Response to anonymous referee 2

September 10, 2019

Marco de Bruine et al.

First of all, we would like to thank the reviewer for his/her comments and the careful reading of our manuscript. There are several main points made, which will be addressed below.

1 General comments

Comment 1

Aerosol-cloud interaction (ACI) is a rather general term, so quite often this could be replaced by a more specific term. For example, "The feedback of ACI on the aerosol population" (page 1, line 4) could be just "The impact of cloud processing on the aerosol population" and the same term could be used also here "Whether ACI increases or decreases the average aerosol size" (page 1, line 16). Please check the whole manuscript.

Response We agree with the reviewer that the general term ACI should be replaced by a more specific description of the processes in play whenever possible. This complements the comment of the other reviewer stating that ACI is a collection of many different processes.

Changes In the revised manuscript we replace instances with a general reference to ACI by a more direct description of the processes we address.

Comment 2

Using the saturation adjustment method (diagnostic cloud water) and assuming a fixed value for supersaturation when calculating cloud activation are significant approximations. Their effects should be at least explained here instead of investigating these in the future (page 7, line 13). Can you really examine aerosol-cloud interactions without explicitly modeling aerosol condensational growth and subsequent cloud activation (prognostic cloud water)? What is the added value of detailed aerosol chemical composition when cloud activation is so much simplified?

Response The long-term goal for DALES is to create a 'virtual lab' to simulate the atmosphere with as few assumptions as possible. We intend to build a model that can study links

between pollution, atmospheric chemistry (including aqueous chemistry) and clouds. The first step towards this goal is the inclusion of an aerosol representation that fits in this framework. This requires a scheme capable of simulating multiple aerosol species. Therefore, we chose to implement an aerosol module following the framework of M7 (Vignati et al., 2014). This comes at the cost of a limited numerical description of condensational growth and activation of cloud droplets, since a chemically-resolving bin scheme would be computationally too demanding.

We agree that by using a fixed value for the supersaturation, the model misses an important feedback between supersaturation and aerosol activation. For this reason, we included sensitivity runs with different values for S as well as a different activation parameterization (Pousse-Nottelman et al., 2015) as a comparison.

Changes In the revised manuscript we will directly address this instead of stating it will be investigated in the future.

The paragraph at the end of Section 3.1.1 (starting at page 7, line 11) is changed accordingly: "As stated above, DALES uses an 'all-or-nothing' cloud water adjustment in which cloud liquid water qc is a diagnostic variable. Therefore, we use a fixed value of supersaturation (S = 0.4%) representative for the simulated case (Derksen et al., 2009). Moreover, the use of a multi-species aerosol scheme comes at the cost of a limited numerical description of condensational growth and subsequent activation. Including both would be computationally too demanding. As a result, the model thus does not capture the competition for moisture between particles (aerosols and cloud droplets) or the role of supersaturation in this process. To asses impact of changing supersaturation on the cloud characteristics in our simulations, we will perform sensitivity simulations with different values of S. Although fixing the value of S is still an approximation, it does allow for an interactive calculation of cloud droplet number concentration based on simulated aerosol."

Regarding the choice for a multi-species aerosol scheme, we address this in the introduction (page 3, line 13-15). However, in the revised manuscript we highlight this again at the beginning of Sect 3 (page 4, line 25) with the following adaptation: "This framework allows for the simulation of an external mixture of multiple aerosol species. In future development, this will be coupled to atmospheric chemistry, including aqueous-phase chemistry. It also allows for the investigation of differences in how cloud processing influences different aerosol species. By using M7, cloud activation can be based on fundamental . . . "

Comment 3

Why did the "runaway activation" (page 8, line 7) were allowed only for the PN activation scheme? For me this looks like a possible reason for the observation that aerosol fluxes for activation (and cloud evaporation) are 12-13 times larger for PN simulations compared with those from KAPPA simulation. This difference is later used as an explanation for several other differences between simulation results. If the difference between activation schemes is related to a technical/numerical reason, then it should be considered as a bug and fixed.

