The silica-carbon biogeochemical cycle in the Bohai Sea and its responses to the changing terrestrial loadings

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Abstract

Silicon (Si) and carbon (C) play key roles in the river and marine biogeochemistry. The Si and C budgets for the Bohai Sea were established on the basis of measurements at a range of stations and additional data from the literature. The results show that biogenic silica (BSi) and total OC in sediments are mainly from marine primary production. Benthic diffusion contributes 49% of total reactive Si input to the Bohai Sea, and smaller amounts come from groundwater (26%), riverine input (17%), surface runoff (7%) and atmospheric deposition (< 1%); the dominant DSi removal from the water column is sedimentation (99%). Rivers contribute 43% of exogenous OC inputs to the Bohai Sea, followed by benthic flux (29%), atmospheric deposition (12%), submarine groundwater discharge (11%), and surface runoff (5%); the dominant outputs of OC are sedimentation (65%) and export to the Yellow Sea (35%). The net burial of BSi and OC represent 3.3% and 1.0% of total primary production, respectively. Primary production has increased by 10% since 2002 as a result of increased river loads of DSi and BSi. Our findings underline the critical role of riverine Si supply in primary production in coastal marine ecosystems.

Key words: Bohai Sea; dissolved silicate; biogenic silica; organic carbon; flux and budget; primary production

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1. Introduction

Diatoms control a large part of primary production in marine ecosystems, up to 50% in the global ocean and more than 75% in coastal waters (Nelson et al., 1995; Rousseaux and Gregg, 2014). The consumption of dissolved silicate (DSi) and production of biogenic silicon (BSi) is mainly controlled by primary production by diatoms (Ragueneau et al., 2000; Tréguer and De La Rocha, 2013).

Although ocean margins cover only 8% of the global ocean area (Berner, 1982), the production and accumulation rates of BSi and organic carbon (OC) in these areas are significantly higher than in the open ocean (Hedges and Kiel, 1995; Tréguer and De La Rocha, 2013). Rivers are the dominant Si and OC source in coastal marine ecosystems, accounting for up to 80% of total exogenous input (Bauer et al., 2013; Regnier et al., 2013; Tréguer and De La Rocha, 2013). However, large parts of the world's coastal marine ecosystems have been changing due to decreasing riverine Si discharge as a result of Si trapping in reservoirs (Humborg et al., 2000; Bernard et al., 2010). The decreasing input of Si may lead to a shift from a system dominated by diatoms to one dominated by non-siliceous phytoplankton (Humborg et al., 1997, 2000; Tréguer and De La Rocha, 2013; Rousseaux and Gregg, 2015), which may influence the functioning of coastal marine ecosystems as a biological pump, especially with respect to the carbon (C) cycle.

The Bohai Sea is a semi-enclosed, shallow shelf water body of the North-western Pacific Ocean, with a surface area of 77,300 km^2 and an average depth of 18 m. A large number of rivers drain into the Bohai Sea, typically with densely populated and industrialized coastal areas. The Yellow River is the largest river draining into the Bohai Sea. Ongoing human activities (dam construction, agriculture and industry) have induced significant changes in the river discharge, sediment load and nutrient concentrations (Gong et al., 2015; Liu, 2015).

A substantial decrease of Si/N ratio in the Bohai Sea is attributed to the reduction of

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the Yellow river discharge (Ning et al., 2010) and dam construction (Liu, 2015; Ran et al., 2015). The water residence time in the Bohai Sea is about 3 years (Liu et al., 2012). Changes of nutrient inputs from rivers in the semi-enclosed Bohai Sea therefore have larger and more long-lasting influence on the ecosystem than in open seas. These processes have changed primary production and phytoplankton composition in the Bohai Sea (Tang et al., 2003; Lin et al., 2005; Ning et al., 2010; Fu et al., 2016). Phytoplankton abundance in the Bohai Sea had decreased in the period of 1959–1999 (Tang et al., 2003) and dominant species succession from diatoms to non-diatoms had also been found in the 1980s and 1990s (Lin et al., 2005). However, the primary production has been increasing (Tan et al., 2011) recently with the progressing of water and sediment regulation in the Yellow River.

There may be a close connection between the changes in nutrient loading and primary production in coastal marine ecosystems (Bernard et al., 2011). The C cycle in shelf seas is also sensitive to changing riverine loading due to anthropogenic perturbations (Li et al., 2014; Woodland et al., 2015), but unfortunately these studies did not include the effects of changing riverine Si export on the C cycle. Most importantly, the water and sediment regulation of the Yellow River since 2002 has greatly changed the nutrient input to the Bohai Sea in the past years, resulting in a large proportion of total annual DSi (30-60%), DOC (36%) and POC (86%) input to the Bohai Sea concentrated during the water-sediment regulation during June-July and a slight increase of the annual fluxes (Wang et al., 2012; Gong et al., 2015; Liu et al., 2015). Thus, the magnitude of nutrient input under a dam-orientated artificial regulation within a short period would have a large influence on the ecosystem, which is an important difference with other coastal systems with non-regulated rivers.

Our understanding of the regional coupled Si-C cycle and ecological effects of changing river loadings in the continental shelves of eastern China is poor. In this paper we establish a reactive Si and organic C budget for the Bohai Sea to analyze the coupled Si-C biogeochemistry; the aim is to quantify the influence of changing

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terrestrial loadings on the Si and C cycles and primary production in the Bohai Sea.

2. Materials and methods

2.1 Sampling and analytical methods

2.2.1 Sample collection and pretreatment

Two campaigns were carried out in spring (May 3 to 24) and autumn (November 2 to 20) of 2012 at several sampling stations in the Bohai Sea and the adjacent area of the Northern Yellow Sea (Figure 1). The spring cruise contained 25 stations while the fall cruise contained 24 stations. Water samples in surface (0.5 m) and bottom water (< 2m from the sea floor) were collected using an oceanography water sampler (Seabird 911 CTD Plus, Sea-Bird Electronics, Bellevue, WA, USA). Ancillary parameters such as temperature and salinity were recorded on board simultaneously. Also, surface (0-1 cm) sediment and core sediment samples (between 20 and 50 cm long) were collected (Figures 1b and 1c) in part of the stations. Sampling expeditions were also carried out at the Lijin Station (Shandong Province) in the Yellow River (Figure 1a) during a full hydrological year in 2013-2014. Water samples were collected once per month at 20 cm below the surface with at least 3 sampling points across the river main channel.

Water samples were filtered with 200 µm Nylon sieves to remove zooplankton, subsequently filtered with 0.45 µm polyethersulfone filters. Filters were pretreated according to the following four steps: cleaned with 1:1000 HCl for 24 h; rinsed with Milli-Q water to achieve a neutral pH; oven-dried at $45^{o}C$ for 72h; weighed after cooling in a dryer with desiccant. Then filters with particulate matter were stored at $-20^{o}C$ for determination of suspended particulate matter (SPM) and BSi, and filtrates were stored at $4^{o}C$ after adding drops of chloroform for determination of DSi. In addition, the pre-weighed water samples were filtered with 0.70 µm GF/F glass-fiber filters (Whatman, Maidstone, UK), which were also pre-cleaned according to the

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following four steps: 1) cleaned with 1:1000 HCl for 24 h; 2) rinsed with Milli-Q water to achieve a neutral pH; 3) burned at 450°C for 4h; 4) weighed after cooling in a dryer with desiccant. The filters were stored at -20°C for determination of suspended particulate organic carbon (POC), and filtrates were stored at -20°C for determination of dissolved organic carbon (DOC) in a later stage. Surface sediment samples (0-1 cm) were collected with a box sediment sampler after removing the overlying water, and then packed into sealed bags and frozen at -20 °C for determination of BSi and total organic carbon (TOC). At the same time, sediment core samples were collected using a sampling tube with an inner diameter of 9 cm at some stations (Figure 1b). Cores were divided into 1 cm intervals after overlying water was collected using syringes (13 mm 0.45 μm, PTFE) with needle tubing. The pore water of each subsample was separated by centrifugation and preserved as above for DSi analyses; finally, subsamples were stored at -20°C before BSi and TOC analysis in a later stage. Replicants (*n*=2) of water samples and sediment samples were chosen from 20% of stations in this study; and the results showed no significant difference between these replicants (p<0.01).

2.2.2 Laboratory analyses

DSi was analyzed with a QuAAtro Autoanalyzer, using the silicomolybdic blue method, with a detection limit of 0.030 μmol L^{-1} and a relative standard deviation < 0.3% (*n*=5). BSi in SPM was extracted by NaOH solution (0.2 mol L^{-1}, 100 °C, 40 min) and corrected for mineral interferences using the Si:Al ratios (Ragueneau et al., 2005), while the BSi content in sediment was measured using the alkaline extraction method (1% Na_2CO_3, 85 °C, extraction during 8 hours, during which the extract is sampled and analyzed every hour) (DeMaster, 1981), with a measurement uncertainty of 0.25% and relative standard deviation < 0.3% (*n*=5). Reactive silicon (RSi) is the sum of DSi and BSi. DOC was determined using a high-temperature catalytic oxidation technique (Zhang et al., 2013) with a TOC analyzer (TOC-C_{CPH}, Shimadzu, Japan); the relative standard deviation is < 2% (*n*=5). For POC determination, 3-5

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drops of 2 mol L^{-1} HCl were added to the sample filters in a closed container with HCl fumes for 24 h to remove inorganic carbon, and then dried at 45°C (Zhang et al., 2013). Subsequently, POC was determined with an elemental analyzer (Euro Vector EA3000, Via Tortona, Milan, Italy) with standard deviation < 10% (*n*=5). TOC in sediments was analyzed with the same elemental analyzer. Before measurement, freeze-dried sediment samples were decalcified using 4 mol L^{-1} HCl and subsequently rinsed with de-ionized water (6–8 times) to achieve a neutral pH, and then pretreated sediments were dried overnight at 60 °C (Hu et al., 2009) for TOC determination. Replicate analyses of one TOC sample (*n*=6) provided a precision of ±0.02 %. DSi was calibrated against nutrient standard sample (Second institute of Oceanography, State Oceanic Administration, China), while TOC was calibrated against Reference Standard Materials (EuroVector, Code E11001).

The software of Surfer 11.0 (Golden Software, inc. USA) and Origin 8.5 (OriginLab Corporation, USA) were used for mapping the concentration patterns in the Bohai Sea.

