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DISCHARGE CHARACTERISTICS OF NO_3^- FOR THE ANALYSIS OF BASINWIDE CIRCULATION OF WATER AND ENVIRONMENTAL POLLUTANTS IN A SMALL RIVER BASIN

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SYNOPSIS

Since water particles can never be labeled according to their traveling paths, separation of discharge components is an extremely difficult matter. Nevertheless it is one of the most important questions to be answered in hydrology, because it is directly relating to a practical problem of estimating effective rainfall in the rainfall-runoff analyses.

This paper investigates the capability of NO_3^- as a tracer of the subsurface flow component. The hypotheses made are that the source of NO_3^- is limited in the thin layer of ground surface, that NO_3^- is supplied to water particles only when raindrops infiltrate to the ground and that it does not change under the ground during the period of flood discharge. The study area is a small mountainous river basin Ai in the central part of Honshu Island of Japan. According to the observations, the peak of NO_3-N concentration of river water did not coincide with the peak of discharge. The NO_3-N concentration peaks usually occurred later than the hydrograph peaks, but there existed some exceptional cases depending upon the rainfall intensity and the antecedent precipitation events. The NO_3-N concentration of surface discharge from the roof tops and the tightly stepped bare playground was found as low as that of rain water. The concentration of well waters was found higher than that of river water at the low flow. These discharge characteristics of NO_3^- were concluded to constitute its potential to serve as a hydrological tracer of subsurface flow.

INTRODUCTION

NO_3-N concentration of natural water has been observed as an indicator of the general status of environmental quality. But no adequate technique of identifying NO_3^- behavior in the nature is yet available. Its identification is difficult because a water particle that goes through a particular path gets the NO_3-N concentration different from another particle through a different path. This fact, however, conversely implies that NO_3-N concentration may act as a tracer to get valuable information about subsurface flow.

The objective of this paper is to demonstrate the capability of NO_3^- existing everywhere in the nature as a tracer to detect the basinwide circulation of water as well as environmental pollutants. This paper especially forcuses on the various aspects of observed concentration of NO_3-N and other water quality indices in precipitation and river water in a small mountainous forestry basin. The groundwater samples from springs and wells were also analyzed.

NO_3^- is a substance present everywhere in the natural environment. Its major sources are the plant residues, animal wastes, domestic sewerages and various

fertilizers applied to agricultural fields. The organic nitrogen compounds are decomposed into NH_4^+ by microbial digestion, and then oxidized into NO_3^- which is stable under normal conditions in the sense that no more oxidation can occur in ordinary environment. The NO_3^- may disappear by plant uptake, by microbial assimilation or by denitrification under anaerobic condition with the presence of organic carbon. None of those phenomena may, however, occur in significant magnitude under the ground deeper than the thin surface layer of humus deposit within the time interval of direct flood discharge from a small river basin, say, less than 10 km^2.

The NO_3^- is also known free from adsorption by soil particles. It perfectly dissolves itself in water and flows absolutely along water. Those characteristics, i.e., limited supply location and invariant quality free from absorption and adsorption, constitute the basis for a hydrological tracer.

Among a variety of tracer materials for hydrological circulation analyses, such as radio isotopes, geochemicals and artificial tracers, the NO_3-N concentration is easily measurable and manageable. Besides, its availability and transport process are similar to many other environmental pollutants wich infiltrate and contaminate groundwater as well as surface water.

Thus the NO_3^- could be used as a practical tracer for analyzing both hydrological infiltration-runoff process and environmental subsurface pollution process. In particular, this paper tries to use NO_3^- to identify the subsurface flow during floods, whose identification is essential to the study of hydrological circulation.

