# Preliminary validation of a $\kappa$-Köhler-based CCN activation model with EUCAARI-IMPACT airborne aerosol observations

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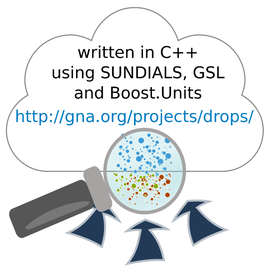
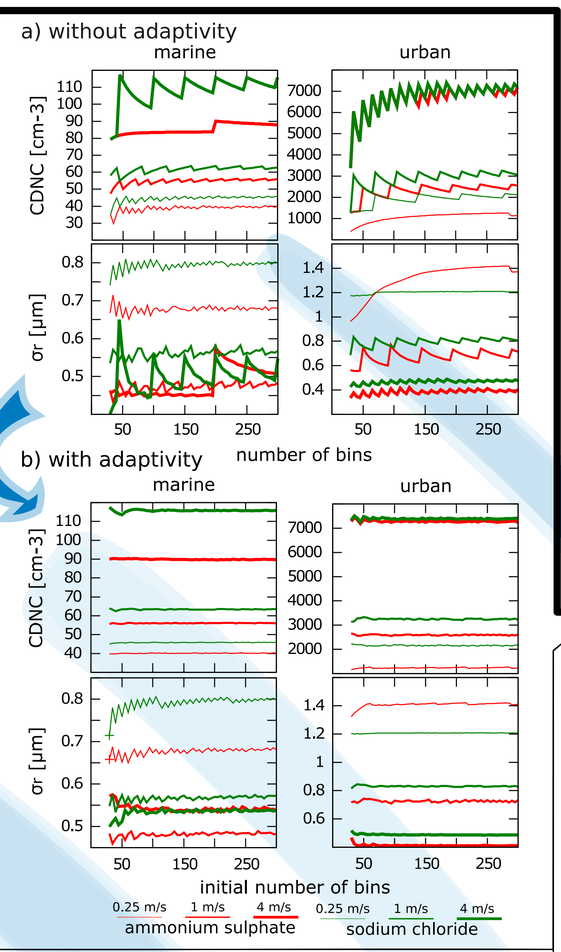
observational data courtesy of:
CNRM/Météo-France: Laurent Gomes, Greg Roberts et al.
CNRS-LaMP: Alfons Schwarzenboeck, Suzanne Crumeyrolle et al.

## CCN ACTIVATION MODEL




The simulations are carried out using a recently developed CCN activation model (Arabas and Pawlowska, 2010).
**The model describes evolution of sizes of an ensemble of solution droplets contained in an adiabatic air parcel being lifted and cooled beyond supersaturation.**
The model utilises the adaptive method of lines (MOL) for numerically solving the so called dynamic equation of aerosol growth by condensation coupled with the heat budget of an adiabatic air parcel. Employment of MOL (Lagrangian representation of particle size evolution) allows accurate description of aerosol chemical composition. That is because particle properties such as mass of solute are retained throughout the computation. Particle composition is represented in the model by a single parameter $\kappa$ using the $\kappa$-Köhler parameterisation (Petters and Kreidenweis, 2007). The rate of growth of a solution droplet at a given supersaturation is derived from the heat and vapour diffusion equations thus the model tracks droplet temperatures explicitly. Model equations are solved employing adaptive timestep adjustments and adaptive size spectrum discretization.

