Representation of CCN activation in numerical models of aerosol-cloud interactions

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LES modelling in collaboration with:
Shin-ichiro Shima (JAMSTEC, Yokohama, Japan)

observational data courtesy of:
S. Crumeyrolle, L. Gomes, G. Roberts, A. Schwarzenboeck et al.
(LaMP, Clermont-Ferrand & CNRS/Météo-France, Toulouse, France)

December 10th 2010, NCAR, Boulder, Colorado, USA

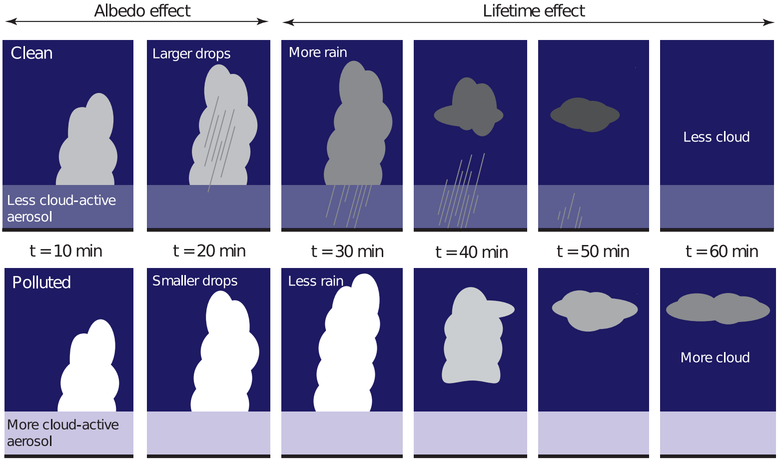
EUCAARI

European Integrated Project
on Aerosol Cloud Climate Air Quality Interactions

- EU's 6th Framework Programme
- 2007–2010, 10M€
- 48 partners from 25 countries, led by University of Helsinki

Plan of the talk

1. **Aerosol-cloud interactions & CCN activation**
2. **CCN activation in a moving-sectional air-parcel model**
   - Basic ideas & the "Drops" model
   - "Drops" model highlights: numerics
   - "Drops" model highlights: physics
   - "Drops" model highlights: implementation
   - (Preliminary) validation against airborne measurements
3. **CCN activation in an LES with the Super-Droplet method**



(figure from Stevens and Feingold, 2009)

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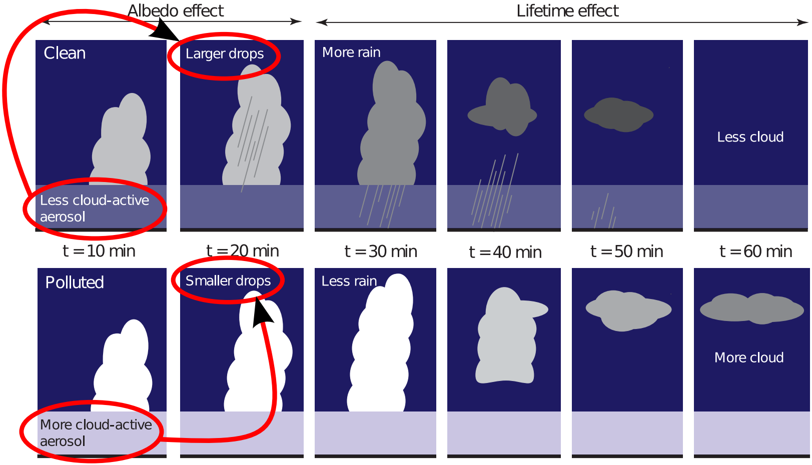

