

## Carbonate scavenging of rare earth elements in the oceans

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### 論 文 内 容 の 要 旨

The oceanic cycling of rare earth elements (REEs) has not seen full explanation. Widely believed explanation is the combined mechanisms of "boundary exchange" and "reversible scavenging" (Siddall et al., 2008; Arsouze et al., 2009; Oka et al., 2009; Rempfer et al., 2011). In this model, REEs are exchanged with continental margin and the particles scavenge REEs from shallower layers of the ocean and release REEs to deeper layers of the ocean. However, there are the unclearness of what is "boundary exchange" and what carries out "reversible scavenging".

It has recently been indicated that REEs are contained in diatom frustules and that the presence of REEs in them could innovate the understanding of the circulation of REEs in the ocean (Akagi, 2013b; Akagi, 2013a). In this model, diatom frustules accumulate REEs in their frustules at the surface layer of the ocean. The diatom frustules settle down to the deeper layer of the ocean, and they are dissolved into the water with REEs. The released REEs are scavenged by particulate carbonate (Akagi, 2013a). Considering REE scavenging processes are keys to finalize the understanding on the REE cycling in the ocean, herein, I focus on the secondary scavenging with carbonate.

This thesis is comprised of three chapters.

In Chapter 1, I reviewed the understanding of the distribution of REEs in the oceans. The vertical distributions of REEs show nutrient-like profile. The mechanism to make these profiles is explained by "boundary exchange" and "reversible scavenging". Previous studies explain the REEs profiles the model combining "boundary exchange" and "reversible scavenging". However, there is few understanding what is "boundary exchange" and what is "reversible scavenging". In the recent study (Akagi, 2013b; Akagi, 2013a), an idea is introduced that diatom frustules drive circulation of REEs. however, the idea is not universally in the oceans because the idea was led at bearing sea where has high productivity of diatoms. Therefore, I introduced an idea to explain the distribution of REEs in the oceans, the idea is combination of "primary scavenging" and "secondary scavenging".

In Chapter 2, I developed a self-consistent algorithm to evaluate the importance of the two scavenging processes. The calculation requires reported vertical profiles of REEs and nutrients (Si or P) and provides the composition of surface plankton (primary scavenging) and REEs partitioning patterns between "secondary scavenger" and seawater. The results of

calculation show us that carbonate and oxide are the main secondary scavengers of L+MREEs and organic matters are the that of HREEs. It is explained that carbonate particles play important role on scavenging REEs in the oceans by calculation.

In Chapter 3, I developed a new experimental method to determine naturally effective partitioning coefficients of REEs between calcite and seawater. The partitioning coefficients of REEs were estimated conventionally by precipitating calcite in solution. The partitioning reaction in the oceans are not precipitation but more like exchange reaction, and kinetic effect disturb the partitioning if the precipitation occurs rapidly. In my study, I estimate the partitioning coefficients by measuring finer calcite particles which are better reacted with seawater. The results of the experiments show a similar feature with partitioning patterns obtained from observational data and these calculated partitioning pattern in Chapter 2.

In Chapter 4, these results of two separate studies, calculation with double scavenging model and experiments under the nearer natural condition than previous studies indicate that scavenging processes are key factors to better understanding the behavior of REEs in the oceanic cycling, and that, among all, carbonate plays role in redistributing dissolved REEs within water columns.