Atelier commun GDR EMIE – GDR SpecMo

«Physico-chimie et caractérisation des atmosphères planétaires »

Villa Clythia – Frejus

25-27 mai 2021

Programme et recueil des résumés

Programme

Mardi 25 Mai		
15:00	Accueil des participants	S. Sobanska/ R. Georges/ H. Tran
15 :10 (15 min)	Evolution récente des contours du GDR EMIE	P. Carçabal /A. Simon
15:25 (40+10)	Conférence inaugurale	Michel Viso (CNES, Paris) *présentiel*
	Break 5min	
Thème Exoplanètes		
16:20 (30+10)	Conférence invitée	Franck Selsis (LAB, Bordeaux) *distanciel*
17:00 (12+3)	Contribution orale	ARFAUX
17:15 (12+3)	Contribution orale	**DUDAS**
Thème Terre		
17:30 -18:10 (30 + 10)	Conférence invitée *"Apport de la modélisation à l'échelle moléculaire pour la compréhension de processus physico chimiques d'intérêt atmosphérique"*	Céline Toubin (PhLAM, Lille) *distanciel*
18 :10 (12+3)	Contribution orale	**GUIMPIER**
18 :25 (12+3)	Contribution orale	GHYSELS
18 :40 (12+3)	Contribution orale	BONHOMMEAU
19:00	Apéritif	
Mercredi 26 Mai		
Thème Système solaire		
9:00 (30+10)	Conférence invitée *"Photochemical hazes in planetary atmospheres: from the solar system to exoplanets"*	Panayotis Lavvas (GSMA, Reims) *distanciel*
9:40 (12+3)	Contribution orale	**COUSIN**
9 :55 (12+3)	Contribution orale	**THIAM**
10:10 – 10:40 (30')	Pause-café	
Thème Terre		
10:40 (30+10)	Conférence invitée *"Chimie photosensibilisée dans les aerosols troposphériques : mythe ou réalité ?"*	Christian George (IRCELYON, Lyon) *présentiel*
11:20 (12+3)	Contribution orale	**COUSSAN**
11:35 (12+3)	Contribution orale	**BECOTE**
11:50 (12+3)	Contribution orale	CHIN
12:00 – 13:30 (1h30)	Repas	
Thème Base de données		
13:30 (30')	Conférence invitée *"Pôle de données et de service AERIS : des données et des outils diverses pour l'analyse des observations dans l'atmosphère terrestre"*	Sébastien Payan (LATMOS, Sorbonne Université) *présentiel*
14 :00-14 :15	Contribution orale	REY
14 :15-14 :30	Contribution orale	**DELAHAYE**
14 :30-14 :45	Contribution orale	**HEARNE**
14 :45-15 :00	Contribution orale	**CHEN**

15:00 – 15:45 (45’) Table ronde	Table ronde *“Bases de données”*	Animateurs : Sébastien Payan et Michaël Rey
16 :15-17 :30	Réunion EMIE-LUMOS	
Jeudi 27 Mai		
Thème Terre		
9:00 (30+10)	Conférence invitée *“Surveillance de la composition atmosphérique depuis l’espace : des émissions de polluants aux pics de pollution”*	Gaëlle Dufour (LISA, Paris-Est Créteil) *distanciel*
9:40 (12+3)	Contribution orale	MIFFRE
9:55 (12+3)	Contribution orale	**TROITSYNA**
10:10 – 10 : 40 (30 ‘)	Table ronde *« méthodologies »*	**Animateurs**
10:40 – 11:00 (20’)	Pause-café	
Thème Système solaire		
11 :00 (30+10)	Conférence invitée “*Latest news from Mars: How did high spectral resolution change our understanding”*	Séverine Robert (BIRA-IASB & UCLouvain) *présentiel*
11:40 (12+3)	Contribution orale	**DESMEDT**
11:55 (12+3)	Contribution orale	**ESTEVES**
12:10 (12+3)	Contribution orale	**COUSTENIS**
12 :25	Conclusion	P. Carçabal / A. Simon
12 :40	Repas	

Recueil des résumés

Etude des brumes dans les atmosphères exoplanétaires: effets de déséquilibre chimique et rétroaction des brumes

Anthony ARFAUX[a], Panayotis LAVVAS[a]

