

LIMA : A 2-MOMENT SCHEME DRIVEN BY A MULTIMODAL POPULATION OF CCN AND IFN

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1 INTRODUCTION

Aerosol particles (AP) modify the cloud micro-structure through their respective capabilities to nucleate droplets or ice crystals. This results from their different nucleation efficiencies, and then affect the microphysical processes, as for instance the delay of formation of precipitating hydrometeors (rain, snow, graupel and hail). Due to the large variability of AP chemical compositions and concentrations, accounting for polydisperse aerosol populations as CCN and/or IN is crucial to limit physical uncertainties in cloud simulations.

Therefore, a 2-moment scheme, LIMA (Liquid, Ice, Multiple Aerosol), focusing on detailed aerosol-cloud interactions, was implemented in the Meso-NH model (Lafore *et al.*, 1998; <http://mesonh.aero.obs-mip.fr>). This scheme is described below, along with the strategy proposed to provide accurate 3D APs for real case studies. The performance of the scheme is shown for both 2D, idealized simulations and a 3D real case of mesoscale convective system.

2 DESCRIPTION OF THE SCHEME

LIMA is based on the ICE3 operational 1-moment scheme (Pinty and Jabouille, 1998), extended to hail, and therefore represents the evolution of mixing ratios for 6 water particle species: cloud droplets, rain drops, pristine ice crystals, snow, graupel and hail. Additionally, the scheme also has prognostic number concentrations for cloud droplets, rain drops and ice crystals.

2.1 Prognostic aerosols

LIMA can distinguish, and handle the competition between, several modes of APs. Each "mode" is characterized by its chemical composition (and their ability to act as CCN, IN, or coated IN), and the modal diameter and width defining its lognormal particle size distribution (PSD). These parameters are fixed for each AP mode throughout the domain and simulation. An example configuration of LIMA is given in table 1. The superimposition of several modes enables a realistic representation of any AP.

For each CCN and IN mode, two prognostic variables are predicted : the number concentration of free aerosols N_{free} , and

activated aerosols N_{acti} , so that the complete aerosol population can always be recomputed. Coated IN in LIMA represent insoluble aerosols with a soluble coating. These particles are first active as CCN to form cloud droplets. Then, when these cloud droplets reach negative temperatures, the particle can act as IN by immersion. Therefore, three prognostic variables are used for coated IN: the number concentrations of free aerosols N_{free} , of activated aerosols in cloud droplets N_{acti} , and nucleated aerosols in ice crystals N_{nucl} .

Mode #	Type	Diameter	Property
1	Sea salt	0.8 μm	CCN
2	Sulfate	0.5 μm	CCN
3	Dust	0.8 μm	IN
4	Dust	3 μm	IN
5	Black Carbon	0.2 μm	IN
6	Organics	0.2 μm	Coated IN

Table 1: Example configuration of LIMA using 6 aerosol modes

The prognostic aerosols are transported by the resolved flow as well as the turbulent motion. They are depleted (transferred from N_{free} to N_{acti} or N_{nucl}) when used as CCN or IN to form droplets and ice crystals as detailed below. When evaporation of cloud droplets or sublimation of ice crystals happen, aerosols are reinjected in N_{free} from the N_{acti}/N_{nucl} reservoirs.

The below-cloud impactation scavenging of aerosols by rain is also carefully treated following Berthet *et al.* 2009.

2.2 CCN activation

The CCN activation parameterization is based on Cohard *et al.* (1998), which was extended to treat the competition between several modes of CCN. The maximum supersaturation is diagnosed from the the convective updraft strength, the radiative cooling, and the growth of droplets by vapor deposition (this last term depends on the aerosol loading).

The number of activated CCN for each mode is then computed as the number of free CCN with a diameter exceeding the critical diameter for the diagnosed maximum supersaturation.

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2.3 *IN heterogeneous nucleation*

IN heterogeneous nucleation is based on the empirical parameterization (EP) of Phillips *et al.* (2008, 2013). This EP is constrained by simultaneous observations of insoluble aerosols in the troposphere, and ice nucleation rates in a continuous flow diffusion chamber under controlled temperature and supersaturation conditions.

One interesting property of this EP is that three types of IN (dust/metallic (DM), black carbon (BC) and organics (O)) are given different nucleating efficiencies. In this EP, the number of nucleated IN also depends on their total surface, with larger particles having a greater nucleation probability.

2.4 *IN-free ice initiation*

Two families of processes can produce pristine ice crystals when no IN are present.

The homogeneous freezing of cloud droplets is described according to Pruppacher (1995). Raindrops instantly freeze at -35°C to form graupel, and, below -40°C , homogeneous freezing of free CCN is parameterized after Kärcher and Lohmann (2002).

