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# Sedimentation and Organic Carbon Burial in the Yangtze River and Hudson River Estuaries: Implications for the Global Carbon Budget

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Abstract One of the most important challenges in global climate change research is balancing the carbon budget within the global carbon cycle. Carbon burial in sediments at the land–ocean interface has been difficult to quantify and model because it represents nonsteady-state boundary conditions that are also affected by human activities. In this study, we document carbon burial rates in the Yangtze River  $(1.6-4.9 \times 10^{12} \text{ gC year}^{-1})$  and Hudson River (1.8–3.6  $\times$  10<sup>10</sup> gC year<sup>-1</sup>) estuaries and integrate our results with carbon burial rates determined by others in the world's 25 largest river-estuarine systems  $(6-11 \times 10^{13} \text{ gC year}^{-1})$ . Our results indicate that carbon burial in estuaries, bays, coves, lagoons, mud flats, marshes, mangroves, and other highly productive or protected lowenergy areas at the land–ocean interface along the entirety of the world's coastlines may serve as an unrecognized sink within the global carbon budget.

Keywords Estuaries · Sediment accumulation · Organic carbon burial · Land–ocean interface - Global carbon budget

### 1 Introduction

For more than three decades, scientists examining the global carbon cycle have been looking for a missing carbon sink of  $\sim$  2 PgC year<sup>-1</sup> to balance the global carbon budget (Bolin [1977;](#page-15-0) Broecker et al. [1979](#page-15-0)). For the past two decades, this missing carbon sink has increased to  $\sim$  3 PgC year<sup>-1</sup>, and it has also been referred to as a "land sink" (Canadell et al. [2007\)](#page-15-0) or ''residual terrestrial sink'' (Houghton and Goodale [2004;](#page-16-0) Prentice et al. [2001\)](#page-16-0). In this study, we used geographic information systems (GIS) and radionuclide

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dating techniques  $(^{137}Cs)$  to examine sediment accumulation and to estimate carbon burial in both the Yangtze River and the Hudson River estuaries over the past 50 years. The Yangtze River estuary represents large river-estuarine–deltaic systems, which rapidly trap and bury carbon in sediments at the land–ocean interface. Geologists search for ancient deltaic areas in the geologic record because over time, this buried carbon forms oil. The medium-sized Hudson River estuary also effectively traps waterborne carbon and sediment due to its estuarine circulation pattern. This circulation pattern is characterized by a net seaward flow at the ocean surface and a net landward flow along the ocean bottom. As a result, the carbon produced in the shelf waters often sinks to the bottom and is thus transported landward across the shelf to accumulate in the marshes, mudflats, bays, and estuaries at the land–ocean interface. In this study, we have also integrated our results with those of others who have examined carbon burial in various estuarine systems worldwide to evaluate the importance of carbon burial at the land–ocean interface within the global carbon cycle.

As ephemeral non-steady-state features, estuaries and other coastal marginal areas are formed at the land–ocean interface during interglacial periods of ice melting and sea-level rise. Our present interglacial period started  $\sim$  18,000 years ago, during which sea level has risen  $\sim$  120 m to cover the world's continental shelves, flood coastal lands, and submerge ancient Greek and Roman cities (Fairbridge [1961](#page-15-0)). During past glacial times when sea level was much lower, major rivers discharged particles coated with carbon films (Mayer [1994\)](#page-16-0), nutrients, and particulate organic carbon (POC) directly into the deep ocean, forming channels on continental shelves and canyons on continental slopes (Broecker [2003\)](#page-15-0). During interglacial times, however, carbon-coated particles and POC are effectively trapped within the land–ocean interface. This trapping results from an estuarine circulation pattern that is governed by external forces (gravity, tides, and winds), internal forces (temperature and salinity pressure gradients), and the Coriolis Effect and is characterized by a net seaward flow of fresher water at the surface and a net landward flow of more saline water along the bottom (Fig. [1\)](#page-2-0). This net landward flow along the bottom extends from across the continental shelf to the tip of salt water penetration in river-estuarine systems (Biscaye [1994\)](#page-15-0), where iron and dissolved organic carbon (DOC) flocculates along with carbon-coated particles, POC, and resuspended sediments to form a zone of turbidity maximum (Schubel [1968;](#page-17-0) Sholkovitz [1976\)](#page-17-0) and a zone of rapid sediment and carbon accumulation.

Many large estuaries, like Chesapeake Bay, have multiple deltas, one forming at its mouth and building landward due to sandy sediments supplied by longshore and acrossshelf transport, and several other deltas forming at the mouths of its tributary rivers, due to the flocculation and deposition of finer-grained sediments (Donoghue et al. [1989](#page-15-0)). In the Savannah River estuary, Olsen et al. ([1989\)](#page-16-0) showed that riverborne particles had a distinctly different  $^{238}Pu/^{239,240}Pu$  ratio compared to the global fallout plutonium ratio measured on oceanic particles. They used this difference to document that riverborne particles were primarily deposited in upstream areas of the Savannah River near its estuarine turbidity maximum and that the sediments rapidly accumulating in the estuary and in Savannah Harbor were marine particles that were transported along shore and landward across the shelf or produced in situ within the estuary. Mulholland and Olsen ([1992\)](#page-16-0) used stable isotope ratios of carbon, nitrogen, and sulfur to determine the source functions for organic carbon in the Savannah River estuary. Their results indicated that over 74 % of the organic particles in suspension or on the bottom of the estuary were of marine origin and that  $\sim$  22 % of the organic material (in bottom sediments) near the tip of salt water penetration was also of marine or estuarine origin. Therefore, particles, carbon,

