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Sediments of the Yenisei River: monitoring of radionuclide levels and estimation of sedimentation rates

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Abstract The Yenisei River, one of the largest rivers in the world, is contaminated with artificial radionuclides released by a Russian nuclear facility producing weapon-grade plutonium, which has been in operation for many years. Examination of Yenisei River sediment samples revealed the presence of artificial radionuclides typical of radioactive discharge from the Mining-and-Chemical-Combine (MCC) nuclear facility: isotopes of europium (^{152}Eu, ^{154}Eu, and ^{155}Eu), caesium (^{137}Cs and ^{134}Cs), ^{60}Co, ^{90}Sr, and transuranium elements. Maximum radionuclide concentrations in sediments remained high as far as 240 km downstream of the MCC. In sediment cores collected upstream of the MCC, γ-spectrometric measurements registered only one artificial radionuclide, ^{137}Cs, with a maximum activity of approx. 8 Bq kg^{-1} dry mass. Sediments of the Yenisei River also contain natural radionuclides. Sedimentation rates in several sections of the Yenisei River were determined using different approaches: the ^{210}Pb dating method and the ratios of artificial radionuclides – ^{137}Cs/^{60}Co and ^{152}Eu/^{154}Eu. With increasing distance downstream of the city of Krasnoyarsk, sedimentation rates increased from 0.88 cm year^{-1} to 1.30–1.51 cm year^{-1}.

Key words river sediments; artificial and natural radionuclides; sedimentation rates; dating methods; Yenisei River

INTRODUCTION

The Yenisei is one of the world's largest rivers, over 3000 km long, flowing into the Kara Sea. The Mining-and-Chemical Combine (MCC) facility, which has been producing weapons-grade plutonium since 1958, is situated on the east bank of the Yenisei River, 60 km downstream of the city of Krasnoyarsk. The MCC has been in operation for many years, and has been contaminating the flood plain soils and sediments of the Yenisei River with both artificial and natural radionuclides (Bolsunovsky *et al.,* 2002, 2007; Klemt *et al*., 2002; Bolsunovsky & Bondareva, 2007).

Various methods have been used successfully to determine sedimentation rates and date sediment in lake ecosystems (Carroll & Lerche, 2003), due to their stable hydrological conditions and, hence, relatively uniform sedimentation rates. The difficulty of using such methods to quantify sedimentary processes in the river channel system is caused by the complexity of hydrological conditions in such environments, especially the Yenisei River, due to the presence of hydroelectric power stations and such seasonal events as ice drifts and floods. On the other hand, the Yenisei River is a unique environment with sediments that contain various artificial and natural radionuclides, which makes it possible to date sediment layers using different methods.

Channel bottom sediments are a unique material that, under certain conditions, records the state of the environment for a particular time period. A popular method used for determining sedimentation rates and the absolute age of sediment layers is the ^{210}Pb dating method (Carroll & Lerche, 2003). Another method for determining sedimentation rates is based on the ratios of ^{137}Cs/^{60}Co and ^{152}Eu/^{154}Eu activity concentrations (Gritchenko *et al*., 2002).

The purpose of this study was to estimate levels of artificial radionuclides in river sediments and to determine sedimentation rates in different sections of the Yenisei River, downstream of the city of Krasnoyarsk.

MATERIALS AND METHODS

About 150 sediment cores were collected from the channel bed of the Yenisei River at different distances downstream of the MCC during 1997–2008. Sediment cores were collected in two main areas: near the MCC (site E15, near the village of B. Balchug, 96 km downstream of Krasnoyarsk) and at a considerable distance downstream (site E20-K, near the village of Kargino, 320 km downstream of Krasnoyarsk) (Fig. 1). The village of B. Balchug is situated about 15 km downstream of where the MCC releases its radioactive water. For comparison purposes, we also collected sediment cores at a position upstream of the MCC, near the village of Esaulovo (site E5, 45 km downstream of Krasnoyarsk) (Fig. 1). Sediments were collected from river branches with not more than 1 m of water above the sediments. The diameter of the corer was 11 cm and it collected cores up to 1.5 m long. Cores were sectioned into 3-cm thick layers, except for the top sediment layer, which was 5–10 cm thick (depending on the core moisture content). The maximum number of layers for a core was 46.

