

## Origin of life: Six environmental requirements to bear life Origin of life: Six environmental requirements to bear life

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The origin of life has been an issue historically paramount to natural science, with improved understanding through time of the environmental conditions in which life was born, including step-wise progress to make life in the laboratory. Since the first attempt to synthesize amino acids by Miller (1953), numerous papers have been written and different models of the origin of life have been proposed.

We think there are six environmental requirements for the origin of life: (1) the presence of water, (2) influx of nutrients from primordial continental crustal materials composed of KREEP/anorthosite, (3) N-fixation system, (4) evaporation-condensation conditions and catalysts for the synthesis of amino acids, (5) a layer of water/clay minerals to shield life from UV radiation, and (6) a H<sub>2</sub> supply for metabolic activity.

The prime habitat for the origin of life to fulfill these requirements is a lacustrine environment within a primordial continent in the Hadean.

CH<sub>4</sub>-H<sub>2</sub>-H<sub>2</sub>O 系の水素同位体システムティクスから推定する蛇紋岩熱水メタンの起源—強アルカリ性白馬八方温泉の解析—  
Origin of ultramafic CH<sub>4</sub>: the CH<sub>4</sub>-H<sub>2</sub>-H<sub>2</sub>O hydrogen isotope systematics of the Hakuba Happo hot spring

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Ultramafic-hosted hydrothermal fluids are characterized by high concentration of H<sub>2</sub> derived from serpentinization. Ultramafic rock is rare in the surface of the Earth today, but is likely to have been an abundant component of the early crust due to higher potential mantle temperature. Ultramafic-hosted hydrothermal ecosystem has attracted attention as a potential habitat of the Earth's earliest microbial community because the earliest chemolithoautotrophs may have utilized molecular H<sub>2</sub> as an electron donor. Along with hydrogen, high concentrations of methane and hydrocarbons have been reported in ultramafic hydrothermal fluid. They are usually <sup>13</sup>C-rich compare to microbially-produced methane, and could have been synthesized by abiotic reactions. However, origin of the methane in the ultramafic hydrothermal system is not clearly understood yet. In this study, we collected fluid samples from ultramafic rocks in Hakuba Happo hot spring, and conducted chemical and C-H isotopic analyses of the CH<sub>4</sub> as well as compounds possibly involved in the methane formation reaction in fluids. Samples from Happo hot spring located on the serpentinite body were directly collected from two drilling wells (Happo #1 and Happo #3). The hot spring water is strongly alkaline (pH>10) and rich in H<sub>2</sub> (201~664 umol/L) and CH<sub>4</sub> (124~201 umol/L). These chemistries were typical of fluids associated with ultramafic rock. Even lower temperature regime, H<sub>2</sub> in Hakuba Happo is derived from serpentinization. We measured the concentrations of dissolved gas and the hydrogen and carbon isotope compositions of H<sub>2</sub>, CH<sub>4</sub>, CO<sub>2</sub> and H<sub>2</sub>O. Hydrogen isotope compositions for Happo #1 were dD-H<sub>2</sub>= -700 permil, dD-CH<sub>4</sub>= -210 permil, dD-H<sub>2</sub>O= -84.5 permil, and those for Happo #3 were dD-H<sub>2</sub>= -710 permil, dD-CH<sub>4</sub>= -300 permil, dD-H<sub>2</sub>O= -84.2 permil. Hydrogen and water at Happo#1 had similar dD values to those at Happo#3, but methane from Happo#1 was approximately 80 permil enriched in deuterium relative to Happo #3. On the other hand, carbon isotope compositions of methane from Happo#1 and Happo#3 were d<sup>13</sup>C= -34.5 permil and -33.9 permil, respectively, and there was almost no difference. The CH<sub>4</sub>-H<sub>2</sub>-H<sub>2</sub>O hydrogen isotope systematics suggests that the most likely production process of Happo #1 methane is the olivine hydration with carbon source, and that biological methane contributes to Happo #3 methane.

