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Partitioning of Black Carbon between ultrafine and fine particle modes in an urban airport vs. urban background environment

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HIGHLIGHTS

- Characterization of the Black Carbon (BC) aerosol in an urban airport vs. urban background environment.
- A scheme to assess the ultrafine BC in the bulk aerosol.
- Separation of BC sources from fossil fuels and biomass burning.

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ABSTRACT

In this work, we characterize the Black Carbon (BC) aerosol in an urban airport vs. urban background environment with the objective to evaluate when and how the ultrafine BC dominates the bulk aerosol. Aerosol optical and microphysical properties were measured in a Mediterranean urban area (Rome) at sites impacted by BC sources including fossil fuels (FF), and biomass burning (BB). Experimental BC data were interpreted through measurement-constrained simulations of BC microphysics and optical properties. A "scheme" to separate the ultrafine BC was experimented on the basis of the relation found between changes in the BC partitioning between Aitken and accumulation mode particles, and relevant changes in particle size distribution and optical properties of the bulk aerosol. This separation scheme, applied to experimental data, proved useful to reveal the impact of airport and road traffic emissions. Findings may have important atmospheric implications. The experimented scheme can help separating different BC sources (FF, BB, "aged" BC) when BC size distributions may be very difficult to obtain (satellite, columnar observations, routine monitoring). Indeed, separating the ultrafine BC from the fine BC may provide significant benefits in addressing BC impact on air quality and climate.

1. Introduction

"Black Carbon" (BC) is the largest component of the atmospheric light-absorbing aerosol, whose effects on human health (WHO, 2012) and climate (Ramanathan and Carmichael, 2008) are still debated. The BC is ambient aerosol material having a unique combination of properties, recently defined by Bond et al. (2013) as a strong visible light absorber with a wavelength independent refractive index, refractory and insoluble in water, with a spherule-like aggregate morphology formed in flames. Flame features characterize BC properties. There is, in particular, a distinct difference in the BC particle number size distribution (PNSD). Recent SP2 measurements by Kondo et al. (2011) showed count median diameters of aerosol in urban areas and in plumes associated with wildfires to be 50–80 nm and 120 nm, respectively. Schwarz et al. (2008) showed that the PNSD of fossil fuels (FF) peaks in the Aitken mode size region (ultrafine BC particles, $D_p < 100$ nm); conversely, that of biomass burning (BB) in the accumulation mode size region (fine BC particles, 100 nm $< D_p <$ 1 μm). Measurements by Petzold et al. (2005) indicated mean number diameters of BC particles produced by aircraft jet engines of about 30 nm.

To address the question concerning what is the contribution of BC to climate forcing and how BC affects human health, it is still necessary to evaluate how BC aerosol properties affect that contribution (Bond et al., 2013; Ramanathan and Carmichael, 2008; Smith et al., 2009). As yet, there is only limited monitoring data of BC for epidemiologic analysis (Smith et al., 2009), or climate modeling validation studies (Aquila et al., 2011; Reddington et al.,

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2013). There is a lack of experimental measurements of the BC sizes due to the difficulty to measure the BC PNSD, especially the ultrafine BC fraction. Large uncertainties are associated with the BC sizes at the emission source, as well as in the ambient atmosphere, and much less is known about the BC particle number concentration. The toxicology of the pure BC and its epidemiology diverge because atmospheric BC has long been considered either as undifferentiated particulate matter (PM) mass (a fine PM component), or as elemental carbon PM, with scarce consideration for relevant BC properties (e.g., differences between ultrafine and fine BC), and interactions with associated toxic species (WHO, 2012; Cassee et al., 2013; Smith et al., 2009). This has resulted in no clear evidence for the relative BC impacts (Bond et al., 2013; WHO, 2012).

