1.2.2 水産生物におけるヨウ素の形態別濃縮係数

Concentration Coefficients of Radioiodine in Different Chemical Forms from Seawater to Fishery Products

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Abstract

Radioiodine takes various chemical forms in the environment. Ion forms of both I^- and IO_3^- were found in the ocean for ^{129}I discharged from the first Japanese commercial nuclear fuel reprocessing plant located in Rokkasho. Since the concentration factor of iodine from seawater to marine products strongly depends on the chemical form of iodine, it is necessary for realistic assessment of radiation dose from the discharged radioiodine via marine products to use the concentration factor of each chemical form of iodine. This study aims to establish the concentration factor of radioiodine in I^- and IO_3^- for marine products (seaweed, shellfish and benthos). In FY 2015, 1) the concentration factors of I^- and IO_3^- for sea cucumber (*Apostichopus japonicus*) were measured using an iodine radiotracer and 2) the chemical form of stable iodine in sea cucumber was analyzed by using X-ray absorption fine structure (XAFS) analysis.

We exposed sea cucumber samples obtained from local fishermen to $^{125}I^-$ or $^{125}IO_3^-$ in seawater using air-tight chambers, followed by collecting the samples at pre-determined time until 72 h after the start of exposure. No feed was administered during the exposure. Three samples were collected at each time, followed by drying in vacuo after removing seawater in their body cavity. The dried samples were measured for ^{125}I concentration by using a NaI auto-counter. The seawater in the chamber was also collected at the time of sea cucumber sampling, and $^{125}I^-$ and $^{125}IO_3^-$ in it were separately measured by their radiochemical separation using an anion exchange column. The exposure experiment was repeated twice excluding exposure time of 48 h in the second experiment, and each results are separately presented in Fig. 1 and 2. As shown in the figures, the intake rate of sea cucumbers in the first experiment was obviously higher than that in the second one. The $^{125}I^-$ concentration in seawater in the first experiment also rapidly decreased in comparison to that in the second one. The $^{125}IO_3^-$ concentration in seawater did not significantly decreased in both experiments.

A single compartment metabolic model of iodine in sea cucumber with separate inputs of $^{125}I^-$ and $^{125}IO_3^-$ was constructed (Fig. 3), and transfer-rate coefficients in it were estimated by fitting the experimental data (Table 1). The transfer-rate coefficients of input for $^{125}I^-$ (k_1) and $^{125}IO_3^-$ (k_2) and of output (k_3) in the first experiment were estimated as 6.E1, 6. and 1.E-1, respectively. The values of k_1, k_2 and k_3 in the second experiment were obtained as 5., 1. and 3E-2, respectively, showing that metabolism of ^{125}I was less active in the second experiment than in the first one by unknown reasons. Concentration factors of $^{125}I^-$ and $^{125}IO_3^-$ in the first experiment was 6.E2 and 6E1, respectively, while they were 2.E2 and 5.E1, respectively, in the second one. Both results showed that the concentration factor of $^{125}I^-$ for sea cucumber was larger than that of $^{125}IO_3^-$. It should be noted that ingestion intake of iodine into sea cucumber was not considered here.

The chemical forms of stable iodine in sea cucumber tissue samples was analyzed by XAFS. Although no clear spectrum was obtained because of low iodine concentration in the samples, it suggested that most of the iodine in sea cucumber tissue was in organic form.

1. 目的

大型再処理施設の海洋放出管から排出される放射性ヨウ素は、海洋中で I^-及び IO_3^-の化学形態を取るため、水産物を介した現実的な被ばく線量を評価する際には、化学形態別の濃縮係数を用いる必要がある。そこで本調査では、海水から六ヶ所村周辺の海産物（緑藻、褐藻、貝類及び底生生物等）への放射性ヨウ素の濃縮係数を化学形態別に（I^-又は IO_3^-）求める。これまでに緑藻及び褐藻の形態別濃縮係数を求めており、平成 27 年度は、マナマコ(*Apostichopus japonicus*) を対象に、化学形態の異なる放射性ヨウ素($^{125}I^-$ 又は $^{125}IO_3^-$)を添加した海水を用いて濃縮係数を求めた。また、大型放射光施設 SPring-8 を用いてマナマコ中のヨウ素の化学形態を検討した。

