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RADIONUCLIDE SOURCES IN THE BARENTS AND KARA SEAS

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INTRODUCTION:

Some of the major sources for radionuclide contamination in the Arctic Ocean are past nuclear explosions and dumping activities on and nearby the island of Novaya Zemlya (Yablokov et al., 1993; Aarkrog, 1994). Prior to the Moscow Test Ban Treaty of August, 1963 approximately 79 nuclear bombs, exceeding 150 megatons yield were detonated by the Soviet military in the atmosphere over Novaya Zemlya. Earlier nuclear testing (1955-61) resulted in the detonation of at least three kiloton yield bombs in shallow water (<100 m) proximal to Chernaya Bay in Southern Novaya Zemlya. Other sources of radioactive contamination near Novaya Zemlya include liquid radioactive wastes dumped in the Barents Sea and solid radioactive wastes in nuclear reactors and barges dumped in the Kara Sea. The potential environmental threats have led to a wide range of marine radioactivity investigations (eg. Strand et al. (1994)) throughout the Barents and Kara Seas, including the present study that focuses on identifying sources, storage areas and delivery rates for radionuclide contaminated sediments in the Novaya Zemlya coastal zone.

METHODS AND MARINE EXPEDITIONS

A preliminary study of radionuclide distributions in Barents Sea sediments was carried out by our research group on board the R/V Geolog Fersman and the R/V Dalniye Zelentsy in the Barents Sea in 1992. These results indicated that levels of artificial radionuclides throughout the Barents Sea were generally consistent with fallout background levels, with one major exception. High levels of ^{239,240}Pu were measured in a single surface sediment collected from the small (1-6 km wide, 15 km long) fjord of Chernaya Bay, the site of several underwater nuclear weapons tests in the 1950s (Smith et al., 1995). Sediment and large volume seawater samples were collected during a subsequent 1993 cruise of the RV Geolog Fersman to Chernaya Bay (Figure 1). During this cruise samples were also collected near a radioactive waste dumpsite in the Novaya Zemlya Trough. At both locations, large volume (50-100 l) seawater samples were passed through KCFC resin cartridges to extract ^{137}Cs and the resins were transported to the BIO laboratory and analyzed using a gamma ray spectrometer. Plutonium and strontium were precipitated from seawater using iron hydroxide and sodium carbonate techniques. One liter water samples were shipped to the IsoTrace Laboratory (Toronto, Ont.) where ^{129}I was measured by accelerator mass spectrometry (Kilius et al., 1992). Sediments were collected using box and gravity (Lehigh) cores, sub-sampled on the ship and returned to the BIO laboratory for radionuclide analyses for ^{210}Pb, $^{239,240}Pu$, ^{241}Am, ^{129}I and gamma emitting radionuclides.

RESULTS AND DISCUSSION

Chernaya Bay

Chernaya Bay is a shallow (< 100 m) embayment on the southern coast of Novaya Zemlya bordered by low elevation (< 50 m) accretionary spits formed by long-shore drift from the southeast. High levels of $^{239,240}Pu$ (2,517 Bq/kg and 11,148 Bq/kg) were measured in surface sediments (0-2 cm) of box cores collected in the fine-grained sediments in the deeper (60-80 m) central region (Sta. 112 and 113) of the fjord (Figure 2). These plutonium levels are similar to those measured in sediments at sites of underwater nuclear weapons tests at Enewetak Atoll in the Pacific Ocean (Nelson and Noshkin, 1973) and at the site of the nuclear weapons accident in Thule, Greenland (Smith et al., 1994). Reduced $^{239,240}Pu$ levels (395 and 1525 Bq/kg) were measured in surface sediments of cores at the northern extremity (Sta. 110) of the fjord and near the 20 m sill (Sta. 114), respectively. $^{239,240}Pu$ levels decrease further to 106 Bq/kg in surface sediments immediately outside the fjord, indicating that Chernaya Bay has been relatively effective in retaining the original inventory of bomb-produced, particle-reactive radionuclides. ^{137}Cs and ^{60}Co levels were much lower than those of $^{239,240}Pu$ with maximum values in surface sediments of 80 Bq/kg and 140 Bq/kg, respectively being measured at Sta. 113.