LIMA also includes the secondary ice production by the Hallett-Mossop process (Hallett and Mossop, 1974). At temperatures between -3°C and -8°C , an ice splinter is produced each time 200 droplets with a diameter between $12\mu\text{m}$ and $25\mu\text{m}$ are riming on graupel (Beheng, 1987).

2.5 *Water deposition and evaporation*

The water condensation/evaporation and deposition/sublimation processes are handled differently depending on the available condensate:

- where only liquid water (cloud droplets) is present, the scheme assumes an equilibrium at water saturation, and thus an implicit adjustment is performed.
- where only frozen particles are present, the scheme explicitly determines the deposition/sublimation rate, following Tzivion *et al.* (1989).
- for mixed-phase conditions, an implicit adjustment to liquid water saturation is first performed. Then, the explicit

condensation/evaporation and deposition/sublimation rates are determined following the scheme of Reisin *et al.* (1996).

2.6 *Other processes*

- The ice-snow conversion was adapted from the explicit parameterization proposed by Harrington *et al.* (1995), to fit single-moment description of snow in LIMA and the generalized gamma particle size distribution of ice and snow in Meso-NH. The critical diameter defining the ice and snow categories is kept at $125\mu\text{m}$.
- LIMA was interfaced with the radiative transfer scheme by Aouizerats *et al.* (2010), and therefore includes the radiative impact of the whole aerosol population.

3 COUPLING TO MACC AEROSOL ANALYSES

To fully benefit from the detailed representation of aerosols possible with LIMA for real case simulations, an accurate, heterogeneous aerosol population initialization (and lateral boundary forcing) is necessary.

3.1 *MACC aerosol analyses*

The Monitoring Atmospheric Composition and Climate (MACC, www.gmes-atmosphere.eu) project combines state-of-the-art atmospheric modeling with Earth observation data to provide information services covering global atmospheric composition and air quality.

The aerosol analyses are run in near-real-time at the ECMWF, at a T255 horizontal resolution with 60 vertical levels, therefore providing an interesting description of the vertical distribution of aerosols. Aerosols are represented by 11 prognostic mass mixing ratios, for sulfates, sea-salts (3 different size bins), hydrophilic and hydrophobic organics and BC, and dust (3 different size bins).

3.2 *MACC to LIMA conversion*

To be used in LIMA, these aerosol data must be converted to number concentrations, and given nucleating abilities. During this conversion,

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assumptions are made (such as the density and modal diameter for the different species) that can have an important impact on the cloud nucleation in LIMA. A proposed choice of nucleating abilities for the different species available in MACC is shown in table 2.

MACC species	LIMA nucleating ability
Sulfate	CCN
Sea salts (3 bins)	CCN
Hydrophilic OM	Coated IN
Hydrophilic BC	
Hydrophobic OM	IN
Hydrophobic BC	
Dust (3 bins)	IN

Table 2: Nucleating properties of the analyzed MACC aerosol types

4 PERFORMANCE OF THE SCHEME

4.1 Impact of IN properties on 2D idealized cold clouds

To show the impact of the aerosol population on heterogeneous ice nucleation in LIMA, an idealized simulation of orographic cold clouds was set-up, and integration was performed for 8 hours with different aerosol loadings. Figures below are 1-hour averages during the last hour of the simulation.

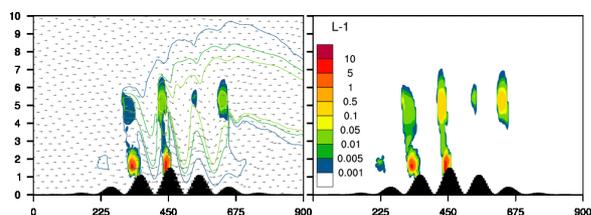


Fig. 1: Ice number concentration (color shading) and activated IN concentration (contours) for the reference simulation (left, IN at $10L^{-1}$) and increased IN (right, $100L^{-1}$).

Fig. 1 shows the ice number concentration as well as the nucleated IN number concentration N_{nucl} for the reference experiment (left). N_{nucl} is clearly transported by the flow from the ice nucleating regions, and therefore the number of available IN is reduced, hindering further ice formation downstream. Fig. 1 (right) shows the ice concentration when the initial IN

concentration was raised to $100 L^{-1}$. Secondary ice production by the Hallett-Mossop process is responsible for the high ice concentrations in lower levels, whereas heterogeneous ice nucleation is dominant in ice formation at colder temperatures (3-8km).

