

Variations in organic carbon loading of surface sediments from the shelf to the slope of the Chukchi Sea, Arctic Ocean

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Abstract

The content of organic carbon (OC) normalized to the specific surface area (SSA) of sediment is widely used to trace variations in OC loading (OC/SSA). This study presents observations of OC/SSA of surface sediments collected in the Chukchi Sea, a typical Arctic marginal sea. Shelf sediments exhibit much higher OC/SSA values than slope sediments in the study area. Compared with OC/SSA values reported from the East Siberian Shelf and Mackenzie River, the slope sediments possess lower OC loading. This abrupt decrease in OC/SSA is mostly related to the lower primary production on slope as well as possible oxidization processes. The results of linear regression analysis between OC and SSA indicate a sedimentary source rock for the OC in the Chukchi Sea sediments. Moreover, shelf sediments with low SSA possess a larger rock OC fraction than slope sediments do. The dataset of the present study enables a more thorough understanding of regional OC cycling in the Chukchi Sea.

Key words: Chukchi Sea, Arctic, surface sediments, organic carbon loading, carbon cycle

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1 Introduction

The Arctic Ocean possesses the largest continental shelf in the world and plays a key role in the global organic carbon (OC) cycle (Chen et al., 2015; Stein and Macdonald, 2004). Under the impact of global warming effect, the Arctic Ocean and surrounding areas are showing greater sensitivity to changes than the other regions (Chen et al., 2015). Global warming will lead to the thawing of permafrost and enhanced the export of soil OC and minerals into aquatic systems. During the transportation and burial of terrestrial OC, a certain fraction of the carbon is converted into CO₂; thus, permafrost thawing is a source of atmospheric CO₂. In addition to affecting the terrestrial environment, warming will also reduce the sea ice extent and keep the water warm (Chen et al., 2015), thus leading to higher oceanic productivity. The oceanic productivity in the Arctic stems mainly from large diatoms (>20 μm; Li et al., 2007), which are more easily transported to the sea bed and are buried in larger numbers than smaller species (Walsh and McRoy, 1986); consequently, a higher burial flux of fresh OC is expected in the euphotic Arctic Ocean.

On geological timescales, the atmospheric CO₂ level is to some extent controlled by the burial efficiency of OC in the marine environment (Lasaga et al., 1985), which is largely dependent

on the OC loading (OC/SSA) of sediments (Blair and Aller, 2012; Hedges and Keil, 1995; Keil et al., 1997). Therefore, tracing the fate of this OC is critical in predicting the feedback of the Arctic Ocean to global warming (Chen et al., 2004, 2015). The OC content and specific surface area (SSA) of the sediments are two of the most widely applied indexes for studying variations in OC loading in sediments (Bergamaschi et al., 1997; Galy et al., 2008; Goñi et al., 2013; Vonk et al., 2015). Bergamaschi et al. (1997) investigated the OC content and SSA of Peru Margin sediments and reported a strong positive correlation between these indexes. Vonk et al. (2015) analyzed the river channel, estuarine and shelf sediments collected from the Mackenzie River system in the North America, revealing that the SSA of surface sediments in these realms shows an increasing trend in the offshore direction.

The Chukchi Sea is a marginal sea of the Arctic Ocean, bounded on the west by the De Long Strait, off Wrangel Island, and in the east by Point Barrow, Alaska, beyond which lies the Beaufort Sea (Polyak et al., 2007). The Bering Strait forms the southernmost limit of the Chukchi Sea, and connects it to the Bering Sea and the Southwest Pacific Ocean. The Chukchi Sea has an area of ~5.95×10⁵ km² and is only navigable for about four months per year (Weingartner et al., 2005). The sea plays a key role in organic carbon cycling in the Arctic Ocean, especially dur-

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ing the melting season when it receives massive amounts of terrestrial OC and marine OC. The OC loading of sediments has a strong influence on flux estimates and predetermines the response of the Arctic Ocean to global warming.