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Les brumes photochimiques sont attendues dans les atmosphères exoplanétaire allant de simples traces presque indétectables à d'importantes quantités capables de modifier fondamentalement leur environnement. L'étude de ces brumes nécessite la prise en compte de multiples phénomènes afin d'obtenir une vision consistante de leurs propriétés. Un de ces phénomènes d'importance majeur est le déséquilibre chimique due notamment au transport et à l'interaction entre les espèces atmosphériques et les radiations provenant de l'étoile. Nous nous intéressons à ce phénomène en comparant les résultats obtenus pour quelques exoplanètes avec, dans un premier temps, un modèle thermochimique et microphysique permettant une détermination préliminaire des paramètres caractérisant les brumes puis, dans un deuxième temps, l'utilisation d'un modèle de déséquilibre chimique se basant sur les paramètres précédemment obtenus. Les résultats montrent, de manière générale, de plus importantes concentrations chimiques entre 1e-2 et 1e-6 bar dues au transport et plus faibles au-dessus de l'altitude de 1e-6 bar dues à la destruction des molécules par les radiations. Cependant, compte tenu de la faible résolution des observations, l'influence du déséquilibre chimique reste indétectable quant à l'ajustement des spectres. La composition chimique de l'atmosphère, ainsi que les brumes, jouent un rôle important dans la détermination du profile thermique qui, à son tour, influence les réactions chimiques et la microphysique des brumes pouvant drastiquement modifier leur répartition, autant en altitude qu'en taille. Cette modification des distributions chimiques et microphysiques implique une modification du champ de radiation qui détermine le profile thermique. L'impact de ces effets rétroactifs entre les constituants atmosphériques et la température sont également étudiés via un modèle couplant le calcul des profiles thermiques, microphysiques et chimiques. La présence de brumes tend à chauffer la partie supérieure de l'atmosphère, faisant ainsi varier la composition atmosphérique et les caractéristiques des brumes. Un autre effet important est la dilatation de l'atmosphère due au chauffage par les particules qui joue un grand rôle dans le spectre de transit. Aux vues de ces résultats, l'utilisation d'un modèle auto-consistent devient une nécessité dans l'étude et la caractérisation des brumes.

Evaporation process of linalool droplet at high relative humidity using acoustic levitation

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In the atmosphere, secondary organic aerosols (SOAs) are well known to impact both air quality and climate change. During their transport, SOAs may react through aging multiphasic processes. These reactions may affect both chemical composition and physical properties of aerosols such as surface tension, hygroscopicity, viscosity and gas-particle partitioning. In the laboratory, the particle aging is usually investigated using two experimental approaches: (i) atmospheric simulation chambers and (ii) flow reactors. However, the complexity of aerosol aging processes occurring at the gas/particle interface is not well described. Recent developments using single particle approaches allow studying various chemical aging processes and microphysical properties at the particle scale[1-4]. In this context, the objective of the present work was to characterize the gas-phase partitioning of individual SOA-like particles. The evaporation behavior of aqueous-diluted linalool single droplet was investigated under high relative humidity using an innovative experimental set-up coupling an acoustic levitation cell with an optical camera and a proton-transfer-reaction mass spectrometer (PTR-MS).

The influence of initial linalool concentration (up to the solubility limit), initial droplet size and RH variation (80-98 %) have been investigated by following size-particle changes (d^2/d_0^2) and the gas-phase composition within the time. Experimental results have been compared to the theoretical "d^2 law" approach[5,6]. The gas-phase composition measured by PTR-MS during the evaporation process has been related to the results obtained from the size variations, bringing new highlights in particle behavior at high relative humidity.

Acknowledgements

Authors thank the University of Bordeaux, ADEME and CNRS-INSU LEFE-CHAT program, for financial support.

References

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Benefit of Molecular Dynamics simulations for investigating physical properties of fluids: Applications to Venus atmosphere and Titan lakes

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Molecular dynamics simulations based on classical force fields are common tools to investigate the properties of biochemical complexes but are also choice approaches to evaluate physical properties of fluids of planetological interest. We present here two recent applications of molecular dynamics to planetology: (i) the modeling of CO_2 and N_2 diffusion within the hellish conditions of Venus atmosphere ($p \approx 90$ bar and $T \approx 700$ K), as a possible explanation for the formation of a chemical gradient of N_2 observed by the probe *Vega-2* [1], and (ii) the estimation of the physical properties (eg, diffusion coefficients and heat capacities) of binary and ternary mixtures involving CH_4, C_2H_6, and N_2 at low pressures ($p \leq 10$ bar) and cryogenic temperatures specific to Titan lakes ($T \approx 90$ K) within the context of the characterization of Titan's alkanofers [2]. We will discuss the relevance of the NVT and NpT thermodynamic ensembles to tackle these problems, present fluctuation formulas of practical use to evaluate some physical properties of fluids or show how some unknown properties like densities of mixtures can be guessed, and eventually conclude on future works these studies call for.

ACKNOWLEDGMENTS

The ROMEO supercomputer center of Champagne-Ardenne and the CINES national supercomputer center (project A0090811976) are gratefully acknowledged for computing resources and technical support.

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Simulation of the hydration of organic compounds relevant to atmospheric aerosols

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Atmospheric aerosols have a strong impact on the climate and on human health: they can absorb or reflect solar radiations; they play an important role in cloud formation; they contribute to the formation of particle matter which badly affect human health. Carboxylic acids, especially glyoxylic acid and pyruvic acid, are very abundant in marine aerosols. In the aqueous phase, glyoxylic and pyruvic acids adopt two forms in equilibrium: The keto form with two carbonyl groups and the gem-diol form that bears two hydroxyl groups on the same carbon. While the gem-diol form is transparent to solar radiation, the keto moiety is light sensitive, thereby contributing through its photochemistry to the production of larger compounds involved in the formation of secondary organic aerosols.
By means of ab-initio simulations well suited to the observation of rare events, our theoretical study on the keto/gem-diol equilibrium sheds light on the hydration mechanism of glyoxylic and pyruvic acids. Our investigation also provides a detailed description of the solvation sites and of their conformational landscape in solution [1].

