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Testing models for predicting the behaviour of radionuclides in aquatic systems

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ABSTRACT

The paper describes the main results of the international EMRAS model testing exercise for radionuclide transport in watershed-river and estuarine systems. The exercises included the following scenarios: multi-point source of ^{3}H discharge into the Loire River (France), radioactive contamination of the Dnieper–Southern Boug estuary (Ukraine), remobilisation of radionuclide contamination from the Pripyat River floodplain (Ukraine) following the Chernobyl accident, release of radionuclides into the Techa River (Russia) and behaviour of ^{226}Ra in the Huelva estuary (Spain).

1. Introduction

Several projects have been launched during the last few decades for the purpose of validating the predictions of radioecological models. Such projects have taken advantage of the substantial amount of experimental data gathered following the accidental introduction of radionuclides into the environment (e.g. from the Chernobyl nuclear power plant accident) to test models for assessing the levels of radionuclides in ecosystem components and the human food chain. In particular, the evaluation of models for predicting the behaviour of radionuclides in the fresh water environment has been the object of international projects such as BIOMOVS (BIOspheric Model VAlidation Study), organised by the Swedish National Institute for Radiation Protection (NIRP), and VAMP (VAlidation of Model Predictions) organised by the International Atomic Energy Agency (IAEA). The results of those projects are summarised in many reports (e.g. IAEA, 2000; BIOMOVS, 1990). The projects addressed the radioactive contamination of lakes, ponds and biota (IAEA, 2000; BIOMOVS II, 1996a, b), the wash-off of radionuclides from experimental plots (BIOMOVS II, 1996c) and the transport of radionuclides from contaminated groundwater to rivers (BIOMOVS, 1990).

The BIOMOVS and the VAMP projects included exercises aimed at improving the reliability of models for predicting the migration of ^{137}Cs in lakes and of ^{137}Cs and ^{90}Sr in rivers. However, such co-operative and extensive model validation studies have not been undertaken for other aquatic systems, such as coastal waters, or for other long lived radionuclides of potential radiological importance for freshwater systems.

The validation of models for predicting the behaviour of radionuclides in the freshwater environment and coastal areas was the object of the working group titled: 'Model validation for radionuclide transport in the system watershed-river and in estuaries'; it was a part of the EMRAS (Environmental Modelling for RAdiation Safety) project organised by the IAEA. The working group activities were aimed at filling some of the aforementioned gaps, taking advantage, not only of the contamination data from the Chernobyl accident, but also of data from other contamination events, such as, the discharge of radionuclides into the Techa River and routine releases of radionuclides into the aquatic environment. The project addressed radionuclides (e.g. tritium) and systems (e.g. coastal areas) that were not studied in previous testing projects.

The present paper summarizes the main achievements of the EMRAS aquatic working group.

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2. Material and methods

State-of-the-art models developed by different organisations were tested by comparison of their predictions with observed data. The descriptions of the environmental scenarios were developed by individual members of the working group who also provided the empirical/observed data for model testing. The five exercises were:

2.1. Wash-off of ^{90}Sr and ^{137}Cs deposits from the Pripyat floodplain, Ukraine

Modellers were asked to predict the time dependent water concentrations of the radionuclides in the Pripyat River following the inundation of the river floodplain in the period after the Chernobyl accident. Modellers were provided with data on the deposition of radionuclides on the floodplain, the time dependent levels of radionuclide concentration in the river water entering the floodplain, the water fluxes and several other morphological, meteorological and hydrological data. Concentrations of radionuclides in the water of the Pripyat River downstream of the floodplain were subsequently supplied to the modellers to allow the performances of the models to be evaluated.

2.2. Radionuclide discharge from the Dnieper River (Ukraine) into its estuary (the Dnieper– Southern Boug estuary) in the Black Sea

Modellers were asked to predict the time-dependent ^{90}Sr and ^{137}Cs concentrations in the water of the estuary. Input information for the modellers was: the deposition of radionuclides of Chernobyl origin, the concentrations of radionuclides in the Black Sea and in the Dnieper and Southern Boug rivers, hydrological, morphological and environmental data (temperature, salinity, pH, etc.). The performances of the models were evaluated by comparing predictions with the observed time dependent radionuclide concentrations in the estuary.

