

## **SIBYLLA Code: Assessment of Water Bodies Contamination and Doses Received By Population Due To Radioactivity Discharges into the Hydrosphere**

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**Abstract.** SIBYLLA code is designed to compute radionuclides concentration in water and bottom sediments of water bodies that can be affected by nuclear facilities during normal operating conditions or accidents. Also SIBYLLA enables conservative assessments of doses resulted from public use of the water-bodies including water-supply. The code contains set of models for water bodies of different types (lakes, ponds, rivers, water-reservoirs, etc). SIBYLLA can be used for radiation safety assessment of a nuclear facility on the all stages of its lifecycle: design, construction, operation, decommissioning. Wide range of sources of radioactive contamination and pathways to the water-bodies can be taken into account – fallouts from the atmosphere, discharges, leakages, wash-out from contaminated catchments, waters of contaminated tributaries.

The code was validated against data on radioactive contamination of eight water-bodies of three different types: the Kiev water-reservoir, a river and three lakes contaminated in the result of Chernobyl accident; the Tygish Lake situated on the axis of East Ural Radioactive Trace; the Techa River; the Tom River influenced by discharges of Siberian Chemical Combine.

The quality of modelling by SIBYLLA code is confirmed by expert council for software accreditation of ROSTECHNADZOR (Federal Environmental, Industrial and Nuclear Supervision Service of Russia), where SIBYLLA was certified in 2016.

**Key Words:** Modelling, radioactive contamination of water-bodies, doses of radioactive exposure, program code.

### **1. Introduction**

SIBYLLA code is designed to calculate concentration of radionuclides in water and bed sediments of rivers, canals, lakes, ponds and water-reservoirs as so as to asses conservatively doses received by population due to use of water-bodies including water supply.

Modelling is enabled both for normal operation of a nuclear facility and for accidents.

Different types of sources of radioactive contamination and pathways to the water-bodies can be taken onto account – fallouts from the atmosphere, discharges, leakages, wash-out from contaminated catchments, waters of contaminated tributaries, and other types of punctual and spatial sources. SIBYLLA can take into account instantaneous sources as so as sources of constant or variable intensity.

The code can be used on the all stages of a nuclear facility lifecycle: design, construction, operation, decommissioning. Also SIBYLLA can be used to support-decision making in case of an accident on a nuclear facility.

SIBYLLA enables modelling for rivers, lakes (ponds) that can be considered perfectly mixed and large lakes that can be subdivided on several parts each of which can be considered

perfectly mixed. The code also can be used for modelling of water systems consisting of the water-bodies of types mentioned above.

Internal exposure pathways taken into account include inhalation of vapour containing tritium, consumption of drinking water, fish, agricultural products that can be contaminated due to watering of cattle or use of contaminated irrigated lands or flood-lands.

External exposure pathways taken into account are swimming, fishing, being at boats, irrigated lands and flood-lands or in the vicinity of water-bodies.

## **2. Brief Description of Included Radioactivity Migration Models**

All of the included models are based on well-known two-chamber model. The chambers are water column and effective layer of bed sediments. The model takes into account radioactive decay, sorption and desorption of radionuclides on suspended particles and bed sediments, sedimentation and resuspension of contaminated suspended matter, diffusion on the water - bottom interface, outflow of radioactivity from a water-body with flowing water or due to technological losses, filtration of radionuclides through bottom and banks.

The models enable computation in cases when values of input parameters are arbitrary functions of time given with the aid of piecewise functions.

Main assumptions of the model are:

- Instant and uniform mixing of radionuclides inside each of the chambers.
- Processes of sorption and desorption of radionuclides are instant, reversible and can be described with a linear function with distribution coefficients. Process of migration of radionuclides can be described by system of differential equations of first order.
- The effective layer is principal for processes of exchange between water column and bed sediments. Thickness of the layer can be measured or estimated.
- Migration of radionuclides on suspended matter consisting of fractions of different grain-size can be described using one fraction with effective grain-size and sorption characteristics.
- Migration of radionuclides on biota can be neglected as quantity of radionuclides accumulated in biota is small comparing to quantity accumulated in abiotic components of a water-body.

SIBYLLA includes three models based on the core two-chamber model: model of a perfectly mixed lake (pond), model of a river (canal), model of a large lake that can be subdivided on several parts each of which can be considered perfectly mixed.

The perfectly mixed lake model is essentially two-chamber. It assumes instant and uniform mixing of radioactivity in the water of the entire lake. A lake (or a pond) can be considered perfectly mixed if characteristic time of water mixing inside the lake is less than modelling time and than characteristic residence time of water in the lake.

