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FORMATION PROCESS OF STREAMWATER CHEMISTRY IN SMALL FORESTED MOUNTAIN BASIN

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SYNOPSIS

This study focussed on investigating the formation process of streamwater chemistry in a small forested mountain basin. Solutes in streamwater were classified into three types, based on the observation and analysis of streamwater and soilwater during a rainfall event. In order to understand the formation process of the streamwater concentration, four typical solutes, SiO₂, SO₄²⁻, NO₃-N and CI were selected, and the relation between temporal variations of runoff components and solute concentrations for both streamwater and soilwater were investigated. The volumetric relative contributions of runoff components, through the different water flow pathways, to the formation of the streamwater concentration of solutes during a short time period, were qualitatively discussed for the first step of mathematical modelling of solute runoff process.

INTRODUCTION

Water and chemical elements circulate in forested basins which can be considered a Soil-Plant-Atmosphere Continuum, so called 'SPAC'. Field observations (1,2,3,4) have suggested that biochemical and physicochemical actions cause rainwater and streamwater chemistry to vary both qualitatively and quantitatively in the runoff process through different water flow pathways. An explanation for this could be that forested basins act as a buffer to control the water runoff process, and to stabilize streamwater chemistry. This buffering action may be the basis of the water purifying function in forested basins. Therefore, the understanding of the circulation process of both water and solutes, and the resultant quantitative prediction of the streamwater chemistry, are necessary for the analysis and measure of practical issues which could include water eutrophication caused by the forest felling for the development or the construction of dam reservoirs. However, effective methodologies

for evaluating dynamics of solute circulation in forested basins have not been established yet.

We believe that mathematical modelling is likely to be one of the most appropriate way to quantify solute circulation in forested basins, based on an understanding of the coupling action of water and solutes in elemental hydrological and hydrochemical processes. Therefore it is important for mathematical modelling of solute runoff process to identify the major runoff component of solute transport at every moment. Then it would be effective to examine the relation between temporal variations of runoff components and stream solute concentration. In this study, field observations of rainfall, throughfall, stemflow, soilwater, streamwater discharge, and their concentrations, were carried out as the first step of model development. Observations were conducted biweekly, and intensively during a rainfall event. From an overall view point, seasonal variations and long term analyses should also be taken into consideration, because the hydrochemical process are highly associated with forest ecosystem. However, this paper focusses on the qualitative investigations of the formation process of the streamwater chemistry, especially during a rainfall event. The relation between the streamwater concentration of solutes and rainwater runoff components as separated by authors' forest water circulation model (FOWAC Model) is investigated. Precise mechanisms of the solute runoff are also discussed.

OBSERVATIONS

The observation site is the Shirakawatani forested experimental basin (33°52'N, 133°40'E) located about 100 km west of Tokushima city in Shikoku island, Western Japan (Fig.1). This site has an area of 23 ha, altitudes between 740-1100 m and a mean slope angle of 21.5°. The predominant tree species is Japanese cedar (*Cryptomeria japonica*) which covers three fifths of the area. The remainder is covered with deciduous broad-leaved forest. The site is underlain by psammite and clay schists with surface brown forest soil. The 'A' horizon is less than 10 cm and 'B' horizon is about between 50-80 cm. Thus, the whole soil layer is almost less than 1 m.

The streamwater level was measured by a float type level recorder at a 90° V-notch weir (S1 in Fig.1), and was converted into streamwater discharge. Rainfall was observed with a tipping bucket type rain gauge also located at S1. Throughfall was observed with two tipping bucket type rain gauges and a throughfall trough, which had an area of 7.2 m², under a 32 year old Japanese cedar (S2 in Fig.1). Stemflow was obtained from the measurement of the rainwater captured with a vinyl collar sealed to the stem of the sample tree at breast height (1.3 m) by means of a tipping bucket type rain gauge.

Observation of common meteorological parameters (air temperature, relative humidity, wind speed, net radiation and solar radiation) were taken 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 sample tree. All parameters except water level and rainfall were measured and recorded on data loggers with a time interval of 10 minutes. The duration of sunshine data was obtained from Ikeda Dam Group Operation Office (Water Resources Development Public Corporation).