To accurately determine the OC loading of sediments in the Chukchi Sea, surface sedimentary samples were collected and their OC content and SSA were analyzed. Spatial variations in OC loading are discussed in detail, and the results of previous studies around the Arctic Ocean are compiled to obtain a full map of OC loading in this area. This work will contribute to a better understanding and prediction of the regional carbon cycle and the amounts of CO₂ emitted from the sediments to the water column in the Chukchi Sea.

2 Materials and methodology

2.1 Sample collection and preparation

Surface sediments were collected during the cruise of the sixth Chinese National Arctic Research Expedition in summer of 2014. A total 23 samples were analyzed in this study, covering the areas from the shelf to the slope, at water depths of 33 m to 3 763 m (Fig. 1). Of these samples, 13 were collected from the continental shelf (<150 m) and the remainder from the shelf break and plateau (150–3 763 m). Sediments were obtained using an iron box-type sampler. The surface sediments (0–2 cm) were subsampled, placed in plastic bags, and stored in a refrigerator at

–20°C. In the laboratory, ~10 g of each sample was freeze-dried. Half of this amount was ground using an agate mortar and pestle and sieved to 80 mesh (187.5 μm) prior to OC analysis to ensure sample homogeneity. The portions of samples that remained unground were used for specific surface area analysis.

2.2 OC loading (OC/SSA) analysis

OC/SSA ((mg C)/m²) is the ratio of OC content ((mg C)/(g dw)) to SSA (m²/(g dw), dw is dry weight). OC content was measured using an elemental analyzer (CHNOS Vario EL III). The dried and homogenized sediments were placed into glass tubes, after which inorganic carbon was removed by adding hydrochloric acid (HCl, 10% v/v) and incubating at room temperature for 24 h. The extra acid was discarded after centrifugation, following which the sediments were dried and homogenized again. About 10 mg dried sediment was weighed and placed in a tin cup. The OC content was calculated from the mass of sediment and the area of the CO₂ peak. The accuracy of OC measurements was assessed by analyzing two marginal sea sediment standards (Chinese National Standard Material, GBW07309 and GBW07333). Analyses in triplicate indicated that the external precision (one relative standard deviation, RSD) was better than 3%.

SSA was determined by nitrogen adsorption using a surface area and porosity analyzer (Micromeritics Tristar 3020). Samples were combusted at 350°C for 12 h to remove the organic matter and water prior to analysis. Subsequently, ~1 g sample was de-

