

Speciation analysis of Gd complexes in river water using HPLC-ICP-MS

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Magnetic resonance imaging (MRI) is now one of the requisite imaging methods for medical diagnostics. Gadolinium (Gd) is used as the contrast agent for MRI to enhance the contrast of an image because of its strong paramagnetic property. Gadolinium-based contrast agents are administered to patients in chelated form, because Gd³⁺ is well known for its toxicity. In recent decades, several studies reported that the apparent positive anomalies of Gd in the REE pattern are shown in rivers running through metropolitan areas [1-3]. It shows that a part of the Gd-based contrast agents is emitted to river water without being processed during the sewage treatments, even though the long-term risks of Gd-based contrast agents for living organisms are not well elucidated so far.

Recently, our group has developed a HPLC-ICP-MS technique using the hydrophilic interaction chromatography (HILIC) column with the water-based eluent for the separation of Gd-based contrast agents [4]. In this study, the presented technique was applied to the speciation analysis to identify Gd-based contrast agents in river water in Japan.

River water samples were collected from the Muko River (Hyogo, Japan) in June 2018 and November 2020 near Sanda City, which is a suburban city with a population of 200,000. The concentrations of Gd in the samples collected in 2018 and 2020 (2018-O and 2020-O) were 97 ng L⁻¹ and 51 ng L⁻¹, and the magnitudes of the anomalies were calculated 39 and 17, respectively.

Gd-species in the river water samples were also investigated using the HILIC-ICP-MS technique with the water-based eluent. As a result, 3 kinds of the macrocyclic type of Gd-based contrast agents were found in 2018-O and 2020-O. They were recommended to be used in Japan. The concentration of these Gd-species makes up 70% of the anthropogenic Gd. The results clearly indicate that a part of Gd-based contrast agents pass through the WWTP without being treated.

[1] Bau and Dulski, *Earth Planet. Sci. Lett.*, **143** (1996). [2] Nozaki et al., *Geochem. Cosmochim. Acta*, **64** (2000). [3] Zhu et al., *Bull. Chem. Soc. Jpn.*, **77** (2004). [4] Okabayashi et al., *Talanta*, **222** (2021).