2.2 Water budget

The water budget of the Bohai Sea (Figure 2) provides the basis for the calculation of the Si and C budgets. The hydrography of the Bohai Sea is largely determined by the Bohai Sea Coastal Current (BSCC) and exchange with the Yellow Sea. River discharge, precipitation, submarine groundwater discharge, surface runoff and evaporation are taken into account in the water budget calculation for the shelf in steady state as follows:

$$Q_R + Q_P + Q_{YTB} + Q_{GW} + Q_{SR} = Q_{BTY} + Q_{EVA} \qquad (1)$$

Where Q are water fluxes ($km^3\ yr^{-1}$), subscripts R, A, YTB, GW, SR, BTY and EVA denote the river discharge, atmospheric deposition, Yellow Sea inflow, submarine groundwater, surface runoff , Bohai Sea outflow and evaporation, respectively (Table

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1, Figure 2). The estimated river input is about 34 km^3 yr^{-1} for the 6 major rivers discharging into the Bohai Sea (Table 1), and precipitation and evaporation amount to 44 km^3 yr^{-1} and 85 km^3 yr^{-1}, respectively, based on Martin et al. (1993) and Lin et al. (2001). The water flux from the Bohai to Yellow Sea (BTY) is 470 km^3 yr^{-1}, and with a reverse flux (YTB) of 442 km^3 yr^{-1} there is a net export 28 km^3 yr^{-1} from the Bohai Sea to the Yellow Sea (Liu et al., 2003a). The submarine groundwater input is about 44 km^3 yr^{-1} based on estimates of submarine groundwater discharge in the Yellow River delta (Peterson et al., 2008). The budget yields an estimate for surface runoff (Q_{SR}) of 1.1 km^3 yr^{-1}, which includes the discharge by small streams not included in the above river discharge. Therefore, the water residence time in the Bohai Sea is about 2.5 years, which is close to the 3 years estimated by Liu et al. (2012).

2.3 Budget of reactive silica and organic carbon

The Si and C budgets of the Bohai Sea are estimated using a steady-state box model, focusing on the reactive Si (RSi, the sum of DSi and BSi) and OC in the water column and accounting for the major hydrological, chemical and biological processes. In this calculation, we use estimates for the fluxes of RSi and OC into and out of the Bohai Sea, i.e. exchange through the Bohai Strait (F_E; F_E = Input to the Bohai Sea (F_{YTE}) - Output to the Yellow Sea (F_{BTY})), riverine input (F_R), surface runoff (F_{SR}) from surficial runoff and small rivers not included in F_R, submarine groundwater discharge (F_{GW}), atmospheric input (F_A), flux from porewaters (F_B) and sedimentation (F_S) (Table1).

Internal processes such as primary production (F_P), regeneration (F_{RC}), respiration and degradation are also taken into account. F_E, F_B, and F_S are based on measurements described in this paper, as well as the major contributions of Yellow River to F_R, while the other fluxes are based on literature values. The various budget terms are discussed in more detail below.

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2.3.1 Riverine input

We estimated the DSi, BSi, DOC and POC fluxes for 6 major rivers discharging into the Bohai Sea (Table 1 and Figure1). Fluxes are calculated with long-time monitoring data. On the basis of monthly data for BSi, POC and SPM, we found that the fraction of BSi and POC in the suspended solids increases exponentially and linearly with SPM concentration (Figure S1), respectively. The Yellow River is the largest river in North China and had been playing a controlling role in the formation of North China Plain, where many studied rivers flow. Assuming that BSi and POC have the same source in general river water (Ran et al., 2015), we use the sediment content and BSi and POC concentrations for the Yellow River to develop regression equations that can be used to calculate BSi and POC concentrations for rivers with missing data:

$$C_{BSi} = a \times C_{TSS}^{\ b} \qquad (r = 0.823, p < 0.001) \qquad (2)$$

$$C_{POC} = c \times C_{TSS} + d \qquad (r = 0.984, p < 0.001) \qquad (3)$$

Where C_{BSi} and C_{POC} represent the BSi and POC concentration in the river (μmol L^{-1}), respectively; C_{TSS} represents the sediment content in the river (mg L^{-1}); a, b, c and d are constants, a (μmol mg^{-1}) = 0.22435; b (unitless) = 0.69235; c (μmol mg^{-1}) = 0,13287; d (μmol L^{-1}) = 243.11. The average ratio DOC: POC in the Yellow River was 0,24, so DOC in other rivers without available data can be estimated from the POC concentration.

2.3.2 Atmospheric deposition

Atmospheric input to the Bohai Sea was calculated from the DSi concentration in precipitation (Martin et al., 1993; Zhang et al., 2004), dry deposition (Zhang et al., 2004), combined with the area of the Bohai Sea. The POC in the air mainly occurs in the particulate matter with grain size < 2.5μm (Chen et al., 1997) and the deposition rate of aerosol is about 0.001 m s^{-1} (Duce et al., 1991); the POC concentration in aerosol in the Bohai Sea was from the base station of Chang island in the Bohai Sea

Strait (Feng et al., 2007), and DOC in the snow and rainfall is assumed equal to the average yearly value based on monthly data from 2014 from Yantai (a city near the Chang island) connected to the southern Bohai Sea (Xu et al., 2016). Rainfall and aerosols have low BSi concentrations and can be neglected as sources (Tréguer and De La Rocha, 2013).

2.3.3 Exchange between the Bohai and Yellow Seas

Water exchange between the Bohai and Yellow Sea is driven by the BSCC in the southwest of Bohai Sea and Yellow Sea Warm Current (YSWC) in the Northern Yellow Sea (Figure 1, Table 1). The RSi and OC fluxes through the Bohai Strait were calculated using the water flux together with the measured RSi and OC concentration data from the Southern Bohai Sea and the Northern Yellow Sea (Table 1). DSi concentrations are from this study for May and November 2012 and Yang et al. (2014) for June and July 2013; BSi concentration data are from this study for May and November 2012; DOC are from this study for May and November 2012 and Zhao et al. (2015) for September 2010; and POC are from this study for May and November 2012 and Shang et al. (2011) for September 2010.

2.3.4 Benthic flux at the sediment-water interface

The benthic flux of DSi at the sediment-water interface was calculated based on Fick's first law (Berner, 1980) according to:

$$J_F = -\varphi \times D_s \times (\partial C / \partial z) \qquad (4)$$

$$D_S = D_0 \times \varphi^{(m-1)} \qquad (5)$$

Where J_F represents the diffusion rate (mmol m^{-2} d^{-1}); φ is the porosity of the sediment (dimensionless, 0.72–0.85, based on Liu et al. (2003b)); D_s is the diffusion coefficient in sediment (m^{-2} d^{-1}); C is the concentration (mmol L^{-1}), z is the depth (m), $\partial C/\partial z$ is the concentration gradient of DSi at the sediment-water interface; D_0 is the molecular

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diffusion coefficient of solute in infinitely diluted solutions (m^{-2} d^{-1}, Li and Gregory (1974)); m is an empirical coefficient (dimensionless, for $\varphi \leq 0.7$, $m = 2$; for $\varphi > 0.7$, $m = 2.5–3.0$) (Ullman and Aller 1982). In addition, the diffusive flux of DSi in the Bohai Sea is also coupled with our study (n=4) and results (n=23) from Liu et al. (2011) for improving our estimation of benthic process (Table 1).

As no direct measurement data are available, we estimated the DOC flux from the benthic flux of DSi to the water column based on the molar ratio of BSi : TOC in surface sediments of 0.56.

2.3.5 Sedimentation

Sedimentation of BSi and OC in the Bohai Sea were calculated from the accumulation rate and the surface area of the sea floor in the Bohai Sea (Ingall and Jahnke, 1994; Liu et al., 2005). Although the sea floor of is not flat, the variation of water depth in most of the Bohai Sea is not large and we assume that the surface area (77300 km^2) is equal to the seafloor area . The accumulation rates were based on the following equations:

$$R_{BSi} = C_{BSi} \times MAR / 28 \quad (6)$$

$$R_{OC} = C_{TOC} \times MAR / 12 \quad (7)$$

where R_{BSi} and R_{OC} represent for the accumulation rates of BSi and OC (mol m^{-2} yr^{-1}); C_{BSi} and C_{TOC} represent for the BSi and TOC content in surface sediments (%); MAR is the mass accumulation rate of the sediment (g m^{-2} yr^{-1}); 28 and 12 are the molar weight of Si and C, respectively.

2.3.6 Submarine groundwater discharge and surface runoff

The submarine groundwater DSi flux into the Bohai Sea was calculated from the water flux obtained from ^{228}Ra and ^{226}Ra mass balance models (Peterson et al., 2008)

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and the DSi concentration in groundwater (Lin et al., 2011). As there are no data on DOC input to the Bohai Sea via submarine groundwater, we assumed that the DOC concentration in submarine groundwater equals to that in rivers based on Barrón et al. (2015).

Similar to the water budget, DSi and OC input from surface runoff and rivers not included in the large river inputs (Table 1) were obtained as a result of the budget calculation.

2.3.7 Primary production

Primary production was estimated from the average primary production in the euphotic layer, obtained by integrating the seasonal data from 1998 to 2008 estimated for the total area of the Bohai Sea by satellite remote sensing technology calibrated against measured in-situ productivity (Tan et al., 2011), in which the chlorophyll-a concentrations and primary production were calibrated against measured data, and results are close to the data from Fu et al. (2016). The rates of DSi uptake by phytoplankton and BSi regeneration rate were calculated using the Redfield ratio (C: Si=106: 15, atom basis, Brzezinski, 1985), which is also close to the molar ratio of BSi: POC (0.12) in the suspended particulate matter in the Bohai Sea from our measured results; OC respiration was calculated according to Wei et al. (2004), who demonstrated that respiration accounted for 78% of primary production in the Bohai Sea.

3. Results

3.1 Distribution of RSi and OC in the water column and sediment

The average DSi concentration in fall exceeds that in spring (Table 2). In spring, DSi concentrations are fairly low in the northeast part of the Bohai Sea, but high in the

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Bohai Strait and Yellow River estuary. In autumn, DSi concentration in coastal areas of the Bohai Sea, especially for the Laizhou Bay and northern part of Bohai Sea, exceeds that in other areas (Figure S2).

