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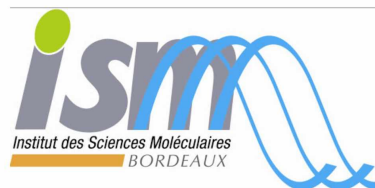
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# Experimental and theoretical study on the capture/desorption of gaseous methyl iodide on sea salt aerosols

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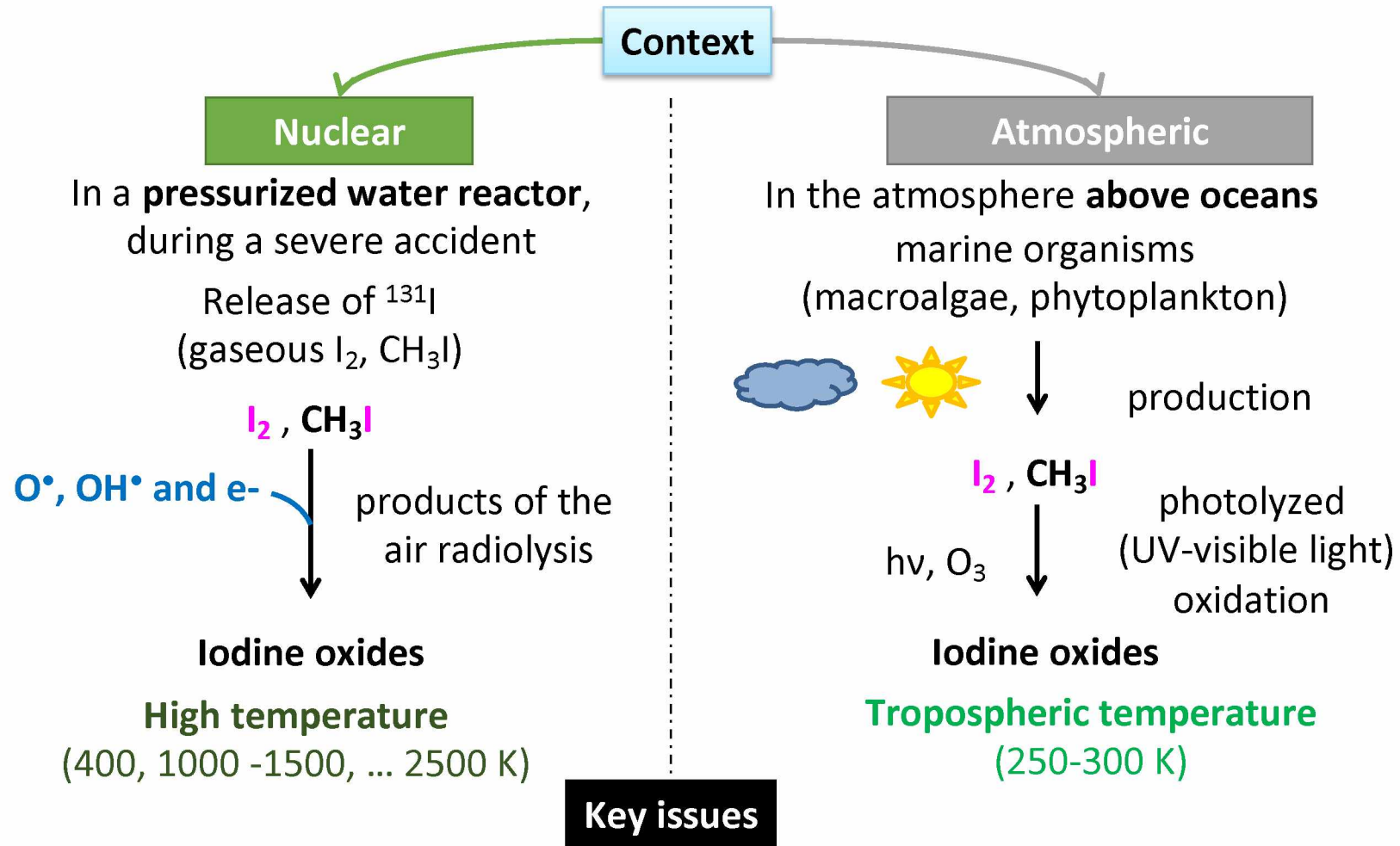
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## Acknowledgments

