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The flux of black carbon to surface sediments on the New England continental shelf

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Abstract—Concentrations and ²¹⁰Pb-derived fluxes of Black Carbon (BC) were deduced in recent ocean margin sediments off northeastern USA to investigate the role of this carbon form in the biogeochemical cycle of this element. BC concentrations in contemporary open shelf sediments ranged between 0.11 and 1.7 mg/gdw. Temporal trends of BC in cores, as well as the pattern of chemical markers of combustion sources, were consistent with anthropogenic fossil fuel combustion dominating the BC input to these sediments in recent times. BC fluxes of 1–2 g m⁻² yr⁻¹ suggest that the northeastern USA shelf sink is of the same magnitude as estimates of the BC production from fossil fuel and biomass burning in the upwind NE USA source region. Identification of about 10% of total organic carbon as BC has implications for sedimentary carbon preservation, as well as several other aspects of the global biogeochemical cycle of carbon. *Copyright* © 1998 Elsevier Science Ltd

1. INTRODUCTION

Black carbon (BC) has been found widely distributed across the surface of the earth in the atmosphere, icecaps, soils, and sediments (summarized in Goldberg, 1985). This omnipresence results from its widespread production in combustion processes and its post-production recalcitrant nature. The ubiquity of BC has significant implications for a myriad of environmental processes. For example, this residue of incomplete combustion has been demonstrated to be an important carrier-phase of pollutants such as polycyclic aromatic hydrocarbons (PAHs) and Pb (e.g., Goldberg et al., 1981; Broman et al., 1990; Gustafsson et al., 1997a; Gustafsson and Gschwend, 1997). BC may also affect climate. BC is the principal light-absorbing aerosol component and thus affects Earth's radiative heat balance (e.g., Charlock and Sellers, 1980).

The potential importance of BC to the global carbon cycle was first pointed out by Seiler and Crutzen (1980). Formation of inert BC from vegetation fires and wood fuel combustion transfers otherwise rapidly cycling biospheric carbon into the long-term geological cycle and thus represents a sink of atmospheric carbon dioxide on short timescales. Estimated BC emission fluxes from vegetation fires were recently used by Kuhlbusch and Crutzen (1995, 1996) to demonstrate that this mechanism of BC formation may indeed explain a significant fraction of the missing carbon in the global carbon balance. Further, the depth profiles of BC in contemporary sediments have been demonstrated to reflect society's time-varying usage of fossil fuel (Goldberg et al., 1981; Wik and Natkanski, 1990; Gustafsson et al., 1997a). Since this predominantly anthropogenic BC is also expected to be of very high preservation potential, it may also constitute a significant fraction of the refractive carbon being currently buried in marine sediments. To make progress in better understanding the many diverse

roles played by BC, we need to investigate the biogeochemical processes affecting the environmental fate of BC itself.

Extensive data exist for the global concentration fields of BC in the atmosphere (summarized in Penner et al., 1993). However, quantitative measurement of BC in soil, sediment, and water samples is scarce. Accumulating evidence is revealing a recalcitrant portion of soil and sediment organic carbon with very high aromaticity (Haumaier and Zech, 1995; Skjemstad et al., 1996) and very old ¹⁴C age (Benoit et al., 1979; Leavitt et al., 1996). This evidence may suggest significant presence of fossil-fuel derived BC in soils and shallow sediments. The continuous BC records of pre-industrial sediments attest to the refractory nature of BC (Griffin and Goldberg, 1981; Herring, 1985; Clark et al., 1989) as does the greater stability towards severe oxidative treatments of BC compared to kerogen in samples from the K-T boundary (Wolbach and Anders, 1989). Based chiefly on North Pacific Ocean data, a first global estimate of the pre-industrial flux of BC to marine sediments has been made, and the magnitude of this flux (10 Tg yr⁻¹) illustrates the potential significance of oceanic sequestering of this carbon form (Suman et al., 1997).

