

Chemical and Optical Properties of Black Carbon Particles (CHEMBC)

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Scientific Background: Black Carbon (BC) is a term used to describe primary carbonaceous particles formed through incomplete combustion processes, which include vehicular traffic, domestic solid fuel burning and biomass burning. BC influences climate directly through positive radiative forcing by absorbing solar radiation and also indirectly by altering the microphysical properties, amount and lifetime of clouds. Compared to greenhouse gases, the atmospheric lifetime of black carbon is relatively low, and thus BC emission controls represent the most likely means by which to mitigate climate change quickly and effectively. However, improved scientific knowledge of the atmospheric processing of BC is required to provide policy makers with the relevant information to implement effective legislation.

Project Objectives: The overall scientific objective of the CHEMBC project was to couple state-of-the-art measurement techniques of the chemical and physical properties of ambient BC with advanced data analysis strategies to greatly improve our understanding of the sources and impact of BC on air quality and climate. The research training objectives focussed on equipping the Fellow with advanced knowledge and skills in multivariate receptor modelling, geographic origin calculations, instrument design and leadership skills.

Work Performed: In the first 2 years of this project (outgoing phase in Toronto), the Fellow developed and applied new data mining techniques to assess the chemical mixing state of BC particles in a single particle mass spectrometer dataset collected in Paris, France. Single particle mass spectrometry is a powerful technique that enables one to investigate if BC exists in 'pure' atmospheric particles as the sole component, or whether other primary and secondary species, e.g., organic aerosol and inorganic ions, are also internally mixed. As part of the CHEMBC project new data analysis techniques have been developed to answer these questions.

An intensive field campaign was also performed in downtown Toronto as part of the CHEMBC project in June 2013. Using a suite of instruments, the Fellow investigated the impact of BC chemical mixing state upon its radiative absorption capacity. Data analysis for this campaign is complete and the results will provide information relevant for modellers aiming to constrain the effect of BC coatings in global climate models.

In the final year of the project (return phase in Cork), a new global library of single particle mass spectrometry datasets was compiled for the first time. Datasets collected between 2002 and 2014 by research groups in Canada, USA, UK, Ireland and Switzerland were merged to produce a single database. The database includes single particle mass spectra collected across North America, Western Europe, East Asia, the Mediterranean, the North Atlantic and the Caribbean. Thus the data represents a cross-section of ground level single particle composition globally. This dataset has been clustered to provide representative particle types that will be useful as an input for other single particle mass spectrometer users to more objectively analyse data collected from future measurements. This development is expected to lead to a more standardised approach to particle mixing state assessment worldwide.

Main Results: The new data mining techniques developed and applied during the outgoing phase have led to a better understanding of BC mixing state in ambient environments. The single particle quantitative approach enabled an advanced apportionment of the sources of BC contributing to poor air quality events in Paris, France (Healy et al., 2013). This enabled a separation of fresh, locally emitted BC and aged BC transported from outside the city.

The Fellow has applied information-theoretic entropy and diversity measures to single particle composition for the first time (Healy et al., 2014b). This new approach enables us to assess quantitatively how ‘mixed’ an ambient aerosol population is using single particle mass spectrometer data. The capacity for BC particles to accumulate water at a given relative humidity (hygroscopic growth) can be used to assess their propensity to form new clouds. Traditionally, it has only been possible to predict hygroscopic growth for bulk aerosol. However, as part of CHEMBC, a new data mining approach has been developed to predict this microphysical property for every single particle measured. This method is uniquely based on single particle composition and produced hygroscopicity estimates that agreed well with measured values (Healy et al., 2014a), thus confirming its potential for wider application in assessing the cloud forming potential of atmospheric particles.

The field campaign in Toronto enabled detailed investigation of the light-absorbing properties of BC particles originating from local vehicular traffic, wildfires in Quebec, and transboundary fossil fuel combustion emissions in the United States. In contrast to previous laboratory studies, enhanced light absorption caused by various coatings on the BC particles was not observed, but significant light absorption was measured in Brown Carbon particles emitted from the wildfires (Healy et al., 2015).

Expected Final Results and Impact: As demonstrated for Paris, quantitative separation of local and transported BC emissions is particularly useful for policy-makers attempting to design and implement effective air quality strategies. From a climate perspective, understanding how BC is mixed with other aerosol species is valuable information as an input for advanced single particle-resolved climate models. The novel diversity metrics developed through CHEMBC will allow climate modellers to investigate the extent to which error in climate models arises from assumptions about BC mixing state, for example. A more direct assessment of BC hygroscopic properties has also been derived from single particle mass spectrometer data for the first time. The field campaign showed that inclusion of BC absorption enhancement in climate models may lead to an overestimation of positive radiative forcing impacts. Finally, the Fellow has fostered multiple new international collaborations and gained leadership skills which significantly enhance his future career prospects as an independent researcher.

References:

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