Interactive comment on “Microphysical processing of aerosol particles in orographic clouds” by S. Pousse-Nottelmann et al.

C. Knote (Referee) christoph.knote@lmu.de
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We would like to thank Christoph Knote for his elaborate and detailed suggestions and comments. There are very helpful in ameliorating the manuscript.

** Synopsis ** The authors describe a parameterization of the processing of ambient aerosol by clouds which they implemented into the mesoscale numerical weather prediction model COSMO. The authors investigate the effects of cloud passage on the development of subsequent clouds downstream by conducting 2-D idealized simulations of the flow over two hills with orographic clouds developing over both. Simulations were made with warm as well as mixed-phase clouds, and employing an existing parameterization of aerosol-cloud interactions (but without aerosol processing). The authors find strong effects on the aerosol population when including aerosol processing into their simulation, ranging from a vertical redistribution due to evaporating precipitation as well as a change in the size distribution due to cloud processing.

** General remarks **
The manuscript is well written, the methodology used is state-of-the-art and explained carefully, findings and conclusions are presented in a concise manner. The topic of the manuscript fits in the scope of Atmospheric Chemistry and Physics. A number of very interesting findings are presented and conclusions are drawn that will be useful to the aerosol community. I have a few concerns listed below which I would like to see addressed, but otherwise recommend publication in ACP.

** Major comments **

1. The authors use the modal approach to approximate the aerosol size distribution. A process acting on the size distribution can change (at most) three parameters: the overall number or mass, the median diameter and the width or shape of the distribution function (the standard deviation). It is well known that the typically observed aerosol size distribution, that is, the result of all processes acting upon the aerosol population, can be well approximated using a superposition of lognormal distribution functions. I doubt, though, that this translates directly into the validity of this approximation when describing single processes acting upon it. Let me give two simple examples:
    - activation to cloud droplets: how can one activate all particles above 35 nm (but none below) into cloud droplets, which implies removing this mass/number from the interstitial size distribution, but end up with a function that is still a lognormal?
      You are right that activation scavenging causes an alteration from log-normal as larger aerosol particles are removed from the modes. The scavenged aerosol mass and number are calculated as described in the model section. They correspond to the tail of the log-normal distribution. This part of the aerosol size distribution is then cut out. After activation scavenging, the remaining aerosol particles are described by new lognormal size distributions, which represents the fundamental assumption of the modal approach. Scavenging thus translates in a decrease of the peak hight of the lognormal distribution and a shift of the mode radius towards smaller sizes. In the M7 module, the standard deviations are hold fixed.
    - the efficiency of below-cloud scavenging by rain (impaction, interception) is a strong, non-linear function of the size of the particle. How does one fold a lognormal with a size-dependent function of scavenging efficiency and end up with a lognormal again?

See above. You are right that below-cloud scavenging causes a deviation from log-normal that is lost when the new log-normal size distribution with reduced number concentration and mode radius is calculated.

I am well aware of the fact that this is a criticism of the modal approach in general and it is not the point made in this manuscript to address this issue. However, the processes involved in cloud processing of aerosols are prime examples of processes that, in my point of view, cannot be accurately described using a modal description of the aerosol size distribution. As investigating the effects of those processes is the aim of the authors, I am left wondering what trust we can have in the results of an investigation of these intricate details of multi-phase interactions between clouds and aerosols when the aerosol description itself is approximated in a way that is unable to reflect the processes of interest. This should at least be discussed in the methods section.
A detailed discussion of the advantages and shortcomings of the modal approach is beyond the scope of our work. Computationally cheaper, the modal approach allows to conduct 3D longterm simulations. Comparing the modal and sectional approach in a global 3D model Mann et al. (2012) found that differences between the two approaches were less than model-observation differences. However, they underline that the size-resolved aerosol properties in modal schemes need to be benchmarked and improved against sectional schemes and observations. Applying a two-moment aerosol bulk microphysics scheme with an explicit treatment of aerosol activation and scaled rates for the other microphysical processes, Lebo and Morrison (2013) obtained similar results between the modal and the sectional approach. The explicit parameterizations of aerosol activation and below-cloud scavenging processes in our model were evaluated against observations by Zubler et al. (2011). We scaled the other aerosol process rates to the cloud microphysical process rates which naturally introduces uncertainties to our results. We added this discussion.

2. The authors present a modeling study and do not compare their results against measurements. The underlying parts of the modeling system have been evaluated in a multitude of ways, but the processes added are based on a number of assumptions which bear considerable uncertainty. Hence, I deem it imperative to identify key parameters of uncertainty and conduct sensitivity studies to understand the uncertainty in the final result. As the authors already suggest in the conclusions that the next step will be realistic 3-D studies, this manuscript is somewhat of a "last chance" (and a low hanging fruit as well!) to derive uncertainties on the process level using a model setup that is simple enough so that clear connections between cause and effect can be made. Some examples: how does a wetter/dryer atmosphere affect these processes? What about very clean vs. very polluted conditions? How does overall aerosol solubility affect cloud development? As far as I understand from Zubler et al. (2011) the standard deviations of the modes are fixed, how does this affect the result? The authors stress the importance of precipitation on aerosol processing, but the warm cloud case is non-precipitating. I suggest adding a precipitating warm-cloud simulation.
We agree that the identification and evaluation of uncertainties of the final results is important. Therefore, we will broaden the present work by sensitivity studies and include the findings in the revised manuscript.

