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This report describes results of field and laboratory studies focusing on the Ob River system. Analyses of suspended particles collected in 1994 in the fresh water portion of the system show the presence of radionuclides (Pu, <sup>237</sup>Np) derived from nuclear fuel reprocessing. Measurements of <sup>129</sup>I in "dissolved" (<0.2 μm) colloidal (1000 NMW - 0.2 μm) and truly dissolved (<1000 NMW) fractions of river water shows that colloidal <sup>129</sup>I represents 4-6% of the "dissolved" fraction in the Ob and 20% in the Taz River. Both "dissolved" <sup>129</sup>I and dissolved organic carbon concentrations decrease towards the Kara Sea, possibly due to colloidal aggregation. Estimates of the <sup>129</sup>I flux to the Kara Sea range from 0.3 - 1 x 10<sup>24</sup> atoms/y. This is equivalent to 0.1 - 0.6%/y of the total <sup>129</sup>I emitted from La Hague and Sellafield over 25 years. Laboratory measurements of radionuclide distribution coefficients (Kd) show that Kd's decrease in the order Am>Co>Cs>I, with values ranging from 2 x 10<sup>5</sup> for Am to 1 x 10<sup>2</sup> for I. The Kd's for Am are strongly affected by the presence of DOC in the Ob River, with two orders of magnitude lower values in the presence of DOC.

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## **FINAL REPORT**

**Transport and fate of radionuclides in the Ob River estuarine system**

**ONR Grant # N000149410**

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## I. OBJECTIVES AND APPROACH

The Ob river system in western Siberia drains into the Kara Sea and may represent an important pathway for the delivery of anthropogenic radionuclides to the Arctic. The Ob system has received radioactive contaminants from global fallout from nuclear weapons testing, local fallout from Novaya Zemlya and perhaps most significantly, discharges from nuclear facilities sited near the Techa, Iset and Tom rivers, which drain into the Ob. In particular, releases of radioactivity from the MAJAK installation near Cheliabinsk and from the Siberian chemical plant Tomsk-7 have contributed anthropogenic radionuclides ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{60}\text{Co}$ , and others) to the Ob and its tributaries. Our research is focusing on the fate of some of these released radionuclides and the possibility of their ultimate transport to the Kara Sea.

We have conducted an integrated field and laboratory study to better understand the concentrations, transport pathways, geochemical fate and biological uptake of anthropogenic radionuclides in the Ob estuarine system. Our focus is on the water column because the transport and fate of particle-reactive radionuclides will be controlled by the transport of colloidal and suspended particles. In particular, colloidal particles (1-200 nm), due to their large surface area, are expected to be an especially effective mode of radionuclide transport in the low salinity region of the system. The aggregation of colloids at higher salinities will produce larger particles whose transport and deposition will differ from colloids. In addition, recent studies have provided new evidence that reactive metals (eg.  $^{241}\text{Am}$ ,  $^{239,240}\text{Pu}$ ) can exist to a significant extent in colloidal form, particularly in particle-rich nearshore and estuarine waters such as the Ob. This has important implications for modeling the transport and fate of anthropogenic radionuclide contaminants in shelf-estuarine waters and thus requires an accurate knowledge of their concentrations and physico-chemical state. Moreover, ion exchange effects as colloidal and suspended particles are transported from lower to higher salinity may be important in controlling the transport of radionuclides such as  $^{137}\text{Cs}$ . Our water column work has been done in conjunction with the field efforts organized in the Ob by Drs. Hugh Livingston and Fred Sayles of the Woods Hole Oceanographic Institution and complements their studies of bottom sediments.

## II. SAMPLE COLLECTION

### A. Station locations

In July-August, 1994, we participated in a cruise on the Ob organized by Drs. Livingston and Sayles. Samples for water, suspended particles, surface sediments, atmospheric aerosols, and aquatic biota, were taken at 12 stations in the Ob and Taz River-estuaries (see Fig. 1; Tables 1,2). These include: stations 1-6 in the Ob River proper; stations 8 and 10 in the Ob River delta; stations 11, 14 and 16 in broad portions of the Ob north of the Arctic Circle and station 13 in the northern end of the Taz estuary. The Taz River served as a control since there is no reported upstream source of artificial radionuclides. Total sample processing at each station was completed within 5-6 hours.

#### B. Large volume pumping sampling (LVPS)

Large volume pumping sampling was conducted at 10 stations. Surface water was pumped on deck through black garden hose and a 0.5  $\mu\text{m}$  prefilter, two  $\text{MnO}_2$  and two  $\text{Cu(Fe)CN}$  cartridges connected in series. All filters and cartridges were hand-carried back, to the lab and ashed in a muffle furnace.

#### C. Colloidal size-fractionated sampling using cross-flow filtration (CFF)

Cross-flow filtration was conducted at 8 stations. This served two purposes: 1) it provided samples of colloidal size-fractionated water, and 2) since the cartridge collection efficiency during LVPS is not well known for freshwater, and could possibly fail for scavenging of filter passing isotopes, the CFF sampling served as a necessary back-up.

Surface water was pumped using an electric Flotec pump through acid-cleaned Bev-a-line tubing connected to a 0.2  $\mu\text{m}$  Gelman Maxi capsule filter. The "dissolved" filter passing fraction ( $<0.2 \mu\text{m}$ ) was connected to a cross-flow filtration system with a 1,000 nominal molecular weight (NMW) spiral wound membrane (polysulfone) in a PVC housing. Cross-flow filtration was conducted in a recirculating mode resulting in collection of colloidal (1,000 NMW - 0.2  $\mu\text{m}$ ) and truly dissolved ( $<1,000$  NMW) size fractions. The wetted parts of the CFF system are PVC, silicon, teflon and polyethylene. The CFF system and filters were acid-leached prior to shipping. A new CFF filter and housing was used at each station in order to eliminate possible cross-contamination between stations. Also, CFF membranes will be acid-leached in our labs to determine any sorptive loss of radionuclides to the CFF filter during processing.

All of the 0.2  $\mu\text{m}$  Gelman filters were hand-carried back to the U.S. The 4-liter colloidal concentrate fractions (1,000 MW - 0.2  $\mu\text{m}$ ) were shipped back. For each station, all of the truly dissolved fraction ( $<1,000$  NMW) was collected in a single 60-liter barrel. These samples were shipped to the U.S. and are stored at the MSRC, SUNY-Stony Brook.

Subsamples (1 liter) of the size fractionated water and particulate matter were collected from this sampling scheme for determination of  $^{129}\text{I}$  at the IsoTrace Laboratory at the University of Toronto by Dr. L. Kilius.

#### D. Surface Sediments

Surficial sediments were collected at all stations using a small box core. Approximately 10 g of surface sediment was subsampled into plastic vials. All surface sediments were hand carried back to the U.S. The surface sediments have been analyzed for  $^{129}\text{I}$  by AMS at the IsoTrace Lab and for several artificial radionuclides by gamma counting. Pu isotope ratios were measured by isotope dilution mass spectrometry.

#### E. Plants and Biota

Samples of seaweed and one fish were collected for determination of  $^{129}\text{I}$  by AMS and other isotopes of interest. The plant and fish samples were hand carried back to the U.S.

#### F. Atmospheric Sampling

A small DC-powered atmospheric sampling pump was used to collect aerosol samples at 10 stations. The 47 mm filter samples were hand carried back for screening for trace metals,  $^{210}\text{Pb}$  and  $^7\text{Be}$ .

#### G. Ancillary Samples

All ancillary samples were hand carried back to the U.S. for analysis. Subsamples of colloidal size-fractionated water were collected for determination of a suite of trace metal contaminants by ICP-MS. Trace metals also will be analyzed in suspended particulate matter ( $>0.2\ \mu\text{m}$ ), large aggregates ( $>53\ \mu\text{m}$ ) and surface sediments. Trace metal sample collection and manipulations were conducted in a portable laminar flow-bench to reduce contamination. Subsamples were collected for determination of organic carbon in dissolved ( $<0.2\ \mu\text{m}$ ), colloidal (1,000 NMW -  $0.2\ \mu\text{m}$ ) and truly dissolved ( $<1000\ \text{NMW}$ ) fractions by high temperature combustion oxidation. Samples for particulate carbon and nitrogen were collected for CHN analysis. Dissolved nutrient ( $\text{PO}_4$ ,  $\text{NO}_3$ ,  $\text{NO}_2$ ,  $\text{SiO}_2$ ) samples were stored acidified and are being analyzed using standard colorimetric methods. The concentration of suspended particulate matter collected using preweighed  $0.4\ \mu\text{m}$  nuclepore filters has been determined gravimetrically.

### III. ANALYTICAL METHODS AND RESULTS

#### A. Large volume pumping

The prefilter cartridges from the large volume pumping retained  $\sim 10\text{--}50\ \text{g}$  of suspended sediment ( $>0.5\ \mu\text{m}$ ) for analyses. These cartridges were ashed at  $550^\circ\text{C}$  for 24 hours and analyzed for  $^{237}\text{Np}$ ,  $^{137}\text{Cs}$  and Pu isotopes, as follows:

$^{237}\text{Np}$  and Pu isotopes were analyzed on  $\sim 1\ \text{g}$  aliquots of the ashed sediment. These samples were supplied to Dr. T. Beasley of the Environmental Measurements Laboratory (New York, NY), who performed the radiochemical separations, followed by isotope dilution mass spectrometry. The latter was carried out by Dr. J. Kelley at Batelle Pacific Northwest Laboratories, (Richland, WA) according to procedures established at Batelle PNL. These data are reported in Tables 3 and 4.

Preliminary radiochemical analyses of Pu and  $^{137}\text{Cs}$  on the  $\text{MnO}_2$  and  $\text{Cu(Fe)Cn}$  impregnated cartridges respectively, that followed the pre-filters in the large volume samples showed poor retention and no further analyses of these fractions were attempted.

$^{137}\text{Cs}$  was measured by non-destructive gamma spectrometry of the cartridge ash using a low background intrinsic germanium detector. The detector was calibrated with NIST Standard Reference Material #1645 (River Sediment), measured in the same geometry as for the samples. The relatively high gamma activity of  $^{137}\text{Cs}$  makes sample self-absorption corrections unnecessary. The standard was measured before and after each group of samples, but not less often than once per month. The  $^{137}\text{Cs}$  activity are given in Table 5, as are  $^{239,240}\text{Pu}$  activities calculated from the  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  atom data (Table 3). Uncertainties are  $1\sigma$  counting errors.

