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East Asian Monsoon History as Indicated by C/N Ratios and δ^{13}C Evidence from the Estuarine Tidal Flat Sediments in the West Coast of Korea

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서해안 염하구 습지 퇴적물의 지화학적 분석(C/N 및 δ^{13}C)에 기반한 동아시아 몬순 변동 연구

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Abstract : Geochemical and physical investigations such as δ^{13}C isotope ratio, carbon/nitrogen (C/N) ratio, magnetic susceptibility (MS), and particle size analyses were carried out on the estuarine tidal flat sediments from the west coast of Korea in order to reconstruct the East Asian summer monsoon variability during the late Pleistocene and Holocene. Our results indicated that the summer monsoon probably peaked around 7,700-7,800 yr BP and then started to decline about 7,400 yr BP in the Korean peninsular, and that the monsoon was relatively weak between 24,000-24,500 yr BP but relatively strong between 18,500-19,500 yr BP during the Last Glacial Maximum. Our estuarine geochemical data have proven to be valuable as a new proxy for detecting the shifts in monsoon strength. This new evidence will be helpful, especially for Korean paleoenvironmental studies with few proxy data archives.

Key Words : East Asian summer monsoon, Korea, late Pleistocene and Holocene, estuarine tidal flat sediments, δ^{13}C and C/N

요약 : 후기 플라이스토신 및 홀로신 시기 동아시아 여름 몬순의 변화를 밝히기 위해 지화학적 분석 방법(탄소동위원소(δ^{13}C), 탄질율(C/N), 대자율, 입도 분석 등)을 활용하여 서해안 염하구 습지 퇴적물을 분석하였다. 연구 결과에 따르면 한반도에서 여름 몬순은 7,700-7,800년 전에 정점이었고 7,400년 전 경부터 약화되기 시작한다. 그리고 마지막 빙하 최성기 중 24,000-24,500년 전 경에는 몬순이 상대적으로 약했고 18,500-19,500년 전 경에는 상대적으로 강했다. 염하구 습지 퇴적물의 지화학적 분석 자료는 과거 여름 몬순의 변동을 밝힐 수 있는 새로운 고환경 자료로서 가치를 갖는다. 특히 고환경 자료가 부족한 한국 학계에 큰 도움이 될 것으로 사료된다.

주요어 : 동아시아 여름 몬순, 한국, 후기 플라이스토신 및 홀로신, 염하구 습지 퇴적물, δ^{13}C와 C/N

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1. Introduction

The East Asian monsoon system dominates the climate of East Asia and plays a significant role in the global atmospheric circulation. Understanding the mechanism of the East Asian monsoon is important to predict future possible climate changes in East Asia and the world. To date, the past variability of the East Asian monsoon has been reported by a number of studies using loess (e.g. An *et al.*, 1990, 1993; Xiao *et al.*, 1995, 1999), lake sediments (e.g. Chen *et al.*, 1999; Peng *et al.*, 2005; Schettler *et al.*, 2006), and cave speleothems (e.g. Wang *et al.*, 2001, 2005; Hu *et al.*, 2008) mainly from China. In Korea, maar lake sediments from Jeju island were analyzed to detect the signals of the East Asian winter monsoon during the late Quaternary and the Holocene (Lim *et al.*, 2005; Lee *et al.*, 2008). However, no study has yet investigated the East Asian summer monsoon in Korea.

Estuarine tidal flat sediments generally have not been the target of paleoenvironmental studies in Korea due to the lack of microfossils and the possibility of sediment reversal. Only the erosional boundaries between Holocene marine mud and late Pleistocene paleosol have been focused on in relation with the past sea-level changes and used as stratigraphic correlation markers along the west coast (e.g. Choi, 2005).