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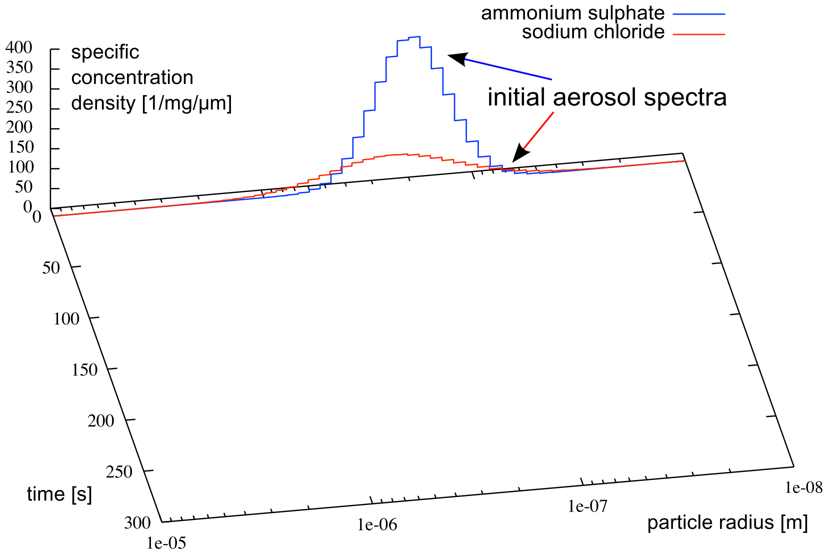
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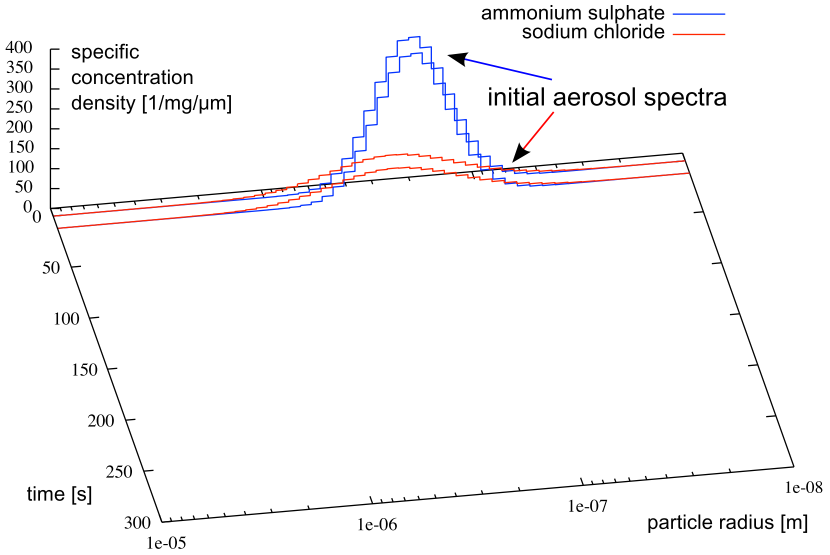
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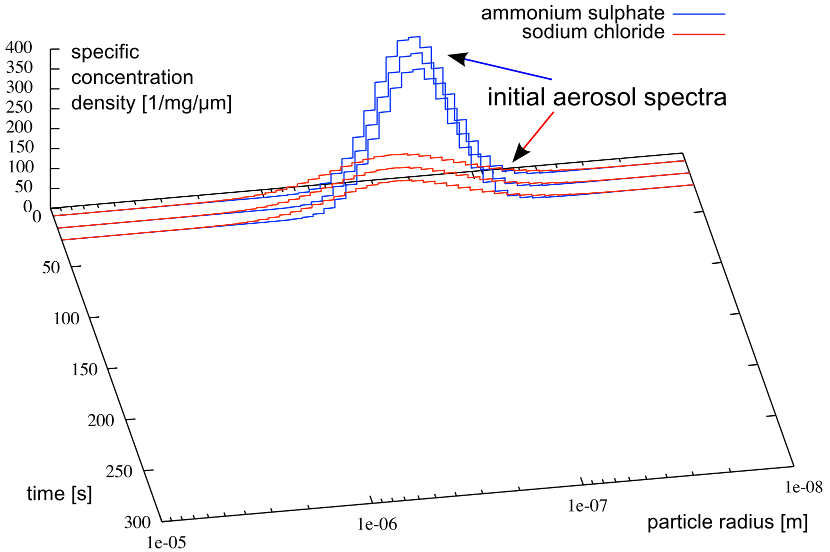
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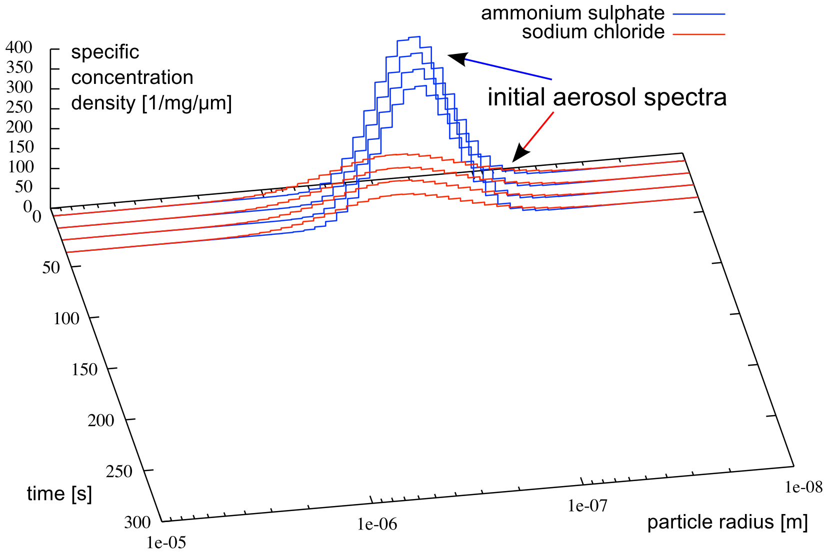
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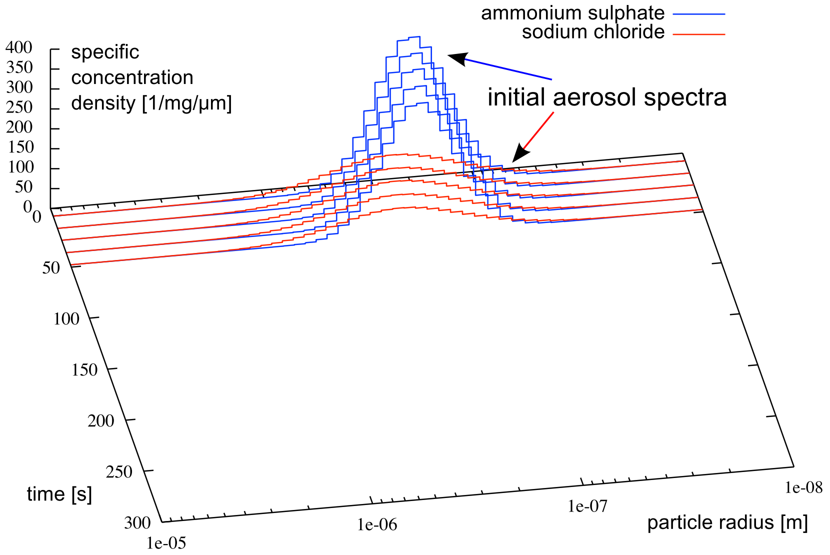
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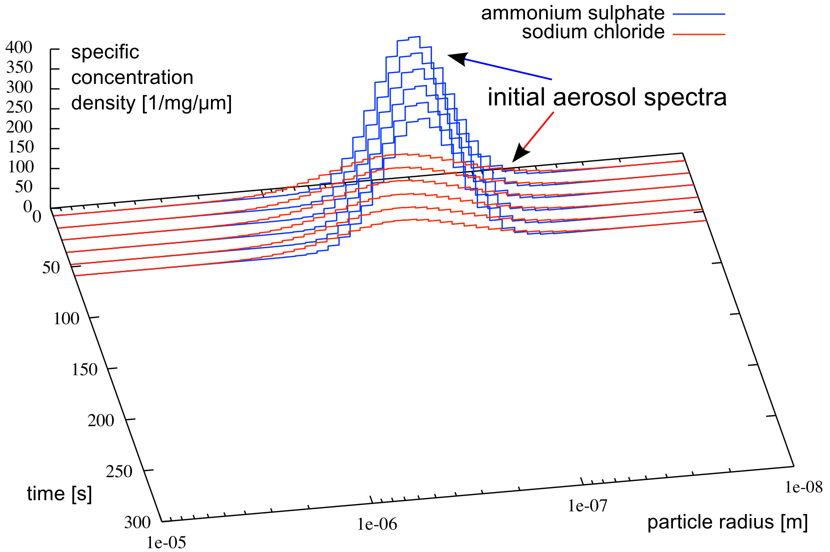
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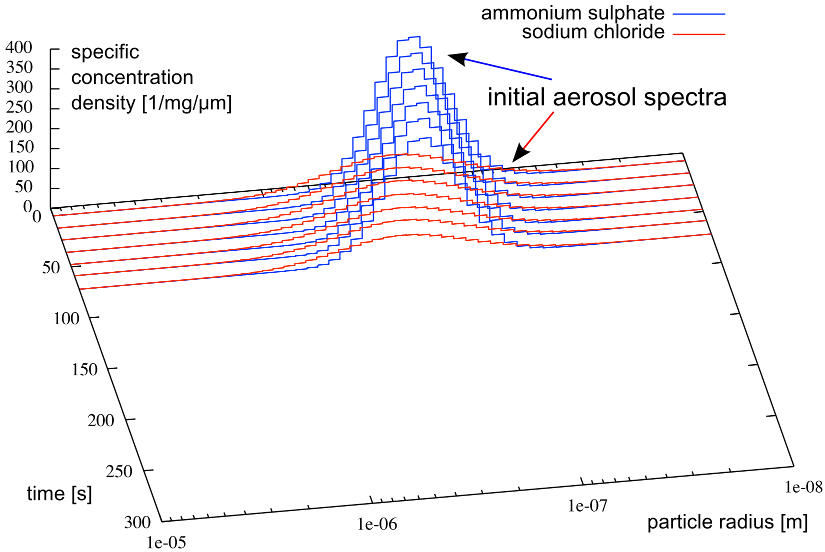
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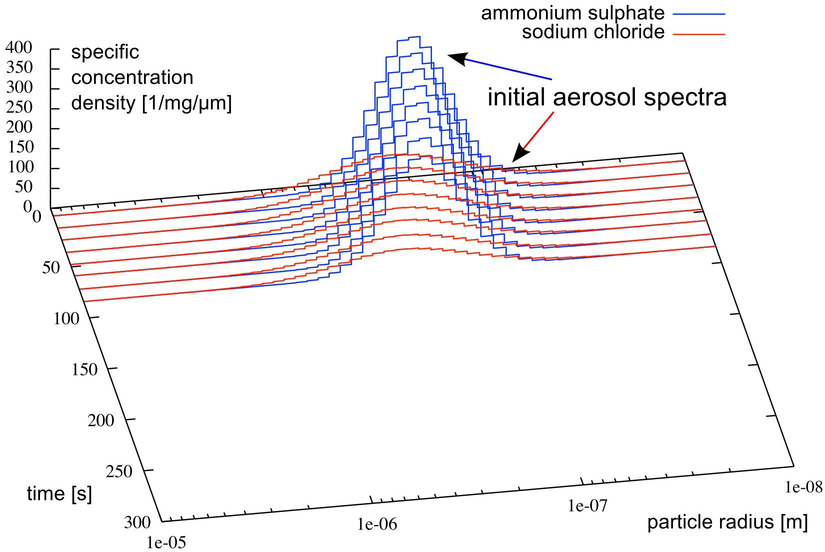
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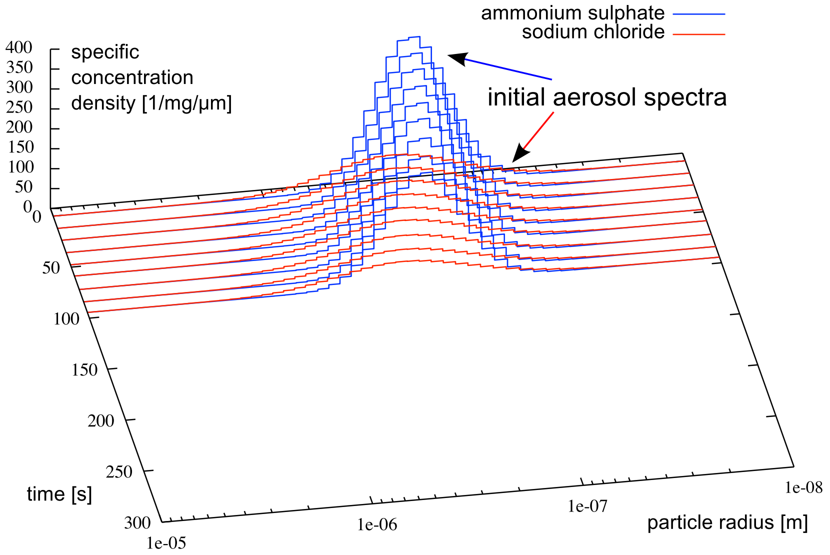
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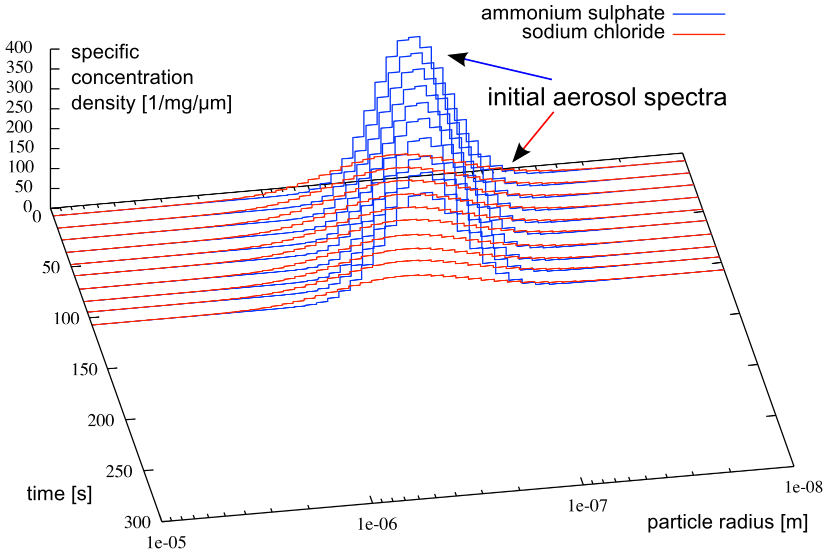
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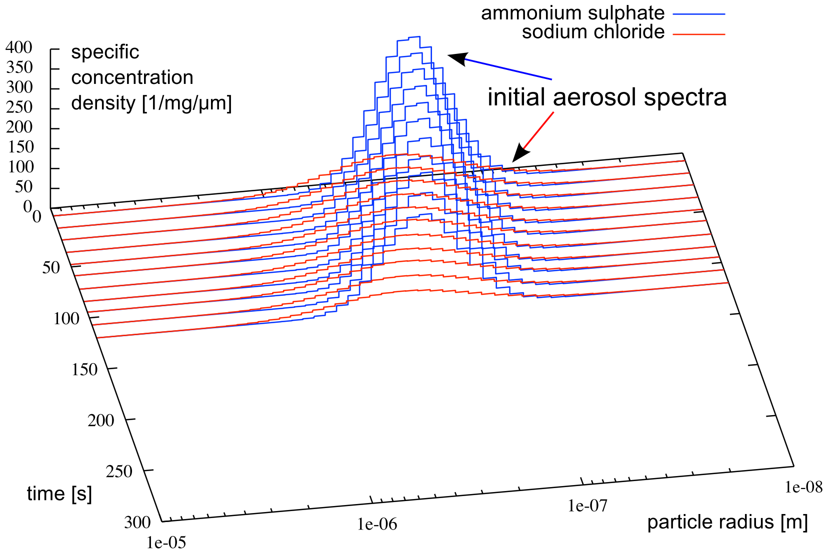
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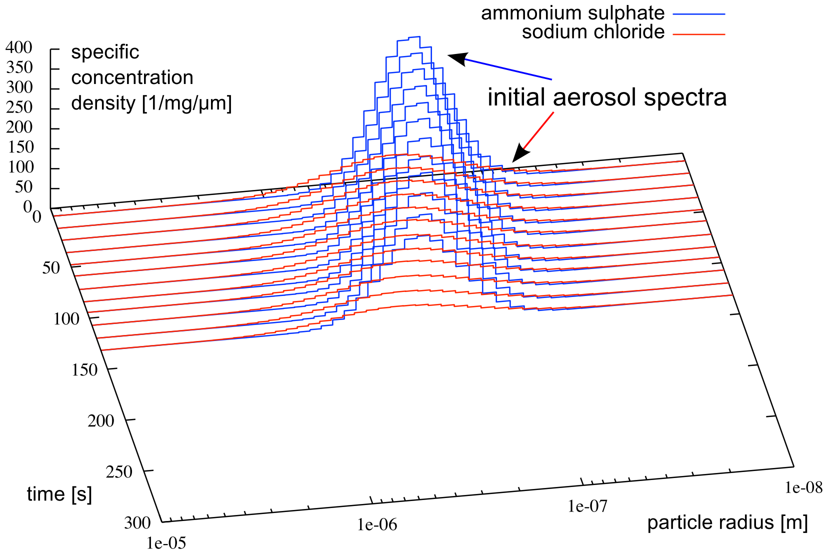
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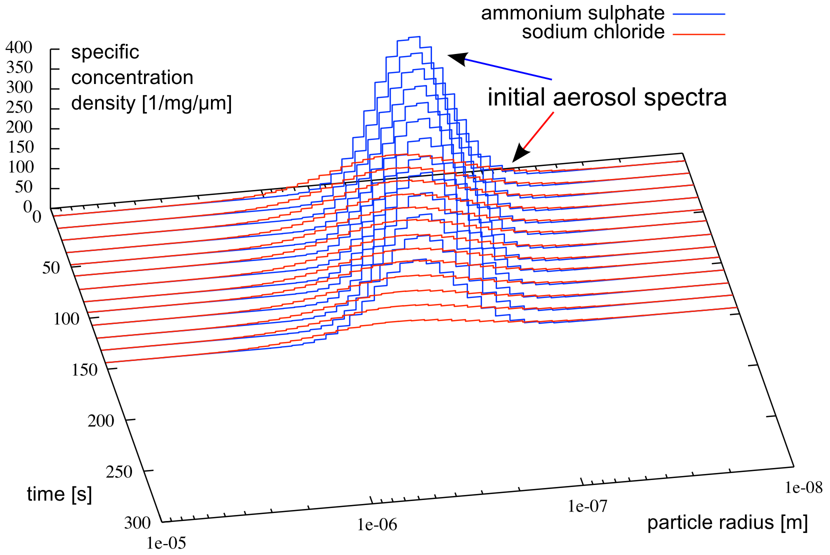
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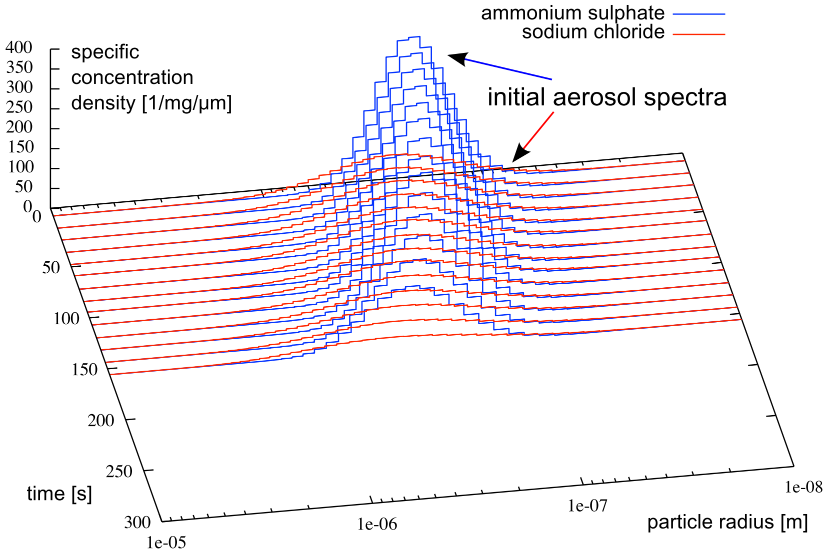
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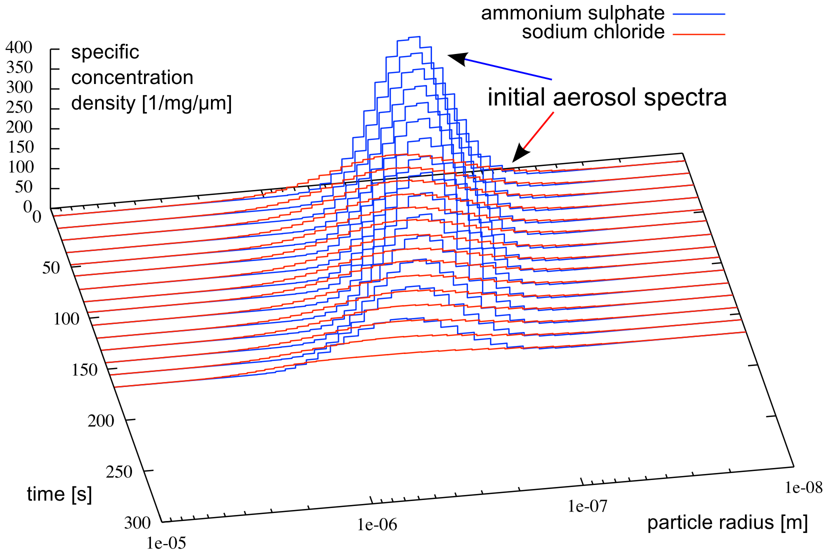
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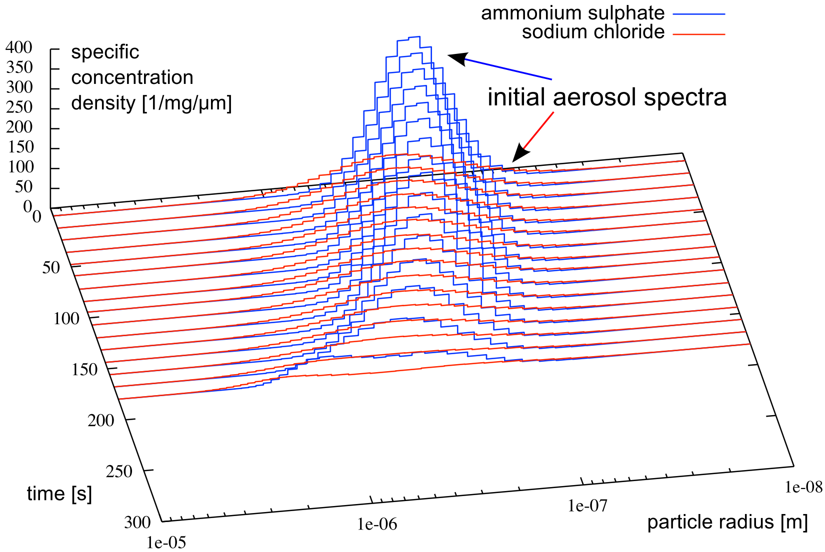
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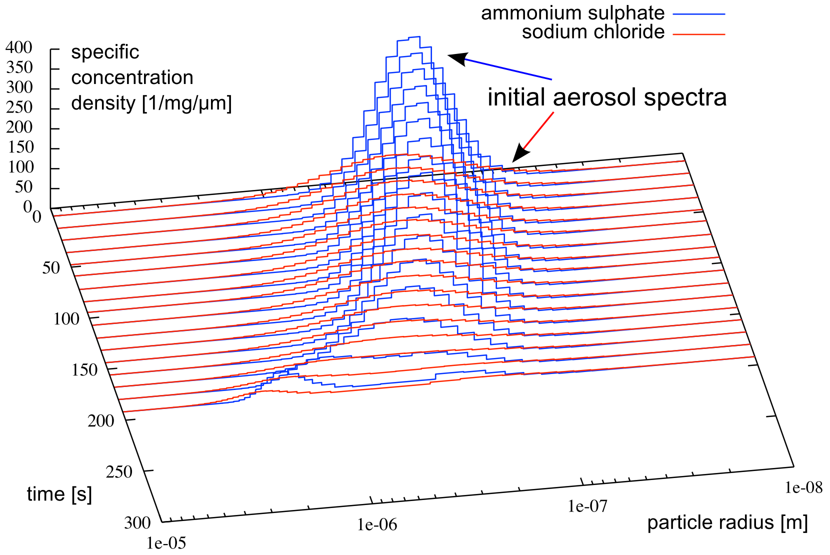
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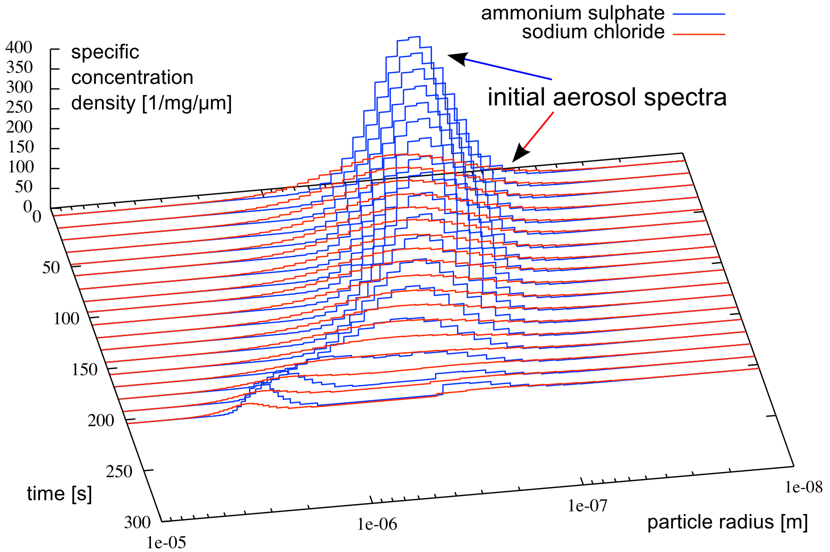
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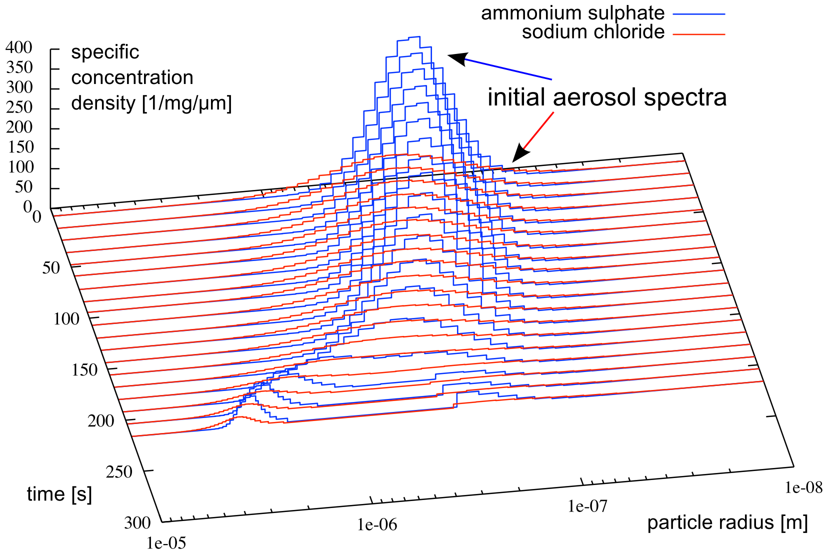
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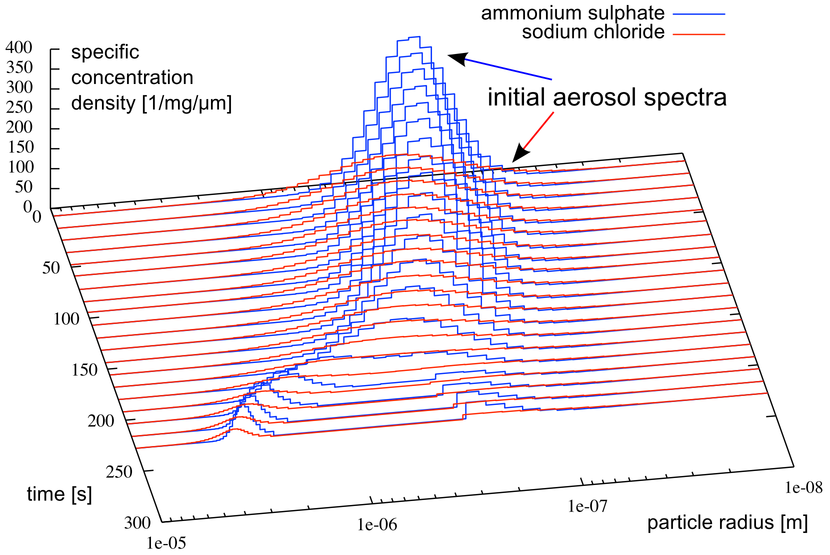

