Cloud-droplet activation from the perspectives of a novel adaptive moving-sectional air-parcel model and the EUCAARI-IMPACT observations

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Institute of Geophysics, University of Warsaw, Poland

data courtesy of LaMP, Clermont-Ferrand, France (**Suzanne Crumeyrolle et al.**)
and CNRS/Météo-France, Toulouse, France (**Laurent Gomes et al.**)
and the **EUCAARI-IMPACT team**

August 19th 2010, JAMSTEC, Yokohama, Japan
August 23rd 2010, MRI-JMA, Tsukuba, Japan

Atmospheric physics at the Univ. of Warsaw

	est.	staff	students BSc/MSc	students PhD
University of Warsaw	1816	6000	55000	2600
Faculty of Physics	1816	330	450	100
Institute of Geophysics	1948	22	20	15
Atmospheric Physics Division	1949	8	8	9

- research:
  - microphysics of clouds and precipitation,
  - cloud modeling,
  - construction of instrumentation for airborne measurements (the UFT ultra-fast temperature probe),
  - aerosol remote sensing and radiative transfer modeling,
  - theoretical hydrodynamics
- educational offer:
  - BSc (3 years), MSc (3+2 years) and PhD (4 years) programmes in physics at the Faculty of Physics

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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
- key aim: halve the uncertainty of the impact of aerosol on climate

Plan of the talk

1. CCN activation and aerosol-cloud-climate interactions
2. CCN activation in a moving-sectional air-parcel model
3. CCN activation in the EUCAARI-IMPACT measurements
4. Summary and outlook

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CCN & aerosol-cloud-climate interactions

Aitken (1875):

- "when water vapour condenses in the atmosphere, it always does so on some solid nucleus"
- "if there was no dust in the air there would be no fogs, no clouds, no mists, and probably no rain"
- "there is probably also something due to the composition of the dust particles; some kinds of dust seem to form better nuclei than others"

Langmuir (1948):

- at the base of a cloud ... thousands of particles [droplets] per cc
- the small particles ... scatter [light] more completely
- a wide distribution of them [particles, IN] in the atmosphere might perhaps have a profound effect upon the climate

Aitken, J.: On boiling, condensing, freezing, and melting, T. Roy. Scott. Soc. Arts, 9, 7–17, (as quoted in Podzimek, J.: B. Am. Meteorol. Soc., 70, 1538–1545, 1989), 1875.

Langmuir, I.: The growth of particles in smokes and clouds and the production of snow from super-cooled clouds, P. Am. Philos. Soc., 92, 167–85, 1948.

CCN & aerosol-cloud-climate interactions

Squires (1958) – ability to precipitate:

- "high droplet concentrations ... assoc. with ... clouds failing to rain"
- "low droplet concentrations ... assoc. with ... release of warm rain"

Twomey (1977) – albedo:

- "pollution acts to increase the reflectance (albedo) of clouds"

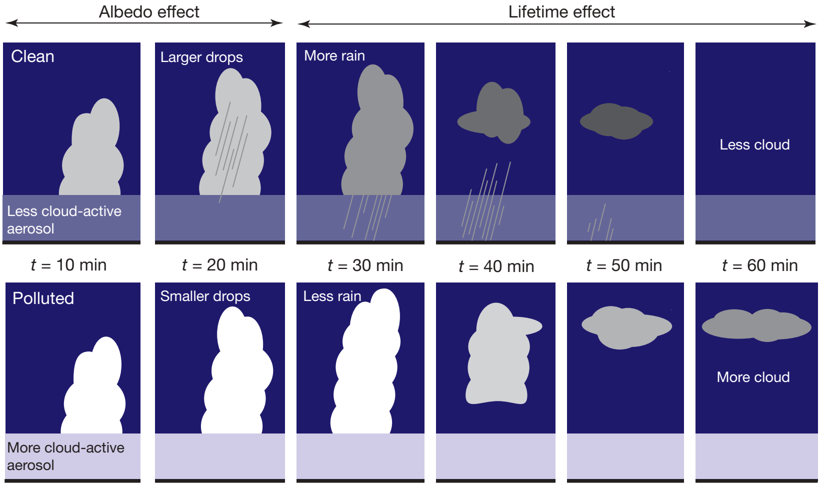
Albrecht (1989) – lifetime:

- "increases in aerosol concentrations over the oceans may increase the amount of low-level cloudiness..."

