

Organic carbon accumulation and sulfate reduction rates in slope and basin sediments of the Ulleung Basin, East/Japan Sea

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Abstract This study investigated the organic carbon accumulation rates (OCARs) and sulfate reduction rates (SRRs) in slope and basin sediments of the Ulleung Basin, East/Japan Sea. These sediments have high organic contents at depths greater than 2,000 m; this is rare for deep-sea sediments, except for those of the Black Sea and Chilean upwelling regions. The mean organic carbon to total nitrogen molar ratio was estimated to be 6.98 in the Ulleung Basin sediments, indicating that the organic matter is predominantly of marine origin. Strong organic carbon enrichment in the Ulleung Basin appears to result from high export production, and low dilution by inputs of terrestrial materials and calcium carbonate. Apparent sedimentation rates, calculated primarily from excess ^{210}Pb distribution below the zone of sediment mixing, varied from 0.033 to 0.116 cm year^{-1} , agreeing well with previous results for the basin. OCARs fluctuated strongly in the range of 2.06–12.5 $\text{g C m}^{-2} \text{year}^{-1}$, these rates being four times higher at the slope sites than at the basin sites. Within the top 15 cm of the sediment, the integrated SRRs ranged from 0.72 to 1.89 $\text{mmol m}^{-2} \text{day}^{-1}$, with rates approximately twice as high in the slope areas as in the basin areas. SRR values

were consistently higher in areas of high sedimentation and of high organic carbon accumulation, correlating well with apparent sedimentation rates and OCARs. The sulfate reduction rates recorded in the basin and slope sediments of the Ulleung Basin are higher than those reported for other parts of the world, with the exception of the Peruvian and Chilean upwelling regions. This is consistent with the high organic carbon contents of surface sediments of the Ulleung Basin, suggesting enhanced organic matter fluxes.

Introduction

In a marine benthic environments, interactions between sediments and water play a fundamental role in the biogeochemical cycling of elements. One important reaction is the mineralization of organic matter during early diagenesis, occurring primarily through biological processes. Organic matter is oxidized by the oxidant yielding the greatest free energy change per mole of organic carbon oxidized (Froelich et al. 1979; Berner 1980). Most organic matter is mineralized in the uppermost sediment layer, with oxygen, nitrate, Mn-oxide, Fe-oxide, or sulfate acting as electron acceptors. Oxidic respiration and sulfate reduction account for roughly half of the total organic matter decomposition in most continental margin sediments, with nitrate and metal oxide reductions playing only minor roles (Jørgensen 1982; Slomp et al. 1997).

In deep-sea sediments where organic matter input is low, oxygen may diffuse down several centimeters, making aerobic decomposition the primary reaction (Reimers and Smith 1986; Jahnke et al. 1989). Nitrate reduction is minimal because of the low concentration of nitrate in the overlying water and porewater (Bender and Heggge 1984). Sulfate concentration is generally high (about 30 mM) in

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the porewater of marine sediments (Capone and Kiene 1988); this is used as an electron acceptor by sulfate-reducing bacteria for organic carbon oxidation. Therefore, bacterial sulfate reduction is a dominant process of anaerobic organic matter decomposition in marine sediments. Sulfate reduction generally accounts for more than 50% of the total organic carbon oxidation in continental margin sediments (Bender and Heggie 1984; Reimers et al. 1992; Canfield et al. 1993). Westrich and Berner (1984) demonstrated that the sulfate reduction rate (SRR) is ultimately dependent on the quantity and quality of organic carbon undergoing mineralization. They also reported that the organic carbon accumulation rate was a primary controlling factor of the rate of sulfate reduction in marine sediments. Schubert et al. (2000) showed that high SRRs were positively correlated with the availability of labile marine organic matter. On the other hand, dilution with less-reactive, terrestrially derived organic matter significantly altered SRR depth profiles.

