Identification of CH_4 and N_2 production processes in deep aquifers associated with the accretionary prism using geochemical and microbiological approaches

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Accretionary prisms are thick layers of sedimentary material piled up at convergent plate boundaries, where oceanic plate has subducted beneath continental plates in the past or current time. An accretionary prism which found in southwest Japan, which is called as the Shimanto Belt, is a typical and highly studied accretionary prism. The Shimanto Belt was mainly formed during the Cretaceous and Paleogene Periods and originated from ancient marine sediments that were deposited on the Philippine Sea Plate. It is known that large amounts of anaerobic groundwater reserved in deep aquifers associated with the accretionary prism. In addition to the anaerobic groundwater, it has been reported that a high concentration of natural gas, mainly methane (CH_4) and nitrogen gas (N_2), are contained in the deep aquifers. However, CH_4 and N_2 production processes in the deep aquifers are poorly understood.

To identify CH_4 and N_2 production processes in deep aquifers of the accretionary prism in southwest Japan, we performed geochemical and microbiological analyses of the groundwater and natural gas samples derived from deep aquifers of the Paleogene accretionary prism in Shizuoka Prefecture (13 sites), the Paleogene accretionary prism in Kyusyu Island (6 sites), and the Cretaceous accretionary prism in Okinawa Island (4 sites), Japan. The component analysis of natural gas samples revealed that CH_4 is the predominant component of most samples. On the other hand, natural gas of 4 sites in Shizuoka Prefecture contained a 15-50% of N_2 as well as CH_4. The stable carbon isotopic signatures of CH_4 suggested that CH_4 in the natural gas samples was of biogenic origin or a mixture of biogenic and thermogenic origins. In contrast, natural gas of 3 sites in a part of coastal region of Shizuoka Prefecture were shown to contain CH_4 of thermogenic origin. Archaeal 16S rRNA gene analysis targeting microbial communities in the groundwater samples revealed the presence of dihydrogen (H_2)-utilizing methanogenic archaea that mainly belong to the order *Methanobacteriales*. Additionally, bacterial 16S rRNA gene analysis showed the presence of H_2-producing fermentative bacteria and denitrifying bacteria belonging to the classes *Alphaproteobacteria*, *Betaproteobacteria*, *Gammaproteobacteria*, *Clostridia*, *Actinobacteria*, *Bacteroidetes*, *Bacilli*, and *Ignavibacteria*. High potential for H_2 and CO_2 production by H_2-producing fermentative bacteria and CH_4 production by H_2-utilizing methanogenic archaea were confirmed by the anaerobic cultures using groundwater samples amended with organic substrates. These microbial CH_4 production potential was confirmed in all sites except for 3 sites in Shizuoka Prefecture, in which the natural gas mainly contained CH_4 of thermogenic origin. In addition to CH_4, a high potential for N_2 production by denitrifying bacteria was observed in the anaerobic cultures using groundwater samples obtained from 4 sites in Shizuoka Prefecture, in which the natural gas contained high concentration of N_2, amended with organic matter as an electron donner and nitrate as an electron acceptors.

Geochemical and microbiological data obtained in this study demonstrated the regional variation of CH_4 and N_2 production processes in deep aquifers associated with the accretionary prism in southwest Japan. In a part of coastal region of Shizuoka Prefecture showing relatively high geothermal gradient, the thermal degradation of organic matter is considered to be a main CH_4 production pathway. In contrast, it was

demonstrated that ongoing biodegradation of organic matter by a syntrophic consortium of H_2-producing fermentative bacteria and H_2-utilizing methanogenic archaea widely contributes to CH_4 production in deep aquifers of the accretionary prism. In addition to CH_4 production, N_2 production through anaerobic oxidation of organic matter by denitrifying bacteria was suggested to be conducted in a part of deep aquifers in Shizuoka Prefecture.

Keywords: Accretionary prism, Deep aquifer, Methane production, Denitrification

