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Compound-specific radiocarbon analysis of atmospheric PAH from a wood smoke-impacted city in Northern Sweden

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Atmospheric polycyclic aromatic hydrocarbons (PAHs) are ubiquitous due to their emission from a variety of combustion sources including motor vehicle exhaust and other fossil fuel combustion and biomass burning. Their potential to detrimentally impact human and ecological health makes it vital to apportion these compounds to emission sources. PAHs also have a wide range in volatility and can be present in both the gas and particulate phase in the atmosphere. However, PAHs present in high concentrations in the atmosphere are not source-specific and can be difficult to trace back to their original emission sources.

Natural abundance radiocarbon analysis offers a trusted method for determining the fraction of modern versus fossil carbon and recent advances in micro-scale techniques have opened the door for compound class-specific radiocarbon analysis (CCSRA) of organic compounds at the low levels present in environmental samples. Atmospheric levels of PAHs provide a particular challenge due to difficulties in obtaining a sample with sufficient mass for radiocarbon analysis. Initial studies in Europe and Japan have shown a wide range of biomass burning contribution from 10% of pooled PAHs for two background sites in south Europe to up to 45% in Tokyo and 50% at a background site in Sweden. Compound-specific radiocarbon analysis (CSRA) of atmospheric PAHs would further enhance the understanding of emission source impacts on ambient concentrations and has not been previously reported.

In this study, CSRA was applied to a set of atmospheric samples collected in Lycksele, a city in Northern Sweden which has frequent episodes of severe atmospheric pollution in the winter. Previous studies carried out in the city have shown that wood combustion for residential heating has a large impact on atmospheric aerosol followed by motor vehicle emissions. This combination provided an ideal location for CSRA to apportion atmospheric PAHs between fossil fuel combustion and wood smoke. In order to maximize sample collection mass, a high volume total suspended particulate matter (TSP) sampler loaded with quartz fiber filters for PM collection and PUFs (polyurethane foam) for volatile species was used in this study. Five different particulate PAH samples were analyzed by CSRA including: phenanthrene, fluoranthene, pyrene, benzo[b+k]fluoranthene and indeno[cd]pyrene+benzo[ghi]perylene whereas phenanthrene was the only compound also analyzed in the gas phase. The percentage wood smoke contribution ranged from 70% for indeno[cd]pyrene+benzo[ghi]perylene to 87% for the PUF phenanthrene and particulate pyrene. There was a distinct molecular-weight trend (r^2 = 0.68) to the fraction wood smoke contribution with higher contribution for the lower molecular-weight PAHs.