The large lake model is multy-chamber. A lake is subdivided on several parts each of which is described by two chambers: water column and bed sediments. Advection and diffusion on borders between the parts of the lake is also taken into account.

The river model is bases on assumption of uniform distribution of radionuclides in any river cross-section (1D model). Each of the cells of computational grid is represented by two chambers: water column and bed sediments. For river sections that can not be considered perfectly mixed SIBYLLA includes a sub-model to estimate conservatively (in accordance

with [1]) maximum concentration of a radionuclide in any cross-section. This enables assessment of doses received by population for river segments located close to a discharge point.

### 3. Supporting Database

Database of the code contains values of necessary constants and information which can be used for parameterisation in case of insufficient input data (see Table I):

- Radioactive decay constants.
- Typical factors of radionuclides wash-off from contaminated basins.
- Typical concentration of suspended matter in water and typical density of bed sediments (dry weight) depending of type of bed sediments (pebble, sand, 3 types of silt).
- Typical values of distribution coefficients between water and suspended matter and between pore water and bed sediments (depending on type of bed sediments).
- Concentration factors for fish, milk, meat and vegetables.
- Average consumption of water, fish, meat, milk and vegetables by representatives of different groups of population.
- Dose coefficients (effective dose and organs / tissues) for different age groups.
- Typical annual occupation time for swimming, fishing, being at boats, irrigated lands, flood-lands or in the vicinity of water-bodies.

TABLE I: LIST OF RADIONUCLIDES PRESENT IN THE SUPPORTING DATABASE

Group	List of radionuclides
All above mentioned data is present	$^{57}\text{Co}$ , $^{58}\text{Co}$ , $^{60}\text{Co}$ , $^{89}\text{Sr}$ , $^{90}\text{Sr}$ , $^{95}\text{Zr}$ , $^{95}\text{Nb}$ , $^{103}\text{Ru}$ , $^{106}\text{Ru}$ , $^{125}\text{Sb}$ , $^{134}\text{Cs}$ , $^{137}\text{Cs}$ , $^{141}\text{Ce}$ , $^{144}\text{Ce}$ , $^{239}\text{Pu}$ , $^{240}\text{Pu}$ , $^{241}\text{Pu}$
Typical value of factor of wash-off from contaminated basin is missing	$^3\text{H}$ , $^{14}\text{C}$ , $^{22}\text{Na}$ , $^{24}\text{Na}$ , $^{46}\text{Sc}$ , $^{51}\text{Cr}$ , $^{54}\text{Mn}$ , $^{55}\text{Fe}$ , $^{59}\text{Fe}$ , $^{64}\text{Cu}$ , $^{65}\text{Zn}$ , $^{76}\text{As}$ , $^{110\text{m}}\text{Ag}$ , $^{129}\text{I}$ , $^{131}\text{I}$ , $^{132}\text{I}$ , $^{133}\text{I}$ , $^{152}\text{Eu}$ , $^{154}\text{Eu}$ , $^{155}\text{Eu}$ , $^{226}\text{Ra}$ , $^{228}\text{Th}$ , $^{230}\text{Th}$ , $^{231}\text{Th}$ , $^{232}\text{Th}$ , $^{234}\text{Th}$ , $^{234}\text{U}$ , $^{235}\text{U}$ , $^{238}\text{U}$ , $^{239}\text{Np}$ , $^{241}\text{Am}$ , $^{242}\text{Cm}$ , $^{244}\text{Cm}$
Typical value of wash-off factor is missing as so as concentration factors for food mentioned above	$^{99}\text{Mo}$ , $^{140}\text{Ba}$ , $^{140}\text{La}$
Typical values of distribution coefficients, wash-off factor and concentration factors for food are missing	$^{129\text{m}}\text{Te}$ , $^{131\text{m}}\text{Te}$ , $^{132}\text{Te}$

### 4. Validation of Radionuclides Transport Models against Observed Data

SIBYLLA was validated against observed data of radioactive contamination of eight water-bodies of three types. The two-chamber model was used for lakes Svyatoe, Tygish, Esthwaite

and Windermere. The Kiev Reservoir was modelled by the multy-chamber model. The river model was used for rivers Plava, Techa and Tom.