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Fig. 1 Schematic drawing of net estuarine circulation at the land–ocean interface indicating that particles and carbon from riverine, marginal, and marine sources are effectively trapped at the land–ocean interface

and particle-reactive contaminants transported from watersheds, introduced via municipal wastewater inputs, produced in situ, or transported landward from marine sources are effectively being trapped at the land–ocean interface due to a rising sea level during our current interglacial period. Today, oil and gas companies use seismic profiling and other geophysical methods to search for ancient estuaries, bays, and deltas in the geologic record as source rocks for hydrocarbons.

Although differences in the biological, chemical, geological, and hydrological characteristics cause patterns of sediment accumulation and carbon burial to be complex and unique within each estuarine system (Wright [1989](#page-17-0)), there appear to be certain generic features that are common to all. Using the established geological concept of ''an equi-librium profile," Olsen et al. ([1993\)](#page-16-0)documented that the accumulation of particles and organic carbon tends to be negligible  $(0-5 \text{ mm year}^{-1})$  in estuarine and coastal areas, where the sediment surface has reached a state of "dynamic equilibrium" with its presentday hydraulic regime. These areas generally occur naturally in river channels, on subtidal banks, and on continental shelves (Type I Areas in Fig. [2\)](#page-3-0). The suspended particulate load tends to bypass these ''equilibrium-surface'' areas through a series of deposition and resuspension events to accumulate at moderate rates  $(1-5 \text{ cm year}^{-1})$  in protected areas, such as marginal coves (Type II Areas in Fig. [2\)](#page-3-0), or at extremely rapid rates  $(5-100 \text{ cm year}^{-1})$  in "non-equilibrium" areas (Type III Areas in Fig. [2\)](#page-3-0), where the sediment surface is temporally out of equilibrium with its physical regime. As illustrated in Figs. 1 and [2](#page-3-0), these ''non-equilibrium areas'' of rapid sediment accumulation and carbon burial can occur naturally in river-estuarine systems, such as around the turbidity maxima, in protected marginal areas, in deltaic environments, or in areas affected by human activities, such as dredged channels, harbor slips, piers, bridges, dams, or other structures affecting the physical regime (Schubel and Meade [1977](#page-17-0); Smith [2007;](#page-17-0) Zhu [2010](#page-17-0)).

Scientists throughout the world have been examining the physics and biogeochemistry of estuaries and using carbon to understand, quantify, and model ecological dynamics at the land–ocean interface (Aller et al. [2008](#page-15-0); Berner [1982;](#page-15-0) Benner et al. [2004;](#page-15-0) Bianchi and Allison  $2009$ ; Dürr et al.  $2011$ ; McKee et al.  $2004$ ). Within the global carbon cycle, the

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Fig. 2 Vertical profiles of cesium-137 (half-life  $=$  30 years) in sediment cores collected in estuaries along the east coast of the United States generally fall into three distinct types. These three types reflect the extent to which the estuarine sediment surface has attained equilibrium with its present wave, current, flow, and sea-level regime. This figure was modified from Olsen et al. ([1993](#page-16-0))

importance of the estuarine water column as a source for  $CO<sub>2</sub>$  to the atmosphere has been well documented (Cai et al. [2006](#page-15-0); Fagan and Mackenzie [2007;](#page-15-0) Green et al. [2006](#page-16-0)).The importance of carbon burial at the land–ocean interface as a sink for atmospheric  $CO<sub>2</sub>$ within the global carbon cycle has been difficult to evaluate, because (1) estuaries are ephemeral non-steady-state features that form at the land–ocean interface during interglacial warming periods, which complicates boundary-condition considerations in modeling carbon source and sink functions; (2) sediment and carbon accumulation patterns are affected simultaneously by both short-term processes (tides, waves, storms, and seasonal changes in freshwater discharge) and long-term processes in association with sea-level rise; and (3) estuaries often experience non-steady-state conditions associated with increasing human population, nutrient supply, and urbanization.

#### 2 Study Areas

### 2.1 Yangtze River Estuary

The Yangtze River is the largest river in China, draining an area of  $\sim 1.8 \times 10^6$  km<sup>2</sup> ( $\sim$ 20 % of the mainland). On a global basis, the Yangtze River ranks fifth for freshwater discharge and fourth for sediment load (Eisma [1998](#page-15-0)). Annual freshwater discharge and sediment load averaged about  $9.0 \times 10^{11}$  m<sup>3</sup> year<sup>-1</sup> and  $4.3 \times 10^8$  mt year<sup>-1</sup>,

<span id="page-4-0"></span>respectively, from 1951 to 2000, as gauged at Datong Station located  $\sim 600$  km upstream from the Yangtze River mouth. The Yangtze River mouth at the land–ocean interface is presently 120 km long and 90 km wide at its outer limit, where the river is bifurcated into the North and South Branches at the Chongming Island, split into the North and South Channels in the South Branch by Changxing and Hengsha Islands, and finally branched into the North and South Passages by the Jiuduansha Shoal (Fig. 3). Tidal height averages 2–3 m with a maximum height of 6 m (Yang [1999\)](#page-17-0).