STUDY AREA

Fig.1 shows the experimental basin used for this analysis. The basin is a small mountainous tributary basin of the Fuji River in the central part of Honshu Island of Japan. The discharge area is 8.4 km^2 of which 86 % is covered by forests of deciduous broad-leaved trees, 4 % is the agricultural land consist of paddy fields, dry fields and mulberry fields and the rest 10 % is residential area, channels and roads. The inhabitants are about 800. The average elevation is roughly 700 m above the mean sea level, the length of main stream is 4.3 km, the average channel slope is 1/9.6 and the highest and the lowest discharges observed during the test period, June 1982 - Sept. 1983, were 23.6 m^3/sec and 0.002 m^3/sec, respectively.

The mean annual precipitation over recent 30 years is about 1200 mm. The rainfall and river water stage are monitored by observation gauges and a weir installed at the site of Kamiigawa Bridge in June, 1983. The weir is made of steal and has 10 m wide double rectangular section. The river water stage is automatically recorded and flow rates are evaluated from stage-flow relationship of the weir. Prior to this installation, a rainfall gauge had long been operating in other site in the basin and the flow rate was measured time to time only manually by stage at the same site without the weir.

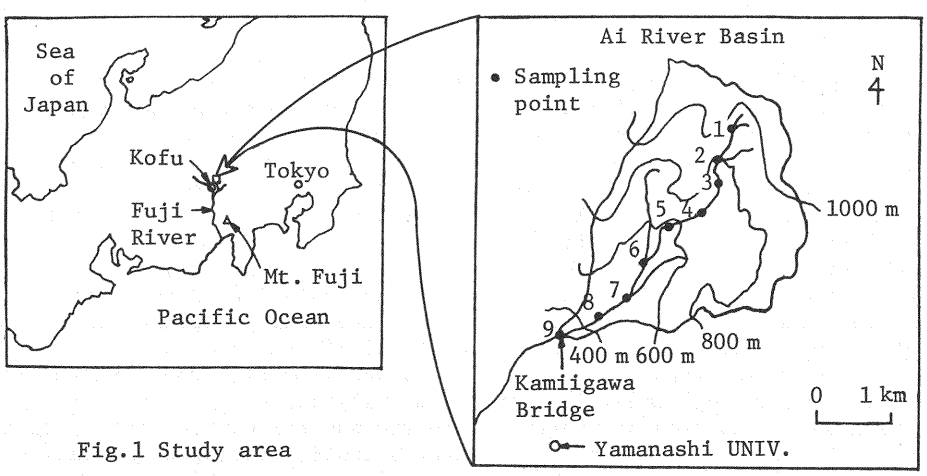

Fig.1 Study area

SAMPLING POINTS AND METHODS OF MEASUREMENT

Nine sampling points shown in Fig.1 were selected along the Ai River to investigate the spatial variation in NO_3-N concentration in the basin. The point No.9 is the key permanent observation station, where waters were sampled continuously for several months to investigate long-term changes of river water quality on clear days as well as short-term changes of storm water quality. Storm waters were collected at intervals varying from 20 min. to few hours on rainy days listed in Table 1. Rain No.8 and No.9 were caused by monsoon fronts and No.10 and No.11 by typhoons.

Sampled waters were filtrated with Whatman's GF/B glass-fiber filters, and NO_3-N concentration was determined by cadmium reduction method. The figures were employed as an average of two measurements when the two coincided within the difference of 0.01 mg/l . In some cases, electric conductivity (E.C.), suspended solids (SS), alkalinity and chemical oxygen demand with potassium per-manganate (COD_{Mn}) were also determined.

Table 1 List of rainfall and runoff data used in this study

Rain No.	Date	Duration hrs	Total rain mm	Maximum intensity mm/hr	Maximum flow m^3/sec	Total runoff mm	Runoff coeff.
1	82.6.20	4	40.0	33.0	0.86	3.2	0.08
2	82.6.21	6	17.5	14.5	0.63	4.6	0.26
3	82.7.8	6	33.5	16.5	0.43	1.6	0.05
4	82.9.24	26	31.4	4.5	-	-	-
5	82.10.8	28	37.9	3.2	-	-	-
6	82.11.10	33	22.7	2.3	-	-	-
7	83.8.2	2	16.0	13.0	0.35	1.0	0.06
8	83.8.9	1	41.5	41.5	1.42	2.6	0.06
9	83.8.10	1	16.0	16.0	0.34	0.7	0.04
10	83.8.15	62	319.5	38.5	23.63	165	0.52
11	83.9.27	39	149.5	17.5	8.38	60	0.40