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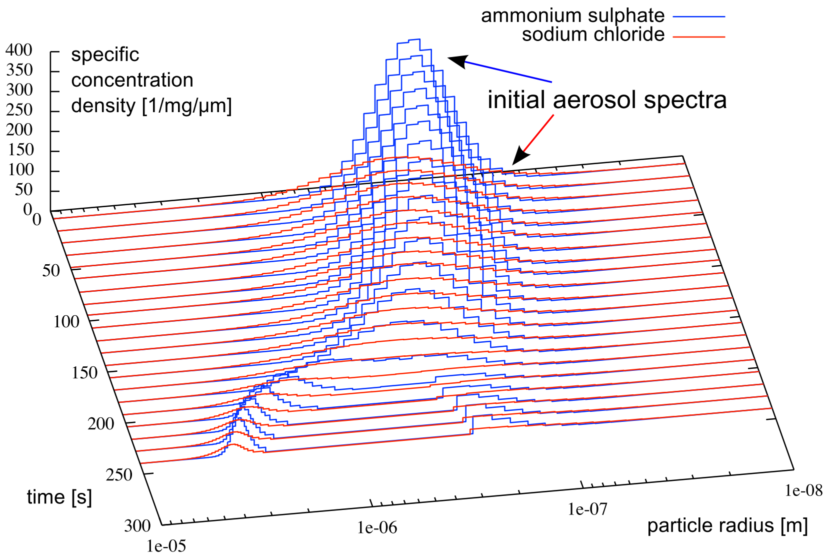
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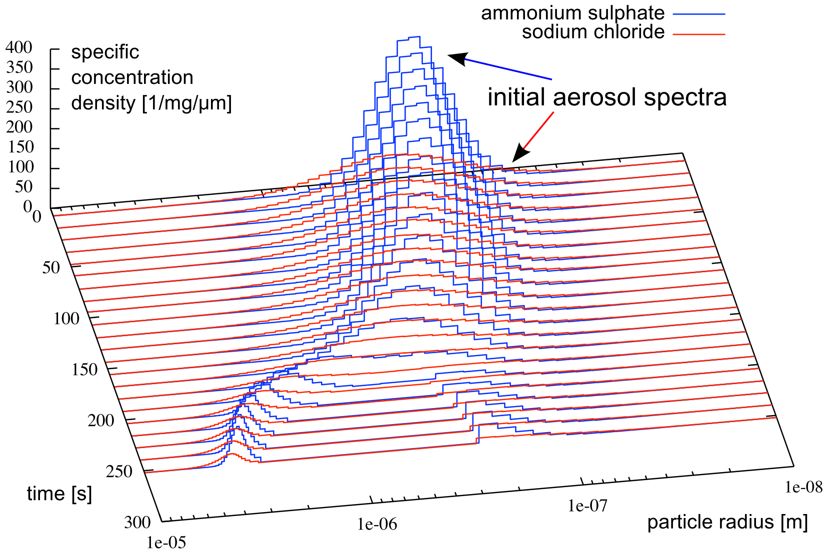
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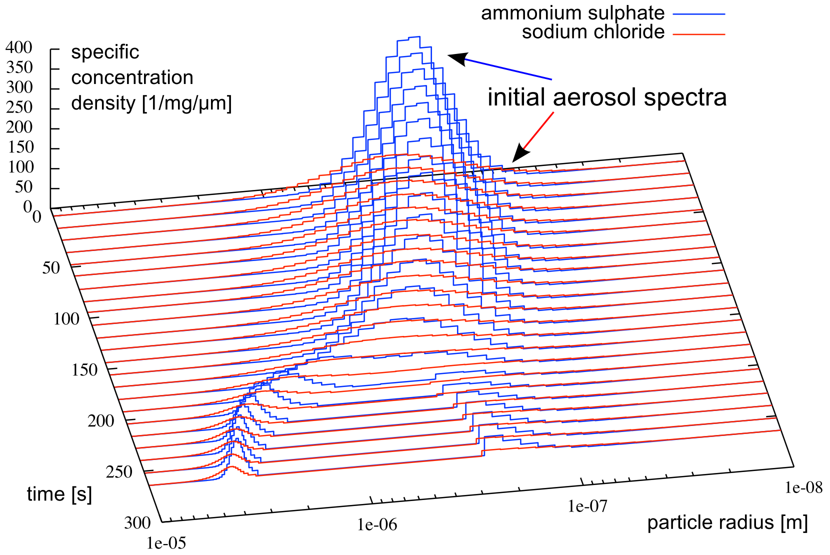
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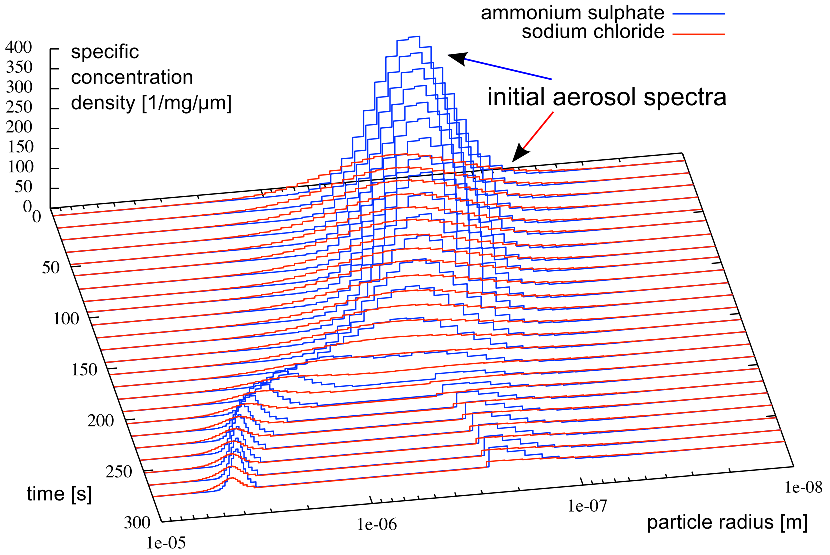
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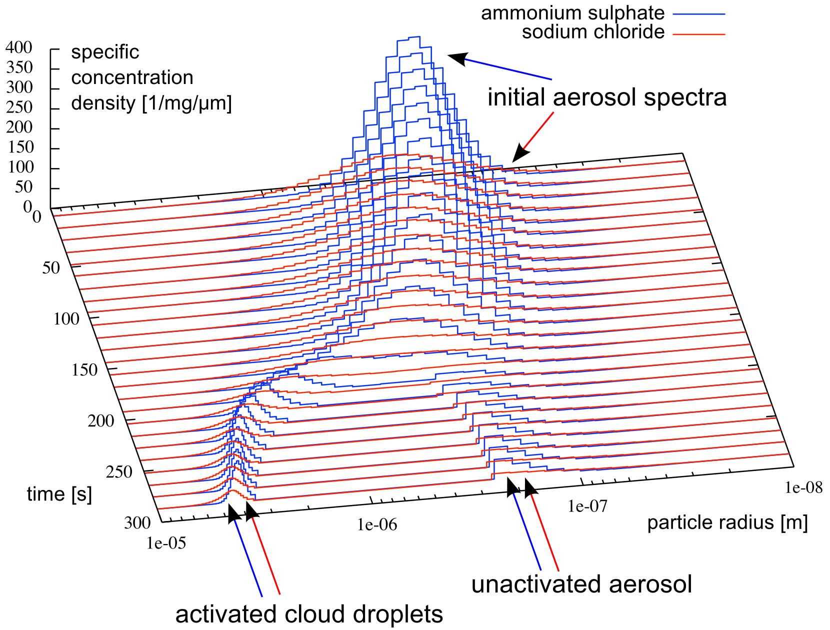
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unactivated aerosol
activated cloud droplets

