Global Simulations of Aerosol Size-Dependent In-Cloud Scavenging

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Summary:

•For fine mode soluble aerosols in mixed phase clouds, the cloud-borne fractions have wide variability, which prescribed fractions cannot capture. A mixed phase cloud-borne fraction of 0.8 for black carbon biased concentrations low by one order of magnitude in ECHAM5-HAM

•For coarse mode insoluble aerosols such as dust the parameterization of impaction scavenging is important for prediction of cloud-borne fractions

•Cloud-borne fractions should be assigned with attention to size/hygroscopicity, and cloud phase.

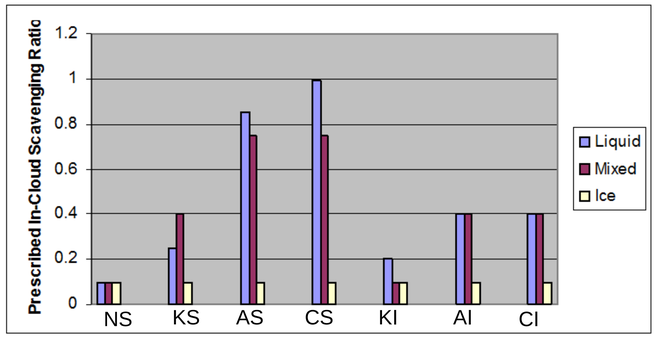
Introduction/Methodology:

Wet deposition processes strongly control aerosol three-dimensional distributions. Global models, such as GEOS-Chem typically treat wet deposition as a first order loss process, which depends on 1) a rate constant for the conversion of cloud water to precipitate, and 2) an aerosol cloud-borne fraction. In GEOS-Chem (Lui et al. (2001)) the soluble aerosol cloud-borne fraction is assumed to be unity for cloud droplets. In actuality, the cloud-borne fraction depends on a variety of parameters such as aerosol size/hygroscopicity, as well as cloud temperature/phase, and supersaturation. Alternatives to the assumption that 100% of in-cloud soluble aerosol is cloud-borne include:

1) prescribed cloud-borne fractions (e.g. Stier et al. (2005))
2) diagnosed cloud-borne fractions based on certain parameters such as updraft velocity, supersaturation, aerosol size/hygroscopity and cloud hydrometeor size(e.g. Croft et al. (2009))
3) prognostic treatments of aerosol mass contained in cloud droplets and ice crystals (e.g. Hoose et al. (2008))

This study compares predicted cloud-borne fractions and aerosol distributions for these three treatments of cloud-borne aerosol. All results presented are for 1-year simulations with the ECHAM5-HAM global climate model (Roeckner et al. (2003), Lohmann et al. (2007)).

Prescribed Cloud-Borne Fractions:



(As a function of cloud temperature and aerosol size)

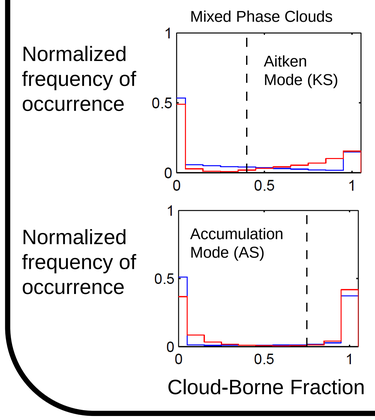
ECHAM5-HAM log-normal modes:

Internally mixed modes:
NS: Nucleation Soluble
KS: Aitken Soluble
AS: Accumulation Soluble
CS: Coarse Soluble

Externally mixed modes:
KI: Aitken inoluble
AI: Accumulation insoluble
CI: Coarse insoluble

Mixed phase cloud: 238K < T < 273K

Diagnosed Cloud-Borne Fractions:

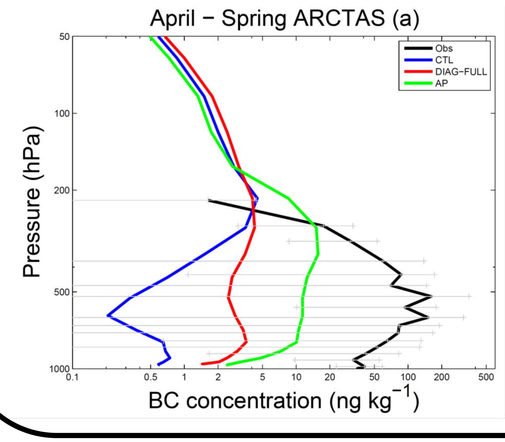


Blue: Fractions for number

Red: Fractions for mass

Dashed: Prescribed fraction of ECHAM5-HAM

Sensitivity of Black Carbon Concentrations to Scavenging:



→ Assumptions related to cloud-borne aerosol fractions can yield an order of magnitude low bias for aerosol concentrations in regions of mixed/ice phase clouds

Simulation acronyms defined below.

