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Below-cloud scavenging: an efficient removal mechanism for super-micron aerosol particles.

J. S. Henzing and P. F. J. van Velthoven
Royal Netherlands Meteorological Institute, The Netherlands, (henzing@knmi.nl)

Introduction
Wet deposition is by far the most important process by which aerosol particles are removed from the atmosphere. In order to realistically simulate aerosol distributions a sufficiently accurate parameterization of in-cloud and below-cloud aerosol scavenging thus needs to be developed for aerosol chemistry models.

Parameterization of wet removal
We have made simulations with the global chemistry transport model TM4, which contains a 12-bin sea salt aerosol scheme, in order to investigate the partitioning of large-scale wet deposition into in-cloud and below-cloud scavenging as a function aerosol particle size. The large-scale in-cloud scavenging through precipitation builds on the work of Roelofs and Lelieveld (1995). For the below-cloud scavenging, the rate of transfer of aerosol particles into falling rain droplets is approximated by the product of aerosol concentration and its scavenging coefficient, Λ:

$$(1)\ \Lambda(r_p) =_0 \int^{\infty} \quad \pi R_p^2 U_t(R_p) \quad E(r_p, R_p) \quad N(R_p)\mathrm{d}R_p$$

where r_p is the aerosol particle radius. The first term under the integral represents the cylinder occupied by a rain droplet with radius R_p falling with (terminal) velocity U_t. The collection efficiency, E, accounts for the fact that not all particles in the swept cylinder are actually collected (Slinn, 1984). We used various published rain droplet size distributions (last term under integral in Eq. 1) to investigate the sensitivity of below-cloud scavenging to the choice of the, generally unknown, rain droplet spectrum. Not only the amount of rain droplets but also rain droplet spectrum depends on the precipitation intensity. For individual aerosol particles, we have calculated the scavenging coefficient as a function of precipitation intensity. For parameterization in our chemical transport model, we replace these computationally expensive calcula-

tions by numerically fast and highly accurate parameterizations. The integration over the aerosol spectrum to obtain the scavenging coefficient for a specific sea salt bin is still done within the transport model itself.

Aerosol size-dependence

Nucleation and accumulation mode particles that have radii smaller than 1 micrometer are effectively scavenged inside clouds, whereas falling rain droplets do not collect these fine particles easily below clouds. The reason for this is that small particles adjust their movement to the induced flow around a falling rain droplet, so that particles move away from falling droplets. This adjustment is very efficient: we find that only about 1% of all accumulation mode aerosol-mass is removed below cloud. This result is in agreement with other studies. Aerosol particles with an anthropogenic source mainly reside in this accumulation mode and in-cloud scavenging is therefore their dominant sink. If one is interested in aerosol column loads only, a good representation of in-cloud scavenging is thus sufficient for these particles.

Coarse mode aerosol particles with radii larger than 1 micrometer are also effectively scavenged by in-cloud processes. Below the cloud, size and mass of these particles (desert dust, sea salt, and road dust) is of crucial importance. A considerable fraction of these particles is too large to avoid collision with a falling rain droplet and/or they are too inert to adjust to the induced flow around the falling droplet. As a consequence, we find that for super-micron aerosol below-cloud scavenging is an equally important removal mechanism as in-cloud scavenging.

Conclusion

We have developed a numerically fast and accurate parameterization for below-cloud scavenging of aerosols in chemistry transport models. We also have also found that for super-micron particles below-cloud scavenging cannot be neglected, contrary to what is often assumed in the literature. This leads to changes in the simulated optical depth distribution of sea salt aerosols.

References

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