SCENARIO W:
WASH-OFF OF ^{90}Sr AND ^{137}Cs FROM TWO EXPERIMENTAL PLOTS

ABSTRACT

Surface water runoff from contaminated land is one of the major processes responsible for the contamination of water bodies. Based on data from the Chernobyl accident, the "Wash-off" scenario was developed to test models concerned with the movement of trace contaminants from terrestrial sources to water bodies. In particular, this scenario provides an opportunity for (1) evaluation of the movement of contaminants from soil to water, (2) calculation of the alteration and migration of contaminants in soil over different time scales, (3) increased understanding of contaminant transport at the process level, and (4) development and use of methods for estimating key parameters.

The "Wash-off" Scenario consists of two experimental plots near the Chernobyl Nuclear Power Plant, one using simulated heavy rain (Plot HR) and one using snow melt (Plot SM). Input data for Plot HR include the following: soil description and properties; hydrographs of rainfall and runoff dynamics; the time of application of rainfall; rainfall amounts, duration, and intensities; soil moisture content before the application of rainfall; regional data on average monthly precipitation and temperature; recorded information on naturally occurring precipitation between May and October 1986; and chemical forms of radionuclides in the soil of the plot prior to the experiments. Input data for Plot SM include the following: soil description and properties; snow storage in the snow melt period of 1988; chemical composition of the snow water; a hydrograph of the runoff dynamics; chemical radionuclide forms in the soil at the end of the experiment; and air and soil temperatures for the plot during the snow melt period.

For both Plot HR and Plot SM, measurements of the vertical distribution of ^{137}Cs and ^{90}Sr in the soil prior to the experiments and the percentages of exchangeable and nonexchangeable forms of each radionuclide at that time are available for comparison to model predictions. For Plot HR, measurements are also available for the total concentration of each radionuclide in surface runoff during each part of the experiment, the percentage of each radionuclide in dissolved and particulate forms for each experiment, and the total amount of each radionuclide lost from the experimental plot during the experiments and 24 hours afterward. For Plot SM, measurements are also available for the total concentration of each radionuclide in surface runoff during the periods of peak flow and minimum flow, the percentage of each radionuclide in dissolved and particulate forms for each time period, and the total amount of each radionuclide lost from the experimental plot. A detailed description of the "Wash-off" Scenario is provided, along with tables of input data and test data.

INTRODUCTION

One of the main processes responsible for the contamination of water bodies is surface water runoff from contaminated land. The large area of land contaminated after the Chernobyl accident has become a continuing source of radionuclides entering the aquatic ecosystem (Konoplev et al., 1992). The "Wash-off" Scenario (Scenario W) is one of three major test scenarios presented by the Post-Chernobyl Data Working Group of BIOMOVS II (Biospheric

Model Validation Study, Phase II) for testing models at the process level (BIOMOVS II, 1996; Konoplev et al., 1996; 1999). Scenario W offers an excellent opportunity to test models concerned with the movement of trace contaminants from terrestrial sources to water bodies by taking into account the chemical form of radionuclides in soil, the effect of wash-off on the transfer of contamination from soil to water, and the geochemical and geophysical processes that affect such transfer. Scenario W is based on the wash-off of ^{90}Sr and ^{137}Cs from two experimental plots, Plot HR (heavy rain) and Plot SM (snow melt), established in the vicinity of the Chernobyl Nuclear Power Plant and contaminated by dry deposition from the Chernobyl accident.

Natural waters are one of the important links in food chains through which radionuclides are transferred to humans. From the soil solution, radionuclides pass to plants and to animals; in addition, the contamination level of surface waters is a major factor governing radionuclide uptake and concentrations in fish. The qualitative characteristics of these processes are functions of parameters such as radionuclide distribution in the soil/water solution, bottom sediments/water, and suspended sediments/water systems. It is particularly important that this scenario includes radionuclide speciation processes in soils. These processes are responsible for changes in the migration characteristics of radionuclides with time and are important in making accurate long-term predictions of the radiation situation, as well as dose assessments for the populations living in contaminated areas.