- Data not available during channel improvement works

OBSERVATION RESULTS AND DISCUSSIONS

1. Seasonal and spatial changes of NO_3^- in river water

Observed seasonal changes of NO_3-N concentration of the river water are shown in Fig.2.a with daily rainfalls at station No.9 on clear days in 1982. The NO_3-N concentration reached to the maximum value in June, decreased in summer and gradually increased again in autumn and winter. The maximum value observed in June may be due to the irrigation water discharged from paddy fields. A slight increase in autumn and winter may correspond to small rainfall but may also be affected by the reduced flow rate. Except in June, the seasonal changes are, as compared with changes caused by rainfalls, small enough to be neglected when considering the short-term effects of rainfalls.

Fig.2.b shows the long-term changes both rainy and clear days in 1983. The overall pattern is similar to that of 1982 in Fig.2.a, but because of intensive sampling efforts, the detail short-term responses of NO_3-N concentration due to various rainfall events are observed. The typhoon which brought 320 mm rainfall on Aug. 15 seems to have considerably affected the NO_3^- status in the basin. It probably washed out the available NO_3^- stock thoroughly and even the aquifer became occupied by the diluted water. This influence continued half a month to reduce the concentration as low as 0.1 mg/l until another rainfall event came over on Sept. 1 . It increased the concentration again but this recovery was not quite enough. The diagram shows that it took another half month to return to the normal status.

Fig.3 shows the spatial changes along the stream on several days in 1982.

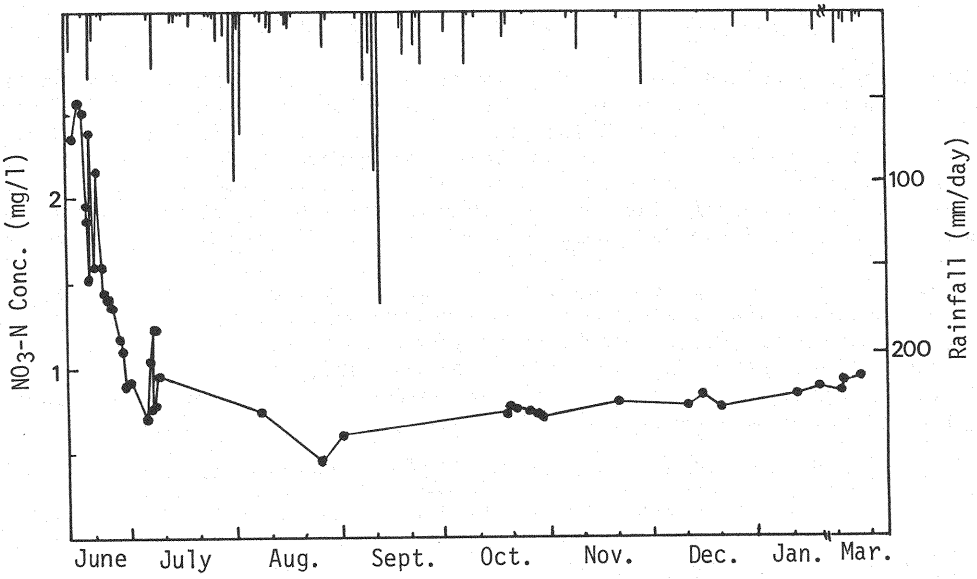

Fig.2.a Daily precipitation series and temporal changes of NO_3-N concentration of river water at Kamiigawa Bridge in 1982.

