REUNION DU GDR SUIE 2021

Villeurbanne, 3-5 novembre

Programme de la réunion plénière 2021 du GDR SUIE

Mercredi 3 novembre

14h00 accueil des participants

14h30 – 15h30 : conférence plénière : *Santé et Black Carbon.* **Dr. Souvet**

15h30 – 16h00 *In situ measurement of second-harmonic generation induced by soot particles exposed to femtosecond laser light.* **M. Joret**, S. Idlahcen, M. Mazur, J. Yon

16h00 – 16h30 *Development of a compact multi-spectral cavity for on-flight aerosol characterisation.* **G. Lefevre**, J. Yon, A. Poux et M. Mazur

16h30 – 17h00 Pause

17h00 – 17h15 *Exploring the spectral optical properties of soot aerosols and the impact of ageing: A mechanistic study in the large CESAM simulation chamber.* **J. Heuser**, C. Di Biagio, A. Bergé, M. Cazaunau, S. Chevaillier, P. Formenti, A. Gratien, M. Maillé, G. Noyalet, E. Pangui, B. Picquet-Varrault M. Zanatta, P. Ausset, T. Bourrianne, R. Ceolato, C. Denjean, P. Decorse, A. Faccinetto, P. Laj, A. Marinoni, D. Massabo, I. Ortega, C. Perruchot, D. Petitprez, P. Prati, L. Renzi, J. Yon, and J.F. Doussin

17h15 – 17h30 *Coupling macroscale and nanoscale simulations of soot produced in a diffusion flame : towards a more realistic simulation of their morphologies.* **J. Morán**, F. Escudero, A. Fuentes, A. Poux, F. Cepeda, L. Gallen, E. Riber, and J. Yon

20:00 Repas du GDR

Jeudi 4 novembre

09h30 – 10h30 : conférence plénière. *How do inorganic species impact the formation and physicochemical properties of atmosphéric aerosols ?* **M. Riva**

10h30 – 10h45 *Mesures en troposphère libre et modélisation à l'échelle régionale des particules de Carbone-Suie.* **S. Tinorua,** C. Denjean, P. Nabat, T. Bourrianne, V. Pont ,F. Gheusi, E. Leclerc.

10h45- 11h15 : pause

11h15 – 11h45 *A la recherche de la dimension fractale des analogues de poussière protoplanétair.* **V. Tobon Valencia**, J.-M. Geffrin, J.-B. Renard, F. Menard, H. Tortel, A. Litman, J. Millie, P. Rannou

11h45 – 12h15 *Mise en évidence de l'auto-absorption et du couplage interne au sein des agrégats fractals.* **C. Argentin**, M.J. Berg, M. Mazur, R. Céolato et J. Yon

12h15 – 14h00 Déjeuner

14h00 – 15h00 : conférence plénière *The record-breaking Australian wildfires season 2019/20: extreme tropospheric and stratospheric trace gases and particulate pollution, a rising smoke-charged vortex in the stratosphere and an exceptional climate impact.* ***P. Sellitto****, B. Legras, S. Khaykin, R. Belhadji, C. Kloss, S. Bucci et al.*

15h00 – 15h30 *Impact of exhaust chemical compositions on the optical extinction of a liquid fueled miniCAST burner.* **M. Daoudi**, P. Schiffmann, A. Faccinetto, P. Desgroux, A. Frobert

15h30 – 15h50 *Consolidating soot optical diagnostics in laminar coflow axisymmetrical flames*. **J. Yon**, J. Moran, A. Fuentes, M. Mazur et F. Liu.

15h50 – 16h15 Pause

16h15 – 17h15 Discussions/Table ronde : la place et le rôle des doctorants au sein du GDR

<u>Vendredi 4 novembre</u>

09h30 – 10h00 : *Complementary shock tube techniques for polycyclic aromatic hydrocarbon chemistry* **F. E. Cano Ardila**, A. Hamadi, N. Chaumeix, A. Comandini

10h00 – 10h30 *Adsorption of Hydrocarbons on Onion-like Carbonaceous nanoparticles.* M. Hanine, E. Liang, S. Lu, Z. Meng, H. Qi, P. Xie, X. Zhou, Y. Zhou, M. Devel, **S. Picaud**, and Z. Wang

10h30 –10h45 Pause

10h45 – 11h15 *The use of optical diagnostics for soot detection in aeronautic combustors.* **J.P. Dufitumukiza**, J. Elias, C. Irimiea, A. Faccinetto, N. Fdida, A. Vincent, D. Carru, A. Ristori, P. Cherubin, A. Lahccen, E. Therssen, A.K. Mohamed, X. Mercier

11h15 – 11h30 *SCIPPER project H2020 : Shipping contributions to inland pollution push for the enforcement of regulations. A presentation.* **B. D'Anna,** G.M. Lanzafame, B. Temime-Roussel, P. Simonen, M. Del Maso, J. Keskinen, A. Hallquist, J. Mellqvist, V. Conde, A. Armangaud, A. Somero, M. Irjala

11h30 – 12h00 Bilan, conclusion des journées

12h00 – 14h00 Déjeuner

Liste des participants

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Santé et Black Carbon

P. Souvet

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Président et Membre fondateur de l'**A**ssociation **S**anté **E**nvironnement **F**rance (ASEF)
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La pollution de l'air est une enjeu sanitaire majeur avec des estimations allant de 40000 à 97000 morts en France par an et un cout global estimé par le Sénat en 2015 de plus de 100Mds par an.