#### 4.1. Validation against Observed $^{90}\text{Sr}$ Concentration in Water of the Tygish Lake

The Tygish Lake is located almost on the axis of the East Urals Radioactive Trace resulted from the accidental explosion of 29.09.1957. Area of the lake ( $6.7 \text{ km}^2$ ), average depth (2 m) and values of other input parameters were taken in accordance with [2].

In [2] one can find data of  $^{90}\text{Sr}$  concentration in summer months of 1960-1996. The concentration decreased 23.3 times from the peak of 1964 to the summer of 1993. The decrease is much more intense than can be explained by radioactive decay only.

Outflow from the lake is almost zero. Thus the concentration decrease must be associated with accumulation of  $^{90}\text{Sr}$  in bed sediments due to sedimentation and diffusion to deeper layers of the bed sediments.

In [2] one can find a statistically obtained regression model for concentration of  $^{90}\text{Sr}$  in water:

$$C_w = 12,66 \cdot \exp(-0,077 \cdot t); R^2=0,86$$

Where  $t$  – time, year;  $R$  – correlation factor.

Modelling with the use of SIBYLLA code was performed for period of 1964 –1996 (see FIG. 1.). 1964 data were used as initial conditions.

Statistically obtained regression model for concentrations computed by SIBYLLA is as follows:

$$C_w = 9,49 \cdot \exp(-0,072 \cdot t); R^2=0,985$$

One can see that self-purification factor obtain on the base of the modelling results ( $0,072 \text{ year}^{-1}$ ) is close to one obtained on the base of the observed data ( $0,077 \text{ year}^{-1}$ ). This indicates that model adequately describes main processes in the lake.

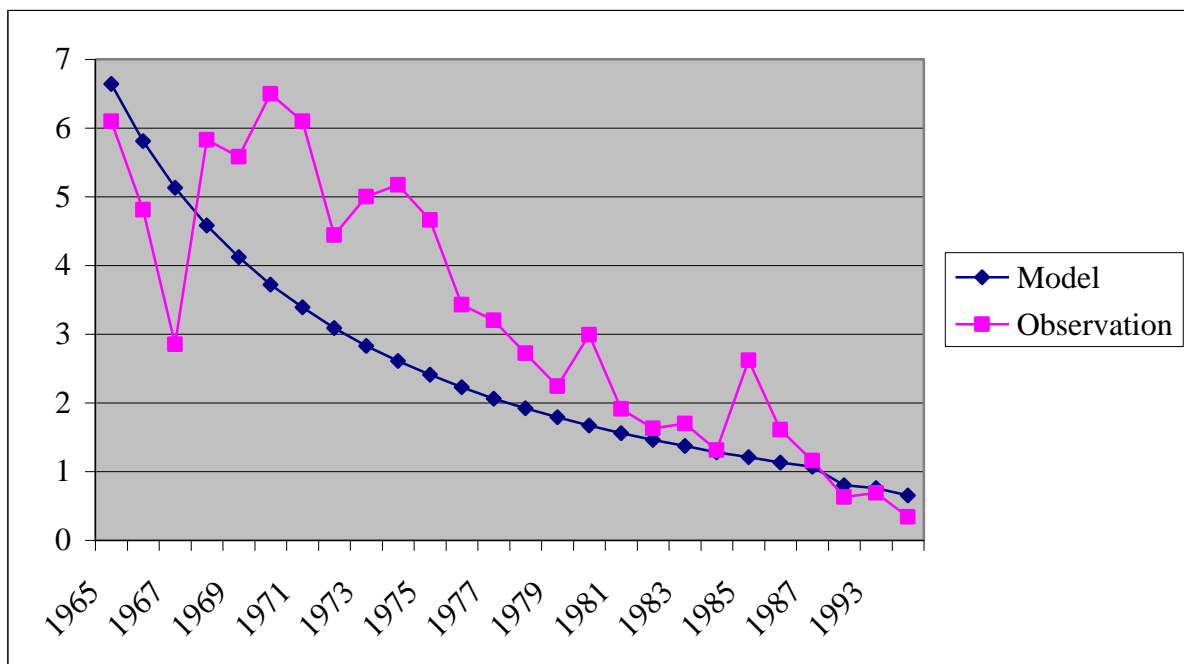


FIG. 1.  $^{90}\text{Sr}$  concentration in water of the Tygish Lake, Bq/l.