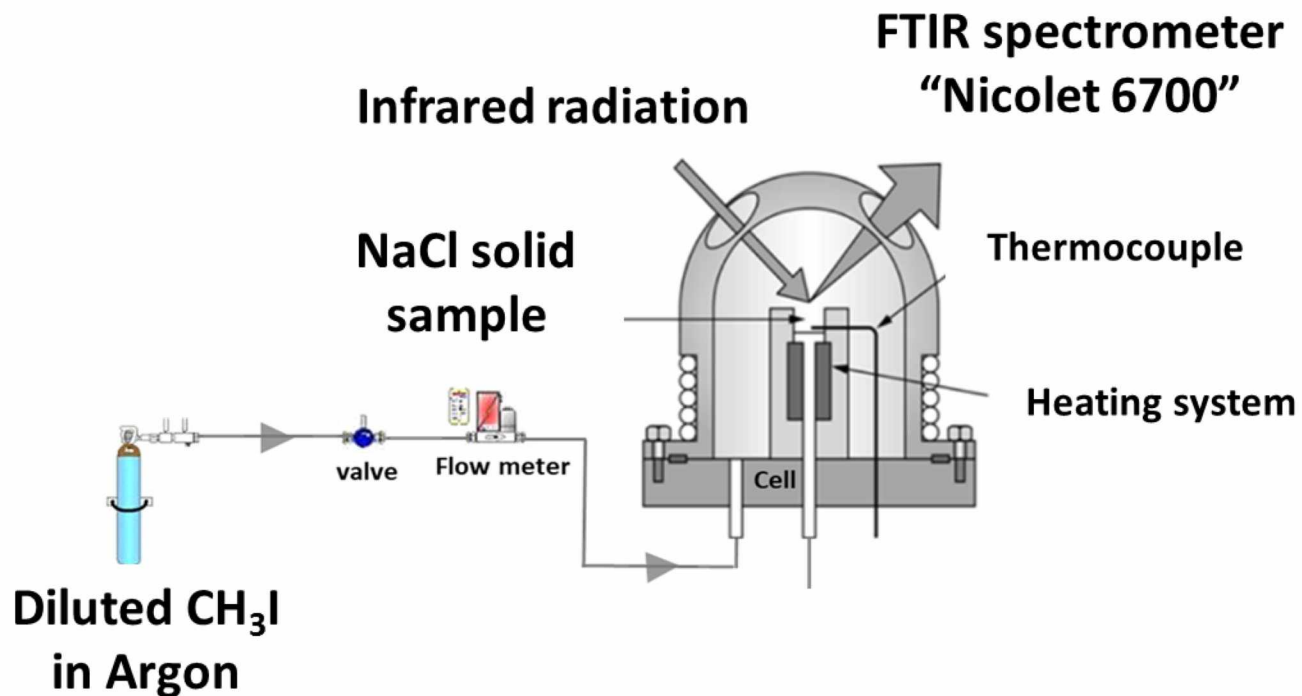
- Labex CaPPA (ANR-11-LABX-0005-01)
- PIA MiRE (ANR-11-RSNR-0013-01)
- Computational ressources: CRI Lille, Slovakia, and CRIANN
- I-SITE ULNE project OVERSEE (contract ANR-16-IDEX-004)



- ✓ How the methyl iodide **reactivity** is influenced by its environment ?
  - **Reactions** with atmospheric aerosols ?
  - **Influence** of the presence of  $\text{H}_2\text{O}$  ?

