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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Between 1965 and 1981, five objects and six naval reactor pressure vessels (RPVs) from four former Soviet Union submarines and a special container from the icebreaker Lenin, all containing damaged spent nuclear fuel (SNF) were dumped in a variety of containments, at four sites in the Kara Sea.

The International Atomic Energy Agency initiated the International Arctic Seas Assessment Project (IASAP) 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 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.

TABLE I. 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	Disposal depth (m)
Abrosimov Fjord	1965	901	Reactor compartment	71°56.03'N 55° 18.15'E	20
		285	Reactor compartment	71° 56.03'N 55° 18.08'E	20
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

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) from the objects listed in Table I. Using the best estimateD discharge scenario, corrosion rates, and methodology of the IASAP Source Term Working Group models [1], 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).

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 [2]. Then, the mass of a radionuclide present in a seawater sample is determined from the product of the radionuclide concentration and the volume of the sample.

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Accelerator Mass Spectrometry (AMS) detection of the long-lived actinide elements is expected to be at least 2-3 orders of magnitude more sensitive than classical radiometric counting methods. 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 [3].

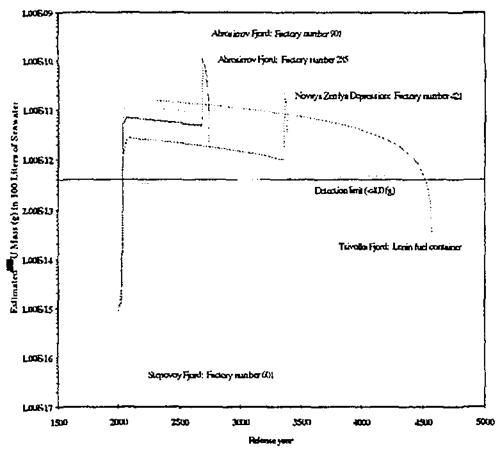
Based on our predicted release rates and dispersion of radionuclides from the dumped objects into the near-field seawater, we have examined the feasibility of measuring ^{59}Ni, ^{99}Tc, and ^{236}U in the waters of the Kara Sea as a tool to monitor the breakdown of containments due to corrosion. The limits of detection for measurement of ^{59}Ni and ^{99}Tc by AMS are about 2-5 fg and 10-100 fg, respectively. The absolute detection efficiency for ^{236}U 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.

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 to the year 5000. Figure 1 shows the result for the ^{236}U. ^{59}Ni is estimated to be present in sufficient quantities to be measured throughout the release period at all five sites. For ^{99}Tc the mass contained in the sample from the area of the submarine 601 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, with the exception of the early release years from the objects 901, 285, and 421 and late release from the Lenin fuel container, ^{99}Tc is estimated to be present in sufficient quantities to be measured throughout the release period. For ^{236}U, from the seawater sample off submarine 601, it is estimated to be of a measurable quantity only after the year 3106. At all other sites however, the ^{236}U is estimated to be present in sufficient quantities to be measured throughout all but the early release years for 901, 285, & 421 and late release from the Lenin fuel container.

For 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 number 601, the ^{99}Tc could become measurable at the upper limit of 100 fg and the ^{236}U could become measurable some 925 years earlier.

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 collected within 1000 m of the 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 601 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 most of 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 601, if the samples are collected 100 m from October 2, 1998the object. Disposal sites with the greatest relative release periods of measurable ^{236}U mass are those containing the units 901& 285 (Abrosimov Fjord), and unit 421 (Novaya Zemlya Depression) and the Lenin fuel container (Tsivolka Fjord). If samples are collected 100 m from the submarine number 601 (Stepovoy Fjord), measurable quantities of ^{236}U are predicted in the year 2181, some 925 years earlier.

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



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