Geophysical Research Abstracts,
Vol. 10, EGU2008-A-03552, 2008
SRef-ID: 1607-7962/gra/EGU2008-A-03552
EGU General Assembly 2008

Radiocarbon (14C) source apportionment of carbonaceous aerosol components in the Asian Atmospheric Brown Cloud

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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 the Asian Brown Cloud 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 monsoon both over the Indian Ocean and in central India. 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. High-volume air samples of total carbonaceous aerosols revealed 14C signals that were similar for N. Indian source and Indian Ocean receptor regions, 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. These novel radiocarbon constraints on the sources of light-absorbing carbonaceous matter aid prioritizing of what combustion processes to target for emission mitigations of these health-afflicting and climate-forcing aerosols in the South Asian region.