Albrecht, B.: Aerosols, cloud microphysics, and fractional cloudiness, Science, 245, 1227–1230, 1989.

Squires, P.: The microstructure and colloidal stability of warm clouds. I. The relation between structure and stability, Tellus, 10, 256–271, 1958.

Twomey, S.: The influence of pollution on the shortwave albedo of clouds, J. Atmos. Sci., 34, 1149–1152, 1977.



(figure from Stevens and Feingold, 2009)

Stevens, B. and Feingold, G.: Untangling aerosol effects on clouds and precipitation in a buffered system, Nature, 461, 607–613, 2009.

Plan of the talk

Cloud droplet formation on aerosol in an air-parcel model

- 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

Mathematical description (elements)

- ordinary differential equation for the rate of change of temperature (adiabatic heat balance, equation of state, hydrostatic equilibrium):

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

- 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

t: time

C: heat capacity

g: gravit. accel.

w: updraft speed

L: latent heat

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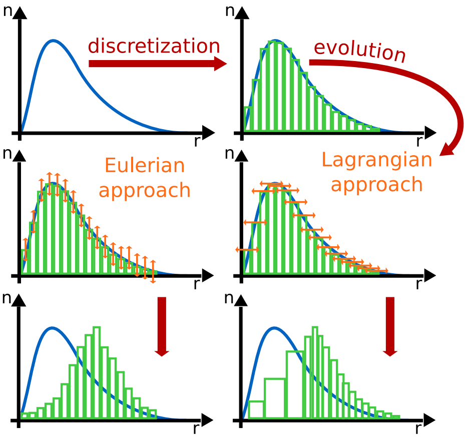
D: vapour diffusivity

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.
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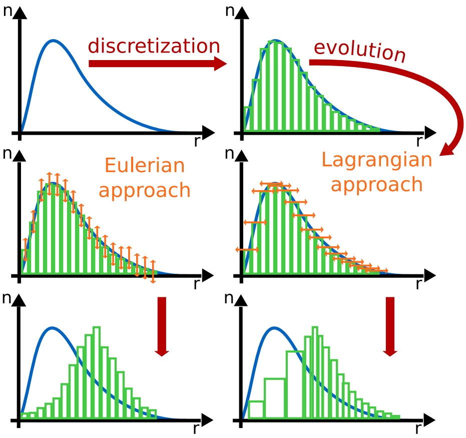

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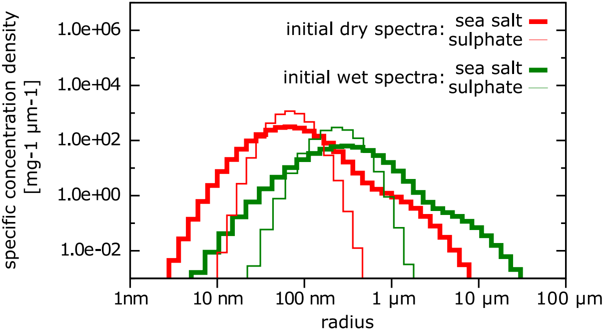
Example: sea-salt sulphate competition

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

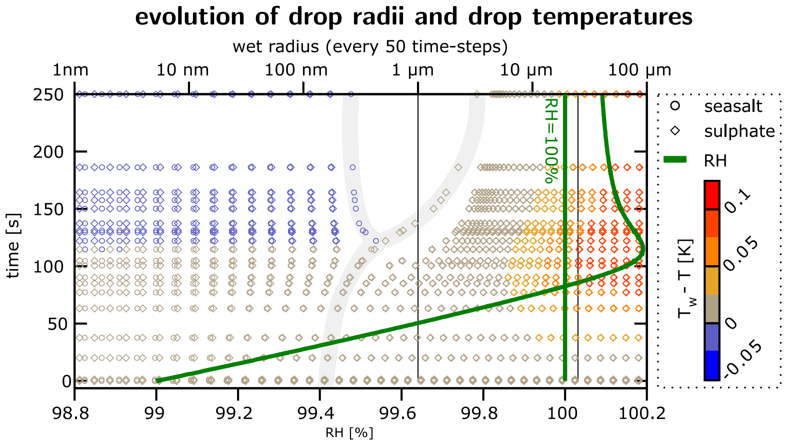


mode	solute	N_m [cm^{-3}]	r_m [μm]	σ_m [1]
film drop	NaCl	51.1	0.10	1.90
jet drop	NaCl	2.21	1.00	2.00
spume drop	NaCl	0.00001	6.00	3.00
accumulation	$(NH_4)_2SO_4$	100	0.08	1.45

- 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.