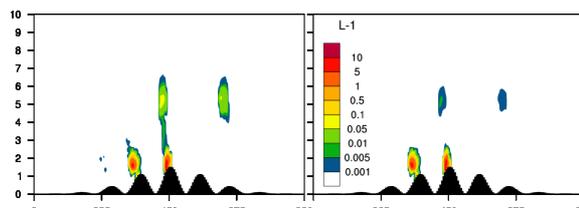


Fig 2: Ice number concentration for pure black carbon (left) or organic (right) IN at $100L^{-1}$.

Fig. 2 shows the pristine ice number concentration for simulations run with one mode of pure black carbon (left) or organic (right) IN with identical size distributions and initial number concentrations ($100 L^{-1}$). It highlights the different nucleating abilities of these species.

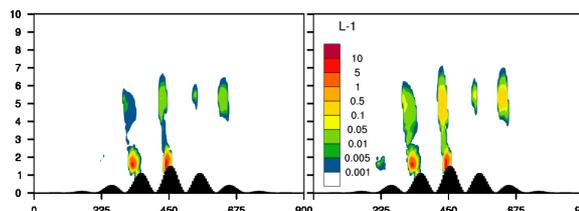


Fig 3: Ice number concentration for pure dust IN at a concentration of $10L^{-1}$, with a modal radius of $0.8\mu m$ (left) and $3\mu m$ (right).

Fig. 3 shows ice concentration from two simulations run with one mode of pure dust IN, with the same concentration ($10 L^{-1}$) but a modal radius of $0.8\mu m$ (left) and $3\mu m$ (right). Here, the biggest particles with a large surface are more efficiently nucleated.

4.2 Impact of aerosol loading and spatial distribution on a 2D idealized squall line

The behavior of LIMA for mixed-phase clouds was examined through idealized 2D squall-line simulations. Simulations were run for 10 hours, and results are presented averaged between 7 and 8 hours of simulation (except for the instantaneous supersaturation over ice).

The warm-phase cloud parameterization in LIMA assumes a water vapor phase in equilibrium at saturation over liquid water. Therefore, no prognostic supersaturation is permitted in this case. In contrast, this constraint is released for the cold phase, where LIMA is able to handle large supersaturation values over ice in strong

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convective updrafts (fig. 4, up to 40% supersaturation for this 2D squall line simulation).

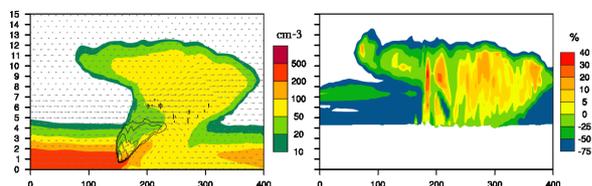


Fig 4: Left: Free CCN (colors) and cloud droplet (contours, same scale from 10 to 500cm⁻³) concentrations. Right: Instantaneous supersaturation over ice where $T < 0^{\circ}\text{C}$, after 8h of simulation.

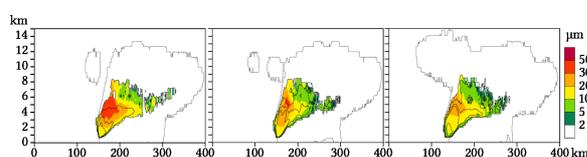


Fig 5: Cloud droplet mean radius for the reference simulation (left), with an additional pollution plume at mid-levels (center) and in low levels (right).

This squall line simulation was also used to test the response of the scheme to an increase in aerosol loading, representing for example a pollution, forest fire, or volcanic ash plume. We found out that the simulations using LIMA are sensitive to the type of aerosol in the plume, but also to the vertical height range at which a pollution plume is released. Fig. 5 highlights such an impact on the mean diameter of cloud droplets for the reference simulation, a simulation, and simulations with an additional pollution plume in the mid-troposphere (around 4km) or in the lower levels.

4.3 Impact of aerosol initialization for a real, 3D heavy precipitating system

To investigate the potential of LIMA for high-resolution real case simulations, a mesoscale convective system which was observed during the HyMeX (www.hymex.org) campaign was studied in this framework. Meso-NH was used at a 2.5km horizontal resolution. It is driven by the atmospheric analyses provided by AROME, the french high-resolution operational model, and by MACC aerosol analyses. The simulations were initialized at 00UTC on 24 September 2012, and run for 12 hours.

Fig. 6 shows the 12-h accumulated rainfall observed by the raingauges, and simulated using the ICE3 operational 1-moment scheme, LIMA with a default, horizontally homogeneous aerosol population, and LIMA initialized and

driven by the MACC aerosols. There is a clear improvement for this case when using the LIMA scheme with realistic aerosols from MACC analyses, although the conversion from MACC data to LIMA CCN and IN has not been carefully calibrated yet. Important differences are also found in the vertical structure of clouds (fig. 7), but weren't compared to the available observations yet.

