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Sources and degradation of organic matter in the surface sediments of the Chukchi Sea: insights from amino acids

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Abstract

In the context of global warming and rapid environment change in the Arctic, the supply of organic matter (OM) has increased significantly and a large amount of OM are buried on the Arctic shelf. Studying the fate of OM in Arctic shelf sediments is crucial to understanding the global carbon sink. As a marginal sea of the Arctic Ocean, the Chukchi Sea is one of the most critical areas where OM is buried. Based on the surface sediment samples collected during the sixth Chinese National Arctic Research Expedition in the summer of 2014 and the Sino-Russian joint Arctic Research Expedition in the summer of 2016, this study takes amino acids (AAs) as the primary tool to explore the source and degradation of OM in the surface sediments of the Chukchi Sea. This study shows that total hydrolyzable amino acid (THAA) concentrations (dry weight) are high, with a mean value of (32.7 ± 15.8) μmol/g. Their spatial distribution is related to primary productivity, hydrodynamic conditions, sediment properties and other factors. The source of OM in the surface sediments of the Chukchi Sea is dominated by diatom-dominated marine productivity, with some input from terrestrial sources. Bacteria, as the main source of the D-enantiomer of AA (D-AA), not only have transforming effect on OM, but their cell walls and remnants likewise supply the OM pool. Based on a series of diagenetic indicators, we conclude that the OM in the surface sediments of the Chukchi Sea has undergone extensive degradation [DI (degradation index) = −0.59 ± 0.44], and the degradation degree in the slope is higher than that in the shelf. This study uses AA to explore the sources and degradation of OM in the sediments of the Chukchi Sea, which facilitates our understanding of OM transport and transformation on the Arctic shelf.

Key words: Chukchi Sea, amino acids, degradation indicator, organic matter

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1 Introduction

In recent decades, global CO_2 emissions have increased, and a series of environmental changes caused by climate warming are particularly evident in the Arctic Ocean (Stroeve et al., 2005; Overland et al., 2014). The degradation of permafrost and the intensification of coastal erosion have greatly increased the input of

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terrigenous organic carbon (Schädel, 2022; Terhaar et al., 2021). In addition, the level of primary productivity in the Arctic Ocean has increased significantly due to rising sea temperatures and receding sea ice (Arrigo et al., 2008; Lawrence et al., 2008). The Arctic has the most extensive continental shelf, accounting for 20% of the global continental shelf. The organic matter (OM) buried on the Arctic continental shelf accounts for more than 70% of the entire Arctic Ocean, and it is an important carbon sink area (Stein and MacDonald, 2004; Chen et al., 2015).

The Chukchi Sea is an important marginal sea of the Arctic continental shelf, serving as a gateway connecting the Arctic Ocean and the Pacific Ocean. The Pacific inflow water can transport 15×10^6–90×10^6 t of suspended matter to the Chukchi Sea every year (Coachman et al., 1975). In addition, the Chukchi Sea is the most productive sea area in the Arctic shelf. The average annual productivity (in terms of C) is 350–500 g/(m^2·a), with peak productivity of up to 800 g/(m^2·a) (Cota et al., 1996; Hill and Cota, 2005; Mathis et al., 2009). In recent decades, the Chukchi Sea has experienced rapid sea ice retreat, leading to increased primary production (Woodgate, 2018; Su et al., 2022). Increased Pacific water inflow (PWI) brings more nutrients into the Chukchi Sea and may also change the structure and size of the phytoplankton community (Zhuang et al., 2016; Ren et al., 2020). OM from upper waters, after settling and transformation, is ultimately preserved in sediments. Biogenic indicators in sediments provide important information for understanding the origin, degradation and preservation of OM. Currently, a large number of biogenic indicators have been studied in the Chukchi Sea, including fatty acids, chlorin and lipid biomarkers (Li et al., 2020; Zhang et al., 2015; Bai et al., 2010). However, these biogenic indicators, which are chemically conservative, indicate the source of OM but fail to reflect the freshness of the OM. The freshness of the OM can have an impact on biomass and nutrient structure, further affecting the structure and efficiency of the biological pump (Gutiérrez et al., 2000; Venturini et al., 2011).

Amino acids (AAs), as important components of organic carbon and organic nitrogen, are abundant in OM with high biological activity. They act as the most labile fraction of OM and serve as indicators for studying the origin and freshness of OM (Dauwe and Middelburg, 1998; Kaiser and Benner, 2008). Due to the labile nature of AAs, their content and composition can change during the degradation of OM. Several diagenetic indicators are established based on this, such as carbon and nitrogen normalized yields of total hydrolyzable AA (THAA-C% and THAA-N%) and the degradation index (DI), which can be used to characterize the degradation state of OM (Cowie and Hedges, 1992; Suthhof et al., 2000; Davis et al., 2009). In addition, bacteria play a crucial role in the transformation of OM in the marine environment (Kawasaki and Benner, 2006). Because the D-enantiomer of AAs (D-AAs) exists almost exclusively in bacterial cell wall peptidoglycan, it can be used as a biomarker for bacteria, and its remnants in sediments are useful for characterizing the bacterial degradation of OM (Kaiser and Benner, 2008; Bourgoin and Tremblay, 2010). Bacteria have a transforming effect on OM. They can convert L-enantiomer of AAs (L-AAs) to D-AAs by racemase or synthesize free D-AAs directly (Zhang et al., 2016). Non-protein AAs such as GABA (γ-aminobutyric) may be formed by the decarboxylation of AAs by microorganisms, and the degree of processing of OM by microorganisms can be inferred through the analysis of changes in their content or ratio (Lee and Cronin, 1982). The synthesis, deposition and burial of AAs in the ocean constitute important transport and transformation processes in the carbon and nitrogen cycles in the ocean. Therefore, the systematic study of AAs and related parameters to investigate the source, degradation level, bioavailability, and contribution and role of microbial activity in processing of OM is important for assessing the fate of OM in the ocean.

