11-12297-2011
• Ovchinnikov and Easter, 2010: 10.1029/2009D012816
• Andrejczuk et al. 2010: 10.1029/2010JD014248
• Shima et al. 2009 10.1002/qj.441
• Feingold et al. 1996: JGR 101 (D16)
The above references have been included in the revised manuscript on page 2, lines 10-15.

In particular, citing some of these works could support or otherwise require rewording of some statements:
• p. 2/line 9/10: “extensive simulations with more detailed and interactive ... schemes ... are relatively sparse”
The sentence “Nevertheless, some examples of such developments include the works of (Andrejczuk et al., 2010; … ” has been added on page 2, lines 14-15.
• p. 2/line 33: “innovative approach” (it is worth clearly stating precisely what is novel here)
This is stated more clearly, page 3, lines 7-9.
• p. 4/lines 6-7: “not a computationally feasible approach”
It now says “However, although such two-dimensional frameworks have been developed (e.g. Lebo and Seinfeld, 2011), the approach is computationally highly demanding for large-eddy modelling applications spanning timescales of days while covering relatively large domains with high spatial resolution, which are pursued here” on page 6, lines 4-8.

We will adjust the manuscript as suggested by the Reviewer.

Lack of model formulation and implementation details

As outlined above, I strongly encourage the authors to increase the level of detail in which the model formulation and implementation is described. Here are some examples:

• p. 4/line 20: How the substepping is implemented? Are the grid-mean values kept constant for all substeps within a timestep (in particular, the supersaturation)?

Substepping is now described on page 5, lines 17-25 in the revised manuscript.

• p. 5/line 13,15: statements seem contrasting: “not knowing the wet droplet diameter exactly” but “bin mean cloud droplet wet diameter” is used, perhaps it is worth summarising clearly what are the variables and constants per bin for each spectrum, and which processes change them – a table would likely give best readability

We have reworded the first paragraph of Section 2.1.2. to avoid confusion. In addition, the sets of prognostic variables and their treatment within the aerosol, cloud droplet and precipitation sizebins is now explicitly mentioned on page 3, lines 27-28; page 6, line 10 and page 7, lines 18-19, respectively.

• p. 5/line 29-30: here the treatment of coalescence is described by just one single sentence without any reference. What are the numerics behind, how the kernels are supplied (if look-up tables, please detail interpolation method)?

Coagulation is now described more accurately on page 4, line 8 – page 5, line 7.

We will increase the level of technical details in the model description, including all the points above raised by the Reviewer. As also requested by Reviewer #1, equations describing the key model processes will be added.

Simulation setup description

The fact that two contrasting cloud regimes are simulated gives a nice opportunity to pick this a criterion for mentioning or not a given simulation parameter. I suggest thus creating a table listing all model parameters that needed to be changed (or where arbitrarily switched on or off) in order to make the simulation depict fog instead of stratocumulus. This could perhaps allow to shorten a bit the setup descriptions in the text, the initial profile given by eq. 1-4 could then be part of the table (why not just cite the relevant equations in the paper in which the DYCOMS profiles where defined). If adaptive timestepping was used, please provide some statistics on the timestep values for the two different setups.If a spinup period is used for model initialisation, please clearly indicate which processes are on or off for how long, and what are other differences between the spinup and the rest of the simulation.

We will include more detailed information about the model setups as well as the spinup configuration. We will also elaborate on how the setup for fog simulations differs from the stratocumulus case. These differences are mainly comprised of model resolution, surface conditions and the input sounding. Adaptive timestep is used – we will add more detailed information about the values to the manuscript.

The description of the fog experiments is revised and now explicitly mentions the differences in the configuration with respect to the stratocumulus case, i.e. on page 14, lines 7-15 and lines 22-23. In addition, the case specific settings for the fog simulations are now divided into a new Section 4.1.1 (regarding the initial aerosol concentration and wind conditions). The spinup is detailed for the stratocumulus case on page 9, lines 20-23, and for the fog case on page 15, lines 1-2. Adaptive timestep is detailed for stratocumulus on page 9, lines 25-26 and the fog case on page 14, line 12.

Also, some of the model features advertised in the beginning of the paper seem not used in the simulations (e.g., condensation of precursor gases and new particle formation mentioned on p. 3/line 30) – please state it explicitly. In contrast, features such as inclusion of the diurnal cycle or the soil energy balance are not mentioned in model description part.

The condensation of aerosol precursors was active in the model simulations, but it's effect was negligible in the current simulations setups. New particle formation was not active, although available. We will mention these and the other features suggested by the Reviewer.

Condensation of aerosol precursors is now mentioned in the model description, page 5, lines 9-10 and new particle formation on lines 27-29. Soil energy balance is given in the case descriptions since its different for stratocumulus and fogs: page 9, line 19 and page 14, lines 15-22. Diurnal cycle is mentioned in the model description on page 8, lines 24-25.

Statements such as "large cloud droplet are considered as drizzle" (p. 8/line 27) or "the surface heat capacity is used as a tunable parameter" (p. 12/line 5) call for numbers.

We will add this information in the manuscript.

Drizzle threshold diameter is mentioned (now on page 11, line 8), Heat capacity is further explained on page 14, lines 20-22

If I understood correctly, presented simulations lack aerosol sources. In contrast, the setups like DYCOMS-II implicitly assume an infinite reservoir of CCN brought in to the domain by advection. If that is correct, this difference is worth mentioning and perhaps discussing.

This is true, as well as the fact that the interpretation of the model simulations indeed does change when switching from the default UCLALES with prescribed CCN concentration to UCLALES-SALSA with a more dynamic description. The latter points more towards a "Lagrangian" simulation in the sense that the depletion of aerosols by clouds and precipitation resembles a domain moving with the flow. We will add discussion of this in the manuscript.

This is added on page 12, lines 22-26.

Aerosol processing nomenclature
Depending on the community "aerosol processing" is associated with different processes if put out of context. Please clearly state, at least in the abstract and introduction, whether chemical processing or collisional processing is addressed. Especially, since condensation of sulphates is mentioned on page 3.

Despite the condensation of aerosol precursors included in the model, for now the model does not have chemical processing. This will be stated more clearly.

This is mentioned on page 7, line 4.

Section scope
The introduction section mentions such, distant from the scope of the paper, matters as challenges in climate modelling, arctic temperatures changes, decrease in fog occurrence in Central Europe. For fellow cloud modellers, the links between those topics and the paper scope might be "obvious", for other members of the GMD audience these will seem puzzling, tough. Please either elaborate on how and why these topics are related with the development of UCLALES-SALSA or keep the introduction closer to the paper scope.

Since this modelling work ultimately aims at providing a research tool to improve the above mentioned features in global and regional climate models, we will keep these topics in the introduction, but will provide more in depth description, as suggested by the Reviewer.

We have clarified the motivation for this discussion on page 2, lines 16-27 and page 3, lines 2-6.

The DYCOMS-II section uses up to three-digit section numbering (e.g., 3.2.1) while the fog case is just divided between two case description and Results subsections. I suggest some work on restructuring the two sections to be more similar in both section numbering and, more importantly, the level of detail.

We will break Section 4 into smaller divisions.

The fog case experiment details are now divided into a new Section 4.1.1

The specific suggestions below will be implemented into the manuscript unless otherwise mentioned.

Specific comments and rewording/correction suggestions

• Paper-wide:

– drizzle → precipitation (in particular in the title, the model is not limited to drizzle and since one of the quantities analysed is the surface precipitation rate, the simulated precipitation is by definition not drizzle) *Done*

– aerosols → aerosol (e.g., in the title, I don't have a strong opinion on it - just a suggestion) *Done*

– computational burden → computational cost *Done*

– high computational burden → resource intensive, etc *Computational cost is now used*

– interactive, fully interactive scheme, interactive description of particles – please explain what you mean exactly (by explaining which models are non-interactive), especially as it is mentioned in the title *The terminology has been made more clear and the title is justified by that UCLALES-SALSA tries to capture the aerosol-cloud coupling explicitly in both directions as well as their impacts on the dynamics. This is stated more clearly on page 3, lines 7-9.*

– please ensure that acronyms are explained on first occurrence (e.g., SALSA is only deciphered in section 2.1) *This is the first occurrence in the text.*

• Abstract:

– line 1: impacts of → impacts on *Done*

– line 1,3: improved over what?, more sophisticated than what?, what kind of observations? Please be precise, please try to cater to a wider community, please make sure that the abstract summarises the presented research – global climate and gaps in observations seem not relevant enough to pop up in the very first sentences of the abstract *We have reworded the abstract to avoid confusion. Global modelling and lack of observations are the most essential key motivation for building the UCLALES-SALSA in the first place so mentioning them in couple of sentences of the abstract is justified.*

– line 4: model, coupled → model (LES), coupled *Done*

– line 5/6: "microphysical model components" is vague – please state if you refer to SALSA or something else as well *Done*

– line 6: "strategies for ... bin layouts" reads awkward, perhaps the keyword discretisation could help to better convey what is meant? I understand bin layout as a parameter of a given simulation, what is perhaps worth mentioning in the abstract is how the modelled particles are classified and which classes of particles are subject to which processes *The word discretization is now used. Going into implementation and process-level details is too much for abstract – these are described thoroughly in the text.*

– line 8: “computational cost of the model acceptable” – this is not only subjective but also likely to be objectively false soon *It now says “as low as possible”*
– line 8: two different cases: one comprising a case with marine stratocumulus . . . → two different simulation setups: the DYCOMS-II marine stratocumulus setup *Done*
– line 9-10: It is shown that, in both cases, . . . *Done*
– line 13: In radiation fogs, the growth *Done*
– line 14: strongly affects → strongly affect *Done*

• Introduction:

– p. 1/line 18: simulators → simulations *Done*
– p. 1/line 19: Moeng 1984 – please either use a few references to support the use of LES for decades or cite a recent review paper (preferably) *References added, page 1, line 21.*
– p. 2/line 7: please mention also particle-based models (in addition to bulk, modal and sectional) *Mentioned, page 2, line 11.*
– p. 2/line 10: “mostly due to their high computational burden” – isn’t it the multi-scale and multidisciplinary nature of clouds that limits us most and not the computer power? *Yes, but building models much more elaborate than what we can afford to use (at least widely) is a fully relevant consideration in the context of this sentence.*
– p. 2/line 25: “fogs also feature many different aspects” – please reword *It now says “Although in many ways driven by the same principles as clouds, fogs also feature many unique aspects considering their evolution.” on page 2, lines 32-33.*
– p. 2/line 30: four references given to support statement that fogs are affected by anthropogenic emissions – please try to keep balance with the paper and journal scope *This issue is not straightforward and the cited references may give the necessary background information for some readers.*
– p. 3/line 6,7: “well-characterised” vs. “findings of” could hint that one is superior to the other, please reword *We have removed “well-characterized” to balance out.*
– p. 3/line “previous model versions” versioning suggests something linear, in this context we are rather faced with multiple diverging branches, please try to avoid the version when giving a precise version number would be tricky *Reworded, page 3, line 17-18.*

• Section 2:

– p. 3/line 16: drizzle/rain → precipitation *Done*
– p. 3/line 17: since the number 10 is just a setting for a particular simulation, perhaps it is worth explaining instead how the bins are laid out (logarithmically?) *Done, page 4, line 3-4.*
– p. 3/line 18: what does the Bergman et al. citation refer to? (10 bins?, Figure?) *The discretization with 10 bins, reference moved to point to this more clearly.*
– p. 4/line 8: “parallel bins” is hard to understand *This is explained more clearly, page 6, lines 10-15.*
– p. 4/line 15: non-chronological order of citations *Corrected.*
– p. 4/line 16: “very fast relative changes” is vague, isn’t it anyhow the stiffness of the governing equations due to presence of multiple size scales that is the crux of the matter? *The subsetpping issues are described in more detail now on page 5, lines 17-25.*
– p. 4/line 32/33: “unwanted discontinuities in ... calculation”, please reword so it is clear what is discontinuous *Reworded, page 6, lines 29-30.*
– p. 5/line 8: I suggest removing the last sentence of this paragraph *Done*
– p. 5/line 9: drizzle → precipitation *Done*
– p. 5/line 12: please rephrase, perhaps referring to statistical moments resolved within each bin would make it clearer *Reworded, page 7, line 6-8.*
– p. 5/line 21: please rephrase so that it is clear that wet diameter is the relevant quantity for condensation and collision, currently the sentence suggests that drizzle condensational growth is critical to produce realistic precipitation *Done, page 7, lines 16-17.*

– p. 5/line 22: does SALSA share the implementation of the Abdul-Razzak and Ghan (2002) scheme with some other (open-source?) model? *It does not.*
– p. 5/line 28: please explain (mathematically) how the bin layout is formulated *This is explained in more detail on page 7, lines 24-27.*
– p. 5/lines 31-32: please refer to the ”aerosol processing” in the sentence *Done, page 7, lines 32-3.*
– p. 5/lines 31-32: perhaps citing Mitra et al. 1992 could be used to support the assumption? *See above.*
– p. 6/line 10: “default version” might mean something different for each user, please be specific *We now refer to this simply by “UCLALES”.*
– p. 6/line 20: “raising the number of prognostic scalars” → “increasing the number of advected scalars from O(?) to O(100) *Done, page 9, line 1.*
– p. 6/line 22: please hint the level of concurrency used – otherwise just the computer type makes the statement very vague *Done, page 9, lines 1-5.*

• Section 3:
– p. 6/line 31: is there any limit on the magnitude of the supersaturation during the initialisation? *Currently no.*
– p. 7/line 22: would one of “Reference case”/”Reference setup”/”Reference run” be more apt than “Default case”? *“Reference case” is now used.*
– p. 7/line 27: from previous statements, I understood model initialisation to be equivalentthe spinup period, here it seems to mean pre-time-zero calculation – please use consistent wording *It now says “model startup”.*
– p. 7/line 28: please reserve the word “parallel” for calculation concurrency
– p. 7/line 28: please define somewhere the “default UCLALES configuration” – again, this might mean different settings to different users *It now says “UCLALES with bulk microphysics”.*
– p. 7/line 29: “default UCLALES” – does it refer to the “default case”, “default configuration” or something else *Referred again simply as UCLALES.*
– p. 8/line 17: the domain-mean plot discussed was likely not the basis for statements“massive shallow convective cumulus elements” or open cells; please clearly define where you discuss the figure *This is now explained more clearly on page 10, lines 26-30.*
– p. 9/line 20: “By the same token” sounds strange to me (but I’m not a native speaker) *Switched to “argument”.*
– p. 9/line 29-31: please reword, “performs this task with very high detail” could be omitted, the use of “beyond” is unclear here *Reworded, page 12, lines 10-12.*
– p. 9/line 32-33: another example where the reader can be puzzled about differencesspinup and model initialisation *Reworded, page 12, lines 12-13.*
– p. 10/line 12: please reword “model with interactive aerosols” *Reworded, page 12, lines 29-32.*
– p. 10/line 24: “to the their” → “to their” *Done*
– p. 10/line 33: I assume this means very small in one simulation and non-existent in the other, please reword Reworded, *page 13, lines 19-20.*
– p. 11/line 5: isn’t coagulation part of the processing *Reworded, page 13, lines 24-27.*
• Section 4:
– p. 11/line 28: “water surface pressure” → saturation vapour pressure? *It now says equilibrium saturation ratio, page 14, line 32.*
– p. 12/line 7-8: “is not available” suggests lack of availability, here it was simply not partthe setup *No, the information was not available.*
– p. 13/line 5: “-radiative” → “-radiation” *Done*
– p. 13/line 5: if “and feedbacks” is needed, please explain how do you differentiate them from interactions *Done, page 16, lines 4-5.*

