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On the Water Uptake and CCN Activation of Tropospheric Organic Aerosols

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Abstract

Aerosol particles introduce high uncertainties to radiative climate forcing. If exposed to a given relative humidity (RH), aerosol particles containing soluble material can absorb water and grow in size (hygroscopic growth). If RH is increased further beyond supersaturation (RH >100%) the particles can act as cloud condensation nuclei (CCN). Aerosol particles interactions with water vapour determine to a large extent their influence on climate. Organic aerosols (OA) contribute a large fraction (20-90%) of atmospheric submicron particulate mass, on the other hand they often consist of thousands of compounds with different properties. One of these properties is solubility, which affects the hygroscopic growth and cloud condensation nucleus (CCN) activation of the organic particles. We investigate the hygroscopic behaviour of complex organic aerosols accounting for the distribution of solubilities present in these mixtures. We use the SPARC method to estimate the solubility distributions of isoprene (IP) and monoterpene (MT) SOA based on their chemical composition, as predicted by the Master Chemical Mechanism (MCM). Combining these solubility distributions with the adsorption theory along with the non-ideal behaviour of organic mixtures, we predict the expected hygroscopic growth factors (HGFs), CCN activation behaviour and the related hygroscopicity parameters kappa for these mixtures. The predictions are compared to laboratory measurements as well as field data from MT- and IP-dominated measurement sites. The predicted solubility distributions do a good job in explaining the water uptake of these two mixture types at high relative humidities (RH around 90%), as well as their CCN activation – including the potential differences between the kappa values derived from HGF vs. CCN data. At lower relative humidities, however, the observed water uptake is higher than predicted on solubility alone, particularly for the MT-derived SOA. The data from the low RHs are further used to discuss the relative importance of surface adsorption and the non-ideality of the organic mixture in governing the water-uptake in these conditions. Finally, the sensitivity of direct and indirect radiative forcing to hygroscopic behaviour of organic particles is tested with the Norwegian Earth System Model (NorESM).