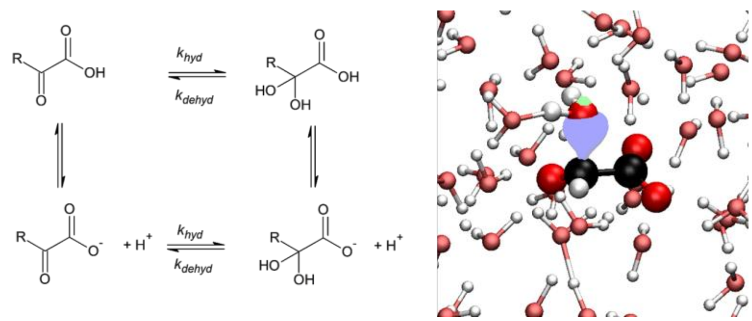

Figure 1 Left: Scheme of the ketone/geminal-diol equilibrium of the R-CO-COOH acid in solution (R=H: glyoxylic acid, R=CH_3: pyruvic acid). Right: Typical configuration for the transition state of glyoxylic acid showing a water lone electron pair (blue and green isosurfaces of a maximally localized Wannier function) interacting with the α-carbon.

ACKNOWLEDGMENTS

This work was supported by the LabEx PALM (ANR-10-LABX-0039-PALM) and by the ANR DYNAAMIX project (ANR-17-CE29-0001-01). This work was granted access to the HPC resources of CINES under allocation DARI-A0060810750 (2019) attributed by GENCI.

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H_2O line shape parameters broadened by CO_2: from the lab to space

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For decades, the composition of planetary atmospheres has been inferred from remote sensing measurements. In the frame of the scientific preparation of the candidate mission to Venus, EnVision with onboard 3 spectrometers, known as the VenSpec suite, probing from the surface to above the cloud decks of Venus, new spectroscopic measurements and calculations are required for H_2O that is one of the target species of the mission. VenSpec-H is designed with a high-resolution spectrometer (R~10000) and will observe the night side of Venus in nadir, spanning the spectral ranges of the transparency windows between 1 and 2.5 microns and therefore enabling the spectral retrievals of various species between 10 and 40 km altitude. Considering the high pressure in the lower atmosphere of Venus and the high abundance of CO_2, line-shape parameters of H_2O broadened by CO_2 are required to infer the H_2O abundances with accuracy.

In 2019, Régalia et al. [Régalia et al, JQSRT 231,126-135 (2019)] published new experimental line-shape parameters of H_2O broadened by CO_2 around 2.7 and 6 µm and Modified Complex Robert-Bonamy (MCRB) calculations of the half-width, line shift, and their temperature dependence in the spectral region from 1300 to 5000 cm^{-1}. In line with the spectral ranges probed by VenSpec-H, new spectra were recorded in the spectral region of 1.88 µm, for which MCRB calculations were performed.

We will present the recently measured spectra at 1.88 µm, the line-shape parameters obtained using a multispectrum fitting procedure and the comparison with the MCRB calculated values. The impact of the line parameters on the Venus' spectra will be assessed.

Can cryogenics be a tool for understanding planetary atmospheres?

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Complexity arises from a chain of extremely simple events. Although we are now able to understand this complexity very imperfectly, particularly that of planetary atmospheres, starting with that of the Earth, we are still very ignorant of all the details of the chemical or physico-chemical reactions that take place within these environments. One of the methods of choice for studying these simple events is cryogenics. Indeed, this science, represented by the trapping in cryogenic matrix of monomers and small polymers, aggregates and complexes, but also by the study of amorphous and crystalline ices, can provide precious information on the very first stages of hydration. The behaviour of these hydrated species under UV-Visible and IR irradiation, three of the main types of radiation reaching the Earth's atmosphere, can then be studied over long timescales (from seconds to days). It is also possible to trace the photoreaction pathways and consequently to compare these results with those obtained from theoretical calculations. These molecules, aggregates and complexes, are trapped in a cryogenic matrix (they are therefore sensitive to this environment), or adsorbed on ice, notably water, and thus constitute excellent molecular models for the understanding of planetary atmospheres [1,2].