Currently, there are relatively few studies on AAs in the surface sediments of the Chukchi Sea, the determination and analysis of the D-AAs are lacking, and relevant data are missing for the western side of the Chukchi Sea (Wang et al., 2008). Therefore, in this study, we determined the L and D enantiomers of AAs, analyzed the composition and distribution characteristics of AAs in the surface sediments of the Chukchi Sea, and investigated the source and degradation state of OM in conjunction with the basic parameters of total organic carbon (TOC), carbon stable isotopes (δ^{13}C), and grain size in the sediments, which provide a basis for understanding the transport and transformation of sedimentary organic matter (SOM) in the Arctic.

2 Materials and methods

2.1 *Study area*

The Chukchi Sea is a typical marginal sea in the Arctic, with an area of approximately 6.2×10^5 km^2, and the continental shelf area accounts for 22% of the total area of the Arctic Ocean shelf. It is one of the Arctic regions undergoing the fastest changes and largest sea ice retreat, with the highest comprehensive primary productivity (Hill and Cota, 2005; Arrigo and van Dijken, 2011). The Chukchi Sea is connected to the North Pacific Ocean through the Bering Strait, bordering Alaska to the east, Siberia to the west, and the Canadian Basin with a water depth of more than 4 000 m to the north.

The PWI is divided into three branches flowing through the Bering Strait into the Chukchi Sea: the east branch is the high-temperature, low-salinity and low-nutrient Alaskan Coastal Water (ACW), which turns east through Barrow Canyon; to the west is the low-temperature, high-salinity and high-nutrient Anadyr Water (AW), which flows northward through Herald Canyon. The middle branch is Bering Shelf Water (BSW), with salinity between that of ACW and AW (Grebmeier et al., 2006; Jones et al., 1998).

2.2 *Sample collection*

Surface sediments were collected during the sixth Chinese National Arctic Research Expedition in the summer of 2014 and the Sino-Russian joint Arctic Research Expedition in the summer of 2016. A total of 22 samples were analyzed in this study, covering the area from the shelf to part of the slope of the Chukchi Sea, at water depths ranging from 33 m to 439 m (Table 1). The surface sediments (0–2 cm) were sampled with a box-corer, placed in plastic bags, and stored in a refrigerator at –20℃, then freeze-dried in the laboratory for analysis. Specific sampling stations are shown in Fig. 1. The sedimentation rates in most of the study area ranged from 0.03 cm/a to 0.37 cm/a, corresponding to approximately 2 cm of surface sediments representing roughly 5 a to 70 a (Cooper and Grebmeier, 2018; Kim et al., 2019), so the effect of sampling year on the samples in this study was negligible.

2.3 *Amino acid analyses*

According to the method described by Fitznar et al. (1999), THAA is pre-column derivatized with O-phthalaldehyde (OPA) and N-isobutyryl-L-cysteine (IBLC), and AA is subsequently measured in the hydrolyzate using high-performance liquid chromatography (HPLC, Agilent 1100). The pretreatment steps of THAA determination refer to the method established by Zhu et al. (2016). In brief, approximately 10 mg of ground and freeze-dried sediments is hydrolyzed with HCl in pre-combusted glass

Table 1. Location, water depth, THAA content, OC content, specific surface area, carbon stable isotopes, THAA-C and DI of the Chukchi Sea surface sediments

Sample station	Latitude/°N	Longitude/°E	Water depth/m	THAA/(μmol·g^{-1})	OC/%	SSA/(m^2·g^{-1})	δ^{13}C/‰	THAA-C/%	DI
Shelf sediments									
LV77–2	68.57	190.09	52.70	51.5	1.68	15.68	−22.4	15.63	−0.20
LV77-3	68.88	187.85	51.00	44.4	2.03	22.11	−21.9	10.75	−0.40
LV77-4	69.20	185.09	48.70	66.1	2.64	27.08	−21.5	12.31	−0.27
LV77-5	69.71	186.79	50.60	49.2	1.93	22.25	−21.8	12.83	−0.34
LV77-6	72.20	186.38	52.72	44.2	2.08	29.61	−21.7	10.64	−0.26
LV77-7	71.18	186.51	43.00	22.1	0.98	12.97	−22.7	11.24	−0.48
LV77-8	69.59	182.52	47.00	32.2	1.00	–	−22.2	15.97	−0.36
LV77-9	69.60	179.85	44.71	22.8	0.53	12.06	−22.7	21.45	−0.25
R02	67.67	−169.00	50.00	34.0	–	–	–	–	−0.51
R03	68.62	−169.00	53.70	49.1	1.11	12.17	−22.7	22.62	−0.06
R05	71.00	−169.00	44.00	23.0	–	–	–	–	−0.54
R06	72.00	−168.98	51.35	43.5	1.47	20.29	−22.1	14.68	−0.53
R07	73.00	−168.97	73.76	46.2	1.47	22.60	−22.0	15.68	−0.38
R08	74.00	−169.00	82.69	29.8	1.27	26.53	−22.3	11.42	−1.12
S02	71.92	−157.46	73.00	48.3	1.72	25.42	−22.7	14.14	−0.49
C01	69.22	−168.14	50.00	30.4	0.89	13.82	−23.6	16.99	−0.54
C03	69.03	−166.48	33.00	18.7	1.06	4.05	−24.4	8.83	−0.40
C05	70.76	−164.74	33.00	3.1	0.10	–	−23.2	15.86	−0.21
Slope sediments									
R09	74.61	−169.03	190.00	20.6	0.86	29.59	−21.9	11.55	−1.15
R10	75.43	−167.90	164.36	13.2	0.56	26.70	−23.8	11.33	−1.55
R11	76.15	−166.20	352.43	18.0	0.79	37.45	−21.6	10.78	−1.62
R12	77.00	−163.89	438.86	9.6	0.50	24.32	−22.7	9.32	−1.33

Note: – represents no data.

