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“Blind” testing of models for predicting the ^{90}Sr activity concentration in river systems using post-Chernobyl monitoring data

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Abstract

Two different models for predicting the time-dependent mobility of ^{90}Sr in river systems have been evaluated using post-Chernobyl monitoring data for five large Belarusian rivers (Dnieper, Pripyat, Sozh, Besed and Iput) in the period between 1990 and 2004. The results of model predictions are shown to be in good agreement (within a factor of 5) with the measurements of ^{90}Sr activity concentration in river waters over a long period of time after the accident. This verifies the relatively good accuracy of the generalised input parameters of these models which were derived primarily from measurements of ^{90}Sr deposited after atmospheric nuclear weapons testing (NWT). For the cases studied here, the simpler AQUASCOPE model performed just as well as the more complex “Global” model which used GIS-based catchment data as an input. The reasons for this are discussed. Exponential decay equations were also curve-fitted to the data for each river to help assess the uncertainties in the predictive models.

Keywords: ^{90}Sr; Model; Chernobyl; River; Pripyat; Weapons testing; Catchment; Runoff

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1. Introduction

A large-scale spatial model (termed here the "Global" model) and a simplified spreadsheet-based model (the "AQUASCOPE" model) have previously been developed to predict the long-term mobility of radioisotopes in river catchments (Smith et al., 2004, 2005). These models were parameterized using a very large data set of ^{90}Sr measurements from European and Asian rivers after atmospheric nuclear weapons testing (NWT). The models are based on studies of the changes in the transport of radioisotopes from the catchments of rivers as a function of time (Monte, 1995; Smith et al., 2000; Cross et al., 2002). Models for the transport of radioisotopes from catchments to rivers have recently been reviewed by Monte et al. (2004).

Values of input parameters for the Global model were obtained using correlations between runoff rates of radioisotopes and the characteristics of each catchment (particularly the catchment coverage by wetland and organic soils). Previous studies have shown that the long-term environmental mobility of radiocaesium and radiostrontium is primarily dependent on the characteristics of the surrounding catchment (Monte, 1997; Smith et al., 2000, 2004). Assessment of these catchment characteristics allows input parameters to be determined for application to new rivers.

The AQUASCOPE model (Smith et al., 2005) uses a simpler approach to predict the runoff of ^{90}Sr from different catchments. In this model, catchments are divided into two categories based on the organic matter content of their soils: "organic" and "mineral".

The aim of this investigation is to test the predictions of these two generalised models against newly available data on ^{90}Sr activity concentrations in five Belarusian rivers and their catchments.

2. Materials and methods

2.1. Mathematical modelling

The mathematical models are based on the assumption that runoff rates of radioisotopes to surface waters are characterised by a fast decrease of recently deposited activity followed by slower transfers during subsequent years. They can be described by the following exponential transfer function (Smith et al., 2004):

$$W_C(t) = \theta \int_{-\infty}^{t} D(\tau)\left(A_1\,\mathrm{e}^{-(\lambda+k_1)(t-\tau)} + A_2\,\mathrm{e}^{-(\lambda+k_2)(t-\tau)} + A_3\,\mathrm{e}^{-(\lambda+k_3)(t-\tau)}\right)\mathrm{d}\tau \qquad (1)$$

where $W_C(t)$ is the concentration of the radionuclide in the runoff water (Bq m^{-3}) at time t and $D(t)$ (Bq m^{-2}) is the radioisotope deposition. A_1, A_2 and A_3 are, independent coefficients, the fast decrease of activity for the first few weeks after initial fallout (at rate k_1), the slow decrease (at rate k_2, timescale years), and a very long-term (at rate k_3, timescale years-decades) runoff fraction, respectively. The scaling factor, θ (m^{-1}), is used to correlate radioisotopes' runoff with characteristics of river catchments in the Global model. The scaling factor is equal to the normalised activity radioisotopes in river water at time zero. λ is the physical decay constant of the radioisotope. Model parameter values are summarised in Table 1.

The Global model studied the relationship between ^{90}Sr runoff and the characteristics of each river catchment, including mean slope, soil texture, mean % carbon content of soils, mean annual precipitation, exchangeable Ca and K content, % of forest areas, % of "*inland water*", % of agricultural lands and Compound Topographic Index (CTI), known as the wetness index. These catchment data were obtained from digital data sets with global coverage which are available online (Post et al., 1982; Verdin and Greenlee, 1996; Batjes, 1996).

