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Radiocarbon-insights into temporal variations in the sources and concentrations of carbonaceous aerosols in the Los Angeles and Salt Lake City Metropolitan Areas

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Increased fossil fuel consumption and biomass burning are contributing to significantly larger emissions of black carbon (BC) aerosols to the atmosphere. Together with organic carbon (OC), BC is a major constituent of fine particulate matter in urban air, contributes to haze and has been linked to a broad array of adverse health effects. Black carbon's high light absorption capacity and role in key (in-)direct climate feedbacks also lead to a range of impacts in the Earth system (e.g. warming, accelerated snow melt, changes in cloud formation). Recent work suggests that regulating BC emissions can play an important role in improving regional air quality and reducing future climate warming. However, BC's atmospheric transport pathways, lifetime and magnitudes of emissions by sector and region, particularly emissions from large urban centers, remain poorly constrained by measurements.
Contributions of fossil and modern sources to the carbonaceous aerosol pool (corresponding mainly to traffic/industrial and biomass-burning/biogenic sources, respectively) can be quantified unambiguously by measuring the aerosol radiocarbon (14C) content. However, accurate 14C-based source apportionment requires the physical isolation of BC and OC, and minimal sample contamination with extraneous carbon or from OC charring. Compound class-specific 14C analysis of BC remains challenging due to very small sample sizes (5-15 ug C). Therefore, most studies to date have only analyzed the 14C content of the total organic carbonaceous aerosol fraction.

Here, we present time-series 14C data of BC and OC from the Los Angeles (LA) metropolitan area in California - one of two megacities in the United States - and from Salt Lake City (SLC), UT. In the LA area, we analyzed 48h-PM10 samples near the LA port throughout 2007 and 2008 (with the exception of summer). We also collected monthly-PM2.5 samples at the University of California - Irvine, with shorter sampling periods during regional wildfire activity and Santa Ana winds from March to August 2013. In SLC, we seasonally collected 48h-PM2.5 samples from October 2012 to February 2014.

We isolated and quantified BC and OC using a thermo-optical analyzer (RT 3080, Sunset Laboratory, Tigard, OR, USA) with the Swiss_4S protocol, and measured the 14C content of BC and OC with accelerator mass spectrometry at UCI's KCCAMS facility. We also measured the concentration and stable isotope composition of total (organic) carbon and nitrogen on the aerosol filters with EA-IRMS (Carlo Erba coupled to Finnigan DeltaPlus).

Preliminary results suggest that in LA, PM10-BC concentrations are on the order of 2-8 ug C/m3. Black carbon is 14C-depleted (FM 0.04-0.21) – indicating that fossil sources dominate emissions. In comparison, OC concentrations were higher (12-17 ugC/m3) and more enriched in 14C (FM 0.54-0.83). In SLC, PM2.5-BC concentrations range from <1 to 3 ug C/m3, with the highest concentrations observed during wintertime inversions. The BC fraction is strongly 14C -depleted (FM 0.06 to 0.12) - indicating a dominance of fossil BC emissions throughout the year. Together, our measurements contribute to a comprehensive quantification of temporal and spatial variations in urban BC, a key uncertainty in constraining BC sources and transport in western North America.