Streamwater was gathered about 100 m upstream from the weir at a time interval ranging from 15 minutes ~ 3 hours, depending on stream discharge. Rainfall, throughfall and stemflow were collected in polyethylene tanks. At S2 in Fig.1, soilwater 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 soilwater. Soilwater was extracted by a suction pump operated using a microtube connected to the porous cup. Chemical determination of water samples is made for nutrients, metal ions, chloride ion, silica, suspended solids (SS), pH and electrical conductivity (EC). Data were collected over the period from September 23 to September 26 in 1992, when a single rainfall event of 145 mm, occurred with a peak rainfall intensity of 38 mmh⁻¹.

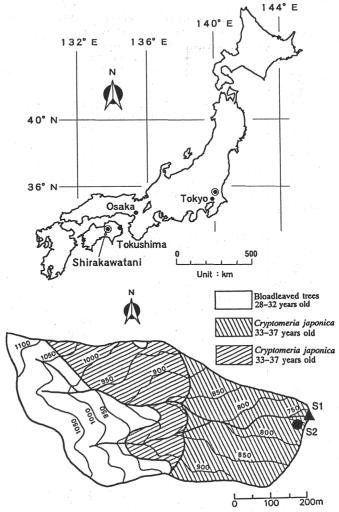


Fig.1 Schematic illustration of the Shirakawatani forested experimental basin

SOLUTE RESPONSE TO RAINWATER RUNOFF

Solute behaviour in streamwater

Temporal variations of streamwater solutes and concerned chemical parameters: NO₃-N, Cl⁷, SO₄², Ca²⁺, Na⁺, K⁺, SiO₂, pH, EC and SS are given in Fig.2. It appears that these can be divided into three groups, based on solute behaviour before and after the peak discharge. Muraoka and Hirata (1) reported similar observation results in the Tsukuba experimental forested basin.

(a) Type A: After the peak discharge, the solute concentration of streamwater recovers to the baseflow level prior to the rainfall event. Elements following this pattern are SiO₂, SO₄², K⁺ and SS. In the time when direct runoff, which consists of surface and rapid subsurface runoff ceases and streamwater is only composed of the groundwater runoff component, the solute concentration returns to the baseflow level. It may be interpreted that the groundwater concentration dominates solute behaviour in streamwater. In the case of SiO₂, K⁺ and SO₄², it appears that the concentrations of direct runoff and delayed subsurface runoff are always less than that of groundwater. However, the recovery patterns of the streamwater concentration after the peak discharge are a little different among them. SS has an opposite pattern that the streamwater concentration at the time of the peak discharge is

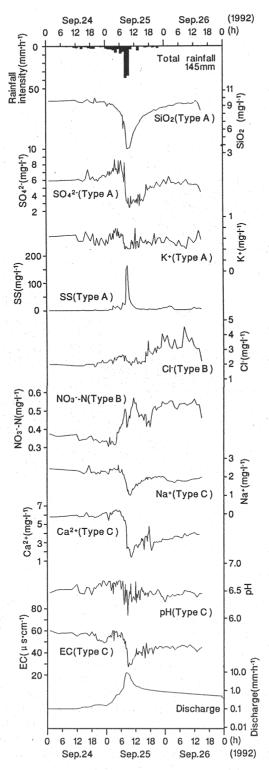


Fig.2 Temporal variations of streamwater solutes

higher than that of base level.

- (b) Type B: When the solute concentration goes up after the peak discharge, it overshoots the baseflow level. Elements following this pattern are NO₃-N and Cl. Delayed subsurface and groundwater runoff components are in general predominant during the recession period. Therefore, these two runoff components may contribute to increasing the concentration of solutes, which is higher than that of baseflow. When groundwater concentration has little variation, the high concentration of streamwater may be explained by the increasing concentration of delayed subsurface runoff components. This hypothesis can be verified by the vertical profile of solute concentrations in soilwater.
- (c) Type C: The solute concentrations during the recession period are smaller than those of baseflow. Elements following to this pattern are EC, pH, Ca²⁺, Mg²⁺ and Na⁺. The solute concentration may return to the baseflow level in future, judging from biweekly observations. This solute behaviour suggests the reduction of the groundwater concentration, but it has not yet been clarified by observations.