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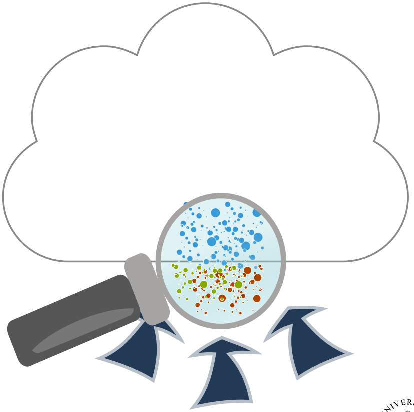
   - Basic ideas & the "Drops" model
   - "Drops" model highlights: numerics
   - "Drops" model highlights: physics
   - "Drops" model highlights: implementation
   - (Preliminary) validation against airborne measurements
3. CCN activation in an LES with the Super-Droplet method

- air-parcel cooled beyond saturation in adiabatic ascent, heated by latent heat release
- condensation/evaporation of water vapour on aerosol particles
- aerosol particles of different size and chemical composition competing for the available water vapour
- droplet surface curvature, temperature and chemical composition effects on the efficiency of diffusion of water vapour



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$$\frac{dT}{dt} = \frac{1}{C_p}\left(-gw - L\frac{dq}{dt}\right)$$

- integro-differential equation for dq/dt:

$$\frac{dq}{dt} \sim \frac{d}{dt}\int r^3 n \, dr$$

- 1-d (compressible) advection equation for conservation of $n(r,t)$:

$$\frac{dn}{dt} = -n\frac{\partial}{\partial r}\frac{dr}{dt}$$

- drop growth law (vapour/heat diffusion, Raoult and Kelvin effects, ...)

$$\frac{dr}{dt} \sim \frac{D}{r}\left(\rho|_{\infty} - \rho|_{\text{drop surface}}\right)$$

symbols:

T: temperature

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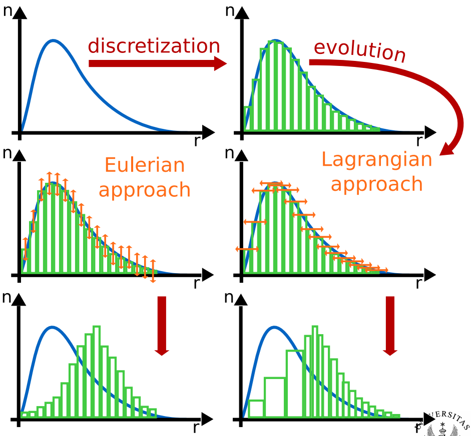
D: vapour diffusivity

ρ: vapour density

Numerical solution with MOL

- moving-sectional technique (Lagrangian, Method Of Lines):
  - approximation of *n* with piecewise constant function (bins, sections)
  - sections are "moving" (i.e. variable position and width)
  - PDE ⇝ system of (stiff) ODEs
  - no numerical diffusion
  - tracking chemical composition easy
- introduced by Howell (1949)
- challenge for the first computers:
  - Swedish BESK: (Mordy, 1959)
  - American SWAC: (Neiburger and Chien, 1960)



Howell, W.: The growth of cloud drops in uniformly cooled air, J. Meteor., 6, 134–149, 1949.
Mordy, W.: Computations of the growth by condensation of a population of cloud droplets, Tellus, 11, 1959.
Neiburger, M. and Chien, C.: Computations of the growth of cloud drops by condensation using an electronic digital computer, in: Physics of Precipitation, edited by Weickmann, H., pp. 191–210, AGU, 1960.

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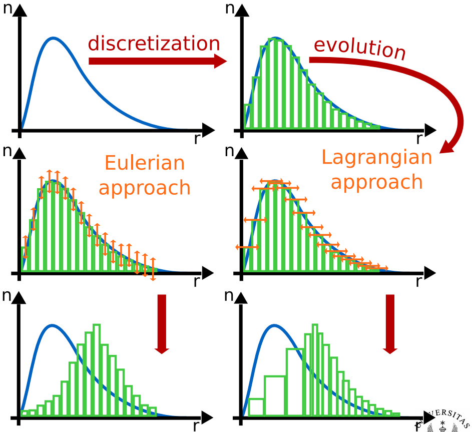

Howell, W.: The growth of cloud drops in uniformly cooled air, J. Meteor., 6, 134–149, 1949.
Mordy, W.: Computations of the growth by condensation of a population of cloud droplets, Tellus, 11, 1959.
Neiburger, M. and Chien, C.: Computations of the growth of cloud drops by condensation using an electronic digital computer, in: Physics of Precipitation, edited by Weickmann, H., pp. 191–210, AGU, 1960.

