Response to anonymous referee 1

September 10, 2019

Marco de Bruine et al.

First of all, we would like to thank the reviewer for the careful reading of our manuscript. His/her comments greatly improved the quality of our manuscript, including a better paper structure. Point-by-point replies to the comments are provided below.

1 Simulation resolution and length

Comment 1

A major concern that I would like to highlight is the limitation of the discussion of sample simulations to a single LES resolution, notably a relatively coarse one for a shallow cumulus case. In the 2011 study of Matheou et al. (doi:10.1175/2011MWR3599.1) it was shown that for the RICO simulation setup used in the present paper, even significantly finer grids and larger domains were not enough to achieve convergence in terms of cloud characteristics (see also Sato et al. 2018, doi:10.1029/2018MS001285). While, arguably, such analysis and discussion is not directly related to the scope of the manuscript, it would be of great value for potential users of the developed aerosol module. Moreover, having the limitations of the resolution in mind, and given the absence of convergence tests in the paper, I strongly encourage the authors to critically revisit all parts of the paper commenting on the match with observations.

Response The other reviewer shared this concern of directly comparing model outcome to observations because of the reasons mentioned by the reviewer. We acknowledge these arguments and revisit all parts of the paper commenting on the match with observations.

Changes In the revised manuscript we will focus the discussion in Sect 5.1 on the behaviour of the different simulations and will not draw conclusions based on direct comparison of observations and model outcome. We would still like to keep the observations as a background against which the different simulation set-ups behave. We will also include a statement about the current lack of convergence tests.

Comment 2

Similar concern applies to the length of the simulation. The original RICO setup featured 24 hour simulations, of which several first hours were treated as spin-up, while output result for model intercomparison was carried out using the last four hours of simulation only. In the present paper, 6-hour long simulations are presented (and the conclusions section enumerating the main findings of the study, comments on processes with multi-day timescales). It is essential to point out this difference, provide the reason for shortening the simulations, and comment on it.

Response The reason we use 6-hour long simulations (with the first 3 hours as spin-up excluded from the analysis) is that we are interested in the evolution of a certain aerosol population within a cloud field. We do not simulate emission of new aerosol during the simulations. In a 24-hour long simulation with substantial wash-out by precipitation but no sources would deplete the aerosol population to unrealistically low levels. In our 6-hour simulations we already lose 20-25% of the aerosol mass.

In the Figures of the model output for the RICO LES intercomparison by Van Zanten et al. (2011) as found on http://projects.knmi.nl/rico/ (last visited: 3 September 2019). We see that metrics like LWP and cloud fraction are more or less stable after 3-4 hours. Therefore, we expect that if we would have a sustained aerosol population in a longer simulation, the results would not be substantially different.

Changes We will add additional information on the choice for 6-hour simulations and expectations of how this would influence the results.

Comment 3

How does the intensive precipitation in the second half of the first hour of the RICO case affects the budget of remaining aerosol, and hence how different are the conditions in which clouds form here with respect to those found in models with infinite CCN reservoir? Please discuss.

Response There is indeed substantial wash-out by the initial burst of precipitation during the spin-up of the simulation. As shown in the figure below, 90.7 and 94.4% of the initial budget remains for the KAPPA and PN simulations respectively. At the end of the simulations this decreases to 76.7 and 79.0% for KAPPA and PN.

Our BASE and BASE30 simulations are examples of a model that implicitly assumes an infinite CCN reservoir that results in clouds with a certain Nc and we describe their results in Sect. 5.1. The ultimate goal for this model is to have a fully coupled simulation which includes emissions. However, in this initial paper, our case remains academic and we highlight the sensitivity of the processing of aerosol to the activation scheme.

Changes In the revised manuscript we will address removal of aerosol and the impact on the simulated microphysics.

