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Isotope forensics unravel regional differences in sources of black carbon aerosol in South Asia

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Black carbon (BC) aerosols from incomplete combustion of biomass and fossil fuel exert a significant influence on the climate system in South Asia, effecting in excess of 1.2 billion people. Models—seeking to advise mitigation policy—are constantly challenged in reproducing observations of seasonally varying BC concentrations in the densely populated regions of South Asia e.g., over the Indo-Gangetic Plain (IGP). The systematic underestimation of the absorption aerosol optical depth over South Asia by a factor of 2-3 in climate models relative to observation-based estimates illustrates the currently large uncertainties. This uncertainty may be related to several different factors, including inaccurate estimates of relative source contributions and emissions of BC.

As part of the South Asian Pollution Experiment 2016 (SAPOEX-16), we investigated wintertime BC and its dual carbon-isotopic signature ($\delta^{13}\text{C}/\Delta^{14}\text{C}$) - diagnosed sources from regional and large-scale background receptor sites — the Bangladesh Climate Observatory at Bhola Island (BCOB), strategically located in the outflow region of the IGP to intercept the integrated IGP emission signal and the remote Indian ocean-based Maldives Climate Observatory at Hanimaadhoo Island (MCOH) to provide an integrated footprint of South Asia, respectively. Our results of "top-down" radiocarbon measurements of atmospheric BC from two regional-receptor sites show that fossil fuel combustion produces $55\pm3\%$ ($\Delta^{14}\text{C}$: -440 ± 6 per mil) of BC emitted from South Asia. In contrast, the $\delta^{13}\text{C}$ signals for BCOB-BC ($\sim -27.2\pm0.2\%$ ₃) are more depleted relative to that of MCOH-BC ($\sim -25\pm0.2\%$ ₃). Using a Bayesian statistical approach, we resolve the isotopic signatures into three source classes namely biomass, coal and liquid fossil fuel. We find the liquid fossil fuel component of BC emissions is $49\pm5\%$ in the IGP versus the larger South Asian footprint of $23\pm9\%$. Our observation is in stark contrast to previous "Bottom-up" emission inventory-based modelling estimates suggesting over $\sim 60\%$ BC from biomass burning in the IGP. This finding brings out the previously unknown sharp contrast in regional emission sources of BC from anthropogenic combustion in South Asia. Observationally-constrained insights on BC sources allows for a more credible scientific underpinning of policy aimed at mitigating the acuteness and scale of ramifications due to BC in South Asia.