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Spatial redistribution of radionuclides within catchments: Development of GIS-based models for decision support systems - SPARTACUS

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1. The main processes determining radionuclide redistribution within catchment

The main processes governing radionuclide redistribution on catchments are presented in Figure 1.1. The radionuclide transport on catchments can take place in soluble form and in particulate form that are carried by surface runoff. The radionuclide transport in soluble form is result of transfer of radionuclides from soil water to the surface runoff as well as desorption from the soil matrix. The solid (particulate) form of radionuclide transport can be regarded to a flow of radionuclides sorbed on suspended sediments which were formed by surface forces and transported by overland water flow.

The radionuclides in soluble form could interact with the suspended sediments and upper soil layer. Radionuclide transfer between surface water and suspended sediments is described by the adsorption-desorption processes. The transfer between surface water and upper soil layer is under the influence of adsorption-desorption and diffusion processes. The deposition of contaminated suspended sediments and the soil erosion are also important pathway of the “surface water – upper soil layer” radionuclide exchange.

Since the wash-off involves radionuclides in the soluble and particulated forms, both runoff and soil erosion processes are of importance. Actually, radionuclide transport on catchments is influenced by all hydrological processes controlling the transport of water.

The radionuclide concentration in water and on suspended particles are related through the distribution coefficient equal to the ratio of equilibrium concentrations of radionuclide in solid and liquid phases. The values of distribution coefficient depend on sorption properties of the soil solid phase and chemical composition of water (hydro-chemical factors) as well as properties and state of the radionuclide in soil (radiochemical factors). The same factors, along with hydro-physical ones (vertical water transport), largely determine the vertical migration of radionuclide in soil and hence time changes in concentrations in different soil layers and radionuclide concentration in groundwater. Thus, the main processes of radionuclides transport on catchment can be divided onto two types:

- *hydrological processes* (transport of water and suspended sediments, erosion/deposition processes);
- *physical-chemical processes* (radionuclide distribution in the soil-water system, “suspended sediments-surface water” system, and “surface water-upper soil layer” system).

The role of these processes in radionuclide redistribution within catchment is considered below.

1.1. Hydrological processes

Runoff formation begins after rain particles reach the surface. During the initial phase of runoff formation, rain energy liberates the soil particles and picks up the particulate and contaminants deposited on the surface and dissolves salts and other chemicals. Runoff generated by precipitation has the following components:

- Surface runoff is overland water flow onto land surface due to a residual of precipitation after all loses have been satisfied.
- Interception is that part of precipitation that wets or adheres to the surface of aboveground objects and vegetation and is returned to the atmosphere by evaporation.
- Infiltration into soils is the principal process controlling the runoff excess, interflow and ground flow.

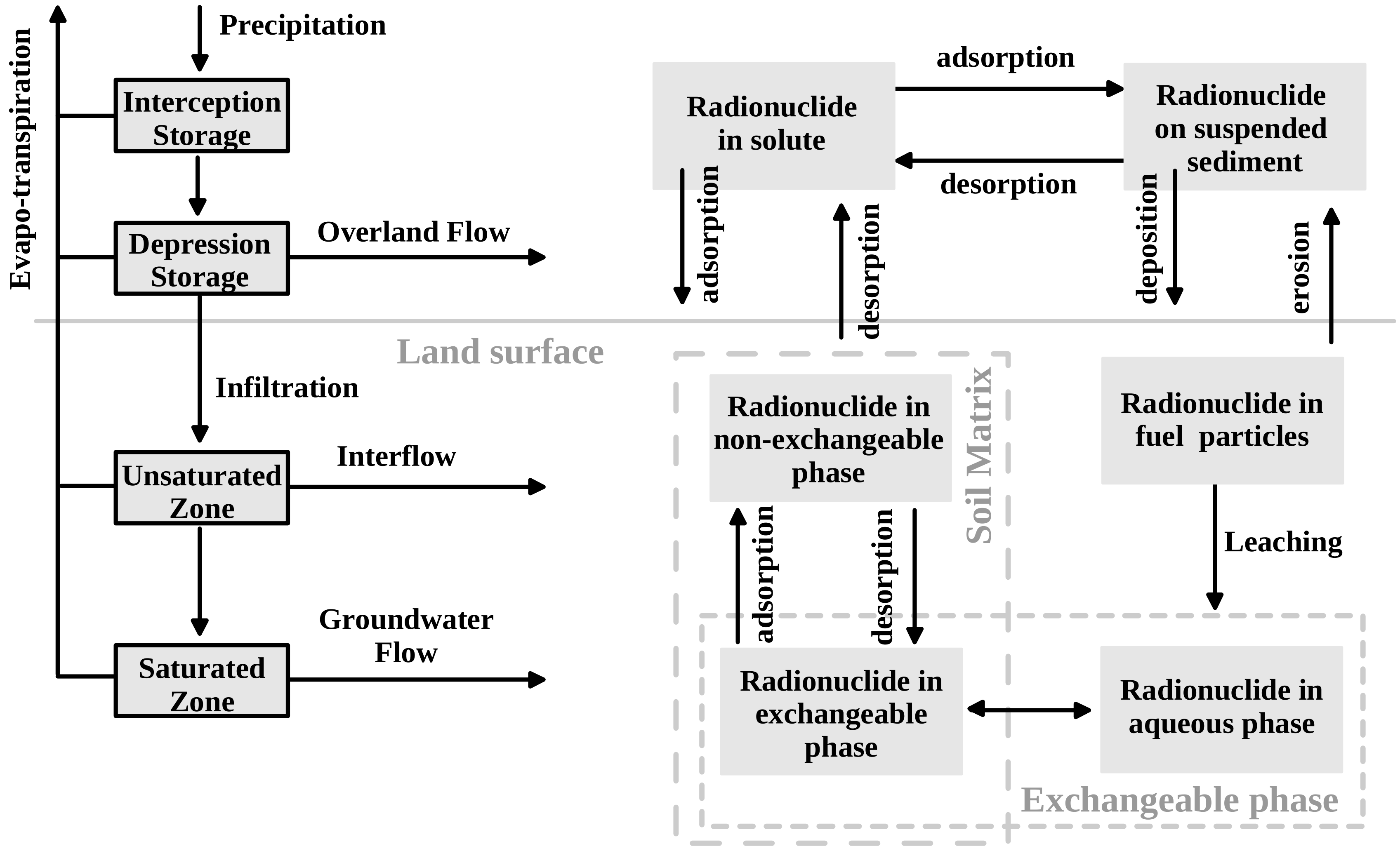

Figure 1.1. Hydrological and physical -chemical processes determining transport of radionuclides on catchment.

? Depression storage is that precipitation that fills the surface depressions, forming small puddles, ponding, or adding to the general wetness of the area. Water stored in the depression storage either evaporates or percolates into the soil zone
? Evapotranspiration represents water loss into the atmosphere by evaporation from both open water surface and soils, while transpiration refers to water drawn from the soil zone by the root systems of plants and vegetation and released to the atmosphere as a part of the life cycle of plants. The direct effect of evapotranspiration on the magnitude of surface runoff is not great.
? Interflow is that portion of the water infiltrating into the soil zone that moves in a horizontal direction due to the lower permeability of subsoils. The amount of interflow is again a residual of infiltration after ground-water recharge, soil moisture storage, and evapotranspiration have been subtracted.
? Groundwater runoff is defined as that part of the runoff contribution that originates from infiltrated precipitation after subtraction of the surface losses – surface runoff, evapotranspiration, and interflow.

The main physical exchange mechanisms of radionuclides are the deposition of contaminated suspended matter onto land surface and soil erosion into overland water. They are controlled by hydraulic factors (e.g., raindrop energy, overland water flow, sediment transport capacity), and depend strongly on the sediment size fractionation (e.g., clay, silt, sand and gravel). Radionuclide diffusion through overland water is a process that accounts for migration phenomena not related to sediment transport. Adsorption and desorption of a radionuclide by the upper soil layer are the main chemical exchange processes. These processes are not always completely reversible and are controlled by geo-chemical reactions of the dissolved radionuclides with the soil.

It is customary to distinguish the following mechanisms of soil erosion that can be considered as complementary to the hydrological processes governing the radionuclide migration on catchments (Hadley et al., 1985):

? Splash detachment and soil splash transport action caused by rain droplets,
? Soil detachment and soil transport of interrill flows or sheet,
? Soil detachment and soil transport of rill flows of overland runoff and subsurface runoff.

The following catchment properties control the soil erosion process (WMO-754, 1992):

? physical structure and chemical properties of soil,
? properties of liquid precipitation (size and energy of droplets, time distribution of rain),
? hydrodynamic properties of surface and subsurface flows,
? relief of catchment area,
? agricultural practices of land use,
? types and seasonal characteristics of vegetation on watershed.

In addition to the above factors intensity of erosion is dependent on inventory of particles capable of moving with run-off and hence the catchment history.

1.2. Physical-chemical processes governing the fate of radionuclide on catchment

The redistribution of radionuclides within catchments is also determining by physical-chemical processes in the system “soil matrix - soil water”, interaction of surface water with upper soil layer, radionuclide transfers between surface water and suspended sediments, leaching radionuclides from fuel particles, radionuclide migration through porous media.

Distribution of radionuclides in the soil-water system is governed by two types of processes: exchangeable sorption and transformation of species in soils. The available models account for the following species in soils: the water soluble phase; the exchangeable phase capable of fast exchange with solution; the non-exchangeable (fixed) phase not involved in the exchange with solution and the insoluble particles incorporating deposited radionuclides (in the case of the Chernobyl accident - fuel particles). The total of water soluble and exchangeable phases is called the mobile phase.

The water soluble fraction of a radionuclide can exist either as cations, as neutrally or as negatively charged complex with dissolved organic substances, or as mineral component of the soil moisture (Borzilov et al., 1989,1993; Konoplev et al., 1988 – 1999; Bulgakov et al., 1990-1999). The cation form of a radionuclide in solution is in equilibrium with the fraction of the radionuclide absorbed onto the solid particles. In the solid phase, radionuclides can be in exchangeable and non-exchangeable phase. In their exchangeable phase the radionuclide is sorbed by an ion exchange mechanism. The non-exchangeable phase consists of radionuclides originate from nuclear fuel particles or are radionuclides absorbed by a mechanism of irreversible sorption (i.e. incorporation into a mineral crystal lattice, formation of radionuclide-organic in soluble compounds etc.)

As a result of leaching from fuel particles radionuclides transfer to solution from which they reversibly transfer to the exchangeable phase. The following process of conversion of the exchangeable phase to non-exchangeable is called fixation and the reverse process of conversion of non-exchangeable phase to exchangeable is remobilization. The instantaneous and reversible transfer of radionuclides from solution to the exchangeable phase is called exchangeable sorption. The mechanism of such sorption for cesium and strontium is cation exchange, since by forms of water migration they belong to the group of cationogenic elements, i.e. occur in natural waters basically as cations (Voitkevich et al., 1990).

Soil particles transfer from soil to overland flow together with radionuclides contained in them. The composition of the overland flow may differ from that of the porous solution, with which soil particles are in equilibrium before entering the runoff. Therefore, sorption-desorption processes in the surface flow can result in redistribution of radionuclides between the solid and liquid phases. The mechanisms of sorption-desorption of radionuclides in the system “suspended particles-overland flow” are the same as the above mechanisms of sorption-desorption in the soil-water system.

It is important to note that the particle size distribution in solid runoff differs from the mechanical soil composition (Konoplev et al., 1996b). As compared to the upper soil layer the suspended material of the runoff is obviously enriched in fine-grained particles. The radionuclides concentration on fine particles are normally somewhat higher that in soil on the average (Pavlotskaya, 1974). This may have a noticeable effect on the activity of the solid runoff if the irreversible sorption of radionuclide has occurred on those soil fractions entrained in the sediment flow.

The exchangeable sorption of radionuclides in the system “upper soil layer - surface water”, like in soil, occurs by the mechanism of cation exchange, though it has its peculiarities. What makes it different is short term of a runoff event, continuos inflow of clean (without radionuclides) rain or snow melt water and different extent of interaction with runoff flow for radionuclides occurring in soil at different distances from the surface. In works (Bulgakov et al., 1990a; Bulgakov et al., 1999) it was shown that the concentration of radiocaesium and radiostrontium in the rain surface flow is not dependent on the rain intensity and runoff rate and is directly proportional to the concentration of analogue-cation. It can therefore be assumed that in the system upper soil layer-surface flow the cation-exchangeable equilibrium sets in. The optimum estimate of the interaction layer with which the runoff is in equilibrium is 0.5 cm. This is the layer where the concentration of the exchangeable form of cations and radionuclides should be determined.

The vertical migration of radionuclides leads to a change in their concentration in soil layers at different distances from the surface. The vertical migration occurs as a result of diffusion and advection of soluble forms of radionuclides in the porous media.

Benes et al. (1992) describe the sorption of the radionuclides by means of two parallel or consecutive reaction for ion exchange with two elements bounds at two different sites on the solid phase. The equation and parameters for all kinetic models were derived for general ion-exchange reactions.

Comans (1992) studied the caesium sorption on potassium and calcium saturated illite. Applying the linearisation method developed by Jannasch et al. (1988) to determine the number of processes, three consecutive reactions can be distinguished: one fast, instantaneous reaction and two distinct slow processes. For investigating the sorption of caesium on time scales of days to weeks which is most relevant for natural systems, two-box and three- box models were suggested and the isotherm of Freundlich was assumed to describe the equilibrium. Intercomparison with experimental data for periods longer than two weeks showed, that reversible reaction on calcium-illite was too slow, whereas the second process (irreversible reaction) was too fast. Therefore, a more complicated three- box model was used (Smith et al., 1996). This model assumed the existence of the easily accessible sorption sites and sorption sites, where the kinetically controlled process are followed by irreversible sorption.

2. Review of existing models of radionuclide transfer processes on catchment

2.1. Leaching of radionuclides from fuel particles

To describe the kinetics of radionuclides leaching from fuel particles several models are used the simplest of which is the first-order kinetics:

$$P(t) = P_0 exp\left(-kt\right)$$

where k is the first order rate constant, day^{-1}; P_0 is the activity of radionuclides in fuel particles in initial depositions; t is time since the accident, days.

In spite of simplified representation of the processes leading to radionuclides leaching from fuel particles, the first order model makes it possible to account for the available experimental dependencies (Konoplev & Bulgakov, 1999).

