

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

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Magnetic resonance imaging (MRI) is a requisite imaging method for medical diagnostics, and thus, the number of MRI examination has been rapidly increasing. To enhance the signal of obtained images, Gd chelates are often used as the contrast agent. In these decades, several studies reported that rivers running through populated and industrial area show apparent positive anomalies of Gd in the REE pattern [1-3]. These studies suggested that the increasing of Gd content in the river water is related to the Gd-based contrast agents for MRI. So far, it is not well known about the long-term risks of Gd-based contrast agents for living organisms. Thus, monitoring of Gd-based contrast agents in river water is required. Recently, our group has developed a high-performance liquid chromatography (HPLC)-ICP-MS technique for the separation of Gd-based contrast agents [4]. In this study, the presented technique was applied for river water to identify and quantify Gd-based contrast agents present in the river water.

River water samples were collected from the Muko River (Hyogo, Japan) in 2018 and 2020. These samples were collected from close by the outlet of the wastewater treatment plant (WWTP) (sample 2018-O, 2020-O), 2 km downstream from the WWTP (2018-D, 2020-D), and 0.5 km upstream from the WWTP (2020-U). The large positive anomalies of Gd in REE pattern were observed from 2018-O and 2020-O, and they were significantly decreased in 2018-D and 2020-D. On the other hand, such Gd anomalies were not observed in 2020-U, indicating that anthropogenic Gd flow into the river from the WWTP.

Gd-species in the river water samples were investigated using the HPLC-ICP-MS technique. As a result, 3 kind of Gd-based contrast agents were found from 2018-O and 2020-O. The concentration of these Gd-species makes up 70% of the anthropogenic Gd. These results clearly indicate that at least a part of Gd-based contrast agents pass through the treatment in WWTP.

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