Abstract of Doctoral Dissertation

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Paleoenvironmental and sedimentary systems within the land-ocean interface reconstructed by biomarker analysis of Miocene sediments from Brunei and Hokkaido, Japan
（ブルネイおよび北海道の中新統堆積岩のバイオマーカー分析による陸-海インターフェイスにおける古環境および堆積システムの復元）

Lipid biomarkers are created by specific marine and terrestrial organisms and are capable of being preserved in the geological record throughout time. They are useful in reconstructing environmental systems as their source organisms respond to changes in the environments such as aridity and precipitation, salinity, temperature and etc. While most studies on paleoenvironmental reconstruction use calcareous fossils such as foraminiferas or diatoms, their preservation can be significantly and negatively affected by diagenesis. Lipid biomarkers have been shown to be well preserved even when microfossils are poorly preserved, giving them an advantageous but underused method in environmental reconstruction. In this Doctoral Dissertation, I use lipid biomarkers to reconstruct environmental and sedimentary systems in the land-ocean interface of Brunei and Hokkaido, Japan. Specifically, I reconstruct how sedimentary processes may create large carbon sinks in sandstones, the changes in hinterland vegetation due to tectono-sedimentary changes during stable climates, and the changes in terrestrial and marine carbon production during global cooling and evolving East Asian Monsoon.

In chapter 1, the effects of sedimentary processes on terrigenous carbon preservation within sandstones were investigated. Sediments rich in organic matter (OM) deposited by sandy floods such as crevasse splay (floodplain) and hyperpycnites that were deposited in the deep-sea were collected from the Bibai Formation exposed in Bibai and the Kawabata Formation exposed in Yubari, respectively, with the former representing early-stage floods while the latter represents later stage floods. OM-rich sediments deposited by tidal reworking were collected from the Belait Formation exposed in Meragang Beach of Brunei. Using pr/ph (pristane/phytane) and C_{27} sterane ratios, terrigenous and marine OM input and transformations from source to sink were analysed. It was found that early-stage floods have high amounts of OM_{terr} but no $OM_{mar.}$ However, as floods flow seawards, some OM is lost due to escape from the flow due to density reversal of less-dense plant deposits compare to seawater. Marine OM is also incorporated into later-stage sandy flows as sea-floor sediments are eroded and incorporated into the sandy flow, with mud-clast being a contributing factor towards their detection and preservation in sandy-deposits. Analysis of later-stage hyperpycnal flows also shows more woody material in denser deposits with more leafy material in less-dense deposits, further suggesting that organic matter distribution is due to density differences between the organic material and the flow. Lastly, in tidal-reworked sediments, two OM-rich facies were identified, which are rippled sandstones with wavy bedding (a type of mud-drape) and trough cross-stratified sandstones. Rippled sandstones have high amounts of TOC%, leaf wax-derived *n*-alkanes, and diatom-derived highly branched isoprenoids (HBI) when compared to OM-poor sandstones. As tides flow, ripples are created, but the flow energy is too high to enable settling of OM. However, before the tide reverses, there is a period of slack/stagnant water that is calm enough to allow OM to be deposited along with mud on the ripples – creating a wavy bed rich in OM. For the trough cross-stratified sandstones, which are created by strong unidirectional water currents, OM is often found on the lee-side of strata. It is believed that small eddies formed in front of the lee-side of ripples trapped OM on the lee-side of the rippled. Unlike rippled sandstones with wavy lamination, no marine OM biomarkers were found, as these sediments were not strongly under the influence of bidirectional currents. These studies suggests that sandstones can become large and important carbon sinks depending on their respective sedimentary processes, in addition to mudstones.