Solute behaviour in soilwater

SiO₂ and SO₄², and NO₃-N and Cl⁻ are adopted for the representatives of Type A and Type B, respectively. Vertical profiles of solute concentrations for Type A (SiO₂ and SO₄²) and Type B (NO₃-N and Cl⁻) are shown in Figs.3(a), 3(b), 3(c) and 3(d), respectively. Referring to the hydrograph illustrated in Fig.2, the times shown in the legend correspond to the pre-event time, the initial rainfall event time, the time of the peak discharge and the recession time, respectively. However, plotted concentrations are not instantaneous values, but average values for the sampling interval time.

(a) Type A: It is clearly found that the concentration of SiO₂ increases with the soil depth, and reduces uniformly over time at each measurement point (see Fig.3(a)). Because SiO₂ originates purely from bed rocks, and the longer the contact time of soilwater with Soil and bed rocks, the higher soilwater concentration will be. Because the solute concentration of groundwater should be higher than that at 50 cm in depth, we can expect the groundwater runoff component to contribute to the baseflow and the direct runoff component to dilute the streamwater.

The maximum concentration of $SO_4^{2^2}$ are found at a depth of 30 cm, and the concentration decreases with the time at most depths except at 10 cm (see Fig.3(b)). The vertical profile of $SO_4^{2^2}$ are different from that of SiO_2 . The temporal variation of the vertical profile could suggest that the deep soil is one of the source of $SO_4^{2^2}$.

(b) Type B: Vertical profiles and their temporal variations are quite different from those of NO. N and Cl in Figs.3(c) and 3(d), respectively. The vertical profile of NO₂-N on September 23 shows that the concentration near the surface is much higher than those at deeper location (See Fig.3(c)). This is because nitrogen is one of essential major elements for the growth of forest plants, and is fixed and accumulated by bacteria in the root zone. During the rising limb of the hydrograph, the concentration reduces to about a half at a depth of 10 cm. This is located in the 'A' horizon of the soil profile. However, at a depth of 20 cm, this trend is reversed. During the recession period, the concentration at a depth of 20 cm increases to about four times of that before the peak discharge. While at 10 cm in depth it decreases to the one third of that at the pre-event time. This may be explained by the downward movement of NO₃-N from the surface layer to the deeper 'B' horizon due to the percolation of rainwater. Rapid subsurface flow which takes place in 'A' hirozon would mainly transport the NO, N during the rising limb of the hydrograph, as shown in the vertical profile at 0300 on September 23. However, the concentrations at the depth deeper than 20 cm increase after the peak discharge (i.e. at 2300 on September 25), delayed subsurface flow which occurs in deeper soil would transport NO₃-N to stream in the place of rapid subsurface flow. Therefore, it is suggested that the main transport medium of NO₃-N changes from rapid subsurface flow to delayed subsurface flow durung a rainfall event. The vertical profile at 1530 on September 26 is similar to that at 2300 on September 25, and the concentration at a depth of 20 cm is higher than before. This shows that the percolating rainwater causes the vertical transport of NO₃-N after the time of peak discharge.