The high NO_3-N concentration was observed at the uppermost point, No.1, and it decreased as going downstream, but increased again in the downstream residential area, No.6 - No.9. The high value at No.1 seems to reflect the fertilizer applied to mulberry fields near by the point. The increase in the downstream is also due to the agricultural and other human activities. On June 17, the paddy fields were flooded and the remarkably high increase was observed in the downstream (o). On August 26, the observed NO_3-N concentration (Δ) was especially low at the midstream points, No.3 - No.5. At the beginning of August, a typhoon brought heavy rain of about 255 mm and most of NO_3-N stock in the soil might have been washed out.

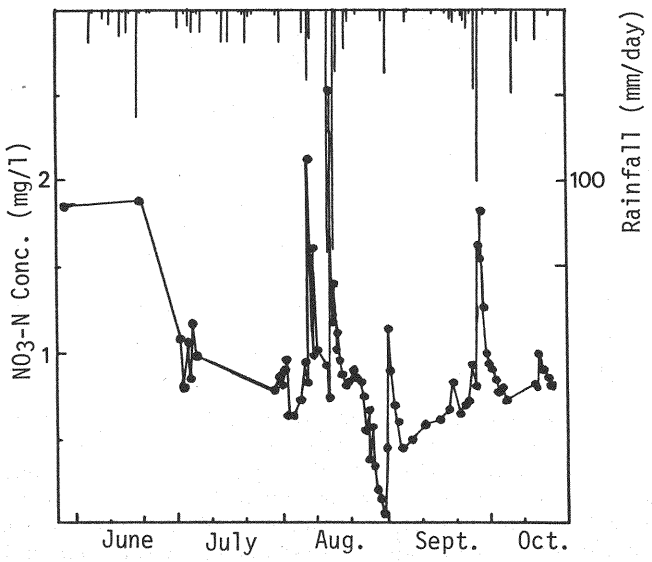

Fig.2.b Daily precipitation series and temporal changes of NO_3-N concentration of river water at Kamiigawa Bridge in 1983.

2. NO_3^- in rain water

Rain water quality was also analyzed to see its range and fluctuation pattern. Rain was collected in a 1 m x 1 m x 10 cm square sampler made of polyvinyl chloride for the rains No.4, 5 and 6. As shown in Figs.4 - 6, their NO_3-N concentrations were found high at the beginning of rainfall and reciprocally related to rainfall intensity, i.e., the stronger the rainfall intensity, the lower the NO_3-N concentration. This relation might be due to difference in the scale of raindrops depending on rainfall intensity. Weak rain may, in large portion, consist of small raindrops which have large surface area per unit volume and more chance to get NO_3^- in the air than the large drops. Regardless of the intensity, however, the concentration of rain water is in the range between 0.0 and 0.3 mg/l which is

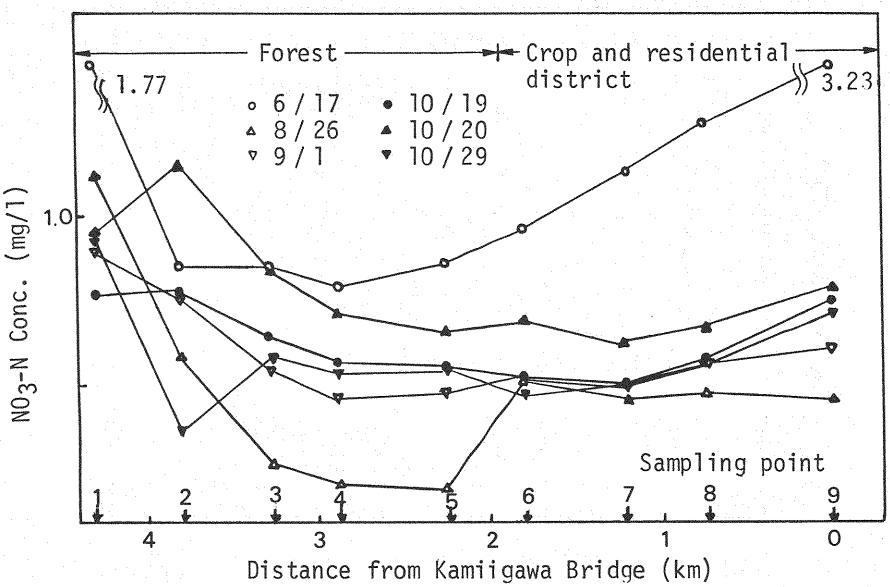

Fig.3 Spatial distribution of NO_3-N concentration of river waters along the Ai River.

significantly lower than that of river water.

