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Biogeochemical profiles in deep sedimentary rocks in an inland fore-arc basin, Central Japan

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ABSTRACT

Microorganisms thrive in the deep subsurface, but their extent and distribution, type and rate of metabolism are not well constrained, at least in part, owing to geochemical, lithological, hydrological and tectonic variations. We obtained deep sedimentary rocks from a tectonically stable inland basin in central Japan by drilling with minimized microbial and geochemical disturbance. Along with hydrogeological and microbial characterizations, geochemical properties of drilled cores from depths between ~200 and ~350 m were investigated. Except for a depth of 202 m, nitrate and/or nitrite were present in pore-water at all depths investigated in the present study. In most of the pore-water samples, the levels of dissolved Fe and H_2S were low. Additionally, the presence of microbially reducible Fe(III) was not evident in most of the core samples. Based on the acetylene blockage method, it was revealed the denitrification potential was high at depths of 302 and 351 m where pore-water was depleted in nitrate and enriched in nitrite, while the denitrification potential was relatively low at depths of 300 and 340 m where nitrate and nitrite were both detected in pore-water. Potential rates of methane production via CO_2 reduction were higher than those via acetate fermentation in the 302- and 350-m deep core samples. In the silty sandstone interval at depths of 340 and 351 m where permeability was relatively low, isotopically light methane of microbial origin was slightly enriched with sodium and chloride in pore-water. As a moderately mineralized fluid similarly enriched in sodium and chloride is entrained in the deeper subsurface with a depth over 1200 m around the drilling site, the increase in dissolved aqueous species is attributed to the remnant of the mineralized groundwater after the circulation of meteoric groundwater. The ubiquity of type of the sedimentary rocks suggests that the biogeochemical processes presented in this study might not be limited to the terrestrial sedimentary basin in central Japan.

1. Introduction

It is becoming increasingly clear that microbes are widespread in deep subsurface settings. It is estimated such microbes constitute as much as half the Earth's total biomass (Parkes et al., 1994; Whitman et al., 1998). Biogeochemical processes within the deep subsurface are of potential importance in controlling rates of chemical reactions and thereby affecting chemical fluxes from subsurface to near surface environments (Bach and Edwards, 2003). In the past decade, our knowledge of the deep biosphere has expanded, at least in part, owing to numerous deep subsurface drillings (Krumholz, 2000; Parkes et al., 2000; Chapelle, 2001), as well as the construction of deep underground tunnels (Pedersen, 1996; Stroes-Gascoyne and Sargent, 1998; Lin et al., 2006). To investigate subsurface microbiology, it appears that biogeochemical, hydrological, lithological and tectonic processes have to be carefully considered to define the subsurface environment (Parkes et al., 2000; D'Hondt et al., 2002, 2004).

The deep subsurface represents one of the most hostile environments for life to thrive. Major factors limiting subsurface life include temperature (Gold, 1992), porosity (Fredrickson et al., 1997) and the availability of nutrients (McKinley et al., 1997). In contrast, it has been recognized that in some subsurface settings such as the Atlantic coastal plain (Balkwill, 1989), Japan Sea (Cragg et al., 1992) and Cascadia Margin (Cragg et al., 1995, 1996), the biomass, cultivable number of microorganisms or potential rate of microbial respiration increases with increasing depth.

Sedimentary basins along active continental margin and island arc are generally disseminated with biogenic and thermogenic methane (Schoell, 1988; Hunt, 1996). Deep subsurface environments where methane production is biologically mediated have not been clearly

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defined partly owing to limited availability of contamination-free core samples, hydrogeochemical uncertainty and complex geologic and tectonic histories including seismic activities, high rates of sedimentation represented by turbidite, uplift and erosion, transgression and regression and so on (Head et al., 2003; Moore et al., 2005).

For the study of subsurface biogeochemical processes, it is critical to evaluate the disturbance of subsurface conditions by tunnel excavation and drilling processes. For deep sediment and rock sampling, rotary drilling and coring, which require drilling fluids, are commonly used to obtain deep subsurface samples (Phelps and Fredrickson, 2002). As drilling fluids typically contain high levels of microbial cells and dissolved O_2, there appears to be a high risk of contamination during deep drilling. Furthermore, it is becoming clear that tunnel excavation results in profound hydrogeochemical disturbance, which leads to the lowering of hydraulic pressure and degassing (Iwatsuki et al., 2007).

To minimize microbial contamination and the disturbance of in-situ geochemical conditions, we applied drilling fluid that was ultrafiltered to remove microbial cells and subsequently deoxygenated to a conventional drilling procedure to obtain core samples from the deep subsurface. We selected a geologically and tectonically stable fore-arc basin with Tertiary sedimentary rocks for drilling. The main purpose of this study is to better understand biogeochemical processes mediated in the deep sedimentary basin by integrating a new drilling technique.

2. Geological setting of the drilling site

The studied site is located in the city of Nasukarasuyama, Tochigi Prefecture, central Japan (Fig. 1). The site is the northern inland part of the Kanto Plain, which is located in the Kinugawa graben. The lithostratigraphy of this area is well characterized along with the K–Ar dating of tuffaceous key beds (Sakai, 1986; Takahashi et al., 1999; Takahashi and Iwano, 2000): The Middle and Late Miocene Arakawa Group, which consists of marine clastic rocks, is unconformably underlain by the Early Miocene Nakagawa Group consisting of terrestrial volcanic rocks (Fig. 2a). After the emergence of the sedimentary units, the Quaternary Kawasaki Group consisting of sand and gravel unconformably deposited under riverine or lacustrine conditions (Sakai, 1986). This inland area was not invaded by seawater during transgressive and regressive episodes within a Quaternary transgressive sequence.