The main processes considered in Scenario W are hydrological and physico-chemical. Hydrological processes include formation of surface runoff as a result of rainfall and the associated wash-off with the surface runoff of suspended soil particles with incorporated radionuclides. Physico-chemical processes that are responsible for radionuclide transfer between soil and water include transformation of the chemical species of a radionuclide, vertical migration of radionuclides in soil, distribution of radionuclides between the solid and liquid phases, and radionuclide transfer from soil to surface runoff. A more in-depth discussion of physico-chemical processes follows in the next section.

A detailed description of the “Wash-off” Scenario is provided below, along with a description of the types of test data available. The Scenario description is followed by tables of information to be used as input data and tables of test data (actual measurements) for comparison with model predictions. Additional information, including the results of the original BIOMOVS II model testing exercise, has been reported by Konoplev et al. (1996; 1999) and BIOMOVS II (1996).

DISCUSSION OF PHYSICO-CHEMICAL PROCESSES INVOLVED

Transformation of Chemical Species

A distinguishing feature of radioactive contamination from nuclear accidents is the chemical speciation of the radionuclides present in atmospheric fallout and deposited on the soil. When the Chernobyl accident occurred, nuclear fuel debris (UO_2 + UO_3), including fission products, was released into the environment. Some of the radionuclides were also deposited onto the ground as aerosol particles, which were formed in the atmosphere due to condensation. The fuel particles are insoluble in water and hence do not participate in exchange processes between the soil and the soil solution.

The rate of radionuclide migration is very sensitive to the chemical speciation of the radionuclides in the soil. In terms of predicting the migration of ^{137}Cs and ^{90}Sr in soil-water systems, it is reasonable to identify the following chemical forms: (1) dissolved cations (A_2), (2) exchangeable forms (A_{2e}), including radionuclides sorbed onto soil or bottom sediments by ion-exchange mechanisms, and (3) non-exchangeable forms, including radionuclides of nuclear fuel particles (A_1) and fixed radionuclides on mineral or organic components of soil or bottom sediments (A_3). Over the course of time, transformation of the chemical species of radionuclides may occur (Figure 1).

Vertical Migration of Radionuclides

Vertical movement of a radionuclide in soil may occur due to infiltration (downward flow) of water during rainfall. The exchangeable fraction (A_2 + A_{2e}) moves in the dissolved state (A_2), with some retardation due to ion exchange. The solid-phase species (A_1, A_{2e}, and A_3) move only with the particles in which they are incorporated.

Distribution of Radionuclide Between Solid and Liquid Phases

Distribution of ^{137}Cs and ^{90}Sr in the soil/soil solution system is governed by ion exchange of those radionuclides in exchangeable forms. The concentration of radionuclide in solution is determined by the sorption capacity of the solid phase and the selectivity and concentrations of the major ions in solution.

Radionuclide Transfer from Soil to Surface Runoff

The following mechanisms are responsible for wash-off of radionuclides in dissolved form by surface runoff from soil (both from heavy rain and from snow melt-induced runoff): radionuclide wash-out from soil solution by interflow (water flow through the soil rather than on top of the soil); radionuclide transfer from the top soil layer to the overland flow due to molecular and hydrodynamic dispersion; and radionuclide desorption from the solid phase of soil by both overland flow and interflow. For the first two mechanisms, the concentration and total amount of washed-out radionuclide is governed mainly by hydrological factors (e.g., rain intensity, hydraulic conductivity of soil) and the radionuclide content in the pore solution as a function of depth. For the third mechanism, the physico-chemical parameters (e.g., soil sorption properties, ion composition of soil solution and rain water) and the vertical distribution of the fraction of radionuclides available for desorption are also of importance.

DESCRIPTION OF THE “WASH-OFF” SCENARIO

Two experimental plots were constructed in areas contaminated by dry deposition resulting from the Chernobyl accident. Most deposition (>95%) occurred between 26 April and 10 May 1986.

