

Geochemical study of Mn and P in modern and Archean hydrothermal systems: its importance to activities of primitive microorganisms

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## 論 文 内 容 要 旨

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	Geochemical study of Mn and P in modern and Archean hydrothermal systems: its		
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	活動の重要性について)		

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## Abstract

Biological activities have greatly altered compositions of atmosphere, hydrosphere and lithosphere on the Earth since the rise of first life around 3.8 Ga. Early biological activities have been studied intensively using bio-essential elements, such as carbon, nitrogen, oxygen and sulfur which are majorly hosted in atmosphere or hydrosphere. However, biological activities are also essentially sustained by bio-essential elements whose major reservoir is lithosphere, such as metals in enzymes and P (phosphorus). For example, Mn (manganese) and P are essential for cyanobacteria's activities, because "Mn cluster (Mn<sub>4</sub>CaO<sub>5</sub>)" is essentially used to split water into oxygen during photosystem II and P is composed of RNA and DNA. Consequently, their supply process from lithosphere, hydrosphere to biosphere have been likely to control biological activities through time. Yet, their transportations from lithosphere, hydrosphere to biosphere have not been certain through time.

In modern ocean, continental weathering of P is known to be major process to transport from lithosphere to hydrosphere. In addition, the following process from hydrosphere to biosphere have been well understood. On the other hands, large uncertainties exist for Mn leaching from lithosphere, and its mobility in hydrosphere and then to biosphere.

The Archean surface environments were considered as different worlds compared to modern Earth. The generally reducing surface environments might have caused different geochemical cycle of Mn. Many researchers consider that deficiencies of continental crusts also had limited P availabilities in the Archean oceans, and thus, limited biological productivities. On the other hands, P and Mn leaching from lithosphere to hydrosphere through submarine hydrothermal activities are not studied.

In chapter 1, geochemical and mineralogical studies were conducted on Mn ores, which were formed by 12 Ma submarine hydrothermal activities in felsic volcanisms in the Hokuroku district, Akita prefecture, Japan. The mobilization of Mn from submarine volcanic rocks during rock/water interaction and evolution of Mn species in hydrothermal fluids were understood. Non-biological and biological processes to precipitate Mn oxide and Mn carbonates were also evaluated. These results clarified Mn leaching from lithosphere, and its mobility from hydrosphere to biosphere in modern environments. In chapter 2, metagenomic, geochemical and mineralogical studies were conducted for chemical precipitates or microbial mats at modern Mn-rich samples at Hachikuro or Mizusawa hot springs in Akita prefecture, Japan. Evidence of oxidation of soluble Mn<sup>2+</sup> by Mn-oxidizing bacteria was obtained. Further, enzyme and Mn-oxidizing genes associated with Mn oxidation were specified. A series of metagenomic analyses robustly proved biologically mediated process oxidized Mn. These biological activities were stimulated by elemental supplies such as copper and Mn, which are essential for

In chapter 3, Mn and P mobilizations from early Archean basaltic ocean floor were examined. Investigations on Archean basaltic ocean floor revealed that it could be an important P sources through submarine hydrothermal activities, and P flux from the basalt into ocean sustained contemporary biosphere. Mn was also confirmed to be significantly leached from the basalt.

biological Mn oxidation, from lithosphere through water/rock interactions by hot spring water.

Combining these results revealed that supply process of Mn and P from lithosphere, hydrosphere to biosphere in modern and Archean environments. In particular, leaching elements from lithosphere has been likely to stimulate biological activities. Consequently, their elemental fluxes have played significant roles of biological evolution through time.

## 論文審査の結果の要旨

塚本雄也は生物進化史におけるリンとマンガンの重要性に着目し研究を行なった。マンガンやリンは、もともと岩石圏に存在し、それが岩石・水反応で岩石から水圏に移動し、最終的に生物圏に取り込まれる。これら移動過程や、生物への取り込み過程、進化しの中での元素サイクルの様子は不明であった。塚本くんはこれらの問題にアプローチするために、以下の3つの課題を設定し、博士論文課題研究を行なった。

- (1)秋田県北鹿地域中新世マンガン鉱床形成時の岩石・水・微生物反応に関する研究。
- (2)遺伝子解析による秋田県八九郎温泉におけるマンガン酸化菌とマンガン酸化酵素の検出。
- (3) 太古代海底玄武岩からのリンとマンガンの溶脱と初期生命活動に対する制約。
- (1)の課題において、秋田県北鹿地域の詳細な地質調査を行い、マンガン鉱床形成地質現場を3次元的に捉えることに成功した。さらにマンガン鉱石中に有機物―轟石―石英―方解石がサブミクロン単位で互層をなす構造を見出した。詳細な TEM, STEM,ラマン分光分析などで、これらは1200万年前の海底熱水活動で噴出したマンガンが、マンガン酸化菌によって鉱物化した痕跡であることを新たに見出された。現生海底のマンガンノジュール形成に微生物活動が関与していたかしばしば議論されるが、本研究によって微生物活動の重要性や証拠が新たに提示されたことになる。
- (2) の課題においては、マンガン酸化物層からマンガン酸化菌特有の遺伝子を取り出すことに世界で初めて成功した。マンガン酸化物が微生物活動によって形成されたことを実証した初めての例である。さらにマンガン酸化酵素であるマルチカッパーオキシターゼを同一試料から検出した。天然におけるマンガン酸化現場での検出は世界で初めてである。
- (3) の課題において、太古代海底玄武岩を海水や熱水との反応の違いによって分類し、詳細な鉱物学的研究を行なった。それによって主に初生リン酸塩鉱物の溶解と再沈殿、完全溶脱の様子が解明された。このことは、大陸がなくても海底からのリンの供給があり、それが初期生物生態系を支えていた可能性を示す画期的な結果を得ることができた。

自立して研究活動を行うに必要な高度の研究能力と学識を有することを示している。 したがって、塚本雄也提出の博士論文は、博士(理学)の学位論文として合格と認める。