キーワード: 蛇紋岩熱水系, メタンの起源, 強アルカリ温泉, 水素同位体組成  
Keywords: ultramafic-hosted hydrothermal system, origin of CH<sub>4</sub>, hyperalkaline, hydrogen isotope composition

## ジャックヒルズジルコン中の包有物から探る冥王代地殻

### Hadean crust inferred from mineral inclusions in detrital zircons from the Jack Hills, Western Australia

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The first 500 million years of the Earth history remain poorly understood. Terrestrial rock records during Hadean era (>4.0Ga) are scarcely preserved, probably due to surface and/or subduction erosion and intense meteorite bombardment. However, clues about conditions during this time can be deduced from detrital zircon grains as old as 4.4 Ga preserved in metasedimentary rocks at Jack Hills in the Narryer Gneiss Complex, Western Australia (e.g. Compston & Pidgeon, 1986; Wild et al., 2001). Jack Hills metaconglomerates deposited in ca. 3 Ga contain detrital zircons with ages continuously spanning from 3.0 to 4.4 Ga. Previous investigations of these grains have suggested the existence of a hydrosphere, granitic continental crust, sedimentary cycling and a thermal excursion by the Late Heavy Bombardment on early Earth (e.g. Valley et al., 2002; Harrison, 2009; Abbott et al., 2012). Especially, granitic mineral inclusions in Hadean detrital zircons from Jack Hills provide strong evidence for the existence of granitic crust on early Earth. On the other hand, in-situ U-Pb dating of monazite and xenotime inclusions in 4.25-3.35 Ga detrital zircons from Jack Hills shows ages with 2.68 Ga or 0.8 Ga, suggesting that the most mineral inclusions are not primary, but suffered from metamorphic/metasomatic overprint during late stage metamorphism (Rasmussen et al. 2011). These results call for a reassessment of mineral inclusions in Hadean detrital zircons.

To better understand the nature of earliest crust on the Earth, we focus on apatite mineral inclusions in Hadean detrital zircons. Chemistry of apatite inclusions in zircon (especially Y<sub>2</sub>O<sub>3</sub> and SrO content) reflects the chemical compositions of the whole rocks and can characterize the host magma (Belousova et al., 2002; Jennings et al., 2011). We performed U-Pb age analyses for Jack Hills zircons using LA-ICP-MS, and a total of 103 mineral inclusions was obtained in 315 Jack Hills zircon grains. The type of inclusions in Hadean zircons identified with EDS and Laser-Raman spectroscopy are as follow; quartz, muscovite, biotite, apatite, albite and REE oxide (monazite?). Although low-abundance of apatite inclusions in detrital zircons from Jack Hills compared to those in granitic rocks suggests a secondary replacement on mineral inclusions proposed by Rasmussen et al. (2011), primary apatite inclusions are observed in detrital zircons from Jack Hills. Most apatite inclusions in zircons show prismatic morphology with no visible cracks. SrO content in apatite determined by EPMA ranges from below detection limit (0.03) to 0.1 wt%. Our preliminary data suggest that both mafic and granitic crust contribute for Hadean detrital zircons from Jack Hills as a source rock. Thus, further geochemical analysis (e.g. oxygen isotopes and Ti-thermometer in zircon, and Pb-Pb ages of apatite inclusions) are required and should provide significant constraints for the earliest crust on the Earth.

キーワード: 冥王代地殻, 碎屑性ジルコン, ジャックヒルズ, 包有物

Keywords: Hadean crust, detrital zircon, Jack Hills, Mineral inclusion

## カナダ・ラブラドル・サグレックブロックの地質と年代 Geology and geochronology of the Saglek Block, northern Labrador, Canada

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The Saglek-Hebron area is located in the northeastern part of the Labrador Peninsula, northeast Canada, and belongs to a coastal, central part of the early Archean terrane, called Saglek Block. The block is the west end of the North Atlantic Craton from the Scotland through the southern part of Greenland to the Labrador. The block contains well-preserved Early to Late Archean suites including the Nulliak supracrustal assemblage ( $>3.73$  Ga), *ca.* 3.73 Ga Uivak TTG gneisses, 3.24 Ga Lister gneiss and *ca.* 2.5 Ga granite. In addition, Collerson (1984) and Collerson and Regelous (1995) found  $>3.9$  Ga zircon cores, and suggested pre-3.8 Ga Nanok Fe-rich monzodioritic gneiss. However, the origin of the  $>3.9$  Ga zircon cores is still obscure: inherited or directly crystallized from the host magma (Schiotte et al., 1989; Krogh & Kamo, 2006). The orthogneisses and supracrustals underwent high-grade metamorphism, locally reaching granulite facies at 2.8-2.7 Ga. In the area, the orthogneisses are predominant, and account for about 80 %. It is considered that the Nulliak supracrustal assemblage and Uivak TTG gneisses are equivalent to the Akilia association and Amitsoq gneiss complex in southern West Greenland, respectively. However, the detailed geology within the supracrustal belts, and the relationship between the supracrustal belts and surrounding orthogneiss complex is still unclear. Additionally, detailed geochronological works, including comprehensive dating with LA-ICPMS and cathodoluminescence for igneous and detrital zircons, still lack. Thus, we made detailed geological maps at 9 areas, including relatively low metamorphic grade areas, amphibolite facies condition in order to reveal the Nanok Gneiss, and find the oldest rocks in this area. Especially, we made detailed sketch maps to describe cross-cutting relationships among orthogneisses and determine the oldest suite in each outcrop. In addition, we conducted LA-ICPMS U-Pb dating of zircons from the orthogneisses.