# 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

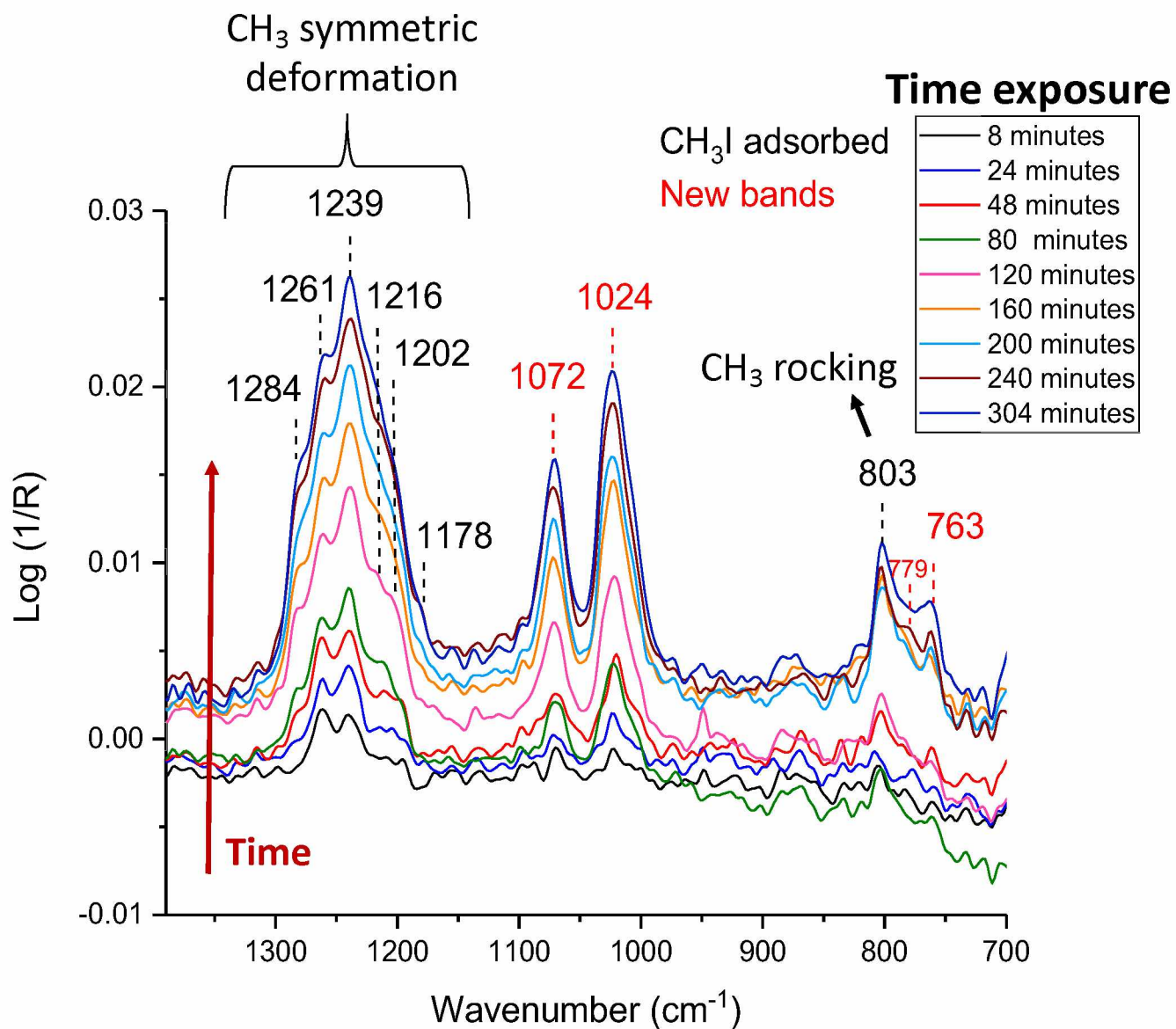
Salt particles	NaCl (powder)
[CH <sub>3</sub> I]	1000 ppm
Total flow	100 ml/min
Temperature	Room Temperature
Humidity	~10% at 20°C
Pressure	100 KPa
Total Exposure time	5 hours

## Spectral recording conditions

Spectral range	4000 to 700 cm <sup>-1</sup>
Resolution	8 cm <sup>-1</sup>
Scans	200
Time for each spectra	8 minutes

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

# Results: DRIFTS Spectra of NaCl when exposed to CH<sub>3</sub>I



- ✓ CH<sub>3</sub>I adsorbed increases as a function of time.
- ✓ New IR bands are observed due to CH<sub>3</sub>I interaction with NaCl - new product formation are observed.
- ✓ No apparent desorption of CH<sub>3</sub>I and new bands are observed when the temperature increased or under Ar flow.
- ✓ The kinetic for interaction between CH<sub>3</sub>I and NaCl follows a 1<sup>st</sup> order law.



# Theoretical study of the CH<sub>3</sub>I microhydration

## Objective

Quantify CH<sub>3</sub>I interaction with water

→ A first step to understand its chemistry

## Methods

Quantum Mechanical calculations  
(Optimization+ Normal modes analysis)

