Sources and impacts of atmospheric aerosol particles on urban and forested environments

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Sources et impacts des particules d'aérosols atmosphériques en environnements urbains et forestiers

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l'Habilitation à Diriger des Recherches

Membres du jury :

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Anne MONOD, Professeure, Aix-Marseille Université (rapporteuse)

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Ecole doctorale Science de la Matière, du Rayonnement et de l'Environnement
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1 Foreword

This document presents a synthesis of my activities of teaching and research since obtaining my PhD title, towards receiving a *Habilitation à Diriger des Recherches* (HDR). This academic qualification is a major milestone in the academic career. Not only as a recognition of research independence and maturity but as a mandatory step towards furthering one of the most amazing aspects of this career, the mentorship and the contribution to the development of the next generation of researchers.

Working on this manuscript has felt like a journey through the "memory lane" mixed with *déjà vu* feelings, reminding me of the enriching (i.e. painstaking) process of writing the PhD thesis back in 2011. Fortunately, the experience has improved significantly since the last time I set out to write such a manuscript. Not only because I ditched Latex around 2013 and have nowadays access to vastly superior spellchecker tools, but because this time I managed to focus on the personal rapports that I had the privilege to gather along the way, from mentors, colleagues, collaborators, and students. The following paragraphs are an attempt to name them, explicitly or implicitly, to the best of my abilities.

It has been a long and winding road to get to this point. The first steps were performed at the Laboratory of Atmospheric Physics at the USP led by Paulo Artaxo. Those were merely two weeks after receiving my PhD diploma, as I was helping to install an SMPS at one of the most inhospitable places known to man, a vehicular tunnel in Downtown Sao Paulo. For the next five years, being in the group was an incredibly enriching experience. Under Paulo's guidance, I was able to work side-by-side with amazing people (Henrique, Alexandre, Luciana, Samara, Fernando, Fabinho, among many others), outstanding collaborations (often under the Amazonian rain) and have my first supervising experiences (Rafael, Glauber, Bruna, Djacinto, etc.).

At the end of my term at USP, I was very fortunate to join LaMP in 2016 as a postdoc. Alfons Schwarzenboeck and Evelyn Freney have been outstanding in providing not only a professional but also a personally welcoming environment that allowed me to quickly adapt to a different place and culture – so much so that we decided to adopt this country ourselves. The help from colleagues there has largely exceeded the scope of the postdoctoral work, and for that, I'll be forever grateful also to Agnès, Aurélie, Regis, Pierre, Maher, Pamela and so many others to cite here. It is also important to highlight the hard work and friendship of colleagues from other institutions who spent a month flying up and down in Southern West Africa during the DACCIWA aircraft campaign. Special attention to the crew of the ATR from SAFIRE and the PIs of different components of that project such as Peter Knippertz and Cyrille Flamant.

As I write these words, it has been nearly 5 and a half years since I joined IMT Nord Europe. The attention and care that I found here have been paramount in making me feel welcomed and aspiring for growth. I am thankful to be here on many different levels:

- I recognize and appreciate immensely the work that Patrice Coddeville and Nadine Locoge have done managing CERI EE, not only from an administrative/financial viewpoint but also through a supportive and attentive approach. This is also strongly applicable to our administrative staff, Véronique F., Hélène and before her, Sandrine.

➢ It is important to highlight the bond and lighthearted exchanges with colleagues whether in day-to-day life in the office, such as Esperanza, Laurent, Thérèse, Anna, Aude, François, during field/ chamber work (Manolis, Marina, Alexandre, Liselotte, Manu, Thierry, etc.) or facing the harshness of teaching fluid mechanics (Jean-Luc, Caroline, Tom and the top folks of the IRP department).

➢ Furthermore, it's crucial to recognize the camaraderie with fellow CERI EE members, including faculty, graduate students, interns, as well as from other CERIs or the broader IMT Nord Europe community. Though too numerous to name individually, their presence, from casual hallway encounters to various meetings, significantly contributes to making this institution a great work environment. A special shoutout to the Brazilian community in the lab that helped make *Festa Junina* the event of the year in Douai.

➢ It is important to highlight the crucial role of community work at the regional level (Labex CaPPA, CPERs CLIMIBIO and ECRIN), nationally or internationally over the past few years. Special mention to fellow thesis advisors (Suzanne Crumeyrolle, Isabelle Chiapello, Olivier Favez, Caroline Marchand, Christopher Pöhlker, Stéphane Sauvage, Sébastien Dusanter) and a particular one to Véronique Riffault, who has not only collaborated closely scientifically and professionally over the years, has guided me (and still does) through the intricacies of the French system, and still has kindly accepted to be the *garante* of this HDR.

➢ Finally, I would like to thank Martial Haeffelin, Anne Monod, and Denis Petitprez to have accepted being the reviewers of this document, and Matthias Beekmann, Maria de Fatima Andrade and Mira Pöhlker for integrating the jury. Last but not least, to the current or former PhD students: Alejandra Velazquez-Garcia, Hasna Chebaicheb, Carolina Ramirez and Olatunde Murana, without whom this HDR would not have been possible.

I dedicate this document to my wife and children, who loved and supported me (on so many levels), whether day-to-day or while absent keeping mass spectrometers alive in the middle of the jungle. Obrigado, vocês são minha vida.

1.1 Document Outline

The manuscript comprises two main sections:

- A description of my professional activities, including supervision, publications, teaching, and administrative tasks.

- A broad overview of scientific achievements, focusing on aerosol research in urban, forested and mixed environments. This section is complemented by research perspectives aiming to improve our understanding of the sources and effects of atmospheric particles.

A detailed Curriculum Vitae and a complete list of publications, communications, and other dissemination activities are included as Appendices.

2 Professional trajectory

I am currently an Assistant Professor at the Centre for Education, Research and Innovation Energy and Environment at the Institut Mines-Télécom (IMT) Nord Europe. This section presents an overview of my professional trajectory since being awarded the PhD title, providing details on student supervision, teaching, scientific collaborations, production and editorial work, as well as administrative responsibilities.

2.1 Career evolution

Soon after receiving my PhD diploma at the Karlsruhe Institute of Technology (KIT), I returned to Brazil to join Prof. Paulo Artaxo's group at the University of Sao Paulo (USP), in May 2011. I was hired as a Research Associate through a PETROBRAS-funded project (termed FONTES, i.e. sources in Portuguese) aiming to quantify the impact of vehicular emissions on Sao Paulo and Rio de Janeiro's atmospheres. My main responsibilities were two-fold: dynamometer test bench measurements on dozens of cars, as well as to set up four observation sites within the Sao Paulo Metropolitan Area (SPMA). Through the project, I also oversaw the acquisition and deployment of unique instrumentation at the time in Brazil, notably two Aerosol Chemical Speciation Monitors (ACSM, Aerodyne) and a Proton-Transfer-Reaction Mass Spectrometer (PTR-MS, Ionicon). The observational strategy consisted of an extensive campaign (at least 1 year) sampling particle filters at every site, and intensive campaigns, deploying online instrumentation, on a rotational basis. The filters were prepared and collected by a technician under my responsibility also hired through FONTES. The project ran from 2011 till 2014, in the meantime I also integrated a range of different projects from the group, including tunnel measurements within SPMA, as well as different campaigns in Amazonia, targeting both pristine, as well as disturbed atmospheres, and combining ground and research aircraft measurements.

After the completion of the FONTES project, I applied for a highly competitive FAPESP (State of Sao Paulo funding agency) post-doctoral grant, being awarded for two years (2014-2016). The grant fitted within the scope of the large-scale Green Ocean Amazon (GoAmazon 2014/5) project. Originally a US Department of Energy (DoE) project, awarded to Prof. Scot Martin at Harvard University, the project grew substantially due to the support of multiple funding agencies (NSF, FAPESP, etc.), thus allowing a wide range of activities and platforms, such as multiple ground sites, aircraft, balloons, and so forth. I was responsible for a number of instruments along multiple sites, measuring mainly aerosol chemical and physical properties, as well as coordinating the T2 site, near Manaus. Albeit professionally extremely enriching, in 2014 I spent a combined 6 months in Central Amazonia, about 3000 km from my wife and young daughter in Sao Paulo. In 2015, due to the looming economic downturn in Brazil, it became clear that the opening of permanent positions would be put to a halt for at least a few years. Combined with the fact that FAPESP limited postdoctoral funding up to 7 years after PhD completion, it was obvious that we needed to move abroad to ensure no salary gaps till an opportunity for a permanent position would arise. Thus, in early 2016 I relocated to a post-doc position at the Laboratoire de Météorologie Physique (LaMP), Clermont-Ferrand, France.

The post-doc at LaMP, starting in April 2016, was inserted into the EU-funded DACCIWA (Dynamics-Aerosol-Chemistry-Cloud Interactions in West Africa) project, coordinated by

Prof. Peter Knippertz (KIT). Under the supervision of Prof. Alfons Schwarzenboeck and Dr Evelyn Freney, my project consisted of studying aerosol formation and composition in Southern West Africa onboard the French ATR-42 aircraft in June-July 2016. The project included as well two other research aircraft, the British Twin Otter and the German Falcon, with complementary payloads and scientific objectives. The successful campaign provided a rich dataset that could be exploited to better understand the role of widespread anthropogenic emissions and atmospheric dynamics on regional climate. At the end of my postdoctoral contract at LaMP, in May 2018, I was offered the assistant professorship position at IMT Nord Europe, in Douai.

The position at IMT Nord Europe was designed to strengthen aerosol research, particularly aerosol mass spectrometry led by Prof. Véronique Riffault. Soon after joining the department, I started teaching at IMT, joining the team responsible for the course "Introduction to Fluid Mechanics", as well as integrating into several research projects allowing me to familiarize myself with the position. Those were very enriching, whether by readily integrating within the regional (CLIMIBIO), or European landscape (ACTRIS, COLOSSAL), as well as providing a first experience in using simulation chamber experiments (INVOC-Dust). In the following years, I furthered those components, combining with student supervision, teaching and responsibilities within and outside IMT Nord Europe. The next sections within this Chapter provide a detailed account of each of those topics.

2.2 Supervision of graduate students and post-docs

To date, I supervised 13 graduate students and post-docs, including three completed PhD theses, as well as two currently ongoing ones. I present below the complete list of supervisions, accompanied by a brief description and my particular role within them.

MSc

Hasna Chebaicheb (2020): "Study of the aerosol dynamics during the summertime in a rural region in the North of France". IMT Nord Europe. Co-supervision with Dr Aude Bourin. Based on observations from the COBIACC field campaign, this M2 project consisted of performing 0-D atmospheric modelling (SSH-aerosol) to simulate secondary organic aerosol formation constrained by field campaign data. I was responsible for data collection, overseeing 0-D model application and interpretation.

Rafael Stern (2014 – 2015): "Physico-chemical characterization of aerosols during 2013 dry season in Central Amazonia". University of São Paulo/INPA. Under the co-supervision of P. Artaxo, this MSc project focused on data gathered during the BUNIAACIC campaign, in Central Amazon, from July-Dec 2013. The analysis included aerosol chemical and physical observations, as well as back-trajectories and coupling with gaseous compounds and meteorological parameters. In addition to being responsible for aerosol composition data, I oversaw the coupling of chemical and physical aerosol properties.

Djancinto M. dos Santos (2013 – 2015): "Vehicular emissions in Sao Paulo: quantification of their effect using receptor models and their carbonaceous content". University of São Paulo. Co-supervised by P. Artaxo, this project has focused on Organic and Elemental Carbon from 1-year filter sampling within the Sao Paulo Metropolitan Area, as part of the FONTES project. Thermogram and correlation analysis have provided interesting insights into primary and secondary sources, as well as the homogeneity of aerosols throughout the city. I oversaw data collection, interpretation and manuscript writing.

PhD

Olatunde Murana (2022 –): "Understanding air masses of anthropogenic and biogenic origin in Paris and Sao Paulo". IMT Nord Europe, funding from ANR (French national research agency) and Labex CaPPA. Direction by V. Riffault, co-supervision by Dr Sébastien Dusanter. Contrasting Paris and Sao Paulo atmospheres, with a particular focus on the coupling of anthropogenic and biogenic emissions on aerosol formation and properties. Analysis was performed through simulation chamber experiments and intensive observation periods in Paris (Jun-Jul 2022) and Sao Paulo (Apr-Jun 2023). My work consisted of writing the PhD funding proposal, project outlining, field campaign, data validation and interpretation and manuscript writing.

Carolina Ramirez Romero (2020 –): "Study of gas and particle phase partitioning of semi-volatile biogenic organic compounds in central Amazon", IMT Nord Europe/Max Planck Institute for Chemistry, funding HdF region and MPI-C. Directed by Prof. Stéphane Sauvage and co-directed by Dr Christopher Pöhlker. Focusing on atmospheric observations and chamber

experiments of semi-volatile organic aerosols, this thesis aims at improving our understanding of secondary organic aerosol formation in Central Amazonia under pristine conditions. My work consisted of writing the PhD funding proposal, project outlining, field campaign, data validation and interpretation, and manuscript writing.

Hasna Chebaicheb (2020 – 2023): "Sources of fine aerosols at various French sites using highly time-resolved multiyear datasets", IMT Nord Europe/INERIS, funding LCSQA. Under the direction of V. Riffault, co-supervision by Dr Caroline Marchand and Dr Olivier Favez, this thesis is centred around multi-annual source apportionment of Organic Aerosols through the use of rolling Positive Matrix Factorization. First dealing in detail with the ATOLL site, and later with 12 sampling sites across continental France. My main contribution has been to project outlining, data validation, interpretation and manuscript writing.

Alejandra Velazquez-Garcia (2019 – 2023): "Chemical and optical properties of particulate pollution in the Lille area, Northern France based on ATOLL observations". IMT Nord Europe/Université de Lille, funding from HdF region and CONACYT. Under the direction of V. Riffault, co-direction of Dr Isabelle Chiapello, and co-supervision of Dr Suzanne Crumeyrolle, this was the first PhD thesis working with the in-situ ATOLL dataset. It has provided extensive multi-annual analysis of aerosol properties and made use of its long dataset for detailed chemical-physical studies, as well as black carbon source apportionment. My work focused on assisting with project outlining, data validation, interpretation, methodology development and manuscript writing.

Florian Wurm (2013 – 2015): "Sources and impacts of Volatile Organic Compounds in the vicinity of Manaus during GoAmazon2014/5 experiment". University of São Paulo. Funding from FAPESP. Under the direction of P. Artaxo, this thesis has focused on gas-phase observations in the vicinity of Manaus, notably contrasting clean and polluted conditions. Despite good progress and interesting results, the student abandoned the PhD program in the third year to return to his native country. My main contribution was to project outlining, analytical developments, deployment, data treatment and interpretation.

Glauber Cirino (2011 – 2015): "Physico-chemical characterization of aerosols during GoAmazon2014/5 experiment: interaction between Manaus and pristine forest emissions". University of São Paulo/INPA, funding LBA. Under the direction of P. Artaxo, the thesis has focused on aerosol properties on multiple sites from the GoAmazon2014/5 project. One of the key results has been an extensive analysis of plume evolution, notably between the site in the vicinity of Manaus (T2), and 70km downwind (T3). My main contribution was observations, data validation, analysis, methodological developments and manuscript writing.

Post-doctoral researchers

Vishnu Murari (2022 –): Characterization of aerosol particles from shipping emissions within the context of PIRATE and SHIPAIR projects. Operating a HR-AMS during the field campaign in Sep-2022 in Dunkirk, he is responsible for data analysis aiming at providing an improved shipping aerosol characterization, notably from the chemical perspective, and contrasting with regional pollution episodes in the region. Co-supervision with V. Riffault and Dr Liselotte Tinel.

Layal Fayad (2021): Development of an innovative VOC sampling system for drone measurements within the ALPAGA project. Methods developed focused on sensors and tedlar bags for offline analysis combined with PTR-Qi-ToF-MS. Co-supervision with Dr Thérèse Salameh.

Antoine Farah (2020 – 2021): Chemical characterization of aircraft engine exhaust within the UNREAL project. Operation of HR-AMS during chamber experiments at CESAM, focusing on primary and secondary aerosols from the combustion of a range of aircraft fuels. Particular focus on the validation of portable oxidation flow reactor compared against the simulation atmospheric chamber on Secondary Organic Aerosol (SOA) formation yield assessment. Co-supervision with V. Riffault.

Evdokia Stratigou (2019 – 2021): Observations of atmospheric composition in a rural site in Northern France. Deployment and data analysis of HR-AMS and PTR-Qi-ToFMS during the COBIACC campaign, including source apportionment through PMF analysis. Co-supervision with V. Riffault and S. Dusanter.

2.3 Participation in MSc and PhD Jurys

I have participated in six PhD committees, two as a member of the defence jury, four as CSI (*Comité de Suivi Individuel*) and two MSc projects. Details of the projects are provided below.

PhD defence jury

Manon Rocco (2021): Analyse des déterminants de la distribution des composés organiques volatils en milieux naturels contrastés, Université Clermont Auvergne, France. Direction: Jean-Luc Baray and Aurélie Colomb.

Beatriz S. Oyama (2015): The role of vehicular emissions on organic aerosol chemical composition in São Paulo, University of São Paulo, Brazil. Direction: Maria de Fatima Andrade.

CSI

Yijie Shi (2023 –): Modeling molecular markers of organic aerosols to assess gas/particle partitioning of organic compounds in the atmosphere, École Des Ponts Paristech, France. Direction: Karine Sartelet and Florian Couvidat.

François Hemeret (2022 –): Constraining the radiative effects of complex aerosol mixtures in southern Africa: an experimental study of their chemical composition and spectral optical properties, Université Paris-Est Créteil, France. Direction: Paola Formenti and Claudia Di Biagio.

Francesco Battaglia (2021 –) : Altération des poussières minérales par les composés organiques volatiles d'intérêt climatique : composition chimique et propriétés optiques de mélanges complexes en fonction du vieillissement atmosphérique, Université Paris-Est Créteil, France. Direction: Paola Formenti.

Yunjiang Zhang (2018): Development of monitoring stations for the measurements of aerosol chemistry and physics in France, Université de Versailles-Saint-Quentin, France. Direction: Valerie Gros.

MSc defence jury

Chang Yuyang (2020): Dust Aerosol Observation (DAO) campaign: Analysis of aerosol events observed at Kashi site. Université de Lille, France. Direction: Philippe Goloub & Qiaoyun Hu.

Antônio T. Bittencourt (2013): Characterization of CCN in São Paulo: the influence of chemical composition and size distribution on their properties, State University of Ceara, Brazil. Direction: Gerson Almeida.

2.4 Collaborative projects

Below, I describe the collaborative projects in integrated, whether as Principal Investigator, work-package coordinator or participant. I present the project acronym, the period that I participated, and partner institutions (the coordination indicated by an asterisk), followed by a short description of its scientific goals, scope, and contextualization of my role and contribution.

IDE (2023 –). Partners: IMT Nord Europe*, Federal University of Espirito Santo*, IMT Alès, Federal University of Minas Gerais, Mines Nancy, Federal University of Ceara, IMT Atlantique among others. The ”Engineering and the objectives of the sustainable development (IDE)” is a French/Brazilian bilateral project focusing on undergraduate student and academic exchange. Focusing on topics linked to climate change, and regrouping research teams on atmospheric composition characterization and mitigation strategies, this project is designed to bring together superior education institutions at multiple levels. I am the coordinator from the French side.

MI-TRAP (2023 –). Partners: NCSR Demokritos*, Technical University Munich, Aarhus University, Institute of Chemical Process Fundamentals (Czech Republic), Politecnico Di Milano, IMT Nord Europe, Wageningen University, among others. Funded under the Climate, Energy and Mobility EU programme, this broad project targets transport-related (road, ship and aircraft) pollution, focusing on innovation developments linked with source apportionment and mitigation strategies. With a particular focus on air quality degradation (notably through solid particle emissions) as well as noise, this project proposes observational studies of 8 urban areas, 5 ports and 5 airports across the EU. I am involved in intensive field campaigns at three sites (two urban sites in Lille and one port site in Dunkirk) and participate in the coordination of the near-real-time source apportionment task.

GAIA (2023 –). Partners: University of Sao Paulo*, National Institute of Space Research (INPE), National Institute of Amazonian Research (INPA), State University of Amazonia, IMT Nord Europe, Max Planck Institute for Chemistry, Max Planck Institute for Biogeochemistry, University of Stockholm, University of California. Focusing on atmospheric composition, reactivity and climate of the Amazon Basin, this thematic FAPESP-funded project (2023 – 2028) aims at combining ATTO extensive observations, including the newly installed automatic elevator for day and night vertical profiles from the ground up to 325 meters, studies on New Particle Formation events, remote sensing, and extensive ship research cruises along the Solimoes, Negro and Amazonas rivers. I participate in the project activities linked to gas and particle observations at ATTO and cruise activities.

FILL (2023 –). Partners: IMT Nord Europe*, Université de Lille, Université du Littoral Côte d'Opale, CNRS. A LEFE-CHAT project (2023 – 2025) focusing on biomass burning, notably the reactivity of a range of furanoids, compounds typically found on vegetation fire plumes. This will be performed by a range of kinetics experiments on pure compounds and on atmospheric chambers, at IMT Nord Europe, ULCO and ICARE. My role on this project focuses on aerosol particles, notably quantification of secondary organic aerosol formation yields, and their chemical and physical properties.

ACROSS-GO (2022 –). Partners: Université Paris-Est Créteil*, Université de Bordeaux, IMT Nord Europe, Université de Versailles-Saint-Quentin, Université Clermont Auvergne, CNRS,

Université d'Aix-Marseille, Météo-France, INRAE, Sorbonne Université, Université de Lille, Université d'Orléans. Focusing on the interaction between anthropogenic emissions, notably from Paris, with biogenic emissions from the city and its surrounding areas. The ACROSS-GO (LEFE-CHAT, 2022 – 2025) aims at integrating urban, peri-urban and forested ground sites within a common framework from a field campaign that took place in the summer of 2022. I was responsible for HR-AMS observations at the forested site, as well as coordinating the WP3 component (Aerosol Properties, Composition and Impacts) together with P. Formenti.

OSEAMS (2022 –). Partners: IMT Nord Europe*, National Centre University (NCU, Taiwan)*. Focusing on tropospheric ozone in southeast Asia, the project combines atmospheric observations and modelling to develop air pollution mitigation strategies, particularly in Taiwan. It is a collaborative research project between ANR and MOST (2022 – 2026), largely based on the scientific exchange between both countries and a field campaign to take place in Taiwan in 2025. My role on the project is to oversee ozone and VOC measurement efforts onboard the drone platform.

PIRATE (2022 –). Partners: IMT Nord Europe*, Université d'Aix-Marseille, École des Ponts ParisTech, AtmoHdF, AtmoSud, AtmoNormandie, INU (Corée du Sud). Aiming at improving pollutant emission estimates from shipping activities in port areas in France, this ADEME-funded project (2022 – 2025) combines measurement campaigns and innovative modelling with real-time data from ships. The latter, developed by collaborators from South Korea, is based on real-time geolocated, and this approach is to be compared with the currently used methodology via annualized emission inventories. Furthermore, the study of their evolution in the near-field to urbanized area scales, in conjunction with better estimates of the impact of maritime traffic, allows for improved pollution mitigation strategies. On this project, I am responsible for aerosol composition during the field campaign in Dunkirk, overseeing the postdoctoral work of V. Murari, and coordinating WP5, responsible for the integration of observational and modelling datasets.

SHIPAIR (2022 –). Partners: École des Ponts ParisTech*, Université d'Aix-Marseille, IMT Nord Europe, Université Grenoble Alpes, AtmoHdF, AtmoSud, AtmoNormandie. This ANR-funded project (2022 – 2026) is a companion project of PIRATE, described above. It focuses on improving our knowledge of shipping-related pollutants emission and dispersion to better tackle their impacts on urban air quality and health. This is based on modelling and observational strategies of regulated and non-regulated, primary and secondary pollutants, notably through field campaigns on three harbours in France (Dunkirk, Marseille and Le Havre). Furthermore, an emphasis of the project is given to associating the health effects of those pollutants, notably through offline oxidative potential with source apportionment. My role on the project is to oversee V. Murari's postdoc, focusing on aerosol composition and sources at the Dunkirk field campaign.

ALPAGA (2021 – 2022). Partners: IMT Nord Europe*. The project was a collaborative effort between atmospheric and computer scientists within IMT Nord Europe (Inter-CERI funding, 2021) to develop a platform for atmospheric observations based on drone technology. In addition to analytical developments inherent to sampling atmospheric constituents at trace level, the project has focused on challenges such as robotics, Internet of Things and Artificial Intelligence. Sampling systems were developed with either sensors or tedlar bags for VOC

sampling, particularly suitable in co-deployment with PTRMS systems. I was the co-PI of the project.

BIOMASP (2021 –). Partners: Université Clermont Auvergne*, University of Sao Paulo*, IMT Nord Europe, Federal University of Uberlandia, Université Grenoble Alpes, Université de Montpellier. This collaborative project between ANR and FAPESP (2021 – 2025) aims to study the impact of biosphere-atmosphere interactions on gaseous and particulate within the Sao Paulo Metropolitan Area. Specifically, the project focuses on ozone and particulate secondary formation, and their impact on health and the biosphere. The project includes flux measurements (CO_2, VOCs), ambient observations at urban and forested sites during extensive and intensive field campaigns, and 0-D and 3-D modelling. I am co-leader of task 3 (Bio-physico-chemical processes and ambient composition) and oversee IMT's instrumental deployment during the field campaign in Apr-Jun 2023.

ECRIN (2021 –). Partners: Université de Lille*, Université du Littoral Côte d'Opale, IMT Nord Europe, Inserm, Institut Pasteur de Lille, CHU Lille, Université de Picardie Jules Verne, Université de Technologie de Compiègne, Université Polytechnique Hauts-de-France, INERIS. The State-Region Plan Contract (CPER) ECRIN (2021 – 2027) includes 25 research laboratories in the Hauts-de-France region, with the objective of better understanding how climate and, more broadly, environmental changes (air quality, water, etc.) linked to human activities, affect our health and biodiversity. I coordinate sub-task 1.1 of the project, the evolution of the physico-chemistry of urban environments, which comprises five partners of the project and counts, at the moment, 24 ongoing and foreseen research activities.