The deposition of sediments in the Yangtze River estuary and delta has resulted in the formation of land, averaging  $\sim 5 \text{ km}^2 \text{ year}^{-1}$  over the last 2,000 years (Yang et al. [2001](#page-17-0)). Presently, over 98 % of the riverine discharge flows through the South Branch (Fig. 3), and as a result, the South Branch outlets are currently the major deltaic depocenters for riverine sediments (Yang et al. [2003](#page-17-0)). Approximately half of the riverine sediments is retained within the estuary and the other half is discharged onto the shelf and transported along the southeast coast of China by alongshore currents (Yang et al. [2000](#page-17-0)). As illustrated in Fig. 3, this study examines sediment accumulation in tidal flat areas (east of the Chongming, Changxing, Hengsha, and Jiuduansha Islands), in deltaic topset bed area (east of the Jiuduansha Shoal), and along a 60 km transect across the topset and foreset beds of the Yangtze River delta. The estimate of organic carbon burial (based on sediment accumulation rate) was confined within a  $2,500 \text{ km}^2$  area.



### **Yangtze River Estuary Study Areas**

Fig. 3 Map of the Yangtze River estuary illustrating its tidal flat areas, as well as its deltaic topset and foreset-bed areas. The tidal flat study areas are the *shaded areas* located east of the 121°50'E line of latitude. The deltaic topset bed study area is located east of the Jiuduansha Shoal (small-boxed area A). The larger box covers a 2,500 km<sup>2</sup> Type III area, which encompasses both the topset and the foreset beds of the Yangtze River delta. The line within the larger box illustrates our deltaic transect

### <span id="page-5-0"></span>2.2 Hudson River Estuary

The Hudson River estuary is one of the most urbanized and industrialized estuaries on the east coast of the United States, draining an area of  $\sim 3.5 \times 10^4$  km<sup>2</sup>. Mean monthly freshwater discharge past the southern tip of the Manhattan Island (Fig. 4) ranges from 150 to 250 m<sup>3</sup> s<sup>-1</sup> during low-flow conditions in the summer to as high as 1,000-1,500 m<sup>3</sup> s<sup>-1</sup> during the spring freshet (Chillrud [1996\)](#page-15-0). Annual freshwater flow averaged  $\sim$  1.7  $\times$  10<sup>10</sup> m<sup>3</sup>. Mean tidal flow varies from 3,000 to 10,000 m<sup>3</sup> s<sup>-1</sup> with maximum tidal current velocities of  $\sim 1 \text{ m s}^{-1}$ . As a partially mixed estuary, an estuarine turbidity maximum (ETM) forms and its position fluctuates with freshwater discharge. During low freshwater flow conditions, the ETM can reach as far as  $\sim 60$  km upstream of the Battery (Geyer et al. [2001](#page-16-0)). During high freshwater flow conditions, the ETM can penetrate as far downstream as the George Washington Bridge,  $\sim$  18 km upstream of the Battery (Woodruff et al. [2001](#page-17-0)). Mean annual suspended sediment load from the freshwater reach of the estuary is 737,000 mt (Wall et al., [2008\)](#page-17-0). The Hudson River estuary also receives  $\sim$  100 m<sup>3</sup> s<sup>-1</sup> of wastewater from sewage treatment facilities, as well as discharges from about 400 industrial sources and 700 combined storm water and untreated sewage outfalls (CSOs). Wastewater can account for as much as 40 % of the freshwater flow during lowflow conditions, and  $\sim$ 90 % of the wastewater discharge occurs in the lower 30 km of the estuary (Sanudo-Wilhelmy and Gill [1999\)](#page-16-0).

In this study, the lower Hudson River estuary is defined as the area extending from its mouth at the Narrows to the George Washington Bridge (Fig. 4). Previous studies have indicated that the riverine suspended particulate load undergoes a series of deposition and



Fig. 4 Map of the lower Hudson River estuary identifying sediment core locations and illustrating the  $137Cs$ vertical profiles representing the three estuarine generic-type areas for sediment and carbon accumulation

resuspension events to ultimately accumulate at extremely rapid rates  $(5-20 \text{ cm year}^{-1})$  in the dredged channels, abandoned harbor slips, and other low-energy areas in the lower Hudson River estuary (Bopp et al. [1993](#page-15-0); Smith [2007;](#page-17-0) Zhu [2010\)](#page-17-0).

# 3 Sites and Methodology

# 3.1 Hudson River Estuary

# 3.1.1 Sediment Sampling

This study used data obtained from more than 80 sediment cores collected in the Hudson River estuary from the early 1970s to 2007 (Fig. [4\)](#page-5-0). Short sediment cores, ranging from 50 to 100 cm in length, were collected using a gravity coring device, and longer sediment cores, ranging up to 250 cm, were collected using vibra- or piston-corers. All the sediment cores were extruded, described, and sectioned within 24 h of collection. The sediment samples were weighed and dried in an oven at 50  $^{\circ}$ C over 24 h, weighed again, ground uniformly using a mortar and pestle, placed in plastic-lined aluminum cans, and analyzed for gamma-emitting radionuclides and sediment total organic carbon content (TOCsed). Sediment water content is determined by using a function of pore water/bottom water ratio and (wet weight-dry weight)/wet weight ratio.

