Flux and budget of BC in the continental shelf seas adjacent to Chinese high BC emission source regions

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Key Points:
- The continental shelf sediments represented a dominant BC sink
- High BC concentrations occurred in the fine-grained mud areas
- BC sequestered in sediments significantly affected the global carbon cycle

Supporting Information:
- Readme
- Figures S1–S6, Texts S1–S3, Tables S2 and S3, and Table S1 caption
- Table S1
- Figure S1
- Figure S2
- Figure S4
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Abstract
This study conducted the first comprehensive investigation of sedimentary black carbon (BC) concentration, flux, and budget in the continental shelves of “Bohai Sea (BS) and Yellow Sea (YS),” based on measurements of BC in 191 surface sediments, 36 riverine water, and 2 seawater samples, as well as the reported data set of the atmospheric samples from seven coastal cities in the Bohai Rim. BC concentrations in these matrices were measured using the method of thermal/optical reflectance. The spatial distribution of the BC concentration in surface sediments was largely influenced by the regional hydrodynamic conditions, with high values mainly occurring in the central mud areas where fine-grained particles (median diameters > 6 μΦ (i.e., < 0.0156 mm)) were deposited. The BC burial flux in the BS and YS ranged from 4 to 1100 μg/cm² yr, and averaged 166 ± 200 μg/cm² yr, which was within the range of burial fluxes reported in other continental shelf regimes. The area-integrated sedimentary BC sink flux in the entire BS and YS was ~325 Gg/yr, and the BS alone contributed ~50% (~157 Gg/yr). The BC budget calculated in the BS showed that atmospheric deposition, riverine discharge, and import from the Northern Yellow Sea (NYS) each contributed ~51%, ~47%, and ~2%, respectively. Therefore, atmospheric deposition and riverine discharge dominated the total BC influx (~98%). Sequestration to bottom sediments was the major BC output pattern, accounting for ~88% of the input BC. Water exchange between the BS and the NYS was also an important BC transport route, with net BC transport from the BS to the NYS.

1. Introduction

Black carbon (BC), also termed elemental carbon (EC) and pyrogenic carbon (PyC) [Hammes et al., 2007; Meredith et al., 2012; Wiedemeier et al., 2013, 2015a; Santin et al., 2015], has been of great interest due to its significant influence on regional/global climate change, carbon cycle, air quality, and public health [Kuhlbusch, 1998; Ramanathan and Carmichael, 2008; Anenberg et al., 2012; Wang et al., 2014a; Zong et al., 2015]. BC is the highly condensed carbonaceous residue derived exclusively from the incomplete combustion of organic matter, such as biomass and fossil fuels [Goldberg, 1985; Masiello, 2004; Hammes et al., 2007; Coppola et al., 2014; Gao et al., 2014]. The combustion process and subsequent resistance toward degradation make BC an omnipresent component in environmental matrices, such as the atmosphere, soils, fresh/sea water, ice, and sediments [Goldberg, 1985; Hammes et al., 2007]. Among them, marine sediments are usually regarded as the ultimate sink for land-based BC via atmospheric and riverine transport [Mitra et al., 2013]. It is estimated that more than 90% of the global BC burial occurs on the continental shelf, though the area accounts for only 10% of the world ocean [Suman et al., 1997; Sánchez-García et al., 2012; Mitra et al., 2013].

Because of the recognized importance of the continental shelf as the major BC sink, increasing attention has been paid to the BC flux and budget in this marine regime [Suman et al., 1997; Gustafsson and Gschwend, 1998; Dickens et al., 2004; Elmquist et al., 2008; Flores-Cervantes et al., 2009; Sánchez-García et al., 2012; Mitra et al., 2013; Sánchez-García et al., 2013; Ali et al., 2014]. Studies of the BC flux and budget contribute significantly toward better understanding of the regional/global carbon cycle, which is a hot topic highlighted by...
the International Geosphere-Biosphere Programme and International Human Dimensions Programme on Global Environmental Change [Swaney and Giordani, 2007; von Glasow et al., 2013]. However, a vast majority of the existing studies were geographically within Europe and America, such as the Northern European Shelf [Sánchez-García et al., 2012], the Gulf of Cádiz [Sánchez-García et al., 2013], the Gulf of Maine [Gustafsson and Gschwend, 1998; Flores-Cervantes et al., 2009], and the Washington Coast [Dickens et al., 2004], and little attention was paid to the continental shelf of Asia (especially for East Asia and South Asia), which envelops major BC emission source regions in the world [Bond et al., 2004; Sánchez-García et al., 2012].

China, with a total population of 1.36 billion (http://www.stats.gov.cn/) in East Asia, has been generally considered as the world’s largest BC emitter [Bond et al., 2004]. The North China Plain (Figure 1a), including the five provinces of Hebei, Shandong, Henan, Anhui, and Jiangsu and the two municipalities of Beijing and Tianjin, has the highest BC emission intensity [Cao et al., 2006; Wang et al., 2012, 2014a]. Specifically, the North China Plain contributes ~36% of the China’s total annual BC emission amount in 2000 [Cao et al., 2006], even if the area there is only ~8% of the Chinese territory. Due to a combination of the prevailing East Asian monsoon (together with its derived extreme weather events, like the dust storms) and large amounts of riverine outflow [Jiang et al., 2010; Wang et al., 2014b], a significant amount of BC released from the North China Plain is predictably injected into the adjacent continental shelf seas, the Bohai Sea (BS) and Yellow Sea (YS). Therefore, the BS and YS are probably important reservoirs for BC from the North China Plain.

To date, only very limited studies on sedimentary BC were conducted in the BS and YS continental shelf surface sediments, with the coverage less than 5% of the total area [Kang et al., 2009; Jiang et al., 2010; Fang et al., 2014]. In the context of such distinct regional features (i.e., high BC emission intensity and strong influences from monsoon and riverine outflow), the nearly complete lack of BC data in the BS and YS continental shelf sediments makes large-scale investigations in this region particularly urgent. Therefore, the present study conducted an extensive sediment sampling campaign in the BS and YS continental shelf. Combined with recent extensive investigations of atmospheric BC in the coastal cities from the Bohai Rim (Table 1), riverine water and seawater samples were concurrently collected to calculate the regional BC budget. The major objectives are (1) to detail the spatial distribution and influential factors of the BC concentration in the BS and YS surface sediments, (2) combining with several sediment properties, to calculate the BC burial flux, (3) applying the BC burial flux to the corresponding areal extension, to estimate the area-integrated sedimentary BC sink flux in the sampling regions, and then extrapolate to the entire BS and YS, and (4) to calculate the regional BC budget as a case study of the multimedium investigated semiclosed BS.
Values of Corresponding Parameters for the Calculation of BC Flux From Atmospheric Deposition in the Semiclosed BS

