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Combined theoretical and experimental investigations of peroxy radical uptake on organic aerosols

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Significant uncertainties are still associated to chemical reaction mechanisms used in atmospheric models, in particular for ROx radicals (OH, HO_2, RO_2). Recent measurements of radicals in forested areas characterized by low NO_x (NO_2, NO) concentrations indicate that models can significantly overestimate peroxy radical concentrations.1,2 These results question the ability of models to correctly simulate the oxidative capacity of the troposphere since peroxy radicals are a main source of the hydroxyl radical (OH), one of the most important oxidative species in the atmosphere.3 One possible explanation is the occurrence of heterogeneous processes (uptake of radicals) on the surface of aerosols that are either misrepresented or not included in models. While recent studies have reported uptake coefficients of HO_2 on different types of aerosols, uptakes of RO_2 radicals have yet to be investigated and the process is not completely understood yet.

Both theoretical and experimental tools have been used to study HO_2 uptake on organic aerosols. A PERCA (Peroxy Radical Chemical Amplifier) instrument capable of measuring HO_2 and RO_2 radical concentrations has been coupled to an aerosol flow tube to quantify HO_2 uptakes on glutaric acid aerosols at different relative humidities. In complement to this experimental work, molecular dynamics combined with ab-initio calculations has been used to model a nanometer size aerosol particle and the sticking process of HO_2 on the organic particle. These theoretical calculations provide insight into the uptake process at the molecular scale. In this presentation, we will emphasize the advantage of using a dual experimental/theoretical approach to investigate the uptake of radical species on atmospheric aerosols.

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