Example: sea-salt sulphate competition

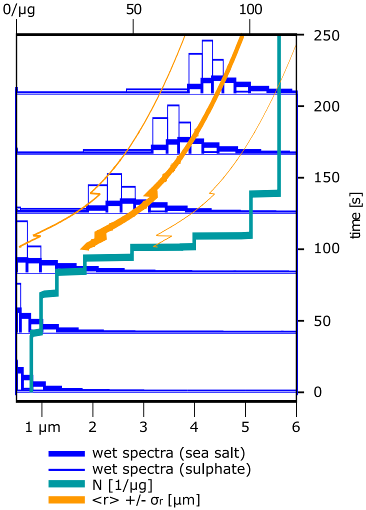


- time-step adjustments (CVODE solver of Cohen and Hindmarsh, 1996)

Cohen, S. and Hindmarsh, A.: CVODE, A stiff/nonstiff ODE solver in C, Comput. Phys., 10, 138–143, (also available as LLNL technical report UCRL-JC-121014), 1996.

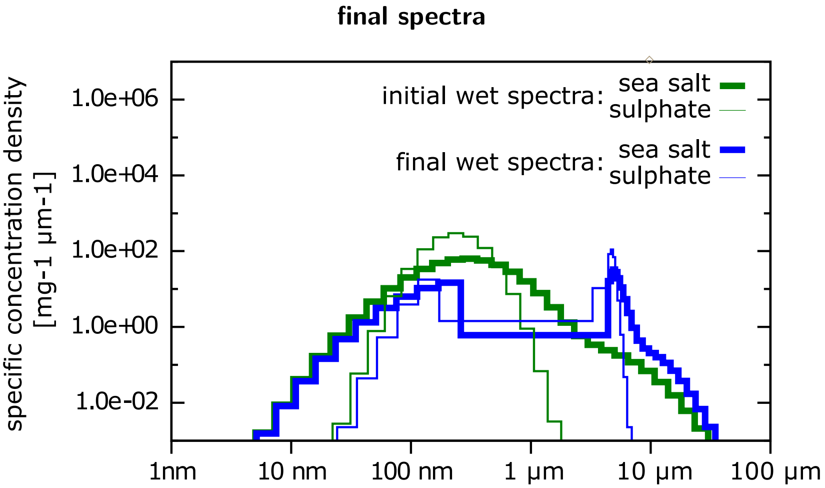
Example: sea-salt sulphate competition

cloud droplet spectrum evolution



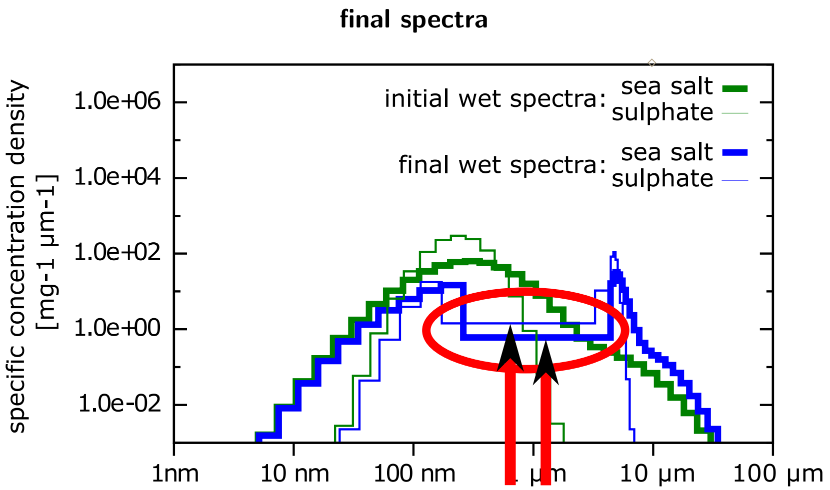
- size and concentration of droplets within the 1–25μm radius range
- droplet concentration (turquoise line) increases till 140 s (35 m), then constant
- average radius (thick orange line) increases continuously with height
- standard deviation approximately constant (thin orange lines: $\pm\sigma_r$)
- sulphate solution droplets dominate in the small-size part of the spectrum (histograms drawn with thin blue lines)
- largest droplets formed on sea-salt (histograms drawn with thick blue lines)