While it is known that the abundances of BC have increased in 20th century sediments (e.g., Griffin and Goldberg, 1983; Renberg and Wik, 1985; Wik and Natkanski, 1990), there are few estimates of contemporary BC mass removal fluxes into such a sink. Recent model calculations suggest that more than 90% of the marine deposition of BC occurs on the continental shelves and that the magnitude of this transfer to shelf sediments is similar to estimates of continental BC emissions (Suman et al., 1997).

In the absence of measurements of BC flux to Atlantic Ocean margin sediments, it was the objective of this work to quantify the sedimentary import fluxes of BC to the continental shelf off northeastern USA. The suggested prominence of the continental shelf as a BC depository and the complete lack of data from Atlantic Ocean shelf sediments makes such work particularly urgent. The resultant BC sink estimate was contrasted with estimates of BC emissions from fossil fuel combustion and

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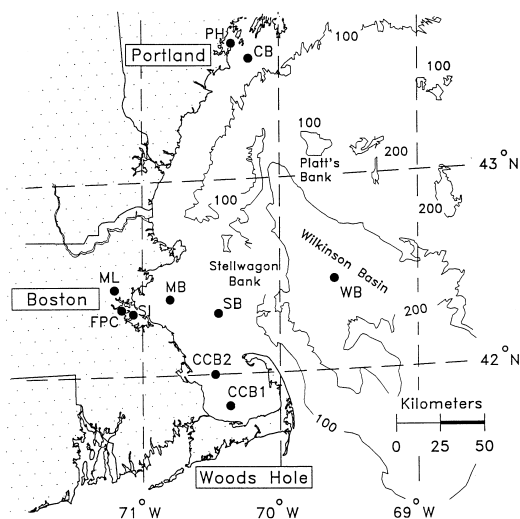


Fig. 1. Locations of sediment sampling stations in the Gulf of Maine.

biomass burning in the upwind USA source region. Since the BC origin (i.e., whether from fossil fuel or biomass-based sources) differentially affects the global carbon cycle, the relative importance of these two sources to the sedimentary BC pool was evaluated using associated PAH source signatures as a diagnostic tool. Finally, the contemporary BC fluxes were considered in the context of selective carbon preservation in marine sediments.

2. EXPERIMENTAL METHODS

2.1 Sediment Sampling

Large-volume box-cores were obtained from locations in the Gulf of Maine during several cruises (Fig. 1). In 1990, two stations were cored in Boston Harbor: at the mouth of Fort Point Channel (FPC) and off Spectacle Island (SI). In 1994, sediments in Portland Harbor (PH), outer Casco Bay (CB), and a station in the open Gulf of Maine, Wilkinson Basin (WB) were sampled, while in 1996, sediments were collected from two sites in Cape Cod Bay (CCB1 and CCB2), Massachusetts Bay (MB), and near Stellwagen Basin (SB). All samples were taken with a Sandia-Hessler type MK3 (Ocean Instruments, San Diego, CA) corer with a 0.25 m² by 0.7 m deep box. Where the sediment-water interface appeared undisturbed, we used acrylic liners (ca. 13 cm diameter) to extract subcores. The subcores were immediately extruded on board and trimmed. Sections for organic compound, total organic carbon (TOC), and BC determination were stored frozen in solvent-rinsed amber glass bottles with Al-covered Teflon-lining of the caps. Sections from parallel subcores for radionuclide analysis were placed in acid-leached plastic jars.

Sediment samples from the Palos Verdes Shelf (PV) were kindly provided by R. Eganhouse (United States Geological Survey, Reston, Virginia). Results from dated sediment samples taken in Upper Mystic Lake, MA (ML) have been previously reported (Gustafsson et al., 1997a) and were included in this study for comparison.

2.2. Quantification of Sedimentary Total Organic and Black Carbon

Our analytical method for measuring TOC and BC has been detailed elsewhere (also referred to as soot carbon, SC, and elemental carbon, EC; Gustafsson et al., 1997a; Gustafsson and Gschwend, 1997). Briefly, the TOC (including both non-BC organic carbon and BC) content of dried sediment was obtained by carbon elemental analysis (PE 2400 CHN; Perkin Elmer Corp., Norwalk, CT) after destruction of

carbonates through mild acidification of the sample in Ag capsules (Elemental Microanalysis Ltd., Manchester, NH). For BC, dried sediment was thermally oxidized at 375°C for 24 h, before acidification and carbon elemental analysis.