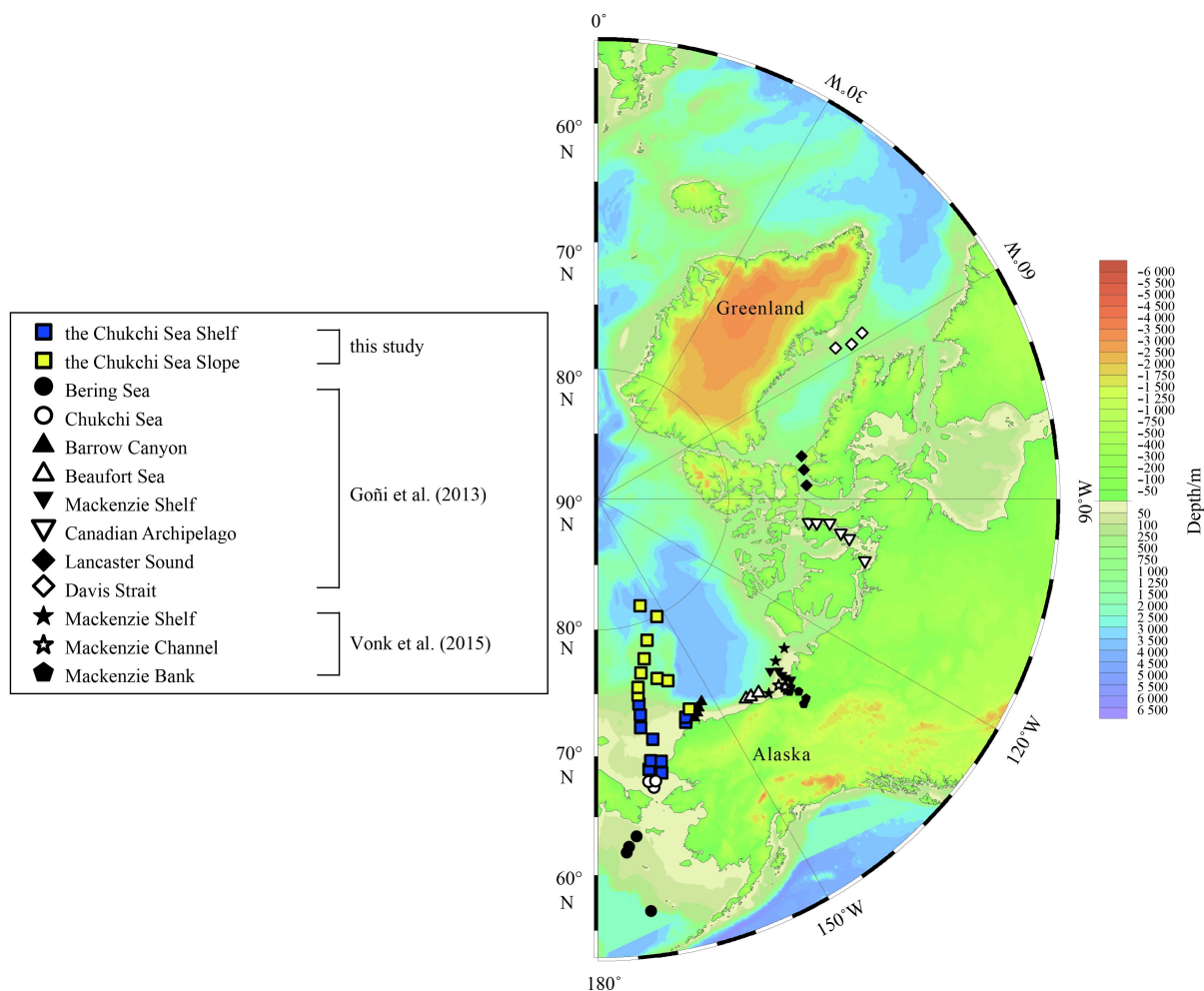


Fig. 1. Map of sample stations this study and locations of data published in the literatures.

gassed at 50°C and 10⁻⁵ Pa for 18 h. Adsorption-desorption isotherms were obtained by subjecting samples to various partial pressures of N₂ at 77 K. The specific surface area was calculated following the Brunauer Emmet Teller (BET) method. The accuracy of SSA measurements was assessed by analyzing Chinese National Standard Materials (GBW130276 and GBW130279). Analyses in triplicate indicated that the standard deviation of these measurements was less than 0.20 m²/g.

2.3 Statistical analysis

Pearson's correlation analysis was performed using SPSS 20.0 software (IBM, USA) in this study. The sampling map was made using the Ocean Data View software, and other figures were produced using OriginPro 8.0 software, respectively.

3 Results

The values of the measured indexes for the surface sediments in the Chukchi Sea are listed in Table 1. The OC contents for all the samples were 3.76–6.51 (mg C)/(g dw) (average 9.30±0.98) (mg C)/(g dw). SSA for these sediments was 4.01–39.65 m²/(g dw), (average 21.15±11.14) m²/(g dw). As the hydrodynamic conditions differ greatly between the shelf and the deep slope/plateau areas, the samples were divided into two categories on the basis of 150 m isobaths: one group of sediments samples was recovered from the shallow shelf region with water depth is less than 150 m; the other was obtained from the slope areas. The shelf sediments exhibited OC values of 3.76–17.19 (mg C)/(g dw) (average 10.32±3.98) (mg C)/(g dw) and 4.01–26.53 m²/g (average 13.35±7.97) m²/g. The calculated OC normalized to specific surface area (OC/SSA) was 0.48–2.60 (mg C)/m² (average 0.97±0.53) (mg C)/m² for these sediments. A positive correlation between the OC content and SSA was observed for the shelf

sediments ($r=0.85$, $p<0.01$, $n=13$). The sediments from the slope area displayed a comparable OC content (4.53–17.51 (mg C)/(g dw), average 7.97±3.56) mg/(g dw) but SSA values two to three times larger (24.32 to 39.65 m²/g, average 31.35±4.5) m²/g. In contrast to the shallow water sediments, there was a weaker linear relationship between OC and SA in the deep-water sediments. The difference in the SSA of sediments also resulted in differences in the OC/SSA values, which were 0.13–0.59 (mg C)/m², average (0.26±0.12) (mg C)/m² (Fig. 2).

