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Radionuclide Bioconcentration Factors and Sediment Partition Coefficients in Arctic Seas Subject to Contamination from Dumped Nuclear Wastes

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The disposal of large quantities of radioactive wastes in Arctic Seas by the former Soviet Union has prompted interest in the behavior of long-lived radionuclides in polar waters. Previous studies on the interactions of radionuclides prominent in radioactive wastes have focused on temperate waters; the extent to which the bioconcentration factors and sediment partitioning from these earlier studies could be applied to risk assessment analyses involving high latitude systems is unknown. Here we present concentrations in seawater and calculated in situ bioconcentration factors for ^{90}Sr, ^{137}Cs, and $^{239+240}$Pu (the three most important radionuclides in Arctic risk assessment models) in macroalgae, crustaceans, bivalve molluscs, sea birds, and marine mammals as well as sediment K_d values for 13 radionuclides and other elements in samples taken from the Kara and Barents Seas. Our data analysis shows that, typically, values for polar and temperate waters are comparable, but exceptions include 10-fold higher concentration factors for $^{239+240}$Pu in Arctic brown macroalgae, 10-fold lower K_d values for ^{90}Sr in Kara Sea sediment than in "typical" temperate coastal sediment, and 100-fold greater Ru K_d values in Kara Sea sediment. For most elements application of temperate water bioconcentration factors and K_d values to Arctic marine systems appears to be valid.

Introduction

High and low level radioactive wastes, the current activity totalling approximately 4.5 PBq (*1*), have been dumped by the former Soviet Union into Arctic waters, particularly the Kara Sea (*1*−*4*). These disclosures have precipitated a series of laboratory and field studies to examine the behavior of long-lived radionuclides in Arctic regions, including interactions with Arctic organisms and sorption to Arctic sediments (*5*−*12*). A recent summary of the radiological impacts estimated for the Kara Sea has concluded that releases from dumped objects have been relatively small and that radiological risks to human and marine animal populations are also small (*13*). To address concerns regarding risks associated with these disposed wastes, modelers require quantitative estimates of radionuclide concentrations in Arctic organisms and sediments. Previously, there have been few measurements of radionuclides in polar waters, and consequently radiological risk assessments have had to rely on data compilations from temperate waters for which there is a more comprehensive database (*14*). Ectotherms inhabiting cold waters have slower metabolic rates and higher lipid reserves than do comparable organisms in warmer waters, and it is not known whether these would influence bioconcentration factors for the radionuclides disposed in the Arctic. In fact, there have been very few systematic comparisons between Arctic and non-Arctic conditions affecting bioconcentration factors and sediment partition coefficients for radionuclides. The question has arisen as to whether radionuclide bioconcentration factors and sediment partition coefficients in the Arctic are different from those derived from studies in temperate ecosystems.

To address the paucity of data regarding bioconcentration factors in Arctic marine organisms, water, sediments, and organisms were collected from the Kara and Barents Seas and analyzed for representative long-lived radionuclides associated with the disposed wastes. We used these data to calculate bioconcentration factors and sediment partition coefficients, which can be used for understanding the biological and geochemical behavior as well as evaluating the risks of the released radionuclides. In this paper, we present in situ bioconcentration factors for select radionuclides in contaminated Arctic seas and compare these values with previously published mean values from temperate waters. This is the first attempt to systematically compare Arctic concentration factors with data collected by different national and international programs, brought together by the International Atomic Energy Agency.

Materials and Methods

Bioconcentration factors were determined for diverse Arctic organisms from the Barents and the Kara Seas (Figure 1). Within the Kara Sea, samples were taken from the Novaya Zemlya Trough, Abrosimov, Tsivolki, and Stepovogo Fjords, and the open Kara Sea (Figure 1). Water, sediment, and biota samples were collected as described elsewhere (*11, 12, 15, 16*). ^{137}Cs concentrations in water, sediment, and biota samples were determined by γ spectrometry using high-resolution HPGe or Ge(Li) detectors, ^{90}Sr concentrations by β spectrometry, and $^{239+240}$Pu by α spetrometry, as described elsewhere (*14*, *15*). In addition to the three radionuclides