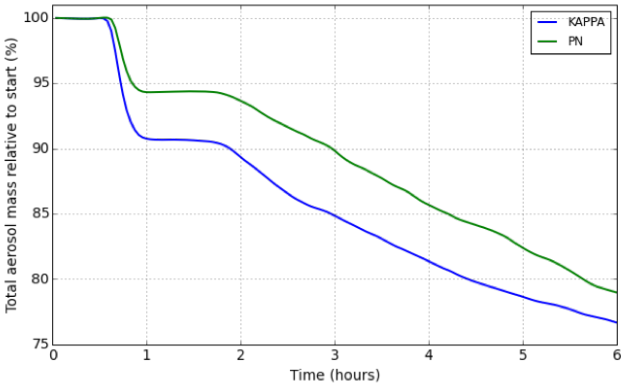

Figure 1: Remaining aerosol budget in the lowest 3 km of the domain relative to the initialisation for the KAPPA and PN simulations.

2 Aerosol processing nomenclature and background information

Comment 4

Aerosol-cloud interaction, as a main theme of the manuscript, is always stated in singular form (i.e., interaction, not interactions). First, in general plural would sounds better in my opinion.

Response & Changes We will follow this suggestion and use the plural form "aerosol-cloud interactions" in the revised manuscript.

Comment 5

Second, it would be worth to elaborate in the paper on the different kinds of interactions, also those beyond the processes covered in DALES-M7. It is striking that aerosol distribution changes through aqueous-phase oxidation are not mentioned in the paper, the mention of chemistry in the penultimate sentence is unclear. Please comment on it and clearly position the capabilities of the introduced model among other available aerosol-cloud interactions modelling frameworks; see, e.g., Ovchinnikov and Easter 2010 (doi:10.1029/2009JD012816) and Jaruga et al. (doi:10.5194/gmd-11-3623-2018) and references therein. Aerosol nucleation processes are also reported to be influenced by clouds (e.g., Wehner et al. 2015, doi:10.5194/acp-15-11701-2015).

Response & Changes We agree, and in the revised manuscript we will add a more elaborate description of the different aerosol-cloud interactions in the introduction, aqueous-phase chemistry in particular. Moreover, in Section 3 we will clarify which of the processes are included in our framework and which are not yet implemented.

Comment 6

On a related note, while the authors claim "resolving most of the turbulence" (worth rephrasing), there is little discussion on how it affects the modelled collisions among aerosol, cloud and precipitation particles - worth mentioning.

Response We agree with the reviewer and add a description of how the resolution in the model compares to the scales involved in particle-level processes like collisions.

Changes In the revised manuscript we rephrased the sentence "resolving most of the turbulence" in response one of the technical comments to page 11, line 14. However, we will add another statement that highlights that although LES is usually considered as a high-resolution simulation, both the spatial resolution of 10m and the temporal resolution of 1s are still too coarse to actually simulate the processes on particle-level for which one would need DNS on the Kolmogorov length-scale of 1mm. These processes therefore remain parameterized in LES.

Comment 7

In general, perhaps putting together a summary of omitted/largely-simplified processes would be a good idea (in-cloud activation, aerosol sedimentation, influence of turbulence on collisions, chemistry, etc)?

Response & Changes As stated in the response to comment 5, we will add a summary of what processes are covered in DALES-M7 and which are not.

Comment 8

Please also make sure it is clear what "explicit" means in different contexts in the paper. In principle, it should be clear (also to readers from neighbouring domains or those focused on largely different scales) what the opposite "implicit" would mean.

Response The meaning of explicit in this paper is "not parameterized", making parameterized the opposite. We acknowledge that by using the term explicit in a manuscript describing numerical methods there is a risk to confuse this with explicit/implicit methods for model integration.

Changes In the revised manuscript, the first mention of explicit aerosol calculation will include an explanation of the opposite being: 'parameterized'. P3, line 12-13 will be adjusted as: "This also allows for explicit calculation of aerosol activation based on the characteristics of the aerosol population, instead of using a parameterization based on i.e. updraft velocity."