#### B. Colloidal size-fractionated sampling

Aliquots of the fractions resulting from the cross-flow filtration of water samples were provided to Dr. L. Kilus of the Isotrace Laboratory, University of Toronto, for measurement

of  $^{129}\text{I}$  by accelerator mass spectrometry. The  $^{129}\text{I}$  data are given in Table 6. Aliquots of these fractions also were analyzed at Stony Brook for DOC using a Shimadzu 5000 TOC analyzer. DOC values in the colloidal (1000 NMW - 0.2  $\mu\text{m}$ ) and truly dissolved (<1000 NMW) were high and variable, suggesting contamination by the cross-flow filtration unit. Only the <0.2  $\mu\text{m}$  DOC values are reported here (Table 7). Uncertainty based on replicate measurements is estimated at  $\pm 2\%$ .

#### C. Surface sediments

Selected surface sediments were analyzed for Pu isotopes and  $^{129}\text{I}$  using the procedures described above. The data are reported in Tables 3, 4 and 6 respectively.

#### D. Ancillary samples

POC and PON were measured on glass fiber filter samples using a combustion method. The filters were dried at 60°C for 24 hours and fumed with concentrated HCl prior to CHN analysis. Quarter filter samples were combusted in a Carlo Erba EA 1108 Elemental Analyzer. Blank estimates were made using both clean filters and those subjected to HCl fuming. Precision is estimated to be  $\pm 5\%$ . The data are given in Table 7.

Suspended particulate concentrations (SPM) were determined by weighing the 0.4  $\mu\text{m}$  Nuclepore filters after drying to a constant weight. The SPM data also are presented in Table 7.

### IV. DISCUSSION OF FIELD DATA

#### A. Pu isotopes and $^{137}\text{Cs}$

Samples taken in 1994 came from three distinct areas of the Ob system: the river south of the Arctic Circle (stations 1-10), the wide portion of the river north of the Arctic Circle (stations 11, 14, 16) and the Taz River draining into the Ob (Station 13). The radionuclide results show geographic variation corresponding to these areas. The atom concentrations and activities in suspended sediments generally have higher values in the stations taken north of the Arctic Circle (Tables 3, 5), and although the data are limited, bottom sediments show a similar trend (Table 3). To a certain extent, these values can be affected by the nature of sediment in suspension, with grain size playing an important role. Both resuspension of bottom sediments and transport of suspended particles from upriver affect the nature of the suspended sediment reservoir at any given location in the river, and it is often easier to discern sources of radionuclides by looking at radionuclide ratios.

The atom ratios of  $^{237}\text{Np}/^{239}\text{Pu}$  and  $^{240}\text{Pu}/^{239}\text{Pu}$  show significant variations that address the sources of these radionuclides to the Ob (Figs. 2, 3). The  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio averages  $0.166 \pm 0.001$  ( $n = 6$ ) in stations in the Ob River proper (Stas. 1-10), increases to  $0.173 \pm .001$  in the delta region north of the Arctic Circle and is greatest ( $0.185 \pm .001$ ) in the Taz River. Values of this ratio from global fallout north of 60°N, as seen in soils, average  $\sim 0.185$ . The distinctly lower values in the Ob are evidence of radionuclide fuel reprocessing Pu with ratios as low as  $\sim 0.04$ , released from the facilities at Cheliabinsk or Tomsk up river.  $^{237}\text{Np}/^{239}\text{Pu}$  ratios also are higher in the Ob River proper (south of the Arctic Circle) and

decrease at Stations 11 and 13 (Table 4). These results provide clear evidence of radionuclides derived from fuel reprocessing in the suspended particle load of the Ob river.

#### B. $^{129}\text{I}$

Concentrations of "dissolved"  $^{129}\text{I}$  ( $<0.2\ \mu\text{m}$ ) range from  $0.65\text{--}2.81 \times 10^9$  atoms/l (Fig. 4). In particular,  $^{129}\text{I}$  levels are elevated furthest into the Ob ( $2.72 \times 10^9$  atoms/l) and gradually decrease toward St. 6 ( $2.33 \times 10^9$  atoms/l). In addition, DOC concentrations also decrease from the Ob River proper toward the estuary (Fig. 5) and are correlated with "dissolved"  $^{129}\text{I}$  ( $r = 0.90$ ,  $n = 10$ ). Levels of  $^{129}\text{I}$  in the river proper are significantly higher than values of  $\sim 0.8\text{--}1.4 \times 10^9$  atoms/l reported for the upper Kara Sea (Raisbeck et al., 1993). Within the delta at St. 10, a maximum is evident ( $2.81 \times 10^9$  atoms/l) and is associated with a maximum in sediment  $^{129}\text{I}$ , POC and SPM (Fig. 6, Table 7). Concentrations further decrease to values of  $1.53\text{--}1.67 \times 10^9$  atoms/l toward the estuary. The lowest "dissolved"  $^{129}\text{I}$  concentration ( $0.65 \times 10^9$  atoms/l) was determined on a sample collected from the Taz River (St. 13), which has no known upstream source.

The colloidal size sampling provides information on the partitioning of "dissolved"  $^{129}\text{I}$  in colloidal and truly dissolved fractions. In particular, the size-fractionated results indicate that  $^{129}\text{I}$  concentrations in the  $<1,000$  NMW fraction range from 64–85% of "dissolved" in the Ob River to 45% of "dissolved" in the Taz River (Fig. 4). Also, the distribution of the  $<1,000$  NMW fraction along the cruise track is similar to "dissolved"  $^{129}\text{I}$ . Concentrations of  $^{129}\text{I}$  determined in the  $1,000\text{--}0.2\ \mu\text{m}$  colloidal fraction range from  $0.084\text{--}0.17 \times 10^9$  atoms/l and are relatively invariant along the cruise track. As a percentage of the "dissolved" fraction, colloidal  $^{129}\text{I}$  represents  $\sim 4\text{--}6\%$  in the Ob and  $\sim 20\%$  in the Taz River. These results indicate that  $^{129}\text{I}$  was concentrated in the colloidal size fraction using cross-flow filtration and that  $\sim 4\text{--}20\%$  of "dissolved"  $^{129}\text{I}$  is partitioned on colloids.

$^{129}\text{I}$  levels in surface sediments range from  $0.64\text{--}10.24 \times 10^8$  atoms/g (Fig. 6). Sediment  $^{129}\text{I}$  concentrations are, on average,  $\sim 200$ -fold greater than "dissolved"  $^{129}\text{I}$  and are correlated with the concentration of suspended particulate matter ( $r = 0.75$ ,  $n = 12$ ) and particulate organic carbon ( $r = 0.81$ ,  $n = 12$ ). These results suggest that  $^{129}\text{I}$  is removed from the water column and deposited in the sediments by uptake on particles, presumably via active biological uptake or passive chemical scavenging.

The water column  $^{129}\text{I}$  results can be used to provide a first-order estimate of  $^{129}\text{I}$  output from the Ob River to the Kara Sea. For this calculation, we use "dissolved"  $^{129}\text{I}$  concentrations, which includes  $^{129}\text{I}$  in true solution and non-settling colloidal matter. Using the "dissolved"  $^{129}\text{I}$  concentration furthest into the Ob as an upper limit ( $2.7 \times 10^9$  atoms/l), the  $^{129}\text{I}$  concentration in the Taz River as a lower estimate ( $0.65 \times 10^9$  atoms/l), and the mean annual discharge of  $1.34 \times 10^4\ \text{m}^3\ \text{s}^{-1}$ , the  $^{129}\text{I}$  flux from the Ob River to the Kara Sea ranges from  $\sim 0.3\text{--}1 \times 10^{24}$  atoms/yr. On a yearly basis, the calculated  $^{129}\text{I}$  output from the Ob River is  $\sim 0.1\text{--}0.6\%$  of the total atoms emitted ( $5 \times 10^{27}$  atoms) from La Hague and Sellafield over 25 years. We estimate that 22% of the total  $^{129}\text{I}$  output from La Hague and Sellafield is now in

the Arctic Ocean and 13% in the Atlantic layer alone. Thus, on an annual basis, the  $^{129}\text{I}$  output from the Ob River corresponds to 0.6-2.5% and 1.1-4.2% of the total  $^{129}\text{I}$  emissions now present in the entire Arctic Ocean and in the Atlantic layer, respectively. We also note that these calculations do not consider temporal variability in  $^{129}\text{I}$  in the Ob River; for example, several samples collected in the Ob in 1993 indicate  $^{129}\text{I}$  concentrations 2-3 x lower than our results (G. Raisbeck, pers. comm.). In order to address this issue, and to improve the spatial coverage in the Ob, we are analyzing samples collected in 1995 to more accurately quantify the flux of  $^{129}\text{I}$  from the Ob River to the Arctic Ocean.

## V. LABORATORY MEASUREMENTS OF DISTRIBUTION COEFFICIENT (Kd)

The partition coefficients (Kds) of  $^{241}\text{Am}$ ,  $^{57}\text{Co}$ ,  $^{131}\text{I}$  and  $^{137}\text{Cs}$  were experimentally determined for sediments from the central Kara Sea basin and from six stations in the Ob River estuary. All values are presented in Table 8. Kara Sea sediment Kds are presented in Fig. 7. Note that the Kds decrease in the order  $\text{Am} > \text{Co} > \text{Cs} > \text{I}$ . Values range from  $< 1 \times 10^2$  for I to about  $1 \times 10^5$  for Am. Cs, of particular interest in the Kara Sea, has a Kd of about  $2 \times 10^2$ . The Kds of these radionuclides in the Ob sediments (determined using filtered Ob River water) are up to an order of magnitude lower, with highest Kds being about  $1 \times 10^4$  (at Stations 8 and 11) (Fig. 8). The Kds tend to be higher for the sediments with smaller grain size, and lowest in the sandiest sediment. We hypothesize that these lower Kds in the Ob are principally due to the extremely high concentrations of dissolved organic carbon in the Ob water (ca. 21 mM). By photo-oxidizing the DOC out of the Ob water, down to concentrations of about 8.1 mM, the Kds of Am increase by 2 orders of magnitude for Station 3 and over one order of magnitude for Station 11 (Fig. 9). The Kds for the other radionuclides are not appreciably affected by the DOC, with the exception of I in Station 11, where a modest increase (approximately 5-fold) in Kd is noted after the DOC is photo-oxidized. Thus, Am would be expected to show much lower Kds in the Ob than in marine systems and perhaps other river systems because the Ob River is enriched in dissolved organic matter which can apparently complex with the Am and reduce its particle reactivity.