Example: sea-salt sulphate competition



bimodal: unactivated aerosol mode & activated cloud-droplet mode

Example: sea-salt sulphate competition



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):

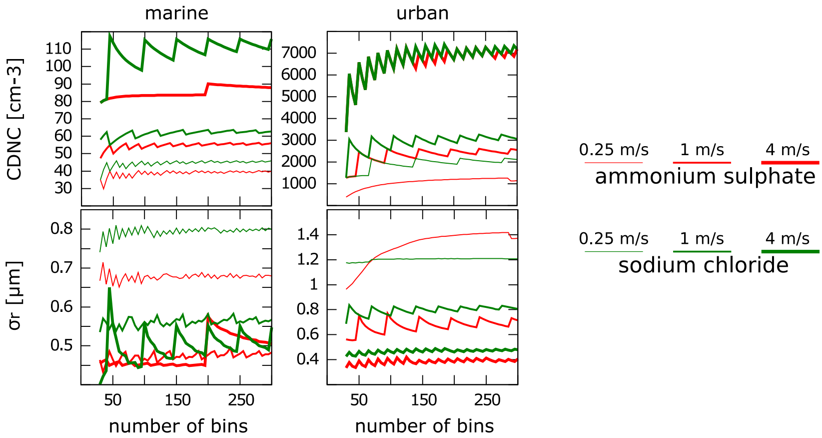
- "...special attention must be paid to the critical sections(s), i.e. size section(s) into which a minimum activation diameter falls."
- "...doubling the particle size resolution improved the results significantly... "

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.

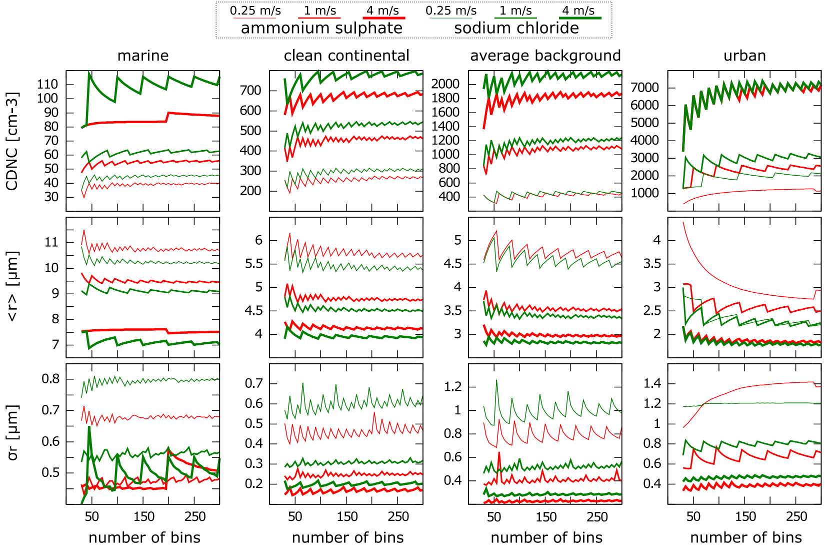
MOL for CCN activation: how accurate?



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?



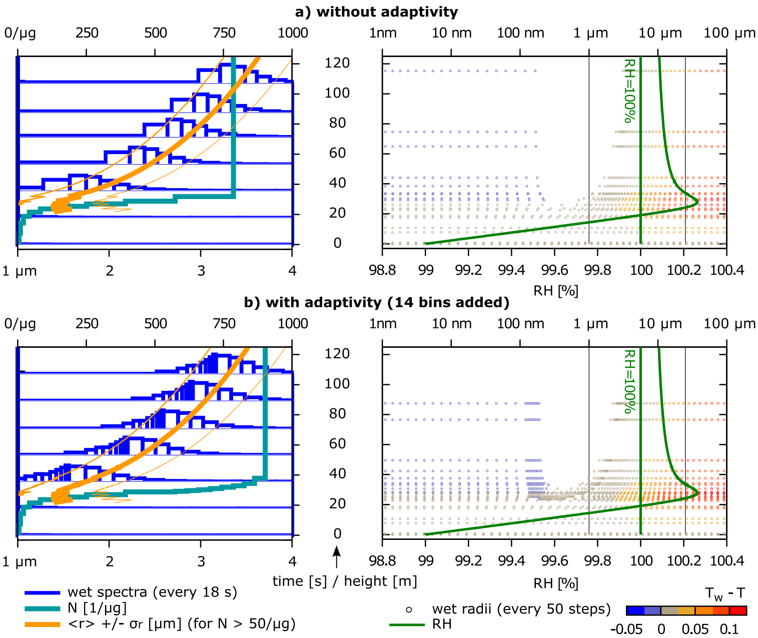
MOL for CCN activation: how to improve?