– p. 13/line 29: “connect the aerosol concentration into fog existence” – I suggest rewording *It now says “couples with fog occurence” on page 16, line 33-34.*

• **Conclusions:**

– p. 13/line 33: “A new large-eddy simulation model” suggests some new fluid dynamics methodology, while the novelty is elsewhere – please reword *Reworded, page 17, line 2.*

– p. 14/line 5: please precise what type of processing (i.e., non-aqueous-chemistry related) *Reworded, page 17, line 7.*

– p. 14/line 27: “observed behavior Price” needs a parenthesis *Done*

• **References:**

– line 23: korolev → Korolev *Done*

– line 28: kokkola → Kokkola *Done*

– line 30: korhonen → Korhonen *Done*

• **Figure 2: please sort out the background colour issue in panel b** *There is in fact no background color issue. In UCLALES-SALSA the condensed water in aerosol particles is classified as “cloud water” which, even though small, becomes visible because of the log-scale of the colormap. We will note this in the caption.*

~~Introducing~~ UCLALES-SALSA v1.0: a large-eddy model with interactive sectional microphysics for ~~aerosols~~aerosol, clouds and ~~drizzle~~precipitation

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Abstract. ~~Aerosol-cloud~~ Challenges in understanding the aerosol-cloud interactions and their impacts ~~of~~ on global climate highlight the need for improved knowledge of the underlying physical processes and feedbacks as well as their interactions with cloud and ~~boundary-layer~~ boundary layer dynamics. To pursue this goal, ~~more~~ increasingly sophisticated cloud-scale models are needed to complement the limited supply of observations of the interactions between aerosols and clouds. For this purpose, a new large-eddy simulation (LES) model, coupled with an interactive sectional description for aerosols and clouds, is introduced. The model, UCLALES-SALSA, builds and extends upon a well characterized LES model (UCLALES) and microphysical model components (SALSA). Novel strategies for the aerosol, cloud and ~~drizzle bin layouts~~ precipitation bin discretization are presented. These enable tracking the effects of cloud processing and wet scavenging on the aerosol size distribution as accurately as possible while keeping the computational cost of the model ~~acceptable~~as low as possible. The model is tested with two different ~~cases: one comprising a case with marine stratocumulus and another~~ simulation setups: a marine stratocumulus case in the DYCOMS-II campaign and another case focusing on the formation and evolution of a nocturnal radiation fog. It is shown that, in both cases, the size-resolved interactions between aerosols and clouds have a critical influence on the dynamics of the boundary layer~~in both cases~~. The results demonstrate the importance of accurately representing the wet scavenging of ~~aerosols~~ aerosol in the model. Specifically, in a case with marine stratocumulus, ~~drizzle formation~~ precipitation and the subsequent removal of cloud activating particles lead to thinning of the cloud deck and the formation of a decoupled boundary layer structure. In radiation fogs, the growth and sedimentation of droplets strongly ~~affects~~ affect their radiative properties, which in turn drive new droplet formation. The size resolved diagnostics provided by the model enable investigations of these issues with high detail. It is also shown that the results remain consistent with ~~an earlier version of the UCLALES model~~ UCLALES (without SALSA) in cases, where the dominating physical processes remain well represented by both models.

1 Introduction

Large eddy ~~simulators~~ simulations (LES) have been used to study the properties of clouds and the boundary layer for a few decades ~~(e.g. ?)~~ (e.g. **????**) . These models solve the low-pass filtered Navier-Stokes equations, i.e. the large energy-containing turbulent eddies are resolved, while the smallest length scales and energy dissipation are parameterized typically using closures based on the Smagorinsky model. This approach provides an attractive compromise between accuracy and computational ~~burden~~cost, which is why LES models have become popular in studies of the properties of ~~boundary-layers~~ boundary layers and clouds.

The typical grid resolution used in LES models (~~$\mathcal{O}$~~ on the order of tens of meters) enables a detailed representation of cloud structure and dynamics. However, the treatment of cloud microphysics is subject to high variability in terms of the level of detail and computational ~~burden~~ cost (**?**). The types of microphysical schemes and their implementation to LES models range from simple one or two moment bulk schemes, where droplet mass is predicted typically through saturation adjustment, with either prescribed or varying droplet number concentrations (**??????**), to more elaborate ones with modal or sectional representations for the droplet size distributions ~~(??) .~~ (**???**) and Lagrangian particle based methods (**?**) . In addition, there has been an increasing trend towards including representations for aerosol particles in these models as well (**???**). However, extensive simulations with more detailed and explicitly interactive aerosol-cloud schemes are as ~~off~~ of yet relatively sparse, mostly due to their high computational ~~burden~~cost. Nevertheless, ~~the~~ some examples of such developments include the works of (**?????**) .

The need for such models is well recognized due to the significant challenges in climate modelling imposed by aerosols and clouds (**?**)~~.~~ , where detailed LES model simulations comprise an essential resource for parameterization develoment. In particular, formation of drizzle and wet scavenging of ~~aerosols~~ aerosol and the associated feedback processes are potentially very important for the dynamics and circulation structures of marine stratocumulus clouds (**????**). Correctly capturing the interactions between aerosol-cloud microphysics and cloud dynamics requires highly detailed microphysical schemes. Moreover, scavenging processes, depending on particle composition and size, are overall rather poorly understood and therefore poorly represented in general circulation models (**??**). Yet, wet scavenging of ~~aerosols~~ aerosol may crucially affect e.g. the transport of black carbon ~~aerosols~~ aerosol from polluted environments to the polar areas (**?**), where it has the potential to significantly affect the future change in arctic temperatures. The main motivation for the LES model development presented in the current paper is indeed to provide a new tool for a better understanding of the above mentioned climate-relevant processes, so that they can eventually be more robustly represented in global models.

Besides cloud processes, another set of topics under research by the LES community is related to the formation and evolution of fogs and the effects of aerosols therein. During the last decades, a clear decrease has been observed in fog occurrence throughout Central Europe (**??**). This has occurred together with improved air quality due to a decreasing trend in sulfur emissions, especially in the case of dense fogs, but this far, a quantitative connection has not been established (**?**). Although in many ways driven by the same principles as clouds, fogs also feature many ~~different aspects~~ unique aspects considering their evolution. (**??**). For example, while cloud droplets are mainly formed at the height of the peak saturation ratio at cloud base, in radiation fogs, one of the most common fog types, the droplet formation is primarily driven by radiative cooling at the top of the developing fog layer or by high supersaturation inside the fog induced by turbulence. Thus there are also marked differences related to the dynamics of the fog layer and it's life cycle as compared to clouds (**?**). Fog properties and their occurrence are strongly affected by aerosol properties and anthropogenic emissions (**????**), although many of the details of these interactions remain poorly understood. Improved knowledge can be pursued through increasingly sophisticated microphysical schemes embedded in LES models. Thus, a case comprising a radiation fog event serves as a well-justified testbed for the model presented in this paper.

Here, an innovative approach is proposed to treat the ~~issues related to~~ microphysical interactions between aerosols and clouds as well as their impacts on boundary layer dynamics within a high-resolution LES model ~~.~~ while keeping the model computationally feasible for long simulation times (few days) and large model domains (tens of kilometers). We build and extend upon a state-of-the-art LES ~~and sectional microphysical models~~ model and a sectional microphysical model (**??**) to create a cloud-resolving framework, where the size distributions of ~~aerosols~~aerosol, clouds and ~~drizzle~~ precipitation are all described with a detailed ~~, fully interactive~~ sectional approach. In particular, the model introduced in this work accurately preserves the characteristics of the aerosol size distribution both in- and outside of clouds~~and fogs~~, making it ideal for studying the impact of removal processes, cloud processing and evaporation on the particle size distribution, as well as the associated feedbacks on cloud properties, ~~drizzle formation and fog life cycle~~precipitation formation and boundary layer dynamics. The model is evaluated by experimenting on two very different cases: one comprising marine stratocumulus clouds based on the ~~well-characterized~~ DYCOMS-II dataset (**?**), and another focusing on a radiation fog event based on the findings of (**??**). The results are compared with earlier studies and ~~previous model versions~~ models with a simple bulk microphysics scheme, and similarities and differences are analyzed and explained in detail.

The new model is described in detail in Section 2 while case descriptions and results for the marine stratocumulus and fog cases are documented in Sections 3 and 4, respectively. Discussion of the model performance and conclusions drawn from the results are reported in Section 5.

2 Model description

2.1 The extended SALSA module

The Sectional Aerosol module for Large Scale Applications (SALSA; **?**) is used as the basis for developing a unified sectional microphysical model for aerosols, clouds and ~~drizzle/rain~~precipitation. The SALSA module, previously employed in the ECHAM (**?**) climate model family, discretizes the aerosol size distribution into 10 size bins according to the dry particle diameter (**?**) as shown in Figure **??**~~(?)~~. The predicted variables for each bin are the aerosol number and compound masses as well as the mass of condensed water, which can be used to determine the bin mean wet particle size. The total diameter range covered by the bins (from 3 nm to 10 μm by default) is divided into subranges, 1a and 2a. This division into subranges aims at minimizing the number of tracer variables. This is achieved by including only those chemical compounds that are significantly abundant in each subrange. Subrange 1a covers the three smallest bins (up to 50 nm) and the particles are assumed to be internally mixed, being composed of ~~compounds that~~ sulfate and organic carbon, which contribute to the growth of newly formed particles~~, i.e. sulfate and organic carbon~~. Subrange 2a includes particles larger than $50\,\mathrm{nm}$ whose composition may comprise all the chemical compounds in the model. The module can be configured to include 7 additional bins (designated 2b) parallel to the bin regime 2a (i.e. same bin diameters), which allow the description of externally mixed particle populations~~so that soluble compounds are~~. In a typical example, soluble compounds would be emitted to 2a ~~while insoluble compounds are emitted~~ and insoluble compounds to 2b. The spacing of the size bins is set logarithmically equidistant within each of the subranges. Further details about the bin ~~layout~~ discretization can also be found in **?**. ~~The spectral resolution given by this bin layout~~ With these settings, the spectral resolution is quite coarse, but does provide a good compromise between computational cost and model performance. Note however, that the numbers given here represent the default settings - the number of bins can be set to be larger, if necessary.

The SALSA module includes detailed methods for solving the key microphysical processes ~~; coagulation , condensation of aerosol precursor gases (sulphate, organics) as well as new particle formation by sulphuric acid. A detailed description of the methods for solving aerosol microphysical processes is given by **??**~~ which are called sequentially. Coagulation is modelled based on the equations in **?** . For particle number this is given as

$$n_{i,t} = \frac{n_{i,t-1}}{1+\Delta t \sum\limits_{j=i+1}^{J} K_{i,j} n_{j,t-1} + \frac{1}{2}\Delta t K_{i,i} n_{i,t-1}}. \tag{1}$$

Similarly, for volume concentration

$$v_{i,t} = \frac{v_{i,t-1}\Delta t \sum\limits_{j=1}^{i-1} K_{j,i} v_{j,t} n_{i,t-h}}{1+\Delta t \sum\limits_{j=i+1}^{J} K_{i,j} n_{j,t-1}}. \tag{2}$$

In the above equations, $K_{i,j}$ is the total coagulation kernel for the colliding particles in bins i and j, $n_{i,t}$ is the particle number concentration in bin i at timestep t ($t-1$ refers to the previous timestep), $v_{i,t}$ is the volume concentration, Δt the length of the timestep and J is the total number of particle bins. Please note that the bin indices $1 \ldots J$ should be interpreted to cover all the bins of all particle categories in the model (aerosol, clouds, precipitation) sorted by increasing particle size.

For coagulation kernels with aerosol particles, we assume brownian coagulation, whose kernel in the continuum regime is given as

$$K^{B}_{i,j} = 2\pi(d_i + d_j)(D_{p,i} + D_{p,j}), \tag{3}$$

where d_i and d_j are the diameters of the colliding particles and $D_{p,i}$ and $D_{p,j}$ their corresponding diffusion coefficients. For the transition regime, the formula by **?** is used. For larger particles, i.e. cloud droplets and precipitation, the convective enhancement of Brownian coagulation and gravitational collection are also included, and given as and

$$K^{GC}_{i,j} = E^{c}_{i,j}\pi(\frac{d_i + d_j}{2})^2 |V_{f,i} - V_{f,j}|, \tag{4}$$

respectively (?) . In the above equations, Sc_i is the particle Schmidt number, Re_j is the Reynolds number, $E^c_{i,j}$ is the collection efficiency and $V_{f,i}$ is the particle fall speed. The latter is parameterized as where ρ_p is the particle/droplet density and ρ_a is the air density. $\rho_{a,ref}$ is a reference air density (given at the STP conditions, 273.15 K and 1000 hPa), g is the gravitational acceleration, γ is the dynamic viscosity of air and β is the Cunningham slip correction factor. The total coagulation kernels in Equations (??) and (??) are obtained as the sum $K_{i,j} = K^B_{i,j} + K^{BC}_{i,j} + K^{GC}_{i,j}$. All the coagulation kernels are currently updated each timestep. However, this is computationally inefficient and the use of lookup tables with billinear interpolation in particle size is planned.

Condensation of water vapour and aerosol precursors gases (currently sulfuric acid and organics) is based on the Analytical Predictor of Condensation (APC) scheme by ? . The scheme first calculates the new vapour mole concentration as

$$C_t = \frac{C_{t-1} + \Delta t \sum\limits_{i=1}^{J} k_{i,t-1} S_{i,t-1} C_{s,i,t-1}}{1 + \Delta t \sum\limits_{i=1}^{J} k_{i,t-1}}, \tag{5}$$

where $k_{i,t-1}$ is the mass transfer coefficient in size bin i based on the current timestep, J is the total number of bins (including all particle categories), $S_{i,t-1}$ is the equilibrium supersaturation and $C_{s,i,t-1}$ is the saturation mole concentration over a flat surface. The new particle mole concentrations for each condensing vapour are then given in a semi-implicit form

$$c_{i,t} = c_{i,t-1} + \Delta t k_{i,t-1}(C_t - S_{i,t-1} C_{s,i,t-1}) \tag{6}$$

While the APC scheme is mass preserving and numerically stable, condensation and evaporation of water vapour on small droplets and especially small aerosol particles requires a very short timestep ($\ll 1$ s) to avoid non-oscillatory solutions. Since this goes beyond the practical range for the applications in this paper, where in general we aim towards a timestep around 1 s, two sets of measures are taken. First, for small aerosol particles with ambient relative humidity (RH) below 98 % the wet size of aerosol particles is determined as an equilibrium solution based on the molalities of different particle species (??) , and the APC equations are solved only above 98 % RH. Second, a simple substepping method is applied with Equations (??) and (??), where the substep length for cloud droplets is user defined and currently set as $\Delta t_c = 0.1$ s. For non-activated aerosol above the 98 % threshold for RH, even further timesplitting was found necessary and the timescale is $\Delta t_a = t_c/10$. The equilibrium saturation ratio is updated for each substepping cycle due to changing droplet/particle size (temperature is kept constant).

Although not used in the context of this paper, new particle formation by sulfuric acid is included in the model. There, the activation-type nucleation is formulated according to ? and the formation rate of 3 nm particles is calculated according to ? .

