

Vertical Distribution of Anthropogenic Radionuclides in Cores from Contaminated Floodplains of the Yenisey River

Brown, J.E.¹, Dowdall, M.¹, Linnik, V. G.², Korobova E.M.², Volosov A.G.², & Standing, W.¹

¹Norwegian Radiation Protection Authority, P.O. Box 55, N-1332 Østerås, Norway.

²Vernadsky Institute of Geochemistry and Analytical Chemistry, Kosygin Street 19, 117975, Moscow, Russia.

INTRODUCTION

The Krasnoyarsk-26 Mining and Chemical Combine (KMCC; currently Zheleznogorsk), has constituted a source of anthropogenic radioactivity to the Yenisey River from commencement of original operations at the site in 1958 through to modern times. The main activities of the KMCC over the years have been plutonium production and the generation of electricity, employing three RBMK-type graphite-moderated nuclear reactors, plutonium reprocessing and the storage of resultant radioactive waste materials. The complex is sited on the east bank of the Yenisey River approximately 50 km northeast and downstream of Krasnoyarsk city in Siberia and discharges of radioactive effluents directly to the river have occurred at several points downstream of this area (JNREG, 2004). The two oldest reactors, which employed direct flow water cooling system, were shut down in 1992 and the third reactor at the site, utilising a closed primary cooling loop, remains in operation today, providing electricity generation for the region. Historically, discharges have comprised of cooling water from the direct flow and closed loop reactors, treated reactor drain water and discharges from the associated KMCC radiochemical facilities (JNREG, 2004). A full overview of the recent discharge history from the mid 1970's to the present is available elsewhere (JNREG, 2004). The hydrological regime of the Yenisey River underwent significant changes after the Krasnoyarsk Hydroelectric Power Plant (KHPP), situated 40 km upstream of Krasnoyarsk city, was completed in 1967. The KHPP influences river flow for about 800 km downstream (to the confluence with the Podkamennaya Tunguska River) and natural sediment load levels are not restored for a further 200 km. The operation of the plant has resulted in water discharges becoming more regular throughout the year, with spring and summer water levels much reduced.

The purpose of the study was to investigate the distribution and variation with depth of anthropogenic radionuclide contamination within alluvial matrices associated with periodic flooding by the waters of the Yenisey River. Furthermore, the association of radioactive contamination with various particle size fractions within the matrices was also studied.

METHODS

Cores were obtained at sites representative of different distances from the KMCC discharge area during field operations that were conducted in 1999 and 2000.

The selected sites constituted both floodplain and insular locations which have undergone different levels of flooding with accumulation of river sediments during the period of KMCC operation. All sites contained measurable amounts of artificial radionuclides as estimated under field conditions by in-situ gamma ray spectrometry using an NaI detector and described in Linnik et al., (2005); Linnik et al., (2006). Samples were taken using a corer of internal

diameter 7.8 cm and of length 30 cm and split into 5 cm increments for analyses. Samples were preserved in field-wet form for mineralogical and sedimentological analyses.

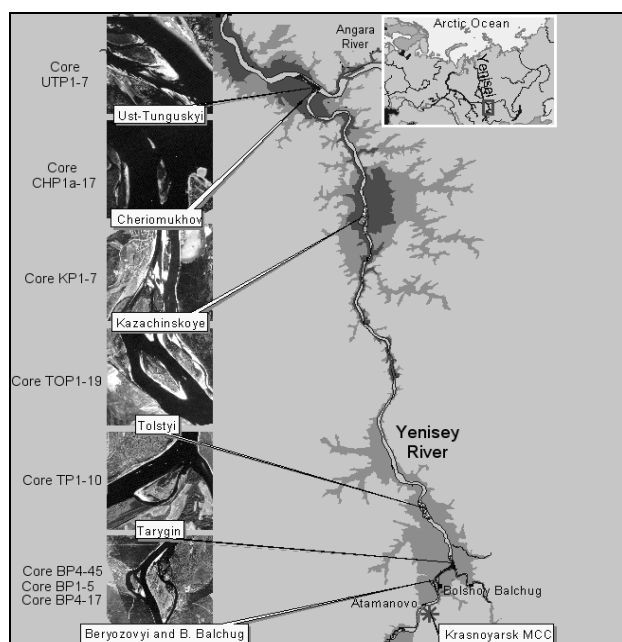


Figure 1. Study area and sampling locations.