SONATA (2021 – 2023). Partners: IMT Nord Europe*, Max Planck Institute of Chemistry - Mainz, University of Sao Paulo. Funded by LEFE-CHAT, the project aims at studying semi-volatile organic aerosols in the Amazon. This is based on ambient observations at the ATTO site, in Central Amazon, during the wet season, having one of the cleanest atmospheres in any continental site globally. Observations include a range of in-situ instruments focusing on gas and particle-phase instruments, whether part of ATTO ongoing measurements (from USP and MPI-C) or IMT's PTRMS coupled with an aerosol inlet (CHARON) in an intensive campaign in April-May 2022. I am the PI of this project.

UNREAL (2020 – 2023). Partners: ONERA*, Université de Versailles-Saint-Quentin, IMT Nord Europe, Université de Franche-Comté, Université de Lille, Université Paris-Est Créteil. Focusing on the atmospheric impacts of aircraft fuel composition, this ANR-funded project aimed at determining the mechanism behind particle nucleation in the aircraft engine exhaust and the physicochemical properties of corresponding emitted aerosol particles. The proposed methodology consists of using portable oxidation flow reactors, thoroughly validated in the project against well-established atmospheric simulation chambers. My role in the project consists of studying primary and secondary aerosol physicochemical properties and on the establishment of the sampling protocol.

ACTRIS (2019 –). Recently established as a European Research Infrastructure Consortium, this project combines more than 100 research institutions from 22 countries providing information on short-lived atmospheric constituents (trace gases, aerosol particles and clouds) and their associated processes. Combining in-situ, remote sensing and atmospheric modelling, the project currently handles over 130 variables from more than 90 different sites in a

homogenized and transparent methodology. Within ACTRIS, I am the PI of in-situ black carbon/aerosol light absorption measurements at the ATOLL site, in Villeneuve d'Ascq. Furthermore, since Feb-2023, I have been responsible for the Working Group on Aerosol Mass Spectrometers as part of the National Facilities Technical and Scientific Forum, aiming to integrate a larger community within and outside ACTRIS on this topic.

INVOC-Dust (2019 – 2020) Partners : IMT Nord Europe*, Université de Versailles-Saint-Quentin, Université Paris-Est Créteil. Focusing on the interaction between natural mineral dust and volatile organic compounds, this LEFE-CHAT-funded project (2018 – 2020) is based on direct sample experiments, simulation chambers and atmospheric modelling. My role was to take part in the experiments at the atmospheric chamber CESAM, particularly studying SOA formation and the corresponding modification of mineral dust physical properties.

COLOSSAL (2018 – 2022). Partners: Institute of Environmental Assessment and Water Research (Spain)*, Paul Scherrer Institute (Switzerland), INERIS, King's College London (UK), National Research Council-Institute for Atmospheric Science and Climate (Italy), University of Galway (Ireland), IMT Nord Europe, Vienna University of Technology-Institute of Chemical Technologies and Analytics (Austria), Norwegian Institute for Air Research (Norway), among many others. An EU-funded Cost action (2017 – 2022) with over 40 partner institutions, the project aims at optimizing and harmonizing fine atmospheric aerosol online measurements. More precisely, COLOSSAL aimed to coordinate overarching analysis to assess European-wise spatial and temporal variabilities, including seasonality, phenomenology and source identification. I participated in activities related to aerosol chemical composition (ACSM), AE33 intercomparisons and phenomenology studies.

CaPPA (2018 –). Partners: Université de Lille*, Université du Littoral Côte d'Opale, IMT Nord Europe. The Laboratory of Excellency of the Physical and Chemical Properties of the Atmosphere (LABEX CaPPA, 2012 – 2024) focuses on aerosols, in particular, their impact on the climate and their formation from precursor compounds. It supports laboratory studies as well as observations (ground, satellite). LABEX CaPPA has co-funded field campaigns (COBIACC, ACROSS) as well as O. Murana's PhD thesis. I participate in the work package WP1 "From gas phase to aerosols: biogenic VOCs as particle precursors", and WP3 "Aerosol observations: Instrumentation and intensive field campaigns".

CLIMIBIO (2018 – 2020). Partners: Université de Lille*, Université du Littoral Côte d'Opale, IMT Nord Europe, Inserm, Institut Pasteur de Lille, CHU Lille). The CPER CLIMIBIO (2015 – 2020) was a multidisciplinary environmental project involving 16 laboratories in the Hauts-de-France region. This project aims to study the evolution of environments and climate, to analyze the impacts of these evolutions on biodiversity, air quality, health, and society and to consider the perspectives and strategies of adaptation to these changes. My role on this project consisted of coordinating the COBIACC field campaign, in June-July 2019.

DACCIWA (2016 – 2018). Partners: Karlsruhe Institute of Technology*, CNRS, DLR, ETH, University of Manchester, Université Clermont Auvergne, Sorbonne Univeristé, Université Paris Diderot, Météo-France, Max Planck Institute for Chemistry, among many others. Funded by the European Union 7th Framework Program (2013 – 2018) to investigate the influence of anthropogenic and natural emissions on the atmospheric composition over South Western Africa and to assess their impact on human and ecosystem health and agricultural productivity.

A large project combining a range of fields and tools such as research aircraft, ground sites, satellite and atmospheric modelling. As a postdoc, my role was to conduct AMS measurements onboard one of the ATR-42 aircraft based at Lomé, Togo.

GoAmazon2014/5 (2014 – 2016). Partners: University of Sao Paulo*, National Institute of Amazonian Research, State University of Amazonia, Max Planck Institute for Chemistry, Harvard University, NOAA Earth System Research Laboratory, University of California, National Center for Atmospheric Research. This is the FAPESP-funded project (2013 – 2018), supporting several Brazilian partners within the GoAmazon project. It aimed to study how Manaus emissions impacted the otherwise pristine Amazonian atmosphere through in-situ and remote sensing, combining ground, research aircraft and satellites. As a postdoc within the project, I was responsible for the T2 site (near Manaus), and also several aerosol physical-chemical measurements upwind (T0z, T0a) and within Manaus (T1).

ATTO (2012 –). Partners: National Institute of Amazonian Research (INPA)*, Max Planck Institute for Biogeochemistry*, Max Planck Institute for Chemistry, University of Sao Paulo, State University of Amazonia, IMT Nord Europe, Federal University of Uberlandia, among many others. The Amazon Tall Tower project started in 2011 as a 60m tower in Central Amazon, about 170 km upwind of Manaus, aiming at studying biogeochemical processes in the Amazon, particularly ecosystem ecology, meteorology, trace gases, and aerosols. Enhanced in 2015 by a 325m tall tower, the site has been a reference point as Central Amazonia's pristine site, integrating a number of other large-scale projects. I participated in this project as responsible for aerosol composition and size distribution (2013 – 2015), and more recently on semi-volatile organic aerosol observations within the intensive observations of the SONATA project.

FONTES (2011 – 2013). Partners: University of Sao Paulo*, Pontifical Catholic University of Rio de Janeiro*. Funded by PETROBRAS (2011 – 2014), this project aimed at quantifying the role of vehicular emissions on air quality degradation in Sao Paulo and Rio de Janeiro. This was based on a multi-site approach, through intensive and extensive observations, combined with dynamometer bench experiments and atmospheric modelling. The project has strongly focused on the specificity of fuel composition (largely ethanol) in those megacities. Hired as a researcher within the project, I coordinated the sampling sites in Sao Paulo and participated in the dynamometer test bench essays conducted in Rio de Janeiro.

SAMBBA (2012 – 2014). Partners: University of Manchester*, National Institute of Space Research (INPE)*, University of Sao Paulo, University of Leeds. Based in southwestern Amazonia (Porto-Velho in the deforestation arc), this project aimed at detailed characterization of the physicochemical properties of biomass-burning aerosols and their corresponding climatic and health impacts. Funded by the NERC-FAPESP partnership (2011 – 2014), SAMBBA is based on a combined approach of ground site, research aircraft (UK's BAE-146), satellites and atmospheric modelling, centred on a field campaign that took place in 2012. My role was to coordinate ground-site observations in Porto-Velho and to conduct PTRMS measurements onboard the BAE-146.

2.5 Publications and presentations

Since my first publication as a post-doc, about a decade ago, I count 72 published peer-reviewed articles in international journals, of which 16 as first or second author. Those have received 2964 citations according to Web of Science, leading to an H factor of 32. Whereas many of those publications have been the result of multi-institution collaborations, I am the first or second author of half of my top-ten cited articles[1]. Conversely, I count 34 presentations at international conferences as first or second author, out of over 120 in total, two invited presentations and 1 book chapter. The complete list of publications and communications is provided in Appendix B.

Figure 2.5.1 depicts the geographical location of citations related to my work, showing to be fairly well distributed across the globe, with higher concentrations around Europe and US, and to a lesser extent China, and Brazil. This is probably due to the combination of total scientific output per region, compounded by a collaborative network with the US (from GoAmazon2014/5), followed by a series of EU-funded projects. Figure 2.5.2 shows a Beamplot from the Web of Science, with year-to-year citations compared against benchmarks. Overall I am placed at the 79th percentile, with first-author publications ranging from the 61st to the 92nd percentile.

Figure 2.5.1 – Geographical distribution of citations according to Web of Science.

[1] https://scholar.google.com/citations?user=5z7UnosAAAAJ&hl=en

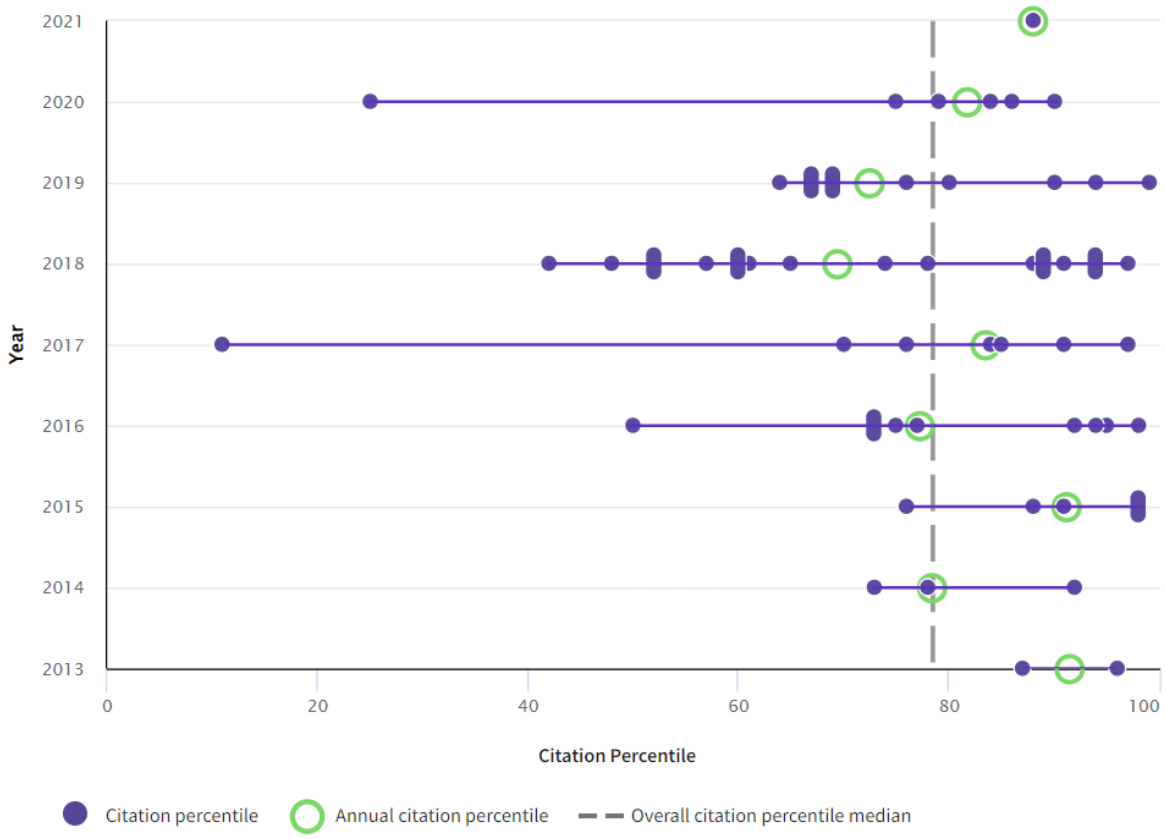

Figure 2.5.2 – Web of Science's Beamplot from 2013 till 2021 (last available year). Citation percentile calculated against a benchmark of "similar" articles. Blue markers indicate individual papers, green markers the annual percentile and grey dashed line the overall citation percentile (79th currently).

2.6 Review and editorial activities

For the past decade, I reviewed 44 manuscripts for 14 different journals, having an average of about 4-5 per year, a rate which I aim to keep despite the increase in responsibilities and diversification of activities in recent years. Manuscript reviews have been for the most part for journals in which I have authored papers, such as Atmospheric Chemistry and Physics, Atmospheric Environment, Environmental Science and Technology, with recent experience on Nature. The weighted distribution of the journal's impact factor is 6.4. The list of journals, completed with the current impact factor and number of reviews is provided in Appendix B. Furthermore, I have been requested to review 8 projects for different funding agencies, ranging from Horizon Europe Marie Curie Actions, European COST Action, DIM Qi2, LEFE-CHAT and large-scale FAPESP Thematic projects. The latter is quite demanding, with requests for yearly report validation through its 5-year duration, and positive evaluation conditioning the receiving of funds. In addition to manuscript and project reviewing, I am part of the editorial boards of Atmosphere and Toxics, both from MDPI. This has allowed me to decide the outcome of 12 manuscripts so far, on the topic of atmospheric chemistry and physics. Despite being quite a time- and energy-consuming activity, I find it very rewarding to participate in the advancement of responsible and high-quality scientific production, may it be through reviewing or editorial work.

2.7 Teaching activities

2.7.1 Lectures

As a post-doc at the University of Sao Paulo, I was invited to participate in the teaching activities of the physics institute, joining my colleagues from the Atmospheric Physics department. They required support for teaching Electromagnetism to 2nd-year physics and meteorology students. The course lasts for 1 semester and includes twice a week theoretical classes, and once a week laboratory. Whereas the activity was very enriching, I could not repeat it for other semesters given that FONTES was ramping up and responsibilities would prevent me from continuing such teaching loads. In 2012 and 2013, I also participated in the IFUSP summer school, teaching modules on aerosol particles and atmospheric chemistry.

At IMT Nord Europe, I teach Introduction to Fluid Mechanics to third-year engineering students. The course, developed by Prof. Jean-Luc Wojkiewicz, lasts for about 6 weeks and follows a hybrid mode, sharing lectures, tutorials (*travaux dirigés* in French), and a MOOC with videos and quizzes. The MOOC is made available to a wider audience during the period of the course and attracts usually about 3000 students. The tutorials are based on a serious game (Mission à Emosson) developed in-house. There, using their computers, the students take the role of an engineer in a dam in Switzerland, facing challenges associated with the industrial setup under the scope of fluid mechanics. Due to the large number of students, up to 340, the tutorial sessions need to be repeated several times (8x2h weekly or more) to ensure a close follow-up and active pedagogical methods (e.g. flipped classroom). Furthermore, the course had to be adapted over the years due to changes in the school's curriculum, whether been offered to all undergrad students, or (only in 2019) limited to students majoring in the energy & environment option. In 2021 we added a support module in vectorial calculus and kinematics to have a more levelized classroom in mathematical tools. I also participated in the meantime at the development of a new serious game (GRAIL), using virtual reality, to complement the currently used one.

In addition to the course described above, I have often engaged in other teaching activities such as the development of MOOC in Air Quality (2020), laboratory classes on Electromagnetism & Optics for 2nd year engineering students (2021), the module of Ecological and Social Transition (TES) for 3rd year students (2022, 2023) and Air Pollution Dispersal and Industrial Risks (2023). Concerning Air Quality MOOC, I created a video about atmospheric observation methodologies, particularly focusing on deployable mass spectrometers. The videos are being finalized and should be ready in the near future to be available to the public. The laboratory classes (about one afternoon per week, for about 5 weeks), lacked personnel in 2021, so I participated sporadically in that class. In the TES module, I participated in tutorials on “environmental controversies”, aimed at stimulating our students to reflect upon, often oversimplified, complex topics. Those revolve around the role of technology and the economy in solutions towards a sustainable future, and the role that future engineers can have. In 2023, in addition to tutorials, Dr Emmanouil Romanias and I created a module on climate change attenuation, ranging from discussions on carbon capture and storage, geoengineering, biofuels and so forth. The course Air Pollution Dispersal and Industrial Risks was aimed at 4th year students combining theory and hands-on activities on Gaussian and Lagrangian models using ALOHA and HYSPLIT.

2.7.2 Supervision of undergraduate students

I have supervised four types of undergraduate activities at IMT Nord Europe: "Introduction to Science" (*Projet de Recherche*), "Extension Program" (*Projet Ouvert*), "Work-study Program on Energetics" (*Alternance*) and "Corporate Internship" (*Stage en entreprise*). Below I present a short description of the activity and the activities that I supervised.

Introduction to Science

This activity aims at introducing the engineers to the scientific method, often exposing them (within the SAGE department) to atmospheric research, whether via data analysis or through some experimental work. The role of the supervisor is to propose a topic, teach the methodology and accompany the students in their development and achievements for about a month.

2019 – "Modelling the sources of atmospheric pollution" with Dr Esperanza Perdrix. The project consisted of making use of source apportionment methods for a range of different atmospheric pollutants from a field campaign that took place previously in Douai.

2020 – "Measuring Particulate Matter in the atmosphere using a drone", with V Riffault and J-L Wojkiewicz. The students should adapt a nacelle into a drone from IMT, and deploy a NOVA PM SDS011 for measuring particulates.

2020 – "Investigating the geographical origin of atmospheric pollutants during the field campaign COBIACC of summer 2019" with A. Bourin and E. Stratigou. This project consisted of combining ambient observations with back-trajectories using the ZEFIR software, to identify potential region sources of different pollutants. They would work with the COBIACC field campaign data.

2021 – "Development of an innovative sampling system for drone measurements", with T. Salameh, L. Fayad and Dr Titouan Verdu. Continuation of drone project from the previous year, that had to be halted due to COVID pandemic. The objective was to develop a nacelle for multi-sensor atmospheric sampling (PM, CO_2 and VOCs). Completed with Arduino programming, the students performed some flights demonstrating the capacity of the system to work to identify pollution plumes.

Extension Program

The Extension Program has a strong component of teamwork and hierarchy, combining engineering issues with general applications, be it resulting in a commercial product, a specific application or raising awareness for specific topics. Here, the students propose a project and work almost independently for about 4 months. The role of the supervisor is to ensure that activities are compatible with expectations, manage potential conflicts, and evaluate the report and final presentation.

2022 – I supervised a group of five students aiming at developing activities with a class of 11-year-old students, particularly those from ULIS (including developmental disabilities). The

objective was to perform activities in space, such as exposing objects to a vacuum or launching water-pressured rockets.

2023 –Working with a classroom of 13-year-olds, the five-student group has performed activities towards raising awareness on climate change and air pollution. Their activities included visiting a recycling centre, clean walks, climate collage game and debates.

Work-study on Energetics

The "Work-study on Energetics" is an undergraduate program in which students share their time almost equally between lectures at IMT Nord Europe, and working as a trainee engineer in a company. The role of the supervisor is to have regular meetings with the employer, read and correct the regular reports, and ensure that both the student and company are respecting the overall objectives of the program. The Work-study program lasts for three years.

2022 – I am responsible for a student working on thermal comfort in buildings. His activities include the calculation of the return on investment of building insulations, solar panels and heat pumps.

2023 – Within the field of renewable energy, the student focuses on photovoltaic and wind power installations. His main activities consist of feasibility analysis, customer relationship and project liaison.

2023 – At a company whose expertise lies in thermal comfort in buildings, the student activities include the cost calculation of renovation or construction work for a range of structures and buildings, including buildings, and car parks among others.

Corporate internship

As faculty members of IMT Nord Europe, we are responsible for overseeing 2-3 students every year during their "Corporate Internship". This means following their activities during the internship (about 2 to 6 months) through meetings and evaluating their reports and final presentations.

2019 – Three third-year students with an internship lasting about 3 months.

2021 – Three third-year students with an internship lasting about 3 months and one fourth-year student with a 6-month internship.

2022 – Three third-year students with an internship lasting about 3 months and two fourth-year students with a 6-month internship.

2023 – Two third-year students with an internship lasting about 3 months and two fourth-year students with a 6-month internship.

2.7.3 Innovative methods

In recent years I have used a number of innovative pedagogical methods including online support through MOOCs (Air Quality, Fluid Mechanics), as well as active pedagogical tools such as serious games and flipped classrooms (Fluid Mechanics) and debates (Ecological and Social Transition). In addition to significantly increasing student engagement, those methods have shown consistently that, combined with lectures, they enhance significantly information retention. Particularly, methods based on collective intelligence, such as Mission à Emosson, Climate Collage or debates, have shown to be a particularly efficient teaching tool, with the caveat that it is not scaled up easily. This is particularly challenging at IMT Nord Europe following the increase in recent years of ingressing students from 140 in 2018 to 340 in 2022. To handle this strong expansion, in addition to multiplying the number of classes to be taught, we had to modify the evaluation method. During my first year, Fluid Mechanics evaluation was based on fixed online quizzes (5% weight), group reports (45% weight) and one final written exam (45% weight). To ensure that the evaluation was representing accurately individual knowledge from different students, and to be able to handle the ever-increasing number of students, I started developing 2019 a "randomized" multiple choice quiz. Using Matlab, I started creating a library of randomized questions. Those variations were either numerical, changing randomly the diameters, velocities, densities and so forth, or symbolic, via creating unique equations and functions for specific problems. This allowed us to ensure that each student had a unique set of questions, following a pre-defined distribution of topics. The new system replaced then the written exam, providing instantaneous correction/feedback and distributing the effort of creating new questions for the library throughout the year. Following requests by colleagues, the package has been adapted into a shareable format, requiring a Moodle platform and, at least for now, a Matlab licence. I plan to have it transposed to an open-source code such as Python to increase its potential reach. IMT organizes a pedagogical workshop yearly (termed MEDIANE) which I participated twice, the first one in 2019 I learned different methodological teaching tools, and the second time, where I gave a seminar on the randomized multiple choice quiz, and co-organized a two-hour workshop on flipped classrooms.

2.7.4 Examinations

As described above, I have worked with several evaluation methods, including written exams, group reports, debates, projects, presentations, and quizzes (static, or the recently developed randomized method). Those evaluations concerned mostly 2nd or 3rd-year IMT Nord Europe students, or the general public when performed within a MOOC.

2.8 Administrative and Community Responsibilities

Internal

As a postdoc at USP and LaMP, I was (co-)responsible for organizing regular (bi-weekly) seminars, aiming at increasing the exchange within the department and providing a platform for junior researchers to present practice talks before attending scientific conferences. Continuing along the same lines, at IMT Nord Europe, from 2019 to 2022 I led a weekly round-table with graduate students and post-docs focusing on paper writing. The idea was that each week, one person would pick a scientific publication in their field, present the science, and provide a critical analysis of the writing style, figures, referencing, and so forth. Publications from Atmospheric Chemistry and Physics (or other open-review journals) were encouraged, providing the students had a chance to evaluate their assessment with those of experienced reviewers. Although it was an enriching and useful experience, due to a busy schedule I had to put a halt on those meetings for the time being.

In early 2019 I took the role of School Representative for Latin America within the Department of International Relationships and Academic Partnership. Whereas it was quite active until the pandemic, afterwards exchanges and visits were shut down, and now it is slowly ramping up once again, culminating with the student/research exchange IDE BRAFITEC program approval in late 2023, and several other activities currently in preparation.

In January 2024 I was nominated coordinator of a Research Axis within the CERI Energy and Environment, termed Observations, Sources, and physico-chemical Processes of the Atmosphere (OSPA). This is the largest of the three Research Axis of the centre, combining about 50 participants (of which 14 faculty members). Responsibilities consist of ensuring optimal communication and work dynamics within the axis, as well as integrating the "COPIL elargi", a coordinating body of the CERI. Furthermore, within IMT Nord Europe, I participated in several admission committees, whether for undergraduate students (about two consecutive days in Paris, interviewing about 10 applicants per day), or personnel, notably the hiring procedure for an engineer position within our department.

External

The list below summarises a range of activities that I performed over the years identified as administrative or community responsibilities.

2023 – Participant on the French Committee of the new state-of-the-art satellite mission Atmosphere Observing System (AOS), funded by CNES, NASA, and JAX. I integrate the working group of Aerosol Particles. Piloted by Juan Cuesta, with the participation of Laaziz El Amraoui, Oleg Dubovik, Michael Sicard, Cyrille Flamant, and Solène Turquety.

2023 – Responsible for ACTRIS National Facilities Technical and Scientific Forum on "Aerosol Mass Spectrometry". The group counts today 17 participants.

2022 – Participated in the Working Group "Clouds and Aerosols in situ" for the preparation of the new research aircraft from SAFIRE. Piloted by Paola Formenti, participation of Alfons Schwarzenböck, Frederic Burnet, and Cyrielle Denjean.

2021 – Co-coordinator of sub-task 1.1 of the CPER ECRIN “The evolution of the physico-chemistry of urban environments”. Co-coordinated with Isabelle Chiapello.

2019 – Co-Convener at European Geophysical Union general conference, session “Atmospheric composition, weather and climate in Sub-Saharan Africa” with Peter Hill, Luis Garcia-Carreras, and Peter Knippertz.

2014 – Participated in the “Volatile Organic Compounds in the Amazon” Working Group, a coming together of atmospheric scientists to discuss observations and measurement techniques involving VOCs under the challenging conditions of the Amazon rainforest. With Kolby Jardine, Emily House, Ana Maria Yanez-Serrano, among 10 others.

3 Research on atmospheric sciences

This section describes research activities that I developed since my PhD. The thread revolves around aerosol particles, focusing on source identification, their properties, and impacts. This Chapter focuses firstly on aerosol source apportionment and characterization on two distinct regions (urban and Amazonian Basin, chapters 3.1 and 3.2, respectively), and then the impact of coupling anthropogenic and biogenic emissions (chapter 3.3). The discussion is mostly based on published work that I led, supervised, or closely collaborated on, highlighting my role in the scientific development of the respective topic.