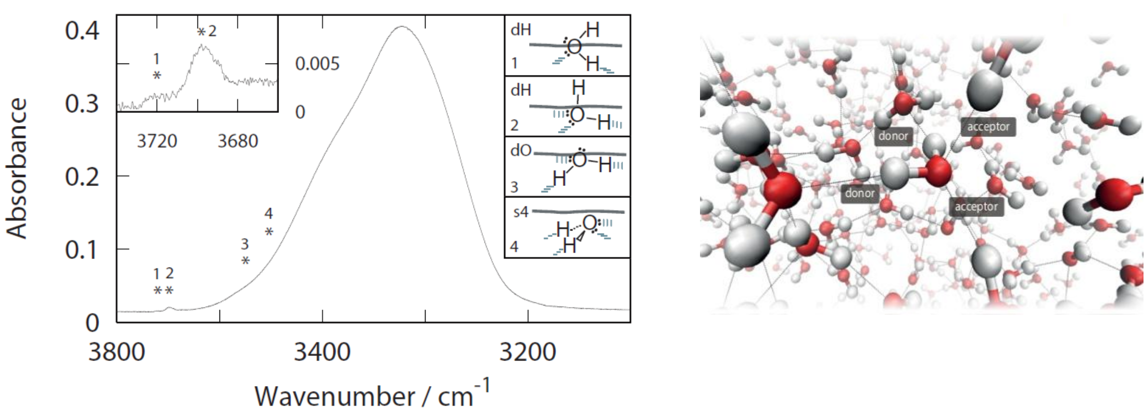

Figure 1 left: OH stretching region of Amorphous Solid Water (ASW); right: 3D view of ASW bulk

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Exploring the tenuous atmospheres of the icy moons

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The atmospheres/exospheres of icy satellites greatly vary from one to the next in terms of density, composition, structure or steadiness [1,2]. Titan is the only satellite with a dense atmosphere comparable in many ways to that of the Earth's in terms of exchanges of energy, matter and momentum. Enceladus' atmosphere manifests itself in large water vapor organic-laden plumes emanating from cracks near the south pole.

The Galilean satellites are known to have thin atmospheres, technically exospheres, produced by ion-induced sputtering and sublimation of the surface materials providing insights on the processes and composition of their surfaces. The O_2 atmosphere at Europa (inferred from UV emissions) is produced by intense radiation bombardment and particles sputtering off the surface, with possible contributions from local H_2O plume sources, while Na and K have also been measured in ground-based data. Callisto's CO_2 atmosphere, first detected by Galileo NIMS, is surprisingly dense in O_2.

Ganymede has a thin O_2 atmosphere, inferred by HST/UV data, and a hydrogen exosphere (from Galileo/UVS). Ganymede is unique due to the presence of an internal magnetic field. Its interaction with Jupiter's magnetosphere (still poorly understood) is powerful enough to create an auroral footprint in Jupiter's atmosphere and an aurora in the exosphere of Ganymede itself. Io's SO_2 atmosphere originates from volcanoes.

ESA's first large Cosmic Vision mission, JUICE, will significantly contribute to our understanding of the atmospheres of the icy satellites, their origin and evolution, as well as the chemical composition of their surfaces, by observing the exospheres of Europa, Callisto and Ganymede through remote monitoring, imaging of the aurora, multi-wavelength limb scans and stellar occultation, and directly by in situ measurements by sensors of the particle packages from low orbits and fly-bys.

On Europa Clipper, the MASPEX instrument for instance will measure trace neutral species, to determine the composition of Europa's atmosphere: major volatiles (such as H_2O, H_2, O_2, and CO_2) and key organic compounds (CH_4, C_2H_2, C_2H_4, C_2H_6, HCN, and CH_3OH) with respect to latitude and longitude, altitude, and local solar time. The data will permit us to distinguish between endogenic and exogenic sources.

I will review the characteristics of the atmospheres of Enceladus, Ganymede and Europa with reference to other larger satellites and discuss future measurements in particular by the JUICE and Europa Clipper missions to the jovian system.

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Evaluation et validation des bases de données spectroscopiques : l'exemple de la mise à jour GEISA-2020

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Le processus de mise à jour des bases de données spectroscopiques afin de fournir aux utilisateurs une base de données qui assure une caractérisation optimale des propriétés spectrales de l'absorption moléculaire est un travail complexe. En s'appuyant sur une solide expérience des activités de calibration/validation au sein du laboratoire, et pour aider à faire les choix les plus pertinents au moment de l'élaboration de la base de données spectroscopiques, la chaîne SPARTE [1] (Spectroscopic Parameters And Radiative Transfer Evaluation) a été développée au LMD, et appliquée pour la première fois à la mise à jour GEISA-2015 [2].
SPARTE repose sur la comparaison entre des simulations de transfert radiatif direct réalisées par l'algorithme 4AOP, utilisant une base de données spectroscopiques donnée en entrée, et diverses observations de télédétection pour plusieurs milliers de situations atmosphériques bien caractérisées. Typiquement, des observations en ciel clair effectuées par deux instruments bien établis sont utilisées pour couvrir une large gamme spectrale : les données de l'instrument IASI (15 à 3,7 µm) et celles des spectromètres à transformée de Fourier (FTS) au sol du réseau TCCON (2,5 à 0,7 µm).
Pour la mise à jour GEISA-2020 [3], la chaîne SPARTE a été utilisée de façon systématique lorsque cela était possible (notamment pour les espèces moléculaires dont la contribution dans les spectres atmosphériques est non négligeable). Nous présentons ici des exemples d'évaluation pour la mise à jour de O_3 à 10 µm, de H_2O entre 1.7 et 1.2 µm et O_2 dans la bande à 1.27 µm.

