論文の内容の要旨

The carbon cycle in the Himalayan river basins on both modern and geological timescales: evidence for a role of CO_2 release from river surface water and chemical weathering

（ヒマラヤの河川流域における現代および地質学的時間スケールの
炭素循環の解明: 河川表層からの CO_2 放出と化学風化が果たす役割について）

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The carbon cycle in terrestrial rivers plays an important part in the Earth-surface environment, transporting vast amounts of material from the land to the ocean and providing essential waters and nutrients for terrestrial biota. On modern timescales of 1-100 years, recent studies have reported that abundant CO_2 generated by biological activity in river basins is released from surface waters to the atmosphere. In contrast, on geological timescales of 10^4 to 10^6 years, chemical weathering of silicate rocks in the basins consumes atmospheric CO_2. In this study, I focused on the Himalayan rivers, where active weathering occurs and large spatial variations in geology and land use are observed between the upper and lower basins. In this setting, the riverine carbon cycle on multiple timescales and its significance within the Earth-surface environment was examined, through the following three studies.

(1) Modern timescale: spatial and seasonal variations in surface water pCO_2.

The Ganges, Brahmaputra, and Meghna rivers together have the second largest water discharge in the world. However, few studies have analyzed the partial pressure of CO_2 (pCO_2) and

CO_2 degassing fluxes in these rivers. I investigated the carbonate systems of these rivers, including spatial and seasonal variations in pCO_2, and determined their potential importance. Although pCO_2 was low in the upper reaches of these rivers, owing to active chemical weathering, values were higher than atmospheric pCO_2 along the lower reaches, where deep soils have developed and where high air temperatures promote active soil respiration. Using a simple mixing calculation, it was found that seasonal variations in these river water carbonate systems are controlled by subsurface water flows, which originate in the lowlands and are influenced by soil respiration. In the rainy season, most of the lowlands are inundated, and the contribution of subsurface flow to river water carbonate systems increases, resulting in higher pCO_2 values. Total CO_2 fluxes from the Ganges and Brahmaputra River waters were calculated to be 0.45-1.7 × 10^{11} mol yr^{-1} and 0.62-2.4 × 10^{11} mol yr^{-1}, respectively. In future research, more detailed spatial and seasonal investigations are required to clarify the role of terrestrial ecosystems in the short-term global carbon cycle.

(2) Geological timescale: chemical weathering and long-term CO_2 consumption reconstructed from major ion chemistry.

The role of Himalayan river systems in the long-term global carbon cycle has been a subject of great interest, especially in the context of past climate change such as global cooling during the Cenozoic. However, there are few reliable geochemical data from the Ayeyarwady River. This study focused on reevaluating chemical weathering in the Himalayan watersheds by carrying out chemical analyses of the composition of dissolved substances in samples taken from the Ayeyarwady, Mekong, and Chao Phraya rivers. Comparisons of water quality showed that, unlike in previous studies, the total alkalinity budgets of the Ayeyarwady are dominated by carbonate rather than silicate weathering. Long-term CO_2 consumption by silicate weathering in the Ayeyarwady is estimated to be only 63-145 × 10^{9} mol yr^{-1}, which is only 10 % of the previous estimate. The results of this study also suggest that all Himalayan watersheds only account for approximately 10 % of the total global CO_2 consumption by silicate weathering. Although further studies are needed, chemical weathering and associated CO_2 uptake in the Himalayas likely played a lesser role in past long-term global cooling than previously thought.

(3) Geological timescale: development of analytical procedures of magnesium and silicon isotope ratio measurement to gain further insight into chemical weathering.

Magnesium and silicon isotope ratios ($\delta^{26}Mg$, $\delta^{30}Si$) are new potential proxies for gaining insight into Mg- and Si-related processes including chemical weathering in river basins. In this study, I examined the availability of $\delta^{26}Mg$ for the carbon cycle, by investigating the spatial and seasonal variations in concentrations and isotope ratios of Mg and Sr in the Ganges, Brahmaputra, and Meghna rivers. $\delta^{26}Mg$ values of these river waters reflected the upstream lithology (dolostone/silicate) throughout the year. The spatial and seasonal variations in the major ion concentrations of the water suggested that the ions may originate from different sources. This result implies that $\delta^{26}Mg$ ratios are the best tool for identifying Mg sources and their chemical reaction histories, which can impact on the global carbon cycle. Additionally, the Ganges, Brahmaputra, and Meghna rivers were found to play an important role in the Mg isotope budget of the ocean, transporting as much as 4 % of the total riverine flux of Mg^{2+} to the ocean, with a $\delta^{26}Mg$ value slightly lower (-1.2 ‰) than the global river average. I also developed a Si separation method, which is essential for $\delta^{30}Si$ measurements. I revised the cation exchange methods reported by a previous study, and conducted repeated pilot studies. The Si recovery rate of this study was 102.3 ± 4.2 % (n = 24), suggesting that separation was successfully carried out in the laboratory environment.