The BSi concentration in fall exceeds that in spring by a factor of four (Table 2). In spring, surface water BSi concentrations are fairly high near the Bohai Bay, Laizhou Bay and Yellow River estuary, and lower in the central part of the Bohai Sea. Relatively high BSi concentrations occur in the central area of the Bohai Sea in the bottom water. In autumn, the BSi concentrations in the Yellow River estuary and other nearshore areas exceed those in the central zone (Figure S2).

The average DOC concentration in fall is slightly lower than that in spring (Table 2). The spatial distributions of DOC are fairly high concentrations in the nearshore and low ones in the offshore areas (Figure S2). The POC concentrations in spring are close to those in fall (Table 2), with fairly high concentration in the western part of the Bohai Sea and the Yellow River estuary (Figure S2).

The average BSi content in surface sediments is 0.4% and the TOC content is 0.3% (Table 3). The spatial patterns of BSi and TOC are high in the mud area of the Bohai Sea and the area adjacent to the Yellow River estuary (Figure S3).

3.2. Budget of RSi and OC in the Bohai Sea

The estimated riverine RSi and OC fluxes are, respectively, 5.0 Gmol yr^{-1} and 39.6 Gmol yr^{-1}, and DSi and DOC account for 54% and 19% of total RSi and OC fluxes, respectively. The estimated deposition flux of RSi in the Bohai Sea is 0.2 Gmol yr^{-1}, primarily (90%) from wet deposition. OC from atmospheric deposition is 10.7 Gmol yr^{-1}, with an important contribution (84%) from wet deposition.

The inputs of RSi and OC from the Yellow Sea into the Bohai Sea are 2.8 Gmol yr^{-1}

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and 100 Gmol yr^{-1}, respectively, while the output fluxes of RSi (3.1 Gmol yr^{-1}) and OC (132 Gmol yr^{-1}) from the Bohai Sea to the Yellow Sea. Thus, net outputs of RSi and OC from Bohai Sea are 0.3 Gmol yr^{-1} and 32 Gmol yr^{-1}, respectively.

Internal cycling of Si and OC are important terms in the budget. Based on primary production in the euphotic zone of the Bohai Sea, C sequestration is 3280 Gmol yr^{-1}, which means that 460 Gmol yr^{-1} of BSi is ingested according to the Redfield ratio, and 2560 Gmol of OC (78%, see 2.3.7) is consumed by respiration. The estimated sedimentation fluxes of BSi and OC are 30 and 60 Gmol yr^{-1}, respectively. Recycling of BSi in the water column amounts to 430 Gmol yr^{-1} of DSi released. Biodegradation and photo oxidation of OC in the water column is about 3220 Gmol yr^{-1}.

The benthic fluxes of DSi and DOC at the sediment-water interface are further important sources of respectively 15 Gmol yr^{-1} and 27 Gmol yr^{-1}. The calculated submarine groundwater discharge into the Bohai Sea (Table 1) amounts to 8.0 Gmol yr^{-1} for DSi and 9.8 Gmol yr^{-1} for DOC. The estimated surface runoff fluxes are 2.1 Gmol yr^{-1} for RSi and 4.9 Gmol yr^{-1} for OC according to Equation (1).

4. Discussion

4.1 Factors controlling RSi and OC in the water column

The DSi distribution is largely affected by the circulation system in the area adjacent to the Bohai Strait. The high DSi concentration in the southeastern part of the Bohai Sea is due to high DSi in the water mass coming in from the Northern Yellow Sea through the Bohai Strait. The DSi distribution is also influenced by the terrestrial input, particularly in the area near the mouth of the Yellow River (Figure S2).

The BSi concentration in the Bohai Sea is similar to that in the other parts of the Eastern China Sea (Liu et al., 2005). BSi contributes to 30% of RSi in the Bohai Sea,

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which is lower than in the Yellow River water (52%, Ran et al., 2015) but higher than that in the Changjiang Estuary (8%; Gao et al., 2013). The rivers draining into the Bohai Sea carry abundant BSi and have a large influence on the composition of RSi. The main reason why BSi in the bottom water exceeds that in the surface water in parts of the Bohai Sea is largely due to sediment resuspension (Liu et al., 2005) and diatom growth after deposition (Wei et al., 2008).

DOC is the dominant form of OC (89%) in the Bohai Sea, which is a little lower than that in the world's oceans (>95%; Reeburgh et al., 1997). The similar spatial distributions of DOC and POC in the Bohai Sea indicates that both are affected by the same processes, such as input from land by rivers, primary production, biological action, sediment resuspension and many other factors. A significant negative correlation between POC concentrations and salinity (r = −0.430, $p < 0.05$, in spring; r = −0.348, $p < 0.01$, in autumn) indicates that POC is largely determined by the terrestrial input in the coastal areas.

The average molar Si : C ratio of BSi and POC in the Bohai Sea of 0.12 is close to that of diatoms in coastal waters (0.13±0.05, Brzezinski, 1985), meaning that BSi is mainly from marine primary production by diatoms. The C : N atomic ratio in SPM ranges from 1 to 10, with an average value of 5, indicating that OC also originates from marine phytoplankton production. This is consistent with the results from Jiaozhou Bay (Liu et al., 2008a) and East China Sea (Liu et al., 2005).

4.2 Factors controlling RSi and OC in sediments

BSi and TOC concentrations are not different among seasons at the 95% confidence level based on the stations at the same position in spring and fall (Figure S3). The BSi content of surface sediments in the Bohai Sea is similar to that in the continental shelves in Eastern China (Liu et al., 2009), but lower than in the northwestern Indian Ocean (Koning et al., 1997), Southern Ocean (Van Cappellen and Qiu, 1997) and the

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equatorial Pacific Ocean (Piela et al., 2012).

High concentrations of both BSi and TOC concentrations in the mud area of the Bohai Sea (Figure S3) suggest that the sediment grain size and hydrodynamic setting have an important influence on the preservation of BSi. BSi content in the sediment is much lower than that in SPM (0.1%-3.0%, average 0.8%), which indicates that BSi in particles has been degraded during sedimentation and burial. Meanwhile, the average Si : C ratio in sediments of 0.56 is much higher than that in suspended particulate matter. This confirms that degradation rate of OC in the ocean is faster than that of BSi (Ragueneau et al., 2000) due to the lower preservation efficiency of autogenetic OC than that of autogenic BSi (Muller-Karger et al., 2005; Tréguer and De La Rocha, 2013).

4.3 Budget of RSi and OC in the Bohai Sea

The RSi budget shows that the benthic flux across the sediment-water interface is the major source of reactive Si in the Bohai Sea water mass, contributing 49% of the total RSi input (Table 1, Figure 3). The next largest source is submarine groundwater, comprising 26% of total inputs. The river input accounts for 17%, and all other inputs are minor (surface runoff, 7%; atmospheric deposition, < 1%). The dominant output fluxes of RSi in the water column is sedimentation, contributing 99% of total RSi removal, while export to the Yellow Sea only accounts for only 1%.

Overall, considering all exogenous input of OC into the Bohai Sea, riverine flux alone accounts for 43%, followed by the benthic flux of DOC, accounting for 29%; the remaining 28% is from atmospheric deposition (12%), submarine groundwater discharge (11%), and surface runoff (5%). The dominant outputs of OC in the Bohai Sea is sedimentation (65% of total output), and the outflow to the Yellow Sea 35%.

The BSi share in river export in total RSi of 46% is much higher than the average

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value for global rivers (15%) (Laruelle et al., 2009). POC comprises 81 % of the riverine OC, which also exceeds the average for world rivers of 40% (Hedges et al., 1997). The Yellow River export to the Bohai Sea is 68% of total exogenous input for RSi and 73% for OC.

The water exchange between the Bohai and Yellow Seas has only a minor influence on the budget of RSi and OC; however, it has an important effect on the distribution, transport, transformation and retention time of RSi and OC.

The benthic recycling of Si in the sediment is a particularly important flux into the DSi pool in the water column, which confirms earlier studies (Van Cappellen et al., 1997; Tréguer and De La Rocha, 2013). The diffusive DSi flux from sediment stems mainly from the BSi dissolution. DSi concentration gradients in the pore water at all studied stations show a diffusion flux from sediment to water column. The diffusion rates vary from 0.38 to 0.62 mmol m^{-2} d^{-1}, similar to previously reported data (Liu et al., 2011). The high benthic flux plays an important role in maintaining the level of primary production in the water column and also results in a concentration gradient, with higher DSi concentration in bottom than in surface waters. The primary production also depends on the euphotic layer depth, vertical mixing and presence of nutrients in the water column, which cause seasonal variation of primary production and RSi and OC in the Bohai Sea.

The DOC in pore water is also an important source of DOC in the water column (Burdige et al., 1999; Barrón et al., 2015). The estimated DOC flux from sediment based on the assumed BSi : TOC ratio of 0.56 is the second largest external source in the Bohai Sea, showing a similar magnitude to riverine loading, which is also found in many coastal areas (Barrón and Duarte, 2015). Another way to estimate the benthic DOC flux is by assuming DOC diffusion rates to be similar to those in bare sediments (0.9 mmol m^{-2} d^{-1}) (Burdige et al., 1999). This yields a DOC flux of 26 Gmol yr^{-1}, which confirms our estimate (27 Gmol yr^{-1}).

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According to the difference between the diffusive flux of DSi and sedimentation of BSi, the net burial flux of BSi is 15 Gmol yr^{-1}, which is 3.3% of the total primary production which exceeds the average value for the world ocean (2.6%) (Tréguer and De La Rocha, 2013). The gross burial efficiency of BSi is 50% in the Bohai Sea, which is in the range of estimated values for the East China Sea (36−97%, Liu et al., 2005) and exceeds the average of 17−20 % in the world's oceans (Bernard et al., 2010; Tréguer and De La Rocha, 2013). The net burial of OC is 33 Gmol yr^{-1} or 1.0% of the primary production, which is also much higher than the 0.3% estimated for the world ocean (Muller-Karger et al., 2005). This indicates that the Bohai Sea is a potential sink for both Si and C.

Our estimates for submarine groundwater and river Si inputs agree with previous study showing that Si inputs from submarine groundwater are 1–2 times higher than those associated with river discharge into the Bohai Sea (Liu et al., 2011). This agrees with data for the Yellow Sea where estimated submarine groundwater DSi and riverine RSi inputs are respectively 4–24 Gmol yr^{-1} and ~23 Gmol yr^{-1} (Kim et al., 2005) and with data for the Mediterranean Sea where estimated submarine groundwater and river inputs are 110 Gmol yr^{-1} and 30-40 Gmol yr^{-1}, respectively (Rodellas et al., 2015). The DOC input from submarine groundwater is also comparable to riverine input, indicating that submarine groundwater is also an important source of DOC in the Bohai Sea.