Comment 9

Could "free aerosol" when referring to out-of-cloud-or-rain-shafts aerosol be named somehow differently? Ambient aerosol?

Response We used the term 'free aerosol' to indicate all aerosol not incorporated in (or captured by, hence the term 'free') cloud and rain droplets. This includes for example interstitial aerosol in clouds or aerosol in the path of falling precipitation. In our opinion, the term 'ambient aerosol' implies that the aerosol is unaffected by cloud processes in any way, which is not the way we intended to use this term here.

3 Statements calling for references

Comment 10

p6/l4-5: "...cloud and rain droplet modes do not have a lognormal shape...", see: Clark 1976 (doi:10.1175/1520-0469(1976)0332.0.CO;2) and Feingold and Levin 1986 (doi:10.1175/1520-0450(1986)0252.0.CO;2)

Response What we meant to say here is that in our model, the assumed distribution for the cloud and raindrop size distributions does not need to have a lognormal shape, but can be different. We did not intend to state here that cloud and rain size distributions are not lognormal, which then indeed would need a reference.

Changes In the revised manuscript, we changed the associated text to be more clear and not imply a certain hydrometeor size distribution: "...cloud and rain droplet modes do not necessarily need to have a lognormal shape..."

Comment 11

p10/l11: "but the measurements were fitted to a bimodal lognormal dist." ? in which work?

Response We based this statement on information from van Zanten et al. (2011) Section 2.2.3 elaborating on the input of models that require an aerosol size distribution.

Changes In the revised manuscript we add the reference and a one-line description: "The aerosol size distribution was measured on aircraft flight RF12, and the measurements were fitted to a bimodal lognormal distribution of aerosols with uniform composition, assuming characteristics of ammonium-bisulfate (see van Zanten et al. (2011), their Sect 2.2.3), despite the marine nature of the environment."

Comment 12

- p11/l7: "corresponds to the actual observed mean values" ? which day, which aircraft, which sensor, which sampling rate, what kind of analysis, which paper...
- p12/l1: "which is in accordance with observations" ? ditto
- p12/l22: "campaign in-situ observations show values" ? ditto

Response All three statements refer to the observations in Fig. 8 in van Zanten et al. (2011) and Fig. 2 in our manuscript. These measurements are an aggregate of 1 Hz FFSSP measurements on flights RF06-RF12 with the NCAR C-130 aircraft. We will better specify these details and make clear this is the data we refer to in the remainder of the section.

Changes We will change the opening of Sect. 5.1 describing this:

"To evaluate the modelled cloud characteristics produced in the different simulations we follow the analysis of vanZanten et al. (2011). Domain-averaged cloud characteristics are shown in Fig. 3, which is constructed to resemble Fig. 8 in vanZanten et al. (2011). Similar to their work we use an aggregate of 1 Hz FFSSP measurements on flights RF06-RF12 with the C-130 aircraft (Rauber et al., 2006). Cloud characteristics are filtered using the condition qc > 0.01 g kg^{-1}, while rain characteristics use the condition qr > 0.001 g kg^{-1}."

4 Paper structure

Comment 13

Several suggestions and comments to the paper structure:

- Section 3.1 is introduced, but there is no 3.2
- Section 5 "Results" should be somehow linked with the setup (as these are not general results)
- Appendix material fits well into the simulation setup section

Response & Changes We will adopt the suggested structure for the paper, which fixes the unnecessary section depth in Section 3. It also clarifies the fact that we discuss the differences between model simulations and cannot directly compare to observation because of model limitations. Lastly, since the description of the simulation set-up is not too long it indeed fits in the main body of the text and we would not have to include an extra short summary of this as we do in Section 4 of the manuscript now.

Comment 14

Code availability section does not need a number (format as acknowledgements)

Response Adjusted.

5 Code availability

Comment 15

In which branch of DALES github repo one can find the code of DALES-M7?