We classified the orthogneisses into eight groups based on the cross-cutting relationships in each outcrop and the distribution of zircon ages:  $\sim 3.90$  Ga, 3.83 Ga, 3.73-3.65 Ga, 3.60 Ga, 3.35 Ga, 2.84 Ga, 2.73 Ga and 2.56 Ga, respectively. The presence of  $>3.9$  Ga zircons provides very important constraint on the formation of felsic continental crust because of the second oldest ages in the world. Collerson (1983) named the pre-3.8 Ga orthogneiss the Nanok Gneiss, but they could not obtain compelling evidence that the  $>3.8$  Ga zircon grains are not inherited/xenocrystic grains. However, the age distribution of oscillatory-zoned zircon grains in the oldest group of the orthogneisses shows presence of older zircons than 3.8 Ga, with the maximum age of 3,956 Ma in  $^{207}\text{Pb}/^{206}\text{Pb}$  age, and apparent lack of 3.7 to 2.7 Ga zircons. In addition, field observation clearly differentiates two orthogneiss suites in an outcrop, and shows the gneiss containing  $>3.90$  Ga zircons is cut by a 3.83 Ga gneiss. The age distribution of the zircons and field occurrence indicates that the old grains,  $>3.9$  Ga, are not inherited or xenocrystic zircons in young (3.7 to 3.8 Ga) orthogneiss but the host orthogneiss were formed at  $>3.9$  Ga. As a result, our geochronological and geological study provides line of evidence of the  $>3.9$  Ga Nanok Gneiss in the Nain Complex.

The Nanok gneiss is the second oldest rock to the Acasta gneiss in the world. The geological relationship that the Nulliak supracrustal rocks are intruded by the orthogneisses implies that the supracrustal rocks also have  $>3.9$  Ga ages, and they are the oldest supracrustal rocks in the world.

キーワード: U-Pb 年代, 初期太古代, ジルコン, 正片麻岩, 表成岩

Keywords: U-Pb dating, Early Archean, zircon, orthogneiss, supracrustals

## カナダラブラドル初期太古代 Nulliak 表成岩中の縞状鉄鉱層の産状と地球化学的研究 Field occurrence and geochemistry of the Eoarchean banded iron formations(BIFs) in the Nulliak Assemblages in Nain Provi

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The earth is the only planet where various life exists ubiquitously, thus it is very important to decode the surface environment in the early Earth in order to understand the origin and early evolution of life. Banded Iron Formation (BIF) is one of the chemical sediments in open sea, and consists of interlayering of white silica layers and black or red iron-rich layers. The BIF occurs from the Early Archean to the Paleoproterozoic, and is a key sediment of deciphering chemical evolution of seawater throughout the Precambrian.

Particularly, only the BIF provides a record of nutrient contents of seawater in the Eoarchean. However, there are found only few pre-3.6Ga supracrustal belts including BIFs, e.g. the 3.71- 3.81Ga Isua supracrustal belt, >3.75Ga Nuvvuagittuq supracrustal belt and the Nulliak supracrustal rocks in the Labrador. Recently, reassessment of comprehensive U-Pb dating of zircons in the Early Archean Uivak gneisses, Labrador suggested the Nulliak supracrustal rocks were formed >3.9 Ga (Shimojo et al., 2012, Mineral. Mag.). This paper presents geological and geochemical data of the BIFs in the >3.9Ga Nulliak supracrustal rocks in the Nain Complex, Northern Labrador, Canada.