<table>
<thead>
<tr>
<th>Coastal Cities</th>
<th>$v_{d}^{b}$ (cm/s)</th>
<th>$W_{p}^{c}$</th>
<th>$C_{BC-TSP}^{d}$ (μg/m³)</th>
<th>$p_{0}^{e}$ (mm/season)</th>
<th>References $^{g}$</th>
</tr>
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<tbody>
<tr>
<td>Tianjin</td>
<td>0.15</td>
<td>2.0 × 10⁵</td>
<td>9.35</td>
<td>65.0</td>
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<td>Tangshan</td>
<td>–</td>
<td>–</td>
<td>10.0</td>
<td>72.5</td>
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<tr>
<td>Huludao</td>
<td>22.6</td>
<td>5.12</td>
<td>13.0</td>
<td>22.6</td>
<td>Xu [2007]; Wang [2006]</td>
</tr>
<tr>
<td>Dalian</td>
<td>3.78</td>
<td>2.28</td>
<td>19.0</td>
<td>383.3</td>
<td>Fan et al. [2014]; Zhao [2011]</td>
</tr>
<tr>
<td>Tuoji Island</td>
<td>2.96</td>
<td>3.30</td>
<td>6.68</td>
<td>32.0</td>
<td>Wang et al. [2014b]; Han and Cui [2014]</td>
</tr>
<tr>
<td>Changdao Island</td>
<td>2.10</td>
<td>5.76</td>
<td>97.0</td>
<td>32.0</td>
<td>Feng et al. [2007]; Han and Cui [2014]</td>
</tr>
<tr>
<td>Dongying</td>
<td>2.50</td>
<td>–</td>
<td>99.7</td>
<td>66.5</td>
<td>Zong et al. [2015]; Lin et al. [2012]</td>
</tr>
</tbody>
</table>

$^{a}$The locations of these coastal cities from the Bohai Rim are illustrated in Figure 1b. $^{b}$Range of 0.1–0.2 cm/s was widely reported in the Bohai Rim [Jurado et al., 2008; Sun, 2010], and the median value of 0.15 cm/s was used in the present work. $^{c}$Data of $W_{p}$ (particle washout ratio, dimensionless) was cited from Jurado et al. [2008]. $^{d}$Only the Tianjin City conducted by Kong et al. [2010] has the BC concentrations in TSP ($C_{BC-TSP}$, μg/m³), PM$_{10}$ ($C_{BC-PM10}$, μg/m³; PM$_{10}$: particles with an aerodynamic diameter less than 10 μm), and PM$_{2.5}$ ($C_{BC-PM2.5}$, μg/m³; PM$_{2.5}$: Particles with an aerodynamic diameter less than 2.5 μm) phase simultaneously. Other cities (except for Tangshan and Dongying) either have the $C_{BC-PM10}$ or have the $C_{BC-PM2.5}$. Because the calculation of the atmospheric BC depositional flux is the $C_{BC-TSP}$-based (see equations (4)–(7)), one needs to obtain the $C_{BC-TSP}$ in these cities. In this study, we assumed that the ratios of $C_{BC-TSP}/C_{BC-PM10}$ and $C_{BC-TSP}/C_{BC-PM2.5}$ were constant in the coastal cities from the Bohai Rim. Thus, the ratios of $C_{BC-TSP}/C_{BC-PM10}$ and $C_{BC-TSP}/C_{BC-PM2.5}$ measured in Tianjin City by Kong et al. [2010] were used to deduce the $C_{BC-TSP}$ for other cities. $^{e}$Data of $C_{BC-TSP}$ was from the references before the “;” (semicolon), and data of $p_{0}$ was from the references after the semicolon. $^{f}$The endash (–) refers to data not obtained.

2. Materials and Methods

2.1. Sample Collection

A total of 191 surface sediment samples (0–3 cm) from the BS and YS were collected using a stainless steel box corer or grab sampler in three cruises deployed by R/V Ke Yan 59 and R/V Dong Fang Hong 2 during 2006–2011. The whole collection of the sediment samples consists of 89 samples from the BS, 53 samples from the northern YS (NYS), and 49 samples from the southern YS (SYS) (Figure 1a, the label of each sediment site is shown in Figure S1 in the supporting information). After collection, they were wrapped in precombusted aluminum foil (450°C for 4 h) and stored at −20°C until analysis.

Riverine discharge and water exchange between the BS and the NYS are two important processes associated with the BS BC budget. Riverine water samples were collected during the wet season (4–22 August 2013). A total of 36 rivers including the most dominant and concerned Yellow River from the Bohai Rim were selected (Figure 1b), and they accounted for more than 95% of the total riverine water discharge into the BS. For the water exchange between the BS and the NYS, Wei et al. [2003] observed that the water exchange between the BS and the NYS generally showed characteristics with water flowing into the BS through the northern Bohai Strait and flowing out of the BS through the southern Bohai Strait. Thus, the site W1 located to the east of the northern Bohai Strait and the site W2 located to the west of the southern Bohai Strait (Figure 1b) could be potentially used to estimate the BC import flux from the NYS and the BC export flux into the NYS, respectively. Considering that the BC concentration in seawater body differed in different water depths and seasons [Flores-Cervantes et al., 2009], the stratified (surface, middle, and bottom) seawater samples were collected over four seasons during 2010–2011. All the collected riverine water and seawater samples were filtered (0.3–2.1 L per piece of filter) in situ through precombusted (450°C for 4 h) and tared 47 mm diameter quartz fiber filters (Whatman) to obtain the total suspended solids for particulate BC (PBC) analysis.

For atmospheric aerosol samples, extensive investigations have been conducted in the coastal cities from the Bohai Rim (Table 1), and they were cited in the present work to calculate the atmospheric derived BC flux to the BS. The detailed aerosol sampling processes and the subsequent BC analytical procedures were not elaborated here. These coastal cities, including Tianjin, Tangshan, Huludao, Dalian, Tuoji Island, Changdao Island, and Dongying, are geographically encircling the entire BS (Figure 1b). That is, they are to a large extent suitable for approaching a more accurate calculation of the BC flux from atmospheric deposition.
2.2. Sample Analyses