MOL's ODE system

$$
\frac{d}{dt}\begin{bmatrix} r_{wl}^{[c,b]} \\ T_{wl}^{[c,b]} \\ q_v \\ \\ p \\ T \end{bmatrix} = \begin{bmatrix} \frac{D(\lambda_D, r_w)}{\rho_l r_{wl}^{[c,b]}} \left[\frac{p\, q_v}{R(q_v)\, T} - \frac{p_{vs}(T_{wl}^{[c,b]}, r_{wl}^{[c,b]}, r_{dl}^{[c,b]})}{R_v T_{wl}^{[c,b]}} \right] \\ \frac{3}{c_l} \left[\frac{dr_{wl}^{[c,b]}}{dt} \frac{l_v(T_{wl}^{[c,b]})}{r_{wl}^{[c,b]}} + \frac{T - T_{wl}^{[c,b]}}{r_{wl}^{[c,b]\,2}} \frac{K(\lambda_K, r_w)}{\rho_l} \right] \\ (q_v - 1)\frac{\pi \rho_l}{3} \sum_c \sum_b N^{[c,b]} \cdot \\ \cdot \left[\gamma \left[r_{wr}^{[c,b]}, r_{wl}^{[c,b]} \right] \frac{dr_{wr}^{[c,b]}}{dt} + \gamma \left[r_{wl}^{[c,b]}, r_{wr}^{[c,b]} \right] \frac{dr_{wl}^{[c,b]}}{dt} \right] \\ -\frac{p\, g}{T R(q_v)} w \\ \frac{1}{c_p(q_v)} \left[\frac{T R(q_v)}{p} \frac{dp}{dt} - l_v(T) \frac{dq_v}{dt} \right] \end{bmatrix}
$$

Geosci. Model Dev. Discuss., 3, 1271–1315, 2010
www.geosci-model-dev-discuss.net/3/1271/2010/
doi:10.5194/gmdd-3-1271-2010

Geoscientific Model Development Discussions

Adaptive method of lines for multi-component aerosol condensational growth and cloud droplet activation

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GMDD

3, 1271–1315, 2010

Adaptive method of lines for CCN activation

S. Arabas and H. Pawlowska

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Abstract | Introduction
Conclusions | References
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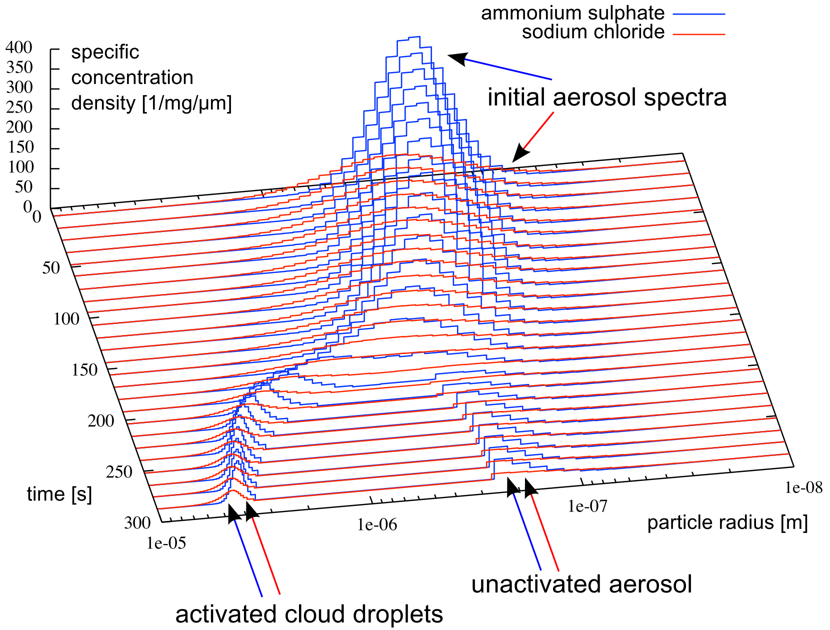
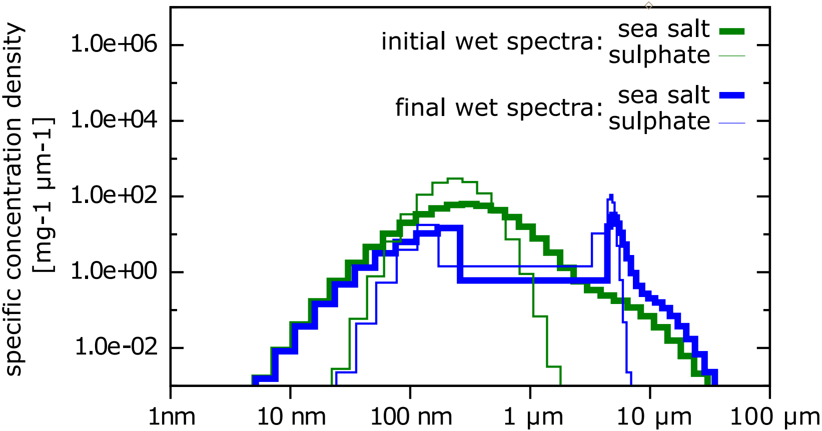
Printer-friendly Version
Interactive Discussion

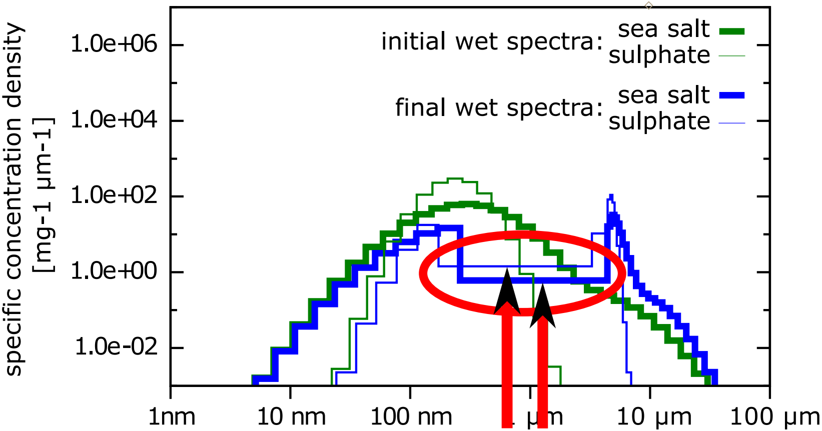
Plan of the talk

1. Aerosol-cloud interactions & CCN activation
2. **CCN activation in a moving-sectional air-parcel model**
   - Basic ideas & the "Drops" model
   - "Drops" model highlights: numerics
   - "Drops" model highlights: physics
   - "Drops" model highlights: implementation
   - (Preliminary) validation against airborne measurements
3. CCN activation in an LES with the Super-Droplet method


ammonium sulphate
sodium chloride
specific
concentration
density [1/mg/µm]
400
350
300
250
200
150
100
50
0
initial aerosol spectra
0
50
100
150
200
250
300
time [s]
1e-05
1e-06
1e-07
1e-08
particle radius [m]
unactivated aerosol
activated cloud droplets



bimodal: unactivated aerosol mode & activated cloud-droplet mode



single sections spanning nearly two "most important" decades

MOL for CCN activation: how accurate?

Takeda and Kuba (1982):

- "It is desirable that the number concentration of nuclei included in one class, specially near the smaller limit of activated nuclei, is very small in comparison with total number concentration of cloud droplets. Otherwise it would have a large influence on total number concentration of droplets ... "

Kreidenweis et al. (2003):

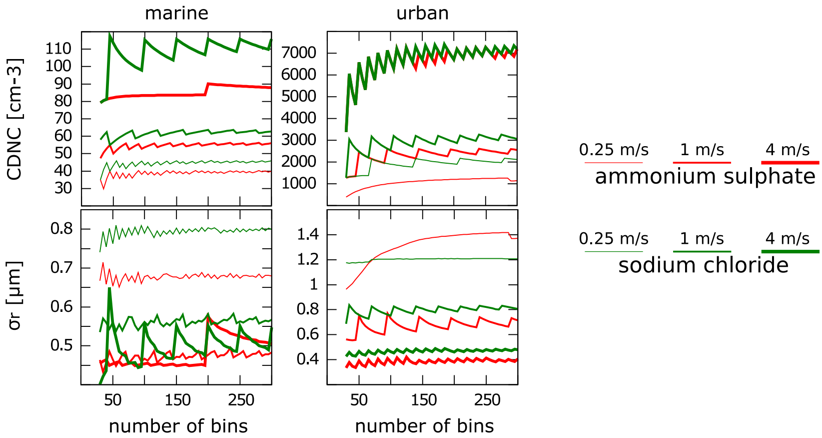
- "The grids ... play a role in determining the number concentration ..."
- "[if critical radius] ... falls just inside or just outside one of the bins, a difference of 30 or more droplets cm^3 can be computed..."

Korhonen et al. (2005):

- "...special attention must be paid to the critical sections(s), i.e. size section(s) into which a minimum activation diameter falls."
- "...doubling the particle size resolution improved the results significantly... "

Korhonen, H., Kerminen, V.-M., Lehtinen, K., and Kulmala, M.: CCN activation and cloud processing in sectional aerosol models with low size resolution, Atmos. Chem. Phys., 5, 2561–2570, 2005.
Kreidenweis, S., Walcek, C., Feingold, G., Gong, W., Jacobson, M., Kim, C.-H., Liu, X., Penner, J., Nenes, A., and Seinfeld, J.: Modification of aerosol mass and size distribution due to aqueous phase SO_2 oxidation in clouds: comparisons of several models, J. Geophys. Res., 108, 4213, 2003.
Takeda, T. and Kuba, N.: Numerical study of the effect of CCN on the size distribution of cloud droplets. Part I. Cloud droplets in the stage of condensationa growth, J. Meteorol. Soc. Jpn, 60, 978–993, 1982.

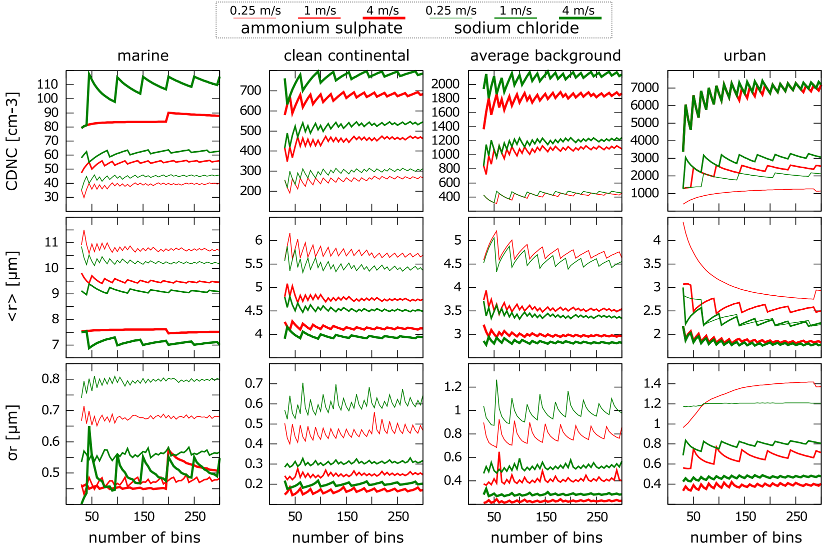
MOL for CCN activation: how accurate?



initial dry aerosol spectra from Whitby (1978)

Whitby, K.: The physical characteristics of sulfur aerosols, Atmos. Environ., 12, 135–159, 1978.

MOL for CCN activation: how accurate?



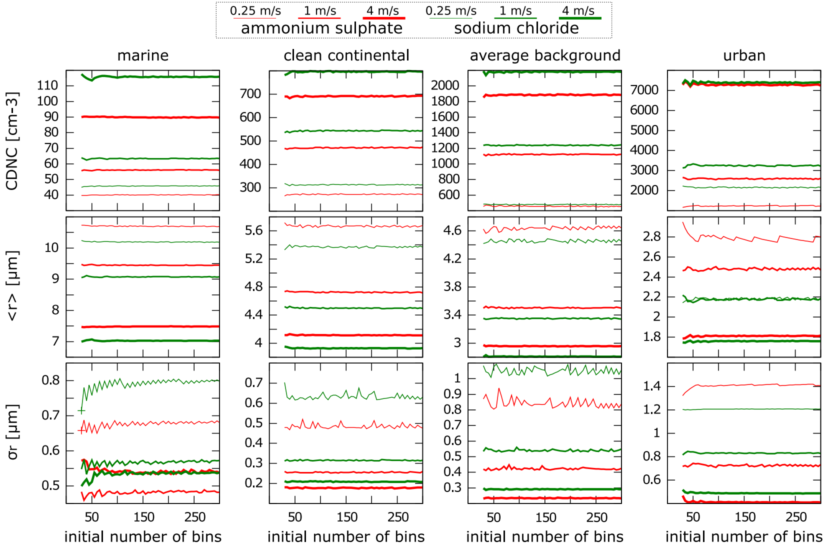
MOL for CCN activation: how to improve?