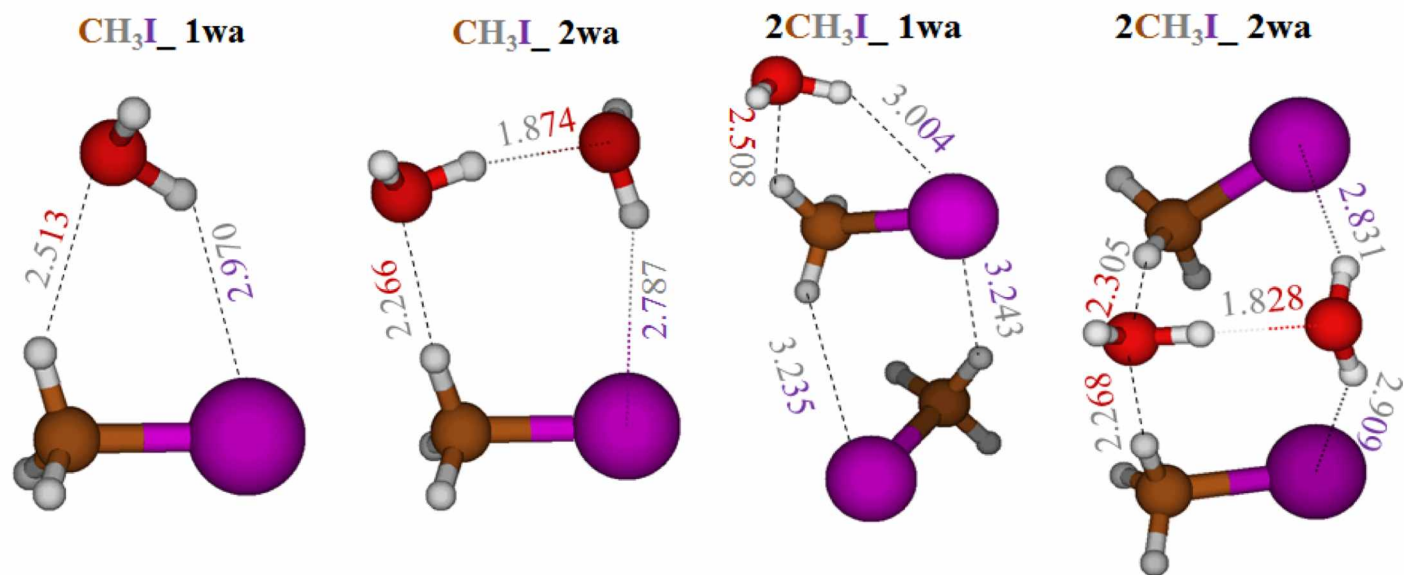
→ ωB97X-D/aug-cc-pVTZ(-PP) level of theory

→ 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.*<sup>1</sup>



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

# Ongoing: Interaction of CH<sub>3</sub>I with a model sea-salt particle

## Objective

How CH<sub>3</sub>I interacts with NaCl (001) ?  
Influence of humidity?

- Support/Explain results from experiment

## Methods

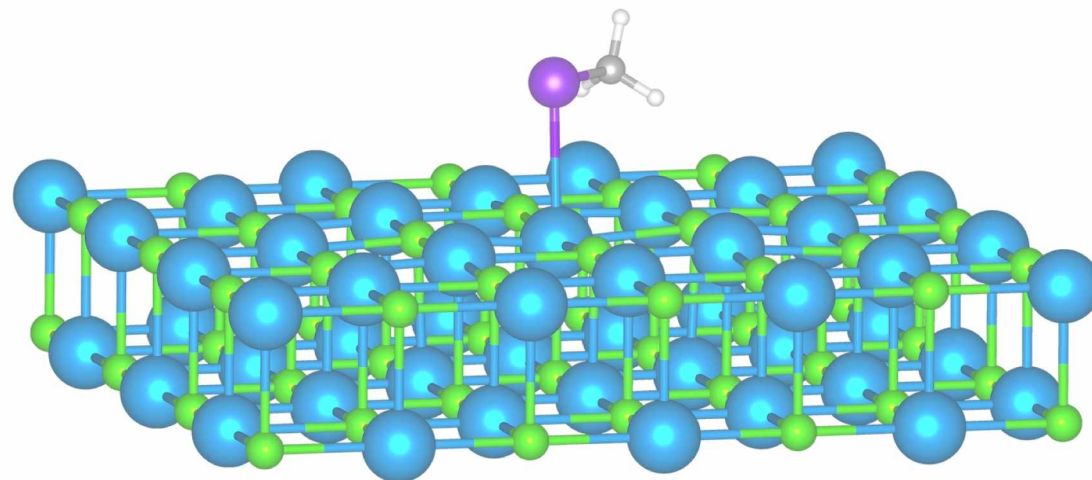
Quantum Mechanical calculations  
(Optimization+ Normal modes analysis)

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

## Results

- Benchmark of the gas phase IR spectra
- CH<sub>3</sub>I/NaCl interaction competes with CH<sub>3</sub>I-H<sub>2</sub>O interaction

Toubin et al, EGU-2020



*Optimized DFT geometry of CH<sub>3</sub>I 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 H<sub>2</sub>O on the reactivity/vibrational frequencies
- Other molecules will be also studied (CH<sub>2</sub>I<sub>2</sub>...)