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THE USE OF ^{59}Ni, ^{99}Tc, AND ^{236}U TO MONITOR THE RELEASE OF RADIONUCLIDES FROM OBJECTS CONTAINING SPENT NUCLEAR FUEL DUMPED IN THE KARA SEA

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Abstract

Between 1965 and 1981, five objects – six naval reactor pressure vessels (RPVs) from four former Soviet Union submarines and a special container from the icebreaker *Lenin*, each of which contained damaged spent nuclear fuel (SNF) – were dumped in a variety of containments, using a number of sealing methods, at four sites in the Kara Sea All objects were dumped at sites that varied in depth from 12 to 300 m

This paper examines the use of the long-lived radionuclides ^{59}Ni, ^{99}Tc, and ^{236}U encased within these objects to monitor the breakdown of the containments due to corrosion Included are discussions of the radionuclide inventory and their release rate model, the estimated radionuclide mass in a typical seawater sample, and the potential for radionuclide measurement via Accelerator Mass Spectrometry (AMS)

Introduction

In the Spring of 1993, Russia released a summary of the former Soviet Union liquid and solid radioactive waste disposal operations, *Facts and Problems Related to Radioactive Waste Disposal in Seas Adjacent to the Territory of the Russian Federation* (Office of the President of the Russian Federation 1993) The *White Book*, as this report was later called, revealed, in part, that five objects – six naval RPVs from four former Soviet Union submarines and a special container from the icebreaker *Lenin*, each of which contained damaged SNF – were dumped at four sites in the Kara Sea, an arm of the Arctic Ocean located east of Novaya Zemlya Table 1 presents a summary of pertinent disposal information for the objects containing SNF dumped in the Kara Sea

The International Atomic Energy Agency (IAEA), as part of their responsibilities to the London Convention of 1972, initiated the International Arctic Seas Assessment Project (IASAP) (Sjoeblom and Linsley 1993) to study the possible health and environmental effects from disposal of these objects One outcome of the IASAP was an estimation of the radionuclide inventory and their release rates from these objects A follow-on concern is the ability to detect the radionuclides released into the water column The focus of the work reported here is the feasibility of using the long-lived radionuclides ^{59}Ni, ^{99}Tc, and ^{236}U encased within these objects to monitor the breakdown of the containments due to corrosion

Radionuclide Inventory and Their Release Rate Model

The inventories used in the calculations of the annual mass release of ^{59}Ni and ^{99}Tc are those reported by the IASAP Source Term Working Group (IAEA 1997) When inventories of ^{236}U computed either by the IASAP Source Term Working Group, or with burnup history from their work, were compared to those prepared by the State Institute of Applied Ecology (SIAE), Moscow (Rubtsov and Ruzhansky 1995), the SIAE predictions of ^{236}U mass were higher at all sites except that of the submarine factory number 601 (Stepovoy Fjord) object Thus,

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to assure an optimistic estimation of the annual mass release of ^{236}U, inventories used in the calculations are from two sources the IASAP Source Term Working Group (Stepovoy Fjord) and the SIAE (all other sites)

Using the "best estimate" discharge scenario, corrosion rates, and methodology of the IASAP Source Term Working Group models (IAEA 1997), the annual activity release (Bq/a) of ^{59}Ni, ^{99}Tc, and ^{236}U were determined for the five disposal sites over the estimated period of release Conversion to annual mass release (g/a) of ^{59}Ni, ^{99}Tc, and ^{236}U was accomplished through the appropriate application of isotopic specific activity (Bq/g) (Brown and Firestone 1986)

Table 1 Pertinent disposal information for the objects containing spent nuclear fuel dumped in the Kara Sea

Disposal site	Year of disposal	Factory number	Dumped object	Disposal coordinates[1]	Disposal depth[2] (m)
Abrosimov Fjord	1965	901	Reactor compartment	71° 56 03' N 55° 18 15' E	20 (10-15)
		285	Reactor compartment	71° 56 03' N 55° 18 08' E	20 (10-15)
Tsivolka Fjord	1967	OK-150	*Lenin* fuel container	74° 26 10' N 58° 36 15' E	50
Novaya Zemlya Depression	1972	421	Reactor	72° 40' N 58° 10' E	300
Stepovoy Fjord	1981	601	Submarine	72° 31 25' N 55° 30 25' E	50 (30)