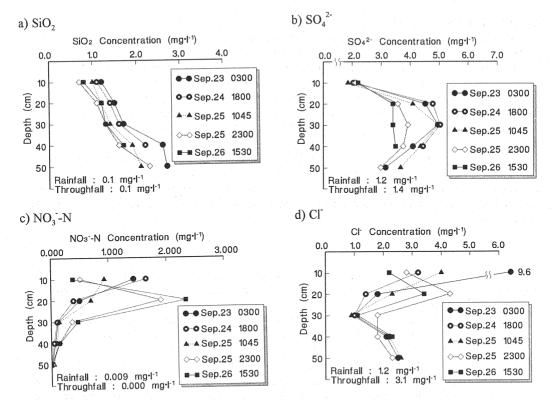


Fig.3 Temporal variations of vertical profile of soilwater solute concentration

The vertical profile of Cl is a little defferent from NO₃-N (see Fig.3(d)). The concentration near the surface is also much higher than those at deeper location on September 23. During the rising limb of the hydrograph, the concentration reduces to about less than a half at a depth of 10 cm. However, the concentrations show little change at other depths (at 1800 on September 24). During the recession period, the concentration at a depth of 20 cm increases to about a twice of that before the peak discharge, while at 10 cm in depth it decreases to the one third of that at pre-event time (at 2300 on September 25). Of course this may be also explained by both the transport of Cl to stream due to rapid subsurface flow and the downward movement of Cl from the surface layer ('A' horizon) to the deeper soil('B' horizon) due to the percolation of rainwater as well as NO₃-N. Therefore, delayed subsurface flow would transport Cl to streamwater in the place of rapid subsurface flow during the recession period.

RESULTS AND DISCUSSIONS

The hydrograph composed of runoff components separated by authors' forest water circulation model (5) (see Fig.4), is compared with the observation in Fig.5. There is good agreement between the observed and estimated runoff discharge. This indicates that the separation model is reasonable. Hereafter the following discussion is based on the result. That is, the following procedure is important for investigating the formation process of streamwater chemistry. First, the hydrograph should be precisely separated in order to identify the key runoff component which transports solutes over time. Subsequently temporal variations in the streamwater concentration should be correlated to the composition of runoff components, incorporating vertical profiles of solute concentrations in soilwater. This is the basis of mathematical modelling of solute runoff process.

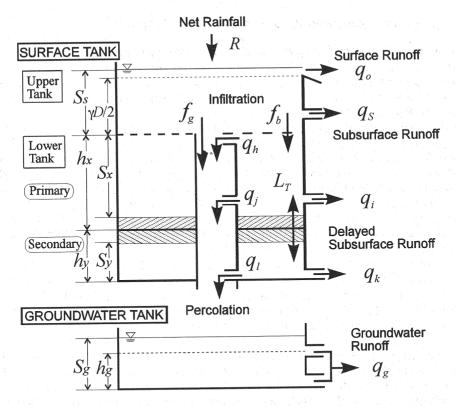


Fig. 4 Schematic illustration of authors' forest water circulation model

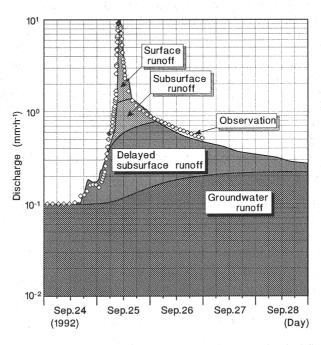


Fig. 5 Runoff components separated by the authors' model

Formation mechanism of streamwater chemistry of SiO₂ and SO₄²⁻ (Type A)

The separated four runoff components, the their composition, and temporal variations of concentrations of SiO₂ and SO₄² in Fig.6. For the clear understanding, sevaral data are selected and plotted. The relation between the streamwater concentration and relative contributions of each runoff component, namely, surface, rapid and delayed subsurface, and groundwater runoffs, to the streamwater discharge are shown as follows.

