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MATHEMATICAL MODELING OF SOLUTE RUNOFF FROM FORESTED BASIN

BY

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SYNOPSIS

A mathematical model for solute runoff from forested basins is developed, based on a tank model comprising a surface tank with soil water storage systems and a groundwater tank. The model takes two elementary processes into account, (a) solute advection due to the rainwater movement and (b) solute exchange between soil aggregate and adjacent soil water. The proposed model is applied to data of streamwater solute concentrations (Cl^- and SO_4^{2-}) obtained from two forested experimental mountain basins (main species *Cryptomeria japonica*). The basic model structure is verified by the observations of streamwater solute concentrations.

INTRODUCTION

It is important to evaluate water and solute mass balance in forested basins for investigating the migration and final destination of nitrogen and phosphorus which cause eutrofication in water environment. The formation process of streamwater chemistry and solute behaviour have been modeled, using a mixture of soil water end-members as called "end member mixing technique" (2,9). Several researchers (1,8) have pointed out that both the vertical profile of solutes in the soil and relative volumetric contributions of runoff components through different water pathways are predominant in the formation process of streamwater chemistry. Much knowledge about hydrochemistry in forested basins has been reported, however, it is not so useful for the quantitative evaluation of solute behaviour and its mass balance in a basin. This is because the rainwater runoff is made up of several elements in hydrological processes, e.g. rainfall interception, transpiration, infiltration, percolation and runoff in a forested basin known as a 'Soil-Plant-Atmosphere Continuum (SPAC)'. It is so complicated that the methodology for expressing the rainwater runoff process have not been established yet. Because rainwater movement may cause solute migration in the soil, it is a key point that elementary processes in hydrological circulation and their interactions in the SPAC should be considered in the modeling of solute runoff process.

We think that the mathematical modeling is an appropriate way to quantify element circulation in forested basins, based on an understanding of the coupling action of water and solutes in hydrological and hydrochemical processes. We have already examined the relationship between temporal variations of rainwater runoff components and streamwater solute concentration, and have confirmed the necessity of the coupling methodology of water and solute circulation (10). In this study, a mathematical model for short-term solute runoff from forested basins is developed for water quality parameters, Cl^- and SO_4^{2-}. They are not biochemically reactive and have high mobility in the soil. The proposed model is applied to the observations of solute runoff obtained from two forested mountain basins in Japan. Estimated temporal variations of streamwater solute concentrations for Cl^- and SO_4^{2-} are consistent with field observations. Based on these results, solute migration in forested basins is discussed.

METHODOLOGY

Modeling of Different Water Pathways (Rainwater runoff module)

A key point for modeling of solute runoff in a forested basin is to describe solute percolation and identify the runoff component which contributes greatly to solute transport in the soil (10). The basic concept of this study is to present a precise description of rainwater migration and solute dynamics. Therefore, it is necessary to separate runoff components, which play their own roles in different layers in the soil, as precisely as possible prior to solute runoff modeling. In order to express rainwater migration in the soil, Hashino et al.(3) developed a rainwater tank model, taking both rainfall interception (11) and transpiration (12) into account. The rainwater tank model consists of a surface tank and a groundwater tank for describing several soil layers which have different rates of infiltration and percolation, and soil water retention capacity as illustrated in Fig.1. The surface tank is composed of an upper tank corresponding to a soil layer called 'A horizon' which has high porosity and hydraulic conductivity, and a lower tank corresponding to a soil layer called 'B horizon' which contributes to soil water retention due to less porosity. Furthermore, the lower tank consists of a primary soil tank where soil water moves rapidly and a secondary soil tank where soil water moves slowly. Between the primary and secondary soil tanks, soil water exchange, L_T is generated by the difference of degree of saturation (S_x/h_x-S_y/h_y). The rates of runoff, infiltration and percolation q_{o1}, q_{o2}, q_s, q_h, q_i, q_j, q_k, q_l, q_g, f_b and f_g are given by the rainwater tank model prior to computation of solute runoff. A detailed description of the rainwater tank model is provided by Hashino et al.(3).

Modeling of Hydrochemical Processes on Solute Migration (Solute runoff module)

he objective of this study is to model solute runoff process during a storm event. Biochemical processes such as mineralization caused by bacteria are not considered, because they may have a less effect on solute migration in the short term. The solute tank model proposed here has two modules for hydrochemical processes in the soil as shown in Fig.2. Substances contained in the soil zone are composed of two types having different responses to rainwater migration. One is the mobile solute which moves freely corresponding to rainwater movement, and the other is the immobile solute adsorbed in soil aggregate, which results in less response to rainwater movement. Both substances are assumed to maintain concentration equilibrium based on solute exchange between soil aggregate as an immobile region and adjacent soil water as a mobile region.