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Trapped-gas selectivity and metastability of gas hydrates.

Claire Petuya[a], Cyrielle Métais[a,b,c], Ludovic Martin-Gondre[c],
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The natural occurrence of large quantities of gas hydrates in deep oceans, permafrost and plausibly in planets or comets of the solar system, is certainly at the origin of numerous developments and researches in fields ranging from physical-chemistry, geosciences or astrophysics to chemical engineering and innovative technologies. Gas hydrates are ice-like systems made of water molecules forming nano-cages stabilized by the encapsulation of foreign molecules. At a fundamental level, the understanding of their physical-chemistry properties (trapped-gas selectivity, kinetics formation mechanism, etc.) is of prime importance to track the evolution of the abundances of species taking part in the compositions of hydrate-bearing deposits not only on Earth, but also potentially on extraterrestrial bodies. The understanding of their molecular interactions plays also a key role requiring the combination of advanced experimental and theoretical approaches, in which Raman spectroscopy brings unvaluable information: the provision of accurate spectroscopic signatures is crucial for evaluating their potential extraterrestrial detection. The presentation will review recent physico-chemical results (e.g. metastability [1,2,3], molecular selectivity [3,4], etc.) relevant in gas hydrates sciences and associated challenges in astrophysics.

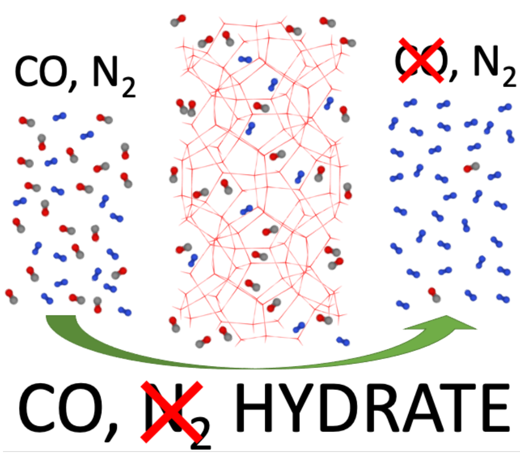

Figure 1 Representation of the CO selective trapping in the mixed CO-N_2 gas hydrate and of the potential sieving role played by the gas hydrate.

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Investigating the atmospheres of Hot Jupiters with the SMAUG experiment

Eszter Dudás[1], Abdessamad Benidar[1], Samir Kassi[2], Michael Rey[3], Vinayak Narayan Kulkarni[4], Christine Charles[5], Robert R. Gamache[6], Robert Georges[1]

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[4]Research School of Physics, Australian, National University, Canberra, ACT, Australia
[5]Indian Institute of Technology Guwahati, Guwahati, India
[6]University of Massachusetts Lowell, Lowell, MA, USA

The SMAUG[1] experimental setup was developed in the frame of the e-PYTHEAS project that focusses on high-temperature spectroscopy of small hydrocarbons. High-temperature infrared spectroscopic data is needed to retrieve temperature and concentration profiles from Hot Jupiter exoplanets. A small dimension Laval nozzle connected to a compact high enthalpy source equipped with cavity ringdown spectroscopy (CRDS) is used to produce high-resolution infrared spectra of polyatomic molecules in the 1.67 μm region. SMAUG can operate according to two complementary working regimes: non-LTE (vibrationally hot and rotationally cold) and LTE conditions, to interpret the complex pattern of highly-excited vibrational states of small hydrocarbons. The results presented are related to methane. A rotational temperature of 39(3) K was measured, while multiple vibrational temperatures corresponding to the different vibrational levels were necessary to reproduce the observed intensities. The results testify for a more rapid vibrational relaxation between the vibrational energy levels constituting a polyad, meanwhile a less efficient vibrational relaxation occurs among the polyads. The spectrum contains a dozen of presumptive candidates for line identification of transitions issuing from the pentad polyad. Supplementary, using a "post-shock CRDS" technique CO and CH4 LTE spectra were recorded at 950 K and 1400 K.

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Clathrate hydrates FTIR spectroscopy: a key in understanding solar system ices

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The formation of clathrate hydrates may play a key role in the stability of gases in many astrophysical bodies, such as comets. They may provide a trapping mechanism responsible for the preservation in the solid state of volatile species at temperatures higher than expected for the pure solid, thus avoiding their early escape. Observations of comet suggest that there is a mechanism still unknown which could explain part of the phenomena linked to cometary activity, in relation to the survival of these volatile molecules [1].

Gas hydrates form and remain stable only in specific temperature and pressure regimes that depend on the nature of the guest molecules [2]. Theoretical phase diagram of clathrate hydrates show that it could be possible to form clathrates at very low pressure (10^{-10} bar) and temperature (< 80 K), but there is a lack of experimental data using these preparation methods [3]. Could clathrate hydrates be formed under such conditions? What if the formation and characterization of these ice-like structures under such conditions could provide valuable experimental evidence for understanding the origin and evolution of comets?