The East/Japan Sea (hereafter, East Sea) is a typical marginal sea in which a large portion of organic carbon is deposited and utilized in the sediments. However, little information is available regarding organic carbon mineralization in this region. Furthermore, sulfate reduction has not been previously studied in the East Sea. Information on organic carbon deposition and sulfate reduction may help clarify organic carbon cycling in East Sea sediments.

Within this context, the aims of this study were to estimate organic carbon accumulation rates and sulfate reduction rates in slope and basin sediments of the Ulleung Basin, East Sea, and to investigate the role of organic carbon deposition in controlling sulfate reduction.

Study area

The East Sea is a semi-enclosed marginal sea between continental Asia and the Japanese archipelago, connected to the open ocean through straits 12–140 m in depth (Chough et al. 2000). The Ulleung Basin, located in the southwestern part of the East Sea, is surrounded by the continental slopes of the Korean Peninsula, the southwestern part of Japan's main island of Honshu, and the Ulleung and Dok islands (Fig. 1). The northern and western margins of the basin are steep ($>10^\circ$), whereas the southern and eastern margins have gentler slopes ($1\sim 2^\circ$) and a broad continental shelf (Park et al. 2005). The basin floor lies at depths of 2,000–2,300 m, the boundary between the continental slope and the Ulleung Basin being approximately 2,000 m in depth. Recent sediments appear to be primarily hemipelagic (Chough 1983); no major rivers drain into the basin along the coast of the Korean Peninsula (Ikehara et al. 1994; Hong et al. 1997).

Materials and methods

Sediment samples were collected using a box corer at sites on the western (site D1) and southern (site B4) slopes, and in the deep basin (sites D2 and D4; Fig. 1). Sampling took place in the period 19 July–1 August 2005, from aboard the R/V *Eardo*. Sediment samples for geochemical analyses were collected from the box cores by means of acrylic core samplers (8 cm i.d.) that were then immediately sealed with polyethylene stoppers. The core samples were transferred to the laboratory onboard the research vessel, where they were sectioned at intervals of 1 or 2 cm and transferred to acid-cleaned polyethylene bottles that were sealed with caps. The sediment samples for measuring organic carbon content, nitrogen content, sediment density, water content, and ^{210}Pb activity were stored at 4°C .

Organic carbon content was determined as the difference between total carbon, measured by a Carlo Erba NA 1500 nitrogen–carbon–sulfur analyzer, and inorganic carbon measured by a Coulometrics inorganic carbon analyzer. Sediment dry bulk density was determined by means of a Micromeritics AccuPyc 1330 density analyzer, and porosity was calculated from water content.

The sulfate reduction rate was determined using the radiotracer method of Jørgensen (1978). Triplicate intact cores (30 cm length and 2 cm i.d.) were collected from the box corer. Then, $2\ \mu\text{Ci}$ of $^{35}\text{SO}_4^{2-}$ was injected at 1-cm intervals, and the cores were incubated for 2 h at in situ temperature. Thereafter, the sediment was sliced into sections, fixed in Zn acetate (20%), and frozen until