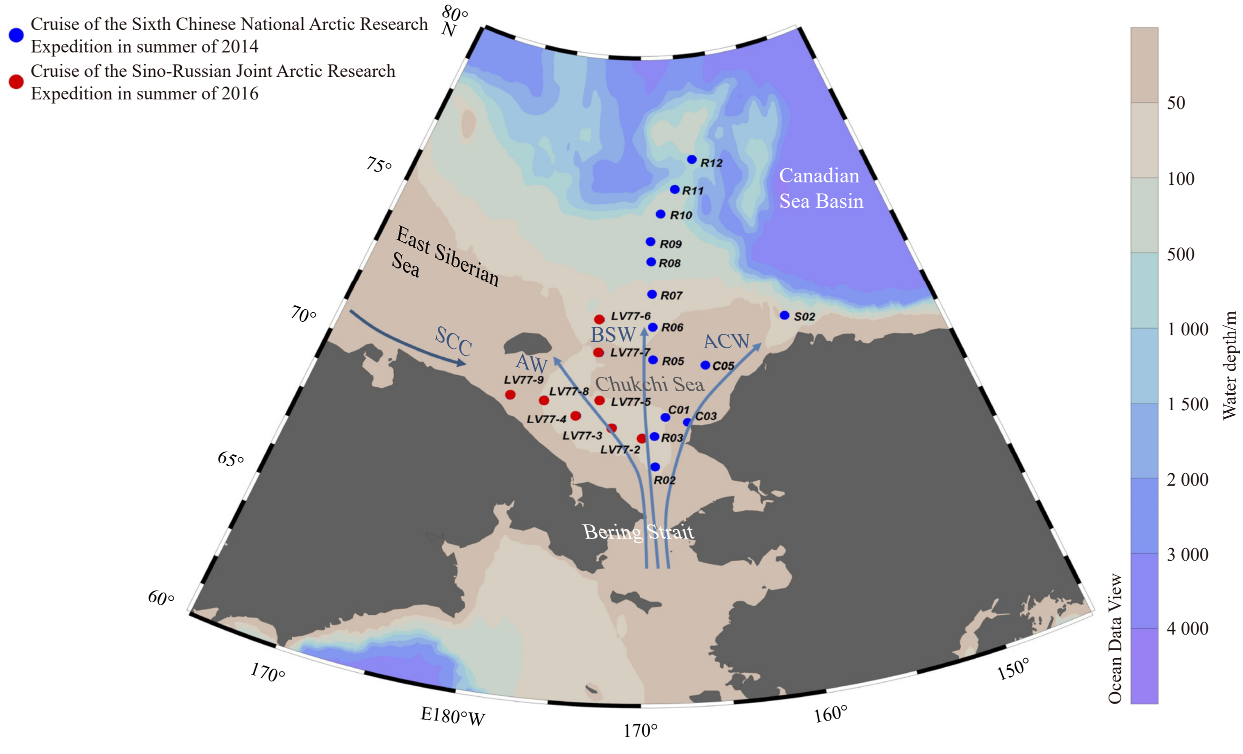

Fig. 1. Map of the Pacific-Arctic oceans showing the sample stations used in this study. Main surface currents are shown. ACW: Alaskan Coastal Water; BSW: Bering Shelf Water; AW: Anadyr Water; SCC: Siberian Coastal Current.

ampoules (450℃, 5 h), sealed in a nitrogen environment and incubated at 110℃ for 24 h in an oven. The cooled samples are neutralized with boric acid buffer and NaOH solution to adjust the pH to 8.5, and then the supernatant is taken for measurement. The detector is a fluorescence detector (excitation λ_{ex} = 330 nm, emission λ_{em} = 445 nm), the chromatographic column is a Phenomenex Hyperclone column (BDS C18, length 250 mm, inner diameter 4mm, filler diameter 5 μm), and the mobile phases are methanol (*A*) and sodium acetate (*B*). Final assay results are corrected for dilution and racemization (Kaiser and Benner, 2005). Asx and Glx are used to denote Asp + Asn and Glu + Gln, respectively, because the corresponding acids are formed by deamination during hydrolysis. Except for AAs such as Asp and Glu whose recovery rate was higher than 100% due to conversion during hydrolysis, the recovery rate of most AAs ranged from 75% to 100%.

2.4 *Other parameters*

Other parameters used in this study, such as specific surface area (SSA), organic carbon (OC), and carbon stable isotopes (δ^{13}C), are derived from published data in the literature (Ye et al., 2021; Wang et al., 2017; Li et al., 2017). SSA was determined by nitrogen adsorption using a surface area and porosity analyzer (Ye et al., 2021). The samples were acidified, dried, and analyzed using an elemental analyzer for OC and an isotope mass spectrometer for δ^{13}C.

2.5 *Diagenetic indicators*

Diagenetic indicators used in this study include the following:

(1) Carbon normalized yield of AA (THAA-C%), defined as the proportion of OC comprising THAA. The calculation is as follows:

$$\text{THAA-C\%} = (C_{\text{THAA}}/\text{OC}) \times 100\%. \tag{1}$$

(2) Degradation index (DI) is a parameter obtained by the principal component analysis (PCA) based on the compositional changes of AA and can be used to indicate the degree of degradation of organic matter. The calculation is as follows:

$$\text{DI} = \sum_i \left[\frac{\text{var}_i - \text{AVG}_{\text{var}_i}}{\text{STD}_{\text{var}_i}} \right] \times \text{fac.coef}_i, \tag{2}$$

where var_i is the molar percentage of AA_i in our data set, and AVG_i and STD_i are the mean and standard deviation of AA_i, respectively, and fac.coef_i is the factor coefficient for AA_i (calculated from PCA analysis) (Dauwe et al., 1999).

