ESTIMATION OF BLACK SEA BASIN CONTAMINATION WITH ^{137}CS AND ^{90}SR OWING TO BOTTOM SEDIMENTS

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Abstract

Contemporaneous contamination of bottom deposits with ^{137}Cs sampled from the currents within the Chornobyl Exclusion Zone is made up to $(0.15—12)\cdot10^{14}$ Bq·km^{-2}. Ratio of ^{137}Cs/^{90}Sr in all samples is 20±2. Initial value of this ratio was approximately 1 (zone of mainly nuclear fuel particles contamination). This fact testifies that ^{90}Sr is now quantitatively removed from bottom sediments. Experimental data of ^{137}Cs leaching from bottom sediments in temporal variation is satisfactory described by logarithmically normal regularity. More than 95 % of ^{90}Sr is leached from the bottom sediments during the 40 days. These data have been described with the application of formal kinetic law. Basing on these data total quantity of radionuclides transoported into the Black Sea Basin during 15 years is estimated to be $2\cdot10^{14}$ Bq for ^{90}Sr, and $2\cdot10^{13}$ Bq for ^{137}Cs.

Introduction

The main source of the radionuclides contamination of the Dnipro reservoirs was the Rivers Prypyat and Dnipro (Table 1). The main radionuclides intake into the River Dnipro system took place because of radioactive aerosol fallout during the active release from the emergency unit of the Chernobyl Nuclear Power Plant (ChNPP). The water and bottom sediments were contaminated with these artificial radionuclides. Formation of bottom deposits contamination with artificial radionuclides of Chornobyl origin within the River Dnipro cascade was generally completed before the summer 1987. During the year after the accident the Rivers Prypyat and Dnipro exported about 80 % of ^{137}Cs and 60 % of ^{90}Sr of their total amount discharged into these rivers during 1986-1989. ^{137}Cs inventory in bottom deposits in 1990 was estimated to be $1.4\cdot10^{14}$ Bq, for ^{90}Sr – to be $4\cdot10^{13}$ Bq (Table 2).

Table 1 – Sources of ^{137}Cs and ^{90}Sr transportation to Kyiv Reservoir, Bq

River	^{137}Cs		^{90}Sr
	Dissolved matter	Suspended particulate matter	
Dnipro	$4{,}70\cdot10^{13}$	$1{,}09\cdot10^{13}$	$2{,}66\cdot10^{13}$
Pryp'yat	$7{,}27\cdot10^{13}$	$1{,}61\cdot10^{13}$	$6{,}01\cdot10^{13}$
Small rivers	$3{,}70\cdot10^{12}$	$2{,}22\cdot10^{12}$	$4{,}44\cdot10^{12}$
Total	$1{,}23\cdot10^{14}$	$2{,}92\cdot10^{13}$	$9{,}12\cdot10^{13}$

The total ^{137}Cs inflow into the Dnipro reservoirs cascade via rivers flow was about $1.7\cdot10^{14}$ Bq from 1986 to 1989 (Table 3). During the same period ^{137}Cs discharge into the Black Sea via dissolved and suspended species was about 10-15 % of the inflow to the cascade (with the exception of the initial phase in 1986). At the same time almost the total amount of ^{90}Sr entering the Dnipro and Prypyat Rivers discharged into the Black Sea (1).

Methods

Bottom deposits were sampled from the currents within left-bank flood-lands of the River Pryp'yat. This area was highly contaminated with hot particles of irradiated nuclear fuel origin. Sampling points are situated at the distance 5-7 km to the north from the emergency unit of Chornobyl NPP. Deposits were dried and consecutively extracted with distilled water, 1 N CH_3COONa, 0.1 N HCL, and 6 N HNO_3 for discrimination of water soluble, exchangeable, mobile and immobile species respectively.

Table 2 – Transportation of ^{137}Cs and ^{90}Sr
to Kyiv Reservoir via Rivers Pryp'yat and Dnipro, Bq

Year	^{137}Cs		^{90}Sr
	Dissolved matter	Suspended particulate matter	
1986 rough estimate	$8{,}14 \cdot 10^{13}$	$4{,}44 \cdot 10^{12}$	$3{,}72 \cdot 10^{13}$
1987	$1{,}63 \cdot 10^{13}$	$1{,}24 \cdot 10^{13}$	$1{,}57 \cdot 10^{13}$
1988	$1{,}30 \cdot 10^{13}$	$5{,}55 \cdot 10^{12}$	$2{,}28 \cdot 10^{13}$
1989	$9{,}06 \cdot 10^{12}$	$4{,}26 \cdot 10^{12}$	$1{,}11 \cdot 10^{13}$
Total	$1{,}20 \cdot 10^{14}$	$2{,}70 \cdot 10^{13}$	$8{,}68 \cdot 10^{13}$