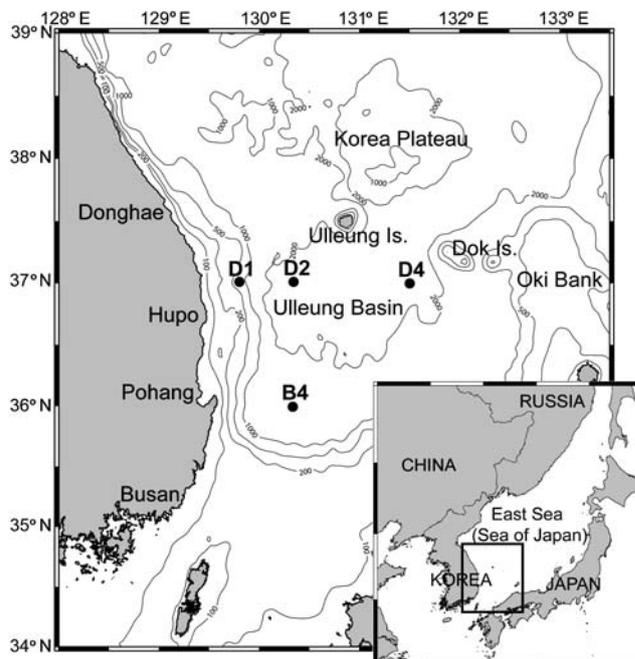


Fig. 1 Locations of the sampling sites (circles) in the Ulleung Basin. Depth contours are given in meters

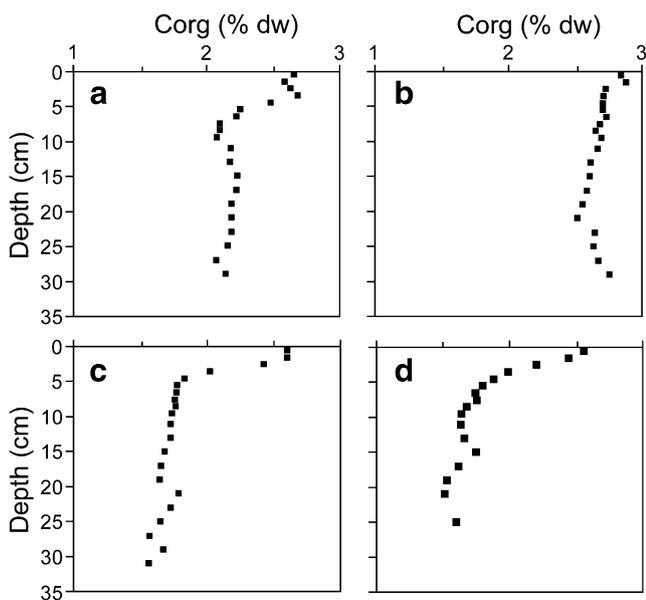


Fig. 2 Downcore profiles of organic carbon contents (% dry weight) at sites B4 (a) and D1 (b) in slope sediments, and sites D2 (c) and D4 (d) in basin sediments

processed in the laboratory. The reduced ^{35}S was recovered by distillation with boiling acidic Cr^{2+} solution, according to the method of Fossing and Jørgensen (1989). All measurements were conducted in triplicate, and standard deviations calculated. Background radioactivity of ^{35}S was $117 \pm 28 \text{ cpm cm}^{-3}$ ($n=8$); SRR detection limits, estimated from the double standard deviation of the blank value (i.e., 56 cpm) according to Fossing et al. (2000), ranged from 3.47 to $4.31 \text{ nmol cm}^{-3} \text{ day}^{-1}$.

Aliquots of dried samples were ground and spiked with a known quantity of ^{208}Po to determine analytical accuracy. These samples were repeatedly leached and dried (three times) in concentrated HNO_3 and $^{6}\text{N HCl}$. Dilute HCl was then added, and the solution separated from the residual solids by centrifugation. For each sample, the dissolved polonium isotopes were plated onto a silver planchet. ^{210}Pb activity was determined by measuring the alpha activity of its granddaughter, ^{210}Po , using a silicon-surface-barrier detector coupled to a multi-channel analyzer. All measurements were conducted at the Korea Basic Science Institute, Daejeon, South Korea.