#### 4.2. Validation against Observed $^{137}\text{Cs}$ Concentration in Lakes Svyatoye, Esthwaite and Windermere.

Lakes Svyatoye (0.23 km<sup>2</sup>) [3, 4], Esthwaite (1 km<sup>2</sup>) and Windermere (8 km<sup>2</sup>) [5] were contaminated as the result of the Chernobyl fallout. Modelling results for Esthwaite Lake one can see below at FIG. 2 (water) and 3 (bed sediments). The results for Windermere Lake one can see at FIG. 4 (water) and 5 (bed sediments).

One can see that computed results of  $^{137}\text{Cs}$  concentration in water are in good accordance with observed data. As for bed sediments the difference is more significant but does not exceed one order of magnitude.

The analysis of observed concentrations in bed sediments shows that discrepancies between different measurements in the same water-body performed approximately in the same time can be very significant (in some cases an order of magnitude). And some of the discrepancies can not be explained by distance between the sampling locations itself. The main reason is that concentration of radionuclides in sediments can vary significantly in space depending on the type of sediments at a sampling location.

Additional uncertainty is added by uncertainty of bed sediments density (which can differ by several times) as SIBYLLA computes concentration per unit volume while measurement results are provided per unit mass. However this uncertainty does not influence accuracy of assessments of doses as for assessments of doses resulted from being at contaminated surface (banks, flood-lands) specific activity of radionuclides per unit area is important.

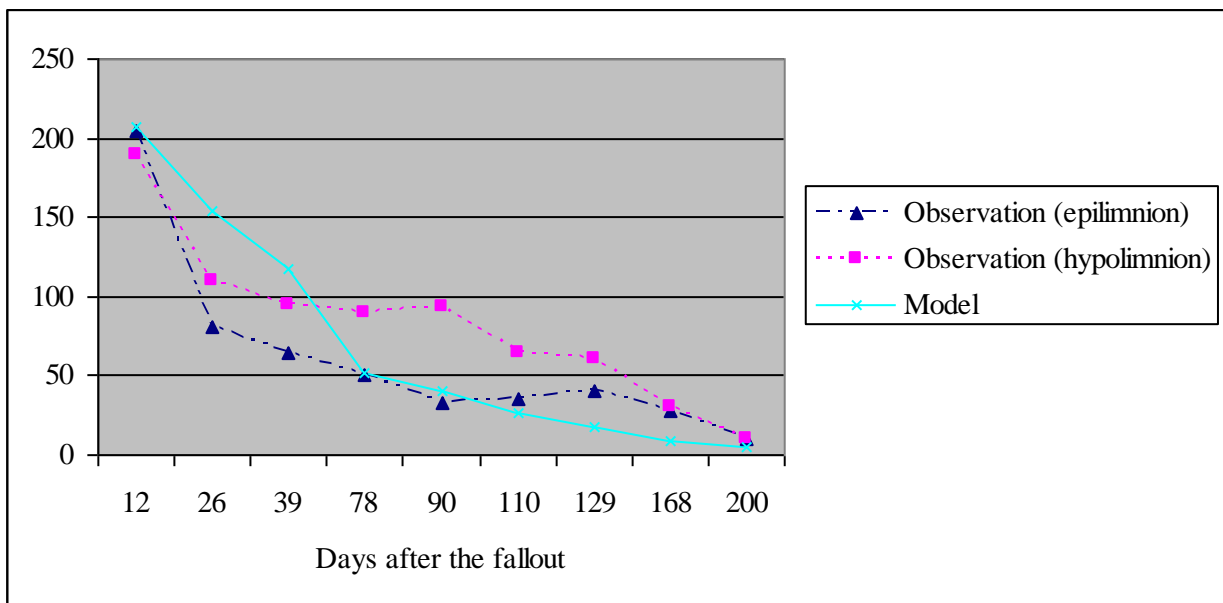


FIG. 2.  $^{137}\text{Cs}$  concentration in water of the Esthwaite Lake, Bq/m<sup>3</sup>.