Arabas and Pawlowska (2010, submitted to GMD):

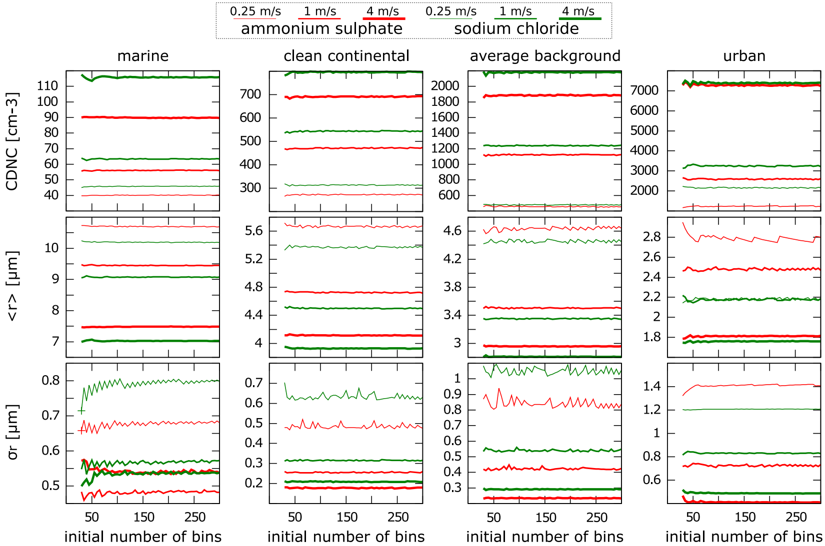
- employ the Adaptive MOL (Wouwer et al., 2001)
- identify and split the critical sections into several smaller ones if needed (i.e. adaptive grid refinement)
- introduce an uncertainty-related parameter for controlling the precision of calculations with regard to spectrum discretization
- suppress the 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.

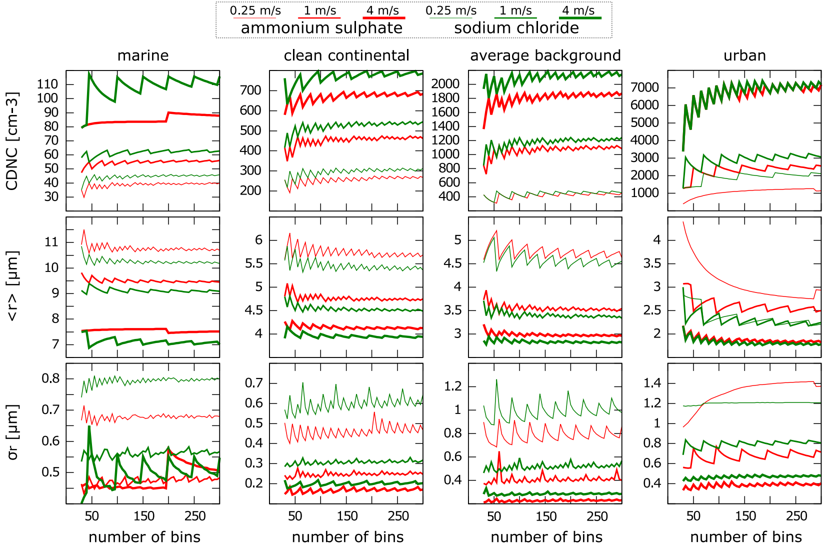
Wouwer, A., Saucez, P., and Schiesser, W., eds.: Adaptive Method of Lines, Chapman & Hall/CRC, 2001.


a) without adaptivity
0/μg 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)
0/μg 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
time [s] / height [m]
98.8 99 99.2 99.4 99.6 99.8 100 100.2 100.4
RH [%]
wet spectra (every 18 s)
N [1/μg]
<r> +/- σr [μm] (for N > 50/μg)
wet radii (every 50 steps)
RH
Tw - T
-0.05 0 0.05 0.1