This decline in particle reactivity of Am is also noted in the bioavailability of Am in Ob River water. The volume-volume concentration factor (VCF) of Am for the euryhaline diatom *Thalassiosira pseudonana* varies from approximately  $3 \times 10^4$  in filtered Ob water to about  $1 \times 10^6$  in filtered Ob water which had its DOC photo-oxidized (Fig. 10). Cs and Co are much less affected. The VCFs of I are extremely low and are not presented in Fig. 10.

Clearly, the high DOC will reduce the bioavailability as well as sediment availability of Am in the Ob (and probably other particle-reactive radionuclides which are subject to complexation by dissolved organic ligands), thereby enhancing its mobility and increasing its probability of being transported to the Kara Sea.

The effects of salinity on VCFs in this diatom are presented in Fig. 11. The VCFs of Am are not significantly affected by salinity over the range of 0 to 30 ppt. However, the VCF of Co decreases about one order of magnitude from 0 ppt to 30 ppt (Fig. 11). The VCF of Cs in the diatoms is as high as 100 at 0 ppt but decreases to 0 at salinities above 0 ppt. Thus, phytoplankton cells in the Ob River can concentrate these radionuclides out of the river water. When they are transported into the Ob estuary these algal cells will lose nearly all their Cs, some of their Co, and none of their Am to ambient water.



## **VI. STATISTICAL INFORMATION**

### **A. Publications**

Moran, S.B., J.K. Cochran, N.S. Fisher and L.R. Kilius. 1995.  $^{129}\text{I}$  in the Ob River. In: Proceedings of the Second International Conference on Environmental Radioactivity in the Arctic. Oslo, Norway, August 21-25, pp. 75-78.

Moran, S.B. and W.L. Woods. 1996. Cd, Cr, Cu, Ni and Pb in the water column and sediments of the Ob-Irtysh Rivers, Russia. Marine Pollution Bulletin (in press).

### **B. Presentations**

Cochran, J.K., Arctic Nuclear Waste Assessment Program Workshop, "Transport and fate of anthropogenic radionuclides in the Ob River system", Woods Hole, MA, May 1995.

Cochran, J.K., ANWAP Workshop on Risk Assessment, "Transport and fate of anthropogenic radionuclides in the Ob River system", Woods Hole, MA, May 1995.

Fisher, N., ANWAP Workshop on Risk Assessment, "Bioconcentration factors and sediment Kds' of long-lived radionuclides in the Ob River and Kara Sea", Sequim, WA, October 1995.

Fisher, N., ANWAP Workshop, "Transport and fate of anthropogenic radionuclides in the Ob River system", Snowbird, UT, May 1996.

### **C. Graduate Students supported**

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Ms. Wendy Woods, University of Rhode Island

Table 1: Ob River station locations and ancillary data.

| Station  | Date<br>(mm/dd/yy) | Location               | Water Depth<br>(m) | Sample Depth<br>(m) |
|----------|--------------------|------------------------|--------------------|---------------------|
| Ob-94-1  | 7/19/94            | 64°56.62'N, 65°40.24'E | 7                  | 3                   |
| Ob-94-2  | 7/20/94            | 65°15.64'N, 65°40.59'E | 6                  | 3                   |
| Ob-94-3  | 7/21/94            | 65°28.86'N, 65°40.66'E | 6                  | 3                   |
| Ob-94-4  | 7/22/94            | 65°47.38'N, 65°52.05'E | 6                  | 3                   |
| Ob-94-5  | 7/23/94            | 66°17.38'N, 66°12.05'E | 6                  | 3                   |
| Ob-94-6  | 7/24/94            | 66°38.35'N, 67°18.47'E | 4                  | 2                   |
| Ob-94-8  | 7/26/94            | 66°48.50'N, 69°26.27'E | 7                  | 2                   |
| Ob-94-10 | 7/28/94            | 66°47.31'N, 70°49.50'E | 5                  | 2                   |
| Ob-94-11 | 7/29/94            | 67°40.92'N, 72°56.87'E | 7                  | 3                   |
| Ob-94-13 | 7/31/94            | 69°05.63'N, 76°43.12'E | 10                 | 4                   |
| Ob-94-14 | 8/01/94            | 68°25.54'N, 73°49.88'E | 10                 | 4                   |
| Ob-94-16 | 8/02/94            | 66°51.54'N, 73°11.90'E | 6                  | 4                   |

Table 2 : Inventory of samples collected for the water column sampling program of the Ob River Expedition.

### Ob River 1994 - Sample Inventory

| Station No. | Sample Type | LVPS-Radionuclides |                       | CFF-Radionuclides |          |           | TM | Atm. | Nuts. | I-129 | DOC | POC | SPM |
|-------------|-------------|--------------------|-----------------------|-------------------|----------|-----------|----|------|-------|-------|-----|-----|-----|
|             |             | Pre*               | MnO <sub>2</sub> -A,B | CuFeCN-A,B        | >0.2 μm* | 1K-0.2 μm |    |      |       |       |     |     |     |
| 1           | water       | 1                  | 2                     | 2                 | 1        | 1         | 5  | 1    | 1     | 6     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 2           | water       | 1                  | 2                     | 2                 | 1        | 1         | 5  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 3*          | water       | 1                  | 2                     | 2                 |          |           |    | 1    | 1     |       |     |     |     |
|             | sediment    | 1                  |                       |                   |          |           |    |      |       | 1     |     |     |     |
| 4           | water       | 1                  | 2                     | 2                 | 1        | 1         | 5  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 5           | water       | 1                  | 2                     | 2                 | 1        | 1         | 5  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 6           | water       | 1                  | 2                     | 2                 | 1        | 1         | 5  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 8*          | water       | 1                  | 2                     | 2                 |          |           |    |      |       |       |     |     |     |
|             | sediment    | 1                  |                       |                   |          |           |    |      |       |       |     |     |     |
| 10          | water       | 1                  | 2                     | 2                 | 1        | 1         | 6  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 11          | water       | 1                  | 2                     | 2                 | 1        | 1         | 6  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |
| 13          | water       | 1                  | 2                     | 2                 | 1        | 1         | 6  | 1    | 1     | 5     | 3   | 1   | 1   |
|             | sediment    | 1                  |                       |                   |          |           | 1  |      |       | 1     |     |     |     |

## Ob River 1994 - Sample Inventory

[illegible]

Table 3: Radionuclide data for suspended and surficial bottom sediment of the Ob River (1994).

| Station               | $^{237}\text{Np}$<br>( $\times 10^7$ ) | $^{239}\text{Pu}$<br>( $\times 10^7$ ) | $^{240}\text{Pu}$<br>( $\times 10^6$ ) | $^{241}\text{Pu}^*$<br>( $\times 10^4$ ) | $^{242}\text{Pu}$<br>( $\times 10^5$ ) |
|-----------------------|--|--|--|--|--|
| -----atoms/g sed----- |  |  |  |  |  |
| Suspended sediment    |  |  |  |  |  |
| 94-1                  | $2.294 \pm 0.081$                      | $4.91 \pm 0.12$                        | $8.03 \pm 0.21$                        | -  | $1.28 \pm 0.18$                        |
| 94-2                  | $2.092 \pm 0.067$                      | $4.25 \pm 0.08$                        | $7.05 \pm 0.17$                        | $8.8 \pm 1.2$                            | $1.39 \pm 0.11$                        |
| 94-4                  | $2.703 \pm 0.085$                      | $5.92 \pm 0.12$                        | $9.80 \pm 0.23$                        | $14.8 \pm 1.2$                           | $1.98 \pm 0.12$                        |
| 94-5                  | $2.365 \pm 0.069$                      | $4.69 \pm 0.10$                        | $7.78 \pm 0.18$                        | $9.1 \pm 1.3$                            | $1.71 \pm 0.12$                        |
| 94-8                  | $4.72 \pm 0.14$                        | $11.0 \pm 0.2$                         | $18.5 \pm 0.4$                         | $27.5 \pm 1.4$                           | $3.95 \pm 0.18$                        |
| 94-10                 | $3.004 \pm 0.088$                      | $6.51 \pm 0.13$                        | $10.8 \pm 0.2$                         | $15.5 \pm 1.0$                           | $2.06 \pm 0.10$                        |
| 94-11                 | $13.10 \pm 0.45$                       | $39.4 \pm 1.0$                         | $68.2 \pm 1.8$                         | $101 \pm 4$                              | $13.7 \pm 0.5$                         |
| 94-13                 | $6.54 \pm 0.22$                        | $28.2 \pm 0.7$                         | $52.1 \pm 1.4$                         | $73.2 \pm 3.0$                           | $11.3 \pm 0.4$                         |
| Bottom sediment       |  |  |  |  |  |
| 94-1                  | $0.258 \pm 0.015$                      | $0.289 \pm 0.010$                      | $0.459 \pm 0.033$                      | -  | $<0.2$                                 |
| 94-5                  | $0.700 \pm 0.027$                      | $1.21 \pm 0.03$                        | $2.04 \pm 0.09$                        | -  | $<0.4$                                 |
| 94-10                 | $0.372 \pm 0.015$                      | $0.236 \pm 0.009$                      | $0.405 \pm 0.033$                      | -  | $<0.3$                                 |
| 94-13                 | $3.02 \pm 0.07$                        | $11.3 \pm 0.2$                         | $20.3 \pm 0.4$                         | $25.8 \pm 2.1$                           | $4.18 \pm 0.19$                        |

\*Decay corrected to 1 January 1995  
Uncertainties are  $\pm 1\sigma$

Table 4: Radionuclide atom ratios for suspended and surficial bottom sediments of the Ob River (1994).

