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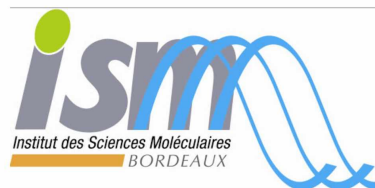
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Experimental and theoretical study on the capture/desorption of gaseous methyl iodide on sea salt aerosols

Hanaa Houjeij^{1,2}, Maxime Infuso^{3,4}, Anne-Cécile Gregoire², Gwenaelle Le Bourdon¹, Laurent Cantrel², Sonia Taamalli³, Florent Louis³, Denis Duflot⁴, Céline Toubin⁴ and Sophie Sobanska¹

¹*Institut des Sciences Moléculaires, Université de Bordeaux, 33405, Talence, France*

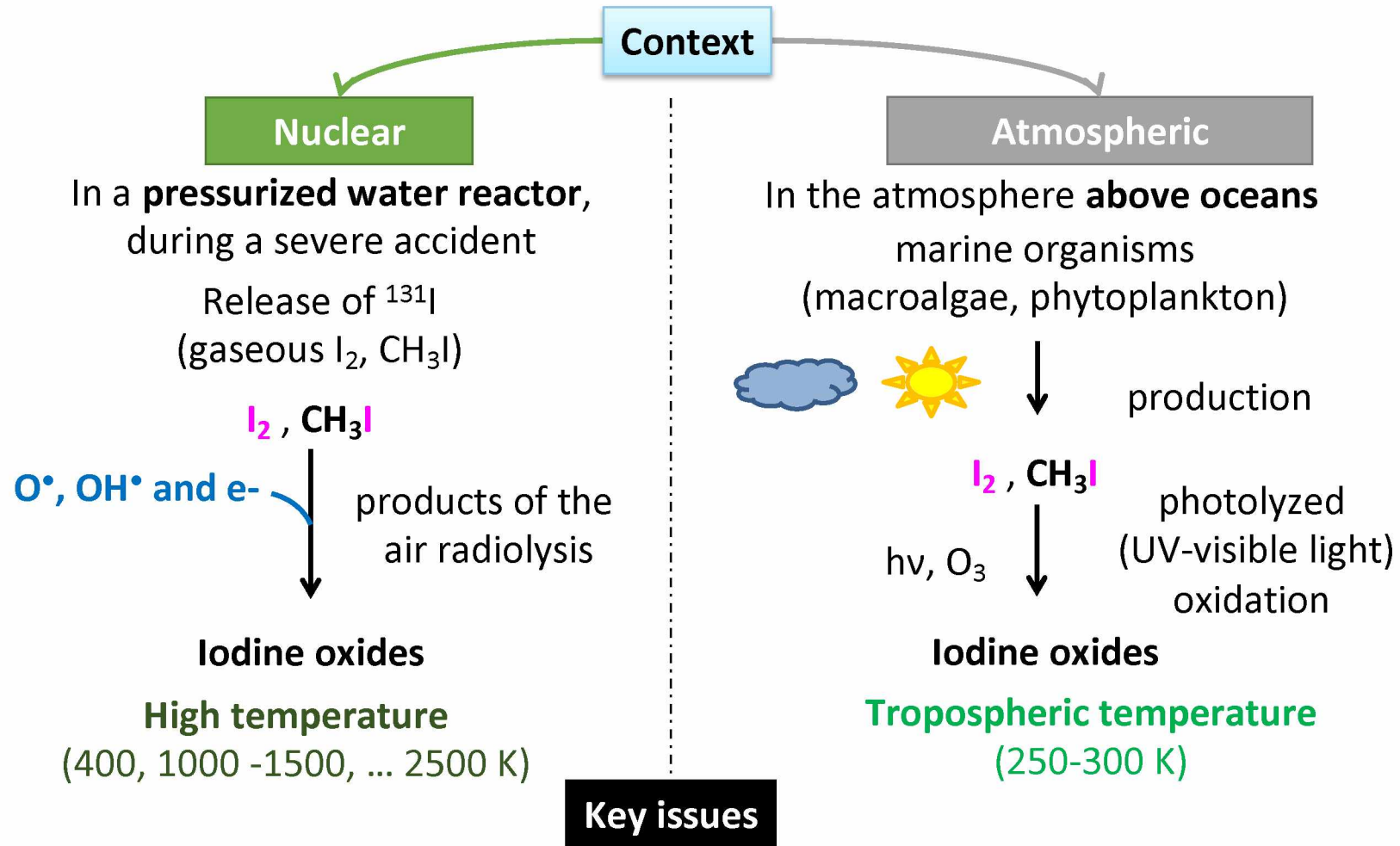
²*Institut de Radioprotection et de Sûreté Nucléaire, PSN-RES, 13115, Saint Paul Lez Durance cedex, France*