FTIR spectroscopic identification of carbon dioxide and methane hydrates have been performed at low temperature (10 K) and pressure (base pressure 10^{-7} mbar) regimes. In an effort to understand the nature of the gas hydrates formed under these conditions, vibrational spectra of distinct gas/ice interactions (clathrate hydrate, gas in/on water ice) were compared. The behaviour of the water crystalline skeleton interactions with the trapped molecules at different temperatures and the influence of various preparation methods will be presented.

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Water vapor measurements in the tropical upper troposphere and lower stratosphere : regional tape recorder, waves and deep convection

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STRATEOLE 2 is a French-American project based on superpressure balloon-borne measurements to study dynamics and processes in the TTL and the lower stratosphere of equatorial regions. One single flight of these balloons (of a duration of about 80 days) can make several turns of the Earth.
Here we present in situ high resolution water vapor measurements at the top of the TTL or the lower stratosphere by the Pico-SDLA diode laser spectrometer on-board the TTL 2 gondola during the first campaign (Dec. 2019 - Feb. 2020).
In this presentation, we analyse the regional tape recorder signal at a constant altitude during the 80 days of flight. Based on comparison between in situ and satellite observations, we discuss the challenges beyond such analysis. We compute an anomaly of the *in situ* water vapor measurements with respect to a regional/temporal satellite-borne mean climatology from Aura MLS v5. The obtained contrast allows the positioning of observations with respect to local climatology and therefore, the identification of singular events responsible for modulation of the local water vapor content : Kelvin wave, packet of gravity waves and possibly deep convection.

Formation & caracterization of methane hydrates in porous clay/sand media.

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Gas hydrates present a very high gas storage capacity at the origin of a broad research activity in the energy area (potential energy resource), the sustainability area (huge carbon dioxide-sequestration opportunities) and also in the environmental area such as geo-hazards, and potential impact on ocean acidification due to climate change (marine methane injection in the hydrosphere) [1,2]. Natural gas hydrates, composed mainly of methane, are found in marine sediments on continental margins and in the permafrost region. Understanding the role of this natural hydrate deposit in the deep carbon cycle and the consequences of a sustained release of methane from its decomposition due to temperature increase caused by climate change is of prime importance. The ocean seafloor, mainly composed of clay and sand, is reproduced at the lab scale to study the influence of such porous matrix on methane hydrate formation. Raman micro-spectrometry coupled to neutron diffraction and spectroscopy, are used to characterize the structural properties, the composition and the formation kinetics of gas hydrates. These original results provide new insights into the formation mechanism in these complex porous media mimicking the natural environment.

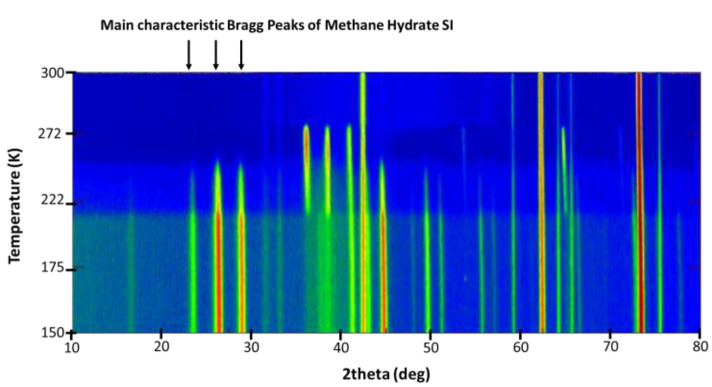

Figure 1 In-situ D20 diffraction (at 2.41Å) of methane hydrate decomposition in montmorillonite/sand sedimentary matrix as function of temperature – from 150K, 1 bar to 300K, 1bar

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Les HEROES de la Spectroscopie Térahertz

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Le domaine térahertz (0.7-10 THz) est une gamme très intéressante pour la caractérisation des objets astrophysiques. Prochainement, les télescopes THz pourront étudier les atmosphères d'exoplanètes avec une sensibilité et résolution spectrale de grande qualité, cependant, les spectres du plusieurs molécules relevant pour l'astrochimie ne sont pas bien connues avec précision dans cette gamme.[1,2] Par exemple, le spectromètre par transformation de Fourier (FTIR) utilisé sur la ligne AILES du centre de rayonnement synchrotron SOLEIL possède une résolution ultime de 30 MHz, alors que la largeur typiques des raies moléculaires sont de l'ordre de quelques MHz dans cette gamme. C'est pourquoi nous avons fabriqué un nouveau spectromètre qui peut mieux exploiter les propriétés de brillance du continuum de l'émission synchrotron dans le projet HEROES.[3] Le principe est d'utiliser un laser moléculaire, qui émet une fréquence THz précise, comme un oscillateur local (LO) pour faire fonctionner un mélangeur (bolomètre a électrons chauds). Le battement hétérodyne entre les deux radiations est détecté dans la gamme radiofréquence (RF), et mesuré précisément (80 kHz de résolution) par un analyseur de spectre RF. Cet instrument permettra la production de bases de données très précises, et pourrait aider la caractérisation des atmosphères d'exoplanètes.