Response The term 'repeated activation' in this work describes activation of new cloud drops in a cloudy gridcel already containing cloud droplets. This repeated activation is prohibited for the k-Kohler scheme, because the modal representation keeps pushing aerosol mass and number to a size above the activation threshold, so there is no mechanism to limit the activation due to numerical diffusion. Without this limit virtually all aerosol would be activated, leading to erroneously high cloud droplet numbers. We termed this process 'runaway activation'.

The PN activation scheme, however, is fundamentally different and uses other mechanisms to limit unrealistic high cloud droplet numbers. The newly activated cloud droplets $\partial N_c \backslash \partial t$ in this scheme are calculated following Eq. (2) in Pousse-Nottelmann et al (2015):

$$\frac{\partial N_c}{\partial t} = \max\left\{\frac{1}{\Delta t}\left[\left(\frac{wN^t_{>35}}{w+\alpha N^t_{>35}}\right)^{1.27} - N_c^{t-1}\right], 0\right\} \tag{1}$$

With w the updraft vertical velocity, Δt the length of the timestep, N_c^{t-1} the number of cloud droplets present, $N^t_{>35}$ the number concentration of soluble/mixed aerosol particles larger than 35 nm and $\alpha = 0.023$ cm^4 s^{-1} an empirically derived constant.

By including updraft velocity w and the existing cloud droplet number N_c^{t-1}, this formulation does include competition for moisture between condensation on existing droplets and activation of new particles. However, the strongest limitation of this formulation is found in the prefactor of 0.1. This prefactor was determined in Zubler et al. (2011a) by comparison of their model outcome against satellite data with respect to the cloud droplet effective radius. The combination of this prefactor and the subtraction of N_c^{t-1} poses such a strong limitation on aerosol activation that 'runaway activation' is not occurring in the PN scheme.

The figure below shows vertical profiles of aerosol activation in terms of aerosol/cloud number. The profiles are normalized individually for each simulation to the maximum of the vertical profile. Note that overall activation in the PN simulation is 12-13 times stronger , as can be inferred from Table 3 and 4 in the paper. However, the vertical distribution of activation in both simulations is similar with a peak near cloud base. Activation above cloud base drops off slightly faster for caused by its dependence on updraft velocity. We therefore conclude that both activation schemes are reasonable and lead to realistic cloud simulations, albeit with widely different aerosol evaporation/activation cycles.

Why the aerosol flux associated with activation and cloud evaporation is so much higher is explained in the reply to the related comment 8.

Summarizing, we deliberately test two valid but fundamentally different cloud-activation schemes to highlight the sensitivity of cloud microphysics to this choice.

Changes In the revised manuscript we will add the above-mentioned formula which is central in the PN scheme and we will better describe why the PN scheme can allow 'repeated activation' without leading to 'runaway activation'.

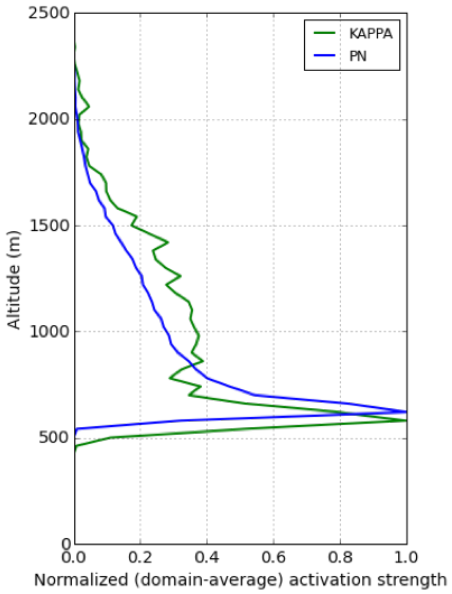


Figure 1: Vertical profile of domain-average aerosol mass-flux to in-cloud aerosol for the KAPPA and PN simulations.