Calculation of sedimentation rate from ^{210}Pb profiles

Apparent sedimentation rates (ASRs) were calculated primarily from the excess ^{210}Pb distribution below the zone of mixing, assuming that sediment flux and the associated excess ^{210}Pb activity have remained reasonably constant on decadal to centennial timescales in the Ulleung Basin. In the surface sediment mixed layer (SML), sediment mixing is assumed to be restricted to the layer of constant ^{210}Pb

activity (DeMaster et al. 1985; Park et al. 1999). For the zone beneath the SML, plotting the excess ^{210}Pb activity as a function of depth gives a curve with a slope proportional to the rate of change in age with depth, or sedimentation rate. The exact relation is $S = -\lambda/b$, where S is the sedimentation rate (cm year^{-1}), λ the decay constant of ^{210}Pb (0.0311 year^{-1}), and b the slope of the curve. Sedimentation rates calculated from profiles of excess ^{210}Pb activity often overestimate actual accumulation rates, because the gradual decrease in mixing efficiency with increasing depth results in a depth profile indicating exponential decay (Niggemann et al. 2007), which can be falsely interpreted as undisturbed sediment accumulation. In the present case, independent measurements of sediment mass accumulation rates were available from an earlier study (1999) using sediment traps in the same region (see below).

Results

On the southern slope (site B4), organic carbon contents were roughly constant at approximately 2.7% in the uppermost 5 cm, and decreased to 2.1% at 10 cm, showing a relatively steady downcore decrease (Fig. 2a). On the western slope (site D1), organic carbon contents varied between 2.5% and 2.9%, with a relatively steady downcore decrease (Fig. 2b). In the basin samples (sites D2 and D4), organic carbon values decreased sharply from 2.6% to 1.7% in the uppermost 5 cm, followed by slight decline below 5 cm depth (Fig. 2c,d). For basin samples, organic carbon content was well correlated with total nitrogen content (Fig. 3; $R^2 = 0.94$), with a mean organic carbon to total nitrogen (OC/TN) molar ratio of 6.98.

Site B4 displayed a subsurface SRR peak of $31.0 \text{ nmol cm}^{-3} \text{ day}^{-1}$ at 3 cm, decreasing to $14.9 \text{ nmol cm}^{-3} \text{ day}^{-1}$ at 15 cm (Fig. 4a). At site D1, SRR peaked at the surface

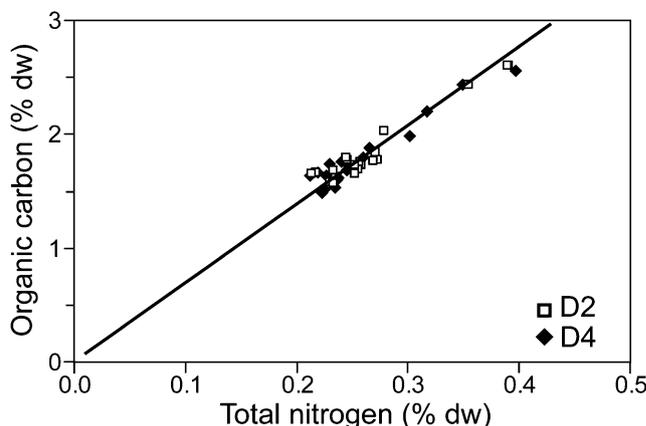


Fig. 3 Organic carbon versus total nitrogen contents at site D2 (squares) and site D4 (diamonds) in basin sediments. The C/N ratio was calculated from the slope of the best-fit regression line (cf. text)

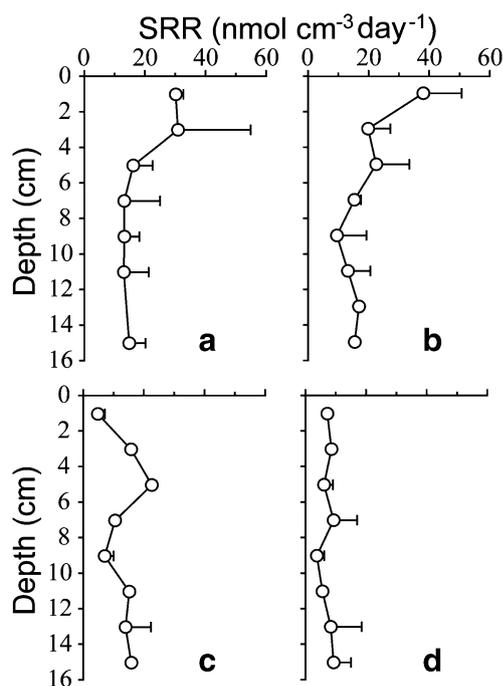


Fig. 4 Downcore profiles of sulfate reduction rates (*SRR*; error bars denote standard deviations) at sites B4 (**a**) and D1 (**b**) in slope sediments, and sites D2 (**c**) and D4 (**d**) in basin sediments

