



## A New Non-Diffusive Scheme of Particle Sedimentation for Use with Global Circulation Models. A Sensitivity Study in the Framework of a Geoengineered Stratosphere.

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The upwind-differencing sedimentation scheme is frequently used within global circulation models for considering aerosol and/or raindrop downward transport. The motivation for the upwind scheme owes to its very moderate computational expense versus the relevance of sedimentation to the modeled system. For tropospheric particles of 10-1000 nm size sedimentation is a negligible process due to their average residence time of about 10 days only, which contrasts with a residence time of up to several years within the stratosphere. Sedimentation has thus been considered to be a key process to the effectiveness of stratospheric geoengineering (Rasch et al., 2008).

In order to explore the sensitivity of the geoengineered stratospheric aerosol to sedimentation, a new non-diffusive formalism has been developed. The method is based on a pseudo-equilibrium approach at the individual grid box interface. Since the flux through the upper boundary differs in general from the flux at the lower boundary the methods still allows for dynamical changes of the particle grid box concentration. However, a pseudo-equilibrium approach tends to produce poor results under transient conditions, for instance during the build up of the geoengineered aerosol plume. To assess the relevance of that point, the new formalism was further combined with the Walcek advection scheme (2000), which has been adapted to account for the nature of sedimentation. The original Walcek scheme may not be used for sedimentation as sedimentation is a non-monotonic process due to the disparity of particle size. The combined scheme assesses the sedimentation flux following Walcek's approach to estimate the gridbox internal gradient and compares it to the flux calculated with the new non-diffusive scheme. The largest value is then chosen. Some limited numerical diffusion as inherent to the Walcek scheme is therefore present.

Global sensitivity runs comparing the upwind with the new non-diffusive scheme as well as with the modified Walcek scheme were performed with the EMAC-model (Joeckel et al., 2006). Microphysical processes were switched off during these runs so as to ensure a monodisperse aerosol. As expected the influence of sedimentation on the concentration of the geoengineered aerosol in the troposphere is negligible. Also the influence of the chosen sedimentation method on the global burden of the aerosol is only marginal for particles of up to 100 nm, but is substantial for 1000 nm particles. However, the new sedimentation schemes limit very effectively the transport of particles to the upper stratosphere, which occurs due to bi-directional numerical diffusion in simpler sedimentation schemes, and thereby tend to concentrate particles at their height of injection and below.

This study thus seems to indicate that sedimentation, even when it is modeled carefully, is not a limiting process to stratospheric geoengineering as long as no substantial large particle formation via coagulation occurs. In the model framework the residence time of small particles is mainly driven by transport within the stratosphere and stratosphere-troposphere exchange resulting in rapid wet removal. However, for particles of all sizes the use of a scheme with limited diffusion is key to limiting the upward transport of particles within the stratosphere. The limitation of upward transport may be relevant when it comes to the interaction of particles with stratospheric chemistry and the radiation balance, as well as in the context of their potential evaporation at higher altitudes.

### References

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