While we recognize that such a thermal oxidation method may include contributions of very recalcitrant non-BC organic matter such as pollen components and dinoflagellate cysts, there are several lines of evidence to suggest that such cross-contamination did not significantly affect our BC measures (Gustafsson et al., 1997a; Gustafsson and Gschwend, 1997). For example, our thermal oxidative conditions removed all but 0.15% of humic carbon (Aldrich Chemical Co.; Milwaukee, WI) and >99.95% of Kimberlite kerogen carbon. Conversely, 95% of the carbon associated with NIST SRM-1650 diesel particulate matter (National Institute of Standards and Technology, Standard Reference Material 1650, Gaithersburg, MD) withstood the treatment. Furthermore, a sediment core profile of BC mimicked the profile of combustion-derived PAHs, while being completely decoupled from the TOC profile. These data suggested that the BC results were not perturbed by inclusion of non-BC components of the TOC pool or from TOC charring during the treatment (Gustafsson et al., 1997a). Finally, BC concentrations (but not TOC) in a variety of continental shelf surface sediments were very well correlated with pyrogenic PAHs (Gustafsson and Gschwend, 1997). If anything, our conditions may err to the side of excluding material that really is BC, since our oxidative thermal cut-off (375°C) is more conservative than other similar methods (340–350°C; Cachier et al., 1989; Kuhlbusch, 1995; Novakov and Corrigan, 1996).

2.3. Quantification of Sedimentary PAHs

Concentrations of PAHs in FPC and SI were taken from published results measured in the same cores (McGroddy and Farrington, 1995). PAH concentrations for ML have also been reported elsewhere (Gustafsson et al., 1997a). In all other samples, a known mass of the wet sediment was transferred to a Soxhlet extractor and spiked with four deuterated PAH recovery standards, spanning a range of aqueous solubilities. The sediments were extracted for 48 h in a 9:1 mixture of methylene chloride and methanol. Extracts were concentrated and exchanged into hexane using Kuderna-Danish or rotary evaporation. The extracts were then charged to a gravity column containing, in order, Na₂SO₄ (anh.) and activated copper (prepared according to Blumer, 1957) to remove residual water and elemental sulfur, respectively, followed by a fully activated silica gel (Takada and Ishiwatari, 1985). The PAHs were eluted in the third fraction with 30 mL 3:1 hexane-toluene and quantified by gas chromatography-mass spectrometry (Hewlett-Packard 5995B). Recoveries of PAH internal standards for both samples and blanks were 65–95% for the entire procedure, and reported concentrations are corrected for these recoveries. PAH contents of the samples were several orders of magnitude above those of the blanks.

2.4. Quantification of ²¹⁰Pb-based Sedimentation Rates

Sediment radionuclides were determined with gamma spectrometry following the method outlined in Gustafsson et al. (1997b). Briefly, dried and homogenized sediments were transferred to plastic jars with tight screw caps, stored to allow ²²²Rn, and hence ²¹⁴Pb, grow-in and counted on a planar geometry low energy Ge detector (Canberra Instruments). Acquired signals were corrected for instrument electronic noise using counts acquired in channels surrounding the peaks and for method blanks from counting empty plastic jars. Counting efficiencies were obtained using standard additions of a calibrated radionuclide standard (pitchblende; US EPA Environmental Monitoring Systems Lab., Las Vegas, Nevada). Excess ²¹⁰Pb activities, used for deriving sedimentation rates, were obtained by subtracting supported levels (obtained from ²¹⁴Pb) from the total ²¹⁰Pb activities.