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A controlled-laboratory experiment to quantify the aerosol-radiation interaction through light backscattering

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As underscored in the latest IPCC report [1], atmospheric aerosols contribute to the Earth's climate through aerosols-radiation interactions (i.e. light scattering and extinction) and aerosols-cloud interactions. Both are still nowadays subject to large uncertainties, mainly due to the complexity of these particles, which exhibit a wide range of sizes, shapes and chemical composition. Examples of aerosols-cloud interaction studies have been given in the oral presentation given at GdR Emie last October, where the remote detection of new particle formation events promoted by mineral dust [2] has been presented by insisting on the required spectral and polarimetric sensitivity for such complex remote sensing observations [3].

In line of this previous presentation, we here study the aerosols-radiation interactions. The interaction of atmospheric aerosols with the electromagnetic field is very complex to study as there is no analytical solution of the Maxwell's equations for such complex-shaped particles. While the physical process of light backscattering by atmospheric aerosols is extremely important as responsible for a net cooling of the Earth's climate, it has surprisingly never been observed in laboratory for atmospheric aerosols. We here present a unique controlled-laboratory experiment [4], developed at iLM, that for the first time addresses exact light backscattering by atmospheric aerosols, as briefly schemed in Figure 1. While no aerosol optical database exists as is the case for greenhouse gases, this controlled-laboratory experiment provides precise spectral and polarimetric fingerprints of diverse aerosols such as mineral dust [4], soot particles [5], or core-shell organosulfates [6]. Concerning the latter, in cooperation with chemical colleagues from IRCELYON (C. George's group), we recently revealed a net decrease in light backscattering by the sulfate aerosol in the presence of organic compounds [6]. Since such organosulfates are the most important secondary organic aerosol precursors in the atmosphere, this finding can be key in quantifying the direct radiative forcing of sulfates in the presence of organic compounds.

In line of the oral presentation given at GdR Emie last October, the proposed presentation will first briefly recall published material concerning new particle formation events promoted by mineral or volcanic dust [2, 3] as an introduction. Then, to open the discussion, examples of aerosols-radiation interactions will be given for mineral dust, soot or organosulfates, by quantifying their light backscattering property for the first time in laboratory [4-6].

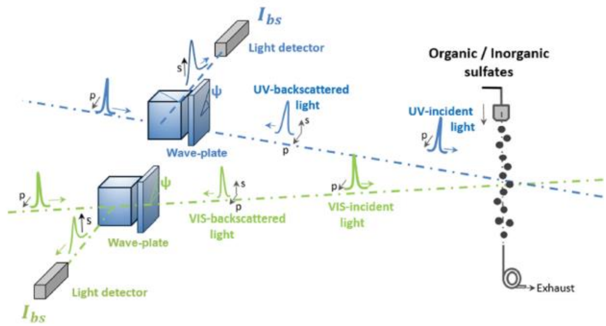

Figure 1 Scheme of our laboratory (UV, VIS) laboratory polarimeter at Pi-backscattering angle (180.0 $\pm$ 0.2°) for particles embedded in ambient air.

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The authors want to thank their chemical colleagues from IRCELYON (C. George's group) for achieved common publications [2,6]. The authors thank GdR Suie for partly funding publication [5], CNRS and Université de Lyon for financial funding.

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Base de données TheoReTS : l'*ab initio* au service de l'atmosphère

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L'étude des propriétés radiatives est d'une importance cruciale pour modéliser correctement les spectres d'émission ou d'absorption de différents systèmes moléculaires. Les demandent croissantes de la communauté atmosphérique/planétologique/astrophysique démontrent à quel point il est nécessaire d'avoir à disposition des listes spectroscopiques fiables, couvrant des plages spectrales très larges et permettant de reproduire les dépendances en température. En particulier, la connaissance précise des intensités des transitions ro-vibrationnelles pour différents systèmes moléculaires (isotopologues inclus) est essentielle pour la modélisation d'atmosphères planétaires. L'information fournie par les bases de données historiques telles que HITRAN ou GEISA est très précise, aussi bien sur les positions que sur les intensités, mais également trop souvent parcellaire et surtout difficilement extrapolable au-delà de 300 K.

C'est dans ce contexte que les calculs *ab initio* avec la construction de surfaces d'énergie potentielle et de moment dipolaire prennent tout leur sens. Avec l'écriture de codes massivement parallélisés et le développement de méthodes d'optimisation poussées, les précisions maintenant atteintes par les calculs variationnels étaient inenvisageables seulement 10 ans en arrière. Aujourd'hui, ils permettent de pallier le manque de complétude dans la plupart des bases actuelles en fournissant toutes les bandes froides et chaudes à une température donnée. De plus, les calculs des intensités pour les transitions les plus fortes sont souvent effectués à la précision expérimentale (quelques %).

Dans cet exposé, la base de données théorique TheoReTS [1] (Theoretical Reims-Tomsk Spectra, http://theorets.univ-reims.fr ou http://theorets.tsu.ru) sera présentée et des résultats seront donnés sur quelques systèmes (CH_4, SiH_4, GeH_4, CF_4, PH_3, C_2H_4, H_2CO, SF_6, NF_3, NH_3 et C_2H_6 de manière préliminaire).