4.4 The uncertainty of budget

We assess the uncertainty of the different sources as the standard deviation of each flux estimate divided by the sum of all Si inputs (30.3 Gmol yr^{-1}) to the Bohai Sea, i.e. $F_R+F_{SR}+F_A+F_E+F_B+F_{GW}$. Since the relative uncertainties of all flux estimates are comparable, the absolute uncertainty is largest for the largest flux. The results of the model simulation show that the diffusive flux is the most important flux in the Si

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budget of the Bohai Sea. This flux is calculated on the basis of pore water DSi concentration in the upper layer sediment and overlying water. The large spatial variability of DSi concentrations causes an uncertainty in the estimate of the benthic flux; we based our estimated absolute uncertainty of 8.5 Gmol yr^{-1} on the standard deviation in the DSi concentration data. This yields an uncertainty of 28% by dividing by the total Si inputs (8.5/30.3), hereinafter). Another uncertainty of the Si budget is associated with the large spatial and temporal variability of RSi concentrations in the southern Bohai Sea and northern Yellow Sea; this uncertainty is at most ~11% (3.3/30.3).

The uncertainty of riverine input is less than 3% (0.9/30.3) of total Si input. It should be pointed out that the measurement of RSi alone may underestimate or overestimate the flux of silica from rivers because it does not include amorphous silica debris (such as aluminosilicate). Dissolution and reverse weathering processes in the estuaries impact the Si budget particularly in the long term (Tréguer and De La Rocha 2013).

The uncertainty of submarine groundwater input is less than 10% (1.8/30.3) even if the groundwater discharge varies from 40 to 47 km^3 yr^{-1}. There are also uncertainties in the atmospheric deposition and surface runoff. However, the contributions of these two processes to the Si budget are small. For example, doubling the assumed atmospheric deposition of RSi, from 0.2 to 0.4 Gmol yr^{-1}, would increase the total input by only < 1% (0.2/30.3).

For uncertainties of OC fluxes we use the same approach, with total inputs amounting to 92 Gmol yr^{-1}. Also, the water exchange between Bohai and Yellow Sea is the most important process influencing on the OC budget, which is affected by the variability of DSi concentrations and currents between southern Bohai Sea and northern Yellow Sea. However, the concentrations of OC in southern Bohai Sea and northern Yellow Sea are relatively close to each other, resulting in small influence on the net OC input. The uncertainty of the diffusive flux of OC calculated on the basis of pore water OC

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concentration in the upper sediment layer and overlying water is 16% (15/92). The estimated uncertainty caused by the spatial and temporal variability of OC in rivers draining into the Bohai Sea is < 10% (9.1/92). The uncertainties associated with atmospheric deposition and submarine groundwater discharge are both less than 10% (6.1/92, 2.0/92), while the spatial variation of TOC content and mass accumulation rate cause an uncertainty of 14% (13/92).

4.5 Response of primary production to changing riverine RSi transport

The DSi concentration in the Bohai Sea had decreased in the period 1980–1990, and has been stable more recently. The average DSi concentration in the Bohai Sea in the early 2000s was only 1/3 of that in 1980s (Tang et al., 2003; Ning et al., 2010; Liu et al., 2011; Liu, 2015). Meanwhile, the DIN concentration increased from 1.7 μmol L^{-1} in the 1980s (Tang et al., 2003) to 5.1 μmol L^{-1} in 2000 (Li et al., 2003) and 10.6 μmol L^{-1} in 2012. Nutrient stoichiometry has changed significantly with molar Si : N ratios varying from 14 in 1980s to 1.5 in 2000 and 0.6 in 2012, respectively. The Bohai Sea has therefore changed from an N limited ecosystem in the 1980s to a Si limited system in recent years, and the BSi production by diatoms now largely depends on available Si (Tang et al., 2003; Ning et al., 2010; Liu et al, 2011).

The Yellow River discharge represents more than 70% of the total freshwater discharge into the Bohai Sea (33 km^3 yr^{-1}, Table 1). Since the water residence time in the Bohai Sea is about 3 years (Liu et al., 2012), changes of riverine Si input in the Bohai Sea would have long-lasting influence on the ecosystem's functioning. Statistical analysis suggests that there is a significant relationship between DSi and RSi flux of the Yellow River in year n-1 and primary production in the Bohai Sea in year n (DSi: $p < 0.005$; RSi: $p = 0.02$) (Figures 4 and 5) reflecting the long residence time of Si. This also suggests that changing terrestrial Si loadings have a direct and long-time influence on primary production in the Bohai Sea.

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Since 2002, the water discharge and sediment load of the Yellow River have increased significantly compared with the late 1990s due to the water and sediment regulation (Figure 4). The monthly DSi flux of Yellow River in July increased 5–10 fold and RSi flux by a factor of 3 since 2002; since the sediment load and water discharge regulation in spring has led to peak events (Figure 4) (Gong et al., 2015; Liu, 2015). Using a factor of 3 increase of RSi river export (from 0.9 prior to 2002 to 4.3 G mol yr^{-1} at present, see Figure 4) with the regression equation in Figure 5 results in an increase of primary production by 10% since 2002 in comparison with the levels in 2000 and 2001. This is confirmed by DOC increasing in the Bohai Sea from 2.1 mg L^{-1} before (Zhang et al., 2006) to 2.6 mg l^{-1} (Chen, 2013) and 3.9 mg L^{-1} (this study) after the Yellow River water-sediment regulation in spring, the TOC concentrations in the Bohai Sea have been increasing in the same period, which indicates that increasing Si loadings may enhance both TOC and DOC levels in the Bohai Sea, particularly in the part close to the river mouth.

5. Conclusions

The distributions of RSi and OC in the Bohai Sea show seasonal and regional variation, and are mainly affected by the riverine input, primary production and water exchange between the Bohai Sea and Yellow Sea. BSi and TOC are mainly from marine primary production, and areas with high BSi and TOC contents in the sediments are mainly in the estuarine and mud areas.

The benthic diffusion in the Bohai Sea is the major source of external Si to the water column, accounting for 49% of the exogenous Si inputs, followed by the submarine groundwater discharge (26%), riverine input (17%), surface runoff (7%), and atmospheric deposition (< 1%). The dominant removal processes of RSi in the Bohai Sea is the BSi sedimentation (99% of total output), and the outflow of RSi to the Yellow Sea accounts for 1%.

The riverine flux contributes 43% of all exogenous OC input to the Bohai Sea, followed by benthic flux of DOC, accounting for 29%; atmospheric deposition (12%), submarine groundwater input (11%), and surface runoff (5%) represents the remaining 28%. The dominant outputs of OC in the Bohai Sea is sedimentation (65% of total output), and outflow to the Yellow Sea contributes 35%. The Bohai Sea is a sink for both Si and C, net burial of BSi and OC in sediments amounting to 3.3% and 1.0% of primary production, respectively.

DSi in the Bohai Sea had decreased and then maintained stable in the last three decades. Earth surface process modified by human activities and riverine load variations change the exogenous Si input and thus primary production. Based on the relation between RSi and primary production in year 2000-2008, primary production in the Bohai Sea has increased by 10% since 2002, as a result of the increasing riverine RSi input from the Yellow River due to water-sediment regulation.

A quantitative mechanistic understanding of the key processes controlling Si flow and preservation of C in the land–ocean continuum is needed. The mechanistic understanding is necessary to parameterize the various processes involving C and Si and their sensitivity to external perturbations at the larger scales of earth system models. At present, this lack of understanding limits our ability to predict the present and future contribution of the aquatic continuum fluxes to the global C and Si budget.

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Appendix A. Supplementary data

Supplementary data to this article, including Figure S1, include the relationships of BSi and POC versus to TSS in the Yellow River; Figure S2, Spatial distributions of DSi (a-d), BSi (e-h), DOC (i-l) and POC (m-p) in the Bohai Sea in surface and bottom water for spring and fall in 2012. Figure S3, Distributions of total OC (a–b) and BSi (c–d) in the surface sediment of the Bohai Sea.

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Table 1. Main fluxes of reactive silica and organic carbon budget in the Bohai Sea.

Flux	Parameters to estimate Si-C fluxes							
Riverine input (F_R)	Water discharge km^3 yr^{-1}	Sediment content mg L^{-1}	DSi µmol L^{-1}	BSi µmol L^{-1}	DOC µmol L^{-1}	POC µmol L^{-1}	RSi flux Gmol yr^{-1}	OC flux Gmol yr^{-1}
Yellow River	22.4 [1]	6868 [1]	81.4±24.7 [2]	70.0±16.3 [2]	252±71 [3]	1034±336 [4]	3.39±0.90	28.8±9.1 [4]
Daliao River	4.31 [1]	1592 [1]	52.7 [5]	37.0	109	455	0.39	2.27
Shuangtaizi River	3.95 [5]	5063 [5]	105 [5]	82.3	220	916	0.74	4.5
Hai River	0.90 [1]	88.9 [1]	29.3 [6]	5.0	61.2	255	0.03	0.28
Luan River	0.60 [7]	100 [7]	87.2 [5]	5.4	61.4	256	0.06	0.19
Daling River	2.06 [8]	8592 [8]	77 [5]	119	332	1385	0.40	3.54
Sum							5.0±0.9	39.6±9.1

Flux								
Atmospheric deposition (F_A)	Precipitation km^3 yr^{-1}		Si concentration in the rainfall µmol L^{-1}	Dry deposition rate of Si µmol m^{-2} yr^{-1}	DOC concentration in the rainfall µmol L^{-1}	Dry deposition rate of POC mol m^{-2} yr^{-1}	RSi flux Gmol yr^{-1}	OC flux Gmol yr^{-1}
	33.9 [9]		4.1±0.9 [10]	72.3 [10]	266±149 [11]	0.02±0.01 [12]	0.2±0.1	10.7±6.1

Flux								
Primary production (F_P)					Primary productivity of carbon g m^{-2} yr^{-1}	Primary productivity of BSi mol m^{-2} yr^{-1}	Si fixed by primary production Gmol yr^{-1}	C fixed by primary production Gmol yr^{-1}
					509±38 [13]	6.0±4.5	460±35	3280±250