The fundamental process is solute advection generated by throughflow and percolation, which takes place with relation to rainwater movement. The rates of solute advection are described as the product of rainwater advection rates and their concentrations. For example, the rate of solute percolation C_xq_j from the primary soil tank to the groundwater tank is expressed as the product of rate

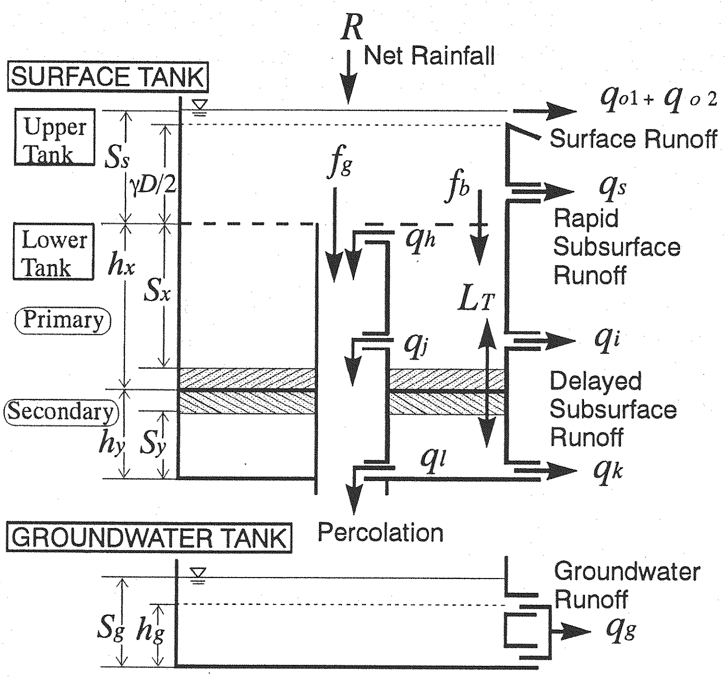


Fig. 1 Schematic illustration of rainwater tank model

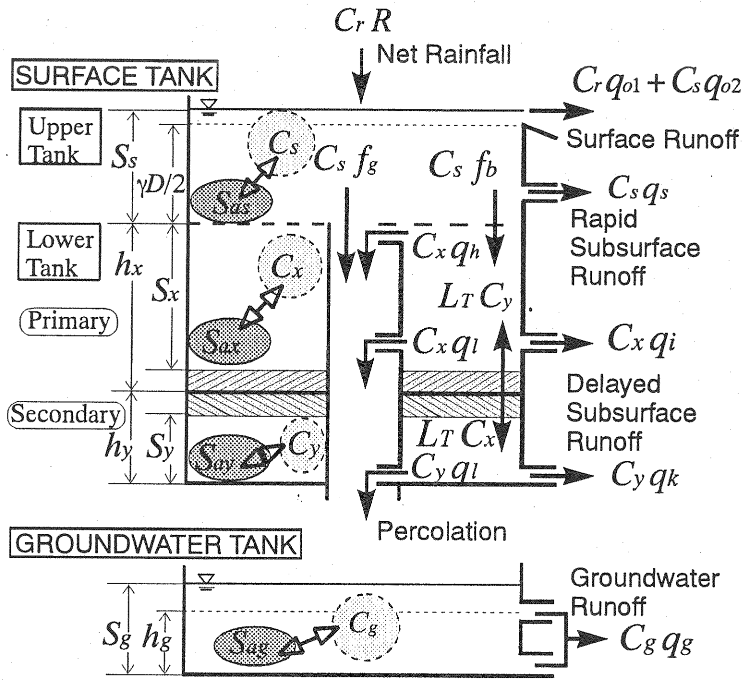


Fig. 2 Schematic illustration of solute tank model

of rainwater percolation q_j and concentration of the primary soil tank C_x as indicated in Fig.2. The rate of solute exchange between mobile and immobile regions is assumed to be proportional to both the difference of concentrations between mobile and immobile solutes and soil water storage within the pores referring to the model proposed by Lapidus et al.(6). For example, that of the primary soil tank $v_x(k_xS_{ax}-C_x)S_x$ is described as the product of the difference of solute concentration $(k_xS_{ax}-C_x)$ and soil water storage S_x. Immobile solute concentration is described using adsorbed solute S_{ax} and a distribution coefficient k_x.

Solute Balance Equations

Solute balance equations for each tank, illustrated in Fig.2, are shown below. They are provided for mobile and immobile solutes in each tank. All equations for mobile region, solute balance is described by advection (throughflow, infiltration and percolation) and solute exchange term between mobile and immobile regions. Those for immobile region, they include only solute exchange term.

Upper tank :

Mobile solute : $$\frac{d(C_sS_s)}{dt}=C_rR-C_rq_{o1}-C_s(q_{o2}+q_s+\mu_bf_b+\mu_gf_g)+\nu_s(k_sS_{as}-C_s)S_s \quad (1)$$

Immobile solute : $$\frac{d[S_{as}(1-\gamma)D]}{dt}=-\nu_s(k_sS_{as}-C_s)S_s \quad (2)$$

Primary soil tank :

Mobile solute : $$\frac{d(C_xS_x)}{dt}=\mu_bC_sf_b-C_x(q_h+q_i+q_j)\mp C_{xy}L_T+\nu_x(k_xS_{ax}-C_x)S_x \quad (3)$$

Immobile solute : $$\frac{d[S_{ax}h_x(1-f_x)/f_x]}{dt}=-\nu_x(k_xS_{ax}-C_x)S_x \quad (4)$$

Secondary soil tank :

Mobile solute : $$\frac{d(C_yS_y)}{dt}=\pm C_{xy}L_T-C_y(q_k+q_l)+\nu_y(k_yS_{ay}-C_y)S_y \quad (5)$$

Immobile solute : $$\frac{d[S_{ay}h_y(1-f_y)/f_y]}{dt}=-\nu_y(k_yS_{ay}-C_y)S_y \quad (6)$$

