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Importance of aerosol composition and mixing state for cloud droplet activation in the high Arctic

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Importance of aerosol composition and mixing state for cloud droplet activation in the high Arctic

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Supplementary material

We developed a new implementation (written in Matlab) of the adiabatic parcel model described by Pruppacher and Klett (1997). It consists of essentially the same equations as the model presented in Leaitch et al. (1986), with the difference that we use the full implicit dynamic diffusional growth equation for particles in each size bin:

$$r\frac{dr}{dt} = \frac{D' M_w e_{sat,w}}{\rho_s RT}\left(S - \frac{1}{1+\delta}\exp\left[\frac{L_e M_w}{RT}\left(\frac{\delta}{1+\delta}\right) + \frac{2\mathrm{M}_w \sigma_{s/a}}{RT(1+\delta)\rho_w r_i} - \frac{\nu \Phi_s \varepsilon_m M_w \rho_N r_N^3}{M_s ?_w \left(r^3 - r_N^3\right)}\right]\right) \quad (1),$$

where:

r - radius of particle

t - time

M_w - molecular weight of water

$e_{sat,\,w}$ - saturation pressure of water vapor above a water surface

ρ_s - density of the droplet solution

R - universal gas constant

T - temperature of the gas surroundings

S - supersaturation

L_e - latent heat of evaporation of water

$\sigma_{s/a}$ - surface tension at solution/air interface

ρ_w - density of pure water

ν - ion number of the salt

Φ_s - osmotic coefficient of droplet solution

ε_m - mass fraction of soluble material

ρ_N - density of dry nucleus

M_s - molecular mass of nucleus material

$r_{N,\,I}$ - radius of dry nucleus

$\delta = T_d / T - 1$, where T_D is the droplet temperature, gives a measure of the heating or cooling caused by latent heat released when the droplet grows or shrinks. It is given by:

$$\delta = \frac{L_e \rho_s}{TK'} r \frac{dr}{dt} \qquad (2)$$

The D' and K' are modified diffusion and thermal conduction coefficients:

$$D' = \frac{D}{\left[\frac{r}{r+\lambda} + \frac{D}{r\alpha_c} \sqrt{\frac{2\pi M_w}{RT}} \right]} \qquad (3)$$

$$K' = \frac{k}{\left[\frac{r}{r+\lambda} + \frac{k}{r\alpha_t \rho c_{pa}} \sqrt{\frac{2\pi M_A}{RT}} \right]} \qquad (4)$$

where:

D - pressure and temperature dependent diffusivity of water vapor in air

λ - constant ($1.2*10^{-7}$ m)

α_c - condensation accommodation coefficient

k - heat conductivity

α_t - condensation accommodation coefficient

M_A - mean molecular mass of air

We simplify the Raoult term in the particle growth equation by identifying it as the water activity.

$$\hat{M}\Phi = -\ln\left(a_w\right)$$

$$\frac{\nu\Phi_s\varepsilon_m M_w\rho_N r_N^3}{M_s\rho_w\left(r^3 - r_N^3\right)} = \nu M \tag{5}$$

Recent research has shown that both of the accommodation coefficients are likely to have values close to unity for pure water surfaces (e.g. Winkler et al., 2004, Winkler et. al., 2006, Morita et al., 2004).

Due to the dependence of δ on dr/dt, the system of ordinary differential equations is implicit, and hence we need to provide both starting values of T, p, S, w and r_i and their time derivatives. There are a few numerical caveats in the model, such as particle sizes shrinking below the dry size or even becoming negative. Unphysical developments like these are avoided by artificially setting the particle size derivatives positive when the droplet size approaches the size of the dry particle.

In contrast to the model of Leaitch et al. (1986) we make no assumptions of log-normal aerosol distributions. Instead, we directly as input use the measured size distributions by the TDMPS. The lower cut-off for the chemical information is 22 nm in diameter, which is well below the size required to act as CCN in the conditions of his study. Chemical mass concentration data from LPI impactor samples are interpolated on to the TDMPS size bins and thereafter converted to number concentration assuming spherical particles.

$$r_i \frac{dr_i}{dt} = \frac{D' M_w e_{sat,w}}{\rho_s RT} \left(S - \frac{1}{1+\delta} \exp\left[\frac{L_e M_w}{RT} \left(\frac{\delta}{1+\delta} \right) + \frac{2\mathrm{M}_w \sigma_{s/a}}{RT(1+\delta)\rho_w r_i} - \frac{\nu \Phi_s \varepsilon_m M_w \rho_N r_{N,i}^3}{M_s \rho_w \left(r_i^3 - r_{N,j}^3 \right)} \right] \right) \tag{6}$$

For an isolated air parcel ascending adiabatically with the vertical speed V, the temperature T, pressure p, supersaturation S and liquid water content are described by the equations:

$$-\frac{dT}{dt} = \frac{gV}{c_{pa}} + \frac{L_e}{c_{pa}} \frac{dw}{dt} \tag{7}$$

$$\frac{dp}{dt} = -\frac{gpV}{R_a T} \tag{8}$$

$$\frac{dS}{dt} = \frac{p}{\varepsilon e_{sat,w}} - (1+S) \left[\frac{?L_e}{R_a T^2} \frac{dT}{dt} + \frac{gV}{R_a T} \right] \tag{9}$$

$$\frac{dw}{dt} = \sum_i 4\pi n_i r_i^2 \frac{dr_i}{dt} \tag{8}$$

In this study, however, we use V=0 and artificially change S in the way the CCN counter does.