Programme LEFE-CHAT	Project Title: Hygroscopic properties of Black Carbon (h-BC)	Years 2019 – 2021
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Context: Black carbon-containing particles (BC), the most strongly light-absorbing components of aerosol, are significant short-lived climate forcer. The BC hygroscopicity, varying with atmospheric aging processes, largely determines (1) their optical absorption through lensing effects and consequently their impacts on direct radiative forcing and snow-covered area (2) their CCN activity, affecting their impacts on formation and properties of clouds and thus their indirect radiative forcing, and (3) their dry and wet deposition rates and thus lifetime in the atmosphere. A number of experimental techniques have been developed to investigate hygroscopic properties of atmospherically relevant particles. Since typical BC mass fractions in submicrometer particles are only a few percentages, in general these techniques cannot provide specific information on BC hygroscopicity.

Objectives / scientific questions: The main objective of the h-BC project is to develop an instrument to measure BC hygroscopicity that could be deployed on both ground-based and airborne platforms. The second goal is to address the following questions: what are the hygroscopicity and the optical properties of BC transported at a high-elevation mountain site, where numerous feedback mechanisms between BC, radiation, clouds and snow have important climate implications? How do they vary with emission sources, transport pathways, meteorological conditions and chemical environment? How can models represent the modification in the optical and hygroscopic properties and the lifetime of BC due to its internal mixing with other components?

Main results:

We have developed a HTDMA-SP2 system that measures how BC grows or shrinks from different initial dry sizes when exposed to changing relative humidity (RH) conditions. The system is shown in Figure 1. The aerosol flow is dried to < 20 % RH and then passed through a first differential mobility analyzers (DMA) to produce quasi-monodisperse aerosols with a specific size; after that, the aerosol flow is humidified to a specific RH and then passed through a second DMA. The aerosol flow exiting the second DMA is then split into two flows, sampled by a CPC and a SP2, respectively. The usage of a Single Particle Soot Photometer (SP2) enables identification of BC aerosol particles, and mobility diameter changes in aerosol particles identified to be BC. It allows to measure the BC hygroscopic growth (GF_{BC}) at RH ranging from 30 to 95 %. A specific software was also developed to invert the measured GF_{BC} distributions into a probability density function (GF_{BC} -PDF).

Performance evaluation was performed by deploying the HTDMA-SP2 at the station of Meteopole-Flux at Toulouse. The quality of GF_{BC} measurements by the HTDMA-SP2 was found to be essentially dependent on the DMA sizing accuracy and the RH control. Figure 2 shows an example of the GF_{BC} and GF_{BC} –PDF for a selected dry size of 130 nm.

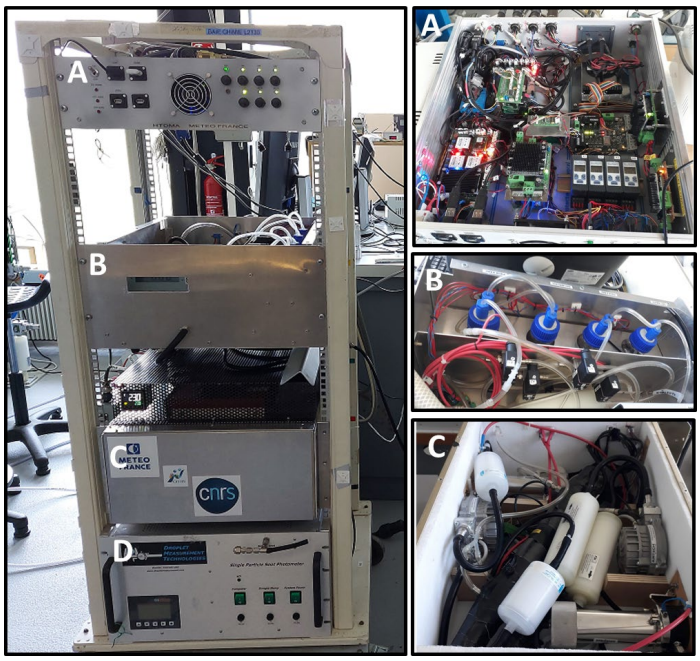
Three-year measurements were conducted at the high-altitude mountain station Pic du Midi de Bigorre in the French Pyrenees (PDM, 42.9°N, 0.14°E, 2850 m a.s.l.). A large set of instrumentation was deployed to characterize the microphysical, chemical and optical properties of BC-containing particles. The expected schedule for the deployment of the HTDMA-SP2 was delayed due to technical issues. This was related to the limited availability of the technical team which was involved in a massive high-priority field campaign not planned at the time of h-BC initiation and to COVID situation which restricted the access at the PDM. In addition, the deployment of the HTDMA-SP2 at the PDM has involved complex adjustment of the RH control of the instrument under these challenging sampling conditions (low pressure, cold temperature). As a result, the measurements were made operational for few days in October 2021.