2.3. ^{3}H migration in the Loire River, France

This exercise was concerned with the assessment of the dispersion of tritium released at different points on the Loire River, spread over about 350 km, and over a period of six months. Modellers were provided with data on water discharges from tributaries, hydrological data and information on tritium discharges from four nuclear power plants. The results were compared with observations of tritium concentration made at Angers, a city on the river.

2.4. Release of radionuclides into the Techa River, South Urals, Russia

The objective of this exercise was to test modelling of the behaviour of ^{90}Sr, ^{137}Cs and $^{239,240}Pu$ in river water and bottom sediments. The scenario was based on data obtained from the Techa River, which was contaminated, mainly in the period 1949–1952, as a result of discharges of liquid radioactive waste into the river.

2.5. Behaviour of ^{226}Ra in the Huelva estuary, Spain

This estuary was affected by radionuclides released from a phosphate factory. In the exercise, the time evolution of the total ^{226}Ra inventory in the bed sediments and the time evolution of the ^{226}Ra concentration in the water column were predicted by to the modellers.

Scenarios (a) and (c) were so-called 'blind' exercises, that is, the modellers were not given access to the observed data in advance of their modelling attempts.

The models used in the present exercises are listed in Table 1. The models have different degrees of complexity, ranging from those based on simple box-type approaches to those that make use of the Navier–Stokes and diffusion-transport equations. Similarly, the processes controlling the interaction of radionuclides with sediments were approached at different levels of detail in the models.

3. Results

This paper summarises the most important conclusions obtained from the aforementioned modelling exercises. Detailed results will be given in an IAEA report to be published later. In this paper, the results of scenarios (b) and (c) have been selected as examples of the project outcome. These provide the basis for the discussion and the conclusions concerning the overall evaluation of model uncertainty. Moreover, these scenarios (b) and (c) exemplify two extremes within the range of possible conditions for the application of models to real circumstances. Scenario (c) concerns the controlled release of tritium to river water. For this scenario sufficient and reliable information and data regarding the hydrological conditions and the input rates of the radionuclide into the river were available for modelling purposes. In contrast, in scenario (b), which considered the contamination of the Dnieper–Southern Boug estuary due to the accidental release of radionuclides into the environment (the Chernobyl accident), the available data was insufficient in that it did not provide the full range of information required to model the system properly. The scenario was complex and the observed data were collected by different institutes according to different criteria. However, the exercise was of great interest because it reproduced, in a realistic way, the situation in which models might be used to assess the consequences of a nuclear emergency. More details of this particular exercise are given in Monte et al. (2006a).

The applications of MARTE (model implemented in the decision support system MOIRA-PLUS), RIVTOX (model implemented in the decision support system RODOS), and the MASCARET and CASTEAUR models for predicting tritium migration through Loire River (scenario (c)) are shown in Fig. 1.

Figs. 2 and 3 show the results obtained from selected models when applied to scenario (b). The spread of the results reflects the various methodological approaches used to model the complex processes occurring in the aquatic environment as well the different values used for the parameters in the models. Compared to scenario (c), the system to be modelled is complex mainly because of the need to account for the interaction of radionuclides with sediments and, in addition, for regional scale models, because of the uncertainty associated with the sub-models for predicting the migration of radionuclides from the catchments of the Rivers Dnieper and Southern Boug to the estuary.