The streamwater concentration of SiO₂ reduces mainly when direct runoff flow comprising surface and rapid subsurface flow takes place in Fig.6 (during the period between time marks ② and ⑤). Thus, this suggests that direct runoff flow which is almost composed of rainwater contains little SiO, and plays a role to dilute streamwater concentration. This is also verified by the relation between the stramwater concentration and relative contributions of each runoff component. In essential, the relation between the streamwater concentration and the ratio of surface runoff to streamwater discharge has a negative correlation in Fig. 7(a). Especially at the time of peak discharge when surface runnoff occupies more than 80 % of streamwater discharge, the streamwater concentration rapidly reduces with the increasing ratio of surface runoff to the streamwater discharge. It is likely that a pre-event water composes the peak flow. Since the pre-event water had been in deeper soil, at least, it would have a higher concentration than surface flow. However, because the experimental basin has a thin soil less than 1 m and 'A' horizon is especially so thin and less than 10 cm that saturated overland flow is likely to occur, the contribution of translstory flow to the streamwater chemistry would be smaller than that of surface flow. Then the reduction of the streamwater concentration would be because of the surface flow which has the short contact time with soil aggregate. Therefore, it is found that the suface runoff component has a major dilution effect on the formation mechanism of the streamwater chemistry. This result is very similar to observations reported by Shimada et al. (6), and Tanaka and Yamazaki (7). On the other hand, the base runoff component, which consists of delayed subsurface and groundwater runoff, contributes to increase of the streamwater concentration after direct runoff has disappeared in Fig.7(c) and (d) (after the time mark (5)).

The relation between the streamwater concentration of SO₄² and relative contributions of each runoff component to streamwater discharge are shown in Figs. 8(a), 8(b), 8(c) and 8(d). The streamwater concentration of SO₄² tends to increase from the time when delayed subsurface runoff occurs (at the time mark ①). In Fig.8(c), the ratio of delayed subsurface runoff to streamwater discharge and the streamwater concentration has an positive correlation. Thus the more delayed subsurface runoff, the higher streamwater concentration. This could suggest that delayed subsurface runoff transports SO₄² to the stream, taking account of the vertical profile of soilwater concentration before peak discharge. Consequently the steamwater concentration starts to decrease with the increase of rapid subsurface runoff. A negative correlation can be found between the streamwater concentration and the ratio of rapid subsurface runoff to streamwater discharge in Fig. 8(b). This implies that the more rapid subsurface runoff, the lower streamwater concentration will be, that is, rapid subsurface runoff would dilute the streamwater concentration. Because the soil at a depth of 10 cm contains little SO₄²⁻ in Fig. 4, therefore, rapid subsurface runoff could not supply SO₄²⁻ to the stream. When surface runoff takes place (during the period between time marks 3and4), the streamwater concentration is much lower than the preevent level. We can find a negative correlation between the streamwater concentratio and the ratio of surface runoff to streamwater discharge in Fig.8(a). Thus surface runoff could strongly dilute the streamwater. During the recession period, it is suggested that the less surface and rapid subsurface runoffs, the higher streamwater consentration in Figs.8(a) and (b). Delayed subsurface runoff also has a role to dilute the streamwater, because 'B' horizon at a depth of 30 cm contains less SO₄²⁻ at 2300 on September 25 than the pre-event (at 300 on September 23).

Formation mechanism of streamwater chemistry of NO₃-N and CI (Type B)

In Fig.6, the concentration of NO_3 -N increases with the increasing ratio of rapid subsurface runoff to streamwater discharge before the time of peak discharge (during the period between time marks ②

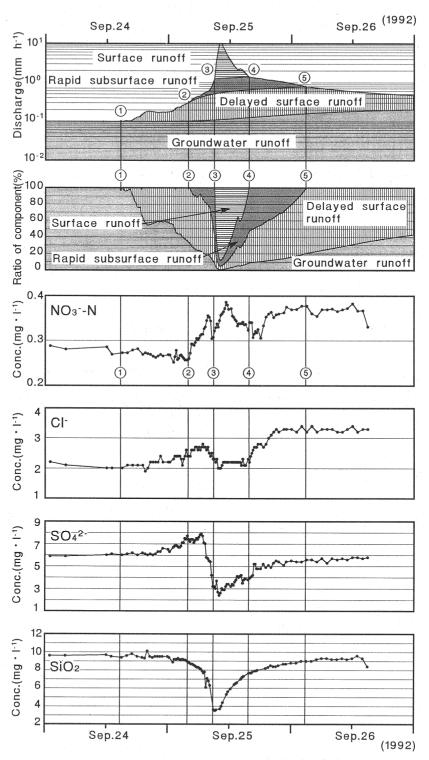
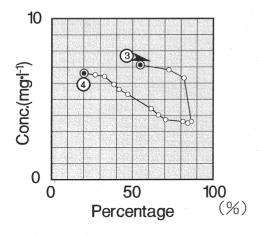
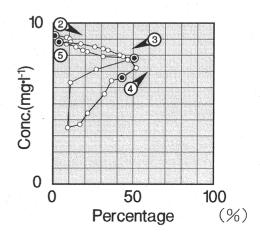