2. 方法

平内町漁業協同組合が漁獲したマナマコを入手し、実験に用いるまで飼育用水槽で 1 か月間飼育した。実験開始時時のマナマコの平均体重は 3.3±1.8 g であり、$^{125}I^-$ 又は $^{125}IO_3^-$を添加した海水を入れたばく露用水槽中に、マナマコ 12 個体を入れ、無投餌で飼育を行った。継時的にマナマコ 3 個体と海水試料を採取し、マナマコ体内 ^{125}I 濃度を測定するとともに、海水中 $^{125}I^-$を陰イオン交換樹脂カラムにより $^{125}I^-$と $^{125}IO_3^-$に分離して測定した。

1 度目の実験では、マナマコをばく露開始後 24、48 及び 72 時間に採取・分析を行ったところ、24 時間でマナマコ中 ^{125}I 濃度は概ね飽和していた。このため、2 度目の実験ではばく露開始後 6、12、24 及び 48 時間に採取・分析を行った。

更に、同時に漁獲されたマナマコを、SPring-8 産業利用 II ビームライン BL14B2 を用いて、ヨウ素 K 吸収端（33.164 keV）の X 線吸収端近傍構造（XANES）測定を行い、マナマコ中の安定ヨウ素の存在形態を調べた。

3. 成果の概要

3.1 水産物における放射性ヨウ素の形態別濃縮係数

1 度目及び 2 度目の実験結果をそれぞれ図 1 及び図 2 に示す。$^{125}I^-$添加海水中の $^{125}I^-$濃度減少速度は 1 度目の実験では 2 度目より速く、一方、マナマコ中 $^{125}I^-$濃度上昇速度も速くなっているが、その原因については不明である。

図 3 に示した代謝モデルにより、海水中 ^{125}I とマナマコ中 ^{125}I の間の移行速度は移行元の濃度に比例するものとし、移行パラメータ求めた。得られた値を表 1 に示す。$^{125}I^-$と $^{125}IO_3^-$について行った 1 度目の実験結果をまとめて解析した結果、k_1=6.E1、k_2=6.及び k_3=1.E-1 となり、^{125}I と $^{125}IO_3^-$の濃縮係数はそれぞれ 6.E2 及び 6.E1 であった。2 度目の実験結果からは、k_1=5.、k_2=1.及び k_3=3.E-2 となり、^{125}I と $^{125}IO_3^-$の濃縮係数はそれぞれ 2.E2 及び 5.E1 であった。得られた濃縮係数には幅があるが、I^-が IO_3^-に比べて吸収されやすいことが明らかとなった。