[1] Disposal site coordinates for all units except those from factory number OK-150 are from the *White Book* (Office of the President of the Russian Federation 1993) Disposal site coordinates for the OK-150 units are from Sivintsev (September 1995)

[2] The disposal depths were provided in May 1993 by the Russian Federation; those in parenthesis were obtained during joint Norwegian-Russian scientific cruises in 1993 and 1994

Estimated Radionuclide Mass in a Typical Seawater Sample

To estimate the concentration of a radionuclide released from a dumped object to seawater, we used a simple equation to predict the average near-field concentration of a radionuclide released from a seabed dumpsite into a diffusive ocean (IAEA 1986) The radionuclide concentration released into seawater on an annual basis is given by

$$C = \frac{Q(3.2 \times 10^{-8})}{\pi R \sqrt{K_v K_h}} \qquad (1)$$

where

- C is the radionuclide concentration in seawater (g/m^3),
- Q is the annual radionuclide mass release rate (g/a),
- R is the distance from the seabed source (m),
- K_v is the vertical diffusivity (m^2/s), and
- K_h is the horizontal diffusivity (m^2/s)

The mass of a radionuclide present in a seawater sample is then determined from the product of the radionuclide concentration and the volume of the sample Expressed as an equation, the mass is given by

$$M = \frac{QV(3.2 \times 10^{-11})}{\pi R \sqrt{K_v K_h}} \qquad (2)$$

where

- M is the radionuclide mass in the seawater sample (g),
- V is the seawater sample volume (l), and

all other terms are previously defined Assuming a sample volume of 100 l, a source distance of R = 1000 m, and diffusivities of $K_v = 1.0 \times 10^{-4}$ m^2/s and $K_h = 1.0 \times 10^{2}$ m^2/s, Eq (2) simplifies to

$$M = Q(1.0 \times 10^{-11}) \qquad (3)$$

where all terms are previously defined Finally, applying Eq (3) to the annual ^{59}Ni, ^{99}Tc, and ^{236}U mass release rates from the dumped objects results in the estimated mass of each available in the sample for analysis

Potential for Radionuclide Measurement via Accelerator Mass Spectrometry

AMS detection of the long-lived actinide elements are expected to be at least 2-3 orders of magnitude more sensitive than classical radiometric counting methods AMS is also a considerably more sensitive and robust measurement technique compared with Inductively Coupled Plasma Mass Spectrometry (ICP-MS) Preliminary studies on the feasibility and sensitivity of using AMS detection demonstrate that detection limits in the order of 10^5 atoms (or < 0 1 fg) are achievable (Fifield et al 1997, Fifield et al 1996) Femtogram limits of detection have also been reported for ICP-MS but sensitivity is critically dependent on very careful optimization of instrument parameters, and on methods of sample introduction and data acquisition

Based on our predicted release rates and dispersion of radionuclides from the dumped objects into the near-field seawater, we have examined the feasibility of measuring ^{59}Ni, ^{99}Tc, and ^{236}U in the waters of the Kara Sea as a tool to monitor the breakdown of containments due to corrosion The limits of detection for measurement of ^{59}Ni ($t_{1/2}$ = 75,000 y) and ^{99}Tc ($t_{1/2}$ = 213,000 y) by AMS are about 2-5 fg and 10-100 fg, respectively The absolute detection efficiency for ^{236}U ($t_{1/2}$ = 23,420,000 y) by AMS is similar to that of other long-lived actinides (10^5-10^6 atoms or 0 04-0 4 fg) although actual detection limits will depend on isobaric interferences from ^{235}U Seawater contains about 3 3 µg/l of uranium or the equivalent of about 0 023 µg/l of ^{235}U With little or no discrimination in the mass-spectrometry, the worst case scenario would limit detection to about 10^7 atoms/l (4 fg/l) of seawater