The only way to estimate parameters of radionuclides leaching from fuel particles in soils is to use experimental time dependence for the part of radionuclides occurring in soil incorporated in fuel particles. Direct measurement of activity of all fuel particles in soil is practically not feasible, but this can be calculated by difference in the part of the exchangeable form of radionuclides in the soil with and without nuclear fuel particles (Konoplev & Bulgakov, 1992; Konoplev & Bulgakov, 1995; Konoplev & Bulgakov, 1999). This method was used for estimating the leaching rate constants for fuel particles in the 30 km zone of the Chernobyl NPP. The most comprehensive lists of the rate constants of radionuclides leaching from fuel particles in different directions and at different distances from the Chernobyl NPP are given in works (Bulgakov, 1998; Kashparov et al., 1997).

2.2. Fixation and remobilization of radionuclide in soil

Most of the existing models account for fixation- remobilization of radionuclides in soils as a regular first order reaction (Benes et al., 1992; Comans & Hockley, 1992; Konoplev & Golubenkov, 1991; Wauters, 1994; Bulgakov & Konoplev 1992). An alternative approach to modeling fixation - remobilization is the diffusion model (Konoplev & Bulgakov, 1995; Bulgakov & Konoplev 1996; Bulgakov & Konoplev, 1999). This model assumes that the transfer of the exchangeable form to non-exchangeable occurs due to diffusion of radionuclides to the volume of soil particles.

Both the first-order model and the diffusion model account with satisfactory accuracy for the experimental data on the kinetics of radiocaesium and radiostrontium fixation, both in suspension and in actual soils (Bulgakov & Konoplev, 1999). A major advantage of the diffusion model is a fewer number of parameters and possibility of their determination in a short-term experiment.

The only method currently used to determine parameters of fixation and remobilization of radionuclides in soils is based on experimental curves. To calculate parameters of the diffusion model, data on radionuclides in soil at two different time moments are required, which makes possible their estimation for a large range of soils (Bulgakov, 1998). By characteristic values of the kinetic parameter of the diffusion model the soils were divided into three groups (Bulgakov, 1998).

To calculate rate constants for fixation and remobilization of radiocesium in models using the first-order kinetic equations much more detailed experimental data are needed.

Therefore, they can be estimated only for several types of soils, which constrains practical application of these models.

In addition to the kinetic fixation parameter, for modeling transformation of species the equilibrium part of the exchangeable (mobile) form has to be known. Methods to estimate this by properties of radionuclides and soils, unfortunately, have not been developed to date. Therefore, for estimation of the equilibrium part of the exchangeable form of radiocesium and radiostrontium experimental data is recommended to be used. For the soils of Belorus, Russia and Ukraine contaminated after the Chernobyl accident there is a large body of such data (see for example, Konoplev et al., 1988; Surkova & Pogodin, 199; Prosyannikov, 1998; Rauret et al., 1996; Petryaev et al., 1993). Consequently, for the catchments of these regions the equilibrium part of the exchangeable form of ^{137}Cs and ^{90}Sr (Bulgakov, 1998) can apparently be estimated with good accuracy.

2.3. Exchangeable sorption

The exchangeable sorption of radiostrontium and radiocaesium occurs by the mechanism of cation exchange. For description of homovalent exchange (exchange of cations with equal charge) in natural systems soil(rock)-water the simplest and most convenient is the Kerr isotherm equation

$$K_c(R/M) \, ? \, \frac{[SR] \, ? \, [M]_w}{[R]_w [SM]} \tag{2.1}$$

where $K_c(R/M)$ is the selectivity coefficient of the exchange of cations of equal valence R^+ and M^+; $[M^+]_w$ and $[R^+]_w$ are the equilibrium concentrations of metal and radionuclide cations in solution, mg-equ/L and Bq/L, respectively; $[SM]$ and $[SR]$ are equilibrium concentrations of metal and radionuclide cations sorbed by soil, meq/kg and Bq/kg, respectively.

Because of non-uniformity of adsorption sites, the effective value of the selectivity coefficient, strictly speaking, is not a constant and depends on other parameters of equation (2.1). To minimize this effect it was suggested to use as competing cations, analogue elements the nearest to the radionuclides by sorption characteristics (Konoplev et al., 1992; Bulgakov et al., 1992). For radiocaesium this is potassium and/or ammonium for radiostrontium - calcium and/or magnesium. This method is called the method of effective selectivity (Konoplev, 1998). Using this method the distribution coefficient of the mobile form is calculated as follows:

$$K_d^{ex} \, ? \, \frac{[R]_{ex}}{[R]_w} \, ? \, \frac{K_c^{eff}(R/M) \, ? \, [M]_{ex}}{[M]_w}$$

where $[M]_{ex}$ is the content in soil (rock) of exchangeable form of analogue cation; $K_c^{ex}(R/M)$ is the selectivity coefficient for exchange of cation R^+ for cation M^+.

A more complex method was proposed by Cremers with co-authors (Cremers et al., 1988; Sweeck et al., 1990; Wauters 1994) and improved by Konoplev (Konoplev, 1998; Konoplev & Konopleva, 1999). It consists in dividing the adsorption sites into two groups: selective and non-selective. The adsorption sites selective with respect to caesium cations are called *FES* (abbreviated from Frayed Edges Sites) and non-selective *RES* (Regular Exchange Sites). The effective distribution coefficient is calculated as the total of distribution coefficients on *FES* and *RES*.

$$K_d^{ex} \, ? \, K_d^{ex}(FES) \, ? \, K_d^{ex}(RES) \tag{2.2}$$

The values K_d^{ex}(FES) and K_d^{ex}(RES), in turn, are determined as:

$$K_d^{ex}(FES) ? \frac{K_c^{FES}(Cs/K) ? [FES]}{[K]_w ? K_c^{FES}(NH_4/K) ? [NH_4]} \tag{2.3}$$

$$K_d^{ex}(RES) ? \frac{K_c^{RES}(Cs/K) ? [K ? NH_4]^{RES}}{[K]_w ? K_c^{RES}(NH_4/K) ? [NH_4]} \tag{2.4}$$

where $K_c^{FES}(Cs/K)$ and $K_c^{RES}(Cs/K)$ are the selectivity coefficients of exchange of cesium and potassium cations on *FES* and *RES*, respectively. K_c^{FES} (NH_4/K) and $K_c^{RES}(NH_4/K)$ are the selectivity coefficients of exchange ammonium-potassium on *FES* and *RES*, respectively; [*FES*] is concentration of selective sorption centers in the soil solid phase, mg-equ/kg; $[K+NH_4]^{RES}$ is the content of potassium and ammonium cations absorbed on *RES*. mg-equ/kg; $[K^+]_w$ and $[NH_4^+]_w$ are potassium and ammonium concentrations in solution, respectively, mg-equ/L.

The product $K_c{}^{FES}(Cs/K)[FES]$ is referred to as exchangeable potential of radiocesium bonding and is designated as RIP^{ex} (Konoplev, 1998). The values K_c^{RES} and K_c^{RES} $(N\text{-}K)$ are normally taken to be unity. As in mineral soils cesium is coupled practically only with *FES*, equations (2.2)-(2.4) can be simplified:

$$K_d^{ex} ? \frac{RIP^{ex}}{[K]_w ? K_c^{FES}(NH_4/K) ? [NH_4]_w}$$

Of the two above methods to calculate the distribution coefficient of the mobile form of radionuclides in soils it is only the method of effective selectivity that can be provided with required input parameters without conducting measurement *in situ*. To calculate the distribution coefficient by this method, the effective coefficients of exchange selectivity, the content of exchangeable cations in soil and the concentration of cations in solution are needed.

The effective selectivity coefficients of exchange $K_c^{ex}(Sr/Ca)$ and $K_c^{ex}(Cs/K)$ are tabulated in work (Bulgakov, 1998). Using these coefficients and given data on soil types map can be drawn for the distribution coefficients of the radionuclide mobile form in the system "soil matrix – soil water" and "water – upper soil layer".

2.4. Formation of radionuclides concentrations on suspended sediments

The content of radionuclide on suspended material in the existing models is associated either with its concentration in the surface soil layer or/and concentration in the runoff water phase (Zheleznyak et al., 1993; Zheleznyak et al., 1996; Konoplev et al., 1999). In the first case, the proportionality coefficient is the enrichment coefficient and in the second - the distribution coefficient. Using the distribution coefficient implies fast achievement of sorption equilibrium in the system suspended material-runoff flow. Using the enrichment coefficient, vice versa, presupposes absence of processes of sorption-desorption in the system suspended material-runoff flow or in other words constant concentration of the radionuclide on soil particles as it transfers to runoff. The first assumption is valid for radionuclides occurring in soil primarily in the exchangeable form and the second - for radionuclides most of which are in the non-exchangeable form. In spite of different initial premises, both calculation methods in the equilibrium models are formally identical and the enrichment coefficient equals to ratio of distribution coefficients in the systems suspended material-runoff and the upper soil layer-porous solution. The enrichment coefficient, however, in most cases is a more physically justified parameter.

As compared to the upper soil layer the suspended material of the runoff is obviously enriched in fine-grained particles. The radionuclides concentration on fine particles are normally somewhat higher that in soil on the average (Pavlotskaya, 1974). It can therefore be expected that the enrichment coefficient is more than unity. The results of laboratory and field experiments on runoff plots, however, suggest that the values of K_e for cesium and strontium are close to unity (Bulgakov et al., 1992; Zubareva et al., 1989). Experiments on determination of K_e in natural runoff are not described in the literature.

The early models for the radionuclide transport in surface water bodies described only the total concentration of the radionuclides in water, and did not distinguish its forms. A first step to improve those models was to develop water-sediment interaction submodels. Codes that include a reasonable mathematical description of the radionuclide interactions with the solid phase, i.e., with the upper soil layer and with suspended sediments, have demonstrated to be more successful in predicting the aquatic transport of radionuclides (see reviews by Codell et al., 1982; Onishi et al., 1981; Santschi and Honeyman, 1989).

3. Governing equations of the RUNTOX model

3.1. Interception

Interception depends on the amount of water stored on the canopy and evaporation from canopy. Interception is modeled by an accounting procedure for canopy storage in which the rate of change in storage is calculated as (Befani, 1975)

$$\frac{?P}{?t} ? \begin{cases} ?R ? E_0, & 0 ? P ? P_{mx} \\ ? 0, & P > P_{mx} \end{cases}$$

where t is the time variable (s); P is the depth of water on canopy (m); P_{mx} is maximum interception storage on canopy (m); R is the rainfall rate (m s^{-1}); E_0 is the potential evaporation rate (m s^{-1}).

3.2. Depression Storage

Depression storage is expressed by (Befani,1982)

$$h_d(t) ? h_{mx} exp?? ?I??1 ? exp?- \frac{Q(t)}{h_{mx} exp?? ?I?}??$$

where h_d is the depression storage (m); h_{mx} is maximum depression storage of a horizontal land surface (m); I is the slope; Q is the cumulative excess rain from the start of the storm (m); ? is the parameter which depends on the surface characteristics.

3.3. Overland Flow

Two-dimensional overland flow equations are obtained by vertically averaging the three-dimensional equations over flow depth and using the above kinematic boundary conditions. These equations consist of a continuity equation and two momentum equations. By denoting q_x and q_y (m^2 s^{-1}) the components of the water discharges per unit width along the horizontal directions x and y, mass conservation yields to

$$\frac{?(? ? ?)}{?t} ? \frac{?q_x}{?x} ? \frac{?q_y}{?y} ? R_{ex}(x,y,t)$$

where t is the time variable (s); x and y are Cartesian coordinates (m); $?(x,y,t)$ is the free surface elevation (m); $?(x,y,t)$ is the bed surface elevation (m); R_{ex} is the rainfall excess rate (m s^{-1}).

The resulting momentum conservation equations in the x- and y-directions are

$$\frac{?q_x}{?t} ? \frac{?[q_x^2 / (? ? ?)]}{?x} ? \frac{?[q_x q_y / (? ? ?)]}{?y} ? u_? R ? ?g(? ? ?)?\frac{??? ? ?}{?x} ? \frac{???}{?x}?? ? \frac{1}{?}(?_x^? ? ?_x^?)$$

$$\frac{?q_y}{?t} ? \frac{?[q_x q_y / (? ? ?)]}{?x} ? \frac{?[q_y^2 / (? ? ?)]}{?y} ? v_? R ? ?g(? ? ?)?\frac{??? ? ?}{? ?y} ? \frac{?? ?}{?y}?? \frac{1}{?}(?_y^? ? ?_y^?)$$

where g is the acceleration of the gravity (m s^{-2}); ? is the density of water (kg m^{-3}); $u_?$, $v_?$ are the rain droplet velocity components at the free surface in x and y directions, respectively (m s^{-1}); $?^?$ is the shear stress at the free surface (kg m^{-1} s^{-2}); $?^?$ is the shear stress at the bed surface (kg m^{-1} s^{-2}); R is the rainfall rate (m s^{-1}). The subscripts x and y are the directional index.

For turbulent flow Reynolds' stress dominates and viscous stress may be negligible. The boundary shear stress can then be approximated by the Manning equation as

$$?_x^? ? ?g\frac{n^2}{(? ? ?)^{4/3}}\frac{q_x}{(? ? ?)}(q_x^2 ? q_y^2)^{1/2}$$

$$?_y^? ? ?g\frac{n^2}{(? ? ?)^{4/3}}\frac{q_y}{(? ? ?)}(q_x^2 ? q_y^2)^{1/2}$$

where n is the Manning roughness coefficient (s $m^{-1/3}$).

The shear stress on the free surface is generally produced by two factors: rain droplets and wind. While the influence of wind on overland flow may be negligible, the impact of rainfall is rather significant. As raindrops fall into flowing water, they generate splashing craters on the free surface, and turbulence in the flow. These can cause energy loss and increase flow resistance. The impact is greater for shallow water and laminar flow but less for deep water and turbulent flow. As the water gets deeper and flow gets more turbulent, the impact of rainfall on flow resistance may be negligible.

Using simple description of relationship between rainfall impact and flow resistance (Zhang and Cundy, 1989) leads to the following expression of surface shear stress due to rainfall

$$?_x^? ? ??R\frac{q_x}{(? ? ?)}$$

$$?_y^? ? ??R\frac{q_y}{(? ? ?)}$$

3.4. Unsaturated Subsurface Flow

Unsaturated zone is divided into three layers to simulate water movement through it. First layer of them is an upper active soil layer. Second layer is a plant root zone. Last layer is an unsaturated zone below the root zone.

The mathematical description of the water transport in the unsaturated subsurface implies assumptions which are commonly done for this type of problems. It is assumed that fluid transport through soil occurs in response to pressure gradients and gravitational body forces followed Darcy's flow equations. The porous medium is considered to be rigid. Pressure gradients in the gas phase to be negligible so that gas pressure remains effectively constant at atmospheric pressure.

