



Mobilization of Radionuclides from Sediments: Potential Sources to Arctic Waters

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INTRODUCTION

Contaminated soils and sediments can act as secondary sources of radionuclides to Arctic waters. In cases where the original source of contamination has ceased or been greatly reduced (e.g., weapons' testing, waste discharges from Mayak and Sellafield) remobilization of radionuclides from previously contaminated sediments increases in importance. With respect to Arctic waters, potential secondary sources include marine sediments contaminated by weapons' testing (Aarkrog 1993; JRNC 1994), by discharges from nuclear installations to seawater, e.g., the Irish Sea (Fjellidal *et al.*, 1995), or by leakages from dumped waste containers (JRNC 1994; Børretzen *et al.*, 1995; Salbu *et al.*, 1995). The major land-based source is run-off from soils and transport from sediments in the catchment areas of the Ob and Yenisey rivers, including those contaminated by Mayak discharges. Remobilization of radionuclides is often described as a secondary source of contamination. Whereas primary sources of man-made radionuclides tend to be point sources, secondary sources are usually more diffuse.

Radionuclides can be transported from the sediment to water phase by chemical, physical and biological processes (Fig. 1). Chemical mobilization includes ion-exchange, leaching and dissolution; physical transport can occur due to natural or anthropogenic resuspension of the sediments, e.g., during flooding, erosion or estuary dredging. Biological processes can effect both chemical and physical (bioturbation) mobilization. The input of radionuclides to the Kara Sea through the Ob-Yensiey estuaries includes ionic species that have been chemically leached from high activity soils in the Upper Techa river (Tronstad *et al.*, 1995), as well as physical transport of contaminated suspended sediments.

The radionuclide speciation and interaction with sediment components will determine the degree to which remobilization of radionuclides can influence levels in other parts of the ecosystem. Transfer fluxes (Bq yr^{-1}) will be a function of both the amount of radionuclides in sediments and the kinetics of transformation processes. With this in mind, the aim of the work presented in this paper was to investigate radionuclide (^{137}Cs and ^{90}Sr) association with sediments and to study possible processes leading to mobilization and transfer to Arctic waters. Investigations include experiments on the transfer of radionuclides from both freshwater sediments and marine sediments to seawater.

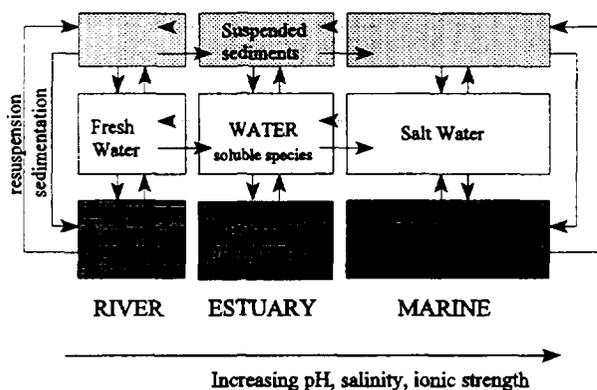


Figure 1: Sources of radionuclides from sediments to river, estuarine and marine waters.

MATERIALS AND METHODS

Experiments were carried out on marine (Kara Sea, Irish Sea, Stepovogo and Abrosimov Fjords), estuarine (Ob-Yenisey) and dirty ice sediments. Total ^{137}Cs and ^{90}Sr concentrations were determined using standard radiochemical techniques. Tracer studies using ^{134}Cs and ^{85}Sr were used to investigate the kinetics of radionuclide adsorption and desorption. All experiments were carried out under aerobic conditions at 4°C , using fresh, wet sediments. The water used in the tracer studies was either Oslo Fjord seawater (salinity 34 psu, pH 8.2) or Akerselv river water (pH 5.4), primarily because site-specific waters were unavailable for most sites. It must be stressed that tracer experiments should be seen as operationally-defined investigations into processes and controlling parameters, rather than an attempt to mimic natural conditions.