Samples were sub-divided into $>63 \mu\text{m}$ (microns) and $< 63 \mu\text{m}$ fractions by a wet sieving process. The $>63 \mu\text{m}$ fraction was dried and weighed to provide the total sand content by mass. The $< 63 \mu\text{m}$ fraction was analysed using a Sedigraph 5100. Mineralogical determinations were made by X-ray Diffraction (XRD). XRD analyses was conducted using a Philips X'Pert X-ray diffractometer with theta-theta goniometer and Cu-tube following sample preparations.

Gamma-emitting radionuclides were analysed using high resolution HPGe detectors (resolution 1.7-2.0 keV at 1.33 MeV, relative efficiency of 20 % or higher). Samples were dried at 105°C , weighed, sieved, homogenised and split into sub-samples for analyses. Standard spectrometric analyses software was used to determine activity concentrations of ^{137}Cs allowing for all usual corrections. For isotopes of plutonium, samples were chemically purified using solvent extraction and ion exchange chromatography techniques following standard radiochemical procedures (Holm, 1984). Samples were electroplated onto stainless steel planchettes before measurement using alpha detectors, e.g. surface barrier detectors or passivated ion implanted silicon detectors.

RESULTS AND DISCUSSION

Variations in grain-size composition within a sampling area were of the same magnitude as variations observed between distinct areas. The samples from the cores taken on Beriozovy Island exhibited quite variable grain-size compositions ranging from sand through to sandy silts. The variation in samples taken from these cores (with distances of 100's of meters between them) is similar to the variation observed between cores from widely spaced geographical locations (extending over approx 300 km). Smectite was the dominant clay

mineral present (forming 21.6 – 79.6 % of the clay mineral assemblage). Illite (7.8 – 35.8%), kaolinite (9.6 – 33.6 %) and chlorite (0-39.5 %) were also identified in the samples.

Surface (0-5 cm increment) ^{137}Cs activity concentrations Beriozovy Island and the floodplain area of Balchug were in the range 254 to 1060 Bq/kg ^{137}Cs (d.w.) with a maximum activity concentration being observed at a site located on a low level floodplain. All cores exhibited subsurface maxima in ^{137}Cs activity levels falling in the range 1550 to 3770 Bq/kg ^{137}Cs (d.w.). The depths of the subsurface maxima were between 7.5 cm and 17.5 cm. Four of 6 cores sampled at locations from Beriozovy Island to Ust-Tunguskyi just downstream of the Yenisey-Angara Confluence exhibited subsurface maxima. The two remaining cores exhibited ^{137}Cs activity concentrations that decreased substantially with depth within the top 15-30 cm of sediment. Maximum surface activities were measured in the core sample taken on Tarygin Island, i.e. 890 Bq/kg ^{137}Cs (d.w.), some 27 km from the KMCC. The maximum sub-surface activity concentration measured was at the site nearest the KMCC, namely at the head of Beriozovy Island where an activity concentration of 2320 Bq/kg d.w. ^{137}Cs was determined.

Surface (0-5 cm increment) activity concentrations in samples from Beriozovy Island and the floodplain area of Balchug were in the range 0.9 – 14.2 Bq/kg d.w. $^{239,240}\text{Pu}$ whereas ^{238}Pu activity concentrations were in the range <0.01 – 1.2 Bq/kg d.w. Two cores from low lying floodplains both exhibited increasing Pu isotope activity concentrations with depth exhibiting maximum activity concentrations of $^{239,240}\text{Pu}$ of 9.3 ± 0.5 Bq/kg (depth = 16 cm) and 5.6 ± 0.1 Bq/kg (depth = 22.5 cm). Pu-239,240 activity concentrations over depth for 6 cores along a stretch of the Yenisey River from Beriozovy Island to to Ust-Tunguskyi just downstream of the Angara confluence exhibited surface activity concentrations in the range 0.7-6.9 Bq/kg d.w. $^{239,240}\text{Pu}$ and <0.1 – 0.8 Bq/kg d.w. ^{238}Pu . In all but two cases, the cores exhibited subsurface activity maxima. The maximum subsurface activity recorded was at Cheriomukhov Island, where an activity concentration of 9.4 ± 0.4 Bq/kg d.w. $^{239,240}\text{Pu}$ was measured at a depth of 22.5 cm.

The non-parametric Spearman rank correlation test was applied to test whether ^{137}Cs activity concentrations were correlated with silt content (% by mass <63 μm). The alternative hypothesis was that there was a correlation between activity concentration and silt content and that this was positive – a one-tailed test being therefore selected. A ρ value of 0.643 was calculated which indicates that the alternative hypothesis was significant at the 0.01 level. In other words, over a stretch of 250 km of the Yenisey River downstream of the KMCC, ^{137}Cs activity concentrations (including all depth increments) are quite strongly influenced by grain-size. Using the Spearman's rank correlation test (one tailed) to consider whether $^{239,240}\text{Pu}$ activity concentrations are dependent on the silt content (% < 63 μm content by mass), a correlation coefficient of 0.572 is observed. This value is significant at the 0.01 level and indicates that activity concentrations at all depths are strongly influence by grain-size.