2.2.1. Analyses of the Sedimentary and Particulate BC

A major problem in quantifying BC is that it is not a well-defined chemical component, but rather exists as a complex concept of combustion/temperature continuum, with widely varying physical and chemical characteristics [Hedges et al., 2000; Masiello, 2004; Hammes et al., 2007; Conedera et al., 2009; Wiedemeier et al., 2015b]. This results in various BC analytical methods developed by multidiscipline researchers for specific studies [Gustafsson et al., 1997; Gélinas et al., 2001; Gustafsson et al., 2001; Song et al., 2002; Elmqquist et al., 2004, 2006; Hammes et al., 2007; Han et al, 2007a; Louchouarn et al., 2007; Khan et al., 2009; Poot et al., 2009; Han et al., 2011; Meredith et al., 2012; Wiedemeier et al., 2013]. In the present work, the wet-chemical treatment combined with thermal/optical reflectance (TOR) detection of Han et al. [2011] was adopted. TOR-BC was operationally defined as the pure carbon evolved from the 2% O2/98% He atmosphere when the filter loaded with sediment residue was analyzed on a Desert Research Institute Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA) [Zong et al., 2015]. The PBC in the riverine/seawater total suspended solids was directly determined on the analyzer after they were acidized with hydrochloric acid fumes at least 24 h to thoroughly remove the inorganic carbon. The TOR method can measure the whole BC continuum, ranging from micron-sized char (1–100 μm) to submicron-sized soot particles (<1 μm) [Han et al., 2007b, 2011, 2012]. The BC fractions in μg/cm3 output from the analyzer were normalized to the initial mass of the analyzed dry sediment or to the volume of the filtered riverine/seawater, and they were reported in mg pure C per g dry sediment (mg C/g dry sediment) or μg pure C per liter water (μg C/L water), respectively [Han et al., 2007a, 2007b]. More detailed procedures regarding the BC separation, quantification and quality assurance and quality control (QA/QC) are available in Text S1.

2.2.2. Analyses of the Total Organic Carbon and Sediment Grain Size

The analytical methods of the total organic carbon (TOC) and sediment grain size were described in detail by our previous work [Hu et al., 2009a]. Replicate analyses of one sample (n = 10) gave a precision of ±0.02 wt % for TOC. For the sediment grain size, the relative standard deviation of the replicate samples was less than 3% (n = 6). The particle sizes were less than 4 μm for clay, 4–63 μm for silt, and larger than 63 μm for sand.

2.3. Calculations of the BC Burial Flux and Area-Integrated BC Sink Flux

To calculate the burial flux of organic components in the surface sediments of a given region, several sediment properties must be taken into account, in addition to the organic component (here it is BC) concentration [Sánchez-García et al., 2012]. The BC burial flux \( \dot{F}_{\text{burial}} \) in μg/cm2 yr was calculated by

\[
\dot{F}_{\text{burial}} = 10^3 \cdot C_{\text{BC}} \cdot \rho \cdot \omega \cdot (1 - \phi)
\]

where \( C_{\text{BC}} \) is the measured sedimentary BC concentration (mg/g) (Table S1), \( \rho \) is the dry density of the sediment samples (g/cm3), \( \omega \) is the sedimentation rate (cm/yr), and \( \phi \) is the sediment porosity (dimensionless).

In equation (1), in addition to our measured \( C_{\text{BC}} \), the other three sediment parameters (i.e., \( \rho, \omega, \) and \( \phi \)) should be considered and selected for reasonable values. For \( \phi \), a range of 0.7–0.8 was widely measured in sediment mixing layers around the world [Sánchez-García et al., 2012, 2013; Ali et al., 2014], and a median value of 0.75 ± 0.05 was used here. For \( \rho \) and \( \omega \), however, we did not choose constant values like \( \phi \), due to the significant differences observed among different subregions/sampling sites in our studying area. Thus, designated values of \( \rho \) and \( \omega \) for each sampling subregion/site were essential. Ranges of 0.84–1.77, 0.72–0.98, and 0.60–1.28 g/cm3 were previously determined for \( \rho \) in the BS, NYS, and SYS subregions, and mean values of 1.30, 0.85, and 0.95 g/cm3 were adopted here, respectively [Li and Yuan, 1991; Zhao et al., 1991; Li and Shi, 1995; Li et al., 2006; Hu et al., 2017a]. For \( \omega \), 99 sites with previously accurately dated values were collected in the BS andYS [Hu et al., 2011a, and references therein], and then the Kriging Interpolation method was used to obtain the \( \omega \) spatial distribution (Figure 2d).

The obtained \( \dot{F}_{\text{burial}} \) is then applied to the corresponding areal extension to calculate the area-integrated BC sink flux in Gg/yr (1 Gg = 109 g), following

\[
\dot{F}_{\text{sink}} = 10^5 \cdot \dot{F}_{\text{burial}} \cdot A = 10^{-2} \cdot C_{\text{BC}} \cdot \rho \cdot \omega \cdot (1 - \phi) \cdot A
\]

where, \( A \) is the area of a given region (km2). There are many criteria for choosing this area, such as the bottom water depth, the sediment characteristics, and the distance between the adjacent sampling sites. Because of the densely distributed sediment sampling sites in the present study, the area was selected depending on the
distance between the adjacent sampling sites. The BS, NYS, and SYS sampling regions were divided into 89, S3, and 49 compartments, respectively. Each of the 191 sampling sites was allocated in the center of one compartment, and the area of this compartment was considered as the area of the centered site represented. This criterion has been widely adopted by other researchers to estimate the mass inventory of persistent organic pollutants (POPs) in surface sediments of the East China Seas [Chen et al., 2006; Lin et al., 2009; Qin et al., 2011]. The BC sink flux of the sampling regions in the BS and YS (~60% of the entire BS and YS) was calculated by summing up the 191 individual BC sink flux listed in Table S1, and the calculation of the BC sink flux in no sampling regions (the remaining ~40%) of the BS and YS are detailed in the following section 3.3. and Text S2.

2.4. Calculation of the BC Budget in the BS

Due to our extensive sampling campaign conducted in multimedium (including the sediments, riverine water, and seawater samples) and other elaborated studies of atmospheric BC by other researchers in the Bohai Rim (Table 1), the BS was the target area to calculate the regional BC budget. The BC inputs into the BS include atmospheric dry and wet deposition ($F_{AI}$, Gg/yr), riverine discharge ($F_{IR}$, Gg/yr), and import from the NYS ($F_{IP}$, Gg/yr). The outputs consist of sequestration to bottom sediments ($F_{sink}$), export to the NYS ($F_{EI}$, Gg/yr), and consumption by degradation/transformation during transport. The difference between the inputs and the outputs is the BC internal sink, i.e., net increase in seawater and consumption by degradation/transformation during transport. The difference between the inputs and the outputs is the BC internal sink, i.e., net increase in seawater ($F_{I}$, Gg/yr). Middleburg et al. (1999) found that degradation of up to 50% of the sedimentary BC may take a timescale of ~10,000 years, thus degradation of BC occurred in the 0–3 cm surface sediments (deposited by recent few years to few decades) collected in the present study was likely to be small and was thus neglected here. The BC budget in the BS (expressed by flux, Gg/yr) can be briefly depicted by the following equation:

\[
\text{Inputs} - \text{Outputs} = \text{Internal sink} \Rightarrow (F_{AI} + F_{IR} + F_{I}) - (F_{sink} + F_{EI}) = F_{IW}
\]

2.4.1. Calculation of the $F_{AI}$

The aerosol BC concentrations measured at the seven coastal cities from the Bohai Rim (Figure 1b and Table 1) were used to calculate the atmospheric depositional flux of $f_{A}$ in $\mu$g/m² season was calculated as follows

\[
f_{A} = f_{DD} + f_{WD}
\]

\[
f_{DD} = 7.78 \times 10^{4} \cdot V_{D} \cdot C_{BC-TSP}
\]

\[
f_{WD} = 10^{-3} \cdot p_{0} \cdot W_{P} \cdot C_{BC-TSP}
\]

where $f_{DD}$ is the seasonal dry depositional flux ($\mu$g/m² season), $f_{WD}$ is the corresponding seasonal wet depositional flux ($\mu$g/m² season), $V_{D}$ is the dry deposition velocity of aerosol (cm/s), $C_{BC-TSP}$ is the measured BC concentration in the collected total suspended particulate (TSP) ($\mu$g/m³) during that season, $p_{0}$ is the precipitation rate (mm/season) of that season, $W_{P}$ is the particle washout ratio (dimensionless), and 7.78 $\times$ 10⁴ and $10^{-3}$ are unit conversion factors that convert cm/s to m/season and mm/season to m/season (here we assumed that one season is equal to 90 days), respectively.