Groundwater tank :

Mobile solute : $$\frac{d(C_gS_g)}{dt}=\mu_gC_sf_g-C_x(q_h+q_j)+C_yq_l-C_gq_g+\nu_g(k_gS_{ag}-C_g)S_g \quad (7)$$

Immobile solute : $$\frac{d[S_{ag}h_g]}{dt}=-\nu_g(k_gS_{ag}-C_g)S_g \quad (8)$$

where t = time (hr); R = rainfall intensity (mmhr^{-1}); C_r = throughfall concentration (mgmm^{-1}); C_s, C_x, C_y, C_g = mobile solute concentrations (mgmm^{-1}); S_{as}, S_{ax}, S_{ay}, S_{ag} = immobile solute concentrations (mgmm^{-}

^{1}); q_{o1}, q_{o2} = rates of surface runoff (mmhr^{-1}); q_s = rate of rapid subsurface runoff (mmhr^{-1}); q_i = rate of delayed subsurface runoff from the primary soil tank (mmhr^{-1}); q_k = rate of delayed subsurface runoff from the secondary soil tank (mmhr^{-1}); q_g = rate of groundwater runoff (mmhr^{-1}); q_h, q_j = rates of percolation from the primary soil tank (mmhr^{-1}); q_l = rate of percolation from the secondary soil tank (mmhr^{-1}); f_b = rate of infiltration into the surface soil tank (mmhr^{-1}); f_g = rate of direct percolation from the upper tank to the groundwater tank (mmhr^{-1}); L_T = rate of soil water exchange between the primary and secondary soil tanks (mmhr^{-1}); S_s, S_x, S_y, S_g = soil water storage (mm); γ, f_x, f_y = porosities; D = saturated soil water capacity of the upper tank (mm); h_x, h_y = saturated soil water capacities of the primary and secondary soil tanks (mm); ν_s, ν_x, ν_y, ν_g = solute exchange coefficients (hr^{-1}); k_s, k_x, k_y, k_g = solute distribution coefficients between mobile and immobile regions; μ_b, μ_g = correction factors for infiltration and percolation fluxes to promote downward movement of solutes and realize high concentration of delayed subsurface and groundwater runoffs which raises streamwater concentration in the recession period of the storm. When L_T takes place from the primary to secondary soil tank, $C_{xy} = -C_x$ in Eq.3 and $C_{xy} = +C_x$ in Eq.5. When the direction of L_T is opposite, $C_{xy} = +C_y$ in Eq.3 and $C_{xy} = -C_y$ in Eq.5. The unit of solute concentration is converted into mgmm^{-1} corresponding to water depth. The subscripts, s, x, y and g in the solute balance equations denote the substance in the upper, primary soil, secondary soil and groundwater tanks, respectively. For taking account of contact time between rainwater and soil aggregate, especially during periods when surface runoff takes place, the rate of surface runoff q_o is divided into two components, q_{o1} and q_{o2} in Eq.1. Component q_{o1} is mainly composed of rainwater. The other component q_{o2} is so called rate of return flow, which originally takes place in the 'A horizon'.

Procedure of Computation and Parameter Identification

A module of rainwater runoff and its model parameters was developed (3). Thus, the rates of throughflow, infiltration and percolation are given by the rainwater runoff module prior to computation of solute runoff. At first, the atmospheric inputs, that is, the sum of solute loads of throughfall and stemflow defined as the product of their runoff rates and solute concentrations are used in the proposed solute tank model. Though they vary temporally, for simplicity, the average concentration of throughfall making up more than seventy five percent during a storm (11) is adopted as representative of the atmospheric input. Computing water and solute balance in each tank by using the rainwater runoff module and Eqs.1-8, mobile and immobile solute concentrations are determined at 1 minute time intervals. Each runoff component transports solutes from various kind of depth in the soil to the stream. Therefore, solute loads of all runoff components are given as the product of the rates of runoff and their concentrations. Finally the streamwater concentration C_{est} is evaluated using Eq.9.

$$C_{est}=\frac{C_r q_{o1}+C_s q_{o2}+C_s q_s+C_x q_i+C_y q_k+C_g q_g}{q_{o1}+q_{o2}+q_s+q_i+q_k+q_g} \qquad (9)$$

where C_{est} = the estimated streamwater concentration (mgl^{-1}); C_r = throughfall concentration (mgl^{-1}); C_s, C_x, C_y, C_g = solute concentrations in each tank (mgl^{-1}) corresponding to rapid and delayed subsurface runoffs and groundwater runoff, respectively; q_{o1}, q_{o2}, q_s, q_i, q_k, q_g = rate of each runoff (mmhr^{-1}).

All model parameters (k_s, k_x, k_y, k_g, ν_s, ν_x, ν_y, ν_g) are identified by the method of least squares. The evaluation function F is defined as the squared sum of the estimated error of streamwater concentration given by Eq.10.

$$F=\sum_{j=1}^{N}(C_{obs,j}-C_{est,j})^2 \qquad (10)$$

where $C_{obs,j}$ = observed streamwater concentration (mgl^{-1}); $C_{est,j}$ = estimated streamwater concentration (mgl^{-1}); N = number of sample data.

Initial values of mobile and immobile solutes, C_{xini}, C_{yini}, C_{gini}, S_{asini}, S_{axini}, S_{ayini}, S_{agini} are unknowns, which should be determined using concentration data of soilwater obtained from the basin before the storms.