Neglecting hysteresis and temperature gradients, movement of moisture through an unsaturated porous medium is described by

$$\frac{?}{?t}??S_w?? \frac{?}{?z}?k_r k\frac{?g}{?}\frac{?H}{?z}?? F_u$$

where t is the time (s); z is depth coordinate (m); ? is the porosity of the medium; ? is the fluid density (kg m^{-3}); k is component of the intrinsic permeability in z-direction (m^2); k_r is the relative permeability $(0 ? k_r ? 1)$; ? is the fluid dynamic viscosity (kg m^{-1} s^{-1}); H is the hydraulic head; F_u represents sources or sinks of fluid in the system (s^{-1}); and S_w is the volumetric saturation.

3.5. Infiltration

The infiltration rate is computed using either a relationship based on the Richards equation, the Svetlichnaya-Befani equation (Svetlichnaya, 1984) or the relationship of Morel-Seytoux (Morel-Seytoux et al., 1989).

3.5.1. Based on the Richards equation

In this case infiltration rate is calculated from the Richards equation as water flux at the land surface

$$Inf ? ?k_r k\frac{?g}{?}\frac{?H}{?z}$$

3.5.2. The Svetlichnaya-Befani equation

Infiltration rate based on the Svetlichnaya-Befani equation (Svetlichnaya, 1984) is defined as

$$Inf ? k_f ?1 ? ?L\frac{? ? ?}{k_f t}?^m exp??a\frac{?}{? ? ?}?1 ? ??? ?$$

$$?(t) ? \frac{R(t)}{2R_a(t)}?1 ? sign?\frac{dR}{dt}??$$

$$? ? ?\begin{matrix}0, & for\ \ Inf ? k_f\\ 1, & for\ \ Inf > k_f\end{matrix}$$

where k_f is the saturated hydraulic conductivity in the z-direction (m s^{-1}); ? is the soil porosity; ? is the initial moisture content; L is the thickness of soil layer (m); R_a is the mean rain intensity from the start of storm (m s^{-1}); a and m are parameters depending on hydrological properties of soil.

3.5.3. *The Morel-Seytoux model*

Infiltration during a rainfall event consists of two phases: infiltration to the time of ponding, t_p, when rainfall rate exceeds infiltration rate; and infiltration that occurs at a rate declining in time as root zone soil water content increases.

Before ponding, infiltration rate equals rainfall rate and excess rainfall rate is equal to zero. After ponding, and while a ponded condition lasts, infiltration rate equals capacity infiltration rate and excess rainfall rate equals rainfall rate minus capacity infiltration rate. In other words, the occurrence of ponding, i.e. the saturation of the soil surface, acts as a threshold determining which physical process should be considered to describe infiltration.

The expression used to compute ponding time is a generalization of the Mein and Larson equation for a variable rainfall intensity derived by Morel-Seytoux (1978, 1984)

$$t_p \; ? t_0 ? \frac{1}{R} ? \frac{S_f}{R^* \; ? \; 1} \; ? W^0 ?$$

where t_0 is the time at the beginning of the current time step, R is the mean rainfall rate during the current time step, S_f is the storage-suction factor, R^* is the normalized mean rainfall rate (ratio of R over the hydraulic conductivity at natural saturation K) and W^0 is the cumulative infiltration depth since the beginning of the current continuous rainfall up to time t_0.

The storage-suction factor is a composite factor affecting ponding and the infiltration process. Its value can be computed as:

$$S_f \; ? \; (? \; ? ?_i) H_c (?_i)$$

where ? is water content at natural saturation, $?_i$ is mean initial water content in an upper soil layer and $H_c(?_i)$ is the effective capillary drive, a quantity which depends but only slightly upon the initial water content.

Under a ponded condition, infiltration rate is equal to the capacity infiltration rate. The capacity cumulative infiltration depth up to time t is computed by the equation:

$$K(t ? t_p) \; ? \; W^t \; ? W_p \; ? \; S_f \ln \frac{S_f \; ? W^t}{S_f \; ? W_p}$$

where W^t is cumulative infiltration depth at the end of current time step, W_p is the cumulative infiltration depth at the time of ponding.

Mean infiltration rate after ponding is simply the incremental infiltration depth during current time step divided by the time step $? t$

$$Inf \; ? \; \frac{W^t \; ? W^0}{? t}$$

3.6. Groundwater Flow

Assume that the Dupuit-Forchheimer assumption (Dupuit, 1863; Forchheimer, 1886) is applicable. This assumption is identified with the assumption that the vertical component of the specific discharge vector is neglected or the resistance to flow in the vertical direction is negligible (Strack, 1984). Then shallow groundwater flow is described by the equation:

$$\frac{?}{?\,t}(S_g H) ? \frac{?}{?\,x}?T_x\frac{?\,H}{?\,x}? ? \frac{?}{?\,y}?T_y\frac{?\,H}{?\,y}? ? \frac{H ? H^?}{c} ? N$$

where t is the time (s); x, y are the Cartesian spatial coordinates (m); H is the hydraulic head (m); T is the transmissivity ($m^2\ s^{-1}$); N is the vertical recharge into the saturated zone ($m\ s^{-1}$); S_g is the storage coefficient (dimensionless); c is the resistance of a semipermeable layer, equal to its thickness divided by its hydraulic conductivity (s); H^* is the hydraulic head at a point separated from the aquifer by a semipermeable layer (m).

The third term on the right-hand side of the equation is added to cover cases of flow in leaky aquifers. For shallow confined flow T equals $K(? - ?)$, for shallow unconfined flow T equals $K(H-?)$, where K is the hydraulic conductivity tensor ($m\ s^{-1}$); ? and ? are the upper surface and bottom surface elevation of the aquifer, respectively (m).

For shallow unconfined flow S_g is the coefficient of phreatic storage, representing the fraction of a unit volume available for storage due to vertical movement of a phreatic surface. For shallow confined flow S_g is defined by expression:

$$S_g ? ?? ?? ?S_s ? ?? ?? ?? g ?m_v ??? ?,$$

where S_s is the coefficient of specific storage (m^{-1}); ? is the porosity (dimensionless); g is the acceleration due to gravity ($m\ s^{-2}$); ? is the density of water ($kg\ m^{-3}$); ? is the compressibility of water ($m^2\ N^{-1}$); m_v is the coefficient soil volume compressibility ($m^2\ N^{-1}$).

3.7. Sediment Continuity

Mass conservation for sediment yields to

$$\frac{?(hS)}{?t} ? \frac{?}{?x_i}(q_i S) ? \frac{?}{?x_i}?hD_{ij}\frac{?S}{?x_j}? ? (1 ? ?)?_b\frac{??}{?t}$$

where x_i is the spatial Cartesian coordinates (m); ? is the porosity of upper soil layer (dimensionless); $?_b$ is the density of soil matrix ($kg\ m^{-3}$); S is the suspended sediment concentration ($kg\ m^{-3}$).

D_{ij} is the coefficient of horizontal dispersion defined by

$$D_{11} ? D_L cos^2? ? D_T sin^2?$$

$$D_{22} ? D_L sin^2? ? D_T cos^2?$$

$$D_{12} \; ? \; D_{21} \; ? \; (D_L \; ? \; D_T)\, sin? \;\; cos?$$

where ? is the angle between the flow direction and the x axis; D_L and D_T are the coefficients of longitudinal and transverse mixing (m^2 s^{-1}), respectively.

3.8. Erosion/Deposition

Change of the bed surface elevation is described by the bed sediment conservation equation (Exner's equation)

$$?_b(1 \; ? \; ?)\frac{??}{?t} \; ? \; q^s \; ? \; q^b$$

where q^s and q^b are the deposition and erosion rates (kg m^{-2} s^{-1}), respectively.

Erosion rate q^b is a sum of splash erosion rate as q_s^b and hydraulic erosion rate as q_h^b

$$q^b \; ? \; q_s^b \; ? \; q_h^b$$

3.8.1. *Splash ero*sion

Splash erosion rate is calculated by formula

$$q_s^b \; ? \; c_f r(h)? \; R_{ex}^{?}$$

where R_{ex} is rainfall intensity (mm h^{-1}); c_f is raindrop soil erodibility coefficient (J^{-1}); $r(h)$ is a reduction factor representing the reduction in splash erosion caused by increasing depth of water; and ? and ? are empirical coefficients (Wicks, 1988).

The function *r(h)* is defined by empirical expression (proposed by Park et al., 1982; to fit the data of Palmer, 1965)

$$r(h) \; ? \; \begin{array}{l} ?\, exp(1 \, ? \, h/d_m), \quad if \;\; h>d_m \\ ? \\ ? \qquad\qquad\quad 1, \quad if \;\; h \, ? \, d_m \end{array}$$

where h is water depth (m), and d_m is median raindrop diameter (m).

The median raindrop diameter is determined from the Laws and Parsons (1943) equation

$$d_m \; ? \; 0.00124 R_{ex}^{0.182}$$

3.8.2. Hydraulic erosion

Hydraulic erosion rate and deposition rate are estimated as being linearly dependent on the difference between the equilibrium concentration and the current sediment concentration

$$q^s \ ? \ max ? 0, w_0 (S \ ? \ S^?) ?$$

$$q_h^b \ ? \ max ? 0, E_r w_0 (S^? \ ? \ S) ?$$

where S is the current local sediment concentration (kg m^{-3}); $S*$ is the concentration at equilibrium sediment transport capacity (kg m^{-3}); w_0 is the particle settling velocity (m s^{-1}); E_r is the overland flow erodibility coefficient.

Particle settling velocity is calculated from particle size and density, assuming the particles have drag characteristics and terminal fall velocities similar to those of spheres. The fall velocity of a sphere is given by (Fair and Geyer 1954, Randkivi 1967, Vanoni, 1975)

$$w_0{}^2 \ ? \ \frac{4}{3} \frac{g d_s}{C_d} \begin{matrix} ? \\ ? \\ ? \end{matrix} \frac{?_s \ ? \ ?}{?} \begin{matrix} ? \\ ? \\ ? \end{matrix}$$

where C_d is the drag coefficient (dimensionless); d_s is the sediment diameter (m); $?_s$ is the density of sediment particle (kg m^{-3}); ? is the density of fluid (kg m^{-3}).

The drag coefficient is a function of particle Reynolds number,

$$C_d \ ? \ \frac{24}{R_n} \ ? \ \frac{3}{\sqrt{R_n}} \ ? \ 0.34$$

in which R_n is the particle Reynolds number, defined as

$$R_n \ ? \ \frac{w_0 d_s}{?}$$

3.9. Transport Capacity

The equilibrium sediment concentration is defined by

$$S^? \ ? \ \frac{p}{(q_x^2 \ ? \ q_y^2)^{1/2}}$$

where p is the particle transport capacity per unit width of flow (kg m^{-1} s^{-1}).

The sediment transport capacity for a given grain size can be calculated by using one of six algorithms: Engelund-Hansen total load equation, Einstein-Brown bedload equation, Bagnold total load equation, Ackers-White total load equation, Yalin bedload equation, and modified Yalin bedload equation.

3.9.1. Yalin bedload equation

The sediment carrying capacity of the runoff flow can be expressed by Yalin’s equation (Yalin,1963), which was reported in the following form:

$$p ? (?_s ? ?)d_s v^? \; 0.635\, s \overset{?}{\underset{?}{?}} 1 - \frac{1}{?s} ln(1 ? ?s) \overset{?}{\underset{?}{?}}$$

where $v^? ? \sqrt{ghI}$ is the shear velocity (m s^{-1}); I is the slope of energy gradient (m m^{-1}); $s ? max ? 0, Y / Y_{cr} ? 1 ?$ is the dimensionless excess tractive force; $? ? 2.45(? / ?_s)^{0.4} \sqrt{Y_{cr}}$; $Y ? v^{?2} / ?(?_s / ? ? 1)gd_s ?$ is the densimetric particle Froude number.

The critical tractive force at which sediment movement begins, Y_{cr}, can be found from the Shields diagram (Shields, 1936), which is based on the particle Reynolds number

$$Y_{cr} ? FR \overset{?}{\underset{?}{?}} \frac{d_s v^?}{?} \overset{?}{\underset{?}{?}}$$

where $?$ is kinematic viscosity of the flow (m^2 s^{-1}); FR is functional relationship.

3.9.2. Modified Yalin bedload equation

In this case, the critical tractive force is defined from the modified Shields curve that can be separated into three parts based on the values of the Yalin parameter Y_m. Regression of these segments gives us the following relationships:

$logY_{cr} ? 0.041(logY_m)^2 ? 0.356\, logY_m ? 0.977$	$Y_m ? 100$
$logY_{cr} ? 0.132\, logY_m ? 1.804$	$100 ? Y_m ? 3000$
$Y_{cr} ? 0.045$	$Y_m ? 3000$

where Y_m is defined as $?? ?_s ? ? ? g\, d_s^3 / ? ?^2 ?^{0.5}$.

3.9.3. Engelund-Hansen total load equation

The formula developed by Engelund and Hansen (1967) for the total bed-material load capacity is:

$$p ? 0.05 ?_s (? ?_b^3)^{0.5} \frac{q_x^2 ? q_y^2}{d_s ? gh ? ?_s ? ? ?^2}$$

where $?_b$ is the bottom shear stress.

3.9.4. Einstein-Brown bedload equation

The Einstein-Brown bedload equation also was obtained from flume experiments under unidirectional flow over well-sorted sediment. With shear stress being replaced by the shear velocity, the Einstein-Brown bedload equation is given here

$$p = 40\rho_s w_0 d_s \left[\frac{\rho v_*^2}{g d_s(\rho_s - \rho)}\right]^3$$

3.9.5. Bagnold total load equation

The Bagnold equation is

$$p = 0.005\rho\rho_s v_*^2 \frac{(q_x^2 + q_y^2)^{0.5}}{gh(\rho_s - \rho)} \exp\left[0.7\left(\frac{Y}{Y_{cr}} - 1\right)\right]$$

3.9.6. Ackers-White total load equation

The Ackers-White total load formula was developed by Ackers and White (1973). Various dimensionless parameters were used by the formula: grain size parameter D_{gr}, mobility number F_{gr}, critical mobility number A_{gr}, and transport parameter G_{gr}. Sediment transport rate is calculated from:

$$p = \rho_s d_s G_{gr} \frac{(q_x^2 + q_y^2)^{0.5}}{h} \left[\frac{(q_x^2 + q_y^2)^{0.5}}{h v_*}\right]^n$$

in which

$$D_{gr} = d_s \left[\frac{g}{\nu^2}\left(\frac{\rho_s}{\rho} - 1\right)\right]^{1/3}$$

$$A_{gr} = 0.17 \qquad D_{gr} \geq 60$$

$$A_{gr} = 0.23\left(D_{gr}\right)^{-0.5} + 0.14 \qquad 1 < D_{gr} \leq 60$$

$$G_{gr} = C_1\left(F_{gr}/A_{gr} - 1\right)^m$$

$$F_{gr} = \left[\frac{v_*^n}{\left(g d_s\left(\rho_s/\rho - 1\right)\right)^{0.5}}\right]\left[\frac{(q_x^2 + q_y^2)^{0.5}}{5.75h \log\left(10h/d_s\right)}\right]^{1-n}$$

In given equations, n is a transition exponent, C_1 and m are respectively the coefficient and exponent in the transport function. Data from flume experiments were used to determine the values of these coefficients. For D_{gr}>60 (d_s>2.5 mm):

$$n = 0$$

$$m ? 1.5$$

$$C_1 ? 0.025$$

For $1<D_{gr}?60$

$$n ? 1.0 ? 0.56\, logD_{gr}$$

$$m ? 9.66 D_{gr} ? 1.34$$

$$logC_1 ? 2.86 logD_{gr} ? ?logD_{gr}?^2 ? 3.53$$

The Ackers-White method is not applicable if $D_{gr}<1$ ($d_s<0.04$ mm).