3.1 Identification and characterization of aerosol particles in urban environments

Much of my research on urban aerosols has focused on two contrasted regions, Sao Paulo, Brazil, and the Hauts-de-France region. For simplicity I have separated the scientific activities of each urbanized area, compiling the scientific context, corresponding actions, and their major outcomes.

Widespread biofuel usage in Brazil

Brazil, in particular the São Paulo Metropolitan Area (SPMA), is characterized by a uniquely high proportion of biofuel usage by road vehicles, especially ethanol from sugarcane. Ethanol was first introduced as a commercial alternative to fossil fuels in Brazil in the early 1980s and its use has strongly increased since the 2000s with the introduction of flexible fuel vehicles (FFV). The FFVs are capable of using any rate of anhydrous ethanol from 20-25 % (mixed with gasoline, also termed gasohol) to pure hydrous ethanol. In the early 2010s, when I first started studying the effects of such a unique fleet on the SPMA's atmosphere, the consumption of gasoline and anhydrous/hydrous ethanol in the Sao Paulo state had a roughly 52% to 48% ratio, a value that remains up to nowadays (EPE, 2022).

To better constrain this unique source profile and improve the understanding of its effects on atmospheric reactivity and composition (particularly on pollutant levels), I worked with a combination of both direct source sampling and ambient measurements. The former was conducted mainly by dynamometer test bench and tunnel measurements, whereas the latter was based on extensive (filter) and intensive (1-3 months) sampling of different sites across the SMPA. Despite strong scientific complementarity between dynamometer test benches and tunnel observations, the former was conducted within the scope of a FONTES (described in project Chapter 2.4), at their R&D centre in Rio de Janeiro, and their results were not allowed to be made public unfortunately. Conversely, tunnel measurements were led by Prof. Maria de Fatima from the Institute of Astronomy and Geosciences (IAG) at the University of Sao Paulo, at two different locations within the SPMA. Janio Quadros (JQ), in the downtown area, being almost exclusively impacted by light-duty-vehicles (LDV, mostly passenger cars), whereas Rodoanel (RA) is a beltway around greater Sao Paulo with significant traffic of heavy-duty vehicles (mostly large trucks). Those experiments, published by Brito et al. (2013), have allowed detailed aerosol particle characterization including elemental and bulk composition, optical properties, and size distribution (Figure 3.1.1). The latter depicts the strong dominance

of ultrafine particles (UFP), with geometrical mean diameters of 48.4 and 38.7 nm, for LDV and HDVs, respectively. Results have also identified the large role of carbonaceous aerosol on both fleet types (ranging from 60% for LDV up to 91% for HDVs), and notably their high Elemental Carbon (EC) content. This can be analysed with their ratio with Organic Carbon (OC), yielding OC:EC ratios of 1.6 (LDV) and 0.5 (HDV). The OC:EC ratio of LDV within the SPMA was considerably lower than the value of 2.4 for LDV in the US, for example (Landis et al., 2007). This result showed a strong contribution of highly absorbing aerosols despite the significant biofuel content, resulting in a Single Scattering Albedo (SSA), namely the ratio of light scattering by light extinction, of 0.5 at 637nm.

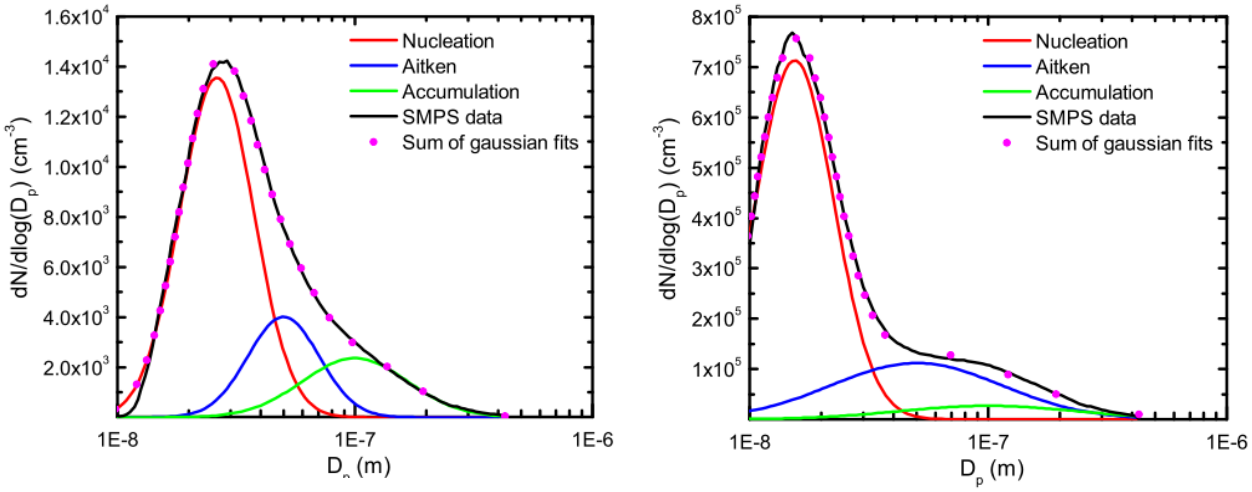

Figure 3.1.1 – Average aerosol size distribution in the JQ (left, dominated by passenger vehicles) and RA (right, dominated by large trucks) tunnels. Reproduced from Brito et al. (2013).

As part of the FONTES project, I oversaw the installation and atmospheric sampling on four sites within the SPMA (Figure 3.1.2, top), for at least 1 full year of continuous filter sampling, combined with intensive observation periods of online instrumentation. The filter analysis of carbonaceous aerosols provided interesting insights into their dynamics and heterogeneities throughout the city. This was the topic of the MSc project that I supervised, carried out by Djancinto Junior Monteiro dos Santos at the Physics Institute of the University of Sao Paulo. The study was based on OC and EC estimated from thermal–optical transmittance analysis, and calculation of secondary organic carbon from variations of OC:EC ratios (e.g. Pio et al., 2011). This combined analysis of four sites yielded interesting analysis, identifying sites as being impacted by comparable carbonaceous populations, except at the Street Canyon, shown in Figure 3.1.2 (bottom). The study has identified the Park and University sites as having OC:EC ratios of 1.9, Downtown as 1.6, and Street Canyon as 0.6 (Monteiro dos Santos et al., 2016). Those results could be compared with tunnel measurements from Brito et al. (2013) indicating that at the downtown site, the OC:EC fractions were comparable to primary LDV, whereas the Street Canyon site had a ratio comparable to HDVs. Conversely, Street Canyon had the lowest fraction of secondary organic carbon (27%), while the other three sites ranged around 40%. This study has also focused on different fractions of OC and EC, estimated according to their characteristic temperature during filter desorption. Although inherent uncertainties are associated with the apportionment of those different fractions (as they might be concentration dependent), interesting results have been obtained for organic pyrolyzed carbon, i.e. the fraction of organics that chars typically above 300°C. This fraction has shown a good temporal correlation between sites, except

Street Canyon, and at the same time had a poor correlation with EC, suggesting a potential biogenic origin, a correlation that has also been suggested previously in the literature.

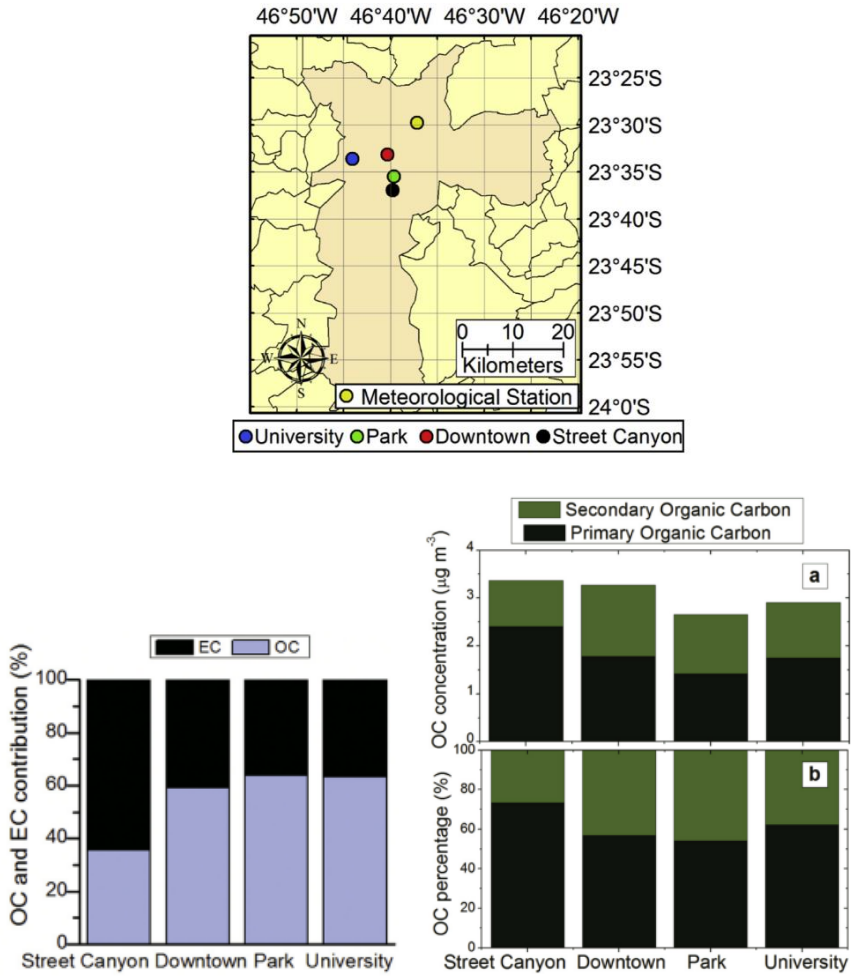

Figure 3.1.2 – Top: Location of four sampling sites for multiple studies of the role of traffic emissions on air quality within the SPMA (plus one meteorological station). Bottom: Average percentage contribution of OC and EC fractions to Total Carbon (left) and estimated Primary and Secondary Organic Carbon (right) concentration (panel a) and relative contribution (panel b). Reproduced from Monteiro dos Santos et al. (2016).

At the University site, I collaborated with Prof. Gerson Almeida, from the State University of Ceara, to characterize aerosol chemical and hygroscopic properties. Those studies were based on my first deployment of the recently acquired ACSM, providing non-refractory submicrometric (NR-PM_1) aerosol chemical composition, in combination with Cloud Condensation Nuclei (CCN) counter (Droplet Measurement Technology). The former volatilizes aerosol particles between 0.1 and 1 µm at 600°C, and ionizes them by 70eV electron impact, being quantified on a quadrupole filter mass spectrometer. The latter counts the number of aerosol particles that "activate", i.e., become a water droplet, by accurately controlling water supersaturation levels through temperature gradients, coupled to an optical particle counter. The droplet activation depends on chemical composition as well as size distribution – the higher the supersaturation level, the more easily smaller aerosols can activate. The objective of the study was to model CCN concentrations based on size distribution and chemical composition. This is

crucial to the understanding of the cloud-aerosol interactions, no studies had been performed at a site so largely impacted by biofuel usage. The results have been shown to work reasonably well with higher supersaturations (R^2 of 0.79, slope 1.03, at 1.13% supersaturation), but struggled at lower levels (R^2 of 0.43, slope 1.22 at 0.23% supersaturation, Almeida et al., 2014). This was associated mainly with poor knowledge of particle mixing and dynamics, notably depending on the hour of the day and the role of injecting fresh particles from traffic in the atmosphere (Figure 3.1.3).

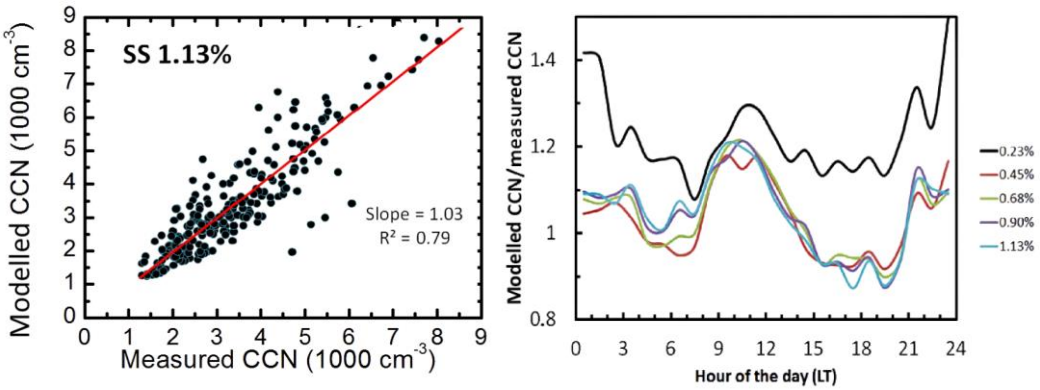

Figure 3.1.3 – Comparison of modelled and measured CCN concentrations at 1.13% (left) and daily variability of modelled/observed CCN concentrations for 0.23, 0.45, 0.68, 0.90 and 1.13% supersaturations (right). Reproduced from Almeida et al. (2014).

Another interesting analysis at the time arose from a collaboration with Prof. Alberto Salvo, from Northwestern University. As an economist, he was analysing how shifts in international sugar prices impacted ethanol usage as biofuel in Brazil. Given that its prices at the pump were not as tightly controlled as fossil fuel, the Brazilians, notably in Sao Paulo State, were used to large price swings over short periods, typically weeks or months. Furthermore, the extensive usage of FFVs allowed consumers to choose at the pump the fuel to consume (ranging from 25% as gasohol up to pure hydrous ethanol). Based on surveys at pump stations, data from oil and sugar industries, coupled with sophisticated economic models, allowed us to use ethanol usage as constraining parameters for atmospheric pollutants conducted at the University site (Salvo et al., 2017). The main results, shown in Figure 3.1.4, depict that increasing gasohol from 30 to 80% in volume (or decreasing ethanol from 70 to 20%) is estimated to lead to a marked increase in the ambient concentration of UFP (defined as particle concentration from 7 to 100 nm) of about 8 700 cm^{-3} and a decrease in Ozone of 8.3 ppb. We interpreted the decrease of the latter as a result of increased nitrogen oxide emissions (with increasing gasoline content) in a VOC-limited regime. Conversely, it was interesting to note the increase in UFP with gasoline usage, but no statistical differences in $PM_{2.5}$ (particulate matter smaller than 2.5 µm), or Black Carbon (BC). The lack of a clear impact of LDV fuel composition on ambient levels of those two parameters was interpreted as i) dominance of HDV on BC levels, in agreement with results from Brito et al. (2013), and ii) probable impact of other sources on $PM_{2.5}$ in addition to LDV tailpipe emissions, like road resuspension, industries, HDVs, natural sources, etc.

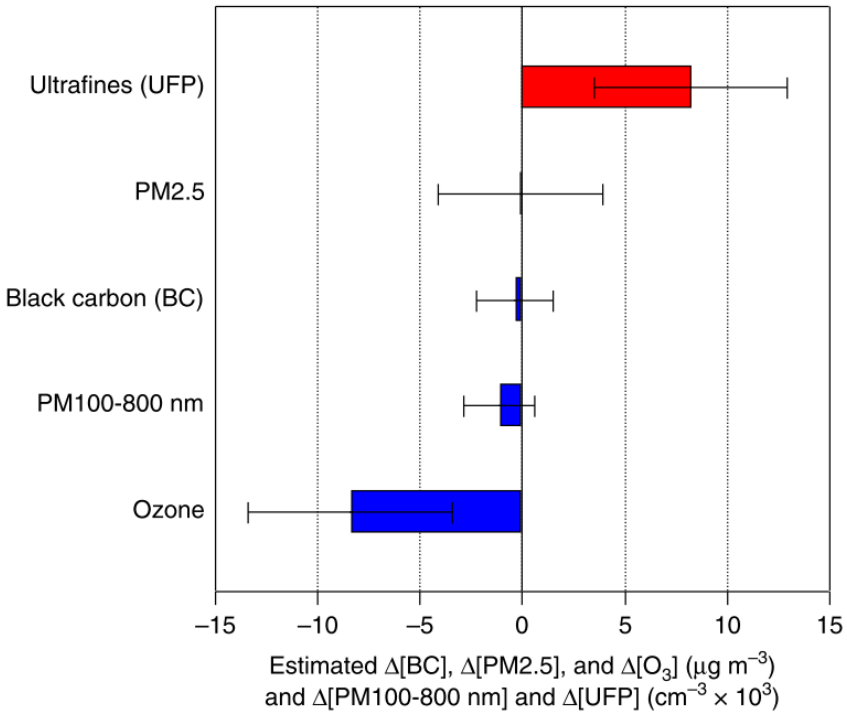

Figure 3.1.4 – Effect of atmospheric concentration in the SPMA as the gasohol share in the flex-fuel fleet rises from 30 to 80 percentage points. Reproduced from Salvo et al. (2017).

In addition to my work on aerosol particles, as a post-doc at the University of Sao Paulo, I developed the research at the group focusing on Volatile Organic Compounds (VOCs), notably with the usage of a Quadrupole Proton-Transfer-Reaction Mass Spectrometer (Q-PTRMS, Ionicon). The latter is a chemical ionization mass spectrometer, using H_3O^+ as a primary ion, providing typically low fragmentation of their analytes through soft ionization. This instrument was aimed at improving understanding of particle precursors (and general reactivity) in Sao Paulo's atmosphere, but also in aiding particle source apportionment studies. Together with ACSM (among other instruments), I deployed the Q-PTRMS at the University and Downtown sites during intensive campaigns. The first one focused on characterizing VOC emission ratios (ER) associated with anthropogenic sources (via correlation with carbon monoxide, CO, during nighttime), allowing to place SPMA in context with other megacities worldwide. This led to the identification, for example, that methanol, acetaldehyde and acetone showed ERs 2-5 times higher in SPMA compared to Mexico City, contrary to other anthropogenic VOCs such as benzene and toluene, which had comparable ER (Brito et al., 2015). Furthermore, the quantification of ER relative to CO allowed us to estimate the biogenic/secondary component of those VOCs, as shown in Figure 3.1.5. The fact that aromatics species (benzene, toluene and C8 aromatics) were almost entirely apportioned to Primary Anthropogenic (PA) was interpreted as result of three aspects: i)homogeneity of sources impacting the site throughout the day (as ER were derived over nighttime), i) negligible fraction of fuel evaporation, and iii) most of aromatics were emitted relatively close to the site, given that CO lifetime largely exceeds those species. Conversely, other species have shown a significant fraction of biogenic/secondary species such as Methanol (associated with significant natural sources), as well as Acetaldehyde and Acetone (secondarily formed from photochemical processes) and isoprene (biogenic

primary emissions). The latter - or at least the compounds contributing to the signal in which isoprene was detected - was estimated to have roughly the same contribution of biogenic and anthropogenic origins at the University site.

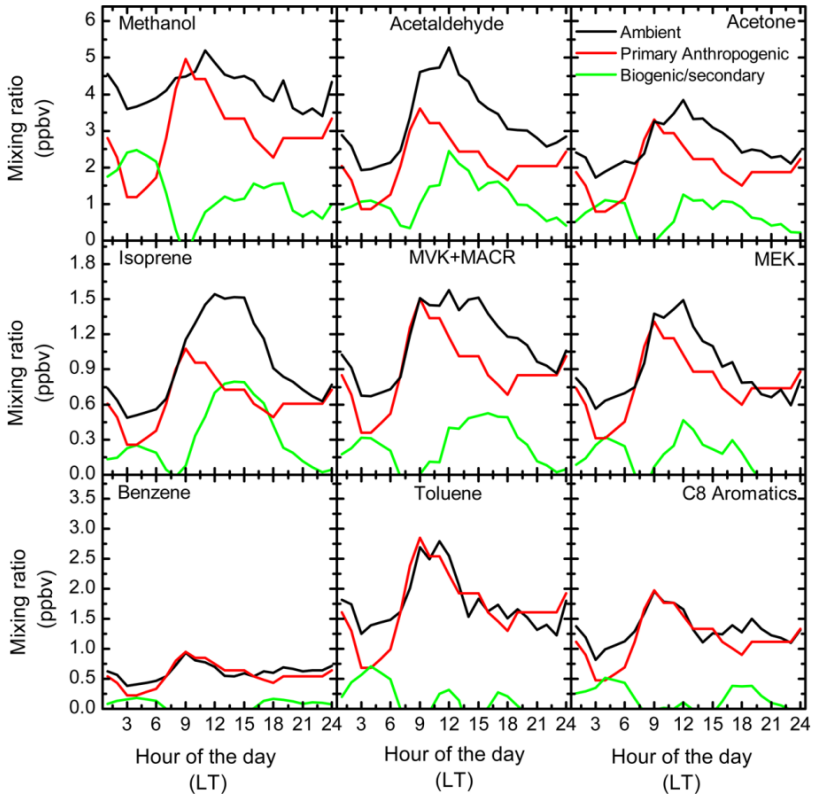

Figure 3.1.5 – Daily profile of mixing ratios of selected VOCs, separated into the components apportioned to primary anthropogenic (red) and Biogenic/secondary (green). Reproduced from Brito et al. (2015).

Still working on source apportionment, at the Downtown site I focused on Organic Aerosols (OA) via Positive Matrix Factorization (PMF), particularly targeting to separate LDV, HDVs and secondary components. This was possible due to the uniquely high usage of biofuel by LDVs, using ethanol as a tracer for passenger vehicles (Brito et al., 2018). The traffic-related factors were termed Hydrogenated Organic Aerosols (HOA), and two aged/secondary factors were combined as Oxygenated Organic Aerosols (OOA). At the site, due to the large contribution of traffic, both HOA factors contributed to over 47% of OA (28% and 19% for LDV and HDV, respectively), with the rest apportionment to OOAs. Once the OA components were identified, they were then used as independent variables on multiple linear regression analyses for a range of pollutants, allowing the construction of a source attribution that could be directly compared with emission inventories, as shown in Figure 3.1.6. Among interesting results is the important role of HDVs on BC, toluene and benzene levels, for example. Considering that HDV represents only 5% of the fleet throughout the city – and in the Downtown area they are mostly municipal buses, the potential for source mitigation for those species is quite high within the SMPA. The analysis also corroborated emission inventories where most of the CO originates from LDVs (estimated here at about 70%), and the comparable

contribution of acetaldehyde from secondary processes as well as from traffic, as reported by Brito et al. (2015) and associated here to LDVs, as expected from their high ethanol content.

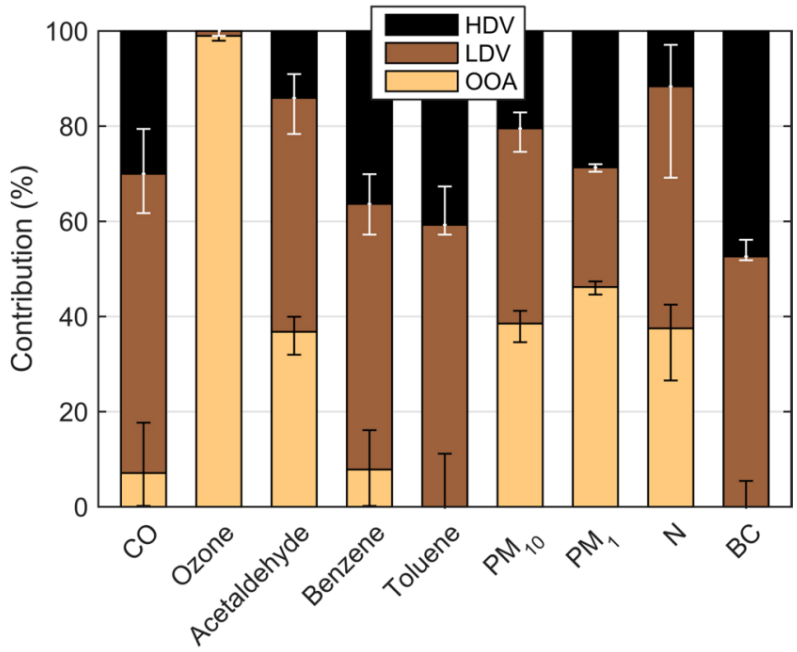

Figure 3.1.6 – Relative contribution of primary (LDV, HDV) and secondary sources (OOA, the sum of OOA-I and OOA-II). Range bars represent the 5th and 95th confidence intervals. Reproduced from Brito et al. (2018).

More recently, already as an Assistant Professor at IMT Nord Europe, I integrated the BIOMASP^{+} consortium, led by Dr Agnes Borbon (LaMP) and Prof. Adalgiza Fornaro (IAG-USP), in which I am the scientific coordinator at IMT. The project, described in Chapter 2.4, aims to study the SPMA atmosphere, particularly focusing on characterizing emissions from the native *Mata Atlantica* biome. This is based on duplicated measurements at the University site and the newly installed Morro Grande, a forested site on the outskirts of SPMA. The project comprises VOC branch levels emission fluxes, an intensive field campaign (a component which I co-lead in the project), extensive filter measurements and atmospheric modelling. The intensive measurements are based on aerosol and gas-phase measurements, with a particular interest in SOA formation processes, through the deployment of PTRMS coupled with a CHARON (CHemical Analysis of aeRosol ON-line) aerosol inlet. This instrument allows for switches between VOC measurements, and semi-volatile organic aerosol particles (volatilization at 140°C) below 1 µm. Our instrument was deployed at the University site and was operated by PhD student O. Murana (presented in Section 2.2). The PhD project aims at contrasting VOCs and semi-volatile aerosol composition between SPMA and Paris. Observations took place in the summer of 2022 (Paris, ACROSS project) and spring of 2023 (SPMA), Figure 3.1.7 depicts preliminary daily variabilities of isoprene and its oxidation products at the University site during the BIOMASP^{+} field campaign. In addition to characterizing the urban atmosphere, and semi-volatile organic aerosols at the University site, the PhD thesis focuses strongly on anthropogenic-biogenic interaction, which will be explored in depth in Chapter 3.3.