| Station                   | $^{237}\text{Np}/^{239}\text{Pu}$ | $^{240}\text{Pu}/^{239}\text{Pu}$ | $^{241}\text{Pu}/^{239}\text{Pu}^*$<br>( $\times 10^{-2}$ ) | $^{242}\text{Pu}/^{239}\text{Pu}$<br>( $\times 10^{-2}$ ) |
|---------------------------|-----------------------------------|-----------------------------------|---|---|
| <b>Suspended sediment</b> |                                   |                                   |   |   |
| 94-1                      | $0.467 \pm 0.020$                 | $0.1636 \pm 0.0020$               | -   | $0.262 \pm 0.036$   |
| 94-2                      | $0.492 \pm 0.017$                 | $0.1657 \pm 0.0023$               | $0.206 \pm 0.027$   | $0.327 \pm 0.026$   |
| 94-4                      | $0.456 \pm 0.015$                 | $0.1654 \pm 0.0018$               | $0.250 \pm 0.019$   | $0.334 \pm 0.020$   |
| 94-5                      | $0.504 \pm 0.018$                 | $0.1657 \pm 0.0019$               | $0.194 \pm 0.028$   | $0.364 \pm 0.025$   |
| 94-8                      | $0.427 \pm 0.012$                 | $0.1679 \pm 0.0017$               | $0.249 \pm 0.012$   | $0.357 \pm 0.015$   |
| 94-10                     | $0.461 \pm 0.016$                 | $0.1660 \pm 0.0016$               | $0.238 \pm 0.014$   | $0.316 \pm 0.014$   |
| 94-11                     | $0.332 \pm 0.010$                 | $0.1731 \pm 0.0010$               | $0.258 \pm 0.008$   | $0.348 \pm 0.009$   |
| 94-13                     | $0.232 \pm 0.010$                 | $0.185 \pm 0.0011$                | $0.260 \pm 0.008$   | $0.401 \pm 0.011$   |
| <b>Bottom sediment</b>    |                                   |                                   |   |   |
| 94-1                      | $0.893 \pm 0.060$                 | $0.159 \pm 0.010$                 | -   | -   |
| 94-5                      | $0.578 \pm 0.026$                 | $0.169 \pm 0.006$                 | -   | -   |
| 94-10                     | $1.58 \pm 0.09$                   | $0.172 \pm 0.013$                 | -   | -   |
| 94-13                     | $0.267 \pm 0.008$                 | $0.180 \pm 0.001$                 | $0.228 \pm 0.018$   | $0.369 \pm 0.015$   |

\*Decay corrected to 1 January 1995

Uncertainties are  $\pm 1\sigma$

Table 5: Radionuclide activities and activity ratios for suspended sediments of the Ob River (1994).

| Station | $^{239,240}\text{Pu}^*$<br>(mBq/g) | $^{137}\text{Cs}$<br>(mBq/g) | $^{239,240}\text{Pu}/^{137}\text{Cs}$ |
|---------|------------------------------------|------------------------------|---------------------------------------|
| 94-1    | $0.072 \pm 0.002$                  | $1.70 \pm 0.20$              | $0.042 \pm 0.005$                     |
| 94-2    | $0.062 \pm 0.001$                  | $2.00 \pm 0.20$              | $0.031 \pm 0.003$                     |
| 94-4    | $0.087 \pm 0.002$                  | $2.40 \pm 0.30$              | $0.036 \pm 0.005$                     |
| 94-5    | $0.069 \pm 0.001$                  | $2.05 \pm 0.33$              | $0.034 \pm 0.005$                     |
| 94-8    | $0.163 \pm 0.004$                  | $1.70 \pm 0.20$              | $0.096 \pm 0.012$                     |
| 94-10   | $0.096 \pm 0.002$                  | $1.50 \pm 0.23$              | $0.064 \pm 0.010$                     |
| 94-11   | $0.587 \pm 0.009$                  | $15.5 \pm 1.5$               | $0.038 \pm 0.004$                     |
| 94-13   | $0.431 \pm 0.011$                  | $8.82 \pm 0.82$              | $0.049 \pm 0.005$                     |

\*Activities calculated from  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  atom concentration data (Table 2).  
All uncertainties are  $1\sigma$  errors.

Table 6: Water column and bottom sediment  $^{129}\text{I}$  results from the 1994 Ob River expedition.

| Station | Water column ( $10^9$ atoms/l)      |                           |  | Sediment<br>( $10^8$ atoms/g) |
|---------|-------------------------------------|---------------------------|--|-------------------------------|
|         | <0.2 $\mu\text{m}$<br>("Dissolved") | <1kD<br>(Truly dissolved) | 1kD - 0.2 $\mu\text{m}$<br>(Colloidal) |                               |
| 94-1    | $2.717 \pm 0.068$                   | $2.133 \pm 0.053$         | $0.102 \pm 0.009$                      | $0.982 \pm 0.088$             |
| 94-2    | $2.577 \pm 0.070$                   | $1.832 \pm 0.047$         | $0.165 \pm 0.012$                      | $3.333 \pm 0.081$             |
| 94-3    | -                                   | -                         | -                                      | $0.641 \pm 0.077$             |
| 94-4    | $2.506 \pm 0.079$                   | $1.832 \pm 0.047$         | $0.139 \pm 0.010$                      | $5.247 \pm 0.219$             |
| 94-5    | $2.535 \pm 0.128$                   | $2.149 \pm 0.055$         | $0.128 \pm 0.008$                      | $6.385 \pm 0.257$             |
| 94-6    | $2.333 \pm 0.093$                   | $1.827 \pm 0.051$         | $0.135 \pm 0.006$                      | $1.898 \pm 0.074$             |
| 94-8    | -                                   | -                         | -                                      | $1.248 \pm 0.062$             |
| 94-10   | $2.807 \pm 0.110$                   | $1.783 \pm 0.048$         | $0.127 \pm 0.008$                      | $10.241 \pm 0.345$            |
| 94-11   | $1.527 \pm 0.100$                   | $1.182 \pm 0.051$         | $0.084 \pm 0.006$                      | $0.778 \pm 0.086$             |
| 94-13   | $0.654 \pm 0.107$                   | $0.296 \pm 0.005$         | $0.130 \pm 0.004$                      | $3.845 \pm 0.144$             |
| 94-14   | $1.675 \pm 0.196$                   | -                         | -                                      | $3.005 \pm 0.271$             |
| 94-16   | $2.068 \pm 0.142$                   | -                         | -                                      | $7.242 \pm 0.227$             |



Table 7: Ob River suspended particle, DOC and POC concentrations (1994).

| Station | Suspended particle <sup>1</sup><br>concentration<br>(mg/l) | POC <sup>2</sup><br>(mmol/l) | PON <sup>2</sup><br>(mmol/l) | DOC <sup>3</sup><br>(mmol/l) |
|---------|--|------------------------------|------------------------------|------------------------------|
| 94-1    | 32.3   | 0.119                        | 0.015                        | 1.00                         |
| 94-2    | 52.9   | 0.153                        | 0.012                        | 1.03                         |
| 94-3    | 38.7   | 0.095                        | 0.012                        | -                            |
| 94-4    | 76.5   | 0.127                        | 0.014                        | 1.01                         |
| 94-5    | 64.7   | 0.147                        | 0.011                        | 1.02                         |
| 94-6    | 58.7   | 0.139                        | 0.013                        | 1.00                         |
| 94-8    | 35.4   | 0.082                        | 0.012                        | -                            |
| 94-10   | 174  | 0.234                        | 0.021                        | 0.95                         |
| 94-11   | 31.7   | 0.124                        | 0.014                        | 0.66                         |
| 94-13   | 33.1   | 0.077                        | 0.010                        | 0.53                         |
| 94-14   | 13.6   | 0.079                        | 0.009                        | 0.84                         |
| 94-16   | 65.1   | 0.173                        | 0.018                        | 0.98                         |

<sup>1</sup> >0.4  $\mu\text{m}$  (Nuclepore filters)

<sup>2</sup> Glass fiber filters

<sup>3</sup> <0.2  $\mu\text{m}$  (Gelman Maxi capsule filter)

Table 8: Kd values for sediment from different stations in the Ob River and Kara Sea.

| Location       | Am                | Co                | Cs                | I                 |
|----------------|-------------------|-------------------|-------------------|-------------------|
| Kara Sea       | $1.1 \times 10^5$ | $1.0 \times 10^4$ | $2.5 \times 10^2$ | $3.0 \times 10^1$ |
| Ob River*      |                   |                   |                   |                   |
| 94-1           |                   | $2.8 \times 10^3$ | $3.0 \times 10^2$ | $1.0 \times 10^2$ |
|                | $2.7 \times 10^3$ | $2.0 \times 10^3$ | $9.0 \times 10^2$ | $2.7 \times 10^2$ |
| 94-3 (+ DOC)** |                   |                   |                   |                   |
|                | $2.0 \times 10^5$ | $2.5 \times 10^3$ | $7.8 \times 10^2$ | $3.0 \times 10^2$ |
| 94-3 (- DOC)   |                   |                   |                   |                   |
| 94-5           | nm                | $5.2 \times 10^3$ | $1.2 \times 10^3$ | $2.1 \times 10^2$ |
| 94-8           | nm                | $1.2 \times 10^4$ | $8.0 \times 10^2$ | $2.5 \times 10^2$ |
| 94-11(+ DOC)   | $7.0 \times 10^3$ | $1.7 \times 10^4$ | $1.7 \times 10^4$ | $2.0 \times 10^3$ |
| 94-11 (- DOC)  | $2.1 \times 10^5$ | $1.3 \times 10^4$ | $1.4 \times 10^4$ | $4.0 \times 10^3$ |
| 94-13          | nm                | $6.0 \times 10^3$ | $3.0 \times 10^3$ | $1.7 \times 10^3$ |