2.1.1 Cloud droplets

In the new extended SALSA, ~~additional size sections for cloud droplets and drizzle are implemented~~ cloud droplets are treated with a sectional description as well (Figure ??). Strictly speaking, to reproduce the evolution of the aerosol size distribution through cloud processing and wet scavenging ~~by precipitation~~ accurately, which is the goal of this work, a two dimensional dry/wet diameter bin system would be required. This is because cloud activation depends essentially on the dry aerosol size distribution, while collision processes and deposition rates depend strongly on the wet particle size. ~~However, the~~ Although such two-dimensional ~~framework is not a computationally feasible approach for applications such as~~ frameworks have been developed (e.g. **?**) , the approach is computationally highly demanding for large-eddy modelling ~~.~~ applications spanning timescales of days while covering relatively large domains with high spatial resolution, all of which are pursued here. As a compromise between accuracy and computational ~~burden~~cost, a unique strategy is proposed, where ~~the cloud droplet bins are defined to be parallel to~~ cloud droplets are described based on the dry size of the activated aerosol (i.e. cloud condensation nuclei, CCN) with the same prognostic bin quantities as for the aerosol bins. The particle diameters at the bin edges for the cloud droplet and non-activated aerosol regimes are set identical within their common size range (specifically, the 2a/b-bins ~~by default) in terms of the dry diameter of the activated cloud condensation nucleus (CCN)~~as a default setting) as shown in Figure **??**. Therefore, each cloud droplet bin is accompanied by a parallel aerosol bin. This way, the ~~properties~~ shape of the aerosol size distribution and the number concentration are preserved upon cloud droplet activation ~~,~~ as well as ~~evaporation of cloud droplets,~~ upon droplet evaporation though subject to the typical uncertainties inherent to the sectional approach (**?**). While the CCN dry diameter is known accurately (to the extent allowed by the spectral resolution of the size sections), the wet size of the cloud droplets, determined by Equations (**??**-**??**), represents a mean over each CCN size class.

~~In the extended SALSA, condensation of aerosol precursors as well as water vapour is solved for all particles and droplets using the analytical predictor method for condensation (?) . However, for water vapour below a relative humidity (RH) of 98 %, the wet size of aerosol particles is determined as an equilibrium solution based on the molalities of different particle species (??) , and the analytical predictor for condensation is solved above 98 % RH. The division of the water vapor condensation process is necessary due to the very fast relative changes in aerosol water content especially in subsaturated conditions and with small particles. Even close to saturation, a very small timestep ($\ll$ 1 s)is required for non-oscillatory solutions with small aerosol particles, which is not practical for the applications of this work. As a solution, a simple substepping method is employed in the condensation procedure, which takes approximately 50 substeps for every host model timestep (typically around 1 s).~~

Two methods are available for simulating the formation of cloud droplets in the extended SALSA. One is the parameterization by **?**, which takes as an input the aerosol properties and updraft velocity (along with atmospheric thermodynamic properties) to determine the maximum supersaturation in a parcel of air and thus the critical particle diameter for activation. Another is based on resolving the wet aerosol particle diameter: once the wet diameter of a particle exceeds the critical diameter corresponding to the resolved supersaturation from the host model, the particle is activated. Since the condensation of water vapour is solved dynamically for high RH, it is preferable to use the latter approach instead of the parameterized one for consistency in terms of the peak supersaturation ~~. This also allows cloud activation~~ and it is the approach used in the experiments of this work. This allows droplet activation also in other parts of the cloud ~~than~~ apart from the cloud base, e.g. due to radiative cooling effects at the cloud top or supersaturation caused by mixing of airmasses. However, if the vertical resolution of the host model is coarse (several tens of meters and above) it becomes necessary to use the parameterized method. With coarse ~~resolution~~ resolutions the supersaturation peak at the cloud base may be underestimated due to averaging effects, which yields underestimated cloud droplet number ~~concentration~~ concentrations (CDNC).

The relatively coarse spectral resolution of the aerosol bins may induce unwanted discontinuities in the ~~cloud activation calculation~~ activation spectrum with increasing saturation ratio due to the particle size discretization. To mitigate these effects, the extended SALSA accounts for the distribution of particle number and mass within the critical aerosol size bin using linearly fitted slopes between the bin centres (**?**) with both of the available methods for cloud activation.

Evaporation and deactivation of cloud droplets is accounted for through the resolved condensation, upon which activated aerosol particles are released back to the aerosol bin regime as illustrated in Figure **??**. For this to take place, a very simple diagnostic is used, where subsaturation with respect to water vapour is required and the cloud droplet diameter should be smaller than ~~20 %~~ 50 % of the critical diameter dictated by the properties of the CCN (or 2 μm at maximum). ~~These thresholds were obtained by physical reasoning and experimentation with the model .~~ Together with the representation of collision-coalescence processes by Equations (**??**)and (**??**), this enables the model to account for aerosol aging inside the clouds. However, please note that chemical processing of aerosol is not presently included in the extended SALSA.

2.1.2 ~~Drizzle~~Precipitation

Due to our strategy of ~~defining~~ describing the cloud droplet ~~size bins according to~~ distribution based on the dry CCN size, the wet ~~radius of the droplets~~ droplet diameter in each bin represents a mean ~~inside each sizebin. Although the~~ over all activated CCN of the corresponding size. Although the wet droplet size can be expected to be somewhat correlated with the dry CCN size, ~~not knowing the wet droplet diameter exactly yields an obvious setback~~ neglecting the variability in the dry/wet size relatioship is an oversimplification when predicting the mass and number of particles ~~considered as drizzle .~~ converted to drizzle droplets. Therefore, a type of autoconversion parameterization is formulated. Here, a lognormal distribution (selected because of mathematical simplicity) is assumed to describe the variation of the droplet wet size within each cloud droplet bin. The mode diameter is given by the ~~bin mean cloud droplet wet~~ known bin mean wet cloud droplet diameter and the geometric standard deviation is set as $\sigma_g^{ac} = 1.2$, which results in a relatively narrow distribution and is similar to the values used for the cloud droplet size distribution in the ~~default UCLALES~~ UCLALES with bulk microphysics. Setting a commonly used threshold diameter for drizzle droplets, $d_0 = 50\ \mu\mathrm{m}$, the number and mass concentrations of newly formed drizzle from the cloud droplet bins are obtained as an integral over the lognormal distribution from d_0 upwards.

The evolution of ~~the drizzle droplet population~~ precipitation is described with an additional set of size bins (Fig. **??**). However, since the growth of the drizzle droplets through ~~condensation and~~ collection processes is critical to ~~produce realistic~~ reach rain drop size and produce realistic surface precipitation rates, the ~~drizzle size~~ precipitation bins are defined according to the wet ~~droplet~~ drop diameter, different from the cloud and aerosol size bins. While the predicted bin properties are again similar to aerosol and cloud droplets, now the aerosol mass (instead of the mass of water) represents a mean for each precipitation size class. This is in contrast with our emphasis of tracking the aerosol size distribution properties, but is an acceptable compromise, since the number concentration of ~~drizzle droplets~~ rain drops is always much smaller than the concentration of cloud droplets or aerosols. Thus, their influence on the shape and chemical composition of the ambient aerosol size distribution upon ~~droplet~~ drop evaporation is not considerably obscured by the averaging effects acting on the properties of the aerosol particles embedded inside the ~~drizzle droplets. The drizzle~~ rain drops. The precipitation bins cover the size range from 50 μm to 2 mm.

This range is divided into 7 (currently fixed) sections with strongly non-uniform spectral resolution: ~~the width of the smallest binsis 5 μm and 1 mm in the largest bins~~up to the diameter of 100 µm (first 3 bins) the bin resolution gradually decreased from 5 µm to 35 µm and above the 100 µm range the resolution decreases from 100 µm to 1 mm.

~~The efficiency of rain collection of particles of different sizes is determined by coagulation kernels, for which Brownian diffusion with convective enhancement and gravitational settling are implemented~~Collection and scavenging of cloud droplets and aerosol particles by precipitation are treated by the coagulation (Equations **??**) and **??**) as well. Aerosol particles collected by ~~drizzle or rain drops accumulate~~ precipitation accumulates the aerosol mass inside the ~~drizzle~~ size bins. Upon evaporation of a drizzle ~~droplet~~or rain drop, it is assumed that a single particle is released (**?**) and it is placed in an aerosol bin with mean diameter closest to the released dry particle size. The size of the released particle is obtained simply based on the mass and the bulk density of the aerosol. This adds the contribution of drizzle formation on the aerosol processing in the model, albeit, again omitting the chemical processing.

2.2 Coupled UCLALES-SALSA

~~The UCLALES (?) model~~ UCLALES (**?**) is a large-eddy model based on the Smagorinsky-Lilly subgrid model. In the doubly periodic domain, advection of momentum variables is based on a fourth order difference equation with time-stepping by the leap-frog method. For scalars, simple forward timestepping is used. Prognostic variables in the UCLALES are the three wind components u, v, and w (with the standard meteorological notation), liquid water potential temperature θ_l and total water mixing ratio ~~r_T~~q_t, plus some additional prognostic scalars depending on the selected thermodynamic level (e.g. rain water). UCLALES contains three thermodynamic levels, which comprise dry, moist and precipitating thermodynamical models, the latter two of which are based on the saturation adjustment method. UCLALES does not include a description for aerosol. Rather, the microphysical processes are driven by a prescribed cloud condensation nuclei (CCN) concentration, taken to represent the cloud droplet number. The drizzle formation is given by **?** as

$$\frac{\partial q_r}{\partial t} = k_c q_c^2 x_c^2, \tag{7}$$

where q_r is the precipitation mixing ratio, q_c is the cloud condensate mixing ratio, $x_c = q_c/N_c$, where N_c is the CCN concentration, and k_c is a coefficient taking into account the droplet size distribution width and non-equilibrium effects (**?**) . Sedimentation of the drizzle and rain drops is determined by sedimentation velocity which depends on the diagnosed droplet size according to Eq (**??**).

Coupling the extended SALSA module into UCLALES yields extensive changes in the thermodynamic core of the model as compared to the ~~default version~~ version based on bulk microphysics, thus adding a new thermodynamic level (Level 4). With the coupled UCLALES-SALSA, condensation and evaporation of water vapour on cloud droplets, rain drops and aerosols is explicitly computed (Eq **??**). Therefore, instead of ~~r_T~~ q_t in case of the saturation adjustment method, Level 4 treats ~~water vapour r_v and condensate r_c mixing ratios~~ q_c, q_r and water vapour mixing ratio (q_v) as separate prognostic variables. This allows non-equilibrium conditions with respect to water vapour in UCLALES-SALSA, in contrast to the ~~default~~ standard UCLALES. θ_l

is retained as a ~~predicted~~ prognostic variable, which allows simple treatment of the latent heat transfer during moist adiabatic transitions.

UCLALES has an option to calculate cloud interaction with radiation using a four-stream radiative transfer solver (**?**). The radiation calculation accounts for the diurnal cycle and takes as an input the total number concentration of cloud droplets and the cloud water content. With UCLALES-SALSA, the total number of droplets and condensate mass are obtained as the sum over the cloud droplet size bins and used to calculate radiative transfer the same way as in ~~the default UCLALES~~ UCLALES (the aerosol fields are not coupled with radiation in the current model version).

~~The~~

2.3 Technical implementation

UCLALES-SALSA is currently implemented under the Fortran95 standard. Output files are written in NetCDF format. For parallel computing the Message Passing Interface (MPI) library is used and the parallellization strategy is based on two-dimensional horizontal blocking of the model domain.

Since the particle number concentrations as well as the masses of different compounds (aerosol species, liquid water) in each particle size bin constitute a prognostic variable, ~~raising~~ the number of ~~prognostic scalars to $\mathcal{O}100$~~ advected scalars is increased from a maximum of 3 in UCLALES to $\mathcal{O}(100)$ in UCLALES-SALSA even with a simple ~~sulphate-based~~ sulfate-based setup. This obviously has ~~an~~ a strong impact on the computational ~~burden - the~~ cost. The model runs at about real-time with a Cray XC30 supercomputer using a decomposition with 8x8 grid points per MPI process. While this is a substantial constraint on the applicability of the model, short 12-24 hour (model time) simulations are ~~feasible~~ still easily performed and in the following sections we will show that the presented methods are necessary to improve our understanding about boundary layer clouds, fogs and aerosols.

3 DYCOMS-II

3.1 Case description and model configuration

The new UCLALES-SALSA is first configured and tested based on the case DYCOMS-II flight RF02 (**?**), which took place off the coast of California in July, 2001. The observations conducted in this case featured a mix of open and closed cell stratocumulus structures, with strong drizzle associated with the ~~first~~former. For the model setup we follow the settings defined by **?**: ~~In~~ in all simulations, the initial profiles of liquid water potential temperature θ_l, total water mixing ratio ~~r_t~~ q_t (taken as supersaturated vapour in the model initialization process), and u and v wind components were specified with the following equations.

$$\theta_l = \begin{cases} 288.3\ \mathrm{K} & z < z_i \\ 295 - (z - z_i)^{1/3}\ \mathrm{K} & z \geq z_i \end{cases} \tag{8}$$

$$\cancel{r}q_t = \begin{cases} 9.45\ \mathrm{g\ kg^{-1}} & z < z_i \\ 3 - 5\left(1 - \exp\left((z - z_i)/500\right)\right)\ \mathrm{g\ kg^{-1}} & z \geq z_i \end{cases} \tag{9}$$

$$u = 3 + 4.3z/1000\ \mathrm{m\ s^{-1}} \tag{10}$$

$$v = -9 + 5.6z/1000\ \mathrm{m\ s^{-1}} \tag{11}$$

In the above, z_i is the initial inversion level set at 795 m. In addition, a large-scale ~~subsidence rate~~ divergence of $3.75 \times 10^{-6}\ \mathrm{s^{-1}}$ is assumed, together with prescribed latent and sensible heat fluxes of 93 and 16 W $\mathrm{m^{-2}}$, respectively.

The simulations span 10 hours~~, the first hour of which~~. The first hour is considered as the spinup period~~. In UCLALES-SALSA,~~ , during which drizzle formation and ~~coagulation of particles are not allowed during this period in order to prevent~~ all the collision processes are turned off, while cloud activation and condensation processes are active. This prevents spurious effects on the cloud properties during the initial buildup of turbulent kinetic energy (TKE) and settling of the boundary layer properties. The simulation domain spanned 5 km into each horizontal direction and 1600 meters in the vertical, with the topmost 200 meters used as a sponge layer, damping unrealistically reflected gravity waves at the model top. The horizontal resolution is set to 50 m while the vertical resolution is 20 m. The model uses an adaptive time step, whose maximum value is set to 1 s. During events of strong mixing in the course of the model run, the timestep was occasionally reduced to about 0.5 s. A more detailed description of the model experiments is given below and their key aspects are summarized in Table **??**. The performance of the UCLALES-SALSA model is evaluated by comparing the results with those ~~from the default UCLALES~~ obtained from similar runs with UCLALES using bulk microphysics as well as field measurements. This can also be contrasted to the model ensemble used in the LES intercomparison in which ~~the default~~ UCLALES was a part of (**?**). Thus we can isolate and characterize the effects induced by the use of an elaborate sectional microphysical scheme for aerosols and clouds.

3.1.1 ~~Default~~ Reference case experiments

The ~~default~~ reference experiments are based on the basic settings in terms of aerosol and cloud microphysics. For the experiment performed with UCLALES-SALSA, designated as LEV4, this means that we use the two-mode lognormal initial aerosol size distribution given in **?**, which is assumed to consist of sulphate aerosol. The total number, geometrical mean diameter and geometrical standard deviation are $125\ \mathrm{cm^{-3}}$, $22\ \mathrm{nm}$ and 1.2 for the first mode and $65\ \mathrm{cm^{-3}}$, $120\ \mathrm{nm}$ and 1.7 for the second mode. In the model ~~initialization~~startup, the size distribution is remapped into the SALSA aerosol size bins. For comparison with the experiment LEV4, a parallel experiment, designated LEV3, is performed with the ~~default UCLALES configuration, including moist thermodynamics and drizzle~~UCLALES configuration using bulk cloud microphysics. However, since ~~the default~~ UCLALES does not contain a description for aerosols, the CCN number concentrations must be prescribed, similar to most other available LES models. In LEV3, the CCN (i.e. cloud droplet) concentration is set ~~as~~ to $55\ \mathrm{cm^{-3}}$, which roughly corresponds to the number of cloud droplets initially produced by LEV4 and is also the number used in other LES simulations based on this particular case (**??**).