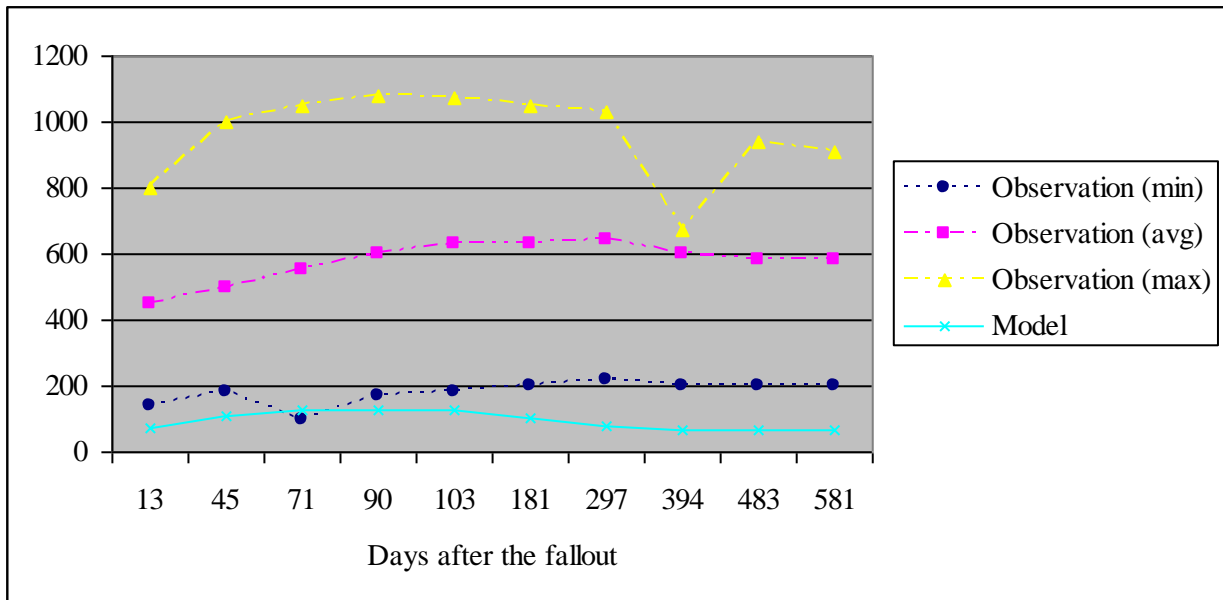


FIG. 3.  $^{137}\text{Cs}$  concentration in bed sediments of the Esthwaite Lake, Bq/kg.

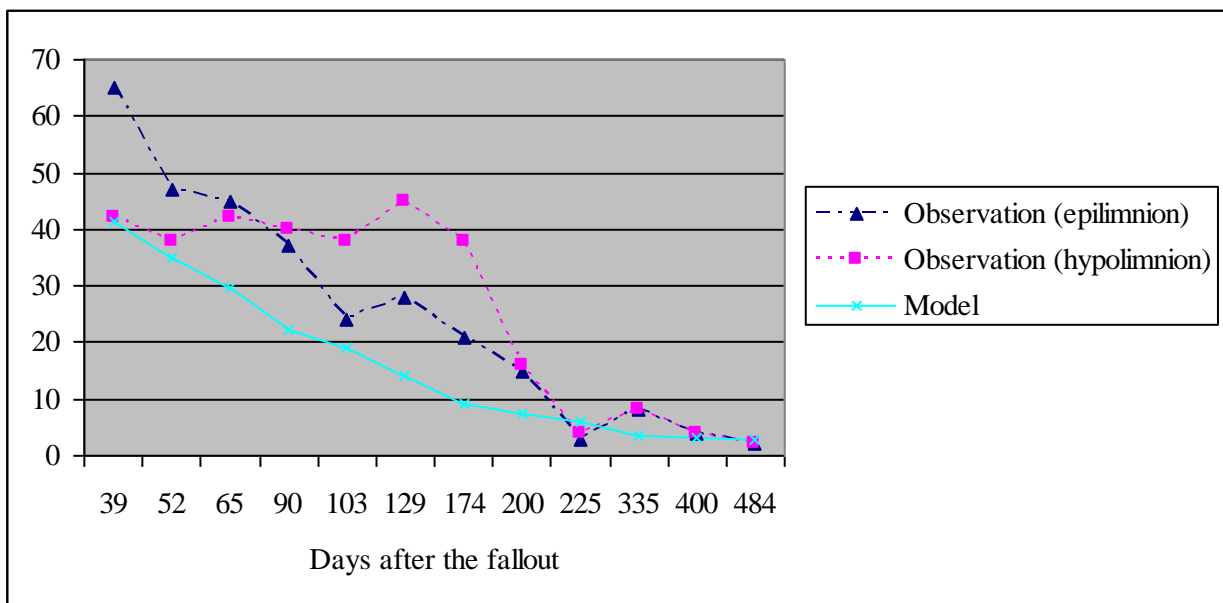


FIG. 4.  $^{137}\text{Cs}$  concentration in water of the Windermere Lake, Bq/m<sup>3</sup>.