L'impact sanitaire de l'exposition à l'exposition aux particules fines 2.5 a été recherché dans de nombreuses études épidémiologiques et ont montré une perte estimée d'espérance de vie à 30 ans de 15 mois due à cette exposition, particulièrement en zone urbaine.

Les cibles sanitaires des Particules sont en premier lieu de système cardio vasculaire qui représente plus de 50% des pathologies liées à cette pollution. Le système respiratoire, le cerveau, le foetus sont également des cibles et le CIRC a classé cancérigènes pour l'homme (groupe 1) les émissions d'échappement des moteurs diesel.

Mais la connaissance des différentes sources et composition qui composent les PM2.5 (carbone suie, particules ultrafines, particules minérales, particules secondaires ...) doit permettre de les corréler plus étroitement aux évènements de santé spécifiques.

Outre l'inflammation systémique et la cascade de réactions chimiques qui génèrent les effets sanitaires, un phénomène de translocation se produit favorisé par la petite taille de la particule comme le montre l'étude réalisé en Belgique montrant la présence dans les urines d'enfants de black carbon dont la concentration est en lien avec l'exposition au trafic routier.

Les études sur le carbone suie montrent des indication « modérée » d'altération de la santé cardiovasculaire (lésions athérosclérotiques, rythme cardiaque, pression artérielle) et une indication « modérée » d'effet sur la santé respiratoire (inflammation pulmonaire). Toutefois les récentes études de cohorte (cohorte Gazel, registre des tumeurs cérébrales du Danemark) suggèrent des associations positives à long terme entre black carbon, mortalité globale, et certains cancers.

- Le carbone suie va servir de cheval de Troie à la fraction organique du carbone (mélange complexe d'aérosols organiques primaires et secondaires) chaque composé pouvant générer des effets sanitaires importants.
- Le carbone suie parait être un bon indicateur des composés toxiques des PM issues de combustion, de la pollution locale. Il devrait être dosé en routine ainsi que certains composés toxiques pouvant composés le carbone organique comme les HAP.

Références

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Contribution of Long-Term Exposure to Outdoor Black Carbon to the Carcinogenicity of Air Pollution: Evidence regarding Risk of Cancer in the Gazel Cohort. Lequy et al EHP March 2021CID: 037005 *https://doi.org/10.1289/EHP8719*

Components of particulate matter air-pollution and brain tumors. Aslak et al 10.1016/j.envint.2020.106046

IN-SITU MEASUREMENT OF SECOND HARMONIC GENERATION INDUCED BY SOOT PARTICLES EXPOSED TO FEMTOSECOND LASER LIGHT

M. Joret[1], S. Idlahcen[1], M. Mazur*[1] et J. Yon[1]

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Abstract
Numerous diagnostics exist giving access to soot properties as volume fraction, particle size, morphology, fine structure, elemental and molecular composition. Most of these techniques relies on the particles' optical and radiative properties, which depend on the particles' maturity. Nevertheless, there are no optical techniques able to inform us about soot surface and atomic structuration at the surface.

A promising approach to gain access to this informationis the use of Non-Linear Optics (NLO), which describes the optical response of a medium to high intensity/femtosecond laser light. With these lasers, effects such as Second Harmonic Generation and Third Harmonic Generation (SHG and THG) can, in theory, be observed, which should depend on the atomic structuration of the medium with a particular emphasis to its surface

Recently, SHG has been detected on different types of molecules that are placed at the surface of particles in aerosol phase (Qian *et al.*, 2018, Zang *et al.*, 2018). However, the particles needed to be coated by specific molecules with a strong SHG response, which would ultimately alter the particles inner structure and thus be contradictory to an application in internal structure analysis. In another study, third-harmonic generation and scattering have been observed on soot particles generated in flames (Zang *et al.*, 2020). Yet, no information is provided on the structure of the soot particles and to the authors' knowledge, no measurement of SHG has yet been carried out on BC particles in-situ in aerosol phase.

We developed and optimized an optical setup in order to measure extremely weak SHG signals in a reliable and repeatable way. Particular care was taken to ensure that generated light is effectively due to light-particle interactions and not due to other phenomena induced in the other elements of the experimental setup. The setup is used to carry out SHG measurements in a 1D laminar sooty flame generated by a porous injector burner similar to a McKenna burner, whose particles properties depend on their position in the flame. The results show indeed a SHG signature from the soot particles and might give access to new information on their atomic/molecular structure in the future.