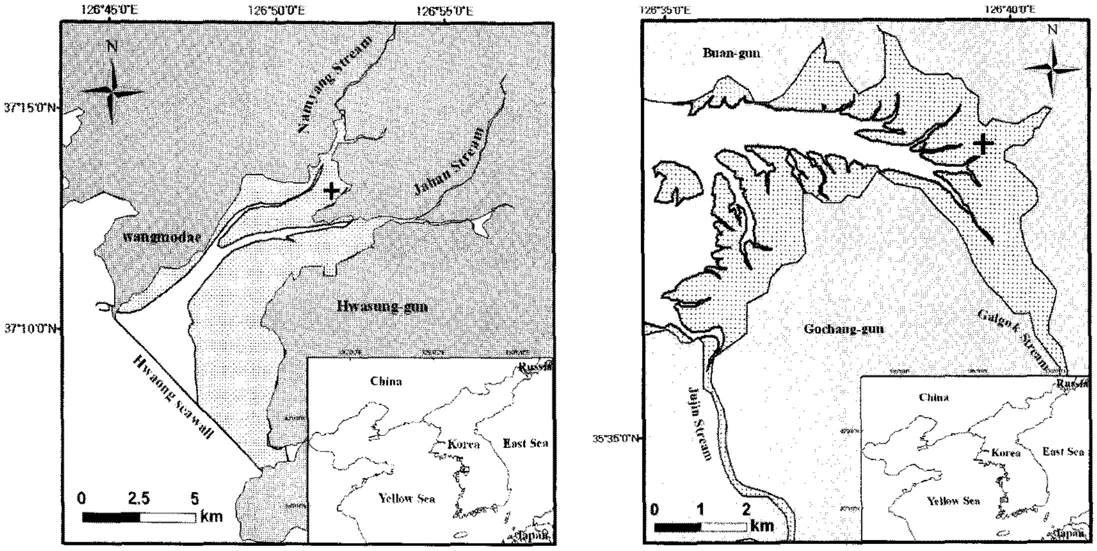
In the estuaries, geochemical signals in the sediment could be indicative of variations in the relative contribution of terrestrial versus marine input. For example, if substantial terrigenous materials are transported by the river flow to the estuarine environment due to intensified summer monsoon, the $\delta^{13}C$ isotope ratio of the sediments will be low and the carbon/nitrogen (C/N) ratio high. In contrast, if sea-level rise leads to an increasing flux of marine organic matter, the opposite geochemical trends will appear. The usefulness of the $\delta^{13}C$ and C/N ratio analyses in revealing the paleoenvironmental changes is based on the significant difference in $\delta^{13}C$ values and C/N ratios between terrestrial organic matter and marine organic matter (Lamb *et al.*, 2006).

In this study, estuarine tidal flat sediments were geochemically analyzed to obtain paleoenvironmental data and test their usefulness in studies on the Korean paleoenvironment. We used several geochemical analysis methods such as $\delta^{13}C$ isotope ratio, C/N ratio, magnetic susceptibility (MS), and particle size. The main investigating tools, $\delta^{13}C$ and C/N ratio analyses, have mostly been used for the reconstruction of relative sea-level change in the coastal environment (Wilson *et al.*, 2005; Lamb *et al.*, 2007; Kemp *et al.*, 2010). However, for this study, they were used to reconstruct the East Asian summer monsoon variability during the late Quaternary and Holocene. A Chinese study from the Pearl River estuary using the same research strategy successfully produced reasonable results for the East Asian summer monsoon during the Holocene (Zong *et al.*, 2006)

2. Materials and methods

In 2008 we recovered 4 m+ long cores at Namyang bay (37°09′ 12″E, 126°46′ 00″N) and Gomso bay (35°35′ 00″E, 126°39′ 24″N) with a vibrating corer (Figure 1). In choosing these coring sites, we tried to get as close to the land in order to avoid excessive influence from the marine effect on the signals of monsoon variation. These cores were returned to the Geography department of Chonnam National University and stored in a 5℃ cold room prior to analysis. Subsequently the cores were split, one half archived and the other half used for analysis.

Radiocarbon chronologies for both sites were



(a) (b)

Figure 1. Map of the study area: (a) Namyang bay and (b) Gomso bay. The locations of coring points are indicated by the crosses.

established by accelerator mass spectrometry (AMS) dating of bulk tidal flat sediment (Table 1). Ten bulk sediment samples were submitted to the National Center for Inter-University Research Facilities of Seoul National University for AMS radiocarbon dating. Calibrated age ranges were determined with the CALIB 6.0 program (Stuiver and Reimer, 1993).

Marine reservoir effect was not considered when establishing the core chronology because of following two reasons. First, the Yellow Sea area including the west coast of Korea shows a relatively low marine reservoir correction factor (ΔR) probably due to the large amount of freshwater input (Southon *et al.*, 2002; Kong and Lee, 2005). The ΔR based on the radiocarbon age of a mollusk shell from the west coast was calculated to be -117±47 years (Kong and Lee, 2005), indicating that the regional reservoir effect in the west coast is much less than the mean global reservoir effect.