(37.7 nmol cm⁻³ day⁻¹), and decreased with depth to reach 15.1 nmol cm⁻³ day⁻¹ at 15 cm (Fig. 4b). In contrast to the slope sediments, high surface SRRs were not observed in the basin sediments. Thus, SRR peaked at 22.6 nmol cm⁻³ day⁻¹ at 5 cm at site D2, and at 9.2 nmol cm⁻³ day⁻¹ at 7 cm at site D4 (Fig. 4c,d). Sulfate reduction rates showed a generally decreasing trend from peak values at the slope sites to lower rates at the basin sites. Indeed, the integrated SRRs within the top 15 cm of the sediment were twice as high in the slope areas (Table 1).

Surface mixed layer depths were identified in terms of sharp changes in the excess ²¹⁰Pb gradient at 4 cm at sites B4, D2, and D4, and at 6 cm at site D1 (Fig. 5). The ASRs varied from 0.033 to 0.116 cm year⁻¹, with higher values in the slope sediments than in the basin sediments. ARS values decreased with increasing water depth, in agreement with previous findings by Hong et al. (1997) for the Ulleung Basin (0.04–0.17 cm year⁻¹). Miralles et al. (2005) proposed an inverse linear relationship between the ASR

and water depth for northwestern Mediterranean margin sediments:

$$\text{ASR (cm year}^{-1}\text{)} = 0.155 - 4.978 \times 10^{-5} \\ \times \text{water depth (m)}$$

This relationship was shown in our study area, too, with only 10% deviation between the observed values and those predicted by the above equation.

The sediment mass accumulation rate (MAR) was calculated as follows:

$$\text{MAR (gm}^{-2}\text{ year}^{-1}\text{)} = (1 - \varphi) \times \text{DBD (gcm}^{-3}\text{)} \\ \times \text{ASR (cm year}^{-1}\text{)} \times 10^4$$

where φ is porosity, and DBD the sediment dry bulk density. We used average values of porosity and DBD below 15 cm at each site for the MAR calculations (Table 1). Calculated MARs ranged from 131 to 481 g m⁻² year⁻¹, and were approximately three times higher in the slope areas than in the basin areas (Table 1).

Organic carbon accumulation rates (OCARs) were calculated by combining data on MARs and average organic carbon contents below 15 cm. Indeed, the latter did not change considerably below this depth (Fig. 2). The calculated OCARs varied from 2.06 to 12.5 g C m⁻² year⁻¹, and were four times higher in the slope areas than in the basin areas (Table 1).

Discussion and conclusions

The organic carbon content in the surface sediments was about 2.6% in the basin, where water depths exceeded 2,000 m. Such high organic matter enrichment has rarely been found in deep-sea sediments, except for the Black Sea that receives large amounts of river discharge (Cociasu et al. 1996; Reschke et al. 2002), and the Chilean upwelling region that has high primary productivity (Schubert et al. 2000; Böning et al. 2005). No major rivers drain into the sea along the Korean Peninsula (Hong et al. 1997), and thus there is little influence of river discharge on sediments in the Ulleung Basin. This is consistent with the OC/TN molar

Table 1 Porosity, dry bulk density (DBD), apparent sedimentation rates (ARS), mass accumulation rates (MAR), organic carbon accumulation rates (OCAR), and depth-integrated sulfate reduction rates (SRR) in slope and basin sediments of the Ulleung Basin

