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Title: Characterizing observed aerosols and their impacts on shallow cumulus clouds during the TRACER field campaign

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Recommendation: Accept after major revisions

This study uses aerosol observations from the TRACER intensive operational period in La Porte, Texas, to identify four dominant aerosol regimes through K-means clustering. Representative aerosol size distributions from these clusters are then used in large-eddy simulations of shallow cumulus clouds with CM1, comparing a bulk microphysics scheme with the super-droplet method. The bulk scheme produces precipitation too early and too strongly relative to both observations and the super-droplet simulations for the considered intercomparison case, which the authors attribute to underestimated cloud-droplet number concentrations which then produce excessive parameterized collision–coalescence rates.

The observational–modeling framework is potentially valuable, but the interpretation of the simulation results would be strengthened by a clearer and more complete discussion of the assumptions underlying the bulk microphysics scheme, together with additional analysis of the cloud-water mass and number budgets and/or one or more targeted numerical experiments to identify the processes responsible for the differences between the simulations.

Major Revisions

1. lines 326-8: "The CDNC variation [between simulations] can be simply explained by the variation in aerosol number concentration; the more available aerosol particles, the higher the domain-mean CDNC.' The key here is "available aerosol." For a specific supersaturation, only the subset of the aerosol particles with critical supersaturations less than the supersaturation will activate. That fraction appears to be very small for clusters 2 and 4, which have total aerosol concentrations over 2000 cm^{-3} yet have CDNCs of about 50 cm^{-3} towards the end of the simulations. That means the "available aerosol" for activation is less than 2.5 %. Even the largest aerosol mode, with GMD of 170

nm has a concentration of 312 cm^{-3}, so only a fraction of this mode is activated. This statement needs to be clarified by stating exactly what is meant by “available aerosol.”

2. lines 341-51: Your discussion based on Figures 7 and 8 might be able to provide more robust conclusions if you analyzed all terms of the CDNC budget as well as of that of cloud droplet mass budget. As it stands, you can only speculate. The results and discussion presented in Section 3.3 also strongly motivate analyzing the budgets of cloud number concentration and cloud water mass.

   As noted by the authors, the low bulk CDNC may reflect not only activation physics and cloud-droplet removal by autoconversion and accretion, but also limited aerosol availability for continued activation aloft. The authors could provide a quantitative cloud-droplet number budget together. Without separating aerosol transport and availability, activation, and subsequent cloud-droplet number loss, the cause of the bulk–SDM differences in CDNC and rain production may be difficult establish.

   As an alternative to performing such analyses, which could be time consuming, you may be able to learn a great deal about the reasons for the differences between bulk and SDM for this case by doing at least one additional simulation, as described in the next comment.

3. This reviewer believes that a particularly useful sensitivity experiment would be to repeat the Total Observed bulk simulation with the Khairoutdinov–Kogan autoconversion process disabled, including both the cloud-water mass transfer and its associated cloud-droplet number sink. In the corresponding SDM simulation, the reported droplet-to-raindrop conversion is essentially zero, whereas the bulk scheme produces substantial conversion. Thus, suppressing bulk autoconversion would remove a process that is negligible in the SDM result and would help isolate the origin of the large CDNC difference. If the bulk CDNC then approaches the SDM value, this would demonstrate that autoconversion and the resulting collision–coalescence sinks largely maintain the anomalously low bulk CDNC. If it remains low, the discrepancy would instead point primarily to aerosol availability, activation, or other aspects of the bulk droplet-number treatment. An intermediate response would quantify the contributions of both mechanisms. This experiment would complement, rather than replace, the requested cloud-droplet number budget.

4. lines 166-68: The bulk scheme uses the parcel-based Abdul-Razzak–Ghan activation parameterization, whereas the SDM represents activation through the

supersaturation histories of individual superdroplets. The manuscript should specify the vertical velocity supplied to the activation parameterization, any imposed subgrid or minimum-updraft treatment, and where (cloud base or above cloud base) activation is calculated.

5. lines 380-81 and lines 166-68: The manuscript should clarify how supersaturation and condensational growth are treated in the bulk microphysics scheme. If saturation adjustment is used, excess water vapor is instantaneously condensed to restore saturation, so supersaturation is not prognosed and condensation is not explicitly controlled by droplet number or surface area. This treatment may be acceptable for some bulk cloud simulations, but it is potentially problematic in an LES comparison with the SDM, where supersaturation fluctuations and finite-rate condensational growth are explicitly represented. The authors should therefore discuss whether differences between the two simulations may partly reflect their fundamentally different treatments of supersaturation, rather than differences in aerosol activation or collision-coalescence alone. A prognostic-supersaturation bulk formulation, such as that of Morrison and Grabowski (2008), could provide a more physically consistent basis for comparison.