Second, our coring sites are located deep inside the estuary close to the sources of terrestrial water. Radiocarbon dates from estuaries often cannot be used in calculating the regional mean ΔR because marine organics sometimes has little influence on them (Heier-Nielsen *et al.*, 1995). Marine samples are supposed to have 100% marine carbon; however, it is unlikely that all the carbons in our sediment were originated from the marine environment since terrestrial freshwater and organics were continuously supplied to the coring sites by the river flow.

Indeed, many previous researches on the west coast and Yellow Sea did not consider the marine reservoir effects (Kim and Kennett, 1998; Kim *et al.*, 1999; Kim and Kucera, 2000). Obviously more data should be accumulated regarding the estuarine reservoir effect in the west coast before this regional effect is explicitly considered for chronologies of tidal sediment cores.

The date from the 400 cm level of NY-1

Table 1. Radiocarbon dates for Namyang (NY-1) and Gomso (GS-2) sediment cores.

Core	Sample Depth (cm)	Material dated	Laboratory No.	$\delta^{13}C$ (‰)	Age (^{14}C yr BP)	Two σ age range (cal yr BP)	Median (cal yr BP)
GS-2	98	sediment	SNU09-R122	-25	2,600±60	2,665-2,849	2,722
GS-2	188	sediment	SNU09-R123	-25	3,200±60	3,325-3,570	3,426
GS-2	288	sediment	SNU10-R124	-25	10,100±80	11,329-12,003	11,688
GS-2	308	sediment	SNU10-R001	-25	14,800±200	17,505-18,566	18,032
GS-2	382	sediment	SNU09-R002	-25	20,820±80	24,486-25,081	24,806
NY-1	80	sediment	SNU09-R206	-25	5,970±50	6,675-6,914	6,807
NY-1	250	sediment	SNU09-R207	-25	6,630±50	7,435-7,578	7,516
NY-1	400	sediment	SNU09-R208	-25	11,540±60	13,257-13,565	13,381

produced a calibrated median age of 13,380 BP, which is too old considering the sea level along the west coast at that time. During marine transgression, mud sediments may have been re-deposited so that marine and non-marine sediments were partially mixed at the bottom section of the cores. The date from the 288 cm level of GS-2 is also too old, probably due to the same reason.

The water and organic contents of both cores were determined by oven drying at 100℃ for 24 hours and combustion at 550℃ for 1 hour. MS and C/N ratios were measured at 5 cm intervals using Bartington Instruments MS2 and an elemental analyzer (FlashEA 1112) at the Korea Polar Research Institute. For isotope analysis, samples were taken at 5 cm intervals from the sediment cores. All samples were treated with 5% HCl to remove carbonates. Carbon isotope ratios in the sediment organics were determined using a GV Isoprime mass spectrometer at Korea Basic Science Institute. Replicate analyses of samples gave a precision of $<\pm 0.2$‰. Particle size analysis was also conducted on the cores at the same sampling interval using a laser granulometer (Malvern master sizer 2000).

3. Results and discussion

$\delta^{13}C$ and C/N ratios from estuarine sediments are useful for detecting the sources of organic materials preserved in them. In our sediments, the main sources of preserved organic material were (1) marine organic material (marine algae and marine dissolved organic carbon (DOC)) brought in by the tide flow, (2) terrestrial C3 plant material transported by the river flow, and (3) freshwater algal matter in situ.

Marine algae have $\delta^{13}C$ values between -16 and -23‰ and C/N ratios usually less than 10 (Meyers, 1994). Marine $\delta^{13}C_{DOC}$ values range from -22 to -25‰ (Peterson *et al.*, 1994). Terrestrial C3 vegetation commonly shows $\delta^{13}C$ values of -21 to -32‰ (Deines, 1980) and C/N ratios of > 12 (Prahl *et al.*, 1980). Marine organic materials and terrestrial C3 plants are therefore differentiable by their differences in $\delta^{13}C$ values. Freshwater algae could also be distinguished from terrestrial C3 vegetation by their low C/N ratios which range from 4 to 10 (Meyers, 1994), even though the $\delta^{13}C$ range of freshwater algae (-25 and -30‰) is included in the range of terrestrial C3 plants (Meyers, 1994).