3.1.2 Sensitivity tests

A set of sensitivity tests are performed to further investigate certain aspects of the model. Experiments designated as LEV4HI and LEV3HI are performed. These are similar to LEV4 and LEV3, but with higher aerosol (or CCN for LEV3HI) concentration (mode number concentrations multiplied by 3), and are utilized to study how the coupling between the model microphysics and dynamics reacts to perturbations in the initial aerosol and cloud properties.

3.2 Results

3.2.1 General features

Figure **??** shows a domain mean time-height plot of the liquid water content (LWC) in the LEV3 and LEV4 experiments. While in the early stages of the simulation the LWC and the macroscopic cloud structure are quite similar between LEV3 and LEV4, after about 4 hours the results start to diverge substantially, marking a clear shift in the boundary layer dynamics. Whereas the LEV3 simulations maintain a solid stratocumulus deck until the end of the simulated period, LEV4 results in a very thin stratiform cloud deck just below the inversion with low LWC and only 5-10 cm^{-3} cloud droplets. However, in the last couple of hours of the simulation, this setting is interspersed by occasional ~~deeper and more massive shallow convective cumulus elements .~~ cumulus elements with base height around 400 m. The thin filaments below the stratiform cloud shown in Figure **??**b are the result of these elements, although they appear weak due to the horizontal averaging (the cumulus clouds were also confirmed from 3-dimensional fields, although not shown here). This is reminiscent of the formation of open cell circulation structures in marine ~~Sc~~ stratocumulus clouds (**?**), which were also observed during RF02 (**?**).

Figure **??** shows the total liquid water path (LWP, taken as cloud droplets plus ~~drizzle~~precipitation) and the rain water path ~~(liquid water interpreted as drizzle in the model)~~ for LEV3 and LEV4. Again, the LWP is fairly similar between the two experiments during the first 4 hours ~~, after which~~ and it also agrees quite well with the observed mean LWP, as shown in the figure. After about 4 hours, LEV4 starts to deviate from LEV3. However, in a later stage, a substantial portion of the total LWP is interpreted as ~~drizzle~~ precipitation in LEV4, while in LEV3 the ~~water mass considered as drizzle~~ mass of precipitation is much smaller. This is ~~seen mainly as~~ mainly due to a diagnostic discrepancy: in LEV4 most of the excess ~~drizzle~~ precipitating droplets reside within the cloud layer ~~in the smallest drizzle bin, which do not yet exhibit notable fall speeds and are evaporated quickly~~ and partition into the smallest bin, where fall speeds are low and droplets quickly evaporate after descending below the cloud layer. This stems from the details in parameterizing drizzle formation. Differences arise for example from the fact that when large cloud droplets (>50 µm) are considered as drizzle in UCLALES-SALSA, they are transferred to the smallest ~~drizzle~~ precipitation bin, beyond which their growth is explicitly modelled (though subject to low bin resolution). Instead in LEV3, a size distribution (based on gamma function) is assumed for ~~the drizzle droplets~~precipitation, which causes at least a part of the ~~drizzle amount~~ precipitating droplets to reach surface-reaching ~~size range very quickly compared to~~ rain drop sizes much faster than in LEV4. Figure **??**a shows that despite the difference in the ~~drizzle~~ rain water path, the surface precipitation rate is of similar order of magnitude between LEV3 and LEV4~~(after considering the spinup period).~~. The results are also for a large part within the observed range as shown in **?** . This is used as the main criterion for setting up the model parameters such as σ_g^{ac}~~for the basic experiments~~. Nevertheless, even after considering the differences in drizzle~~amount~~, it is evident, that the boundary layer and cloud properties in LEV4 shift towards a very different state as compared to LEV3.

3.2.2 Boundary layer structure

In the LEV4 experiment, the boundary layer shows somewhat more stratified characteristics than that in LEV3. Figure **??** plots the domain mean vertical profiles of potential temperature, water vapour and liquid water mixing ratios. Especially towards the end of the simulation, LEV4 shows a rather distinct division of the ~~boundary-layer~~ boundary layer into two separate mixing regimes. This decoupling of the cloud-driven layer (see for reference the conceptual models e.g. in **?**) is evident in both the potential temperature as well as water vapor mixing ratio, with the sharpest gradient taking place around 400 m height, following the criteria defined in **?**. In LEV3 the temperature profile is weakly stable as well after 9 hours of simulation, but less so than in LEV4. In particular, the water vapor mixing ratio in LEV3 does not show the same separation as LEV4.

It is typical for a stratocumulus topped boundary layer to shift towards a decoupled structure in the morning as shortwave heating by the rising sun begins to offset the longwave cloud top radiative cooling and therefore reduces cloud driven mixing (**?**). Although wind shear may also affect the entrainment and cloud top static stability (**?**), it is not surprising that the sharpest transition in LEV4 occurs around 5 hours into the simulation, which is also close to sunrise at the assumed location. It is also noted that after the initial shift, the decoupled structure is subject to positive feedbacks as it reduces the supply of moisture from the surface to the cloud layer, which further reduces the cloud top radiative cooling and thus the cloud driven mixing. This also weakens the cloud top inversion, allowing transport of heat to the upper mixed layer by entrainment. Due to the stable layer at the decoupling interface, heat transferred to the cloud-driven layer is not efficiently mixed, resulting in even more pronounced decoupling of the cloud layer. By the same ~~token~~argument, a larger portion of moisture released from the surface by the latent heat flux is confined to the surface layer, thus contributing to the relatively high water vapour mixing ratio in LEV4 as compared to LEV3.

3.2.3 Role of microphysics and drizzle

Even though LEV3 and LEV4 simulations are subject to identical external forcings, LEV3 does not show as abrupt changes in the simulated cloud layer nor the ~~boundary-layer~~ boundary layer structure as LEV4 does, indicating that something makes the LEV4 ~~boundary-layer~~ boundary layer more susceptible to undergo the decoupling process. As discussed next, the reason for the initial perturbation towards this different state can be traced back to the representation of microphysics and ~~drizzle.~~ precipitation.

Figure **??** shows the surface precipitation rate in LEV3 and LEV4 simulations as well as the rate of removal of sulphate aerosol embedded inside precipitating droplets in UCLALES-SALSA, illustrating the ~~models~~ model's ability to resolve the aerosol wet scavenging process. The UCLALES-SALSA performs this task with very high detail: the size distribution of aerosols is preserved through activation scavenging ~~, beyond~~ after which droplet growth and subsequent drizzle generation favors large soluble particles.

~~The~~ The aerosol scavenging by cloud activation is clearly visible as a reduction in aerosol number concentration ~~due to cloud activation~~ in LEV4 ~~is clearly visible immediately after the model initialization~~ already during the model spinup as shown in Figure **??**. After a couple of hours of ~~drizzle~~ precipitation formation in LEV4, the consequences of aerosol scavenging by ~~activation and drizzle~~ drizzle and rain fallout become visible in the ~~below cloud layer as the aerosol number concentration decreases quite rapidly~~ below-cloud layer as well. Scavenging by precipitation is treated as a coagulation process between the rain drops and aerosols both in the ~~cloud and in the~~ in-cloud and below-cloud layers. ~~Since the scavenging is size resolved, and favors larger particles,~~ Upon collision, the mass of the aerosol particle is moved to the rain drop bin in question, and removed from the aerosol bin along with the corresponding number concentration. The reduction in the number of potential CCN ~~and thus CDNC is strongly reduced with time. This~~ indirectly supports continuing production of drizzle ~~droplets, which eventually cover~~ through reduced competition for water vapour between cloud droplets. Eventually, drizzle covers a considerable fraction of the total droplet concentration within the stratiform cloud layer. The scavenging of particles and the ~~consequent~~ reduction in cloud water content due to drizzle start to weaken cloud top radiative cooling already during the first few hours of the simulation in LEV4. In contrast, in LEV3 such ~~transition towards~~ a transition towards a thinner cloud layer with lower cloud water content does not take place, because of the lack of representation for aerosol scavenging. This marks a distinct change in the interpretation of the model, since the use of prescribed CCN concentration in LEV3 implies an infinite supply of particles advected to the domain. For LEV4 this is not true and in the absense of emissions (aerosol emissions are not implemented in this model version) the aerosol is gradually depleted by scavenging, which is more reminiscent of the model domain moving with the flow (Lagrangian modelling approach).

Because of the low aerosol concentration and sufficient amount of water available in the model initial state, the presented case favors considerable drizzle production. Considering the ~~interactive~~ detailed description of particles and the ~~detailed~~ aerosol removal mechanisms included in the model, the results shown here are not scientifically surprising, but are used to demonstrate the model's ability to reproduce the transitions in ~~boundary-layer~~ boundary layer and cloud structure due to microphysical interactions. Interestingly, ~~the~~ the reduced cloud water and the drizzle maintained by the depletion of aerosol, as shown by Figure **??** for LEV4~~results on LWP and rain water path show quite similar features as those obtained~~ , resemble the corresponding effects shown by (**?**) for low aerosol simulations performed with a cloud ~~system resolving modelwith interactive aerosols (?)~~ resolving model. However, UCLALES-SALSA provides the means for more detailed investigations ~~about~~ of the impact of the particle size distribution and composition on cloud dynamics and aerosol-cloud interactions, which justifies the added complexity and computational demand. This is demonstrated ~~in more detail by Figure ??~~by Figure **??**, showing the ~~relative change in particle number concentrations for individual bins (combining both non-activated aerosol and activated CCN particles) in the in-cloud and below-cloud layers. It is shown that after the spinup period in the below-cloud layer, the smaller particlesare removed by precipitation collection and, in part, also by activation in air parcels caught in updrafts. However, the number of the largest particles present increases, which is due to evaporating drizzle below the cloud. These droplets have had time to collect additional aerosol mass during their growth, which upon the droplet evaporation is released as a single large particle, preserving~~

~~the mass of the aerosol. Note that while~~ size distributions of activated and non-activated particles for two heights right after the 1-hour spinup and after 8 hours into the simulation. It clearly shows the impact of activation on the large diameter end of the distribution in the beginning of the simulation, as well as the fact that the total distribution (activated plus non-activated particles) in the cloud layer corresponds well to the dry aerosol distribution at lower levels. After 8 hours of simulation, the depletion of activation sized particles is evident as well, together with a small increase in coarse mode particles at low levels due to particles released from evaporating drizzle and rain drops. An additional example is given by Figure **??**, showing the relative change ~~appears large, the initial number of particles in these bins is relatively small, about $1-10$ cm^{-3}. Inside the cloud the particles are removed by activation , cloud collection and the subsequent wet deposition. Since the~~ in particle number concentrations for individual bins averaged over the in-cloud and below-cloud layers. The effects of cloud activation and scavenging by drizzle and rain are clearly seen here as well. In particular, the increase of large particles in the below-cloud layer due to rain evaporation is seen here much more clearly than in Figure **??**.

Since UCLALES-SALSA includes a variety of processes that directly influence the size distribution and composition of aerosol particles, this also affects the distribution and variation of the mass of soluble material inside cloud droplets. This, at least in the initial phase of droplet formation, contributes to ~~the~~ their growth rate which may affect ~~drizzle~~ precipitation formation. In this context, the detailed description of the evolution of the aerosol size distribution provided by the model also enables the investigation of aerosol particle emissions, e.g. giant sea salt particles, and their influence on ~~the~~ cloud properties and ~~drizzle~~precipitation.

3.2.4 Impact of initial particle concentration

Since it is apparent that drizzle formation and the subsequent impacts of particle scavenging yield the divergence of results between the LEV3 and LEV4 simulations, it is necessary to test how changing the particle number concentrations affects the results. This is done simply by repeating the LEV3 and LEV4 experiments with particle concentrations multiplied by three, designated as LEV3HI and LEV4HI. With higher particle concentrations, the precipitation reaching the surface is very small ~~or non-existent~~ in both simulations, which suppresses the wet scavenging effect in LEV4HI. As a result, the cloud properties in LEV4HI remain quite close to those in LEV3HI during the simulated period. This is seen in the domain mean profiles of LWC and the boundary layer thermodynamical properties, shown in Figure **??**, which are indeed remarkably similar between the two experiments. This shows that in conditions where the additional processes and interactions in the new UCLALES-SALSA are not dominating the ~~boundary-layer~~ boundary layer and cloud evolution, the results remain physically consistent with the more simple model versions. It should be noted though, that if the LEV4HI simulation would be continued over an extended period of time, the supply of moisture by the (constant) latent heat flux and the effects of cloud processing ~~and coagulation~~ on the aerosol size distribution would eventually create drizzle and rain, which would then lead to a similar situation as seen in the experiment LEV4. It has been shown that maintaining a steady-state cloud structure requires aerosol replenishment from multiple sources, including aerosol emissions (**?**). Although there is some aerosol replenishment through mixing from the free ~~atmosphere~~ troposphere in our model experiments, this is not enough to maintain the cloud deck over prolonged periods of time. Considering the outcomes of the experiments LEV4 and LEV4HI, the model results here are consistent with the findings of **?** regarding the effects of aerosol replenishment. Thus, implementation of aerosol emissions into UCLALES-SALSA is part of our future plans.

4 Simulating fog formation and evolution

4.1 Case description and model configuration

To demonstrate the versatility of UCLALES-SALSA, the model is configured according to the conditions from a radiation fog event that took place at the UK Met Office research site at Cardington in the night of $12-13^{\mathrm{th}}$ of February, 2008 (**??**). Simulations using different aerosol concentrations and horizontal wind profiles are ~~introduced~~ described to illustrate the potential effect of aerosol and wind shear on the properties of radiation fog.

In addition to adapting the model initial conditions (temperature, humidity and wind profiles, aerosol size distributions) for this particular case, the main differences in the model configuration here, as compared to the DYCOMS-II case in Section 3, have to do with spatial and temporal resolution and the surface forcing. Here, the model is run with a very high resolution, vertically spanning $1.5\ \mathrm{m}$ in the lowest $150\ \mathrm{m}$. Above, the resolution is gradually decreased so that the model top is at approximately $800\ \mathrm{m}$ with the total number of levels being 165. The horizontal resolution is $4\ \mathrm{m}$ in each direction and the domain covers an area spanning $320\ \mathrm{m}$ by $640\ \mathrm{m}$. The timestep is set to $1\ \mathrm{s}$ as in the stratocumulus case. However, the adaptive timestep reduces down to around $0.2\ \mathrm{s}$ during the simulation. This is somewhat less than the minimum adaptive timestep length in the stratocumulus case, which is expected due the higher horizontal resolution used here.

In contrast to the experiments in Section 3, the surface heat fluxes are not prescribed, but are determined with a simple parameterization for soil energy balance, which is coupled with the radiation scheme (**?**) . Moreover, the scheme accounts for the heat transfer between surface and deeper soil. For the latent heat flux the surface is assumed to be saturated with respect to water. This can be assumed to be a fairly good approximation until the fog dissipation phase, when the evaporation from the warming surface can deplete the water from the surface layer and the assumption of a saturated surface may overestimate the latent heat flux. Due to the simplicity of the surface energy balance model, to produce reasonable surface cooling rates with respect to observations (**?**) , the equation for surface heat capacity was tuned to yield values on the order of $1000\ \mathrm{J\,kg^{-1}\,K^{-1}}$, which also depends on the soil water fraction.

