**High Resolution Late-Holocene Terrestrial Biomarkers Records in southernContinental shelf area of the Korean Peninsula: Climate and Marine Environmental Implications during the last 5kyr**

Sangmin Hyun1

Corresponding author

Email: [smhyun@kiost.ac.kr](mailto:smhyun@kiost.ac.kr)

Nayeon Kang1

Email: [nayeon@kiost.ac.kr](mailto:nayeon@kiost.ac.kr)

JinHyung Cho2

Email: [jcho@kiost.ac.kr](mailto:jcho@kiost.ac.kr)

(Institutional address)

1Marine Environmental Research Center, Korea Institute of Ocean Science and Technology (KIOST), Yeongdo-gu, Hean-ro 385, S-Korea

2Marine Security Research Center, Korea Institute of Ocean Science and Technology (KIOST), Yeongdo-gu, Hean-ro 385, S-Korea

**Abstract**

High-resolution records for carbon isotopes of organic matter and *n*-alkane compounds were investigated in two gravity cores (SJP15-2 and SJP15-4) taken from the southern continental shelf of the Korean peninsula to evaluate the variation in influxes of terrestrial organic compounds and their linkage to paleoclimate and marine environmental changes since the last 5 kyr. The total organic carbon contents were < 1%, and the carbon isotope(13Corg) ratio rangedfrom approximately −21‰ to -22‰ and, they did not highly fluctuate throughout the two cores. However, the vertical distributions of total terrestrial biomarkers, long-chain *n*-alkanes (*n*C25-35), and individual *n*-alkane compounds exhibited distinctive fluctuations. There are three switching points that discriminate patterns of excursion and distribution at ca. 4.5 ka, 3.0 ka, and 1.8ka. Several *n*-alkane combined indices such as average chain length (ACL), carbon preference index (CPI), and paleovegetation index (P*aq*), were coincident with these switching points, implying that the supply of terrestrial biomarkers was strongly associated with environmental changes at the source area. In particular, the ratios of *n*C31/*n*C27 and *n*C31/*n*C29showcoincident excursion pattern with lower ratio between 3.0 ka and 1.8 ka, implying that this short-term event-like record (STER) was associated with wetter climate conditions, and thus paleovegetation and paleoclimate variation. Comparison with previous data of the detrital quartz from the East China Sea and aeolian dust in the Cheju (Jeju) Island, South Korea, and Dongge cave oxygen isotope records indicates strong synchronicity with those switching points, suggesting that paleoclimate system of the East Asian region may have influenced the sediment records of study area since the last 5 kyr. Therefore, our high-resolution *n*-alkane data are very useful for reconstructing past climatic records, and East Asian monsoonand regional records could be associated with the paleoclimate variations of the study area.

Keywords

13Corg, *n*-alkanes, paleoclimate, compound-specific isotope, East Asian monsoon

**Introduction**

Continental shelf and coastal areas are important locations where accumulated terrestrial materials and *in situ* marine materials can be easily preserved. This means that terrestrial and oceanic records are mixed and reflect past climatological and oceanographic variations. Identification of the sources of terrestrial biomarkers in marine environments and the factors controlling their distributions is important for understanding the global biogeochemical cycle of biomarkers as well as local oceanographic and paleoclimatic environmental changes (Hu et al. 2006; Hyun et al. 2006; Jeng 2006). Such sediments are likely to contain high concentrations of oceanic and terrestrial materials (Stein1990). However, it is difficultto determine the contributions of certain types of biomarkers to bulk organic matter because most biomarkers are mixed with terrestrial and marine materials. Owing to the complex nature of organic compounds in shallow continental marine environments, various parameters (stable isotope ratios of organic carbon [δ13Corg] and total nitrogen [δ15NTN], carbon/nitrogen ratio [C/N ratio], and *n*-alkanes have been widely used to determine the sources of organic matter (Minoura et al. 1997; Naidu et al. 2000; Cloern et al. 2002; Usui et al. 2006; McKirdy et al. 2010).

As one of the most significant terrestrial organic materials, *n*-alkanes have been widely used to determine organic matter sources and climatic variation in various locations (Jeng 2006; Eglinton and Eglinton 2008; Yamamoto et al. 2010; Chu et al. 2014). In particular, because high-molecular-weight long-chain *n*-alkanes such as *n*C27, *n*C29, and *n*C31 are major constituents of vascular land plants (Sikes et al. 2009; Yamamoto et al. 2010; Bush and McInerney 2013), sedimentary *n*-alkanes are potentially important for determining the sources of organic matter and related environmental changes, including paleoclimate variation. The abundance of these waxy hydrocarbons reflects the amount of organic matter transported to lake beds, alluvial deposits, marine sediment, and rivers from the surrounding land, making it possible to determine their contribution to the terrestrial organic matter flux. Therefore, these proxies provide insight into organic matter sources, past environmental changes, including vegetation changes (Zhang et al. 2006; Yamamoto et al. 2010), and local and globalpaleoclimatic environmental changes (Horikawa et al. 2010; Seki et al. 2006; Street et al. 2013; Li et al. 2013; 2015).

As a component of global climate changes, the East Asian monsoon system and its variation have been investigated through the various proxy data (Nagashima et al. 2013; Wang et al. 2014; Tada et al. 2016). In particular, variation of the East Asian Summer Monsoon (EASM) and East Asian Winter Monsoon (EAWM) has been a focused because many studies have shown that the monsoon signal and its intensity differsfrom among scientific datasets (Li et al. 2013; Wang et al. 2014; 2020). Some signals show coincident variations, while others are inconsistent in terms of EASM and EAWM intensity (An 2000; Wang et al. 2020). Regarding the Holocene climatic variation, it is strongly coupled with enhanced and/or reduced precipitation due to the latitudinal shift of EASM intensity. This earlier study indicates that the oscillation of EASM front results in deepest northwestward penetration make enhanced EASM during the period 3.5-2.0 kyr, and southeastward retreat (weak EASM) occurred during the period 2.0-0 kyr (Wang et al. 2020). Also this precipitation changes supported by the other proxies of eolian dust that transported in accordance to the westerly jet (WJ) path over the East Asia region (Nagashima et al. 2013).