Site	Water depth (m)	Porosity	DBD (g cm ⁻³)	ASR (cm year ⁻¹)	MAR (g m ⁻² year ⁻¹)	OCAR (g C m ⁻² year ⁻¹)	SRR (mmol m ⁻² day ⁻¹)
B4	1,570	0.81	2.60	0.077	380	9.37	1.89 (8.28) ^a
D1	863	0.84	2.59	0.116	481	12.5	1.85 (8.10)
D2	2,208	0.85	2.64	0.048	190	3.19	1.32 (5.78)
D4	2,143	0.85	2.64	0.033	131	2.06	0.72 (3.15)

^a Corresponding rates of organic carbon remineralization (g C m⁻² year⁻¹) are shown within parentheses

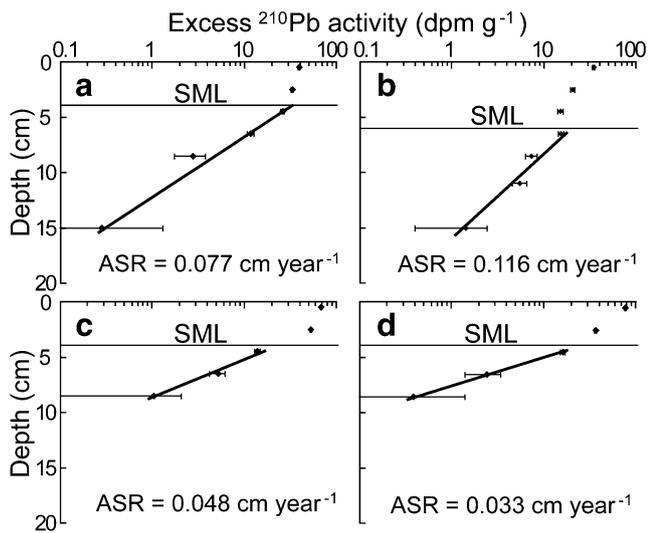


Fig. 5 Downcore profiles of excess ^{210}Pb activity (*error bars* denote standard deviations) at sites B4 (**a**) and D1 (**b**) in slope sediments, and sites D2 (**c**) and D4 (**d**) in basin sediments. The *horizontal lines* indicate the surface mixed layer (SML). *ASR* Apparent sedimentation rate

ratio of 6.98 estimated for these basin sediments in the present study, very similar to the Redfield ratio, and therefore suggesting that the organic matter deposited in the basin is predominantly of marine origin.

Time-series sediment traps have been deployed at 2,100 m water depth in the Ulleung Basin from December 1998 to January 2000 to estimate the annual flux of particulate matter (KORDI 2003). This was found to be $122 \text{ g m}^{-2} \text{ year}^{-1}$, similar to the MAR ($131 \text{ g m}^{-2} \text{ year}^{-1}$) estimated for site D4 in the present study. The location of one sediment trap was only about 50 km from site D4. Thus, the MAR obtained for the Ulleung Basin in this study agrees well with the sediment trap data of KORDI (2003).

The organic carbon flux estimated by the time-series sediment trap for the Ulleung Basin was $8.8 \text{ g C m}^{-2} \text{ year}^{-1}$ at a water depth of 1,020 m from December 1998 to January 2001 (KORDI 2003), which is three times higher than that in the Bering Sea (Takahashi et al. 2000), and comparable with that of the Chilean upwelling region (Hebbeln et al. 2000). The high organic carbon content in the Ulleung Basin probably reflects the high export production in this region. How such high export production can be sustained in the basin is currently under investigation (Yoo, personal communication). The high organic carbon content in the Ulleung Basin may also be at least partly due to reduced dilution by calcium carbonate. Chen et al. (1995) suggested that carbonate saturation depths for aragonite and calcite in the East Sea were 300 and 1,300 m, respectively. Calcium carbonate contents in Ulleung Basin sediments were generally less than 1% in the present study, which would have caused a relative enrichment in organic carbon.