Mobilization from Freshwater Sediments

The potential for mobilization of radionuclides from sediments following transfer from a freshwater to a saltwater environment was studied. Labelled river waters (20 ml) were added to Ob and dirty ice sediment samples (equivalent to ca. 1 g dry weight). After 1 week contact time, distribution coefficients (K_d ml g^{-1}) were measured in fresh water (3 x 20 ml extraction) and after transfer to saltwater (3 x 20ml extraction). Extractions were carried out sequentially: labelled sediments were shaken with water for 1 hour, then separated by high-speed centrifugation.

Sorption and Mobilization from Marine Sediments

Rates of change in sediment-water K_d (ml g^{-1}) were measured as a function of contact time in static seawater-sediment tank studies. Radionuclide mobilization from sediment components was studied using sequential extraction after different contact time of labelled seawaters. The extraction agents included: seawater; 1M NH_4Ac (pH 7); 1M NH_4Ac (pH 5); and 7M HNO_3 . In samples having high enough "natural" contamination levels (i.e., Irish Sea and Stepovogo) the distribution of ^{137}Cs and ^{90}Sr between sediment extraction fractions was compared with that of the ^{134}Cs and ^{85}Sr tracers.

RESULTS AND DISCUSSION

Mobilization from Freshwater Sediments and Soils

Run-off from contaminated sediments and soils through the Ob-Yenisey estuaries represents a source of contamination in Arctic waters (JRNC, 1993; JRNC, 1994). During "normal" conditions the transfer of both ^{137}Cs and ^{90}Sr with suspended sediments is not thought to represent a significant fraction of the activity inputs to the Kara Sea. However, two main sources of sediment transport can occur under episodic events, namely, "dirty" ice (i.e., ice containing sediments) and suspended sediments transported with flood waters. In both cases, freshwater sediments are transported into marine waters, hence are subject to changes that can influence mobility of radionuclides associated with sediments. Preliminary analysis of ^{90}Sr in surface sediments from the Ob-Yenisey area indicated enhanced levels in Ob river sediments and in sediment samples taken from dirty ice 600 km NNW of the estuary mouth (up to 41 Bq/kg). These levels are higher than the average for open Kara Sea sediments (< 2 Bq/kg) (JRNC 1993).

Experiments using Ob and dirty ice sediments labelled with ^{134}Cs and ^{85}Sr tracers showed that K_d varied enormously between freshwater and seawater: K_d in freshwater was up to factor of 1000 higher than in seawater (Fig. 2.). The mobilization of ^{134}Cs and ^{85}Sr after transfer of sediments from freshwater to seawater may reflect the effect of increased pH, ionic strength, salinity and/or concentration of exchangeable elements (i.e., alkali or alkaline earth metals). Previous studies with tracers showed that reducing the pH of seawater decreased K_d for both ^{134}Cs and ^{85}Sr , so pH is unlikely to be controlling factor (JRNC, 1993). In the case of ^{85}Sr , high stable Sr concentrations in seawater (8 mg/l) will promote mobilization through isotopic exchange. Of course if Sr is not in an exchangeable form (e.g., carbonate forms, or fuel particles), mobilization may not be so extensive.