   Morrison, H., and W. W. Grabowski, 2008: Modeling supersaturation and subgrid-scale mixing with two-moment bulk warm microphysics. Journal of the Atmospheric Sciences, 65, 792–812, `https://doi.org/10.1175/2007JAS2374.1`

6. lines 386-88: "This difference is another counterintuitive discrepancy between the two simulations given that the bulk simulation gets an infinite supply of aerosols while the availability of aerosols is limited in the SDM run (i.e., Sect. 2.2)." As noted in the comment for lines 326-8, the key here is "availability of aerosols." For example, one could have a very large concentration of very small and/or hydrophobic aerosols, yet no activation in otherwise identical conditions. Or even have exactly the same size distributions of aerosols, yet encounter different supersaturations, so that many aerosols are activated (become "available" to become cloud droplets) in one case compared to the other. This statement should be revised to reflect other factors that control the "availability" of aerosols to activate.

7. lines 399-407: The explanations for the differences between the bulk and SDM runs in this paragraph are not convincing. There are many differences between the simulations with the bulk scheme and the SDM scheme. These include

not just that aerosols are processed and advected in the SDM run but not in the bulk model which leads to differences in the aerosol profiles in the later parts of the simulations, but also that supersaturation is prognosed in the SDM scheme and implicitly diagnosed by the ARG scheme in the bulk model, the collision-coalescence process is explicit in SDM but parameterized in the bulk scheme, the hydrometeor mixing ratios are advected using an Eulerian method in the bulk scheme but all particles are transported using a Lagrangian scheme in the SDM model. I am not sure what to recommend, other than that this paragraph needs to be completey revised based on more extensive analyses and/or numerical experiments, or simply omitted.

Minor Revisions

1. line 15: “particles sized around 180 nm” – state whether diameter or radius.
2. line 18: “these aerosol clusters” – how many clusters? Two or four?
3. line 60-61: As your study and many others show, numerous factors in addition to aerosol abundance play roles in convection and precipitation. Therefore, references to some key review articles might be helpful here, especially in regards to the roles of aerosols as well as meteorology in determining cloud properties.
4. line 64: Please state which cloud properties Huang et al. studied.
5. line 144: Change “weighed” to “weighted.”
6. line 154: Change “Based on the results from the aerosol clustering above” to “Based on the results from the aerosol clustering described in section 3.1.”
7. line 155: Change “double periodic” to “doubly periodic”
8. line 156: Change “50-m horizontal resolution” to “50-m horizontal grid size” because resolution and grid size are not the same. Same for “vertical resolution is 30 m.”
9. line 174-75: You wrote “The collision kernel of Hall (1980) is chosen for this study.” More accurately, Hall (1980) provides/tabulates collision *efficiencies* for the hydrodynamic-gravitational collision kernel. For completeness, you could also mention this applies to laminar flows.
10. line 177: Please clarify “Subgrid-scale velocity fluctuation is accounted for in SDM but subgrid-scale supersaturation fluctuation is not.” Accounted for in terms of which processes? And for context, please mention how the same

processes are accounted for in the Abdul-Razzak and Ghan (2000) aerosol activation scheme.

11. line 205-6: "The large-scale advections were modified slightly to better match observations." Observations of what? Please state.

12. line 218: Should "deflection point" be "elbow point"?

13. Figure 7: I assume that observed radar reflectivity is available. This could be used for comparison to the simulations.

14. Figure 9, lines 396-8: Point measurements of convective rainfall are not useful for comparison to domain means of simulations due to large spatial and temporal variability of convective rainfall. Observed radar reflectivity if from a scanning radar, would be much more useful. Conclusions drawn from the observed point measurements are almost certainly not reliable or robust and should be omitted from the text.

15. line 379: "In-cloud-mean cloud mass is 29% higher on average in the SDM run throughout the simulation." Cloud mass needs to be defined: is it liquid water path? or liquid water content at some level?

16. line 381-82: "This tendency of lower cloud mass in the bulk run, alternatively, can be a result of numerical diffusion." You should state what field(s) you are referring to. If you are referring to the bulk hydrometeor fields, then yes, numerical diffusion could be a factor, but there are many other possible factors as well. It seems you might be referring to numerical diffusion in a bin scheme (lines 434-7). However the bulk scheme is not a bin scheme, so this specific type of numerical diffusion would not apply to the bulk model.