学位論文の要旨

Abstract of Thesis

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学位論文題目 Title of Thesis（学位論文題目が英語の場合は和訳を付記）

Spatial and temporal evaluations of fulvic acid iron in Takahashi and Asahi Rivers in Okayama Prefecture, Japan
（岡山県高梁川と旭川におけるフルボ酸鉄の時空間評価）

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Iron (Fe) is an essential micronutrient in aquatic environments, playing a crucial role in supporting primary productivity and maintaining ecosystem balance. It is essential for phytoplankton growth, photosynthesis, and nitrogen fixation, which form the foundation of aquatic food webs. In many regions, Fe availability limits primary production, influencing nutrient cycling, species composition, and energy transfer through the food web. Additionally, Fe participates in biogeochemical processes such as redox reactions, organic matter degradation, and mineral cycling, contributing to water quality and ecosystem stability. Riverine input is a key source of Fe in coastal environments, and humic substances like fulvic acids play a crucial role in binding terrigenous Fe to enhance its mobility. This interaction forms fulvic acid iron (FAFe) complexes, enhancing Fe solubility and facilitating its transportation from the land to fluvial systems. Because of the critical role Fe plays in aquatic environments, numerous studies on its global distribution and dynamics have been conducted. Although some regions reported high Fe concentrations in riverine systems, studies on Japanese rivers documented much lower levels. Besides, despite the extensive research on riverine dFe, investigations specifically addressing FAFe remain limited, likely because of the time-intensive and complex nature of FA extraction processes. Furthermore, in surface waters, Fe has a limited residence time because the thermodynamic stability of Fe (III) is highly sensitive to changes in environmental conditions. For example, Fe (III) rapidly undergoes hydrolysis, scavenging, and precipitation in response to pH variations, resulting in its removal or transformation. Besides, seasonal anthropogenic activities and natural factors can significantly influence variations in riverine FAFe concentrations.

Moreover, in Japan, extensive urbanization and industrial activities have transformed watersheds, potentially threatening downstream ecosystems, including the Seto Inland Sea (SIS), a historically productive fishing and aquaculture zone. These human activities substantially affect the riverine variation and mobility of Fe to coastal environments. For instance, the creation of reservoirs and dams traps micronutrients including Fe, thereby altering its bioavailability through biogeochemical processes such as oxidation and precipitation. This human intervention disrupts the riverine Fe supply, potentially hindering primary productivity and ecological balance downstream.

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In addition, previous studies that analyzed riverine dissolved iron concentrations often employed low-frequency sampling over short durations, which may be insufficient to capture the stable variations in dissolved iron concentrations in rivers. Against this backdrop, this study applied a high-resolution sampling approach by employing systematic, long-term, and spatially diverse field work water sampling measures to analyze the spatial variations and temporal behavior of FAFe in Takahashi and Asahi Rivers, which drain into the SIS. Such approaches are crucial for identifying Fe hotspots, understanding its seasonal dynamics, and may help in developing strategies for ecosystem conservation and the sustainable management of water resources. Therefore, the main objectives of this thesis were to:
Evaluate the temporal behavior of FAFe in Asahi River located in Okayama Prefecture, and
assess the spatial variations in FAFe in Takahashi River, located in Okayama Prefecture.

To achieve the above objectives, weekly water sampling was conducted at the Asahi River from January 2022 to December 2023, and monthly water sampling was carried out at the Takahashi River from January 2022 to March 2024. During these activities, water samples were collected, and the physicochemical parameters of the rivers were measured on-site. Laboratory experiments were conducted to determine the concentration of FAFe. Temporal and spatial analyses were performed to identify seasonal trends and pinpoint hotspots along the rivers. Additionally, multivariate regression models were employed to evaluate the impact of multiple watershed variables on FAFe variability.

The results showed significant trends with higher FAFe concentrations observed in spring ($p<0.001$) and summer ($p<0.05$), suggesting seasonal anthropogenic inputs from terrestrial sources coinciding with peak hydrological events in the two watersheds. In Takahashi River, spatial analysis revealed a higher distribution of FAFe concentrations in the tributaries than in the mainstream, which was likely influenced by land use dynamics within the watershed. In Asahi River, upland fields significantly influenced FAFe concentrations ($p<0.01$) through runoff with abundant supply of NO_3^- and PO_4^{3-}. Meanwhile in Takahashi River, the regression models revealed forestland ($p<0.01$), upland fields ($p<0.001$), paddy fields, and urban commercial LULC ($p<0.01$) as the predominant sources of FAFe; conversely, transport land and other land uses ($p<0.05$) were negatively correlated with FAFe concentration. Moreover, principal component analysis revealed a close relationship between FAFe variability and soil types such as Helvic Acrisols, Haplic Andosols, Humic Cambisols, Gleyic Andosols, and Rhodic Acrisols. Additionally, sedimentary rocks, including sandstone, mudstone, and green tuffs, exhibited a significant relationship with FAFe (r = 0.8). Understanding the complex relationships between FAFe concentrations and environmental factors within the catchment is essential for evaluating the intricate environmental impacts of iron's biogeochemical cycling within aquatic ecosystems. In conclusion, spatial monthly and weekly measurements of FAFe concentration in this study identified hotspots along the river and significant trends showing distinct seasonal variations that short-term and low frequency sampling might not have captured. These spatial and temporal variations in the two rivers resulted from a complex interplay of multifaceted factors, including land use activities, hydrological regimes, and geomorphological characteristics of the watersheds.