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授与した学位	博士
専攻分野の名称	理学
学位授与番号	博甲第 7059 号
学位授与の日付	2024年 3月 25日
学位授与の要件	自然科学研究科 地球惑星物質科学専攻
	(学位規則第4条第1項該当)
学位論文の題目	Exploring the potential role of transitional elements to understand the past and the future of the Earth's biosphere (地球生物圏の過去と未来を理解するための、遷移元素の潜在的役割に関する研究)
論文審査委員	教授 小林 桂 教授 田中亮吏 教授 牧嶋昭夫 准教授 国広卓也教授 KathleenA.Campbell
学位論文内容の要旨	

In the first part of the thesis (from chapter 1 to 3) the experimental procedures for the development of sequential ion exchange chromatography techniques for isolating transitional metals from geological matrices are presented. These element isolation techniques were evaluated by applying them to the geological reference materials and the evaluation results are also presented. In addition to the sequential element separation techniques individual methods were developed to efficiently isolate Ni and Cr from geological matrices. These individual separation methods are presented in separate chapters (chapter 2 and 3).

In the second part of the thesis, the sample preparation techniques for obtaining microbial metal isotope fractionation were evaluated and the results are presented (chapter 4). For the evaluation cyanobacterial cultures grown in the laboratory were used and they were treated with reagents to remove the adsorbed metal fraction. Then the effectiveness of the treatment was evaluated by measuring and comparing the elemental abundances and isotopic variations on treated and the untreated samples.

Third and final part of the thesis is dedicated to understand the role of transitional elements in shaping the Earth's biosphere. This has been carried out by following an experimental approach to understand the evolution of biogeochemical N cycle and atmospheric oxygenation since these two processes are thought to be controlled by the availability of particular transitional metals. A past literature review is presented to set up the background for the particular problem in chapter 5. Then the experimental results for the determination of the ability of cyanobacteria to use urea as a nitrogen source are presented (chapter 6). Based on the experimental results a hypothesis was build up to explain the coevolution of biological N cycle and the atmospheric oxygenation of the Earth.

## 論文審査結果の要旨

The evolutionary trajectory of life on Earth presents a complex enigma, driven by a multitude of interconnected biological and abiological processes. Despite rigorous research efforts, a definitive explanation remains elusive.

Mr. Ratnayake conducted a comprehensive investigation into the intricate interplay between transition metals and nitrogen-containing molecules within cyanobacteria, organisms that played a pivotal role in the oxygenation of the early Earth. The research findings are presented in four distinct chapters.

This doctoral dissertation unfolds in four distinct parts. The first section investigates the evolutionary trajectory of biological nitrogen fixation and its relationship to Earth's oxygenation, focusing on the unresolved interplay between transition metals and bacterial nitrogen sources. The second section details advancements in analytical methods for transition metals, specifically those of nickel and chromium isotope analysis. The third section delves into experimental studies of both biological and non-biological processes that fractionate transition metal isotopes in cyanobacteria cultures, highlighting the current challenges in differentiating these processes using such isotopes in natural environments. Finally, the fourth section presents an experimentally determined model for the Great Oxidation Event, based on the urea and nickel requirements of cyanobacteria in a simulated Paleozoic seawater medium.

This newly proposed model necessitates further validation through numerical simulations and geological evidence. Nevertheless, the original methodology of this dissertation and the novel discussion of the role of nitrogen-containing molecules and transition metals in the evolution of the Earth's biosphere are sufficient for a doctoral dissertation in terms of its academic quality.