3.10. Overland Radionuclide Transport in Solute

The complex process of radionuclide transport in soluble phase is affected by many factors such as advection, diffusion and adsorption-desorption processes. The species transport equation is established by writing a mass balance over a stationary control volume through which the fluid is flowing. When diffusion effects are significant, the use of Fick's law results in the appearance of additional terms. The complete radionuclide transport in the aqueous phase by overland flow is described by the equation

$$\frac{?(hC)}{?t} ? \frac{?}{?x_i}(q_iC) ? \frac{?}{?x_i}?hD_{ij}\frac{?C}{?x_j}? ? ?hC ? a_shS?k_d^sC ? C_s?? Z_?a_b?k_d^b\frac{?_b}{?}C ? C_b?$$

where C is the volumetric radionuclide activity in aqueous phase (Bq m^{-3}); C_s is the radionuclide activity in exchangeable phase on suspended sediment (Bq kg^{-1}); C_b is the volumetric radionuclide activity in exchangeable phase in upper soil layer (Bq m^{-3}); Z_* is the thickness of active upper soil layer (m); ? is the radionuclide decay constant (s^{-1}); k_d^s and k_d^b are the partition coefficients for "water-suspended sediment" and "water-upper soil layer" system, respectively; a_s and a_b are the exchange rates for "water-suspended sediment" and "water-upper soil layer" system (s^{-1}), respectively.

3.11. Overland Particulate Radionuclide Transport

Radionuclide transport on suspended sediment is described by the following advection-diffusion equations with the sink-source term describing erosion-deposition exchange processes

$$\frac{?\,(hSC_s)}{?\,t} ? \frac{?}{?\,x_i}(q_iSC_s) ? \frac{?}{?\,x_i}?hD_{ij}\frac{?\,SC_s}{?\,x_j}?? ?\,hSC_s ? a_shS\,?k_d^sC ? C_s?? \frac{1}{?_b}q_bC_b ? q_sC_s$$

$$\frac{?\,(hSC_s^f)}{?\,t} ? \frac{?}{?\,x_i}(q_iSC_s^f) ? \frac{?}{?\,x_i}?hD_{ij}\frac{?\,SC_s^f}{?\,x_j}?? ?\,hSC_s^f ? \frac{1}{?_b}q_bC_b^f ? q_sC_s^f$$

$$\frac{?(hSC_s^p)}{?t} ? \frac{?}{?x_i}(q_iSC_s^p) ? \frac{?}{?x_i}?hD_{ij}\frac{?SC_s^p}{?x_j}?? ?hSC_s^p ? \frac{1}{?_b}q_bC_b^p ? q_sC_s^p$$

where C_s^f and C_s^p are the radionuclide activity in fixed phase and fuel particles on suspended sediment (Bq kg^{-1}), respectively; C_b^f and C_b^p are the volumetric radionuclide activity in fixed phase and fuel particles per soil solid volume in upper soil layer (Bq m^{-3}), respectively.

3.12. Contamination of Upper Soil Layer

Contamination of the active upper soil layer is described by the equations

$$\frac{?}{?t}(Z_?C_b) ? a_bZ_??k_d^b\frac{?_b}{?}C ? C_b? ? \frac{1}{1? ?}?\frac{1}{?_b}q_bC_b ? q_sC_s?$$

$$\frac{?}{?t}(Z_?C_b^f) ? ?\frac{1}{1? ?}?\frac{1}{?_b}q_bC_b^f ? q_sC_s^f?$$

$$\frac{?}{?t}(Z_?C_b^p) ? ?Z_?(? ? ?_p)C_b^p ? \frac{1}{1? ?}?\frac{1}{?_b}q_bC_b^p ? q_sC_s^p?$$

where $?_p$ is the first-order constant of radionuclide leaching from fuel particles (s^{-1}). The last equation is described the leaching of radionuclides from the fuel particles and erosion/deposition processes for the fuel particles.

The leaching of radionuclides from the fuel particles is described by the first term on right-hand side of the third equation.

3.13. Unsaturated Species Transport

Assuming that the ion-exchange reaction transferring activity between aqueous and solid exchangeable phases has a sufficiently short timescale, we will consider this reaction as instantaneous, which is described by a linear equilibrium isotherm. Transfers of activity to and from the fixed phase are modelled by first-order rate constants $?_{sf}$ and $?_{fs}$, respectively, as shown in Figure 3.1 (Konoplev et al., 1992b).

Ignoring chemical diffusion in the solid phase, the equation for advective-dispersive transport of radionuclide activity in the exchangeable phase in unsaturated media may be written in the form

$$\frac{?C_u}{?t} ? \frac{?}{?z}??S_wD_z\frac{?C_u^l}{?z}? ? \frac{?}{?z}(vC_u^l) ? ?C_u ? (1? ?)(?_pC_b^p ? ?_{fs}C_u^f ? ?_{sf}C_u^s)$$

where C_u is the radionuclide activity per total volume (Bq m^{-3}); C_u^l is the volumetric radionuclide activity in aqueous phase (Bq m^{-3}); C_u^s is the volumetric exchangeable sorbed radionuclide activity in solid phase (Bq m^{-3}); C_u^f is the volumetric radionuclide activity which is fixed in mineral lattice (Bq m^{-3}); $?_{sf}$ and $?_{fs}$ are first-order rate constants of transfers radionuclide activity to and from the fixed phase.

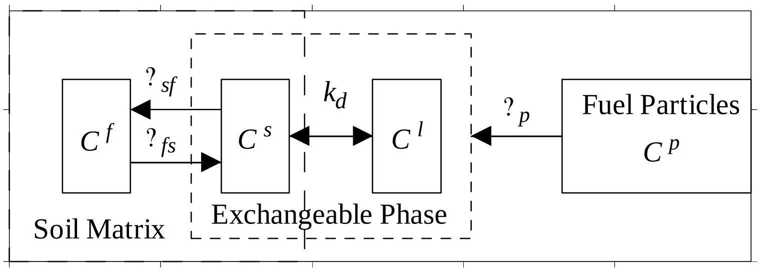

Figure 3.1. Schematic representation of the kinetic sorption model for system ‘water – soil matrix’.

D_z is the dispersion coefficient, defined by:

$$?S_wD_z\ ?\ ?_L|v|\ ?\ ?S_w?D_0$$

where $?_L$ is the longitudinal dispersivity (m); $|v|$ is the magnitude of the Darcy velocity in z-direction (m s^{-1}); D_0 is the molecular diffusion coefficient (m^2 s^{-1}); and ? is the tortuosity.

The total radionuclide activity is defined by:

$$C_u\ ?\ ?S_wC_u^l\ ?\ (1\ ?\ ?)C_u^s$$

Volumetric phase activities are interrelated through solid-aqueous partition coefficient k_d^u, according to relationship

$$k_d^u\ ?\ \frac{C_u^s?}{C_u^l?_b}$$

The transport of activity in the fixed phase is described by

$$\frac{?C_u^f}{?t}\ ?\ ?_{sf}C_u^s\ ?\ (?_{fs}\ ?\ ?)C_u^f$$

3.14. Groundwater Species Transport

The equation for advective-dispersive transport of radionuclide activity in the exchangeable phase by shallow groundwater flow may be written in the form

$$\frac{?}{?t}?TC_g?\ ?\ \frac{?}{?x_i}?T?D_{ij}^g\frac{?C_g^l}{?x_j}?\ ?\ \frac{?}{?x_i}(Q_iC_g^l)\ ?\ ?TC_g\ ?\ (1\ ?\ ?)T(?_{fs}C_g^f\ ?\ ?_{sf}C_g^s)\ ?\ F_{sg}$$

where x_i are the Cartesian spatial coordinates (m); C_g is the radionuclide activity per total volume (Bq m^{-3}); C_g^l is the volumetric radionuclide activity in aqueous phase (Bq m^{-3}); C_g^s is the volumetric exchangeable sorbed radionuclide activity in solid phase (Bq m^{-3}); C_g^f is the volumetric radionuclide activity which is fixed in mineral lattice (Bq m^{-3}); F_{sg} is the species source-sink (Bq m^{-2} s^{-1}).

The discharge vector Q is defined as

$$Q_i \; ? \; ?TK_i \frac{?H}{?x_i}.$$

For shallow confined groundwater flow T equals $(? \text{-} ?)$, for shallow unconfined flow T equals $(H\text{-}?)$. D_{ij}^g is the dispersion tensor, defined by:

$$?D_{ij}^g \; ? \; ?_T |v| ?_{ij} \; ? \; (?_L \; ? \; ?_T)\frac{v_i v_j}{|v|} \; ? \; ??D_0 ?_{ij}$$

where $?_L$ and $?_T$ are the longitudinal and transverse dispersivities (m), respectively; $?_{ij}$ is the Kronecker delta; $|v|$ is the magnitude of the Darcy velocity.

The total radionuclide activity is defined by:

$$C \; ? \; ?C_g^l \; ? \; (1 ? ?_T)C_g^s$$

where $?_T$ is the total porosity of the medium; and volumetric phase activities are interrelated through solid-aqueous partition coefficient k_d^g, according to relationship

$$k_d^g \; ? \; \frac{C_g^s ?}{C_g^l ?_s}$$

where $?_s$ is the soil solid density (kg m^{-3}).

The transfers radionuclide activity to and from the fixed phase is described by

$$\frac{?C_g^f}{?t} \; ? \; ?_{sf} C_g^s \; ? \; (?_{fs} \; ? \; ?)C_g^f$$

$$C_g^f(0,x) \; ? \; C_f^0(x),$$

where C_f^0 is the initial activity in the fixed phase.

4. Comparisons with existing solutions

4.1. Dam Break Problem

Consider a wide channel having a barrier placed across its width. Let h_1 and h_2 be respectively the height of the water upstream and downstream. At time $t=0$ the barrier is suddenly removed. The flow consists of bore traveling downstream and a rarefaction wave traveling upstream. The analytical solution of this problem is given in Stoker (1957):

$$h(x,t) = \begin{cases} h_1 & \text{if } \dfrac{x}{t} \le -\sqrt{gh_1} \\ \dfrac{1}{9g}\left(2\sqrt{gh_1} - \dfrac{x}{t}\right)^2 & \text{if } -\sqrt{gh_1} \le \dfrac{x}{t} \le u_m - \sqrt{gh_m} \\ h_m & \text{if } u_m - \sqrt{gh_m} \le \dfrac{x}{t} \le s \\ h_2 & \text{if } s \le \dfrac{x}{t} \le \infty \end{cases}$$

where h_m and u_m are given in terms of velocity of propagation of the shock s by

$$h_m = \frac{1}{2}\left(\sqrt{1 + \frac{8s^2}{gh_2}} - 1\right)h_2$$

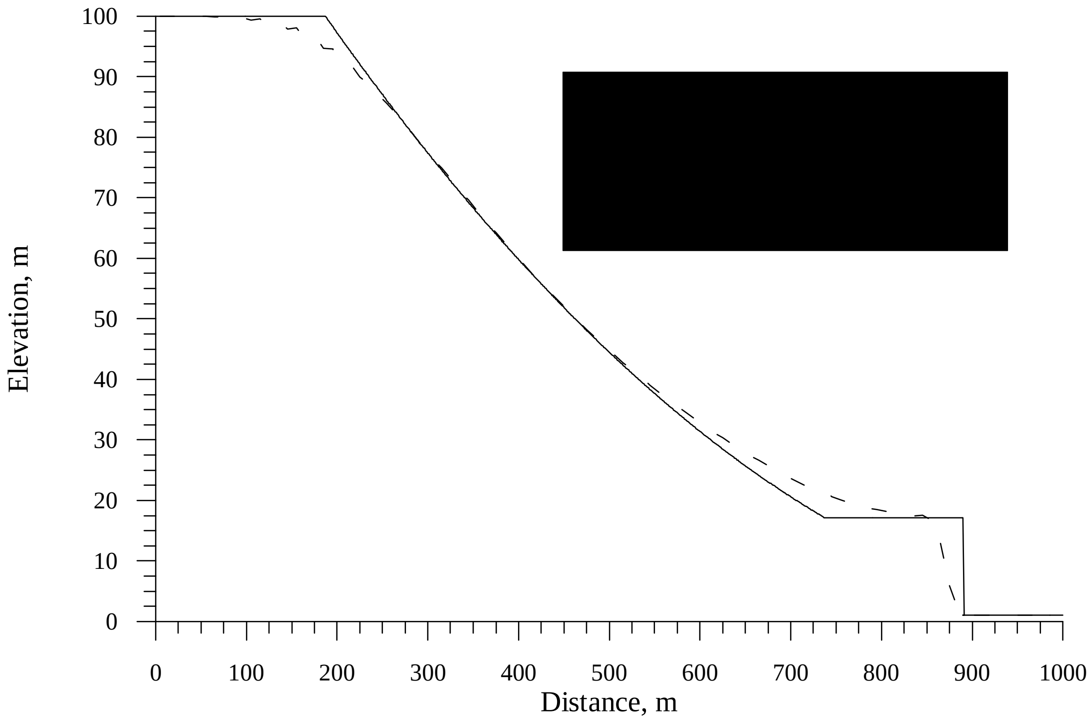
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Figure 4.1. Comparison of RUNTOX Numerical Solution to the Analytical Solution of the Dam Break Problem After 10 seconds.

$$u_m = s - \frac{gh_2}{4s}\left(1 + \sqrt{1 + \frac{8s^2}{gh_2}}\right)$$

and s is the positive real solution of the equation

$$u_m + 2\sqrt{gh_m} - 2\sqrt{gh_1} = 0$$

The initial conditions are defined as

$$h_0 = \begin{cases} h_1 & if \quad x \le 500 \\ h_2 & if \quad x > 500 \end{cases}$$

$$u_0(x) = 0$$

To test the ability of the RUNTOX code to solve correctly the Saint Venants equations, the RUNTOX is applied to the dam break problem. The water depth before the dam is 100 m, whilst outside the dam it is 1 m. Comparison of analytical solution with numerical result of the evolution of the flow at time t=10 s is shown in Figure 4.1.