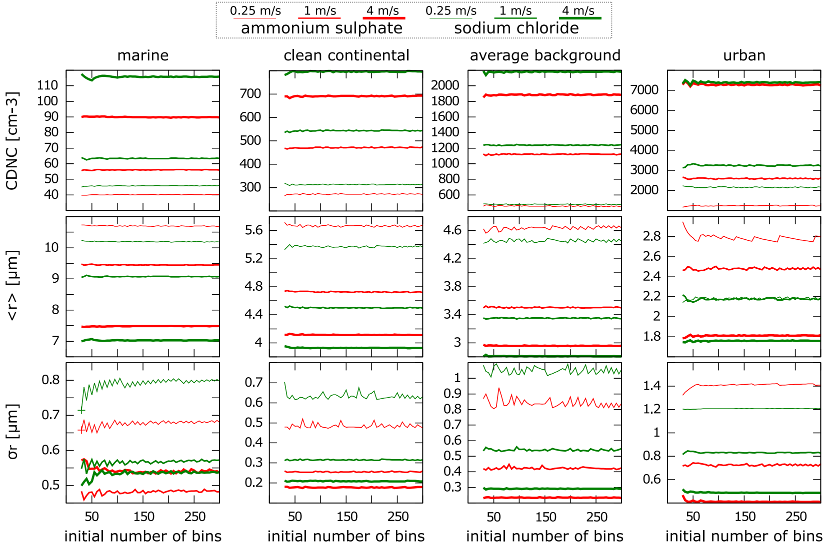
aMOL for CCN activation: it works!



aMOL for CCN activation: adaptivity off



aMOL for CCN activation: adaptivity on



A few words more about the implementation

- aerosol chemical composition using the κ-Köhler parameterization (Petters and Kreidenweis, 2007)
- implemented in C++ using Boost.units (Schabel and Watanabe, 2008)
- ODE integration using SUNDIALS/CVODE (Hindmarsh et al., 2005)
- helper numerics (root-finding etc) from GSL (Galassi et al., 2009)
- source code and a manual in an electronic supplement of the paper (GNU GPL license, journal with open-access policy)
- ... and a user-friendly web-based interface (CLI as well)

Galassi, M., Davies, J., Theiler, J., Gough, B., Jungman, G., Alken, P., Booth, M., and Rossi, F.: GNU Scientific Library Reference Manual - Third Edition (v1.12), Network Theory Ltd., 2009.

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.

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

Schabel, M. and Watanabe, S.: Boost.Units: Zero-overhead dimensional analysis and unit/quantity manipulation and conversion, in: Boost Library Documentation, (available at `http://www.boost.org/doc/libs/`), 2008.

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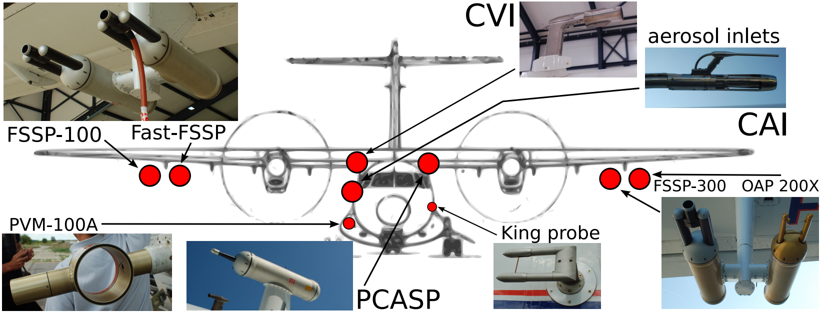
EUCAARI Intensite Measurement Period At Cabauw Tower (IMPACT)

- 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$^\circ C$), CPCs, nephelometer, ...