OBSERVATIONS

Study sites are Shirakawatani experimental forested basin (33°52'N, 133°40'E) located about 100 km west of Tokushima City in Shikoku Island, Western Japan (Fig.3(a)) and Tsukuba experimental forested basin (36°12'N, 140°9'E) located about 70 km northeast of Tokyo. Shirakawatani basin has an area of 23 ha, altitudes range between 740-1100 m and has a mean slope angle of 21.5°. The predominant tree species is Japanese cedar (*Cryptomeria japonica*) which covers sixty percent of the area. The remainder is covered with deciduous broad-leaved forest. The site is underlain by psammite and clay schists with a surface of brown forest soil. The 'A horizon' is less than 10 cm and 'B horizon' is between 50-80 cm. Thus, the whole soil layer is mostly less than 1 m.

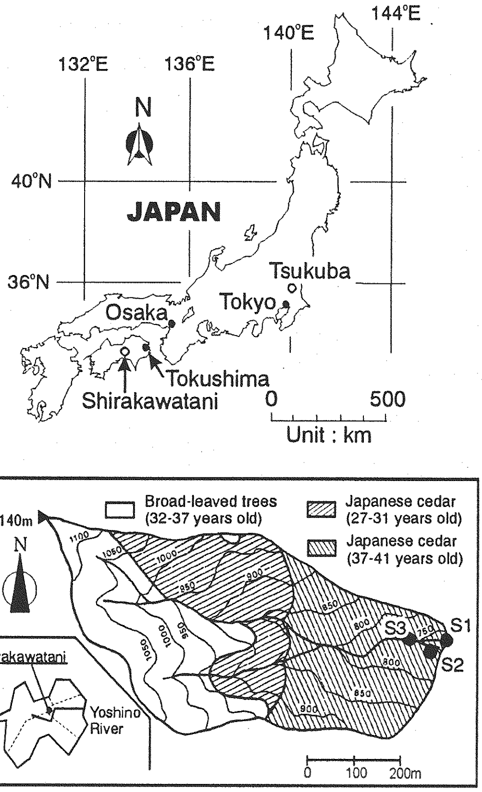
The streamwater level was measured by a float type level recorder at a 90° V-notch weir (S1 in Fig.3(a)), and was converted into streamwater discharge. Rainfall was measured with a tipping bucket rain gauge at S1. Throughfall was recorded with two tipping bucket rain gauges and a throughfall trough, which had an area of 7.2 m^2, under a 32 year old Japanese cedar (S2 in Fig.1). Stemflow was obtained from measurement of rainwater captured with a vinyl collar sealed to the stem of the Japanese cedar at breast height (1.3 m) by means of a tipping bucket rain gauge.

Meteorological parameters (air temperature, relative humidity, wind speed, net radiation and solar radiation) were measured about 2 m above the canopy using instruments mounted on a 22 m observation tower. Soil moisture potential (suction) was monitored by tensiometers at depths of 10, 20, 30, 40, 50, 60 and 70 cm adjacent to the Japanese cedar. All parameters except water level and rainfall were measured and recorded on data loggers with a time interval of 10 minutes. Data of duration of sunshine was obtained from Ikeda Dam Group Operation Office (Water Resources Development Public Corporation).

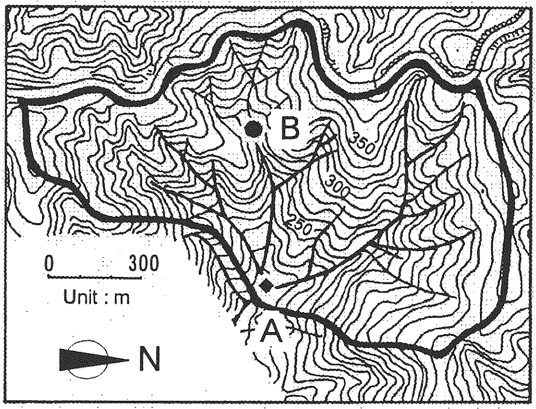
Streamwater was gathered about 100 m upstream from the weir (S3) at time intervals ranging from 15 minutes to 3 hours, depending on streamwater discharge. Rainfall, throughfall and stemflow were collected in polyethylene tanks at S1 and S2. At S2 in Fig.3(a), soil water was gathered at depths of 10, 20, 30, 40 and 50 cm. A polyethylene tube with an unglazed porous cup of 2 cm in diameter and 5 cm in length was used for collecting soil water. Soil water was extracted five times by a suction pump operated using a microtube connected to the porous cup in a storm event. Chemical analysis of water samples was made for NO_3^--N, SiO_2, Cl^-, SO_4^{2-}, K^+, Na^+, Ca^{2+}, pH, electric conductance (EC), and suspended solids (SS). Data was collected over the period from September 23 to September 26 in 1992, during which time a single rainfall event of 145 mm, occurred with a peak rainfall intensity of 38 $mmhr^{-1}$.

The other experimental basin is the Tsukuba experimental forested basin (Fig.3(b)) with an area of 67.5 ha (8). The altitude of the basin ranges from 200 m - 380 m. The bed rock of the basin consists of granite and gneiss, above which exists weathered granite. The forest surface soil is composed of volcanic loam. The basin is covered with Japanese cedar (*Cryptomeria japonica*), Japanese cypress (*Chamaecyparis obtusa*) and burned pine (*Pinus densiflora*), and some areas with broad-leaved trees. There are three main streams which converge at the exit of the basin.

