I-129 LEVELS IN MARINE ENVIRONMENT ALONG THE SLOVENIAN COAST

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Abstract

^{129}I is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear power reprocessing plants increases. A radiochemical neutron activation analysis method was developed to measure the concentration of ^{129}I in environmental and biological samples. The method was validated using the IAEA-375 Soil, FC98 Seaweed and NIST 4357 Ocean Sediment. The method was applied to analyze $^{129}I/^{127}I$ isotopic ratios in the marine environment of Slovenia. The results found were in the range from 1.3 to $55.4 \cdot 10^{-9}$ μq g^{-1} for seawater, from 104 to $127 \cdot 10^{-9}$ μq g^{-1} for blue mussel, from 334 to $471 \cdot 10^{-9}$ μq g^{-1} for alga *Fucus virsoides* and from 72 to $256 \cdot 10^{-9}$ μq g^{-1} for marine sediment.

Keywords: Algae, Coastal Waters, Fallout, Radionuclides, Sediments

1 Introduction

The only stable natural iodine isotope is ^{127}I and the total amount of this element in the Earth's crust was estimated to be $8.6 \cdot 10^{15}$ kg of which nearly 70 % resides in marine sediments and 28 % in sedimentary rocks. The marine environment, i.e. the oceans, is the major source of iodine with average concentrations of around 60 μg L^{-1} iodine in seawater. The biogeochemical cycling of iodine is driven with its volatilization from oceans and soil to the atmosphere in the form of iodinated hydrocarbons of which methyl-iodide predominates. From the atmosphere the iodine is washed out to the marine and terrestrial environment by wet (precipitation) and dry (aerosol) depositions [1].

^{129}I ($T_{½} = 1.57 \cdot 10^7$ years) is the only natural radioactive isotope of iodine, which is formed in nature by two processes. The cosmogenic ^{129}I is produced in the atmosphere by the interaction of cosmic rays with xenon isotopes and the fissiogenic ^{129}I by spontaneous fission of uranium in the lithosphere. For the pre-nuclear era (no addition of anthropogenic ^{129}I to the environment) an $^{129}I/^{127}I$ isotopic ratio of about $1.5 \cdot 10^{-12}$ has been estimated. The quantity of ^{129}I in the pre-nuclear age ocean was ~100 kg. Since 1945 anthropogenic production of ^{129}I started which shifted the natural isotopic ratio for 3 to 6 orders of magnitude in favour of ^{129}I.
The main sources of ^{129}I are nuclear fuel reprocessing plants.

To our knowledge the ^{129}I level has not been measured in any biological or environmental sample from the Mediterranean area (Adriatic Sea). The aim of our work was to investigate the distribution of ^{129}I in the marine environment of Slovenia.

2 Methodology

2.1 Sampling and preparation
First sampling of alga (*Fucus virsoides*) and sediment was performed in 2005 and another sampling in 2009 including seawater, alga (*Fucus virsoides*) and blue mussel (*Mytilus galloprovincialis*). Alga and blue mussel were dried by freeze dryer to constant mass and homogenized. Seawater and sediment samples were analysed as collected.

2.2 Determination of ^{129}I and ^{127}I
Radiochemical neutron activation analysis method (RNAA) was used for the determination of ^{129}I [2] and ^{127}I [3] in environmental samples. Environmental samples contain very low amounts of ^{129}I therefore pre-concentration of iodine from up to 100 g of alga, blue mussel and sediment and up to 8 L of seawater are needed. Irradiation of sample, combustion in an oxygen atmosphere and extraction of iodine with $CHCl_3$ followed. Induced radioisotopes were measured on a HPGe detector. The chemical yield for the whole procedure was determined spectrophotometrically and by using the ^{126}I activity.

3 Results and Discussions

The method was applied to analyze $^{129}I/^{127}I$ isotopic ratios as well as ^{129}I and ^{127}I concentrations in the marine environment of Slovenia. The results found for analysed samples collected in 2005 and 2009 are summarised in Table 1. There are no literature dates for ^{129}I and ^{127}I concentrations in blue mussel. Values found in analysed seawater and sediment samples are in agreement with values found in literature for areas that are not under the influence of direct liquid discharges of ^{129}I from nuclear fuel reprocessing plants. ^{129}I and ^{127}I concentrations found in analysed alga collected in 2005 and 2009 (Table 1) are in the same range. The ratio of $^{129}I/^{127}I$ found for alga *Fucus virsoides* is up to 10^{-9}, which is one order of magnitude higher than along the coast of China, up to two orders of magnitude lower than in the Baltic Sea, which is influenced by direct liquid discharges from La Hague and Sellafield, and up to four orders of magnitude lower than in the vicinity of the La Hague reprocessing plant.

Tab. 1. Range of ^{129}I and ^{127}I in marine environment of North Adriatic Sea

Sample	Number of samples	Year of sampling	Range	
			^{127}I (μg g^{-1} dry weight)	^{129}I (10^{-9} μg g^{-1} dry weight)
Seawater	6	2009	0.052 – 0.070	1.3 – 55.4
Alga (*Fucus virsoides*)	3	2009	267 – 470	334 – 452
	5	2005[a]	371 – 448	362 – 471
Blue mussel (*Mytilus galloprovincialis*)	3	2009	9.9 – 14.6	104 – 127
Sediment	4	2005	59.3 – 76.8	72 – 256

[a] published in 2008 [4]

The data of this study represent a survey of ^{129}I in the marine environment of Slovenia. The most likely source of ^{129}I are nuclear fuel reprocessing plants in La Hague and Sellafield, which are known to be the major sources of ^{129}I in the environment of North Europe. ^{129}I is transferred to the atmosphere and washed out to the marine environment of Slovenia by precipitation, so it is of atmospheric-precipitation origin.

References

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