4. Discussion and conclusions

Generally, the basic components of state-of-the-art models for predicting the behaviour of radionuclides in the abiotic compartments of aquatic systems are: (1) hydrological sub-models (and related transport-diffusion sub-models), (2) sub-models for predicting the interaction of radionuclides with sediments and suspended matter and (3) sub-models for predicting the migration of radionuclides from contaminated catchments. The

Table 1
List of tested models

Institute and country	Model	Scenario	References	Note
IMMSP, Institute of Mathematical Machine and System Problems, Ukraine	COASTOX	a	Zheleznyak et al. (1992,1997)	Implemented in RODOS decision support system
	THREETOX	b	Margvelashvili et al. (1997)	
	RIVTOX	c d e	Bäverstam et al. (1997) and Zheleznyak et al. (1992,2001)	
University of Seville, Spain	University of Seville	a b e	Periañez et al. (1996)	Customised versions for each specific application
ENEA, Ente per le Nuove Tecnologie l'Energia e l'Ambiente, Italy	MARTE	a b c e	Monte et al. (2006a) Monte et al. (2006b) Monte et al. (2001)	Customised versions for applications (a), (b) and (e); application (c) was performed by using the model version implemented in the decision support system MOIRA-PLUS
Uppsala University, Sweden	COASTMAB	b	Håkanson (2005)	
EDF, Electricité de France, France	MASCARET	c e	Luck and Goutal (2003)	
IRSN, Institut de Radioprotection et de Sûreté Nucléaire, France	CASTEAUR	c d	Beaugelin-Seiller et al. (2002)	
IGE, Institute of General Energy, Ukraine	UNDBE	c	Sizonenko (1998)	
AEP, Atomenergoproject, Russia	TRANSFER-2	d	Kryshev et al. (2007)	
ISDAE, Institute of Safety Development of Atomic Energy, Russia	CASSANDRA	d	Kryshev et al. (2007)	
Institute of Experimental Meteorology SPA "Typhoon", Russia	GIDRO	d	Kryshev et al. (2007)	

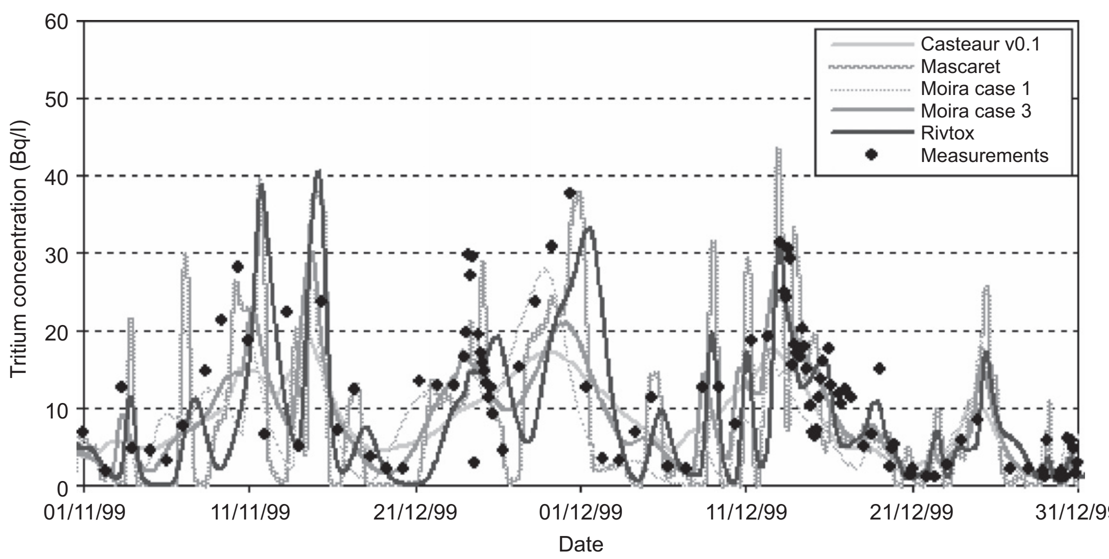

Fig. 1. Comparison of observed ^{3}H concentrations in the Loire River at Anger with predictions from models MARTE (MOIRA-PLUS), RIVTOX, MASCARET and CASTEAUR. To simulate the average monthly water fluxes of the river, generic default functions were used for Run 1 of MOIRA, whereas site-specific functions for the hourly averages of the water fluxes were used for Run 3. The exercise involved a time-varying, multi-point source of ^{3}H discharge over a river length of approximately 350 km.

uncertainty in the model predictions is related to the features of these components.

