FINAL REPORT PROGRAM LEFE

Program LEFE-CHAT	Project Title : BlAck Carbon Optical aging (BACON)	Years 2019 – 2021

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Context: Black carbon (BC) is a significant component of atmospheric aerosols and the major contributor to aerosol absorption and direct radiative effects. However, large uncertainties still exist in understanding and representing the spectral optical properties of BC in models and remote sensing algorithms.

Objectives / scientific questions: the objective of the BACON project is to study the spectral optical properties of particulate BC in the solar domain and their variability in relation to atmospheric aging. The project aims to answer two scientific questions in particular: 1. What are the mass extinction, absorption, and scattering coefficients (respectively MEC, MAC, MSC, in m^2 g^{-1}), the simple scattering albedo (SSA) and the complex refractive index (of particulate BC and how do these properties change during aging due to the presence of organic and inorganic coatings on BC particles? 2. What is the relationship between these properties (their intensity and spectral variation) and the composition of BC aerosols during ageing?

Main results: the BACON project is based on original experiments in the 4.2 m^3 CESAM simulation chamber (Wang et al., 2011). The environment of the CESAM chamber enables us to reproduce realistic conditions (i.e., particle lifetime and concentrations, multiphase chemistry, irradiation) and allow aerosols to be generated, held in suspension, aged, and deposited under controlled conditions. The chamber is equipped with state–of–the–art instrumentation for the measurement of particle number size distribution (SMPS, OPCs), spectral optical properties (both via ex situ commercial instrumentation, such as nephelometer, aethalometer, extinction cavities, and dedicated in situ spectroscopy lines measuring from UV–Vis to IR wavelengths) and chemical composition (both online (ACSM) and offline by means of analysis on collected filter samples). For the BACON experiments, the CESAM set up was upgraded with the SP2 (Single Particle Soot Photometer, collab. IGE and CNR-ISAC) and with a CPMA (Centrifugal Particle Mass Analyser, collab. CORIA) for detailed characterization of the BC refractory mass concentration (SP2) and particle effective density (CPMA), necessary to properly constrain the BC mass fraction in aerosols and particles' physical properties.

In order to study the optical properties of the BC and to be able to compare/combine the results with most of the literature, a standard generation method is used during CESAM experiments. The generation of particulate BC is performed by a commercial miniCAST burner (JING model 5200, 6204C). The miniCAST is currently one of the most widely used in laboratory experiments, recognised as a very stable and reproducible BC generator producing particles with a wide range of morphological, optical and chemical properties. The miniCAST allows the generation of BC nanoparticles via a propane–nitrogen diffusion flame assisted by an air jet.

The simulation experiments carried out during BACON enabled us to develop and optimize protocols for the injection, ageing and analysis of the physical-chemical properties of the primary soot particles generated by the incomplete combustion of propane. A series of experiments have been then conducted at the CESAM simulation chamber in order to study the spectral optical properties of particulate BC and the impact of aging on these properties. Those include aging in: 1/ dark dry and humid conditions with high and low soot mass concentrations; 2/ irradiation in dry/humid conditions; 3/ irradiation in the presence of humidity, ozone, and SO_2 inducing the formation of sulphate aerosols and formation of internal/external mixtures with BC; 4/ presence of Secondary Organic Aerosol (SOA) generated by the ozonolysis of alpha-pinene inducing formation of internal/external mixtures with BC. Figure 1 shows an example of temporal evolution of the particle number concentration size spectrum measured in the CESAM chamber for minicast soot aerosols aged under dry dark conditions over 22 hours.

The analysis of the ensemble of observations from the BACON experiments suggests that it is possible to spectrally resolve the variation of the optical properties of BC–containing aerosols as a function of different aging

conditions. The analysis of the optical and physical–chemical measurements supports the idea that differences in composition, shape and size translate in variations of the spectral optical properties of the aerosols, as observed in a repeatable manner for our different experiments. An illustrative example is provided in Figure 2 where the evolution of the mass absorption coefficient (MAC, absorption per unit of aerosol mass) and Single Scattering Albedo (SSA, ratio of scattering over extinction) at 630 nm for soot aged in presence of ozone and alpha–pinene is shown. The internal mixing of soot with SOA generated from the ozonolysis of alpha–pinene induces a measurable and significant increase of both the MAC (from 7 to 11 m^2 g^{-1}) and the SSA (from 0.3 to 0.48), showing that the capacity of particles to absorb and scatter visible radiation increases considerably through internal mixing with organic material. The complete results from the BACON experiments are still under analysis. These results are expected to be published soon