Experimental Plot HR

The 625-m^2 experimental plot is located 7 km from the Chernobyl Nuclear Power Plant, 0.6 km from the village of Benevka on the oxbow of the Pripyat River. It is defined on three sides by dikes and on the lower side by a collection chute. The plot size is 25 m x 25 m; the mean grade of the slope is 5%. The observations were carried out during an experiment conducted on October 14, 1986. In this experiment artificial heavy rain applications were used to simulate natural runoff. Based on randomly selected soil samples, the total contamination in the top 10 cm of the plot just prior to the experiments was determined to be 1.4 ± 0.1 MBq m^{-2} of ^{137}Cs and 1.8 ± 0.4 MBq m^{-2} of ^{90}Sr [± standard deviation (SD)]. Descriptions of the soil and of Experimental Plot HR are provided in Tables 1 through 3.

Intense artificial rain was used to simulate the formation of natural surface runoff on the plot. This was done for two main reasons. First, there was an urgent need at the time to predict the contamination of water bodies due to runoff caused by rainfall, flooding, and snow melt from areas of contaminated soil. However, there was a lack of rainfall during the period immediately after the accident, and it was not possible to wait until there was natural surface runoff. Second, in the 30-km zone there is poor surface runoff formation; the mean annual runoff coefficient is ~15%.

The observations were made during an experiment conducted on 14 October 1986. The artificial heavy rain was applied by pointing a fire hose up in the air and letting the water come down. A second artificial rainfall application was used to test the dependence of radionuclide wash-off on initial conditions (i.e., before the first rainfall application, the soil was dry, whereas before the second application it was wet). A hydrograph of the runoff dynamics is given in Figure 2. Characteristics of the artificial rain applications are given in Table 4. The chemical composition of the artificial rain is not representative of the actual rain in the region, in that the artificial rain contained more dissolved salts than actual rain would have contained. The intensity of the artificial rain was representative of actual rain that can cause surface runoff. The applicability of data obtained with artificial rain to actual environmental conditions has been discussed by Bulgakov and Konoplev (1992) and Bulgakov et al. (1992).

Experimental Plot SM

The 1000-m^2 experimental plot is located on a north-facing slope near the town of Chernobyl. It is defined on three sides by dikes and on the lower side by a collection chute. The plot size is 50 m x 20 m; the mean grade of the slope is 4%. The total contamination of the plot was determined to be 480 ± 155 kBq m^{-2} of ^{137}Cs and 270 ± 48 kBq m^{-2} of ^{90}Sr (± SD). The observations were made in winter and spring during the snow melt period of 1988 (March). A hydrograph of the runoff dynamics of Plot SM is provided in Figure 3. Descriptions of the soil, the plot, and the snow water are provided in Tables 1 - 2 and 5 - 9.

EXPERIMENTAL DETAILS

The contamination of the plots was determined by taking randomly selected soil samples; each sample was 180 cm^2 in area and 10 cm in depth. Runoff samples were collected in plastic bottles and filtered with a 0.45 μm filter. The ^{137}Cs content in soil samples, filters, and water samples was determined through gamma spectrometry without preconcentration using a Canberra gamma counter with a semiconductor detector (Makhonko et al., 1985). The ^{90}Sr content was determined with a radiochemical analysis that used carbonates of precipitation (Sereda and Shulepko, 1966). All samples were processed at the Institute of Experimental Meteorology, SPA "Typhoon," in Obninsk, Russia.

TEST DATA AVAILABLE

1. Endpoints for Experimental Plot HR
   - (a) Vertical distribution of ^{137}Cs and ^{90}Sr in the soil immediately prior to the experiments on 14 October 1986 (Table 10).
   - (b) Percentages of exchangeable and non-exchangeable forms of each radionuclide at that time (Table 10).
   - (c) Concentrations of ^{137}Cs and ^{90}Sr in surface runoff during each experiment (Table 11).
   - (d) Percentages of ^{137}Cs and ^{90}Sr in dissolved and particulate forms for each experiment (Table 11).
   - (e) Total amounts of ^{137}Cs and ^{90}Sr lost from the experimental plot during the experiments and 24 hours afterward (Table 12).