It should be noted that as tritium does not interact with sediments, the only processes that control the migration of this radionuclide are diffusion and transport by the water current. Therefore, exercise (c) was an important test for assessing the reliability of the hydrological and diffusion-transport modules being used in state-of-the-art models. The simulations produced using the MARTE model were essentially based on the assessment of the water balance in different consecutive 'segments' of the river and on the evaluation of the average depth and width of each river segment as functions of the water flux (see IAEA, 2001). As Fig. 1 shows, the results from the model MARTE are consistent with those obtained using the models RIVTOX and MASCARET that are based on one-dimension shallow-water fluid dynamics and advection–dispersion equations to simulate water flow and radionuclide transport and by the 'box-type' model CASTEAUR. This 'blind test' exercise showed that the behaviour of radionuclides that hardly react with suspended matter and sediments can usually be predicted to acceptable levels of accuracy by

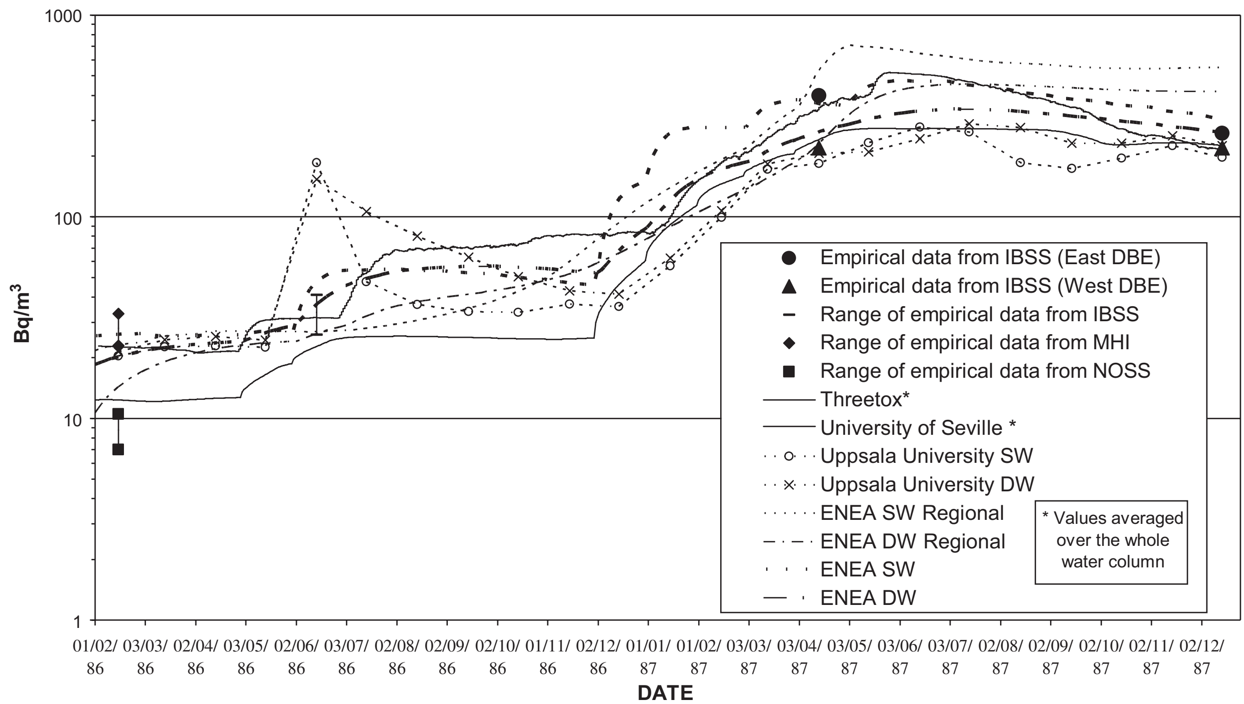

Fig. 2. Comparison of model predictions with observed values of ^{90}Sr collected by different institutes in the Dnieper–Southern Boug estuary: IBSS (Institute for Biology of the Southern Seas, Sevastopol, Ukraine), MHI (Marine Hydrophysical Institute, Sevastopol, Ukraine), NOSS (Nikolaevskaya Oblast Sanitary Station, Nikolaev, Ukraine). THREETOX is a module of the decision support system RODOS. Predictions from ENEA and Uppsala University models were supplied for both surface water (SW) and deep water (DW). The model 'ENEA regional' was used to predict the concentration of ^{90}Sr in the estuary from the deposition of the radionuclide on to the contaminated area around Chernobyl. The contribution of ^{90}Sr to the estuarine water from the Dnieper River was evaluated by an application of model MARTE to the whole basin of the river.