Comment 4

Validating simulations against observations is not as straightforward as expected in this work (e.g. page 10, line 2). LES inputs (aerosol size distributions and composition, atmospheric variables, etc.) are not fully synchronized with the cloud and rain observations, so one-to-one comparison is not fair. I would recommend reformulating/removing all such direct comparisons.

Response We agree with the reviewer's comment that direct comparison of model results and observations is problematic. However, we still believe that the observations of cloud characteristics are useful to be included as a qualitative validation in terms of order of magnitude.

Changes Stimulated also by the comments of both reviewers, we will discuss the results in the revised manuscript mainly in terms of model behaviour and sensitivity, and stick to a more academic approach.

2 Specific comments

Comment 5

P7, Eq. 5: This equation is not valid for hygroscopicity parameter, because some species-specific hygroscopicity parameters are zeros. Did you really used this equation (and how)? This equation can give unrealistic hygroscopicity parameter values (divide by zero) and in that case all calculations should be updated. The correct way to calculate the mode mean hygroscopicity parameters is volume fraction weighted average.

Response We thank the reviewer for pointing out this error in the source code of the model. The mentioned equation (5) was applied, while species with the addition that occurrences of $\kappa = 0$ were left out of the summation preventing division by zero. Nevertheless, the equation is incorrect and will be replaced by the volume-mean average as:

$$\varphi_k = \frac{\sum_i V_i \varphi_i}{\sum_i V_i}, V_i = \frac{m_{i,k}}{\rho_i} \tag{2}$$

For a mode mean aerosol density ρ_k, equation (5) does hold as the occurrences of ρ_i in the numerator cancel out.

$$\rho_k = \frac{\sum_i V_i \rho_i}{\sum_i V_i} = \frac{\sum_i \frac{m_{i,k}}{\rho_i} \varphi_i}{\sum_i \frac{m_{i,k}}{\rho_i}} = \frac{\sum_i m_{i,k}}{\sum_i m_{i,k}/\rho_i} \tag{3}$$

The resulting equation was reused by replacing ρ by κ, for which this cancellation obviously does not happen.

Fortunately, the simulations with the corrected mode mean hygroscopicity only show minor differences. There is only a small differences between the volume and mass-mean average hygroscopicity due to the dominance of sea salt aerosol in the ACS and COS modes. Likewise, the main species in the AIS mode are sulfate (SO4) and organics (POM) which have a similar density (1841 and 1800 kg m^{-3} for SO4 and POM respectively). So in the AIS mode, the mass and volume-mean are comparable as well.

Changes New simulations will be performed using the correct calculation of the volume-mean. The revised manuscript will be updated with the results and figures from the corrected simulations. These modifications are minor and do not affect the results or interpretation.

Comment 6

P12, L27: Maybe the above-mentioned possible bug in hygroscopicity parameter could explain why KAPPA simulations produce much lower cloud droplet number concentration (CDNC) compared with that from the PN simulation? Many other explanations are based on this difference in CDNC (e.g. page 14, line 15-), so a clear explanation is required in any case.
Also, why the interactively calculated CDNCs are so low compared with the available aerosol concentration, and why CDNC seems to be independent of the selected cloud supersaturation? Why does CDNC from the PN simulation decrease with altitude?

Response New simulations were performed using the corrected calculation of the mode volume-mean hygroscopicity parameter. As noted above, the error in the calculation did not cause substantial differences in the cloud characteristics.

The low CDNC in the KAPPA simulation are the direct result of only allowing activation once. As soon as clouds are present in a grid cell new in-cloud activation is prohibited to avoid the 'run-away activation' discussed above. The activated aerosols here are distributed over the whole cloud, which leads to low CDNC without extra in-cloud activation. The changes in S between 0.2 and 1.0% do not change this heavy dilution of CDNC. In the PN simulation, the formulation of activation also severely limits how much of the available aerosol is activated as discussed in the general comment concerning the 'runaway activation'. Both simulations show a decrease of CDNC with altitude as most activation takes place near cloud base.

Changes In the revised manuscript, we will discuss the ratio between aerosol concentration and CDNC and the decrease with altitude for CDNC as mentioned above.

