

論文の内容の要旨

論文題目 **Re-evaluation of impact on global carbon cycle by variations in terrestrial weathering and nutrient cycles**

(陸の風化と栄養塩動態の変動が炭素循環におよぼす影響の再評価)

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Carbon cycle via rivers is one of the key processes connecting carbon reservoirs of the continents and the atmosphere and needs to be assessed to fully understand climate change, because riverine carbon cycle and climate are strongly interacting each other. Rivers transport carbon largely derived from chemical weathering from the terrestrial carbon reservoir to the ocean. In a conventional view, only silicate weathering has the role as a long-term CO₂ sink from the atmosphere and is thought to be a very slow-reactive process that stabilizes the Earth's environment only in a geologic timescale, typically in a million years. In this view, the role of other carbon sources to rivers such as carbonate weathering and organic matter has been almost neglected. However, in shorter timescales, these carbon sources can have a rather large impact on climate than silicate weathering. In addition, the cycling of nutrients, which is intimately linked to the cycle of carbon, is also important and can impact on the short-term carbon cycle. The present thesis evaluates two feedback processes of riverine carbon and nutrient cycles to climate change and its impact on global carbon cycle, by field survey and by model simulations,

depending on the timescales of targeted processes.

First, in a very short timescales such as the hundred-year timescale of the ongoing global warming, carbonate weathering may respond to climate change and can act as an additional sink of CO₂. I present the importance of carbonate weathering in short-term CO₂ consumption by a comparison of river water on small subtropical watersheds of aluminosilicate dominated Iriomote Island and carbonate-based Ishigaki Island, southwestern Japan. Contribution of carbonate weathering on higher CO₂ consumption was also found in rivers in Chugoku Region. Rivers in Ishigaki Island exhibited more than 10 times higher alkalinity than rivers in Iriomote Island, which was even higher than the mean value of major rivers in the world, such as St. Lawrence and Mississippi. This high alkalinity was the result of carbonate dissolution enhanced by soil originated CO₂. In spite of the relatively small surface area of carbonate depositions, an averaged alkalinity in Kotoh River and Takahashi River was more than four times higher than Oze River and Yoshii River, indicating a strong influence of carbonate area, even in the whole watershed scale. From the perspective of CO₂ flux between the land and the atmosphere, more than half of riverine alkalinity is considered as CO₂ consumed by chemical weathering. Carbonate weathering, which is driven by soil-derived CO₂, can be further enhanced with the increasing of global warming and mitigate CO₂ release from soil to the atmosphere. With the progress of ocean acidification, the enhanced consumption of CO₂ by carbonate weathering will not be compensated for and should be counted as an anthropogenic sink. As a result of this high inorganic carbon input, the ratio of dissolved CO₂ and nutrients of the river water in Iriomote and Ishigaki Islands was higher than that of marine organic matter such as the Redfield's Ratio, making the coastal seas locally a potential source of CO₂ for the atmosphere. The ratio was generally lower in rivers in Chugoku Region and even lower in dam lake waters. Reflecting biological activities in lake water, concentration of nutrient elements was quite depleted in lake water, and the reduction of nutrient concentration was also found in the downstream waters, too. Dam lakes are acting as an absorptive storage of riverine nutrient elements, especially that of P and Si.

Secondly, a large change in riverine transport of organic matter can also have an important impact on climate. Using an earth system model, this study also addressed a feedback process of riverine biogeochemical cycles in the glacial-interglacial cycles. In the past 800

thousand years and before industrialization, the largest variations in atmospheric CO₂ concentration ($p\text{CO}_2$) occurred in connection with the glacial cycles that characterized Earth's climate over this period. One curious feature of at least the last four glacial-interglacial cycles is that atmospheric $p\text{CO}_2$ reached about the same upper limit of 280 ppm during peak interglacial periods and about the same lower limit of 180 ppm during peak glacial periods. Here, I show using a numerical model of earth system that enhanced shelf sediment weathering during glacial sea-level low stand tends to raise $p\text{CO}_2$ even after carbonate compensation and thus stabilize $p\text{CO}_2$ from further reduction. This is because not all nutrients from weathering will be utilized by biology but more importantly because the spatial distributions of carbon and phosphorus from weathering become decoupled in such a way that carbon is preferentially stored in the upper ocean and phosphorus in the deep ocean. In addition, the C:P ratios in continental margin sediments are generally much higher than the Redfield ratio due to preferential remineralization of phosphorus in shelf sediment diagenesis. When these factors are accounted for in the model, the input of organic matter, which corresponds to the observed negative shift in ocean $\delta^{13}\text{C}$ during glacial periods, raises $p\text{CO}_2$ by approximately 5-10 ppm. The same mechanisms operating in the opposite directions during interglacial high stand tend to lower $p\text{CO}_2$ and stabilize it from further increase. The impact of sea level-driven continental shelf exposure and submersion of CO₂ is therefore a negative feedback that may have contributed to limiting the variation of Pleistocene $p\text{CO}_2$ to the observed 100 ppm range.