2. Endpoints for Experimental Plot SM
   - (a) Vertical distribution of ^{137}Cs and ^{90}Sr in the soil as of 15 March 1988 (Table 13).
   - (b) Percentages of exchangeable and non-exchangeable forms of each radionuclide at that time (Table 13).
   - (c) Concentrations of total and dissolved ^{137}Cs and ^{90}Sr in surface runoff at peak flow for 18, 20, and 24 March 1988 and at the minimum flow on the night of 20-21 March 1988 (Table 14).
   - (d) Percentages of ^{137}Cs and ^{90}Sr in dissolved and particulate forms for each time period (Table 14).
   - (e) Total amounts of ^{137}Cs and ^{90}Sr lost from the experimental plot as of 31 March 1988 (Table 15).

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TABLES OF INPUT DATA

Table 1. Soil characteristics of the experimental plots.

Characteristic	Plot HR	Plot SM
General description	Alluvial acid sod	Cultivated sod podzolic soil
Vegetation	mixed grasses on fallow land	mixed grasses on fallow land
Effective water-holding capacity of the 0 - 10 cm layer of soil, %	45 ± 6	30 ± 3
Hydraulic conductivity[b] (velocity of downward percolation of water in the soil profile), mm min^{-1}	0.34 ± 0.07	0.31 ± 0.03
Cation exchange capacity (CEC) of the upper 1 cm of soil, meq g^{-1} dry weight	0.15 ± 0.02	0.07 ± 0.01

[a] Not applicable.
[b] Hydraulic conductivity was calculated as the difference between rainfall intensity (mm min^{-1}) and runoff intensity (mm min^{-1}) during special experiments using artificial rain on 1-m^2 plots.

Table 2. Soil descriptions for the experimental plots.

Soil Layer	Depth (cm)	pH	Humus Content (%)	Soil Type
Plot HR				
Root zone	0 to 8	5.1	8 - 12	sandy loam
A1 Horizon	8 to 15 - 20	4.6 - 4.8	4 - 6	sandy loam
B Horizon	15 - 20 to 40 - 50	4.6	1 - 2	sandy loam
Plot SM				
Root zone	0 to 5	5.8	4 - 5	sandy loam
Plowed layer	5 to 15	5.8	1 - 1.5	sandy loam
B Horizon	15 - 30	5.0 - 5.2	0.3 - 0.8	loamy sand - sand

Table 3. Characteristics of Plot HR.

Characteristic	Value	
Water table depth (m)	0.5 to 5 (yearly average, 1.5)	
Standing water depth during artificial rain application (mm)	5 to 10	
Measured natural precipitation at runoff plot during 1986 (mm)		
May	20.5 (est.)	
June	39.5	
July	19.5	
August	60.1	
September	44.6	
October	26.4	
Biomass density		
wet weight (g m^{-2})	510 ± 200	
dry weight (g m^{-2})	130 ± 40	
Vertical distribution of soil density in experimental plot HR		
Layer (cm)	0-5	5-10
Volumetric density (g cm^{-3} dry weight)	1.00	1.49
Porosity (%, cm^3/cm^3)	49.5	42.6
Chemical forms of radionuclides in the soil of plot HR prior to artificial rain application		
Radionuclide	^{90}Sr	^{137}Cs
Mobile form[a] (%)	25	7
Non-exchangeable form (%)	75	93

[a] Amount in solution and extracted in 1N ammonium acetate solution.

Table 4. Characteristics of the artificial rain applied to Plot HR.

Characteristic	Value	
pH	7.2	
Ionic composition of water (mg L^{-1})		
Ca^{2+}	55	
K^{+}	5	
Na^{+}	17	
Mg^{2+}	12	
HCO_3^{-}	170	
Start of rain application	16:29	17:22
Duration (min)	25	16
Rain amount (mm)	33.2	21.8
Rainfall intensity (mm min^{-1})	1.33	1.37
Soil moisture content before rain (%, g H_20 g^{-1} soil wet weight)	16	-

Table 5. Characteristics of Plot SM.