3. It is unclear from the manuscript how the transport of aerosol mass incorporated in strongly sedimenting hydrometeors (rain, graupel, ...) is treated numerically. During one time step in the model (10 s), a rain drop falling with 5-10 m/s will fall through several layers in the model. Hence at least in the operational cloud microphysics scheme in COSMO (not the 2-moment scheme used here) a special semi-implicit scheme is used to describe the sedimentation of precipitation. How is sedimentation of hydrometeors treated in the model,

and do the authors apply the same method for the in-rain / in-graupel aerosol mass (e.g. as we did in Knote and Brunner, 2013)?
The parameterization of hydrometeor sedimentation is based on corresponding number and mass weighted mean fall velocities as described in Seifert and Beheng, 2006. Within one model time step, hydrometeors may fall through several vertical model layers as the sedimentation flux calculations are done on a smaller time step. We scale the corresponding flux to the in-hydrometeor sedimentation flux.

The authors showed that there are intricate connections between the locations where the different processes take place. If the in-rain aerosol mass does not fall in the same way the rain drops do - e.g. because the in-rain sedimentation is diagnosed from the precipitation flux - the location at which it is released upon evaporation, for example, is wrong. As the authors present vertical redistribution of aerosol mass as one of the major findings, the underlying methods need to be described in more detail.
In our model, in-rain aerosol mass and rain drops fall in the same way. Therefore, the locations of rain drop evaporation and the release of a new aerosol particles upon rain drop evaporation coincide. We will describe this in more detail.

** Minor comments **
2412, 10: is the standard deviation fixed in this implementation of M7 or is it a prognostic variable?
We added:
"In M7, the aerosol number and mass concentrations are prognostic variables whereas the standard deviations are hold fixed."

2412, 11ff: the aerosol module does not include the major inorganic ions nitrate (NO3-) and ammonium (NH4+), neither does it consider thermodynamic equilibration of these semi-volatile compounds and organics (OC) with the gas-phase. NO3-, NH4+ and OC together typically constitute the majority of observed aerosol mass. The authors should at least briefly mention these omissions and discuss possible influences on the results.
We added:
"The M7 aerosol module does not account for nitrate and ammonium aerosol components as it does not include a treatment of the thermodynamic equilibration of these semi-volatile aerosol compounds with the gas phase. However, these constituents may represent an important part of the aerosol population and modify the surface properties of other aerosol particles due to coatings increasing aerosol growth and activation to cloud droplets."

2415, 4-5: this sentence reads as if the authors would know its bad but keep it for the sake of consistency. If this is the intention, it would be helpful to hint to the reader what the problem is so he/she can understand possible effects on the results. Otherwise please rephrase.
We changed it to:
"To be consistent with the standard parameterization of cloud droplet activation in the model we maintain this calculation of the updraft velocity in our simulations though not imperative at a horizontal resolution of 2 km."

2415, 11-12: see major comment - how can one cut off the tail of a lognormal and still keep a lognormal?
See answer to major comment.

2416, 26: unclear formulation: how is the scavenging by rain happening locally in a grid cell influenced by the amount of rain reaching the ground if precipitation (and hence the incorporated particles) are prognostic quantities? Please explain more thoroughly.

We changed it to:
“The parametrization is based on the scavenging coefficient following the equations in Zhao and Zheng (2006) and Croft et al. (2009), as described by Zubler et al. (2011a). In order to consider only the final removal of aerosol particles from the atmosphere by surface reaching hydrometeors, Zubler et al. (2011a) multiplied the rate change of the tracer mass and number densities due to scavenging by rain (or snow) by the fraction of precipitating rain (or snow) reaching the surface. In the new aerosol processing scheme, not only the surface reaching hydrometeors, but all precipitating rain drops and snow flakes may scavenge aerosol particles. In this scheme, the scavenging coefficients are thus independent from the fraction of surface reaching precipitation.”

2416, 28ff: the reasoning that the authors omit graupel "because of the large sedimentation velocity" is not convincing as rain has even higher sedimentation velocities. Please explain and rephrase in the manuscript.
We changed it to:
“Scavenging by falling graupel is omitted.”

2420, 19-21: that is a good start - regarding the sensitivity studies mentioned in the major comments it might be interesting not only to vary the overall level of "pollution", but also consider more realistic vertical profiles (e.g. how does a dirty boundary layer influence cloud development in cleaner upper levels vs. equally dirty upper levels).
We will evaluate this suggestions for the sensitivity studies.

2424, 24-26: why do the authors need to set standard deviations, are they not given by M7?
In the setup used for this study, the standard deviation (sigma) of the Aitken and accumulation mode are adjusted to the observations. (sigma= 1.59 for the nucleation mode, sigma=2.13 for the Aitken mode, sigma=1.61 for the accumulation mode, and sigma= 2.0 for the coarse mode).

Fig 17: the isolated peak of high cloud droplet number concentrations in a) (around 750 km distance / 3500m height) seems odd - I guess this is due to the overall low liquid water content in the cold case?
You are right that the liquid water content is rather low in the cold case. The isolated peak in LWC is not taken into account in our results.

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