3 Results

3.1 *Composition and distribution of THAA*

The distribution of THAA in the surface sediments of the Chukchi Sea is shown in Fig. 2a. THAA concentrations (dry weight) range from 3.1 μmol/g to 66.1 μmol/g, with a mean value of (32.7 ± 15.8) μmol/(g·dw). The highest value was measured at station LV77-4 (66.1 μmol/g) on the western side of the Chukchi Sea, while the lowest one was measured at Station C05 (3.1 μmol/g) near the Alaska coast. In general, the distribution of THAA varies widely in the shelf, with THAA values on the western side being higher than those on the eastern side, and with hot spots occurring in the region between 72°N and 74°N. The THAA concentrations in the slope are significantly lower than those in the shelf, with a more stable distribution.

The composition of THAA in the surface sediments of the Chukchi Sea slope and the shelf is similar, with Gly accounting for the highest molar percentage, followed by Asx (Asp+Asn), Ala, Ser, Glx (Glu+Gln), and Thr. In addition, Leu, Lys, Ile, and Phe each account for a lower percentage (less than 5% on average); Tyr, Met, and GABA each account for a very low percentage of about 1% (Fig. 2b). Among these, the proportions of Ala, Arg, Thr, and GABA are significantly higher in the slope than in the shelf, while Gly, Leu, Phe, Ile, and Lys have relatively higher proportions in the shelf than in the slope.

3.2 *Range of diagenetic indicators*

THAA-C% in the surface sediments of the Chukchi Sea ranges from 8.83% to 22.62% with a mean value of 13.70% ± 3.62%. The highest value is found at Station R03 on the south side of the Chukchi Sea (22.62%), while the lowest value is found at Station C05 near the Alaskan coast (8.83%). THAA-C% is relatively low in the slope and has its distribution varies greatly in the shelf (Fig. 3a).

The DI values range from −1.62 to −0.06 with a mean value of −0.59 ± 0.44. The highest value is found at Station R03 in the shelf (−0.06) while the lowest one is recorded at Station R11 in the land slope (−1.62). The distribution trend of DI values is different from that of THAA-C%, with a clear decreasing trend with increasing latitude (Fig. 3b). In addition, the D/(D+L)AA value, which represents the molar ratio of the D-enantiomer to the D and L-enantiomers of AAs (total AAs), ranges from 0.04 to 0.13, with an average value of 0.06 ± 0.03, and shows an upward trend with increasing latitude (Fig. 3c).

4 Discussion

4.1 *Factors influencing the composition and distribution of THAA*

AAs in marine sediments mainly originate from the depos-

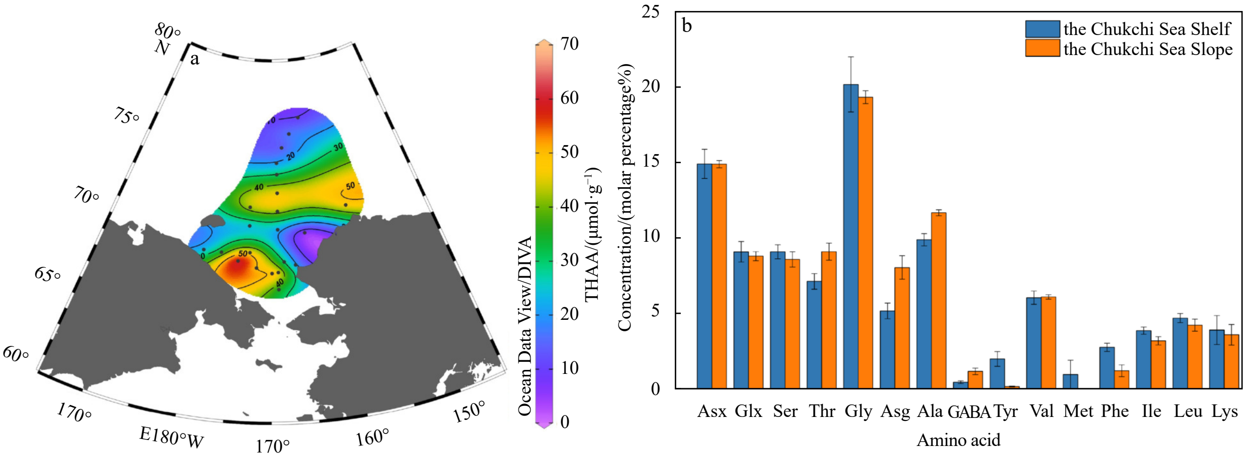

Fig. 2. Distribution (a) and composition (b) of amino acids in the surface sediments of the Chukchi Sea. Error bars, mean standard deviations.