# 3.1.2 Gamma Spectrometric Analysis

All sediment radionuclide activities were determined by nondestructive gamma spectrometry. Most recently, these analyses were conducted using an ultra low-background, high-resolution Canberra (Be5030) Broad Energy Germanium planar detector, equipped with a Genie 2000 MCA (Canberra) microprocessor and an interactive peak-fit software package. The measured vertical distribution of cesium-137 ( $^{137}Cs$ , half-life = 30 years) in sediment cores was used to evaluate the rates of sediment and carbon accumulation in the lower Hudson River estuary. Most of the <sup>137</sup>Cs delivered to the Hudson River watershed occurred via global fallout from nuclear weapons testing starting in the later 1940s, which exhibited a peak in 1963. In addition, there was a significant coolant water release of  $137Cs$ in 1971 from the Indian Point Nuclear Power Plant (Buchanan, NY) that also served as a sediment stratigraphic marker for dating purposes (Chillrud [1996\)](#page-15-0).

# 3.1.3 Carbon–Hydrogen–Nitrogen (CHN) Analysis

Sediment total organic carbon content  $(TOC_{\text{sed}})$  was determined using loss-on-ignition for samples collected in the 1970s, and a Perkin-Elmer 2400 Carbon, Hydrogen, and Nitrogen (CHN) analyzer was used for more recent samples. The loss-on-ignition method estimated organic matter concentration by the weight loss-on-ignition at  $375 \text{ °C}$  for samples taken on precombusted and preweighed glass-fiber filters (GFC, 1.0 micron pore size).The CHNanalyzed sediment samples were acidified using 2–5 ml of 1 N HCl and then dried for 24–48 h at 60  $\degree$ C to remove the inorganic fraction of carbon in the sediments. The CHN analyzer was calibrated using Acetanilide (71  $\%$  C and 10  $\%$  N) as the standards. Errors in the measured  $\text{TOC}_{\text{sed}}$  values were calculated using the standard deviation of the measured standard values.

### 3.1.4 Bathymetric Change Analysis

Sediment accumulation patterns in the lower Hudson River estuary were also determined by comparing changes in bathymetry between 1930 and 2001, using two sets of navigational charts (sounding images) obtained from U.S. National Oceanic and Atmospheric Administration (NOAA).The navigational charts were geo-referenced to the same geographic coordinate system using ERDAS IMAGINE 9.1 software and then digitized using ESRI ArcGIS 9.2 software, to obtain the bathymetric information. These navigational charts covered our entire study area and were used to estimate sediment accumulation rates in the lower Hudson River estuary over the same time frame covered by our  $137Cs$  dating technique.

### 3.2 Yangtze River Estuary

A total of 19 navigational charts of the Yangtze River estuary from 1958 to 2001 were obtained from the Shanghai Sea Route and China Maritime Survey Bureaus to conduct bathymetric change analysis. These navigational charts spanning different time periods were digitized using MapInfo 6, processed using Surfer 3 and overlaid one another to determine the sediment accumulation rates on the tidal flat, on the deltaic topset bed, and along the transect over time. The sediment total organic carbon content  $(TOC_{sed})$  in the Yangtze River estuary has been reported by others, ranging from  $0.5$  to  $1.5\%$  (Lu et al. [2006;](#page-16-0) Wu et al. [2007\)](#page-17-0).

### 4 Results and Discussion

#### 4.1 Sediment Accumulation in the Yangtze River Estuary

Results from bathymetric change analysis described above indicate that sediments have been rapidly accumulating both laterally (progradation) and vertically (accretion) in the Yangtze River estuary over the last 40 years (Fig. [5](#page-8-0)). The growth rates of tidal flat surface areas have ranged from  $\sim$  3 to 13 km<sup>2</sup> year<sup>-1</sup> (Fig. [5a](#page-8-0)). The sediment accumulation rate of the deltaic topset bed ranged from 5 to 20 cm year<sup>-1</sup> (Fig. [5b](#page-8-0)). Vertical sediment accretion rates across the deltaic topset and foreset beds ranged from 5 to 20 cm year<sup>-1</sup>, and the horizontal progradation of the entire Yangtze River delta varied from 6 to 12 km between 1958 and 1998 (Fig. [5c](#page-8-0)).

The data illustrated in Fig. [5](#page-8-0) also indicate a decreasing trend in the rates of tidal flat growth and deltaic topset bed accretion. This decreasing trend probably reflects a reduction in the sediment load delivered to the Yangtze River estuary over the past 50 years. Previous research by Zhu ([2003\)](#page-17-0) documented that the sediment load at Datong Gauging station decreased by  $\sim$  25 % from 1951 to 2000 (Fig. [5d](#page-8-0)). Zhu [\(2003](#page-17-0)) also showed that a strong linear correlation ( $R^2 = 0.8$  and  $P < 0.01$ ) existed between the sediment load delivered to the East China Sea and the accretion rate of deltaic topset bed within the Yangtze River estuary. Previous research by Yang et al. ([2005\)](#page-17-0) also documented response of delta intertidal wetlands to decrease in riverine sediment supply. They suggested that when the deposition rate is higher than sinking rate (compaction rate  $+$  subsidence rate), it leads to increase in intertidal wetland elevation, which is in line with results from this study. Despite the decreasing trend in the riverine sediment loads to the estuary over the past 50 years (Fig. [5d](#page-8-0)), our results indicate that the average sediment accumulation rate