4.2. Two-Dimensional Groundwater Flow

Consider a two-dimensional groundwater hillock spreading over an impervious horizontal base in a two-dimensional rectangular (15 cm by 10 cm) domain. The purpose of the problem is to demonstrate how RUNTOX propagates the phreatic surface in two dimensions. The groundwater hillock spreading over an impervious bottom is described by the Boussinesq's equation

$$\frac{\partial h}{\partial t} = \frac{k}{m}\frac{\partial}{\partial x}\left(h\frac{\partial h}{\partial x}\right) + \frac{k}{m}\frac{\partial}{\partial y}\left(h\frac{\partial h}{\partial y}\right) \qquad (4.1)$$

where t is the time; x and y are the spatial coordinates; k is the hydraulic permeability; h is the hydraulic head; m is the storage coefficient.

Sokolov (1956) provided an analytical solution of the equation (4.1) which can be written in form

$$h(t,x,y) = -\frac{m}{8k}\frac{(x - x_0)^2 + (y - y_0)^2}{t - t_0} + c\left|t - t_0\right|^{-1/2}$$

where x_0, y_0, t_0, and c are arbitrary constants.

Parameter values used in numerical simulations are

$m=0.02$

$k=0.05$ cm/h;

$t_0=1$ h;

$x_0=0$ cm;

$y_0=0$ cm;

$c=20$ cm $\text{h}^{1/2}$.

The whole right and upper boundaries are maintained at a prescribed head gradient of

$$\left.\frac{?h}{?x}\right|_{x?15\ \text{cm}} ? ?\frac{3}{2}(1?t)^{?1}$$

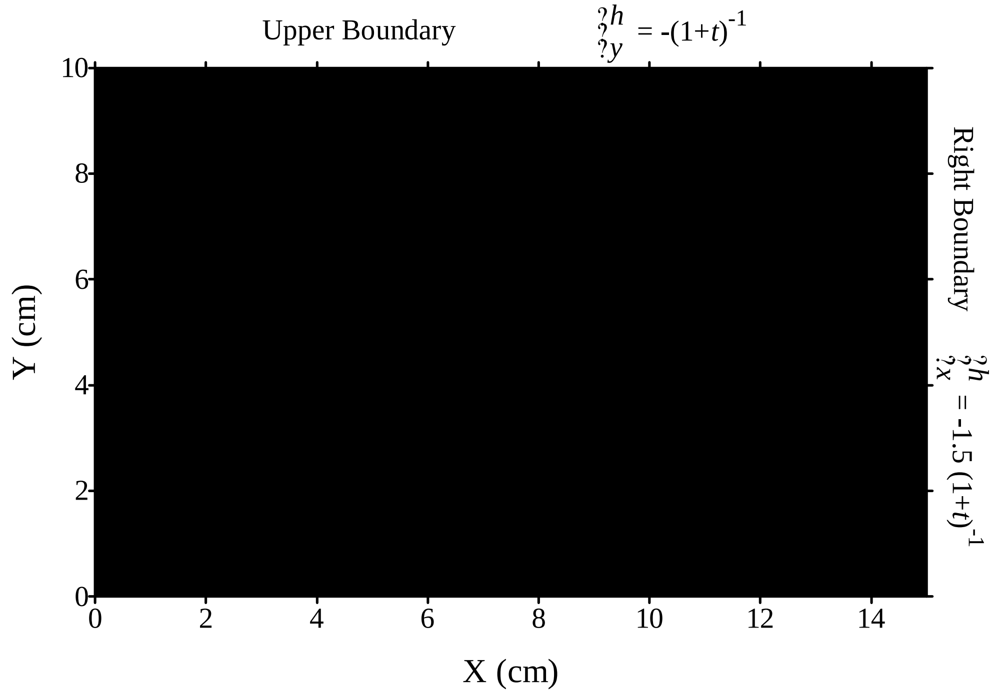

Figure 4.2. Two-Dimensional Groundwater-Flow Problem Geometry, Discretization, and Boundary Conditions.

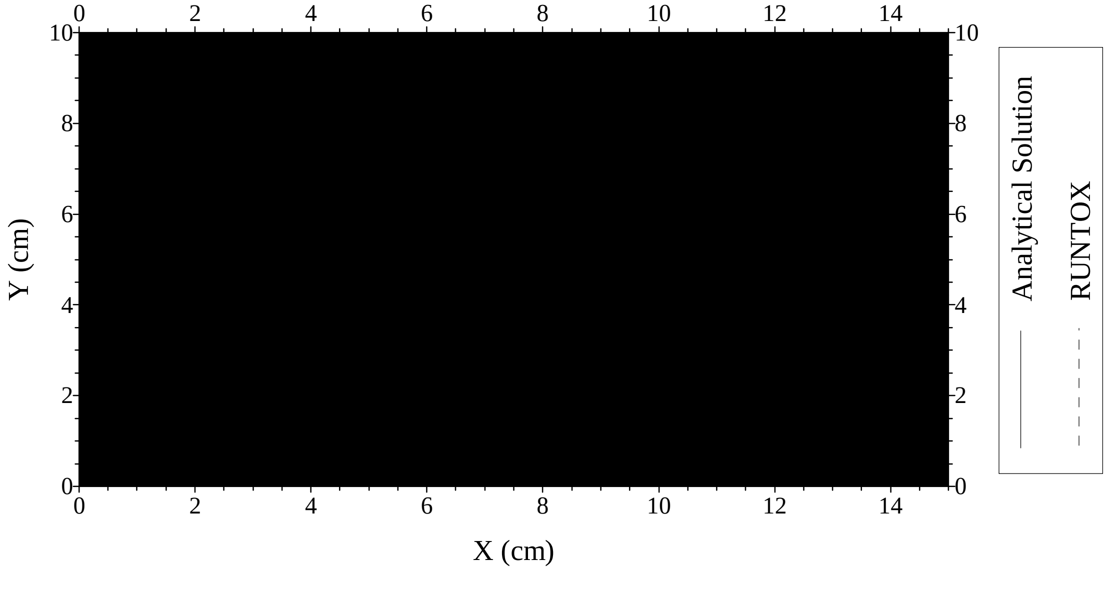

Figure 4.3. Comparison of Numerical Solution to the Analytical Solution After 2 Hours of Groundwater Hillock Spreading

$$\left.\frac{?h}{?y}\right|_{y?10\ \mathrm{cm}} ? ?(t?1)^{?1},$$

respectively. All other boundary surfaces are defined as zero-flux surfaces. Figure 4.2 provides a schematic of the problem and illustrates the discretization and boundary condition placement.

Initial conditions for the equation (4.1) are

$$h(0,x,y) ? ?\frac{1}{20}(x^2 ? y^2) ? 20$$

The hydraulic head distribution predicted by RUNTOX after 2 hours is compared to the analytical solution in Figure 4.3.

4.3. Unsaturated Species Transport

Ross et al. (1982) proposed to consider the transport in a one-dimensional, semi-infinite, horizontal tube of partially saturated soil as a test problem. The object of this problem is to determine the species concentration field in a tube of soil that has a uniform initial species concentration and moisture content and in which the upstream boundary is maintained at a prescribed moisture content and a prescribed solute concentration. Transport processes include both advection and hydrodynamic dispersion as functions of moisture content. Semi-analytical solution for the one-dimensional flow domain represented in this unsaturated transport problem was provided by Philip (1955). Semi-analytic solution for same transport problem was obtained by Smiles et al. (1977).

A saturation commencing at time zero is assigned on the left boundary of 20-cm horizontal soil tube, and a constant liquid pressure of 93,157.21 Pa was specified on the right boundary. The species boundary conditions are 1.0 g/L species concentration commencing at time zero at the left boundary and 0.1 g/L at the right boundary. The initial conditions are a liquid pressure of 93,157.21 Pa and species liquid concentration of 0.1 g/L. Values of other parameters used in simulations are given in Table 4.1.

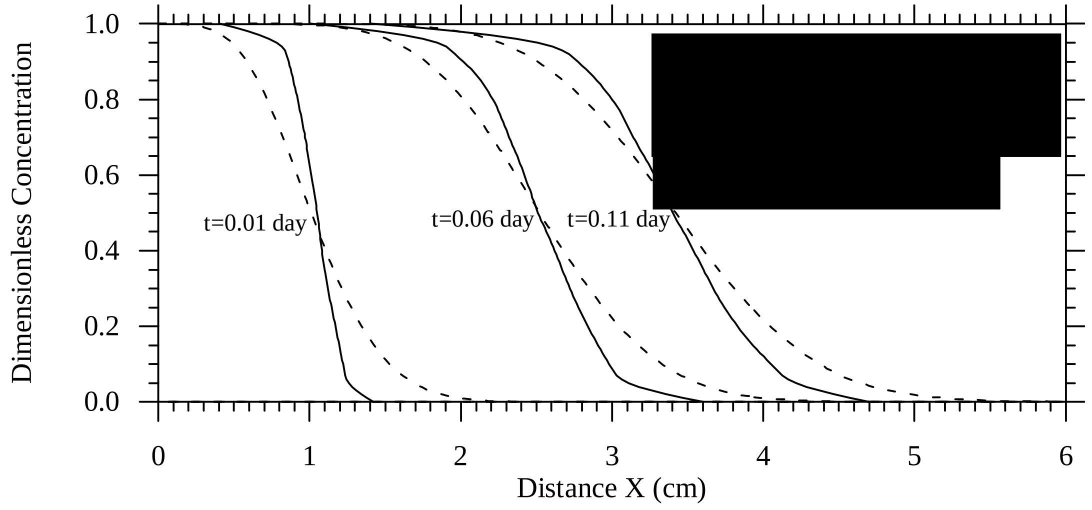

Figure 4.4. Comparison between dimensionless solute concentrations versus distance at various times generated by semi-analytic solution and RUNTOX code.

Table 4.1. Values of parameters used in simulations.

Item	Value
Diffusive porosity	0.45
Hydraulic conductivity, cm/day	1.0
Molecular diffusivity, cm^2/day	1.0
Tortuosity	1.0

Comparison between RUNTOX solution and semi-analytic solution at various time moments are shown in Figure 4.4 as dash and solid lines, respectively. Solute concentration obtained by RUNTOX code is converted into a dimensionless concentration by scaling it between the values at each boundary using a formulae $c=(c-c^n)/(c^0-c^n)$.

5. Comparison with experimental data

5.1. Application of RUNTOX model to the Log Plosky catchment

Data obtained from UHMI and OSU for rain-induced sediment yield events in 1970 and 1980 at agricultural catchment, Log Plosky (0.085 sq.km), located in the Butenya river basin were used to test the RUNTOX model. Length of the catchment from its most remote part up to the closing outlet is equal 0.54 km. Average width of the catchment is 0.19 km. Average slope of the catchment equals 24.7%, average slope of thalweg is 22.6%. Land surface of the Plosky broad gully is quite flat and has prolated from the south on north shape. The catchment boundaries are feebly marked so errors in determination of the catchment area are possible. The catchment has dark-grey loam soils which are planted with wheat, corn and vegetable in rotation and tilled in spring.

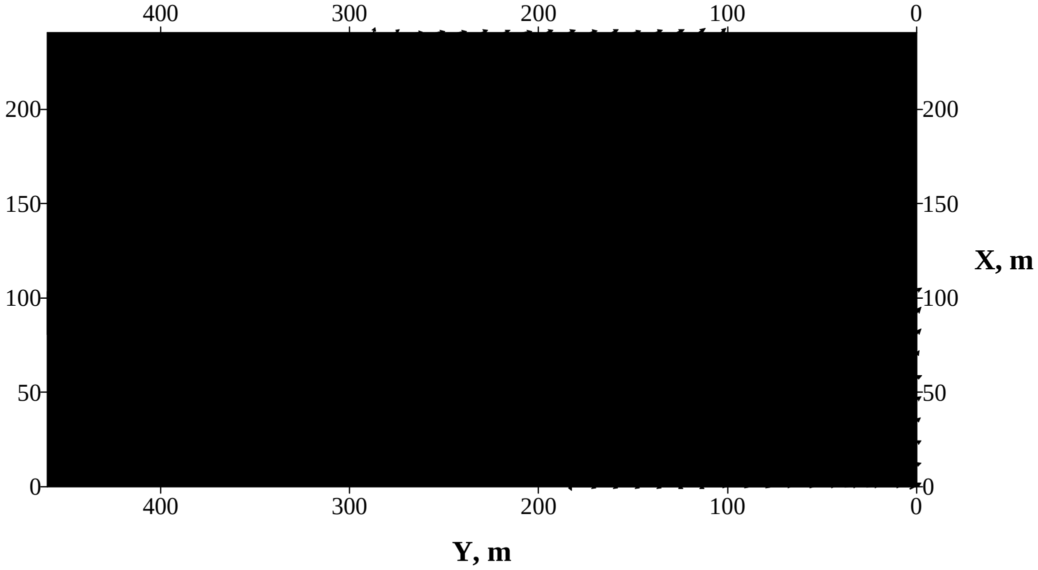

Figure 5.1. Velocity field of the overland flow after 30 min. rainfall begin for the 13 June 1970 event at the Log Plosky catchment.

Catchment was simulated using 11.5 m ? 11.5 m model grid squares and a computational time step of 1.0 min. The Morel-Seytoux model was used to calculate infiltration rate. Depression storage losses and interception were computed from the Befani models. The transport capacity of the flow were determined by using the Engelund-Hansen total load equation for the 13 June 1970 event and the Engelund-Hansen total load equation and the Yalin bedload equation for the 16 August 1980 event.

For graphical comparison, the computed hydrographs and temporal variation of sediment concentrations for these storm events were plotted along with the corresponding observed hydrographs and sediment concentrations in Figure 5.2-Figure 5.3. Field measurements showed that the fraction of sediment particles with a particle diameter from 0.01 mm up to 0.05 mm was prevailed (more than 70%). In numerical simulations, the particle diameter of the suspended sediment particles was assigned to 0.01 mm if the transport capacity was computed by using the Yalin bedload equation and 0.07 mm if the Engelund-Hansen equation was used to calculate the transport capacity of the overland flow.