The AMS limits of detection have been compared against the estimated mass of ^{59}Ni, ^{99}Tc, and ^{236}U expected in a 100 l seawater sample collected at a distance of 1000 m from the dumped objects as a function of the release year through the year 5000 Figures 1, 2, and 3 depict these results for the ^{59}Ni, ^{99}Tc, and ^{236}U, respectively Table 2 presents a summary of the year in which the ^{59}Ni, ^{99}Tc, and ^{236}U are estimated to be initially released and present in sufficient quantity for measurement by AMS Regardless of the range-in-detection limit, with the exception of the initial release years from the submarine factory 421 (Novaya Zemlya Depression) object, ^{59}Ni is estimated to be present in sufficient quantities to be measured throughout the release period at all five sites In the case of ^{99}Tc, the range-in-detection limit has a significant impact only on the measurable sample mass from the area of the submarine factory number 601 (Stepovoy Fjord) object The ^{99}Tc mass contained in a 100 l seawater sample from the area of the submarine factory number 601 (Stepovoy Fjord) object is estimated to be of a measurable quantity only at the lower limit of 10 fg and in all but the early release years At all other sites, regardless of the range-in-detection limit, with the exception of the early release years from the submarine factory number 901 (Abrosimov Fjord), 285 (Abrosimov Fjord), and 421 (Novaya Zemlya Depression) objects and late release from the *Lenin* fuel container (Tsivolka Fjord), ^{99}Tc is estimated to be present in sufficient quantities to be measured throughout the release period For the ^{236}U, the mass contained in a 100 l seawater sample from the area of the submarine factory number 601 (Stepovoy Fjord) object is estimated to be of a measurable quantity only after the year 3106 However, at all other sites, the ^{236}U is estimated to be present in sufficient quantities to be measured throughout all but the early release years from the submarine factory number 901 (Abrosimov Fjord), 285 (Abrosimov Fjord), and 421 (Novaya Zemlya Depression) objects and late release from the *Lenin* fuel container (Tsivolka Fjord)

For 100 l seawater samples collected a distance of 100 m from the dumped objects, the ^{59}Ni, ^{99}Tc, and ^{236}U mass is predicted to increase tenfold In the case of samples collected from the area of the submarine factory number 601 (Stepovoy Fjord) object, the ^{99}Tc could become measurable at the upper limit of 100 fg and the ^{236}U could become measurable some 925 years earlier

Figure 1 Comparison of Estimated ^{59}Ni Mass (g) in 100 Liters of Seawater from Various Disposal Sites in the Kara Sea versus Release Year

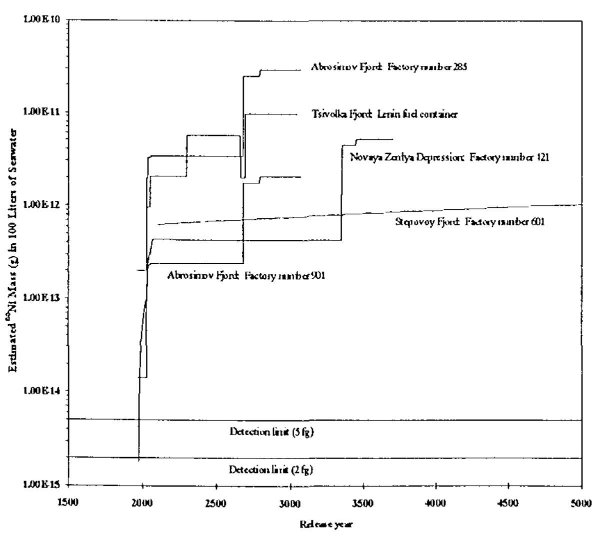

Figure 2 Comparison of Estimated ^{99}Tc Mass (g) in 100 Liters of Seawater from Various Disposal Sites in the Kara Sea versus Release Year