Flux								
Water exchange (F_E)	Water discharge km^3 yr^{-1}		DSi concentration µmol L^{-1}	BSi concentration µmol L^{-1}	DOC concentration µmol L^{-1}	POC concentration µmol L^{-1}	RSi flux Gmol yr^{-1}	OC flux Gmol yr^{-1}
Output to YS (F_{BTY})	470 [14]		4.33±2.02 [15]	2.20±1.56 [16]	251±82 [17]	29±3.0 [18]	3.1±1.7	132±40
Input to BS (F_{YTE})	442 [14]		4.27±2.29 [15]	1.95±1.34 [16]	207±68 [17]	20±5.7 [18]	2.8±1.6	100±33

Flux								
Flux from porewater (F_B)					Diffusion rate of DSi mmol m^{-2} d^{-1}	Diffusion rate of DOC mmol m^{-2} d^{-1}	Benthic DSi flux Gmol yr^{-1}	Benthic DOC flux Gmol yr^{-1}
					0.53±0.30 [19]	0.95±0.54 [20]	15±8.5	27±15

Flux								
Sedimentation (F_S)		Accumulation rate g cm^{-2} yr^{-1}	BSi content %	TOC content %	Accumulation rate of BSi mol m^{-2} yr^{-1}	Accumulation rate of TOC mol m^{-2} yr^{-1}	BSi flux Gmol yr^{-1}	TOC flux Gmol yr^{-1}
		0.1–0.6 [21]	0.2–0.7	0.1–0.7	0.2–0.8	0.2–1.8	30±12	60±13

Flux								
Submarine groundwater discharge (F_{GW})	Water discharge km^3 yr^{-1}				DSi µmol L^{-1}	DOC µmol L^{-1}	DSi flux Gmol yr^{-1}	DOC flux Gmol yr^{-1}
	40.4–46.7 [22]				183±41.6 [23]	226±47 [24]	8.0±1.8	9.8±2.0

[1] Ministry of Water Resources of the People's Republic of China, 2013; [2] From this study of Kenli

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station in the Yellow River in 2013–2014 (n=12); [3] Wang et al. (2012) and this study (n=24); [4]Wang et al. (2012) and this study (n=23); [5] Liu et al. (2009); [6] Liu et al. (2008b); [7] Li and Feng (2007); [8] Dou et al. (2014); [9] Lin et al. (2001); [10] Zhang et al. (2004) (n=25); [11] Xu et al. (2016) (n=10); [12] Duce et al. (1991), Chen et al. (1997) and Feng et al. (2007) (n=4); [13] Tan et al. (2011) (n=11); [14] Liu et al. (2003a); [15] Data for the southern Bohai Sea are from Yang et al. (2014) and this study (n=35); [16] Data for the southern Bohai Sea are from this study (n=37); [17] Data for the southern Bohai Sea are from Zhao et al. (2015) and this study (n=34); [18] Data for the northern Yellow Sea are from Shang et al. (2010) and this study (n=46); [19] From Liu et al. (2011) and this study (n=27); [20] Calculated based on the Si/C in the surface sediment; [21] Hu et al (2016) (n=25); [22] Peterson et al. (2008); [23] Liu et al. (2011); [24] Average value of DOC in the rivers flowing into the Bohai sea (n=6); The unidentified data are from this study and calculations; surface runoff (non-river compartment) (F_{SR}) and other internal processes are results of the budget calculation; Data following ± are standard deviations, and n is the number of data collected from this study and references.

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Table 2. Reactive silicon and organic carbon concentrations in surface water (0.5 m), bottom water (< 2m from the sea floor) and average for water column in the Bohai Sea in 2012.

Season	Layer	RSi		OC	
		DSi	BSi	DOC	POC
		μmol L^{-1}			
Spring	Surface	4.0±4.2	0.7±1.0	265±100	35±14
	Bottom	3.6±3.7	1.5±1.5	352±193	35±13
	Whole layer	3.8±3.9	1.1±1.3	327±167	35±13
Fall	Surface	7.0±3.0	3.8±4.0	225±49	24±12
	Bottom	7.6±3.3	4.2±4.2	206±39	33±12
	Whole layer	7.3±3.1	4.0±4.0	217±47	28±19

Results are presented as mean ± standard deviation. The number of samples (n): surface in spring, n=22; bottom in spring, n=22; surface in fall, n=21; bottom in fall, n=20.

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Table 3. Biogenic silica and total organic carbon contents of the surface sediment and core sediment in the Bohai Sea.

Sample and station [a]		BSi (%) [b]		TOC (%) [b]	
		Range	Average	Range	Average
Surface sediment [c]	Spring	0.29–0.61	0.41±0.12	0.10–0.66	0.31±0.20
	Fall	0.20–0.69	0.39±0.15	0.10–0.67	0.35±0.19
Core sediment [d]	B45	0.34–0.59	0.45±0.07	0.13–0.84	0.36±0.14
	B49	0.20–0.46	0.28±0.05	0.10–0.62	0.23±0.13
	B61	0.42–0.93	0.59±0.11	0.10–0.75	0.45±0.15

[a] See Figure 1 for the location of the stations; [b] percentage of sediment by weight (%). [c] The number of surface sediment samples (*n*): spring, *n*=7; fall, *n*=18. [d] The slices number of core sediment samples: B45, *n*=35; B49, *n*=34; B61, *n*=41.

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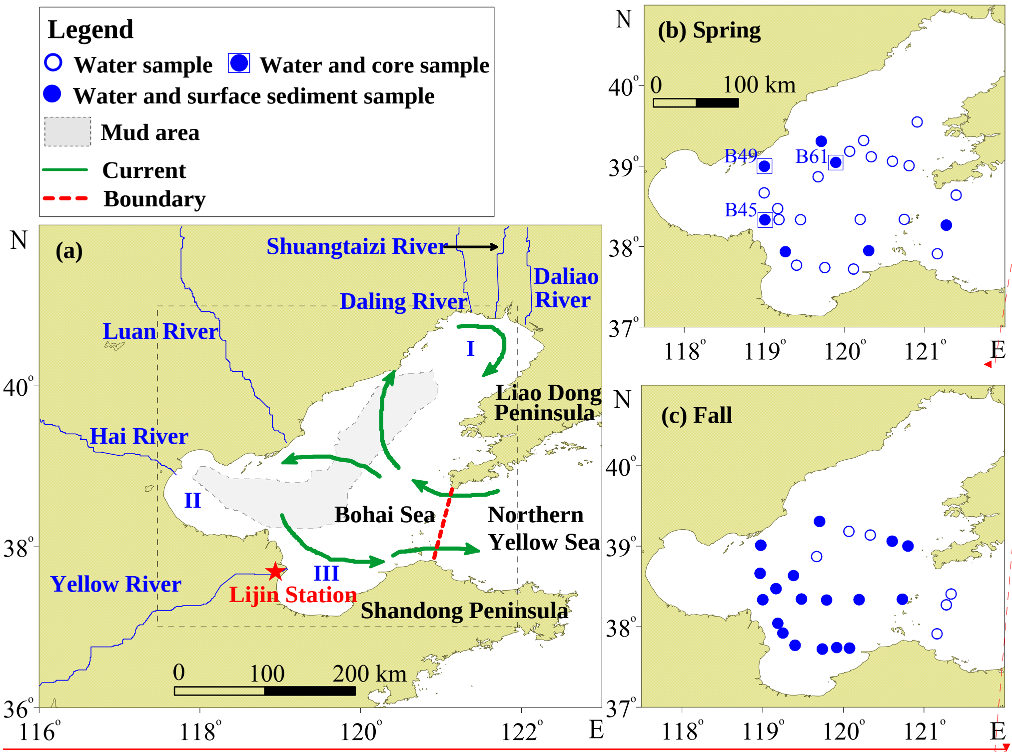

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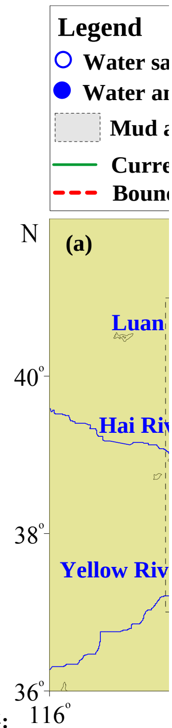

Figure 1. Rivers, mud area, circulation system and sampling stations in the Bohai Sea (I = Liaodong Bay, II = Bohai Bay, and III = Laizhou Bay), and Mud area and circulation system are redrawn according to studies of Hu et al. (2012) and Sündermann and Feng (2004).

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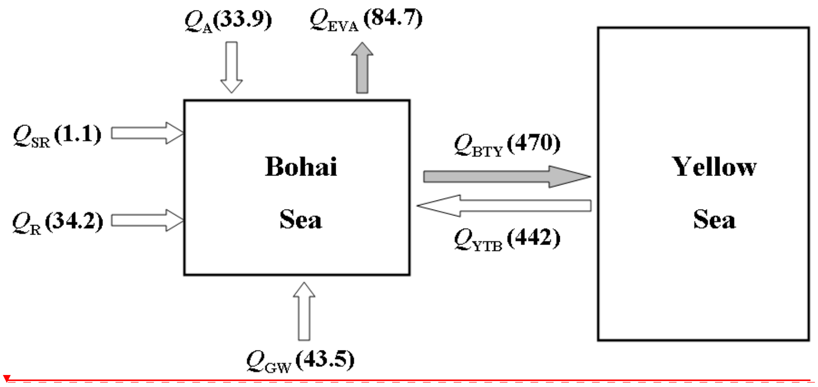

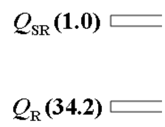

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Figure 2. Water budget in the Bohai Sea. Fluxes in km^3 yr^{-1}. Subscripts R, A, YTB, GW, SR, BTY and EVA denote the river discharge, atmospheric deposition, Yellow Sea inflow, submarine groundwater, surface runoff , Bohai Sea outflow and evaporation, respectively.