In chapter 2, the effects of uplifting and expansion of low-land environments on vegetation patterns during stable climates were investigated. Samples were collected from shallow-water Belait Formation outcrops (about 12 Ma) exposed in Meragang Beach and deeper offshore Miri Formation outcrops (about 10 Ma) exposed in Batong of Brunei, North Borneo. These were deposited during a period of continuous uplifting and lowland-expansion with stable climates as shown from $\delta^{18}O$ and mg/ca (magnesium/calcium) paleotemperature reconstructions from the South China Sea (Hutchison et al., 2000; Yang et al., 2021). Cadalene, which is an angiosperm biomarker commonly found in *Dipterocarpacaea* plants are more abundant in younger and distal offshore outcrops. This is contradictory to modern analogues where terrestrial OM is less abundant in distal marine settings than in proximal settings, showing that facies change was not a control in their distribution over longer geological timescales. Since younger outcrops were deposited during a period with a larger low-land hinterland area, it is probable that the increased accommodation space allowed the growth of more cadalene-rich *Dipterocarpaea* plants. Diterpenoids, which are found in conifers, are also in higher abundances in younger outcrops. Since temperatures were stable from 12 to 10 Ma, cooling climates cannot be attributed to their expansion. However, as we see in modern day vegetational patterns of Borneo, conifers are abundant in mountains with high elevations. Thus, it is interpreted that their expansion is related to higher mountain tops created due to continuous uplifting of the Bornean hinterland.

In chapter 3, the effects of increased aridity/reduced precipitation caused by global cooling and strengthening East Asian Winter Monsoon on the Japanese Island and Sea between 15 and 9 Ma was investigated. Samples were taken from mudstones of the Kawabata Formation exposed along the Soumokumaisawa and Higashiyama rivers in Yubari, Japan, which were deposited during the above climatic phenomenas. Leaf wax *n*-alkanes show a slight decrease in younger sediments while gymnosperm-derived diterpenoids show an increase relative to angiosperms. These suggest reduced river discharge and cooling-driven vegetation changes respectively. This in in contrast to a study done by Wang et al. (2001) where pollens in South Japan show an increase in angiosperms, suggesting a warming climate. Thus, it is probable that cooling affected North Japan first as the Subpolar Front migrated southwards. The Average Chain Length (ACL), Carbon Preference Index (CPI), Aquatic Macrophyte Index (P_{aq}) and long-chain *n*-alkanes ratios do not show significant variation throughout the studied interval, suggesting that aridity may have been weak. This is also in contrast with a study done by Peng et al. (2016) where an increase in the above proxies showed two drying intervals between 14.5 – 12.5 Ma and 10 – 4 Ma. These suggest that aridification was stronger in Central Asia than in the Japanese Islands. An increase in C_{27} sterane ratios and HBI concentrations also shows an increase in younger sediments. These increases coincide with lower pr/ph ratios and higher concentrations of a biomarker similar to rearranged gammacerane, though the latter may also have terrestrial origins. A plot of MTTCI (chroman index) against pr/ph ratios also shows a shift from semi-saline or freshwater conditions to normal marine-like conditions during deposition of samples with high marine OM. Additionally, HBI shows a good correlation with gammacerane concentrations ($R = 0.55$) and a pr/ph ($R = - 0.66$), showing that diatoms grow when bottom waters are saline and/or anoxic/dysoxic respectively. These changes in primary productivity could be due to a shift from calcareous to silicate-rich environment of the North Pacific during the Miocene but could also be due to periodic intrusion and upwelling of nutrient-rich Oxygen Minimum Zone (OMZ) deep waters (Lübbers et al., 2019; Tada, 1994). Statistical analysis also shows a very poor correlation between leaf wax *n*-alkanes and HBI. These results show that changes in the Japanese Islands' terrestrial OM production due to aridification is not as intense as Central Asia, while primary productivity is dependent on oceanographic processes rather than terrigenous input from the Japanese Islands.

In conclusion, lipid biomarkers were used to reconstruct the changes in the land-ocean interface that is shaped by the linkage between climate, sedimentary processes and systems and Earth's biosphere. We found that sedimentary processes can influence terrigenous OM preservation in the marine environments, often preserving more organic carbon than mudstones – providing a new, unconventional carbon sink. We also found that vegetation change in the hinterland is not only related to climate change, but also local tectono-sedimentary changes. Lastly, we found that global cooling would change riverine terrigenous OM delivery and vegetation in North Japan earlier than South Japan, while the physico-chemical properties of Japan Sea and diatom-primary productivity mainly controlled by oceanographic processes rather than riverine discharge.