論文内容の要旨

Biogeochemical processes of suspended particles in the North Pacific

（北太平洋における懸濁粒子の生物地球化学的過程に関する研究）

岩本　洋子

Oceanic suspended matter (SPM) consists of a variety of components, some of which have external sources (e.g. riverine and/or atmospheric input), and some of which are produced internally (e.g. biological activity or non-biogenic precipitation). Because of its relatively long oceanic residence time and large surface area-to-volume ratio, the SPM plays an important role in regulating the chemical composition of seawater *via* the sorption and desorption of trace elements and other substances. The physical and chemical properties of SPM reflect the characteristics of the water mass, including the interaction with biota and/or atmospheric input, and are able to be a clue for elucidation of the ocean biogeochemical processes.

The North Pacific is the characteristic ocean of seasonal and spatial variation of atmospheric mineral dust inputs. Mineral dust aerosols are transported over the North Pacific especially in spring season by the westerlies and deposit onto the ocean surface. It was also reported that the primary production of the subarctic North Pacific are limited by iron, which can be derived by atmospheric mineral dust.

However, there is still only a little knowledge of the SPM, as well as ultimate fate of atmospheric mineral dust particles in the North Pacific. In order to understand biogeochemical processes in the North Pacific, the impact of the sporadic event, such as Asian dust storm, on the SPM in the surface North Pacific is need to be evaluated. In this study, using both bulk and single particle chemical analysis techniques, size and elemental compositions of the SPM collected in the North Pacific area is characterized. The objectives of this study are

1) To obtain general knowledge of spatial variation in number, volume and chemical properties of the SPM collected in the North Pacific during summer, when the influence of atmospheric Asian dust transport was minimal.
2) To characterize size and chemical composition of individual mineral particles in the SPM.
3) To understand the impact of atmospheric mineral dust aerosols on the composition of SPM.
4) To understand how the phytoplankton bloom initiated by the addition of iron from sporadic atmospheric dust event affects the SPM.

SPM were obtained from the North Pacific seawater during the several cruises carried out in summer of 2004 to 2007 on board R/Vs Hakuho-maru (KH-04-3/SEEDSII iron fertilization experiment, KH-05-2 and Leg 1 of KH-06-2) and Mirai (MR07-04 and MR07-05), which belong to the Japan Agency for Marine-Earth Science and Technology (JAMSTEC). In the R/V Tansei-maru (JAMSTEC) KT-07-7 cruise, simultaneous observations in the surface ocean and atmospheric boundary layer over the semi-pelagic western North Pacific were carried out in spring season.

The bulk elemental composition of SPM for biogenic (Si and Ca) and crustal (Al, Ti and Fe) was determined by an X-ray fluorescence spectrometry. Individual particles were also analyzed using an electron probe X-ray micro analyzer and characterized by size and elemental composition (Mg, Al, Si, P, S, K, Ca, Ti, Mn, Fe and Ba). The single particle method for studying SPM provide information unobtainable by those used only based on bulk analysis, for example, the methods make it possible to calculate the number and volume concentrations and size distributions. The elemental data obtained based on single particle analysis shows that the particles are decidedly heterogeneous, and is valuable for understanding the composition of small particles because the results based on the bulk chemical analyses are mainly influenced by the small number of larger particles.

The spatial variation in number, volume and chemical composition of the SPM collected during summer in the North Pacific are significantly affected by primary productivity. The number and volume of SPM are approximately 3 and 5 times higher in subarctic high productive regions than subtropical low productive regions. Organic types particles were most dominant in SPM number, and particles generated by biogenic skeletal material, such as opal or carbonate shell, contributed to the SPM volume in all oceanic areas. The mineral particles are, however, distributed throughout the oceanic regions in the North Pacific, and their relative abundances in the total SPM was 5-15% by number and about 2-7% by volume. The size distribution of the suspended mineral particles in the SPM was similar to that of the atmospheric mineral aerosols, indicating that wherein background level of mineral dust occurs in the marine boundary layer even in summer.

