Rev 2: LIMA paper for GMD

Major Comments
1. Model Physics and Equations

The use of "the prognostic evolution of a three-dimensional (3-D) aerosol population" in the abstract is somewhat confusing.
"3-D" was referring to true three-dimensional aerosol fields in contrast to single value homogeneous aerosol fields which are often used to treat the indirect effect (Cs^k law for CCN activation and Meyers' formula for heterogeneous ice nucleation). We propose "the prognostic evolution of an aerosol population" which still keeps the idea of heterogeneous aerosol concentrations and properties as in the real world.

It is also not clear why the authors chose a factor of 1.2 times the rain water mixing ratio to demarcate the boundary between conditions without accretion and self-collection and those with these processes.
This factor was suggested indirectly by Berry and Reinhardt (1974) scheme (hereafter BR74). These authors made the distinction between the time T_2 needed for a characteristic radius of the rain spectrum to reach the value of 50 µm (and thus to accumulate a rain mixing ratio of L) and the time $T_H \approx 1.2*T_2$ at which a hump shows up on the rain spectrum (with BR74 notations). During the T_2-T_H transition, the autoconversion rate (L/T_2) of BR74 is supposed to include spuriously cloud droplet accretion and raindrop self-collection. This is why the application of the explicit parameterizations of rain accretion and self-collection are delayed until a "well-formed" rain mixing ratio reaches as least $1.2*L$. At this point, the production rate of raindrop concentration by autoconversion is also modified as explained in Cohard and Pinty (2000a).

I am also concerned about the lack of prognostic supersaturation in the model, especially given all of the effort that has clearly been taken to include a more physical representation of the ambient aerosol population.
The scheme doesn't allow for supersaturation over water (adjustment to strict saturation) while supersaturation over ice is free and unconstrained as illustrated in Fig. 6 of the manuscript. The mixed-phase case is such that again, the water-vapor mixing ratio is bounded by the saturation value over water.
There are several arguments to support this choice:
1- Supersaturations are probably much less than 1% in warm unpolluted clouds (see Morrison and Grabowski, 2007); the peak value is confined in the first tens of meters above cloud base where condensation on droplets competes with CCN activation. So cloud layers with expected supersaturated conditions over water are probably barely resolved most of the time when simulating clouds at convection-resolved scales with a vertical grid spacing of 100-200 m (so with the exception of moist LES when simulating stratus and stratocumulus clouds). A mean grid spacing of 200 m implies 75 model levels approximately to describe the gravity waves in the atmosphere which propagate well above the tropopause.
2- To the authors' knowledge, supersaturation over water is not measured in clouds, meaning that there is no possible experimental check of simulation results. Probably the supersaturation field would show a lot of fluctuations too, especially at the cloud base interface where air is highly turbulent. In passing, a mean supersaturation at model resolution may not be appropriate to represent an instantaneous peak value for activation schemes.
3- For a cloud base at 850 hPa, a large supersaturation of 1% corresponds to an uncertainty of 1% of the water vapour mixing ratio at saturation that is 0.17 g/kg (equiv to 0.31 K) at T=293

K and 0.045 g/kg (equiv. to 0.15 K) at T=273 K. These values correspond to a modest buoyancy acceleration ($\approx g^*\Delta\theta/\theta$) of 0.01 and 0.005 m/s^2 respectively.

4- The equation of the "water vapour deviation to saturation" (hereafter $\delta q_v(t)$) is a first order differential equation, given in Lebo et al. (2012), Morrison and Grabowski (2008) or Reisin et al. (1996) but with a complex forcing term involving water vapour and temperature tendencies. This equation can be integrated analytically with initial condition $\delta q_v(0)$ at the beginning of the time step. Then $\delta q_v(t)$ is averaged over the running time step Δt as suggested earlier by Sakakibara (1979). As a result, $\delta q_v(t)$ is a redundant variable to be used to compute condensation and evaporation rates in the t, t+Δt interval but without historical feedback (or temporal filter to mitigate successive $\delta q_v(t)$ samples) as the true prognostic variable is still the water vapour mixing ratio $q_v(t)$.

5- The true difficulty is more to define a saturation level (and hence the supersaturation over water in the presence of pristine ice crystals) in the case of mixed phase clouds (see Reisin et al., 1996). In this situation we adopted the Reisin's scheme but with the constraint of no supersaturation over water (a positive supersaturation over ice is inherent to the situation). This is necessary to remain consistent with the treatment of the warm clouds.

In conclusion, we don't claim that estimating the supersaturation over water to compute a condensation rate, is useless. However we feel that errors arising in the computation of water vapour and temperature tendencies (see above) and the weak difference of buoyancy when considering supersaturation or not, is far below known model errors due to model numerics or more basically to the order physical processes are integrated in cloud schemes.

One thing that is not address in the paper is how collisional processes affect the number of activated CCN and nucleated IFN. As collisions occur, the number of particles should decrease. Thus, the reactivation of evaporating drops, for example, may be incorrect if such a process is not accounted for.