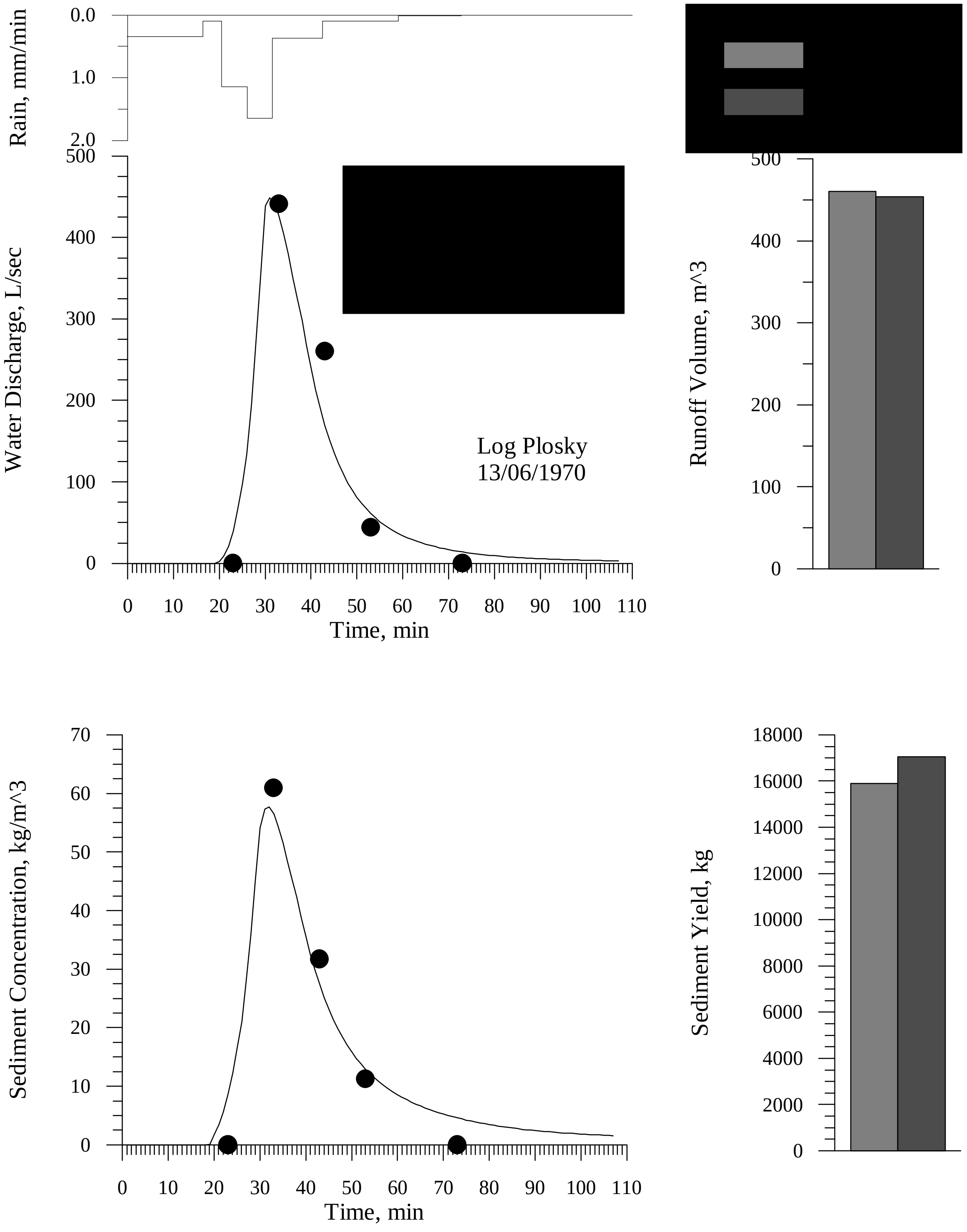

Figure 5.2. Comparison of measured and simulated water and sediment discharges for the 13 June 1970 event at the Log Plosky catchment.

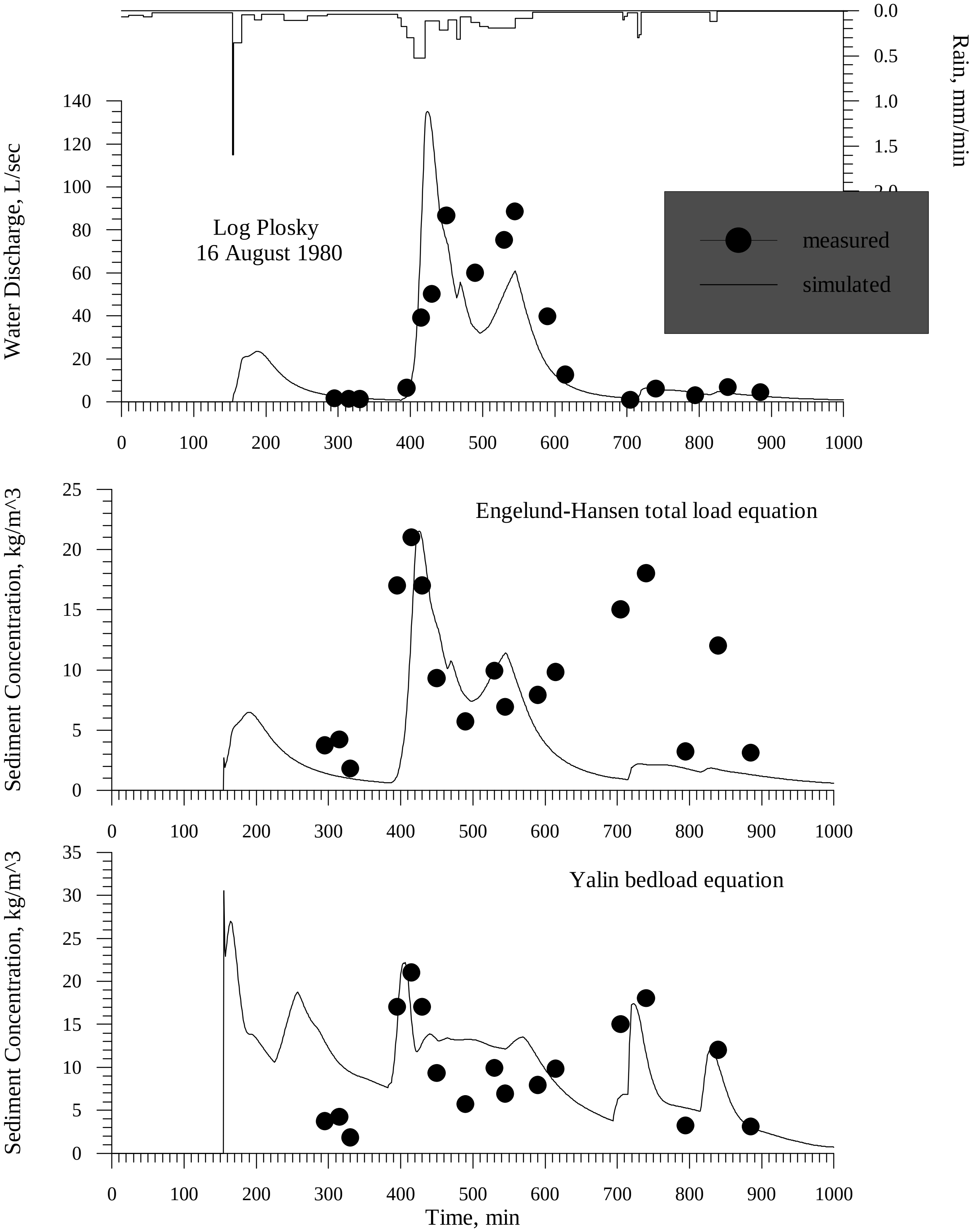

Figure 5.3. Comparison of measured and simulated water and sediment discharges for the 16 August 1980 event at the Log Plosky catchment.

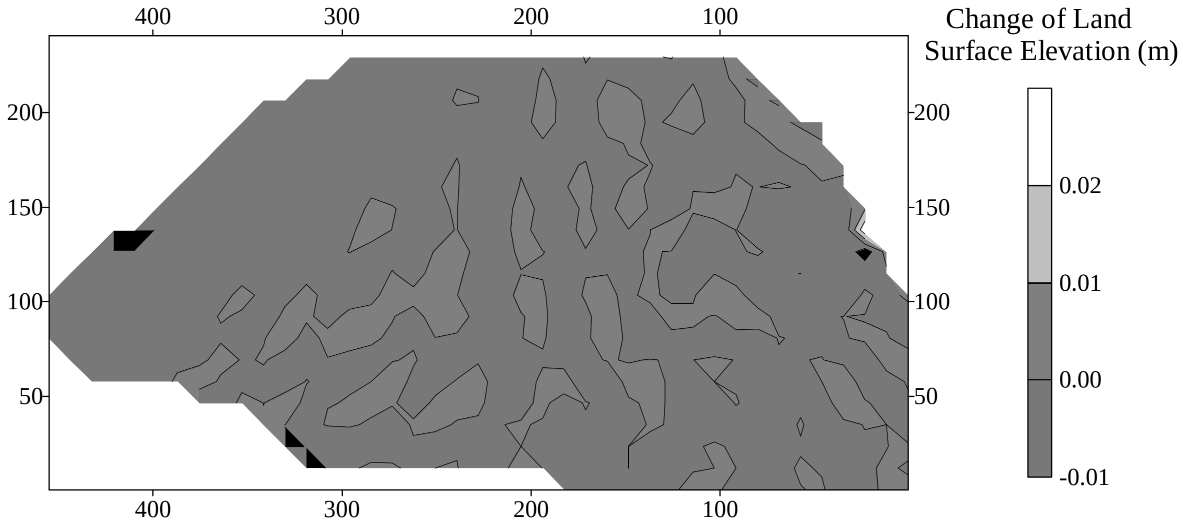

Figure 5.4. Change of land surface elevation after the 13 June 1970 event at the Log Plosky catchment.

Figure 5.2 shows good agreement between the simulated and observed hydrographs and sediment concentration for the 13 June 1970 event. Corresponding velocity field of the overland flow and division between the sedimentation and erosion areas are given in Figure 5.1-Figure 5.4, respectively. Light to dark gray indicate sedimentation, while diagonal cross stands erosion.

Figure 5.3 shows not such good agreement between simulated and observed hydrographs and sediment concentration for 16 August 1980 event as in previous case. Note that sediment concentration is strongly dependent on transport capacity equation, especially in the domain of small velocity of overland flow.

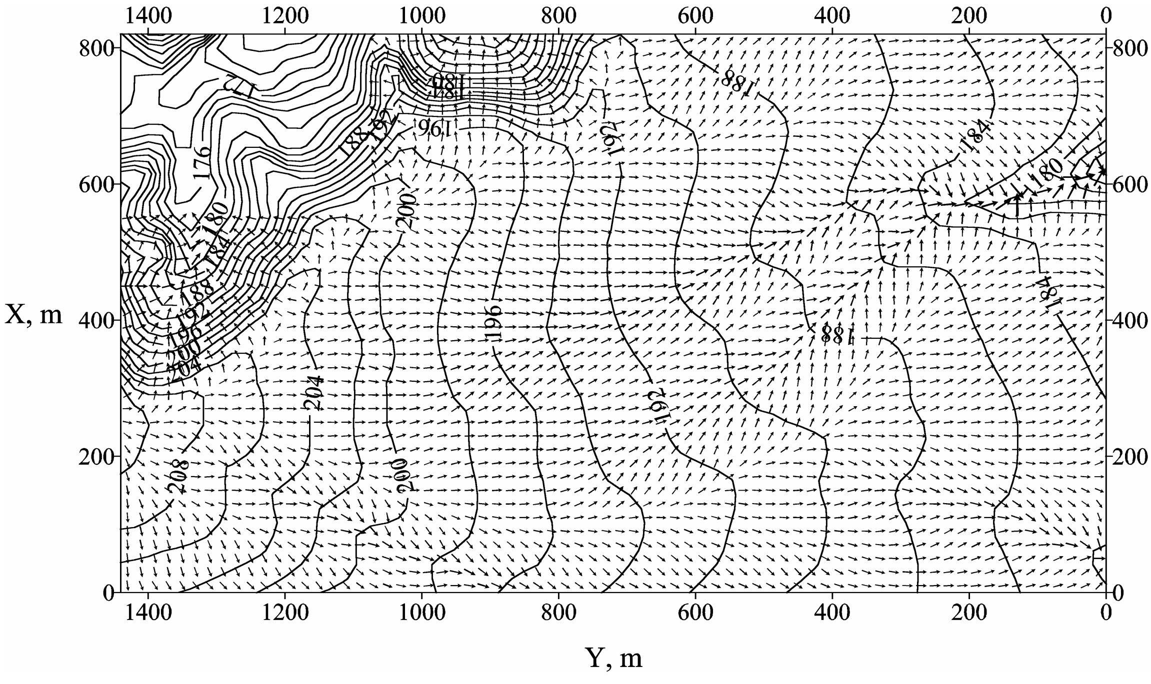

Figure 5.5. Elevation map and velocity field of overland flow for the 28 August 1993 event at the Log Pridorozhny catchment.

5.2. Application of RUNTOX model to the Log Pridorozhny catchment

To test ability of the RUNTOX model to simulate radionuclide redistribution within catchment, data obtained from UHMI and OSU for rain event in 1993 at Log Pridorozhny catchment was used. The Log Pridorozhny is agricultural catchment located in upper right-bank of the Butenya river basin. Area of the catchment equals to 0.40 sq.km. Length of the catchment from its most remote up point to the closing outlet is equal 1.21 km. Average width of the catchment is 0.33 km. Average slope of the catchment equals 22.7%, average slope of thalweg is 19.0%. Land surface of the Pridorozhny catchment is flat and has prolated from the north on south shape (see Figure 5.5). The catchment boundaries are feebly marked. Left and right boundaries of the catchment are small earth banks along field and near road shelterbelt forests. The thalweg is sharp marked in lower part of the catchment and passes into ravine.