The annual atmospheric BC depositional flux of $F_{A}$ in Gg/yr in the entire BS was then calculated by equation (7):

\[
F_{A} = \sum_{i=1}^{4} \left[ F_{AI} \times (7.7 \times 10^{10}) \right] / 10^{15}
\]

where $i = 1–4$ represent spring, summer, autumn, and winter, respectively. $F_{AI}$ is the averaged seasonal atmospheric depositional flux ($\mu$g/m² season) of the seven coastal cities, 7.7 $\times$ 10¹⁰ is the area of the BS (m²) (Hu et al., 2009b), and the denominator 10¹⁵ is the unit conversion factor that converts $\mu$g/yr to Gg/yr. The values of the corresponding parameters for the calculation of the atmospheric BC depositional flux are listed in Table 1.

2.4.2. Calculations of the $F_{IR}$, $F_{IP}$, $F_{EI}$, and $F_{IW}$

$F_{R}$ was calculated by summing up the 36 riverine BC fluxes, with each obtained by multiplying the annual water discharge ($Q_{w}$, m³/yr) (Table 2) by its corresponding PBC concentration ($C_{PBC,TSP}$, $\mu$g/L) (Table S2).

\[
F_{R} = 10^{-12} \cdot \sum_{i=1}^{36} \left[ C_{PBC,TSP} \cdot Q_{w} \right]
\]
3. Results and Discussion

3.1. BC Concentration and BC/TOC Ratio in the BS and YS Surface Sediments

BC was detected in all the BS and YS surface sediments, and its concentration ranged from 0.02 to 3.55 mg/g, with an arithmetic mean value of 0.85 ± 0.57 mg/g. The BS, NYS, and SYS subregions had BC concentration ranges of 0.02–1.75 mg/g, 0.14–3.55 mg/g, and 0.25–2.22 mg/g, with mean values of 0.69 ± 0.40 mg/g, 1.01 ± 0.78 mg/g, and 0.96 ± 0.47 mg/g, respectively (Tables 3 and S1). The mean BC concentrations measured in the BS and YS surface sediments were comparable to those of the previously reported continental shelf marine systems, such as 0.76 ± 0.29 mg/g for the coast of Taiwan [Hung et al., 2003], 0.71 ± 0.23 mg/g for the East China Sea [Hung et al., 2011], 0.65 ± 0.46 mg/g for the Gulf of Maine [Gustafsson and Gschwend, 1998], and 1.10 ± 0.50 mg/g for the coast of Pakistan [Ali et al., 2014]. Besides, they were also comparable to those of the pelagic/deep-sea regimes, which had mean BC concentrations of 0.45–1.62 mg/g [Smith et al., 1973; Middelburg et al., 1999; Lohmann et al., 2009] (Table 3).

Figure 2a shows the spatial distribution of the BC concentrations in the BS and YS. The most remarkable characteristic was the existence of the isolated high BC concentrations (1–2 mg/g) in the central part of each subregion. These central areas were under the influence of cyclonic cold eddies, resulting in the deposition of large amounts of fine-grained sediment particles with median diameters >6 μm (i.e., <0.0156 mm) (Figure 2c) and thus the formation of mud patches (Figure 1a) [Zhu and Chang, 2000; Li et al., 2005]. Therefore, BC with high fine-particle affinity could cotransport to such areas and was ultimately sequestered in the weak hydrodynamic sedimentary environment. Some POPs, such as OCPs (organochlorine pesticides) and PAHs (polycyclic aromatic hydrocarbons), have been documented with high
concentrations in these areas as well [Hu et al., 2009b, 2011b; Lin et al., 2011; Qin et al., 2011]. The similar spatial distribution of BC and POPs in these areas suggested that BC might strongly affected the distribution and transport of these POPs, which was consistent with the results found by Hung et al. [2011] in East China Sea surface sediments. In contrast to the high BC concentrations, low BC concentrations (<0.5 mg/g) occurred in the eastern BS (i.e., the Laotieshan Channel in the northern Bohai Strait), the eastern NYS, the Haizhou Bay, and the southwestern SYS. Those areas were mainly characterized by sandy and even gravel/sandy gravel sediments (Figure 2c), due to strong tidal currents with relatively high energy [Zhu and Chang, 2000]. Such hydrodynamic condition is not conducive toward the deposition and the subsequent sequestration of micron-/submicron-sized BC particles. Based on the above discussions, it can be inferred that the hydrodynamic transport and depositional mechanisms are key factors affecting the spatial distribution of the BC concentration in the BS and YS continental shelf. This can be also demonstrated by the significant positive correlations between the BC concentration and the median diameters (MD) of grain size (Table S1) in both the three subregions (r = 0.66–0.72, p < 0.01) and the total area (r = 0.60, p < 0.01) (Figure 3).

Apart from the hydrodynamic conditions, riverine discharge is an extra factor influencing the distribution of the BC concentration. Recently, Jiang et al. [2010] found that riverine discharge was the main source of BC for the intertidal zones and nearshore areas of the Bohai Bay (Figure 1b). The delivered BC was then confined within the Bohai Bay and became of high concentration there due to the weak water exchange. Unlike in the Bohai Bay, riverine discharge did not lead to such high BC concentration in the Yellow River Estuary of