<span id="page-8-0"></span>

Fig. 5 Illustrations documenting changes in a Yangtze River tidal flat growth rates, b deltaic topset bed accumulation rates, c deltaic sediment surface elevation and foreset-bed progradation rates, and d changes in the riverine sediment load. These illustrations indicate rapid sediment accumulation, both vertically and laterally, in the Yangtze River estuary over a 40-year time period (1958–1998) and a decreasing trend in the riverine sediment load delivered to the estuary from 1951 to 2000

 $(\sim 10 \text{ cm year}^{-1})$  is still very high. Although there were upstream agriculture and levee developments during 1951–2000 (Yang et al. [2004](#page-17-0)), there were no dredging activities in the North Passage area of the Yangtze River estuary (Fig. [3\)](#page-4-0), and the Three Gorges Dam was not yet constructed. As a result, sediment accumulation during our time period of study primarily reflected natural processes. Therefore, results from this study may provide baseline information that can be used to assess the impacts of urbanization and anthropogenic activities, such as large-dam construction and harbor dredging, on sediment dynamics and accumulation patterns within the Yangtze River estuary. Previous studies by Yang et al. ([2005\)](#page-17-0) and Zhu ([2006\)](#page-17-0) have shown that the construction of the Three Gorges Dam in 2001 significantly decreased the riverine sediment supply to the Yangtze River estuary.

### 4.2 Sediment Accumulation in the Lower Hudson River Estuary

Sediment accumulation patterns and rates were, respectively, determined in the lower Hudson River estuary, over a 70-year time frame using bathymetric change analysis (Fig. [6\)](#page-9-0) and radionuclide dating techniques (Fig. [7\)](#page-10-0). Figure [6](#page-9-0) was made based on the changes in bathymetry over time, indicating sediment accumulation (shoaling) and erosion

<span id="page-9-0"></span>(deepening). Channel deepening could be due to sediment erosion or human activities, such as dredging. Dredging creates temporary sediment accommodation space, which leads to sediment accumulation. Therefore, the actual long-term deepening is likely due to sediment erosion. Figure [7](#page-10-0) shows cesium-137 and organic carbon vertical profiles in two sediments cores collected in two locations, representing two types of sedimentation regimes. As indicated in Figs. 6 and [7,](#page-10-0) the areas of sediment accumulation determined by bathymetric change analysis often coincided with the ''non-equilibrium areas'' that were determined by the  $137$ Cs sediment-depth profiles.

Previous studies have shown that  $\sim 1.6 \times 10^6$  mt of fine-grained sediments are annually accumulating in the lower Hudson River estuary (Bokuniewicz and Ellsworth [1986;](#page-15-0) Chillrud [1996](#page-15-0)). Ellsworth ([1986\)](#page-15-0) estimated that system-wide accumulation is 0.5–5  $\times$  10<sup>5</sup> mt year<sup>-1</sup> greater than the sediment input from the riverine and marine sources. This deficit could potentially be balanced by bottom erosion 4.4  $\times$  10<sup>5</sup> mt year<sup>-1</sup> (Klingbeil and Sommerfield [2005](#page-16-0)). Olsen et al. [\(1984\)](#page-16-0) quantified the source functions for this total annual sediment input using cesium and plutonium isotopic tracers. Their results indicated that  $\sim$  1.1  $\pm$  0.3  $\times$  10<sup>6</sup> mt year<sup>-1</sup> of fine-grained sediments were derived from



Fig. 6 Map of the New York/New Jersey Harbor (within the lower Hudson River estuary) illustrating changes in the sediment surface (shoaling or deepening) over a 70-year time span. In this urbanized estuary, dredging activities create a sediment surface far below its equilibrium depth, causing rapid sediment accumulation and carbon burial. As a result, the amount of sediment accumulating in the estuarine channel is balanced by the amount of sediment that is annually dredged from the channel ( $\sim 1 \times 10^6$  mt year<sup>-1</sup>)

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**Vertical 137Cs and Carbon Profiles in Type II and Type III Area Sediment Cores** 

Fig. 7 Vertical distributions of  $^{137}$ Cs and sediment organic carbon content (TOC<sub>sed</sub>) in a sediment pistoncore (collected during 1975 in a previously dredged channel) and in a sediment vibra-core (collected during 2007 in a protected marginal cove). The large  $137Cs$  activity peak observed in both sediment cores was associated with a major coolant water release from the Indian Point Nuclear Power Facility in 1971

riverborne sources;  $\sim 0.15 \pm 0.1 \times 10^6$  mt year<sup>-1</sup> were derived from sewage solid inputs and in situ primary production; and  $\sim 0.35 \pm 0.35 \times 10^6$  mt year<sup>-1</sup> were transported landward into the estuary from marine sources. Records from the U.S. Army Corps of Engineers indicate that  $\sim 1 \times 10^6$  mt of sediment were annually dredged from the main harbor channels to maintain "steady-state" navigable harbor depths (Panuzio [1965](#page-16-0)). If we assume that the remaining  $\sim 0.6 \times 10^6$  mt of the total sediment input (1.6  $\times$  10<sup>6</sup> mt) are accumulating in non-dredged areas of the lower Hudson River estuary, then the, nonet sediment accumulation has been balanced by channel dredging activities over the past 70 yrs as determined by our bathymetric change analysis in the main channel. The organicrich and highly contaminated dredged spoils were previously dumped onto the continental shelf. Due to the landward transport of dredged spoils across the shelf bottom and their subsequent accumulation in protected areas onshore, these dredged spoils are no longer discharged offshore and are now used for landfill (Swanson et al. [2004](#page-17-0)).