Sedimentation rates were determined by fitting the excess ²¹⁰Pb profiles in sediment strata where exponential falloffs were observed (e.g., Berner, 1980; Fig. 2). Hence, regions of near-constant excess ²¹⁰Pb activities such as the zone closest to the sediment-water interface (e.g., 0–7 cm in PH and 0–5 cm in CCB1) were not included in the curve fit. Similarly, zones of near-constant or even maxima in excess

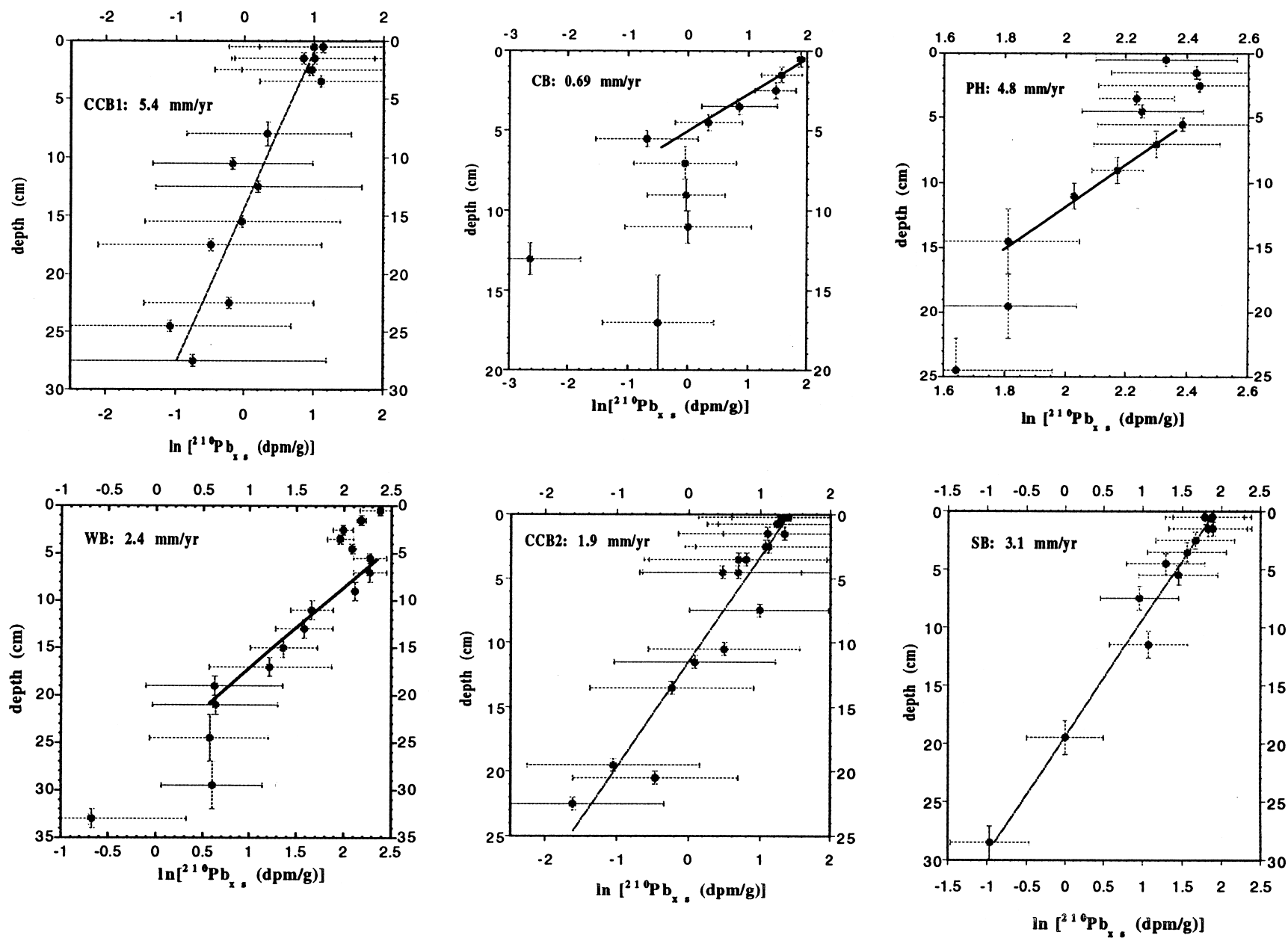


Fig. 2. Sediment profiles of excess ^{210}Pb activity used for deducing sedimentation rates.