The Nulliak supracrustal rocks comprises ultramafic rocks, mafic rocks, BIF, chert, carbonate rocks, conglomerate, and paragneisses. They underwent the amphibolite to granulite facies metamorphism in the Archean. Especially, the metamorphic grade reached the granulite facies in the western side of the Handy Fault, including the Pangertok Inlet, whereas the metamorphic grade of the eastern side never exceeded amphibolite facies, including the Big Island and Nulliak Island. There are two types of BIFs in the area: thin BIF layers associated with mafic rocks, and BIF layers interlayered with carbonate rocks, respectively. The former is a typical Algoma-type BIF, but the latter is associated with shallow-water carbonates, and uncommon in the Archean supracrustal belts. Mineral assemblages of the BIFs are similar each other, and are magnetite + quartz + actinolite + cummingtonite, which are typical of an amphibolite facies assemblage for BIFs (Klein, 2005, Am. Mineral.).

Preliminary chemical analyses show that these BIFs contain >1wt% Al<sub>2</sub>O<sub>3</sub> and relatively high abundances of HFSE (e.g. up to 20ppm Zr). Their high abundances suggest that detritus input was common in the sedimentary environments of the Nulliak supracrustals. On the other hand, their rare earth element (REE) patterns display seawater and hydrothermal fluid-like patterns, namely positive La and Eu anomalies, and superchondritic Y/Ho ratios. In addition, they contain high Cr, Ni, Zn, Sr and Ba (>50ppm) contents. Particularly, the high abundances of Ni and Zn suggest that the >3.9 Ga seawater were enriched in the transition metals due to high hydrothermal activities or alteration of ultramafic magmas, analogous to the other Archean BIFs, <3.8 Ga (e.g. Konhauser et al., 2009, Nature, Mloszewski et al., 2012, EPSL).

キーワード: 初期地球, 初期太古代, 縞状鉄鉱層 (BIFs), 希土類元素

Keywords: the Early Earth, Eoarchean, Banded Iron Formations(BIFs), Rare Earth Elements



## 太古代中期のクリバービル縞状鉄鉱層の側方変化: DXCL2 掘削報告 2 Lateral variation of Mesoarchean Cleaverille Iron Formation: DXCL2 drilling preliminary report 2

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クリバービル縞状鉄鉱層はピルバラクラトンにおいても有名な太古代中期の縞状鉄鉱層であり, 当時の海底や表層環境復元する上で重要な情報をもたらしてくれる。我々は2007年9月(DXCL1)および2011年11月(DXCL2)にこの縞状鉄鉱層および下位の地層についての掘削を行い新鮮な試料を取得した。また, 2012年8月に陸上に露出する縞状鉄鉱層・頁岩層の詳細地質図および柱状図を作成し, 一枚一枚の地層の対比をもとに縞状鉄鉱層の側方変化についての調査を行った。

クリバービル層の層序は, 下位の黒色頁岩および少量の火山砕屑岩層と上位の縞状鉄鉱層からなる。層厚約400mであり, それぞれ, 300m, 100mほどになる。クリバービル層は下位の枕状玄武岩に整合的に重なり, リザードヒル層(66ヒル部層)の石英質砂岩, 黒色・白色チャート, チャート角礫層が不整合で重なる。最上部およびリザードヒル層は褶曲軸に沿っており, それより上位の地層は見られない。デキソン枕状玄武岩の下位には熱水活動を記録するデキソンアイランド層がある。

CL3 コアは黒色頁岩層から縞状鉄鉱層にかけて, 210 mの連続コアを取得した。地層は大きく4つの部分に区分され, 下位から1) 黒色頁岩層(CL1 コアサンプルと類似), 2) 淡緑色?白色層状シデライト層, 3) 縞状マグネタイト・チャート互層, 4) 黒色頁岩・チャート互層からなる。コア観察によると黒色頁岩層には斜交葉理を含む火山岩砂岩を挟み, その上位は淡緑色?白色シデライト層はチャート基質にシデライト結晶がラミナを形成する。部分的に球状シデライトもあり初期堆積時の組織が残る。マグネタイト層は淡緑色?白色シデライト層上位において, 徐々に量比を増して重なり, マグネタイト濃集層になる。マグネタイト濃集層は数ミリの厚さでシデライト層と互層しており, 堆積時に形成されたかどうかは今のところ判断ができない。陸上露頭においては風化が著しくシデライト層は確認できない。