Figure 2. Spatial distribution of the (a) measured BC concentration, (b) BC burial flux, (c) median diameters (MD) of grain size, and (d) collected sedimentation rate in the Bohai Sea and Yellow Sea continental shelf surface sediments.
<table>
<thead>
<tr>
<th>Study Area</th>
<th>n</th>
<th>Range</th>
<th>Mean ± SD</th>
<th>C&lt;sub&gt;BC&lt;/sub&gt; (mg/g)</th>
<th>Range</th>
<th>Mean ± SD</th>
<th>C&lt;sub&gt;TOC&lt;/sub&gt; (mg/g)</th>
<th>BC/TOC Ratio (%)</th>
<th>Range</th>
<th>Mean ± SD</th>
<th>F&lt;sub&gt;burial&lt;/sub&gt; (μg/cm&lt;sup&gt;2&lt;/sup&gt; yr)</th>
<th>Mean ± SD</th>
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<td>Bohai Sea</td>
<td>89</td>
<td>0.02-1.75</td>
<td>0.69 ± 0.40</td>
<td>0.47-13.74</td>
<td>3.81</td>
<td>2.19</td>
<td></td>
<td>0.8-34.2</td>
<td>19.7</td>
<td>8.2</td>
<td>10-1100</td>
<td>260</td>
<td>± 252</td>
<td>TOR</td>
</tr>
<tr>
<td>Northern Yellow Sea</td>
<td>53</td>
<td>0.14-3.55</td>
<td>1.01 ± 0.78</td>
<td>1.34-10.86</td>
<td>5.20</td>
<td>2.76</td>
<td></td>
<td>4.3-45.2</td>
<td>19.8</td>
<td>8.4</td>
<td>4-412</td>
<td>100</td>
<td>± 84</td>
<td>TOR</td>
</tr>
<tr>
<td>Southern Yellow Sea</td>
<td>49</td>
<td>0.25-2.22</td>
<td>0.96 ± 0.47</td>
<td>1.24-15.67</td>
<td>4.76</td>
<td>2.65</td>
<td></td>
<td>4.4-38.9</td>
<td>22.8</td>
<td>10.1</td>
<td>6-276</td>
<td>67</td>
<td>± 51</td>
<td>TOR</td>
</tr>
<tr>
<td>Yellow Sea</td>
<td>102</td>
<td>0.14-3.55</td>
<td>0.99 ± 0.65</td>
<td>1.24-15.67</td>
<td>4.99</td>
<td>2.72</td>
<td></td>
<td>4.3-45.2</td>
<td>21.2</td>
<td>9.4</td>
<td>4-412</td>
<td>84</td>
<td>± 72</td>
<td>TOR</td>
</tr>
<tr>
<td>Bohai Sea and Yellow Sea</td>
<td>191</td>
<td>0.02-3.55</td>
<td>0.85 ± 0.57</td>
<td>0.47-15.67</td>
<td>4.43</td>
<td>2.55</td>
<td></td>
<td>0.8-45.2</td>
<td>20.5</td>
<td>8.9</td>
<td>4-1100</td>
<td>166</td>
<td>± 200</td>
<td>TOR</td>
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<tr>
<td><strong>Previous Studies</strong></td>
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<tr>
<td>Continental Shelf Regimes</td>
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<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Coast of Taiwan, China</td>
<td>35</td>
<td>0.36-1.90</td>
<td>0.76 ± 0.29</td>
<td>1.04-17.10</td>
<td>6.33</td>
<td>3.67</td>
<td></td>
<td>5.1-39.8</td>
<td>15.3</td>
<td>8.0</td>
<td>267-8067</td>
<td>1103</td>
<td></td>
<td>CTO-375</td>
</tr>
<tr>
<td>East China Sea, China</td>
<td>13</td>
<td>0.33-1.16</td>
<td>0.71 ± 0.23</td>
<td>0.70-6.11</td>
<td>2.85</td>
<td>1.52</td>
<td></td>
<td>14.1-47.1</td>
<td>29.0</td>
<td>9.1</td>
<td>86-190</td>
<td>131</td>
<td>± 38</td>
<td>CTO-375</td>
</tr>
<tr>
<td>Gulf of Maine, USA</td>
<td>10</td>
<td>0.11-1.73</td>
<td>0.65 ± 0.46</td>
<td>1.5-22.5</td>
<td>12.7</td>
<td>6.6</td>
<td></td>
<td>3.1-14.6</td>
<td>5.7</td>
<td>3.5</td>
<td>86-190</td>
<td>131</td>
<td>± 38</td>
<td>CTO-375</td>
</tr>
<tr>
<td>Swedish Continental Shelf</td>
<td>120</td>
<td>0.58-17.66</td>
<td>2.41 ± 2.18</td>
<td>4.8-168.0</td>
<td>49.1</td>
<td>28.2</td>
<td></td>
<td>1.7-47.1</td>
<td>4.6</td>
<td>2.0</td>
<td>267-8067</td>
<td>1103</td>
<td></td>
<td>CTO-375</td>
</tr>
<tr>
<td>Gulf of Cádiz, SW Spain</td>
<td>15</td>
<td>0.10-1.10</td>
<td>0.40 ± 0.30</td>
<td>0.50-14.0</td>
<td>8.8</td>
<td>2.6</td>
<td></td>
<td>2.0-15.0</td>
<td>5.1</td>
<td>3.4</td>
<td>42-285</td>
<td>120</td>
<td>± 76</td>
<td>CTO-375</td>
</tr>
<tr>
<td>Gulf of Cádiz, SW Spain</td>
<td>15</td>
<td>0.10-2.30</td>
<td>0.90 ± 0.80</td>
<td>0.50-14.0</td>
<td>8.8</td>
<td>2.6</td>
<td></td>
<td>0.9-34.0</td>
<td>11.0</td>
<td>10.0</td>
<td>21-633</td>
<td>245</td>
<td>± 205</td>
<td>BPCA</td>
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<tr>
<td>Coast of Pakistan</td>
<td>285</td>
<td>0.60-2.50</td>
<td>1.10 ± 0.50</td>
<td>2.8-28.6</td>
<td>11.7</td>
<td>6.4</td>
<td></td>
<td>5.3-28.8</td>
<td>12.2</td>
<td>7.5</td>
<td>183-766</td>
<td>330</td>
<td>± 155</td>
<td>CTO-375</td>
</tr>
<tr>
<td>Pelagic Regimes</td>
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<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pacific Ocean</td>
<td>24</td>
<td>0.01-0.99</td>
<td>0.45 ± 0.31</td>
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<td>n/a</td>
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<td></td>
<td>n/a</td>
<td>n/a</td>
<td></td>
<td></td>
<td>0.002</td>
<td>± 0.2</td>
<td>H&lt;sub&gt;2&lt;/sub&gt;O&lt;sub&gt;2&lt;/sub&gt;</td>
</tr>
<tr>
<td>South Atlantic Ocean</td>
<td>23</td>
<td>0.40-1.70</td>
<td>1.02 ± 0.26</td>
<td>2.50-44.70</td>
<td>14.1</td>
<td>12.3</td>
<td></td>
<td>2.4-34.7</td>
<td>13.2</td>
<td>9.2</td>
<td>0.50-7.80</td>
<td>22.7</td>
<td>± 1.73</td>
<td>CTO-375</td>
</tr>
<tr>
<td>Madeira Abyssal Plain</td>
<td>5</td>
<td>0.74-2.85</td>
<td>1.62 ± 0.85</td>
<td>1.82-11.39</td>
<td>6.09</td>
<td>4.36</td>
<td></td>
<td>21-40</td>
<td>31.8</td>
<td>7.6</td>
<td>n/a</td>
<td>n/a</td>
<td>CTO-375</td>
<td>Middelburg et al. [1999]</td>
</tr>
</tbody>
</table>