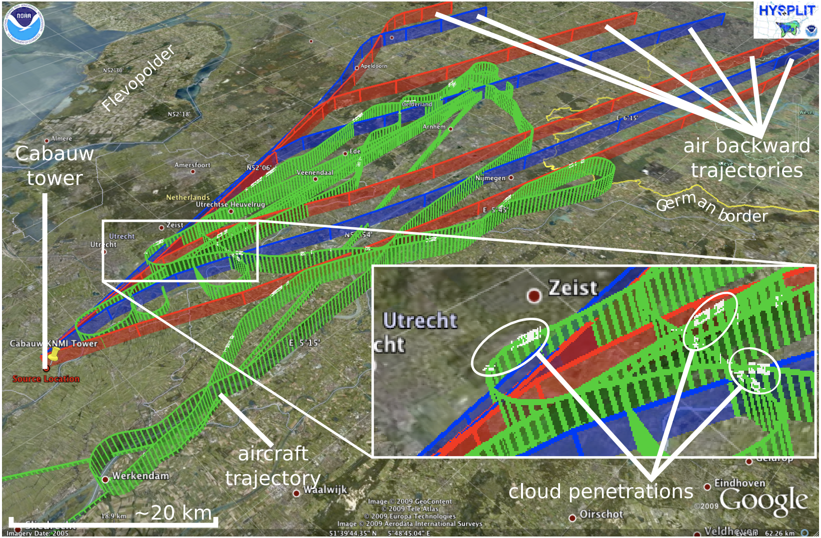
Case study: marine Sc vs. continental Cu

two flights in air-masses of contrasting characteristics:

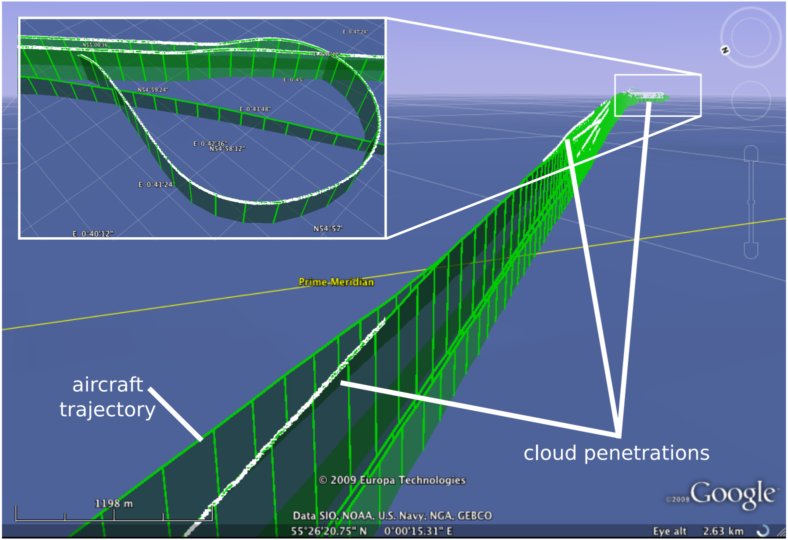
- RF49, May 13th 2008:
  - flown over The Netherlands,
  - polluted continental air,
  - $\sim$ 5 minutes of penetrations of Cu clouds

- RF51, May 15th 2008:
  - flown over the North Sea,
  - relatively pristine marine air,
  - $\sim$ 50 minutes of penetrations of a deck of Sc clouds

Case study: RF49 (May 13th) Cu above NL



Case study: RF51 (May 15th) North Sea Sc



Case study: marine Sc vs. continental Cu

CCN activation relevant measurements:

- near cloud base:
  - dry aerosol size spectrum
  - wet aerosol size spectrum
  - CCN count
  - vertical velocity
  - basic meteorological parameters
- in cloud:
  - cloud droplet size spectrum

Case study: marine Sc vs. continental Cu

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  - CCN count (model output)
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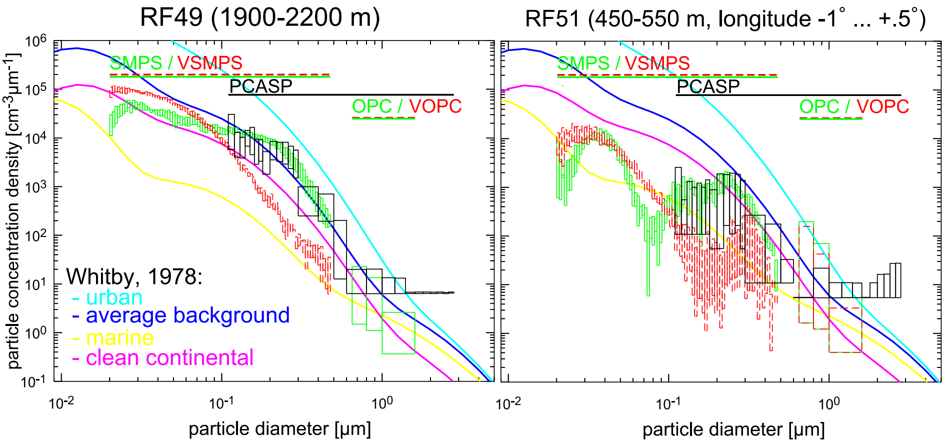
Case study: marine Sc vs. continental Cu

aerosol measurements below cloud base[2]