Characteristic	Value
Soil density by layer, g cm^{-3} dry weight	
0-0.5 cm	1.55
0.5-1 cm	1.62
1-2 cm	1.53
2-3 cm	1.66
3-5 cm	1.53
0-10 cm	1.55
Soil porosity (0 - 10 cm)	37.5 % (cm^3 per cm^3)
Snow storage in the snow melt period	23.5 mm
Depth of freezing (January and February)	60 - 100 cm
pH of snow water	5.3
Cation composition of snow water (mg L^{-1})	
Ca^{2+}	1.5
K^+	1.3
Na^+	1.9
NH_4^+	1.4
Mg^{2+}	4.4

Table 6. Chemical forms of the radionuclides deposited with atmospheric fallout in Plot SM.[a]

Radionuclide	Mobile form[b], %	Non-exchangeable form, %
^{90}Sr	7.2	92.8
^{137}Cs	29.2	70.8
^{144}Ce	11.1	88.9
^{106}Ru	15.4	84.6

[a] Samples were taken daily from fallout collectors beginning on 26 April 1986, conserved by a special method, and stored. The chemical forms were determined in the laboratory during 1988. Samples were dried and stored in sealed containers to ensure that the chemical forms measured were the same as were deposited. The values are integrated estimates based on the total activity received by the plot over the entire time of initial deposition.
[b] Amount in solution and extracted with ammonium acetate.

Table 7. Chemical radionuclide forms in the soil of Plot SM at the end of the experiment (5 April 1988).

Radionuclide	Mobile form[a], %	Non-exchangeable form, %
^{90}Sr	70.6	29.4
^{137}Cs	7.9	92.1
^{144}Ce	4.9	95.1
^{106}Ru	8.1	91.9

[a] water-soluble + exchangeable.

Table 8. Air and soil temperature for Plot SM during the snow melt period of 1988.

Date (March 1988)	Air temperature, °C			Soil temperature (0 - 2 cm), °C		
	Time			Time		
	9:00	15:00	21:00	9:00	15:00	21:00
15	-6.5	1.3	-0.6	-3.7	1.3	-1.7
16	-3.6	0.4	3.9	-2.8	0.1	0.5
17	3.7	11.8	6.9	1.3	1.0	1.0
18	2.3	4.1	0.5	0.3	4.2	-0.8
19	-2.5	1.5	-1.4	-0.7	2.0	-2.0
20	-2.5	2.9	1.4	-1.0	2.4	-0.5
21	-4.6	4.4	-2.1	-2.2	8.4	-0.5
22	1.0	2.5	1.4	0	2.4	0.3
23	1.9	4.5	2.8	1.1	5.1	1.1
24	2.1	4.4	3.9	1	3.4	1.6
25[a]	0.8	7.5	4.6	0	9.3	2.5

[a] Runoff ceased after 25 March 1988.

Table 9. Variations in atmospheric precipitation at Plot SM during the snow melt period of March 1988.

Date (March 1988)	Precipitation, mm
15-21	0
22	2
23	1
24	5
25	1
26	0
27	1.5
28-31	0

TABLES OF TEST DATA

Table 10. Vertical distribution of ^{137}Cs and ^{90}Sr in the soil of Plot HR.

Soil layer, cm	Total (Bq g^{-1} dry weight)	Lower bound[a]	Upper bound[a]	Exchangeable %	Lower bound	Upper bound	Non-exchangeable %
^{137}Cs							
0 - 0.5	253	237	269	6.3	5.7	6.9	93.7
0.5 - 1	16.2	11.5	20.9	7.4	6.7	8.1	92.6
1 - 2	2.2	1.6	2.8	9.1	8.2	10	90.9
2 - 3	1.1	0.9	1.3	12.3	9.8	14.8	87.7
3 - 5	0.72	0.42	1.02	16.7	13.4	20	83.3
5 - 10	0.14	0.07	0.21	14.2	11.4	17	85.7
^{90}Sr							
0 - 0.5	308	264	352	21.1	19	23.2	78.9
0.5 - 1	28.2	20	36.4	32.6	29.3	35.9	67.4
1 - 2	3.3	2.4	4.2	48.5	43.7	53.4	51.5
2 - 3	2.4	2	2.8	58.3	46.6	70	41.7
3 - 5	1.3	0.7	1.9	69.2	55.4	83	30.8
5 - 10	0.4	0.2	0.6	75	60	90	25

[a] In Tables 10 - 15, the lower and upper bounds refer to the 95% confidence intervals on the mean values.

