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題目(和文)	熱帯の富栄養化した養殖域におけるリンおよびリン酸の酸素同位体比を用いた生物地球化学的研究
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Thesis Title: Biogeochemistry of phosphorus and application of oxygen isotopes of phosphate in a eutrophic tropical mariculture area （熱帯の富栄養化した養殖域におけるリンおよびリン酸の酸素同位体比を用いた生物地球化学的研究）

Thesis Summary

Mariculture, or fish farming in coastal areas, has become a major source of food and livelihood in developing countries especially in Southeast Asia. However, intensive, extensive and unregulated mariculture activities are also sources of deterioration of water and sediment quality due to excess nutrients and organic matter (eutrophic conditions) from wasted feeds and fish by-products. An example is the mariculture area in Bolinao and Anda, two towns sharing semi-enclosed coastal waters in Northern Luzon, Philippines. Eutrophic conditions in Bolinao have been sustained despite the regulation of fish farm structures in this area after a major fish kill event in 2002. To examine the reasons for the persistent eutrophic conditions and understand their implications to management of mariculture activities to prevent future occurrences of algal blooms, hypoxia and fish kills, it is indispensable that the nutrient dynamics of nitrogen (N) and phosphorus (P) are understood in these areas. The goal of this study is to examine the reason of sustained eutrophic conditions in Bolinao by determining the sources and cycling of phosphorus (P), which is the major component of fish feed-derived wastes, using biogeochemical indices including a novel tool, the analysis of oxygen isotopes of phosphate ($\delta^{18}O_p$).

Spatio-temporal observations showed that mariculture areas exhibited high concentrations of dissolved inorganic nitrogen (DIN) especially ammonium (NH_4^+), and dissolved inorganic phosphorus (DIP, or phosphate) coming from decomposition of uneaten and undigested fish feeds, and fish excretions. Compared to the conventional Redfield ratio (N/P of 16), these materials are enriched in P relative to N, resulting in low N/P ratios (~6.6) of the regenerated nutrients. DIP in the water was higher during the dry season than the wet season possibly due to enhanced accumulation of regenerated nutrients inside the embayment due to the flow pattern. Temporal analysis of satellite images showed that while fish farm structures in Bolinao have been regulated, the structures in Anda continued to increase in number. This has contributed to fish farm-derived organic matter and regenerated nutrients enriched in P that can get advected to Bolinao waters with the residual currents during the dry season. These factors sustained the DIP enrichment and created an N-limited condition that is highly susceptible to sporadic algal blooms whenever N is supplied from freshwater input during the wet season.

Nutrient uptake experiments using natural zooxanthellate corals, seagrasses and seaweeds (macroalgae) were conducted to evaluate the influence of phosphate uptake by biological organisms on oxygen isotopes of phosphate ($\delta^{18}O_p$) in the surrounding water. Live samples of corals *Acropora digitifera, Porites cylindrica* and *Heliopora coerulea*, seagrasses *Cymodocea rotundata, Enhalus acoroides* and *Thalassia hemprichii,* and seaweeds *Gracilaria*

firma, *Padina minor* and *Ulva lactuca* were incubated in nutrient-enriched aquaria. For corals, the $\delta^{18}O_p$ usually increased with time towards the equilibrium value with respect to oxygen isotope exchange with ambient seawater, but sometimes became higher than equilibrium value at the end of incubation. Results indicated that the magnitude of the isotope effect associated with DIP uptake depended on coral species with the greatest effect in *A. digitifera* and the smallest in *H. coerulea*. Although isotope effect during DIP uptake by corals is different among coral species, it is greater than the isotope effect during DIP uptake by seagrasses and macroalgae in general. However, even within the same coral species, large differences in the isotope effect are observed. This is probably due to the fact that multiple processes with different isotope effects operate simultaneously in the incubated coral holobionts. These differences in the isotope effect by various coastal organisms should be considered when the $\delta^{18}O_p$ will be applied to the coastal ecosystem to separate the external DIP source signal and internal cycling.

The different end-member sources of phosphate to the Bolinao and Anda mariculture area were identified by analyzing the oxygen isotopes of phosphate of different types of environmental samples. Based from $\delta^{18}O_p$ signatures, freshwater (rivers, 14.4 ± 0.2 ‰; groundwater, 14.8 ± 1.6 ‰) and fish feed (21.8 ± 0.4 ‰) are two contrasting end-member sources of phosphate to the mariculture areas. Sediment porewater (21.3 ± 0.2 ‰) has similar isotopic signature as fish feeds which may indicate that decomposed feeds are the sources of nutrients to the porewaters, and may also suggest equilibrium values. The contribution of rivers is manifested in the lower $\delta^{18}O_p$ of samples collected at the surface waters during the wet season. Most of the samples from mariculture areas have $\delta^{18}O_p$ values closer to the fish feed signature which confirms that fish feed are the major sources of DIP to the mariculture area.

In this study, therefore, the overall mechanism of sustaining the eutrophic conditions and the occurrences of algal blooms, hypoxia and fish kills was found out. In addition, this study demonstrated the applicability of the analysis of $\delta^{18}O_p$ in eutrophic coastal areas with mariculture influence, and contributed new $\delta^{18}O_p$ signatures for end-member sources. Inclusion of $\delta^{18}O_p$ in future modelling studies may be approached provided that the major isotope effects are incorporated in a quantitative manner. Nonetheless, the identification of feeds as the major source of phosphate in the mariculture areas using the $\delta^{18}O_p$ technique may be helpful in striving for management efforts in the application of feeds.

Contents