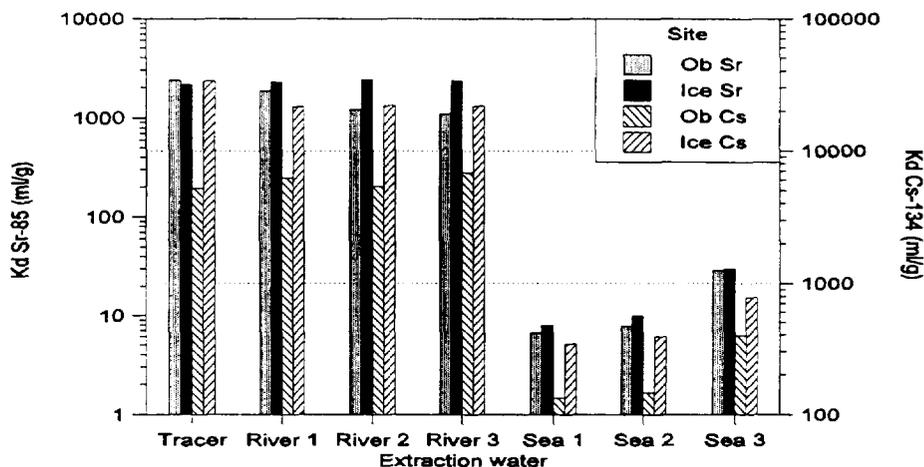


Figure 2. Change in K_d following sequential extraction (1 hr) of labelled Ob and Dirty ice sediments (1 wk contact time) with freshwater and saltwater ($n=3$, STD < 5%). Ob samples collected 400 km upstream from estuary mouth, Dirty ice collected 600 km N from estuary mouth

Sorption and Mobilization from Marine Sediments

In Arctic waters, radionuclides can be transferred from the water phase to bottom sediments by a number of processes, including sorption of ionic species, aggregation of colloids and precipitation of mineral particles or biological materials. For many radionuclides, sediments represent the major sinks in the ecosystem, the distribution between water and sediments being determined by the physical and chemical properties of the radionuclide, the water and the sediment. Tracer uptake studies to sediments showed that K_d for ^{85}Sr was much lower than for ^{134}Cs , 11 ± 2 and 950 ± 350 , respectively, after 1 month contact time. Both static and shaken systems indicated that sediment type influenced the degree and rate of ^{134}Cs fixation (Fig. 3). K_d for ^{134}Cs appeared to be correlated with sediment cation exchange capacity and loss on ignition (organic content). Gentle shaking increased binding rates of ^{134}Cs by a factor of 10 compared to the static experiments. However, after 1 month contact time, there was reasonable agreement between K_d in static and shaken water-sediment systems.

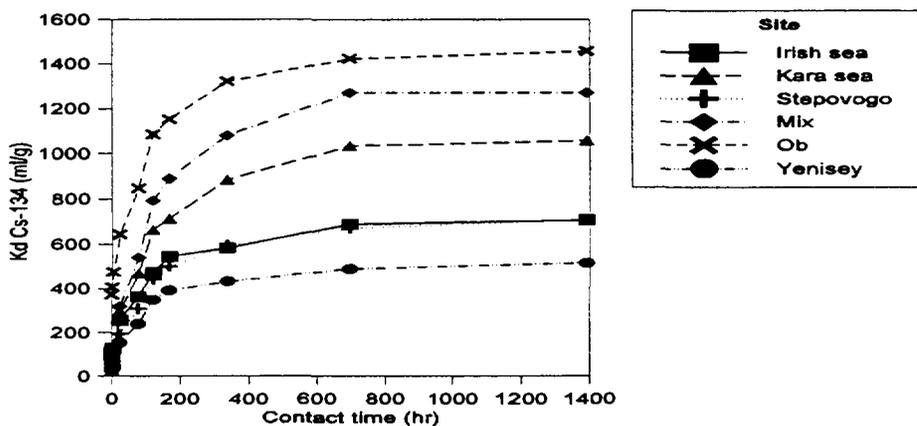


Figure 3: Change in K_d for ^{134}Cs as a function of contact time in static seawater-sediment system (ca. 10g dry weight sediment, added wet, to 2000 ml seawater).

Sequential extraction studies showed that ^{85}Sr was easily extracted from sediments, whereas ^{137}Cs was rapidly and strongly fixed to sediment components (Fig. 4). Comparison of tracer distributions with results from extraction studies on Irish Sea and Stepovogo sediments gave very similar distributions for ^{137}Cs and the ^{134}Cs tracer. However, ^{90}Sr was rather more strongly-bound to the sediment than ^{85}Sr , showing a significantly higher extraction into NH_4Ac (pH 5) than was observed for the tracer. This could reflect a time-dependent sorption reaction, e.g., incorporation of ^{90}Sr in the carbonate or amorphous fraction in marine sediments. Differences between the tracer and the "natural" ^{90}Sr may also indicate different source terms (i.e., non-ionic for the ^{90}Sr) or different removal mechanisms to sediments (e.g., biological cycles rather than ion-exchange).