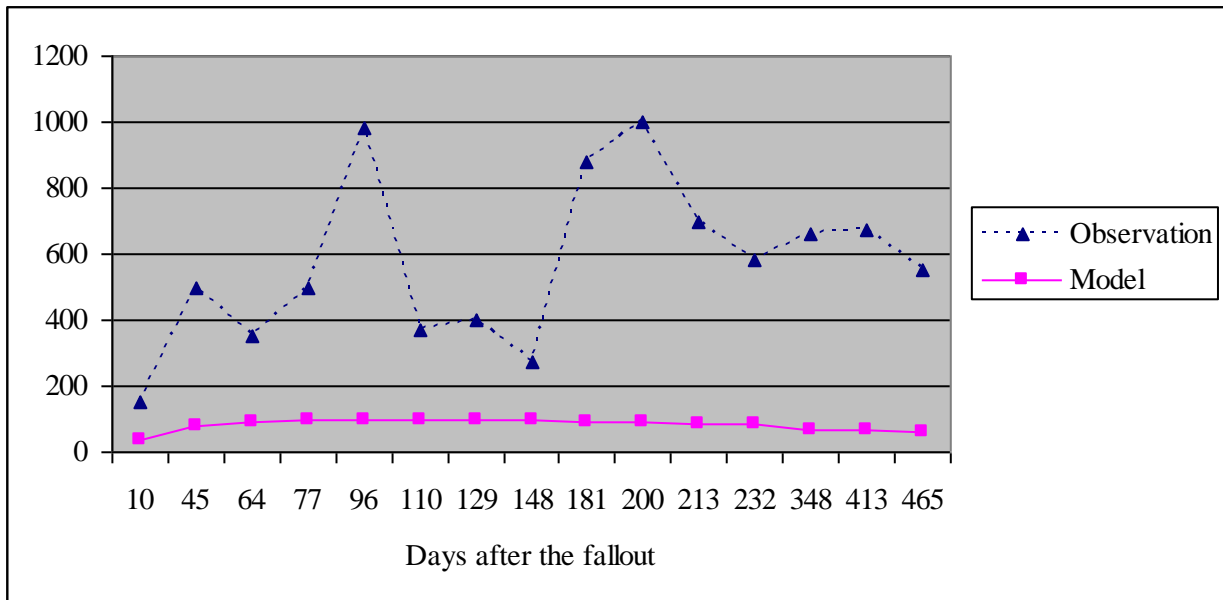


FIG. 5. <sup>137</sup>Cs concentration in bed sediments of the Windermere Lake Bq/kg.

#### 4.3. Validation against Observed <sup>137</sup>Cs Concentration in Water of Plava River

As a result of Chernobyl fallout the so called «Tula Radioactive Trace» was formed in the Tula region of Russia. The Plava River crosses the trace. In [6] one can find observed data of <sup>137</sup>Cs concentration in water and bed sediments in July (low water) and October (period of rains) of 1992. Using July data as initial conditions, concentration <sup>137</sup>Cs in October was computed for a river segment 56.5 km long. The segment includes settlements Mescherino (starting point), Sergieva Sloboda (18.5 km), Krapivna (56.5 km). Data of the Plava River basin contamination were taken from [7].

Difference between modelling results (Sergieva Sloboda 16.8 Bq/m<sup>3</sup>; Krapivna 5.77 Bq/m<sup>3</sup>) and observed data (Sergieva Sloboda 17.39±9.0 Bq/m<sup>3</sup>; Krapivna 7.77±3.9 Bq/m<sup>3</sup>) are within the error of measurements.

#### 4.4. Validation against Observed <sup>239</sup>Np Concentration in Water of the Tom River

Discharges of the Siberian Chemical Combine (SCC) in the Tom River take place through its right-hand tributary — the Romashka River. In [8] one can find observed concentrations of <sup>239</sup>Np on suspended matter of the Tom River in August 2008 for two locations near the right bank: Location 1 (8 km downstream from the Romashka mouth - 106±5 Bq/m<sup>3</sup>), Location 2 (20 km - 43±3 Bq/m<sup>3</sup>). Direct-flow reactors that were the main source of <sup>239</sup>Np discharges were stopped 05.07.2008.

On the first stage of validation concentration in Location 1 was computed for unitary (1 Bq/s) discharge of <sup>239</sup>Np. The result was used to assess intensity of discharges. The assessed intensity (1.29 · 10<sup>13</sup> Bq/year) is in sufficiently good accordance with observed data (see Table II).