Comment 7

P14, L30 "None of the simulations scores best on all metrics ...": direct comparison of observations and LES simulations is not that simple, but if observations were considered as the truth, would the new KAPPA framework be far from best? Although diagnostic cloud water is accurately predicted, it fails to predict cloud droplet number.

Response The comparison to the observation will be given much less weight in the revised version of the manuscript. However, we still would not argue that the KAPPA framework is far from the best, because that would imply that correctly simulating CDNC is more important than the other metrics.

Changes In the revised manuscript we focus the discussion on how the model outcome changes due to different assumptions and parameterizations and refrain from making statements based on direct comparison with observations. We will highlight that it is difficult to improve all the cloud metrics as follows from the outcome of the different aerosol activation schemes.

Comment 8

P16, L13: Aerosol fluxes for activation (and cloud evaporation) are 12-13 times larger in PN simulation compared with those in KAPPA simulation. The given explanation is based on different autoconversion strengths so that in the KAPPA simulation a larger fraction of cloud water becomes rain before evaporation and is therefore not counted as cloud evaporation, right? If this is the reason, then why cloud-to-rain conversion process strengths are so similar? At least for me, this looks more like a bug than a physically realistic process (see the related general comment). Because meteorology is similar for both PN and KAPPA simulations, there is no physical reason for the large difference between cloud activation fluxes.

Response This paragraph was thoroughly revised. Importantly, the statement: "...the same cloud water is distributed over more but smaller cloud droplets" was incorrect and removed. We are convinced however that the large differences between the two simulations (KAPPA PN) are not caused by a bug. Our conviction is based on two arguments: (1) the meteorological differences which are shown below, and (2) the fundamentally different approach to activation in the two schemes, which allows a higher Nc in the PN scheme than the KAPPA scheme.

Fig 3 panel (b) indicates that the clouds in the PN simulation hold more water than in the KAPPA simulation. By only showing conditional sampled cloud characteristics, the differences between the KAPPA and PN were somewhat hidden. To better illustrate the differences between the simulations, we refer to the figures below. In the leftmost 2 panels, we see that the domain-average cloud water is substantially higher in PN compared to KAPPA (up to +250%). This higher domain-average water load is not only the result of the increased liquid water content in individual clouds as follows from Fig 3, panel (b) in the manuscript. In the rightmost 2 panels, we show that the cloud cover in PN is higher as well. Moreover, by combining Fig 3, panel (d) with data from Tables 3 & 4 we observe that these clouds produce similar amounts of precipitation at the surface and consequently re-evaporate more water.

In conclusion, the PN simulation does produce more clouds, containing more water, but leads to a similar amount of precipitation reaching the surface. These extra clouds thus dissipate and re-evaporate more water back to the atmosphere. This, in combination with activation in the PN simulation leads to the substantially higher aerosol fluxes in the clouds.

Changes In the revised manuscript we will summarize this overview of the difference in meteorology in Section 5.1 and refer to it when discussing the aerosol microphysics in Section 5.2.