Response Currently, DALES-M7 is not on the DALES github repository. Instead, everything can be found at the link stated in the code availability section: http://doi.org/10.5281/zenodo.3241356. DALES-M7 is based on the 4.1 branch, which also is the one used for the BASE and BASE30 simulations in this work. This line of development of DALES is currently in progress and still an unfinished research line. After completion, we intend to merge this branch into the main DALES repository (version 4.2).

Comment 16

Is M7 an external dependency or was it incorporated into (or reimplemented?) DALES codebase?

Response It is incorporated in the DALES code base.

Comment 17

What is the license of M7? Is it compatible with DALES?s GPL? Which version of M7 was used/incorporated/reimplemented?

Response There is no GPL defined for M7. Moreover, in this work we only implemented the aerosol representation used by M7. We excluded the dynamic processes of M7, such as nucleation, coagulation and condensation. This will, however, be part of future development of the model.

6 Minor or technical comments

Comment 18

p1/l4: “The feedback of ACI on the aerosol population remains relatively understudied” ? within the abstract, please concentrate on describing the contents of the paper, and not motivation.

Response & Changes In the revised manuscript we wemoved the following sentences containing motivation of this work: “These models combine a spatial resolution high enough to resolve cloud structures with domain sizes large enough to simulate macroscale dynamics and feedback between clouds. However, most research on ACI using LES simulations is focused on changes in cloud characteristics. The feedback of ACI on the aerosol population remains relatively understudied."

Comment 19

p1/l18-19: please clarify if "larger" refers to size or mass

Response The aerosol size comparison in the last part of the abstract refers to aerosol size (i.e. radius).

Changes Revised manuscript is adjusted to explicitly mentions this: "Analysis of typical aerosol size associated with the different microphysical processes shows that aerosols resuspended by cloud evaporation have a radius that is only 5 to 10% larger than the originally activated aerosols. In contrast, aerosols released by evaporating precipitation are an order of magnitude larger".

Comment 20

p2/l10: "missing atmospheric context" ? please rephrase

Response Adjusted

Changes Sentence in revised manuscript changed to: "…process-based small-scale simulations (e.g. Roelofs, 1992) describe the microphysical processes in high detail, but cannot model the effect of aerosol-cloud interactions on the macroscale thermodynamics and structure of a cloud."

Comment 21

p2/l28: given the paper discusses aerosol-cloud interactions, mentioning also 2D-bin (e.g., Lebo and Seinfeld 2011, doi:10.5194/acp-11-12297-2011) and particle-based methods (e.g., Grabowski et al 2019, doi:10.1175/BAMS-D-18-0005.1) would be apt

Response Indeed, the 'traditional' choice of bin vs. bulk is complemented by particle-based methods like the libcloudph++ by Arabas et al. (2015) or the similar 'superdroplet' method (Riechelmann et al., 2014; Hoffmann et al., 2019). We will also add a reference to the overview paper of Grabowski et al., 2019) as it is a very good illustration of the current status of modelling aerosols and clouds in LES. The extensive 2D-bin method by Lebo & Seinfeld (2011) deserves a mention here as well.

Changes In the revised manuscript we will add references to these methods in the text to inform the reader of these alternative numerical frameworks to study aerosol-cloud interactions.

Comment 22

p3/l3-4: recent advances in representing aerosol in LES are not limited to these two works! please be more comprehensive or rephrase

Response We aimed here to elaborate on LES models that include aerosol frameworks with the focus on multiple aerosol species and/or (aqueous-phase) chemistry.

Changes We will rephrase this paragraph in the revised manuscript to better specify that we focus on aerosol modules in LES simulations including multiple aerosol species. We added Jaruga and Pawlowska (2018) to the discussion as their extension to the libcloudph++ library opens up a range of possibilities to include and interactively calculate multiple aerosol species.