The typical $\delta^{13}C$ and C/N ranges presented above revealed the past coastal environmental changes. For example, the relatively high δ^{13}C values and low C/N ratios of the sediments indicated a large contribution of marine inputs to the sediment organics, implying either a higher sea level or a weaker summer monsoon, whereas relatively low δ^{13}C values and high C/N ratios indicate a large contribution of terrestrial inputs to sediment organics, implying the opposite situation. On the other hand, high δ^{13}C values and low C/N ratios probably resulted from the substantial supply of autochthonous organic matters originating from the death and decay of freshwater algae and the minimal influx of terrestrial organics due to weakened summer monsoon rainfall. MS and particle size analyses also provided valuable information on the influx of terrigenous materials and assisted in the interpretation of the δ^{13}C and C/N results.

1) The Namyang core (NY-1)

Calibrated radiocarbon dates of 80 cm and 250 cm depth were 6,800 cal yr BP and 7,520 cal yr BP, respectively, showing very high sedimentation rates. The base of the core (400 cm) was dated to 13,380 cal yr BP, which seems to be too early considering the sea level at that time. This may be due to the mud redeposition during marine transgression as mentioned earlier.

The results of geochemical analyses are shown in Figure. 2. In Zone 1 (360-230 cm, ?-ca. 7,400 cal yr BP), δ^{13}C values were relatively low and C/N ratios relatively high compared to the upper part of the sediment core. The geochemical composition of this zone implied a period of stronger summer monsoon, during which a relatively large amount of terrestrial organics were brought into the site. The lowest δ^{13}C value and the highest C/N ratio at a depth of around 300 cm implied that the Asian summer monsoon was the strongest at the corresponding time. The tree-ring Δ^{14}C record (Stuiver *et al.*, 1998), mainly reflecting the variations in solar activity and consequently the change in the strength of summer monsoon, indicates that the summer monsoon was prominently intensified around 7,800-7,700 yr BP in the northern hemisphere. Therefore, the age of the 300 cm level was estimated as 7,800-7,700 yr BP. The large peak of particle size at 300 cm was also indicative of the culmination of the summer monsoon, which would have temporarily widened the river channels and supplied coarse materials to the coring site. An *et al.* (2000) suggested that the peak of East Asian summer monsoon precipitation was asynchronous in central and eastern China because of the seasonality change related to orbitaly induced insolation change. According to their map showing the position of the East Asian monsoon maximum over time, the central west coast of Korea experienced this maximum between approximately 7,000-8,000 yr BP, indicating the geochemical peak in our result is related to the East Asian monsoon maximum in the Korean peninsular.

In Zone 2 (230-95 cm, ca. 7,400 cal yr BP-6,900 cal yr BP), δ^{13}C values were higher and C/N ratios were lower than in the previous zone, indicating that the influx of terrestrial organics was reduced as the summer monsoon became weaker. The tree-ring Δ^{14}C record showed that the solar activity cycle began to fade around 7400 yr BP, implying the start of a weak summer monsoon. In the core section above Zone 2, δ^{13}C and C/N ratios did not show any changes reflecting the intensified summer monsoon, even though the tree-ring Δ^{14}C record implied it after about 6,900 yr BP. This discordance between the geochemical results and the tree-ring Δ^{14}C record was probably related to the change in sea level, which culminated at around 7,000-6,000 yr BP in the west coast of Korea. Increased marine organic inputs due to sea-level culmination may have offset increased influx of terrigenous materials