\*Measurements made with bottom sediment collected at the stations indicated

\*\*+DOC = Kd measured in natural Ob River water; - DOC = Kd measured in UV-irradiated Ob River water

nm = no measurement

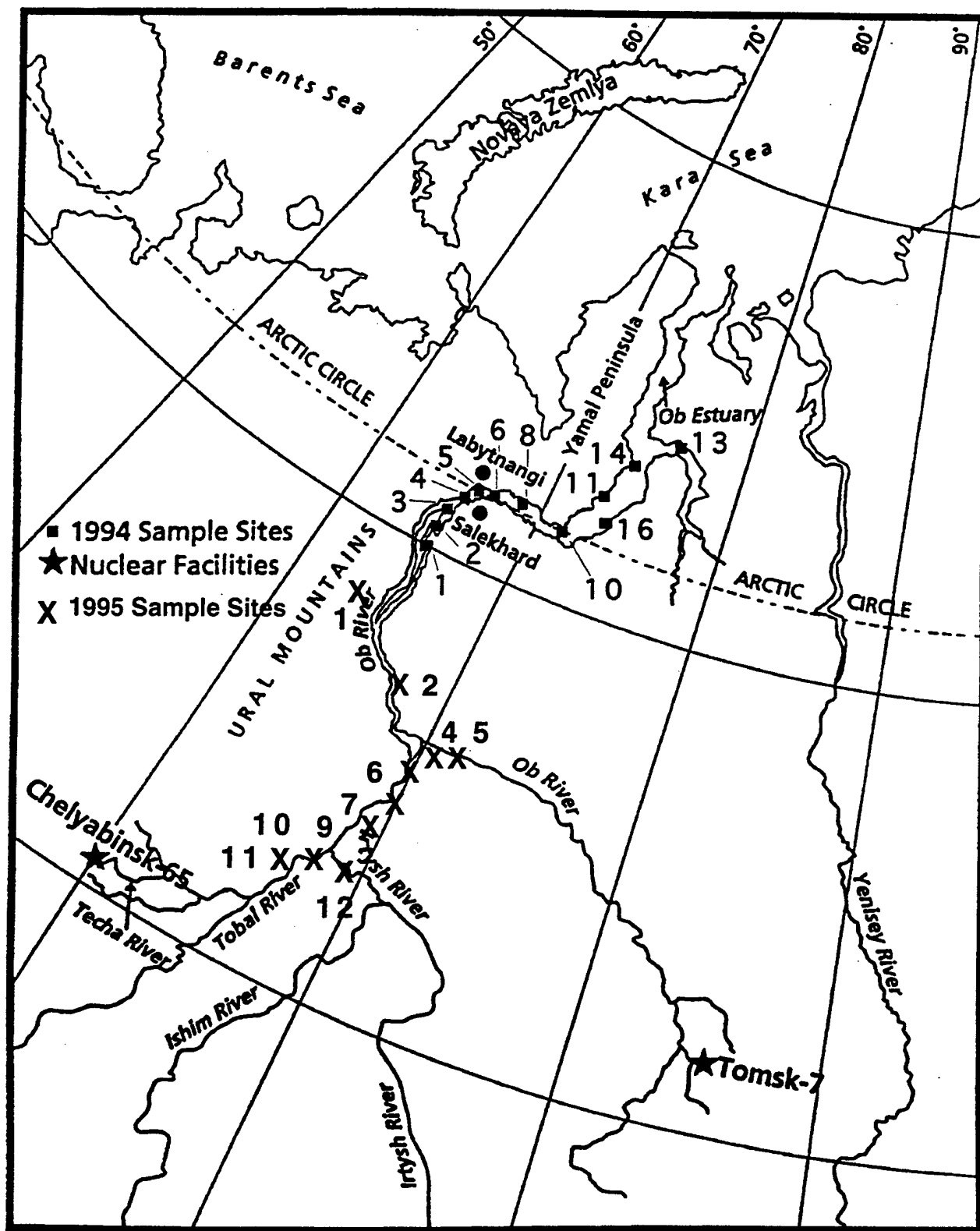


Fig. 1. Map showing stations sampled in the Ob River system. Stations marked with a filled square were sampled in 1994 and are discussed in this report. Samples indicated with an "x" were sampled in 1995 with continuation ANWAP funding.

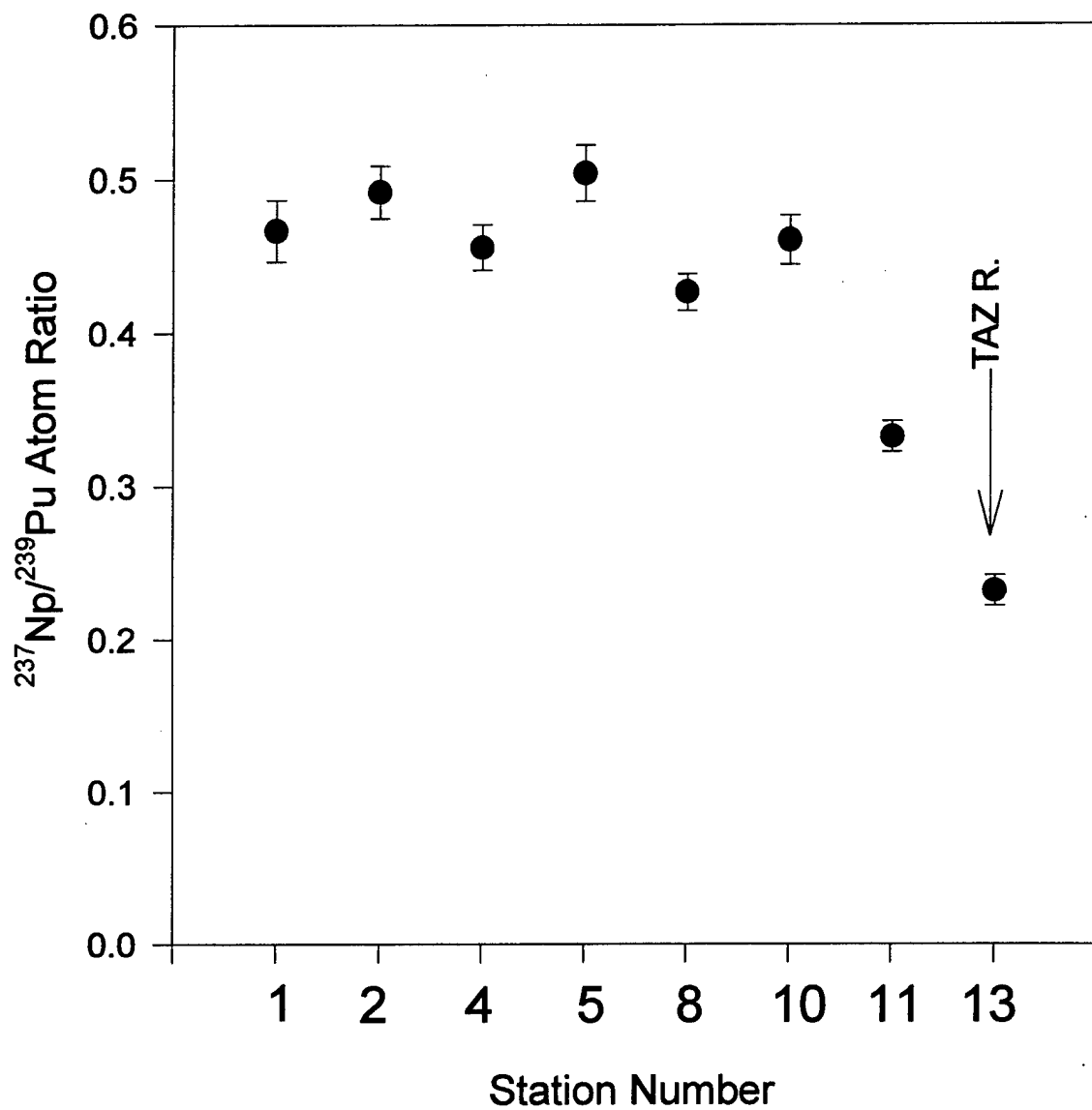


Fig. 2.  $^{237}\text{Np}/^{239}\text{Pu}$  atom ratio in suspended sediment ( $>0.5\ \mu\text{m}$ ) filtered from Ob River water in 1994.

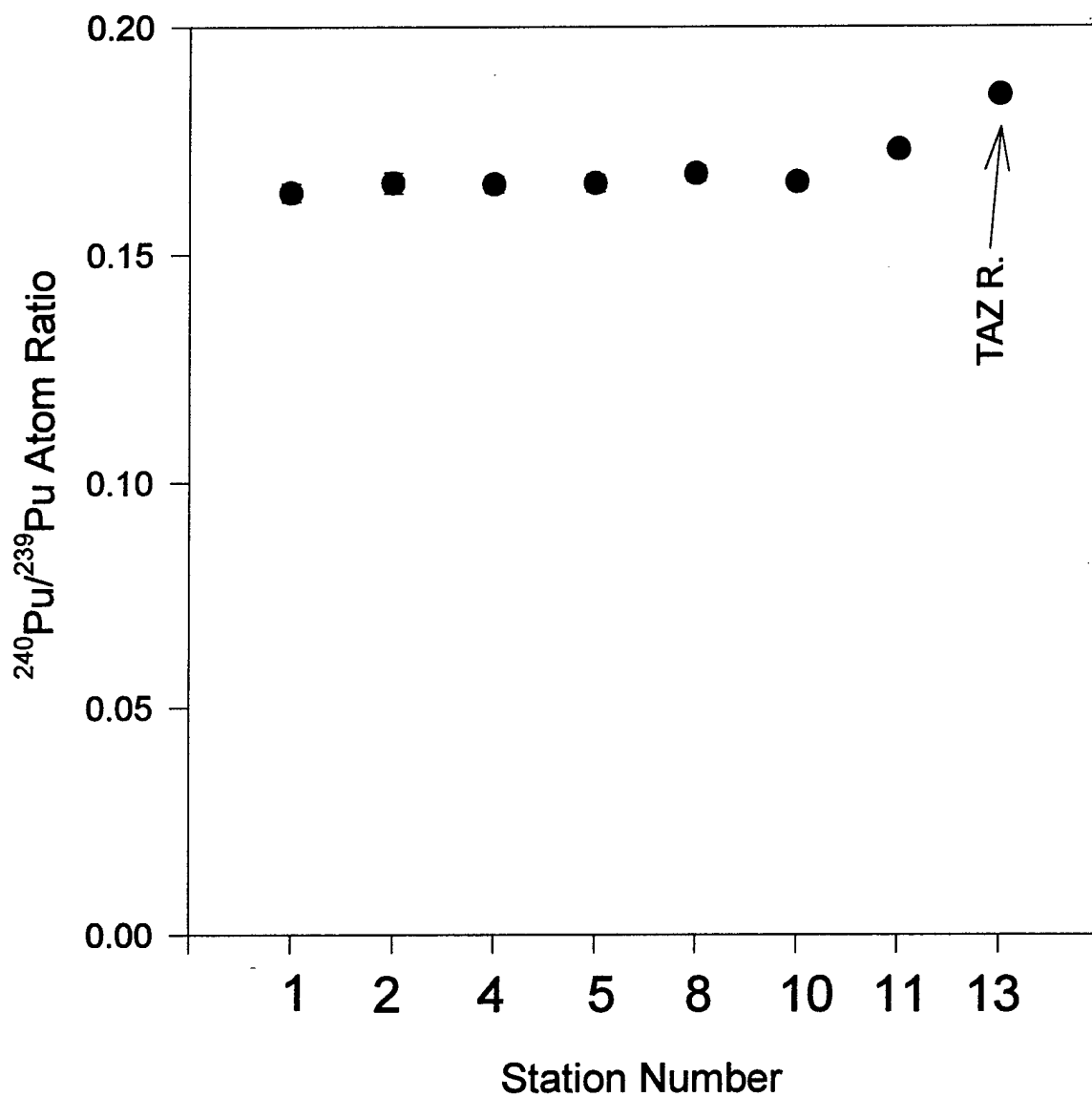


Fig. 3.  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio in suspended sediment ( $>0.5\ \mu\text{m}$ ) filtered from Ob River water in 1994.