3. NO_3^- in well and spring water

To estimate the NO_3-N concentration of groundwater, water samples were collected from four springs and ten domestic wells. The springs are in the upstream area of station No.1 and the origins of Ai River. The wells are located near by the study area and none is operating now for daily domestic use.

The observed NO_3-N concentration of four springs was 0.83, 0.87, 0.87 and 1.28 mg/l, a little higher than that of the river water. But they decreased to 0.23 or 0.57 mg/l as they trickled few hundred meters downstream from the springs. Trickling water may be easily affected by microbial absorption in the river bed. The NO_3-N concentration of domestic wells widely differed from 0.45 to 10.0 mg/l depending on their maintenance status and inner vegetation. Thus, they do not provide the information of typical NO_3-N concentration of groundwater.

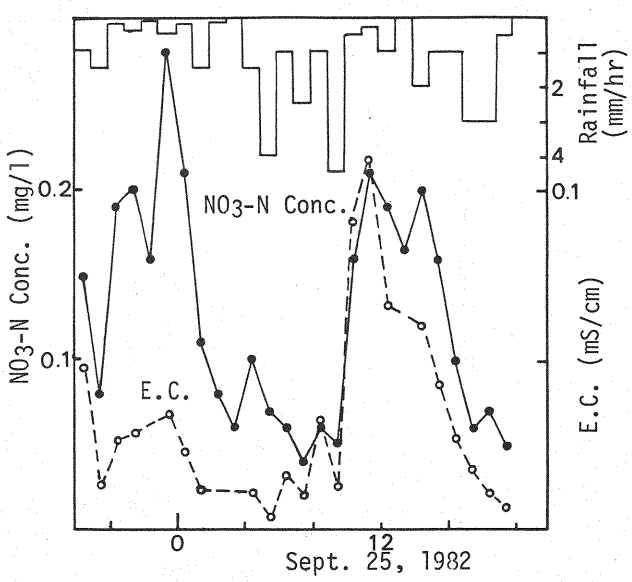

Fig.4 Rainfall intensity and corresponding NO_3-N concentration and electric conductivity of rain water at Rain No.4.

In order to get the representative NO_3-N concentration of groundwater, samples from well-managed wells for municipal water-supply were examined. Their NO_3-N concentration was between 1.7 and 3.1 mg/l, and was 2.3 mg/l on an average. This value was even higher than that of the river water at rain. Because none seems affecting underground to increase the NO_3-N concentration, this difference might be introduced in the river water during flowing down. The magnitudes may depend on their flow routes reflecting the basin topography and geology.