Our study area is within the East Asian monsoon belt; therefore, the sediments of this study may reflect past climatic environmental variations in the scope of the East Asian region. As long-chain *n*-alkanes from this studied core are likely to reveal changes in terrestrial influx by river and atmosphere under the influence of East Asian monsoon climate, we investigate *n*-alkanes variability recovered from southern inner continental shelf. Few studies have used *n*-alkane signatures to track mid-Holocene climate changes in study area. The purpose of the present study is the reconstruction of past *n*-alkane influxes and their relationships with *in situ*paleoceanographic variation and paleoclimate variabilities in neighboring areas since the mid-Holocene.

**Geological and oceanographicsetting**

The southern coastal area of the Korean peninsula is characterized by a complex geomorphology and shallow water depth. It is expected that large influxes of terrestrial material have been supplied from large and small river systems such as the Seomjin River in study area (Fig. 1). The Seomjin River, located on the extreme southern coast of Korea, is characterized by a large outward flow of freshwater throughout the year, with an annual discharge of approximately 7.2 × 108 m3 of fresh water to Gwangyang Bay (Korea Ministry of Construction, KMC 1980). The average suspended sediment accumulation rate in the river mouth is ~17.8 mg/L in the bottom layer and 4.8 mg/L in the surface layer, which results in an estimated deposited mass of 0.8 × 106 tons per year (Kim and Kang 1991). Considering the sea level changes of southern coastal area of Korean peninsula, it has been considered that the sea level was reached at present level around the 6~7kabefore (Park et al. 2000; Yoo et al. 2014), therefore Yeosu Bay, a major catchment area of the Seomjin River, has been subject to sediment accumulation since the early Holocene.

Besides direct supply from the neighboring inland area, there are other sediment transportations resulting from the regime of the East Asian monsoon belt. Materials could be transported through air, and suspended materials could be transported by the Korean Coastal Current (Fig. 1). Therefore, the high sedimentation rate of the studied cores makes it possible to reconstruct high-resolution records reflecting past monsoon signals as well as *in situ* local paleoceanographic variation.

Although the two cores used in this study were taken from different locations, the lithological characters were very similar and contain paleoclimatic information such as input of terrestrial biomarkers. To acquire information and improve our understanding of the mid-Holocene paleoclimate variation, we gathered Holocene paleoclimatic data from cores acquired from the Cheju Island, South Korea (Lim and Matsumoto 2008), and a coastal area of the Yangtze River, China (Wang et al. 2020).

**Methods**

A total three gravity piston core samples were collected from the outer part of Seomjin River, inner shelf of the South Sea of Korean Peninsula using a gravity piston core (Fig. 1).After sedimentological study and sampling for carbon 14C ages sediment samples were collected. Based on the sedimentary feature, the radio carbon 14C ages samples were determined using sedimentary organic matters at Beta-isotope (Table 1). From the two piston core, total 57 samples were collected for the analysis of total organic carbon (TOC), and carbon and nitrogen isotopes, *n*-alkanes. TOC, carbon (13Corg) isotopes in organic matter, and *n*-alkanes were analyzed to determine the time-dependent variation of source of the organic matter and its distribution.

Isotopic composition of sedimentary organic matter was determined after treatment with 1M HCl overnight to remove carbonate materials and then subsequently rinsed with deionized water three times before freeze drying.The percentage of total carbon and nitrogen as well as their isotopic composition (δ13Corg) from the carbonate free samples were determined using EA-CF-IRMS.The isotope ratios of standards in the laboratory was measured three times on three separate occasions and quoted an instrumental measurement precision of ±0.01‰ for carbonand ±0.03‰ for nitrogen. To ensure quality control, each analysis run included international standards.All analytical process was conducted at the Iso-analytic in UK.

For the analysis of *n*-alkanes, lipids were extracted from dried sediment (about 10 g) over three extraction cycles using a DIONEX Accelerated Solvent Extractor (ASE 200) at 100ºC and 1500 psi with dichloromethane (DCM)/methanol (MeOH) (95:5). The extracted lipid was saponified with 1N KOH/MeOH, and neutral lipid was separated by hexane extraction. The extracted neutral lipid was further separated into four fractions using silica gel column chromatography: F1 (hydrocarbon), F2 (aromatic hydrocarbon), F3 (ketones), and F4 (polar compounds). The F1 fractions were eluted with hexane (100%), hexane/DCM (2:1, v/v), DCM (100%), and DCM/MeOH (95:5, v/v), to analysis of *n*-alkanes.

Gas chromatography (GC) was used for the analysis of *n*-alkanes in the F1 fraction using a Hewlett Packard 6890 series GC unit, with a split/splitless injector operated in a pulsed splitless mode, and electronic pressure control systems and a flame ionization detector (FID). A CP Sil5CB column (60 m × 0.32 mm i.d.) was used with the oven temperature programmed at 50–120ºC at 30ºC/min, 120–320ºC at 7ºC/min, and then held at 320ºC for 50 min. Prior to the quantification of lipids, a known aliquot of an internal standard, 5α-Cholestane, was added to the hydrocarbon fraction. Individual aliphatic (*n*-alkane) compounds were identified and quantified using the retention times of *n-*alkanes and the mass spectrum of selected ions from calibration standards at concentrations ranging from 10 to 200 μg/ml.