The inventory of artificial radionuclides in Chernaya Bay sediments is almost entirely retained in the upper 20 cm of the sediment column. Sediment-depth distributions of $^{239,240}Pu$, ^{137}Cs and ^{60}Co typically exhibit a plateau or sub-surface maximum in the upper 5 cm and then decrease to background levels by the 15-20 cm level. In contrast, ^{210}Pb levels in Chernaya Bay sediments are typical of uncontaminated, fine-grained marine sediments and can therefore be used to constrain the sedimentation and biological mixing (biodiffusion) rates (Smith et al., 1986; 1995). The application of a single layer, bio-diffusion model to radionuclide profiles in core 113 results in good agreement with measured profiles for parameter values given in Figure 3 where a constant input flux was assumed for ^{210}Pb and an impulse input flux in 1957 was assumed for the artificial radionuclides. These results indicate that sedimentation rates in Chernaya Bay are low (< 0.1 cm/y) and that the sediment-depth distribution of radionuclides is governed mainly by biological mixing processes. Elevated levels of $^{239,240}Pu$ (4.2 Bq/m^3) were also measured in unfiltered bottom water in Chernaya Bay, owing either to the release of dissolved $^{239,240}Pu$ from bottom sediments and/or to the resuspension of plutonium-enriched, surficial sediments.

Sediments in Chernaya Bay are characterized by low initial $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ atom ratios of 0.030 and 0.0012, respectively, which are reduced from ratios of 0.18 and 0.015, respectively typical of global fallout (Smith et al., 1995). These low ratios are caused by low yields of ^{240}Pu and ^{241}Pu in the relatively inefficient nuclear tests conducted in Chernaya Bay. The low yield of ^{241}Pu has resulted in a $^{241}Am/^{239,240}Pu$ activity ratio of 0.05, which is also considerably lower than the value of 0.18, typical of modern sediments contaminated by atmospheric fallout. The $^{241}Am/^{239,240}Pu$ activity ratio in surface sediments increases with increasing distance from its value of 0.05 in Chernaya Bay to fallout levels of 0.30 at distances of 150 km into the Barents Sea. $^{241}Am/^{239,240}Pu$ activity ratio data provides a cost-effective method for estimating the fractional quantity of Chernaya Bay plutonium transported into the Barents Sea, assuming end member mixing between sediments contaminated separately by atmospheric fallout and by nuclear tests in Chernaya Bay.

N1 Dumpsite - Novaya Zemlya Trough

Following the work conducted in Chernaya Bay the RV Geolog Fersman proceeded to the N1 Dumpsite in the Novaya Zemlya Trough (Figure 1). Sediment cores and seawater samples were collected at well-defined positions proximal to a sunken barge reported in the Yablokov Report to have been carrying 118 Ci of solid radioactive wastes when it was scuttled in 1980. ^{137}Cs and $^{239,240}Pu$ activities in surface sediments (Figure 4) are typically 20 Bq/kg and 1 Bq/kg, respectively, and represent values that are typical of fallout levels in the Barents and Kara Seas (Strand et al., 1994). Excess ^{210}Pb and artificial radionuclides are generally retained in the upper 10 cm of the sediment column. ^{137}Cs and $^{239,240}Pu$ activities of 14 Bq/m^3 and 0.02 Bq/m^3, respectively, measured in deep water from the dumpsite region represent combined inputs from atmospheric nuclear tests, Chernobyl and Atlantic water contaminated by releases from the Sellafield Reprocessing Plant. Contamination of this water from reprocessing plant inputs is also evidenced by the relatively high $^{129}I/^{137}Cs$ atom ratios of 70-80 measured in bottom water that are in excess of levels of < 10 that are typical of atmospheric fallout (Raisbeck et al., 1993). There is no evidence for enhancement of radioactivity levels in sediments or seawater proximal to the barge to indicate recent leakage of radioactive contaminants from the dumpsite.