Table 11. Concentrations of ^{137}Cs and ^{90}Sr in runoff water from Plot HR.

	(1)16:29-17:22			(2) 17:22 and next 24 hours		
	Mean	Lower bound	Upper bound	Mean	Lower bound	Upper bound
^{137}Cs						
Total, Bq L^{-1}	28.7	19	38.4	15.3	7.9	22.7
% dissolved	38	21	55	44	31	57
% particulate	62	45	79	56	43	69
^{90}Sr						
Total, Bq L^{-1}	66.8	58.6	75	61.1	51.4	70.8
% dissolved	81	75.5	86.5	86.9	78.6	95.2
% particulate	19	13.5	24.5	13.1	4.8	21.4

Table 12. Total amount of radionuclide washed off from Plot HR (Bq).

Radionuclide	Mean	Lower bound	Upper bound
^{137}Cs	6.80×10^4	3.70×10^4	9.90×10^4
^{90}Sr	4.70×10^5	3.50×10^5	5.90×10^5

Table 13. Vertical distribution of ^{137}Cs and ^{90}Sr in the soil of Plot SM.

Soil layer, cm	Total (Bq g^{-1} dry weight)	Lower bound[a]	Upper bound[a]	Exchangeable %	Lower bound	Upper bound	Non-exchangeable %
^{137}Cs							
0 - 0.5	36	29	43	5.6	5	6.2	94.4
0.5 - 1	14	10	18	7.8	7	8.6	92.2
1 - 2	3.7	2.7	4.7	16.2	14.6	17.8	83.8
2 - 3	1	0.7	1.3	10	8	12	90
3 - 5	0.45	0.3	0.6	11	8.8	13.2	89
5 - 10	0.1	0.06	0.14	20	16	24	80
^{90}Sr							
0 - 0.5	12	11	13	56.5	50.9	62.2	43.5
0.5 - 1	4.2	2.8	5.6	69	62.1	75.9	31
1 - 2	2.8	2.2	3.4	76.4	68.8	84	23.6
2 - 3	1.7	1.4	2	76	60.8	91.2	24
3 - 5	1.4	1	1.8	85	68	102	15
5 - 10	0.35	0.22	0.48	86	68.8	103	14

Table 14. Concentrations of ^{137}Cs and ^{90}Sr in runoff water from Plot SM.

	18 March			20 March			20-21 March			24 March		
	Mean	Lower bound	Upper bound	Mean	Lower bound	Upper bound	Mean	Lower bound	Upper bound	Mean	Lower bound	Upper bound
^{137}Cs												
Total, Bq L^{-1}	3.5	2.1	4.9	2.5	1.5	3.5	2.7	1.9	3.5	2.4	1	3.8
Dissolved, Bq L^{-1}	2.2	1.2	3.2	2.2	1.4	3	2.5	1.8	3.2	1.5	0.8	2.2
Particulate, Bq g^{-1}	3.7	1.6	5.8	12.5	3.5	1.5	11	5.2	16.8	9.3	3.2	15.4
% dissolved	62.9			88			92.6			62.5		
^{90}Sr												
Total, Bq L^{-1}	17.7	12.8	22.6	17.7	13.6	21.8	27.1	18	36.2	43.2	33.7	52.7
Dissolved, Bq L^{-1}	14.7	10.3	19.1	15.8	12.1	19.5	25.5	16.9	34.1	39.3	32.5	46.1
Particulate, Bq g^{-1}	3.1	1.7	4.5	95	67	123	106	64	148	40.6	8.6	72.6
% dissolved	83.1			89.3			94.1			91.0		

Table 15. Total amount of radionuclide washed off from Plot SM (Bq).

