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Radiocarbon source apportionment of carbonaceous aerosol components of South Asian Atmospheric Brown Clouds

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Light-absorbing carbonaceous matter constitutes one of the largest uncertainties in climate modeling. The high concentrations of black carbon – soot - in South Asian Atmospheric Brown Clouds lead to strong atmospheric heating and large surface cooling that is as important to regional climate forcing as greenhouse gases, yet the sources of these aerosols are not well understood.

Emission inventory models suggest that biofuel/biomass burning accounts for 60-90% of the sources of these aerosol components whereas measurements of the elemental composition of ambient aerosols compared with source signatures point to combustion of fossil fuel as the primary culprit. However, both approaches acknowledge large uncertainties in source apportionment of the elusively-defined black carbon. This study approached the sourcing challenge by applying microscale radiocarbon measurements to aerosol particles collected during the winter in North India, Central India and over the Indian Ocean receptor (The Maldives). The radiocarbon approach is ideally suited to this task as fossil sources are void of 14C whereas biomass combustion products hold a contemporary 14C signal.

In a first pilot study in 2006, high-volume air samples of total carbonaceous aerosols revealed 14C signals that were similar for a Central Indian source region site located in Sinighad, near Pune, and an Indian Ocean receptor (the Maldives), consistent with the absence of any significant formation of secondary organic aerosols, with a 60-70% contribution from biomass combustion and biogenic sources. Isolates of elemental or soot carbon fractions varied between 40-70%, depending on isolation method. A subsequent 15-month continuous probing of these two sites in 2008-2009 confirmed an average contribution of two-thirds from contemporary sources to the TOC. The 14C-data revealed a stronger contemporary signal arriving to the Maldives in May-June, presumably due to biogenic secondary organic aerosols. A period of stronger fossil contributions was also found near Pune in mid to late summer.

The first 14C data for northern India revealed for two sites (Hisar and Manora Peak) that the total carbonaceous aerosols were only 30% from fossil-fuel origin.

These novel radiocarbon constraints on the sources of light-absorbing carbonaceous matter aid prioritizing what combustion processes to target for emission mitigations of these health-afflicting and climate-forcing aerosols in the South Asian region.