SSA reflects both the physical properties of the sediment (grain size) and the adsorption capacity of the organic matter. As shown in Fig. 3, when considering the SSA for all the sediments studied herein, a positive correlation between SSA and water depth in all the Chukchi Sea sediments can be observed ($r=0.55$, $p<0.01$, $n=23$). This increasing trend of SSA mainly reflects hydrodynamic sorting of the sediments: finer sediment particles with a high specific surface area are more easily carried to the deep ocean than are coarse particles.

4 Discussion

4.1 Spatial variations in OC loading for the Chukchi Sea surface sediments

OC/SSA has been widely used to trace variations in the OC loading of sediments in various aquatic systems, such as the river basins (Galy et al., 2008; Vonk et al., 2015), estuaries, and shelf regions (Blair and Aller, 2012; Goñi et al., 2000, 2005, 2013; Vonk et al., 2015). The OC/SSA values of shelf sediments in the Chukchi Sea are comparable to those of Mackenzie River sediments (Fig. 3; Goñi et al., 2013; Vonk et al., 2015; this study). However, for sampling locations in the slope area of the Chukchi Sea, the OC/SSA values of sediments are lower than those of shelf sedi-

Table 1. Location, water depth, specific surface area and OC content of the Chukchi Sea surface sediments

Sample station	West longitude/(°)	North latitude/(°)	Water depth/m	SSA/m ² ·(g dw) ⁻¹	OC/(mg C)·(g dw) ⁻¹	OC/SSA/(mg C)·m ⁻²	Rock organic carbon/%
Shelf sediments							
C03	69.03	166.48	33.00	4.05	10.55	2.60	44.3
CC6	68.24	167.13	42.28	4.01	6.00	1.50	77.8
C04	71.01	166.99	45.00	12.81	12.79	1.00	36.5
CC4	68.13	167.51	48.90	6.83	5.81	0.85	80.4
C01	69.22	168.14	50.00	13.82	8.94	0.65	52.2
R06	72.00	168.98	51.35	20.29	14.65	0.72	31.9
CC3	68.10	167.90	52.50	4.14	3.76	0.91	-
R03	68.62	169.00	53.70	12.17	11.14	0.92	41.9
CC2	67.90	168.24	57.60	5.26	5.34	1.01	87.5
S01	71.62	157.93	62.94	15.56	10.70	0.69	43.6
S02	71.92	157.46	73.00	25.42	17.19	0.68	27.2
R07	73.00	168.97	73.36	22.60	14.68	0.65	31.8
R08	74.00	169.00	82.69	26.53	12.65	0.48	36.9
Slope sediments							
R10	75.43	167.90	164.36	26.70	5.58	0.21	15.8
S03	72.24	157.08	169.20	29.51	17.51	0.59	-
R09	74.61	169.03	190.00	29.59	8.63	0.29	10.2
R11	76.15	166.20	352.43	37.45	7.85	0.21	11.2
R12	77.00	163.89	438.86	24.32	4.99	0.21	17.6
R14	78.63	160.43	761.37	33.70	4.53	0.13	19.4
C13-5	75.20	159.18	941.76	29.25	7.50	0.26	11.7
C14	75.40	161.30	2 091.80	33.66	9.13	0.27	9.6
SIC03	81.08	157.66	3 634.20	29.24	5.33	0.18	16.5
SIC06	79.98	152.63	3 763.00	39.65	8.62	0.22	10.2