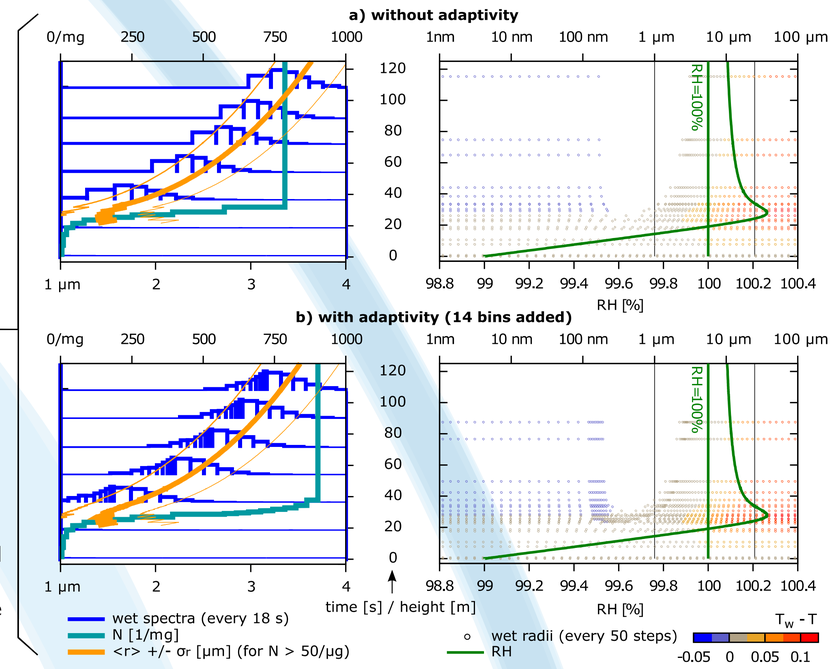
Particle size spectra are approximated in the model using histograms composed of bins whose position and width evolve with time. During activation the largest aerosol particles grow rapidly and the shape of the size spectrum in the region between the unactivated aerosol mode and the activated cloud-droplet mode becomes poorly represented. The adaptive spectrum discretization scheme improves the accuracy of the numerical solution by splitting any overly wide bins into several smaller bins while the model equations are being integrated. The figure to the right presents a comparison of aerosol spectra evolution as calculated in two model runs with adaptive size spectrum discretization toggled off (a) and on (b). The plots to the left depict how the shape of the cloud droplet spectra (blue histograms, bottom x-axis), the average size of the droplets and the standard deviation of their sizes (orange lines, bottom x-axis), and the total droplet concentration (turquoise lines, top x-axis) differ between the two model runs. Enabling adaptivity allows the model to better resolve the shape of the left-hand side slope of the droplet spectra, what in this example causes the final concentration to be about 15% higher than in the model run without adaptivity. The evolution of sizes of all particles (i.e. unactivated and activated) is summarised in the plots to the right by plotting the evolution of the position of bin boundaries (small circles, top x-axis) every 50 model time steps (of variable length). The colour scale corresponds to the difference between the drop and air temperatures. The evolution of the relative humidity (RH) is plotted with the green line (bottom x-axis).



