ÉCOLE DOCTORALE GALILÉE
Université Sorbonne Paris Nord

Contrat doctoral – ED Galilée

Titre du sujet : Mechanistic insights into water-mediated pre-nucleation of aerosols

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- Discipline : Physique
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- Domaine de recherche : Physico-chimie de l'atmosphère.
- Mots clés : Nucleation, aerosols, action spectroscopy, mass spectrometry, molecular structure, Nuclear magnetic resonance.

Context: Aerosol particles – solid and liquid particles suspended in the atmosphere – significantly influence both climate and human health. The latest Intergovernmental Panel on Climate Change assessment report indicates that aerosol particles are responsible for the largest uncertainties in current climate models. This uncertainty is mainly due to the limited knowledge about the early stage of New Particle Formation, where gas-phase molecules form clusters of about 2 nm in diameter[1]. To improve models used to assess the environmental and climate impacts of aerosols, it is imperative to understand how a few acid and base molecules interact with water as a mediator. It is crucial to precisely quantify the chemical composition of the critical nucleus, beyond which aerosol growth occurs spontaneously.

[1] Tröstl et al. Nature, 533, 527 (2016).

Objectives: The objective of this thesis is to provide a molecular-level understanding of the early stages of nucleation, from a few molecules in the gas phase to the formation of a critical nucleus. Particular attention will be paid to the role of water during these early growth stages. State-of-the-art spectroscopic techniques, combined with quantum chemical calculations, will be used to determine the structures of these clusters. Hydrated clusters of organic acids and amines will be generated using an innovative source developed by the Laser Physics Laboratory's Biomolecules and Spectroscopy group[2]; their structures will subsequently be probed by action spectroscopy in the gas phase. The obtained vibrational spectra will be compared with quantum chemical calculations performed on the MAGI high-performance computing facility. This structural study will also be conducted in collaboration with Debora Scuderi at the Institute of Physical Chemistry, utilizing the CLIO (Centre Laser Infra-rouge d'Orsay) free-electron laser and ultra-high-resolution mass spectrometry. This will provide insights into clusters obtained via electrospray ionization—another gas-phase transfer technique that is likely to lead to slightly different structures. In parallel, liquid-phase structural studies will be performed via Nuclear Magnetic Resonance at the Bioorganic, Biophysics, Biomaterials Chemistry for Health laboratory (CB3S). Real-time monitoring of the growth from isolated molecules to dimers, trimers, and then pre-nucleation clusters of a few nanometers will be performed using DOSY (Diffusion Ordered Spectroscopy). The main precursors studied will be organic acids (dicarboxylic acids such as oxalic, malonic and succinic acids) and amines.

[2]Badri et al. Rev. Sci. Instrum. 96, 065206 (2025).