Marine Boundary Layer Size Distributions:

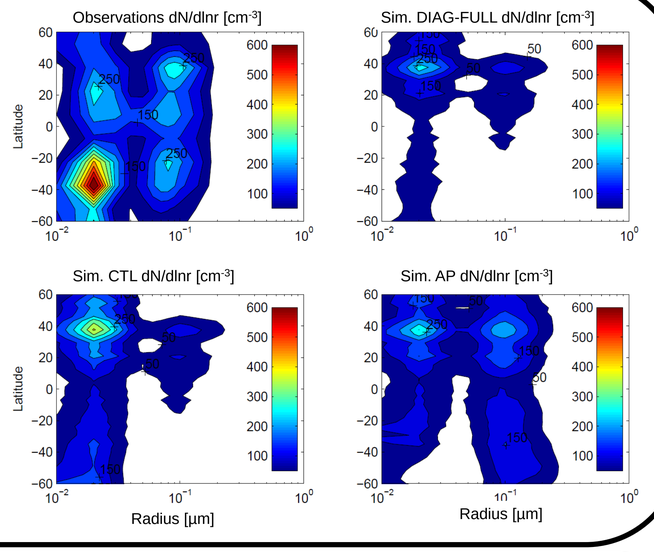
CTL: Standard model with prescribed cloud-borne aerosol fractions

DIAG-FULL: Diagnostic size-dependent uptake into cloud hydrometeors by nucleation and impaction scavenging

AP: In-droplet and in-crystal aerosol mass treated as prognostic species

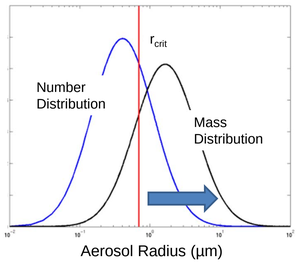
Observations from Heintzenberg et al. (2000)

→ Prognostic scheme predicts accumulation mode in closest agreement with observations.



Size-Dependent Uptake into Cloud Hydrometeors:

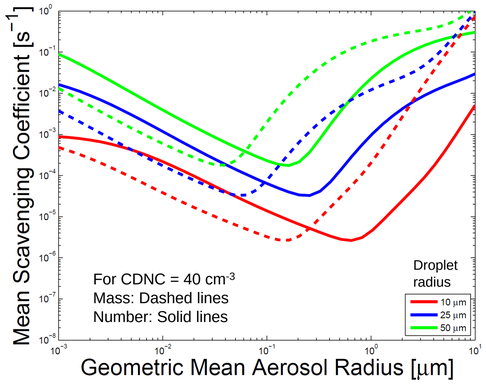
Nucleation Scavenging:
Assume each cloud droplet and ice crystal scavenge one aerosol. Schematic below shows fractional scavenging of mass distribution is greater than for the number distribution, scavenge all mass for r > r_{crit}.



$$r_{crit} = r_g\left[\exp(\sqrt{2}\ln\sigma_g \bullet erf^{-1}(1-(2\bullet(CDNC+ICNC)\frac{frac_j}{N_{>35nm}})))\right]$$

where $frac_j$ is the fraction of soluble mode j number having radius > 35nm
$N_{>35nm}$ is the number of aerosols having radius > 35 nm for all soluble modes
r_g and σ_g are parameters of the lognormal distribution, *erf* is the error function.

Impaction Scavenging:
Scavenging coefficients are found in look-up tables as a function of aerosol size, cloud droplet/ice crystal size and cloud droplet/ice crystal number concentration (CDNC/ICNC).



(Similar approach for aerosol-ice crystal collisions)

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Acknowledgements: NSERC CRSNG