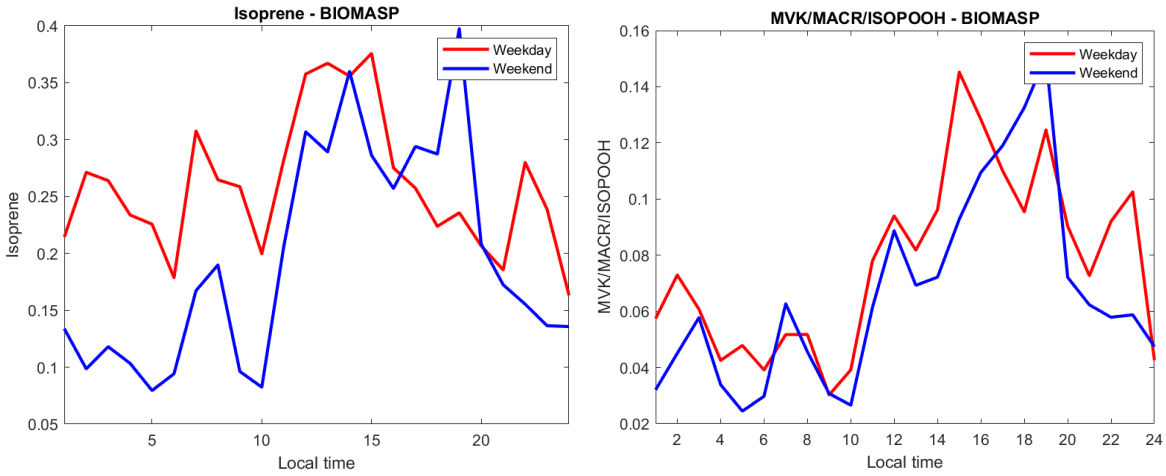

Figure 3.1.7 – Median concentrations in ppb of isoprene (left) and its oxidation products MVK+MACR+ISOPOOH (right) for weekdays (red) and Sundays and public holidays (blue). Data from the BIOMASP+ intensive campaign from April 21st till May 26th 2023 at the University site.

The Hauts-de-France region

The Hauts-de-France region, in the north of France, has over 6 million inhabitants and is the second most densely populated after the Parisian region of Île-de-France. Furthermore, the region combines higher than national average emissions of both agricultural and combustion (industrial, transport) pollutants, such as ammonia (NH_3) and nitrogen oxides (NO_x), respectively (Hauts-de-France, 2020). The surface averaged NH_3 emission in the HdF region is 15.7 kg ha^{-1} (11.4 kg ha^{-1} nationally), and for NOx is 32.3 kg ha^{-1} (17.5 kg ha^{-1} nationally). Furthermore, the industrial sector's contribution to NOx emissions is twice the national average (21.7% and 10.7%, respectively). In addition to locally emitted pollutants, the HdF region is located at the crossroads of air masses transporting pollutants from a range of locations, including short, medium and long-range origins such as Paris, Benelux, UK, Germany and Eastern Europe (Waked et al., 2018).

To better understand the sources and characteristics of aerosol particles in this region, I have led or participated in several research efforts in the region, whether through intensive, short-period campaigns or through extensive measurement periods. The latter notably at the ATOLL (Atmospheric Observations in Lille) site, at the top of the P5 building from the University of Lille, in Villeneuve-d'Ascq. The site is located 6 km southeast of Lille's downtown area, without significant local aerosol sources. Aerosol in-situ observations started at this site in 2014, with continuous measurements of aerosol composition in 2016 by V. Riffault. Those measurements were based on the deployment of a quadrupole ACSM and an Aethalometer (AE33, Magee), the latter becoming my responsibility as Principal Investigator (PI) in 2021. The site integrated at that time the French ACTRIS (Aerosols, Clouds and Trace gases Research Infrastructure), and is currently undergoing the process of integrating European ACTRIS as a National Facility.

The first thesis that I supervised involving ATOLL was of Alejandra Velazquez-Garcia, already addressed in Chapter 2.2. In a nutshell, the goal of the thesis was to provide a first

overview of aerosol particles at the ATOLL site, as well as linking their optical and chemical properties, measured by LOA and IMT, respectively. Figure 3.1.8 below depicts some of the datasets from the thesis, namely $PM_{2.5}$ and PM_{10} observations from the air quality agency ATMO HdF at the "Lille Fives" site. Data shows the strong seasonality of both aerosol concentration as well as the dominance of coarse vs fine mode, the former, particularly during some pollution events in spring and autumn (Velazquez-Garcia et al., 2023).

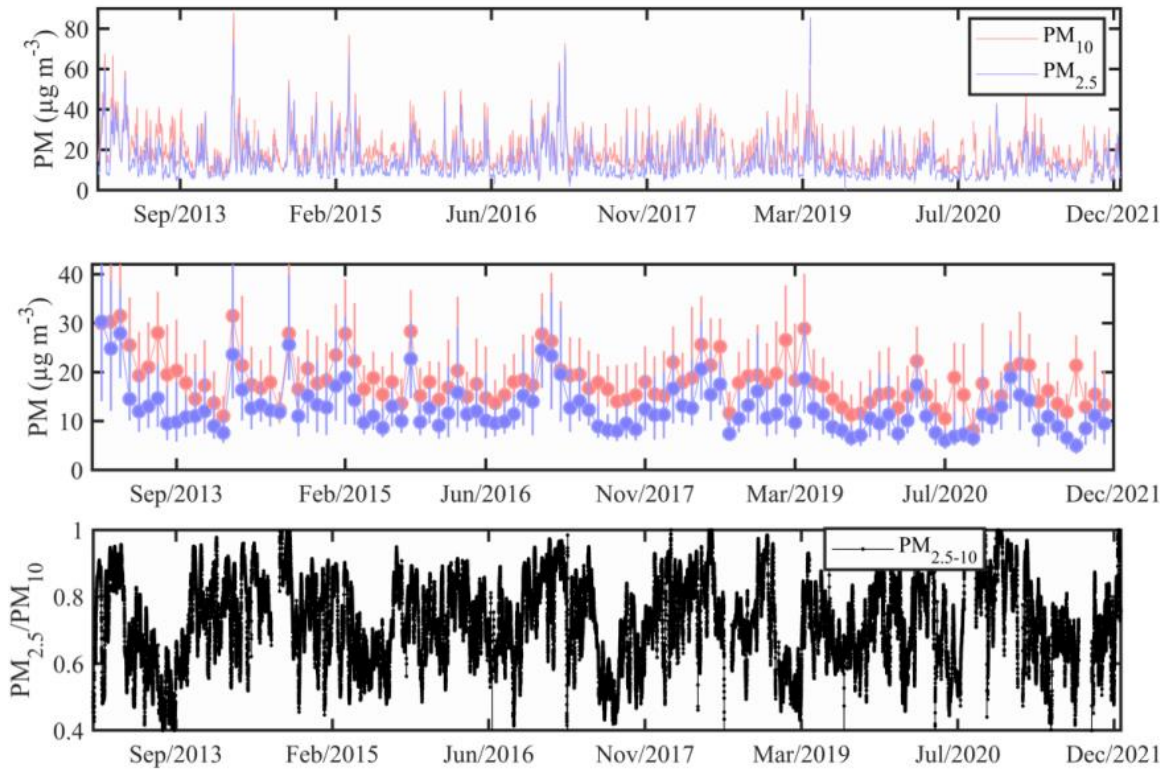

Figure 3.1.8 – PM_{10} and $PM_{2.5}$ time series from the ATMO HdF "Lille Fives" station in Lille for the ATMO HdF "Lille Fives" station in Lille for the period Jan/2013 – Dec/2021. Top: Hourly concentrations; Middle: Monthly concentrations (median and 25th – 75th percentile); Bottom: Hourly time series of the $PM_{2.5}$ /PM_{10} ratio. Reproduced from Velazquez-Garcia (2023).

The first step towards connecting chemical and optical properties can be through composition-dependent mass efficiencies. Those applied here are to light extinction (termed MEE), scattering (MSE) and absorption (MAE), defined as:

$$\sigma_{ext,\lambda} = \sum_j MEE_{j,\lambda} \times C_j \qquad \text{eq. 1}$$

Where $\sigma_{ext,\lambda}$ is the total extinction (scattering or absorption) for a given wavelength λ and C_j is the concentration of species j. Equation 1 is, of course, a strong simplification of matter-radiation interactions, neglecting the effect of different aerosol sizes, mixings, coatings, non-sphericity and other non-idealities. However, given the extensive dataset (30-min averages over four years of data), it was important to validate this simple assumption within the thesis. Those results allow, furthermore, to be applied in other optical-mass closure methods, particularly via remote sensing such as LIDARs, sunphotometers, etc., and to estimate the relative contribution of different aerosol species to their optical effects. Equation 1 can thus be converted into a multiple linear regression (MLR), where optical properties and species concentrations are used to calculate corresponding mass efficiencies. It was interesting to retrieve, for example, an MEE

of ammonium sulfate (AS) comparable between ATOLL and Po valley, Italy, 5.0 m^2 g^{-1} and 4.4 m^2 g^{-1}, respectively, whereas OA was almost 3-fold higher (1.8 m^2 g^{-1} at ATOLL and 6.1 m^2 g^{-1} in Italy). Such a large difference can be speculated to originate from a comparable AS (regional) population in Central Europe, whereas OA can be more locally influenced and thus have different compositions and sizes. We have also studied the effect of composition on SSA, indicating that lower SSA levels were dominated by carbonaceous aerosols, as expected, and that higher SSA values were linked with ammonium nitrate (AN) peaks, as shown in Figure 3.1.9.

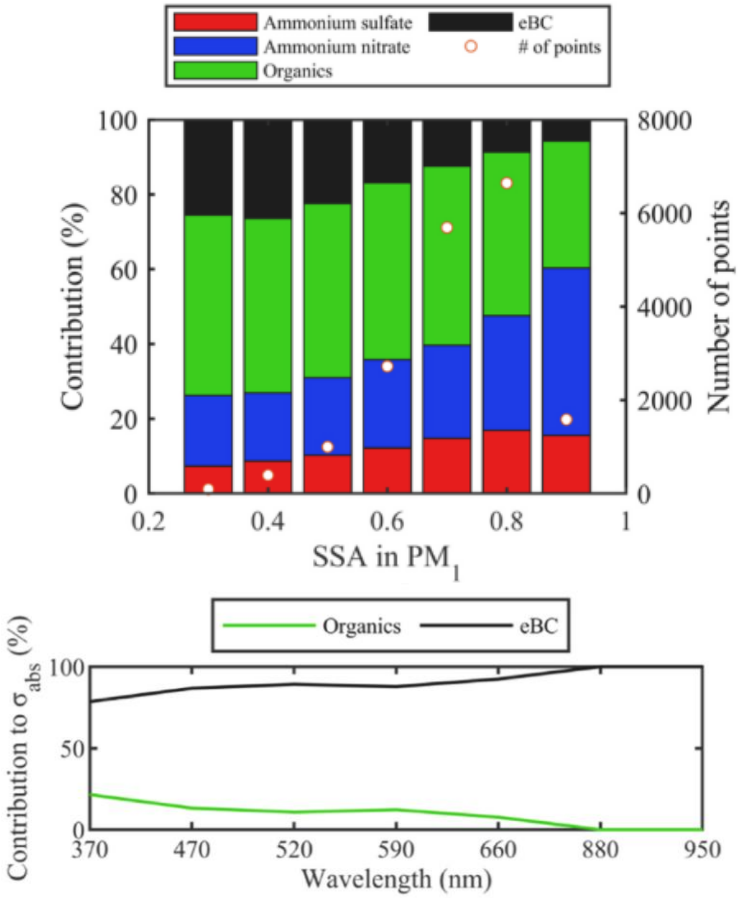

Figure 3.1.9 – Top: PM_1 fractional composition (left axis) and the number of observations (dots, right axis) according to SSA at 550 nm. Bottom: Averaged relative contributions of PM_1 chemical species to the aerosol absorption for multiple wavelengths. Data from the ATOLL site for Jul-17 till Dec-19. Reproduced from Velazquez-Garcia et al. (2023).

The relative contribution depicted above can also be analysed for scattering and absorption, and for multiple wavelengths. Results show generally an equally distributed contribution of organics, AS and AN on aerosol light scattering, a surprising dominance of carbonaceous aerosols on light extinction (in agreement with fairly low SSA at the site of about 0.7), and an increasing contribution of organics to absorption with decreasing wavelengths, up to 23% at 370 nm (Figure 3.1.9, bottom). This fraction of light-absorbing organics is termed Brown Carbon (BrC), which remains an important source of uncertainty on the aerosol-radiation interaction due to the complexity associated with organic makeup and atmospheric dynamics.

To study more in detail the sources of BC and BrC at the ATOLL site, we developed a method entitled INTERPLAY (iN-siTu obsERvations, hysPLit, And emission inventorY). As

the name suggests, the method combines back-trajectories and land information to explore air mass history and better interpret the atmospheric composition and its properties. The main advantage is that it is based on computationally inexpensive Lagrangian modelling, being able to gain important insights from large datasets, such as multi-annual observations. The method consists of integrating emissions of a given species along individual trajectories to quantify the relative contributions (including information such as distance, and travelled time) to the receptor site. In the thesis, we applied INTERPLAY to BC, given that it is expected to be relatively stable in the atmosphere within the time frame back-trajectories considered here (72 hours). Results from INTERPLAY can be found in Figure 3.1.10, showing the dominance of UK and BENELUX areas as main contributors to BC concentration at ATOLL (Figure 3.1.10a), with notably shipping lanes across the Channel and North Sea, as well as Paris. The method further estimates only about 8% of BC originates from Lille (Figure 3.1.10b), and a surprising 25% contribution from shipping (Figure 3.1.10c), complemented by typically expected sources such as traffic (31%) and residential (21%). If split between summer and winter, residential becomes the dominant source in the former, whereas shipping dominates in the latter (both at 38%). Despite being located about 60 km from the coast, we estimate that quite low SSA at the ATOLL site can be associated with intense shipping activities in the Channel and North Sea (combined with canals and other in-land shipping transport).

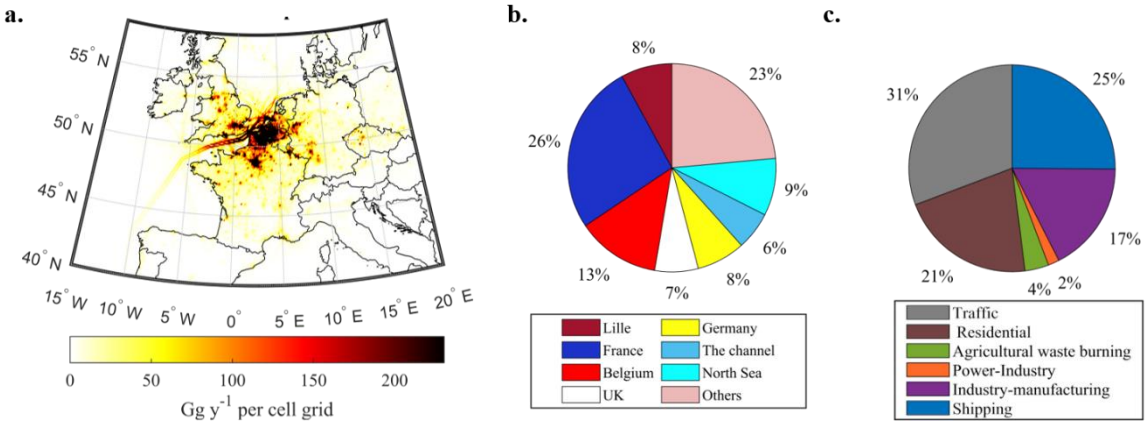

Figure 3.1.10 – a) Accumulated contribution of BC to ATOLL in Gg y-1 per cell grid and its regional (b) and sectorial (c) distribution over the period Dec 2016 – Dec 2019. Reproduced from Velazquez-Garcia et al., submitted.

Through INTERPLAY, we could associate, for example, different Angstrom Absorption Exponents (AAE) measured in situ with different BC sources such as Traffic, Residential, Shipping and Industry. Although it is generally assumed that AAE for Residential is 2, and for the other sectors (usually) closer to 1, our data shows more mixed values, of residential at 1.45 and the three others at about 1.35. This result is interpreted as the effect of atmospheric ageing and mixing in the atmosphere. To further understand the effect of aerosol ageing on its spectral absorption, and thus BrC contribution, we have focused on well-studied sources of Residential and Traffic and further separated between fresh (emitted < 24 h before arriving at ATOLL) and aged (>24 h). Then, the multiannual database of these four components was used in an MLR to explain BrC calculated in situ from the AE33. Results are shown in Figure 3.1.11, indicating that although BC residential Fresh and Aged contribute equally to BC, the contribution to BrC

is largely dominated by fresh emission (<24h), thus indicating a significant decrease in organic absorption with atmospheric ageing. Thus we provide evidence suggesting of fast decrease of BrC in the atmosphere in Central Europe with a significantly shorter lifetime (24h) compared to BC (7 days), impacting total aerosol radiative forcing in the region. Furthermore, BC source attribution nowadays is performed via the so-called "Aethalometer model", assuming that AAE equals 1 and 2 originate from fossil fuel and wood combustion, respectively. Therefore, such consumption of BrC in the atmosphere within 24 h leads to a reduction of associated AAE, and therefore a potentially strong underestimation of regional residential heating and an overestimation of traffic.

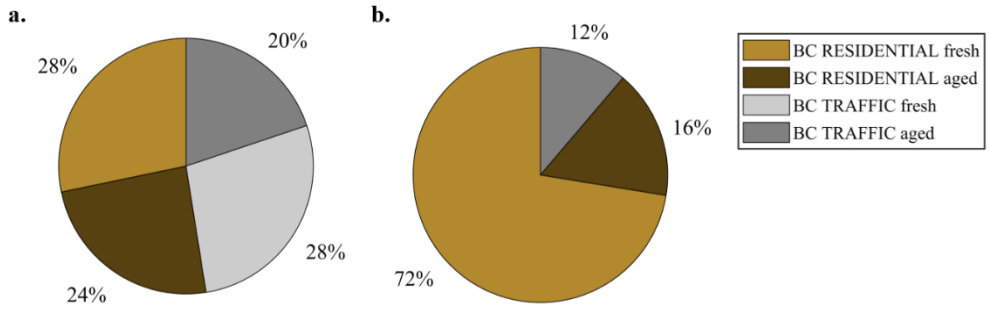

Figure 3.1.11 – a) Relative contribution to BC according to fresh (<24 h) and aged (> 24) residential and traffic sectors. b) Relative contribution to BrC absorption calculated at 470nm. Reproduced from Velazquez-Garcia et al., submitted.

In addition to the INTERPLAY method, identifying BC sources at the ATOLL site, I have participated in near-source studies focusing on quantifying the role of shipping emissions on air quality degradation, namely the projects SHIPAIR (ANR, PI Dr Yelva Roustan, CEREA) and PIRATE (ADEME AQACIA, PI V Riffault). I also lead in the latter the work package on result integration between observations, inventories and numerical models. Together with V. Riffault and my Dr Liselotte Tinel, we supervise the post-doctoral work of Dr Vishnu Murari. He is responsible for aerosol observations at the Dunkirk harbour, particularly using our High-Resolution Aerosol Mass Spectrometer (HR-AMS, Aerodyne), during the field campaign that took place in Sep 2022. Currently, V. Murari is studying aerosol composition and OA sources through PMF analysis. Preliminary results have identified an extremely high contribution of carbonaceous aerosols emitted from the ships (OA 66%, BC 20%, SO_4 11% and negligible amounts of NO_3 and NH_4) and with an organic component following a spectral signature as HOA, typically associated with vehicular emissions. The emission factors derived here will be used to improve estimates of the role of shipping emissions on coastal cities, particularly in the Channel.

Further focusing on OA sources through PMF analysis, I currently supervise the PhD work of Hasna Chebaicheb. Contrary to other source apportionment studies presented previously, the technique used in this thesis relies on rolling PMF, a technique recently developed at the Paul Scherrer Institute (PSI) and commercially available (Source Finder Professional, Datalystica Ltd.). Typical PMF has an inherent limitation of having fixed factors across the entire dataset. This creates some issues on longer datasets, particularly multi-seasonal, since it is expected some degree of source profile variation, notably between cold and warm seasons. Therefore,

the traditional PMF method requires longer datasets to be split into three-month sub-sets, having the drawback of running multiple independent PMF analyses, being labour-intensive, and prone to discrepancies between them. Therefore, rolling PMF analyses long-term datasets in a single iteration via a sliding time window (typically about a month), thus enabling, to some extent, inter and intra-seasonal variations of OA source profiles. To ensure homogenization across the dataset, the analysis is quite strongly constrained, whether by reference spectra of primary factors (such as HOA), via correlation with external tracers, and signal intensity on specific tracer ions. To identify the reference factors, a seasonal "classical" PMF must be performed for at least 1 year of data so that site-specific factors can be retrieved. At ATOLL, we selected the reference year to be from Oct-2016 to Aug-2017, yielding four factors, namely HOA, BBOA (biomass burning OA) related to biomass combustion, and two OOA factors, a less-oxidized OOA (LO-OOA) and a more-oxidized one (MO-OOA). A one-year rolling PMF was then performed for this reference year, prior to the full dataset analysis. This 1-year analysis, one of the first main results from the thesis of H. Chebaicheb, integrated into a large European overview of 22 datasets in Europe led by Andre Prevot's group from PSI (Chen et al., 2022). The resulting analysis, summarized in Figure 3.1.12, depicts PM_1 levels and composition, including the identified OA factors at each site.

Results have shown that, excepting Krakow and Bucharest, ATOLL (identified as "Lille" here) has the highest PM_1 for the period (14.0 µg m^{-3}), being dominated by OA (particularly OOA factors) and AN, associated with traffic and agricultural activities, in agreement with the analysis of Velazquez-Garcia et al. (2023). As discussed at the beginning of this section, those sources are indeed relevant in the HdF region, which compounded with favourable weather conditions, is often responsible for significant pollution events. The role of chemical species/sources on pollution episodes at ATOLL has been studied in more detail following the first analysis described above when a four-year dataset has been processed using rolling PMF (Chebaicheb et al., 2023). The full dataset (October 2016 through December 2020) uses the same reference spectra for the primary factors (HOA and BBOA), however with 28-day windows and seven-day shifting (instead of 7-day windows and 1-day shifting for 1-year analysis) for increased computational efficiency. Considering 50 PMF iterations of bootstrapping for increased robustness, a total of 11050 PMF analyses were performed for the entire dataset.

Figure 3.1.13 depicts PM_1 chemical composition and mass concentration for all seasons at ATOLL during the 2016-2020 period. It is important to note that the 10 % highest PM_1 level nearly reaches 30 µg m^{-3} in winter, in contrast with summer (15 µg m^{-3}). Excepting summer, values above the 30^{th} percentile (roughly 5 µg m^{-3}) show an increasing dominance of NO_3 with aerosol loading, having an important role in pollution events, while maintaining fairly stable contributions of SO_4 and OA, albeit with a larger fraction being OOA factors. In summer, the composition during pollution episodes depicts a different trend compared to other seasons. For example, SO_4 relative contribution decreases with increasing PM_1, thus not being significant during pollution events, however being driven by an increasing contribution of OOA fractions. It is interesting to note that in this work, we have identified that, during summertime, LO-OOA particularly is associated with biogenic SOA, whereas in wintertime, the same factor is associated with aged BBOA. During summertime, thus biogenic SOA represents about a third of OA, whereas, in wintertime, wood combustion is about half of OA at the ATOLL site.

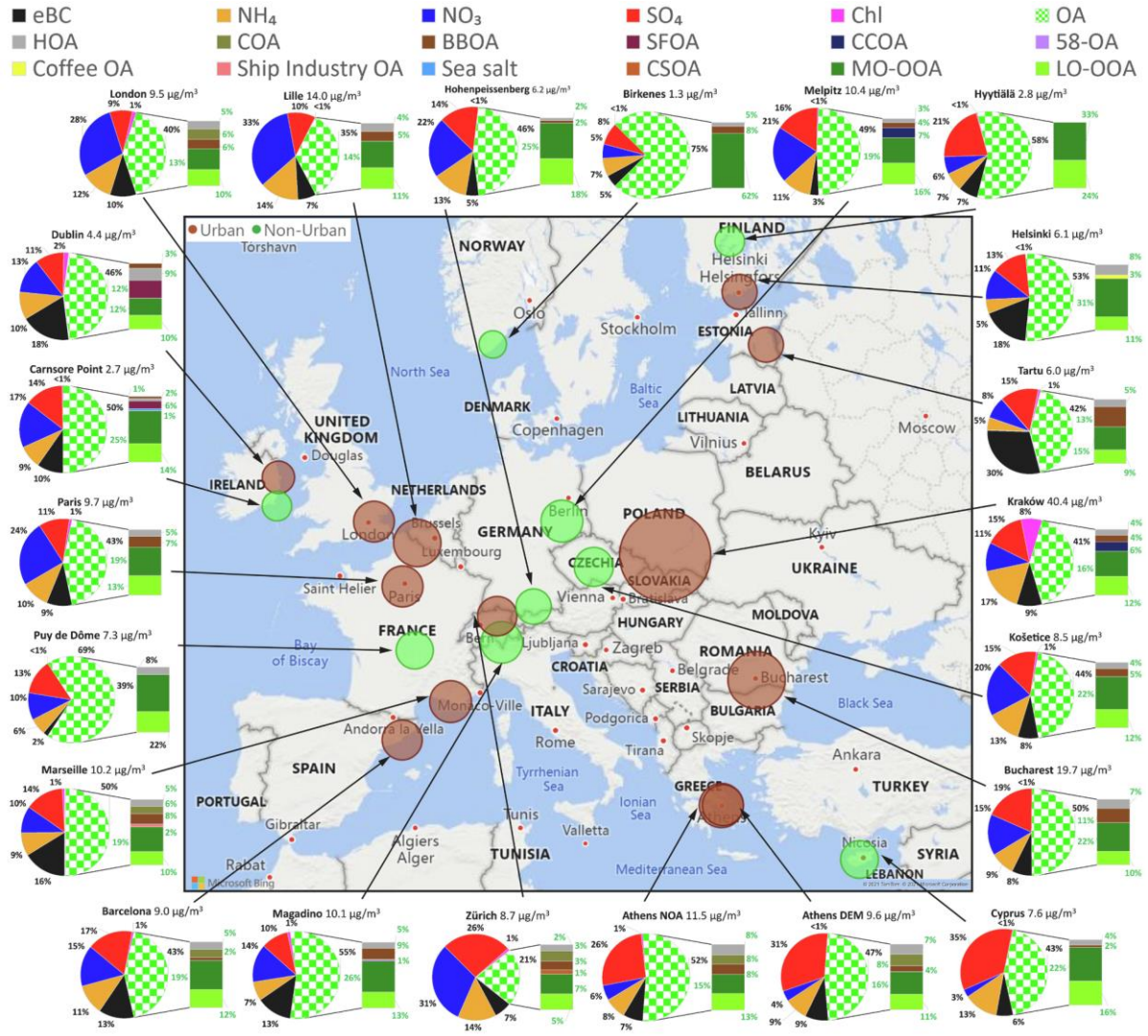

Figure 3.1.12 – PM_1 mass fractions of non-refractory inorganic species, BC and OA covering all seasons. The size of the markers corresponds to the PM_1 mass concentration. The brown colour of the marker indicates an urban site, while the green marker indicates a non-urban site. The checkered green/white shading of the pie charts denotes the OA fraction in PM_1, and the bar charts represent the contributions of each OA factor to the total OA mass. COA stands for cooking-like OA, CCOA for coal combustion OA, SFOA for solid fuel OA (mainly mixture of coal and peat combustion), 58-OA a specific source associated with Amines, and CSOA for cigarette smoke OA. ATOLL is identified here as "Lille". Reproduced from (Chen et al., 2022).