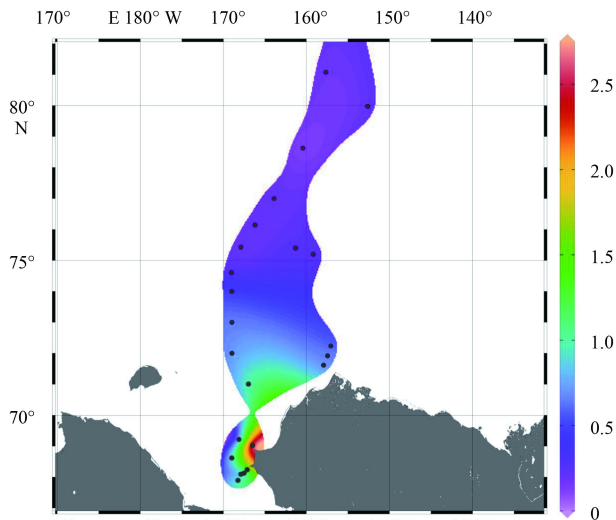


Fig. 2. The spatial distribution of OC loading ((mg C)/m²) in the Chukchi Sea surface sediments.

ments (Fig. 3). The variations in OC/SSA in the Chukchi Sea sediments indicate decreasing OC loading from the inner shelf to the outer shelf and slope, despite the increasing sediments SSA values. This result suggests that the OC loading varies during sediment transport. Similar results were reported in the Canadian Archipelago by Goñi et al. (2013).

Many factors influence the OC loading of deep marine sediments, such as local productivity, vertical fluxes in OC to the benthos, and exposure to effective oxidants (Blair and Aller, 2012; Hedges and Keil, 1995). The abrupt decrease in OC/SSA from the shelf to the slope in the Chukchi Sea, as detected in the present study, is likely to be controlled by multiple factors.

First, the spatial variations in the upper-layer primary production may be responsible for the shift in OC loading in the Chukchi Sea. As indicated by the level of integrated chlorophyll *a*

in surface total suspended material, the high primary production occurs farther south in the nutrient-rich areas of the northern Bering Sea and southern Chukchi Sea (average 470 (g C)/(m²·a); Grebmeier et al., 2006), in comparison, the deep Chukchi Sea shows a much lower primary production than the nutrient-replete waters (Hill and Cota, 2005). This finding was further verified by the mass and particulate OC fluxes in the Canadian Basin, which is located nearby the Chukchi Sea (Hwang et al., 2008, 2015). Therefore, it is likely that the relatively low vertical flux of OC to the sediments is responsible for the low OC loading in the slope areas of the Chukchi Sea.

Second, the changes in OC/SSA may also be related to the OC loss due to *in situ* oxidation reactions at the sediment-water interface. For example, Goñi et al. (2013) investigated the OC/SSA variations in the sediments along the North American Arctic margin (Fig. 1), and found that OC/SSA was negatively correlated with the thickness of Mn oxyhydroxides at each coring sites. In the present study, the thickness of sediments was 2 cm, representing approximately 13 to 5 000 years of sedimentation in the Chukchi Sea assuming a sedimentation rate of 1.45–0.004 mm/a (Lin et al., 2016). In general, a lower sedimentation rate will result in a prolonged exposure duration of sediment to the aquatic environment, and thus enhanced oxidation of OC. Therefore, the OC loading in the sediments will gradually decrease as the sedimentation rate decreases from the shallow to deep water.

In addition, oxidation during the lateral transport of sediments may affect OC/SSA. Recently, the importance of OC lateral transport in the deep ocean was emphasized (Keil et al., 2004), especially in areas of the Arctic Ocean covered by the ice (Fahl and Nöthig, 2007; Hwang et al., 2008; Hwang et al., 2015). Fahl and Nöthig (2007) pointed out that the lateral input accounted for 42%–64% of the total flux to the sea bed on the Lomonosov Ridge. Hwang et al. (2008, 2015) used radiocarbon isotope data of trap sediments collected from the Canadian Basin to trace the lateral transport of particulate OC, and concluded that lateral particle supply was a predominant feature of the particle-flux processes in this basin. Unfortunately, no data for trap sediments

