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Depth distribution of trace elements in pore water collected from Japan Sea sediments associated with methane hydrate

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Methane hydrate occurs in the deep ocean floor, and also in the permafrost layers of Siberia and Alaska. In Japan, relevant gas hydrate studies have been carried out in Nankai Trough since the 1990s and in the Japan Sea since 2004.

This study focuses on the trace element concentrations, with a special reference to iodine, in pore water samples collected from Japan Sea sediments. The long-lived iodine isotope, ^{129}I, which is produced by the spallation of atmospheric and by spontaneous fission, was also measured in order to provide an estimation of the age of iodine.

Marine sediment samples were collected from methane hydrate areas of Umitaka Spur and Joetsu Knoll, Japan Sea, during the cruise with R/V Marion Dufresne (MD179; MH21 Research Consortium) in 2010. The depths of the sediment cores obtained with a giant piston corer reached maximum depths of about 40 mbsf, which is much deeper than previous piston core samples (down to 8 mbsf) recovered by R/V Umitaka-maru on previous cruises in the region. Pore waters were extracted from the sediment soon after recovery. The concentrations of iodine, bromine and some trace metals (such as Mn, Sr, Ba) were later determined by ICP-MS. Samples having a high iodine concentration were selected for ^{129}I analysis. For this, iodine from samples was extracted by solvent extraction and precipitated as AgI. ^{129}I/^{127}I ratios were determined by accelerator mass spectrometry (AMS) at MALT, the University of Tokyo.

Our results show that iodine concentrations in pore water increase constantly with depth in almost all cases. In particular, samples on the Umitaka Spur showed a markedly high increase of iodine concentrations, i.e. the concentration at 35 mbsf was 0.4 mM, which is about 800 times higher than the average iodine concentration in the ocean. Bromine concentration in pore water also increased with the depth, but not as much (only about 1.7 times seawater). On the other hand, profiles show a decreasing trend with depth in pore water chlorine concentrations. This may be explained by the dilution effect with non-saline water produced by the dissociation of methane hydrate.

Analytical results for ^{129}I/^{127}I ratio in pore waters deeper than 3 mbsf ranged from 0.20 - 0.35 x 10^{-12}. These values were much lower than the background value of ^{129}I/^{127}I ratio of 1.5 x 10^{-12} in the ocean in the pre-atomic age. Considering the ^{129}I systematics, iodine age for the samples was estimated to be 30-45 Ma. This age is almost the same or older than the age when the Japan Sea was formed. The results of lower ^{129}I/^{127}I ratios and increasing iodine concentration in pore water with depth suggest that iodine did not originate from shallow marine sediments. The iodine must be derived from deeper layers, such as old sediments rich in organic matter and iodine which were deposited in the sediments at the opening of Japan Sea. Alternatively, since the iodine ratios indicate minimum ages, it may be possible that iodine was released from sediment layers which were formed before the formation of Japan Sea.

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Keywords: iodine, methane hydrate, Japan Sea, radioisotope ratio