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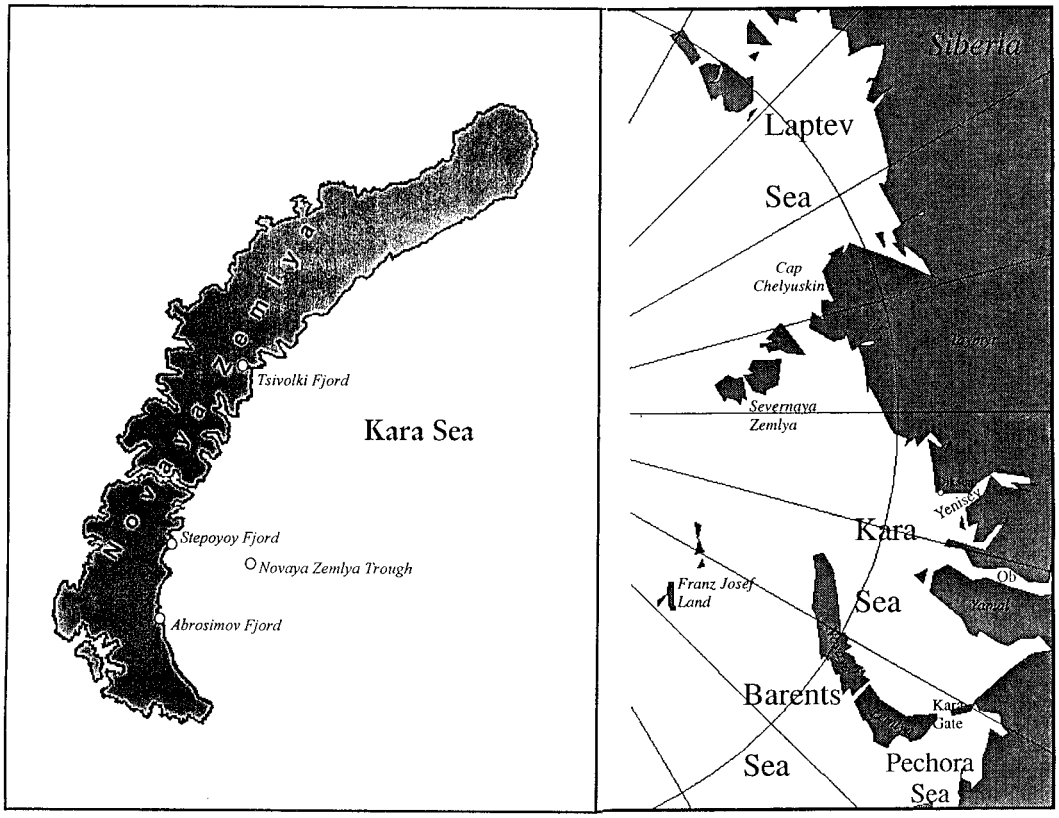

FIGURE 1. Map of study site area, including details of Novaya Zemlya.

^{90}Sr, ^{137}Cs, and $^{239+240}$Pu, calculations were made for trace elements with radioisotopes that are prominent in the radioactive wastes disposed in the Kara Sea (^{59}Ni, ^{63}Ni, ^{60}Co, ^{106}Ru, ^{129}I, ^{125}Sb,^{141}Ce, ^{154}Eu, ^{155}Eu, and ^{210}Pb) (*1, 4*). Stable element concentrations were determined by graphite furnace AAS and neutron activation analysis (*15*).

Bioconcentration factors were determined by dividing the metal or radionuclide concentration in organisms by the dissolved (<1 μm) metal or radionuclide concentration in ambient seawater. Thus, concentration factors were computed as moles of element (or Bq radioactivity) g^{-1} wet wt organism/moles element (or Bq radioactivity) mL^{-1} water. For fish, macroalgae, seabirds, and seals, surface water concentrations were used; for the invertebrates, which are benthic, bottom water concentrations were used. For those elements where there were no measured concentrations in Arctic waters, dissolved concentrations in North Atlantic surface waters were used. Because the suspended particle loads in Arctic waters are typically low ($\leq$1 mg L^{-1}) (*17*) most of the elements/radionuclides in the water column should be in the dissolved phase (even for the most particle-reactive elements), and thus the bioconcentration factors for organisms in these waters could be assumed to be essentially equivalent to the bioaccumulation factors (element g^{-1} organism/total element mL^{-1}).