Radionuclide	Mean	Lower bound	Upper bound
^{137}Cs	1.00×10^5	5.00×10^4	1.50×10^5
^{90}Sr	1.10×10^6	8.00×10^5	1.40×10^6

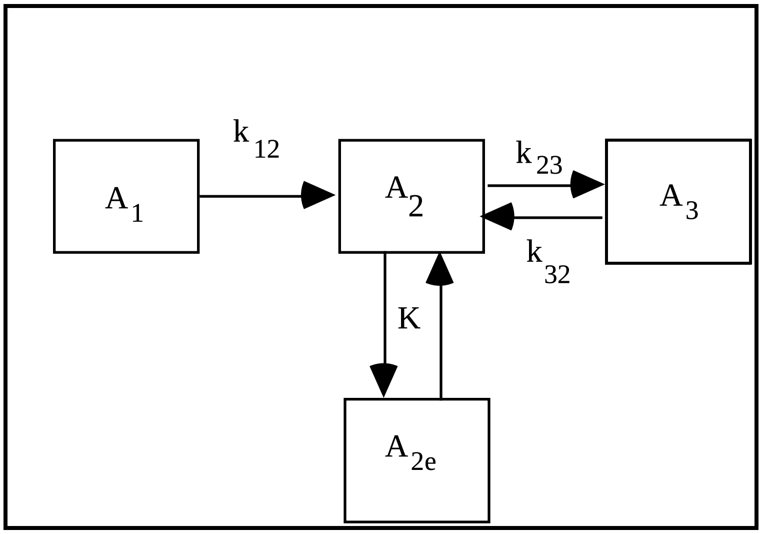

Figure 1. Schematic representation of the transformation processes of radionuclide chemical species (Konoplev et al., 1996). The chemical forms are represented as follows: A_1, radionuclides of nuclear fuel particles (non-exchangeable); A_2, dissolved cations; A_{2e}, exchangeable forms, including radionuclides absorbed onto soil or bottom sediments by ion-exchange mechanisms; and A_3, non-exchangeable radionuclides fixed on mineral or organic components of soil or bottom sediments. K is the constant for the ion-exchange equilibrium, and k_{ij} is the rate constant for the process at issue (i-j). The chemical forms may be considered in units of either mass or activity. The units of the ion-exchange equilibrium constant K will depend on the choice of concentration units for A_2 and A_{2e}. Radioactive decay has been omitted from the diagram because the time scale of the wash-off process being studied here is considerably smaller than the half-lives for the radionuclides in question.