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DEVELOPMENT OF A COMPACT MULTI-SPECTRAL CAVITY FOR ON-FLIGHT AEROSOL CHARACTERISATION

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KEYWORDS : Nanoparticles, Multiwavelength optical cavity, RDG-FA

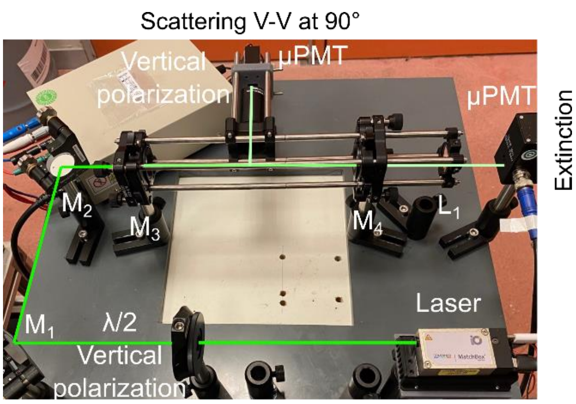
ABSTRACT

Black carbon (BC) is known to be a health and environmental problem [1,2]. These nanoparticles are formed during the incomplete combustion of hydrocarbons, for example during industrial fires. To understand their impacts, in-situ experimental analysis of smoke is crucial. UAVs are an ideal mean of providing such temporally and spatially resolved measurements, however current systems are limited. Indeed, a wide variety of techniques exist to provide information on the concentration, size, optical properties, morphology or organic content of CB particles, but the most comprehensive technologies are generally expensive and only available at laboratory scale.

One way to obtain information on concentration, size and optical properties resolved in time and space is the simultaneous measurement of extinction and scattering coefficients. In the present study, a multi-spectral optical cavity (405, 450, 520 nm) allowing the measurement of extinction and scattering in vertical-vertical polarization at 90° is developed. The aim of this cavity is to increase the optical path in a reduced space in order to finally be embedded in a UAV to study smoke plumes or for air quality monitoring. One of the first steps in this development is the installation of the cavity and its calibration. Indeed, the determination of the reflection coefficients of the mirrors constituting the cavity and the calibration of the sensors are a crucial step for the quality of the measurements.

Once the calibration of the experimental device is completed, laboratory measurements on standard aerosols are carried out to validate the method. For this purpose, extinction and diffusion measurements (in the cavity) are carried out and the information extracted is compared with measurements carried out on reference devices. The MiniCAST soot particle generator that is used as a reference generator, was used as a standard aerosol generator [3].

The data analysis is carried out using the works of Dobbins et al. [4], who developed in 1991 a theory for the scattering of light by aggregates called RDG-FA (Rayleigh - Debye - Gans for Fractal Aggregates). According to Köylü [5], this theory can be used to calculate the absorption, scattering and extinction coefficients for aggregates. Thus, an inversion method of the optical measurements (with the experimental setup) allowing the evaluation of parameters such as concentration, size, optical properties and information related to the content of organic compounds via a least square fit has been set up and will be presented.



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5. U. O. Ko¨ylu¨ and G. M. Faeth, J. Heat Transf. **116**, 152 (1994).

Exploring the spectral optical properties of soot aerosols and the impact of ageing: a mechanistic study in the large CESAM simulation chamber

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Atmospheric aerosol particles have both direct and indirect influences on the climate. The majority of aerosol particles is generally considered to produce negative direct radiative forcing meaning they are considered to force cooling of the Earth-atmosphere system. Certain types of particles are instead found to exert a positive radiative effect, in particular the strong light absorbing BC-containing particles (BC, Black Carbon). At present, the radiative properties of BC-containing particles, which determine their interaction with atmospheric radiation, still have significant uncertainties and remain difficult to represent in climate models and remote sensing retrieval algorithms. This is due to the poor knowledge on how these spectral optical properties vary at emission and modify during atmospheric aging in link with the variability and modification of the particle physicochemical properties and mixing state. Particularly, little knowledge exists on the spectral complex refractive index (CRI) of BC-containing aerosols, the basic intensive parameter describing the capacity of particles to absorb and scatter radiation.

A set of original experiments are performed in the atmospheric simulation chamber CESAM (https://cesam.cnrs.fr/) at LISA, Créteil, in order to advance our understanding of the variability of radiative properties of BC-containing particles in relation to the processes of formation and atmospheric ageing. CESAM allows the simulation of different ageing processes as it provided lifetimes of more than 24h for submicron aerosol particles and it enables the measurement of the physicochemical properties (i.e. size distribution, composition, effective density and morphology) at the same time as the spectral optical properties by combining in-situ measurement, ex-situ measurement by commercial instrumentation (e.g. cavity attenuated phase shift spectroscopy, nephelometer, aethalometer) and off-line analysis of aerosol samples collected out of the chamber. The latter for example were used to retrieve morphological information via transmission electron microscopy that will be used in future to initiate shape-advanced optical calculations to retrieve the CRI of the aerosol particles.

Two CESAM measurement campaigns on BC-containing aerosols were performed in February and May of 2021. Soot aerosols generated from the incomplete combustion of propane from a commercial diffusion flame soot generator (miniCAST model 6204 TYPE C by JING) were chosen as proxy for the BC-containing aerosols. Experiments at CESAM were set up in order to explore (i) the change of optical properties for fresh soot particles generated with different fuel/oxygen rates and therefore varying sizes and OC/EC contents and (ii) the change of the soot optical properties during aerosol ageing. Both physical aging to evaluate the influence of particle morphological restructuration in dry condition, and chemical/photochemical aging in humid conditions were simulated to evaluate the changes due to illumination and the formation of non-absorbing inorganic and organic coating on the particles. The inorganic coating was obtained by making the soot aerosols to react with sulfur dioxide (SO_2) at 35% RH in absence and presence of ozone (O_3), while the organic coating was produced by interaction with Secondary Organic Aerosols (SOA) produced in the chamber by the reaction between α–pinene, a natural Volatile Organic Compound (VOC), and O_3, as an oxidant. In this presentation we are giving an overview of the 2021 campaigns and the perspectives for future data analysis and chamber experiments.