Analytical quality control was carried out to make sure that the results are consistent, comparable, within specified limits of precision and accurate.*n*-alkane standard mixtures were injected to the GC/MS three times to have an absolute retention time of each individual compounds. Individual compounds of *n*-alkanes in samples were identified and quantified based on the retention time and mass spectrum of selected ions with the calibration standards. Samples were spiked with a surrogate standard (n-Triacontane d62). After final concentration, the extracts were spiked with internal standard (5α-Cholestane). A 70% to 90% surrogate recovery was seen from all samples.

After*n*-alkaneanalysis, the fractions were used for the individual *n*-alkane carbon isotope (13CALK) analysis using the GC/IRMS at Woods Hole Oceanographic Institute (WHOI), USA. Atotal of 1 *μ*l of the *n*-alkane concentration was injected into a fused silica capillary column (30 m \* 0.32 mm i.d.), with helium used as carrier gas. The GC oven temperature was programmed with a stepwise increasefrom 50 to 310ºC at rates of 30ºC, 6ºC, and 9ºC/min.All 13CALK values were checked for precision and accuracy to interpret small differences in 13CALK values. The standard material (*Methyl heneicosanoate*, C22H44O2) was used and the precisions of its13CALK values obtained by GC/IRMS analyses were reported as the standard deviations (σ) of populations of at least three analyses. The standard deviation of GC/IRMS data for *n*-alkanes was usually less than 0.3‰.

**Results and discussion**

**Sediment characteristics, age model, and stratigraphy**

In order to track any past records from the core sediment,it is fundamental to understand the sediment characteristics. This meanssedimentological examination is the first step to identifying past environmental changes. The examination begins with age determination on the sediment column, which may clarify high-resolution time-dependent environmental records in terms of terrestrial influx from nearby land areas as well as *in situ* oceanographic variations.

Based on the detailed examination of the sedimentary structure in this study, the core sedimentsreveal typical hemipelagic sediment characteristics, having massive structures and being composed of homogenous clay materials. According to the carbon-14(14C) age results, which were obtained by radiocarbon age dating at Beta Analytic Inc. (Miami, FL, USA), the age at the bottom of the core did not exceed 6 kacalyrBP (Fig. 2). Based on this14C age determination, the sedimentation rates of the two cores were very similar and quite high, it was72 cm/kyr for core SJP15-2 and 71 cm/kyr for core SJP15-4, respectively, exhibiting typical features of coastal andhemipelagic sediment. This high sedimentation rate preserves valuable information regarding climatic changes at nearby locations through the terrestrial *n*-alkane flux. The results of 14C ages for the two cores are summarized in Table 1.

The sedimentary features and sedimentation rates preserved in these two cores suggesting that the sedimentary environment did not change significantly throughout deposition since the last 5 ka. This may imply that the sediments preserve continuous *in situ* oceanographic and climatic records. In particular, coring site belongs to East Asian monsoon belt, it can provide monsoon climatic signal as well as local environmental changes. The high sedimentation rate makes it possible to track the influx of terrestrial organic materials since the mid-Holocene.

**Variations of total organic carbon (TOC, %), δ13Corg, and δ15NTN**

Overall, TOC contents in the sediments of the two cores were not highly variable, ranging from 0.6% to 0.9% (average = 0.71%, n = 53). In particular, the core sediment of SJP15-2 (inner shelf) showed a narrow range of TOC variation, remaining almost constant throughout except for several upper samples, which showed a slight decrease. The TOC contents of SJP15-4 (outer shelf) differed slightly from SJP15-2, showing an increasing upward tendency until 3 ka, and then increasing again from 2.5 kato 1.8 ka(Fig. 3). Although there were slight fluctuations in the TOC contents in both cores, these fluctuations mainly originated from biogenic source productivity changes.A C/N ratio has been used to determine the source of organic matter even though it could not provide critical criteria where they are come from. For example,when the C/N ratio of organic matter > 12 indicates a terrigenous source, whereas a ratio < 12 indicates a marine source (Stein 1990).Given that the C/N ratios of organic matter in both cores were < 10, larger portion of organic matters were derived from *in situ* bio-productivity.The surface distribution of TOC from the nearby areas reflects a mixture of marine and terrestrial sources, and the terrestrial portion of organic matter reaches up to 80% of total materials (Badejo et al. 2017). Also large portion of terrestrial organic matter were contained in the sediment of thesouthern part of the coastal area (Hyun et al. 2011). Therefore, the TOC contents of the two sediment cores may come from a mixture of terrestrial and marine sources.

Like TOC contents and C/N ratios, 13Corg and 15NTN values have also been used to determine the sources of organic carbon. In present study, δ13Corgvalues ranged from −22‰ to −21‰ (average = −21.4‰, *n* = 41) in both coresediments, showing typical marine productivity-related organic carbon sources when considering its endmember(Table 2). The 13Corg value of marine particulate organic matter ranges from −22‰ to −18‰ (Middelburge and Nieuwenhuize, 1998), whereas lower values between −33‰ and −22‰ are considered to reflect terrestrial origins (Pancost and Boot 2004). Because lighter ratios indicate marine sources (the marine endmember value is approximately −20‰ and the terrestrial endmember value is −27‰), the values in the two sediment cores indicate a large portion of biogenic organic carbon input. In our study, the values were very close to the marine endmember indicating that most of the organic carbon was produced by *in situ* marine bioproductivity.

Wide range of δ13Corg values have been obtained around the Korean peninsula as shown in Table 2. Surface sediments from the surrounding areas showed that relatively high amounts of terrestrial organic carbons were supplied (Hyun et al. 2011; Badejo et al. 2017). The heavier values of δ13Corg upward in SJP15-4 may be attributable to a higher portion of marine sources or to diminishing terrestrial organic carbon. However, in the inner shelf, core SJP15-2 showed a relatively narrow range of δ13Corg, implying a higher portion of terrestrial organic carbon compared to SJP15-4. Similar δ13Corg values were recognized in nearby surface sediments, indicating mixtures of terrestrial and marine organic matter, while the δ13Corg values of the Seomjin River predominantly indicate a terrestrial endmember (Badejo et al. 2017). The results indicate that the organic matter deposited in the study area was delivered not directly from the Seomjin River but was mixed with biogenic sources and other origins. A notable depletion of δ13Corg in surface sediments was observed around the catchment areas of Gwangyang Bay and Yeosu Bay, which had relatively low C/N ratios of < 10 (Hyun et al. 2011);data from the neighboring areas indicates that organic carbon has been deposited from various sources, and the mixture of terrestrial and oceanic sources differed from their site.

