Seasonal variations of black carbon physical properties influenced by different sources in London urban environment

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A recent comprehensive assessment of the contribution by black carbon aerosol (BC) to total climate determined it to be +1.1 W m^{-2}, and is deemed to be the second most important human emission in terms of its climate-forcing in the present-day atmosphere [Bond et al., 2013]. One of the most important and fundamental constraints on modelling BC climate forcing is determining its source strength. BC can be emitted from different combustion related sectors, i.e. they are often classified as fossil fuels for transportation; biofuels for industrial and residential uses including solid unprocessed biofuel, and open burning of biomass. The motivation to treat the source-attributed BC separately is due to its complex physical properties being largely source dependent, such as the BC size, mixing state, optical and hygroscopic properties. The variety of BC physical properties from sources can lead to considerable variations and uncertainties when modelling the BC direct forcing effect on climate and also the indirect forcing due to their different behaviours when interacting with clouds.

In urban environments, the BC from diesel fuel such as vehicle emission has been widely reported and been incorporated in most of the regional models. Although a few studies have emphasized the importance of emissions from residential solid fuel burning (i.e. from wood burning and cooking) in UK winter time [Allan et al., 2010; Liu et al., 2011], the properties of BC associated with these sources and their differences compared to that from vehicle emissions have not been well characterized.

This study reports results of continuous BC measurements from the Clean Air for London (ClearfLo) campaign for two major intensive observation periods (IOPs) during 2012. Both IOPs lasted for four weeks and were conducted in winter (11st Jan.- 08th Feb.) and 2012 summer (18th Jul.-22nd Aug.) respectively, the latter being during the period of the London Olympic Games. The experimental site was located in the grounds of a school in North Kensington, which is representative of a typical London urban background environment. A long term source attribution study in London based on the positive matrix factorization (PMF) analysis of AMS data shows that London urban environment in winter is significantly influenced by the source of solid fuel burning, such as wood burning for residential heating. In contrast vehicle emissions almost form the only BC source in summer. In the present study physical properties of BC were characterized using a single particle soot photometer. By assuming a core-shell model of coated BC, the relative coating thickness of the aprticles, which is the entire BC particle size (D_p) divided by refractory BC core size (D_c) - D_p/D_c, was also determined using Mie theory.

The physical properties of BC exhibited seasonal differences, summarized in Figure 1. The winter experimental period in London was generally more polluted than summer, with almost twice the BC mass loading on average. Both summer and winter showed a morning rush hour peak of BC mass, whereas an additional evening peak showed in winter. The BC particles in winter, influenced by solid fuel burning, showed larger core size (geometric mean mass diameter D_{geo}170-205nm) than in summer (D_{geo}155-180nm). The summer BC, dominated by vehicle emissions, showed consistent core size distribution throughout the experiment without any diurnal variation, however an apparent diurnal trend of BC core size was observed when influenced by solid fuel burning. The winter-time BC showed significantly thicker coatings than in summer (as reflected by D_p/D_c). The thickest BC coatings in summer were observed between 14:00-15:00 corresponding with the maximum photochemical reactions; however the strongest mixing occurred at midnight in winter. The work we will also present a methodology using Mie calculations to estimate the mass absorption cross section (MAC) and the absorption enhancement due to the coating for different sources of BC and relate these to different photochemical ages.

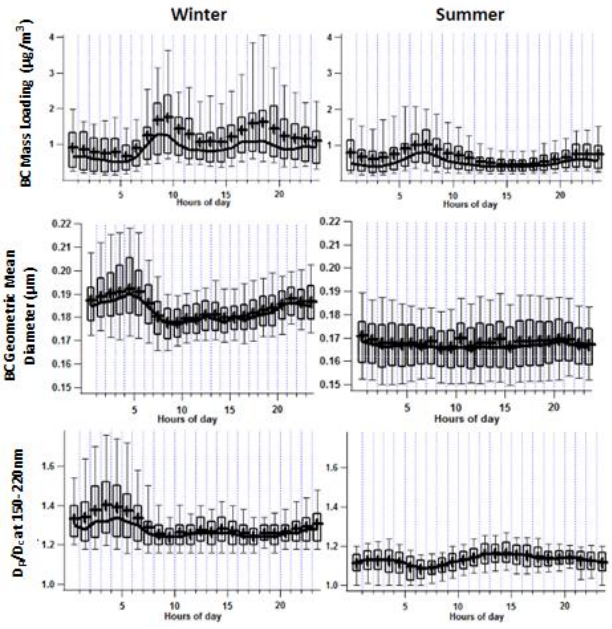

Figure 1. The diurnal trends of BC physical properties in winter and summer, from top panel to the bottom: BC mass loading, BC geometric mean diameter and D_p/D_c at 150-220nm

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