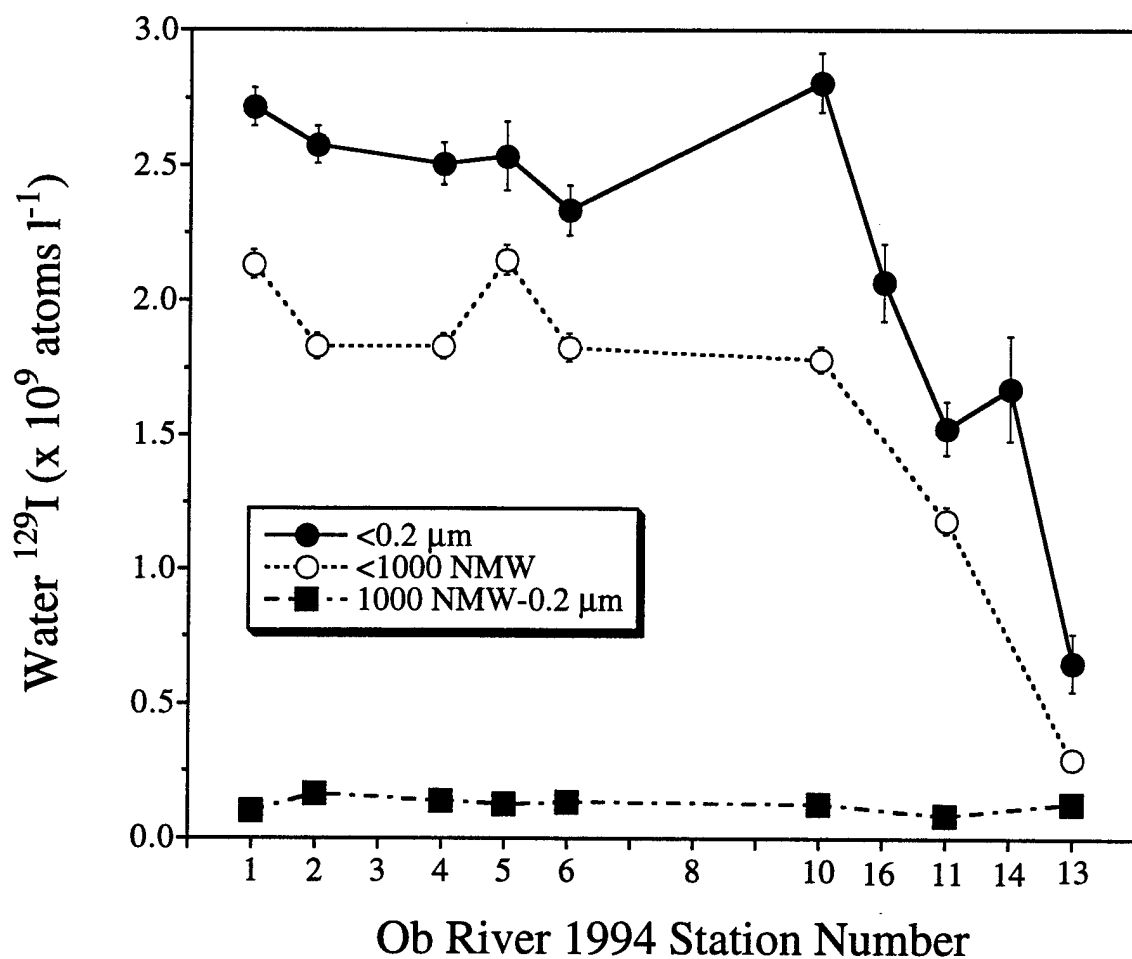


Fig. 4.  $^{129}\text{I}$  in size-fractionated samples of Ob River water collected in 1994. Samples were filtered through  $0.2 \mu\text{m}$  filters to collect "dissolved"  $^{129}\text{I}$  then through cross-flow filtration to separate colloidal  $^{129}\text{I}$  (1000 NMW -  $0.2 \mu\text{m}$ ) from truly dissolved ( $<1000$  NMW).

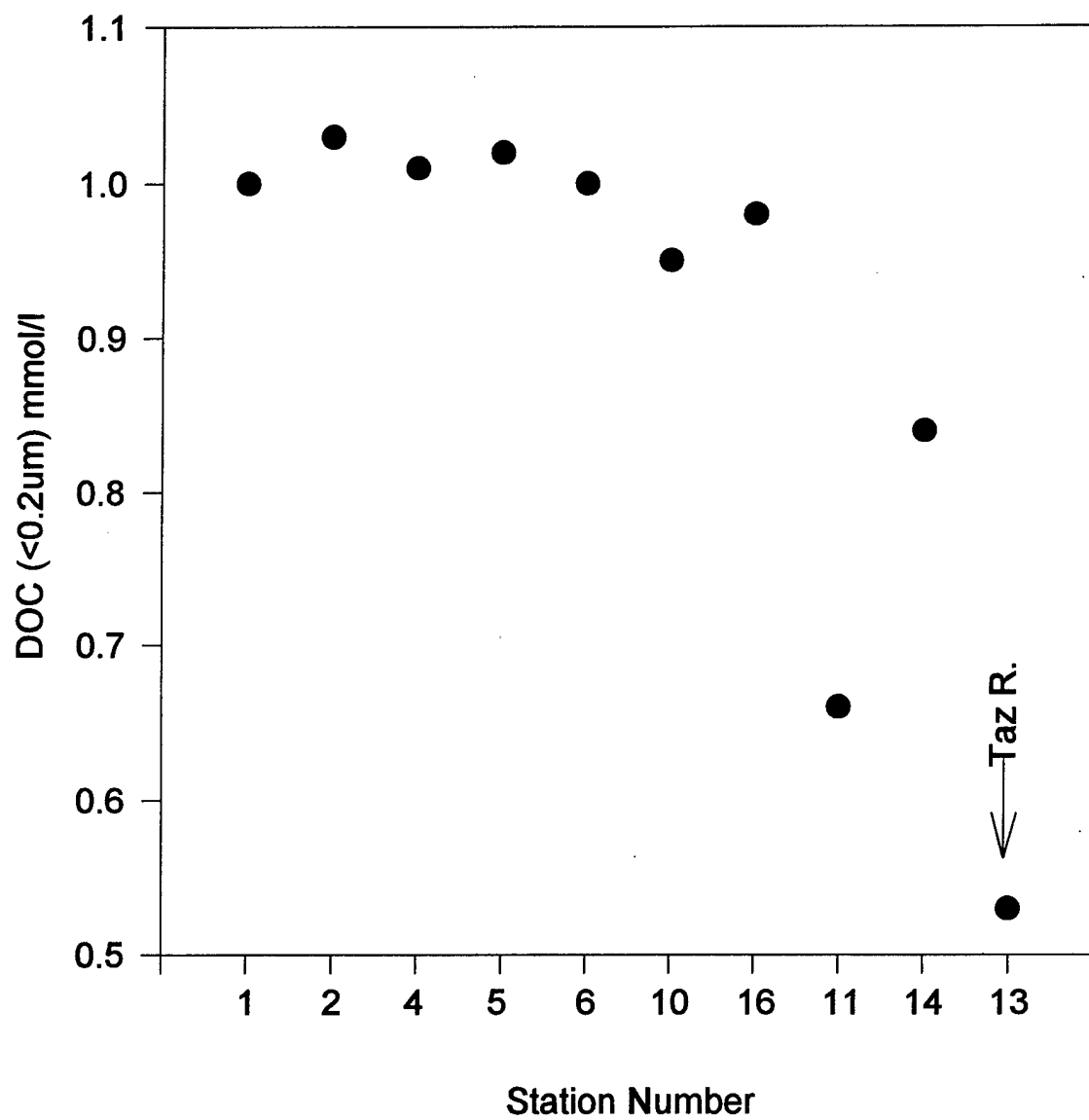


Fig. 5. Dissolved organic carbon (<0.2  $\mu\text{m}$ ) in the Ob River.