The vertical profiles of  $^{137}Cs$  and TOC<sub>sed</sub> for two selected sediment cores are illustrated and described in Fig. 7. Both sediment cores exhibit a large peak in  $137Cs$ , which was associated with a major release of coolant water in 1971 from the Indian Point Nuclear Power Plant (Buchanan, NY). Correlation of this 1971 reactor peak with the vertical distribution of  $137$ Cs in the sediments of the piston-core collected in 1975 indicated an average sediment accumulation rate of  $\sim$  15 cm year<sup>-1</sup> in the previously dredged channel. The vertical distribution of TOC in the sediments of the 1975 piston-core ranges from 2 to 4 %. Correlation of the 1971 reactor peak with the vertical distribution of <sup>137</sup>Cs in the sediments of the vibra-core collected in 2007 indicated an average sediment accumulation

rate of  $\sim$  2 cm year<sup>-1</sup> in the protected marginal cove near the George Washington Bridge. The global  $137Cs$  peak was not observed in either of the two sediment cores likely due to mixing. The vertical distribution of TOC in the sediments of the 2007 vibra-core also ranges from 2 to 4 %. The  $137Cs$  peak in both cores did not reflect changes in sediment grain size or composition as indicated by the measured vertical distribution of  ${}^{40}$ K (halflife =  $1.27 \times 10^9$  years). Although the <sup>40</sup>K data are not presented in Fig. [7,](#page-10-0) previous work by Olsen ([1979\)](#page-16-0) has shown that  ${}^{40}K$  is a good indicator of sediment grain size and composition (i.e., quartz-rich sandy sediments are characterized by low  ${}^{40}$ K activities).

### 4.3 Sediment Organic Carbon Burial in the Yangtze River Estuary and the Hudson River Estuary

Sediment organic carbon burial  $(C_{\text{Burial}})$  in both the Yangtze River and the Hudson River estuaries is a function of area (A), sediment accumulation rate (SAR), sediment bulk density (SBD), sediment total organic carbon content  $(TOC_{\text{sed}})$ , and its sediment burial efficiency(SBE).

$$
C_{\text{Bural}} = A \times \text{SAR} \times \text{SBD} \times \text{TOC}_{\text{sed}} \times \text{SBE}
$$

The sediment carbon burial efficiency (SBE) is the only unknown parameter in the above equation that is needed to calculate total organic carbon burial  $(C_{\text{Burial}})$  in the sediments for both the Hudson and the Yangtze River estuaries. Because of the uncertainties associated with carbon remineralization and burial rates, we decided to use a sediment carbon burial efficiency of 100 % for Type III areas where sediment accumulation and carbon burial are  $>5$  cm year<sup>-1</sup>, as supported by our results in the Hudson River estuary and by other studies in the Yangtze River estuary, such as Lu et al. ([2006](#page-16-0)). For Type II areas (marginal coves, with sediment accumulation rates of  $1-5$  cm year<sup>-1</sup>), we also used a carbon burial efficiency of 100 %, which is also supported by our results in the Hudson River estuary. For Type I areas (in equilibrium with sea-level rise), we expect that the only carbon buried in these dynamic resuspension environments would be carbon trapped in the pores of mineral grains, where it is unavailable for remineralization (Mayer and Johnson [1994\)](#page-16-0).

In the Yangtze River estuary, our study area encompassed the deltaic topset and foreset beds (see large box in Fig. [3](#page-4-0)), which also represented Type III areas. As a result, we expect that  $\sim$  100 % of the carbon deposited on the Yangtze River delta is also retained in the sediments, which is supported by Lu et al. [\(2006\)](#page-16-0), who showed no appreciable loss of organic carbon with depth in the sediment cores that they collected in the Yangtze River estuary. To calculate the total organic carbon burial  $(C_{\text{Burial}})$  in the Yangtze River estuary, we used the following parameters: a Type III area  $(A)$  of 2,500 km<sup>2</sup>; an average sediment accumulation rate (SAR) of 10 cm year<sup>-1</sup>, an average of sedimentation rates on the deltaic topset bed and along the transect (Fig. [3\)](#page-4-0); a sediment bulk density (SBD) of 1.3 g  $cm^{-3}$ (Yang et al. [2001](#page-17-0)); a range of sediment organic carbon content (TOC<sub>sed</sub>) of 0.5–1.5 % (Lu et al. [2006](#page-16-0); Wu et al. [2007](#page-17-0); Zhu et al. [2006](#page-17-0)); and a sediment carbon burial efficiency (SBE) of 100 %.This calculation indicates that the total organic carbon burial in non-equilibrium Type III areas of the Yangtze River estuary and delta  $(2,500 \text{ km}^2)$  ranged from 1.6 to  $4.9 \times 10^{12}$  gC year<sup>-1</sup>. The total organic carbon burial rate over the entire East China Sea Shelf including the Yangtze River estuary and delta was  $7.4 \times 10^{12}$  gC year<sup>-1</sup>, as determined by Deng et al. ([2006](#page-15-0)). As a result, it appears that the total organic carbon burial in our small study area at the river mouth could account for as much as 66 % of the total organic carbon buried on the entire East China Sea Shelf.

