

BLACK CARBON IN THE GULF OF MAINE: NEW INSIGHTS INTO INPUTS AND CYCLING OF COMBUSTION-DERIVED ORGANIC CARBON

by

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ABSTRACT

Emissions of black carbon (BC), the soot and char formed during incomplete combustion of fossil and biomass fuels, have increased over the last century and are estimated to be between 8 and 270 Tg BC/yr. BC may affect problems as diverse as global warming, human health, carbon cycling in ecosystems, and pollutant dynamics. However, currently there is substantial uncertainty with respect to the fate of BC released to the environment. To increase our understanding of BC's fate and effects, modifications to the Chemo-Thermal Oxidation method at 375 °C (CTO 375), a current method used to quantify BC in marine sediments, were made to measure BC concentrations in, and fluxes out of, the water column in the Gulf of Maine (GoM), a representative coastal area downwind of important BC sources of the Northeastern United States. In addition, an alternative method to infer BC concentrations in seawater by observing pyrene fluorescence losses (PFL) in spiked samples was developed. Average concentrations measured in the GoM were 5 and 4 µg/L using the modified CTO 375 and PFL methods, respectively. Although these two methodologies involve independent observations, correspondence between the modified CTO 375 and PFL methods suggested that the isolated material was both highly sorptive and refractory. These concentrations also suggested that (a) up to 50% of the "molecularly uncharacterized" particulate organic carbon (POC) in surface seawater is BC; (b) the presence of this recalcitrant organic carbon may explain why some POC is not recycled to CO₂ during its transport to depth and even within the sediment beds below; and (c) hydrophobic pollutants like polycyclic aromatic hydrocarbons (PAHs) and dioxins would have their "bioavailabilities" controlled by sorption to BC. The observed BC spatial distributions and average water-column export fluxes near 10 gBC/m² year imply that most of the BC is carried offshore by wind and is accumulated in the coastal zone. Finally, sediment records of CTO-375-derived BC inputs into the GoM for the past 100 years were consistent with historical changes in fossil and biomass fuel emissions to this area.

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