Table 1. Concentrations and Sediment Fluxes of Black Carbon on the Continental Shelf

Sample depth (cm)	Latitude (N)	Longitude (W)	TOC (mg / gdw)	BC (mg / gdw)	BC/TOC	ϕ	ω (mm/yr)	Flux-BC (g/m ² yr)	1,7-/2,6- DMP
Harbors and Suburban Lake									
FPC	7-9	42°21'22"	71°02'41"	50.9±0.7	6.61±0.84	0.130	10.77 16.0	23	
SI	0-1	42°19'46"	70°59'34"	28.8±2.0	2.72±0.40	0.094	10.79 13.3	4.7	
PH	2-3	43°39'63"	70°14'32"	37.0±4.3	1.76±0.56	0.048	0.69 4.8	6.5	0.90
ML	10-12	42°26'	71°06'	133±1	3.68±0.16	0.028	0.89 210	11	0.60
	20-22	42°26'	71°06'	119±1	6.94±0.14	0.058			1.20
Gulf of Maine Proper									
CB	2-3	43°36'94"	70°09'31"	12.5±3.5	0.72±0.10	0.058	0.31 0.69	0.86	0.96
WB	2-3	42°38'00"	69°36'26"	22.5±0.5	0.71±0.30	0.031	0.73 2.4	1.2	0.93
CCB1	2-3	41°51'18"	70°17'54"	11.1±0.2	0.34±0.13	0.031	0.59 5.4	1.9	
	4-5	41°51'18"	70°17'54"	8.8±0.6	0.29±0.03	0.033			
	18-21	41°51'18"	70°17'54"	7.8±0.2	0.32±0.05	0.042			
CCB2	2-3	42°03'30"	70°24'36"	13.0±0.2	0.42±0.04	0.032±0.003	0.48 1.9	1.0	
	8-10	42°03'30"	70°24'36"	7.8±2.5	1.14±0.42	0.146±0.071			
MB	2-3	42°22'00"	70°54'00"	1.50±0.7	0.11±0.07	0.073			
SB	2-3	42°20'33"	70°23'30"	22.3±0.4	0.75±0.01	0.034	0.73 3.1	1.6	
	4-5	42°20'33"	70°23'30"	19.8±0.8	1.73±0.40	0.087			
Palos Verdes Shelf, off Los Angeles, CA									
PV	2-4	33°42'31"	118°21'09"	29.9±2.8	1.20±0.18	0.040	30.78 310-15 7-10		
	18-20	33°42'31"	118°21'09"	44.6±3.8	1.35±0.12	0.030			

¹MCGRODDY (1994); ²SPLIETHOFF and HEMOND (1996); ³EGANHOUSE, R.B., pers. comm. (USGS, Reston, VA)

²¹⁰Pb activities at some subsurface depth (e.g., 7–11 cm in CB and 4–7 cm in WB) were also excluded before deducing the sedimentation rates.

3. RESULTS AND DISCUSSION

3.1. Total Organic Carbon and Black Carbon

3.1.1. Concentrations of black carbon in sediments

The BC content of these contemporary shelf sediments ranged from 0.11 to 6.6 mg/gdw (gdw = gram dry weight sediment) with a geometric mean of 1 mg/gdw ($n = 15$; Table 1). The highest BC concentrations were found closer to urban sources. BC concentrations in the Gulf of Maine Proper spanned 0.11–1.7 mg/gdw (Table 1). The BC concentrations found on the New England continental shelf are similar to the contents that we found in shelf sediments off Palos Verdes (PV) California (Table 1). Our BC concentrations were also similar to the limited data available from other corresponding sites (Table 2).