³*Laboratoire PC2A, Université de Lille, 55655 Villeneuve d'Ascq Cedex, France*

⁴*Laboratoire PhLAM, Université de Lille, 55655 Villeneuve d'Ascq Cedex, France*

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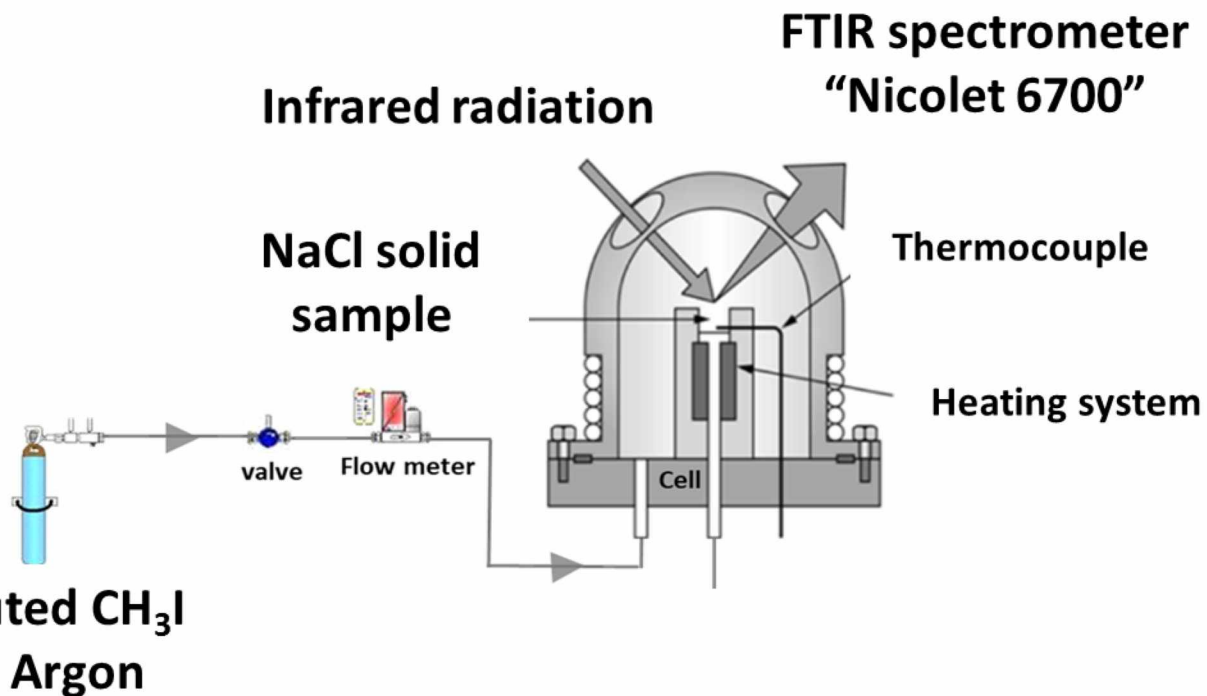
- Labex CaPPA (ANR-11-LABX-0005-01)
- PIA MiRE (ANR-11-RSNR-0013-01)
- Computational ressources: CRI Lille, Slovakia, and CRIANN
- I-SITE ULNE project OVERSEE (contract ANR-16-IDEX-004)



- ✓ How the methyl iodide **reactivity** is influenced by its environment ?
 - **Reactions** with atmospheric aerosols ?
 - **Influence** of the presence of H_2O ?

Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFT) experiments

- ✓ To probe Chemical and structural information for solid surfaces exposed to gaseous species
- ✓ Provide semi quantitative analyses for kinetics, adsorption-absorption and surface reaction
- ✓ Reproduce atmospheric conditions (P, T)
- ✓ FTIR spectra reprocessing in Pseudo absorbance ($\log(1/R)$)



Experimental conditions

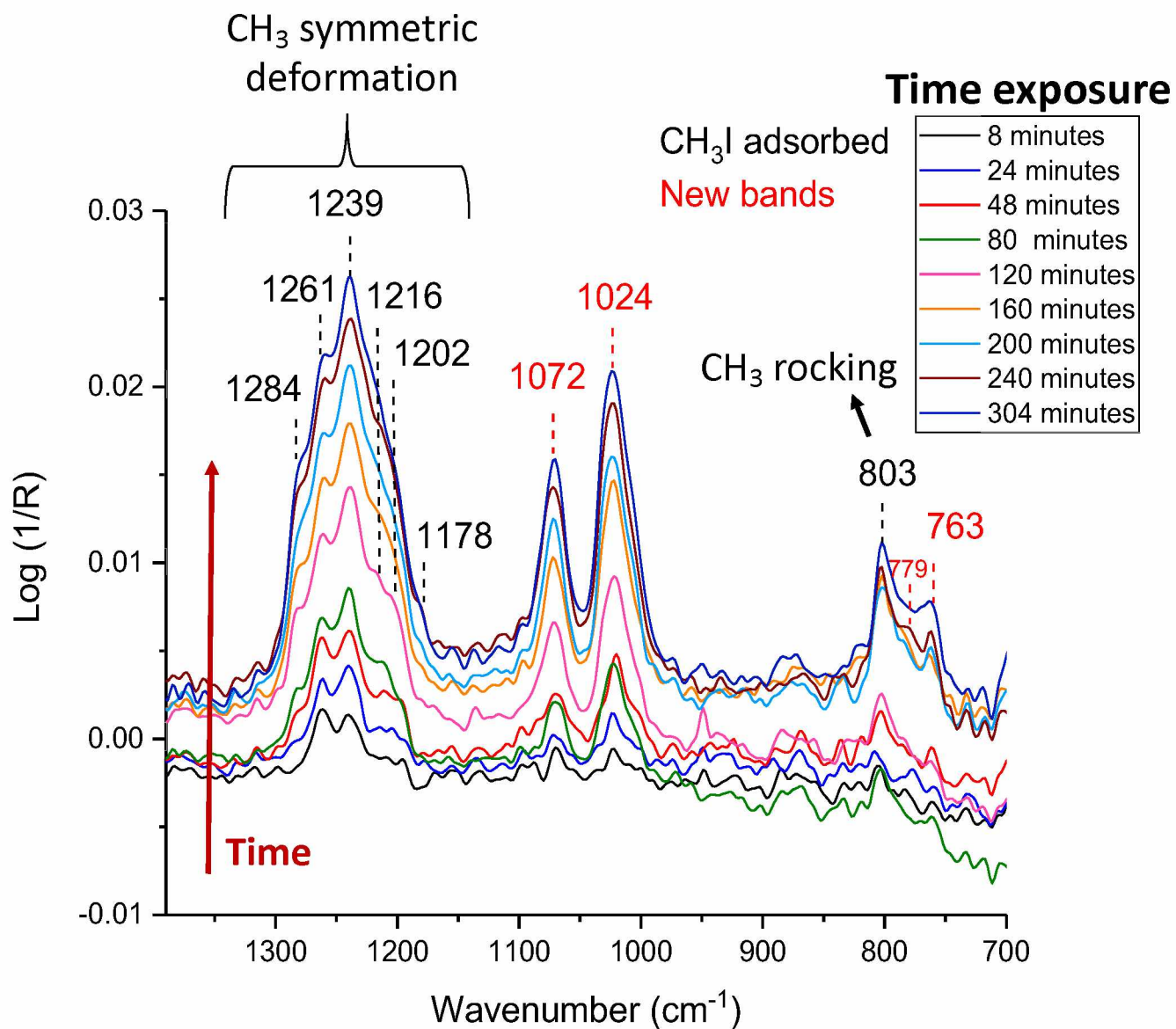
Salt particles	NaCl (powder)
[CH ₃ I]	1000 ppm
Total flow	100 ml/min
Temperature	Room Temperature
Humidity	~10% at 20°C
Pressure	100 KPa
Total Exposure time	5 hours