The aerosol model doesn't consider the growth by gas deposition & coagulation, the sedimentation, and Brownian & phoretic diffusion. A single aerosol CCN or IFN is released each time a cloud droplet or an ice crystal evaporates. This is done in proportion of the different sources of activated CCN or nucleated IFN. This is indeed an approximation for the IFN as some ice crystals may form by the Hallett-Mossop mechanism or by homogeneous freezing of cloud droplets. The complete evaporation of the raindrops produces neither giant CCN nor IFN.

Please also review Equation 7.

Eq. 7 is correct but Eq. 6 is not. We apologize for that. The denominator should be written with factor $(\psi_1\omega + \psi_3 dT/dt)^{1/2}$ instead of $(\psi_1\omega)^{1/2}$ as usual. Here we recall that the source of supersaturation is twofold, an upward transport of humidity in the case of convective air parcels, but also a total cooling rate (third term in Eq. 4) which is dominant in the case of fog (infrared cooling). This last term was given in the appendix of Morrison and Grabowski (2008). To avoid confusion with the derivation made by Pruppacher and Klett (1997) and by Cohard et al. (2000), we simply added "… with B the Beta function. The derivation of Eq. 6 includes the cooling rate term of Eq. 4 which is often neglected in previous works."

Similarly, is the power of 20 on the first equation on Page 7780 correct? The power is 12 in the cited reference. Perhaps this is a unit conversion difference? If so, please make this clear in the paper.

The power of 20 is necessary to convert cm into m. The original power 12 must be increased by 8 to convert σ_c and d_c in MKS units as the total power of these parameters is 4. We added

"… The 10^{20} and 10^{6} factors account for unit conversion as Berry and Reinhardt (1974) original expressions are not MKS." to draw attention to this unit conversion.

Please check the units of K1 and K2; the units appear to be incorrect/inconsistent.
We thank the reviewer for the detection of this typographical error. K1 should be in s^{-1} while unit of K2 is the good one (this explains why there is an additional power 6 to the Long's original expression to convert cm into m). This is corrected now.

The approximation to the solution of Equation 8 was at first not clear to me. Perhaps being a bit more thorough would help the reader through the derivation, e.g., something as simple as "using the following assumption: for $x<<1$, $1-e^{x}\approx x$*".*
The first order expansion approximation is described in the appendix. The derivation is easy to follow. Reference to the appendix is made in section 2.4.2 which follows section 2.4.1 where Phillips' parameterization is introduced. Note that the same approximation is suggested in the original paper of Phillips et al. (2008).

On page 7784, the production of graupel at temperatures below -35° C is confusing. I think what you are saying is that frozen raindrops are added to the graupel category in the model at such temperatures. I think it is important to separate model assumptions from physics because, for example, graupel is formed via riming and is not necessarily a frozen raindrop.
The reviewer is true as basically freshly frozen raindrops are not graupel, but graupel embryos until riming rubs out the origin of the particles. As done in the majority of the cloud schemes, frozen raindrops are transferred into the graupel category. In the scheme all surviving raindrops are frozen when they reach -35°C.
We modified the text on page 7784: "the freezing of the raindrops is instantaneous at temperatures below -35°C, and frozen raindrops are added to the graupel category."
We modified the text on page 7785: "They include the light riming of snow with cloud droplets, the wet/dry growth of graupel when collecting other hydrometeors, and the accretion of rain and aggregates. The freezing of raindrops upon contact with an ice crystal leads also to the formation of graupel as frozen drops are not a separate ice category."

Lebo, Z. J., Morrison, H., and Seinfeld, J. H.: Are Simulated Aerosol-Induced Effects on Deep Convective Clouds Strongly Dependent on Saturation Adjustment?, Atmos. Chem. Phys., 12, 9941–9964, 2012.
Morrison, H., and W. W. Grabowski, 2008: Modeling supersaturation and sub-grid scale mixing with two-moment warm bulk microphysics. J. Atmos. Sci., 65, 792-812.
Reisin, T., Levin, Z., and Tzivion, S.: Rain production in convective clouds as simulated in an axisymmetric model with detailed microphysics. Part I: description of the model, J. Atmos. Sci., 53, 497–519, 1996.
Sakakibara, H. (1979), A scheme for stable numerical computation of the condensation process with large time step, J. Meteorol. Soc. Japan 57, 349-353.

2. Test Cases: *I have a few concerns with the test cases chosen in this work.*
(a) Orographic Case: *While I understand that the simulations are intended to be illustrative and for proof of concept, the model resolution seems a bit large (i.e., 5 km in the horizontal). Given that these are 2D simulations with a bulk scheme, I am not sure why a higher resolution was not chosen. I bring this up because I am concerned that some of the microphysical characteristics may be different if one used a higher resolution, e.g., 500 m. Moreover, where did the sounding come from for this case? The very low tropopause (i.e., near 400 mb) seems extremely low. The description of the simulation length is confusing;*

please explain more clearly how the results are presented in the figures and how long the simulations were. In general, I think it would be useful to explain the findings in the context of what we know about orographic clouds from observational and previous modeling efforts. For example, "indicate that black carbon is a more efficient nucleating agent than organics" makes it appears as though this is a result of the current work when in fact this has been established in prior works.