3.1.2. Sources of black carbon in modern sediments

While BC in ancient sediments clearly reflect the geological record of vegetation fires, there is accumulating evidence to suggest that modern deposition of BC to aquatic sediments is heavily influenced by fossil fuel combustion. Temporal BC trends in several lacustrine cores, representing input over the past century, follow the historical development of fossil fuel consumption in those areas (Goldberg et al., 1981; Renberg and Wik, 1985; Gustafsson et al., 1997a). At the same time, vegetation fires in the USA have decreased since European colonization (Goldberg, 1985).

The sedimentary imprint of combustion products is further evident in the sedimentary records of PAHs emitted from all combustion processes (e.g., Ramdahl et al., 1983). Widely distributed sediment cores indicate a large increase in PAH concentration, of pyrogenic origin, in 20th century sediments

(e.g., Hites et al., 1977; Prahl and Carpenter, 1979; Gschwend and Hites, 1981; Gustafsson et al., 1997a). The predominance of fossil fuel combustion over residential wood combustion (and presumably also over boreal forest fires), as the major anthropogenic source of sedimentary BC and associated PAHs was further examined by us using ratios of 1,7-dimethylphenanthrene to 2,6-dimethylphenanthrene (1,7-/2,6-DMP). This ratio is known to be 0.5–0.6 for diesel particles and urban dust, whereas it reaches 2.4 in emissions from combustion of oak wood and 9 in emissions from pine burning (Benner et al., 1995). In sediment from Portland Harbor, Casco Bay, and Wilkinson Basin, we found this PAH ratio to be between 0.9 and 1.0 (Table 1), implying 80–95% of the BC-associated PAHs were derived from fossil fuel combustion. Gustafsson et al. (1997a) found that this ratio increased somewhat downcore in the Mystic Lake and reached a value above 2 at depths representative of the first half of the 20th century; a time when wood combustion may have played a relatively more significant role as an energy source than today (Hottle and Howard, 1971).

3.2. Black Carbon Accumulation Fluxes

We also used the observed surface BC inventories with sedimentation rates deduced from excess ²¹⁰Pb profiles (Fig. 2) to evaluate the sedimentary sink for BC. The derived sedimentation rates (<1–6 mm/yr; Table 1) in our study are typical of coastal and continental shelf sediments. Were steep BC concentration gradients to exist, this approach could yield an underestimation of the surface sediment BC fluxes due to biological mixing. Although only investigated in a few of the marine cores, there was relatively little change in BC content with depth (at most a factor of three) over the shallow sediment depths analyzed.

The fluxes of BC in the ten dated sediment cores ranged from 1 to 20 g m⁻² yr⁻¹ (Table 1). The highest fluxes were found in

Table 2. Black Carbon Concentrations and Fluxes in Surface Sediments

<i>Region</i>	<i>BC</i> (mg/gdw)	<i>BC/TOC</i> (%)	<i>Flux-BC</i> (g m ⁻² yr ⁻¹)	<i>Reference</i>
<i>Pelagic Regimes</i>				
N. and S. Pacific Ocean	0.009-1	n/a	0.0001-0.001	SMITH <i>et al.</i> (1973)
<i>Continental Shelf Regimes</i>				
Santa Barbara Basin, CA	0.4	n/a	0.4	GRIFFIN and GOLDBERG (1975)
Saanich Inlet, British Columbia	0.7	n/a	0.9	GRIFFIN and GOLDBERG (1975)
Lake Michigan	1.6	n/a	1.3 ^b	GOLDBERG <i>et al.</i> (1981)
Gulf of Panama	n/a	n/a	1-3 ^a	SUMAN (1986)
W. Med.Sea	0.2-1.6	5-38	1 ^a	LIM and CACHIER (1996)
Gulf of Maine	0.3-1.7	3-9	1-2	This study
<i>Near-Urban Sites</i>				
Mouth of Providence River, RI	18	n/a	14	GOLDBERG <i>et al.</i> (1977)
Palos Verdes Shelf, off Los Angeles, CA	1.2	4	7-10	This study
New England Harbors and Suburban Lake	2-7	3-13	5-23	This study

^aSUMAN *et al.* (1997)

^bassuming $\phi=0.9$ and $\rho=2$ g/cm³ in this lacustrine sediment.

the harbor and near-urban lake core (all within 10 km of an urban center), spanning a range of 5–20 g m⁻² yr⁻¹. When combining the BC concentrations with the sedimentation rates at each site (not available for MB), a remarkably tight range of BC sediment fluxes (1–2 g m⁻² yr⁻¹) was obtained for the five nonurban Gulf of Maine shelf stations.