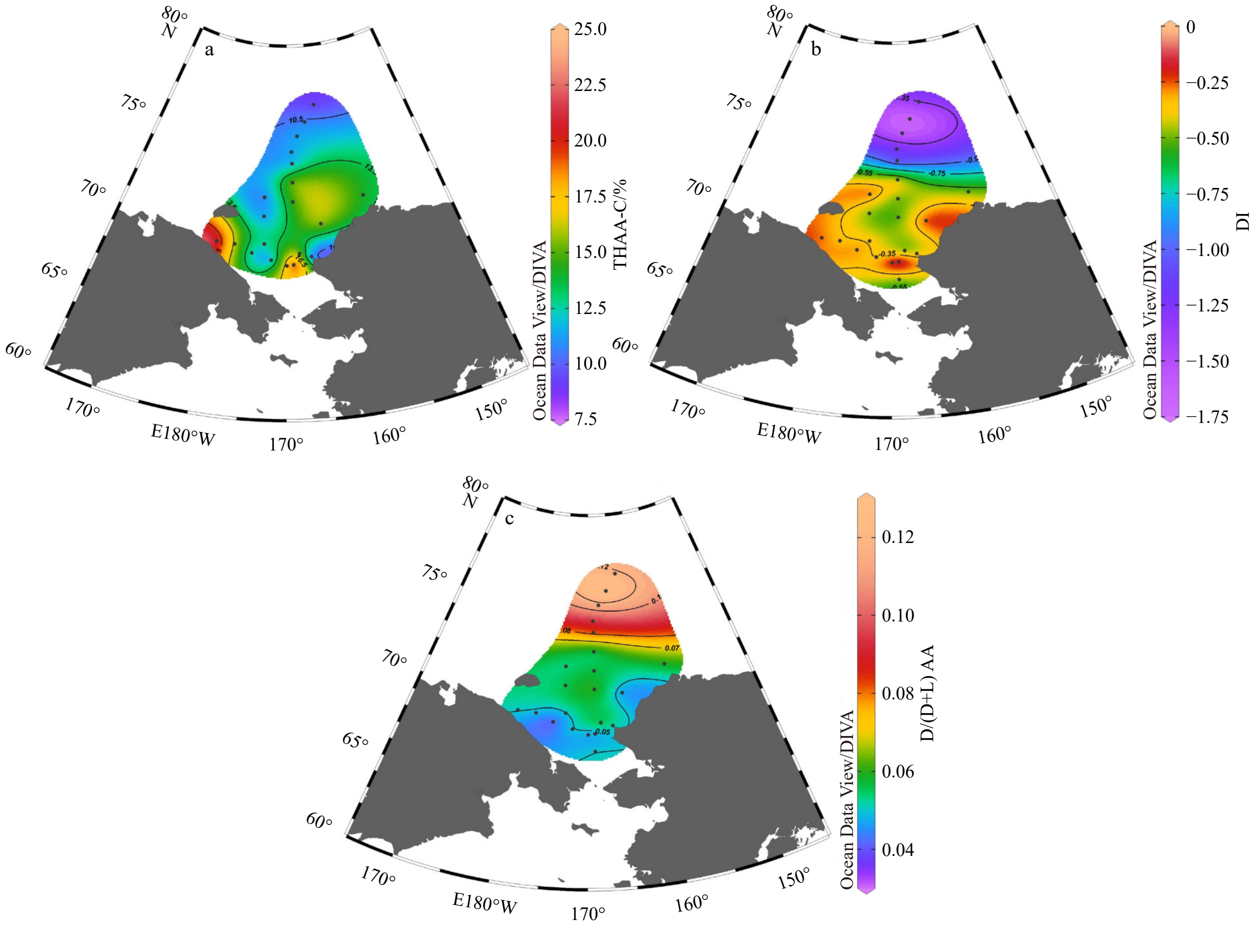

Fig. 3. Variation in THAA-C (%) (a), DI (b), and D/(D+L)AA (c) in surface sediments in the Chukchi Sea.

ition of upper water particles, and their distribution is related to the productivity level of upper water, hydrodynamic conditions, and sediment preservation conditions, with varying degrees of influence from region to region.

The composition of THAA is dominated by Gly, Asp, Ala and Glu, which is consistent with the results reported in most marine sediments (Lomstein et al., 2006; Vandewiele et al., 2009; Chen et al., 2018; Alkhatib et al., 2012). The variation in AA composition may be due to differences in the responsiveness of individual AAs to biological degradation, causing different levels of accumulation (Bourgoin and Tremblay, 2010). The productivity of diatoms in the Chukchi Sea can account for more than 70% of the total primary productivity, and the output efficiency of its sedimentation into sediments can reach 20% to 32% (Moran et al., 2005; Lepore et al., 2007; Yu et al., 2012; Zhuang et al., 2020). Diatom cell walls are rich in Gly, Ser, and Thr, and their enrichment may be related to their close association with biogenic silica in diatom cell walls and diatom shells, thus protecting them from degradation (Lee et al., 2000; Ingalls et al., 2003).

The PWI carries large amounts of nutrients through the narrow Bering Strait to the Chukchi Sea that gradually settle onto the Chukchi Sea shelf (Woodgate et al., 2005). The decrease in nutrient supply is caused by the gradual decrease in THAA concentration from south to north in the southern Chukchi Sea. The eastern part of the Chukchi Sea is mainly affected by low-salinity and low-nutrient ACW, while the western side is significantly influenced by high-salinity and high-nutrient AW (Walsh et al., 1989). The differences in nutrients carried by the two flows lead to differences in the productivity between the areas through which they each flow. In the western side of the Chukchi Sea especially, the THAA concentration reaches the highest value (66.1 μmol/g) under the influence of AW. In addition, the receding sea ice and limitation of reduced light during the melting period cause a burst of phytoplankton, and together with the large amount of nutrients carried by PWI, the primary productivity of the Chukchi Sea increases significantly (Stein and Fahl, 2000; Hill and Cota, 2005). Some studies have found that the Chukchi Sea has high productivity and a high particle sinking flux, which is another reason for the large area of high THAA values on the Chukchi Sea shelf (Bates et al., 2005; Moran et al., 2005). Compared with the shelf, the productivity of the Chukchi Sea slope is much lower, less affected by PWI, and OM is degraded to a certain extent during the subsidence process, thus resulting in a significantly lower THAA concentration in the slope.

In addition, AAs in sediments are also affected by sediment grain size. The smaller the grain size of the sediment, the larger the SSA allowing for greater adsorption of OM. As shown in Fig. 4, there is a good correlation between THAA concentration and SSA in the Chukchi Sea shelf and slope, indicating that the adsorption on the sediment surface plays an important role in the distribution of THAA. Although the results of the Chukchi Sea slope are not statistically significant, it can still be found that the sediments in this region, despite having a large SSA, still have a much lower THAA concentration than the shelf, which further indicates a lower productivity leading to a reduced supply of fresh OM from the surface.