CONCLUSIONS:

1. Elevated levels of artificial radionuclides as great as 15,000 Bq/kg for $^{239,240}Pu$, 250 Bq/kg for ^{137}Cs and 100 Bq/kg for ^{60}Co were measured in sediments in Chernaya Bay which have been contaminated by several nuclear tests conducted in the 1950s.

2. Sediment-depth distributions of $^{239,240}Pu$ and other artificial radionuclides are consistent with results from biodiffusion models that are constrained by ^{210}Pb sediment-depth distributions. These results indicate that sedimentation rates in Chernaya Bay are low (< 0.05 cm/y) and that downward transport of radioactive contaminants is governed

mainly by sediment mixing or bioturbation with the entire inventory retained in the upper 20 cm of the sediments

3. Chernaya Bay plutonium is distinguished by low initial $^{240}Pu/^{239}Pu$ and $^{241}Pu/^{239}Pu$ atom ratios of 0.030 and 0.0012, respectively and a $^{241}Am/^{239,240}Pu$ activity ratio of 0.05 (compared to 0.3 in fallout) which provides a method for tracking its dispersion over distances of 100 km into the Barents Sea.

4. Artificial radionuclide levels in sediments and seawater near a sunken barge loaded with radioactive wastes in the Novaya Zemlya Trough are similar to background fallout levels in the Kara Sea and provide little evidence for the release of radioactive contaminants from the dumpsite.

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Figure 1. Cruise track

Figure 2. Chernaya Bay

Figure 3. Radionuclide sediment profiles at Station 113 in Chernaya Bay are consistent with results from a biodiffusion model for a sedimentation rate, ω, of 0.05 cm/y and a biodiffusion coefficient, D_b of 0.15 cm^2/y indicating that downward transport of radionuclides is governed mainly by biological mixing.

Figure 3. Plutonium transport from Chernaya Bay is reflected by a decrease in the $^{239,240}Pu$ signal, normalized to % (clay + silt) component of the sediments, with increasing distance seaward from Chernaya Bay. The increase in the $^{241}Am/^{239,240}Pu$ ratio with increasing distance up to 100 km reflects mixing of sediments contaminated with Chernaya Bay fallout ($^{241}Am/^{239,240}Pu$ = 0.05) and atmospheric fallout ($^{241}Am/^{239,240}Pu$ = 0.30)

Figure 4. Large volume water samples were collected in the vicinity of the barge dumpsite in the Novaya Zemlya Trough (see Figure 1) using a 100 l sampling bottle. Radionuclide bottom water concentrations around dumpsite location are typical of levels measured throughout the Kara Sea and Novaya Zemlya Trough and show no indication of enhanced releases of radioactivity from the barge.

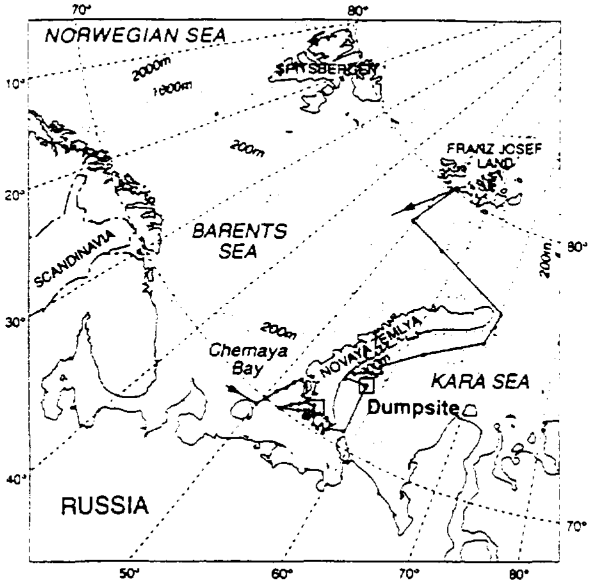

Figure 1: Cruise track for R/V Geolog Fersman in September 1993. The location of Chernaya Bay and the site of the dumped vessel are marked.