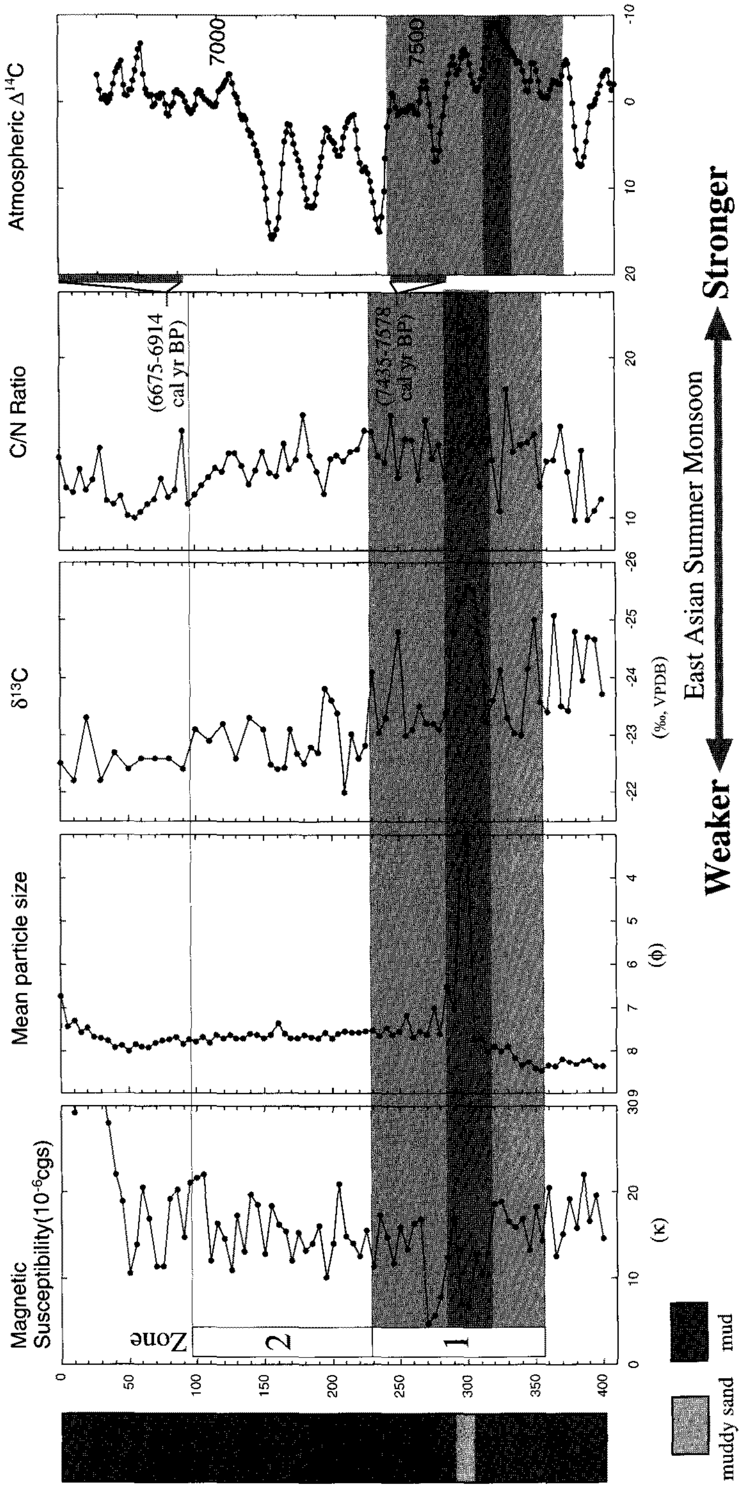


Figure 2. Geochemical and physical characteristics of Namyang sediment (MS, $\delta^{13}C$, C/N, and grain size) and atmospheric $\Delta^{14}C$ (1000-yr moving average) (Stuiver *et al.*, 1998). Note that reverse x-axes were used for particle size, $\delta^{13}C$, and atmospheric $\Delta^{14}C$.

induced by summer monsoon intensification.

The high MS in the uppermost part of the core had no important meaning, probably due to the pedogenesis after the construction of the Hwaong seawall. MS were consistently low throughout the core. The relatively low values between 320-260 cm might have resulted from a reduced supply of terrigenous weathered materials or eolian materials from inner Asia as the winter monsoon weakened.

Our geochemical results in Zones 1 and 2 showed a similar trend to that of the oxygen isotope records from stalagmites in Dongge cave in China (Wang *et al.*, 2005) and the tree-ring $\Delta^{14}C$ record, which are related to the solar output (Figure 2). Thus, we can conclude that changes in solar output as well as orbital forcing were strongly associated with shifts in the East Asian summer monsoon in Korea during the early to mid-Holocene. In summary, the summer monsoon intensified from the early Holocene, probably peaked around 7,700-7,800 yr BP, and then started to decline about 7,400 yr BP in the Korean peninsular.

2) The Gomso core (GS-2)

The Gomso core consisted of oxidized terrestrial sediments (400-300 cm) made up of late Pleistocene paleosol and marine sediments (300-0 cm). Therefore, the chemical analysis results differed widely between the upper and lower sections. The 288 cm level of marine sediments was dated to 11,690 cal yr BP, which also seems to be too early as with the Namyang core. During marine transgression, disturbed terrestrial sediments may have become mixed with marine sediments around the boundary. Marine sediments do not demonstrate any clear geochemical patterns reflecting paleoenviron mental changes, so we can only discuss the results of terrestrial sediments here.