*SD refers to standard deviation.
*This column shows the BC quantification methods used in these studies: TOR = thermal/optical reflectance, CTO-375 = chemical thermal oxidation at 375°C for 14–24 h, BPCA = benzene polycarboxylic acids (molecular marker method), and H<sub>2</sub>O<sub>2</sub> = peroxide oxidation.
*n/a refers to not analyzed.
the Laizhou Bay (Figures 1b and 2a), although the Yellow River delivered large quantities of sediments to the BS [Wang et al., 2007]. Sediments in the Yellow River Estuary were mainly derived from the low-populated and thus low-polluted loess plateau, with a previously measured BC concentration in the loess as low as 0.10–0.15 mg/g (using the same TOR method as the present study) [Han et al., 2007b]. Moreover, the lower reaches of the Yellow River are distinguished by the “aboveground river,” meaning that its riverbed is higher than the ground elevation outside banks [Hu et al., 2009b]. BC particles driven by surface runoff cannot be easily transferred from ambient farmlands and cities into the Yellow River. As a consequence, the large flux of “clean” terrestrial sediments could result in a dilution effect on BC and other trace components (such as OCPs and PAHs) in sediments at the Yellow River Estuary [Hu et al., 2009b; Qin et al., 2011].

In the BS and YS surface sediments, the TOC concentration varied from 0.47 to 15.67 mg/g, and averaged 4.43 ± 2.55 mg/g. Because BC is a purely terrestrial component, extracting BC from the TOC of the marine sediments would promote a more accurately evaluation of the marine primary productivity [Verardo, 1997]. In the present study, BC accounted for 1–45% of the TOC, with a mean value of 20.5 ± 8.9% (Tables 3 and S1). The spatial distribution of the BC/TOC ratio resembled that of the BC concentration, with high values also mainly occurring in the central mud areas (Figure S2). The range of the BC/TOC ratio obtained in the BS and YS continental shelf was also comparable to that for the other continental shelf and pelagic regimes, which reported ranges of 1–47% and 2–40%, respectively (Table 3). Such a high BC/TOC ratio indicated that the terrestrial BC indeed constituted a significant fraction of the sedimentary TOC pool, which might disturb our understanding of the true marine organic carbon burial.

3.2. BC Burial Flux in the BS and YS Surface Sediments

Compared with the BC concentration, the BC burial flux was more favorable because it was not affected by the varying inputs and thus the dilution effect of the major detrital matter [Elmquist et al., 2007, 2008; Sánchez-García et al., 2012]. The BC burial flux in the BS and YS ranged from 4 to 1100 μg/cm² yr, and averaged 166 ± 200 μg/cm² yr. The relatively high relative standard deviation (~120%) suggested that the BC burial flux in the BS and YS had a large spatial differentiation (Figure 2b). For the subregions, the BS had the highest BC burial flux, reaching up to 260 ± 252 μg/cm² yr, followed by that in the NYS (100 ± 84 μg/cm² yr) and the SYS (67 ± 51 μg/cm² yr) (Tables 3 and S1).

Similar to the spatial distribution of the BC concentration, high BC burial flux (>100 μg/cm² yr) also occurred in the central fine-grained mud areas (Figure 2b), manifesting the role of shelf mud depositional process on

![Figure 3](image-url)

Figure 3. Correlation between the BC concentration and the median diameters (MD) of grain sizes in both (a–c) the subregions and (d) the total area.
the fate of BC. But particularly notable was the differences observed between them. Specifically, as indicated by the two cyan arrows in Figure 2b, the BC burial flux rather than the BC concentration decreased seaward with increasing distance from the coastline in the BS and NYS. Such a decreasing seaward pattern for the BC burial flux resulted, to a large extent, from the significant influence of the outflow by the land-based sources, as clearly evidenced by the same decreasing seaward pattern for the sedimentation rates (Figure 2d). Additionally, the Yellow River Estuary had the lowest BC concentration but the highest BC burial flux, with the mean value exceeding 600 μg/cm² yr, 4 times higher than that of the rest of the BS (~150 μg/cm² yr). Thus, it was obvious that the Yellow River played an important role in delivering BC to the BS (see section 3.4.), and the observed threefold higher BC burial flux in the BS (260 ± 252 μg/cm² yr) than that in the YS (84 ± 72 μg/cm² yr) might be largely attributed to the abundant input by the Yellow River.

The mean BC burial flux in our studying area (166 ± 200 μg/cm² yr, TOR) was slightly higher than those in the Gulf of Maine (131 μg/cm² yr, CTO-375) [Gustafsson and Gschwend, 1998], but nearly a magnitude lower than that in the extensive investigated Swedish Continental Shelf (1103 μg/cm² yr, CTO-375) [Sánchez-García et al., 2012]. Although nearly a magnitude discrepancy was observed among these typical continental shelf regimes, the BC burial fluxes there were still 2–4 orders of magnitude higher than that in the pelagic regimes, such as 0.09 μg/cm² yr (H₂O₂) for the Pacific Ocean and 2.27 μg/cm² yr (CTO-375) for the South Atlantic Ocean [Smith et al., 1973; Lohmann et al., 2009] (Table 3). Such low BC burial fluxes in pelagic sediments were mainly resulted from the extremely low sedimentation rates (<10 cm/kyr), which was a key parameter involved in the BC burial flux calculation. The overwhelmingly high BC burial fluxes in the continental shelf regimes over the pelagic regimes indicated the prominence of the continental shelf as the major BC repository [Gustafsson and Gschwend, 1998]. Therefore, to better understand the global BC budget or cycle, future investigations should focus more on the continental shelf regimes, such as the present BS and YS, as well as the world’s largest continental shelf system “the Arctic” [Sánchez-García et al., 2012].

Despite the above comparison conducted, however, it was worth noting that different methods used by the researchers might result in the various BC burial fluxes. This has been demonstrated by a method comparison study conducted by Sánchez-García et al. [2013], who found that the BPCA-based BC burial flux in the Gulf of Cádiz (southwest Spain) surface sediments varied between 21 and 633 μg/cm² yr (mean, 245 ± 205 μg/cm² yr) and the CTO-375-based BC burial flux was relatively smaller, varying from 42 to 285 μg/cm² yr (mean, 120 ± 76 μg/cm² yr) (Table 3). Therefore, the comparison of the BC burial flux would make more sense when considered the different methods used. This implied that developing a standard BC analytical method to a single sediment medium would be of great significance. But as we know, it is still a great challenge because of the varying physical-chemical properties of the BC combustion continuum [Hedges et al., 2000; Masiello, 2004; Hammes et al., 2007; Conedera et al., 2009; Wiedemeier et al., 2015b].