COUPLING MACROSCALE AND NANOSCALE SIMULATIONS OF SOOT PRODUCED IN A DIFFUSION FLAME: TOWARDS A MORE REALSITIC SIMULATION OF THEIR MORPHOLOGIES

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Abstract
Experimental studies have suggested that soot particles formed along different streamlines in a diffusion flame may have different morphologies. However, the uncertainties linked to the sampling and their analysis makes difficult to quantify those differences. In this work, this problem is explored from a numerical point of view by coupling macroscopic simulations of the flame by using typical CFD codes (CoFlame, Eaves *et al.*, 2016) with nanoscopic simulation of their aggregation (MCAC, Morán *et al.*, 2020). This innovative coupling is *one-way* and consists in extracting the macroscopic Lagrangian trajectories of particles based on CoFlame simulations by considering the related temporal evolution of the flame temperature and the soot particles-gas mass transfer rates. This information is subsequently used by the Monte Carlo Discrete Element Model (MCAC) which then simulate the Brownian motion of the particles, their aggregation, their surface growth as well as nucleation and oxidation (including fragmentation) all along the residence time in the flame.

Simulations show remarkably larger and more locally compact aggregates formed near the wings of the flame as compared to the centerline of the flame. Analyzing extreme morphological parameters such as anisotropy, monomers overlapping and coordination numbers reveals the complex morphology of soot including the occurrence of chain-like aggregates along the different considered streamlines. These different morphological parameters lead to the identification of considerably different soot aggregates structures. Maximum effective numbers of primary spheres are observed near the wings of the flame with values as high as 59. Maximum anisotropy coefficients as large as 15 have been observed for aggregates having a quasi-linear chain-like structure. Primary particle overlapping as high as 90% can exist when analyzing local values (averaged within an aggregate), while population averaged values are not larger than 50%. These aggregates show larger local compacity than those studied in previous premixed flames (Morán *et al.*, 2021).

References
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Morán, J., Poux, A., & Yon, J. (2021). Impact of the competition between aggregation and surface growth on the morphology of soot particles formed in an ethylene laminar premixed flame. *Journal of Aerosol Science*, 152, 105690.

How do inorganic species impact the formation and physicochemical properties of atmospheric aerosol ?

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Atmospheric aerosol can make up a substantial fraction of ambient particulate matter, both in polluted urban areas and in cleaner rural and remote regions. Therefore, they can impact the Earth's radiative balance, stratospheric processes, and human health at local, regional, and global scales. Aerosols are made up of a complex mixture of inorganic and organic species with a wide range of functionalities and volatilities, making them one of the most challenging components of the atmosphere to characterize. Once organic and inorganic compounds are emitted into the atmosphere, they can undergo complex chemical and physical processes. While chemical processes involving inorganic and organic species have been investigated, the impacts of these interactions on the physicochemical properties of atmospheric aerosols, such as their hygroscopicity, viscosity, diffusion of water, acidity remain to be determined.

Mesures à long terme en troposphère libre et modélisation à échelle régionale des particules de Carbone-suie

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Le Carbone-suie (BC), émis naturellement par les feux de biomasse mais aussi de façon anthropique par les véhicules diesels et le chauffage urbain, a la particularité de réchauffer l'atmosphère à travers sa capacité d'absorption. Son forçage radiatif est estimé à 0.1 W.m² avec une incertitude très significative (± 0.4 W.m²) (IPCC AR6 WG1, 2021). Ce forçage s'effectue à travers l'interaction du BC avec le rayonnement et les nuages, dont l'intensité varie avec le degré de mélange du BC avec d'autres espèces chimiques (mélange interne) et la vapeur d'eau (hygroscopicité) (Bond and Bergstrom 2006, Fierce et al., 2016). Aujourd'hui les estimations du forçage radiatif considèrent rarement les effets du mélange interne et de l'hygroscopicité du BC compte tenu de leur complexité. Le manque d'études menées en troposphère libre rend notre connaissance sur les propriétés du BC lors de son transport et de son vieillissement dans l'atmosphère incomplète, ce qui entraîne une surestimation de sa concentration dans les modèles climatiques d'environ un facteur 2 (Swartz et al., 2013). L'objectif de cette étude est de contribuer à mieux comprendre la variabilité spatio-temporelle des propriétés optiques et hygroscopiques du BC, en vue d'obtenir une meilleure représentation du cycle de vie du BC dans les modèles de climat et qualité de l'air.