The uppermost lighter ratios of δ13Corg and δ15NTN in SJP15-2 are associated with lower organic carbon contents. One simple explanation for this is the presence of inorganic nitrogen, presumably as ammonium absorbed onto clay, in the surface sediments, which can interfere with the identification of organic matter sources when applying the C/N ratio (Kang et al. 2007; Hu et al. 2009). This assumption is supported by the 15NTNvalues (Fig. 3), which had a similar distribution to that of the C/N ratio. The observed variation in 15NTN could be the result of a change in the nitrate supply, which could be in the form of nitrate fertilizer around the catchment area and a marine nitrate source in the inner shelf study area. Because the onset ofδ13Corgincrease has occurred at 4.5 ka and 2.5kain SJP15-4, it may reflect a stepwise increase of productivity. Since the TOC and δ13Corg excursions coincide, the lower C/N ratio of ~8 supports *in situ*bioproductivity. Even though stepwise increase of δ13Corg in SJP15-4 were observed and interpreted as bio-productivity variation, terrestrial biomarker such as *n*-alkanes also could be supplied. Because δ13Corgvalues are not an end-member of biogenic source, it was mixed marine and terrestrial organic matter.

**Concentration and distribution of terrestrial *n*-alkanes**

In addition to δ13Corg and δ15NTN, terrestrial *n*-alkanes provide detailed information on source-related organic compound characteristics because each *n*-alkane compound can reflect a specific source. For example, among the long-chain *n*-alkanes, *n*C29 and *n*C31 are predominately derived from terrestrial plant waxes, whereas the mid-chain *n*-alkanes *n*C20 and *n*C23 are indicative of different floating macrophyte groups (Visco et al. 1993; Fricken et al. 1998). This means that both middle (*n*C21, *n*C23) and longer (*n*C29, *n*C31) chain lengths can be traced to aquatic and terrestrialsources with excellent confidence.In a broad sense, the overall distribution of *n*-alkane concentrations is consequence of vegetation changes, thus climatic changes in source area.

An overall distribution of *n*-alkanes is characterized by predominant odd-number long-chainalkanes concentration (*n*C27-33). The vertical distribution of *n*-alkanes between the two cores was harmonious, but it was slightly different in terms of the concentration of mid-chain *n*-alkanes (*n*C22 and *n*C24). The total *n*-alkane concentration (*n*C25–*n*C35) showed large fluctuations, ranging from 1272 to 274 µg/g (average = 728 µg/g, n = 27) in core SJP15-2, and from 3056 to 339 µg/g (average = 773 µg/g, n = 30) in core SJP15-4, respectively(Fig. 4).However, in detail, the distribution was characterized bya distinctive increase in the total concentration from 3.0ka to 1.8 ka in core SJP15-4, and from 3 ka to 2.6ka with a slight increase at the layers of 4.3 ka in core SJP15-2. The timing of onset of increase in several individual *n*-alkanes are different from each other. Individual *n*-alkane such as *n*C29, *n*C31, *n*C33, *n*C35in two core start to increase at 3.0 ka, but these four *n*-alkanes group differ from other group that shows distinctive increase from 3.0 ka. Given that long-chain *n*-alkanes such as *n*C29 and *n*C29 + *n*C31 are considered proxies for paleoclimate variation,the excursions of these *n*-alkanes can provide information on forest and/or grassland vegetation because they are derived from terrestrial higher plant waxes (Eglinton and Hamliton 1967).Therefore, the major changes in above mentioned long-chain odd-carbon-numbered *n*-alkanes reflect shifts in terrestrial vegetation at the source area, as well as shifts in the photosynthetic pathway. This variation directly can be interpreted as vegetation community changes, thus climate changes. The abrupt increasing excursion of even numbered*n*-alkanes, *n*C22 and *n*C24occurred around4.5ka in the cores SJP15-4 is very meaningful (Fig. 4). This may imply some submerged vegetation has been flourished in this interval.Also, the strong increase of nC24 appeared at 1.8 ka in core SJP15-4 can be explained as flourish of floating macrophyte.

There is a distinctive short-term event-like (STER) features in the total *n*-alkane excursions in the two cores. The concentration of most *n*-alkane compound increase at 3.0 ka, but in detail it’s excursion peaked at ca. 2.5 kain core SJP15-2, and peaked at ca. 1.8 ka in SJP15-4 (Fig. 4). Each *n*-alkane compound has a specific excursion with time differences and different concentrations. For example, the mid-chain *n*-alkanes (*n*C22 and *n*C24) showedabnormal increase at ca. 4.5ka, and centered at ca.4.3 ka. Similarly, the increase of mid-chain *n*-alkanes (*n*C21, *n*C23)has been observed during mid-late Holocene at the Swamp Lake of USA, and this mid-chain increase was explained by an increase in the abundance of aquatic plants and availability of shallow water habitat in rising lake level (Street et al. 2013). Therefore, increase of mid-chain *n*-alkanes occurred around 4.5 ka in our study might be associated with temporal environmental changes due to the flourishing of aquatic plantsunder the shallow water environment.