Spectral recording conditions

Spectral range	4000 to 700 cm ⁻¹
Resolution	8 cm ⁻¹
Scans	200
Time for each spectra	8 minutes

The C-I stretching band at 572 cm⁻¹ is not accessible, because the spectra is cut in the range below 700 cm⁻¹.

Results: DRIFTS Spectra of NaCl when exposed to CH₃I



- ✓ CH₃I adsorbed increases as a function of time.
- ✓ New IR bands are observed due to CH₃I interaction with NaCl - new product formation are observed.
- ✓ No apparent desorption of CH₃I and new bands are observed when the temperature increased or under Ar flow.
- ✓ The kinetic for interaction between CH₃I and NaCl follows a 1st order law.

Theoretical study of the CH₃I microhydration

Objective

Quantify CH₃I interaction with water

→ A first step to understand its chemistry

Methods

Quantum Mechanical calculations
(Optimization+ Normal modes analysis)

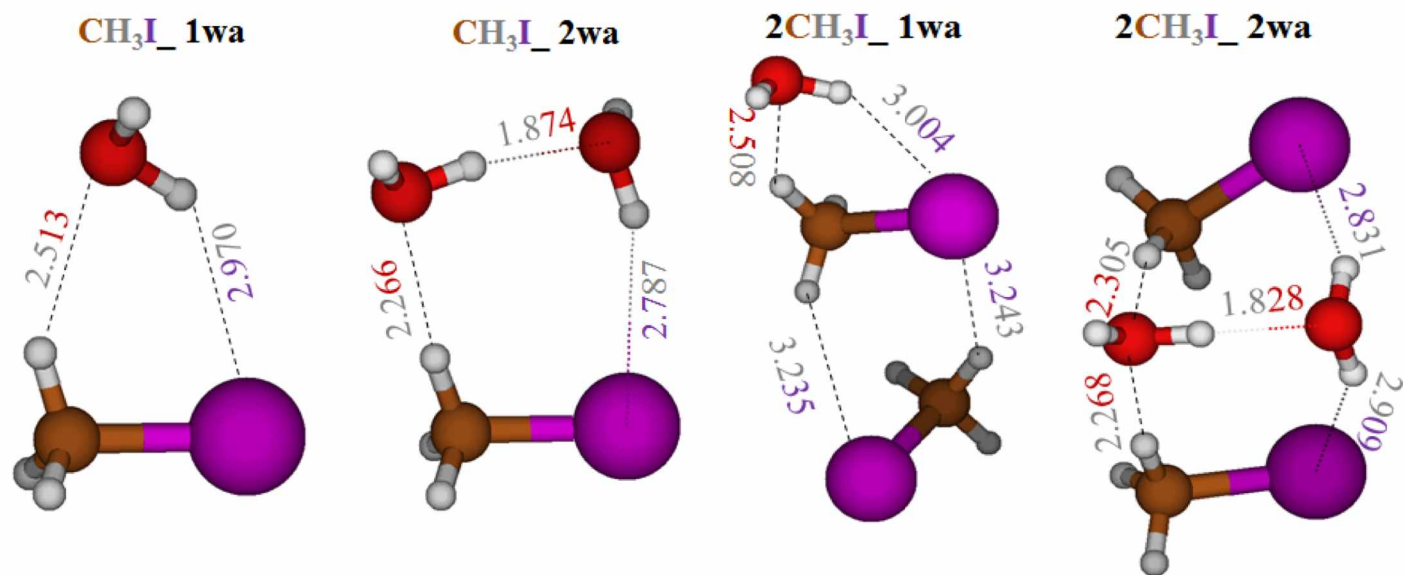
→ ωB97X-D/aug-cc-pVTZ(-PP) level of theory

