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

Maxime Infuso, Hanaa Houjeij, Céline Toubin, Anne Cecile Gregoire, Gwenaelle Le Bourdon, Laurent Cantrel, Sonia Taamalli, Florent Louis, Denis Duflot, Sophie Sobanska

► To cite this version:

Maxime Infuso, Hanaa Houjeij, Céline Toubin, Anne Cecile Gregoire, Gwenaelle Le Bourdon, et al.. Experimental and theoretical study on the capture/desorption of gaseous methyl iodide on sea salt aerosols. European Geoscience Union - General Assembly EGU 2020, May 2020, EN LIGNE, France. hal-03107892

HAL Id: hal-03107892

<https://hal.science/hal-03107892>

Submitted on 12 Jan 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Copyright

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, Gwenaëlle 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*

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 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...)