Un important dispositif de mesures dédié à la caractérisation des propriétés des aérosols a été déployé à la station de recherches ACTRIS-SNO CLAP du Pic du Midi de Bigorre afin de fournir les premières mesures à long terme en troposphère libre de BC. Le travail repose en grande partie sur l'utilisation de la technique SP2 (Single Particle Soot Photometer) qui permet de mesurer la concentration de BC et son état de mélange ainsi que d'un HTDMA-SP2 qui permet de quantifier le facteur de grossissement hygroscopique du BC. Ces mesures ont été complétées par des mesures des propriétés intrinsèques des aérosols, à savoir leur concentration, leur taille, leur composition chimique, leur morphologie et leurs propriétés optiques.

Des simulations climatiques, pilotées par une réanalyse (ERA5), ont été menées en parallèle grâce au modèle régional ALADIN-Climat afin de comparer les propriétés du BC simulées avec les données expérimentales. Les premières analyses des données expérimentales montrent une variabilité saisonnière du coefficient d'absorption massique (MAC) du BC et un mélange interne plus important en été qu'en hiver. Les comparaisons mesures/modèle montrent quant à elles une sous-estimation globale de la concentration en BC, ainsi qu'une surestimation de l'albédo de simple diffusion (SSA), particulièrement au printemps et en été.

A la recherche de la dimension fractale des analogues de poussière protoplanétaire

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Cette étude se concentre sur la mesure d'analogues de poussière protoplanétaire ici à partir de sept agrégats, similaire à des agrégats de suie, ayant une dimension fractale (Df) entre 1,5 et 2,8 et une orientation aléatoire [1]. A partir de ce champ diffusé, différentes propriétés peuvent être étudiées telles que la fonction de phase ou le dégré de polarisation linéaire. En exprimant la fonction de phase en fonction de $q = 2kSin\left(\frac{\theta}{2}\right)$, θ étant l'angle de diffusion, et k le nombre d'onde, on peut trouver une pente qui donne la Df, cette technique est dite Analyse de Guinier [2]. Différents auteurs ont étudié cette pente en déterminant les zones dans laquelle la Df peut être trouvée. Par exemple quand il s'agit d'une agrégation de type *Diffusion limited agregation* (DLA) la pente est entre $1/10a$ et $1/a$ (a étant le rayon du monomère de l'agrégat) [3], mais si l'agrégation est de type *Diffusion Limited cluster aggregation* (DLCA), la zone pour déterminer la pente change [3]. Nos agrégats ont été mesurés et étudiés dans les différentes zones que la littérature propose avec de multiples longueurs d'ondes, nous avons également testé un fit à partir d'un filtre de Butterworth. Néanmoins, nous avons rencontré des difficultés pour trouver la valeur exacte de la Df. Cette présentation propose ainsi une discussion ouverte sur la détermination de la Df d'agrégats basée sur les mesures et simulations que nous avons réalisées.

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MISE EN EVIDENCE DE L'AUTO-ABSORPTION ET DU COUPLAGE INTERNE AU SEIN DES AGREGATS FRACTALS

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Résumé

La théorie de Rayleigh-Debye-Gans (RDG) pour les agrégats fractals (FA) est, du fait de sa simplicité, fréquemment utilisée dans le calcul des propriétés radiatives des nano-agrégats. Néanmoins, elle reste très restrictive dans son utilisation. En effet, elle impose un indice optique proche de celui d'un milieu transparent et un diamètre des sphérules primaires négligeable devant la longueur d'onde. Si ces conditions ne sont pas remplies, des facteurs correctifs sont à appliquer, $\boldsymbol{A}$ pour la diffusion avant (dans le sens de propagation de la source lumineuse) et $\boldsymbol{h}$ pour l'absorption. Ces corrections ont été montrées pour différents descripteurs morphologiques fins (Gangue, revêtement, recouvrement (Liu *et al.*(2016))), mais aussi pour différents indices optiques (Sorensen *et al.*(2018)). Dans certains cas, la RDG-FA peut commettre des erreurs dans sa prédiction des sections efficaces pouvant atteindre jusqu'à 50% (Yon *et al.*(2014)). Bien que ces erreurs aient été observées pour différents paramètres morphologiques et optiques, jusqu'à présent aucune explication physique détaillée des phénomènes sous-jacents n'a été proposée.

L'objectif de cette étude est de comprendre l'origine physique de ces déviations en analysant le champ électrique interne des agrégats. En utilisant une approche phaseurielle dite « en tranche », nous montrons pour différents paramètres morphologiques des agrégats que deux phénomènes sont à l'origine de ces déviations : l'auto-absorption de la lumière par les particules et les « point-chauds » du champ électromagnétique interne se produisant au niveau du contact entre les sphérules. Ces phénomènes présentent une tendance universelle qui nous permet de proposer un modèle semi-empirique basé sur nos observations pour calculer les termes correctifs $\boldsymbol{A}$ et $\boldsymbol{h}$.

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The record-breaking Australian wildfires season 2019/20: extreme tropospheric and stratospheric trace gases and particulate pollution, a rising smoke-charged vortex in the stratosphere and an exceptional climate impact.