Comment 23

p6/l13-17: unlike in a basic single-particle model as κ-Köhler, activation in clouds happens on populations of particles and with complex supersaturation dynamics related to small-scale fluctuations and drop-growth feedback, please acknowledge what is simplified when just considering a critical supersaturation

Response We indeed acknowledge that by using a direct calculation based on κ-Köhler and using a fixed value for supersaturation leaves out the competition for moisture between non-activated aerosol and existing droplets.

Moreover, by directly translating supersaturation to particle activation, we implicitly assume that the equilibration time of the droplets is instantaneous or at least considerably shorter than the model timestep. This might lead to an overestimation of activated droplets as some particles would activate at a certain supersaturation but did not have enough time to grow to the critical radius yet. This would be better captured by a numerical framework that directly calculates the condensational growth.

Changes We will add this discussion after the description of the activation routine, to the paragraph on page 7, line 12 where we discuss the supersaturation.

Comment 24

p7/l12: please clarify if this is peak or equilibrium in-cloud supersaturation

Response As discussed for the previous comment, in our model we assume that aerosols/droplets equilibrate instantaneously with the supersaturation of the environment. This implies that there is no difference between the two. However, in the KAPPA activation scheme we only activate once and assume all subsequent water surplus condenses on the cloud droplets, so this value would refer to the supersaturation maximum at the cloud base.

Comment 25

p8/l7: first mention of KAPPA, not introduced as an acronym before, please define

Response We remove the reference to the KAPPA simulation here, as this part of the text does not yet refer to the exact simulations performed in this work, but to the activation scheme in general.

Changes Changed sentence to: To avoid this 'runaway activation' in the κ-Köhler-based scheme, activation in a cloudy grid cell is allowed only once.

We also changed PN in this paragraph to PN15 for consistency as we refer to the complete work by Pousse-Nottelmann et al. (2015) here, not the simulation.

Comment 26

p9/l6: final → last

Response Adjusted

Comment 27

p9/l11: add "and" before "is calculated"

Response Adjusted

Comment 28

p9/l22:"their Eq. 4" → "Eq. 4 therein"

Response Adjusted

Comment 29

p10/l18: being over an ocean is not the point, the point is from where the wind blows and how far from the sources it is

Response Agreed, we specified why the dominance of sea salt aerosol is to be expected here.

Changes Sentence change to: "The aerosol population mainly consists of sea salt particles, as expected for this ocean region with trade winds blowing from the open ocean."

Comment 30

p10/l22: shouldn't the concentrations be expressed in the units of mg^{-1} (to reduce variation from density changes)

Response One of the main figures in our manuscript is Fig 1, which is made to resemble Fig. 8 in Van Zanten et al. (2011). Here, the values are expressed per unit volume. For consistency between figures and values stated in the text we opted to use the units of cm^{-3} here as well.

Comment 31

p11/l14 "beautifully display the richness"... please refrain from vague statements

Response Agreed.

Changes Text adjusted to be more to-the-point and precise: "These cross-sections display the internal variability within the LES model domain that results from the high spatial resolution."

Comment 32

p12/l8: "at left" $\rightarrow$ "at the left"

Response Adjusted

Comment 33

p14/l22: "mighty" $\rightarrow$ "might"

Response Adjusted

Comment 34

p16/l15: perhaps worth commenting on how in-cloud activation was modelled (or neglected)

Response This paragraph is thoroughly revised. Reviewer 2 commented that the difference in cloud processing between PN and KAPPA required more explanation. This manuscript is adjusted to include a description of cloud microphysics in Sect 5.1 and refer to this in Sect 5.2 which will include the difference in activation between the two simulations. For a full description see the response to comment 8 of reviewer 2.

Comment 35

p19/l13: are 4 significant digits needed? p20/l5: ditto

Response & Changes Accuracy of all radii mentioned in paragraph 5.2.2 and Table 5 reduced to 1 nm.

Comment 36

References: please be consistent in using journal name abbreviations vs. full journal names

Response & Changes All journal names now abbreviated using Caltech Library Services (www.library.caltech.edu/reference/abbreviations)