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Determination of ^{129}I in environmental samples by AMS and NAA using an anion exchange resin disk

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Abstract

We have developed a new extraction method for the measurement of ^{129}I by accelerator mass spectrometry (AMS) utilizing an anion exchange resin disk. In comparison to traditional methods such as solvent extraction and ion exchange, this method provides for simple and quick sample handling. This extraction method was tested on soil, seaweed and milk samples, but because of disk clogging, the milk samples and some of the seaweed could not be applied successfully. Using this new extraction method to prepare samples for AMS analysis produced isotope ratios of iodine in good agreement with neutron activation analysis (NAA). The disk extraction method which take half an hour is faster than previous techniques, such as solvent extraction or ion exchange which take a few hours. The combination of the disk method and the AMS measurement is a powerful tool for the determination of ^{129}I. Furthermore, these data will be available for the environmental monitoring before and during the operation of a new nuclear fuel reprocessing plant in Japan.

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Keywords: Anion exchange resin disk; ^{129}I; Accelerator mass spectrometry; Neutron activation analysis; Soil; Seaweed; Milk

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1. Introduction

The long-lived iodine isotope, ^{129}I ($T_{1/2} = 1.57 \times 10^7$ yr), is naturally produced by cosmic ray-induced spallation of xenon in the atmosphere and by spontaneous fission of uranium in the geosphere. Anthropogenic ^{129}I has been released from nuclear weapon tests, nuclear accidents such as the Chernobyl accident and spent nuclear fuel reprocessing plants, which is main global source of ^{129}I at present date [1]. A new nuclear fuel reprocessing plant will be operated in the northern part of Japan, and some amount of ^{129}I is expected that ^{129}I to be released into the environment from this plant. Evaluating the level of ^{129}I in environmental samples in Japan before its operation would contribute to studies on iodine circulation, radiological exposure and utilization as a tracer.

Accelerator mass spectrometry (AMS) and neutron activation analysis (NAA) are available for the measurement of ^{129}I, both of which require chemical extraction of iodine from environmental samples before the measurement. With AMS, it is possible to measure many samples at the same run, it is desirable necessitating a simple and quick preparation method compatible with AMS measurement. The preparation technique using the anion exchange resin disk for the extraction of Pu is a well known as simple and quick method [2]. In this study, we have developed a simple and rapid chemical separation method for the measurement of ^{129}I in environmental samples using an anion exchange resin disk.

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2. Experimental

In order to investigate the applicability of AMS measurement by using an anion exchange resin disk, environmental samples were measured by both AMS and NAA. Because the iodine isotopic ratio was high around the vicinity of Tokai nuclear fuel reprocessing pilot plant due to its emission of ^{129}I, environmental samples were collected in Hokkaido, Iwate, Akita, Hyogo and Oita which are far from Tokai pilot plant and not downwind. The sampling locations are shown in Fig. 1.

2.1. AMS measurement

2.1.1. Anion exchange resin disk

A 3M Empore^{TM} anion exchange-SR resin disk (diameter: 47 mmϕ, thickness: 0.5 mm, composition: 90 wt.% resin particle and 10 wt.% fiberillated PTFE, resin: styenedivinylbenzene copolymer, functional group: strongly basic quaternary ammonium, surface area of resin: 350 m^2/g, average particle size; 12 μm) was used in the measurement. The disk was successively washed before the extraction to remove any contamination with 10 mL of methanol, 15 mL of H_2O, 15 ml of 1M NaOH to convert the resin to OH^- form and finally 15 mL of H_2O (three times).

2.1.2. Sample preparation

2.1.2.1. Soil. Soil samples were sliced into two sections, a surface-layer from 0 to 5 cm depth and a subsurface-layer from 5 to 20 cm depth. Dried soil samples (30 g) were placed in a quartz tube. A pre-combustion at 150 °C for 10 min was done before the high temperature combustion to avoid a too violent reaction. A stream of oxygen and nitrogen gas was passed through the tube and heated up to 750 °C in an electric tube oven. During the heating, iodine evaporated from the soil and was then adsorbed on charcoal. Iodine was eluted from this charcoal by NaOH alkali solution (6.0 mol/L) and we measured iodine concentration of its solution using the inductively coupled plasma mass spectrometry (ICP-MS) for the subtraction of iodine carrier ratio. Iodine carrier (4 mg) was added and its solution passed through the 3M Empore^{TM} anion exchange resin disk for the adsorption of iodide (I^-) at a flow rate of 220 mL/min. After the disk was washed with 20 mL of H_2O, iodide was eluted by the 15 mL of 1N HNO_3. The chemical yield of this procedure was more than 80%. The flow chart of this procedure is shown in Fig. 2.

2.1.2.2. Seaweed. Wakame (*Undaria pinnatifida*) samples were collected from Hokkaido and Iwate and konbu (*Laminaria*) samples were collected from Akita, Hyogo and Oita. Because seaweed contains enough iodine for the determination by AMS, the sample preparation of seaweed was carried out without the addition of iodine carrier. A combustion method, similar to that used for soil samples, was used for seaweed samples. Since not enough iodine was collected through combustion, iodine was also extracted from a residual material. The residual material

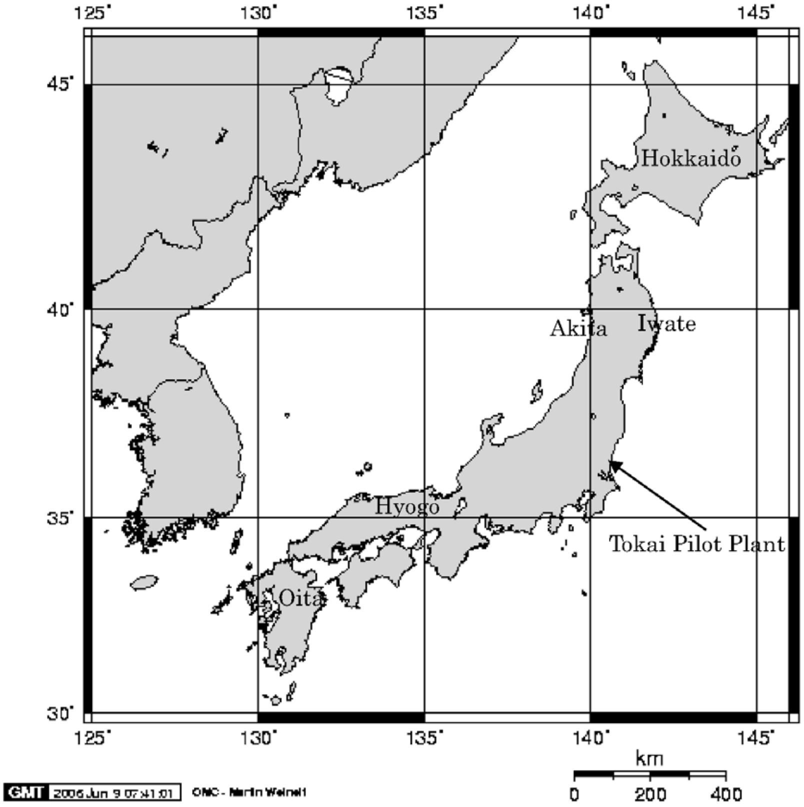

Fig. 1. Location of the collection sites.