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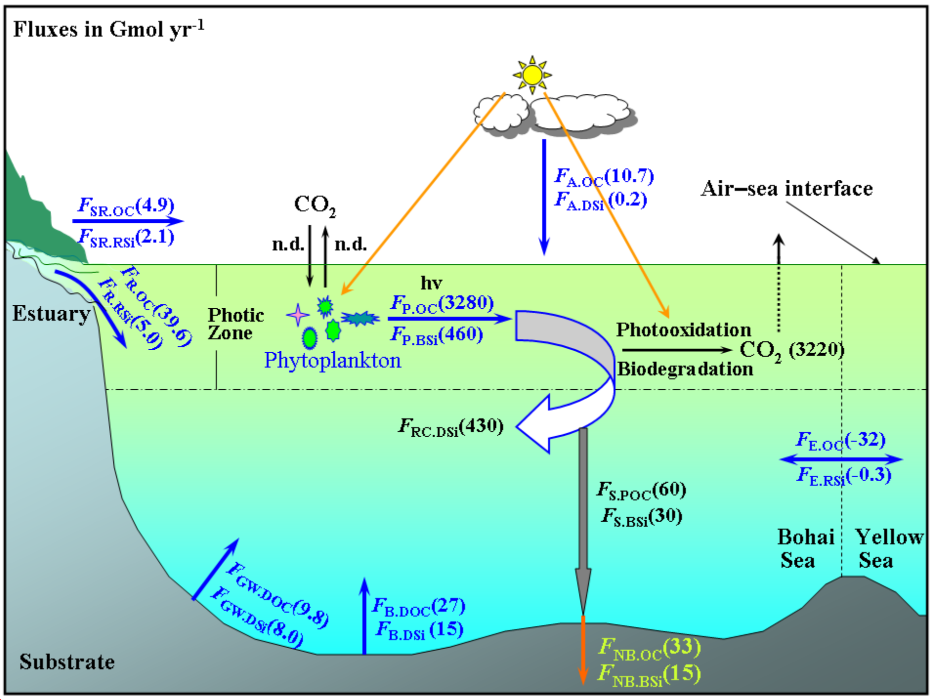

Figure 3. Fluxes of reactive silica (DSi and BSi) and organic carbon (DOC, POC) in the Bohai Sea. F_A: Atmospheric deposition; F_B = Benthic diffusion flux; F_R: River input; F_E = Water exchange from Yellow Sea to Bohai Sea (F_E = F_{YTE} - F_{BTY}; negative fluxes denote outflow from Bohai Sea to Yellow Sea); F_{GW} = Submarine groundwater discharge; F_{NB} = net burial; F_P = Primary production; F_{RC} = Internal recycle; F_S = Sedimentation; F_{SR} = Surface runoff (small rivers not included in F_R); n.d.: No data available.

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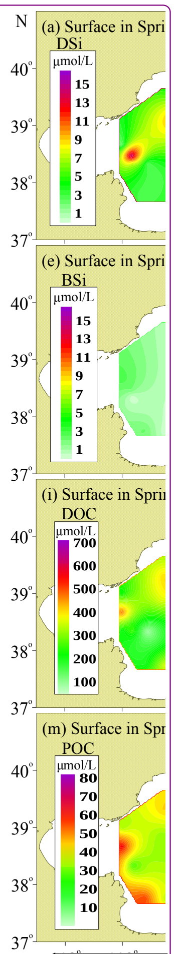

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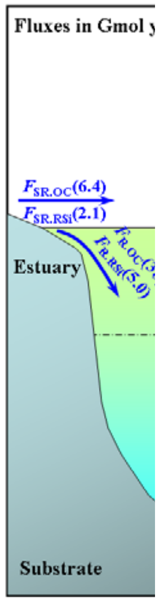

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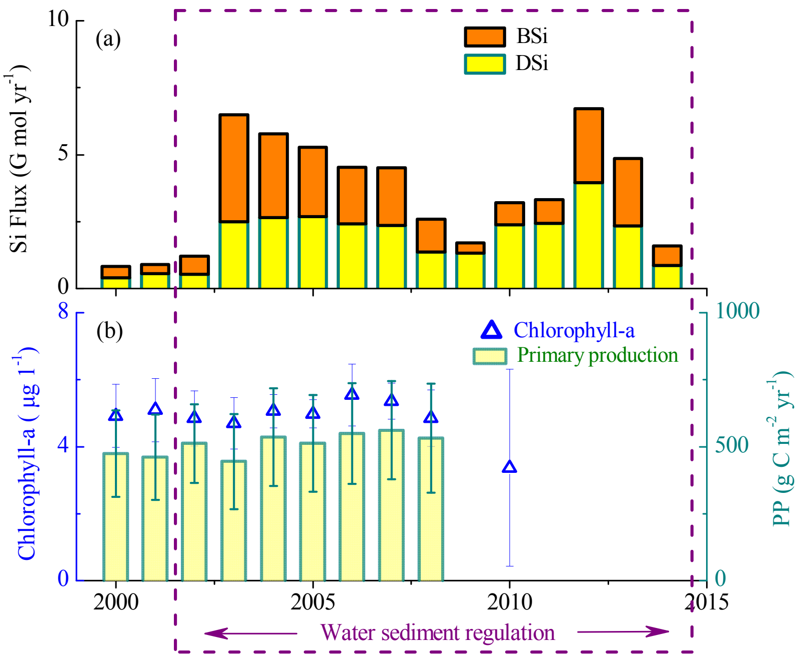

Figure 4. Data for Lijin station in the lower Yellow River for the period 2000-2015 representing (a) DSi and BSi fluxes; (b) data for chlorophyll-a and primary production in the Bohai Sea with standard deviation. DSi in the Yellow River is from Gong (2015), Ran et al. (2015) and this study. BSi data is calculated by Equation 2. Chlorophyll-a and primary production (*PP*) in 2000-2008 of the Bohai Sea are from Tan et al. (2011), data for 2010 are from Chen et al. (2013) and Zhao et al. (2015).

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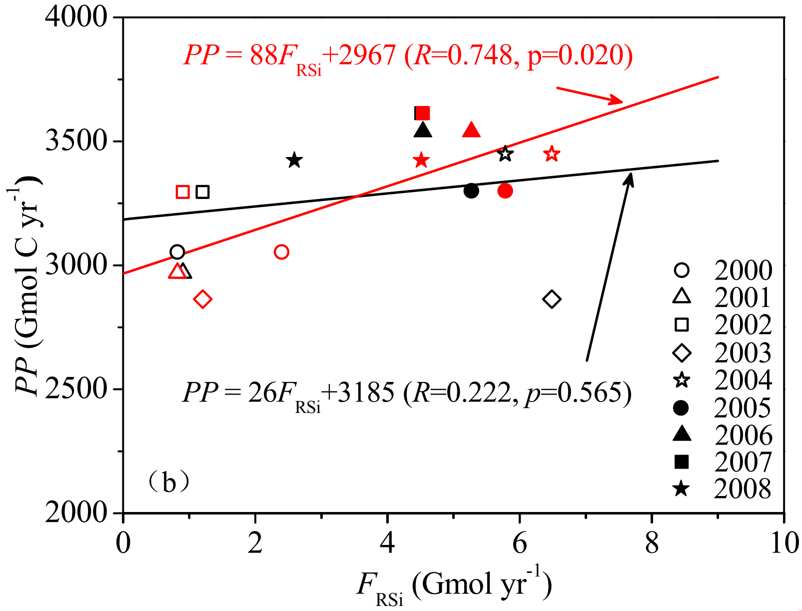

Figure 5. Relationship between annual RSi loading of the Yellow River and primary production in the Bohai Sea obtained by linear regression (black markers represent *PP* and silica loading in the same year, red markers represent *PP* in year n and RSi loading for year n-1. PP data correspond to the data in Figure 4 recalculated to Gmol C yr^{-1}. The regression equation is $PP=a\,F_{RSi}+PP_0$, with *PP* is the primary production (Gmol C yr^{-1}) and PP_0 is the intercept, representing the background primary production from all RSi sources except the Yellow river; F_{RSi} is the RSi flux of the Yellow River (G mol yr^{-1}); *a* is a constant (Gmol Gmol^{-1}).

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SUPPLEMENTARY INFORMATION

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The silica-carbon biogeochemical cycle in the Bohai Sea and its responses to the changing terrestrial loadings

Jun Liu[1,2], Lex Bouwman[3,4], Jiaye Zang[1], Chenying Zhao[1], Xiaochen Liu[3], Xiangbin Ran[1,3]

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4. PBL Netherlands Environmental Assessment Agency, Bilthoven, 3720 AH, The Netherlands

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Correspondence to: Xiangbin Ran (rxb@fio.org.cn)

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Table S1. Biogenic silica and total organic carbon contents (%) of the surface sediment in the Bohai Sea.

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SUPPLEMENTARY FIGURE

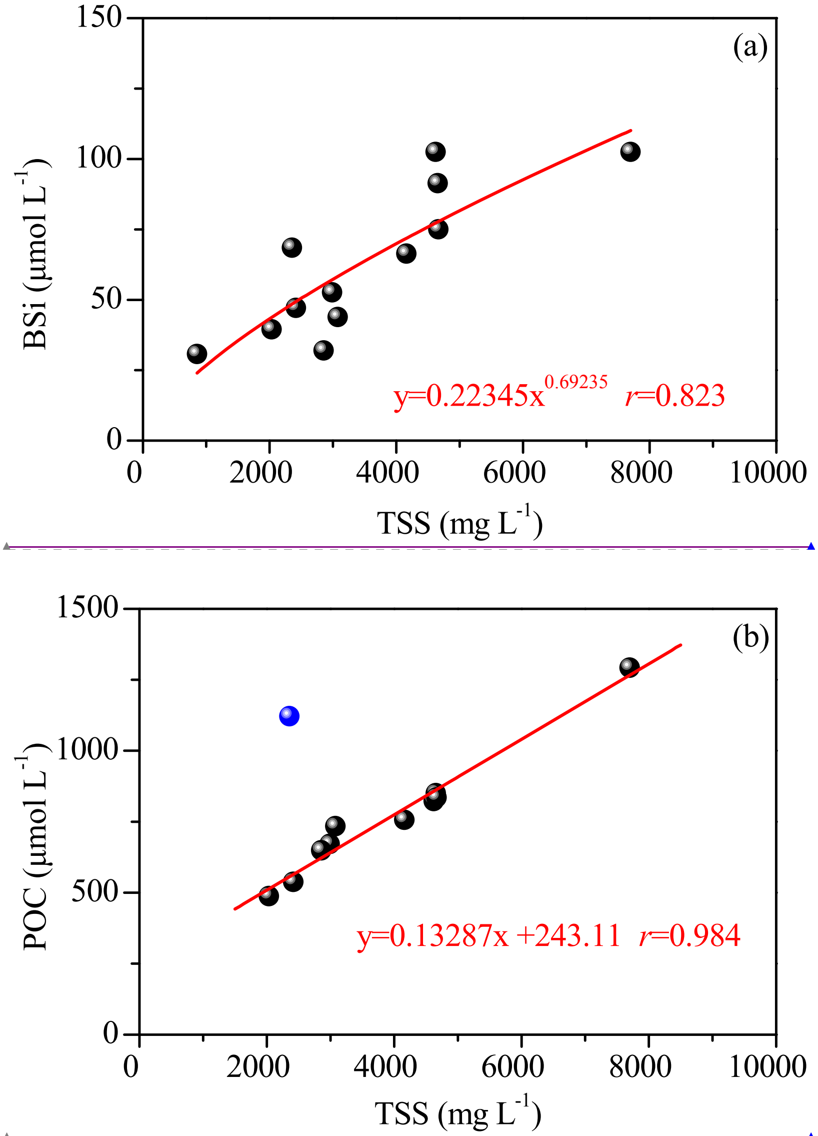

Figure S1. Relationship between BSi and TSS in the Yellow River (a). Relationship between POC and TSS in the Yellow River (b) (The blue cycle represents data not used for the regression equation).