Table 1
Parameters for AQUASCOPE and Global models

User input parameters	
D_c (Bq m^{-2})	Deposition to catchment required for all models
AQUASCOPE	Classify catchment as “organic” or “mineral”. $\theta = 1$ m^{-1}
Global model	% Inland water land cover class from GIS. Calculate θ from Eq. (2)

“AQUASCOPE” model parameters

	A_1	A_2	A_3	k_1 (y^{-1})	k_2 (y^{-1})	k_3 (y^{-1})
Organic catchment	0.8	0.03	0.005	16	0.09	0
Mineral catchment	0.8	0.005	0.003	16	0.09	0

“Global” model parameters

	A_1	A_2	A_3	k_1 (y^{-1})	k_2 (y^{-1})	k_3 (y^{-1})
	0.984	0.0123	0.0037	16	0.09	0

θ calculated from Eq. (2) using land cover info from GIS data

	Dnieper	Sozh	Besed	Iput	Pripyat (Mozyr)
Inland water (%)	0.26	0	0	0	0.76
θ (m^{-1})	0.59	0.55	0.55	0.55	0.66

Previous results (Smith et al., 2004) of correlations between characteristics of river catchments and scaling factors, θ (m^{-1}), showed a highly significant positive correlation ($r = 0.84$ for ^{90}Sr NWT) with the “*inland water*” land cover category:

$$\theta = 0.141\,(\%\text{ of ``inland water''}) + 0.55 \tag{2}$$

The category indicates all forms of surface standing or running water in catchments. In the case of the Global model, this relationship was used to predict the value of θ for the Belarusian rivers studied here (Table 1).

The simplified AQUASCOPE models are based on similar exponential transfer functions and model parameters were estimated from the same database of both ^{137}Cs and ^{90}Sr in river waters in a wide range of European countries after different fallout events (Smith et al., 2005). The AQUASCOPE model parameters, however, do not include as much information concerning catchment characteristics as the Global GIS-based model. The parameter values for the AQUASCOPE model were simply determined by fitting exponential functions to measurements of radioisotope runoff in various river catchments having different coverages of organic, boggy soils. The catchments are classified as either “organic” or “mineral”. In this study, all of the catchments were classed as mineral except the Pripyat for which the classification was organic, based on (independent) evaluation of the soil types in the catchment. On the basis of the worldwide organic soil carbon data set (Post et al., 1982), however, the mean organic carbon content is 4.82% which would define it as “mineral” in the AQUASCOPE model (less than 10% mean organic carbon content; Smith et al., 2005). This is discussed further in Section 3 below.

2.2. *River ^{90}Sr concentrations and deposition data*

The monitoring data of ^{90}Sr activity in river waters of the Dnieper, Pripyat, Sozh, Besed and Iput after the Chernobyl fallout were obtained by workers at the laboratory of Hydrogeology NAS Belarus and the Belarusian Committee on Hydrometeorology (Kudelsky et al., 1997, 2002). The sampling points are shown in Fig. 1. These data of ^{90}Sr activity in river water include only the dissolved form which is the dominant form due to the high solubility of this radioisotope. Samples of river waters have been taken four times per year in different seasons but only average annual values of ^{90}Sr activity for each river were used for this study (Table 2).