3.3. BC Sink Flux in the Entire BS and YS

The area-integrated sedimentary BC sink flux in the entire BS and YS was the sum of the BC sink flux in regions with sampling and regions with no sampling, with each accounting for ~60% (~271,100 km²) and ~40% (~186,900 km²) of the total area (~458,000 km²). The BC sink flux of the sampling regions was calculated by summing up the BC sink flux values of the 191 sampling sites (Table S1). For the latter, due to the lack of investigations, the BC concentration could only be deduced from our present work. The rationality for the BC concentrations inference and the calculations of the BC sink fluxes in these no sampling regions are detailed in Text S2 and Table 4. According to the calculations, the BC sink flux in the entire BS and YS was estimated to be ~325 Gg/yr. For the subregions, the BC sink fluxes were sorted in the order of BS (~157 Gg/yr) > SYS (~127 Gg/yr) > NYS (~41 Gg/yr). Although the area of the BS (~77,000 km²) was only one fifth that of the YS (~381,000 km²), the BS and YS had a similar BC sink flux, suggesting that the source intensity of BC input into the BS was significantly higher than that of the YS.

It should be noted that there is still some uncertainty associated with the calculation of the sedimentary BC sink flux. Specifically, the uncertainty in the calculated BC sink flux was predominantly based on the errors in the measured CBC (0–13%), referenced ρ (9–22%), and φ (7%) values. As illustrated in Text S3, an uncertainty of ~26% was determined based on the propagation of the errors in these three parameters. The BC sink flux
was also widely reported by researchers in some other marine regimes. For instance, Gustafsson and Gschwend [1998] and Lohmann et al. [2009] reported that the BC sink fluxes were 400–800 Gg/yr (CTO-375) in the New England continental shelf off the northeastern USA (380,000 km2) and ~480–700 Gg/yr (CTO-375) in the total South Atlantic Ocean (~40,838,000 km2), respectively. In addition, a similar value of ~300 Gg/yr (CTO-375) and much lower value of ~13–26 Gg/yr (CTO-375 and BPCA) were separately found in the Swedish Continental Shelf (155,342 km2) and the Gulf of Cádiz (10,788 km2) [Sánchez-García et al., 2012, 2013].

Because the BC sink flux was proportional to the calculated area (see equation (2)), a comparison among different regions would make sense when accounting for the varying accumulation areas. When considered, the BC sink fluxes in the continental shelf regimes were of similar magnitude but significantly higher than those of the pelagic regimes.

Despite the observed differences among various regions in terms of BC sink flux, BC accumulation in marine sediments has an identical effect. That is, BC sequestered in marine sediments transfers carbon from the fast-cycling bioatmospheric carbon cycle (i.e., more labile organic carbon) into the slow-cycling geological carbon cycle (i.e., more refractory organic carbon), at least on timescales of hundreds to thousands of years [Lohmann et al., 2009]. Thus, the formation of BC disturbs the global carbon budget and balance [Masiello, 2004]. With regard to the present study concretely, the well-calculated BC sink flux combined with the extensive investigations of BC in other closely related multimedium (including the atmosphere, riverine water, and seawater) makes it possible for us to calculate the regional BC budget and then better understand the regional BC source-to-sink processes and geochemical behavior.

3.4. Calculation of Regional BC Budget: A Case Study of the Semiclosed BS

The BC budget calculated in the BS is clearly shown in Figure 4. The total BC flux into the BS was ~182 Gg/yr, with riverine discharge, atmospheric deposition, and import from the NYS each accounting for ~85 Gg/yr, ~93 Gg/yr, and ~4 Gg/yr. The riverine discharge contributed ~47% of the total BC input. Of particular note was that the Yellow River alone contributed ~82% (~70 Gg/yr) of the total riverine BC influx. A similar proportion was also obtained with the PAHs by Xia [2007], who found that in summer the Yellow River alone contributed ~75% of the total riverine PAHs influx to the BS. The similar proportion of the Yellow River-derived BC and PAHs in the total riverine influx suggested that BC and PAHs in this region were mainly derived from coemission sources, or the released PAHs could be adsorbed onto BC particles during transport, which were consistent with the results found by Hung et al. [2011] in East China Sea surface sediments.

Atmospheric deposition, including wet deposition and dry deposition, was of comparable importance (~51%) as riverine discharge. The atmospheric deposition showed strong seasonal variations (Figures S3–S5). Specifically,

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Table 4. Calculations of the Area-Integrated Sedimentary BC Sink Flux in the Entire Bohai Sea and Yellow Sea Continental Shelf Surface Sediments

<table>
<thead>
<tr>
<th>Studying Area</th>
<th>A (km²)</th>
<th>Fsink (Gg/yr)</th>
<th>Sediment Types</th>
<th>A (km²)</th>
<th>Mean CBC (mg/g)</th>
<th>Mean ω (cm/year)</th>
<th>Fsink (Gg/yr)</th>
<th>A (km²)</th>
<th>Fsink (Gg/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bohai Sea</td>
<td>61,800</td>
<td>133</td>
<td>STY</td>
<td>7,600</td>
<td>0.83</td>
<td>0.57</td>
<td>11.7</td>
<td>77,000</td>
<td>157</td>
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<td></td>
<td></td>
<td>YT</td>
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<td>6.0</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>YS</td>
<td>2,100</td>
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<td>3.1</td>
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<td></td>
<td></td>
<td>TY</td>
<td>1,700</td>
<td>1.10</td>
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<td>3.5</td>
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<td></td>
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<tr>
<td>Northern Yellow Sea</td>
<td>53,000</td>
<td>39</td>
<td>FS</td>
<td>14,400</td>
<td>0.24</td>
<td>0.13</td>
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<td>71,000</td>
<td>41</td>
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<tr>
<td></td>
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<td></td>
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<td>2,300</td>
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<td>0.6</td>
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<td></td>
<td></td>
<td>MFS</td>
<td>1,300</td>
<td>0.24</td>
<td></td>
<td>0.1</td>
<td></td>
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</tr>
<tr>
<td>Southern Yellow Sea</td>
<td>156,300</td>
<td>102</td>
<td>FS</td>
<td>69,100</td>
<td>0.25</td>
<td>0.12</td>
<td>4.9</td>
<td>310,000</td>
<td>127</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>STY</td>
<td>53,800</td>
<td>0.93</td>
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<td>14.3</td>
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<td>MFS</td>
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<td>0.24</td>
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<td>1.15</td>
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<td>5.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>S</td>
<td>15,400</td>
<td>0.15</td>
<td></td>
<td>0.7</td>
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</tr>
<tr>
<td>Entire Bohai Sea and</td>
<td>271,100</td>
<td>274</td>
<td>STY</td>
<td>186,900</td>
<td>0.50</td>
<td></td>
<td>50.7</td>
<td>458,000</td>
<td>325</td>
</tr>
<tr>
<td>Yellow Sea</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

aSediment types are referred to Li et al. [2005]. STY, YT, YS, TY, FS, MFS, and S stand for sediments of sand-silt-clay, clayey silt, clayey sand, silty clay, fine sands, middle fine sands, and sands, respectively.

bThe selected values of ω are referred to in Text S2.