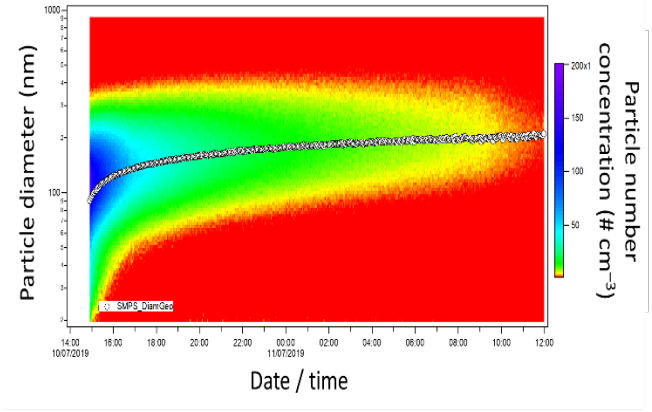

Figure 1: Time series of the particle number concentration size spectrum in the CESAM simulation chamber for minicast soot aged for 22 hours under dry dark conditions. The evolution of the median geometrical diameter of the size distribution is also shown (dots in the figure).

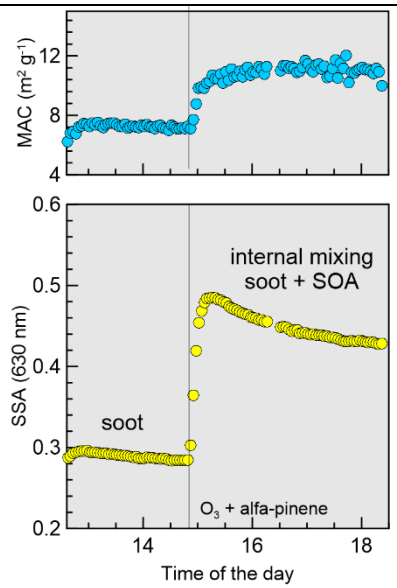

Figure 2: Evolution of the Mass Absorption Coefficient (MAC) and Single Scattering Albedo (SSA) at 630 nm of soot aerosols during an aging experiment involving the internal mixing with Secondary Organic Aerosols (SOA) generated from the ozonolysis of alpha-pinene.

Future of the project: the BACON activity helped build the bases for the development of a larger project aimed at studying the spectral optical properties of carbonaceous aerosols based on CESAM simulation chamber experiments: the B2C project (Constraining the direct radiative effect of Black and Brown Carbon on climate: an innovative experimental study on their spectral optical properties) funded by ANR (2020–2024). Indeed, the BACON project has allowed the PI to develop her scientific network, by establishing solid and fruitful collaborations with experts in BC aerosol characterization of physical and chemical properties, and advanced optical modelling approaches. The scientific activity around the study of spectral BC optical properties in simulation chambers actively contributes to foster scientific exchanges within the GDR SUIE (https://www.gdr-suie.cnrs.fr/) research group.

Number of publications, communications and theses:

- ✓ 3 articles under preparation
- ✓ 1 postdoc (M. Zanatta, 7 months Nov 2020 – May 2021)
- ✓ 1 PhD Thesis (J. Heuser) started in January 2021 in the framework of the BACON continuation (ANR B2C)
- ✓ 7 oral communications in national and international workshops and conferences (Journée métrologie des suies, GDR Suies annual meeting, Journées thematiques du GDR Suie, PM2022, 1st Actris Science conference, EGU General Assembly)
- ✓ 4 poster communications in national and international workshops and conferences (LEFE Colloque de Chimie Atmospherique, European Aerosol Conference, Workshop ACTRIS-FR, 1st Actris Science conference)

Data availability: relevant data (both experimental temporal series and retrieved spectral optical properties) obtained from simulation chamber experiments will be distributed freely and permanently in the EUROCHAMP-2020 database hosted by the AERIS data and service centre (https://data.eurochamp.org/).