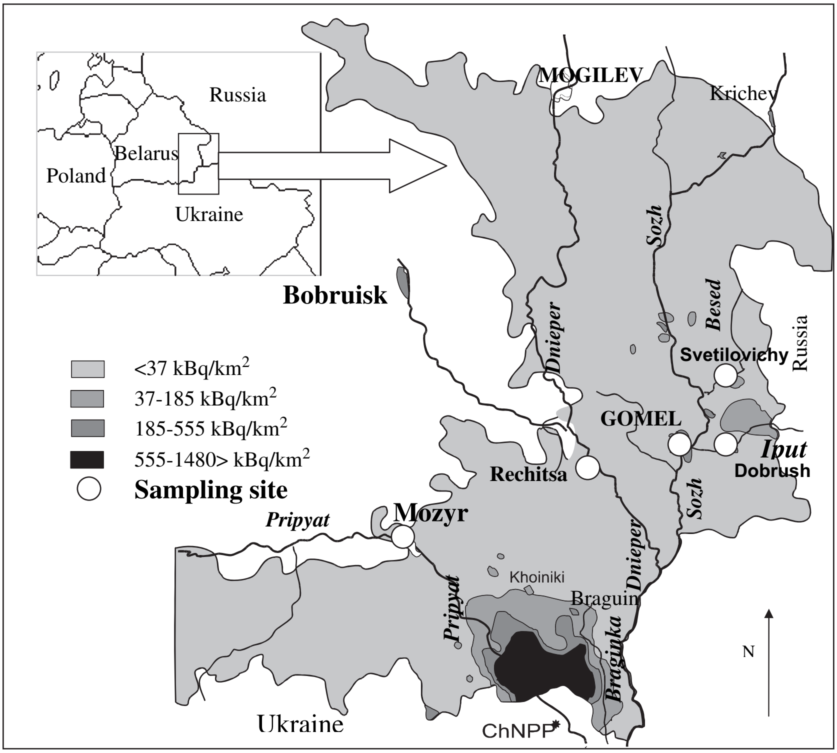


Fig. 1. Map of areas of Belarus contaminated by ^{90}Sr with river water sampling points indicated. The Chernobyl Nuclear Power Plant (Ch NPP) is in Ukraine, just south of the Belarus–Ukraine border.

Table 2
^{90}Sr activity concentration in the investigated river systems

Year	River and sampling point						
	Braginka (Gden)	Pripyat (Chernobyl)	Dnieper (Rechitsa)	Pripyat (Mozyr)	Sozh (Gomel)	Iput (Dobrush)	Besed (Svetilovichy)
Annual average ^{90}Sr activity concentration in river waters (Bq m^{-3})							
1990	216	780	46	83	111	136	231
1991	490	1000	37	37	70	123	197
1992	687	440	28	30	42	50	66
1993	1197	850	18	32	65	65	120
1994	2200	930	21	32	46	56	83
1995	1490	330	21	17	30	56	185
1996	1665	340	18		28	30	32
1997	1140	250	22		38	34	48
1998	2040	300	20		40	70	60
1999	2675	500					
2001					28	13	
2002	730				49		45
2003	1700		9		26	43	38
2004	2550		24	15	23	31	35

The Braginka and Pripyat (Chernobyl) data sets were not used (see text).

All sampling points of the ^{90}Sr activity are situated outside the Chernobyl 30 km zone. We excluded from the study data from the Pripyat at Chernobyl and the Braginka which are both in the exclusion zone. This was because in the former case, the models assume an approximately homogeneous distribution of fallout in the river catchment, but the Pripyat at Chernobyl receives significant inputs from the very small part of the catchment in the exclusion zone. In the case of the Braginka, the catchment is wholly within the exclusion zone and may be significantly influenced by the dissolution of the very high fraction of ^{90}Sr (ca. 90%) which was deposited in the form of fuel particles. The increase over time in ^{90}Sr activity concentrations in the Braginka (Table 2) is likely to be due to dissolution of these fuel particles and consequent release of ^{90}Sr.

The average ^{90}Sr activity deposition to each catchment $D(t)$ (Bq m^{-2}) was estimated using the average deposition of ^{137}Cs to each catchment (Kudelsky et al., 1998) and the ^{90}Sr:^{137}Cs ratio in fallout. The ratio of ^{90}Sr:^{137}Cs which was deposited depends on the distance from Chernobyl nuclear power plant (Ch NPP) and can be described (Mück et al., 2002) by the following relationship:

$$\frac{^{90}\mathrm{Sr}}{^{137}\mathrm{Cs}} = 0.37\,\mathrm{e}^{-0.017d} \tag{3}$$

where d is distance from Chernobyl Nuclear Power Plant (Ch NPP) in km. Note that the deposition of ^{90}Sr relative to ^{137}Cs is also dependent on direction from the reactor (Mück et al., 2002). For these catchments outside the 30-km zone and to the north of the reactor, no correction to Eq. (3) for direction is needed (Mück et al., 2002), though there remain significant uncertainties in the estimation of ^{90}Sr fallout which play an important role in overall model uncertainty.

For the Global model, scaling factors, θ (m^{-1}), were calculated by inputting values of the percentage "*inland water*" land cover class for each catchment into Eq. (2). For the AQUASCOPE model, all river catchments were classed as "mineral" except the Pripyat. The Pripyat catchment has a high degree of coverage by organic soils.

Note that no ^{90}Sr activity data are available for the first weeks after fallout, therefore the data could not test the fast runoff (first exponential term) part of the models.