Arabas and Pawlowska (2010, GMDD):

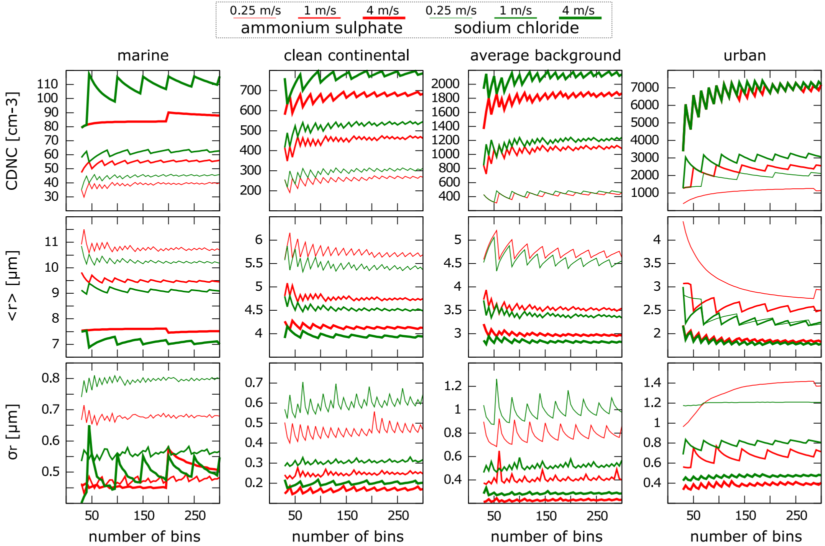
- employ the Adaptive MOL
- identify and split the critical sections into several smaller ones if needed (i.e. adaptive grid refinement)
- introduce an uncertainty-related parameter for controlling the precision of calculations with regard to spectrum discretization
- suppress the sensitivity to the bin-number choice

Arabas, S. and Pawlowska, H.: Adaptive method of lines for multi-component aerosol condensational growth and cloud droplet activation, Geosci. Model. Dev., (submitted), 2010.

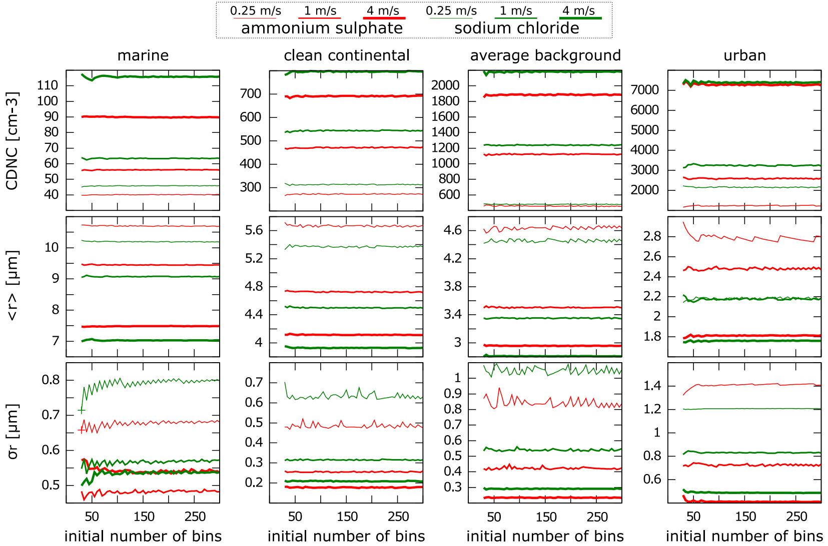
aMOL for CCN activation: it works!

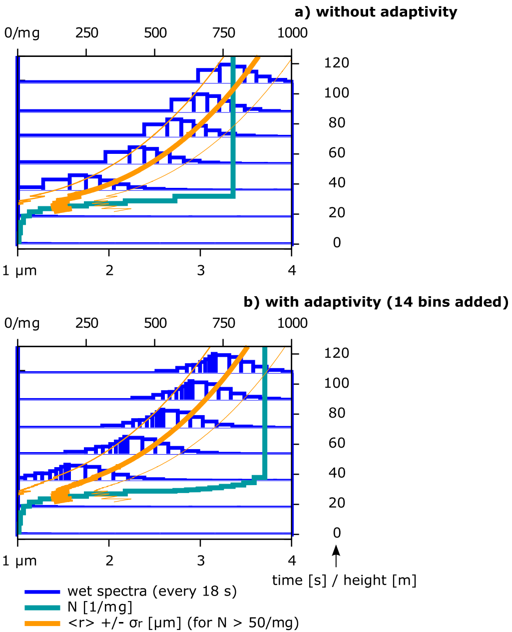


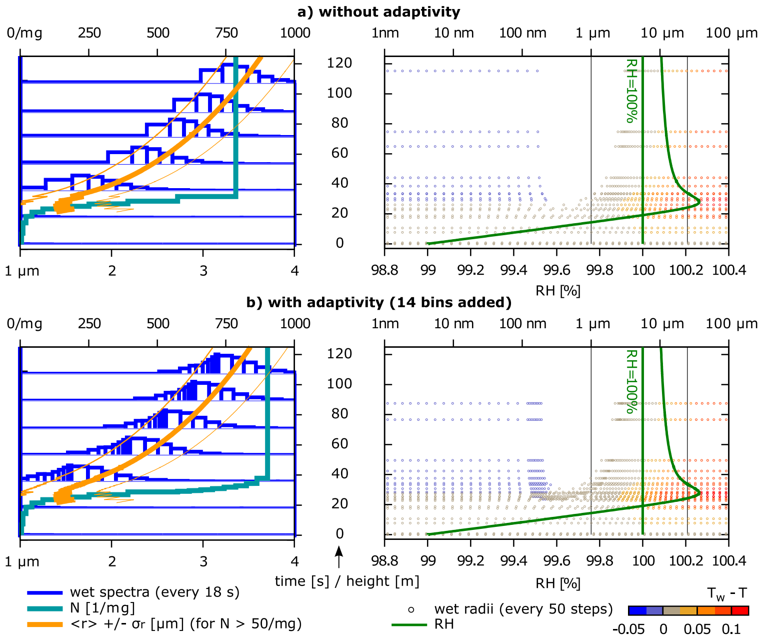
aMOL for CCN activation: adaptivity off



aMOL for CCN activation: adaptivity on




a) without adaptivity
0/mg
250
500
750
1000
120
100
80
60
40
20
0
1 μm
2
3
4
b) with adaptivity (14 bins added)
0/mg
250
500
750
1000
120
100
80
60
40
20
0
1 μm
2
3
4
time [s] / height [m]
wet spectra (every 18 s)
N [1/mg]
<r> +/- σr [μm] (for N > 50/mg)


a) without adaptivity
0/mg
250
500
750
1000
1nm
10 nm
100 nm
1 μm
10 μm
100 μm
120
100
80
60
40
20
0
RH=100%
1 μm
2
3
4
98.8
99
99.2
99.4
99.6
99.8
100
100.2
100.4
RH [%]
b) with adaptivity (14 bins added)
time [s] / height [m]
wet spectra (every 18 s)
N [1/mg]
<r> +/- σr [μm] (for N > 50/mg)
wet radii (every 50 steps)
RH
Tw - T
-0.05
0
0.05
0.1

Plan of the talk

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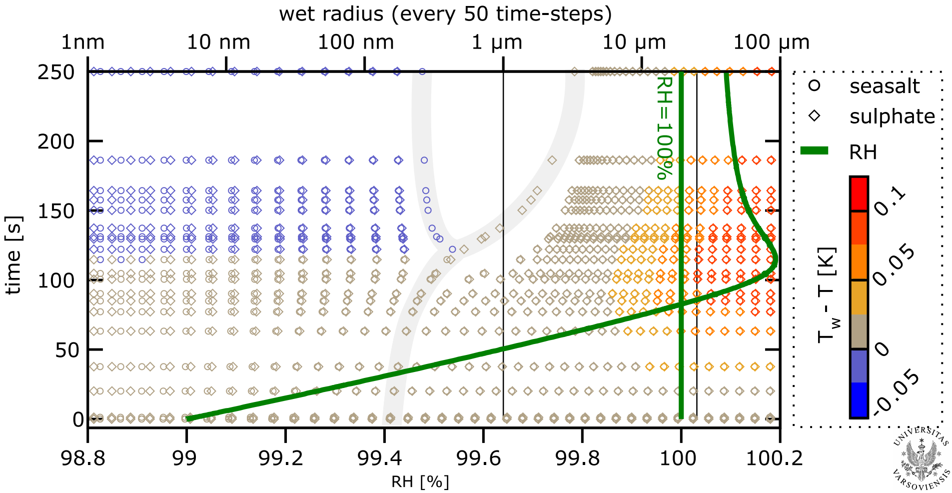
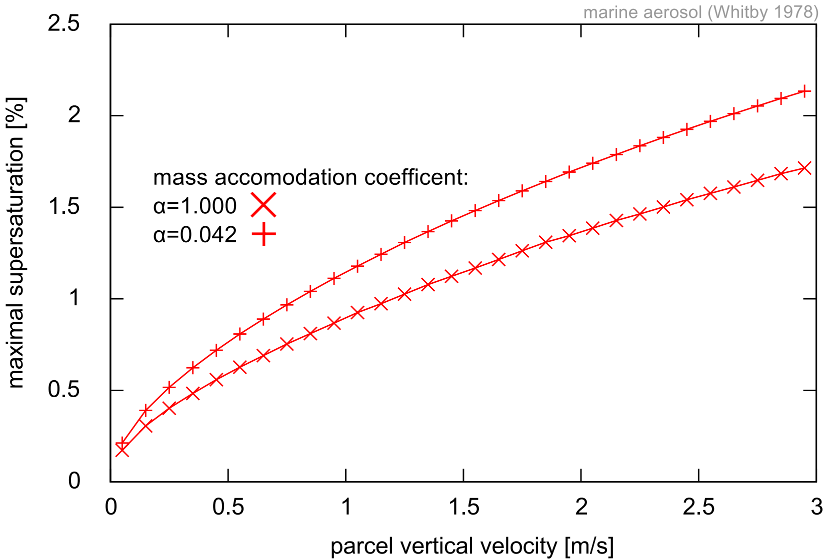
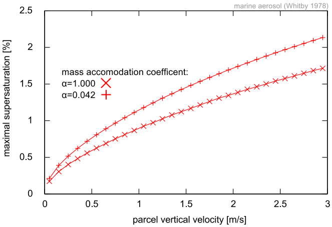
3. CCN activation in an LES with the Super-Droplet method