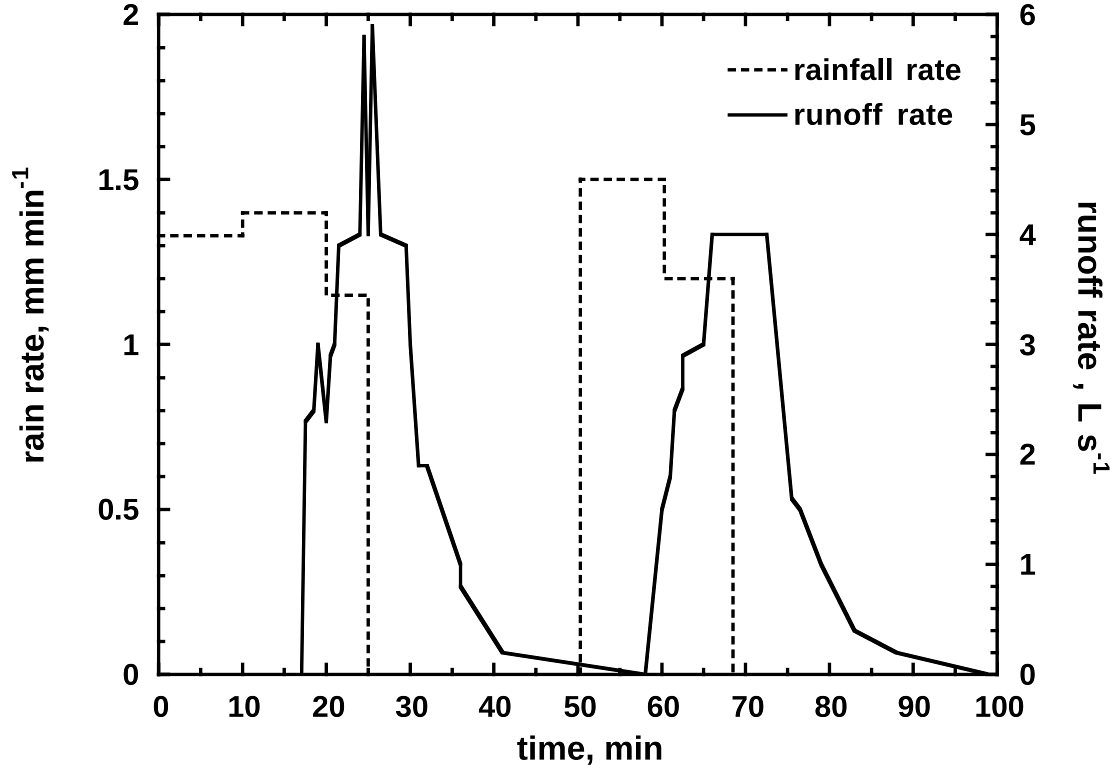

Figure 2. Hydrograph of the runoff dynamics for Plot HR (Konoplev et al., 1996).

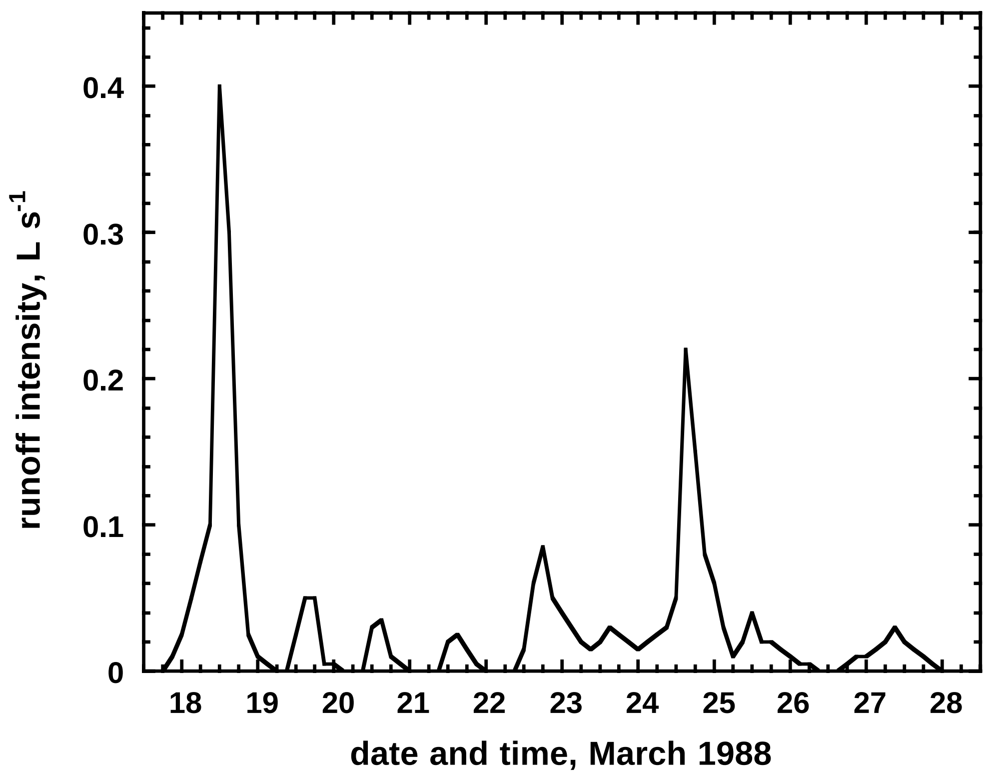

Figure 3. Hydrograph of the runoff dynamics for Plot SM (Konoplev et al., 1996).