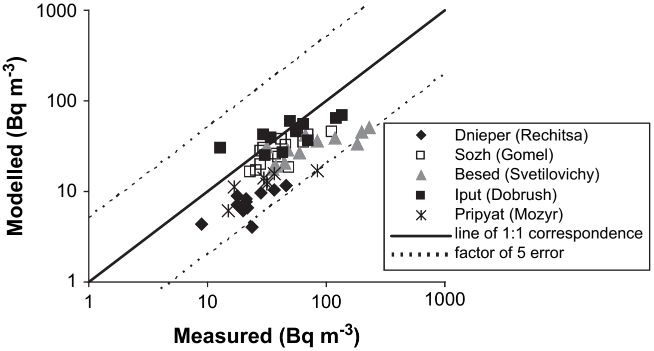
3. Results and discussion

The best method of assessing the quality of predictive models is by the "blind" comparison of predictions with measured data. Fig. 2a shows such comparison for the Global model. The model gave good predictions of the ^{90}Sr activity concentrations in these rivers: 95% of predictions fall within a factor of 5 of the measured values. However, it should be noted that there appeared to be a bias in the model in that it tended to under-estimate ^{90}Sr activity concentrations (most predicted values were less than measured).

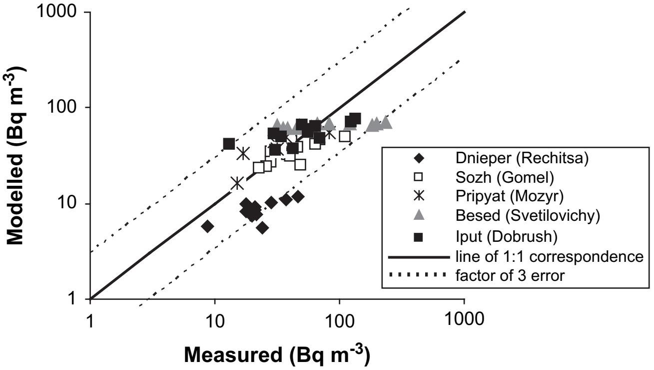
AQUASCOPE model results showed similarly good agreement with measurement data, but there was also a bias towards under-prediction. Almost all values were within the range showing factor of 3 error in model predictions (Fig. 2b). It is perhaps surprising that the simpler AQUASCOPE model performs just as well as the "Global" model since the latter has a more sophisticated way of estimating the runoff of ^{90}Sr in relation to land cover class. There are, however, significant uncertainties in this relation: the inland water land cover class may not be a good indicator of ^{90}Sr runoff in all catchments since the ^{90}Sr runoff is likely to be related to a number of land cover classes. There is, of course, further uncertainty in the use of the global land cover data to estimate actual land cover classes. In the test we have carried out here, increased model complexity has not led to improved predictive power owing to uncertainties in parameter estimation.

In order to illustrate the best possible model performance, we also carried out a curve-fitting exercise by fitting Eq. (1) to the data using a least-squares fit of model parameters to the

(a) Global model



(b) AQUASCOPE model



(c) Fitted model

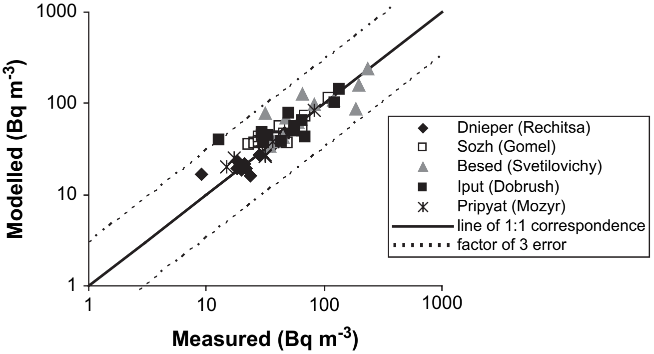


Fig. 2. Validation of (a) Global model; (b) AQUASCOPE model; and (c) Fitted model against ^{90}Sr Chernobyl measured data. The estimated errors are shown by the dotted lines.

