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Investigation of the behavior of tropospheric relevant compounds at the interface gas/organic acid aerosols: An ONIOM QM/MM study

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The uptake of atmospheric gaseous oxidant such as O_3 or the ROx (OH, HO_2, RO_2) family, have a strong impact on the oxidative capacity of the atmosphere. [1], [2] Last decade, few studies have been carried out on the uptake of such compounds on atmospheric aerosol. However, the large variety of organic compounds provides uptake coefficients with a wide range of order of magnitude. [3], [4] Furthermore, the uptake resulting from the combination of different processes (mass accommodation, bulk diffusion, reactivity), the detailed understanding of such a process is not always accessible through experiments. Theoretical tools such as quantum mechanics (QM) combined with Molecular Mechanics (MM) is one way to investigate separately the different processes.

The ONIOM hybrid QM/MM method [5] allows to study the reactivity of few molecules in a large system. In our group, a methodology using this computational method have been developed in order to estimate the reactive uptake of gaseous compounds onto organic aerosol particles. In this presentation, reactive uptake of HO_2 and O_3 onto glutaric acid and oleic acid aerosols respectively will be discussed. Comparisons will be addressed with gas phase theoretical reaction rates and with experimental data.

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