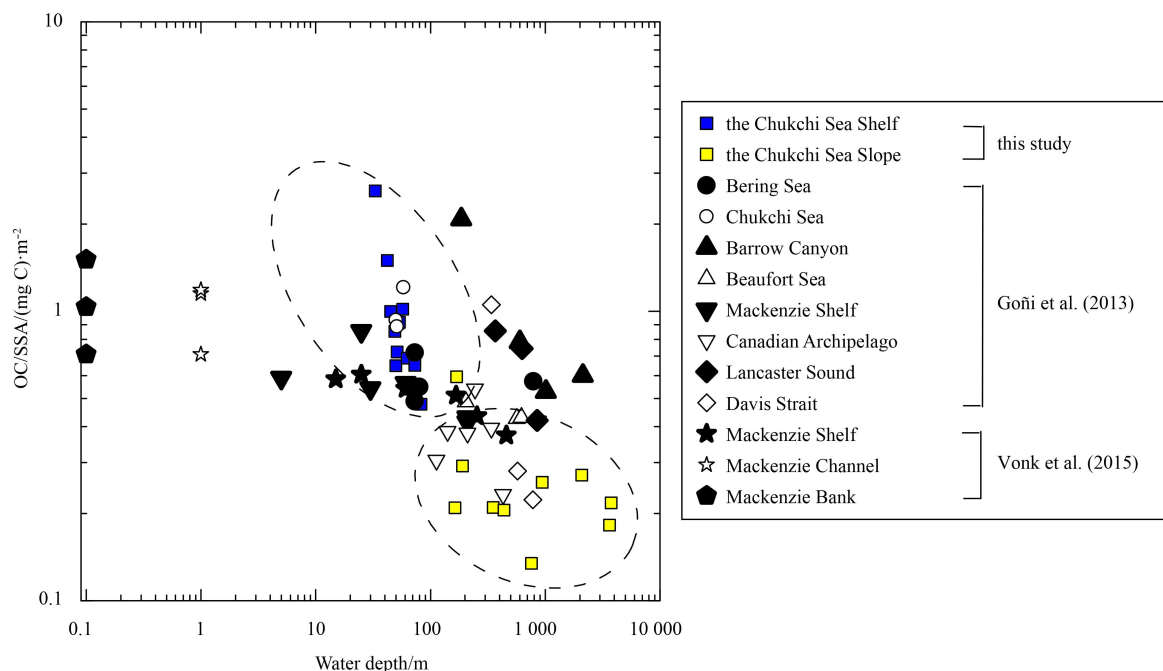


Fig. 3. The variations in organic carbon loading in the Arctic sediments along the sampling water depth.

in the Chukchi Sea have been reported to verify the occurrence of this process. To compensate for this limitation, the SSA and grain size of sediments can be used to trace the sediment transport mechanism: the ocean current from the Bering Strait moved fine (high SA) sediments to the slope of the Chukchi Sea (Wang et al., 2015). In addition, dissolved oxygen in the Chukchi Sea was at normal levels without the occurrence of hypoxic events (Wang et al., 2015). These two facts might suggest suitable environmental conditions for OC oxidization by the effective oxidants during lateral transport, consequently resulting in low OC loading in the deep stations. However, it should be noted that for an improved quantitative evaluation of the relative significance of all the listed mechanisms, further investigations focusing on suspended matter and core sediments should be carried out in the future.

4.2 Possible contribution of rock organic carbon and compilation of literature studies

Figure 4 displays the correlation between OC and SSA in the Arctic sediments, as reported in the present and previous studies. The correlation between OC and SSA in the Chukchi Sea surface sediments is different to that reported in previous studies in other regions (Bergamaschi et al., 1997; Hedges and Keil, 1995). The OC axis intercept for sediment from water depths of <150 m and >150 m is approximately 4.0 mg/(g dw) and 1.0 mg/(g dw), respectively (Fig. 4). Regression analysis of OC and SSA in North American Arctic sediments shows a comparable intercept with that of station above 150 m in the Chukchi Sea; however, there was no strong correlation between OC and SSA in the Mackenzie River sediments.