The OCARs estimated in the Ulleung Basin also showed high values, exceeding $2 \text{ g C m}^{-2} \text{ year}^{-1}$; this has rarely been found for deep-sea sediments (below 2,000 m), except in the Black Sea deep basin and in intense upwelling regions (Calvert and Price 1983; Hay 1988; Böning et al. 2004). The OCARs obtained from the deep basin of the Black Sea were similar to, or two to three times higher than those in the Ulleung Basin (Hay 1988; this study). Studies of Peruvian and Namibian upwelling regions found OCAR values that were an order of magnitude higher than those of the Ulleung Basin (Calvert and Price 1983; Böning et al. 2004; this study). Although the MAR estimated for the northern Gulf of Mexico was similar to that for the Ulleung Basin, the OCAR was much lower, due to the lower organic carbon content (Yeager et al. 2004).

Table 2 Comparison of the integrated sulfate reduction rates (SRR) of the study area and other continental margins

Region	Water depth (m)	SRR ($\text{mmol m}^{-2} \text{ day}^{-1}$)	References
Continental slope			
East Sea	863–1,570	1.85–1.89	This study
Chile upwelling	1,000	4.9	Thamdrup and Canfield (1996)
Peru upwelling	502	25.5	Fossing (1990)
Benguela upwelling	855–1,488	0.18–1.39	Ferdelman et al. (1999)
Black Sea	400–1,500	0.2–2.3	Lein et al. (1997)
East China Sea	637–1,498	0.095–4.81	Lin et al. (2002)
Washington State	465–630	0.65–1.3	Devol and Christensen (1993)
Gulf of Main	250–290	0.36–2.7	Christensen (1989)
Gulf of Mexico	1,086–2,008	0.038–2.45	Lin and Morse (1991)
Basin (continental rise)			
East Sea	2,143–2,208	0.72–1.32	This study
Chile upwelling	2,000	2.4	Thamdrup and Canfield (1996)
Peru upwelling	2,650	5.2	Fossing (1990)
Benguela upwelling	2,060–3,013	0.14–0.37	Ferdelman et al. (1999)
Black Sea	2,000–2,100	0.1–1.4	Albert et al. (1995), Lein et al. (1997)

The integrated SRRs were consistently higher in areas of high sedimentation and high organic carbon accumulation, which showed good correlation between ASR and OCAR. The integrated SRRs varied with the overlying water depth, decreasing at greater water depths, as found in previous studies (Table 2). For example, Lein (1985) proposed that SRRs were inversely related to water depth, and Christensen (1989) showed that SRRs varied exponentially with water depth, decreasing tenfold between 50 and 300 m. Thus, depth is a critical factor when comparing and interpreting the results of different studies.

For slope sediments at water depths of 863–1,570 m, the integrated SRRs were 1.85 and 1.89 mmol m⁻² day⁻¹, lower than those reported for Peruvian and Chilean continental slope sediments (Table 2). Except for those areas, however, the integrated SRRs recorded in the present study were comparable to, or higher than those of other continental slope sediments. In Ulleung Basin sediments taken from basin sites with water depths of around 2,200 m, the integrated SRRs were 0.72 and 1.32 mmol m⁻² day⁻¹, lower than those of Peruvian and Chilean continental rise sediments, comparable to those of the Black Sea, but higher than those of continental rise sediments in the Benguela upwelling region (Table 2).

Based on the discussion presented above, it is evident that the sulfate reduction rates recorded in basin and slope sediments of the Ulleung Basin are higher than the values reported to date for other deep-sea sites, with the exception of the Peruvian and Chilean upwelling regions. This is consistent with the strong organic matter enrichment documented in Ulleung surface sediments (2.6–2.9% organic carbon), a very unusual finding for deep-sea sediments suggesting enhanced organic matter fluxes.

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