The partition coefficients, or K_d values, of elements/radionuclides for sediments were determined for surface sediments from different regions of the Kara Sea. The K_d values were determined by dividing the moles element (or Bq radioactivity) of each gram (dry wt) of sediment by the moles element (or Bq radioactivity) in 1 mL of bottom water. The K_d values of selected radionuclides in Kara Sea sediments determined in laboratory and shipboard experiments are also presented (*11*).

TABLE 1. Seawater Concentrations of Radionuclides and Trace Elements in Surface and Deep Waters of Northern Seas Used for Calculations of Bioconcentration Factors and Sediment K_d Values[a]

radionuclide/ metal	area	concn (mBq L^{-1} or ng L^{-1}) surface water	deep water
^{90}Sr	Barents Sea	4	3.4
	Kara Sea	5	
^{137}Cs	Barents Sea	5	5.3
	Kara Sea	6	
$^{239+240}$Pu	Barents Sea	0.004	0.009
	Kara Sea	0.005	
Ni	N. Atlantic	120	360
Co	N. Atlantic	1.5	1−17
Eu	N. Atlantic	0.25	0.10
Ru	N. Atlantic	<0.005	0.005
Ce	N. Atlantic	7	11
Sb	N. Atlantic		207
Pb	N. Atlantic	4.1	25.9
I	N. Atlantic	60000	60000

[a] Concentrations of trace elements from Donat and Bruland (*20*).

Results and Discussion

The concentration of dissolved ^{90}Sr, ^{137}Cs, and $^{239+240}$Pu in surface water from the Kara and Barents Seas are given in Table 1. Bottom water concentrations of these radionuclides are also shown as are surface and deep-water concentrations of other elements in the North Atlantic which were used in calculations of sediment K_d values. Surface water concentrations of each of the three radionuclides are very similar between the two bodies of water, indicating no evidence of locally elevated concentrations of any of these contaminants in the dissolved phase. Further, surface and deep water concentrations of ^{90}Sr and ^{137}Cs generally do not differ appreciably (Table 1).

TABLE 2. Bioconcentration Factors of ^{90}Sr, ^{137}Cs, and $^{239+240}Pu$ in Marine Organisms Collected from the Kara and Barents Seas[a]

organisms	^{90}Sr	*n*	^{137}Cs	*n*	$^{239+240}Pu$	*n*
brown macroalgae[b]	182 ± 48	2	124 ± 124	12	1.6 ± 0.7 × 10^4	5
IAEA value	5		50		2 × 10^3	
molluscs[c]	nd		63 ± 42	13	nd	
IAEA value			30			
crustaceans[d]	nd		52 ± 53	5	nd	
IAEA value			30			
fish muscle[e]	4.1 ± 2.4	7	146 ± 68	11	<500	7
IAEA value	2		100		40	
echinoderms[f]	nd		62 ± 40	5	nd	
seabird muscle[g]	nd		414 ± 352	7	nd	
seabird liver	nd		530 ± 307	5	nd	
seal muscle	0.4−1.2	2	13−70	2	nd	
seal liver	0.2−3		nd			

[a] Values are means ± 1 SD. There were no significant differences between concentration factors for each radionuclide in organisms within each taxonomic group or between similar organisms from different regions; therefore, the results for each group were pooled. The recommended concentration factors for these radionuclides in similar organisms from temperate marine waters are also presented for brown macroalgae, crustaceans, bivalve molluscs, and fish (*14*). Not determined: nd. [b] Species analyzed include *Fucus vesiculosus, Laminaria saccharina, Ascophyllum nodosum, Palmaria palmata, Odonthalia dentata,* and *Chorda filum.* [c] Species analyzed include *Chlamys islandica, Buccinum* sp., *Macoma calcarea, Yoldia hyperborea, Tridonta borealis, Ciliatocardium ciliatum, Modiolus modiolus, Mytilus edulis*, *Portlandia arctica*, and *Musculus niger.* [d] Species analyzed include *Hyas araneus, Pagurus pubescens,* and *Mesidothea entomon.* [e] Species analyzed include Arctic cod *Gadus morphua morphua,* haddock *Melanogrammus aeglefinus,* seithe *Pollachius virens,* seacat *Anarhichas lupus*, plaice *Pleuronectes* sp., redfish *Sebastes marinus,* and ray *Raja radiata.* [f] Species analyzed include sea urchin *Strongylocentrotus* sp., seastar *Asterias rubens*, ophuiroid *Stegophiura nodosa,* holothurian *Cucumaria frondosa,* and ophuiroid *Ophiothrix fragilis.* [g] Species analyzed include *Larus marinus, Larus canus, Stercorarius skua, Mergus merganser, Tringa erythropus, Calidris minuta, C. maritima, Somateria mollissima,* and *Cepphus grylle.*