→ Standard reaction enthalpy ($\Delta_r H_{298K}^\circ$)

→ Standard Gibbs free reaction energy ($\Delta_r G_{298K}^\circ$)

Results

Weak interaction, in agreement with *Ito et. al.*¹



Molecular complex	$\Delta_r H_{298K}^\circ / \text{kJ mol}^{-1}$	$\Delta_r G_{298K}^\circ / \text{kJ mol}^{-1}$
CH ₃ I + 1 H ₂ O	-8.1	14.8
CH ₃ I + 2 H ₂ O	-33.1	25.5
2 CH ₃ I + 1 H ₂ O	-14.1	29.9
2 CH ₃ I + 2 H ₂ O	-53.3	43.5

Ongoing: Interaction of CH₃I with a model sea-salt particle

Objective

How CH₃I interacts with NaCl (001) ?
Influence of humidity?

- Support/Explain results from experiment

Methods

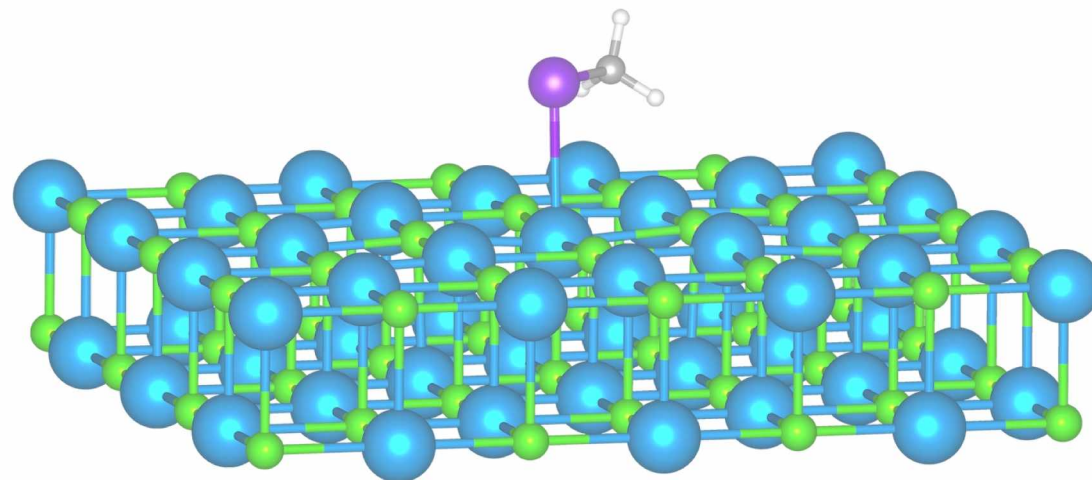
Quantum Mechanical calculations
(Optimization+ Normal modes analysis)

- QM/QM' ONIOM (DFT/PM7) (Gaussian)
- Periodic plane-wave DFT calculations (Quantum Espresso)

Results

- Benchmark of the gas phase IR spectra
- CH₃I/NaCl interaction competes with CH₃I-H₂O interaction

Toubin et al, EGU-2020



Optimized DFT geometry of CH₃I adsorbed on a NaCl(100) surface from a QM/QM' ONIOM (DFT/PM7) calculation

Expectations

- AIMD to unravel the reactivity on the surface (diffusion, coverage influence)
- Effect of H₂O on the reactivity/vibrational frequencies
- Other molecules will be also studied (CH₂I₂...)