Employment of novel tools for the continuous characterization of the carbonaceous fraction in ambient aerosol

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Part of this programme and projects: GAW-CH Science

Keywords: atmospheric aerosol particles; aerosol climatic effects; aerosol source apportionment; carbonaceous aerosol; novel aerosol instrumentation

1. Project description

Total aerosol carbonaceous mass (TC) is a major constituent of the atmospheric fine aerosol particle mass. However, this fraction is not yet continuously monitored at GAW sites with an adequate time resolution. Adding a TC measurement is thus crucial to complement the existing measurement programs for a comprehensive interpretation of the impact of aerosols on our climate. Currently, the visible light absorbing, more elemental part of carbonaceous aerosols is permanently monitored by means of various filter based Aethalometer type instruments. However, no information on the organic carbon and especially the non-visible-light absorbing carbon is available. Within Switzerland, offline measurements of OC and EC via Sunset devices are rare, routinely performed for a few NABEL stations at low time resolution (*i.e.* every 12th day). Daily data for OC and EC concentrations at the Jungfraujoch (JFJ) site are only available for a handful of measurement campaigns.

Commercial thermo-optical methods for characterization of carbonaceous aerosol are expensive and not ready for continuous standalone operation. To fill this gap, the “fast thermal carbon totalizator” (FATCAT) has been developed at FHWN, an in-situ carbonaceous aerosol measurement system for long-term monitoring of TC. FATCAT collects aerosol particles on a sinter metallic filter, which is subsequently heated to 800°C under an oxidizing atmosphere. Further oxidation of the evolved carbonaceous material is achieved by a catalyzer located downstream of the heating unit. TC detection is achieved by means of a NDIR CO_2 measurement. The fast-heating cycle of 50 seconds allows a low limit of detection (LoD) of 0.1 and 0.2 µg of carbon (µg-C) at the altitude of European lowlands and the JFJ observatory, respectively. At the reduced atmospheric pressure of the JFJ, which also limits the sampling flow of the device to 6 lpm, this is enough to measure TC concentrations with a LoD=0.3 µg-C/ m^3 using a time resolution of two hours.

FATCAT has been deployed at the Sphinx observatory aerosol inlet for unattended long-term measurements since 2020, generating a high time-resolution TC dataset. The TC measurement performed by FATCAT consists of analyzing thermograms of the evolving carbon capture from an aerosol sample. Aerosol particles containing more refractory, crystalline carbon from fossil fuel combustion, e.g. from diesel engine emissions, are less volatile and carbon tends to evolve at higher temperatures (see, e.g., Figure 1), whereas aerosol particles from biomass burning and biogenic aerosols containing more volatile organic carbon tend to decompose at lower temperatures. Therefore, source specific fingerprints for various combustion emissions are distinguishable and FATCAT allows different carbonaceous aerosol sources to be distinguished.

During 2022, FATCAT was also included in the carbon measurement campaign organized by PSI. This campaign included measurement instruments for characterizing the different fractions of carbonaceous aerosol, as well as measurement of gas-phase organic carbon. Other devices for measuring carbonaceous aerosol include eBC measurements via Aethalometer (deployed at JFJ by PSI), organic mass using ToF-ACSM (deployed at JFJ by PSI), and a further TC measurement via TCA08 (deployed by Aerosol d.o.o.). Figure 2 shows the TC time series for the first semester of 2022 as well as for the carbon campaign in August and September 2022. After the carbon campaign (October 2022), measurements were interrupted for the rest of the year to replace a component. The constant deployment at JFJ has shown that the current prototype can be deployed for approximately 26,000 hours of operation (equivalent to 3 years of continuous operation) with minimum maintenance.

Currently, the different measurement techniques that were used to measure the carbonaceous aerosols during the August campaign are being compared by colleagues at PSI. We are also working on the further development of the FATCAT. An exciting topic is the investigation of the information content hidden in the shape of the thermograms. It has been shown that this can be used to distinguish between different anthropogenic and biogenic aerosol sources. We also plan to submit a technical paper on this new method and its advantages.

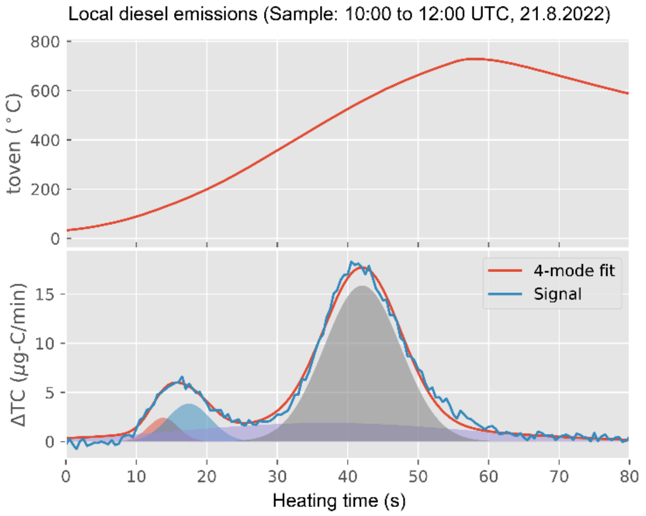

Figure 1. Total carbon thermogram for a datapoint containing a large amount of carbonaceous aerosol from local diesel emissions measured at the JFJ Sphinx laboratory. The top panel shows the filter temperature during analysis. The lower panel shows the differential total carbon signal (blue line) and a 4-mode fit (red line and filled curves). The red, blue, violet, and grey fitted components originate from materials with increasingly higher temperature stability. The red and blue component is identified with the higher volatility fraction of condensed organic compounds. The grey component shows the higher thermal stability fraction. The presence of diesel emissions was confirmed by the custodians of the Sphinx laboratory.

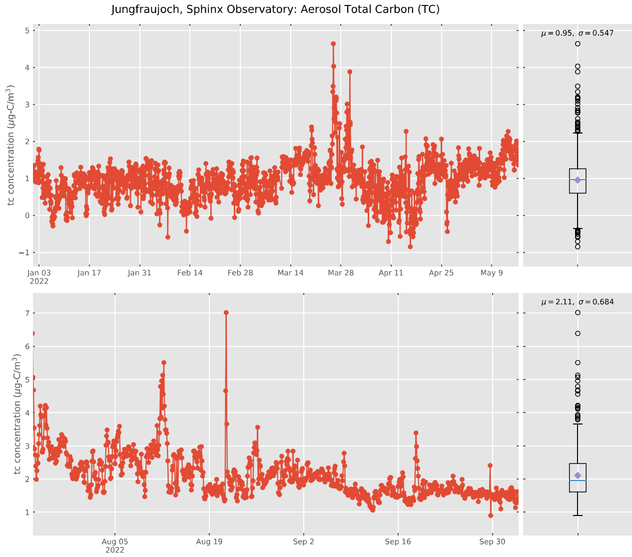

Figure 2. Uninterrupted time series and box plot for total carbon, measured at the JFJ observatory by means of FATCAT, from January through May 2022, and during the carbon campaign organized by PSI in August and September 2022. In the period between these two datasets FATCAT measurements were interrupted for instrument calibration at the FHNW. The large sharp increase of signal on August 21st corresponds to the diesel emissions event shown in Figure 1.

Collaborating partners/ networks

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Dr. C. Hüglin, Dr. S. Henne, Dr. M. Steinbacher, and Dr. S. Reimann, Empa, Dübendorf
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