論文名：

Environmental study of the behavior of trace elements radiocesium and arsenic in relation to minerals —with special reference to radiocesium behavior in the Fukushima River basin（要約）

新潟大学大学院自然科学研究科

氏名　萩原　大樹

Radiocesium contamination derived from the Fukushima Daiichi nuclear power plant (FDNPP) accident and arsenic contamination in groundwater in Inner Mongolia are serious environmental issue related to mineral in sediments. To understand behavior of both elements in environment, considering factors, for example, sorption and desorption of contaminants on the minerals, organic matter, microbes, water chemistry is important. In this study, I have conducted the several investigations of behavior of minerals with contaminants in river system.

The deposited radiocesium, which caused by FDNPP accident in the Fukushima river basin is transported in the river systems by soil particles and redistributed in the downstream areas. Although predicting the behaviors of minerals that adsorb radiocesium and of radiocesium dissolved in river water within the river systems is essential, the dominant mineral species that adsorb radiocesium have not yet been comprehensively identified. During transportation, part of the radiocesium accumulated on the riverbed at the estuary. However, inventory of radiocesium deposited in the riverbed and natural process of radiocesium release from soil to solution is not well known.

The purpose of this study was to identify mineral species by investigating the ^{137}Cs distribution and the mineral species in each size fraction that are found in the bedload sediments from an upstream reservoir to an estuary within the Tomioka river basin. To quantify the radiocesium deposition on the riverbed in the Odaka River estuary, I investigated the relationship between the physical characteristics of the river bottom sediments and the areal distribution of the radiocesium inventory. Additionally, to investigate fundamental characteristics of radiocesium sorption especially to organic matter in natural process, the radiocesium release experiments were performed using natural indigenous microbial assemblages and contaminated natural sediment samples in Fukushima.

Results show that in the fine sand sediment, which is the dominant fraction in terms of the radiocesium quantity in the river bedload, the radiocesium concentrations of the felsic and mafic minerals are comparable to that of micas. The mafic minerals contain 62% of the radiocesium in the fine sand fraction in the upstream area, while the felsic minerals contain the highest quantities of radiocesium in the downstream area. The radiocesium inventory at the brackish water area is higher compared with that of the freshwater and estuarine marine areas. Moreover, the particle size distribution in the brackish water area shows a high proportion of silt and clay particles compared to the distribution in other areas. The increase in the radiocesium inventory in

this area is attributed to the fine particle sedimentation due to the negligible velocity of the river flow and the flocculation caused by salinity variation. The possibility of uptake radiocesium by using bacteria activity from Fukushima local contaminated soil is indicated. The quantity of radiocesium released accounts for 8 % of radiocesium adsorbed to contaminated soil sediments after using heat treated soils with batch experiment repeated 2 times.

The observations indicated that the quantification of the mineral species and the ^{137}Cs concentration of each size fraction are critically important in predicting the behaviors of the minerals and radiocesium within the Fukushima river basin in the future. Especially, hornblende for radiocesium transporting media was newly emphasized other than vermiculite and clay minerals which have been previously mentioned. About three years after the FDNPP accident, 15%–40% of the total amount deposited in the river catchment is found in the Odaka river estuary. This inventory is necessary for evaluating radiocesium transport in the future. Otherwise, thermal effects to organic materials were clarified and radiocaesium is extracted from contaminated soil by microbial activity with addition of culture media.

Arsenic contamination in groundwater has been potentially recognized as serious environmental problems. High arsenic concentrations in groundwater were detected in the village of Ershe, Jianshe but were not detected in the village of Qishe, Fengle in Inner Mongolia. In this study, core samples from two comparable regions were mineralogically examined. The principal minerals present in the drill core samples are quartz, micas, feldspars, calcite and clay minerals. Variation of these mineral species does not correlate with depth but to lithofacies; minerals were arsenic-rich in muddy sediments and arsenic-poor in sandy sediments. The arsenic content shows a positive correlation with total Fe_2O_3, V and loss of ignition (LOI). In order to identify the chemical forms of arsenic present, a selective sequential extraction (SSE) method was carried out. The results of the SSE experiment suggested that in highly reducing environments, arsenic that is adsorbed on Fe oxyhydroxides is released. Extremely black sand was found in deep drill core samples. To investigate the cause of black sands, inorganic and bacterial culture experiments were carried out. The black color in sands is due to the Fe sulfide greigite evolved from the Fe oxyhydroxide film coating the sand grains. This change in color occurred upon reaction with H_2S gas most probably associated with bacterial activity. Transmission electron microscope (TEM) observations clarified that the sulfide structure of black sand grains is that of greigite. In summary, based on water chemistry, geology, mineralogical characteristics, and geomicrobiological features, the arsenic release mechanisms in Ershe and Qishe are discussed. Then, a possible model was proposed that includes formation of Fe sulfides around sand grains from SO_2^{-4} under strongly reducing conditions.

In summary, the behavior of trace elements of radiocesium and arsenic in relation to minerals for environmental contamination was closely examined in river system.