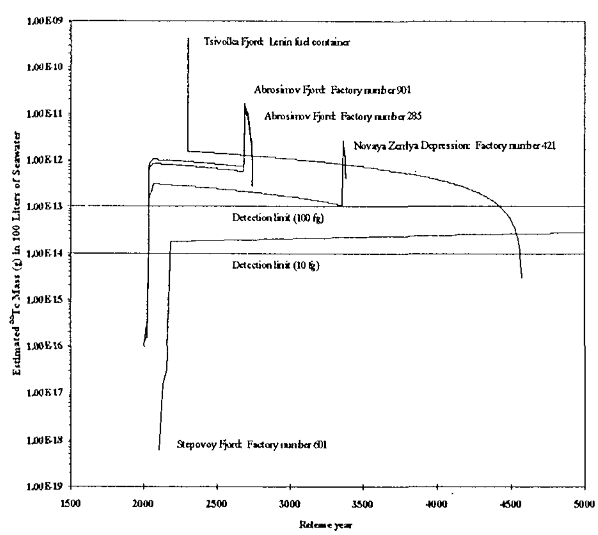

Figure 3 Comparison of Estimated ^{236}U Mass (g) in 100 Liters of Seawater from Various Disposal Sites in the Kara Sea versus Release Year

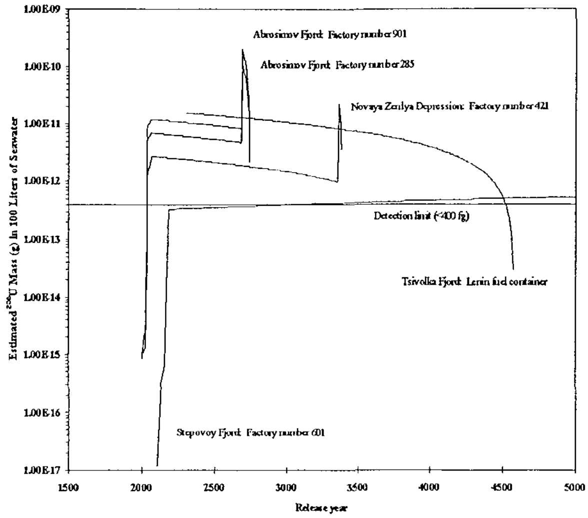

Table 2 Summary of the years in which the ^{59}Ni, ^{99}Tc, and ^{236}U are estimated to be initially released and present in sufficient quantity for measurement by AMS for the object containing spent nuclear fuel dumped in the Kara Sea

		Year of initial release or AMS measurable mass							
		^{59}Ni			^{99}Tc			^{236}U	
			AMS			AMS			AMS
Disposal site	Factory number	Release	2 fg limit	5 fg limit	Release	10 fg limit	100 fg limit	Release	<400 fg limit
Abrosimov Fjord	901	1966	1966	1966	2016	2039	2039	2016	2039
	285	1966	1966	1966	2016	2039	2039	2016	2039
Tsivolka Fjord	OK-150	2028	2028	2028	2303	2303	2303	2304	2304
Novaya Zemlya Depression	421	1973	1974	1975	2004	2035	3035	2004	2035
Stepovoy Fjord	601	2106	2106	2106	2106	2181	-	2106	3106

Conclusion

If AMS techniques for measurement of the long-lived radionuclides ^{59}Ni, ^{99}Tc, and ^{236}U achieve the detection limits we have considered, then measurement of these radionuclides encased within the objects containing SNF dumped into the Kara Sea is a valuable tool to monitor the breakdown of the containments due to corrosion Seawater samples of 100 l volume collected within 1000 m of the five sites are predicted to yield measurable quantities of ^{59}Ni throughout virtually the entire release period Whether the ^{99}Tc mass collected from the area of the submarine factory number 601 (Stepovoy Fjord) object is measurable depends on the range-in-detection limit At the lower limit of 10 fg, ^{99}Tc is estimated to be present in sufficient quantities to be measured throughout essentially the entire release period at all sites At the upper limit of 100 fg, measurable quantities of ^{99}Tc mass are also predicted from the submarine factory number 601 (Stepovoy Fjord) object if the samples are collected 100 m from the object Disposal sites with the greatest relative release periods of measurable ^{236}U mass are those containing the submarine factory number 901 (Abrosimov Fjord), 285 (Abrozimov Fjord), and 421 (Noyaya Żemlya Depression) objects and the *Lenin* fuel container (Tsivolka Fjord) If samples are collected 100 m from the submarine factory number 601 (Stepovoy Fjord) object, measurable quantities of ^{236}U are predicted in the year 2181, some 925 years earlier

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