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Characterizing black carbon and brown carbon aerosols by their optical and microphysical properties in an urban Mediterranean area.

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Sub-micrometer carbonaceous aerosols in the atmosphere are of serious concern for air pollution and climate change. They consist of particles with diameters from a few to several hundred nanometers made of organic carbon (OC), black carbon (BC), plus additional trace elements. Combustion sources contribute to primary carbonaceous aerosols (BC- and OC-rich) in a overwhelming way. Combustion also contributes to secondary particles, i.e. particles deriving from chemical transformation of primary particles, or from gas-to-particle transformation of gaseous emissions (mainly OC-rich). Combustion carbonaceous aerosols show huge number concentrations in the ultrafine particle range (UFPs, with diameters < 100 nm); they also show relatively large light absorption, whose spectral behaviour depends on the relative BC-to-OC abundance. The combination of these parameters is therefore important to identify and characterize such particles.

In this work, we discuss an original approach to characterize BC and OC aerosols based on their spectral optical and microphysical properties. Total absorption (σ_a) is decomposed in the sum of three terms: an absorption due to dust (σ_{aD}), a spectrally constant absorption (σ_{aBC}) due to BC, and a spectrally variable absorption (σ_{aOC}) due to "brown carbon". σ_{aD} is evaluated through a proper combination of scattering angstrom exponent, single scattering albedo, and particle number concentration. σ_{aOC} is obtained as the fraction of the dust-free absorption ($\sigma_a - \sigma_{aD}$) increasing with increasing Absorption Angstrom exponent. σ_{aBC} is finally calculated as $\sigma_a - \sigma_{aD} - \sigma_{aOC}$, and used to obtain an equivalent BC mass concentration (BC_{eq}). Through Mie theory simulations, particle diameters of aerosol populations with large absorbing OC equivalent contents (OC_{eq}) or BC_{eq} contents are calculated. This approach is applied to a dataset of spectral light absorption (3 wavelength PSAP) and scattering (3 wavelength nephelometer) measurements in the visible region, and (butanol-based CPC) measurements of total particle number concentrations (N). Measurements were carried out in 2010-2011 in the Central Mediterranean area of Rome (Italy) at three sites representative of conditions ranging from rural background to urban pollution to aircraft/road traffic emissions.

Findings show σ_{aOC} to be surprisingly similar to σ_{aBC} regardless of measurement site and season. For both σ_{aBC} and σ_{aOC}, accumulation mode particles with 100-300 nm diameters show significant contributions, in particular at the urban background site. The role of UFPs is as well not negligible, in particular at the aircraft traffic site, where N is of the order of $10^6 cm^{-3}$: here, σ_{aBC} apportioned to soot mode particles, and σ_{aOC} apportioned to Aitken mode particles reach, respectively, up to 20 and 10 Mm^{-1}. Future evaluations of the OC_{eq} and BC_{eq} mass absorption efficiency varying with varying particle size down to UFPs become necessary to unravel the true role of brown carbon aerosols, this role being here indicated as potentially comparable to the one of BC_{eq}.