The long-term data analysis of spectral aerosol optical properties indicates that BC was the predominant absorption component of aerosols at PDM throughout the three years. Microscopic analysis of aerosol particles collected on grids showed the presence of a variety of BC morphology and mixing state, with a

dominant number of attached-type BC particles often mixed with sulfate and organics. Significant light-absorption enhancement of BC was observed due to this internal mixing. We discovered a large seasonal contrast in BC properties with higher BC mass concentration and absorption enhancement in summer than winter, largely driven by the complex influence of BC emission sources, transport pathways, atmospheric dynamics and photochemical processing.

In parallel, we compared three state-of-the-art methods for measuring BC concentrations deployed continuously at the PDM over the three years: a recently upgrated aethalometer AE33, a thermal-optical analyser Sunset and a single-particle soot photometer SP2. Significant deviations in the response of the instruments were observed, which were explained by incorrect correction factors, interferences from dust particles transported at PDM and the different particle size range covered by the instruments.

The evaluation of the representation of BC properties in the regional climate model ALADIN is ongoing using the three-year dataset acquired in the project. First estimates show that the average BC mass concentration in the simulations was larger by a factor ~3 than the observed concentrations. BC absorption was greatly underestimated in the model by a factor ~2. The reasons for these biases are still under investigation and are likely due to emission inventories, size-dependent wet removal process and/or model treatment of BC optical properties.



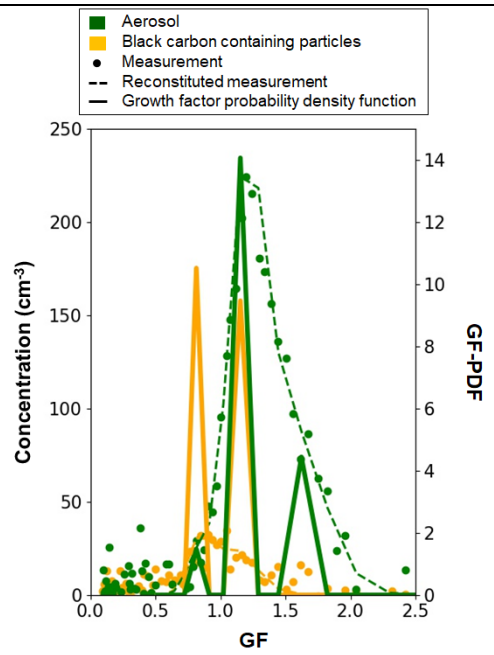


Figure 1. Picture of the HTDMA-SP2 system.

Figure 2. Example of GF and GF-PDF of BC and aerosol at 90%RH for a selected dry size of 130 nm measured at the Meteopole-Flux site.

Future of the project: The project now focuses on the exploitation of the results acquired in the project for modelling purposes. This includes model-observations intercomparisons with the regional climate model ALADIN and the chemical-transport model MOCAGE.

The HTDMA-SP2 system flew on the ATR-42 research aircraft during the 2022 ACROSS campaign, providing data on BC hygroscopicity in Paris urban plumes. Various projects including the deployment of the HTDMA-SP2 for characterizing BC hygroscopicity in biomass burning plumes have been submitted for a start in September 2023, if selected.

Publications, communications and theses

Publications:

S. Tinorua *et al.*, Two-year measurements of black carbon at the high-altitude mountain site of Pic du Midi Observatory in the French Pyrenees, *submitted to ACP*.

S. Tinorua *et al.*, Intercomparison of three methods for measuring black carbon concentration at a high-altitude research station in Europe, *in preparation for AMT*.

Communications: IAC, GDR Suie, AEI, media visit for television and press releases in newspapers.

Students: 1 PhD thesis (Sarah Tinorua, 2019-2021) + 4 student internships.