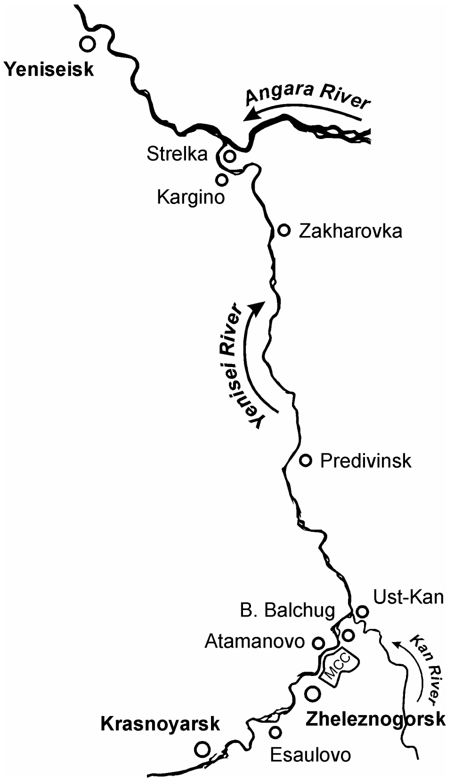

Fig. 1 Map of the south of the Krasnoyarsk Territory (Russia), showing the MCC facility and settlements near where sediment samples were collected. Scale 1 : 2 800 000.

Measurements of activity concentrations of γ-emitting nuclides (including ^{241}Am) were conducted at the Institute of Biophysics SB RAS using a Canberra γ-spectrometer (USA) coupled to a GX2320 23% hyperpure germanium (HPGe) co-axial detector. Gamma-spectra were processed using the CANBERRA GENIE-2000 software (USA). Radiochemical determination of actinides (^{238}Pu, ^{239,240}Pu, ^{241}Pu, ^{241}Am, ^{243,244}Cm) in sediment was performed at the RPA RADON (Moscow) (Bolsunovsky *et al.*, 2002, 2007). Activity concentrations of radionuclides in sediment layers were decay corrected to the sample collection dates.

LEVELS OF ARTIFICIAL RADIONUCLIDES IN SEDIMENTS OF DIFFERENT RIVER SECTIONS

Gamma-spectrometry measurements showed artificial radionuclides typical of the radioactive discharge from the MCC: isotopes of europium (^{152}Eu, ^{154}Eu, and ^{155}Eu), caesium (^{137}Cs and ^{134}Cs), ^{60}Co, and the transuranium element ^{241}Am. The vertical distribution of radionuclides in the sediment cores is complex and there are several concentration minimums and maximums due to different amounts of radionuclides released by the MCC and due to variations in global fallout. Since 1997, we have collected sediment samples from the same position at the Yenisei River branch near the MCC at site E15. The maximum ^{137}Cs activity concentrations (up to 2000 Bq kg^{-1}) were registered in the middle and lower sections of the E15 core (Fig. 2). Gamma-spectrometric analysis also revealed very high ^{241}Am concentrations (up to 64 Bq kg^{-1}) in the middle and lower sections of the core, although the location of the peaks in the middle section of the core differ for two radionuclides. For the sediment core collected at site E20-K, the maximum ^{137}Cs activity concentrations (up to 900 Bq kg^{-1}) were recorded also in the middle and the lower parts, with relatively low concentrations at the surface. ^{241}Am (up to 10 Bq kg^{-1}) was also detected in core E20-K (Fig. 2).