Streamwater level was measured by a float type level recorder at A as shown in Fig.3(b) and turned into streamwater discharge. Rainfall was measured with a tipping bucket rain gauge near site A; and 10 minutes rainfall intensity was measured. Throughfall in the forest was gathered by pot type rain gauges at plots of Japanese cedar, Japanese cypress and broad-leaved stands. Streamwater and rainfall were collected at A. Meteorological parameters were provided by Kakioka Geomagnetism



(a) Shirakawatani Experimental Forested Basin



(b) Tsukuba Experimental Forested Basin

Fig. 3 Maps of study sites

Table 1 Storm events selected for analysis

Event	Date	Rainfall amount (mm)	Maximum rainfall intensity (mmhr^{-1})
Event T-1	Jun.13 - Jun.16, 1985	48.0	4.5
Event T-2	Jun.29 - Jul.3, 1985	136.5	16.5
Event T-3	Jun.25 - Jun.28, 1986	26.5	8.5
Event S-1	Sep.24 - Sep.26, 1992	145.0	38.0

Observation Station located 6.3 km northwest of the basin. Data was collected in three storm events in 1985 and 1986 listed in Table 1.

An itemized list of water quality parameters includes NO_3^--N, SiO_2, Cl^-, SO_4^{2-}, K^+, Na^+, Ca^{2+}, pH, electric conductance (EC), and suspended solid (SS).

RESULTS

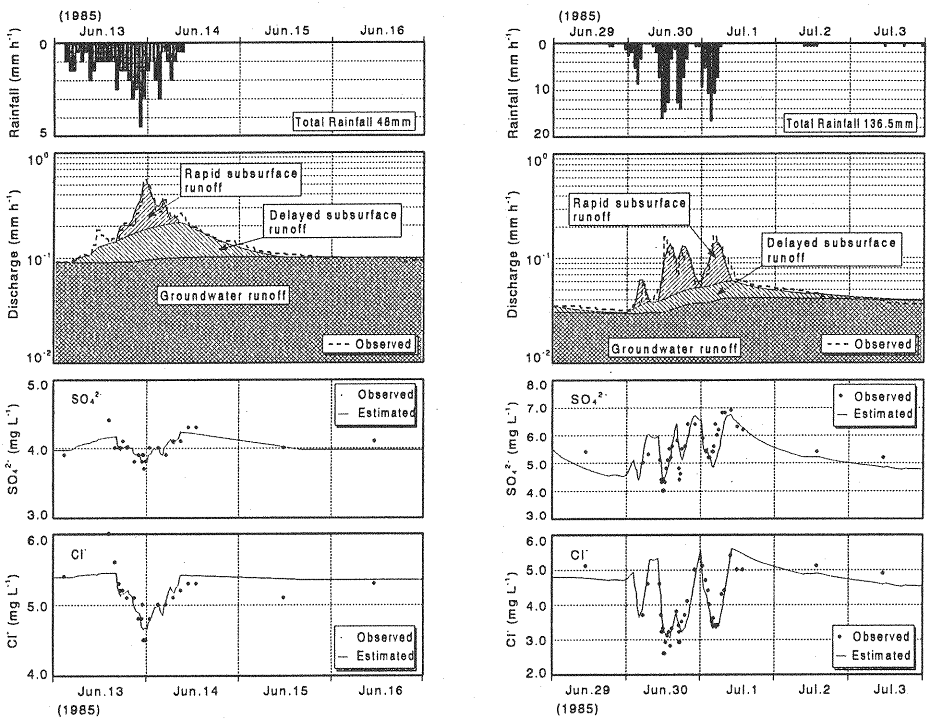
Two anions, Cl^- and SO_4^{2-} were selected as water quality parameters in both basins, because they are not biochemically reactive and are readily mobile in the soil. Thus, we can expect they are readily mobile in the soil. All of the parameters identified for each basin are listed in Table 2.

Tsukuba Experimental Forested Basin

Hyetograph, hydrograph and temporal variations of Cl^- and SO_4^{2-} concentrations in streamwater for three storm events are shown in Figs.4(a), 4(b) and 4(c). Unique values of model parameters were identified for three storm events. But the term of solute exchange between mobile and immobile regions had a minor effect on solute balance in the groundwater tank, and parameters ν_g and k_g were not necessary in all events. Rainfall intensity was not so high and rainfall amount was 48 mm in Event T-1. Temporal variation of streamwater discharge is expressed well by the model. Separation of runoff components may be valid taking into account the separation points on the recession curve. In this case, a surface runoff component is not present. Both solute concentrations decrease near the time of peak discharge, thus the patterns of solute behavior resemble each other. Significant correlation of streamwater solute concentrations between estimated and observed can also be seen in Fig.4(a). Similar illustrations obtained for Event T-2 are shown in Fig.4(b). In this case, rainfall intensity was not high, but the rainfall amount of 136.5 mm was the biggest for the three events. Though the event has four peaks in the hydrograph, estimated streamwater discharge correlates well with the observations. The separation of runoff components are also valid. Temporal variations of streamwater concentrations of Cl^- and SO_4^{2-} are also estimated well by the model. It is found that streamwater concentration at peak discharges increases with time as shown in Fig.4(b). In Event T-3, rainfall intensity was not so high in comparison with Event T-2, and rainfall amount was only 26.5 mm. But the slope of hydrograph is very steep, because it rained intensively in Event T-3 as shown in Fig.4(c). Streamwater discharge is expressed well by the model and separation of runoff components also may be valid. A different temporal pattern of SO_4^{2-} can be seen at the time of peak discharge where streamwater concentration is higher than that of pre-storm in comparison with the other two events. However, significant correlations between estimated and observed streamwater concentrations are found in Fig.4(c).