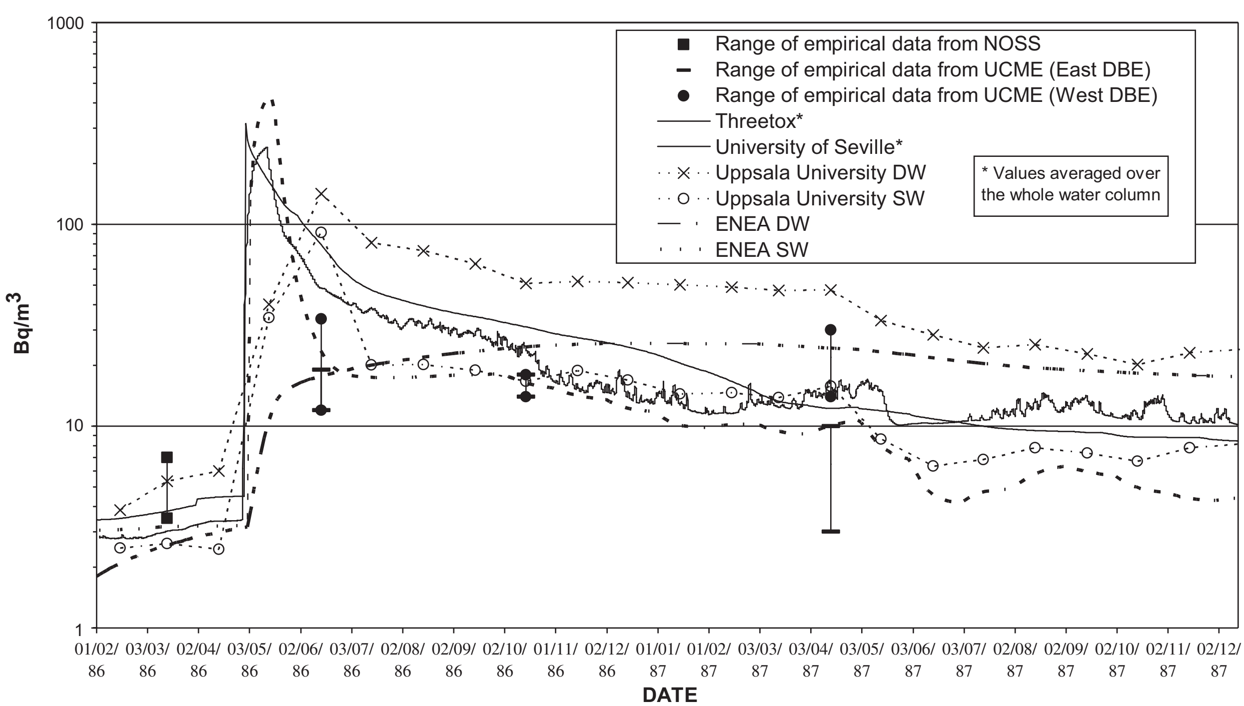

Fig. 3. Comparison of model predictions with observed values of ^{137}Cs collected by different institutes in the Dnieper–Southern Boug estuary: NOSS (Nikolaevskaya Oblast Sanitary Station, Nikolaev, Ukraine) and UCME (Ukrainian Centre for Marine Ecology, Odessa, Ukraine). Predictions from ENEA and Uppsala University models were supplied for both surface water (SW) and deep water (DW).

models even if they are based on relatively simple hydrological sub-models.