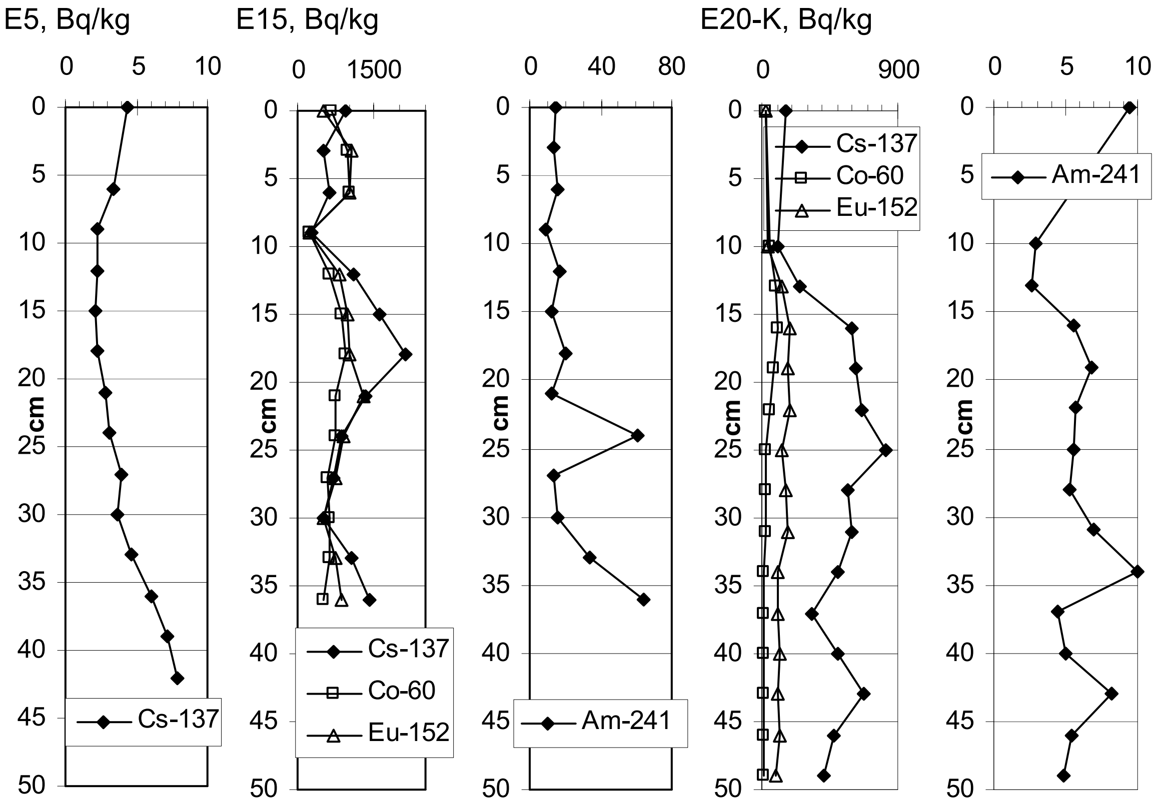

Fig. 2 Representative vertical distribution profiles of radionuclides in channel bed sediments (dry mass) of the Yenisei River near the villages of Esaulovo (E5), B.Balchug (E15) and Kargino (E20-K). See Fig. 1 for locations

The fact that some sediment layers contained high ^{241}Am concentrations could be indicative of high concentrations of other transuranium elements. Further analysis of the sediment layers that contained maximum levels of radionuclides, including ^{241}Am, indicated that the Yenisei River sediment samples had local anomalous hotspots with high activity concentrations of transuranium elements ($^{239,\ 240}$Pu and ^{241}Am), which were 100 times higher than their global fallout levels. Results of previous measurements of transuranium elements, such as ^{241}Pu and ^{237}Np, in Yenisei River sediments show that their levels remain high as far as 200 km downstream of the MCC (Bolsunovsky *et al.*, 2002; Bolsunovsky & Bondareva, 2007). The highest level of $^{243,\ 244}$Cm isotopes registered in the sediment of the Yenisei River was 21.4 Bq kg^{-1} dry mass (Bolsunovsky *et al.*, 2007), which is more than twice the maximum levels reported for soils sampled near the Chernobyl Nuclear Plant. The presence of sediment layers containing abnormally high actinide levels (Bolsunovsky *et al.*, 2002, 2007; Bolsunovsky & Bondareva, 2007) may be indicative both of the mobile behaviour of transuranium radionuclides in the river ecosystem and of continued disposals of artificial radionuclides by the MCC.

As mentioned above, we also collected sediment cores at a position upstream of the MCC, at site E5. In these sediment cores, γ-spectrometric measurements registered only one artificial radionuclide that is also present in the MCC radioactive discharge – ^{137}Cs (Fig. 2). The maximum activity of ^{137}Cs (7.9±0.6 Bq kg^{-1} dry mass) occurred in the lowest sediment layers (39–45 cm) of the sediment cores collected at E5 (Fig. 2). This level of ^{137}Cs can be accepted as the background level of radioactive contamination of sediments in this part of the Yenisei, due to global fallout of ^{137}Cs from nuclear tests. Subsequently, we can use these values obtained for site E5 as a reference to analyse sediments collected from the Yenisei sections near the MCC discharges: the hydrological conditions in this river section are stable as the distance between the village of Esaulovo and the MCC discharge point is just 40 km and there are no large tributaries flowing into the Yenisei along this reach.