In the Hudson River estuary, it appears that  $\sim$  100 % of the organic carbon deposited is retained within the sediments of Type II  $(1-5 \text{ cm year}^{-1})$  and Type III  $(5-20 \text{ cm year}^{-1})$ areas, which is supported by the no appreciable loss of organic carbon with sediment depth (see Fig. [7](#page-10-0)). The observed variations of organic carbon in these sediment cores were primarily associated with changes in sediment grain size. To calculate the total organic carbon burial ( $C_{\text{Burial}}$ ) in the lower Hudson River estuary, we used the following parameters: Type II and III areas (A) of 20  $km^2$ , extending from the Narrows to the George Washington Bridge; an average sediment accumulation rate (SAR) of 4.5 cm year<sup>-1</sup>, an average of sediment accumulation rates in Type II and Type III areas; a sediment bulk density (SBD) of 1 g  $cm^{-3}$  (based on our lab measurements); a sediment organic carbon content (TOC<sub>sed</sub>) of 2–4 % (based on our lab measurements); and an organic carbon burial efficiency (SBE) of 100 % as there was no loss of organic carbon with depth in the sediment cores. This calculation indicated that the total organic carbon burial in the lower Hudson River estuary ranged from 1.8 to 3.6  $\times$  10<sup>10</sup> gC year<sup>-1</sup>.

### 4.4 Carbon Burial in Estuarine Environments and its Global Implications

Previous studies on global carbon cycle have provided a variety of useful perspectives on organic carbon burial in different ocean regimes (Berner [1989](#page-15-0); Hedges and Keil [1995;](#page-16-0) Schlunz and Schneider [2000](#page-17-0)). To investigate the amount of carbon burial in estuarine and deltaic environments worldwide, we have combined our results for the Yangtze and Hudson River estuaries with the results for other river-dominated estuaries in Table [1](#page-13-0). Because the types of depositional environments for sediment accumulation and carbon burial in these 25 largest river-estuarine systems are unknown, we used a range of sediment carbon burial efficiencies (SBE) between 50 and 100 % to determine the potential range of total carbon buried in these river-estuarine systems. As indicated in Table [1](#page-13-0), the 25 largest rivers in the world (in terms of sediment discharge) deliver  $\sim 6.5 \times 10^{15}$  g year<sup>-1</sup> of sediments to their respective estuarine and coastal environments. If we assume a sediment organic carbon content of 1.75 %, based on the average of the high end of  $TOC<sub>sed</sub>$  for the Yangtze River estuary (1.5 %) and the low end of  $TOC_{\text{sed}}$  for the Hudson River estuary (2 %), and use a range of 50 and 100 % for the sediment carbon burial efficiency, then the total carbon burial within these 25 largest river-estuarine systems would range from 6 to  $11 \times 10^{13}$  gC year<sup>-1</sup>.

Globally, rivers discharge about  $\sim 2 \times 10^{16}$  g year<sup>-1</sup> of fluvial sediment to the land– ocean interface (Meade [1996](#page-16-0)). The POC associated with this fluvial input has been estimated to be  $\sim 2.5 \times 10^{14}$  gC year<sup>-1</sup>, which represents about half of the total organic carbon discharged by rivers (Meybeck [1982](#page-16-0); Schlunz and Schneider [2000\)](#page-17-0). Less than 5 % of the riverine particles delivered to the coastal margin reach the deep sea (Hedges [1992](#page-16-0)). This illustrates the effectiveness of the two-layer estuarine circulation pattern, which results in significant amounts of shelf sediment and carbon being transported landward during interglacial times and rapidly buried at the land–ocean interface, where the sediment surface is out of equilibrium with its physical regime (Olsen et al. [1993\)](#page-16-0). The rapid accumulation of carbon-coated particles and the burial of organic carbon in these natural or human-impacted "non-equilibrium" areas removes TOC<sub>sed</sub> from exposure to oxygenated surface water, captures it within the sedimentary environment by rapid deposition and burial, and causes low-energy areas at the land–ocean interface to be net sinks for atmospheric  $CO<sub>2</sub>$ .