Table 3 - ^{137}Cs inventory in the River Dnipro reservoirs in 1990

Reservoir	Square, km^2	^{137}Cs inventory, Bq	Density of contamination $Bq \cdot km^{-2}$
Kyiv	922	$9{,}62 \cdot 10^{13}$	$1{,}04 \cdot 10^{11}$
Kaniv	675	$1{,}30 \cdot 10^{13}$	$1{,}92 \cdot 10^{12}$
Kremenchuh	2250	$1{,}63 \cdot 10^{13}$	$7{,}40 \cdot 10^{9}$
Dniprodzerzhynsk	567	$2{,}26 \cdot 10^{12}$	$4{,}07 \cdot 10^{9}$
Zaporizhya	410	$1{,}26 \cdot 10^{12}$	$3{,}07 \cdot 10^{9}$
Kahovka	2150	$6{,}40 \cdot 10^{12}$	$2{,}96 \cdot 10^{9}$
Total	6974	$1{,}32 \cdot 10^{14}$	

Dynamics of water leaching was studied by passing the distilled water through deposits during 40 days (Fig. 1). Aliquots were tested every day during the first week, every other day during the second week, and further weekly.

Results and Discussion

Tested deposits were differing in chemical properties and radionuclide contamination (Table 4). Contemporaneous contamination of bottom deposits with ^{137}Cs sampled from the currents within the Chornobyl Exclusion Zone is made up to $(0.15—12) \cdot 10^{14}$ $Bq \cdot km^{-2}$. Ratio of $^{137}Cs/^{90}Sr$ in all samples is 20±2. Initial value of this ratio was approximately 1 that corresponded to zone of mainly nuclear fuel particles contamination. This fact testifies that ^{90}Sr is now quantitatively removed from bottom deposits.

Quantum of water soluble species of ^{137}Cs is between 0.18 and 0.26 %. About 1 % of ^{137}Cs is corresponded to exchangeable species and 2 % is related to mobile form. The main part of ^{137}Cs (up to 90 %) is immobile and inconvenient for leaching with concentrated HNO_3.

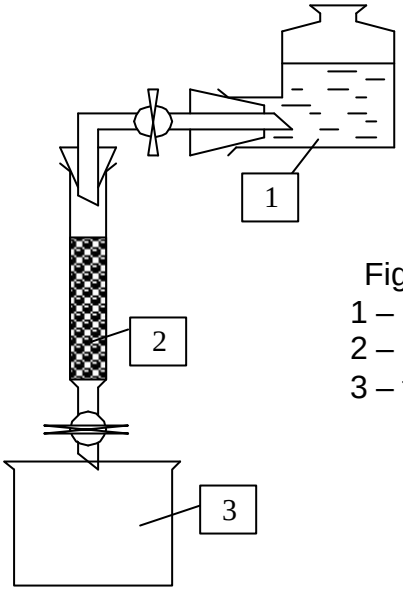

Fig. 1 – Scheme of water leaching in dynamic conditions:
1 – glass with distilled water,
2 – burette with sample,
3 – testing glass.

Up to 40 % of ^{90}Sr in bottom deposits is related to water soluble form. The rest part of ^{90}Sr is exchangeable. No more than 2.5 % of ^{90}Sr is corresponded to immobile species. So mobility of ^{90}Sr is substantially higher than of ^{137}Cs.

Leaching of deposits during 40 days with distilled water in dynamic conditions leads to quantitatively removing of ^{90}Sr from the solid phase. At the same time no more than 4 % of ^{137}Cs is transferred to water phase.

Table 4 – Chemical properties of bottom sediments

Characteristic	Unit	Mineral	Gleyish	Organic
Mg^{2+}	mg-eq/100 g	0.14	0.60	-
Ca^{2+}	mg-eq/100 g	2.38	6.8	-
Na^{+}	mg-eq/100 g	0.051	0.077	-
K^{+}	mg-eq/100 g	0.02	0.085	-
H^{+}	mg-eq/100 g	2.19	7.3	14.3
SEB	mg-eq/100 g	4.74	15.6	32.6
pH_e		5.4	3.57	4.9
pH_w		5.1	3.3	4.5
^{137}Cs	$MBq \cdot m^{-1}$	15	330	1200
^{90}Sr	$MBq \cdot m^{-1}$	0.68	16.5	67

Dynamics of leaching of ^{137}Cs is satisfactory described with logarithmical normal regularity (Fig. 2):

$$N = k_B \ln t + A, \quad (1)$$

where N corresponds to part of radionuclide leached during the time t, k_B is rate constant of radionuclide transformation from the solid phase of deposit to water soluble species, and A is parameter related to physical-chemical properties of solid phase. Value of k_B is between 0.25 and 0.86 (central tendency is 0.51). Value of A is between 1.7 and 5.5 y^{-1} and central tendency is 3.3 y^{-1}. Half time of removing of ^{137}Cs from bottom deposits (more than 300 years) is substantially higher than its half-life. So ^{137}Cs is fast retaining by the solid phase of sediments and not able for transportation by the aqueous way. Basing on these data we can assess that during

1987–2003 near 8 % of ^{137}Cs was removed from bottom deposits that corresponds to $1 \cdot 10^{13}$ Bq.