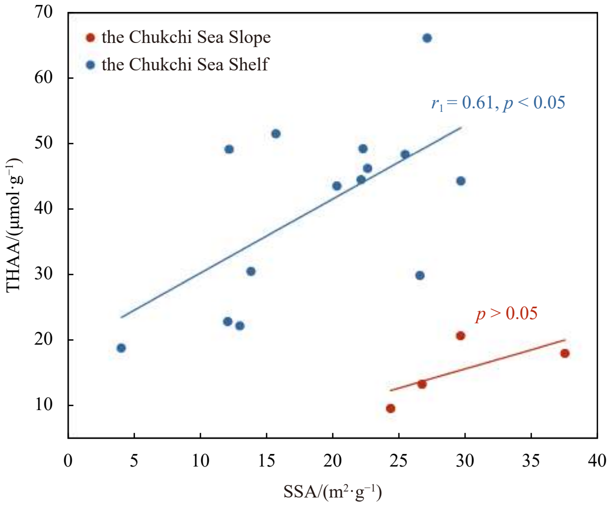

Fig. 4. Correlation between THAA concentration (μmol/g) and SSA (m^2/g) from the Chukchi Sea shelf and slope.

4.2 *Amino acids as indicators of OM sources*

The δ^{13}C‰ values in this study vary between −24.4‰ and −21.5‰ (Fig. 5), which are all within the range of published data in the Arctic Ocean (Goñi et al., 2005; Tesi et al., 2014). As shown in Fig. 5, there is a gradual increase in δ^{13}C‰ with depth in the Chukchi Sea shelf, indicating a gradual decrease in terrestrial sources and a gradual increase in marine sources. The presence of a minimum value of δ^{13}C (−24.4‰) near the coast of Alaska may be the result of coastal erosion leading to some input of terrigenous materials. In addition, although no large rivers are flowing into the Chukchi Sea, terrigenous materials transported by the Yukon River are entrained into the ACW and transported through the Bering Strait to the Chukchi Sea (Macdonald et al., 2002). The high percentage of terrigenous materials present in the Chukchi Sea slope is most likely caused by ice rafting. Terrigenous materials from riverine input or resuspension are caught onto sea ice near the coastal zone and transported to distant shores by drifting sea ice and icebergs, and sink when the sea ice melts (Sakamoto et al., 2005; Xu et al., 2017).

Furthermore, in addition to geochemical parameters, some biomarkers such as AAs also provide further evidence for the origin of OM in sediments. It has been shown that Gly, Ser and Thr are enriched in diatom cell walls, while calcareous organisms are enriched in Asp (Ingalls et al., 2003; Hecky et al., 1973). Asp/Gly ratios and Ser+Thr (molar percentage) can be used to distinguish the OM origin of diatoms and calcareous organisms (Ittekkot et al., 1984). The sediments in the Panama Basin, dominated by calcareous materials, have a high Asp concentration (23.7%) and high Asp/Gly ratio (1.88). However, the diatomaceous material composition of the Drake Passage is characterized by a predominance of Gly (17.7%) and a low Asp/Gly ratio (0.62) (Müller et al., 1986). In this study, the mean value of the Asp/Gly ratio in the surface sediments of the Chukchi Sea is 0.74 ± 0.09 (<1), and the mean value of Ser+Thr is 16.47 ± 0.87. Our data clearly show a predominance of diatomaceous OM in the sediment compared to calcareous bio-derived OM. The results are consistent with previous studies in the Laptev Sea in the Arctic (Asp/Gly = 0.70 ± 0.03, Ser+Thr = 17.5 ± 0.5) (Dittmar et al., 2001). In addition, we found a significant correlation between the Asp/Gly ratio and δ^{13}C‰ ($r = -0.46$, $p < 0.05$), which further confirms the important contribution of diatom-dominated marine productivity to the OM in the Chukchi Sea.

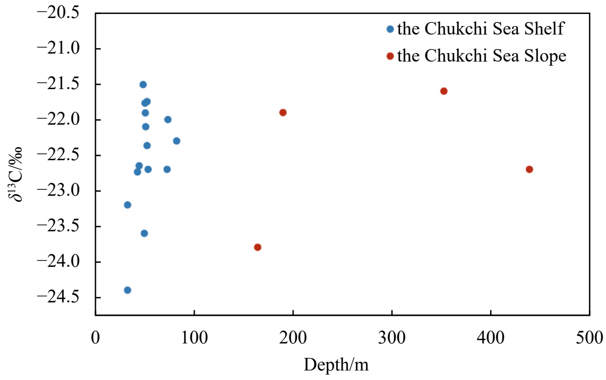

Fig. 5. The relationship between δ^{13}C(‰) and the water depth (m) in the surface sediments of the Chukchi Sea.

Bacterial cell walls and their remnants in sediments have Ala signals (Mayer et al., 1995). The detection of Ala signals in the surface sediments of the Chukchi Sea shelf and slope (around 10%) indicates that bacteria and their remnants are also a source of OM in the sediments. D-AAs are a major component of bacterial cell walls and are not produced by algae or vascular plants (Wu et al., 2007; Chen et al., 2018). Compared with L-AA, D-AA is not easily biodegraded and accumulates during the diagenesis of OM (Nagata et al., 1998; Zhang et al., 2016). The enrichment of D-AA in the surface sediments of the Chukchi Sea similarly demonstrates the contribution of bacteria or OM that has been modified by bacteria to the OM pool.

By combining information from a series of parameters, it can be concluded that the source of OM in the Chukchi Sea is dominated by diatom-dominated marine productivity, with some input from terrestrial sources. Bacteria not only have a transforming effect on OM, but their cell walls and remnants likewise supply the OM pool. However, the specific contribution values are not reflected in the AA results and need to be further described in conjunction with other studies.