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Prédiction des énergies de résonance d'attachement électronique

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La synthèse de molécules complexes dans les atmosphères planétaires est le résultat de processus physicochimiques non triviaux. Il est aujourd'hui admis que les électrons lents (< 20 eV) ont la capacité d'altérer le milieu irradié en induisant des réactions permettant de produire des espèces complexes. La collision d'un électron de faible énergie avec une molécule peut induire des phénomènes physicochimiques variés : dissociation, ionisation, isomérisation, réactivité, etc., via la formation d'un anion transitoire formé de la molécule cible et de l'électron projectile, et pouvant évoluer vers un état dissociatif selon des processus sélectifs.

Étudier l'interaction de ces électrons avec des molécules organiques élémentaires peut permettre d'identifier de nouvelles voies de chimie de synthèse, notamment dans les atmosphères planétaires.

Cependant, les méthodes numériques pour traiter les anions temporaires reposent souvent sur des approches semi-empiriques trop simplifiées, ou au contraire sur des méthodes ab-initio avec un coût numérique très élevé.

Nous présentons ici une méthode basée sur la TD-DFT afin de prédire les énergies d'attachement électronique pour des électrons de basse énergie. Cette méthode peut s'utiliser pour des molécules de grande ou petite taille et ne nécessite pas de recours à un quelconque paramètre empirique. Un benchmark sur plusieurs dizaines de molécules et de résonances montre un très bon accord avec l'expérience.

CH_3I-N_2, -O_2 and -air line-broadening coefficients for the ν_6 fundamental at room temperature

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Methyl iodide is the principal iodide-containing compound of the terrestrial atmosphere and serves as a route of transfer of iodine from seawater to the troposphere. As it is easily photolyzed, producing reactive iodine, that can react with ozone, the concentration of CH_3I should be carefully monitored. For this purpose atmospheric sounding from satellites can be employed, and the ν_6 band that falls into the 11μm transparency window is a suitable candidate for detection.

To supplement the existing experimental data [1-3] for a wider range of rotational quantum numbers ($0 \leq J \leq 70$ and $K \leq 20$) required by spectroscopic databases, the calculations are performed of line-broadening coefficients by a semi-classical (SC) and a semi-empirical (SE) methods for CH_3I-N_2, CH_3I-O_2, and CH_3I-air. In the SC computations electrostatic, induction and dispersion interactions are accounted for, as well as the atom-atom interactions. Comparisons of calculations with available measurements [1-3] are provided and discussed.

ACKNOWLEDGMENTS

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Ground-based remote sensing of greenhouse gases in the atmospheric column using laser heterodyne radiometers (LHR)

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Monitoring of vertical concentration profiles of key atmospheric trace gases, in particular greenhouse gases (GHGs), is essential for our understanding of regional air quality and global climate change trends. In this context, infrared (IR) laser heterodyne radiometers (LHR) have been developed for ground-based remote measurements of GHGs in the atmospheric column [1-6]. The sunlight traverses the Earth's atmosphere and undergoes absorption by atmospheric species (molecules and aerosols). The shape of the ground-based measured absorption spectrum of the molecular absorber contain information on its vertical concentration distribution. By de-convoluting this spectral signal (absorption line shape and depth) through a retrieval algorithm, the target gas abundance at different altitudes can be retrieved (Fig. 1).

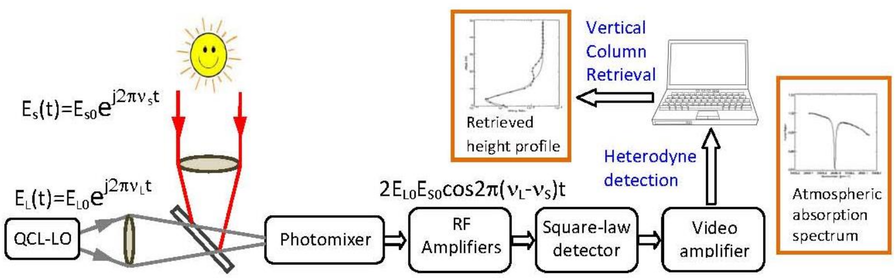

Figure 1 Principle schematic of a LHR. E_S is the sunlight electric field and E_L the laser LO electric field.

Near-IR (~1.5 µm) and mid-IR (~8 µm) LHRs have been recently developed in the present work. Field campaigns have been performed on the roof of the platform of IRENE in Dunkerque (51.05°N/2.34°E). The developed LHR instruments as well as the preliminary results of their applications to the measurements of CH_4, N_2O, CO_2 (including $^{13}CO_2/^{12}CO_2$), H_2O vapor (and its isotopologue HDO) in the atmospheric column will be presented and discussed.

Acknowledgments The authors thank the financial supports from the LABEX CaPPA project (ANR-10-LABX005), the MABCaM (ANR-16-CE04-0009) and the MULTIPAS (ANR-16-CE04-0012) contracts, as well as the CPER CLIMIBIO program.

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