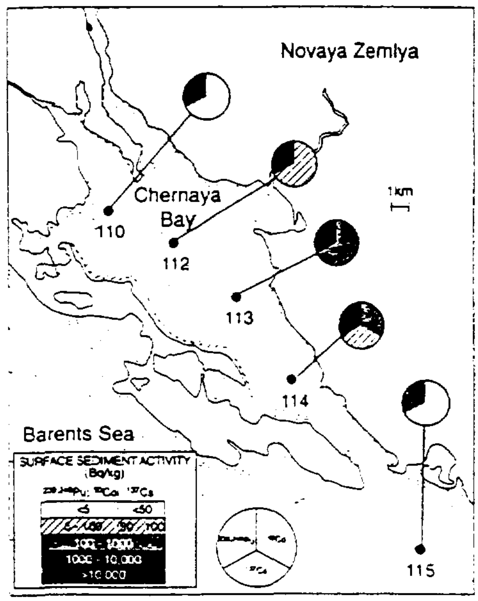

Figure 2: Levels of Cesium-137, Plutonium-239,240 and Cobalt-60 measured in surface sediments in Chernaya Bay

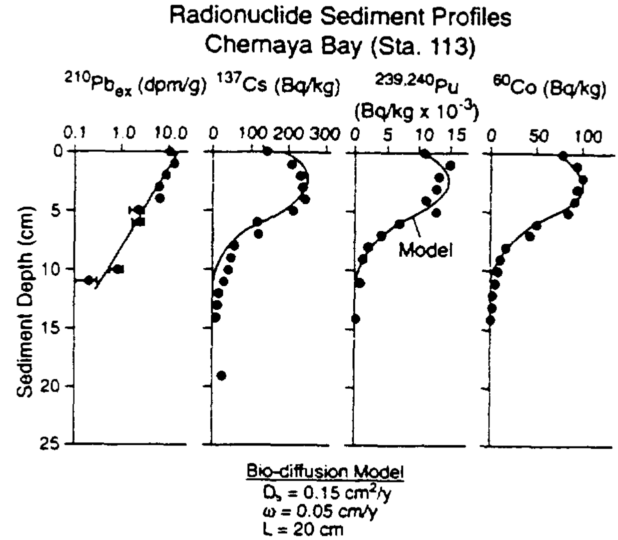

Figure 3. Radionuclide sediment-depth profiles (Sta. 113) in Chernaya Bay are simulated by biodiffusion model using steady-state flux of ^{210}Pb, and 1957 pulsed input of artificial radionuclides. Plutonium concentrations are among the highest ever recorded in the marine environment.

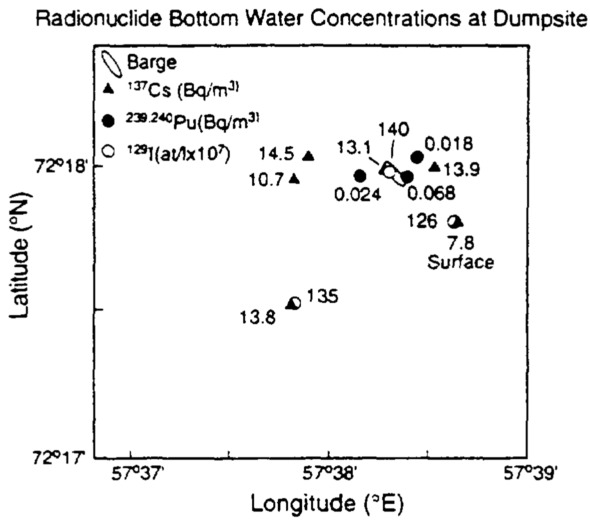

Figure 4. Elevated ^{137}Cs levels in bottom compared to surface water are due to older age of deep water in Novaya Zemlya Trough labelled by higher levels of Sellafield and Chernobyl tracers. Bottom water activities reveal no evidence for radioactivity releases from the dumpsite.