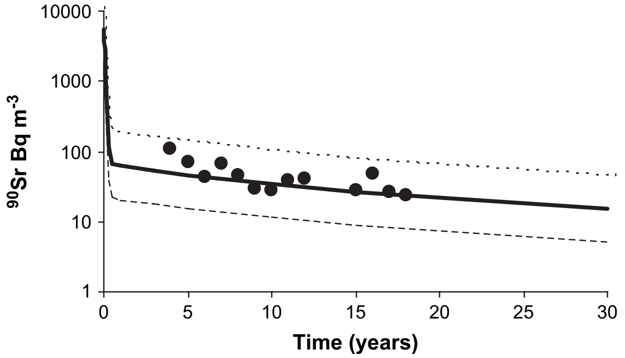
empirical data. The results of this curve-fitting exercise are shown in Fig. 2c. As expected, the Fitted model performed better than the two predictive models indicating that there is still room for improvement in model predictions. Such improvement is, however, likely to be difficult since uncertainty in model predictions is driven largely by uncertainty in input parameter values (these generally applicable models are driven by global parameters which may not capture local site specific conditions accurately) and by uncertainty in estimation of the fallout to each catchment.

Example predictions of the AQUASCOPE model for the Sozh and Pripyat rivers are given in Fig. 3a,b. Fig. 3b shows the different model predictions for the Pripyat assuming that the catchment is classed either as mineral or as organic. Both models give predictions within their respective error ranges, but the ‘‘organic’’ catchment assumption predicts better. It may be that local evaluation of the catchment characteristics (giving ‘‘organic’’ classification) is more accurate than our evaluation based on the world soil carbon data set (giving ‘‘mineral’’). It should, however, be noted that in the world data set the Pripyat does show higher coverage of both organic and sandy soils than the other river catchments – a high proportion of sandy soils may also influence ^{90}Sr runoff.

4. Conclusions

It has been shown that general models can predict radioactivity (in particular ^{90}Sr activity concentrations) with reasonable accuracy in river water over long times after fallout. These models could therefore be useful in predicting the consequences of future fallout events and

(a) River Sozh at Svetilovichy



(b) River Pripyat at Mozyr

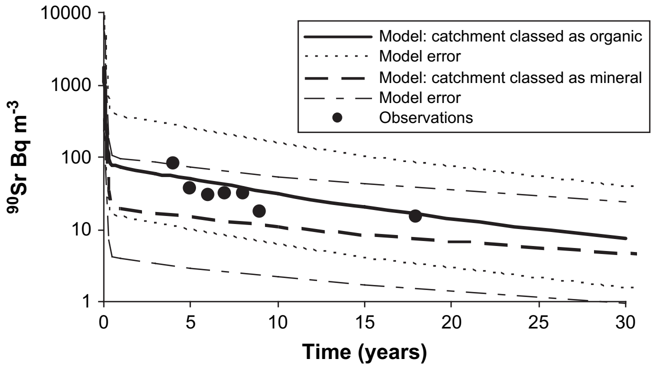


Fig. 3. Examples of values predicted by the AQUASCOPE model compared with ^{90}Sr measured data in (a) the River Sozh at Svetilovichy and (b) the River Pripyat at Mozyr.

risk assessment scenarios. The models are applicable to European rivers and probably other rivers at similar latitudes with similar ranges of catchment characteristics.

It is interesting to note that the simpler AQUASCOPE model performed just as well as the GIS-based Global model – the latter is based on a global data set of catchment coverage by the "inland water" land class category (assumed to be a surrogate of catchment coverage by organic, boggy soils). The AQUASCOPE model was a simplification of this approach where the runoff of ^{90}Sr is based only on an evaluation of the "mineral" or "organic" nature of the catchment. The more complex Global approach has not improved predictive power in this case. This is due to the high uncertainty in the relationship between ^{90}Sr runoff and land cover class as determined by the available global land cover data, and to the high uncertainty in ^{90}Sr deposition estimates.

As we have shown, both modelling approaches can be used to identify whether catchments may be vulnerable to radionuclide deposition, the length of time elevated concentrations might persist, and whether countermeasures would be needed after an accident. The high uncertainties in estimating the ^{90}Sr deposition, and the correct model parameter values, mean that predictions are only within a factor of 5 of the measured data. Given these inevitable uncertainties, however, we think it unlikely that generally applicable predictive models can be made significantly more accurate than this.

It was suggested (Cross et al., 2002; Smith et al., 2004) that the environmental behaviour of ^{90}Sr was similar after the NWT and Chernobyl fallout events (excluding the areas where fuel particles dominated the ^{90}Sr deposition after Chernobyl). This study has confirmed this by showing that the runoff of Chernobyl deposited ^{90}Sr can be accurately predicted using models developed from NWT deposited ^{90}Sr.

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