The settings for microphysics in the UCLALES-SALSA run are kept similar to those used in the DYCOMS-II case (Section 3), with the exception that drizzle formation ~~was~~ is switched off in the fog simulations. This is justified due to the fact that the liquid water content in the fog remains relatively low and the sedimentation of cloud droplets ~~was~~ is the main sink of cloud water. ~~While~~ This setting also conforms with the model setup by **?** . In addition, while droplet number concentrations were prescribed in the simulations performed by **?**, ~~here,~~ in UCLALES-SALSA the droplet activation is computed based on the growth of the aerosol particles to sizes larger than their critical ~~radius at~~ diameter at the water vapour supersaturation, which is resolved by the model. ~~This is important~~ While this method for cloud activation was also used in the experiments in Section 3, it is particularly important here, since in radiation fogs the droplet formation is mainly driven by the radiative cooling at the top of the fog layer. Solving the condensation equation (Eq **??**) also allows the evaporation of cloud droplets inside the fog if the water supersaturation falls below the ~~water surface pressure~~ equilibrium saturation ratio in the smallest cloud droplet bins. This process can reduce the number of droplets and has been found to take place also in clouds (**??**).

~~In contrast to the experiments in Section 3, the surface heat fluxes are not prescribed, but are determined with a simple parameterization for soil energy balance, which is coupled with the radiation scheme (?) . Moreover,~~ Note, that no spinup period in terms of the configuration of the microphysical processes is used since here it generally takes a few hours from the ~~heat transfer between surface and deeper soil is approximately taken into account (?) . For the latent heat flux the surface is assumed to be saturated with respect to water. This can be assumed to be a fairly good approximation until the fog dissipation phase, when the evaporation from warming surface can deplete the water from surface layer and the assumption of saturated surface may overestimate the latent heat flux. Due to the simplicity of the surface energy balance model , the surface heat capacity is used as a tunable parameter to reproduce reasonable surface cooling rate in comparison with observations~~start of the model run for the fog to emerge.

4.1.1 Experiments

The impact of aerosols on fog formation is first investigated by three parallel experiments with zero initial horizontal wind velocities, which differ in their initial particle concentration. As the information ~~of~~ about aerosol concentration is not available for ~~the chosen simulation~~this study, we use a bimodal aerosol size distribution with mean sizes of $50\ \mathrm{nm}$ and $150\ \mathrm{nm}$. In all simulations the number concentration in the Aitken mode is kept ~~in~~ at $1000\ \mathrm{cm}^{-3}$ while the number of accumulation mode ~~aerosols~~ particles is increased consecutively so that the accumulation mode particle concentrations are 200, 400 and $800\ \mathrm{cm}^{-3}$ in experiments A200, A400 and A800, respectively. An additional experiment, A400W is then presented, where the model is initialized with the horizontal wind data from (**?**). The list of experiments is summarized in Table 2.

4.2 Results

Similar to the observation-based reports by **?** and LES studies (**??**), the fog layer investigated here undergoes distinct thermodynamical transitions during its evolution. Initially, the fog forms near the surface in a very stable layer due to the longwave cooling effect. As the fog-layer encroaches upwards and more droplets are activated at the fog top layers, its optical thickness increases which reduces the radiative cooling effect at the surface. At the same time the peak of radiative cooling at the fog top region becomes more pronounced. Figure **??** shows the evolution of the fog droplet concentration (sampled at 10 meters height) and the growth of the fog layer thickness. For the experiments A200, A400 and A800 (initialized with zero horizontal wind) the increase in the number of droplets due to the increasing aerosol concentration is clearly seen. ~~Higher~~ A higher initial aerosol concentration yields an increased fog layer depth, but the differences between the experiments are minor. This is due to the stability of the temperature profile, which suppresses the mixing especially with low aerosol ~~concentration~~concentrations, as show in Figure **??**.

In the early morning there is a transition from stable to almost neutral temperature stratification inside the fog (Fig. **??**). Higher aerosol concentrations promote increased optical thickness of the fog layer, which leads to faster formation of the neutral temperature profile. This is qualitatively similar to the results presented in **?** and is attributed to the reduction in the surface longwave cooling effect with optically thick fog layers and to the supply of heat from the soil. As can be seen from Figure **??**, the earlier formation of a neutral temperature profile with higher aerosol load further enhances the aerosol effect on fog droplet concentration (04 UTC in A800) through a positive feedback similar to what has been found to take place at the top of fog, where the increase in droplet concentration enhances radiative cooling which again feeds back as a higher supersaturation and enhanced particle activation (**?**).

Figure **??** shows the profiles of radiative cooling rate and the water vapour supersaturation as a function of time for the three experiments. As expected, the peak radiative cooling is indeed found near the top of the fog layer. Moreover, the intensity of the cooling increases with increasing aerosol concentration, owing to the higher optical depth: in A800 the peak cooling rate is approximately $7\ \mathrm{K\,hr^{-1}}$ and in A200 $4\ \mathrm{K\,hr^{-1}}$. This is in agreement with the range of values reported in **?**. The peak water vapour supersaturation is found at the same altitudes as the strongest radiative cooling. However, ~~larger particle concentration depletes~~ as larger particle concentrations deplete the available water vapour more efficiently, ~~resulting in highest supersaturation to occur~~ the highest supersaturations occur in the experiment with the lowest particle ~~concentrations~~concentration (A200).

These findings illustrate the ability of ~~the~~ UCLALES-SALSA to provide a realistic description of not only the thermodynamic and microphysical properties of fogs, but also the ~~aerosol-fog-radiative~~ aerosol-fog-radiation interactions and feedbacks on the dynamics. The results from the experiments A200, A400 and A800 compare quite well with those reported in **?**. This includes the rate of growth of the fog layer depth, despite the fact that their simulations were initialized with ~~the~~ non-zero horizontal wind profiles. However, the growth rate is considerably lower than ~~the observed~~in the observations, where the fog top reaches about ~~a~~ $100\ \mathrm{m}$ within 7 or 8 hours from the first appearance of the fog (see figure 5 in **?**). For UCLALES-SALSA, this is presumably because of the lack of shear generated turbulence. Wind shear has been shown to be very important in controlling the turbulence characteristics inside radiation fogs (**?**). Thus, in the additional experiment A400W, ~~the~~ UCLALES-SALSA is initialized with an approximately similar wind profile as in (**?**). Interestingly, in this case the growth of the fog layer corresponds much more closely to the observed, as shown by the dashed line in Figure ~~**??**~~**??**. The wind shear present in A400W (Figure **??**) yields vertical mixing, which strongly enhances the droplet production within the fog layer even at the initial phase (Figure **??**). The mixing and perturbations in radiative heating, as compared to the zero-wind experiments, produce the neutral temperature stratification quite quickly and the strength of the inversion at the top of the fog is also slightly reduced, as shown by Figure **??**. This allows more rapid growth of the fog layer, the depth of which reaches over $150\ \mathrm{m}$ by morning. This is even deeper than suggested by the observations, and can be attributed to e.g. missing advection effects or possible differences in the initial moisture or temperature profiles. At the same time the increased mixing enhances droplet activation and decreases the differences ~~between different aerosol concentrations~~caused by changing the initial aerosol concentration (not shown).

~~These~~ The results point towards the importance of a detailed representation of the microphysical processes ~~.~~ in cases of fog formation. In particular, the ~~size resolving~~ size-resolving microphysics in UCLALES-SALSA ~~does well in reproducing~~ result in a peak number concentration in the fog droplet size distribution ~~. The peak number concentrations are generally found for droplet diameters between 20 µm and~~ at approximately $25\ \mathrm{\mu m}$ in terms of the wet diameter, which agrees ~~well with measurements (?) .~~ with the observed range between $20\ \mathrm{\mu m}$ and $25\ \mathrm{\mu m}$ based on the measurements presented in **?** . This has many positive implications, since realistically capturing the droplet growth is important for representing the droplet sedimentation, which is an essential driver for the fog evolution. ~~As the~~ The droplet number concentration ~~simulated by UCLALES-SALSA also~~ in the experiment A400 (Figure **??**) agrees quite well with ~~observations, together these processes contribute to accurate estimation of the liquid water content and fog optical depth as well as their vertical distribution . These are then~~ the observed range ($20\,\mathrm{cm}^{-3}$ to $60\,\mathrm{cm}^{-3}$) illustrated in **?** as well before the fog layer rises to form low level stratus in the morning. Similar values are also seen for A400W in the development phase before midnight. However, after midnight the droplet number concentration increases substantially due to activation of even smaller particles as the mixing intensifies because of the wind shear and reduced stability. The resulting high droplet concentrations owe at least in part to the fact that detailed information about the aerosol size distribution was not available. Nevertheless, it is clear that these microphysical aspects are directly linked to the fog and ~~boundary-layer dynamics. Increased~~ boundary layer dynamics. An increased fog optical depth due to an increased droplet concentration will delay fog evaporation in the morning after sunrise, which thus ~~connect~~ couples the aerosol concentration ~~into fog existence~~with fog occurrence. However, to fully evaluate the aerosol effect on fog lifetime, a more detailed land surface scheme is needed to correctly simulate the latent heat flux and atmospheric water content after sunrise.

5 Conclusions

~~A new large-eddy simulation model coupled with a fully interactive bin-microphysical scheme for aerosols and clouds~~ The implementation of a novel bin-microphysics scheme for aerosol, clouds and precipitation in an LES model was presented. The coupled model is based on well-established components: the UCLALES large-eddy simulation model and the SALSA aerosol model, extended with cloud droplets and ~~drizzle~~rain. The bin system for ~~aerosols~~ aerosol and clouds follows a unique approach, where the size bins are defined according to the dry particle size for both activated and non-activated particles in an attempt to hold detailed information about the aerosol size distribution both in ambient air and within clouds. This also enables an elaborate description of the effects of cloud processing through collision-coalescence on the properties of the aerosol population as well as a size and composition-resolved simulation of the wet scavenging of ~~aerosols~~aerosol.

The model was tested and evaluated using two well-characterized cases which have also been simulated with LES models in previous work: one comprising marine stratocumulus clouds from the DYCOMS-II campaign and another based on measurements of a radiation fog event in Cardington, UK. For the stratocumulus experiments, ~~the~~ UCLALES-SALSA initially produced very similar cloud and boundary layer properties as other LES model versions, many of which rely on bulk microphysics and prescribed particle or droplet concentrations. However, after about 5 hours, UCLALES-SALSA shifted towards a very different ~~boundary-layer~~ boundary layer state, as compared to the ~~default LES~~standard version of UCLALES, resulting in a thin stratiform cloud deck at the top of a decoupled layer instead of a solid stratocumulus cloud layer. This shift was attributed to the wet removal of aerosol particles ~~with drizzle~~through precipitation, which eventually led to a decrease in cloud droplet number and water content. This enhanced the susceptibility of the boundary layer to undergo a significant decoupling~~when~~, which was triggered by the change in radiation budged during sunrise, which then yielded even more dramatic shift in the cloud properties, forming a feedback loop. Such behavior was not reproduced by the ~~default~~ standard UCLALES nor by most of the models used in **?**, which is due to the use of prescribed microphysical properties and the lack of interactions treated by the model. While the transition in the cloud properties simulated by ~~the~~ UCLALES-SALSA resembles ~~that related to~~ closed-to-open cell transitions in marine stratocumulus, it is noted that the rather small model domain (5×5 km) is much too small to represent the circulation dynamics and feedbacks closely related to the real-world mesoscale morphological transitions. Nevertheless, the results are encouraging and show that the model may very well provide the necessary new information related to ~~aerosol-cloud-drizzle~~ aerosol-cloud-precipitation interactions in future studies to explain the observed stratocumulus characteristics.

In another set of experiments, the skill of the model in simulating fog formation and development was shown. The model was able to capture the evolution of the fog radiative properties and the resulting changes in the thermodynamical profiles. While increasing the initial aerosol concentration had only slight impact on the growth of the fog layer depth, larger particle ~~concentration~~ concentrations did clearly affect the rate of evolution of the temperature profile, which showed a transition from very stable ~~conditions to~~ conditionsm to an eventually almost neutral profile. This is qualitatively in agreement with the observed behavior ~~**?**~~ (**?**) . While the growth of the fog-layer depth was clearly underestimated, as compared to observations, when the model was initialized with zero wind speeds, setting a realistic wind profile resulted in a growth rate very similar to the observations. With horizontal wind present, the formation of a neutral temperature stratification is even more pronounced than with zero wind conditions, and even more resembles the observed properties. **?** identified advection and drainage flows as plausible explanations for the discrepancy between their model and observations. The results presented in this study also bear these deficiencies and are also affected by other shortcomings, such as the surface scheme which is most likely ~~over simplified~~over-simplified. The remaining differences between the radiation fog simulated by UCLALES-SALSA and the observations notwithstanding, the results of this study still make a strong point for a very detailed representation of aerosol and cloud microphysics in simulating the fog evolution.

The need for high-resolution models that can accurately simulate the ~~interactions between~~ effects of aerosol-cloud interactions on both aerosols and clouds ~~in both ways~~ and couple these effects to the dynamical features of the atmosphere is clearly highlighted by the current challenges e.g. in climate research. UCLALES-SALSA provides these abilities making it a highly sophisticated, yet computationally relatively efficient alternative to investigate the role of ~~aerosols~~ aerosol in marine stratocumulus clouds or fogs, or the process of wet scavenging. Although the model is currently limited to warm clouds only, implementation of ice processes is on the way and will be published in a separate paper. Work is currently done also to add treatment of semivolatile aerosol species in the model and to couple the aerosol fields with radiation computation. This will extend the repertoire of the model ~~also~~ towards more elaborate studies of the aerosol-cloud interactions as well as towards ice and mixed-phase clouds, whose representation in climate models and the deficiencies therein have recently started to attract more widespread interest.

Code availability

The model ~~code is available upon request and will be made more generally available by the time of release of the final version of this paper .~~ source code and input files needed to reproduce the simulations presented in this paper can be downloaded from Github at https://github.com/UCLALES-SALSA.

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Table 1. ~~Model~~ The DYCOMS-II model experiments with their key configuration details.

Experiment	SALSA	Particle number (cm^{-3})*
LEV3	off	55
LEV4	on	190
LEV3HI	off	165
LEV4HI	on	570

*This is the prescribed CCN concentrations when SALSA is not used, otherwise the total initial aerosol number concentration.