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Exceptionally strong fires occurred during the 2019/20 Australian bushfire season (also called the Australian Black Summer). For many aspects, this can be considered a record-breaking event and a new reference for fire impacts on the ecosystems and the atmosphere. These mega forest fires burnt an unprecedented area of about 5.8 million hectares, which is more than 20% of whole Australian temperate forests amount. This historically-relevant fire season was active from September 2019 to March 2020. The concentrations of many tropospheric pollutants were enhanced, in the Southern Hemisphere, since the early phases of the fire season. By the way, the intensity of the fires escalated and had a peak in intensity starting from New Year's Eve 2019/20 to early January 2020, and led to extreme pyro-convective clouds events and the formation of a self-sustained smoke-charged vortex that polluted the stratospheric trace gases and aerosol composition at the Hemispheric spatial scale. In this talk, we will present the corpus of information and studies we have collected since the early phases of this event. The atmospheric aerosol and trace gases perturbations linked to this event will be shown using satellite and ground-based remote sensing as well as atmospheric modelling, with a particular accent on water vapor, ozone and smoke aerosols. We also could track dispersing plumes of less abundant pollutants, like nitrous acid, using satellite observations, and we will quickly discuss this aspect in the talk. The core of the talk will be on the striking evidence of the formation of a self-maintained smoke vortex during the most intense phases of the pyro-convective events. This highly stable vortex persisted in the stratosphere for several weeks and, due to the absorption of solar radiation within the absorbing aerosol plume, lifted a confined bubble of smoke to not less than 35 km altitude. Finally, we will discuss the planetary-scale blocking of solar radiation by the smoke associated to this event, which is found larger than any previously documented wildfires and of the same order of magnitude as the radiative forcing produced by moderate volcanic eruptions.

Impact of exhaust chemical compositions on the optical extinction of a liquid fueled miniCAST burner

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Soot generators are widely used test subjects and calibration sources. While electro erosion devices and propane fueled CAST burners were the work horses for nano soot particle generation, we focus in this experimental work on a liquid fuel-based miniCAST soot generator (model 5201 D). More specifically, its exhaust chemical composition and how it can affect optical measurements in the UV-VIS and near IR range. Standard Diesel B7 fuel was burned over a range of globally fuel-lean to fuel-rich conditions. Optical extinction in the UV region increased quasi-proportionally with fuel-to-air ratio. Different means of sample pretreatment have been employed to dissociate solid and volatile particles from gas phase with the aim to quantify their respective contribution to the measured optical spectra. Global gas phase HC concentration and qualitative chemical analysis of soot samples are analyzed using ex-situ time of flight secondary ion mass spectrometry.

CONSOLIDATING SOOT OPTICAL DIAGNOSTICS IN LAMINAR COFLOW AXISYMMETRICAL FLAMES

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Abstract

The detailed study of soot particle formation in academic flames (laminar, stationary) is still necessary to improve the kinetic models of soot particle formation. The information is more accessible and parametric studies are possible (effect of ventilation, fuel, pressure...). The optical techniques allowing the extraction of information concerning soot particles are numerous and rely on different mechanisms. Among these techniques, light extinction, scattering and laser induced incandescence are the most used. The collected signals are interpreted in terms of volume fraction and size by using light-particle interaction models such as RDG-FA. The results thus obtained can suffer from the inaccuracy or the unsuitability of these models and work is to be carried out in this direction. However, the results can also suffer from strong assumptions (for example the complex index of soot considered uniform) or from experimental constraints that can limit, for example, the spatial resolution.

In this presentation, we present two recent advances in optical diagnostics that have been developed and applied to a laminar diffusion flame (Gülder burner fed with ethylene). The first one consists in exploiting the spectral dependence of the attenuation imaging (Line Of Sight Attenuation) in order to remove the uncertainty of the optical index variation driven by the evolution of its maturity. The second one consists in proposing an original experimental configuration of angular scattering (Horizontal Planar Angular Light Scattering) in order to access size information with an improved spatial resolution. Finally, we show the benefit of coupling these two techniques in order to access all the following quantities: temperature, volume fraction, maturity, absorption function, numerical concentration of particles, diameter of primary spherules.

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Complementary shock tube techniques for polycyclic aromatic hydrocarbon chemistry

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Particulate matter (PM) emissions from combustion devices constitute a serious threat for human health and the environment. Mitigation techniques aimed at reducing such emissions are often based on CFD calculations for design of optimized engines and turbines or fuel reformulation. For these techniques to be effective, accurate chemical kinetic models of particle chemistry are required for coupling with the fluid dynamic components of the codes. Particle formation is a very complex, multi-step process starting with the formation of the polycyclic aromatic hydrocarbons, PAHs, the particle building blocks. Kinetic studies on PAH chemistry are then essential to develop accurate and predictive particle kinetic models.

The high-purity single-pulse shock tube at the laboratory ICARE has been coupled to gas chromatographic systems for the measurement of species profiles at high-temperatures and high-pressures typical of modern combustion devices. The specific characteristics of the apparatus allow measurements of relatively large PAH products, up to four-rings, in pyrolytic highly diluted conditions. A case study will be presented where toluene (100-200 ppm) is pyrolyzed at temperatures of 1200 – 1700 K, pressure of around 20 bar, and reaction time of 4 ms. The data have been used to validate a detailed chemical kinetic model focused on the PAH chemistry which is used to improve our fundamental understanding of the reaction network leading to key multi-ring structures.