3.2 マナマコ中の安定ヨウ素の化学形態分析

XANES スペクトル測定結果より、マナマコに含まれるヨウ素の大部分が有機態であることが示唆された。

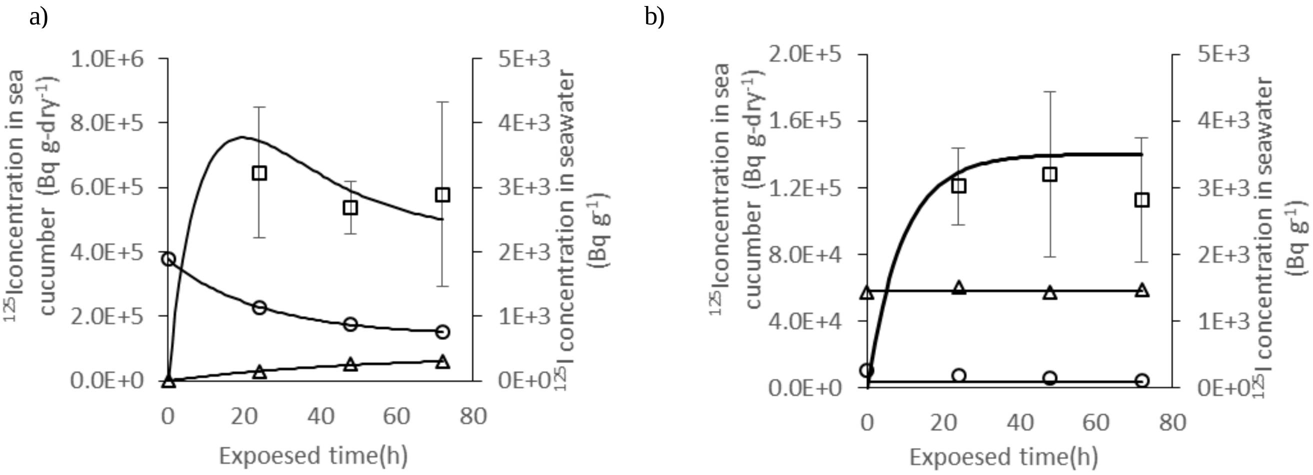

Fig. 1 Measured ^{125}I concentration in sea cucumber and seawater during the first experiment. Open square shows ^{125}I concentration in sea cucumber (Bq g-dry^{-1}). Open circles and triangles show $^{125}I^-$ and $^{125}IO_3^-$ concentrations in seawater (Bq g^{-1}). Solid lines show estimated value by the model.

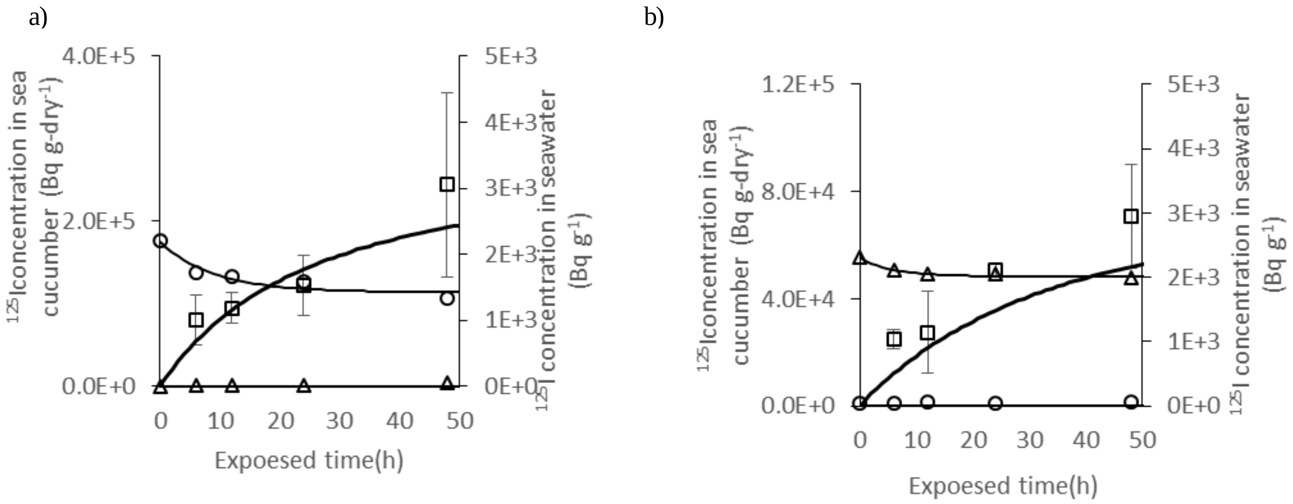

Fig. 2 Measured ^{125}I concentration in sea cucumber and seawater during the second experiment. Open squares show ^{125}I concentration in sea cucumber (Bq g-dry^{-1}). Open circles and triangle shows $^{125}I^-$ and $^{125}IO_3^-$ concentrations in seawater (Bq g^{-1}). Solid lines show estimated value by the model.

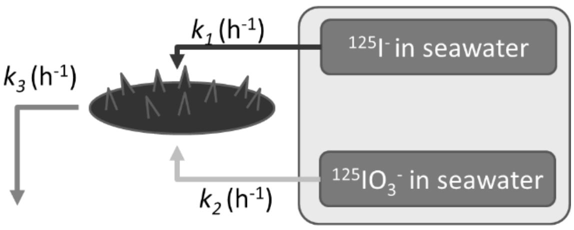

Fig. 3 Metabolic model of ^{125}I in sea cucumber.

Table 1 Obtained transfer-rate coefficients.

	k_1(h^{-1})	k_2(h^{-1})	k_3(h^{-1})
1st experiment	6.E1	6.	1.E-1
2nd experiment	5.	1.	3.E-2