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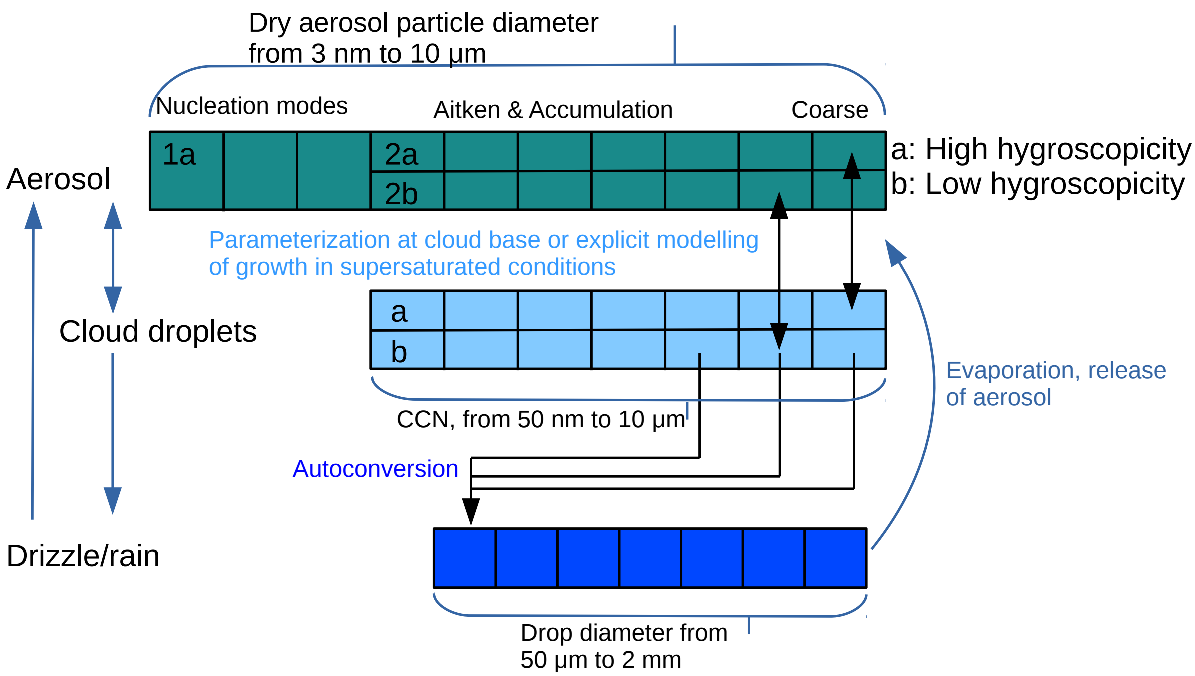

Figure 1. Schematic representation of the bin ~~layout~~ system and processes included in the extended SALSA module. Aerosol ~~and cloud droplet~~ bins (green) cover the size range from 3 nm to 10 µm, separated into bin regimes 1a, 2a and 2b (see text). Cloud droplet bins (light blue) are parallel ~~with each other and follow~~ to the aerosol bins in terms of the ~~same size range for~~ dry ~~particle~~ CCN diameter above 50 nm (~~from 3 nm to 10 µm~~)i.e. ~~In contrast,~~ the ~~drizzle~~ aerosol bin regime 2a/b). Precipitation bins (dark blue)~~are~~, defined according to the wet diameter of the droplet~~and~~, cover the size range between 50 µm and 2 mm.

Table 2. Radiation fog model experiments with their key configuration details. N_{acc} is the number concentration of accumulation mode particles.

Experiment	N_{acc} [cm^{-3}]	Wind profile
A200	200	zero
A400	400	zero
A800	800	zero
A400W	400	Porson et al. (2011)

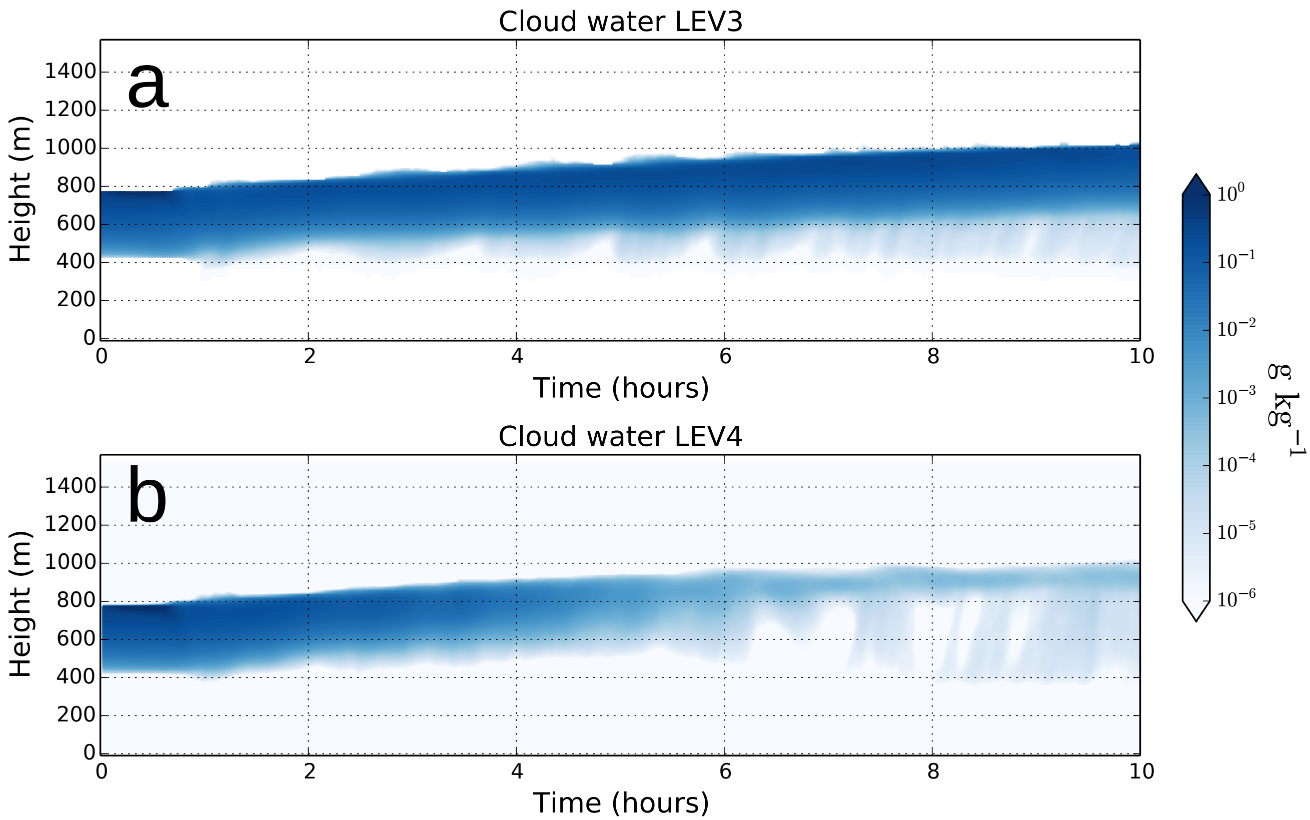

Figure 2. Time-height cross section of the cloud water content for LEV3 and LEV4 simulations in g kg^{-1}.

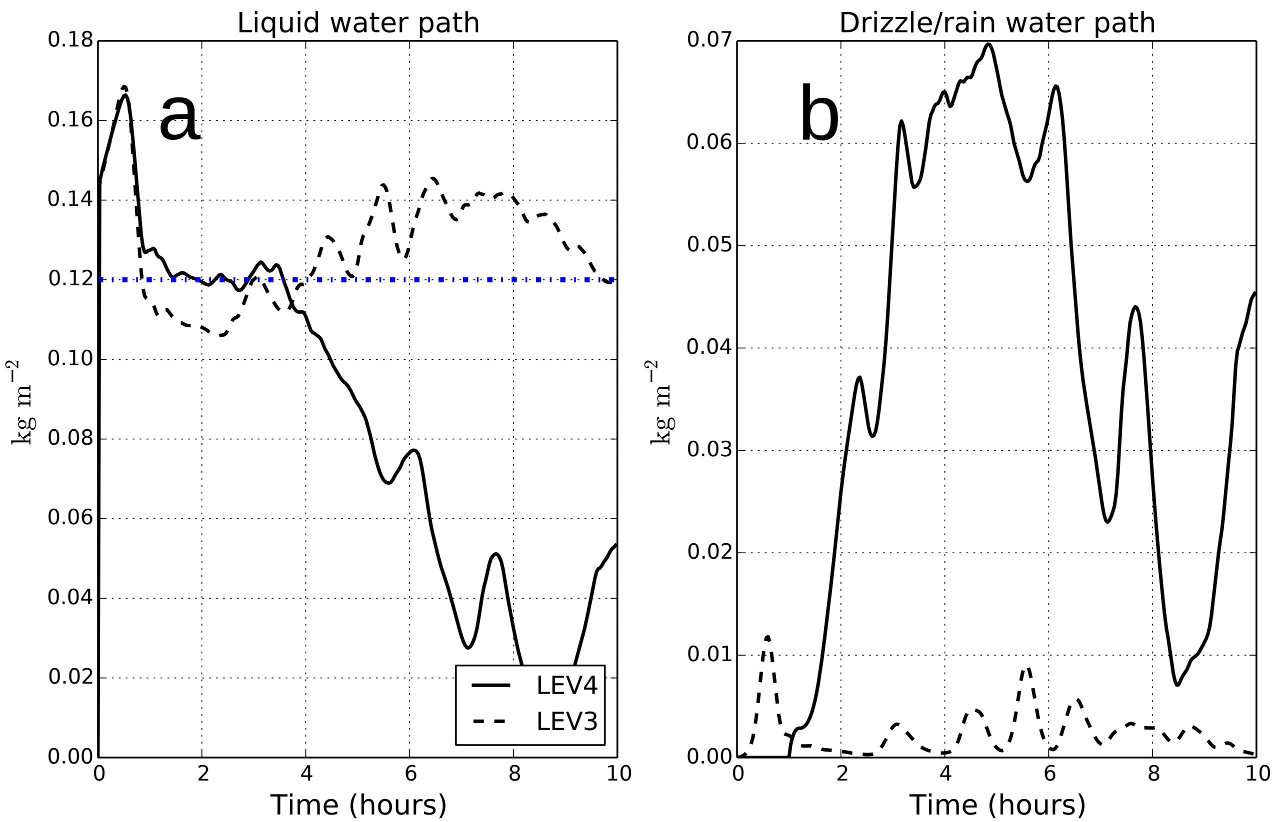

Figure 3. a) Liquid water path, interpreted as the total mass of water, including both cloud droplets and drizzle. b) Rain water path, taken as the water mass diagnosed ~~in drizzle and rain drop~~ from precipitation bins only. Results from LEV3 are shown with a dashed line while those from LEV4 are shown with a solid line. The horizontal blue dashed line in panel a) represents the observed flight mean liquid water path at 120 $\mathrm{g\,m^{-2}}$ as reported by **?** .

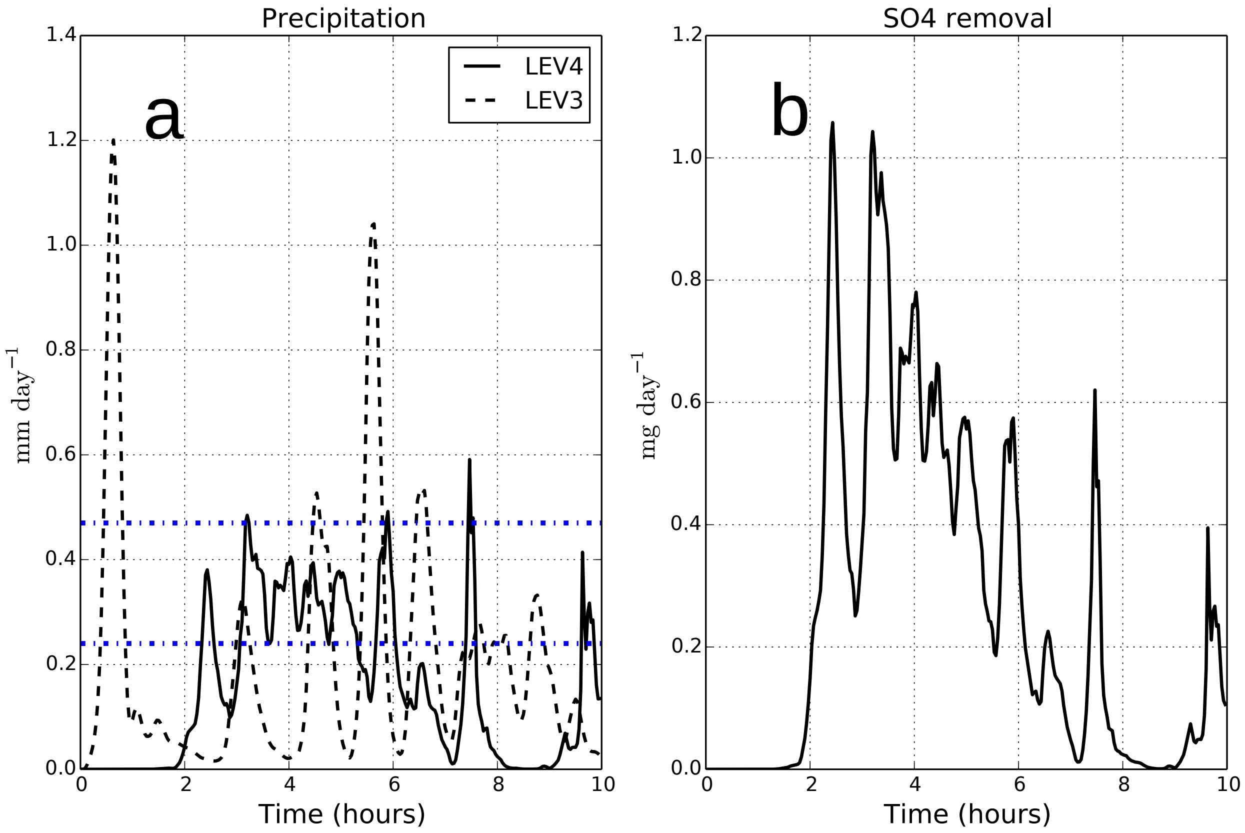

Figure 4. a) Surface precipitation rate in $\mathrm{mm\,day}^{-1}$ and b) removal rate of sulphate embedded in precipitating drops in $\mathrm{mg\,day}^{-1}$. Results from LEV3 are shown with a dashed line while those from LEV4 are shown with a solid line. The blue horizontal lines in panel a) indicate range of observed values shown in **?** .