4.3 *Diagenetic characteristics of sedimentary OM*

Some AA indicators, such as THAA-C%, THAA-N%, DI, and D/(D+L)AA, are often used to indicate the degradation status of OM in marine sediments (Dauwe and Middelburg, 1998; Wu et al., 2007). AAs have high biological activity and are easily used by microorganisms and preferentially degraded during the degradation process. Therefore, the THAA-C% value usually decreases with the degradation of OM, and is more sensitive to indicating OM in the early diagenetic state (Davis et al., 2009). THAA-C%value is lower in the slope (10.75% ± 0.87%) than that in the shelf (14.44% ± 3.67%) (Fig. 3a). It may be related to the source of OM. The low overlying productivity of the slope leads to a lack of fresh OM input, which reduces the freshness of OM. However, when considering AA changes alone or in simple ratios to reveal degradation of OM, the results are susceptible to environmental factors (Wang et al., 2018).

In addition, because D-AA, a major component of bacterial cell walls, accumulates during OM diagenesis, the D/(D+L)AA values can also indicate the degree of degradation of OM in sediments. It has been shown that D-Ala is present in all bacterial cell walls and can be used as a powerful tracer to estimate the contribution of bacteria to OM (Tremblay and Benner, 2009; Bourgoin and Tremblay, 2010; Kawasaki et al., 2011). GABA is a non-protein AA, which is produced via the bacterial degradation of OM. The strong correlation between D-Ala (%) and GABA (%) also confirms the important role of bacteria in the degradation of OM (Fig. 6). The D/(D+L)AA values in the surface sediments of the Chukchi Sea gradually increase from south to north, indicating a gradual increase in the degree of OM degradation (Fig. 3c).

The DI is used to characterize the degradation status of OM in particulate matter and sediments by using the fact that the composition of AA changes during the degradation of OM. More negative DI values indicate a higher degree of OM degradation (Dauwe and Middelburg, 1998; Alkhatib et al., 2012). The DI values in the surface sediments are all less than 0, indicating a high degree of OM degradation in the Chukchi Sea. The mean DI value in the slope of the Chukchi Sea is −1.41 ± 0.19, which is significantly lower than that in the shelf (0.41 ± 0.22) (Fig. 3b). This is due to its relatively low productivity and less supply of OM, coupled with the transport of OM to the slope and the more adequate degradation during subsidence in deep water, resulting in a high degree of OM degradation in sediments in this region. Many studies have confirmed that sediment grain size is an important factor affecting the degradation of sedimentary OM (Zhang et al., 2012; Bao et al., 2016; Chen et al., 2021). DI was negatively correlated with the SSA of sediments (Fig. 7a). This indicates that the finer grains have a higher degree of degradation, which may be related to hydrodynamic sorting processes. The narrow and shallow terrain near the Bering Strait makes the current velocity increase here, and the hydrodynamic disturbance of the sediment is stronger (Qiu et al., 2007). Area with strong hydrodynamic has higher scouring and transport capacity. Fresh biogenic coarse grains brought from the Bering Sea, which are rich in nutrients, are trapped here and exhibit low OM degradation (Wang et al., 2008). The fine grains are transported in suspension to the far shore, gradually settling and further degrading. In addition, the drift of sea ice can also transport the fine grains entrained in the sea ice by resuspension to the far shore (Eicken et al., 2005). Although the fine grains have more obvious sea-source characteristics (Fig. 7b), during the resuspension and transportation of sediments, the terrestrial OM adsorbed on the fine-grained sediments degrades more slowly than the sea-source OM and can be dispersed and accumulated more effectively (Zonneveld et al., 2010; Hu et al., 2012). In addition, a strong negative correlation is also observed between DI and D-Ala% (Fig. 7d). This implies a close link between the degradation status of OM and the degree of bacterial processing. Bacteria are more likely to attach to fine-grained sediments (Fig. 7c), which further explains the high degree of degradation of fine grains.

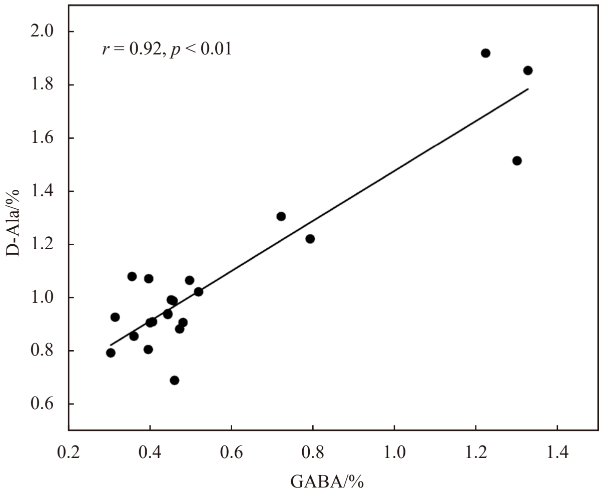

Fig. 6. Correlation between D-Ala(%) and GABA(%) in the surface sediments of the Chukchi Sea.

Several indicators in this study exhibit somewhat different distributions because different degradation indicators apply to different stages of OM diagenesis. For example, THAA-C% is more sensitive in the early degradation of OM, but the results are easily affected by environmental biases, while DI values are more applicable to the middle and late stages of OM formation, but may be affected by the source of OM. Therefore, the study of the degradation state of OM in the surface sediments of the Chukchi Sea requires a comprehensive comparative analysis using various indicators. In general, the OM in the Chukchi Sea shelf in this study is fresher than that in the slope and has a greater relationship with the high primary productivity overlying it. Fresh OM is not only a potential nutrient supply for benthic organisms, but also affects the structure and efficiency of Arctic biological pumps, reflecting the conservation status of OM. In the future, due to reduced light confinement and increased inflow of warm Pacific water, increased productivity of the Chukchi Sea will have more supply of fresh OM In addition, the Chukchi Sea shelf water will heat up, enhancing bacterial activity, which in turn accelerates the degradation of fresh OM and releases more CO_2 back into the water column (Palmer et al., 2014; Grebmeier et al., 2018; Meiners et al., 2008). AAs, as indicators to explore the source and freshness of OM, are expected to be used in combination with other indicators in future research on the fate of OM in the Arctic Ocean and its response to environmental changes.