縞状鉄鉱層はクリバービル海岸の東端部に広く分布しており, この地域の500 mにおよぶ海岸線の詳細地質図(1/100)と柱状図作成を行い, 側方変化を調査した。層厚1 mほどの頁岩層も50 m側方では数 cmの厚さになることもあり, 約50?80 mで層厚が変化する地層であることが明らかになった。

中期太古代のクリバービル縞状鉄鉱層の形成には, 1) 大量の有機物層(有機物はシアノバクテリアの沈殿物), 2) 熱水起源のチャート層, 3) シデライト層の形成が伴っている。つまり, 地層は陸源の影響がない遠洋場だが, 熱水活動によって鉄が供給されるが, シデライトとして沈殿し, 側方へ約50 mオーダーで変化が起こる嫌気性な海底で形成したと思われる。

キーワード: 太古代, 縞状鉄鉱層, 層状チャート, 黒色頁岩, 熱水活動, ピルバラ

Keywords: Archean, BIF, bedded chert, black shale, hydrothermal activity, Pilbara

## 南インド、ダルワール超層群における後期太古代のS-MIF化学層序 S-MIF Chemostratigraphy of the Late Archean In the Dharwar Supergroup, South India

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Earths tectonic and climatic systems may have fundamentally changed through the late Archean period, which is characterized by major deposition of banded iron formation (BIF) and appearance of stromatolite reef along continental margins. The earliest known glaciation (~2.9Ga) is recorded in the Mozaan Group of South Africa (Young et al., 1998). Also, in the earliest Proterozoic, Snowball Earth event is recorded in the Huronian Supergroup of Ontario, Canada (~2.4Ga; Young et al., 2001). In accord with the climate change, mass-independent fractionation of sulfur isotopes (S-MIF) demonstrated that Earth atmosphere and ocean was oxygenated at around 2.3 Ga from virtually oxygen-free environment (Farquhar et al., 2000). Before the oxidation event, the S-MIF signature changed dramatically: minimum D33S at around 2.9 Ga, subsequent large D33S variation culminated at 2.5 Ga and its sudden drop at the end of Archean. Moreover, D33S-D36S relation shows characteristic ratio of roughly -0.9 in the Archean period. Change of this D36S/D33S relation may reflect the perturbation of atmospheric chemistry. But there is an active debate about the cause of the large D33S variations and D33S-D36S relation through the Archean period.

We studied late Archean volcano-sedimentary sequence of the Dharwar Supergroup, distributed in the Chitradurga Schist Belt, Western Dharwar craton. Our new field mapping and zircon U-Pb dating allows us to reconstruct detailed lithostratigraphy (Hokada et al., 2012). The lower unit (post-3.0 Ga) consists of basal conglomerate, stromatolitic carbonate, silici-clastics with diamictite, chert/BIF and pillowed basalt in ascending order, all of which are older than 2676 Ma magmatic zircon ages from dacite dyke intruded into the topmost pillowed basalt. The upper unit unconformably overlies the pillow lava, and consists of conglomerate/sandstone with ~2600 Ma detrital zircons, komatiite lava, BIF and silici-clastic sequence with mafic volcanics.

Sulfur isotope analysis of extracted sulfide of these sedimentary rocks show a clear MIF and D33S-D36S correlation. The lower group of the Dharwar Supergroup shows D36S/D33S slope of -1.52, middle group shows -1.20, and upper group shows -0.96. This trend is similar to those reported from Pilbara-Kaapvaal equivalents, thus could be a global signature. Moreover, a marked change of D36S/D33S is observed across a diamictite layer (Talya conglomerate) between the lower and the middle group. If this diamictite was glacial in origin, these changes in sulfur isotopes may indicate the link between some transition in atmospheric chemistry and Earth's surface environmental change.