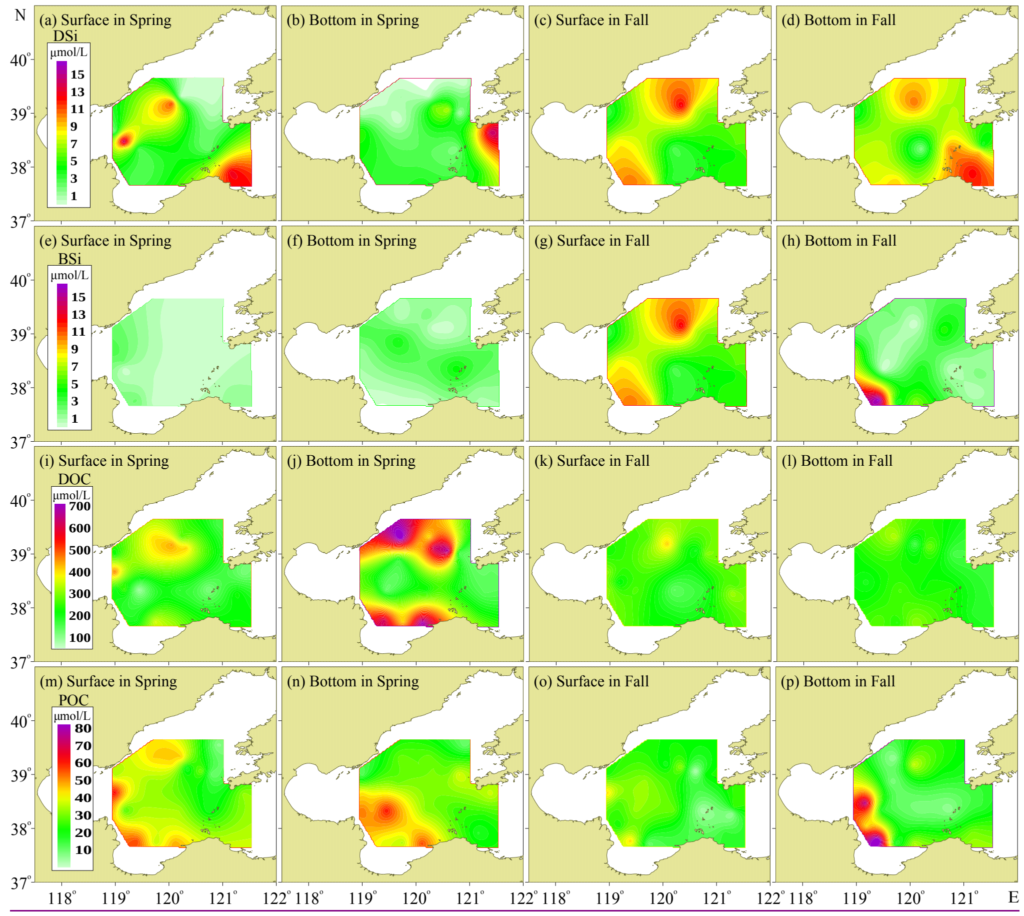

Figure S2. Spatial distributions of DSi (a-d), BSi (e-h), DOC (i-l) and POC (m-p) in the Bohai Sea in surface and bottom water for spring and fall in 2012.

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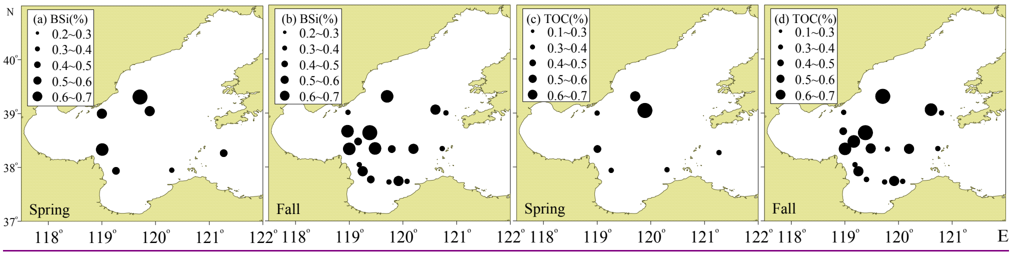

Figure S3. Distributions of total OC (a–b) and BSi (c–d) in the surface sediment of the Bohai Sea.

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The major supply of dissolved silicate (DSi) comes from b

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The Yellow River is the largest river emptying into the Bohai Sea with freshwater and sediment. More recently, enhanced human activities have greatly changed nutrient concentration and composition in the Yellow River (Ning et al., 2010; Gong, 2012; Liu, 2015).

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For instance, dDam construction has caused temporal variations decreased of Si transport by the Yellow River (Liu, 2015; Ran et al., 2015), and distorted nutrient stoichiometry (Tang et al., 2003; Ning et al., 2010; Liu et al., 2011),).

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; the dominant species changed from small cell diatoms (*Chaetoceros*) in 1960s to dinoflagellates (*Noctiluca miliaris*) and then large cell diatoms (*Navicula*) (Tang et al., 2003; Wei et al., 2008), recently;

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a substantial decrease of Si/N ratio in the Bohai Sea is aloso attributed to the reduction of the Yellow river discharge (Ning et al., 2010).

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Furthermore, The water and sediment regulation of the Yellow River since 2002 may enhance primary production by increasing export of water and sediment to the Bohai Sea. cChanges of nutrient inputs from rivers in the semi-enclosed Bohai Sea have larger and more long-lasting influence on the ecosystem than in other open seas because the water residence time in the Bohai Sea is about 3 years (Liu et al., 2012).

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Recent studies also pointed out the sensitivity of s

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input to coastal marine ecosystems and

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consequences

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(Li et al., 2014; Woodland et al., 2015)

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affected by rivers

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(Ragueneau et al., 2010)

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which would provide a new insight on the land-sea interaction

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Sampling expeditions were also carried out at the Lijin Station (Shandong Province) at the Yellow River (Fig. 1a) during a full hydrological year in 2013-2014. Water samples were collected for DSi, BSi, DOC and POC measurements once per month at 20 cm below the surface with at least 3 sampling points across the river main channel. Water samples were pretreated as described above.

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overlap areas with other river basins

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large

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and has large overlap areas with other river basins

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Again, the Yellow River has played a controlling role in the formation of North China Plain, where many studied rivers flow.

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Again, the Yellow River had played a controlling role in the formation of North China Plain, where many studied rivers flow.

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Diatom growth? Or soils?

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Ratio, or is SPM concentration in yellow River used for the other rivers?

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in rivers generally stems from[BL1]

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BSi/POC and SPM[BL2]

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in the other rivers are similar as that in

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Therefore, The BSi and POC concentrations for rivers with missing or scant data were could be estimated with the following

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in snow and rainfalls

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from Willey et al. (2000)

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the distribution of DSi in surface water is differentsimilar to from that in bottom water, and

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in the surface water

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the southeastern part, particularly in

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; the distribution of bottom water DSi is relatively homogeneous except for

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with the exception of particularly

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high values in the Bohai Strait

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; bottom water DSi concentration in coastal areas of the Bohai Sea exceeds that

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with is fairly highlower leve

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Laizhou Bay and Bohai Strait

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t than in coastal areas

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The BSi concentrations are similar to those of DSi; in fall t

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distribution of BSi in the bottom water differs from that in the surface water, with

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distribution of

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in surface water is similar also different fromto that in bottom water, with fairly high

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are reasonable higher

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页 13: [106] 带格式的	RT	2016/4/29 11:27:00

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页 13: [107] 批注 [BL9]	Bouwman, Lex	2016/4/28 11:52:00

BSi or DSi? If DSi, move to first paragraph

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; bottom water BSi[BL3] concentration is fairly lower with the exception of particularly high value in the Yellow River estuary

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are of the same order of magnitude as inputs.similar

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of the Bohai Sea, particularly

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is an important component (

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would be affected by sediment resuspension (Liu et al., 2005) and diatom bloom after settling[BL4] (Wei et al., 2008), which may be t

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页 15: [114] 带格式的	RT	2016/4/29 11:27:00

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页 15: [115] 带格式的	RT	2016/4/29 11:27:00

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页 15: [115] 带格式的	RT	2016/4/29 11:27:00

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页 15: [115] 带格式的	RT	2016/4/29 11:27:00

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页 15: [115] 带格式的	RT	2016/4/29 11:27:00

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页 15: [116] 删除的内容	RT	2016/4/29 11:11:00

(>95%)

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in the world's oceans (Reeburgh et al., 1997), and a little less so (89%)

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页 15: [117] 带格式的	RT	2016/4/29 11:27:00

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页 15: [117] 带格式的	RT	2016/4/29 11:27:00

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页 15: [118] 删除的内容	RT	2016/4/29 11:14:00

being

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Our results show a

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页 15: [120] 带格式的	RT	2016/4/29 11:27:00

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indicating

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concentrations in the coastal areas exceed those in the high salinity waters and that the distribution of POC

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The POC distribution is also affected by sediment resuspension in ocean margins (Zhu et al., 2006), which may explain why POC in bottom water is generally higher than in surface water.

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This

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s

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is consistent with the results from Jiaozhou Bay (Liu et al., 2008a) and East China Sea (Liu et al., 2005).