MOL's ODE system

$$
\frac{d}{dt}\begin{bmatrix} r_{wl}^{[c,b]} \\ T_{wl}^{[c,b]} \\ q_v \\ p \\ T \end{bmatrix} = \begin{bmatrix} \frac{D(\lambda_D, r_w)}{\rho_l r_{wl}^{[c,b]}} \left[\frac{p\, q_v}{R(q_v)\, T} - \frac{p_{vs}(T_{wl}^{[c,b]}, r_{wl}^{[c,b]}, r_{dl}^{[c,b]})}{R_v T_{wl}^{[c,b]}} \right] \\ \frac{3}{c_l} \left[\frac{dr_{wl}^{[c,b]}}{dt} \frac{l_v(T_{wl}^{[c,b]})}{r_{wl}^{[c,b]}} + \frac{T - T_{wl}^{[c,b]}}{r_{wl}^{[c,b]\,2}} \frac{K(\lambda_K, r_w)}{\rho_l} \right] \\ (q_v - 1) \frac{\pi \rho_l}{3} \sum_c \sum_b N^{[c,b]} \cdot \left[\gamma \left[r_{wr}^{[c,b]}, r_{wl}^{[c,b]} \right] \frac{dr_{wr}^{[c,b]}}{dt} + \gamma \left[r_{wl}^{[c,b]}, r_{wr}^{[c,b]} \right] \frac{dr_{wl}^{[c,b]}}{dt} \right] \\ -\frac{p\, g}{T R(q_v)} w \\ \frac{1}{c_p(q_v)} \left[\frac{T R(q_v)}{p} \frac{dp}{dt} - l_v(T) \frac{dq_v}{dt} \right] \end{bmatrix}
$$


wet radius (every 50 time-steps)
1nm
10 nm
100 nm
1 μm
10 μm
100 μm
250
200
150
100
50
0
time [s]
98.8
99
99.2
99.4
99.6
99.8
100
100.2
RH [%]
RH=100%
seasalt
sulphate
RH
T_w - T [K]
0.1
0.05
0
-0.05


marine aerosol (Whitby 1978)
mass accomodation coefficent:
α=1.000
α=0.042
maximal supersaturation [%]
parcel vertical velocity [m/s]
0
0.5
1
1.5
2
2.5
3



Laaksonen et al. (2005):

- "... the use of mass accommodation coefficient **values lower than 1** in cloud models together with the rigorous droplet growth theory is inconsistent and **should be avoided**."

Fountoukis et al. (2007):

- "**Optimal [measurement-model] closure is obtained if** the water vapor uptake coefficient is **equal to 0.06**, but can range between 0.03 and 1.0."

Fountoukis, C., Nenes, A., Meskhidze, N., Bahreini, R., Conant, W., Jonsson, H., Murphy, S., Sorooshian, A., Varutbangkul, V., Brechtel, F., Flagan, R., and Seinfeld, J.: Aerosol–cloud drop concentration closure for clouds sampled during the International Consortium for Atmospheric Research on Transport and Transformation 2004 campaign, J. Geophys. Res., 112, D10S30, 2007.

Laaksonen, A., Vesala, T., Kulmala, M., Winkler, P., and Wagner, P.: Commentary on cloud modelling and the mass accommodation coefficient of water, Atmos. Chem. Phys., 5, 461–464, 2005.

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3. CCN activation in an LES with the Super-Droplet method

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EUCAARI

User- & developer-friendly

- implemented in C++ using Boost.units
  (zero-overhead dimensional analysis at compile-time)
- ODE integration: SUNDIALS/CVODE (Hindmarsh et al., 2005)
- helper numerics (Köhler curve inversion, . . .): GNU Scientific Library (GSL)
- source code and a manual in an electronic supplement of the paper,
  public CVS at: http://gna.org/projects/drops/
- ... and a user-friendly web-based interface (CLI as well)

Hindmarsh, A., Brown, P., Grant, K., Lee, S., Serban, R., Shumaker, D., and Woodward, C.: SUNDIALS: Suite of Nonlinear and Differential/Algebraic Equation Solvers, ACM Transactions on Mathematical Software, 31, 363–396, 2005.

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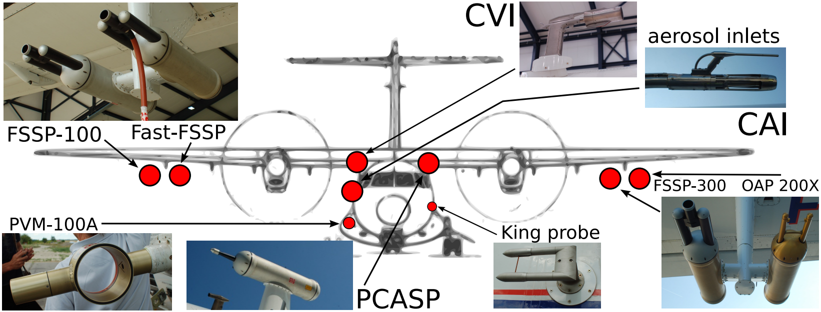
EUCAARI

EUCAARI Intensite Measurement Period At Cabauw Tower (IMPACT)

- 30 days, May 2008, The Netherlands
- ground-based remote-sensing and 200m-tower in-situ observations
- two research aircraft, one helicopter platform, 50 research flights
- >20 institutes from around Europe incl.:

  Royal Netherlands Meteorological Institute; Météo France; Institute for Tropospheric Research in Leipzig; Universities of Berlin, Bonn, Clermont-Ferrand, Delft, Heidelberg, Helsinki, Köln, Manchester, Utrecht, Warsaw, Wageningen; Research Centre Jülich; Leosphere; Institute of Atmospheric Sciences and Climate in Bologna; TNO; Energy Research Center of the Netherlands; Dutch National Institute for Public Health and Environment
- more info: http://www.knmi.nl/eucaari/

SAFIRE[1] ATR-42: cloud/aerosol probes

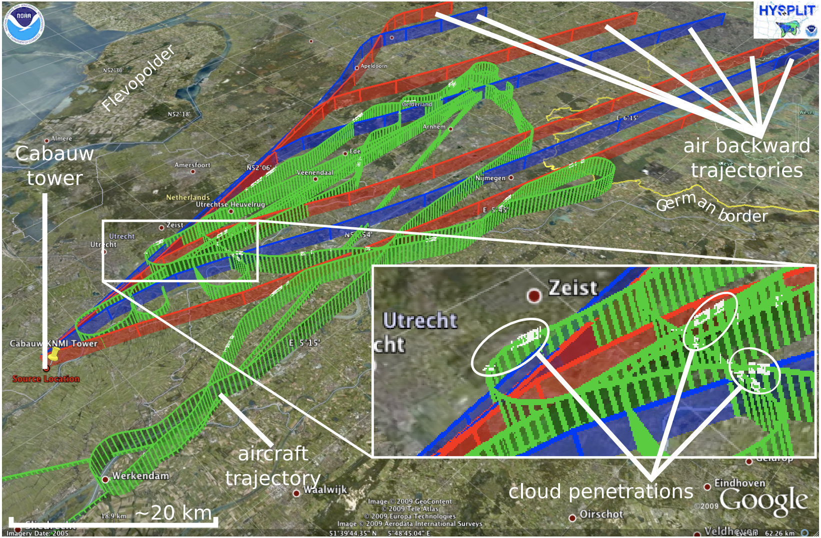


[1] Service des Avions Français Instrumentés pour la Recherche en Environnement (www.safire.fr)

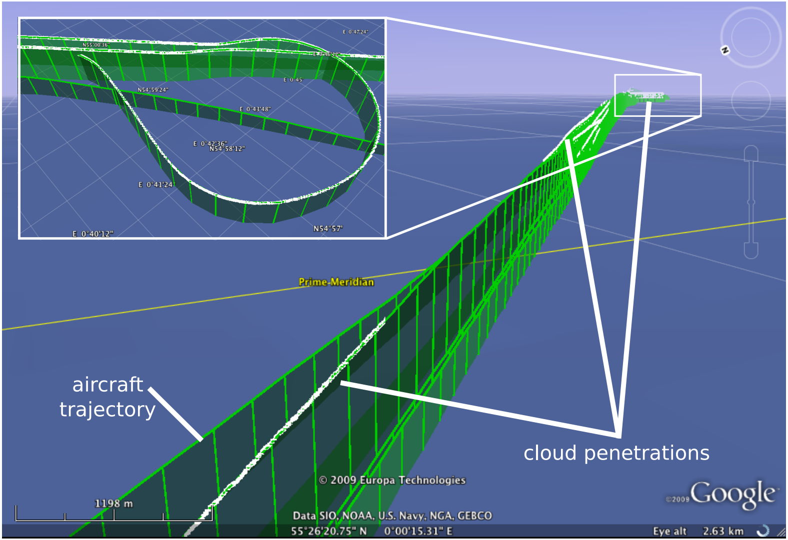
SAFIRE ATR-42: aerosol instruments (int.)

aerosol mass spectrometer, CCN counter, two pairs of optical and scanning mobility aerosol size spectrometers (one pair connected through a heater set at 280° C), CPCs, nephelometer, . . .

RF49 (May 13th) Cu above NL



RF51 (May 15^{th}) North Sea Sc



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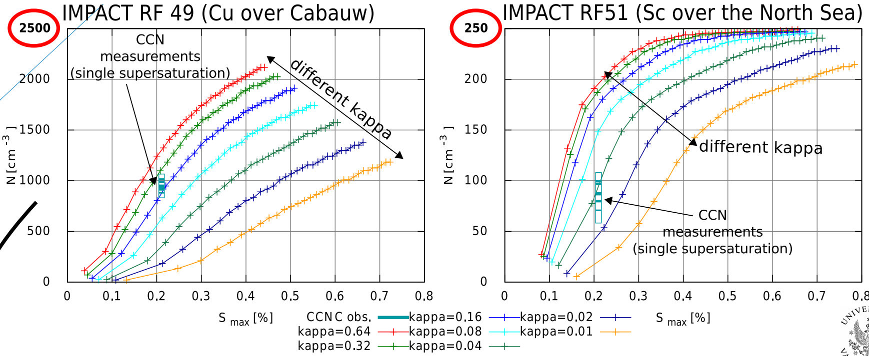
EUCAARI

- model input: SMPS+OPC aerosol spectra & p,T,RH
- model output vs. CCNC measurements (single supersaturation)
- aerosol chemical composition using the κ-Köhler parameterisation (Petters and Kreidenweis, 2007)



CCN activation parametrization for LES

Petters, M. and Kreidenweis, S.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, 2007.