The comparison of each numbered *n*-alkane concentration of the two core can provide more detailed time-dependent variations (Fig. 5).Long-chain *n*-alkane such as *n*C29, *n*C31 are predominantly present throughout the two core. However, mid-chain *n*-alkane such as *n*C24~26 are differ from each other. Theconcentration of mid-chain *n*-alkane in SJP15-4 is relatively higher than that of SJP15-2 at the time of 1.70 ka, whereas it is less at 2.26 ka than that of SJP15-2 at 2.33 ka(Fig. 5). Therefore, as shown in Fig. 4 and Fig. 5, each *n*-alkane concentration has been differentiated fromtime to time in each core site, implying time-dependent mid-chain *n*-alkane fluctuation in accordance toclimatic and vegetation changes.

Badejo et al. (2017) examined terrestrial organic matter inputs based on *n*-alkane concentrations from the surface sediment in study area, and suggested large inputs of materials from the mouth of the Seomjin River to the inner shelf area, with a slight variation that could be due to an indirect supply. According tothat results, the influx of terrestrial material was higher near the Seomjin River mouth, decreasing further offshore. Therefore, various *n*-alkane compounds have been expected to being supplied from a nearby source by Seomjin River and possible faraway mainland sources.

As combined proxies, spatiotemporal distribution of ACL and the CPI of the *n*-alkanesalso have been used to determine the characteristics of organic carbon since they canprovide valuable information on *n*-alkane sources(Sikes et al. 2009; Jia et al. 2015; Li et al. 2015). It is known that the distribution of ACL has been linked to the geographical distribution of fluvial,aeolian inputs and source regions (Poynter and Eglinton 1990). Therefore, it is possible to delineate*n*-alkane sources by identifying environmental differences, even though the usefulness of this approach is limited to a source-specific *n*-alkane distribution (Jeng 2006).

In our study, there was a narrow and distinctive variation in the ACL with a similar trend between two core sediments.Furthermore, a clear difference between the two cores appears around ca. 2.3ka, where one of three switching points occurs(Fig. 6). Overall, values of ACL in our core start to decrease at 3 ka, and it recovered at 1.8 ka. Given that ACL refer to average number of carbon atoms per molecule based on the abundance of the odd-carbon-numbered higher plant *n*-alkane (Poynter and Eglinton 1990), this lowered interval implies the variabilities of alkane distribution, i.e., it might be related to wetter climaticcondition. One possible reason for the variation in the distribution is the ACL variation of *n*-alkanes in different plant species.Based on this decreased ACL interval (STER), we infer that our study area also experienced a susceptible East Asian monsoon climatic changes within this short-term period.

CPI values of the waxes also provide additional information on *n*-alkane source because it originates from terrestrial plant community. Usually it shows wide range of variation; terrestrial higher plants usually have values of 5~10 (Hedges and Prahl 1993), lower values < 1 is thought to oil contamination in modern sediments (Jeng 2006; Seki et al. 2006). In the present study, the average CPI of 3.06 (range = 1.74–4.93) in core SJP15-2, and 3.2 (range = 1.77–4.38) in core SJP15-4, respectively. The highest value (4.93) occurred at 1.8 ka at SJP15-2 close toSeomjin River/inner shelf area, possibly associated with an influx of *n*-alkanes from a terrestrial leaf wax source. A decrease in the CPI value was apparent in the 3.0–2.3-ka layer in both cores, with values of < 3 (Fig. 6). A previous study suggested that CPI values < 3 indicate oiled sediments (Farrington and Tripp, 1977). However, it is difficult to evaluate whether core sediments were contaminatedor not, because the used sample is not modern, this is beyond the scope of our discussion.

Like ACL and CPI, the P*aq* is indicative of paleovegetation changes (Yamamoto et al., 2010). Sike et al. (2009) used this paleo *n*-alkane indexto detect the pattern of paleovegetation as follows: P*aq* = (C23 + C25)/(C23 + C25 + C29 + C31). This formula indicates the relative abundance of C3 plants with respect to C4 plants. Increasing P*aq* values indicate an increasing ratio of C23and C25. A P*aq* value < 0.1 corresponds to terrestrial plants; 0.1–0.4 corresponds to emergent macrophytes; and 0.4–1 corresponds to submerged macrophytes (Ficken et al. 2000). In our study, the values ofP*aq*does not exceed 0.4, but it includes increased intervalfrom 3.0 kato1.8ka in two cores with a peakat2.3ka(Fig. 6).Thus, the increasing tendency from 3.0 ka to 2.3 ka indicates an expansion of C3 plants. This means that the P*aq* is strongly associated with the ACL and CPI, both of which indicate climatic and paleo-vegetation changes.

Similarly, *n*C31/*n*C23, *n*C31/*n*C25, *n*C31/*n*C27and*n*C31/*n*C29ratios also widely used as an indicator of past environmental changes such as paleovegetation and paleoclimate variation(Hanisch et al. 2003; Xie et al. 2003; Zech 2006; Horikawa et al. 2010). Two distinct boundaries were observed at 3.0kaand 1.8ka in above mentioned four indices (Fig. 6).In particular, the *n*C31/*n*C29 ratio is used to discriminate *Sphagnum* from other plants because *n*C29 is relatively scarce in *Sphagnum*. This interval from 3.0 ka to 1.8ka corresponds well with the vertical distribution of *n*-alkanes, increasing TOC (%), and decreases in the ACL and CPI. All of these proxies show a clear change between 3.0 ka and 1.8 ka, supporting vegetation and climate changes.A striking depleted STER corresponds to wetter/humid climatic conditions. This climate condition supported by expansion of C3 plant, as increase of P*aq* as shown in Fig.6. All of multiple proxies of these ACL, CPI and P*aq* are very useful to understand paleovegetation and paleoclimate variations in study area.