There was also substantial agreement between these BC fluxes and those previously reported for surficial ocean margin sediments elsewhere, all in the range 0.4–3 g m⁻² yr⁻¹ (Table 2). In contrast, the mean BC flux in pelagic sediments of the Pacific Ocean was estimated at 0.0006 g m⁻² yr⁻¹ (Smith *et al.*, 1973; Griffin and Goldberg, 1975). The homogeneity in the BC fluxes to contemporary continental shelf sediments suggests that these fluxes may lend themselves to areal extrapolation to estimate the shelf sedimentary sink of BC on the continental shelf.

3.3. The Black Carbon Budget of the New England Continental Shelf

Our surface sediment fluxes provide an opportunity to estimate the magnitude of the BC sink on the continental shelf off northeastern USA. Taking the area of this shelf region as 3.8×10^5 km² from published estimates (Buesseler *et al.*, 1985/86), we estimate that 0.4–0.8 Tg yr⁻¹ of BC is currently being sequestered in the continental shelf sediments off northeastern USA (Table 3).

The estimated sedimentary black carbon flux may be contrasted to the fossil fuel emission of BC from the upwind North American continent. Since the zonal wind pattern in this region is predominantly westerlies, it may be anticipated that long-range transport from the entire northeastern USA may impact this offshore area. In fact, the surface ocean fluxes of similarly pyrogenic PAHs into the western North Atlantic have been estimated to be significant relative to the fossil fuel emissions of such substances from the northeastern USA (Gustafsson *et al.*, 1997c). For the purpose of developing a mass-balance for combustion-derived PAHs, Simonich and Hites (1994) suggested a broad definition of the northeastern USA, which included an area hosting 52% of the population. Applying this same scaling to the estimates by Penner *et al.* (1993) of the entire USA fuel combustion emission of BC yields an anthropogenic BC production in this region of 0.2–0.6 Tg yr⁻¹ (Table 3). While this emission flux is remarkably similar to our estimate of the shelf sediment flux, the comparison is complicated by the differences in BC analytical methodology utilized. The Penner *et al.* (1993) study is based on BC data from optical methods, which await to be intercompared with thermal methods such as the one employed in the present study.

Our BC sink estimates may also be contrasted with atmospheric BC emissions from burning of northeastern USA boreal and temperate forests of 0.1–0.2 Tg yr⁻¹ (see Table 3). This estimate does not include residential wood combustion, which

Table 3. Comparison of BC Fluxes in Surface Sediments with Emission Estimates

<i>Region</i>	<i>Type of Flux</i>	<i>Method of Estimation</i>	<i>Regional BC Budget</i> (Tg BC yr ⁻¹)	<i>Source</i>
Continental Shelf off NE USA	Surface Sediments	²¹⁰ Pb-derived fluxes of surface inventories	0.4-0.8	This study
NE USA	Fossil fuel combustion	BC/SO ₂ measurements coupled with known SO ₂ emissions	0.2-0.6 ^a	PENNER <i>et al.</i> (1993)
NE USA	Vegetation Fires	BC/CO ₂ from control burns coupled with known CO ₂ emissions from temperate and boreal forest	0.1-0.2	KUHLBUSCH and CRUTZEN (1995)

^aEstimate based on scaling entire US fossil fuel combustion emissions with the portion of the population living in the NE USA (52%), following the approach suggested by SIMONICH and HITES (1994) for similarly combustion-derived PAH emissions.

