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Contribution of gaseous and particulate species to droplet solute composition at the Puy de Dôme, France

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Abstract. Chemical reactions of dissolved gases in the liquid phase play a key role in atmospheric processes both in the formation of secondary atmospheric compounds and their wet removal rate but also in the regulation of the oxidizing capacity of the troposphere. The behavior of gaseous species and their chemical transformation in clouds are difficult to observe experimentally given the complex nature of clouds.

During a winter field campaign at the summit of the Puy de Dôme (central France, 1465 m a.s.l), we have deployed an experimental set-up to provide a quantification of phase partitioning of both organic (CH_3COOH, HCOOH, $H_2C_2O_4$) and inorganic (NH_3, HNO_3, SO_2, HCl) species in clouds.

We found that nitric and hydrochloric acids can be considered close to Henry's law equilibrium, within analytical uncertainty and instrumental errors. On another hand, for NH_3 and carboxylic acids, dissolution of material from the gas phase is kinetically limited and never reaches the equilibrium predicted by thermodynamics, resulting in significant sub-saturation of the liquid phase. On the contrary, S^{IV} is supersaturated in the liquid phase, in addition to the presence of significant aerosol-derived S^{VI} transferred through nucleation scavenging.

Upon droplet evaporation, a significant part of most species, including S^{IV}, tends to efficiently return back into the gas phase. Overall, gas contribution to the droplet solute concentration ranges from at least 48.5 to 98% depending on the chemical species. This is particularly important considering that aerosol scavenging efficiencies are often calculated assuming a negligible gas-phase contribution to the solute concentration. Our study emphasizes the need to account for the in-cloud interaction between particles and gases to provide an adequate modeling of multiphase chemistry systems and its impact on the atmospheric aerosol and gas phases.

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