キーワード: 非質量依存同位体分別, 硫黄同位体, ダルワール超層群, 後期太古代, 氷河期

Keywords: mass independent fractionation, sulfur isotope, Dharwar supergroup, late Archean, glaciation

## 太古代 (30億年前) 微化石群の生活環復元と分類の試み

## Life cycle reconstruction and taxonomy of Archean (3.0Ga) microfossil assemblage

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西オーストラリア・ピルバラ地塊の Farrel Quartzite (30億年) に産する黒色チャートは、その多くが長径 40 ミクロン以上で時には 100 ミクロンに達する微化石を多数含む。これらは、岩石薄片中における見かけの形から、これまで防錐状 (spindle-like) としてきたが、実際はレンズ状でその赤道面に沿ってツバ状の突起 (古生物学用語ではフランジ) を有するので、Flanged Lenticular Microfossils (略称 FLM) と演者は最近勝手に呼んでいる。FLM はその形とサイズ、産状の多様さにおいて、太古代のものとしては極めて特異である。なお FLM は有機質であり、HF-HCl 分解で抽出できること、すなわちいわゆる acritarch として扱えることも付け加えたい。

演者は FLM とその他の形態の微化石を計 2000 個体以上観察してきたが、その結果、以下のような重要な標本と産状を確認した。1) 内部に複数の小球体をもつもの、2) 小球状微化石とレンズ状微化石が混在したコロニー様集合体の存在、3) 球状体を放出しているように見えるもの、4) ダンベル状・鎖状構造。FLM と呼んでいる構造物の生物起源性 (biogenicity) については、これまで様々な手法を用いて検証してきたが、そのいずれの結果もポジティブであり、複数の公表論文に対する反論もない。したがって以上述べた特徴や産状を、生物学的 (すなわち生活環に対応する形態変化や増殖プロセスに対応させて) に解釈することは許されるだろう。あくまで仮説であるが、FLM は少なくとも 2 種類の微生物に対応し、そのうち一方は多分裂により、そしてもう一方は通常の 2 分裂で増殖したと推定される。鎖状構造は後者の連続した増殖の結果形成されたものであろう。そしてこの仮想的な 2 種類のいずれか、あるいは両方とも休眠胞子を形成した可能性は高い。このような解釈は“妄想”にすぎないかもしれない。しかしながら、30 億年前の地層、さらには 34 億年前の地層に FLM が含まれていることは確実であり、その生命初期進化における意義 (それがまだ全く分かっていない) の重要性は揺るぎようもないと考える。

キーワード: 太古代, 微化石, 生活環, 分類学, ピルバラ

Keywords: Archean, microfossils, life cycle, taxonomy, Pilbara



## In-situ iron isotope analyses of pyrites from 3.5 to 3.2 Ga sedimentary rocks of the Barberton Greenstone Belt, Kaapvaal

## In-situ iron isotope analyses of pyrites from 3.5 to 3.2 Ga sedimentary rocks of the Barberton Greenstone Belt, Kaapvaal

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The Barberton Greenstone Belt (BGB), South Africa consists of volcano-sedimentary successions which, were deposited between 3.5 and 3.2 Ga, and is subdivided into three groups: the Onverwacht, Fig Tree, and Moodies groups (Viljoen and Viljoen, 1969). The Barberton Greenstone Belt underwent relatively low-grade tectonothermal events after the deposition, suitable to estimate the surface environmental events and biological evolution in the Middle Archean. Several putative morphological fossils (filamentous and spheroidal type) and trace fossils were reported from the Hooggenoeg and Kromberg formations in the Onverwacht Group (e.g. Engel et al., 1968; Walsh and Lowe, 1985; Schopf, 1992, 1993, 1999; Furnes et al., 2004; Glikson et al., 2008; Javaux et al., 2010).