The BC sink flux in the entire Bohai Sea and Yellow Sea is calculated as the sum of the BC sink flux in these three subregions.
among the total ~93 Gg/yr of BC, the spring, summer, autumn, and winter season each accounted for ~20 Gg (~22%), ~40 Gg (~43%), ~18 Gg (~19%), and ~15 Gg (~16%) (Figure S4). It was obvious that the summer season alone contributed nearly half of the annual atmospheric derived BC to the BS, which was to a large extent attributed to the overwhelming high precipitation occurred in summer (mean, ~400 mm) (Table 1). As shown in Figure S5, the wet deposition accounted for ~90% (~36 Gg) of the summer seasonal BC flux, and the dry deposition contributed only the remaining ~10% (~4 Gg). Additionally, the slightly higher BC flux from spring might relate to the regional prevailing extreme weather events, like the dust storms, which has been observed in Changdao Island (Figure 1b) [Feng et al., 2012].

The atmospheric deposition and riverine discharge two BC input patterns dominated ~98% of the total BC influx, and the BC import from the NYS (~2%) could be considered negligible. As a typical shallow continental shelf sea strongly affected by riverine discharge, such a high proportion of BC derived from atmospheric deposition could be attributed to the prevailing winter and summer East Asian monsoon [Feng et al., 2012; Wang et al., 2014b], which passes through the Chinese high BC emission source regions (i.e., the North China Plain) [Wang et al., 2012]. Lin et al. [2011] recently found that the source pattern of the pyrogenic 4–6 ring PAHs in the BS surface sediments correlated well with the PAHs emission feature of the aerosol PM$_{2.5}$ (i.e., particles with an aerodynamic diameter less than 2.5 μm) samples collected from the upwind Beijing (Figure 1a), also demonstrating the significant influence of atmospheric deposition on the input of combustion-derived substances into the BS.

As for the outputs, ~86% (~157 Gg/yr) of the input BC was sequestered to bottom sediments, so sequestration was the major BC output pattern. The semiclosed (resulting in weak water exchange) and shallow (mean water depth of 18 m) characteristics of the BS [Hu et al., 2009b] as well as the short residence time of BC particles in the continental shelf water column (~2 months) [Flores-Cervantes et al., 2009] might be responsible for such high BC burial/sequestration potential in the BS sediments. Based on the estimated surface-ocean export flux, Flores-Cervantes et al. [2009] found that the sediments in the Gulf of Maine captured ~80% of the BC in that body of seawater, which was well in agreement with the present study. These two studies both revealed the “sink” role of the continental shelf sediments for terrestrial BC preservation. In addition to sequestration, export to the NYS was also an important route for the BS BC output, accounting for ~12% (~21 Gg/yr) of the total BC influx. When compared with the BC flux import from the NYS and export to the NYS, the net BC transport flux over the Bohai Strait reached up to ~17 Gg/yr and was from the BS to the NYS (Figure 4), which was consistent with the net particle transport pattern over the Bohai Strait obtained by the grain size trend analysis [Cheng et al., 2004]. Because the net transport particles were mainly the Yellow River fine particles, the consistent transport pattern indicated that BC was likely to be adsorbed onto fine particles, and suspended fine particles in the seawater matrix probably acted as an important carrier phase for BC transport in the marine systems. BC transport between the BS and the NYS also indicated that seawater was also an important BC reservoir that should receive coequal attention along with several known BC reservoirs (such as the atmosphere, soils, and sediments) [Jaffe et al., 2013; Masiello and Louchouarn, 2013]. The net increase of the BC flux in seawater (~4 Gg/yr) accounted for ~2% of the total BC influx, implying that BC concentration in the BS seawater was at a state of relatively stabilized equilibrium or saturated.

There are also several studies of the BC budget conducted in geographically separated continental shelf regimes, such as the New England Continental Shelf, the Gulf of Maine, and the Northern European Shelf [Gustafsson and Gschwend, 1998; Flores-Cervantes et al., 2009; Sánchez-García et al., 2012]. All these studies found that the sedimentary soot-BC sinks were of the same magnitudes as with estimates of the upwind
atmospheric deposition/emission of soot-BC. It is worth noting that the above direct comparison was complicated for several aspects. For instance, different BC quantification methods were used with regard to the BC sources (TOR and sinks (CTO-375)). In addition, there was a lack of consideration for the additional riverine input of soil soot-BC that remained on the land. Despite of these facts, these studies all clearly demonstrated the significant roles of atmospheric deposition on BC delivery from sources to sinks and the deposition of continental shelf sediments for terrestrial BC preservation, which matched well with the present findings.

4. Conclusions

This study conducted the first comprehensive investigation of BC in multimedium to constrain the sedimentary BC concentration, burial flux, sink flux, and budget in the BS and YS continental shelf regimes, which are adjacent to Chinese high BC emission source regions (i.e., the North China Plain). The major conclusions include the following:

1. High BC concentrations mainly occurred in the central fine-grained mud areas, while low values took place in the sandy and even gravel/sandy gravel regions. This indicated that the spatial distribution of the BC concentration was largely influenced by the regional hydrodynamic conditions.
2. The BC burial flux in the BS and YS ranged from 4 to 1100 μg/cm² yr, and averaged 166 ± 200 μg/cm² yr, which within the burial fluxes reported in other shelf regimes, but they were 2–4 orders of magnitude higher than that of the pelagic/deep-sea regimes, indicating the prominence of the continental shelf as the major BC sink. The BC burial flux exhibited a distinct decreasing seaward pattern with increasing distance from the coastline in the BS and NYS, which reflected the significant influence of the outflow of land-based sources.
3. The area-integrated sedimentary BC sink flux in the entire BS and YS was ~325 Gg/yr, with the BS, NYS, and SYS subregion each accounting for ~157 Gg/yr, ~41 Gg/yr, and ~127 Gg/yr. BC sequestered in these marine regimes transfers carbon from the fast-cycling bioatmospheric carbon cycle into the slow-cycling geological carbon cycle and thus disturbs the regional and even global carbon budget and balance.
4. The BC budget was calculated in the semiclosed BS. The total BC flux into the BS was ~182 Gg/yr. Of which, riverine discharge, atmospheric deposition, and import from the NYS contributed ~85 Gg/yr, ~93 Gg/yr, and ~4 Gg/yr, respectively. Atmospheric deposition and riverine discharge played comparable importance in delivering BC to the BS, and they dominated ~98% of the total BC input. As for the outputs, sequestration to bottom sediments was the major BC output pattern, accounting for ~86% of the input BC. The water exchange between the BS and the NYS resulted in ~17 Gg/yr of BC net transporting from the BS to the NYS.

Acknowledgments

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