**Individual carbon isotopes of *n*-alkanes (13CALK) and sources**

The *n*-alkane compound-specific isotopic ratio (13CALK) has higher potential for complementing *n*-alkane distributions and providing more reliable *n*-alkane provenances because *n*-alkane isotopes show no fractionation during photo-oxidation and microbial degradation (Collister et al. 1994; Ahad et al. 2011). This means that different sources result in different characteristics for 13CALK values. Previous research showed that 13CALK values for C4-derived *n*-alkanes were all within the range of −18.5‰ to −24.5‰, and those for C3-derived *n*-alkanes were more depleted, ranging from approximately −31.4‰ to −38.6‰ (Collister et al. 1994). For this reason, 13CALK has been widely used for compound source discrimination in various environments (e.g., Jeng 2006; Ahad et al. 2011).

In this study, the entire range of odd-numbered *n*-alkane carbon isotopes (13CALK) showed overall comparable excursions (Fig. 7).The distribution and the entire range of odd-numbered 13CALK does not show homogenous excursion, but it shows large fluctuation at around 1.8 ka boundary. Judging from the criteria reported previous result, our 13CALKratio ranged from about -21‰ to -24‰ is regarded asintermediate ratio of C3 and C4 plants, thusas mixture of C3 and C4-drived consequence. In particular, the 13CALK excursions are very complicated and characterized by an abrupt positive shift of 13*n*C21 and13*n*C33 and negative shifts of 13*n*C2313*n*C25in SJP15-4 at 18ka.The excursions of these individual 13CALK values were decoupled in several compounds such as*n*C23 and *n*C33. Among floating macrophytes groups, such as *n*C21, *n*C23, and *n*C25, seems to differ from each other, and also the excursion of this group can be distinguished from emerged land macrophytes, such as *n*C29, *n*C31, and *n*C33 (Fig. 7). As mentioned previously,because of the different sources of *n*-alkane isotopes have different values for 13CALK, the wide variation of 13CALK values in our study suggests major paleo-vegetation community change. As far as we focused on compound-specific 13CALK between 3.0 ka and 1.8 ka, it is quite complicated to compare each *n*-alkane compound provenance.As indicated in Fig. 7, compound-specific 13CALKmay show some switching points at about 3.0 ka and 1.8 ka as combined indices shown in Fig 6. This means that *n*-alkane compound changes would be matched well with its corresponding 13CALK.

As *n*-alkane compound of *n*C29 and *n*C31 are illustrated, their compound-specific 13*n*C29and 13*n*C31 showsimilar excursion with other proxies between 3.0 ka and 1.8 ka (Fig. 8).Even though both concentrations of *n*C29 and *n*C31show rather homogenous excursion between two core site,13*n*C29and 13*n*C31 are quite different from those of *n*C29 and *n*C31.The difference of compound-specific *n*-alkane source can be expressed asㅿ13CALK. Relatively large difference in 13CALK (around -1.5) appeared around 2.0ka in SJP15-4 may imply differences of vegetation community at this site, and correspond to paleovegetation change from C4 plants to C3 plants. As mentioned, this interval is regarded as wetter climatic interval, so depleted13*n*C31 in SJP15-4may support paleo-vegetation community changes. As mentionedpaleovegetation index, P*aq*, also indicate C3 plant expansionduring STER period. Therefore, compound-specific ㅿ13*n*CALK strongly indicative of variations of compound source and supports individual *n*-alkane excursion. Furthermore, compound-specific *n*-alkane can providemore detailed information on paleoclimatic and paleovegetation changes in our study area.

**Implication for paleoclimate variations in East Asia**

In East Asian region, the most well-known studies on paleoclimate variation during the past glacial–interglacial periods were carried out using stalagmites, which showed evidence of strong coincident variation with climatic variation in high-latitude ice core studies and East Asian monsoon variation (Xiao et al. 1999; Wang et al. 2005; Li et al. 2015). In these studies, *n*-alkanes proxies were also widely used to track past paleoclimate variation during the Quaternary period (Li et al. 2013; 2015; Street et al. 2013;Hyun et al. 2017). Several previous records have focused on Holocene climate changes by using indirect proxies such as detrital quartz in Yangtze River off inner shelf area (Wang et al. 2020), and influx of quartz as an indicator ofaeolian dust in Cheju Island, Korea (Lim et al. 2005; Lim and Matsumoto 2008). These studies used proxies associated with the East Asian monsoon system. Therefore, detecting any of these climatic events at a high-resolution scale is important for detailed assessments of paleoclimate variation in the East Asian monsoon area.

The concentration of *n*-alkanes, the *n*C31/*n*C27 proxy, and the δ13Corg values in this study show a distinguished variation since the mid-Holocene. Because thesekinds of combined proxies imply humid conditions in East Asia (e.g., Li et al. 2013), our data showing a decreasing interval between 3.0 ka and 1.8ka may correspond to a wetter climatic condition as indicated by Li et al. (2013) in East Asian region. However, a climatic event-like wetter condition during the late Holocene (3.0–1.8 ka) has not reported yet from the nearby areas. Therefore, it is also necessary to discuss with other paleoclimatic indicator studied previously. One of similar result can be found from the detrital quartz from the Yangtze River, off East China Sea (Wang et al. 2020) (Fig. 9). This earlier study indicated that enhanced and/or reduced EASM intensity strongly associated with southward shift of EASM precipitation during this interval.

As a paleoclimate monsoon variation, our *n*-alkane data clearly show distinguished variation in STER. The total *n*-alkane concentration started to increase from 3 ka, then peaked at 2.5 ka in SJP15-2 and at 1.8ka in SJP15-4, then was terminated at 1.8 ka in both cores. Individual *n*C22 and *n*C24 records showed an abrupt increase between 4.5 ka and 3.8 ka in SJP15-4. These mid-chain *n*-alkanes that originate from floating plants indicate that floating macrophytes flourished during this STER. Previous works in nearby areas have shown that the climatic environment during the Holocene enabled the transport of aeolian dust and a high flux of terrestrial *n*-alkanes (Lim et al. 2012; Badejoet al. 2017). Relatively large amounts of terrestrial leaf wax might have been supplied to the present study area, enhancing the total *n*-alkane concentration. Concerning the switching points of 3.0 ka and 1.8 ka, they coincide with that of previous data from Wang et al. (2020) and Lim et al. (2008).

