

Geochemical evidence of fermentative methanogenesis around 2.7 Ga submarine hydrothermal vents

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Archean volcanogenic massive sulfides (VMSs) are often overlain by sedimentary rocks enriched in organic matter. It has been uncertain as to which microorganisms were responsible for such high organic production. To constrain microbial activities around the Archean hydrothermal vents, geochemical studies were performed on carbonaceous rocks associated with 2.7 Ga volcanogenic massive sulfide deposits (Potter, Potterdoal, and Morley Occurrence) in Ontario, Canada. Samples near VMSs contain organic carbon up to 23.0 wt % C (type 1), although samples from distal zone contain less organic carbon ranging from 0.0 to 5.2 wt %C (type 2). Those organic carbon were moderately graphitized, indicated by low H/C ratios and high G peak by Raman spectroscopic analyses of extracted kerogen. Carbon isotope compositions of type 1 kerogen range from -48 to -24 per mil, suggesting high activities of methanogens and methanotrophs. On the other hand, carbon isotope compositions of type 2 kerogen range from -34 to -23 per mil, suggesting production of organic matter by non-hydrothermal microbial community. H₂ concentrations in ore-forming fluids at three sites were estimated by thermodynamic calculations based on sulfide mineralogy. The estimated H₂ level is in the same range of modern submarine hydrothermal fluids at Guaymas Basin and Lost City. Such H₂ level did not allow high production of CH₄ (thus, high organic production) by hydrogenotrophic methanogens or abiotic processes. TEM observation revealed that type-1 kerogen was more graphitized and sulfurized in nano-scale. This indicates that fermentative CH₄ production was promoted by interaction of primary sedimentary organic matter with H₂S-rich hydrothermal fluids. Such fermentative CH₄ were discharged into oceans and then remineralized by methanotrophs around the vents. This new recycling model of carbon species explains observed high concentrations of organic carbon, together with extreme ¹²C-enrichment, around the 2.7 Ga VMSs. Similar carbon isotope “anomaly” is found in hanging wall sediments of Miocene Kuroko deposits in Japan, implying submarine hydrothermal vents have been cradles of fermentative methanogens through the Earth history.