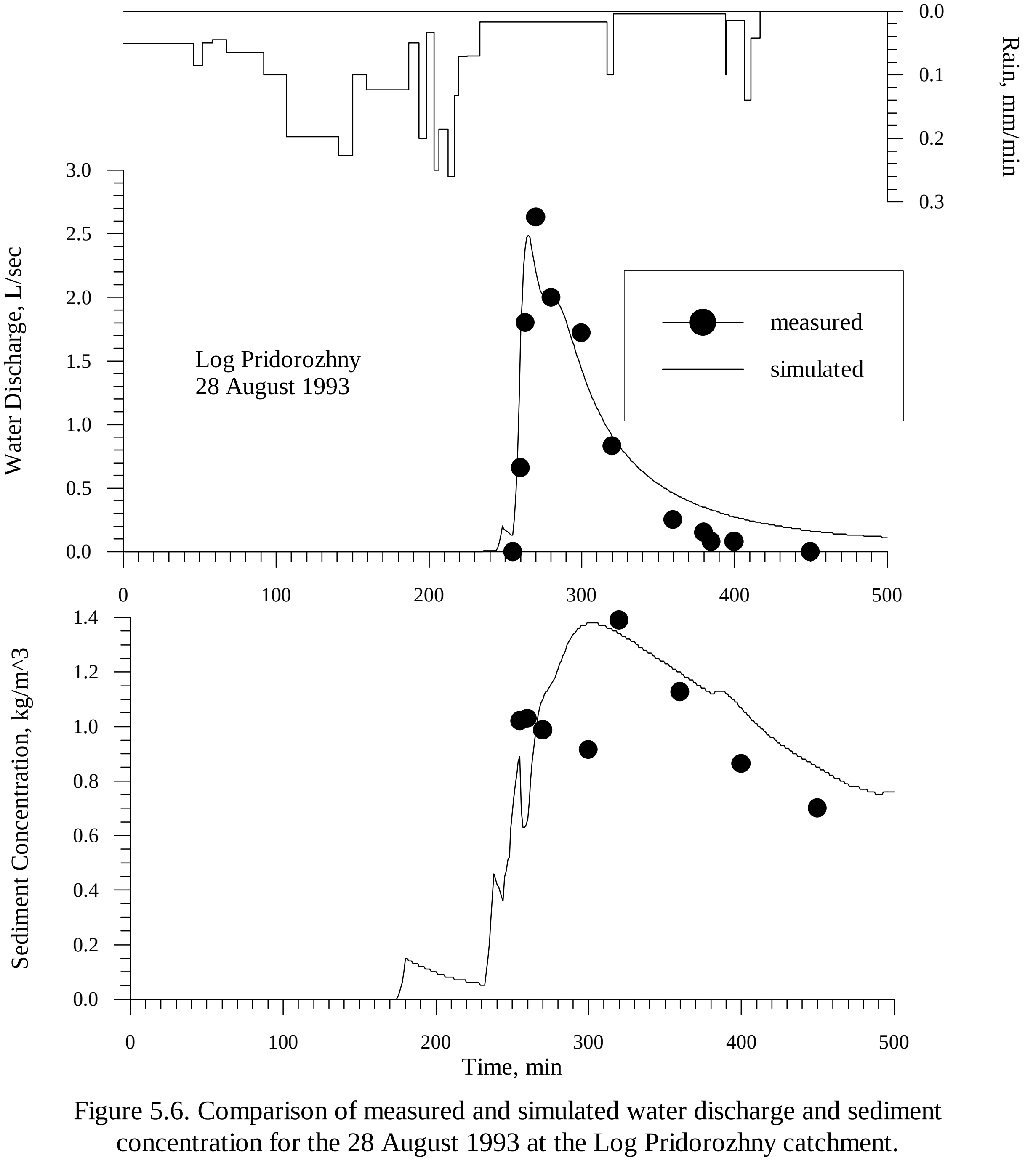

Figure 5.6. Comparison of measured and simulated water discharge and sediment concentration for the 28 August 1993 at the Log Pridorozhny catchment.

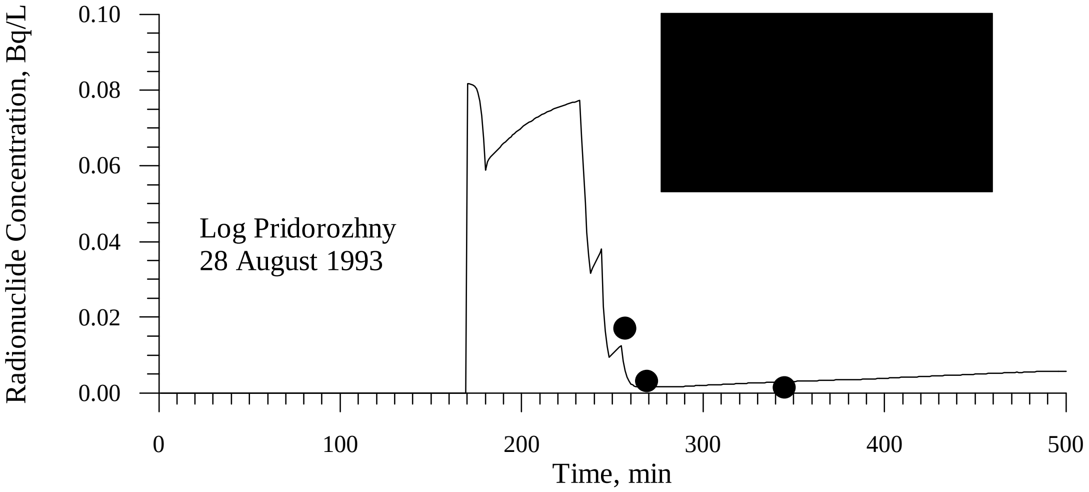

Figure 5.7. Comparison of measured and simulated radionuclide concentrations in solute for the 28 August 1993 event at the Log Pridorozhny catchment.

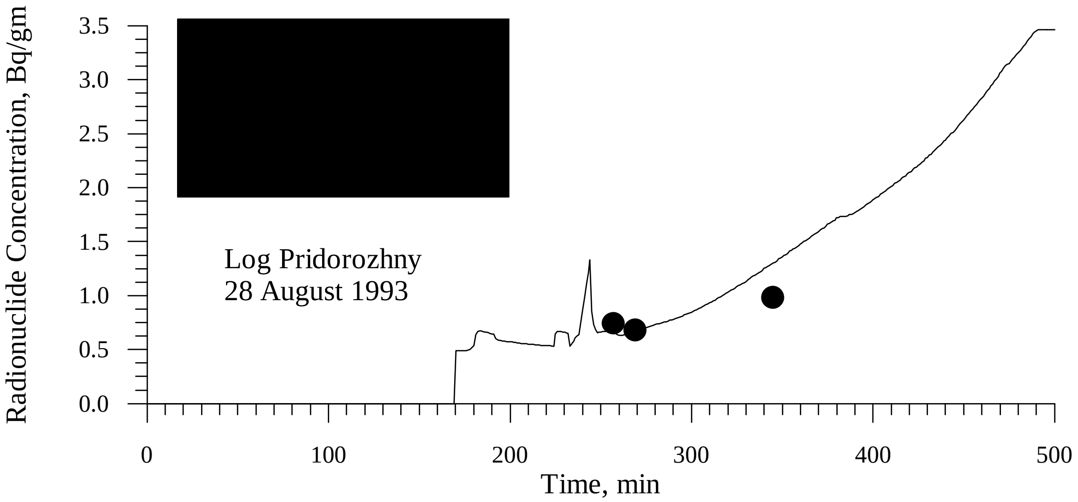

Figure 5.8. Comparison of measured and simulated radionuclide concentrations on suspended sediments for the 28 August 1993 event at the Log Pridorozhny catchment.

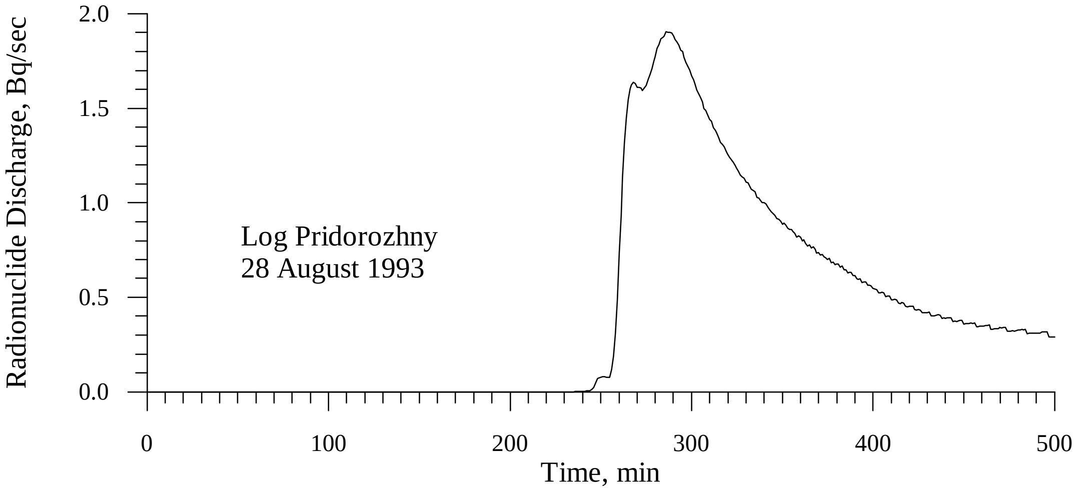

Figure 5.9. Discharge of ^{137}Cs on suspended sediments for the 28 August 1993 event at the Log Pridorozhny catchment.

The catchment has leached and podzolised chernozem soils which are planted with wheat, corn and vegetable in rotation and tilled in spring. Depth of the groundwater table is about 20-40 m below the land surface.

Catchment was simulated using 20 m ? 20 m model grid squares and a computational time step of 1.0 min. The Morel-Seytoux model was used to calculate infiltration rate. Depression storage losses and interception were computed from the Befani models. The transport capacity of the flow was determined by using the Engelund-Hansen total load equation. Depth of upper soil layer is assigned to 2 cm.

Comparison between computed and observed water discharges and sediment concentrations is shown in Figure 5.6.

It has been assumed that the catchment was uniformly contaminated by radionuclides. Land surface contamination by ^{137}Cs was up to 1.7 Ci per sq. km. Activity of ^{137}Cs in the exchangeable plus soluble phases was equal to 5% of total activity of ^{137}Cs. Figure 5.7-Figure 5.8 show agreement between computed and measured concentrations of ^{137}Cs in solute and on suspended sediments. At the runoff beginning the thin film of water is formed on land surface. Due to exchange processes between this thin water film and upper soil layer radionuclide concentration in this water film become high. Concentration of radionuclide in the water film is compatible with radionuclide concentration in soil water. But effect of these first minutes of runoff beginning on radionuclide discharge from catchment is negligible.

Discharge of ^{137}Cs on suspended sediments is given in Figure 5.9. Total discharge of ^{137}Cs (soluble phase plus on suspended sediments) is practically coincident to radionuclide discharge on suspended sediments. Therefore, the aqueous phase of ^{137}Cs can be neglected for modeling radionuclide wash-off from catchment.

Discharge of ^{137}Cs in subsurface water at the 50-cm depth from the Log Pridorozhny catchment for the 28 August 1993 event is shown in Figure 5.10.

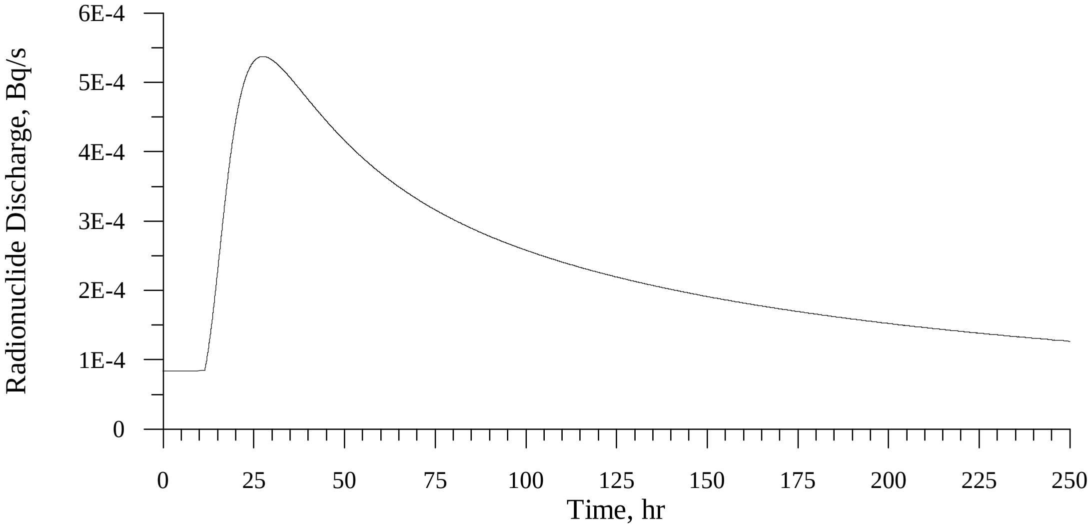

Figure 5.10. Discharge of ^{137}Cs at the 50-cm depth below land surface for the 28 August 1993 event at the Log Pridorozhny catchment.

6. Sensitivity analysis

Consider the effect of model parameter variations on the simulation results of runoff discharge, erosion/deposition processes, and radionuclide redistribution within catchment.

6.1. Effect of roughness coefficient

To evaluate sensitivity of water discharge and runoff volume to the Manning roughness coefficient, numerical simulations were conducted with various values of the roughness coefficient for 13 June 1970 event at the Log Plosky catchment.

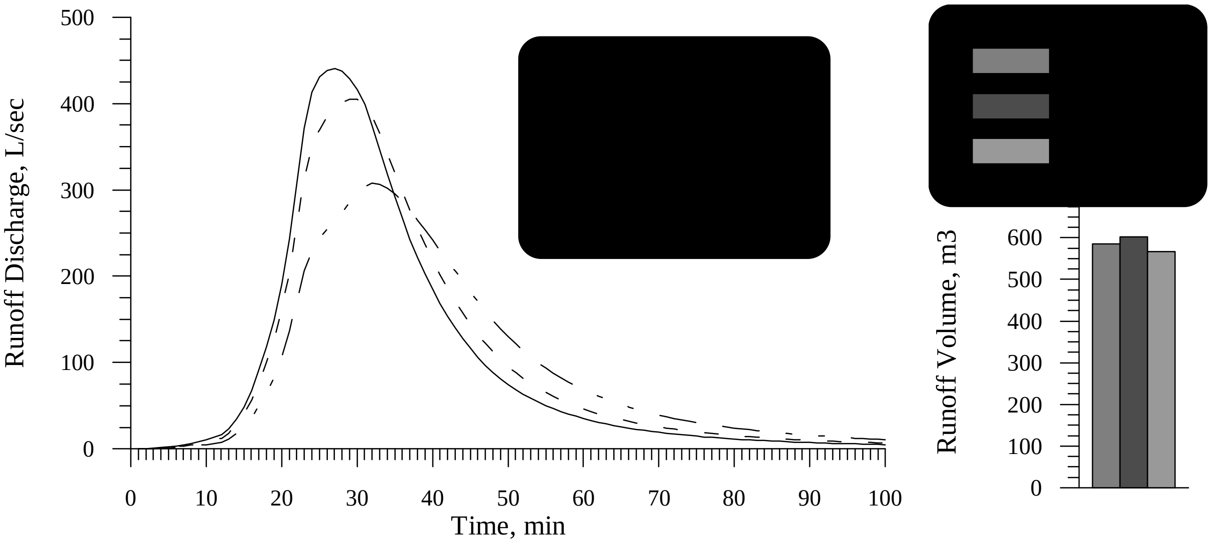

Figure 6.1. Effect of the Manning roughness coefficient on water discharge and runoff volume.