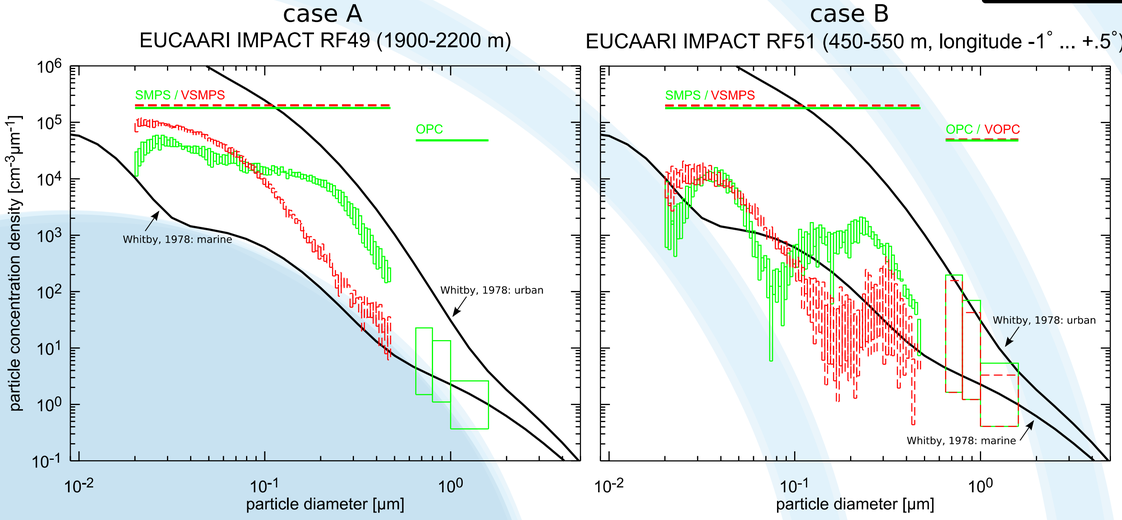

The figure above presents a summary of multiple model runs. The four upper and the four lower plots correspond to simulations with adaptivity toggled off (a) and on (b), respectively. Model was run with different initial aerosol size spectra (left and right columns, tri-modal lognormal distributions from Whitby (1978), see plots below), different aerosol composition and vertical velocity of the air parcel (key above ), and different initial number of bins (x-axis). The total concentration of cloud droplets, and the width of the cloud droplet size spectrum at the end of a model run is given on the y-axis. Toggling adaptivity on results in suppression of the sensitivity of the model results to the discretization parameter.

## INTRODUCTION

Cloud condensation nuclei (CCN) are the particles of atmospheric aerosol that take part in formation of cloud droplets. CCN activation is the first stage of the condensational growth of cloud droplets in which the hydrated aerosol particles reach sizes beyond their critical values ensuring their further growth. The number of activated particles depends on the size and composition of aerosol particles, as well as on evolution of the ambient water vapour supersaturation. In the present study an air-parcel CCN activation model is used to derive the number of activated particles as a function of maximal supersaturation. The model is initialised with measured aerosol size spectra. Predicted droplet concentrations are compared with measured CCN concentrations.