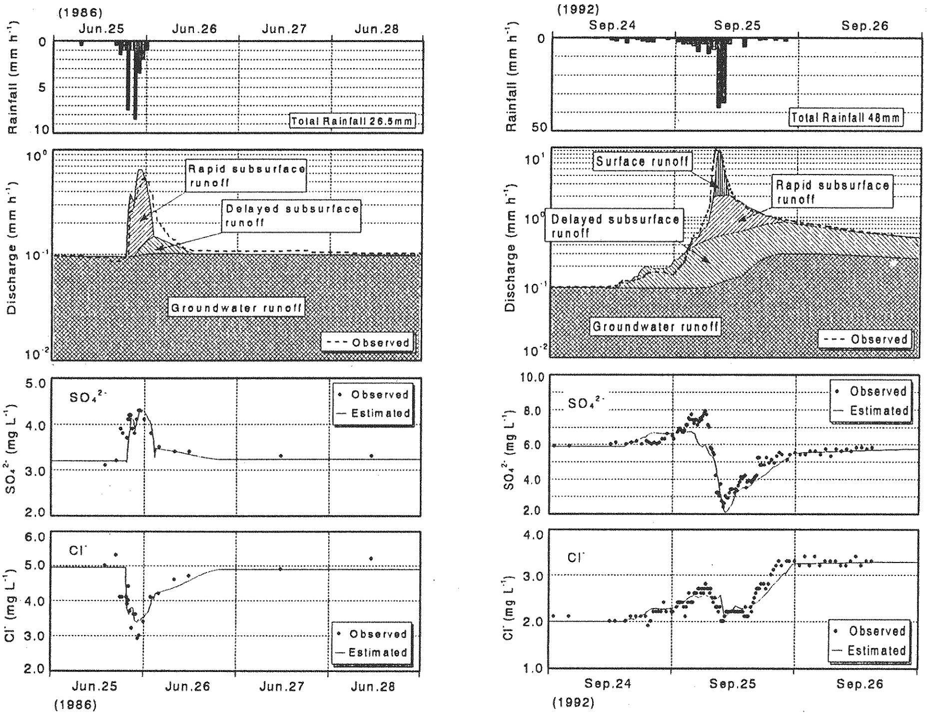
Table 2 Identified parameters and initial values

Substances Parameters	Event T-1		Event T-2		Event T-3		Event S-1	
	Cl^-	SO_4^{2-}	Cl^-	SO_4^{2-}	Cl^-	SO_4^{2-}	Cl^-	SO_4^{2-}
ν_s			0.429	0.400			1.116	2.022
k_s			0.088	0.200			0.131	0.106
ν_x			2.722	4.973			0.002	0.003
k_x			0.144	0.250			0.219	0.222
ν_y			0.002	0.009			0.002	
k_y			0.110	0.248			0.247	
C_{xini}	5.980	5.250	5.500	7.610	4.340	4.120	2.200	7.858
C_{yini}	5.580	5.100	5.450	4.540	4.540	3.800	2.200	7.774
C_{gini}	4.700	3.800	4.950	4.530	4.950	3.200	2.100	5.911
S_{asini}	36.40	17.50	40.90	20.00	35.20	22.00	30.50	31.92
S_{axini}	41.40	21.00	38.30	30.40	30.10	16.40	20.00	152.01
S_{ayini}	50.80	20.50	49.60	18.30	41.20	15.30	18.00	
μ_b							2.721	
μ_g							2.546	



(a) Event T-1 (Jun.13-Jun.16, 1985)

(b) Event T-2 (Jun.29-Jul.3, 1985)



(c) Event T-3 (Jun.25-Jun.28, 1986)

(d) Event S-1 (Sep.24-Sep.26, 1992)

Fig. 4 Hyetograph, hydrograph and temporal variations of streamwater solute concentration

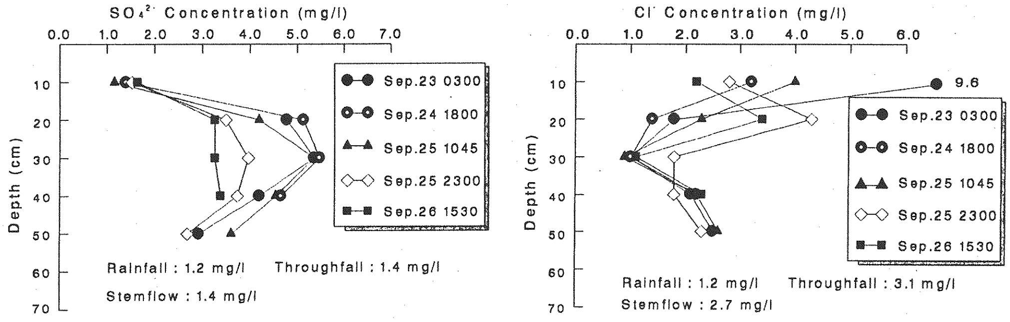
Shirakawatani Experimental Forested Basin