Furthermore, sulfur and phosphorus, mainly of marine biogenic origin, were associated with the mineral particles in the SPM compared to mineral dust aerosols. The single particle analysis shows that bio-related elements (sulfur and phosphorus) were detected more often in the organic type SPM than other trace elements. These results suggest that chemical composition of suspended mineral particles is significantly modified by adsorption or aggregation of organic particles.

The simultaneous observations in the surface ocean and atmospheric boundary layer over the semi-pelagic western subarctic North Pacific were carried out in the spring season. During the observation, mineral dust aerosols transported from the Asian arid area, Kosa, was observed in the marine atmospheric boundary layer. The atmospheric observation suggests that mineral dust aerosols were scavenged by sea fog that appeared over the observation area, and deposited onto the ocean surface. At the ocean surface, both Al and Fe concentration in SPM were significantly increased by deposition of the atmospheric dust aerosols. The average concentrations of Al and Fe of SPM in the surface water were approximately four to five times higher than that during the non-Kosa period. Furthermore, the Al and Fe concentrations during spring were higher than those collected in the same region during the summer by more than 2.5 and 0.7-2.7 times, respectively. The atmospheric deposition of mineral dust particles onto ocean surface from one Kosa-event was directly calculated to be 270 mg m^{-2} $event^{-1}$.

Average diameter of the suspended mineral particles in the mixed layer was 2.1 ± 1.6 μm, similar to that collected during the summer. Average settling velocity of suspended mineral particles was estimated to be 0.35 m day^{-1}, and residence time in the mixed layer (15-40 m) was estimated as 40 –110 days. However, in the summer season, number and volume concentration of suspended mineral particles in the mixed layer was less than 20% of that of the spring season. It is considered that strong seasonal bloom of phytoplankton, especially diatoms are predominant in the spring bloom in the western subarctic North Pacific, may lead to formation of the aggregates, and the suspended mineral particles are incorporated in the aggregates and scavenged rapidly into deep water.

The SEEDS II iron fertilized experiment conducted in the western subarctic North Pacific, one of the HNLC (High Nutrient low Chlorophyll) areas, shows that SPM increased significantly after the iron fertilization in response to the increase in primary production. Throughout the study, Si-rich, Ca-rich and Organic particles were dominant and their number increased inside the fertilized patch; these particles accounted for 21%, 13% and 58% of the particles examined, respectively. There was consistently higher percentage of Ca-rich particles and lower percentage of Si-rich particles inside the patch than outside of it in number, but both types of these particles apparently occupied a larger volume inside the patch than outside of it. Organic particles, that showed having peaks in smaller diameter particles, increased apparently inside the patch with time after iron fertilization. These results suggest that the increase in suspended particles following the iron enrichment was due to a combination of detrital material and live phytoplankton. The experimental studies also provide better understanding of the process and how different particle types respond to the changing conditions during a plankton bloom.

The atmospheric deposition flux of the dissolve iron flux at the Kosa-event was estimated at 130 - 230μg m^{-2} $event^{-1}$, and comparable to that of SEEDS II iron fertilization experiment. The deposition flux is found to have a potential to supply enough amount of iron to lead phytoplankton bloom in HNLC (High Nutrients Low Chlorophyll) waters. This result shows that a sporadic supply of dissolved iron leading to phytoplankton bloom can naturally occur in the western subarctic North Pacific.

This thesis study shows that understanding of the physical and chemical properties in SPM are useful approach to elucidate oceanic biogeochemical cycles, including atmospheric material transport and ecosystem change by sporadic atmospheric event, quantitatively. Further investigations are needed to characterize the morphology of SPM as well as the physical and chemical interactions, such as adsorption and/or dissolution of SPM and associated trace elements. Aggregation/disaggregation processes are also important in understanding the scavenging processes of SPM from the surface into deep waters, in order to establish a relationship between SPM and vertical settling of particles.