In contrast, the predictions of models for representing the behaviour of radionuclides that interact strongly with sediment and soil particles are associated with significant levels of uncertainty. The blind test exercise (a) showed that the models in the exercise properly predicted the remobilisation of strontium from the contaminated Pripyat River floodplain but that they significantly overestimated the remobilisation of radiocaesium (Monte et al., 2006b). The modelling of the complex dynamics of extreme hydrological events, such as inundation of large flood plains, was also recognised as an important source of uncertainty for reactive radionuclides. Indeed, the concentration of remobilised radionuclides in water depends on the time of inundation and on the proportion of water flowing over more or less contaminated areas. In many circumstances, these quantities cannot be predicted with sufficient accuracy and, therefore, have a major influence on the overall uncertainty of model predictions. Generally, the sub-models for assessing the interaction of radionuclides with sediments and the migration from the catchments are the most significant sources of uncertainty. These conclusions were confirmed by the results of exercises (d) and (e) which also dealt with this type of complex problem.

From exercise (b) it can be noted that, in spite of the range of variability of the model predictions (a range that has the same order of uncertainty as the measured concentrations of radionuclides in water), the models reliably predicted the different time behaviour of radiocaesium and radiostrontium in estuary waters. It is well known that these radionuclides show significantly different degrees of mobility in the aquatic environment. The potential for radiocaesium migration from the contaminated catchment of the estuary is significantly lower than that of radiostrontium. Therefore, the direct deposition of ^{137}Cs was a significant source of contamination of the estuary. Due to the relatively rapid dynamics of the estuarine water, radiocaesium concentrations showed a marked decline with time. The direct initial atmospheric deposition of radiostrontium on to the estuary was negligible and the increase of radiostrontium concentration in water was due to the delayed transport to the estuary from the heavily contaminated area around Chernobyl. It is interesting to note that the ENEA model applied at a regional scale (the radionuclide flux to the estuary was calculated by an application of the model MARTE to the whole basin of the Dnieper River) gave results that were in the same range as the results of the other models. In fact, the assessed models are ‘behavioural’, that is they reproduce the different time behaviours of the concentrations of radionuclides in water although the results are spread over almost one order of magnitude. This characteristic of the models, even if the results may be affected by uncertainties, is essential for applications aimed at supporting the management of the aftermath of a nuclear accident.

The exercise has shown the benefits of using several models to predict the behaviour of the same environmental system and contamination scenario (multi-model approach). This approach is of special value when the environmental processes are complex and when there are, therefore, difficulties in selecting unambiguous site-specific values of model parameters. In these circumstances, different hypotheses and approaches may be used by modellers to represent the same situation. The exercises performed in the working group showed that a multi-model approach can be useful for the management of complex problems in environmental assessment. Through this approach the conclusions that obtain the greatest degree of consensus among modellers are made evident and the aspects that are subject to dispute and which should therefore be handled carefully also become clear.

References

Bäverstam, U., Fraser, G., Kelly, G.N. (Eds.), 1997. Decision making support for off-site emergency management. In: Proceeding of the Fourth International Workshop, Aronsborg, Sweden, October 1996. Radiat. Prot. Dosim. 73, 315.

Beaugelin-Seiller, K., Boyer, P., Garnier-Laplace, J., Adam, C., 2002. CASTEAUR: a simple tool to assess the transfer of radionuclides in waterways. Health Phys. 84, 539–542.

BIOMOVS, 1990. On the validity of environmental transfer models. In: Proceedings of a Symposium, October 8–10, 1990, Swedish Radiation Protection Institute, Stockholm.

BIOMOVS II, 1996a. Validation test for carbon-14 migration and accumulation in a Canadian Shield Lake. Technical Report no. 14. Swedish Radiation Protection Institute, Stockholm, Sweden, p. 40.

BIOMOVS II, 1996b. Assessment of the consequences of the radioactive contamination of aquatic media and biota. Technical Report no. 10. Swedish Radiation Protection Institute, Stockholm, Sweden, p. 70.

BIOMOVS II, 1996c. Wash-off of Sr-90 and Cs-137 from two experimental plots: model testing using Chernobyl data. Technical Report no. 9. Swedish Radiation Protection Institute, Stockholm, Sweden, p. 32.