The Arakawa Group, which crops out along the Arakawa River, consists of four conformable formations: the Kobana, Ogane, Tanokura and Irieno Formations (Fig. 2a). The Kobana Formation (~200 m in thickness) is mainly composed of silty sandstone, while the Ogane Formation (~300 m in thickness) includes shale and silty sandstone. The main rock types of the Tanokura Formation (~210 m in thickness) and the Irieno Formation (~70 m in thickness) are silty sandstone or sandy siltstone. All formations contain a variety of tuffaceous layers in terms of their grain size ranging from fine ash to coarse pumice, as well as their thicknesses ranging from several millimeters to ~60 m. Paleobathymetric reconstruction based on paleontological analyses of diatoms, benthic foraminifers and mollusks suggests that the upper Arakawa Group, which is the main target of the present study, underwent a single upward bathymetric transition from the middle bathyal to outer sublittoral environment (Yanagisawa, 2003).

3. Methodology

3.1. Description of a drilling program

As shown in Fig. 1, drilling points were chosen between the Arakawa and Egawa Rivers (36°42′34″, 140°3′45″, 196.78 m above sea level). A seismic survey around the drilling points was conducted prior to drilling. As consistent with previous field observations, there were no evident faults and folding, and the strata, the strike of which is in the N–S direction, dips toward the west at ~5–10°. Dipping is considered to occur after deposition of the sedimentary sequence, but a geologic event associated with the slight dip is currently unknown.

To obtain a large amount of groundwater for use as drilling fluid, a relatively shallow borehole called Kr-2 was drilled into the lower Kawasaki Group at a depth of ~90 m below ground level (mbgl), where the water table was at a depth of ~60 mbgl. Hereafter, we refer to “meters below ground level” simply as m, unless otherwise stated. Near the Kr-2 borehole, a 352-m deep borehole called Kr-1 was drilled

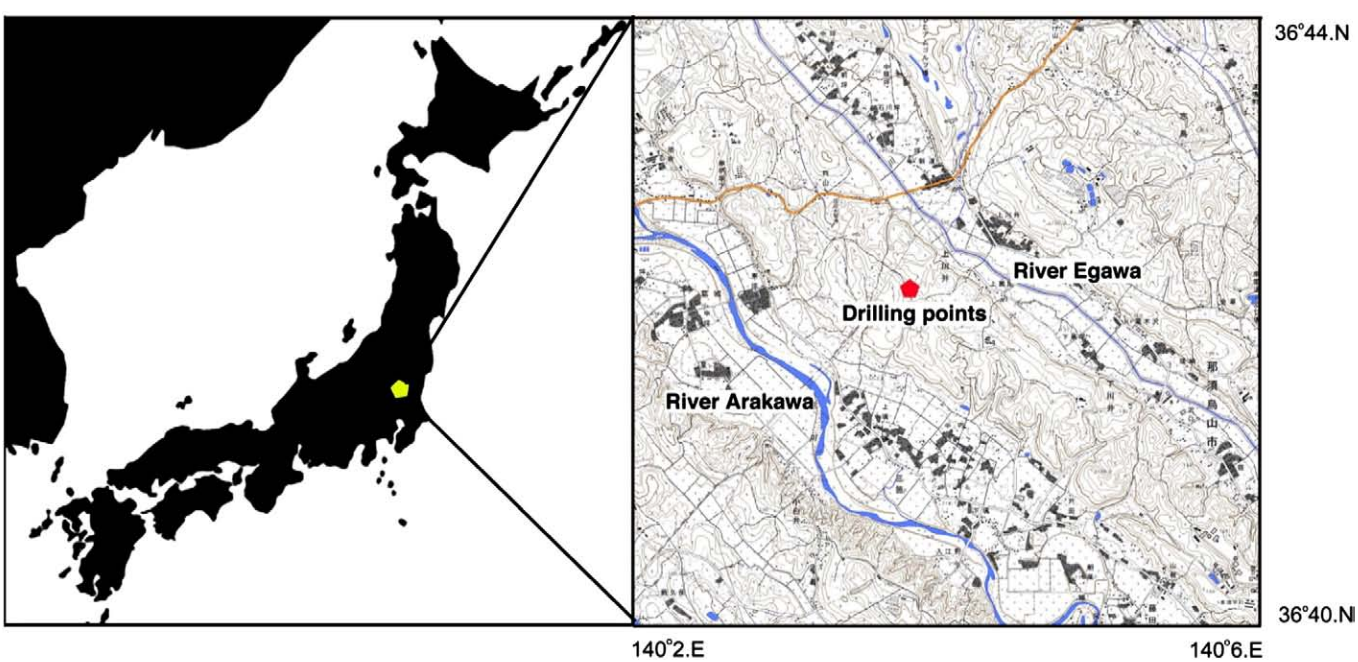

Fig. 1. Location of drilling points (Kr-1 and Kr-2) in Japan (left). The topographic map (right) is a part of a 1:25,000 scale map of Kitsuregawa published by the Geographic Survey Institute.