SEDIMENTATION RATES AND DATING OF SEDIMENT LAYERS IN DIFFERENT SECTIONS OF THE RIVER

Sedimentation rates in different parts of the Yenisei River are shown in Table 1. The model for unsupported ^{210}Pb was used to estimate the sedimentation rate (v = 0.88±0.09 cm year^{-1}) at site E5. The ^{210}Pb dating method cannot be used to determine the age of sediments downstream of the MCC as they contain ^{152}Eu, which emits X-rays of energy 45.4 keV, overlapping the ^{210}Pb γ-radiation energy (46.5 keV). Thus, to determine the sedimentation rate at other locations, we used the following ratios of artificial radionuclides: ^{137}Cs/^{60}Co and ^{152}Eu/^{154}Eu. The initial values of these ratios are assumed to be constant, changing with time due to different half-lives of the isotopes. Based on this, we determined sedimentation rates of cores collected downstream of the MCC discharge point: site E15 (96 km downstream of Krasnoyarsk) and site E20-K (320 km downstream of Krasnoyarsk). Sedimentation rates for two cores at site E15 (1.05 and 1.26 cm year^{-1}) were estimated from the ^{137}Cs/^{60}Co ratio (Table 1). For this area, the regression line showed a weak correlation (R^2 = 0.33) between the values of the ^{152}Eu/^{154}Eu ratio and depth, so this ratio was not used to determine sedimentation rate. The sedimentation rates obtained for site E15 (Table 1) are reasonably close to the value calculated using the ^{210}Pb dating method for the area upstream of the MCC (site E5, 0.88±0.09 cm year^{-1}). Thus, as stated above, the hydrological conditions in this section of the river are indeed stable. Sedimentation rates for site E20-K were also calculated from the ratios of the artificial radionuclides ^{137}Cs/^{60}Co and ^{152}Eu/^{154}Eu. The sedimentation rates calculated using the ^{137}Cs/^{60}Co ratio and the ^{152}Eu/^{154}Eu ratio were similar (v = 1.30±0.08 cm year^{-1} and v = 1.51±0.09 cm year^{-1}, respectively) (Table 1). The sedimentation rate at site E20-K is greater than at sites E5 and E15 due to the fact that site E20-K is located downstream of the inflow of a large eastern tributary of the Yenisei – the Kan River (see Fig. 1). The water discharge of the Kan River amounts to 10–15% of the discharge of the Yenisei at this location. Moreover, the Kan River has a higher suspended sediment concentration than the Yenisei River.

Table 1 Sedimentation rates for cores collected from sites along the Yenisei River.

Area, site	Distance from Krasnoyarsk (km)	Sedimentation rate (cm year^{-1})	Calculation method
Esaulovo, E5	45 km	0.88±0.09	^{210}Pb
B. Balchug, E15-1	96 km	1.26±0.08	^{137}Cs/^{60}Co
E15-2		1.05±0.11	^{137}Cs/^{60}Co
Kargino, E20-K	320 km	1.30±0.08	^{137}Cs/^{60}Co
		1.51±0.09	^{152}Eu/^{154}Eu

The history of contamination of channel bed sediments by ^{137}Cs from the MCC was examined using the estimated sedimentation rates. The highest floods for the Yenisei River of the past 50 years occurred in 1966, 1988 and 2006. The MCC also performed emergency discharges of radionuclides (including ^{137}Cs) into the river, but the exact dates of these events are unknown. During extreme floods, sediments contaminated with radionuclides from the MCC are remobilized, and subsequently deposited on the channel bed and flood plain surface further downstream, which causes the formation of new peaks of ^{137}Cs and other radionuclides in sediments. The highest ^{137}Cs activity concentrations for site E15 are found in the sediment layers dated as 1964 and 1982–1984 (Fig. 3(a)). Taking into account calculation errors and uncertainties, these concentration peaks can be equated with the high floods of 1966 and 1988. The highest ^{137}Cs activity concentrations in the core from site E20-K, collected at a considerable distance downstream of the MCC, are recorded in the layer dated as 1987 (Fig. 3(b)), and this peak can be also associated with the 1988 flood.