The results of geochemical analyses on terrestrial sediments are presented in Figure 3. In the terrestrial sediments, the gravel percentages, MS results and C/N ratios exhibited similar variations but the $\delta^{13}C$ values were negatively correlated with the former three. In Zones 1 (390-378 cm) and 2b (366-353 cm), the gravel percentages, MS results and C/N ratios were consistently low and the $\delta^{13}C$ values high, compared to the other zones. In contrast, the opposite trends appeared in Zones 2a (378-366 cm), 2c (353-337 cm) and 3 (328-304 cm).

The high $\delta^{13}C$ values and low C/N ratios, gravel percentages and MS results indicated that the sediments probably accumulated in a small local swamp distant from the main channel, which may have been created as the Asian summer monsoon weakened. The high $\delta^{13}C$ values and low C/N ratios of the sediment were presumably associated with considerable inputs of autochthonous algal matters and reduced influx of terrestrial organics due to the weakening of the summer monsoon. The low gravel percentage and MS also implied that only a little terrigenous material could be transported by dwindling river flows to the site. Considering the clarity of the results, the river flow does not seem to have influenced the sediment in Zone 1 (390-378 cm), which was most likely accumulated in the environment of an entirely isolated swamp. On the other hand, it is possible that the sediments in Zones 2b (366-353 cm) and 2d (337-328 cm) were somewhat affected by the river flow close to the site.

The low $\delta^{13}C$ values and high C/N ratios, gravel percentages and MS results in Zones 2a, 2c and 3 indicated that substantial terrigenous organic matters with coarse materials were transported into the site as the river channel was widened due to the strengthened summer monsoon.

According to relative sea level data from around the world, the timing of the Last Glacial

