Regional variation of CH_4 and N_2 production processes in the deep aquifers of an accretionary prism

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Methane (CH_4), reserved in the subsurface environments such as natural gas and methane hydrates, have recently attracted considerable attention as the important greenhouse gas and energy resource. It is generally considered that CH_4 in subsurface environments is mainly generated by methanogenic archaea (biogenic origin) or through thermal degradation of organic molecules in the sediments (thermogenic origin). The processes of CH_4 production have been investigated by geochemical and microbiological studies at various surface and subsurface environments. Furthermore, it has been reported that a large amount of CH_4 is present in the deep aquifers associated with accretionary prism.

Accretionary prisms are thick sediments that are formed at a convergent plate boundary, and it is observed around the world. In Southwest Japan, accretionary prism is distributed in a wide region from the coastal area of the Pacific Ocean side to the mountainous area. Since the sediment contains layers of water-bearing permeable sandstone, groundwater is anaerobically reserved in the deep aquifers. In addition to the groundwater, it has been reported that a large amount of the natural gas, mainly CH_4 and nitrogen gas (N_2) is present in the deep aquifers. However, the processes of CH_4 and N_2 production in the deep aquifers associated with accretionary prism is poorly understood. The objectives of this study were to identify the origin of the CH_4 that is reserved in the deep aquifers on the basis of the carbon stable isotope analysis. We also determined the processes and potential of microbial CH_4 and N_2 productions using 16S rRNA gene analysis and culture experiments.

The groundwater and natural gas derived from the deep aquifers of accretionary prism were collected from 14 deep wells situated in Shizuoka Prefecture, Japan. CH_4 was the predominant component of the natural gas derived from deep aquifer of the coastal and middle areas of the accretionary prism (>96 vol.%). In contrast, the natural gas collected from deep aquifer of the mountainous area included a considerable amount of N_2 (23-50 vol.%) as well as CH_4. The stable carbon isotope analysis of CH_4 in the natural gas and the DIC in the groundwater, mainly bicarbonate, showed that CH_4 included in the natural gas of the coastal area is thermogenic origin. On the other hand, it was suggested that the natural gas of the middle and mountainous areas mainly contains CH_4 of biogenic origin. Next generation sequencing analysis of bacterial and archaeal 16S rRNA genes showed the dominance of methanogenic archaea, fermentative bacteria, and denitrifying bacteria in the groundwater. Furthermore, high potential of CH_4 production through anaerobic degradation of organic substrates by syntrophic consortium of H_2-producing fermentative bacteria and H_2-using methanogenic archaea was observed from anaerobic cultivation using the groundwater samples collected from the middle and mountainous areas of the accretionary prism. In addition, high potential of N_2 production by denitrifying bacteria was also confirmed from anaerobic cultivation using the groundwater samples collected form the mountainous area.

From these results, it was suggested that CH_4 is produced by a thermogenic process especially in the deep aquifer of the coastal area associated with the accretionary prism, and H_2-producing fermentative bacteria and H_2-using methanogenic archaea contribute to significant CH_4 production in the deep aquifer of the middle and mountainous areas. Our results also suggest that N_2 production

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by denitrifying bacteria occurs particularly in the deep aquifer of the mountainous area.

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