In conclusion, ^{90}Sr is much less strongly bound to marine sediments than ^{137}Cs , and can be chemically mobilized through ion exchange with elements in seawater. Radiocaesium is strongly and rapidly fixed to sediments. Discharges of ^{137}Cs to surface sediments (i.e., from dumped containers) would be expected to be retained in sediments to a greater extent than discharges to sea-waters. Physical mobilization of sediments, for example resuspension, may be of more importance for transport of ^{137}Cs than for ^{90}Sr .

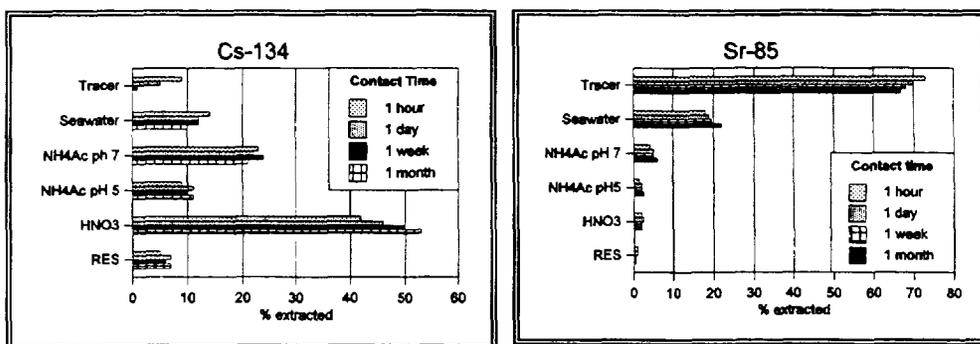


Figure 4. Tracer study showing extractability of ^{134}Cs and ^{85}Sr tracers associated with marine sediment as a function of contact time (c.a. 0.5 g dry weight to 20 ml seawater, shaken for 30 mins then stored at 4°C). The sediment sample was collected 300 km North of the Ob-Yenisey estuary mouths.

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REFERENCES

- Aarkrog, A. 1993. Radioactivity in polar regions - main sources. In: P.Strand and E.Holm (eds): Environmental Radioactivity in the Arctic and Antarctic. NRPA: Østerås, 15-33.
- Børretzen, P., H. Lien, H. Fjellidal, D.H. Oughton, B. Salbu. 1995. Mobility of radionuclides in sediments from Abrosimov and Stepovogo Fjords. In: Radioactivity in the Arctic. This meeting.
- Fjellidal, H., H. Lien, D.H. Oughton, B. Salbu. 1995. Mobility of Radionuclides in Irish Sea sediments. In: Radioactivity in the Arctic. This meeting.
- Joint Russian-Norwegian Collaboration (JRNC). 1993. A survey of artificial radionuclides in the Kara sea. Results from the Russian-Norwegian 1992 expedition to the Barents and Kara seas. NRPA: Østerås.
- JRNC. 1994. Radioactive contamination at dumping sites for nuclear waste in the Kara Sea, Results from the Russian-Norwegian expedition to the Kara Sea. NRPA: Østerås.
- Salbu, B., A.I. Nikitin, P. Strand, G.C. Christensen, V.B. Chumichev, B. Lind, H. Fjellidal, T.D. Selnæs, A.L. Rudjord, M. Sickel, N.K. Valetova and L. Føyn. 1995. JRNC on radioactive contamination from dumped nuclear waste in the Kara Sea - Results from the 1994 expedition to Abrosimov and Stepovogo Fjords. In: Radiactivity in the Arctic. This meeting.
- Trondstad, E., A. Nøren, D.H. Oughton, L. Skipperud, B. Salbu, A.P. Clacher, G.N. Romanov, P. Strand, and G.C. Christensen 1995. Mobilization of ^{90}Sr and other radionuclides from Mayak and Chernobyl soils and sediments. In: Radioactivity in the Arctic. This meeting.