Isotopic studies of sulfur and carbon of biogenic pyrites and organic carbons suggested activities of methanogen, sulfate-reducing bacteria and photosynthetic bacteria at 3.4 Ga (Ueno et al., 2006; Kakegawa and Ohmoto, 1999; Shen et al., 2001, 2009; Ueno et al., 2008; Philippot et al., 2007). On the other hand, it is well known that dissimilatory iron reduction (DIR) is one of the earliest metabolisms on Earth (Vargas et al., 1998; Lovley, 2004), but the evidence for the microbial DIR is still uncertain in the Archean (i.e. Craddock and Dauphas, 2011; Yamaguchi et al., 2005). We performed in-situ iron isotope analyses of individual pyrites in the sedimentary rocks from the BGB, using femtosecond laser ablation multi-collector ICP-MS technique (fs-LA-MC-ICP-MS) to find isotopic evidence for the microbial activity. We obtained a large variation of iron isotope values from -1.9 to +3.6 permil in  $\delta^{56}\text{Fe}$  values for 139 pyrite grains in 24 samples: 7 cherts from the Hooggenoeg Complex, 10 cherts from the Noisy Complex, 2 cherts from the Kromberg Complex, 1 sandstone from the Fig Tree Group, and 4 sandstones from the Moodies Group, respectively. The  $\delta^{56}\text{Fe}$  values in pyrites from the Hooggenoeg Complex show positive values, whereas those from the Noisy Complex show a wide variation from positive to negative  $\delta^{56}\text{Fe}$  values. One of the main differences between these Complexes is their depositional environments. The Hooggenoeg Complex was considered to be deposited in deep-ocean, whereas that of the Noisy Complex was shallow. The negative  $\delta^{56}\text{Fe}$  value of pyrites with a nadir down to -1.9 permil in the Moodies Group indicates the occurrence of microbial DIR in the middle archean shallow sea.

Keywords: Barberton Greenstone Belt (BGB), iron isotope, microbial dissimilatory iron reduction, pyrite, middle archean

## 原生代海洋化学環境の復元：海洋生物化学循環モデルからの制約

## Conditions required for Proterozoic oceanic chemistry: Constrains from an ocean biogeochemical cycle model

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During the Mesoproterozoic Eon (~1.6-1.0 Ga), oceanic interior below euphotic layer had been kept in pervasive anoxic condition. Such reducing condition has been considered a corollary of a weakly oxidized atmosphere at that time (Holland, 2009, GCA).

Accumulating geochemical data, such as iron speciation, reveal that the pervasive anoxic and ferruginous conditions in the ocean interior have been prevailed during the mid-Proterozoic, and sulfidic waters are restricted around continental margins. However, the atmospheric oxygen level ( $pO_2$ ) in the Proterozoic has not been well constrained, and it remains unclear exactly what biogeochemical conditions are necessary to explain the redox structure in the Proterozoic ocean interior.

Here we constrain the conditions for Proterozoic ocean redox structure by use of a marine biogeochemical cycle model in which C-N-P-O-S-Fe coupled marine biogeochemical cycles are adequately taken into account. The sensitivity experiments with respect to  $pO_2$  demonstrate that pervasive anoxia and euxinia would appear when  $pO_2 < 0.14$  atm and  $< 0.12$  atm, respectively. An expansion of anoxic environments in the ocean interior significantly stimulates the sulfate reduction. As a consequence, the pyrite precipitation into marine sediments is promoted, giving rise to a low sulfate condition ( $SO_4 < 5$  mM) when  $pO_2 < 0.11$  atm. We also found that, under  $pO_2 < \sim 0.02$  atm, a scarcity of sulfate results in the anoxic but non-sulfidic (namely low  $O_2$  and low  $H_2S$ ) condition (i.e., ferruginous conditions). Systematic sensitivity experiments regarding  $pO_2$  and chemical weathering rate on land unequivocally show that the conditions for pervasive euxinia are very limited, implying that widespread ferruginous condition would be a plausible consequence of low  $pO_2$  and high burial efficiency of pyrite during the Proterozoic eon. Sensitivity experiments with respect to other factors affecting long-term oceanic redox state (e.g, sea-level stand, settling rate of particulate organic matters in water column) indicate that the essential biogeochemical consequences are not changed by such factors.

These quantitative results would provide insight into further understanding of the Earth's redox history and its stabilization mechanism(s) from a perspective of the biogeochemical dynamics.

キーワード: 原生代, 生物地球化学, アノキシア, ユーキシニア

Keywords: Proterozoic, biogeochemistry, anoxia, euxinia, ferruginous

## 酸素量で制御された後生動物の初期進化と絶滅 Early metazoan evolution and extinction controlled by oxygen levels

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Organic-molecular dissolved-oxygen index above and below storm wave base, from Cryogenian to Cambrian marine sedimentary rocks in Australia and China records three rises in dissolved oxygen levels. The first rise in dissolved oxygen levels coincides with molecular diversification of animals in the early Ediacaran, the second rise with appearance of large Ediacaran animal fossils, its drop with extinction of Ediacaran metazoa, and the third rise is coeval the explosion of Cambrian metazoa. Our evidence for widespread dissolved-oxygen changes synchronizing with macroevolution and extinction suggests that the global dissolved oxygen level in the sea had controlled the evolution of metazoans during the Neoproterozoic-Phanerozoic transition. The first rise in dissolved oxygen levels in the early Ediacaran is newly found and consistent with molecular diversification of animals in the early Ediacaran.