River	Sediment discharge <sup>a</sup> $(10^6 \text{ mt year}^{-1})$	Sediment discharge rank	Organic carbon burial $(10^{12} \text{ gC year}^{-1})$	References
Amazon, Brazil	1,150	1	4.5	Showers and Angle (1986)
Yellow, China	1,100	2	$3.1 - 6.2^d$	Coynel et al. $(2005)$
Ganges-Brahmaputra, Bangladesh	1,050	3	7.2	Galy et al. (2007)
Yangtze, China (selected estuarine area)	480	$\overline{4}$	$1.6 - 4.9$	This study
Yangtze, China $(estuary + shelf)$	480	$\overline{4}$	7.4	Deng et al. $(2006)$
Irrawaddy, Burma	260	5	$1.4 - 2.8$ <sup>b,d</sup>	Bird et al. (2008)
Magdalena, Colombia	220	6	ND <sup>c</sup>	
Mississippi, USA	210	$\tau$	$2 - 4^d$	Bianchi et al. (2007)
Godavari, India	170	8	0.6	Balakrishna and Probst (2005)
Mekong, Vietnam	160	9	ND	
Red, Vietnam	160	9	ND	
Orinoco, Venezuela	150	11	1.7	Coynel et al. $(2005)$
Purari/Fly, New Guinea	110	12	0.52	Aller et al. (2008)
Mackenzie, Canada	100	13	$\overline{c}$	Dittmar and Kattner (2003)
Parana/Uruguay, Brazil	100	13	ND	
Salween, Burma	100	13	$1.4 - 2.8^{b,d}$	Bird et al. (2008)
Pearl, China	80	16	0.5	Ni et al. (2008)
Copper, USA	70	17	ND	
Choshui, Taiwan	66	18	$0.25 - 0.5$ <sup>d</sup>	Goldsmith et al. (2008)
Yukon, USA	60	19	<b>ND</b>	
Amur, Russia	52	20	0.9	Nakatsuka et al. (2004)
Indus, Pakistan	50	21	ND	
Zaire, Zaire	43	22	$\overline{c}$	Coynel et al. $(2005)$
Liao, China	41	23	ND	
Danube, Romania	40	24	ND	
Niger, Africa	40	25	ND	
Yenisey, Russia	5		0.17	Dittmar and Kattner (2003)
Lena, Russia	11		0.47	Dittmar and Kattner (2003)
Ob, Russia	16		0.31	Dittmar and Kattner (2003)
Hudson, USA	1.6		$0.02 - 0.04$	This study

<span id="page-13-0"></span>**Table 1** Sediment discharge ( $10^6$  mt year<sup>-1</sup>) and organic carbon burial ( $10^{12}$  gC year<sup>-1</sup>) in selected rivers worldwide

<sup>a</sup> Data from Milliman and Meade ([1983](#page-16-0)) and Meade ([1996](#page-16-0))

**b** Is an averaged value

<sup>c</sup> No data (ND)

<sup>d</sup> Organic carbon burial was calculated using the total riverine POC input and assuming a 50–100 % carbon burial efficiency

### 5 Conclusions

Within the global carbon cycle, it has been well established that remineralization of POC and DOC within the estuarine and shelf water column serves as a source for atmospheric  $CO<sub>2</sub>$ . The importance of carbon burial in sediments at the land–ocean interface as a potential sink for atmospheric  $CO<sub>2</sub>$  has not been extensively addressed, because (1) the amount of organic carbon that is input to the system is derived from various sources, including riverine, marine, wastewater, and in situ productivity; (2) the amount of carbon buried at the land–ocean interface is spatially and temporally heterogeneous; and (3) estuaries and other systems at the land–ocean interface have been and continue to be significantly perturbed by urbanization and other human activities. Although each estuarine system (large or small) throughout the world has its own physical, chemical, biological, and geological features and processes, they are all characterized by a two-layer estuarine circulation pattern that promotes the landward transport, trapping, and accumulation of carbon-coated particles and POC at the land–ocean interface.

Our integrative results, spanning over 50 years, indicate that carbon is being rapidly buried in estuarine areas, where the sediment surface is currently out of equilibrium with its physical regime. These non-equilibrium areas can occur naturally, such as in the Yangtze River estuary, where vertical accretion of deltaic topset beds and horizontal progradation of foreset beds can be as high as 20 and 300 m  $year^{-1}$ , respectively. These non-equilibrium areas can also occur anthropogenically, such as in dredged-harbor areas within the Hudson River estuary, where the accumulation of carbon-rich sediments can be as high as  $15-20$  cm year<sup>-1</sup>. The fine-grained and carbon-coated particles that escape estuarine systems are generally transported both *alongshore* on the shelf and *landward* into other protected low-energy coastal areas. For example, the fine-grained sediments escaping the Amazon River delta are generally transported northward and landward, forming muddy beaches and accumulating in protected coastal areas as far north as Venezuela (Smoak [2009\)](#page-17-0). In addition, nutrient-rich estuarine water discharged onto the shelf can stimulate in situ production of POC, which sinks to the bottom and is transported landward back into estuaries or other coastal marginal areas.

Although carbon burial in the world's 25 largest river-estuarine–deltaic systems  $(\sim 0.1 \text{ PgC year}^{-1})$  is small relative to the oceanic sink  $(\sim 2 \text{ PgC year}^{-1})$  (Sabine et al. [2004\)](#page-16-0), these 25 systems only account for a small areal component of the entire world's coastline. Future work involving a global network of estuarine and coastal scientists working in their own backyards may provide a platform for quantifying the extent of carbon burial along the entirety of the world's coastline. It is also important to determine whether carbon assimilation and accumulation in these areas have increased during the past 50 years in association with the increased loadings of agricultural and wastewater derived nutrients.

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