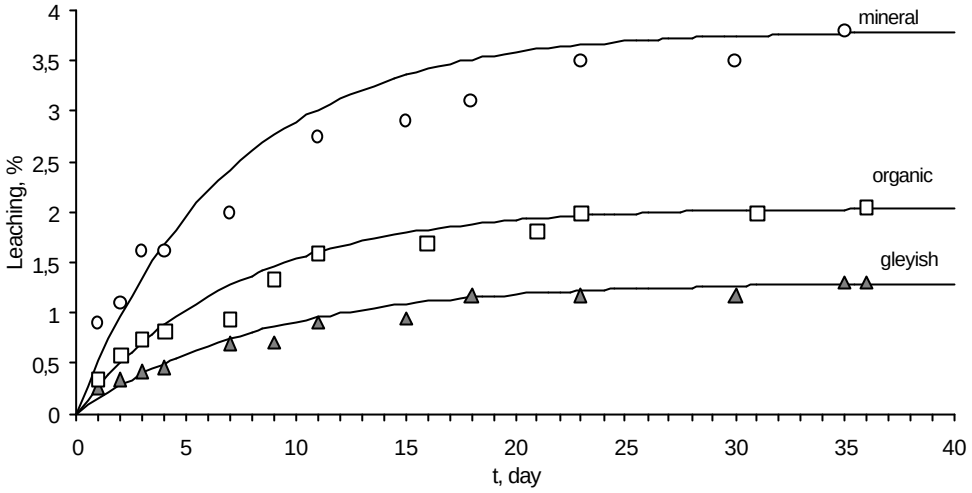

Fig. 2 -Temporal dynamics of ^{137}Cs leaching from bottom sediments

Dynamics of ^{90}Sr leaching is subjected to kinetic law of the first order (Fig. 3):

$$N = N_0(1 - e^{-k_B t}), \quad (2)$$

where N_0 related to the bulk content of ^{90}Sr in bottom deposit, k_B is rate constant of radionuclide transformation from solid phase of sediment to water soluble species, t is time of leaching. When N_0 = 100 %, value of k_B of ^{90}Sr is $(4.2–10) \cdot 10^{-6}$ s^{-1}. Half-time of ^{90}Sr removing from the solid phase of deposit is between 2 and 4 days. Thus ^{90}Sr in general is exported from bottom sediments via the River Dnipro cascade to the Black Sea that corresponds to $1 \cdot 10^{14}$ Bq.

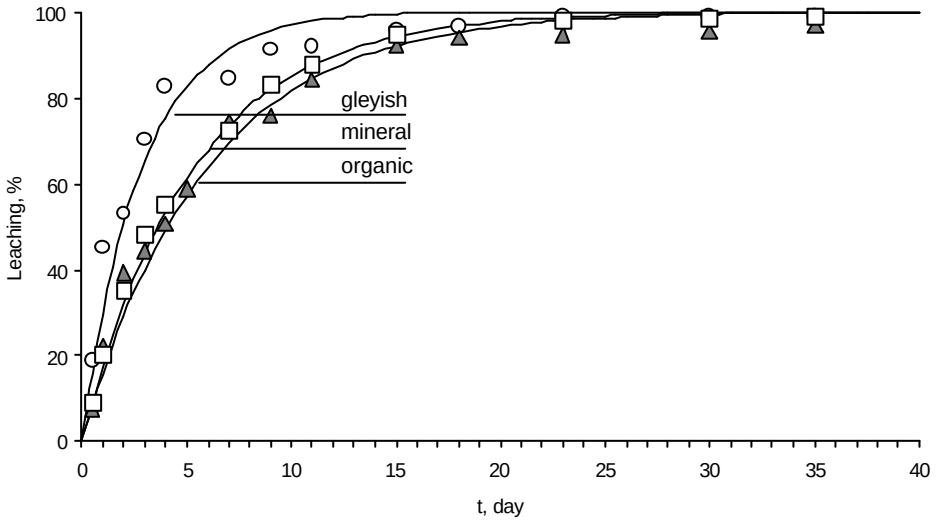

Fig. 3 - Temporal dynamics of ^{90}Sr leaching from bottom sediments

In earlier investigations we have assessed that near $6 \cdot 10^{11}$ Bq of ^{137}Cs and $7 \cdot 10^{12}$ Bq of ^{90}Sr is exported from contaminated columbines to Black Sea Basin in dissolved species. Thus the total export from different sources is assessed to $2 \cdot 10^{14}$ Bq of ^{90}Sr and $2 \cdot 10^{13}$ Bq of ^{137}Cs.

Conclusions

Now ^{90}Sr is practically completely removed from bottom deposits and transported via the River Dnipro system to the Black Sea.

About 10^{14} Bq of ^{90}Sr been transported to the Mediterranean Basin is corresponded to contaminated bottom sediments.

^{137}Cs is fast retaining by the solid phase of bottom sediments. No more than 8 % of ^{137}Cs was removed from bottom sediments during 1987-2003 that corresponds to $1 \cdot 10^{13}$ Bq.

The total export from different sources is assessed to $2 \cdot 10^{14}$ Bq of ^{90}Sr and $2 \cdot 10^{13}$ Bq of ^{137}Cs.

References

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