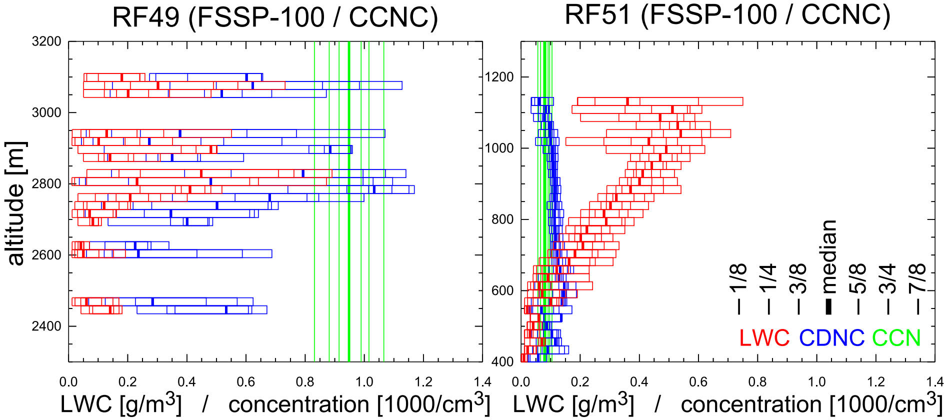


- min–max ranges indicated
- VSPMS and VOPC were connected with the inlet through a heater set at 280° *C*

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

Case study: marine Sc vs. continental Cu

cloud-droplet (FSSP) and CCN measurements[3]



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

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

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4. **Summary and outlook**

Summary:

- Development of an accurate CCN activation model

Paper accepted for public review & discussion at:
http://www.geosci-model-dev-discuss.net/
(model code and a manual available at the journal website)

Arabas & Pawlowska 2010: Adaptive method of lines for multi-component aerosol condensational growth and cloud droplet activation

- Preliminary statistical analysis of IMPACT airborne measurements of parameters relevant to CCN activation

- Assessment of closure between the model and the measurements

- Employment of the parcel model for improving CCN activation parameterization for use in models in which it is a sub-scale process (e.g. LES)

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Arabas & Pawlowska 2010: Adaptive method of lines for multi-component aerosol condensational growth and cloud droplet activation

- Preliminary statistical analysis of IMPACT airborne measurements of parameters relevant to CCN activation

Outlook (ongoing work):

- Assessment of closure between the model and the measurements
- Employment of the parcel model for improving CCN activation parameterization for use in models in which it is a sub-scale process (e.g. LES)

Summary:

- Development of an accurate CCN activation model

Paper accepted for public review & discussion at:
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ご清聴ありがとうございました。

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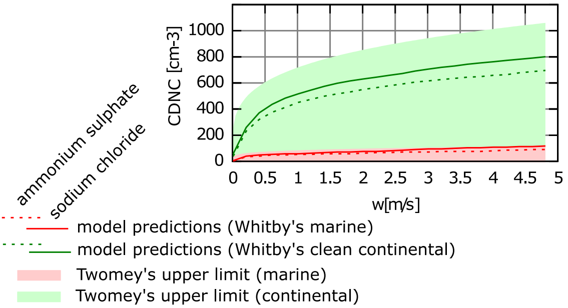
Thank you for the invitation!

Example usage / tentative validation

comparison with the Twomey's approximate upper-limit solution (1959):



E: dew-point elevation [K]
w: vertical velocity [cm/s]
$CNDC$: drop conc. [cm^{-3}]
k, c: aerosol parameters

marine: $k = 1/3,\ c = 125/cm^3$

continental: $k = 2/5,\ c = 2000/cm^3$

$$E_{max} < \left[\frac{1.63\times10^{-3}\cdot w^{3/2}}{c\cdot k\cdot \mathrm{B}(3/2,k/2)}\right]^{\frac{1}{k+2}}$$

$$CDNC < c\cdot(E_{max})^k$$

may be used for creating look-up tables/parametrizations for models run at time/space resolutions where CCN activation is a sub-scale process

Twomey, S.: The nuclei of natural cloud formation. Part II: The supersaturation in natural clouds and the variation of cloud droplet concentration, Geofis. Pura Appl., 43, 243–249, 1959.

MOL's ODE system

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