Håkanson, L., 2005. A new general model predicting radionuclide concentrations and fluxes in coastal areas from readily accessible driving variables. J. Environ. Radioactiv. 78, 217–245.

IAEA, 2000. Modelling of the Transfer of Radiocaesium from Deposition to Lake Ecosystems. IAEA-TECDOC-1143. International Atomic Energy Agency, Vienna, Austria.

IAEA, 2001. Generic models for use in assessing the impact of discharges of radioactive substances to the environment. Safety Reports Series no. 19. International Atomic Energy Agency, Vienna, Austria, p. 216.

Kryshev, I., Boyer, P., Dzyuba, N., Krylov, A., Kryshev, A., Nosov, A., Sanina, K., Zheleznyak, M., 2007. Radioactive contamination of the Techa River by ^{90}Sr, ^{137}Cs and ^{239,240}Pu (South Urals, Russia). Scenario and model testing. EMRAS Aquatic Working Group Final Report 〈http://www-ns.iaea.org/projects/emras/emras-aquatic-wg.htm〉.

Luck, M., Goutal, N., 2003. Système Mascaret: Note de principe de l'outil Tracer (transport de traceurs) et des modules de qualité d'eau O2, Biomass, Eutro, Micropol et Thermic. Rapport EDF-LNHE HP-75/03/047/A.

Margvelashvili, N., Maderich, V., Zheleznyak, M., 1997. THREETOX: a computer code to simulate three dimensional dispersion of radionuclides in stratified water bodies. Radiat. Prot. Dosim. 73, 177–180.

Monte, L., Van deer Steen, J., Bergström, U., Gallego Díaz, E., Håkanson, L., Brittain, J., 2001. A model-based computerised system for management support to identify optimal remedial strategies for restoring radionuclide contaminated aquatic ecosystems and drainage areas. In: European Commission, Radiation Protection Fourth Framework Programme (1994–1998) Project Summaries. EUR 19792 EN (2001), Brussels, pp. 360–363.

Monte, L., Håkanson, L., Perianez, R., Laptev, G., Zheleznyak, M., Maderich, V., Angeli, G., Koshebutsky, V., 2006a. Experiences from a case study of multi-model application to assess the behaviour of pollutants in the Dnieper–Bug Estuary. Ecol. Modelling 195, 247–263.

Monte, L., Periañez, R., Kivva, S., Laptev, G., Angeli, G., Barros, H., Zheleznyak, M., 2006b. Assessment of state-of-the-art models for predicting the remobilisation of radionuclides following the flooding of heavily contaminated areas: the case of Pripyat River floodplain. J. Environ. Radioactiv. 88, 267–288.

Periañez, R., Abril, J.M., García-León, M., 1996. Modelling the dispersion of nonconservative radionuclides in tidal waters. Part 1: conceptual and mathematical model. J. Environ. Radioactiv. 31, 127–141.

Sizonenko, V.P., 1998. Increasing accuracy of the box model. In: Babovic, V., Larson, L.C. (Eds.), Hydroinformatics '98—Proceedings of the Third International Conference on Hydroinformatics/Copenhagen/Denmark/24–26 August 1998, vol. 1. A.A. Balkema, Rotterdam, pp. 225–230.

Zheleznyak, M., Demchenko, R., Khursin, S., Kuzmenko, Y., Tkalich, P., Vitiuk, N., 1992. Mathematical modeling of radionuclide dispersion in the Pripyat–Dnieper aquatic system after the Chernobyl accident. Sci. Total Environ. 112, 89–114.

Zheleznyak, M., Shepeleva, T., Sizonenko, V., Mezhueva, I., 1997. Simulation of countermeasures to diminish radionuclide fluxes from Chernobyl zone via aquatic pathways. Radiat. Prot. Dosim. 73, 181–186.

Zheleznyak, M., Heling, R., Raskob, W., 2001. Hydrological dispersion module of the decision support system RODOS. In: Proceedings of Conference on ECORAD-2001, Aix-en-Provence, France, 3–6 September 2001.