Despite the detailed and essential kinetic information that can be obtained with the shock tube – gas chromatography combination, this laboratory-based technique has some limitations due to the condensation of large species and reduced capabilities to separate isomers of large PAHs. To overcome such limitations, a miniature high-repetition-rate shock tube (ICARE-HRRST) has been designed and constructed for use at Synchrotron facilities. The ICARE-HRRST was coupled to the double imaging photoelectron photoion coincidence spectroscopy (i^2PEPICO) as developed at the DESIRS beamline of the Synchrotron SOLEIL in France. The main features of these techniques will be presented together with the experimental results obtained on the pyrolysis of 0.1 % toluene at temperatures of around 1480-1490 K and pressures of around 7.4-7.6 bar. In particular, the presence of a large number of PAH species was detected in the mass spectra compared to the laboratory-based studies, up to m/z of around 350. For the main species, the kinetic profiles were derived as function of time. In addition, experiments were performed to derive the photoelectron spectra for identification of PAH compounds.

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Adsorption of Hydrocarbons on Onion-Like-Carbonaceous nanoparticles

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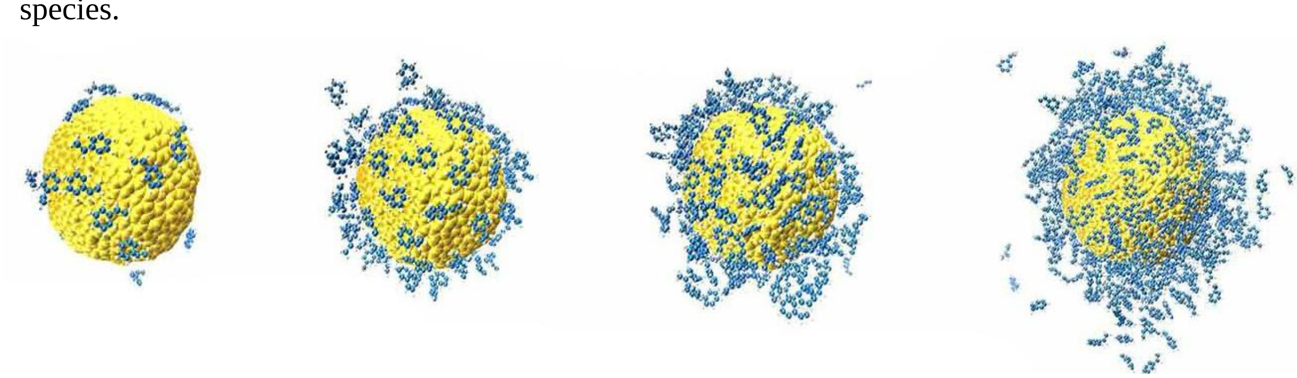
ABSTRACT Molecular dynamics simulations are used to characterize the adsorption process of aliphatic and aromatic hydrocarbon molecules on onion-like carbonaceous nanoparticles mimicking nanograins which are present in the interstellar medium (ISM) as well as atmospheric soot primary nanoparticles. These atomistic simulations are based on the reactive AIREBO interaction potential model to describe the interaction between the adsorbate and the substrate, and both hydrogenated and dehydrogenated molecules are considered in the calculations.

More specially, we focus on the influence of the carbonaceous nanoparticle on the aggregation process of the hydrocarbon molecules and compared the behaviors around the nanoparticle and in the gas phase without any nanoparticle.

The results of the simulations reveal the role that carbonoceous nanoparticles may play as selective catalysts providing reaction substrates for the formation of interstellar PAHs, large-fullerenes and soot particles from adsorption and subsequent aggregation of various hydrocarbon species.



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The use of optical diagnostics for soot detection in aeronautic combustors

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In order to design the Next Generation of aircraft combustors that emit low quantities of gaseous and particulates (soot) pollutants, it is necessary to have representative combustion conditions establish in aircraft engines and experimental tools that can capture complex phenomena participating in these combustion settings. Thus, nowadays, propulsion systems integrity and development depend on the ability to create a theoretical model that describes working principles with the most significant detail, as well as to practically demonstrate that proposed models function on small and large scale representative systems. Therefore, semi-industrial test rigs equipped with suitable optical diagnostics offer a glimpse into combustion processes. Furthermore, soot formation is still among today's combustors' milestones, hence the necessity to use optical diagnostics for its characterization and detection.

In this work, Laser Induced Incandescence (LII) is adapted and developed for detecting soot particles in aeronautic combustors. Laser Induced Fluorescence is also used to identify the regions with various polycyclic aromatic hydrocarbons (PAH) classes. These two optical techniques are already well known, but their implementation on large scale combustors is not straightforward due to the various technical challenges. This study targets the development of planar in-situ laser-based methods serving to map soot precursors and soot particles in aircraft combustors. Figure 1 shows LII at 1064 nm coupled with LIF at 532 nm for monitoring of soot and its precursors, respectively. A progressive approach is followed for implementation of the optical techniques, where LII/LIF is first tested and evaluated in a laminar diffusion flame stabilized on a coflow burner at atmospheric pressure. The optical configuration and the working parameters, such as the laser sheet generation and dimensions, adequate laser fluence for LII and LIF, were evaluated in the CH_4/air flame at atmospheric pressure to estimate uncertainties in the case of the use of an extended laser sheet. Additionally, two optical configurations of LII (spectral and image) were tested during a collaboration between ONERA and PC2A laboratory, aiming at preparing the LII technique for the implementation on the MICADO test rig[1].