After that on the base of assessed intensity of discharges concentration in the Location 2 was computed (32.8 Bq/m<sup>3</sup>). The value is also in sufficiently good accordance with observed data (43±3 Bq/m<sup>3</sup>).

TABLE II: ANNUAL DISCHARGES OF <sup>239</sup>NP FROM SCC TO THE TOM RIVER, BQ [8, 9].

2002	2003	2004	2005	2006	2007	2008
$8,14 \cdot 10^{12}$	$6,23 \cdot 10^{12}$	$7,51 \cdot 10^{12}$	$1,30 \cdot 10^{13}$	$1,46 \cdot 10^{13}$	$12,6 \cdot 10^{12}$	$7,15 \cdot 10^{12}$

#### 4.5. Validation against Observed $^{137}\text{Cs}$ Concentration in the Kiev Reservoir

Two main pathways of the Kiev Reservoir contamination by  $^{137}\text{Cs}$  were taken into account. First one is the direct fallout on the surface of the reservoir in the days following the Chernobyl accident [10]. Second is the inflow of contaminated river water (Dnieper and Pripyat) in the following months [10]. The Kiev Reservoir was subdivided into 5 parts for the multy-chamber model. One can see that modelling results are in sufficiently good accordance with data observed in October 1986 (see Table III). In 50% cases the difference are within the measurement errors. None of the modelling results differ from the observed data more than 3 times.

TABLE III:  $^{137}\text{CS}$  CONCENTRATION IN DIFFERENT PARTS OF THE KIEV RESERVOIR (FROM NORTH TO SOUTH) IN OCTOBER 1986.

Symbolical designation of the part of the reservoir	Bed sediments, Bq/kg		Water, Bq/m <sup>3</sup>	
	Observation	Model	Observation	Model
Upper path of the reservoir	12210±4070	11618	1036±370	421
«Strakholesie»	7770±2590	6722	-	207
«Sukholuchie»	5180±1850	5725	281±102	144
«Hlebovka»	4440±1480	4515	385±133	154
Lower path of the reservoir	2950±740	4189	414±133	159

#### 4.6. Validation against Observed $^{137}\text{Cs}$ and $^{239,240}\text{Pu}$ Concentrations in the Techa River

At present the dam 11 is considered to be the Techa River head. In [11, 12] one can find observed data on radioactive content for following cross-sections of the river: 44 km from the river head (Muslyumovo), 78 km (Brodokalmak), 207 km (Zatechenskoye) (see Table IV). Assessments of hydrological characteristics of the river and estimations of values of coefficients influencing migration of the radionuclides are also adduced in [11, 12].

Modelling was performed for the part of the river from 44 km to 207 km with assumption that values of model parameters does not vary in time (steady-state approximation). Observed concentrations at Muslyumovo were used as boundary conditions. Results are shown in Table IV. One can see that modelling results of  $^{137}\text{Cs}$  concentration differ from observations 1.3 – 2 times. As for  $^{239,240}\text{Pu}$  the difference is 1.1 – 2.3 times. The differences do not exceed uncertainty of the input data. First, measurement error of concentration in water of the Techa River was 14 - 50% for  $^{137}\text{Cs}$  and 19 - 33% for  $^{239,240}\text{Pu}$  [11]. Second, uncertainty of characteristics of the river is also significant. For example discharge of the river can vary 10 times within a year. Inter-year variability is also significant. For example annual discharge in 2007 and 2009 differ almost 2.5 times. Velocity of the river flow and depth also vary significantly along the river and in time.

TABLE IV: TECHA RIVER. MODELLING RESULTS AND OBSERVED DATA.



Concentration		78-кМ		143-кМ		207-кМ	
		<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>239,240</sup> Pu	<sup>137</sup> Cs	<sup>239,240</sup> Pu	<sup>137</sup> Cs
Water, Bq/m <sup>3</sup>	Observation	0,16±0,05	310±110	0,12±0,04	120±30	0,073±0,024	70±10
	Model	0,21	279	0,118	144,8	0,067	74,4
Bed sediments, Bq/kg	Observation	17±6	-	3,2±1,1	590±170	1,2±0,7	200±70
	Model	2,92	3847	1,63	729	1,10	213

## 5. Conclusions

It was performed 63 comparisons of computed and observed concentrations in water (see FIG. 6. and Table V). Relative standard deviation of modelling results from observed data is 48.9%. The computed concentrations of radionuclides in water are in good accordance with observed data. The deviation of modelling results in both directions are equiprobable enough (55.6% and 44.4%). This let one assume that SIBYLLA provides unbiased assessments.