Doubts in the BC microphysical properties (the BC PNSD) reduce the understanding of the role of BC on climate (Pierce et al., 2007; Jacobson, 2010; Reddington et al., 2013). It is well-known that the Single Scattering Albedo (SSA) – a key parameter to understand the warming or cooling effect of atmospheric aerosol (Haywood and Shine, 1995) – for one BC particle decreases with decreasing particle diameter and increasing imaginary part of refractive index (e.g., Moffet and Prather, 2009). The absorption coefficient (σ_a) has a similar tendency. When the bulk BC aerosol is considered, the relation between the BC PNSD and SSA is however not obvious, especially for mixed aerosol. Factors influencing the SSA vs PNSD relation have been previously investigated (Khatri et al., 2014; Kahnert and Devasthale, 2011; Garland et al., 2008). Garland et al. (2008) found a SSA_{532} decreasing tendency with decreasing (number mode) diameter; the diameter being the same, different SSA values were found (but not discussed). Khatri et al. (2014) showed that SSA can get changed depending on the size distribution of mixed components, the aerosol size distribution being capable to bring a considerable change in SSA even without any change in chemical composition. They also observed that the aerosol size distribution effect on SSA could be one of the important reasons for observed low values of SSA near source regions. Kahnert and Devasthale (2011) analyzed SSA for different particle sizes and morphologies, such as lacy aggregates and compact aggregates, and concluded that there is no dependency of SSA on particle shape for particle diameters <100 nm (Aitken mode). SSA was also used to characterize the composition of BC aerosols (Jacobson, 2010; Cappa et al., 2009; Garland et al., 2008; Clarke et al., 2007). Cappa et al. (2009)'s results indicate SSA increasing with increasing σ_a when BB aerosol (wood smoke) was large compared to FF aerosol; conversely, SSA was found to decrease with σ_a (down to 0.5) for FF aerosol (ship emissions from the Boston harbor). Garland et al. (2008) found larger values of SSA during the "intense smoky period" (BB aerosol), low SSA indicating strong emissions of light absorbing carbon. Clarke et al. (2007) analyzed the relation linking the dry SSA to the σ_a(530 nm) for BB and pollution aerosol, and suggested that larger SSA values correspond to larger particles for a given σ_a, and that when SSA data are grouped by diameter, the strongest dependency occur for the smallest particle sizes. In a comprehensive study, Jacobson (2010) concluded that while BB aerosol globally cool the climate, FF aerosol contributes significantly to global warming. Albeit all these studies have investigated the issue, it appears as still there is the need to understand how BC effects on health and climate can get changed by changes of the BC size distribution.

In this work, we characterize the Black Carbon (BC) aerosol in an airport/urban vs. urban background environment aiming at experimenting a "scheme" to separate ultrafine BC particles from other BC aerosols. The argument is analyzed by interpreting experimental measurements carried out in a Mediterranean urban area (Rome) at sites representative of airport and road traffic emissions, and urban background, through measurements-constrained Mie simulation of BC microphysical and optical properties in the visible region.

2. Experimental

2.1. Measurement sites

Measurements were carried out in Winter 2011 (Dec 2011–Feb 2012) and Spring 2012 (May–June) at two sites located in the southern urban outskirt of Rome (Italy) in the Mediterranean coastal area (Fig. 1). The ARPT site is located in very close proximity (less than 1 km, North) to a medium-size city airport (*Ciampino*). In between the site and the airport there is an urban road; to the west, a larger traffic road; to the East, an urban agglomeration. Because of the breeze regime, the site is downwind to planes that took off from 9 p.m. to 10 a.m. (UTC), aircrafts flying from 4 a.m. to 9 p.m. The UBG site is in the suburban background, within 5 km from the Rome city boundary and 40 km from the Mediterranean coast (inside the ISAC-CNR Rome Atmospheric Supersite); it is surrounded by residential areas where wood is abundantly burned in winter for heating.

At both sites, instruments were set-up on the same inlet system. External air was pumped in the cabin into a stainless steel tube (length = 4.0 m) by an external pump ensuring a Reynolds number <2000 (laminar flow). The cabin was conditioned at 20–25 C to dry sample air based on the difference between air temperature and dew point.

2.2. Measurement of optical properties

Scattering measurements were performed by an integrating nephelometer (Ecotech, mod.Aurora 3000) operated to measure the dry aerosol scattering coefficient (σ_s) at three wavelengths, 450, 520, 635 nm. A 3-wavelength particle soot absorption photometer (PSAP, Radiance Research) was operated to measure the dry aerosol absorption coefficient, σ_a, at three wavelengths, 467, 530, 660 nm. Raw PSAP data were corrected after the iterative procedure described by Bond et al. (1999), Virkkula et al. (2005) and Virkkula (2010). The procedure requires wavelength resolved σ_s, which were extrapolated from dry scattering data by means of the measured Scattering Angstrom Exponent (SAE). All measurements with $\sigma_a < 1$ Mm^{-1}, $\sigma_s < 10$ Mm^{-1} were eliminated to reduce experimental errors. Additional details on the procedure used to correct PSAP and nephelometer data were given by Costabile et al. (2013).

2.3. Measurement of particle number concentration

A butanol-based Condensation Particle Counter (CPC, TSI mod.3022A) was used to measure the particle number concentration, N. The lowest cut off particle diameter (D_p) of the 3022A CPC is D_{p50} (50% of particle detection) at 7 nm, and D_{p90} at 15 nm. The N_7 (total number concentration of particles larger than 7 nm) can be measured up to 10^6 cm^{-3}. All measurements with $N_7 > 3 \times 10^5$ cm^{-3} were eliminated to reduce experimental errors.

Co-located measurements of particle number size distribution were carried out by a Scanning Mobility Particle Sizer. Mobility Particle Size Spectrometers are based on the electrical mobility diameter depending on particle shape. The influence of different diameters resulting from the different sizing techniques was investigated in a number of previous studies (e.g., Hinds, 1999; De Carlo et al., 2004), and relative measurement standards were harmonized (Wiedensohler et al., 2012).