In Shirakawatani basin, parameters ν_g and k_g in the groundwater tank also were not identified as well as in Tsukuba basin, but ν_y and k_y in the secondary soil tank and special correction factors μ_b and μ_g were identified only for Cl^-. Hyetograph, hydrograph and temporal variations of streamwater concentration of solutes during a storm (Event S-1 : September 24-26, 1992) in Shirakawatani experimental forested basin are illustrated in Fig.4(d). It rained very intensively (maximum intensity was about 38 $mmhr^{-1}$) during the storm, and the amount of rainfall was 145 mm. Estimated temporal variation of streamflow discharge correlates with the observed results and the separation of runoff components may be valid based on several separation points on the recession curve of the hydrograph. Surface runoff is apparently identified around the time of peak discharge. In the event, temporal variations of streamwater concentrations of Cl^- and SO_4^{2-} are quite different; the SO_4^{2-} concentration declines until the time of peak discharge and almost recovers to the pre-storm level at the end of the storm. However, the Cl^- concentration increases especially after the time of peak discharge, while it decreases slightly a little before the time of peak discharge. Significant correlation between estimated and observed streamwater concentrations of Cl^- and SO_4^{2-} were obtained during the storm.

DISCUSSION

In Tsukuba basin, a vertical profile of soil water concentration of Cl^- before and after a storm event (Event T-3) was observed by Hirata et al. (12). Before the storm, it was almost uniform and its concentration was about 5.0 mgl^{-1} in the soil zone corresponding to B and C horizon. Initial values of soil water concentrations C_{xini}, C_{yini} and C_{gini} listed in Table 2 are slightly less than the observed data. But they are, at least qualitatively, consistent with the observed data, and valid initial values of soil water concentration may be obtained in Event T-3. Because Cl^- concentration decreases significantly when rapid subsurface runoff is dominant during the storm, it is suggested that rapid subsurface runoff mainly containing rainwater contributed to the dilution of the streamwater. Consistent results can be found in other storm events. Therefore, the model proposed could express roles of each runoff component in solute transport and the resultant behaviors of solute concentration in the streamwater. In the case of SO_4^{2-}, temporal variations of streamwater concentration illustrated in Figs.4(a), 4(b) and 4(c) differ from those of Cl^-; the role of the rapid subsurface runoff component was the same as that of Cl^-, however, the delayed subsurface runoff component contributed to rising of streamwater concentration of SO_4^{2-} in all storms. It may be suggested that the delayed subsurface runoff component carried a large amount of SO_4^{2-} to the streamwater, and thus the soil tanks were a potential source of SO_4^{2-}. Initial values of soil water concentration identified for each soil tank listed in Table 2 are consistent with the hypothesis of streamwater chemistry, because initial values of mobile solute concentration, C_{xini} and C_{gini} correspond to delayed subsurface and groundwater runoffs, respectively. Initial value of the delayed subsurface runoff was apparently greater than that of groundwater runoff especially in Event T-2 and T-3 as shown in Table 2. Therefore, the initial condition of soil water may have much influence on the formation of streamwater chemistry. Because unique values were identified for each parameter through the three events, some discrepancies between estimated and observed streamwater concentration are found in Figs.4(a), 4(b) and 4(c). This may be the other reason why streamwater chemistry greatly depends on initial conditions of soil water chemistry and the initial conditions were not given correctly because information about soil water is little.

In Shirakawatani basin, the same kind of parameters as in Tsukuba basin were identified for SO_4^{2-}. Temporal pattern of streamwater concentration was, in principle, the inverse of the hydrograph. But we can see two remarkable features where, 1) streamwater concentration was raised until rapid subsurface runoff took place and 2) streamwater concentration recovered to the pre-storm level at the end of the storm. This may suggest that the delayed subsurface runoff component contributed to the rise of streamwater concentration until rapid subsurface runoff took place, and the main source of SO_4^{2-}



(a) SO_4^{2-} (b) Cl^-

Fig. 5 Temporal variation of vertical profile of soilwater concentration

had already disappeared on the recession limb of the hydrograph. Identified initial concentrations of mobile solutes, C_{xini} and C_{yini} in the primary and secondary soil tanks, respectively were higher than that of the groundwater tank. Vertical profiles of soil water concentration of SO_4^{2-} are shown in Fig.5(a). Here, an apparent decline of the SO_4^{2-} concentration at a depth of 30 cm corresponding the 'B horizon' before and after the time of peak discharge can be noticed. The above may prove temporal change of streamwater concentration of SO_4^{2-} and high initial values of soil water concentration in the primary and secondary soil tanks.

