Laboratoire Inter-universitaire des Systèmes Atmosphériques

**PhD Student Opportunity at University Paris-Est Creteil, France, deadline 31th July 2020. Start between 01/09/2020 and 01/01/2021**

# Constraining the direct radiative effect of « *black carbon* » and « *brown carbon* » on climate: an innovative experimental study on their spectral optical properties (funded ANR project B2C)

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It is now well recognized that atmospheric aerosols contribute directly to global climate change through the scattering and absorption of solar (SW) and longwave (LW) radiation in the atmosphere (IPCC, 2013). Most aerosol species (sulphates, sea salts, mineral dust, ...) are mostly scattering in the SW domain (absorption < 10-20%) and their direct effect is negative, i.e. they induce a cooling of the Earth-atmosphere system by increasing the planetary reflectivity. This cooling effect opposes and partly mitigates the warming due to greenhouse gases. On the other hand, particulate "black carbon" (BC) and "brown carbon" (BrC), generated by the incomplete combustion of fossil fuels or biomasses or by atmospheric reactions of gaseous pollutants, strongly absorb light in the solar spectrum, which induces a warming effect. BC and BrC contribute to more than 80% of the absorption by aerosols and represent the main source of global warming after carbon dioxide, both globally and regionally (e.g., Bond et al., 2013; Feng et al., 2013).

The direct radiative effect of BC and BrC, although very large and expected to increase in the next decades due to the increase in fire frequency and growing anthropization, remains to date one of the largest uncertainties in the assessment of global climate forcing (IPCC, 2013). The level of scientific understanding of the fundamental mechanisms of aerosol-radiation interactions for these species remains very low, resulting in the inability to represent the spectral optical properties of BC and BrC in climate models (Boucher et al., 2016; Samset et al., 2018). These properties are: the complex refractive index (m=n-i.k; the intensive parameter describing the aerosol scattering and absorption capacity), the absorption, scattering, and extinction cross sections (MAC, MSC, MEC, $m^2g^{-1}$), and the single scattering albedo (SSA, the ratio of particle scattering to extinction). At present, one of the limiting factors remains our inability to represent the spectral optical properties of BC and BrC under ambient conditions (i.e. as a function of RH) and their possible modifications according to different atmospheric ageing processes (heterogeneous reactions with gaseous species, mixing, ...), as well as the link with the particle chemical composition. Although these properties have been widely documented in the field, the challenge is currently to move from an empirical knowledge to a systematic description for a better representation of these properties in climate models and satellite restitution algorithms. Satellite measurements are indeed key for detecting and quantifying the presence of these aerosols in the atmosphere on a regional and global scale, so to estimate their impact on air quality.

The objective of this thesis work is to spectrally resolve the ambient optical properties of BC and BrC aerosols and to understand their variability in relation to formation processes/conditions and atmospheric ageing. This work will be based on original experiments performed at the CESAM atmospheric simulation chamber developed at LISA (http://cesam.cnrs.fr/; Wang et al., 2011, Di Biagio et al., 2014, 2019), a 4.2 m3 chamber where aerosols can be generated, aged and deposited in a controlled manner. The BC and BrC aerosols will be generated by combustion of gaseous and liquid fuels by means of commercial generators (miniCAST) and by aqueous and non-aqueous phase reactions in situ in the CESAM chamber. Different BC and BrC ageing processes will be simulated (aerosol mixing, organic and inorganic coating formation). Multiple on-line measurements will be made on the chamber and laboratory analyses will be performed on aerosol samples taken during experiments to characterize the full range of physical, chemical, morphological and optical properties of the particles. In particular, the work will take advantage of the recent instrumental developments of the CESAM chamber, notably the new UV-Vis spectrometer which allows the measurement of the aerosol extinction spectra at high spectral resolution and at different levels of relative humidity in situ in the chamber in the SW domain, in addition to the LW spectrometer already operational in CESAM. The acquired experimental data will be combined with numerical modelling (Mie, core-shell Mie, RDG-FA Rayleigh-Debye-Gans theory for Fractal-Aggregates) to determine the essential optical properties of BC and BrC over the whole UV-Vis-LW spectrum.

**Context**: This thesis work is part of the B2C (Black and Brown Carbon) project which has been funded by the French National Research funding Agency (ANR) for the period 2020-2024 and of the EUROCHAMP-2020 activity (https://www.eurochamp.org/). The B2C project aims at reducing uncertainties in the estimation of the radiative effect of carbonaceous aerosols on climate. The primary objective of the project is to elucidate the dependence of the spectral optical properties of BC and BrC on the physico-chemical properties of the particles, as well as their modifications during atmospheric transport in order to improve their representation in climate models and satellite restitution algorithms.

**Expected results.** The study will allow characterizing the spectral absorption properties of particulate black and brown carbon and to relate these properties to the chemical composition, size and morphology of the particles and their modifications during atmospheric ageing through numerical parameterizations. These parameterizations based on laboratory experiments will be transferred to French climate models, in particular to the IPSL LMDz model (used by the IPCC) and satellite restitution algorithms such as those developed at LISA. This work is based on a new state-of-the-art tools from the CESAM chamber (new UV-Vis cell) to answer one of the questions identified as a priority by the IPCC international committee: what are the spectral optical properties of carbonaceous aerosols? How do these properties vary according to the source / combustion process and their residence time in the atmosphere? Answering these questions will allow major advances in estimating the radiative forcing of these major aerosol species on the global climate.

**Hosting laboratory**. The thesis will be realized at LISA (Laboratoire Interuniversitaire des Systèmes Atmosphériques, http://www.lisa.u-pec.fr/fr) at the campus of the University of Paris-Est Creteil (https://www.u-pec.fr/), in the suburbs of Paris. The student will integrate the MEREIA group, a dynamic team of scientists working on many aspects of atmospheric chemistry and physics.

**Possible collaborations:** This thesis work is articulated around measurement campaigns in the CESAM simulation chambers to which several French and European research laboratories will participate by providing specific expertise on the physico-chemistry of aerosols, under the umbrella of the B2C and EUROCHAMP-2020 projects. The PhD student will be led to interact and collaborate with these different actors, which will contribute crucially to his/her scientific enrichment. The MEREIA group is also very actively involved in collaborations with several foreign research groups working on the properties of BC and BrC through experiments in the CESAM chamber. The PhD student will be able to take advantage of this scientific dynamism within the MEREIA group and interact in a privileged way with several actors of the international community working on the same research topic.

**Profile and skills**: The candidate must have a master's degree in chemistry or physics and have a good knowledge of atmospheric sciences. Knowledge of radiation transfer and/or organic chemistry will also be appreciated. The candidate must have programming skills (R, IDL, Python,...), capacities in statistical analysis of data sets, a good level of English (oral and written), as well as a taste for teamwork and should also show strong motivation.

**Please send applications including CV, motivation letter and name of one or two reference persons to** Jean-Francois Doussin (Jean-Francois.Doussin@lisa.u-pec.fr) and Claudia Di Biagio (Claudia.Dibiagio@lisa.u-pec.fr)

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