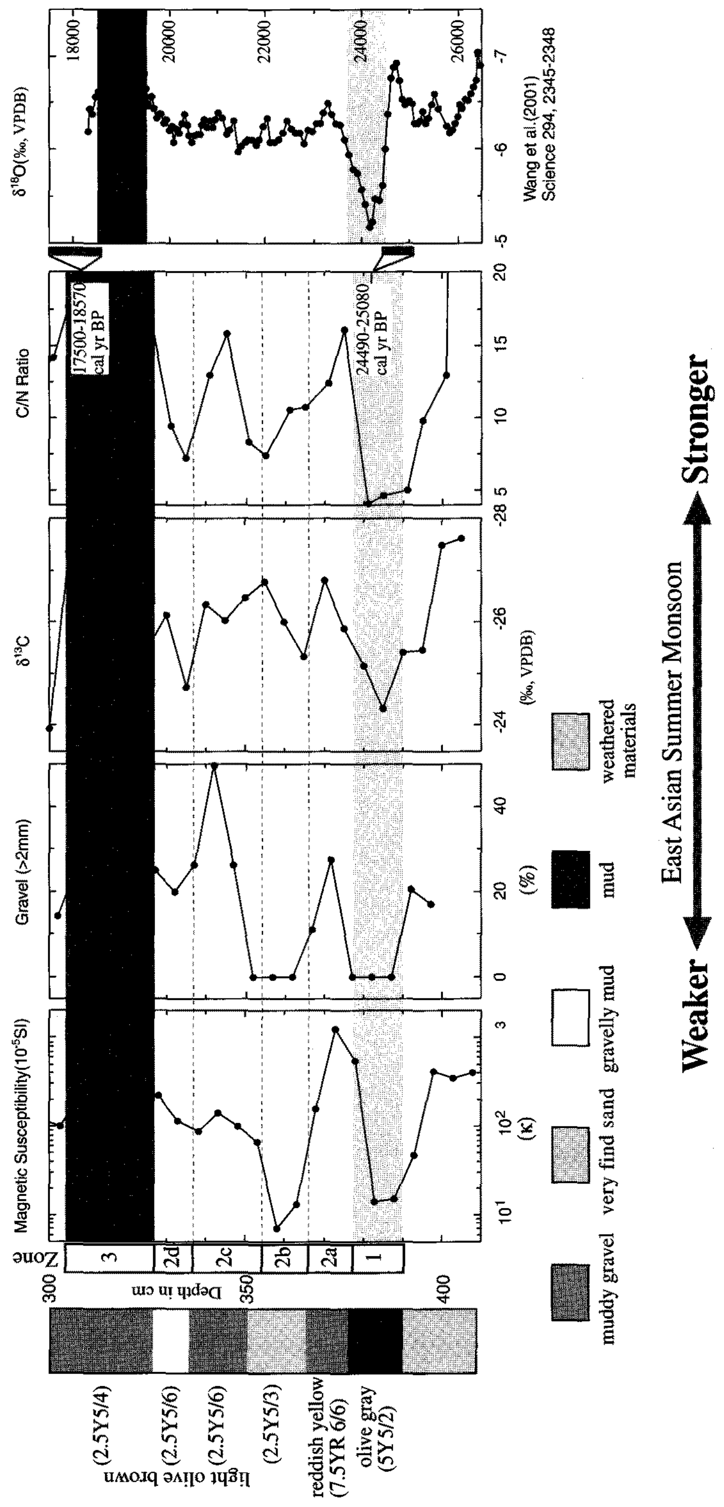


Figure 3. Geochemical and physical characteristics of Gomso terrestrial sediment (MS, $\delta^{13}C$, C/N, and gravel percentages) and oxygen isotope record from Hulu Cave (Wang *et al.*, 2001). Note that reverse x-axes were used for $\delta^{13}C$ and $\delta^{18}O$ from Hulu Cave.

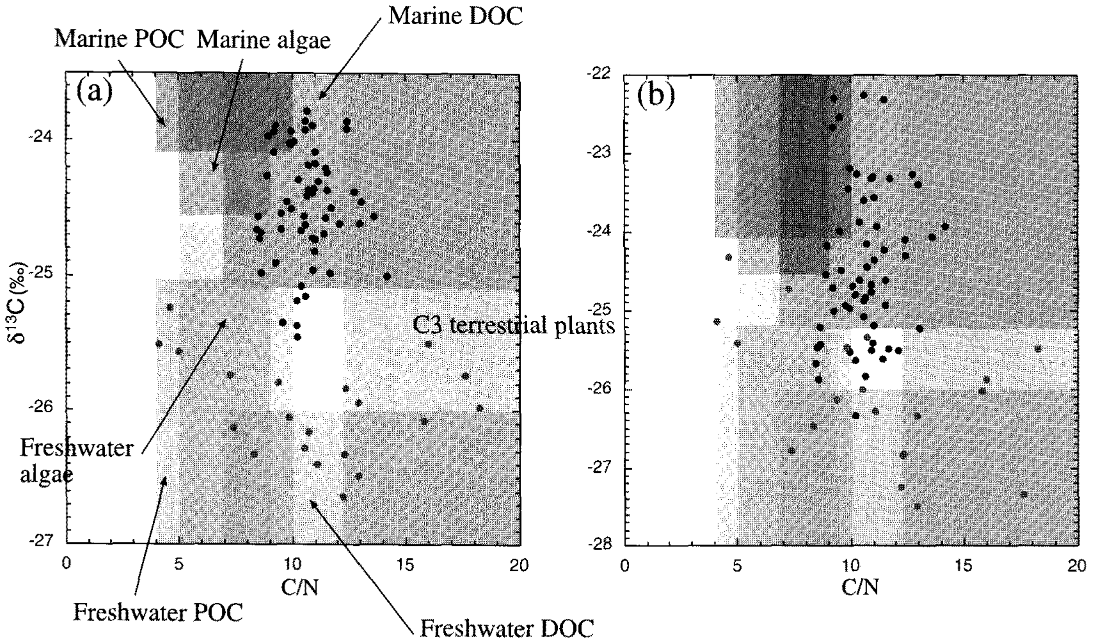


Figure 4 Comparison between typical $\delta^{13}C$ and C/N ranges for organic inputs to coastal environment (Lamb *et al.*, 2006) and $\delta^{13}C$ and C/N values of Gomso sediment (this study): (a) Smoothed $\delta^{13}C$ data (5-point moving average) and (b) non-smoothed $\delta^{13}C$ data. Terrestrial and marine sediment samples are indicated by red and black dots, respectively.