Following the analysis at the ATOLL site, H. Chebaicheb has performed a large multi-site rolling PMF analysis in continental France. Based on collaboration with research institutions and air quality networks, rolling PMF has been processed mostly in-house through a remarkable effort, providing a large overview of aerosol particle sources and composition in France. The temporal coverage and sites are presented in Figure 3.1.14 below, the large temporal overlap and homogeneity of source apportionment methods are expected to provide a strong robustness of the factor analysis. Results have identified interesting seasonal variations across different regions, some sites (i.e. in Paris) with a strong contribution of cooking-like OA, and generally

a large fraction of OOA factors, compatible with findings at ATOLL (Chebaicheb et al., *in preparation*).

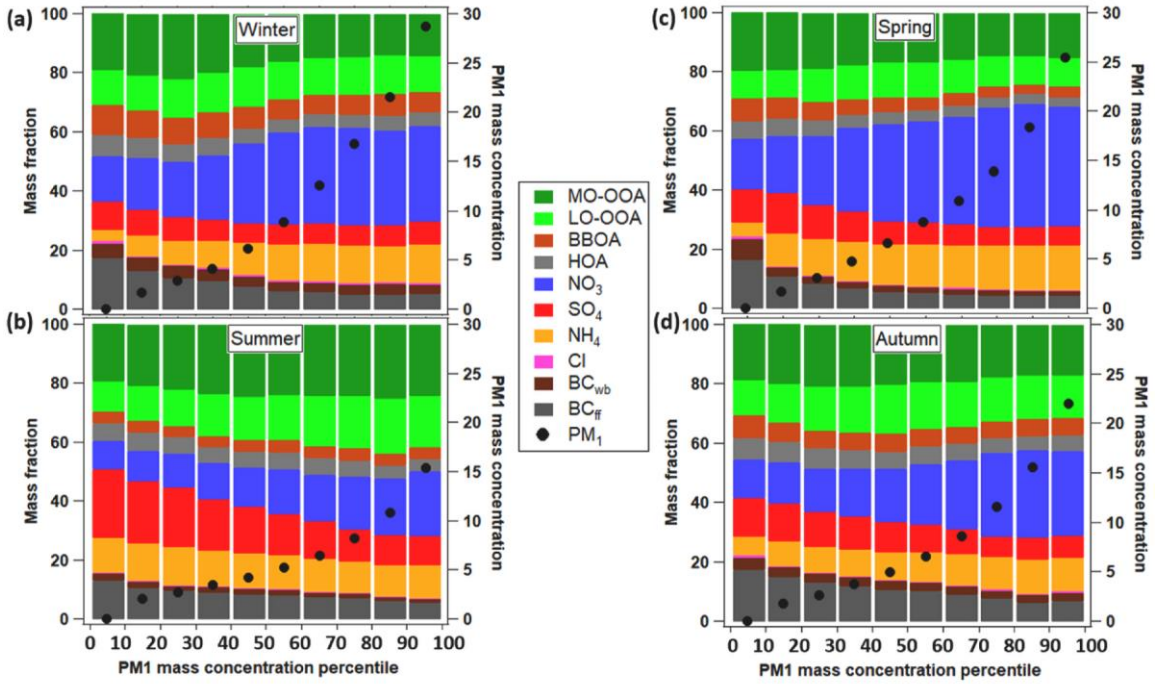

Figure 3.1.13 – Seasonal mass fraction of PM_1 species as a function of mass concentration, a) in winter, b) in summer, c) in spring, and d) in autumn. Reproduced from Chebaicheb et al. (2023).

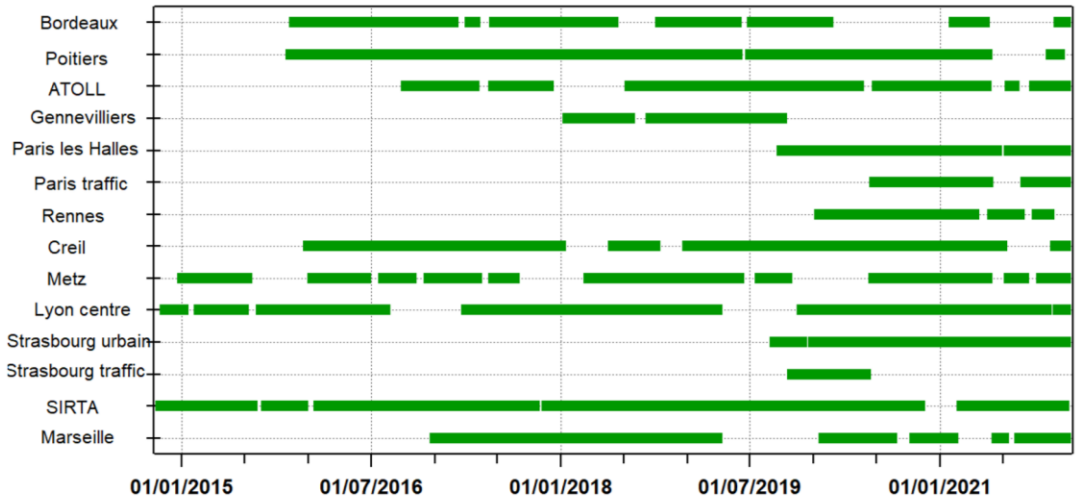

Figure 3.1.14 – Temporal coverage of rolling PMF analysis across multiple sites in France. Sites are integrated here whether through collaborations with research institutions such as LSCE (SIRTA), LCE (Marseille) or air quality networks. Reproduced from Chebaicheb et al., in prep.

Concluding remarks

As a post-doc at the University of Sao Paulo, I had the opportunity to enter the topic of aerosol research, having previously only worked on mass spectrometry development for organic gaseous components. The activities were intense, integrating tunnel measurements, conducting dynamometer chamber experiments and running ambient observations on four different sites across the SPMA, including extensive filter sampling and deploying online instrumentation such as ACSMs and PTR-MS, in addition to a range of aerosol microphysical properties and gaseous pollutants. Those activities resulted in several important studies of such unique vehicular fleet, fueled with 50% biofuel. Those ranged from providing a first characterization of the emitted aerosol composition and physical properties (Brito et al., 2013), the corresponding ambient aerosol hygroscopicity (Almeida et al., 2014) and the dynamics of carbonaceous aerosols within SPMA (Monteiro dos Santos et al., 2016). Furthermore, for the first time presenting the emission profile of Oxygenated VOCs attributed to traffic (Brito et al., 2015), quantifying the effect of biofuel on UFP and Ozone (Salvo et al., 2017), and using ethanol as a tool to distinguish the impact of LDVs and HDVs on atmospheric pollutants (Brito et al., 2018). More recently, as an Assistant Professor at IMT Nord Europe, I continued working on urban environments, whether in Sao Paulo, through the currently ongoing BIOMASP^{+} project, or in the HdF region, in the north of France. Here, much of the activities related to the French ACTRIS site ATOLL through the PhD thesis of A. Velazquez-Garcia and H. Chebaicheb, focus on a multi-annual analysis of aerosol physico-chemical properties (Velazquez-Garcia et al., 2023), light-absorbing aerosols (Velazquez-Garcia et al., *submitted*) and OA sources (Chen et al., 2022; Chebaicheb et al., 2023).

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3.2 Amazonian aerosols: from biomass burning to the pristine atmosphere

Introduction

The Amazon Basin holds the largest rainforest in the world, spanning over 5.5 million km^2. For context, this is about 30% larger than the surface of all European Union countries combined, or about 9 times of France. The forest is estimated to hold about 100 billion metric tons of carbon, usually acting as a carbon sink, however with a noticeable decrease in resilience over the past 20 years (Boulton et al., 2022). This is attributed to forest degradation due to deforestation, urbanization and global climate change, the latter not only affecting air temperatures but also rainfall patterns and the frequency and intensity of drought events in recent years. It is important to note that the forest is also home to about 30 million people, with a fast-growing population. Those are mostly found in the east and south of the forest, and in Manaus, a 2 million people city in the heart of the forest, surrounded by thousand kilometres of pristine forest.

Figure 3.2.1 depicts a map of South America indicating two areas where I concentrated my research. Porto Velho (PVH) is located within the "deforestation arc", in Southwestern Amazonia and represents an area that was largely degraded over the past 20 years. The TT34-ZF2 is in Central Amazonia and about 40 km upwind (northeast) of Manaus, complemented by the ATTO (Amazon Tall Tower Observatory) site, further northeast, at about 170 km of Manaus. In one of the first studies that I participated in the Amazon, we contrasted both regions, which provides interesting insights into Amazonian aerosols. During biomass burning season, for example, 24h $PM_{2.5}$ reached up to 370 µg m^{-3} in PVH (of which 22 µg m^{-3} were BC), whereas TT34-ZF2 peaked at 16 µg m^{-3} (Artaxo et al., 2013). Correspondingly, due to proximity to forest fires in PVH, the average $PM_{2.5}$ dry season concentrations were 10 times higher than during the wet season (33.0 and 3.3 µg m^{-3}, respectively), whereas in TT34-ZF2 the ratio was only about 2.5 times (3.4 and 1.3 µg m^{-3}).

In late 2012 and early 2013, I conducted my first ACSM observations in the Amazon at PVH (dry season) and TT34-ZF2 (wet season), yielding respectively NR-PM_1 levels of 12.9 µg m^{-3} and 0.4 µg m^{-3}, about 80-93% comprised of organics (Figure 3.2.1, bottom). Furthermore, interestingly, SSA at 637 nm in both regions was observed at the lowest (0.83) at TT34-ZF2 during the wet season, when the anthropogenic impact is considered negligible. In contrast dry season at the same site yielded an SSA of 0.89, and throughout the year at PVH around 0.93. At the time, the fairly low SSA at TT34-ZF2 was associated with a higher concentration of Primary Biogenic Organic Aerosols (PBOA), however, we identified later to be associated with sporadic transatlantic transport of biomass-burning (BB) plumes from Central Africa, strongly enriched with BC (Holanda et al., 2020; Saturno et al., 2018). In the following, this chapter will be separated into specific studies that I performed whether focusing on fresh BB aerosols (mostly at PVH), or Central Amazon whether pristine forest or in the environs of Manaus.

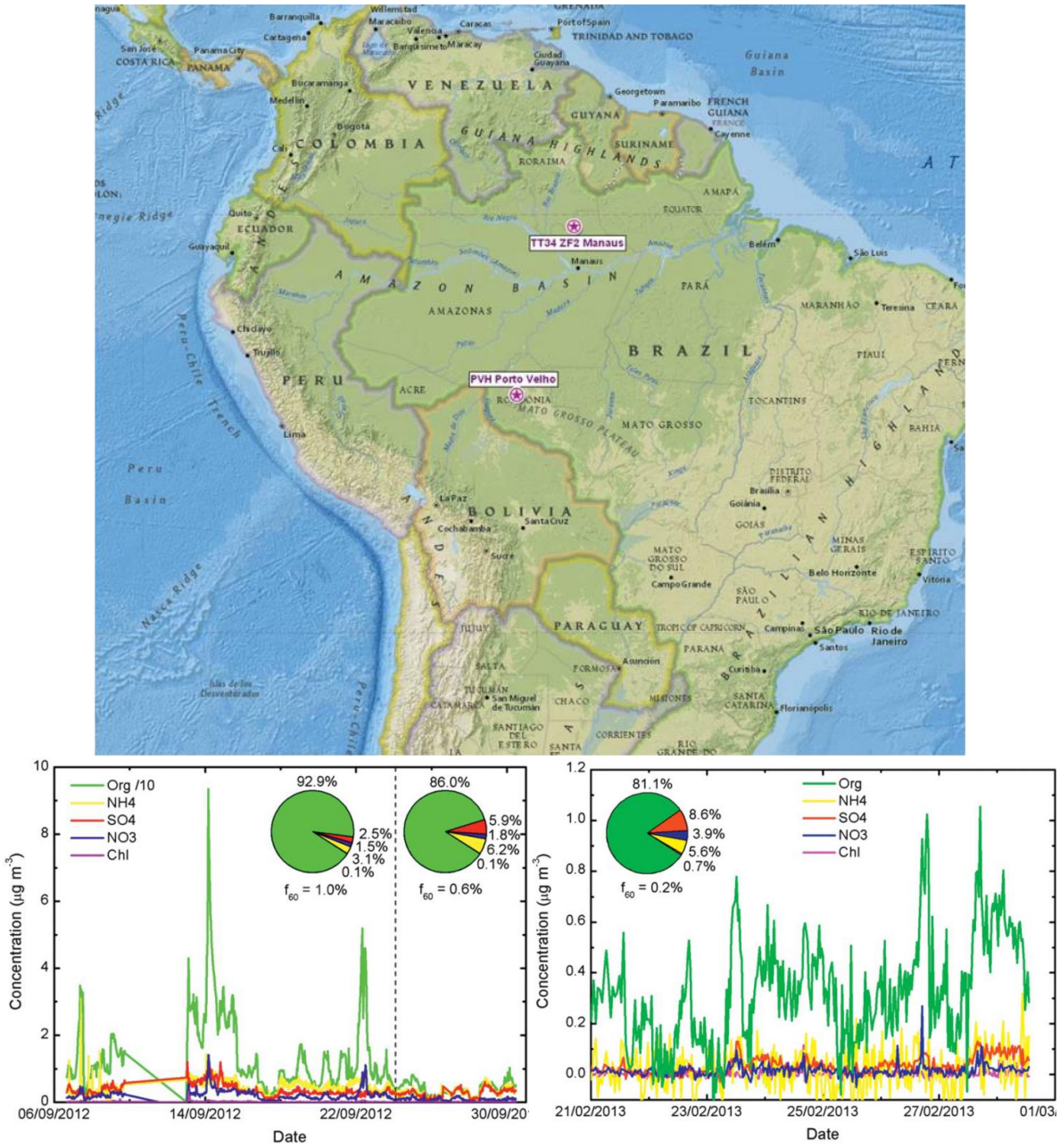


Figure 3.2.1 – Top: Map indicating the location of TT34-ZF2, near Manaus, and PVH, in the vicinity of Porto Velho. Bottom: Aerosol composition during the dry season and transition to wet season (dashed line) at PVH (left) and wet season at TT34-ZF2 (right). Reproduced from Artaxo et al. (2013).

Fresh biomass burning emissions

Following the first observations described above, the PVH site has been studied in more detail within the scope of the South American Biomass Burning Analysis (SAMBBA) project, a Brazilian/UK collaboration. As described in Chapter 2.4, the project combined ground and aircraft measurements to better characterize biomass-burning plumes and their effect on the Amazonian atmosphere. As a postdoc at the University of Sao Paulo, I had the opportunity to integrate both components of the project, overseeing aerosol observations at the PVH site, and

operating the PTR-MS from David Oram (National Centre for Atmospheric Science, UK) onboard the research aircraft. Due to the extensive amount of forest fires in the regions surrounding the ground site, I could study how BB aerosols transformed as they oxidized in the atmosphere, both chemically and physically (Figure 3.2.2). The ground observations have shown a strong dominance of OA within the fresh plumes (90% of OA within PM_1), one of the highest ratios among forested sites worldwide, typically impacted by higher fractions of BC or sulfate. Results have shown as well that despite strong changes in chemical composition, increasing the OA oxygen content (oxygen to carbon ratio from 0.2 up to 0.7), negligible changes are observed in the aerosol size distribution, apart from dilution, keeping a dominant accumulation mode at 270 nm. In that study, we estimated that already 3h downwind of the fire, the OA signal was dominated by an oxidized BBOA, depicting quite small amounts of f60, a typically used diagnostic tracer of BB on aerosol mass spectrometry.

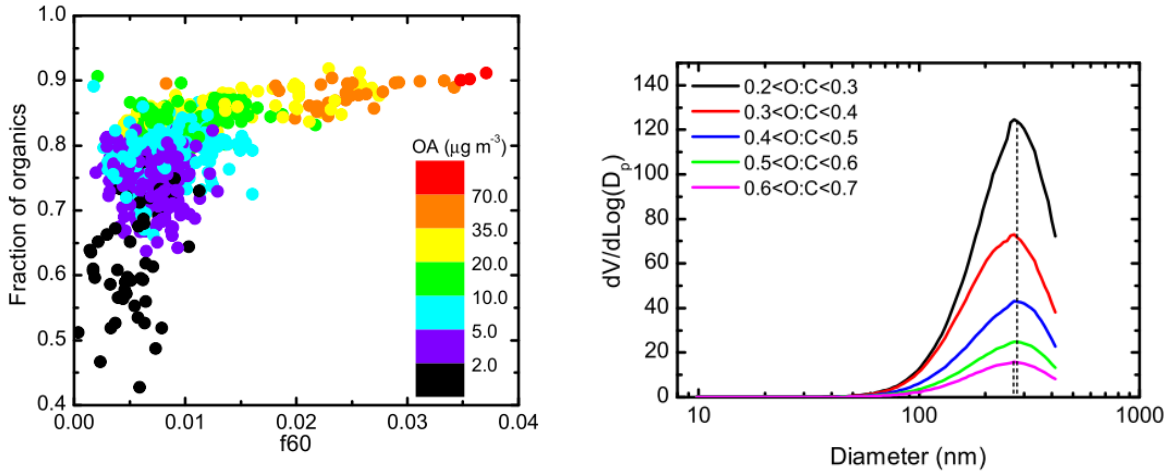


Figure 3.2.2 – Physical and chemical characterization of BB aerosols at PVH during the SAMBBA experiment. Left: Fraction of OA within PM_1 (NR-PM_1 + BC) versus f60, a diagnostic tracer for fresh BB aerosol particles. Right: aerosol volume distribution according to the corresponding oxygen-to-carbon ratio in the OA. Reproduced from Brito et al. (2014).

The PVH ground site has also been the topic of health-focused studies. The analysis that I collaborated more closely was in a partnership between the physics and chemistry institutes of the University of Sao Paulo, and the Biochemistry department at the Rio Grande do Norte Federal University. This was based on offline analysis of high-volume filters, where several species were quantified, including Polycycle Aromatics Hydrocarbons and organic tracers, and particulate matter toxicity was quantified via in-vitro tests (de Oliveira Alves et al., 2015). Results obtained high levels of Levoglucosan, a tracer for wood combustion reaching up to 8% of PM_{10}, in agreement with previous observations of Brito et al., (2014). Based on the dataset created here, I performed Principal Component Analysis, isolating the BB component and allowing us to identify this source's profile, including a range of well-known toxic species such as Retene and Benzo[a]pyrene. Furthermore, despite an average PM_{10} level of 30.2 µg m^{-3} (within the recommended by the World Health Organization at the time), lung cancer risk already exceeded significantly recommended values due to high carcinogenic potential, as depicted in Figure 3.2.3. This study showed that the high toxicity of wood combustion must be

considered in environmental policies in the region, impacting regularly a population of over 10 million people.

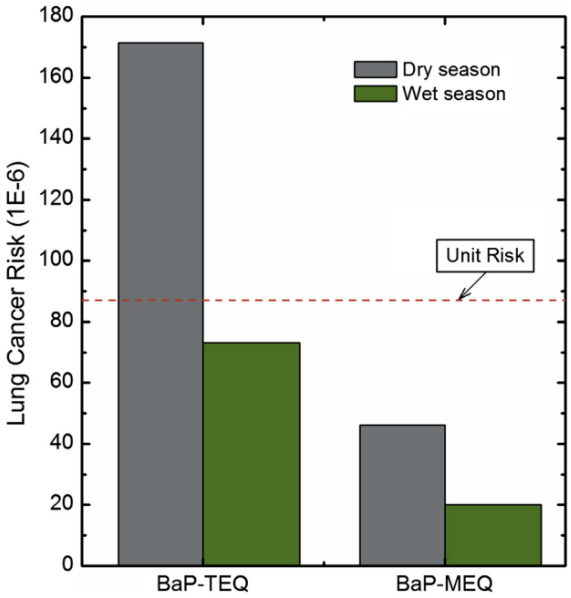


Figure 3.2.3 – Assessment of the lifetime lung cancer risk (LCR) from the BaP-TEQ (carcinogenic potential) and BaP-MEQ (mutagenic potential) during the dry and wet seasons at the PVH site. Reproduced from de Oliveira Alves et al. (2015).

The aircraft component of SAMBBA aimed to characterize as well for the first time the impact of BB emissions on higher layers of the atmosphere, whether it was on the gas or particle phase. Figure 3.2.4 depicts an idealized scheme for pollutant mixing, advection and plume injection (top) and presents a transect flight from east to west (bottom). The scheme presents that as a result of surface heating, the buoyant turbulence increases during the day through the convective mixing layer. Morning mixing destabilizes the residual layer, and the growth of the mixed layer is inhibited by a statically stable entrainment zone. The lifting condensation level, leading to patchy cumulus fields, is above the mixed layer top, within the entrainment zone. Typically, a maximum horizontal wind speed, known as the trade wind inversion is present above the entrainment zone.

The aerosol pollution layer was deeper in the east (~ 3 km) than the west (~ 1.5 km), reflecting a regional contrast in soil moisture and surface insolation. Enhancement of gaseous pollutants above the boundary layer was likely transported via deep moist convection which could remove aerosols via wet scavenging. Conversely, horizontal wind shear inhibited mixed layer growth and the vertical ascent of smoke plumes from fires. The transect flight could capture the regional contrast in pollutant vertical distribution and control meteorological parameters. Via a reduction in altitude of the mixing layer altitude (following sharp changes in equivalent potential temperature, θ_e), one can also see the increase in RH above the mixed layer top. Conversely, a wind speed maximum is present at 5–6 km, coincident with the entrainment zone. Together, this structure can explain the capping of aerosol particles below the first wind-speed jet, notably their height differences between the regions. Furthermore, black carbon mass loadings were much greater in the east than west 1.5 vs. 0.85 μg m^{-3}, despite comparable CO and aerosol light scattering. This supports the relatively low Black Carbon emission in the

vicinity of the PVH site, previously reported by Artaxo et al. (2013) and Brito et al. (2014), suggesting mostly smouldering fires, in contrast to the east, a drier region, likely dominated by flaming regimes.

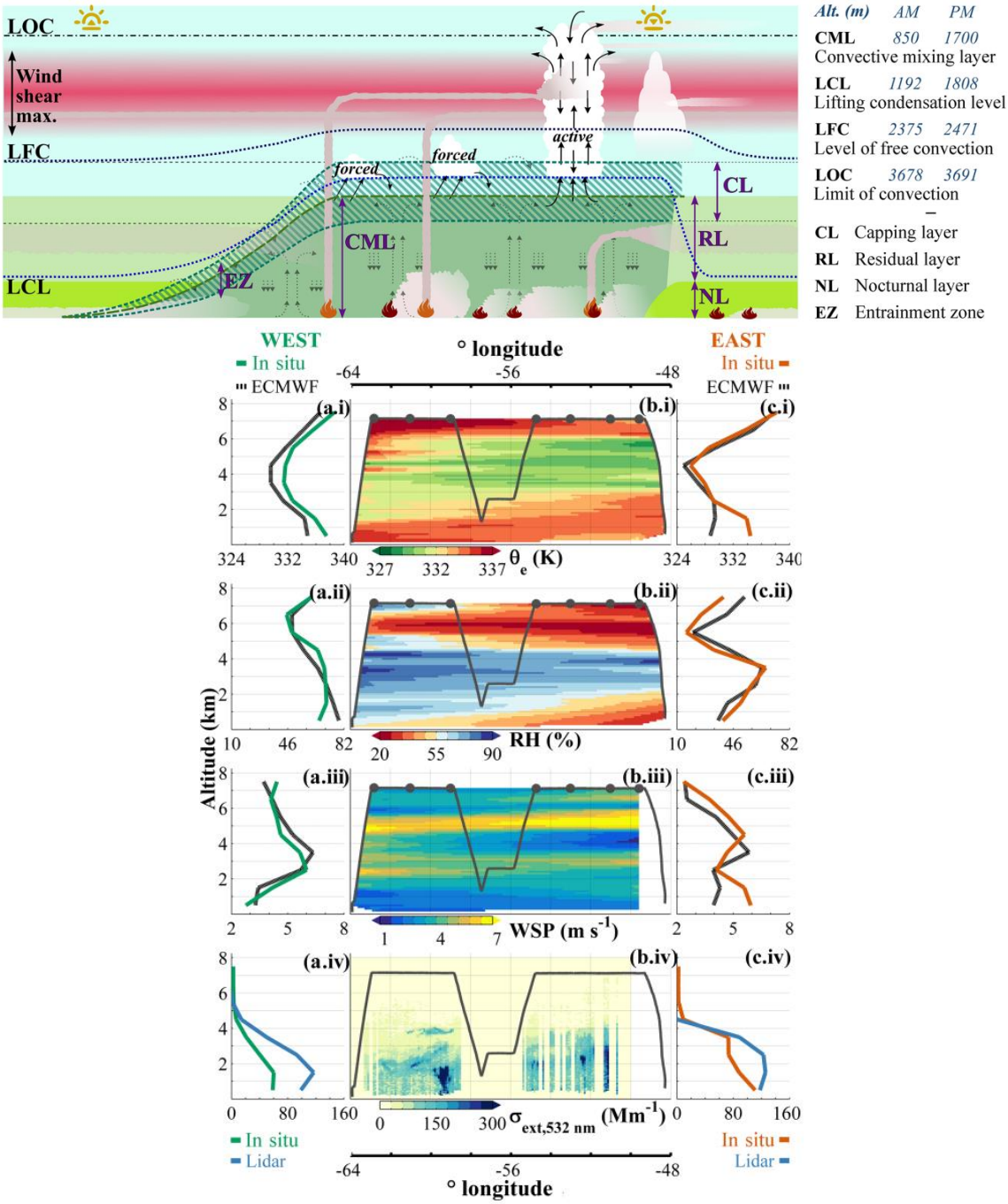


Figure 3.2.4 – Top: Schematic of the typical diurnal development of the convective boundary layer. Red flame symbols indicate more smouldering fires and orange symbols more flaming combustion. Bottom: Summary of the west-to-east regional gradient in equivalent potential temperature (i), relative humidity (ii), horizontal wind speed (iii) and aerosol extinction (iv). The regional gradient is emphasised by the side panels which show the median thermodynamic and aerosol extinction profiles for all western (a) and eastern flights (c). Reproduced from Darbyshire et al. (2019).