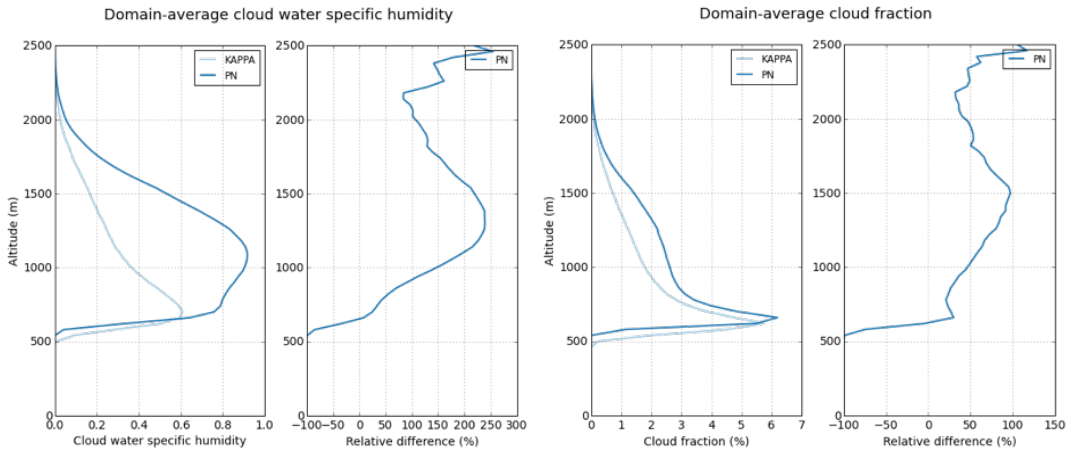


Figure 2: Vertical profile of domain-average (left) cloud liquid water specific humidity and (right) cloud fraction for the KAPPA and PN simulations.

Comment 9

P18, L32-: Average median radius of activated aerosols are different for the KAPPA and PN simulations, and the explanation is related to "stronger cycling of aerosol through the clouds in the PN simulation". What about the effect of supersaturation? It is fixed (0.4%) for KAPPA, but depends on updraft velocity for PN. Lower supersaturation in the PN case could explain the difference in median radius.

Response Nc is higher in the PN simulation than in the KAPPA simulation. This implies that a larger fraction of the aerosols activate. Since both schemes assume that activation of the aerosols progresses from large to small, the higher Nc in the PN simulation goes together with the activation of more small aerosols.

The combination of a higher Nc and a larger average in-cloud aerosol size can therefore not be caused by a lower (effective) supersaturation, and must be the result of the changes of the aerosol distribution by cloud processing.

3 Technical corrections

Comment 10

P1, L3: "feedback between clouds"?

Response & Changes Description made more specific as follows:

These models have a spatial resolution high enough to resolve clouds and associated microphysics. This is combined with domain sizes large enough to simulate macroscale dynamics and mesoscale cloud structures.

Comment 11

P1, L10: "in this pristine ocean environment virtually all aerosols enter" - not all aerosols, but those that activate, right?

Response The purpose of this sentence is to point out that the aerosol (mass) in the cloud droplets is the result of activation. We agree that "in the cloud (phase)" can be understood differently as "in the cloud". This can then imply both activated and interstitial aerosol which is not what we intended to say here.

Changes In the revised manuscript, the sentence is changed so that it is emphasized that we mean the aerosol mass in cloud droplets:

"We find that in this pristine ocean environment virtually all aerosol mass in the cloud droplets is the result of the activation process, while in-cloud scavenging is relatively inefficient."

Comment 12

P2, L17: "which influence further ACI"

Response & Changes In the revised manuscript, this general reference to ACI by a more detailed description as follows:

"Moreover, processing of the aerosol population by one cloud influences the microphysical processes in subsequent clouds. For example, when one cloud depletes the aerosol population by wash out, this might lead to larger clouds droplets in the subsequent cloud formed on the depleted aerosol population. The might lead to faster rain formation and an even further depletion of the aerosol population. This underlines the non-linear character of the interaction between aerosols and clouds and the need to simultaneously simulate the clouds and the aerosol population."

Comment 13

P2, L25: Please clarify "bulk" and "numerical" methods.

Response & Changes Changed text to exclude specific terms like 'bulk' that refer to the way models represent cloud and/or aerosols. This is elaborated upon in the next paragraph.

Sentence change to: "Although methods based on a fixed cloud droplet number, or fixed (infinite) ambient aerosol concentration are almost completely replaced by methods that do consider the aerosol size distribution in a prognostic way. Aerosol composition, however, is often assumed to be uniform."

Comment 14

P2, L33: There is also an ECHAM version with SALSA microphysics.

Response & Changes The reason for including a reference to ECHAM here is to point to models using M7. There indeed is a version of ECHAM with SALSA, but to our knowledge, M7 is still the default microphysics scheme, even in the most recent cycle of the 'ECHAM family' ECHAM-HAMMOZ.

Comment 15

P2, L34: Why "However" here?