Fig.6 Temporal variations of hydrological and hydrochemical parameters

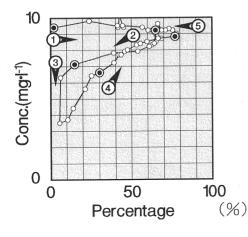
(a) Surface runoff



(b) Rapid subsurface runoff



(c) Delayed subsurface runoff



(d) Groundwater runoff

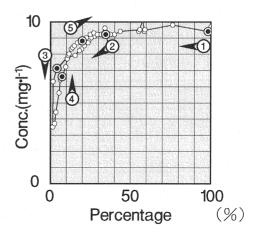
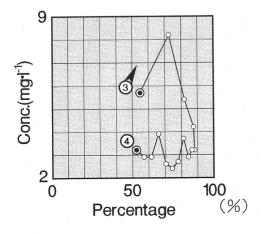
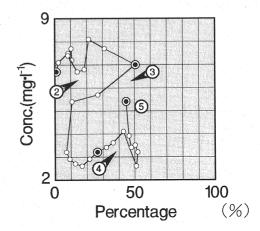


Fig. 7 The relation between the streamwater concentration of SiO₂ and the ratio of each runoff component to streamwater discharge for 24-26 September 1992.

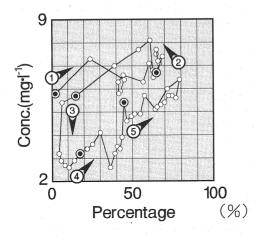
(a) Surface runoff



(b) Rapid subsurface runoff



(c) Delayed subsurface runoff



(d) Groundwater runoff

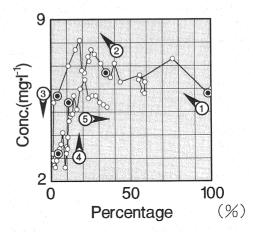


Fig. 8 The relation between the streamwater concentration of $SO_4^{\ 2}$ -N and the ratio of each runoff component to streamwater discharge for 24-26 September 1992.

and ③). However, the concentration turns to decrease with the increasing ratio of surface runoff to streamwater discharge (during the period between time marks ③ and ④). On the contrary, the increasing ratio of rapid subsurface runoff does not cause the increase of the streamwater concentration after peak discharge. The rapid increase of the concentration results from the increasing ratio of delayed subsurface runoff to streamwater discharge.

The similar relations as Figs. 7(a), 7(b), 7(c) and 7(d) are illustrated in Figs. 9(a), 9(b), 9(c) and 9(d). It is clearly found in Fig.9 (a) that surface runoff fundamentally dilutes streamwater (during the period between time marks 3 and 4), and is most effective at the time of peak discharge as for SiO2. As in the case of rapid subsurface runoff, the streamwater concentration increases with the inceasing ratio of rapid subsurface runoff to streamwater discharge during the rising limb of the hydrograph (during the period between time marks 2 and 3). On the contrary, Fig. 9(b) indicates that the streamwater concentration decreases with the increasing ratio of rapid subsurface runoff to streamwater discharge during the recession period (during the period between time marks 4) and (5). In other words, this suggests that the same runoff component plays quite opposite roles before and after the time of peak discharge. Therefore, it is suggested that rapid subsurface flow contains much NO₃-N, but the major runoff component which transports NO₃-N has already changed after the time of peak discharge. As shown in Fig. 3(c), the concentration of NO₃-N near the ground surface (at 10 cm in depth) is higher than those in the deeper soil (at 20 or 30 cm in depth) before the time of peak discharge. However, the percorating rainwater transports the NO₃-N from the surface layer to the deeper soil. Because of this, the NO₃-N concentration in the deeper layer goes up during the recession period in Fig.3(c). On the other hand, the ratio of delayed subsurface runoff to streamwater discharge and the streamwater concentration of NO₂-N have a negative correlation until the end of rapid subsurface flow in Fig. 9(c)(at the time mark ⑤). However, the relation turns to be positive after that time. This suggests that the delayed subsurface runoff takes part in the transport of NO₃-N during the recession period. That is, it may be verified that the transport of NO₃-N during the recession period is largely due to delayed subsurface runoff.