Lim et al. (2008; 2015) demonstrated that paleoclimatic changes in Korea may be associated with the East Asian monsoon and the D-O cycle based on the input of aeolian quartz records from the Cheju Island, South Korea. In these result, the paleovegetation of Cheju Island was strongly linked with East Asian monsoon climatic variation, and the quartz influx has been influenced by the pathway change of the westerlies. In our study, event-like distinctive interval between3.0 ka and 1.8 ka may correspond to long range of global climatic variation, and might be associated with East Asian monsoon environmental changes since the mid-Holocene (Fig. 9).The clear decrease of influx of aeolian quartz begin at about 3.3 ka, and it recovered at around 2.2 ka. The timing of this fine quartz input is rather earlier than our *n*-alkane record (3.0~1.8 ka). Nevertheless, assuming that decrease influx of fine quartz as diminished westerlies, and thus more moisture, in turn, this decrease influx of fine quartz may bring vegetation changes.

Large amounts of terrestrial leaf-wax *n*-alkanes would have been transported, particularly during glacial periods and the Holocene. Due to the dry conditions during glacial periods in East Asia, terrestrial *n*-alkanes were transported far across the Pacific Ocean (Ohkouch et al. 1997) and the East Sea (Yokoyama et al. 2006). Also, aeolian dust from mainland China could be transported via the westerlies to Cheju Island (Lim et al. 2005; Lee et al. 2014) and even in present study area. A large amount of terrestrial leaf wax could have been derived from nearby land areas. Therefore, our record of *n*-alkane concentrations and their carbon isotopes reflects information about not only the *in situ* paleo plant community but also allochthonous terrestrial *n*-alkanes which were associated with east Asian monsoon climatic variation. Allochthonous leaf wax could be transported from nearby and faraway places, and these delivery variations could have been associated with local climatic changes, as well as East Asian monsoon variation.

**Conclusions**

The time-dependent distributions of sedimentary 13Corg (‰) and *n*-alkanes of organic matterwere investigated to link paleoclimate and oceanographic changes since the mid-Holocene. The proxies (TOC, 13Corg, *n*-alkanes, 13CALK), representative terrestrial biomarkers, and long-chain*n*-alkanes exhibited distinctive variations. In particular, 4.5 ka, 3.0 ka, and 1.8 ka are switching points, indicating that the input of terrestrial biomarkers has been changed in accordance with paleoclimate variation in East Asia.

The other proxies, including ACL, CPI, and P*aq*, also strongly coincide with the excursions of long-chain *n*-alkanes. In particular, *n*C31/*n*C27 was lower during 3­–1.8 ka, implying relatively wetter climatic conditions. Compound-specific 13CALKindicates more clearly distinguished switching boundaries, and show clear difference of compound source between 3.0 ka and 1.8 ka. A synthetic comparison with previous works indicated that most paleoclimate proxies of stalagmites, fine quartz, and detrital quartz show coincident variation, and are well matched with our *n*-alkanes proxy. Therefore, this study provides information on paleoclimate variation that might have been regulated by centennial-scale dynamics such as the East Asian monsoonas well as local paleoclimate effects since the mid-Holocene in the southern inner continental shelf of the Korean peninsula. In this study, specially, distinctive wetter climate condition during Late-Holocene (3.0 ka ~1.8 ka)has been prevailed in southern inner shelf of Korean peninsula.

**Abbreviations**

KCC: Korean Coastal Current; TWC: Tsushima Warm Current; STER: short-term event-like record; EASM: East Asian Summer Monsoon; EAWM: East Asian Winter Monsoon

**Declarations**

**Availability of data and material**

The dataset supporting the conclusions of this article is available in the Korea Institute of Ocean Science and Technology. Please contact author for data requests.

**Funding**

This work was supported by KIOST grant (PE99775) and K-IODP program (PM61460)

**Competing interests**

The authors declare that they have no competing interest.

**Authors’ contributions**

SM (the first author)designed and proposed this study, and also made manuscript. NY carried out the experimental study and arranged the data. JH draw the figure and discussed the content. All authors read and approved the final manuscript.

**Authors’ information**

SM have been interested in paleoclimatology and paleoceanographic fields for more than twenty years, also he published several papers with similar methods of n-alkanes and its isotopic study. Recently he changed his interest into Holocene period because Holocene holds more important knowledge which is close to environmental state between nature and human beings. As a principal scientist in KOST, he like to contribution on the research of Holocene paleoenvironmental study through this kind of work. This is the first manuscript to PEPS, and willing to more contribute to this journal.

**Acknowledgement**s

The authors thankanonymous reviewers for their critical reading of the original document. The first author thanksto Dr.Irino and Wang, Hokkaido University, and Dr. Lim, Korea Institute of Geoscience and Mineral Resources (KIGAM), for their valuable provide of raw data and comments and suggestions, in particular. This study was conducted with support from Korea Institute of Ocean Science and Technology (KIOST) grants (PE99775). The Korea International Ocean Discovery Program (K-IODP) program (PM61460) also supported this study.

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**Figure Legends**

Figure 1. Sampling locations of the two core sediments collected from coastal area of southern continental shelf of Korean peninsula. Previous works cited in this study were indicated as black dots (left). (A) is Dongge cave (Dykoski et al. 2005), (B) offYangtze River (Wang et al. 2020), and (C) is Cheju Island, Korea(Lim et al. 2005). KCC is Korean Coastal Current, TWC is Tsushima Warm Current, respectively.