The accuracy of modelling is acceptable for assessments of doses from public use of the water-bodies. The quality of modelling by SIBYLLA code is confirmed by expert council for software accreditation of ROSTECHNADZOR (Federal Environmental, Industrial and Nuclear Supervision Service of Russia), where SIBYLLA was certified in 2016.

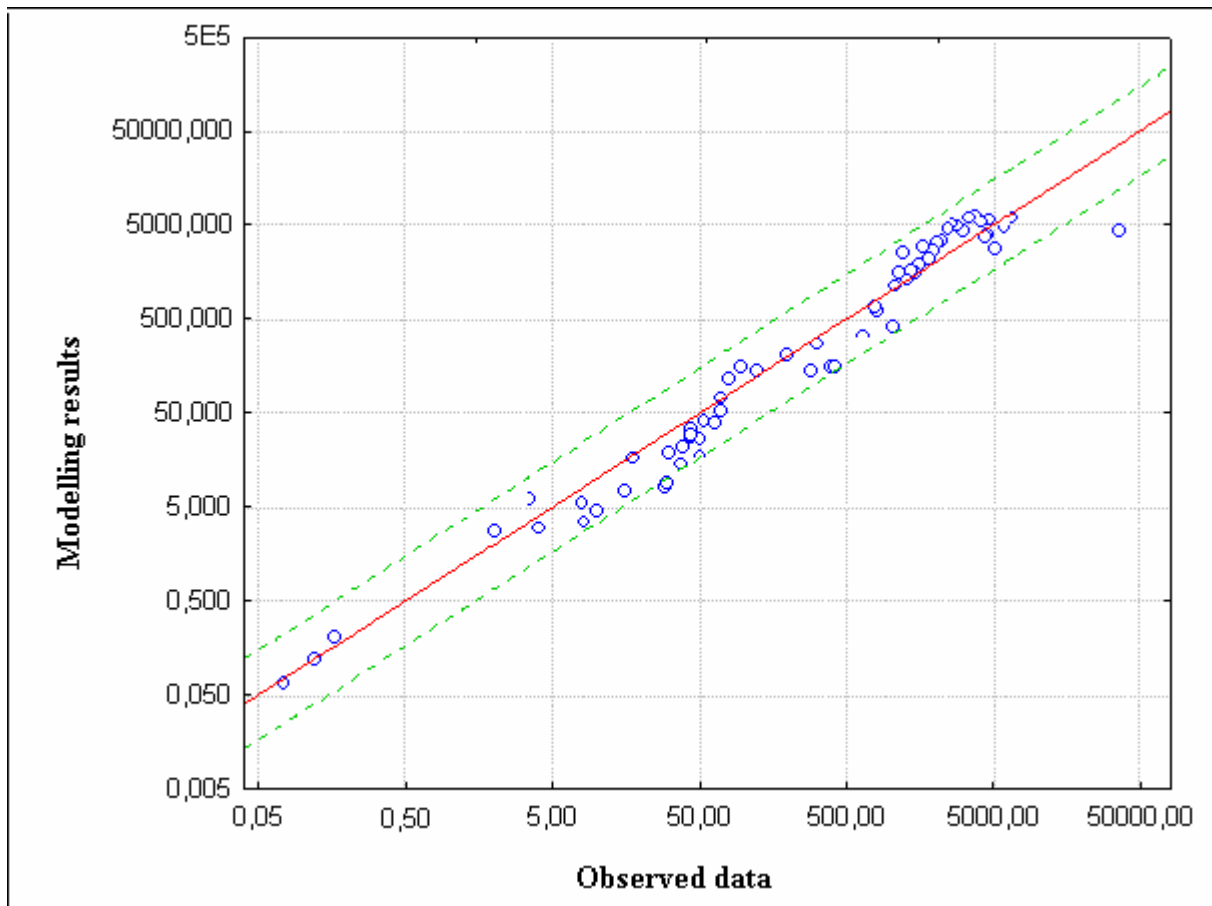


FIG. 6. Validation of modelling results against observed concentration in water, Bq/m<sup>3</sup>.

TABLE V: DIFFERENCE BETWEEN MODELLING RESULTS AND OBSERVED CONCENTRATION IN WATER.

Difference	> 1.5 times	> 2 times	> 3 times	> 5 times	> 10 times
Portion of cases	47,6%	19,0%	4,76%	1,59%	0%

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