The concentration factors of ^{90}Sr, ^{137}Cs, and $^{239+240}Pu$ in macroalgae, bivalve molluscs, diverse crustaceans, sea birds, and marine mammals are given in Table 2. Because different species within each group did not differ significantly in their concentration factors for each radionuclide, they were pooled and means were generated. Concentration factors for ^{137}Cs in sea bird muscle tissue were about an order of magnitude higher than ^{137}Cs concentration factors computed from earlier work with different bird species in the Arctic Ocean around Svalbard (*18*). Cs concentration factors were also higher than in other organisms in our study, probably because the birds are in contact with seawater primarily for catching prey and cannot depurate themselves of accumulated contaminants through desorption in the way that other marine organisms can. The bird tissue may also reflect ^{137}Cs obtained from terrestrial sources. The relatively high variability observed in the concentration factors of all organisms is not unexpected, given the variability in species, age, and sex of the animals analyzed, and, in some cases, their mobility, which could greatly affect their exposure history if they move between regions with different contaminant levels.

Table 2 also compares the mean concentration factors with mean recommended values from the International Atomic Energy Agency (*14*), which are based largely on field and laboratory studies using temperate water organisms. Where comparisons are possible, it appears that concentration factors are somewhat higher in organisms in the Arctic than in organisms in temperate waters, although it is emphasized that there is considerable variability in the data. Further, the database for the Arctic is still very limited compared to a much wider database for temperate organisms. Explanations for higher bioconcentration factors in Arctic organisms are not obvious. Since concentration factors under steady-state conditions equal influx rates divided by efflux rates (*19*), it is possible that the slower digestion and metabolism of cold water animals results in slower efflux rates than in warmer water species. However, this possibility does not apply to macroalgae which accumulate radionuclides only from the dissolved phase. Arctic values are clearly higher for Sr and Pu than the IAEA recommended values derived from temperate water studies in which there was considerable spatial variability among the data used in the IAEA compilation (*14*).

Several laboratory studies have investigated the effects of temperature on radionuclide accumulation in Arctic organ-

TABLE 3. Partition Coefficients (K_d Values) for Radionuclides and Trace Elements in Arctic Surface Sediments[a]

radionuclide/ metal	area	K_d	type of data
^{60}Co	open Kara Sea	2 × 10^6	field
	open Kara Sea	1 × 10^4	expt
	Abrosimov Fjord	1.2 × 10^3	expt
	Stepovogo Fjord	1.9 × 10^6	expt
^{90}Sr	open Kara Sea	40	field
	Tsivolki Fjord	60	field
	Stepovogo Fjord	200	field
	Barents Sea	50	field
	Abrosimov Fjord	15−130	expt
	open Kara Sea	4−12	expt
^{137}Cs	Tsivolki Fjord	4 × 10^3	field
	Stepovogo Fjord	2−20 × 10^3	field
	Abrosimov Fjord	6 × 10^3	field
	open Kara Sea	2 × 10^3	field
	open Kara Sea	2 × 10^3	field
	open Kara Sea	4 × 10^3	field
	Novaya Zemlya Trough	0.5−4.3 × 10^3	field
	open Kara Sea	200	expt
	open Kara Sea	300	expt
	open Kara Sea	300−700	expt
	open Kara Sea	1 × 10^3	expt
	Abrosimov Fjord	1 × 10^3	expt
	Abrosimov Fjord	190−800	expt
$^{239+240}Pu$	Eastern Kara Sea	1.5 × 10^5	field
	Western Kara Sea	8 × 10^4	field
	Central Kara Sea	1.2 × 10^5	field
^{241}Am	Stepovogo Fjord	1.4−3 × 10^6	field
	open Kara Sea	1.2 × 10^6	expt
	open Kara Sea	0.1−3 × 10^6	expt
Ni	open Kara Sea	0.8−1.6 × 10^5	field
	Novaya Zemlya Trough	1.3 × 10^4	field
Ru	Abrosimov Fjord	1.8−4 × 10^4	expt
Sb	open Kara Sea	2−200 × 10^2	field
I	open Kara Sea	40	expt
Ce	open Kara Sea	9 × 10^6	field
Eu	open Kara Sea	6 × 10^6	field
	Abrosimov Fjord	0.8−1.6 × 10^5	field
Pb	open Kara Sea	4 × 10^6	field

[a] Field-based values were calculated as Bq (or mol) g^{-1} dry wt sediment divided by Bq (or mol) mL^{-1} bottom water. Experimental values are site-specific; they were determined with sediment from specific regions and are derived from measured partitioning of γ-emitting radionuclides between dissolved and particulate phases following the basic protocol given in Fisher et al. (*21*), as described in Carroll et al. (*11*) and Salbu et al. (*15*).