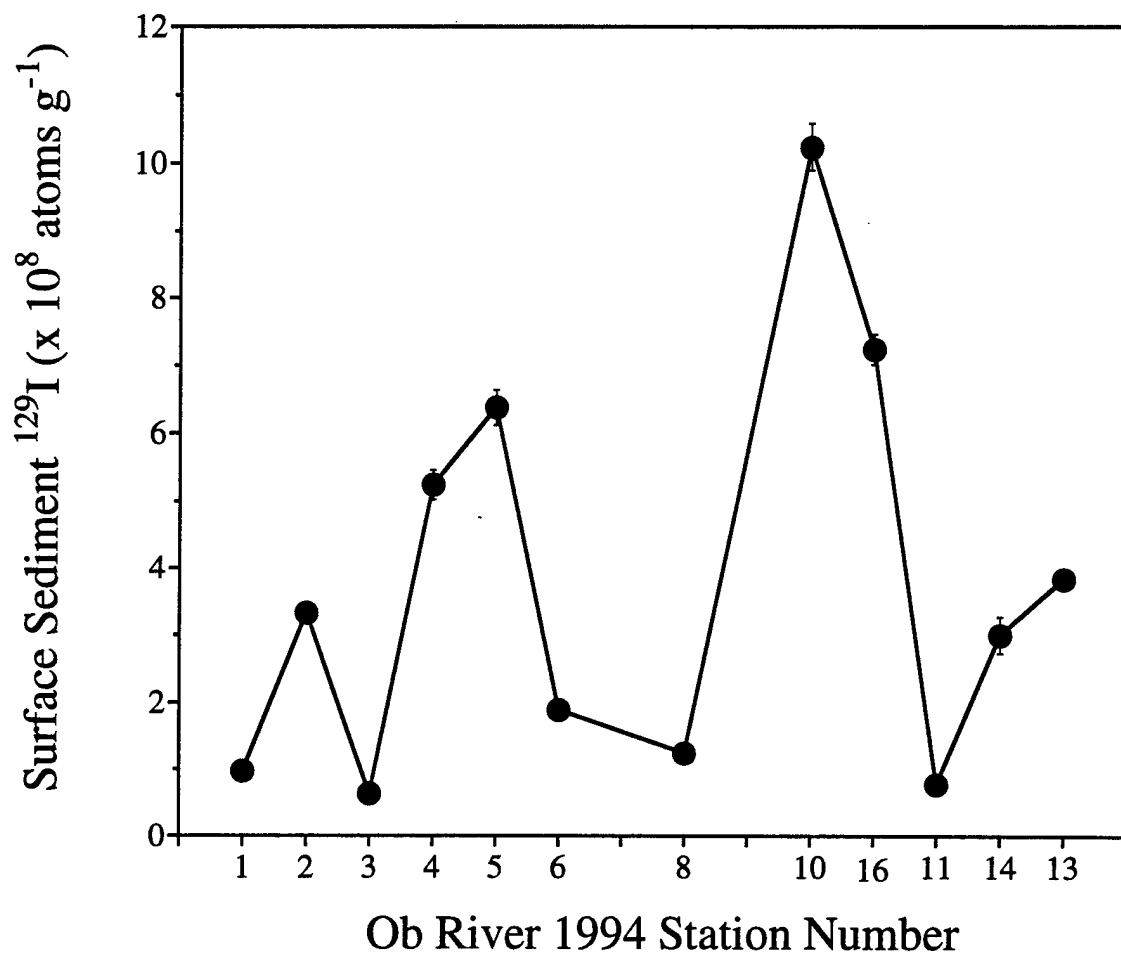


Fig. 6.  $^{129}\text{I}$  concentration in surficial bottom sediments collected in the Ob River in 1994.



# Kara Sea Kd Values

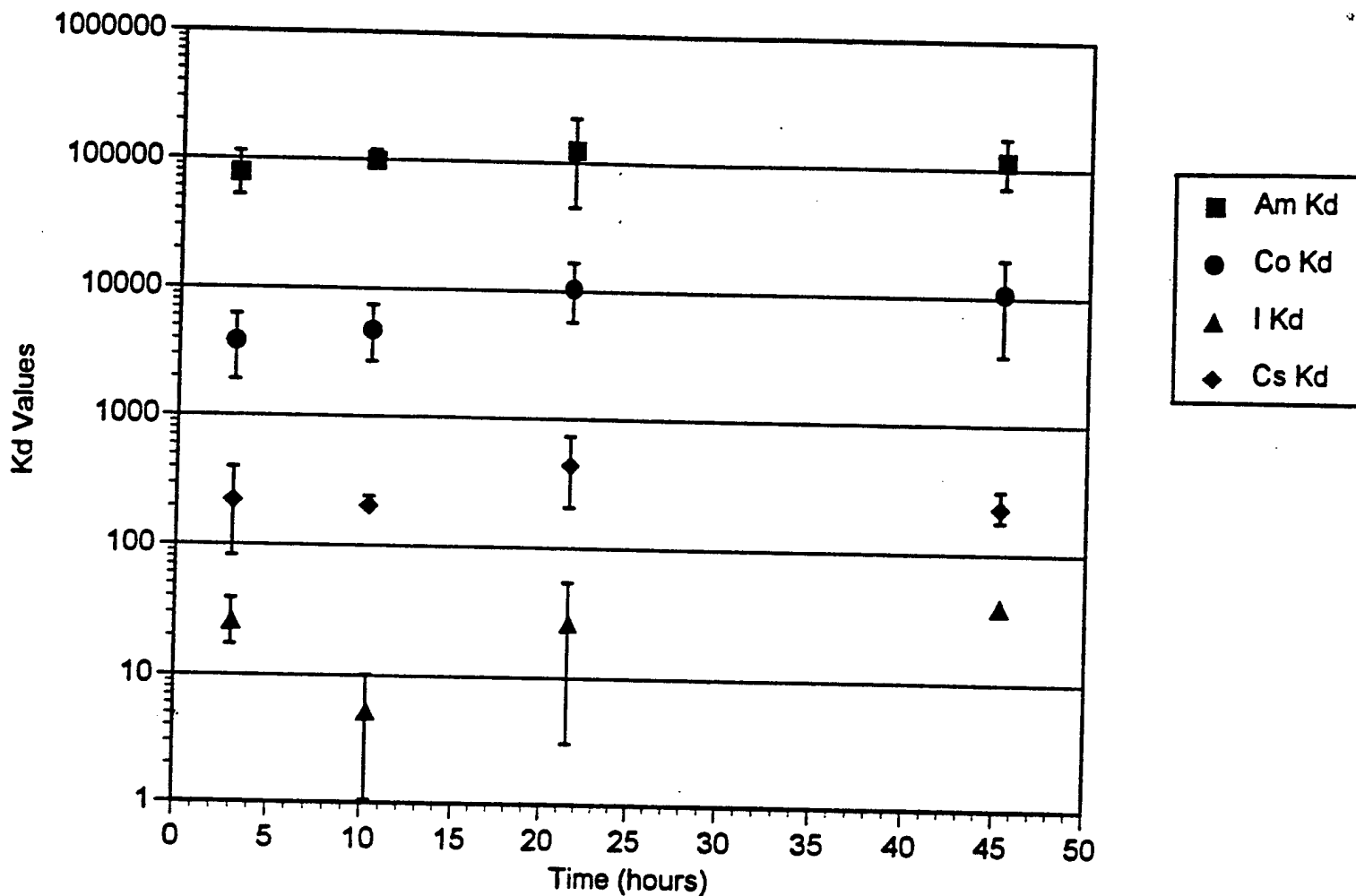


Fig. 7. Kd values of Am-241, Co-57, I-131, and Cs-137 for Kara Sea sediment from the central basin. Note that equilibrium was reached within one day.

# Ob River Kd Values

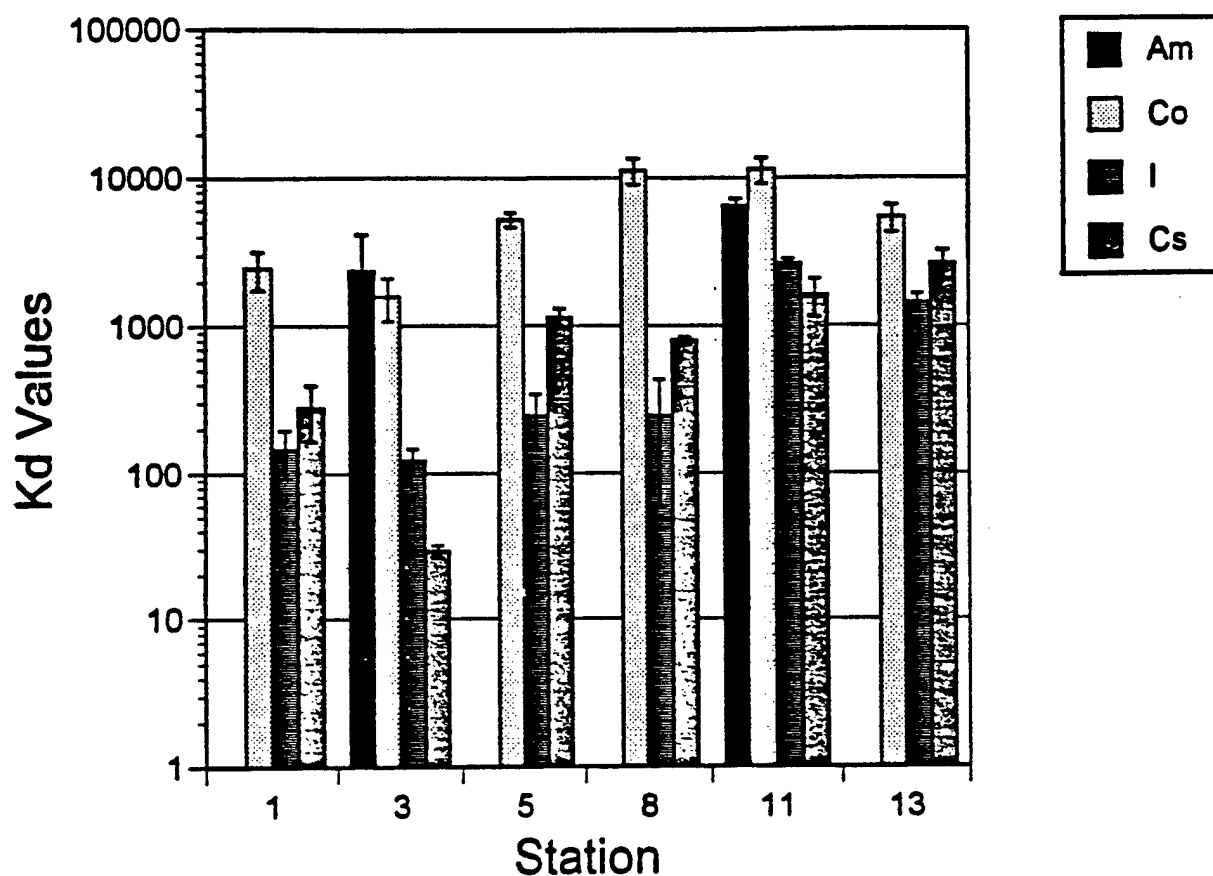


Fig. 8. Kd values of Am-241, Co-57, I-131, and Cs-137 for Ob River sediments from 6 stations, assessed in filtered Ob River water. Stations 1 and 3 were sandy sediments, Stations 8, 11, and 13 were muddy (smallest grain size) and Station 5 was mixed.

