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Simulation of trace gas redistribution by convective clouds - Liquid phase processes

Y. Yin, D. J. Parker, and K. S. Carslaw
School of the Environment, University of Leeds, Leeds, U.K.

Abstract. A two-dimensional dynamic cloud model with detailed microphysics and a spectral treatment of gas scavenging was used to simulate trace gas vertical redistribution in precipitating continental and maritime clouds. A general picture of gas transport in such clouds has been developed by examining the sensitivity to a range of parameters, including cloud dynamic and microphysical structure, gas solubility, and the method of calculating gas uptake by droplets. Gases with effective Henry's law constants (H^*) ranging from zero to greater than 10^9 mol dm^{-3} atm^{-1} were simulated. The abundance of highly soluble gases in the uppermost parts (top 1 km or so) of continental precipitating clouds was found to be as much as 20-50% of that of the insoluble tracer under conditions where the mixing ratio of the tracer was approximately 5% of its boundary layer value. The abundance of highly soluble gases was approximately 6 times higher in the uppermost parts of the continental cloud than in the maritime cloud, due to differences in wet removal efficiency in the two cloud types. A fully kinetic calculation of gas uptake, as opposed to assuming Henry's law equilibrium, was found to have a significant effect on gas transport, with the abundance of highly soluble gases in the uppermost parts of the cloud being a factor of 5 lower in the equilibrium simulations. The temperature dependence of the Henry's law constant was also found to be an important parameter in determining the abundance of soluble gases at cloud top, with the abundance of moderately soluble gases being as much as 70% lower when the temperature dependence of H^* was included. This reduction in abundance was found to be equivalent to increasing the temperature-independent solubility by a factor of 7. The vertical transport of soluble gases could be parameterized in large-scale models by normalizing against the transport of tracers. However, our results suggest that there is no straightforward scaling factor, particularly if small concentrations of highly soluble gases in the upper troposphere need to be defined.

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