Based on the same flights as described in the study above, we analysed also gas-phase distribution throughout those large pollution plumes above the Amazon rainforest, however looking at the oxidative capacity of the atmosphere, notably using isoprene and its oxidation products as a proxy (Santos et al., 2018). Using CO levels, and their ratios with O_3, flight tracks were categorized as background (BG), Fresh Plume (FP) or Aged Plume (AP). Notably, the oxidation of isoprene is higher in fresh smoke plumes at lower altitudes (~ 500 m) than in aged smoke plumes. Furthermore, during the fresh plume regime, the estimated vertical profile of OH through the isoprene oxidation ratio showed evidence of an increase in the oxidizing power in the transition from the planetary boundary layer to the cloud layer (1000–1500 m). Those ranges indicate a significant change above and inside the cloud decks due to edge effects on photolysis rates, which have a major impact on OH production rates. Figure 3.2.5 presents cross sections (according to altitude and hour of the day) of isoprene mixing ratios and its oxidation ratio for BG, FP and AP. This presents quite high levels of isoprene on the upper layer in the presence of fire plumes, contrary to BG conditions, associated with fire activity promoting vertical transport, and increased isoprene release associated with the heat from fires affecting nearby vegetation. In general, higher isoprene oxidation values were observed throughout the atmosphere for AP, compared to FP, or BG. However the latter depicts an interesting profile along the cloud layer (1200–2000 m), where isoprene oxidation increases by 94 %. As discussed previously, this is associated with higher OH due to photolysis increase due to cloud edge effects.

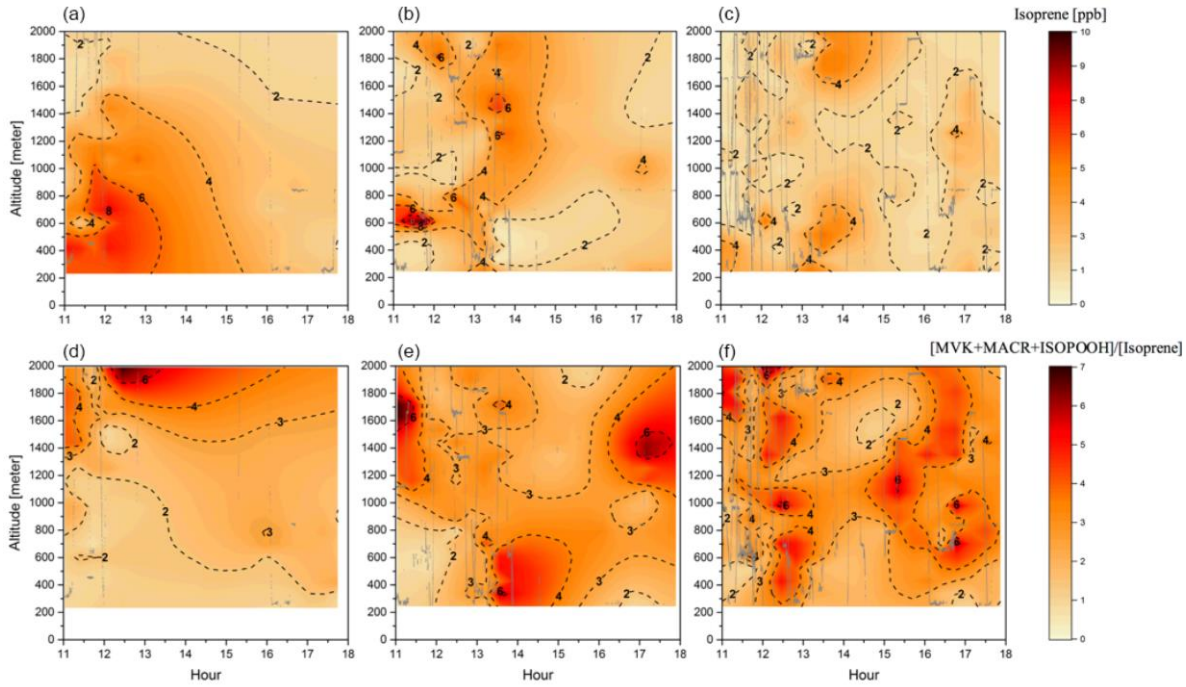


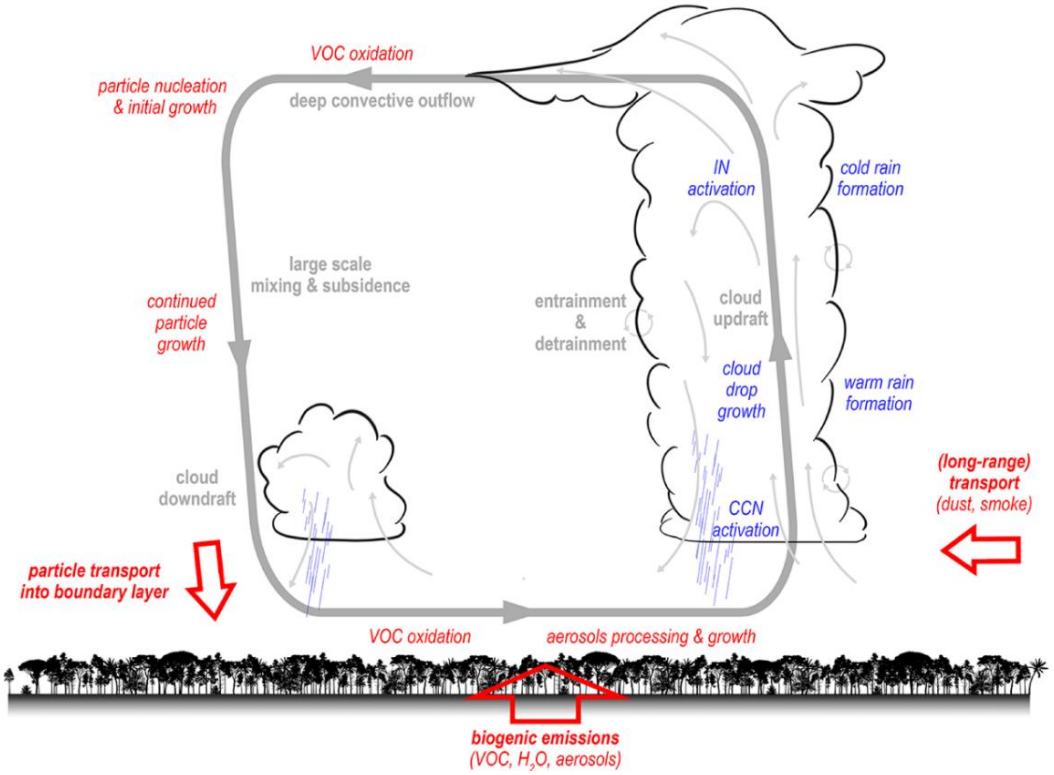
Figure 3.2.5 – Cross section of the isoprene mixing ratio (ppbv) (a–c) and the [MVK+MACR+ISOPOOH] / [isoprene] ratio (d–f) for the three different groups: background environment (on the left), fresh smoke plume (t < 2 h, middle), and aged smoke plume (t > 2 h, on the right). Hour is in local time. Reproduced from Santos et al. (2018).

Pristine Central Amazon

At the end of the SAMBBA project, I focused my research in Amazonia on the region of the TT34-ZF2, performing observations at this site, as well as at the ATTO and in the environs of Manaus, during GoAmazon14/5. At TT34-ZF2 I supervised Rafael Stern's master's studies focusing on linking aerosol physical and chemical properties in Central Amazon. Recently, we re-visited the dataset, quantifying the contribution of different OA types (BBOA, IEPOX-SOA, OOA) and inorganics on optical properties, and a manuscript is under preparation for submission on the topic. Furthermore, in 2013, I participated in the Brazil–UK Network for Investigation of Amazonian Atmospheric Composition and Impacts on Climate (BUNIAACIC) campaign at the TT34-ZF2. During that time, our observations integrated a study led by colleagues from the University of Manchester focusing on aerosol hygroscopicity and the distribution of bioaerosols using fluorescence techniques (Whitehead et al., 2016), and from the Federal University of Sao Paulo focusing on the dynamics of aerosol size distribution (Rizzo et al., 2018). The former identified quite a clean atmosphere with a total number aerosol concentration of only 260 cm^{-3}, dominated by OA (>80%), and with a low hygroscopicity (κ from 0.12 to 0.18) and significant daily variability in bioaerosols (200 L^{-1} during the day, up to 1200 L^{-1} during nighttime). The analysis published by Rizzo et al. has shown that New Particle Formation (NPF) events were rarely observed at ground level (about 3% of the days only), and the important role of convective rain in removing accumulation mode aerosol and injecting freshly formed Aitken mode.

Given the small number of UFP primary sources in the remote aerosol forest, and extremely unfrequent NPF at ground level, the origin of aerosol sources in the Amazon has been a strong research topic for at least the past 10 years. Through a combination of aircraft observations and ground observations at the ATTO site, we proposed in an article in Nature that NPF events at higher altitudes (in the Free Troposphere, FT) combined with downdrafts associated with convective rains were a significant source of aerosol numbers at ground level (Wang et al., 2016). The scheme proposed by our recent long-term reanalysis of ground and aircraft data by Franco et al. (2022), and the figure from Wang et al. (2016) showing the dominance of nucleating particles at higher altitudes are reproduced here in Figure 3.2.6.

It is interesting to note that, to date, multiple studies have identified this upper troposphere NPF and downdraft as an important source of nucleation-sized aerosols, other studies have identified primary emission from the forest as a potential source. This was notably studied in collaboration with Alexander Laskin's group at the Pacific Northwest National Laboratory (PNNL). Through cycles of hydration and de-hydration of coarse-mode filters, we have identified strong fragmentation of PBOA, increasing the aerosol number by a few orders of magnitude, and fragment sizes ranging down to tens of nanometers, potentially smaller however limited by equipment (China et al., 2016). Focusing more in detail on the composition of the fragments, we quantified that up to 30% of the aerosol-containing sodium in Central Amazon originated from PBOA, and not sea salt as originally thought (China et al., 2018). Due to the high hygroscopicity, those sodium-containing fragments are expected to have a significant influence on cloud formation within the Basin.



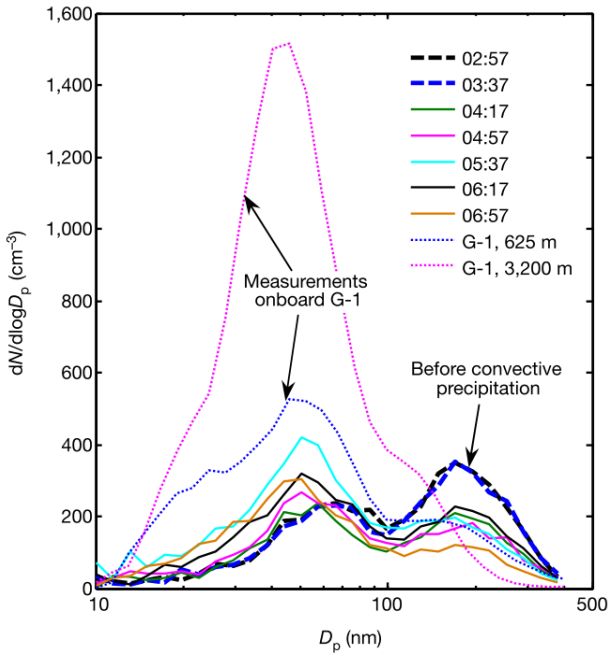


Figure 3.2.6 – Top: Scheme depicting aerosol cycling in the course of deep convection, with an upward transport of VOCs and aerosol particles, new particle formation in the free troposphere with initial particle growth, followed by the downward transport of sub-50 nm particles into the planetary boundary layer. The three red arrows represent the main aerosol source categories in the Amazon, (i) primary and secondary particle formation from local and regional biogenic sources, (ii) long-range transport of dust, smoke, and other aerosols, and (iii) the vertical transport of sub-50 nm particles from aloft. Scheme reproduced from Franco et al. (2022). Bottom: Aerosol number size distribution based on aircraft (identified as G-1) and at 60m tower at the ATTO site, reproduced from Wang et al. (2016).

Evolution of Manaus plumes within the Amazonian atmosphere

The GoAmazon14/5 was a large project funded mainly by the US and Brazil (Martin et al., 2017). Initially a Department of Energy (DoE) proposal by Prof. Scot Martin (Harvard University), the project received multiple targeted funding, including a collaborative injection from the US National Science Foundation (NSF) and the Sao Paulo State funding agency (FAPESP), the latter funding my 2014-2016 postdoc. During the observational period, Artaxo's group contributed by performing measurements at multiple sites, including upwind of Manaus (ATTO and TT34-ZF2, termed T0a and T0t in the project), downtown Manaus (T1), downwind of Manaus, about 8 km from the city, across the negro river (T2) and to some observations at the supersite T3, about 70 km from Manaus (Figure 3.2.7). This project was quite an intense experience, where I ran or oversaw some instrumental deployment across most of the sites. I operated, for example, the size distribution measurements at T0a (which integrated Wang et al. (2016) analysis, among other studies), conducted aerosol chemical composition and microphysics properties at T0a, I installed and coordinated the T2 site (including operation of ACSM, PTRMS, aerosol microphysics, gaseous pollutants) and operated an ACSM at T3, particularly in between intensive operating periods when no other aerosol mass spectrometer was operational.

During that time, I supervised the thesis of Glauber Cirino, with a focus on the evolution of Manaus plume, particularly on aerosol physicochemical properties. To achieve some robust results, we had to provide a detailed characterization of the atmospheric dynamics at the T2 site, notably by an intense river-breeze effect provided by an 8 km wide river between the city and the sampling site. Through a combination of extensive back-trajectory calculation local wind analysis and pollutant levels, we identified periods when air masses from Manaus intersected both T2 and T3 sites. This allowed for a direct comparison of the anthropogenic masses during both wet and dry seasons, with very distinct background aerosols as previously discussed. Among the main findings of Cirino et al. (2018), we estimated that the transport between T2 and T3 lasted on average about 4-5h, having a significant formation of both secondary organic (40%) and inorganic (30%) aerosols, the latter being mainly in the form of sulfate. Conversely, the SSA at 637 nm of the plume increased from 0.70 to 0.83 during transport from both sites, corresponding to the increase in organics and sulfate. To have also sites in the Amazon that were shifting between in- and out-of-plume was a good setup to study isoprene oxidation pathways (and their dependency on pollution levels), discussed in the next section.

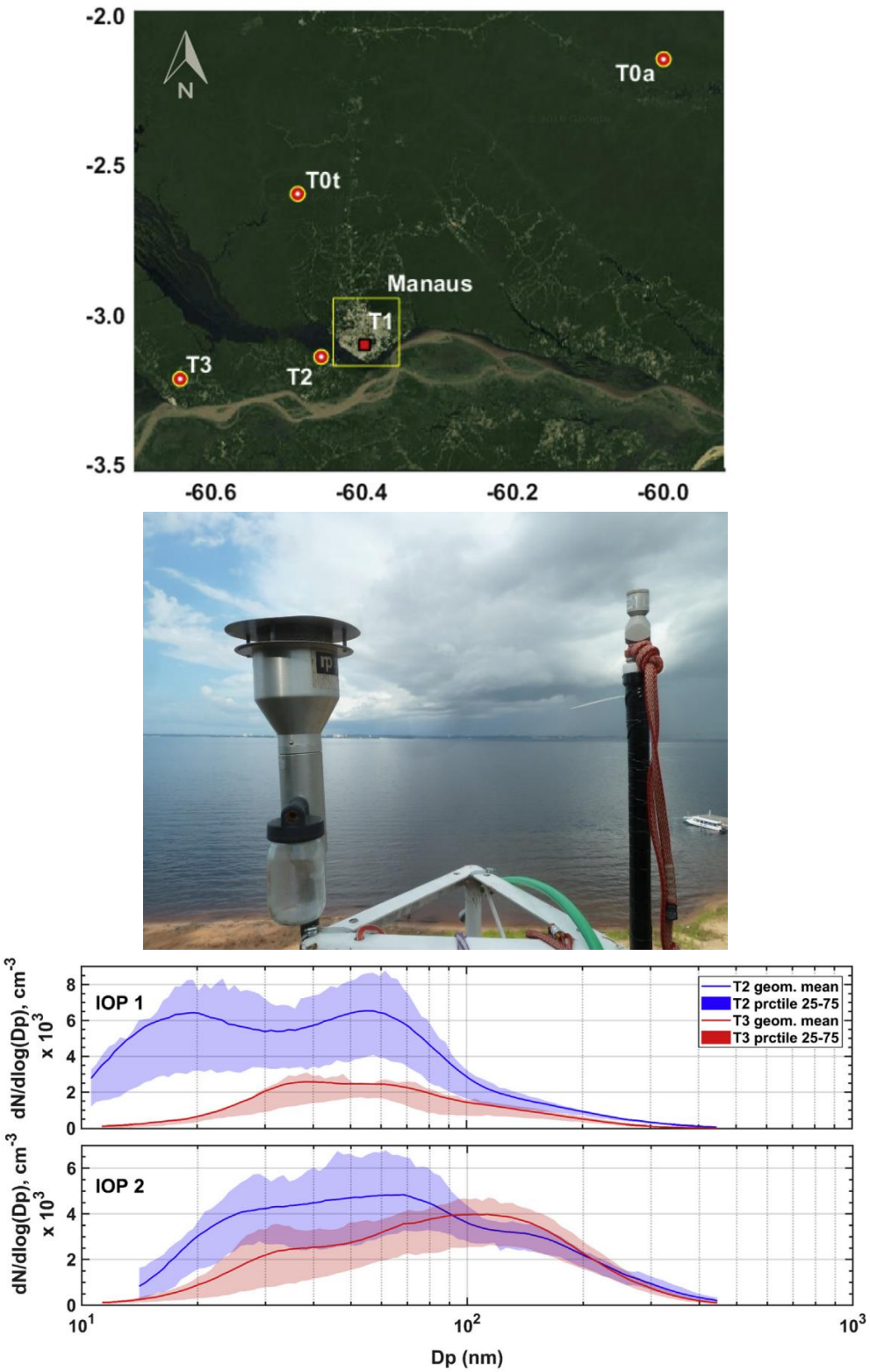


Figure 3.2.7 – Top: Locations of GoAmazon2014/5 research sites in the environs of the city of Manaus. Middle: View from aerosol inlets from the T2 site overlooking Manaus across the Negro river. Bottom: Mean particle number size distribution at T2 (blue line) and T3 (red line) for the wet (IOP 1, top) and dry (IOP 2, bottom) seasons, considering selected periods under the influence of the Manaus urban plume. Shaded areas represent the 25th and 75th percentiles. The top and bottom figures are reproduced from Cirino et al. (2018)

Concluding remarks

This chapter has described work linked with the Amazonian atmosphere, notably contrasting Southwestern and Central Amazonia. The first one is located within the deforestation arc and is strongly impacted by biomass burning. Stemming from my studies on aerosol composition, I contributed to studies providing a general aerosol physical and chemical overview (Artaxo et al., 2013), as well as a detailed description of plume ageing and their corresponding physical evolution (Brito et al., 2014), or associated toxicity (de Oliveira Alves et al., 2015). The aircraft component of the SAMBBA project has allowed us to gain interesting insights into the large regional pollution haze during biomass burning season, focusing on atmospheric meteorological dynamics control on particles (Darbyshire et al., 2019) or gas-phase, specifically focusing on atmospheric reactivity through isoprene oxidation (Santos et al., 2018). In the pristine Amazonian atmosphere, I contributed to studies looking at aerosol dynamics (Rizzo et al., 2018), identifying the potential role of free tropospheric NPF events as the source of condensation particles at the ground level (Franco et al., 2022; Wang et al., 2016), or the potential source of PBOA also as UFPs (China et al., 2018; Whitehead et al., 2016). Finally, my role in the GoAmazon2014/5 project was described (Martin et al., 2017), including the PhD supervision of G. Cirino, studying the evolution of aerosol particles between T2 and T3 sites (Cirino et al., 2018). The research in the pristine Amazon continues nowadays mainly through SONATA, a LEFE-CHAT-funded project that I coordinate aiming at studying in detail isoprene oxidation mechanisms at the ATTO site, which will be described in detail in the next section.

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3.3 The coupling of anthropogenic and biogenic emissions on the atmosphere

With an estimated budget of 500 Tg $year^{-1}$, isoprene emission is comparable to that of methane. Unlike methane, which has an atmospheric lifetime of nearly a decade and is relatively well mixed through the atmosphere, isoprene does not travel far from its source before being oxidized. It reacts rapidly with atmospheric oxidants, especially the hydroxyl radical (OH). This compound, although being identified as a relevant atmospheric species of biogenic origin and thoroughly studied for over 60 years (e.g. Rasmussen and Went, 1965), was only recognized as a relevant SOA forming species in the 21st century through a ground-breaking work at the University of Antwerp, through analysis of Amazonian aerosol filters (Claeys et al., 2004). In the late 2000s and early 2010s, much has been developed on analytical tools, leading towards the identification of some of the pathways that led to methyltetrol species identified in the seminal work of 2004. As I started my postdoc at the University of Sao Paulo in 2011, much was still unclear about its main oxidation pathways and consequent role in SOA formation.

Figure 3.3.1 presents a simplified schematic diagram of the current understanding of isoprene photooxidation. Organic peroxy radicals (ISOPOO), produced by OH attack and O_2 addition to isoprene, are reacted along nitric oxide (NO) or hydroperoxyl (HO_2) pathways or can undergo isomerization if NO and HO_2 concentrations are sufficiently low, the latter being identified even more recently (e.g. Praske et al., 2018) and still subject to strong analytical limitations. Through reaction with NO (at ambient levels > 50-100 ppt), isoprene reaction is the "classically understood route", leading to formaldehyde (HCHO), methacrolein (MACR), methyl vinyl ketone (MVK) and organic nitrates (ONIT), and further oxidizing to (low-yield) SOA forming species such as glyoxal, methylglyoxal, methacryloyl peroxynitrate (MPAN), methacrylic acid epoxide (MAE) and hydroxymethylmethyl-α-lactone (HMML). It is important to note that from an aerosol mass spectrometry perspective, this pathway leads to SOA that has no unique tracers, and thus impossible to identify uniquely. Therefore, it is generally grouped under generic terms such as Semi-Volatile Oxygenated Organic Aerosols (SV-OOA) and Low-Volatility OOA (LV-OOA).

The HO_2 route, favoured under pristine conditions (NO<50 ppt), leads to organic hydroperoxides (ISOPOOH), whose oxidations lead to isoprene epoxydiols (IEPOX), and effectively form the species detected by Claeys et al. work cited earlier with high yields, such as 2-methyltetrols, C5-alkene triols, 3-methyltetrahydrofuran-3,4-diols (3-MeTHF-3,4-diols), among others. Those compounds do present unique tracers under aerosol mass spectrometry (notably methyl-furan, at ion $C_5H_6O^+$), and are generally identified as IEPOX-SOA (Hu et al., 2015). As for the higher generation products of the 1,6 H-shift, species such as C4 and C5-LVOC are expected. It is understood that particle-phase species derived from the 1,6 H-shift do not have unique tracers on AMS and thus are typically detected under the same generic names as OOA. It is important to note that the isoprene oxidation scheme shown in Figure 3.3.1 represents its current understanding, having been significantly developed over the last 10 years.

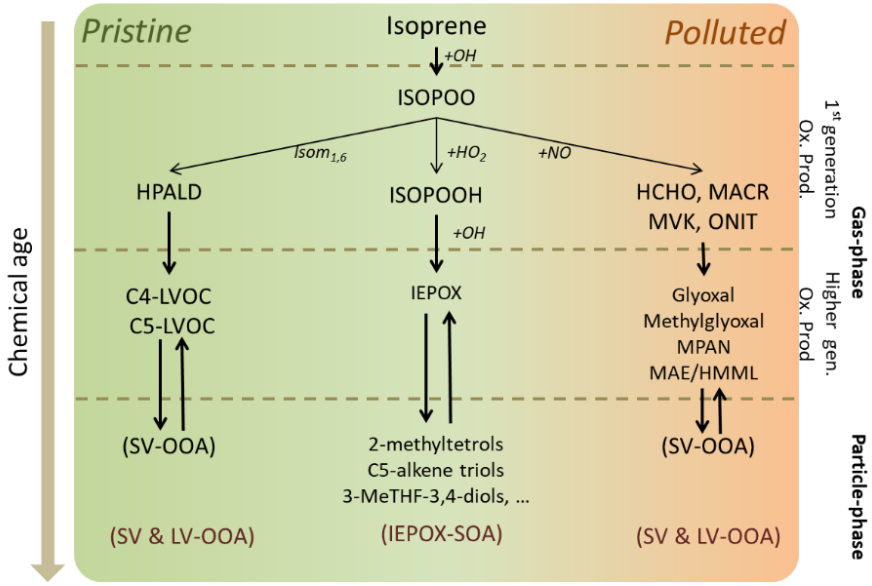

Figure 3.3.1 – Schematic diagram of isoprene photooxidation. In the presence of NO, isoprene oxidation favours the NO route. At low NO levels, ISOPOO can be scavenged along the HO_2 pathway, or undergo isomerization when NO and HO_2 concentrations are sufficiently low. The HO_2 route can lead to isoprene epoxydiols (IEPOX), an efficient aerosol-forming compound.