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EUCAARI

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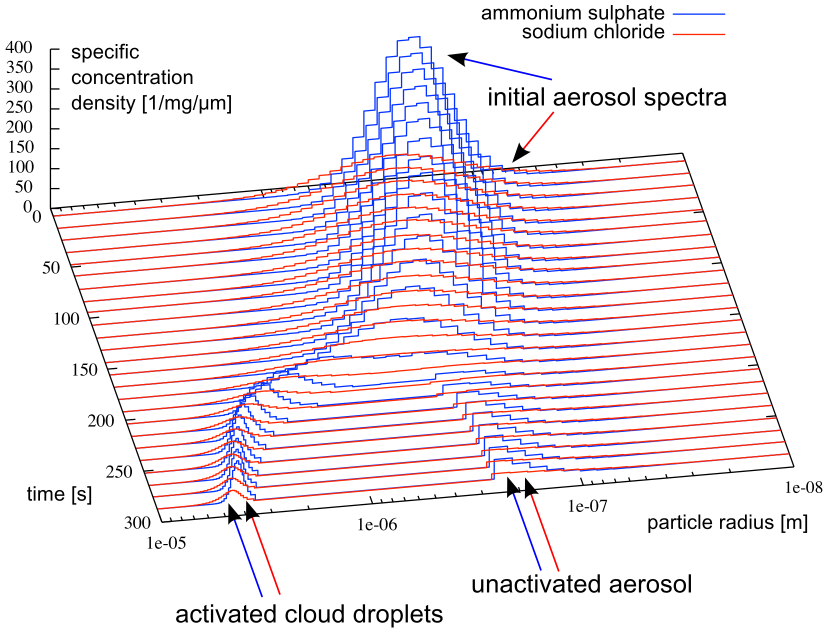

ammonium sulphate
sodium chloride
specific
concentration
density [1/mg/μm]
400
350
300
250
200
150
100
50
0
initial aerosol spectra
0
50
100
150
200
250
300
time [s]
1e-05
1e-06
1e-07
1e-08
particle radius [m]
unactivated aerosol
activated cloud droplets

coupling aerosol spectrum evolution with:

- parcel model:
  ⇝ grid: dry radii
  ⇝ constants (per bin): concentration
  ⇝ variables (per bin): wet particle radii (+ temperatures)
- LES:
  ⇝ grid: dry radii (+ LES grid)
  ⇝ constants (per bin): concentration (+ sources + coalescence)
  ⇝ variables (per bin): wet radii & position coordinates (x,y,z),

"Super Droplet" microphysics for LES (Shima et al., 2009)

Particle-based approach **resolving explicitly CCN activation**, condensational and collisional growth (Monte-Carlo scheme) & gravitational sedimentation including drizzle and rain precipitation

Shima, S., Kusano, K., Kawano, A., Sugiyama, T., and Kawahara, S.: The super-droplet method for the numerical simulation of clouds and precipitation: A particle-based and probabilistic microphysics model coupled with a non-hydrostatic model, Quart. J. Roy. Meteor. Soc., 135, 1307–1320, 2009.

coupling aerosol spectrum evolution with:

- parcel model:
  - ⇝ grid: dry radii
  - ⇝ constants (per bin): concentration
  - ⇝ variables (per bin): wet particle radii (+ temperatures)
- LES:
  - ⇝ grid: dry radii (+ LES grid)
  - ⇝ constants (per bin): concentration (+ sources + coalescence)
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  - ⇝ grid: dry radii (+ LES grid)
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RICO cumuli: CReSS-SDM vs. Fast-FSSP

- simulations:
  - dynamics: Cloud Resolving Storm Simulator (Tsuboki and Sakakibara, 2006)
  - microphysics: Super Droplet Method
  - set-up: GCSS RICO case (vanZanten et al., 2010)

Tsuboki, K. and Sakakibara, A.: Large-Scale Parallel Computing of Cloud Resolving Storm Simulator, Lecture Notes in Computer Science, 2327/2006, 463–471, 2006.

vanZanten, M., Stevens, B., Nuijens, L., Siebesma, A., Ackerman, A., Burnet, F., Cheng, A., Couvreux, F., Jiang, H., Khairoutdinov, M., Lewellen, D., Mechem, D., Noda, A., Shipway, B., Slawinska, J., Wang, S., and Wyszogrodzki, A.: Controls on precipitation and cloudiness in simulations of trade-wind cumulus as observed during RICO, J. Adv. Model. Earth Sys., (submitted), 2010.

RICO cumuli: CReSS-SDM vs. Fast-FSSP

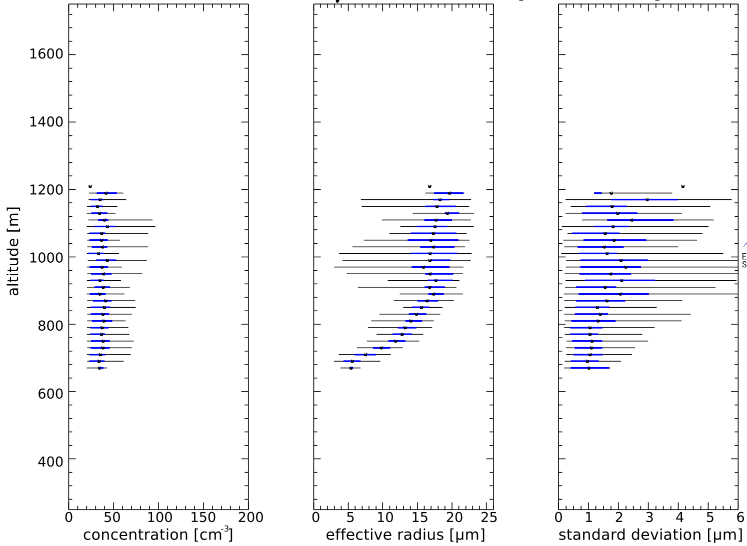
- measurements:
    - 8h-long C-130 flights from Antigua (5-10% penetrating clouds)
    - Fast-FSSP cloud droplet spectrometer (Brenguier et al., 1998)
    - height-resolved statistics as in Arabas et al. (2009)

NCAR

Arabas, S., Pawlowska, H., and Grabowski, W.: Effective radius and droplet spectral width from in-situ aircraft observations in trade-wind cumuli during RICO, Geophys. Res. Lett., 36, L11 803, 2009.

Brenguier, J.-L., Bourrianne, T., Coelho, A., Isbert, J., Peytavi, R., Trevarin, D., and Weschler, P.: Improvements of Droplet Size Distribution Measurements with the Fast-FSSP (Forward Scattering Spectrometer Probe), J. Atmos. Oceanic Technol., 15, 1077–1090, 1998.

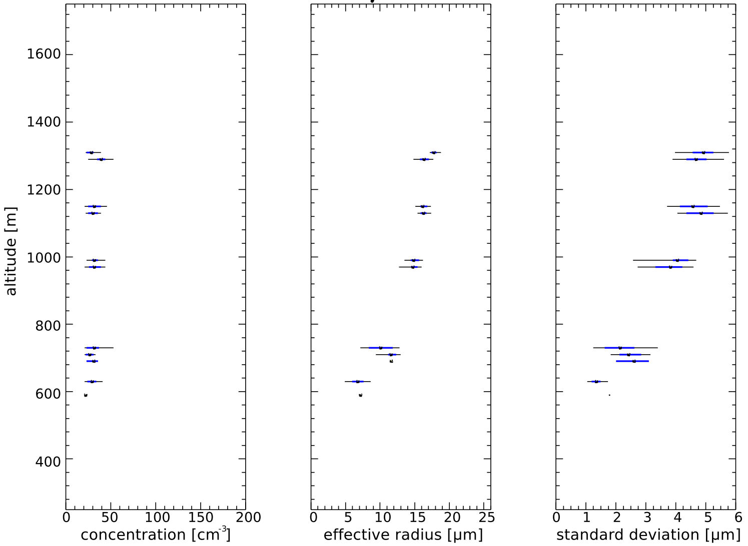
RICO cumuli / LES+Super Droplet[2]



EARTH SIMULATOR

[2] calculations done on The Earth Simulator at JAMSTEC

RICO cumuli / Fast-FSSP data[3]



[3] observational data: CNRS/Météo-France; NCAR

Thank you for your attention!

Parcel model details and code:

http://geosci-model-dev-discuss.net/3/1271/2010/

http://gna.org/projects/drops/

Acknowledgements:

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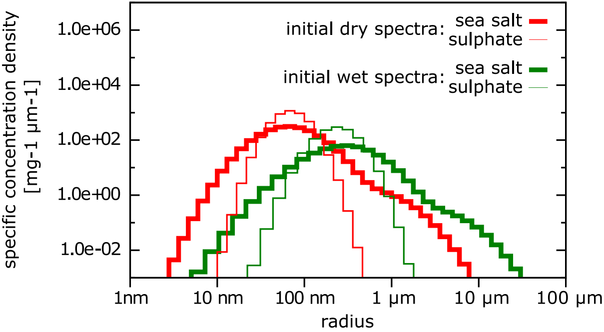
initial condition (set-up akin to the one of Ghan et al., 1998)



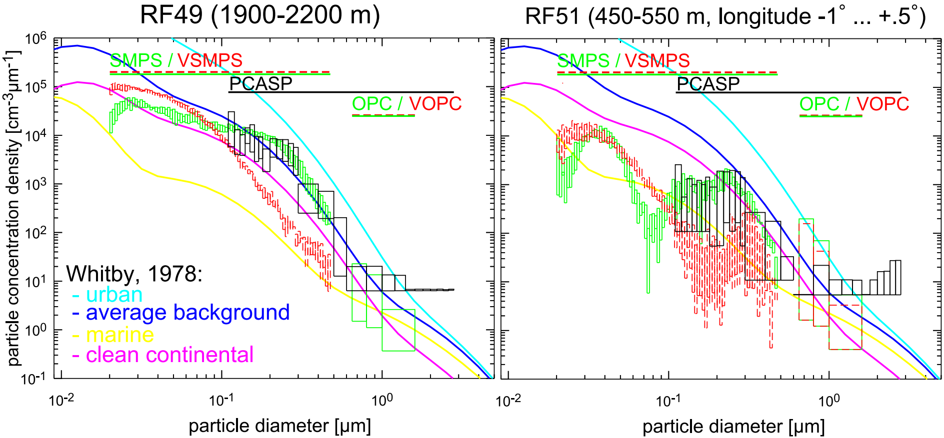
mode	solute	N_m [cm^{-3}]	r_m [µm]	σ_m [1]
film drop	NaCl	51.1	0.10	1.90
jet drop	NaCl	2.21	1.00	2.00
spume drop	NaCl	0.00001	6.00	3.00
accumulation	$(NH_4)_2SO_4$	100	0.08	1.45

- 280K, 1000 hPa, RH=99%
- solution droplets at equilibrium (wet spectra)
- sulphate: single-mode lognormal, $(NH_4)_2SO_4$
- sea-salt: tri-modal lognormal, NaCl
- 45 bins spaced linearly in logarithm of radius

Ghan, S., Guzman, G., and Abdul-Razzak, H.: Competition between sea salt and sulfate particles as cloud condensation nuclei, J. Atmos. Sci., 55, 3340–3347, 1998.

Case study: marine Sc vs. continental Cu

aerosol measurements below cloud base[4]

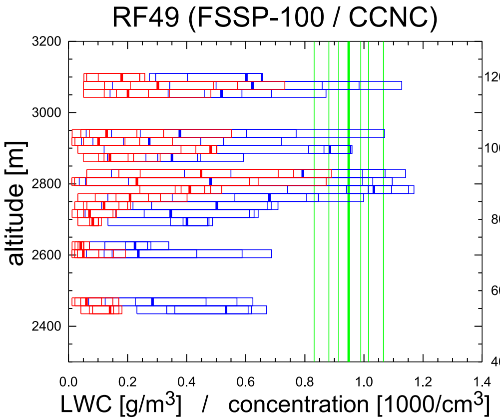


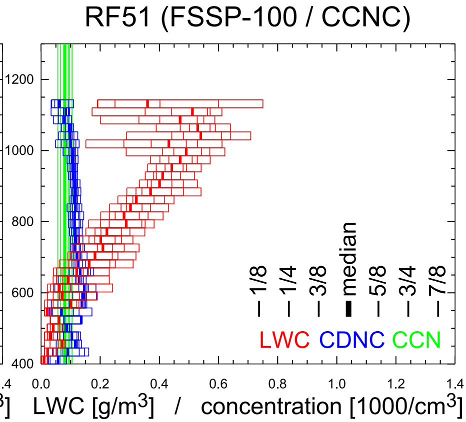
- min–max ranges indicated
- VSPMS and VOPC were connected with the inlet through a heater set at 280° C

[4] data courtesy of LaMP, Clermont-Ferrand, France

Case study: marine Sc vs. continental Cu

cloud-droplet (FSSP) and CCN measurements[5]





- CCNC operated at 0.21% supersaturation
- FSSP-100 classifies droplets into 15 size bins in the 1–25μm radius range

[5] data courtesy of CNRS/Météo-France, Toulouse, France