## Effects of DOC on Kd

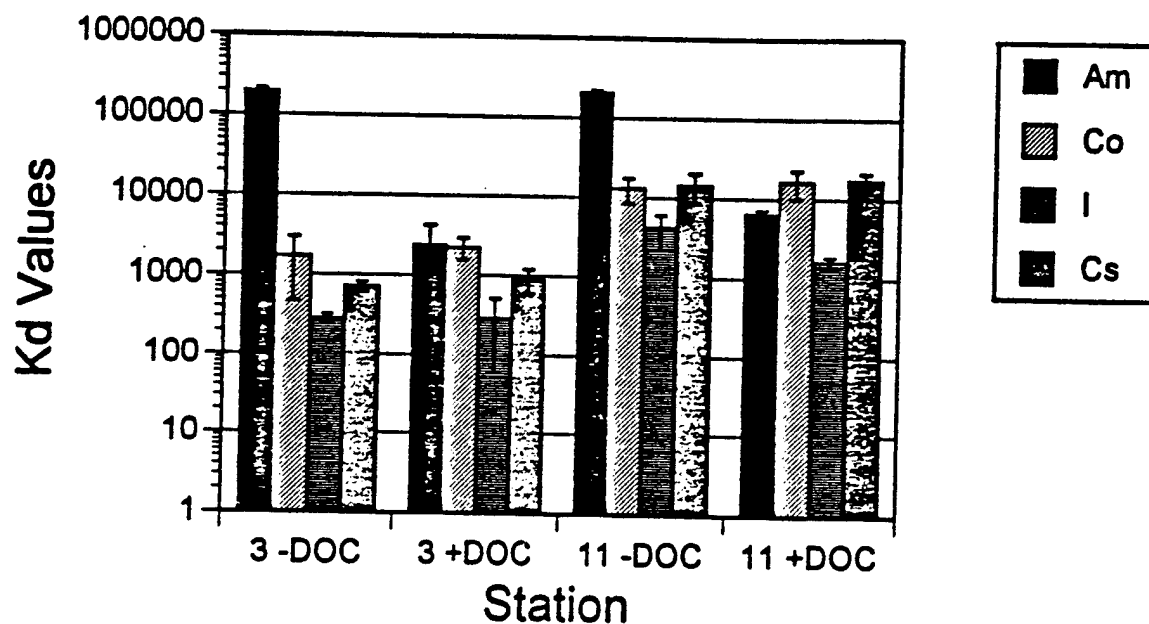


Fig. 9. Kd values of Am-241, Co-57, I-131, and Cs-137 for Ob River sediments from 2 stations, assessed in filtered Ob River water with and without UV irradiation (DOC removal). Note that only the Kd for Am was significantly affected by DOC photooxidation.

## Effects Of DOC on VCF

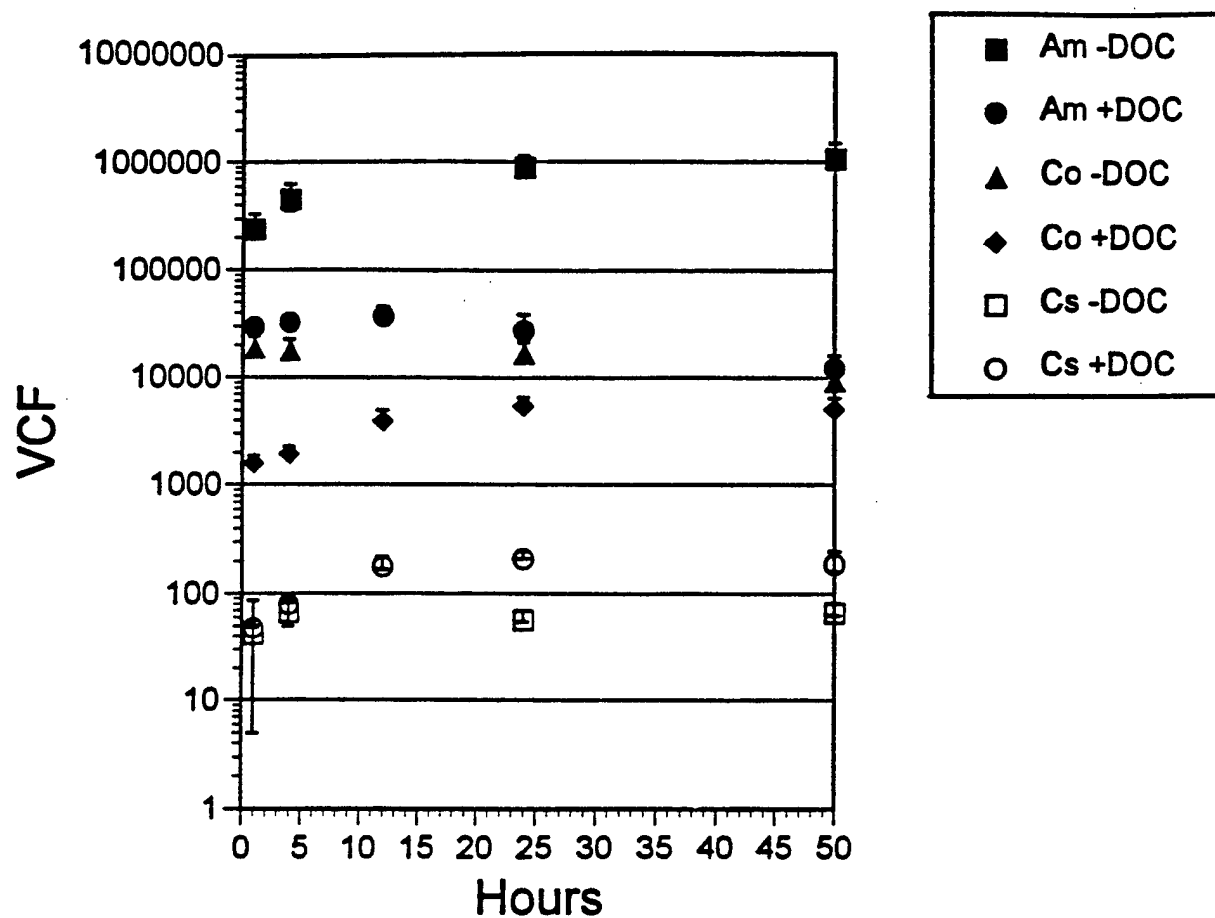


Fig. 10. Volume-volume concentration factors (VCFs) of Am-241, Co-57, and Cs-137 in the diatom *Thalassiosira pseudonana* (clone 3H) in filtered Ob River water with and without UV irradiation. Note the large effect of UV treatment on the VCF of Am.

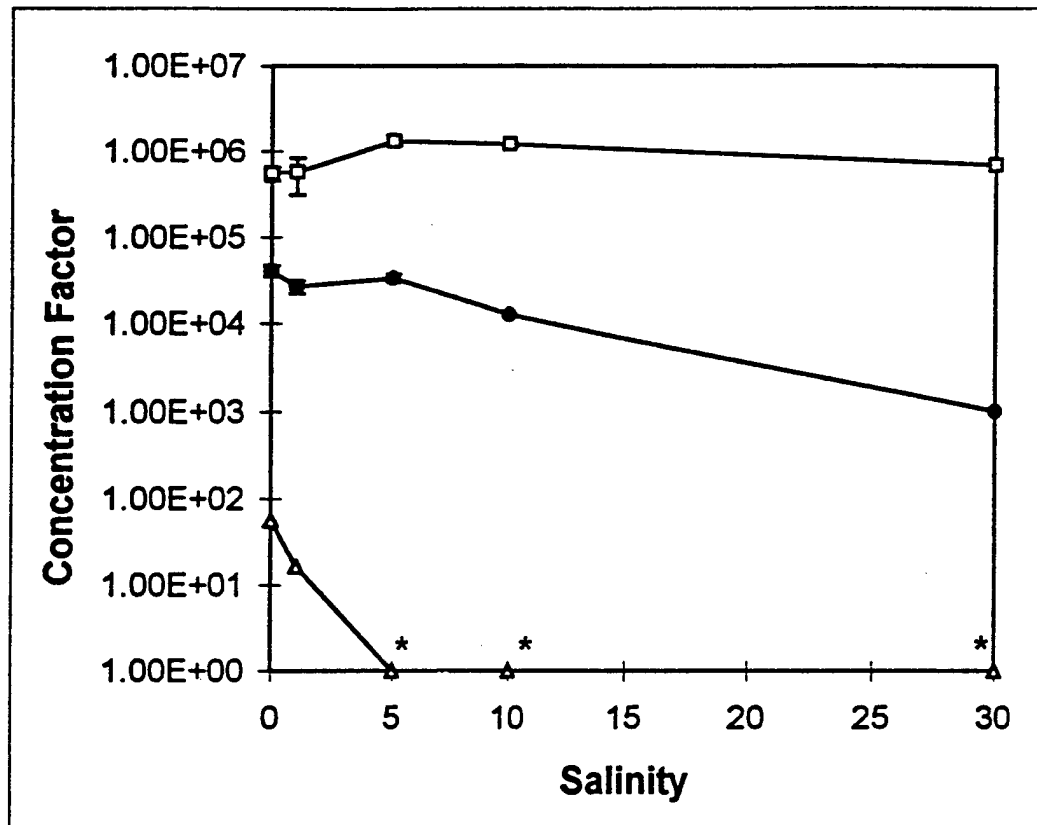


Fig. 11. Volume/volume concentration factors (at time of apparent equilibrium) of  $^{241}\text{Am}$  (□),  $^{57}\text{Co}$  (●), and  $^{137}\text{Cs}$  (Δ) in the diatom *Thalassiosira pseudonana* at different salinities (ppt). Data points are means of triplicate cultures  $\pm$  1 SD; where error bars are not visible, the errors were smaller than symbols used to denote means. \* denotes concentration factors < 1.