The reviewer is right to notice that the scheme is more adapted to high resolution cloud simulations. So even for this illustrative an academic 2D case, the simulations were redone at 1 km resolution. The conclusions regarding the ability of LIMA to produce different ice crystals concentrations depending on the IFN size, number concentration, and chemical composition, are unchanged.

The radiosounding used to initialize the simulation is similar to the sounding observed at 00UTC on 8 March 2004 from the Jungfraujoch case description by Muhlbauer and Lohmann (2008, their figure 8a).

Following this suggestion as well as a similar remark from Reviewer 1, we modified the manuscript to better explain that the higher nucleation probability of black carbon compared to organics is inherent to the parameterization by Phillips and not a result of our study.

(b) Squall Line Case*: As noted for the orographic case, it is unclear why the resolution is relatively coarse, i.e., exceeding 1 km. These simulations should be extremely short and thus it might be worth improving the grid spacing to better represent the processes that are important for the cases selected. Again, the results should be presented in the context of what we know about squall lines to better demonstrate the capabilities of the model.*

The reviewer is right to question again about a better model resolution but the purpose of this study was not to perform a detailed analysis of the squall line dynamics and processes, but rather to illustrate the behaviour and abilities of LIMA. We chose a resolution around 1km, which is currently used in the french operational meso-scale weather forecasting system AROME, and is representative of configurations used for 3D, real-case simulations.

Again, the purpose was not to study the dynamics of the squall lines, the strength of the cold pool and so the sensitivity of the aerosols to rain evaporation. We believe that the most original issues of the simulations were clearly to show the effects of a strong external aerosol forcing in a well organized cloud system. Here the perturbation depends on the aerosol type (CCN or IFN) and the atmosphere layer at which these aerosols are released. To summarize the results, we showed that LIMA is able to represent the impact of an aerosol plume depending on both its altitude and the aerosol type and size. In this case, the impact may be important on cloud composition, but less so regarding accumulated ground precipitations. However, we expect that other cloud types (such as fog) may behave differently.

Two forthcoming papers will focus on the initialization of aerosols from the near-real-time analyses of the MACC project, from the ECMWF, and on a detailed evaluation of the cloud representation of LIMA using the microphysical observations from the HyMeX campaign for heavy precipitating MCSs.

Minor Comments

1. P7768, line 20 : The sentence was deleted following the reviewer's suggestion.

2. P7769, line 3 : The sentence was changed to “The experiments show that LIMA responds well to the complex nature of aerosol-cloud interactions (...)”.

3. P7769, lines 14-17 : The unclear sentence was changed to : “The complex interactions between aerosol particles, clouds and precipitation strongly affect the evolution of the atmosphere and its dynamics at all temporal and spatial scales. Accounting for this interplay is important for high-resolution cloud modelling (aerosols influence the precipitation-forming processes in clouds) and for climate forcing (aerosols influence the radiative-convective equilibrium in many ways), as analyzed by Rosenfeld et al. (2008).”

The reference to Rosenfeld (2008) seems right, but there is a problem with the URL linking. We will check that this is corrected in the final version. The correct URL for this reference is : http://www.sciencemag.org/content/321/5894/1309.short

4. P7769, line 25 : Corrected “aerosol plumes” according to the reviewer's suggestion.

5. P7770, lines 10-11 : References added.

6. P7770, lines 16-17 : Manuscript corrected according to the reviewer's suggestion.

7. P7771, line 17 : Corrected according to the reviewer's suggestion.

8. P7772, lines 2-5 : The confusing sentence was changed to : “The nucleation of aerosol particles is dependent on water vapour amounts brought by vertical updrafts. The resolution of the vertical motion is therefore an essential point in the computation of nucleation processes (Morrison and Grabowski, 2008).”

9. P7773, line 15 : The term “mode” is used for the aerosols in LIMA, because they explicitly correspond to modes of the total aerosol population. For example, a bi-modal aerosol population will be represented in LIMA by two modes with different size distribution parameters. LIMA considers three categories of aerosols : CCN, IFN and coated IFN. Therefore, we chose to keep the current terminology.

10. P7781, line 13 : Manuscript corrected according to the reviewer's suggestion.

11. P7782, line 7 : Manuscript corrected according to the reviewer's suggestion.

12. The manuscript was corrected so that all diameters are denoted by a lower-case “d”. In the previous version, an upper-case “D” was indeed used for hydrometeors.

13. Precisions regarding the averaging and times were added in the figure captions.

14. Since we are aware that english is not our mother tongue, we had the manuscript checked for correctness by an English teacher, a native English speaker, before it was submitted to GMDD.