The first study that I participated on the topic was led by the University of Manchester during the SAMBBA campaign (Allan et al., 2014), a project addressed in the previous chapter. During that study we analysed the vertical profiles of OA spectral signature under a range of pollution levels due to biomass burning, proposing the use of a "diagnostic tracer" from AMS based on the relative signal of $C_5H_6O^+$ within total OA (termed f_{82} from the unit resolution mass of the species). This analysis has been extended to a range of other datasets, including my observations at ATTO and compiled into the reference work from the Jimenez group from the University of Colorado (Hu et al., 2015).

As described in Chapter 3.2, during GoAmazon2014/5, we collaborated with the Martin group at Harvard University. One of the topics of interest was to have a joint analysis between T0, T2 and T3 looking at the effects of urban plumes on isoprene oxidation mechanisms. Through cryogenic trap experiments, we quantified for the first time at T3 ambient levels of ISOPOOH (HO_2 route) and MVK/MACR (NO route). Based on those observations, we managed to estimate ISOPOOH levels at T3 of about 0.6 ppb, compared to 0.9 to 1.2 ppb for MVK+MACR. With this data, we managed to estimate that the shift between both regimes in the Amazon occurs at about 30-50 ppt of NO (Figure 3.3.2). Further studies of GoAmazon2014/5 on the topic where I participated included combined AMS at T3 and our ACSMs at T0 and T2, whether via PMF analysis of OA spectra, identifying IEPOX-SOA (de Sá et al., 2017), flow reactor experiments coupled to the AMS at T3 and our ACSM at T2 (Palm et al., 2018), and modelling of the Manaus plume fed by our data at T0a and contrasted with aircraft data (Shrivastava et al., 2019). Based on results obtained from GoAmazon, Ben Langford (Centre for Hydrology and Ecology) has recently led a re-analysis where we combined isoprene oxidation product fluxes obtained during BUNIAACIC with tracers of anthropogenic activities that we measured to estimate the seasonality of ISOPOOH vs MVK+MACR at the TT34-ZF2 (Langford et al., 2022). This study has identified that for

Central Amazon, acetonitrile (ACN) and BC could serve as proxies for NO, and corresponding concentrations where ISOPOOH and MVK+MACR are equally formed equates to about 0.22 ppb of ACN and 0.4 µg m^{-3} of BC.

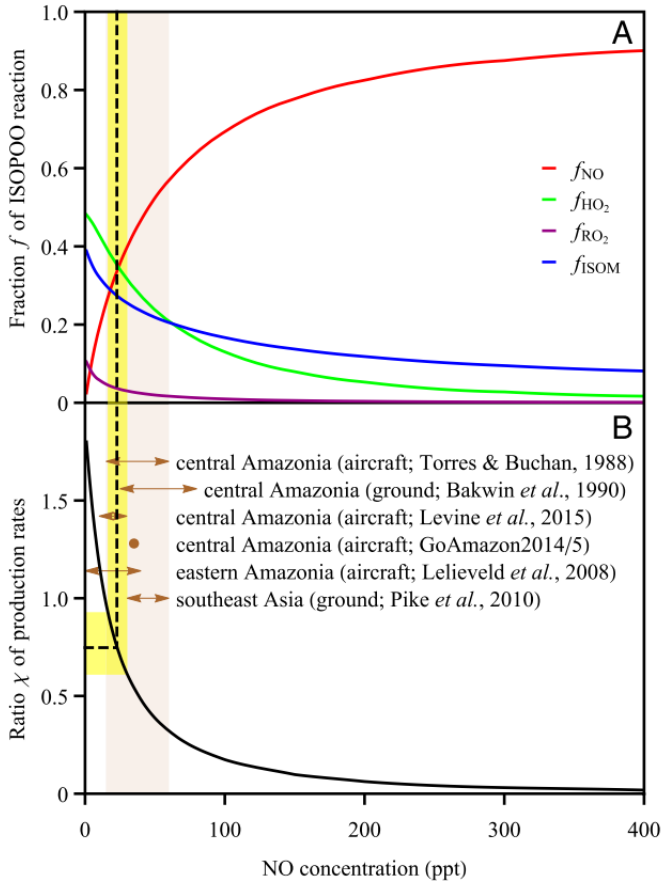

Figure 3.3.2 – Modeled dependence on NO concentration of (A) fractional contributions f_{HO2}, f_{NO}, f_{RO2}, and f_{ISOM} to ISOPOO reactive loss and (B) the ratio χ of the production rate of ISOPOOH to that of MVK + MACR. The brown arrows represent the reported ranges of NO concentration for central Amazonia, eastern Amazonia, and southeast Asia under background conditions. The dashed line projects the central value of χ for background conditions onto the effective NO concentration, illustrating the possible relative contributions of different reaction pathways of ISOPOO. The yellow region shows the same as the dashed line but for confidence interval in χ. Reproduced from Liu et al. (2016).

As addressed in Chapter 2.1, in 2016 I relocated to the LaMP, for a postdoc integrated into the EU-funded DACCIWA project. The underlying objective was to characterize the potential role of anthropogenic emissions on precipitation over southern West Africa, a region home to 350 million people with strong growth. My role in the project was to operate a compact time-of-flight aerosol mass spectrometer onboard the French ATR-42 aircraft during the field campaign that took place in June-July 2016, on 26 flights lasting over 70 h (Flamant et al., 2018b). Analysis of interest from the dataset integrated atmospheric circulation over the Gulf of Guinea focusing on aerosol particles and other anthropogenic pollutants (Flamant et al., 2018a; Deroubaix et al., 2019). Further studies have focused on detailed analysis of aerosol particle absorption enhancement (the so-called "lensing effect"), where we combined AMS composition and sizing information, aerosol light absorption and refractory black carbon (Denjean et al., 2020). It is interesting to note that in addition to significant anthropogenic

emissions, the region also has a significant forest cover, as depicted in Figure 3.3.3 (top). This motivated me to look into details on the anthropogenic-biogenic coupling, particularly on the formation of biogenic SOA particularly sensitive to anthropogenic pollution (and speciated through AMS), namely IEPOX-SOA, presented earlier in this chapter, and particulate Organic Nitrates (pON), typically associated with nighttime NO_3 oxidation of VOCs of biogenic origin.

The first step of the analysis was to provide an overview of background conditions and compare them with in-plume levels. The latter was identified through three criteria: aerosol number concentration above 2500 cm^{-3}, HOA factor from AMS explaining over 70% of OA and distance from source city <110 km. Conversely, background conditions were defined as aerosol number concentration below 1300 cm^{-3} and HOA < 70% of OA. We have identified for example that aerosol species typically presented a factor of roughly 2-5 enhancement within the plume, such as BC: 0.3 vs 0.7 µg m^{-3}, OA: 3.1 vs 6.6 µg m^{-3}, SO_4:1.7 vs 2.9 µg m^{-3}, NO_3: 0.1 vs 0.5 µg m^{-3}. Conversely, gaseous pollutants were not so strongly enhanced (e.g. NO_x had an in-plume median value < 3 ppb, compared to background 0.3 ppb, and O_3 was comparable at about 30 ppb). This indicates the large fraction of particulate pollution in megacities in the region (Abidjan, Lomé, Accra), strikingly different emission patterns compared to European cities for example, generally more marked by NOx. When looking into IEPOX-SOA and pON, both also present a marked increase within plume (0.9 vs 1.8 µg m^{-3} and 0.2 vs 0.4 µg m^{-3}, respectively), although their relative contribution to OA remains fairly constant (~30 and 6% for IEPOX-SOA and pON, respectively). Figure 3.3.3 (middle) depicts the background and in-plume species concentrations by distance from the source. It is interesting to note the dynamics of different species whether decreasing (BC and CO via dilution, or aerosol number also via coagulation), while others remain fairly constant or depict some increase due to in-plume formation such as SO_4, OA and IEPOX-SOA. To better study the dynamics, IEPOX-SOA (and its fraction on OA) is analysed according to SO_4 levels, indicating a net increase within the plume (Figure 3.3.3 bottom). As will be shown later, SO_4 (or rather the acidity) can play a major role in the reactive uptake of gaseous IEPOX, and thus the formation of this highly relevant secondary species under tropical regions.

After being recruited at IMT Nord Europe in Sep-2018, I started to place together a research project planning to study isoprene chemistry with next-generation instrumentation. Through a LEFE-CHAT project entitled SONATA (Semi-volatile Organic Aerosols from biogenic precursors in the Amazon) and the jointly supervised thesis of Carolina Ramirez Romero between IMT Nord Europe and Max Planck Institute of Chemistry (MPI-C), we deployed the Quadrupole ion guide Time-of-Flight Proton-Transfer-Reaction mass spectrometer (PTR-Qi-TofMS) from April 4 till May 2nd in 2022 at the ATTO site (Figure 3.3.4, top). The instrument was further coupled with an aerosol inlet (CHARON), briefly discussed in Chapter 3.1. The interest of the project of returning to Central Amazon was to study how pristine SOA formation processes might be affected by pre-existing aerosol properties, including long-range transport of BC-rich aerosols from African fires, or natural and anthropogenic SO_4.

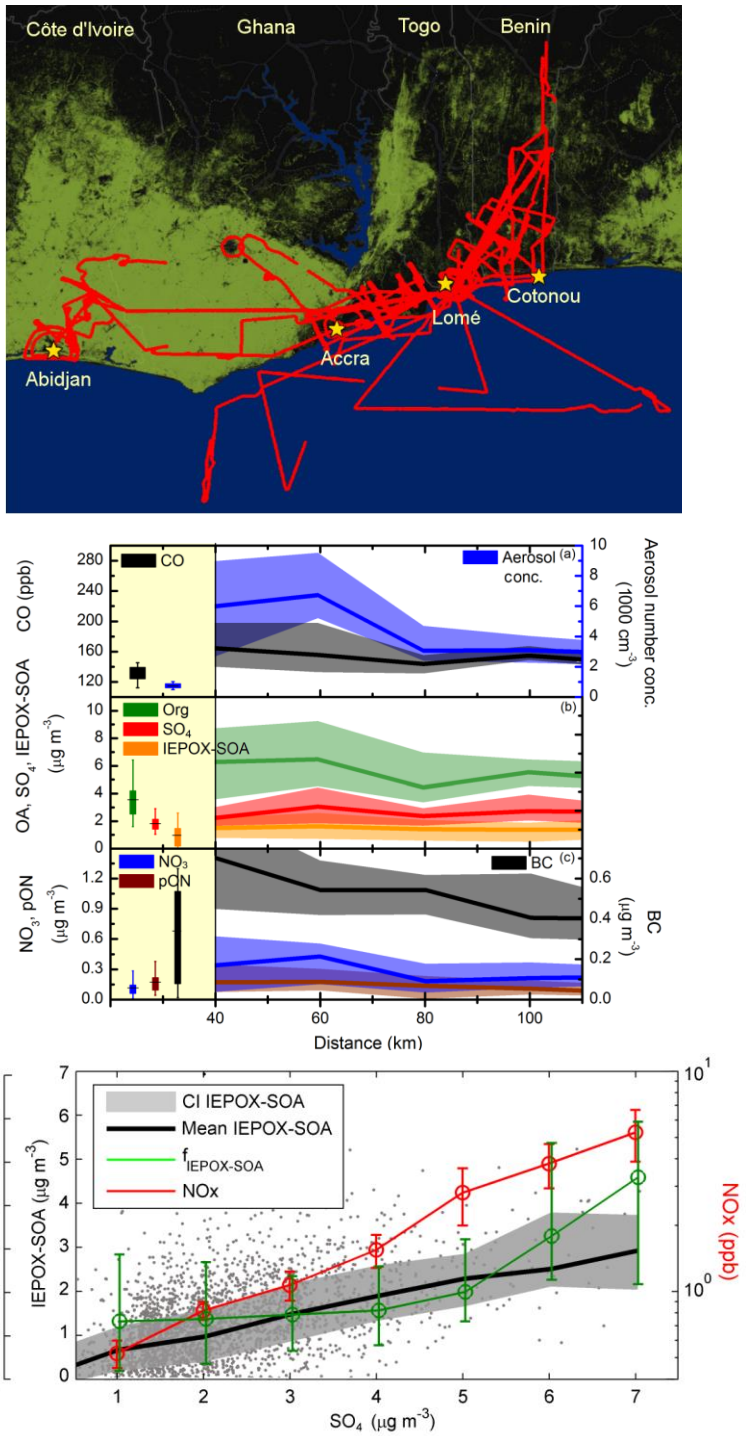

Figure 3.3.3 – Top: ATR42 trajectories (in red) during DACCIWA for altitudes below 2000 m overlaid the forest cover (in green), non-forested areas (black) and water surface (blue). Middle: Regional background (marked in yellow) and in-plume concentrations for CO and aerosol concentration (a); OA, SO_4 and IEPOX–SOA (b); and NO_3, pON, and BC (c). Bottom: Scatterplot between IEPOX–SOA concentration and SO_4. The black line and grey area represent the mean and 5 and 95 % confidence intervals of the mean, respectively. Red and green markers are mean NOx and $f_{IEPOX–SOA}$, respectively. Reproduced from Brito et al. (2018).

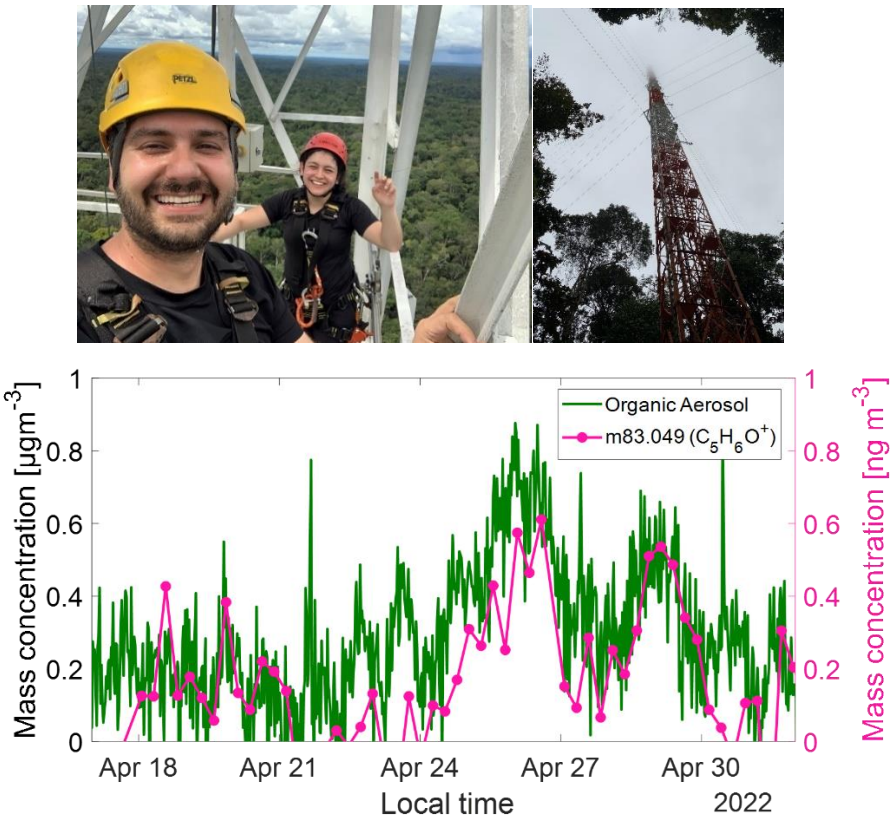

Figure 3.3.4 – Top: Climbing the ATTO tower with C. Ramirez. Bottom: Observations during the SONATA campaign at 80m at the ATTO site of OA (measured from ACSM) and methyl-furan, a tracer of IEPOX-SOA using CHARON-PTRMS. Reproduced from Ramirez et al., in preparation for Atmospheric Chemistry and Physics journal.

In addition to typical H_3O^+ ionization switching through gas and particles, we also operated the PTRMS-CHARON during SONATA in NH_4^+ mode, targeting, among other compounds, ISOPOOH, allowing for direct measurements of first oxidation isoprene products and their particle-phase counterparts. The first half of the campaign sampled below the canopy (at about 4m agl) and the second half at 80m agl. Figure 3.3.4 (bottom) depicts OA and $C_5H_6O^+$ in the particle-phase mode, associated with IEPOX-SOA. The field campaign had also periods of out-of-basin BC intrusion providing rich contrasting atmospheric conditions and allowing for deeper insights into Biogenic SOA formation in the Amazon. Whereas IEPOX-SOA has been identified through the particle-phase $C_5H_6O^+$, other tracers were also observed, however without being able to directly assign a precursor or process. Thus, after SONATA, we have conducted a range of experiments at the DouAir Chamber, at IMT Nord Europe. Developed by Dr Sébastien Dusanter and Prof. Alexandre Tomas, this Teflon chamber has nearly 9 m^3. There, we have generated (so far) three types of SOA, namely Limonene (through O_3 reaction), representative of typical monoterpene SOA, non-IEPOX isoprene SOA and IEPOX-SOA. Those three OAs are thought to be the main components of aerosol particles in pristine Amazon (Shrivastava et al., 2019), and their identification through unique tracers in CHARON will allow source apportionment analysis to be conducted not only for SONATA but also a range of biogenically impacted sampling sites. In order to generate isoprene SOA, we have used tetramethylethylene ozonolysis to generate OH, and AS seed particles as condensing surfaces. Led by both PhD students C. Ramirez and O. Murana, relative humidity and aerosol acidity were shown to modify SOA types between IEPOX and non-IEPOX isoprene SOA, as plotted

in Figure 3.3.5 (top). With this, we have obtained a range of tracer ions for each SOA, as shown in Figure 3.3.5 (bottom), for IEPOX-SOA.

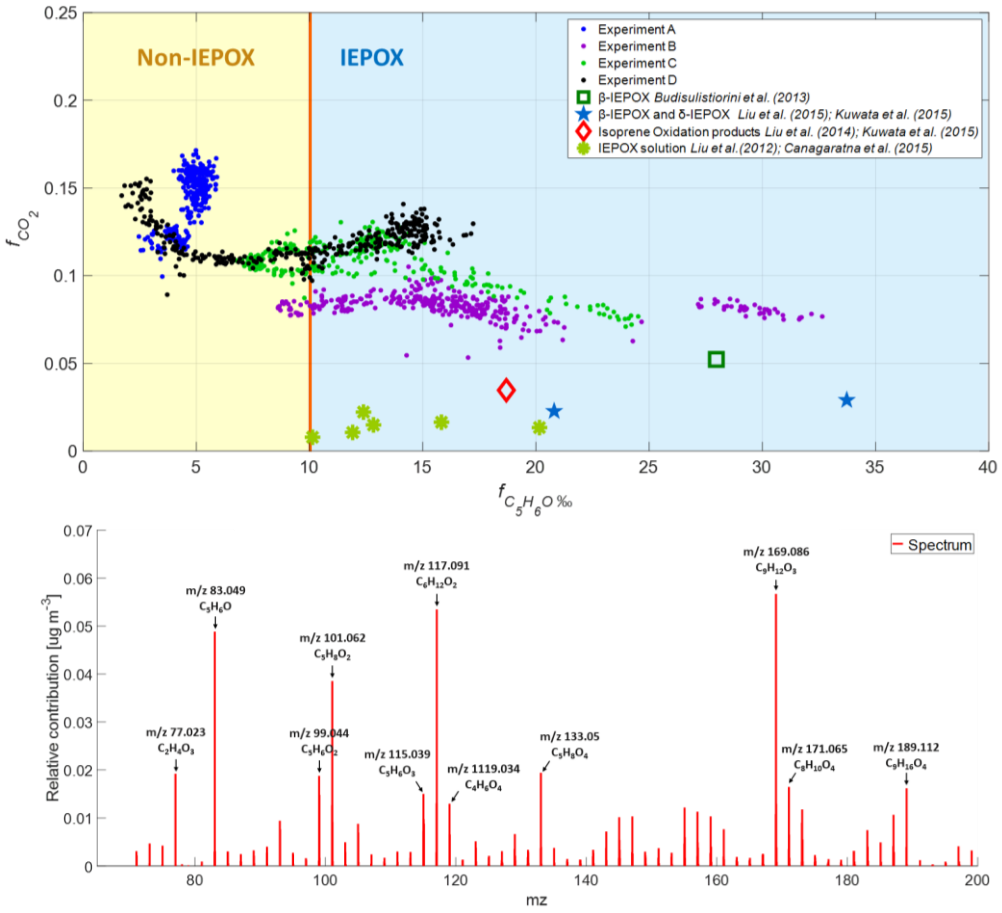

Figure 3.3.5 – Top: changes in IEPOX and non-IEPOX isoprene SOA formation during the experiments at the DouAir chamber at IMT Nord Europe. Identification was performed via HR-AMS diagnostic tracers f_{C5H6O} and f_{CO2} as proposed by Hu et al. (2015). Bottom: PTRMS-CHARON aerosol spectrum of IEPOX-SOA from chamber experiments. Reproduced from Ramirez & Murana et al., in preparation for Atmospheric Measurement Techniques journal.

Atmospheric modelling depicts a very enriching analysis to study biogenic SOA formation under a range of anthropogenic influences. For example, in a study led by colleagues from LaMP focusing on SOA formation above the Mediterranean (Freney et al., 2018), we combined AMS measurements, comparable to the DACCIWA campaign, however, integrated with a sophisticated aerosol modelling component (Polyphemus) from the CEREA. Interestingly, the model could successfully replicate the measured OA during the flights and predicted roughly 20% of OA to be pON. Albeit somewhat higher than estimated by the instrument (~ 5%, expected to be a lower limit), the contribution of this type of aerosol depicts a quite relevant biogenic SOA source in Europe. Furthermore, measurements in the continent have shown that, even in high isoprene areas, IEPOX-SOA is not the dominating route. Although NOx levels are the main hypothesis, neutralized aerosols could also play an important role. This is highlighted in the study above. Although isoprene emissions are 2.5 times higher than those of monoterpenes, isoprene-derived SOA represents about 15 to 35 % of the simulated OA, which

is lower than the monoterpene-derived SOA which represents 35 to 40 %. Combined with observations, this indicates that isoprene oxidations are mainly following the low SOA yield non-iepox route.

The study of biogenic SOA under anthropogenic influence has also been one of the scientific objectives of other projects that I have led or participated in the recent years. Notably, I coordinated the COBIACC (Campaign for the observation of aerosols and their precursors in Caillouël-Crépigny) campaign. The observations, funded by CLIMIBIO CPER and Labex CaPPA projects, took place in June and July 2019 and combined four air-conditioned housings with over 20 instruments from 7 partners looking at atmospheric dynamics, composition and reactivity at a rural site in northern France. To study SOA formation processes, I combined in-situ observations with box modelling. The latter was mainly conducted within the scope of the post-doctoral project of E. Stratigou (particularly HR-AMS and PTR-Qi-ToF-MS) or SOA tracers from filters. The box model for the COBIACC campaign (SSH-Aerosol, based on the Polyphemus) was initially developed during the M2 research project of H. Chebaicheb (supervised by Aude Bourin and myself), which I later developed using full atmospheric conditions. Figure 3.3.6 depicts COBIACC results, from modelling (top) and filter analysis (bottom). Both results indicate a predominance of monoterpene SOA (86% for a particular stagnant day, the 22nd of July 2019), and 65% throughout the campaign by filters. Interestingly, the latter also depicts the negligible contribution of IEPOX-SOA to OA, with only methylglyceric acid (i.e. non-IEPOX) contributing to Biogenic SOA formation.

More recently, in June and July 2022, the ACROSS (Atmospheric ChemistRy Of the Suburban foreSt) campaign took place in the environs of Paris, notably in a forested remote site (50 km southwest of downtown Paris), and a peri-urban and an urban sites. The project coordinator is Christopher Cantrell through “Make Our Planet Again” (MOGPA) funding, at LISA. The 12 project partners were then funded through an LEFE-CHAT project, of which I coordinated the aerosol in-situ component, together with Dr Paola Formenti. At the forested site, we operated the HR-AMS and PTR-Qi-ToFMS coupled with CHARON inlet (the latter integrating O. Murana’s PhD thesis). The interest of the site, comparable to the GoAmazon2014/5 project, is to have contrasting clean vs. polluted air masses, particularly focusing on the oxidation of biogenic species and their mixing with anthropogenic emissions from Paris. Although still in the early stages of scientific analysis, the dataset has shown some interesting variability, particularly of pON derived from the HR-AMS, shown in Figure 3.3.7. Further analysis will be conducted on a comparable strategy as COBIACC, however, this time enhanced by PTRMS-CHARON measurements.

