Contrats doctoraux 2026

Titre du projet de thèse : Exploring ammonia adsorption at air–water and air–ice interfaces on the molecular level

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Résumé du projet de thèse (en 20 lignes maximum) :

Ammonia (NH_3) is the most abundant alkaline trace gas in the atmosphere and plays a central role in controlling the pH of clouds and aerosols. It is also an important precursor of nitrogen oxides such as nitrous oxide (N_2O), and its interactions with acidic trace gases are key drivers of nucleation and secondary aerosol formation in the upper troposphere. Despite this relevance, a unified mechanistic understanding of how proton transfer influences NH_3 uptake on ice—particularly the role of hydrogen bonding and the formation of solvation shells at the air–ice interface—remains lacking. Moreover, the behavior of alkaline trace gases such as NH_3 at this interface has received little attention.

Co-adsorption studies at atmospherically relevant temperatures are scarce and have largely focused on acidic gases. Carbon dioxide (CO_2), for example, exhibits significant uptake on ice, which increases strongly as temperatures approach the melting point due to enhanced interfacial disorder. Understanding CO_2 uptake on ice in the presence of NH_3 is therefore essential for assessing the potential contribution of snow and ice to the carbon cycle.

In atmospheric chemistry, the partially liquid-like nature of the air–ice interface has prompted extensive debate, particularly regarding the validity of describing interfacial reactions using aqueous-phase kinetics and mechanisms. Because hydrogen-bond strength strongly influences interfacial chemical interactions, this aspect will also be examined in the context of the relevant adsorbates.

This thesis aims to deliver a molecular-level understanding that complements ongoing experimental work by our collaborators Thorsten Bartels-Rausch and Markus Ammann at PSI within the IRP project CETIMPAI. Using a combination of classical and quantum mechanical simulation approaches (CMD, AIMD, QM/MM, etc.) in collaboration with Ivan Gladich (DiSPeA, University of Urbino), the project will examine the co-adsorption of ammonia at the air–ice interface, resolve its specific interactions with dangling OH groups, and provide a detailed molecular description of interfacial structure and dynamics. Depending on the progress of the research, the development or application of machine-learning potentials may also be explored.

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Remarques/commentaires supplémentaires :