Dual carbon isotope characterization of total organic carbon in wintertime carbonaceous aerosols from northern India

Srinivas Bikkina1, August Andersson1, M. M. Sarin2, R. J. Sheesley1, E. Kirillova1, R. Rengarajan2, A. K. Sudheer2, K. Ram1,3, and Örjan Gustafsson1

1Department of Environmental Science and Analytical Chemistry and the Bolin Centre for Climate Research, Stockholm University, Stockholm, Sweden, 2Planetary and Geosciences Division, Physical Research Laboratory, Ahmedabad, India, 3Institute of Environment and Sustainable Development Banaras Hindu University, Varanasi, India

Abstract Large-scale emissions of carbonaceous aerosols (CA) from South Asia impact both regional climate and air quality, yet their sources are not well constrained. Here we use source-diagnostic stable and radiocarbon isotopes (δ13C and Δ14C) to characterize CA sources at a semiurban site (Hisar: 29.2°N, 75.2°E) in the NW Indo-Gangetic Plain (IGP) and a remote high-altitude location in the Himalayan foothills (Manora Peak: 29.4°N, 79.5°E, 1950 m above sea level) in northern India during winter. The Δ14C of total aerosol organic carbon (TOC) varied from −178‰ to −63‰ at Hisar and from −198‰ to −19‰ at Manora Peak. The absence of significant differences in the 14C-based fraction biomass of TOC between Hisar (0.81 ± 0.03) and Manora Peak (0.82 ± 0.07) reveals that biomass burning/biogenic emissions (BBEs) are the dominant sources of CA at both sites. Combining this information with δ13C, other chemical tracers (K+/OC and SO42−/EC) and air mass back trajectory analyses indicate similar source regions in the IGP (e.g., Punjab and Haryana). These results highlight that CA from BBEs in the IGP are not only confined to the atmospheric boundary layer but also extend to higher elevations of the troposphere, where the synoptic-scale circulations could substantially influence their abundances both to the Himalayas and over the downwind oceanic regions such as the Indian Ocean. Given the vast emissions of CA from postharvest crop residue combustion practices in the IGP during early Northeast Monsoon, this information is important for both improved process and model understanding of climate and health effects, as well as in guiding policy decision aiming at reducing emissions.

1. Introduction

Carbonaceous aerosols (CA) cause a large but poorly constrained forcing of the climate [Ramanathan and Carmichael, 2008; Bond et al., 2013; Stocker et al., 2013] and are additionally implicated as the culprit of widespread respiratory and cardiovascular diseases [Kim et al., 2015, and references therein]. CA is an important component of the Atmospheric Brown Clouds (ABC) that seasonally envelope the Indo-Gangetic Plain (IGP), where CA affect the hydrological cycles, storm frequencies, Himalayan glacier melting and agricultural crop yields through dimming [Menon et al., 2002; Ramanathan et al., 2005; Venkataraman et al., 2005; Bollasina et al., 2011; Brown, 2014]. The combination of large emissions of light-absorbing CA from the IGP and strong solar irradiance in the tropics, makes the ABC influence on regional climate, particularly significant in South Asia [Chung et al., 2005; Ramanathan and Lelieveld, 2010].

Effective strategies to mitigate the climate and health problems of CA rely on identifying and characterizing the sources. Bottom-up emission inventories (EI) are key input to climate and chemical transport models. However, these EIs are typically associated with uncertainties in the range of 2–4, and similar large offsets have also been observed when comparing bottom-up-based models with top-down observations [Gustafsson et al., 2009; Zhao et al., 2011; Bond et al., 2013; Cohen and Wang, 2014]. These uncertainties are, in part, due to differences in the suggested relative contribution to CA from biomass burning/biogenic emissions (BBEs) vis-à-vis fossil-fuel combustion (FF-comb) sources. The EI of CA for South Asia suggest a dominance of BBEs over FF-comb sources [Bond et al., 2004; Venkataraman et al., 2005; Chen et al., 2013]. On the contrary, observation-based source apportionment of BC suggests relatively larger contribution from fossil sources [Sheesley et al., 2012; Kirillova, 2013; Bosch et al., 2014; Krishnakant et al., 2015].

The studies associated with the Indian Ocean Experiment campaign in the late 1990s concluded, based primarily on various elemental ratios such as EC/TC and using end-member compositions from other continents, that FF-comb was the dominant source class [Novakov et al., 2000; Mayol-Bracero et al., 2002].