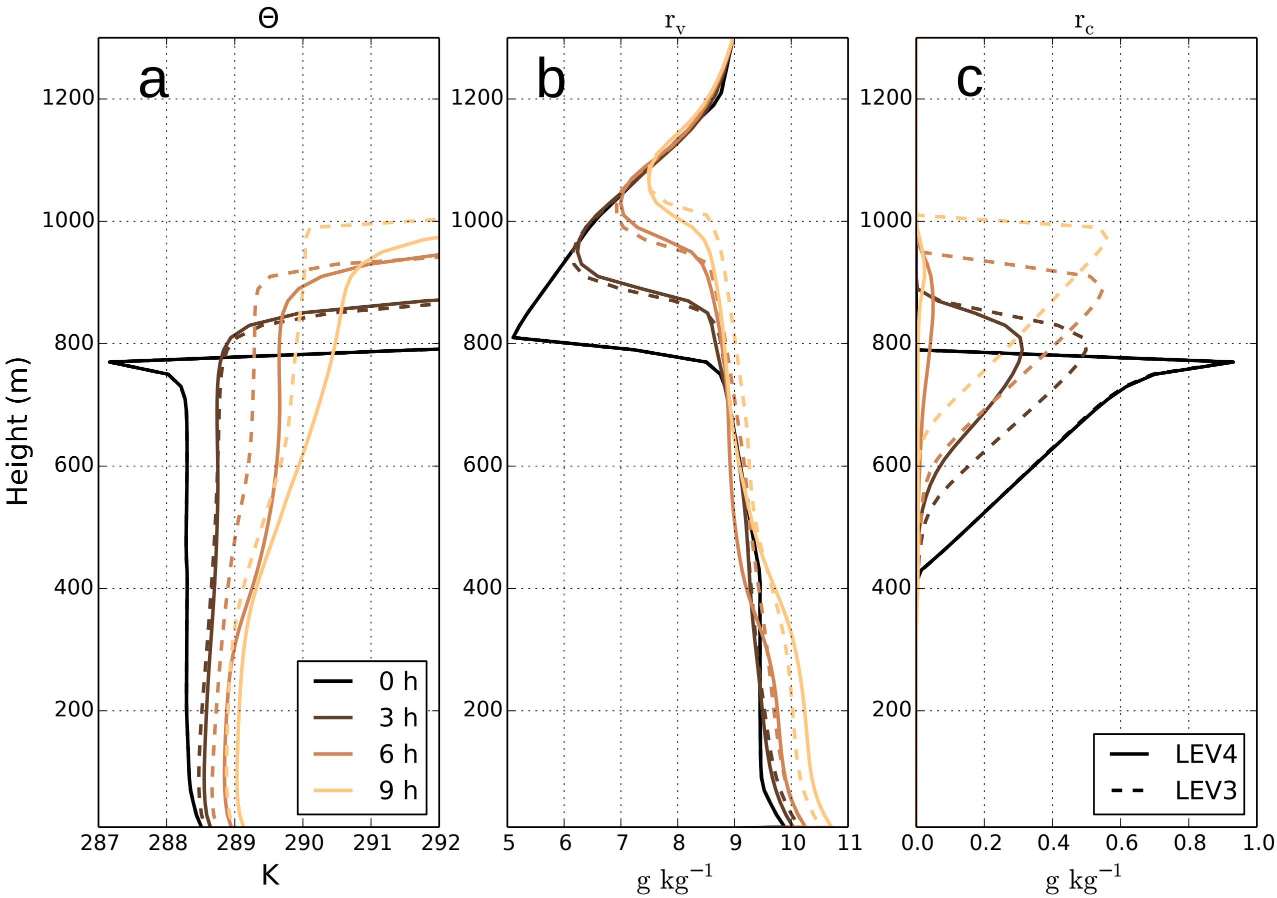

Figure 5. Domain mean vertical profiles of a) potential temperature, b) water vapour mixing ratio and c) liquid water mixing ratio. Data is plotted in 3 hour intervals from the initial state of the model to 9 hours into the simulation (from black to orange). Results from LEV3 are shown with a dashed line while those from LEV4 are shown with a solid line.

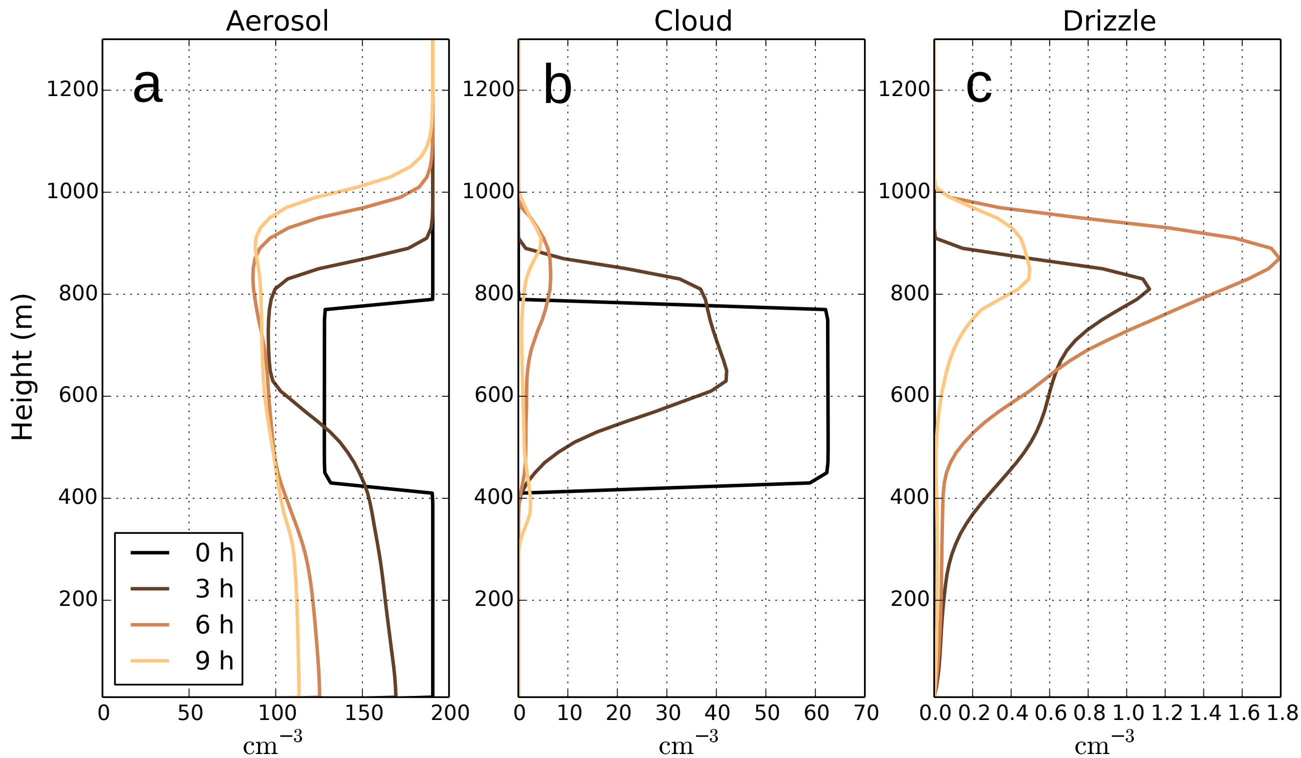

Figure 6. LEV4 domain mean profiles for a) aerosol, b) cloud droplet and c) drizzle number concentrations, plotted in 3 hour intervals from the initial state of the model to 9 hours into the simulation (from black to orange).

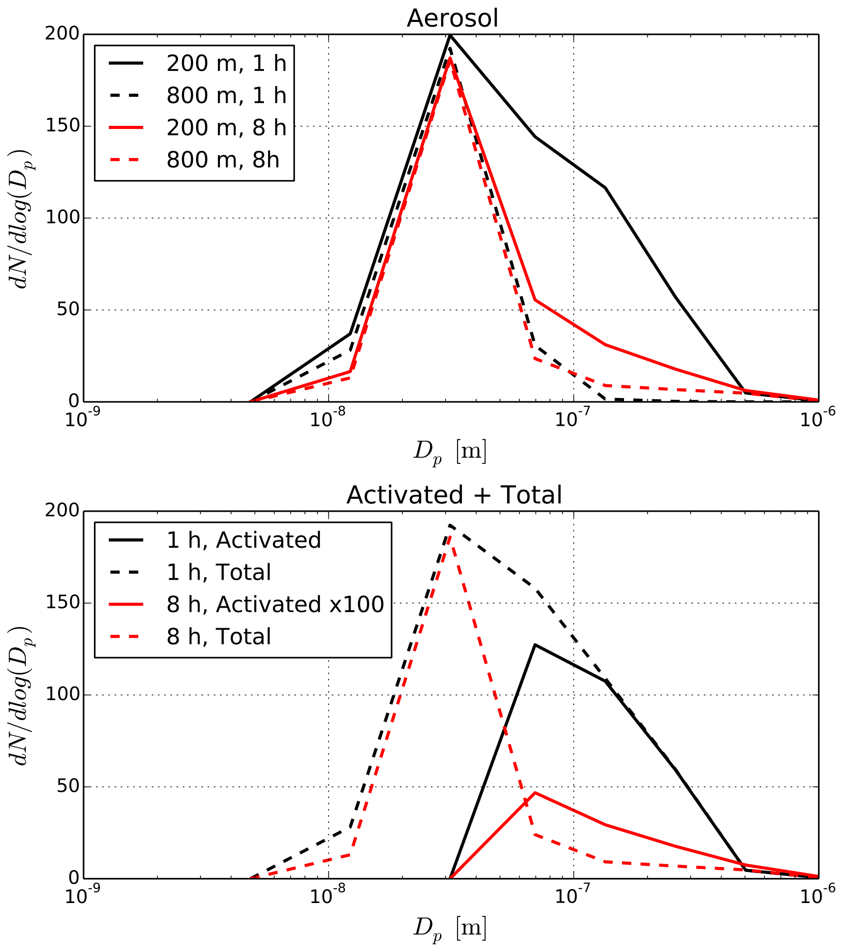

Figure 7. Size distributions of a) dry/interstitial aerosol after spinup (1h, black) and after 8 hours (red) averaged over the domain at 200 m (solid) and 800 m (dashed) heights, and b) activated (solid) and total (activated + interstitial) aerosol size distributions after the spinup (black) and after 8 hours (red) sampled at 800 height. Please note that the concentration of activated particles at 8 hours is multiplied by 100 to be visible in the figure. D_p is the particle diameter.

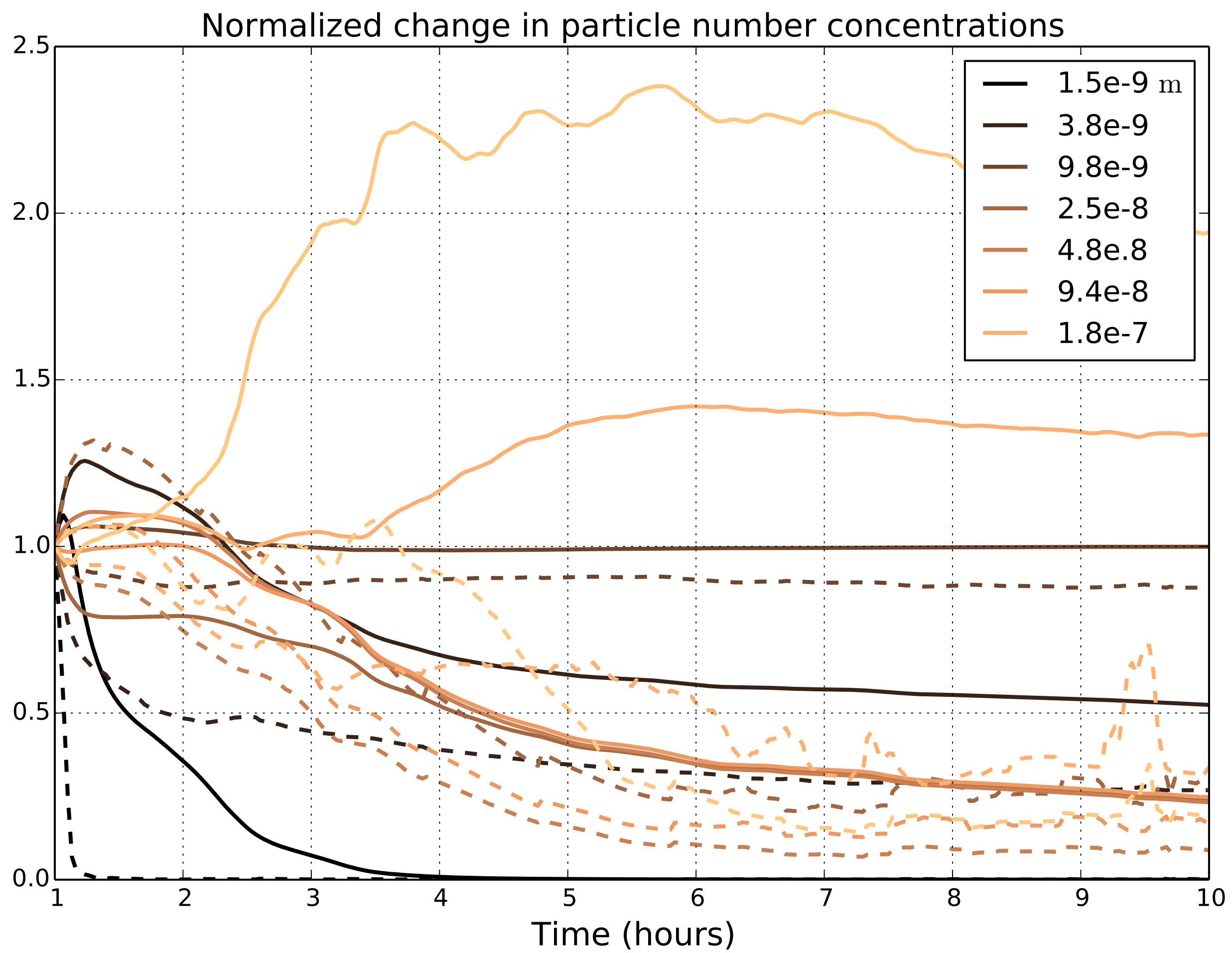

Figure 8. Normalized change in particle number concentration in each size bin. The concentrations are presented as a domain average from the i) below-cloud layer (solid lines) and ii) in-cloud (dashed lines). In the latter case, the sum of the number of interstitial particles and activated CCN is presented for each bin. The two largest size bins are not shown because of very small absolute concentrations in this case. The legend gives the lower limit diameter of the presented size bins. Data is show from the end of the spinup period. Number concentrations from this time are used as the normalizing factor for each bin.

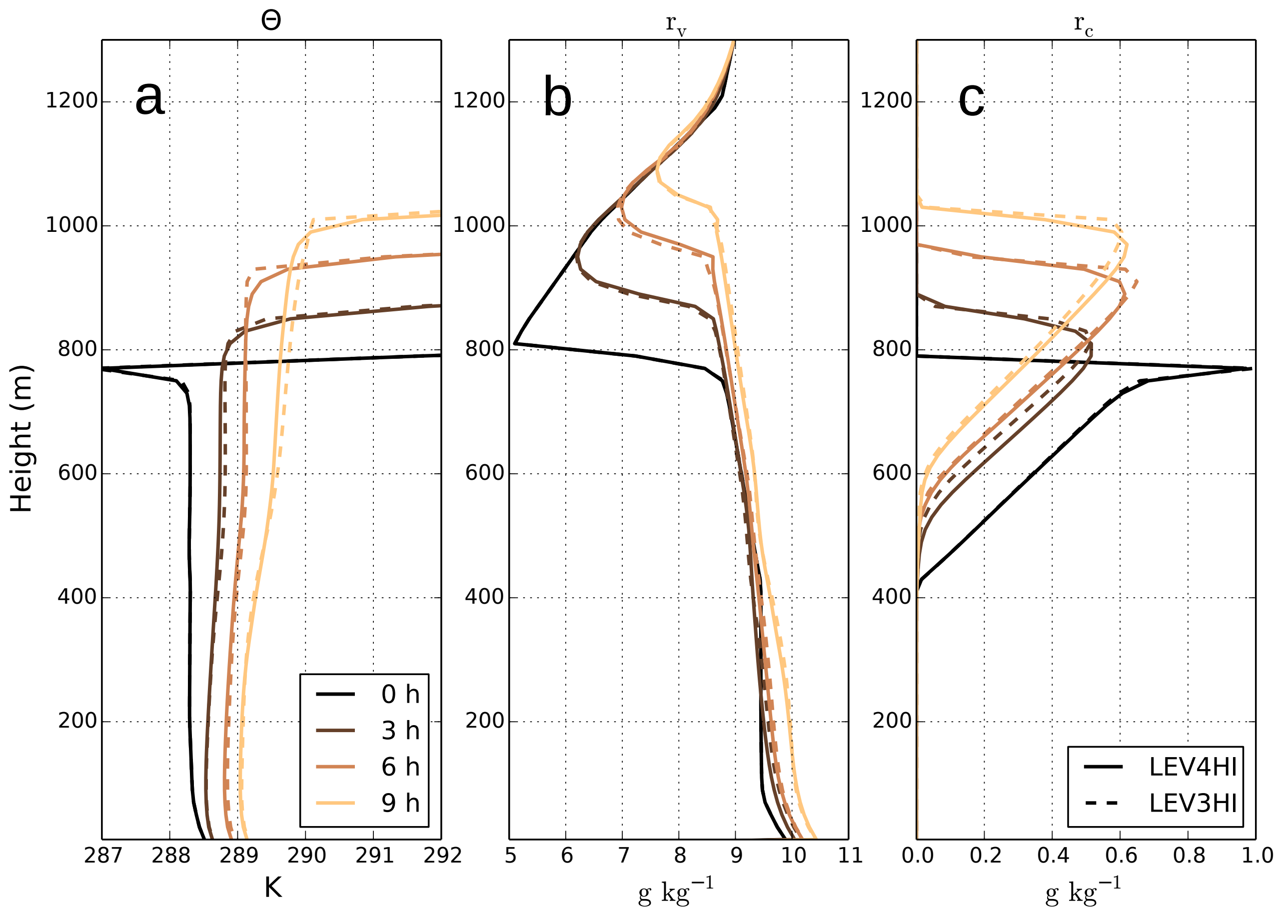

Figure 9. Similar to Figure 5, but for the experiments LEV3HI and LEV4HI with high aerosol number concentrations.