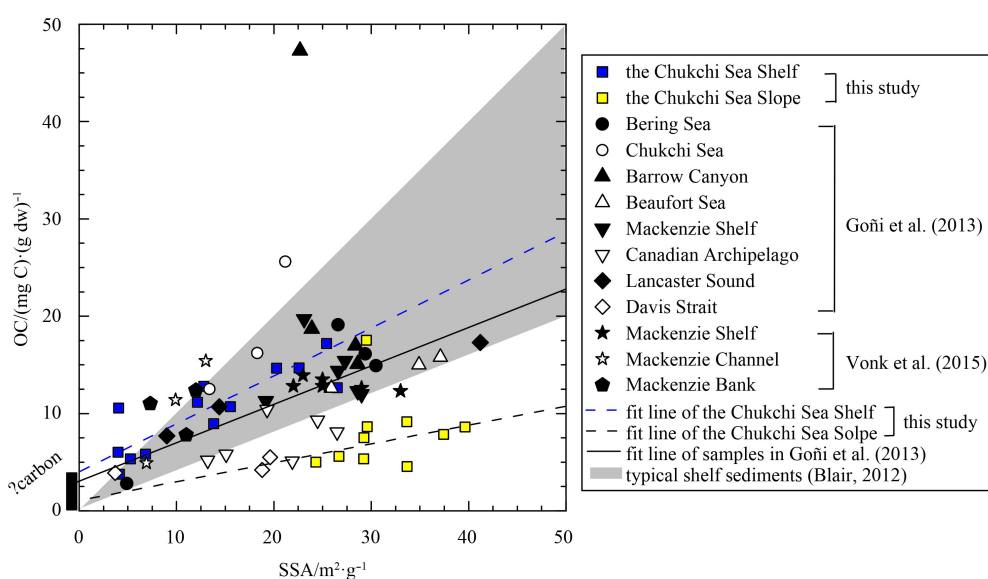


Fig. 4. The correlation between OC and SSA of the sediments in the Chukchi Sea and other regions of the Arctic Ocean.

The three OC-SSA regression lines for all the samples in the Arctic Ocean to zero SSA indicated that a fraction of OC remained independent of SSA (Fig. 4), i.e., some of the OC in the sediment did not originate from adsorption. A positive intercept on the OC axis was also observed for Peru Margin and Washington coast sediments (Bergamaschi et al., 1997; Keil et al., 1994), and was ascribed to OC from plant tissues and sedimentary rock. Plant tissue is a significant contributor to the OC in the coarser fraction of Washington coastal sediments (Keil et al., 1994). Sedimentary rock OC is a dominant source of OC in rivers and marginal seas, such in the Ganga-Brahmaputra basin (Galy et al., 2008), and the Yellow River system (Tao et al., 2015). Vonk et al. (2012) analyzed the organic carbon isotope composition of West Siberian sediments, and found that nearly half of the sedimentary rock OC returned to the atmosphere during the sediment transport. For the Chukchi Sea sediments, the intercepts may represent the contribution from rock organic carbon rather than the plant tissues, because plant debris only accumulates in in-shore regions of the Siberian margin, whereas mineral-bound OC is transported to offshore regions (Tesi et al., 2016).

Based on the intercept on the OC axis and the OC content, it is possible to perform a simple calculation of the amount of rock carbon as a percentage of total OC (Table 1). Rock OC constituted an average of $53.2\% \pm 23.4\%$ of total OC in shelf sediments, decreasing to only $13.6\% \pm 5.3\%$ in slope sediments. The higher

fraction of rock-derived OC in the shelf area indicated a high input of terrestrial OC. However, a previous study based on the stable carbon isotopes concluded that most OC was derived from the marine production (Stein and Macdonald, 2004). This discrepancy in results might reflect the isotopically depleted nature of phytoplankton in the polar oceans (Goericke and Fry, 1994; Rau et al., 1989).

5 Conclusions

The OC content and SSA values of the Chukchi Sea surface sediments provide novel insights into the fate of sedimentary OC exported from terrestrial permafrost and marine production. Hydrodynamic sorting is the dominant control on the spatial distribution of the SSA of the sediments. In contrast, the OC content in these sediments is likely affected by multiple factors, such as primary production in the upper ocean, oxidation of OC during transport and *in situ* oxidation of OC in the sediments. The intercepts of regression analysis between OC and SSA revealed that rock organic carbon constitutes a high percentage of total OC, especially in shelf stations. The outcomes of this study will improve our knowledge of the regional carbon cycle in the Chukchi Sea.

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