5 Conclusions

This study shows that THAA concentrations (dry weight) in the surface sediments of the Chukchi Sea are high with a mean value of (32.7 ± 15.8) μmol/g. The THAA concentration on the west side of the Chukchi Sea is higher than that on the east side, with higher values in the shelf than in the slope. Its distribution is related to the productivity level, hydrodynamic conditions, sediment properties and other factors of the sea. The source of OM in the surface sediments of the Chukchi Sea is dominated by diatom-dominated marine productivity, with some input from terrestrial sources. Bacteria, as the main source of D-AA, not only have transforming effect on OM, but their cell walls and remnants likewise supply the OM pool. They are an important regulator in the process of SOM transport and transformation. Combining a series of diagenetic indicators, we conclude that the OM in the surface sediments of the Chukchi Sea has undergone extensive degradation (DI = −0.59 ± 0.44), with the degree of degradation being higher in the slope than in the shelf. The degree of OM degradation is related to multiple factors such as overlying primary productivity, bacterial transformation, and sediment properties. In this study, AAs are used to explore the source and degradation degree of OM in the sediments of the Chukchi Sea, which provide certain indications for understanding the migration and transformation of OM on the Arctic shelf.

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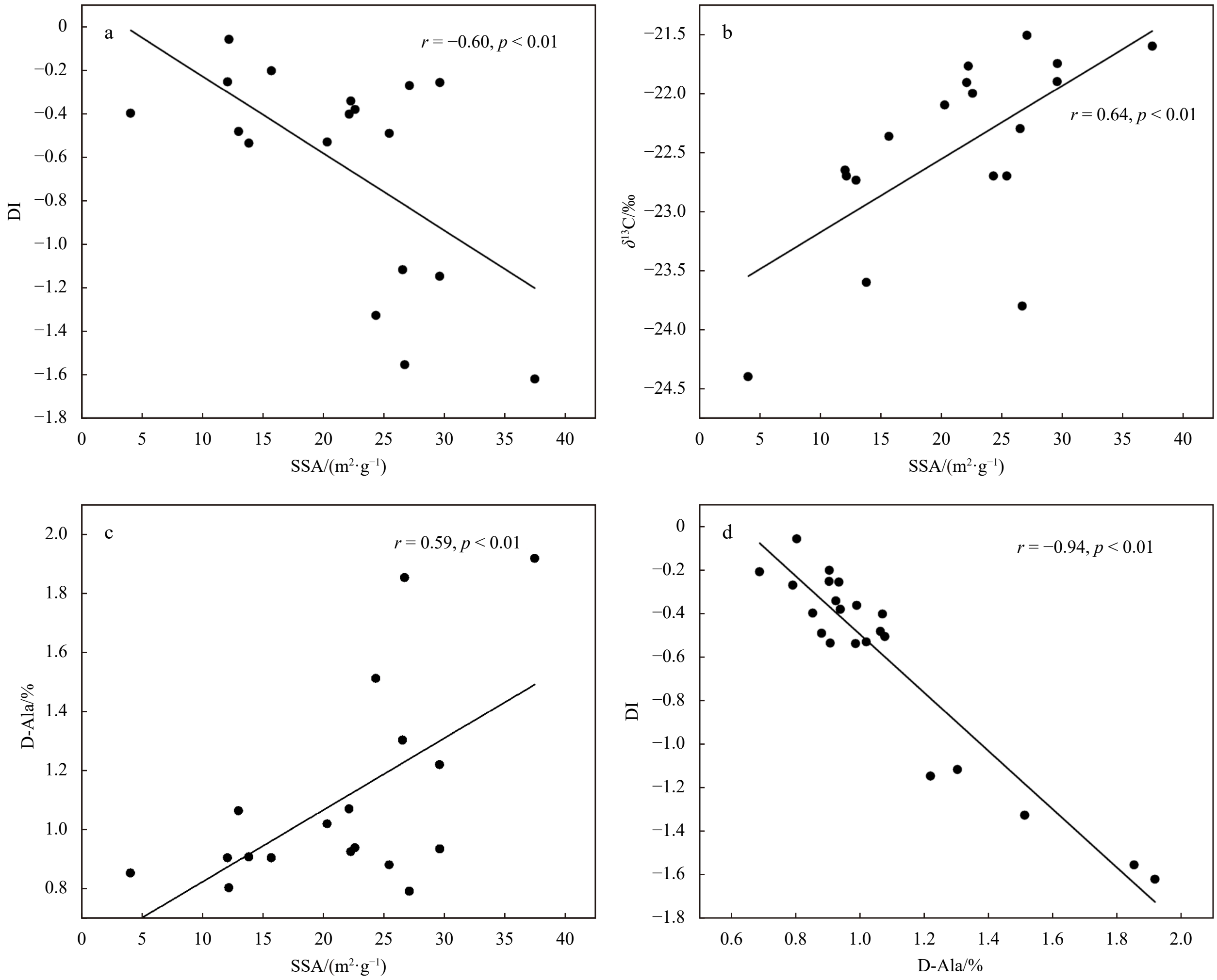

Fig. 7. Correlation between SSA (m^2/g) and DI (a), $\delta^{13}C$ (‰) (b), and D-Ala (%) (c), respectively, and D-Ala (%) vs. DI (d) in the surface sediments of the Chukchi Sea.

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