Maximum (LGM) is constrained between 26,500 and 19,000 yr BP (Clark *et al.*, 2009). Our GS-2 data therefore could show the change in relative strength of East Asian Monsoon during the entire LGM period. The $\delta^{18}O$ values of stalagmites from Hulu cave, China (32°30′ N, 119°10′ E) (Wang *et al.*, 2001) can be used to confirm the shifts of the LGM Asian monsoon indicated by our results (Figure 3). According to Wang *et al.* (2001), the Asian summer monsoon was conspicuously weak between 24,000-24,500 BP, corresponding to the Heinrich event 2(H2) (Bond *et al.*, 1993) but was relatively strong between 18,500-19,500 BP. In Figure 3, Zone 1 matches the colder-drier period (24,000-24,500 BP) and Zone 3 does the warmer-wetter period (18,500-19,500 BP). The trends shown in Zones 1 and 3 generally followed the Hulu cave $\delta^{18}O$ record whereas the results in Zone 2 were only weakly correlated with the corresponding Hulu $\delta^{18}O$ record.

3) $\delta^{13}C$ values and C/N ratios in the Gomso core

As aforementioned, the main sources of organic material preserved in the sediments were marine organic material, terrestrial plant material, and freshwater algal matter. In Figure 4, the C/N ratios and $\delta^{13}C$ values in the Gomso core are compared with typical $\delta^{13}C$ and C/N ranges for organic inputs to coastal environments (Lamb *et al.*, 2006). The smoothed $\delta^{13}C$ values of the Gomso core samples (Figure 4a) fit into (Lamb *et al.*, 2006)'s classification more clearly than the non-smoothed $\delta^{13}C$ values (Figure 4b). The range of $\delta^{13}C$ values differed conspicuously between the terrestrial (-25.5~-23.7‰) and marine (-26.7~-25.3‰) sediments. Our marine samples were mostly positioned in the category of marine DOC, whereas the terrigenous samples were in C3 terrestrial vegetation or freshwater algae/ particulate organic carbon (POC) (Figure 4a).

Terrigenous samples can be divided into two different types in terms of their organic sources: autochthonous organic matter produced by freshwater algae has relatively low C/N ratios, while organic matter from C3 terrestrial plants has high C/N ratios. Thus, these geochemical values enable the more important organic source during the sediment accumulation to be determined. Eight samples (C/N ratios: 6-10) belonged to the category of freshwater algae/POC, and nine (C/N ratios: 12-19) to C3 terrestrial plants (Figure 4a).

As shown in Figure 4a., the organic materials in the Gomso sediments could be categorized by the $\delta^{13}C$ and C/N results. This information was useful in revealing the past sedimentational environment and detecting the shifts in monsoon strength. The west coast of Korea features a number of small estuaries with both the Holocene and late Pleistocene deposits. Therefore, the study of estuarine sediments along the west coast can comprehensively reveal the past variability of the East Asian monsoon because it is possible to correlate research data from different estuaries and thus obtain a great deal of reliable information on paleoenvironmental changes.

4. Conclusion

We analyzed estuarine tidal flat sediments to reveal past environmental changes in the Korean peninsular and attempted to test if estuarine sediments from the west coast of Korea could provide plausible paleoenvironmental records. Geochemical and physical methods such as $\delta^{13}C$ values, C/N ratios, MS data, and particle size analyses were used to reconstruct the East Asian summer monsoon variability during the late Quaternary and Holocene.

An analysis of the Namyang core (NY-1) revealed that the East Asian summer monsoon intensified from the early Holocene, probably peaked around 7,700-7,800 yr BP, and then started to decline about 7,400 yr BP in the Korean peninsular. Our results were strongly supported by the oxygen isotope records from stalagmites in the Dongge cave in China and the tree-ring $\Delta^{14}C$ record. The geochemical data from the Gomso core (GS-2) showed that the monsoon was relatively weak between 24,000-24,500 yr BP but relatively strong between 18,500-19,500 yr BP during the LGM. This difference in humidity between the two periods was also well indicated by the Hulu cave $\delta^{18}O$ record.

Estuarine geochemical evidence has not been used as reliable proxy data in Korea due to the chronological uncertainty of tidal flat sediments in the west coast. However, as supported by oxygen isotope records from caves in China and the tree-ring $\Delta^{14}C$ record, the estuarine geochemical data presented here proved to be valuable as a new proxy to help reconstruct the past changes in the coastal environment and detect the shifts in monsoon strength. Many attempts to find reliable paleoenvironmental data archive in Korea have failed because of the long history of heavy human activity. The usefulness of our estuarine geochemical data was partially limited by chronological uncertainty; nevertheless, this new evidence will be very valuable for Korean paleoenvironmental studies with few proxy data archives.

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