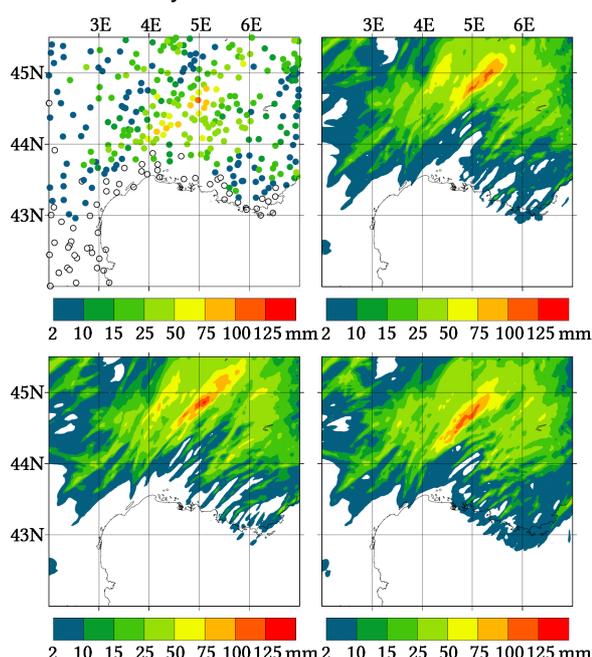


Fig 6: 12-h accumulated rainfall at 12UTC, 24 sept 2012. Rain-gauge obs (top left), simulations with ICE3 (top right), LIMA with default aerosols (bottom left), LIMA with MACC aerosols (bottom right).

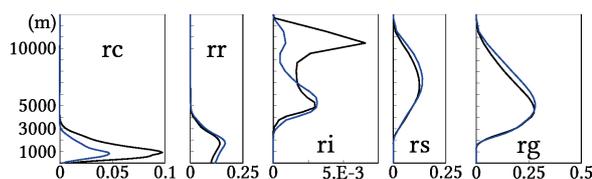


Fig. 7: Vertical profiles of hydrometeors mass mixing ratios (g.kg^{-1}) for simulations with LIMA and default (black) or MACC (blue) aerosols.

5 CONCLUSION AND PROSPECTS

A new 2-moment scheme, LIMA, was implemented in the Meso-NH non-hydrostatic research model. LIMA provides a detailed representation of the aerosol-cloud interactions, especially through a refined CCN activation and IN nucleation and the prognostic evolution of a multimodal aerosol population.

LIMA represents well the impact of varying aerosol chemical properties, concentrations, or

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size distributions, on the droplet activation and ice nucleation, which then have an impact on the cloud structure and dynamics.

For real case simulations, a strategy to initialize and drive LIMA with realistic aerosols provided by the near-real-time MACC analyses and forecasts was established. A first test conducted for a mesoscale convective system over southeastern France shows the potential of LIMA for both forecasting applications and process studies.

5.1 Future improvements

Several possibilities are under consideration to extend the LIMA scheme.

Considering the ice phase treatment, the inclusion of ice crystal habits in LIMA could provide better representation of radiative effects, or more accurate vapour deposition rates and fall speeds. Other options include the addition of an intermediate “large ice crystals” between pristine ice and snow, to better represent the transition, or adding prognostic number concentrations for all ice hydrometeors.

5.2 Evaluation and calibration of the scheme

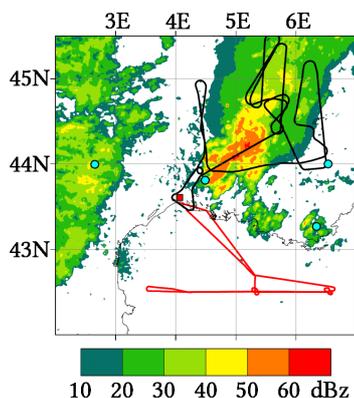


Fig. 8: Radar reflectivities at 07UTC 24 sept. 2012, ATR42 (red) and Falcon20 (black) flight tracks, doppler radars (blue dots) and Raman lidar (red square).

The calibration and evaluation of LIMA will first be conducted with data from the HyMeX campaign, as illustrated on fig. 8.

Aerosol loadings and characteristics provided by the aircraft-based observations by the ATR42 and the ground-based Raman lidar will help to calibrate the conversion of MACC aerosol analyses into LIMA CCN, IN and coated IN

populations.

Aircraft data acquired onboard the Falcon20 (both in-situ microphysical observations and RASTA doppler radar retrievals), combined with ground-based polarimetric radar observations will help evaluate the scheme and check the simulated cloud structures.

Then, the evaluation of LIMA (with MACC data) for contrasted cloudy situations, such as fog, stratocumulus and cirrus covers, will be undertaken in the open-source Meso-NH model (<http://mesonh.aero.obs-mip.fr/>).

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