Response & Changes To emphasize that the fixed distribution shape is the simplification that is made to achieve the previously mentioned computational efficiency.

Comment 16

P5, L14: "of the originating free aerosol mode"

Response & Changes Sentences are rearranged to clarify cause and effect:

"This modal approach leads to the implicit assumption that the in-hydrometeor aerosol mass is assumed homogeneously distributed across the cloud or rain drop distributions, i.e. aerosol concentrations do not change with hydrometeor size. As a result, size (and mass) information of the originating free aerosol mode is lost once aerosols are incorporated in cloud and raindrops."

Comment 17

P6, L19: S is saturation ratio, right?

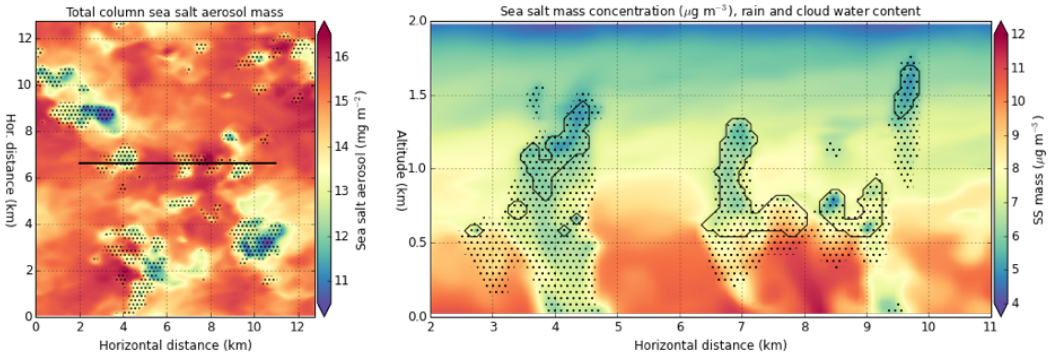
Response Correct.

Changes In the revised manuscript, we will highlight this in the description of Eq. (1), but opt to keep using the term supersaturation in the main body of the text.

Comment 18

P11, Fig. 2: The unit of sea salt mass concentration is more likely micro than milligrams per cubic meter. Also, would it be possible to separate clouds and precipitation or otherwise indicate cloud base height to the vertical cross section?

Response & Changes The unit is corrected in the revised manuscript (μg m^{-3}). Cloud (outline) and rain (hatching) liquid water is now indicated separately in the figure as shown below.



Comment 19

P11, L7: "κ-KAPPA"

Response & Changes Typo corrected to "KAPPA".

Comment 20

P13, Fig. 3 (and Fig. 4): Altitude range could be increased to show also cloud tops.

Response & Changes Cloud tops in our simulation do not reach much further than 2500 m. We left out the upper- most part of the vertical profile here because the statistics in Fig. 3 can be misleading at the highest levels because very few clouds reach that altitude. We chose the vertical range in Fig. 4 to be consistent with Fig. 3. Nevertheless, we will increase the

altitude range to include all cloud tops in both figures.

Comment 21

P17, L11-12: Unclear sentence

Response & Changes We have rewritten the sentence to immediately make clear that we compare the fate of the in-rain aerosol vs. the fate of rainwater itself:

"The abovementioned balance between the two sink processes for in-rain aerosol (i.e., resuspension vs. sedimentation) is substantially different than for the rainwater itself, in which 93 (KAPPA) or 83% (PN) of the falling precipitation evaporates leading to the resuspension of only 50-55% of the in-rain aerosol mass."

Comment 22

P19, Table 5 and related text: Maybe 1 nm accuracy would be good enough?

Response & Changes Agreed, we adopt the suggested accuracy of 1 nm.

Comment 23

P27 -: Journal names should be abbreviated

Response & Changes We checked the complete list of references and abbreviated all journal names using Caltech Library Services (www.library.caltech.edu/reference/abbreviations)

Comment 24

P28, L25: Manuscript is already published in GMD

Response & Changes Changed the reference to the final version: Kurppa et al. (2019)