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Cell Model of In-cloud Scavenging of Highly Soluble Gases

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Transport of soluble gases in clouds is an integral part of the atmospheric transport of gases and is important for understanding the global distribution pattern of soluble trace gases. In the present study we investigated mass transfer during absorption of highly soluble gases such as hydrogen peroxide H_2O_2 and nitric acid HNO_3 by stagnant cloud droplets in the presence of inert admixtures.

Diffusion interactions between droplets, caused by the overlap of depleted of soluble gas regions around the neighboring droplets, are taken into account in the approximation of a cellular model of a gas-droplet suspension whereby a suspension is viewed as a periodic structure consisting of the identical spherical cells with periodic boundary conditions at the cell boundary. Using this model we determined temporal and spatial dependencies of the concentration of the soluble trace gas in a gaseous phase and in a droplet and calculated the dependence of the scavenging coefficient on time. It is shown that scavenging of highly soluble gases by cloud droplets leads to essential decrease of soluble trace gas concentration in the interstitial air. We found that scavenging coefficient for gas absorption by cloud droplets remains constant and sharply decreases only at the final stage of absorption. This assertion implies the exponential time decay of the average concentration of the soluble trace gas in the gaseous phase and can be used for the parameterization of gas scavenging by cloud droplets in the atmospheric transport modeling.

In the calculations we employed gamma size distribution of cloud droplets. It was shown that despite of the comparable values of Henry's law constants for the hydrogen peroxide and the nitric acid, the nitric acid is scavenged more effectively by cloud than the hydrogen peroxide due to a major affect of the dissociation reaction on nitric acid scavenging. We obtained also the analytical expressions for the "equilibrium values" of concentration of the active gas in a gaseous phase and for the total concentration in the liquid phase for the case of the hydrogen peroxide and nitric acid absorption by cloud droplets.

The developed cell model of in-cloud scavenging of highly soluble gases or parameterizations based on its results can be easily integrated into online coupled meteorology-chemistry or climate-chemistry models, where the cloud processes and chemical transformation of atmospheric pollutants are considered together with two-way interactions.