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There are no differences of both

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of RSi

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页 16: [128] 删除的内容	Liu009Liu	2016/4/13 16:54:00

and

页 16: [128] 删除的内容	Liu009Liu	2016/4/13 16:55:00

, contributing

页 16: [129] 删除的内容	Liu009Liu	2016/4/13 16:57:00

99% and

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to Si removal in the budget, respectively

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surface runoff (8%)

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atmospheric deposition (5%)

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are

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contributes the remaining

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页 17： [137] 带格式的	RT	2016/4/29 11:27:00

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页 17： [138] 批注 [BL11]	Bouwman, Lex	2016/4/28 11:52:00

I believe this is the first time this is stated; how important s this

页 17： [139] 批注 [BL12]	Bouwman, Lex	2016/4/28 11:52:00

I believe this is the first time this is stated; how important s this

页 17： [140] 批注 [BL13]	Bouwman, Lex	2016/4/28 11:52:00

What is additional?

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diagenetic alteration[BL5] of aluminosilicate minerals and

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and diagenetic alteration[BL6] of aluminosilicate minerals (Loucaides et al., 2008; Krom et al., 2014)

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, representing an additional external source[BL7]

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, and lead to seasonal variations of RSi and OC in the water column, conversely

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based on the assumed BSi : TOC ratio of 0.56.

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of

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on

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BSi

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. This ratio of large BSi sedimentation flux to BSi production (3.3%)

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of

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36–97%

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world ocean (

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,

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shows

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ed

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nutrient

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the

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we

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Series of study have showed that Relative contributions from submarine groundwater and is comparable to riverine Si input are similar to those in the Yellow Sea (Kim et al., 2005) and Mediterranean Sea (Rodellas et al., 2015), a semi-closed sea like the Bohai Seain some coastal regions.

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页 18: [162] 带格式的	RT	2016/4/29 11:27:00

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in the Yellow Sea reported by

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(

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; also, Si input from

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, in the Mediterranean Sea

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a semi-closed sea like the Bohai Sea

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I think the Mediterranean is totally different from Bohai so I deleted this

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Our estimate for submarine groundwater DSi input exceeds riverine input, and this agrees with the above studies (Liu et al., 2011; Rodellas et al.,

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Sediment? Why sedi

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process impacting on

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on

页 19： [177] 删除的内容 Bouwman, Lex 2016/4/28 11:44:00

may arouse

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28% uncertainty of total Si input estimation (the relative deviation (SD) of F_B divided by total Si input SD/(F_R+F_{SR}+F_A+ F_E+F_B+F_{GW}FA),

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similar for following calculation

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Further, biologic and hydrologic conditions are the most important factors influencing the diffusive flux (Berner, 1980). Thus, sediment as a source of DSi needs to be tested by massive field measurements.

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introduced by

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ly

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. These spatial and temporal variations will introduce

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SD

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flux

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uncertainty in the calculation of the RSi output from the Bohai Sea

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of total input

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might

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be

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were rather

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sediment

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arouses

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of uncertainty of total OC input estimation

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Another

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of the OC budget is introduced by

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flowing

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, which would introduce

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uncertainty of total OC input

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of

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of total OC input.

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may arouse

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uncertainty of total OC output estimation

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silica

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Si concentration
in the rainfall

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DOC concentration
in the rainfall

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Dry deposition
rate of Si

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Dry deposition
rate of POC

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μmol L^{-1}

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μmol m^{-2} yr^{-1}

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μmol L^{-1}

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mol m^{-2} yr^{-1}

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Gmol yr^{-1}

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Gmol yr^{-1}

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4.1±0.9 [10]

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72.3 [10]

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75±34[11]

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0.02±0.01[12]

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0.2±0.1

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4.2±2.2

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Primary productivity
of carbon

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Primary productivity
of BSi

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Si fixed by
primary production

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C fixed by
primary production

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g m^{-2} yr^{-1}

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mol m^{-2} yr^{-1}

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Gmol yr^{-1}

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Gmol yr^{-1}

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509±38 [13]

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6.0±4.5

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460±35

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3280±250

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DSi concentration

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BSi concentration

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DOC concentration

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POC concentration

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RSi flux

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μmol L^{-1}

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μmol L^{-1}

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μmol L^{-1}

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μmol L^{-1}

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Gmol yr^{-1}

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Gmol yr^{-1}

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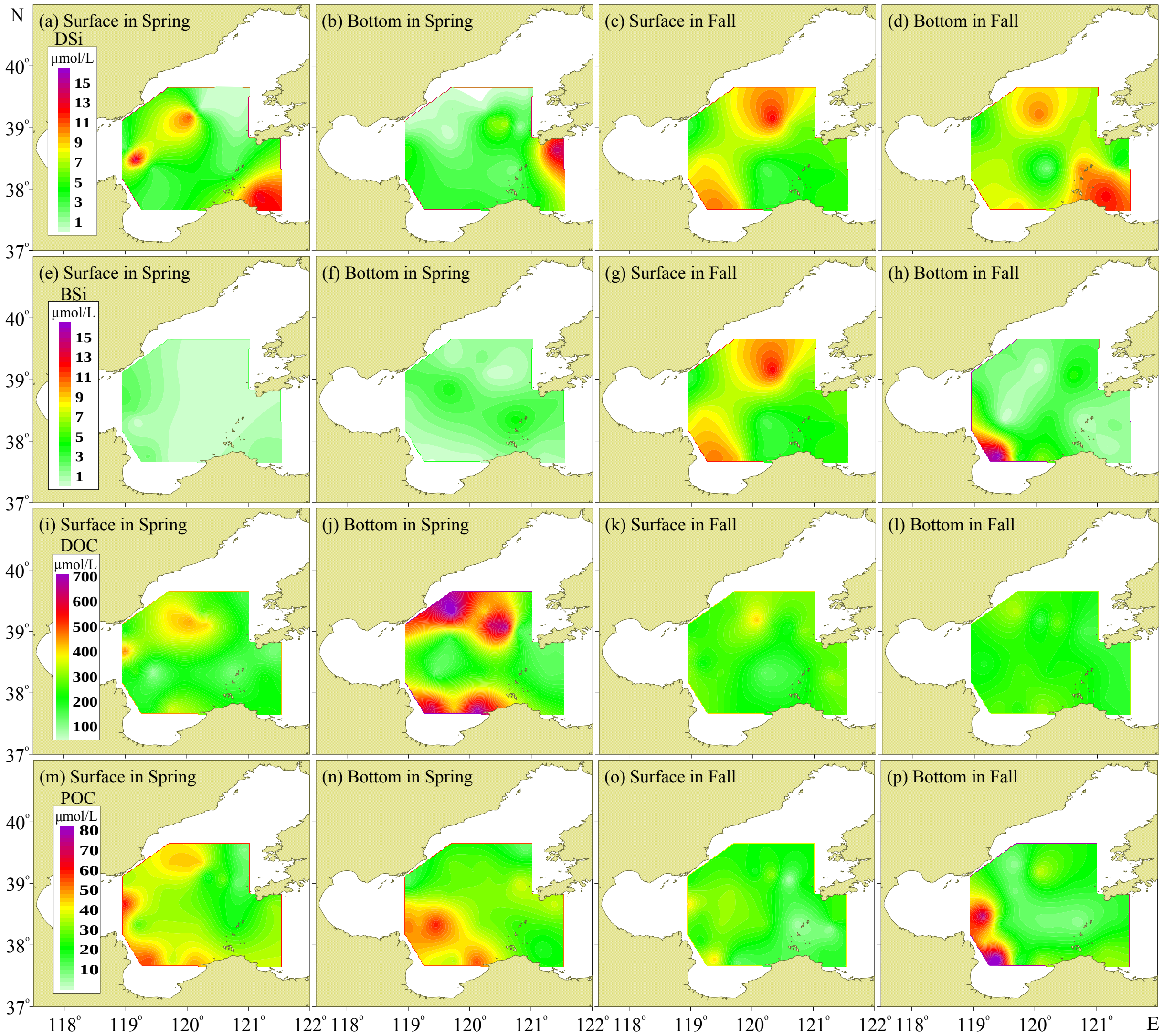

Figure 3. Spatial distributions of DSi (a-d), BSi (e-h), DOC (i-l) and POC (m-p) in the Bohai Sea in surface and bottom water for spring and fall in 2012.

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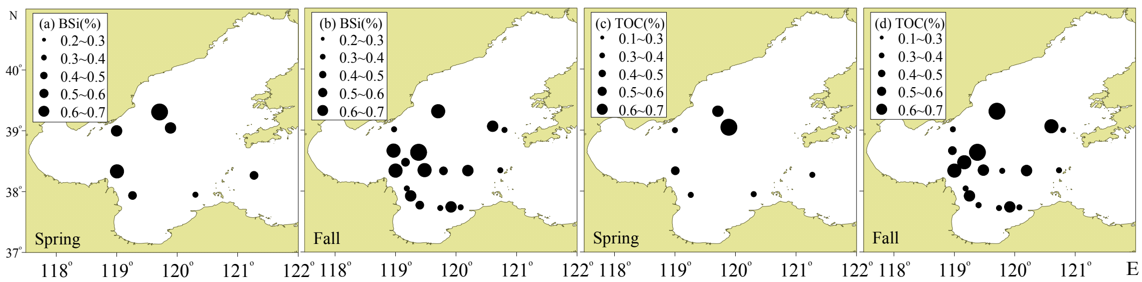

Figure 4. Distributions of total OC (a–b) and BSi (c–d) in the surface sediment of the Bohai Sea.

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SUPPLEMENTARY TABLE

Table S1. Biogenic silica and total organic carbon contents (%) of the surface sediment in the Bohai Sea.

Spring			Fall		
Station	BSi	TOC	Station	BSi	TOC
B36	0.31	0.20	B39	0.20	0.10
B45	0.53	0.31	B41	0.46	0.50
B49	0.41	0.10	B42	0.40	0.17
B50	0.61	0.48	B43	0.53	0.45
B61	0.41	0.66	B45	0.54	0.53
B65	0.37	0.26	B47	0.56	0.37
B71	0.29	0.19	B49	0.29	0.10
			B50	0.60	0.67
			B53	0.42	0.50
			B54	0.27	0.19
			B63	0.69	0.60
			B64	0.35	0.55
			B65	0.41	0.44
			B66	0.28	0.21
			B68	0.21	0.10
			B69	0.41	0.44
			B70	0.21	0.17
			YD01	0.27	0.24

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