The ^{238}Pu : $^{239,240}\text{Pu}$ activity ratio for global fallout is in the range 0.03-0.04 (Sholkovitz, 1983). Pu discharge data from the KMCC have been present in a recent ISTC project (Vakulovsky, 2001) for the period 1994-2000. The information from this source is of limited use because the ^{238}Pu data are reported as “less than” values. However, the ratios can be seen to not exceed 0.28 and if one extracts a mean ratio based on the average discharges in the period 1994-2000 inclusive (i.e. a ^{238}Pu value of 50 MBq/year and a mean $^{239,240}\text{Pu}$ value of 725 MBq/year) a ratio of 0.069 is derived. Some high ^{238}Pu : $^{239,240}\text{Pu}$ activity ratios are observed for some of the samples that may be indicative of the early years of operation of the KMCC although, without

a reported discharge chronology this is impossible to verify. High ratios have been observed primarily in subsurface samples. At one station on Beriozovy Island, elevated ^{238}Pu : $^{239,240}\text{Pu}$ activity ratios were observed in the surface sediments falling to global fallout levels at depth. Other stations, in the vicinity but on the Balchug floodplain express ratios that are indicative of a global fallout source of Pu or a mixture of global fallout inputs and other sources.

CONCLUSIONS

Sediment samples from cores exhibit quite variable grain-size compositions ranging from silts to sandy silts, the grain-size not varying with depth for most sampled cores with the exception of one core taken on the Balchug floodplain. Clay mineral assemblages are highly variable within the study area, smectite being the dominant clay mineral present in most of the clay samples although percentages as low as 21.6 % were measured in some cases. Cs-137 activity concentrations were in the range 23-3770 Bq/kg (d.w.) with numerous cores exhibiting subsurface maxima which may be related to the historical discharge profile from the KMCC. Cs-137 activity concentrations (including all samples) were closely correlated with the fraction of silt (% by mass < 63 μm) present in the cores. Pu-239,240 activity concentrations were in the range <0.01 to 14.2 Bq/kg (d.w.) with some cores exhibiting elevated concentrations of Pu at their base. Pu-239,240 activity concentrations (including all samples) were correlated with the fraction of silt present. The ^{238}Pu : $^{239,240}\text{Pu}$ activity ratios suggested a complex system although a Pu signal discernable from global fallout could be observed in numerous samples in the KMCC near zone.

REFERENCES

- Holm, E. (1984). Review of alpha-spectrometric measurements of actinides. *Int. J. of Applied Rad. Isotop.*, 35, pp.285-290.
- JNREG (Joint Russian Norwegian Expert Group) 2004. Impacts on man and the environment in northern areas from hypothetical accidents at Krasnoyarsk Mining and Chemical Industrial complex, Russia. In: W.J.F. Standring, Editor, Programme of the Joint Norwegian–Russian Expert Group for Investigation of Radioactive Contamination in Northern Areas, Østerås, 2004 (2004), p. 97.
- Linnik V.G., Brown J.E., Dowdall M., Potapov V.N., Surkov V.V., Korobova E. M., Volosov A.G., Vakulovsky, S.M., Tertyshnik, E.G. (2005). Radioactive Contamination of the Balchug (Upper Yenisey) Floodplain, Russia in Relation to Sedimentation Processes and Geomorphology. *Science of the Total Environment*, 339, pp.233-251.
- Linnik, V.G., Brown, J.E., Dowdall, M., Potapov, V.N., Nosov, A.V., Surkov, V.V., Sokolov, A.V., Wright, S.M. and Borghuis, S. (2006). Patterns and inventories of radioactive contamination of island sites of the Yenisey River, Russia. *Journal of Environmental Radioactivity*, 87 (Issue 2), pp. 188-208.
- Vakulovsky, S.M. (2001). Estimation and prediction of the consequences for the environment and population of radioactive contamination of the River Yenisey by discharges of the Krasnoyarsk Mining and Chemical Industrial Complex. 1st Annual Report for ISTC Project 1404. SPA “TYPHOON”, Obninsk, Russia, pp77.
- Sholkovitz, E.R. (1983). The geochemistry of plutonium in fresh and marine water environments *Earth Science Review*, 19, pp.95-161.