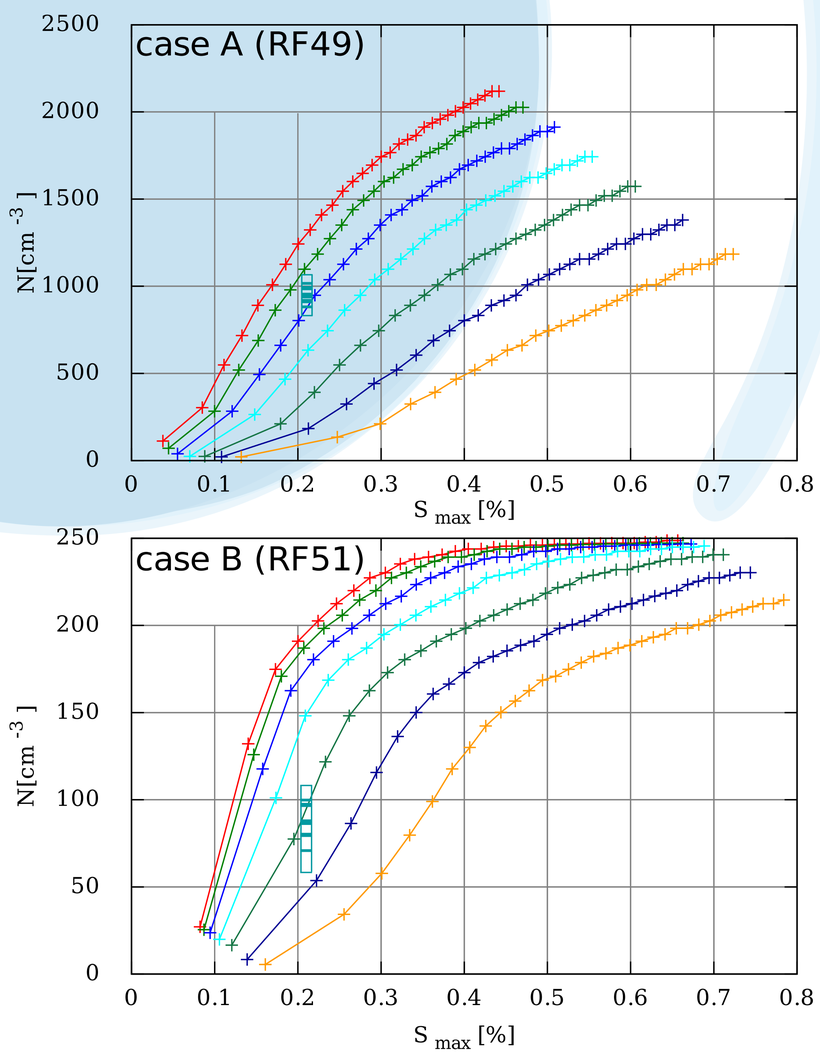



## OBSERVATIONAL DATA




Current analysis is based on the in-situ aerosol observations made on the SAFIRE ATR-42 aircraft during the EUCAARI IMPACT field campaign (see Crumeyrolle et al., 2010, for details). The aerosol size spectrum within the 10-250 nm radius range was measured using a scanning mobility particle sizer (SMPS), larger particles of radii up to 0.65 $\mu m$ were measured using an optical spectrometer (OPC). **Two sets of model input parameters are derived from the aircraft observations during research flights RF49 (May 13th, flight over Cabauw) and RF51 (May 15th, flight over the North Sea ).** Data from these two flights were chosen, for they represent distinct aerosol characteristics of relatively polluted and pristine air masses. The figures to the left present size spectra obtained with SMPS and OPC when the aircraft was flying below cloud base (vertical bars indicate minimum-maximum ranges of measured values). The red dashed lines correspond to measurements obtained with an additional pair of SMPS and OPC connected to the aerosol inlet through a heater set at 280°$C$. The black curves are the trimodal lognormal distributions from Whitby (1978) used in the simulations discussed above.

## MODEL VS. OBSERVATIONS




**Figures to the left present results of simulations run with the measured aerosol size spectra used as model input** (spectra averaged over ca. 200 s.). The initial layout of the histogram bins in the model corresponds to the layout of SMPS and OPC size classes. Aerosol is assumed to be initially in equilibrium with the measured ambient humidity, and the initial spectrum of dry aerosol is found by inverting the $\kappa$-Köhler curve. Simulations were run with different values of $\kappa$ and different vertical velocities leading to different maximal supersaturations. All simulations were run until reaching about 150 m above cloud base where the concentration of activated droplets does not change anymore. **The final concentrations are plotted as a function of the maximal supersaturation** reached during the air parcel ascent. Consequently, one CCN activation spectrum (i.e. cumulative number of CCN as a function of supersaturation) is plotted for each value of $\kappa$. **The ranges of CCN concentrations measured at the single supersaturation of 0.21% on the aircraft are indicated with turquoise bars** representing 7 percentiles of the frequency distribution of measured values ($1/8$, $2/8$, ... $7/8$ of data points, thickest line $\rightsquigarrow$ median). In the case of polluted air (flight RF49 over The Netherlands) the best agreement with measurements is found for $\kappa$ between 0.16 and 0.32 – typical values of $\kappa$ for continental Europe (Pringle et al., 2010). Much lower values of $\kappa$ are needed for agreement with measurements in the case of marine air (flight RF51 over The North Sea). However, marine aerosol is expected to be characterised by higher hygroscopicity and thus higher values of $\kappa$ than for the continental one. One reason for this discrepancy might be the invalidity of the implied assumption that the aerosol is internally mixed and may be described by single value of hygroscopicity parameter $\kappa$ in the whole particle size range.

## OUTLOOK

Further analyses are aimed at:

- relating the value(s) of $\kappa$ with the results of measurements of aerosol volatility and composition
- performing the comparison using data from other flights as well as ground-based observations at the Cabauw tower (where CCN concentrations were measured at several supersaturations)
- assessing the closure among model-predicted and measured parameters of cloud-droplet size spectrum
- synthesising parcel model results into LES-suited parametrisations of cloud droplet spectrum parameters such as droplet concentration, cloud droplet spectral width at cloud base, etc, expressed as a function of the supersaturation, the $\kappa$ parameter, and aerosol size spectrum parameters

**References**:
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