キーワード: 酸素, 進化, 絶滅

Keywords: oxygen, evolution, extinction

## 数値計算によるエディアカラ紀後期の炭素同位体比異常の原因の解読 Numerical modeling to evaluate carbon cycle changes in the Ediacaran for identifying the cause of the Shuram excursion

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Ediacaran is one of the most important periods, because some environmental changes are proposed (e.g. Oxidation, nutrient and carbon cycle) before the Cambrian explosion and macroscopic multicellular metazoan first appeared and their sizes became drastically large. Therefore, quantitative carbon cycle changes in Ediacaran period need to decode in order to compare of environmental changes and evolution.

We assumed box model that there were two carbon reservoirs in Ocean and fluxes are taken as the first order reaction of each reservoir (Rothman et al., 2003; Ishikawa et al., 2012). Thus, we could estimate both  $d1$  and  $d2$  by changes of parameters to trace analyzed  $d13C_{carb}$  and  $d13C_{org}$  curves from drilling core samples in Three Gorges through the Ediacaran to the early Cambrian (Tahata et al., 2012; Kikumoto et al., 2013; Ishikawa et al., 2012). The  $d13C_{carb}$  in Three Gorges shows negative excursions in Gaskiers glaciation (ca. 580 Ma), Shuram excursion (ca. 570-550 Ma) and Precambrian/Cambrian boundary (ca. 542 Ma). On the other hand, the  $d13C_{org}$  in Three Gorges show constant ca. -30 per mill in early Ediacaran and correlation to  $d13C_{carb}$  after Shuram excursion.

The parameter sets suggested carbon cycle changes in Ediacaran period. This Reconstructed Three Gorges carbon cycle quantitatively estimated carbon cycle changes in these periods. The results indicate the rate of remineralization need to increase before the Shuram excursion and the rate of organic carbon burial increase to ca. 100 times in the late stage of Shuram excursion. The increase of remineralization might indicate step-by-step changes of dominant metabolism from anaerobic respiration to aerobic respiration. In addition, the change of organic carbon burial is possibly consistent with the first appearance of mobile metazoan and zooplankton.

Keywords: Ediacaran, Shuram excursion, carbon cycle change

## 西南日本の上部三畳系深海底堆積岩から発見されたイジェクタ層の地球化学的特徴 Geochemical characterization of impact ejecta layers from the Upper Triassic deep-sea deposits in Southwest Japan

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Anomalously high platinum group element (PGE) concentrations have been reported for Upper Triassic (middle Norian) deep-sea sediments in the Sakahogi section, central Japan, which have been interpreted to be derived from an extraterrestrial impact event that formed the 100 km Manicouagan crater in Canada. The Late Triassic PGE anomalies have been identified in deep-sea claystone layers at three new bedded chert sections in Southwest Japan: (i) Unuma section in the Inuyama area, Mino Terrane, (ii) Hisuikyō section in the Kamiaso area, Mino Terrane, and (iii) Enoura section in the Tsukumi area, Chichibu Terrane. At each of these sites, the Late Triassic claystone layers are characterized by high PGEs abundances, coincident with minor enrichments of Ni and Cr, and abundant Ni-rich magnetite grains and microspherules. These claystone samples have high PGE concentrations of up to 7.0-38.1 ppb Ir, 13.2-65.1 ppb Ru, and 18.0-27.5 ppb Pt, which are comparable to that observed at the Sakahogi section. Given that PGEs are highly depleted in continental crust of the Earth relative to solar abundances, these anomalously high PGE abundances may have resulted from a large extraterrestrial impactor, similar to the Chicxulub impact event at the Cretaceous/Paleogene boundary. Redistribution of PGEs under reducing conditions can also result in PGE enrichments in marine sediments, but the Sakahogi claystone samples have unique geochemical signatures such as anomalously high Os contents (11.4 ppb) with unradiogenic Os. These lines of geochemical evidence are consistent with a significant extraterrestrial input to the claystone, which can account for the anomalously high PGEs concentrations.