^bEstimate based on scaling global BC production from forest fires in the temperate and boreal zone, taking into account that only 20% is believed to be emitted to the atmosphere (KUHLBUSCH and CRUTZEN, 1995), with an estimate of the areal extent of NE USA (3.3 10⁶ km² or 8%; RAND McNALLY World Atlas, 1992).

is believed to be a significant contributor to atmospheric PAHs in this region (Ramdahl *et al.*, 1983; Benner *et al.*, 1995). There are also uncertainties regarding the fate of BC fire residues that initially remain on the ground, as opposed to being emitted into the atmosphere (e.g., Kuhlbusch and Crutzen, 1995, 1996). Nevertheless, the relative BC source strengths are consistent with the sedimentary dimethylphenanthrene ratios, strongly suggesting that fossil sources are more significant than wood burning (Table 1).

These budget calculations indicate that a major fraction of the atmospherically released BC from the northeastern USA may find itself buried in the continental shelf sediments off the East coast of the USA. While atmospheric deposition is likely to dominate the input of combustion-derived substances away from the continent (e.g., Gustafsson *et al.*, 1997c), river and urban runoff may locally be an important transport route (e.g., O'Malley *et al.*, 1996). Similar to our findings, Suman *et al.* (1997) recently suggested that about 40% of global BC emissions may be found in continental shelf sediments. The somewhat higher transfer coefficient (sediment sink-to-continental source ratio) found in our study might be anticipated based on the westerly zonal wind patterns off the eastern side of a continent that was considered here.

3.4. A Role for Black Carbon in Sedimentary Carbon Removal

Our measures of continental shelf BC may be considered in the context of the ocean carbon cycle. Over 80% of the annual

marine carbon burial (total of 90 Tg/yr) is believed to take place in continental shelf sediments (Berner, 1989). The BC fraction of the total organic carbon in the surficial margin sediments examined here ranged from 3% to above 10% (Table 1). This ratio was rather invariant (geometric mean value was 5%) in the upper sections of the sediments and reflects the importance that BC may play in carbon preservation. This BC fraction increased in the few deeper sections that were analyzed. This may be due to either selective BC preservation during early diagenesis (e.g., Wolbach and Anders, 1989) or to a changing BC input function, mirroring the switchover from coal to cleaner burning gas and oil as primary fuel that took place in this region of the world around the time represented by these deeper sediment sections (Hottle and Howard, 1971).

Further, our estimate of the BC-to-TOC ratio is similar to other recent estimates in the literature. Lim and Cachier (1996) reported that BC represented 5–38% of TOC in Mediterranean Sea surface sediments (Table 2). Based on ancient sediment data, Suman *et al.* (1997) calculated the pre-industrial global burial of BC in marine sediments to be on the order of 10 Tg/yr. This estimated BC burial takes place predominantly on the shelf and corresponds to 11% of estimated global marine sediment burial of TOC (Berner, 1989).

A present-day sequestration of BC in continental shelf sediments on the order of 5–10% or more of TOC would have important implications for our understanding of the biogeochemical cycles of carbon and associated elements. It may help to explain the occurrence of ¹⁴C-poor organic matter in shallow

margin sediments (Benoit et al., 1979; McNichol et al., 1988). A simple mixing calculation of 10% fossil BC with 90% modern carbon in surficial margin sediments yields an overall TOC age of nearly 1000 years, which corresponds well with TO¹⁴C observations in rapidly accumulating coastal sediments (Benoit et al., 1979; McNichol et al., 1988). This microparticulate aromatic residue of incomplete combustion might also in part explain why it is difficult to account for a high fraction of sedimentary TOC by conventional biochemical analysis (Hedges et al., 1988). Significant input of such land-based carbon emissions to marine sediments would complicate vertical models of carbon preservation potentials from primary production. Further, the portion of the current BC depositions that result from forest fires and wood fuel burning represents a net sink for CO₂ and conversely a net source for atmospheric O₂ (Kuhlbusch and Crutzen, 1995). Hence, interglacial variations in such atmospheric gases induced by changing biomass burning may be reflected in the sedimentary BC record (Verardo and Ruddiman, 1996; Suman et al., 1997). Further work on BC fluxes in recently deposited sediments would benefit from ¹⁴C analysis of the isolated BC fractions. Such information would contribute further towards understanding the relative importance of fossil and biomass sources to this contemporary BC sink.

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