Cl^- concentration continued to increase with the time except around the time of peak discharge. This is a quite different feature of streamwater chemistry in Shirakawatani basin from that in Tsukuba basin. Vertical profiles of soil water concentration of Cl^- are shown in Fig.5(b). The figure represents a decrease of Cl^- in the surface soil zone and an apparent increase in the deeper soil zone. From 0300 on September 23 to 1800 on September 24, a very high concentration at a depth of 10 cm was found because of little water in the surface soil zone. At a depth of 10 cm, after a drastic decrease of Cl^- concentration until 0300 on September 23, it was found that the concentration increased just before the time of peak discharge and decreased after that. On the other hand, an apparent increase of the concentration at a depth of 20 cm can be seen, but there was little change at a depth of 50 cm. The following hypotheses may be suggested; 1) delayed and rapid subsurface runoffs contained much Cl^-, at latest, before the time of peak discharge, and 2) solute concentration of groundwater runoff should be higher after the time of peak discharge than before. An intensive downward movement of Cl^- from the upper tank to the primary soil tank and the groundwater tank is clearly necessary to express the rapid increase of the concentration of streamwater as shown in Fig.4(d). Therefore, it seemed that correction factors to promote downward solute fluxes due to infiltration and percolation, μ_b and μ_g, which were greater than 1.0, were identified only for Cl^- in this basin. As hydrochemical processes have their own characteristics, and spatially differ, it is essentially difficult to precisely express hydrochemical processes using a lumped modeling method such as the tank model. However, from a viewpoint of civil engineering, it is necessary for water resources managers to quantify hydrochemical processes and to predict streamwater chemistry as an integrated result in a basin. It may be valuable to attempt to express the processes taking advantage of a lumped modeling method. In the present, because information about formation processes of soil water and streamwater chemistries is little, more information is necessary to verify the authors' methodology. Details of the model structure and its validity should be investigated using several sets of data.

It has been reported that forest trees affect throughfall and stemflow chemistries (7), but in principle, average throughfall concentration was adopted for evaluating solute loads of rainwater as atmospheric inputs. Because very intensive acid rain may have a considerable effect on streamwater chemistry, solute migration in the rainfall interception process as modeled by the authors' group (5) also should be quantified for consistent analysis of solute migration in forested basins.

CONCLUDING REMARKS

For quantitative analyses of solute migration in forested basins, a mathematical model for describing the solute runoff process in the soil was developed. The model takes two hydrochemical processes into account; solute advection due to throughflow, infiltration and percolation and solute exchange between mobile and immobile regions in the soil. Two anions; Cl^- and SO_4^{2-}, which are not biochemically reactive and readily mobile, were used as water quality parameters. The model was applied to observed streamwater concentrations of Cl^- and SO_4^{2-} in two forested basins in Japan, and attempted to describe the solute runoff process in the basins. Temporal variations of streamwater discharge were simulated well by the model and the four runoff components separated by the model also appear to be reasonable. Temporal variations of streamwater concentration estimated by the model correlated well with the observed data. Model parameters identified were consistent with temporal variation of streamwater concentration and vertical profiles of soil water concentration obtained from Shirakawatani Basin. Because there were only the data of the vertical profile of soil water concentration for Shirakawatani basin, the precise structure of the model and mechanism of solute migration could not be identified. They should be investigated further, based on the relationship between temporal variation of the vertical profile of soil water chemistry and solute concentration in each tank. In the future, the model proposed may be extended to incorporate biochemical processes for analyzing seasonal changes of streamwater and soil water chemistries.

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APPENDIX - NOTATION

The following symbols are used in this paper :

C_{est}, C_{obs} = estimated and observed streamwater concentrations;

C_r = rainfall concentration;

C_s, C_x, C_y, C_g = mobile solute concentrations in upper, primary soil, secondary soil and groundwater tanks;

C_{sini}, C_{xini}, C_{yini}, C_{gini} = initial values of mobile solute concentrations in upper, primary soil, secondary soil and groundwater tanks;

C_{xy} = mobile solute concentration of soilwater exchange flux;

D = depth of surface soil (corresponding to 'A horizon');

F = evaluation function for parameter identification;

f_b = percolation rate from upper tank to primary soil tank;

f_g = percolation rate from upper tank directly to groundwater tank;

f_x, f_y = porosities of primary and secondary soil tanks, respectively;

h_g = threshold of depth that unconfined groundwater runoff takes place;

h_x, h_y = depths of primary and secondary soil tanks, respectively;

k_s, k_x, k_y, k_g = solute distribution coefficients in upper, primary soil, secondary soil and groundwater tanks, respectively;

L_T = soilwater exchange rate;

N = number of observed data for parameter identification;

q_g = rate of groundwater runoff from groundwater tank;

q_h = rate of steady percolation from primary soil tank to groundwater tank;

q_i = rate of delayed subsurface runoff from upper tank;

q_j = rate of temporary percolation from primary soil tank to groundwater tank when water storage in primary soil tank exceeds field capacity;

q_k = rate of delayed subsurface runoff from secondary soil tank;

q_l = rate of steady percolation from secondary soil tank to groundwater tank;

q_{o1} = rate of surface runoff which has little contact time with soil aggregate;

q_{o2} = rate of surface runoff which has long contact time with soil aggregate;

q_s = rate of rapid subsurface runoff from upper tank;

R = net rainfall intensity;

S_s, S_x, S_y, S_g = water storages in upper, primary soil, secondary soil and groundwater tanks, respectively;

S_{asini}, S_{axini}, S_{ayini}, S_{agini} = immobile solute concentrations in upper, primary soil, secondary soil and groundwater tanks, respectively;

t = time;

γ = porosity of upper tank;

μ_b, μ_g = correction factors for infiltration and percolation fluxes of solutes; and

ν_s, ν_x, ν_y, ν_g = solute exchange coefficients in upper, primary soil, secondary soil and groundwater tanks, respectively.

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