TABLE 4. Comparison of Mean K_d Values for Radionuclide/Metal Binding to Kara Sea Sediments and Coastal Sediments from Temperate Regions

element	mean K_d for Kara Sea sediment (range)	mean K_d for temperate region sediment (*14*) (range)
Ni	1×10^5 (1×10^4–2×10^5)	1×10^5 (2×10^4–5×10^5)
Co	1×10^5 (1×10^3–7×10^5)	2×10^5 (2×10^4–1×10^6)
Sr	1×10^2 (4×10^0–3×10^2)	1×10^3 (1×10^2–5×10^3)
Ru	3×10^4 (2×10^4–4×10^4)	3×10^2 (1×10^2–3×10^3)
Sb	2×10^3 (2×10^2–2×10^4)	1×10^3 (2×10^2–5×10^3)
I	4×10^1	2×10^1 (5×10^0–1×10^2)
Cs	1×10^3 (1×10^2–2×10^4)	3×10^3 (1×10^2–2×10^4)
Ce	9×10^6	2×10^6 (1×10^5–1×10^7)
Eu	7×10^5 (8×10^4–6×10^6)	5×10^5 (1×10^5–2×10^6)
Pb	5×10^5 (5×10^4–5×10^6)	2×10^5 (5×10^4–1×10^6)
Pu	1×10^5 (8×10^4–2×10^5)	1×10^5 (1×10^4–1×10^6)
Am	1×10^6 (1×10^5–3×10^6)	2×10^6 (1×10^5–2×10^7)

isms. In a study of brown macroalgae (*Fucus vesiculosus*), some elements, particularly essential metals (Co, Mn, Zn), were bioconcentrated to a greater extent at 12 °C than at 2 °C (*8*). In the clam *Macoma balthica* the assimilation efficiency of ^{241}Am from ingested phytoplankton was found to be significantly lower at 2 °C than at 12 °C (*5*). In the sea star *Asterias forbesi* the assimilation efficiency and retention of Am and Co following ingestion of clams were significantly greater at 2 °C than at 12 °C, but temperature had no effect on uptake or retention of these elements or Cs from the dissolved phase (*6*). In the brittle star *Ophiothrix fragilis* metals were accumulated at a slower rate from the dissolved phase at 2 °C than at 12 °C, but efflux rates were comparable at the two temperatures (*7*). While questions still remain about the real effect temperature has in regulating radionuclide uptake and retention, aside from some exceptions temperature does not appear to substantially affect equilibrium concentrations factors of radionuclides in nature.

The sediment K_d values for different radionuclides and elements are presented in Table 3. The K_d values varied less between locations than between elements, suggesting that the geochemical characteristics of the sediments had less influence on the partitioning of the elements than did properties of the elements themselves. However, because different regions within coastal seas, including the Kara Sea, typically have sediments with different chemical composition, it is expected that K_d values should vary spatially and perhaps temporally in areas which become anoxic seasonally. Consequently, it is appropriate to consider ranges of K_d values as well as mean values in radionuclide transport models and risk assessment analyses. Mean K_d values are compared with mean values for temperate waters in coastal sediments in Table 4. For most elements, these values are within a factor of 2 or 3 of each other, indicating that there is nothing unique about Kara Sea sediments with respect to metal partitioning and that the sediments had sufficient time to equilibrate with the released radionuclides. However, mean K_d values in the Kara Sea were about 1 order of magnitude lower for Sr and 2 orders of magnitude higher for Ru than those reported for temperate systems (Table 4). This may, however, reflect the very limited data currently available for Sr and Ru in Kara Sea sediment. It is unlikely that polar and temperate regions would display differences in equilibrium partitioning of metals to their sediments, particularly because bottom waters even in temperate regions are often ≤4 °C.

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