A new model framework for bulk – interface partitioning and chemistry of aqueous aerosols

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Chemical reactions occurring at the interface of aqueous aerosols play an important role in atmospheric chemistry, with effects such as the accelerated oxidation and photolysis of several key compounds, e.g., S(IV), NO_2 and O_3. Nevertheless, major uncertainties still persist, for example, the interfacial concentration remains undetermined, and this is key to understanding the interfacial reaction effects. The interfacial concentration can be determined through a bulk – interface partitioning coefficient (Kp), which is the ratio between the concentration of a compound in the interface and in the bulk. A new model framework (CAPRAM-HET2.0) was developed which estimates the bulk – interface partitioning coefficient through a relationship between surface tension and solubility, based on the partitioning approach introduced by Pegram and Record (2006). In total, 4532 species were partitioned, including radical species. Highly soluble species, such as H_2O_2 and HNO_3, partition very weakly to the interface, with Kp values of 1.3×10^{-3} and 7.3×10^{-4}, respectively. High bulk – interface partition coefficients are calculated for species with very low water solubility, including inorganic compounds such as NO, NO_2 and O_3, with Kp values of 2.7×10^{3}, 6.0×10^{2} and 6.3×10^{2}, respectively, and also organic compounds such as medium and long aliphatic chain hydrocarbons (C4 and above). The Kp of organic species increased with number of C atoms, and the addition of certain functional groups such as the nitrate group. The CAPRAM-HET2.0 model framework was added to the current gas- and aqueous-phase chemistry mechanism (MCM – CAPRAM), and box model simulations were performed in order to determine the impact of the enrichment or derichment of compounds at the gas-aerosol interface on tropospheric chemistry.