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Analytical Technique of Long-Lived Radioiodine Released from Fukushima Daiichi NPP for Monitoring Marine Pollution

Shiho Asai [1], Aya Sakaguchi [2], Yoshiaki Kon [3], Takuma Horita [4],
Makoto Matsueda [5], and Chikako Chong [1]

[1] National Metrology Institute of Japan (NMIJ), National Institute of Advanced Science and Technology (AIST), [2] Faculty of Pure and Applied Sciences, University of Tsukuba
[3] Geological Survey of Japan, National Institute of Advanced Science and Technology (AIST)
[4] Sector of Fukushima Research and Development, Japan Atomic Energy Agency, [5] Collaborative Laboratories for Advanced Decommissioning Science (CLADS), Japan Atomic Energy Agency

Summary

Concentrations of radionuclides in seawater around Fukushima Daiichi Nuclear Power Plant which generated more than 1.2 Mt of radioactively contaminated water has been monitored since the nuclear accident. Among the radionuclides found in the contaminated water, ^{129}I, radioiodine, can be one of the radionuclides to be monitored due to its long half-life of 1.57×10^7 years. Despite the importance of the measurement of ^{129}I, conventional analytical methods based on ICP-MS imposes cumbersome pretreatment, leading to insufficient number of measured data for the assessment of the contamination situation. In this study, we proposed a simple and rapid analytical technique using laser ablation ICP-MS which allows direct measurement of ^{129}I incorporated in AgCl by adding $AgNO_3$ to about 10 mL of seawater sample.

To efficiently recover I from a seawater sample, we focused on the solubility product constants of AgI. As it is about 2 million times smaller than that of AgCl, AgI can preferentially coprecipitate with AgCl by adding $AgNO_3$ to seawater sample. This enables efficient concentration of ^{129}I which can be incorporated in AgI (^{127}I, natural abundance: 100%) and AgCl. The amount of $AgNO_3$ added was optimized based on the prediction of the precipitation percentage of AgI calculated from the precipitation equilibrium among Cl^-, Br^-, I^- and Ag^+. Here, the concentration of ^{129}I in seawater sample was assumed to be 0.09 Bq/L (1.4×10^{-5} mg/L), which is designated as a target level for ^{129}I measurement of the conventional method. The result showed that the concentration of ^{129}I in the precipitate formed by adding 0.1 mg of $AgNO_3$ was around 0.5 mg/kg, which is sufficient concentration for the measurement of LA-ICP-MS.

For the measurement of low level of ^{129}I by LA-ICP-MS, removal of the interference caused by isobar ^{129}Xe that can be found in Ar gas used for the plasma source of ICP is required. We thus employed triple quadrupole ICP-MS (ICP-MS/MS) to eliminate such interference inside the instrument by colliding interference sources with gas molecules. With ICP-MS/MS, the integrated counts at m/z 127 (^{127}I) in the simulated AgI precipitate was around 108-109 counts, whereas those at m/z 129 were negligibly low. This shows that ^{129}I in samples with less than 10^{-8} of $^{129}I/^{127}I$ (atomic ratio) can be detected by the proposed technique using LA-ICP-MS/MS.