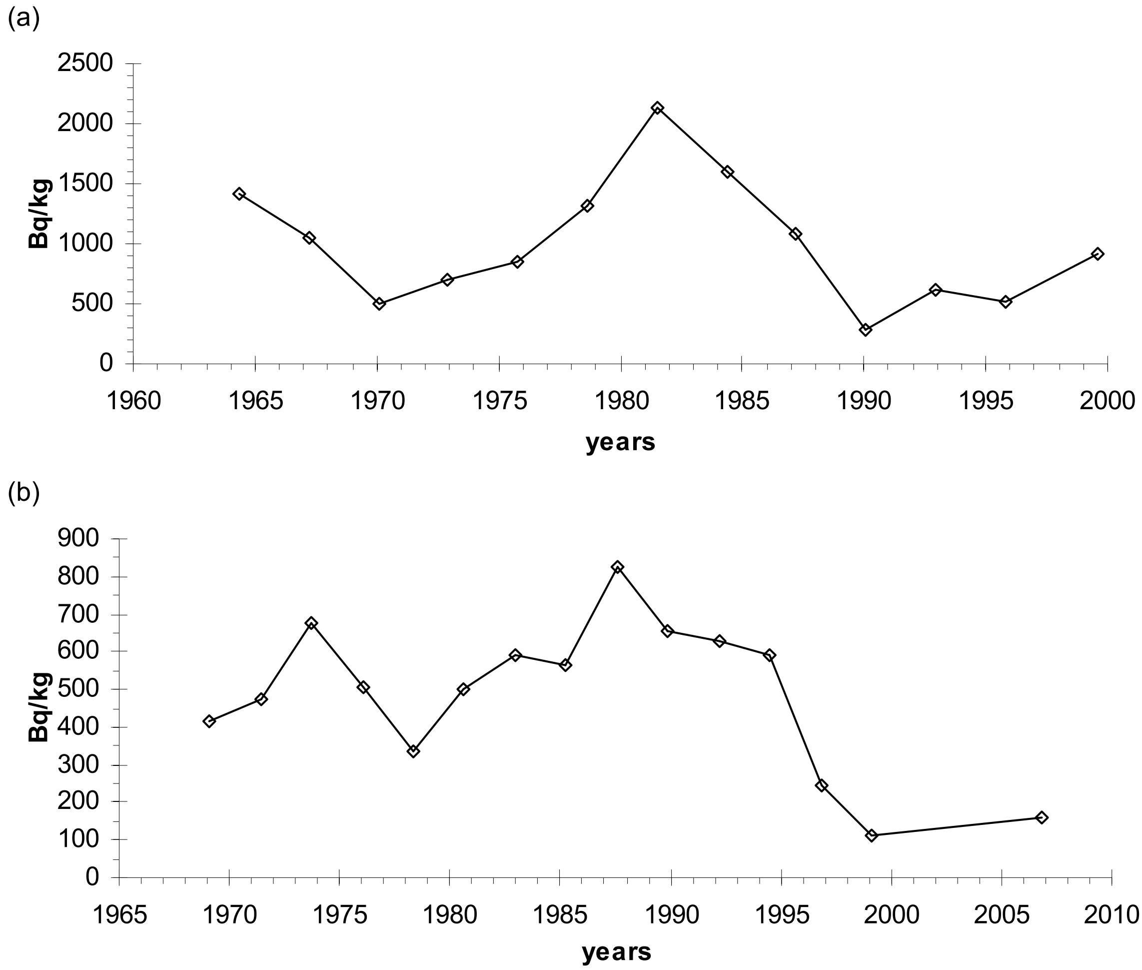

Fig. 3 ^{137}Cs contamination from the MCC nuclear facility of the sediments of the Yenisei River for site E15 (a) and site E20-K (b) based on estimated sedimentation rates and the record of known high flood events.

CONCLUSION

The examination of channel bed sediment samples collected from the Yenisei River revealed the presence of artificial radionuclides characteristic of radioactive discharges from the MCC facility: isotopes of europium (^{152}Eu, ^{154}Eu and ^{155}Eu), caesium (^{137}Cs and ^{134}Cs), ^{60}Co, ^{90}Sr and transuranium elements. Radionuclide concentrations in sediment layers remained high as far as 240 km downstream of the MCC. The vertical distribution of radionuclides in sediment cores was very complex, and downcore variations were due to different amounts of radionuclides released by the MCC and due to variations in the hydrological conditions in the river. For sediment cores collected at a site upstream of the MCC, there was only one artificial radionuclide, ^{137}Cs, with a maximum activity of approx. 8 Bq kg^{-1} dry mass. This level of ^{137}Cs can be assumed to represent the background level of radioactive contamination of sediments in this part of the Yenisei River, due to global ^{137}Cs fallout from nuclear tests. Sedimentation rates in several sections of the Yenisei River were estimated using different radioisotope methods. In the reference area, situated upstream of MCC, the estimated sedimentation rate was 0.88 cm year^{-1}. Similar sedimentation rates were obtained for the site just downstream of the MCC discharge point (1.05–1.26 cm year^{-1}), while rates calculated for the site situated at a considerable distance from the MCC (320 km and 240 km downstream of Krasnoyarsk and the MCC, respectively) were somewhat higher (1.30–1.51 cm year^{-1}). The obtained sedimentation rates provided a basis for dating the layers of sediments collected at the different river sections below the MCC nuclear facility. For example, the highest ^{137}Cs concentrations in Yenisei channel bed sediments can be tentatively dated to 1966 and 1988 – the years of two extreme flood events.

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