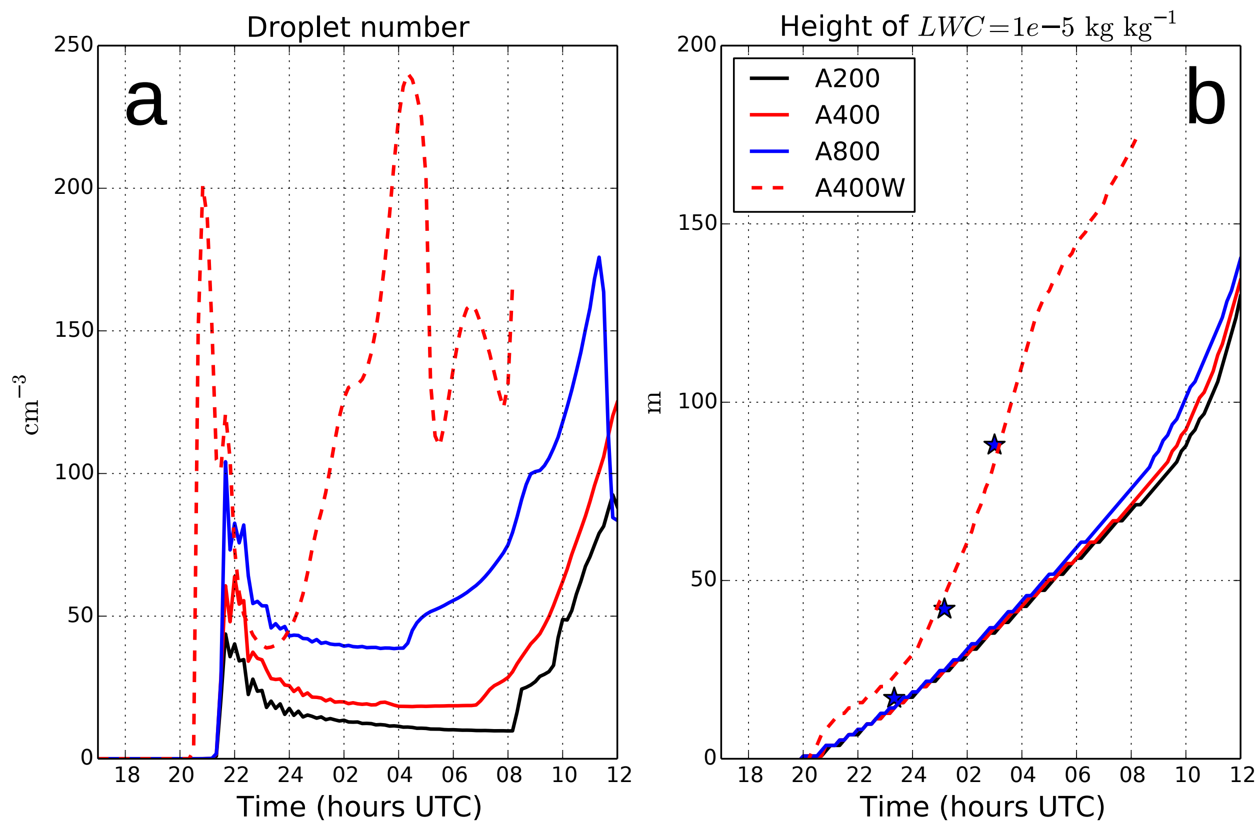

Figure 10. a) Fog droplet number concentrations sampled at approximately 10 m height and b) the height of the fog top layer interpreted as the 1×10^{-5} kg kg^{-1} isoline for liquid water ~~mixing ratio~~content. Observed values of the fog layer depth based on tethered balloon data given by **?** are shown with blue star symbols in panel b).

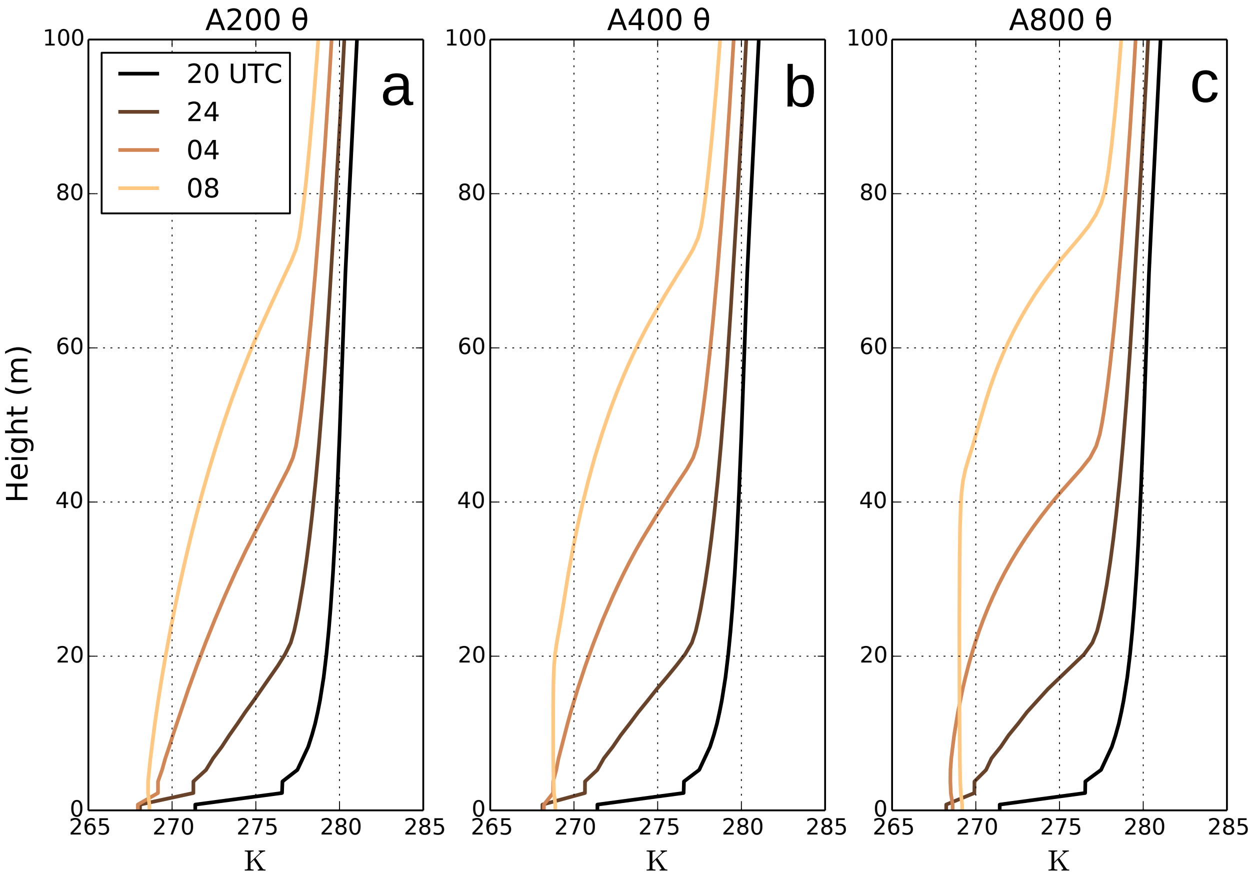

Figure 11. Domain mean profiles of potential temperature in 4 hour intervals starting from the formation of the fog layer (from black to orange) for the experiments a) A200, b) A400 and c) A800.

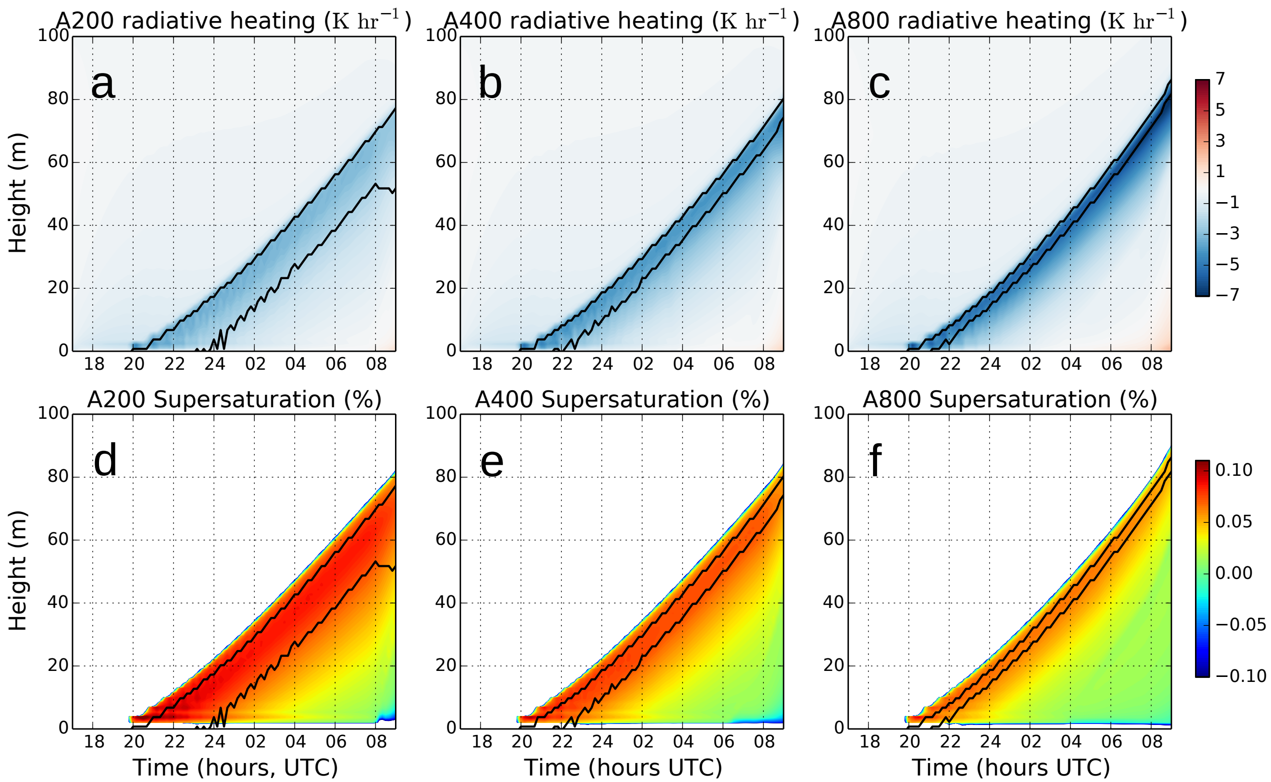

Figure 12. a)-c) Radiative heating in $\mathrm{K\,hr^{-1}}$ for the experiments A200,A400 and A800, respectively. d)-f) Water vapour supersaturation in per cent for the same experiments. The upper and lower black curves give the $0.01\ \mathrm{g\,kg^{-1}}$ and $0.1\ \mathrm{g\,kg^{-1}}$ isolines for the liquid water mixing ratio, respectively.

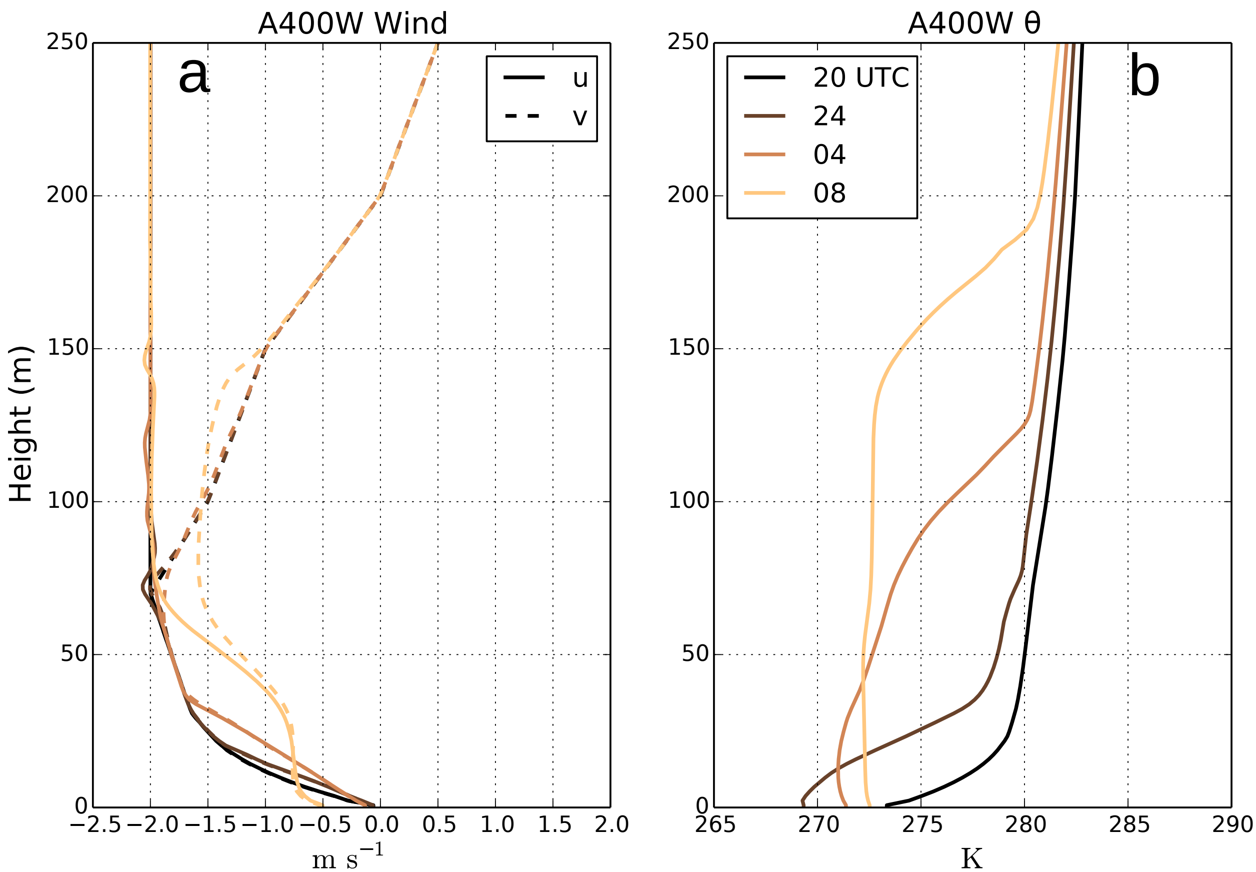

Figure 13. a) Domain mean profiles of u and v wind components and b) the potential temperature for the experiment A400W in 4 hour intervals from the formation of the fog layer (from black to orange).