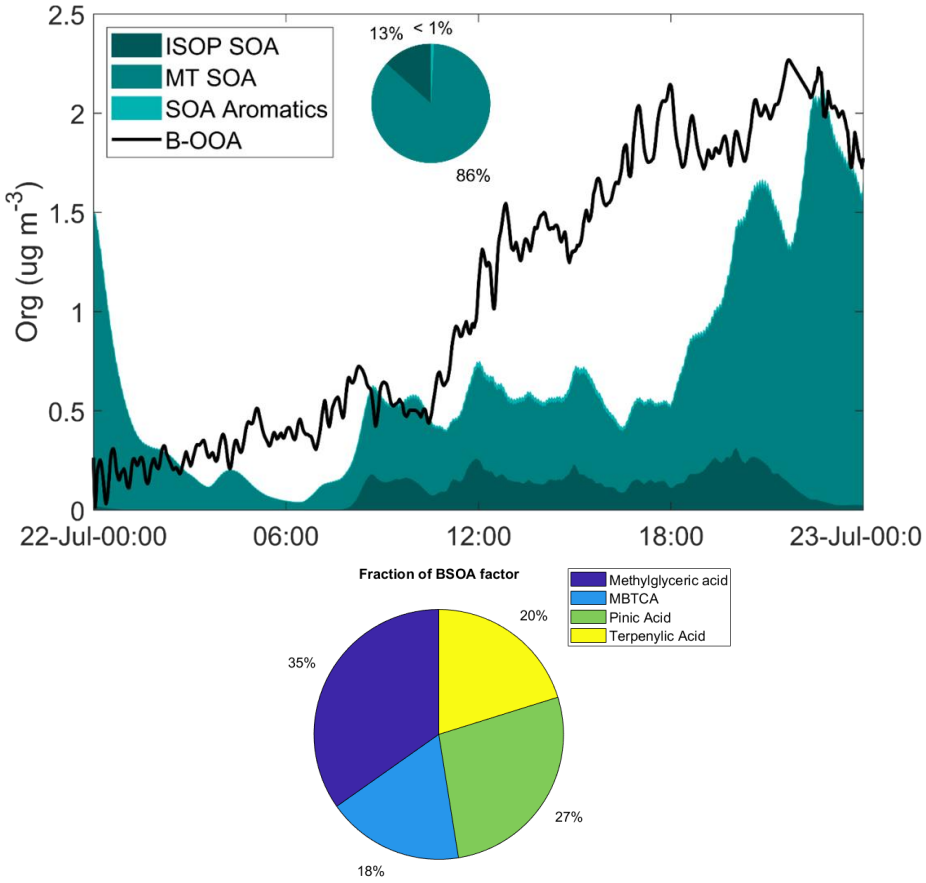

Figure 3.3.6 – Top: SSH-Aerosol simulation results for 22 Jul 2019 during the COBIACC campaign. Modelled SOA from isoprene, monoterpenes and aromatics and comparison with biogenic SOA factor (B-OOA) from HR-AMS PMF analysis (Stratigou et al., in prep.). Bottom: Multiple linear regression of OA based on different tracers of BSOA. Reproduced from Brito et al., in prep.

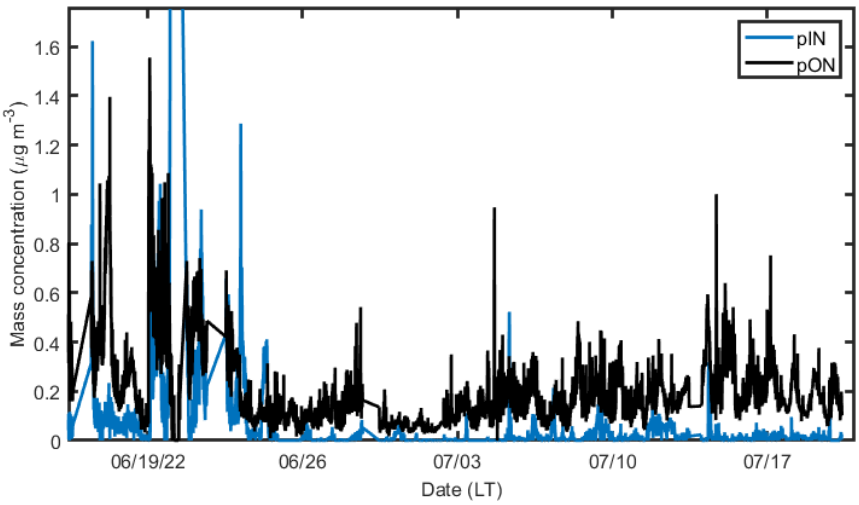

Figure 3.3.7 – Inorganic and Organic nitrate (pIN and pON, respectively) during the ACROSS field campaign. Reproduced from Brito et al., in prep.

Concluding remarks

The coupling between anthropogenic and biogenic compounds is a complex and ever more relevant topic, both by ongoing decrease of primary pollution levels, and modifications in biogenic emission profiles due to increased frequency, duration and intensity of droughts and heat events due to global climate change. This chapter has focused on how different oxidation pathways from biogenic precursors – which are often abundant in most urban environments – can be modified by the presence of anthropogenic components, may it be in the gas phase (NO, or nitrate radicals, for example), or in the changes of the particle-phase (hygroscopicity, acidity), affecting uptake and thus SOA formation. The most studied species are typically isoprene (and IEPOX-SOA being the most well-known product affected by anthropogenic levels), as well as organic nitrates, often associated with nighttime monoterpene oxidation.

I first started working on the topic in the Amazon, whether in biomass-burning impacted areas (Allan et al., 2014), downwind of Manaus (Liu et al., 2016) or pristine regions (Hu et al., 2015; Langford et al., 2022). Those have been later applied to studies in other regions, such as southern West Africa (Brito et al., 2018), and the Mediterranean Basin (Freney et al., 2018). More recently, I have been working on the topic through a range of ongoing studies including rural northern France, with an important modelling component, and, including PTRMS-CHARON observations, at ATTO, Sao Paulo, and Paris, associated with SONATA, BIOMASP+ and ACROSS projects, respectively.

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3.4 Perspectives

This Chapter has provided an overview of my research since the completion of my PhD work, focusing on urban, forested and mixed atmospheres, topics on which I plan to continue to work. Here, instead of focusing on the short-term, I will provide broad lines of what I expect to conduct research in a mid to long-term context. I expect those fields to be impacted by the following main points:

Analytical developments

Continued development of analytical tools for improved characterization of the chemical species found in the atmosphere. Much has been done in the last two decades on deployable mass spectrometers. Continued improvement of inlet systems, ultra-high-resolution mass spectrometers, and coupled/hybrid techniques will likely continue to allow us to pierce more and more into a molecular characterization of highly relevant, albeit in very low quantities, compounds. This will push further and further into semi-volatile compounds, bridging the gap of secondary organic aerosol formation. Better characterizing those species, that we started to scratch the surface through PTRMS-CHARON, will lead to improved parametrizations for secondary atmospheric processes. Within CPER ECRIN, we are currently acquiring a VOCUS mass spectrometer, allowing us to sample a wider range of compounds. I plan to continue working with next-generation instruments to continue pursuing analytical improvements leading to a better understanding of atmospheric processes and their impacts.

Aerosol source apportionment studies focusing on alternative fuels and non-exhaust emissions

Following current plants to scrape new combustion vehicles across Europe in a decade, as well as strong constraints on reducing fossil fuel usage, is likely to impact significantly the emission profile from the transport sector, road, shipping or aircraft. Assessing its impacts shall require combined statistical methods, including a range of specific tracers for better quantification of the future generation of energy matrices for the sector. I plan to continue making use of state-of-the-art statistical tools, such as Rolling PMF, in combination with other online instruments towards improved aerosol source apportionments. In that context, the ATOLL site, and more generally the work developed within the framework of ACTRIS, will be extremely valuable under homogenized, high-quality controlled extensive datasets. Other projects, such as UNREAL, looking at aerosol emission through new engine technology, shall continue to be explored in the foreseeable future.

Modified atmospheric chemistry resulting from shifts in NO_x levels

As a result of climate-friendly policies forcing a strong reduction of fossil fuel combustion described above, it is expected that nitrogen oxides will decrease significantly, in a comparable fashion as SO_2 or CO have done in the not-so-distant past. The effect will likely shift quite significantly atmospheric oxidations, notably organic peroxides, towards "low-NO_x" and auto-oxidation chemistry in urbanized areas. Improved modelling of urban atmospheres will rely on observations constraining isomerization pathways, significantly under-studied compared to "high" and "low-NO_x". Continuing the work on quantifying IEPOX vs. non-IEPOX SOA, I

expect to be able to characterize those formation mechanisms attributed nowadays to “remote” sites, for a more complete view of isoprene chemistry under relevant atmospheric conditions.

Impacts of climate change

If, on one hand, the air is expected to be cleaner in urbanized areas due to the reduction in fossil fuel consumption, it will also undoubtedly be negatively affected by episodic events of uncontrolled forest fires during heat and drought spells. Much work will have to be done on how to prevent those events, but also to accurately assess their climatic and health effects. Making use of long-term in-situ observations, combined with remote sensing (ground and satellite), I plan to continue the work developed during SAMBBA and the PhD work of Alejandra Velazquez-Garcia and be able to provide an improved characterization of the climatic effects of forest fire plumes across Europe. Those will focus, for example, on their spectral absorbance (Brown Carbon content), chemical composition, lifetime, hygroscopicity and so forth. Conversely, I expect to continue collaborating in the Amazon forest, and study aerosol population modification induced by Climate Change, may it be from changes in fluxes of mineral dust from the Sahara, forest fires (in or out of the basin), vegetation emission profile during climatic extremes and so forth. For that, observations at the ATTO tower will provide crucial information concerning Central Amazon.

Put together, the points discussed above point towards ongoing challenges in the field of atmospheric research, particularly focusing on aerosol particle sources and effects. Those are associated largely with current and expected changes in global climate, whether induced via policies to mitigate it or as direct results of its effects.

List of Figures

Appendix

A. CV Détaillé

Etat Civil

Nom : FERREIRA DE BRITO

Prénoms : Joel

Née : le 30 Décembre 1982 à Sao Paulo, Brésil

Nationalité : Brésilienne et Italienne

Situation familiale : Marié, 2 enfants (2012, 2015)

Situation professionnelle : Enseignante-chercheur à l'Institut Mines-Télécom Nord Europe (CERI Energie Environment) depuis le 03 Septembre 2018

Adresse professionnelle : 941 rue Charles Bourseul, 59500 Douai

Téléphone : 03 27 71 26 09

Courriel professionnel : joel.brito@imt-nord-europe.fr

Thématique de recherche

Composition de l'atmosphère terrestre, particulièrement les particules fines en suspension, leurs caractéristiques physico-chimiques et impacts.

Parcours

- 2018 – Enseignant-chercheur à l'Institute Mines Télécom Nord Europe (CERI EE), Douai, France.
- 2016 – 2018 - Post-doctorat à Université Clermont Auvergne (LaMP), Clermont-Ferrand, France.
- 2011 – 2016 - Post-doctorat à Université de Sao Paulo (Artaxo group), São Paulo, Brésil.
- 2007 – 2011 - PhD en Physique Atmosphérique, *« A lightweight high-sensitivity chemical mass spectrometer for organic compounds »*, Karlsruhe Institute of Technology (KIT), Karlsruhe, Allemagne. Direction : Johannes Orphal.
- 2005 – 2007 - Master en Acoustique et Vibrations, *« La méthode de retournement temporel dans le domaine de l'audible »*, Institut National des Sciences Appliquées (INSA), Lyon, France.
- 2001 – 2005 - Bachelor en physique, University of Campinas, Brésil.

Enseignement et encadrement pédagogique

Enseignement :

- 2023-2024, *Dispersions des pollutions atmosphériques et risque industriels en Master 1,* IMT Nord Europe, France.

- 2022-2024, *Transition Ecologique et Sociétale en Licence 3 (Travaux Dirigés)*, IMT Nord Europe, France.
- 2021, *Electromagnétisme/Optique en Licence 2 (Travaux Pratiques)*, IMT Nord Europe, France.
- 2020, *MOOC Air Quality session*, IMT Nord Europe, France.
- 2018-2024, *Mécaniques des fluide en Licence 3*, IMT Nord Europe, France.
- 2013, *Summer School IFUSP*, University of Sao Paulo, Brésil.
- 2012, *Summer School IFUSP*, University of Sao Paulo, Brésil.
- 2011 (1 semestre), *Electromagnétisme en Licence 2 Physique/Météorologie*, University of Sao Paulo, Brésil.

Encadrement des doctorants ou des étudiants de master

- 2022-, Olatunde Murana (PhD), *Understanding air masses of anthropogenic and biogenic origin in Paris and Sao Paulo*, IMT Nord Europe, France. Direction: Véronique Riffault.
- 2020-2023, Hasna Chebaicheb (PhD), *Sources of fine aerosols at various French sites using highly time-resolved multiyear datasets*, IMT Nord Europe /INERIS, France. Direction: Véronique Riffault.
- 2020-, Carolina Ramirez Romero (PhD), *Study of gas and particle phase partitioning of semi-volatile biogenic organic compounds in central Amazon*, IMT Nord Europe, France/Max Planck Institute, Germany. Direction : Stéphane Sauvage.
- 2019-2023, Alejandra Velazquez-Garcia (PhD), *Chemical and optical properties of particulate pollution in the Lille area, Northern France based on ATOLL observations*, IMT Nord Europe/LOA Université de Lille, France. Direction : Véronique Riffault.
- 2019, Hasna Chebaicheb (M2): *Etude de la dynamique des aérosols atmosphériques pendant l'été dans une région rurale au nord de la France*, IMT Nord Europe, France.
- 2011 – 2015, Glauber Cirino (PhD): *Physico-chemical characterization of aerosols during GoAmazon2014/5 experiment: interaction between Manaus and pristine forest emissions*, University of São Paulo & INPA, Brésil. Direction : Paulo Artaxo.
- 2013 – 2015, Florian Wurm (PhD): *Sources and impacts of Volatile Organic Compounds in the vicinity of Manaus during GoAmazon2014/5 experiment*, University of São Paulo, Brésil. Thèse arretée au 3eme année. Direction : Paulo Artaxo.
- 2014 – 2015, Rafael Stern (Master): *Physico-chemical characterization of aerosols during 2013 dry season in Central Amazonia*, University of São Paulo & INPA, Brésil.
- 2013 – 2015, Djancinto M. dos Santos (Master): *Vehicular emissions in Sao Paulo: quantification of their effect using receptor models and their carbonaceous content*, University of São Paulo, Brésil.

Encadrement de post-doctorants

- 2022-, Vishnu Murari, *Characterization of aerosol particles from shipping emissions in Dunkirk*, IMT Nord Europe, France.

- 2021, Layal Fayad, *Development of an innovative VOC sampling system for drone measurements,* IMT Nord Europe, France.
- 2020-2021, Antoine Farah, *Chemical characterization of aircraft engine exhaust based on chamber experiments,* IMT Nord Europe, France.
- 2019-2020, Evdokia Stratigou, *Inorganic vs organic secondary aerosol production in north of France within the context of climate change,* IMT Nord Europe, France.

Encadrement projets ouverts (L3)

- 2022 – Encadrement d'une équipe de cinq élèves en travaillant avec une classe ULIS au sein de l'école Saint-Jean à Douai. L'objectif du projet était de les sensibiliser à la problématique du spatial de façon ludique, avec des jeux, démonstration du vide et une fusée.
- 2023 – Avec l'objectif de sensibiliser une classe de 4eme sur la problématique du changement climatique et de la pollution de l'air, cette équipe de 5 élèves a conduit des expériences, des visites à une déchetterie, des clean walks, des jeux de société et des débats.

Participation à jury de thèse ou master :

- 2022, François Hemeret (comité suivi thèse) : *Constraining the radiative effects of complex aerosol mixtures in southern Africa: an experimental study of their chemical composition and spectral optical properties*, LISA, France. Direction : Paola Formenti.
- 2021-2022, Francesco Battaglia (comité suivi thèse) : *Altération des poussières minérales par les composés organiques volatiles d'intérêt climatique : composition chimique et propriétés optiques de mélanges complexes en fonction du vieillissement atmosphérique*, LISA, France. Direction : Paola Formenti.
- 2021, Manon Rocco (jury de thèse) : *Analyse des déterminants de la distribution des composés organiques volatils en milieux naturels contrastés*, UCA, France. Direction : Jean-Luc Baray.
- 2020, Chang Yuyang (jury de Master): *Dust Aerosol Observation (DAO) campaign: Analysis of aerosol events observed at Kashi site.* LOA, France. Encadrement : Philippe Goloub et Qiaoyun Hu.
- 2018, Yunjiang Zhang (comité suivi thèse) : *Development of monitoring stations for the measurements of aerosol chemistry and physics in France*, LSCE, France. Direction : Valerie Gros.
- 2015, Beatriz S. Oyama (jury de thèse) : *The role of vehicular emissions on organic aerosol chemical composition in São Paulo*, University of São Paulo, Brésil. Direction : Maria de Fatima Andrade
- 2013, Antônio T. Bittencourt (jury de Master): *Characterization of CCN in São Paulo: the influence of chemical composition and size distribution on their properties,* UEC, Brésil. Encadrant: Gerson Almeida

Production scientifique

Web of science: https://www.webofscience.com/wos/author/record/1772419

ORCID: 0000-0002-4420-9442

- Facteur H : 32 (Web of Science) avec 2964 citations.
- 72 articles de rang A dont 16 en tant que 1er ou 2ème auteur.
- 16 présentations dans des congrès scientifiques internationaux (1er auteur), dont 2 invitées.
- 1 chapitre de livre.

Projets de recherche

- Participation à 23 projets de recherche dont 8 en tant que coordinateur de projet (BRAFITEC IDE, LEFE-CHAT SONATA, InterCERI ALPAGA), responsable scientifique (ANR BIOMASP, FAPESP GAIA, PETROBRAS FONTES) ou responsable de tâche/sous-tâche (CPER ECRIN, ADEME PIRATE, LEFE-CHAT ACROSS).

Révision et prise de décision scientifique

- Révision de 8 projets (H2020 Curie, EU Cost Action, DIM Qi2, LEFE-CHAT, FAPESP Thematic projects)
- Révision de 44 articles pour 14 journaux à comité de lecture
- Membre du corps éditorial de Toxics (MDPI) et Atmosphere (MDPI), prise de décision sur 11 articles.

Comités scientifiques

- 2023 - Pilotage du groupe de travail en Spectrométrie de masse d'aérosols dans le cadre du NF Forum ACTRIS.
- 2023 - Comité français du programme satellitaire Aerosol Observation System (AOS, CNES/NASA/JAX), WG Aérosol (piloté par Juan Cuesta, LISA).
- 2022 - GT "aérosols et nuages in situ" pour la préparation du nouveau jet "ANVOLE"/SAFIRE (piloté par Paola Formenti, LISA).

B. Production Scientifique

B.1 Articles dans des revues internationales à comité de lecture

1. Chebaicheb, H., de Brito, J. F., Chen, G., Tison, E., Marchand, C., Prévôt, A. S. H., Favez, O., and Riffault, V.: Investigation of four-year chemical composition and organic aerosol sources of submicron particles at the ATOLL site in northern France, Environmental Pollution, 121805, https://doi.org/https://doi.org/10.1016/j.envpol.2023.121805, 2023.

2. Crumeyrolle, S., Kontkanen, J. S. S., Rose, C., Velazquez Garcia, A., Bourrianne, E., Catalfamo, M., Riffault, V., Tison, E., de Brito, J., Visez, N., Ferlay, N., Auriol, F., and Chiapello, I.: Measurement report: Atmospheric new particle formation at a peri-urban site in Lille, northern France, Atmos Chem Phys, 23, 183–201, https://doi.org/10.5194/acp-23-183-2023, 2023.

3. Velazquez-Garcia, A., Crumeyrolle, S., de Brito, J. F., Tison, E., Bourrianne, E., Chiapello, I., and Riffault, V.: Deriving composition-dependent aerosol absorption, scattering and extinction mass efficiencies from multi-annual high time resolution observations in Northern France, Atmos Environ, 119613, https://doi.org/10.1016/j.atmosenv.2023.119613, 2023.

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5. Savadkoohi, M., Pandolfi, M., Reche, C., Niemi, J. V, Mooibroek, D., Titos, G., Green, D. C., Tremper, A. H., Hueglin, C., Liakakou, E., Mihalopoulos, N., Stavroulas, I., Artiñano, B., Coz, E., Alados-Arboledas, L., Beddows, D., Riffault, V., De Brito, J. F., Bastian, S., Baudic, A., Colombi, C., Costabile, F., Chazeau, B., Marchand, N., Gómez-Amo, J. L., Estellés, V., Matos, V., van der Gaag, E., Gille, G., Luoma, K., Manninen, H. E., Norman, M., Silvergren, S., Petit, J.-E., Putaud, J.-P., Rattigan, O. V, Timonen, H., Tuch, T., Merkel, M., Weinhold, K., Vratolis, S., Vasilescu, J., Favez, O., Harrison, R. M., Laj, P., Wiedensohler, A., Hopke, P. K., Petäjä, T., Alastuey, A., and Querol, X.: The variability of mass concentrations and source apportionment analysis of equivalent black carbon across urban Europe, Environ. Int., 178, 108081, https://doi.org/https://doi.org/10.1016/j.envint.2023.108081, 2023.

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B.2 Articles de rang A soumis

- "Apparent Tropospheric Observation of Criegee Intermediate Oligomerization Reaction Signatures" by Caravan et al. Submitted to Nature Geosciences.
- "Assessment of light-absorbing carbonaceous aerosol origins and properties at the ATOLL site in Northern France" by Velazquez-Garcia et al. Submitted to Aerosol Research.

B.3 Chapitres de livre

- P. Artaxo, H. Barbosa, G. Jorge, J. F de Brito; E.T. Sena, L. V. Rizzo, G. Cirino, A. M. Yanez-Serrano. A Amazônia em processo de mudanças ambientais. In: Tercio Ambrizzo; Livia Dutra; Pedro Jacobi. (Org.). Ciência das mudanças climáticas e sua interdisciplinaridade. 1ed. São Paulo: Annablume, 2015, v. 1, p. 39-56.

B.4 Communications invitées

- On the sensitivity of isoprene oxidation route to pollutants and its atmospheric impacts. Groupe Français de Cinétique et Photochimie, Rennes, France, 2019.

- Atmospheric chemistry and dynamics throughout the Amazon Region, measurements in pristine, deforested and urban environments. Brainstorm Coffee Farm Conference, Campinas, Brazil, 2013.

B.5 Communications dans des conférences à comité de revue en 1er ou 2ème auteur

A ce jour, je compte environ 120 communications dans des conférences à comité de revue. La liste ci-dessous est restreinte aux travaux en 1er ou 2ème auteur.

1. C. Ramirez-Romero, J.F. de Brito, S. Dusanter, M. Jamar, A. Tomas, H. Bouzidi, A. Lahib, L. Fayad, M.A. Franco, S. Carbone, S. Wolff, A. Edtbauer, J. Williams, P. Artaxo, C. Pöhlke and S. Sauvage. Characterization of Secondary Organic Aerosol formation under forested pristine conditions. European Aerosol Conference, Malaga, Spain, 2023. *Oral*

2. J.F. de Brito, A. Oliveira, M. Staudt, S. Souza, A. Borbon and A. Fornaro. The interaction between vegetation and human emissions and their effect on air quality in Sao Paulo in a context of climate change. Dialogues franco-lusophones « Villes en temps de crises démocratiques et climatiques », Paris, France, 2023. *Oral*

3. C. Ramirez-Romero, J.F. de Brito, S. Dusanter, M. Jamar, A. Tomas, H. Bouzidi, A. Lahib, L. Fayad, M.A. Franco, S. Carbone, S. Wolff, B. A. Holanda, L. A. Kremper, A. Edtbauer, J. Williams, P. Artaxo, C. Pöhlker and S. Sauvage. Laboratory characterization of SOA tracers from biogenic precursors using a Proton Transfer Reaction Mass Spectrometry coupled to a CHARON inlet. 3rd Innovation In Atmospheric Measurement Techniques, virtual, 2023. *Oral*

4. S. Dusanter, J. F. de Brito, A. Lahib, A.Tomas, M. Jamar, C. Cantrell, V. Michoud, V. Daele, A. Kukui, C. Xue, J. Houny. Observations of trace gases above & below a forest canopy during ACROSS. European Geophysical Union, Vienna, Austria, 2023. *Oral*

5. J. F. de Brito, P. Formenti, S. Dusanter, A. Tomas, L. Alleman, E. Perdrix, P. Espina, V. Riffault, C. Yu, C. Di Biagio, A. Gratien, L. Hawkins, B. D'Anna, J. Kammer, A. Monod, J.E. Petit, S. Deshmukh, L. Poulain, V. Michoud, and C. Cantrell. Contrasting aerosol composition in and out of Paris plume during the ACROSS campaign at the Rambouillet supersite. European Geophysical Union, Vienna, Austria, 2023. *Oral*

6. A. Velazquez-Garcia, J.F. de Brito, S. Crumeyrolle, E. Tison, E. Bourrianne, I. Chiapello, V. Riffault. Studying light-absorbing aerosol properties in Northern France through combined in-situ observations, emission inventory, and individual back-trajectory analyses. International Aerosol Conference, Athens, Greece, 2022. *Poster*

7. C. Ramirez-Romero, J.F. de Brito, S. Dusanter, M. Jamar, A. Tomas, H. Bouzidi, A. Lahib, L. Fayad, M.A. Franco, S. Carbone, S. Wolff, A. Edtbauer J. Williams, P. Artaxo, C. Pöhlker and S. Sauvage. Secondary Organic Aerosol formation under pristine rain-forest conditions. International Aerosol Conference, Athens, Greece, 2022. *Poster*

8. H. Chebaicheb, J.F. de Brito, O. Favez, C. Marchand and V. Riffault. 5-year analysis of submicron aerosol characteristics and organic aerosol source apportionment of in northern France. International Aerosol Conference, Athens, Greece, 2022. *Oral*

9. A. Farah, J. F. de Brito, M. Cazaunau, R. Barrellon-Vernay, A. Berge, C. Focsa, J.-F. Doussin, I. Ortega, V. Riffault. Impact of aircraft fuel chemical composition on the physicochemical properties of primary and secondary particulate matter emissions. European Aerosol Conference, virtual, 2021. *Poster*

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B.6 Rapporteur de publications

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- Environment Science & Technology (5, IF : 11.4)
- Atmosphere (4, IF : 3.1)
- Environnement Pollution (3, IF : 10.0)
- Journal of Environmental Sciences (3, IF : 6.8)
- Atmospheric Pollution Research (2, IF : 4.8)
- Earth and Space Science (2, IF : 3.7)
- Environmental Science and Pollution Research (2, IF : 5.2)
- Journal of Geophysical Research (2, IF : 5.2)
- Acta Amazonica (1, IF : 1.1)
- Building and Environment (1, IF : 7.1)
- Environmental Science: Atmospheres (1, IF : N/A)
- Environmental Science: Processes & Impacts (1, IF : 5.3)
- Journal of Environmental Science and Technology (1, IF : 3.5)
- Nature (1, IF : 60.9)