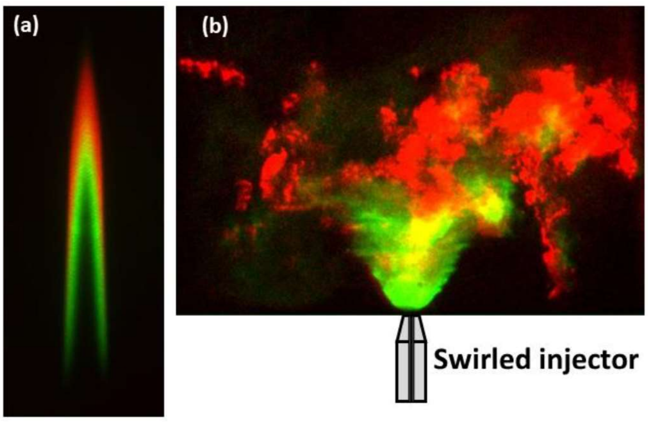

Figure 1: (a) Overlay of LII (red) and LIF-PAH (green) on CH_4/air laminar diffusion flame at atmospheric pressure. (b) Overlay of LII (red) and LIF-PAH (green) obtained from Jet A-1/air combustion on MICADO test rig at p= 7.14 bar.

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SCIPPER PROJECT H2020
SHIPPING CONTRIBUTIONS TO INLAND POLLUTION PUSH FOR THE ENFORCEMENT OF REGULATIONS A PRESENTATION

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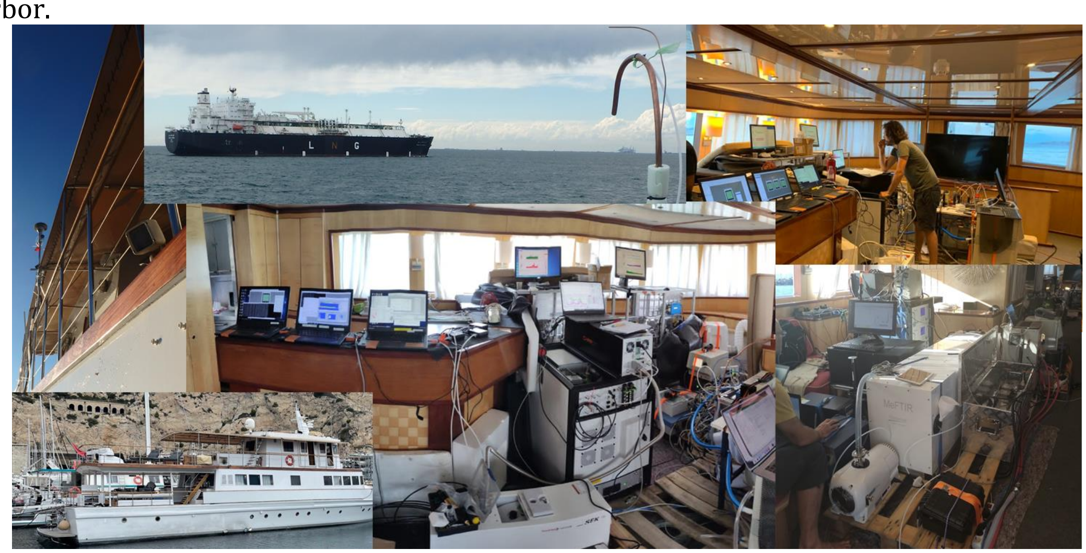
Shipping represents the largest global cargo transport mode, serving more than 80% of total freight transport. While vessels exhibit comparatively low fuel consumption per unit of cargo-distance, they produce high specific emissions of NO_X, SO_X and PM. Most of these occur rather close to the shore and degrade air quality (AQ) in coastal areas. This has been investigated in several studies using atmospheric chemistry models and field experiments. These showed that marine exhaust may be responsible up to 20-30% of ambient NO_2 and similar fractions of nitrate and sulphate PM over the southern North Sea coast.

Since 1 January 2015, the equivalent fuel sulphur content (FSC) in Sulphur Emission Control Areas (SECAs) dropped from 4.5% m/m down to 0.1% m/m. On 1 January 2020 a maximum FSC of 0.5% will globally be enforced outside SECAs. In the EU, additional requirements for ferries and ships at berth go beyond IMO regulations. This creates requirements to monitor the compliance of ships with sulphur regulations separately in port areas and in the open-seas.

SCIPPER is based on five field measurement campaigns (Gothenburg, Hamburg or Rotterdam and Marseille, English Channel), one mirror campaign in Hong Kong and long-term monitoring data SCIPPER will assess the suitability, operational capacity, and cost-effectiveness of various monitoring methods.

In July 2021 SCIPPER consortium deployed state of the art instrumentation in Marseille harbor and at Fos-sur-Mer to assess ship emissions impacts using drones, sniffer boat, permanent sites in the harbor.



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