Figure 2. Sedimentation rate of the two cores. Both core show homogenous mud sediment and have a similar sedimentological characteristics and high sedimentation rate. A.S.R means Average Sedimentation Rate (cm/kyr).

Figure 3. Vertical distributions of TOC (%), C/N ratio, 13Corg(‰)and15NTN(‰) in two cores. Data were drawn by three-point moving average. Distinctive three switching points (4.5, 3.0 and 1.8 ka) in their excursionare shown by dotted lines.

Figure 4. Vertical*n*-alkane distribution in two cores. The total concentration of *n*-alkanes (*n*C25-35) is calculated by sum from each*n*C25-*n*C35. Due to rather specific excursion in even-numbered *n*-alkanes distribution, odd and evennumber *n*-alkanes profiles are illustrated together.

Figure 5. Representative individual*n*-alkane concentrations in studied two core. These time-dependent differences in concentration were attributed either to changes in paleo-plant population or supply changes of *n*-alkanes due to the paleoclimate variation.

Figure 6.Average chain length (ACL), carbon preference index (CPI) and paleo-vegetation index (P*aq*).Several representative combined *n*-alkanes indices were also illustrated. These distinctive changes inproxies were very clear between 3.0ka and 1.8ka, suggestingwetter climate condition and increase of C3 plant during this time.

Figure 7. Vertical profiles ofodd-numbered compound-specific carbon isotope. The excursions are quite different in 13*n*C21, 13*n*C23, 13*n*C27, 13*n*C33, 13*n*C35 suggesting provenance changes due to the paleo-vegetation change or/and supply change due to climatic changes.

Figure 8. Comparison of single *n*-alkane compounds (*n*C29, *n*C31) and their compound-specific carbon isotope. The difference of compound-specific*n*-alkanes between two 13*n*C31, 13*n*C29 is show as \*13*n*C31-29.

Figure 9. Synthetic comparison with previous works.(A) Dongge cave (Dykoski et al. 2005), (B) Yangtze River inner shelf(Wang et al. 2020),(C) Cheju Island, Korea(Lim et al. 2008),(D) SJP15-2 core(This study),(E) SJP15-4 core (This study).

Tables

Table 1. The results for 14C age.

Table 2. Comparison of average values of carbon isotope with previously reported values from around the Korean peninsula.

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Table 1. 14C age data used in this study. | | | |  |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| Core ID | Latitude (°N) | Longitude (°E) | Core depth (cm) | Material | 14C age (conventional) |  | Two sigma(σ) age range | | Median (cal yr BP) |  |
| SJP15-2 | 34.6780 | 127.8873 | 74-76 | Organic sediment | 2110±30 BP |  | Cal BC 200-45 | (Cal BP 2150-1995) | 2072 |  |
|  |  |  | 252-254 | Organic sediment | 2930±30 BP |  | Cal BC 1220-1020 | (Cal BP 3170-2970) | 3070 |  |
|  |  |  | 428-430 | Organic sediment | 5000±30 BP |  | Cal BC 3930-3875 and Cal BC 3805-3705 | (Cal BP 5880-5825) (Cal BP 5755-5655) | 5779 |  |
| SJP15-4 | 34.5213 | 128.1092 | 74-76 | Organic sediment | 2640±30 BP |  | Cal BC 830-790 | (Cal BP 2780-2740) | 2760 |  |
|  |  |  | 252-254 | Organic sediment | 3480±30 BP |  | Cal BC 1885-1735 and Cal BC 1715-1695 | (Cal BP 3835-3685) (Cal BP 3665-3645) | 3708 |  |
|  |  |  | 422-424 | Organic sediment | 4260±30 BP |  | Cal BC 2910-2875 | (Cal BP 4860-4825) | 4843 |  |

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Table 2. Carbon and nitrogen isotope values with average values | | | | |  |  |  |  |  |  |
|  |  |  |  |  |  |  |  |  |  |  |
| Sampling area | |  | Highest 13C (‰) | Lowest 13C (‰) | Average 13C (‰) | Highest 15N (‰) | Lowest 15N (‰) | Average 15N (‰) |  | References |
| East China Sea & Okinawa Trough | |  | -19.60 | -24.10 |  | 6.30 | 3.50 |  |  | Kao et al. (2003) |
| East China Sea, core sediment | |  | -20.70 | -25.49 |  | 6.60 | 3.30 |  |  | Hyun et al. (2006) |
| East Sea core sediment\* | |  |  |  | -24.60 |  |  | 3.90 |  | Khim et al. (2008) |
| Jiaozhou Bay, Qingdao, China | |  | -23.40 | -26.10 |  |  |  |  |  | Wang et al. (2006) |
| Gwangyang Bay, Korea | |  | -20.47 | -25.86 | -21.50 | 8.57 | 4.37 | 6.99 |  | Hyun et al. (2011) |
| Masan Bay, Korea | |  | -19.97 | -22.69 | -20.76 | 7.80 | 3.93 | 5.73 |  | Hyun et al. (2011) |
| East shelf of East Sea, Korea | |  | -20.48 | -21.36 | -21.05 | 9.80 | 4.44 | 5.96 |  | Hyun et al. (2011) |
| Seomjin off, and southern shelf | |  | -20.26 | -25.86 | -21.47 | 8.57 | 4.37 | 6.72 |  | Hyun et al. (2013) |
| Inner continental shelf, Korea | |  | -20.26 | -25.86 | -21.47 | 8.57 | 4.37 | 6.72 |  | Badejo et al. (2017) |
| southern continental shelf, Korea\*\* | | | -21.00 | -21.79 | -21.39 | 5.94 | 2.81 | 4.12 |  | This study |
| \* East Sea core sediment is 96EBP4, and the average values were from the data during the glacial period | | | | | | |  |  |  |  |
| \*\* Southern continental shelf, core sediment | | | |  |  |  |  |  |  |  |