The streamwater concentration of Cl shows the similar temporal pattern to that of NO_3 -N in Fig. 6. After rapid subsurface runoof takes place (from the time mark ②), the streamwater concentration tends to increase. When surface runooff occurs (during the period between time marks ③and④), the concentration decreases and returns to the baseflow level. It would be because surface and subsurface runoffs have a similar concentration. After surface runoff ceases (from the time mark ④), the reduction of rapid subsurface runoff and the increase of delayed subsurface runoff cause the increase of the streamwater concentration. The relations between the streamwater concentration and the ratio of each runoff component to streamwater discharge are illustrated in Figs. 10(a), 10(b), 10(c) and 10(d). These show almost the same patterns as NO_3 -N.

CONCLUDING REMARKS

It has been difficult to discuss the precise mechanism of solute runoff in previous studies because of difficulties such as the separation of the hydrograph. In this study, the formation process of the streamwater chemistry has been qualitatively investigated, based on the separation of the hydrograph, the temporal variation of the vertical profile of the solute concentration in the soilwater, and the relation between the streamwater concentration and the composition of runoff components in streamwater. The authors considered the separation of the hydrograph, and the resultant evaluation of several water flow pathways from the surface to stream, to be key points for the mathematical modelling of solute runoff. We therefore adopted the following methodology. First, the hydrograph was separated into four runoff components, using our hydrological water circulation model for a rainfall event. Then, temporal and spatial variations of water flow pathways were routed, and the effective runoff component for the solute transport and its water flow path was identified over time. On the other hand, the vertical profile of the solute concentration in the soil is monitored during a rainfall event. Their temporal and spatial variation identified the location and movement of the solutes. Finally, the investigation of the relation between

(%)

Percentage

(a) Surface runoff (b) Rapid subsurface runoff 0.6 0.6 Conc.(mg·l-1) Conc.(mg·l-1 0.3 50 100 (%) 50 100 0 Percentage (%) Percentage (c) Delayed subsurface runoff (d) Groundwater runoff 0.6 0.6 Conc.(mg·l⁻¹) 0.3 0.3 100 (%) 50 50 100

Fig. 9 The relation between the streamwater concentration of NO₃-N and the ratio of each runoff component to streamwater discharge for 24-26 September 1992.

Percentage

(b) Rapid subsurface runoff (a) Surface runoff 5 5 Conc.(mg·l-1) Conc.(mg·l-1) 1 100 (%) 50 100 50 0 0 (%) Percentage Percentage (d) Groundwater runoff (c) Delayed subsurface runoff 5 5 Conc.(mg·l-1) Conc.(mg·l-1) 1 100 50 100 50 0 0 (%) (%) Percentage Percentage

Fig. 10 The relation between the streamwater concentration of Cl and the ratio of each runoff component to streamwater discharge for 24-26 September 1992.

the streamwater concentration and the composition of runoff components in streamwater, enabled us to evaluate the relative volumetric contribution of each runoff component to the formation of thestreamwater concentration. The authors think that this is the most important information for mathematical modelling of solute runoff in forested basins. This knowledge which approaches the internal mechanism in the soil, is essential for the successful modelling of the solute balance in forested basins. In future works, the authors intend to develop a mathematical model for quantitatively describing the temporal variation of the streamwater concentration, based on qualitative information obtained here.

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