Figure 6.1 shows increase of the peak time and increase of damping of hydrograph peak magnitude with increase of the Manning roughness coefficient. Variation of the roughness coefficient does not have a large effect on the runoff volume.

6.2. The sensitivity to diameter of sediment particle and transport capacity equation

Effect of particle size of suspended sediments on sediment concentration and sediment yield are shown in Figure 6.2-Figure 6.3. Numerical simulations were conducted with various values of the sediment size by using the Engelund-Hansen total load equation and Yalin bedload equation for 13 June 1970 event at the Log Plosky catchment.

Figure 6.2-Figure 6.3 illustrate that simulation results of catchment erosion processes strongly depend on which load equation used for calculating sediment transport capacity. The Yalin bedload equation and Engelund-Hansen total load equation produce different results. In first case, a sediment yield and sediment concentration increase as sediment particle size increases. For the Engelund-Hansen equation, a sediment yield and sediment concentration decrease with sediment size increases.

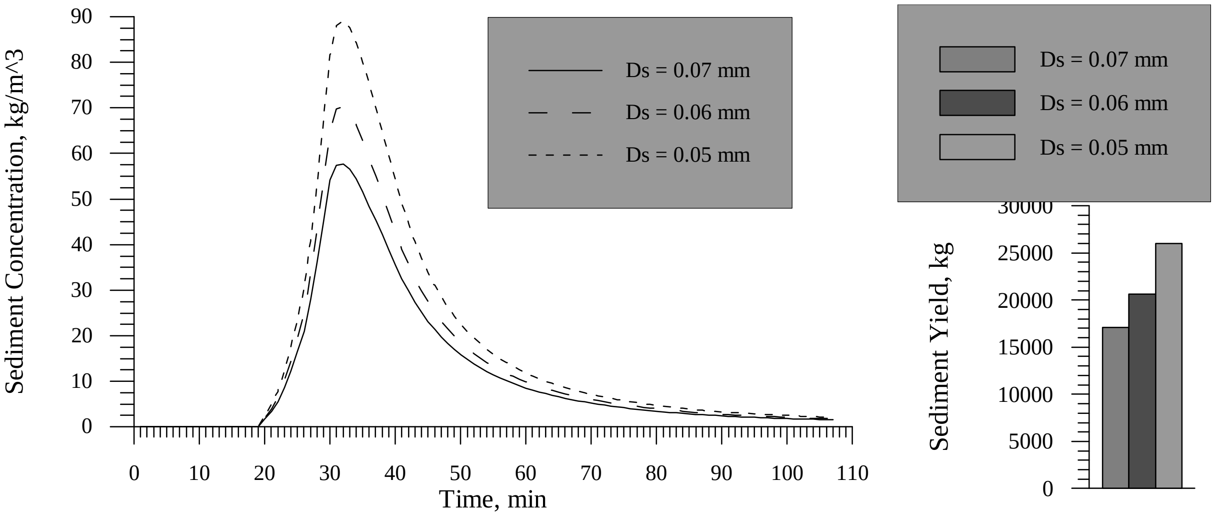

Figure 6.2. Effect of particle diameter on sediment concentration and sediment yield calculated by using the Engelund-Hansen total load equation.

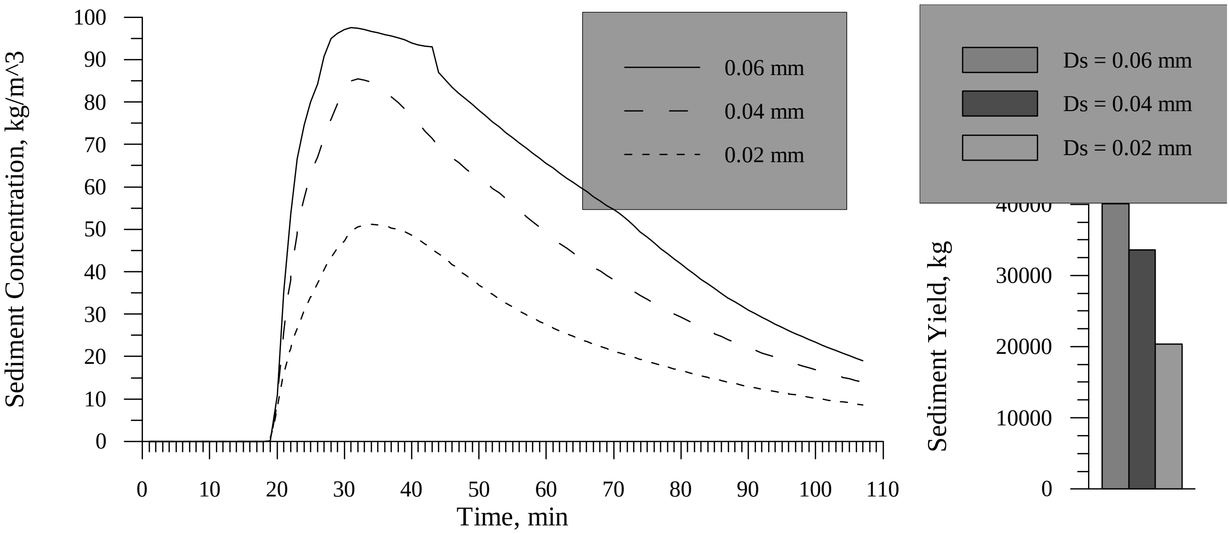

Figure 6.3. Effect of particle diameter on sediment concentration and sediment yield calculated by using the Yalin bedload equation.

6.3. Effect of the erodibility coefficient

Dependence of suspended sediment concentration and sediment yield on the erodibility coefficient is given in Figure 6.4. Numerical simulations were conducted with various values of the erodibility coefficient for 13 June 1970 event at the Log Plosky catchment. Transport capacity of the overland flow has been determined by using the Yalin bedload equation for sediment size equals to 0.02 mm.

Figure 6.4 shows decrease of magnitude of peak sediment concentration and sediment yield with decrease of the erodibility coefficient.

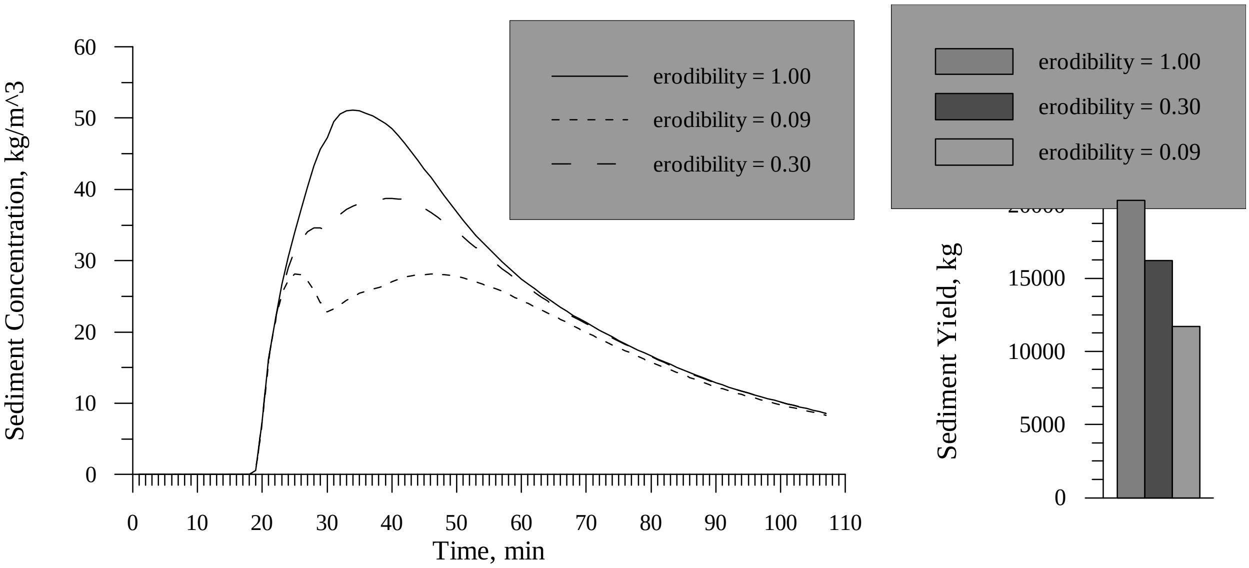

Figure 6.4. Effect of the erodibility coefficient on sediment concentration and sediment yield.

6.4. Effect of partition coefficient in system “water-upper soil layer”

Sensitivity of ^{137}Cs activity in overland water and on suspended sediments to a variation of the partition coefficient is presented in Figure 6.5-Figure 6.6. Simulations were conducted for the 28 August 1993 event at the Log Pridorozhny catchment.

Figure 6.5-Figure 6.6 indicate decrease of ^{137}Cs activity in overland water and on suspended sediments with increase of the partition coefficient for the system “water – upper soil layer”.

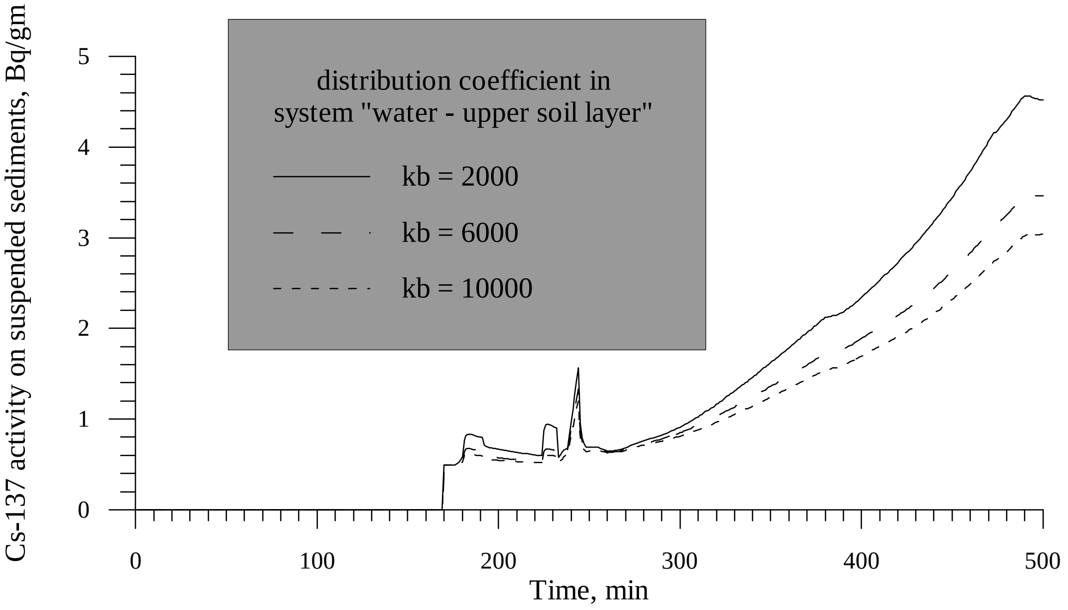

Figure 6.5. Effect of partition coefficient in the system “water-upper soil layer” on ^{137}Cs activity on suspended sediments.

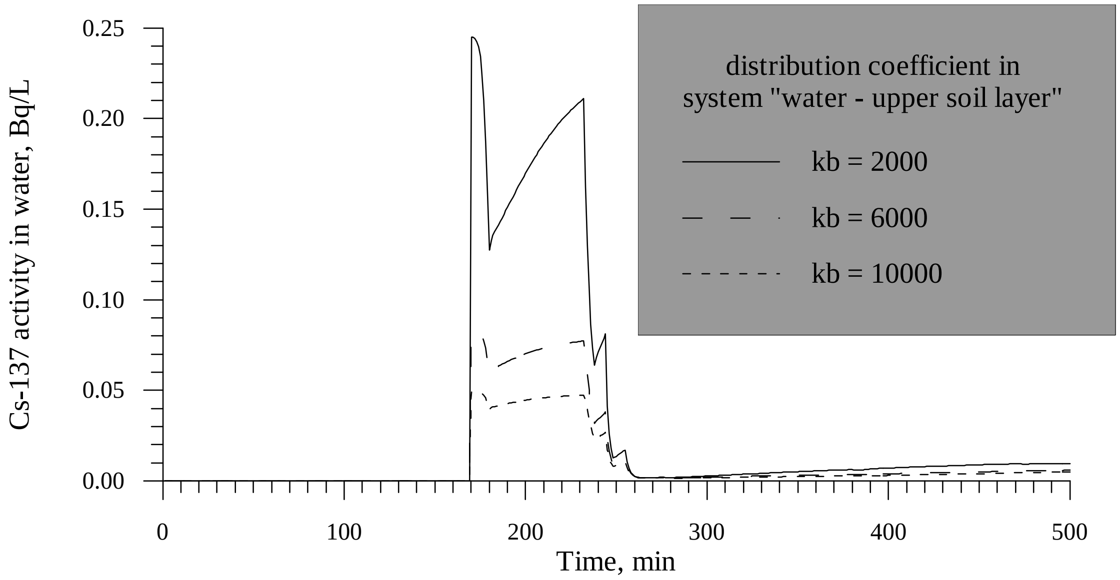

Figure 6.6. Effect of partition coefficient in the system “water-upper soil layer” on ^{137}Cs activity in overland water.

7. Conclusions

? Comparison of numerical results with measured data demonstrates the ability of the RUNTOX model to simulate transport of Cs at the catchment scale, including transport of radionuclides in soluble phase and radionuclides adsorbed to sediment particles.

? Discharge of activity of ^{137}Cs in soluble phase is negligible in comparison with activity of ^{137}Cs on suspended sediments. Thus, adequate modeling of erosion/deposition processes is very important for prediction of ^{137}Cs wash-off from catchment.

? Erosion/deposition models, describing hydraulic erosion/deposition as a kinetic transfer process, are strongly dependent on transport capacity relationship has being used.

? Secondary ^{137}Cs contamination of surface water throughout wash-off from catchment strongly depends on sediment discharge from this catchment. Therefore, reduction of secondary radionuclide contamination of surface water can be accomplished by soil-conservation practices such as strip cropping and no-till planting in agricultural zones, erosion control of construction sites, and by sedimentation ponds in urban areas.

? Sensitivity analysis shows that use of chemical fertilizers increasing soil properties related to transfer ^{137}Cs into fixed phase allows prevent secondary contamination of groundwater. Limitation and optimization of irrigation on arid lands are also very important to reduce contamination of groundwater by radionuclides.

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