EGU21-1094
https://doi.org/10.5194/egusphere-egu21-1094
EGU General Assembly 2021

Theoretical core spectroscopy of molecules interacting with ice surfaces

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Ice plays an essential role as a catalyst for reactions between atmospheric trace gases. The uptake of trace gases to ice has been proposed to have a major impact on geochemical cycles, human health, and ozone depletion in the stratosphere [1]. X-ray photoelectron spectroscopy (XPS) [2], serves as a powerful technique to characterize the elemental composition of such interacting species due to its surface sensitivity. Given the existence of complex physico-chemical processes such as adsorption, desorption, and migration within ice matrix, it is important to establish a theoretical framework to determine the electronic properties of these species under different conditions such as temperature and concentration. The focus of this work is to construct an embedding methodology employing Density Functional (DFT) and Wave Function Theory (WFT) to model and interpret photoelectron spectra of adsorbed halogenated species on ice surfaces at the core level with the highest accuracy possible.

We make use of an embedding approach utilizing full quantum mechanics to divide the system into subunits that will be treated at different levels of theory [3].

The goal is to determine core electron binding energies and the associated chemical shifts for the adsorbed halogenated species such as molecular HCl and the dissociated form Cl- at the surface and within the uppermost bulk layer of the ice respectively [4]. The core energy shifts are compared to the data derived from the XPS spectra [4].

We show that the use of a fully quantum mechanical embedding method, to treat solute-solvent systems is computationally efficient, yet accurate enough to determine the electronic properties of the solute system (halide ion) as well as the long-range effects of the solvent environment (ice).

We acknowledge support by the French government through the Program "Investissement d'avenir" through the Labex CaPPA (contract ANR-11-LABX-0005-01) and I-SITE ULNE project

OVERSEE (contract ANR-16-IDEX-0004), CPER CLIMIBIO (European Regional Development Fund, Hauts de France council, French Ministry of Higher Education and Research) and French national supercomputing facilities (grants DARI x2016081859 and A0050801859).