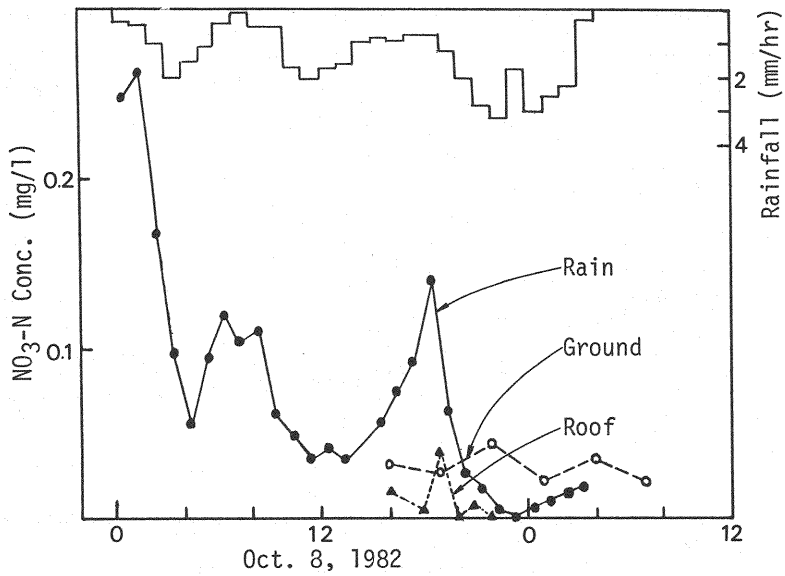

Fig.5 Rainfall intensity and NO_3-N concentration of rain water at Rain No.5, as compared with NO_3-N concentrations of surface discharges from the sport ground and from the roof.

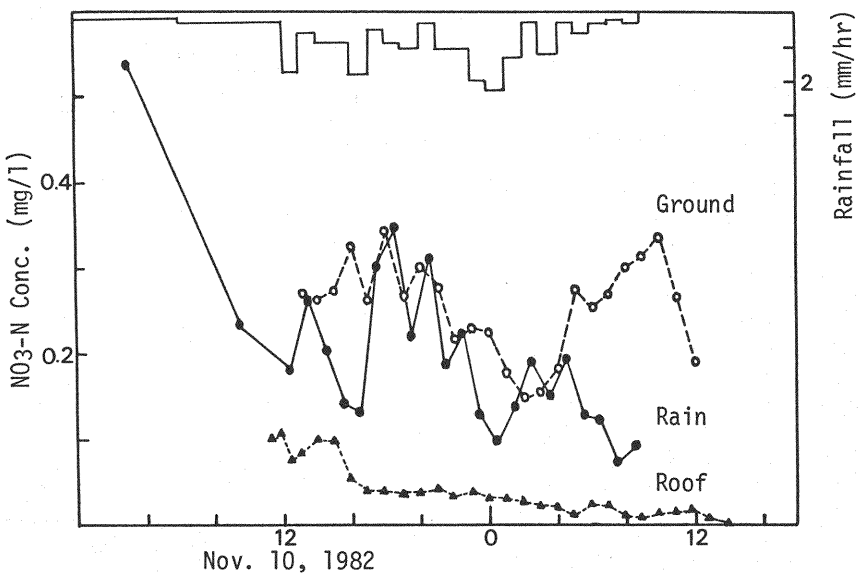

Fig.6 Rainfall intensity and NO_3-N concentration of rain water at Rain No.6, as compared with NO_3-N concentrations of surface discharges from the sport ground and from the roof.

4. NO_3^- in surface flow water

It was not possible to collect the actual surface flow water in forest region. But the overland flow on a bare sport ground in the Yamanashi University campus and drain water from the roof were sampled as the examples of surface flow water. Results are shown in Figs.5 and 6 compared with rain water. Water from the bare sport ground, which easily produces excessive rainfall, was recognized to have the NO_3-N range similar to rain water, although its changing pattern was different. In contrast, waters from the roof strangely showed lower NO_3-N concentration than rain. Because the samples were collected after they flew through a 15 m water

drain pipe, its quality might be affected physicochemically and biochemically on the wall of the pipe. This phenomenon may be similar to that observed in the upstream trickling water near the springs. Thus, the NO_3^- content in water flowing over the ground surface as thin film flow is concluded as low as that of rain water which is much lower than water flowing through the soil.

5. NO_3^- in subsurface flow water

Subsurface water exuding from the bottom of a slope of 7 m length in the Yamanashi University was analyzed as an example of subsurface flow water, because actual subsurface flow was not available. The slope gradient is 1 / 1.3 and is covered by weeds of several centimeters height. Fig.7 shows NO_3-N concentration change caused by a 43.5 mm rainfall with average NO_3-N concentration 0.006 mg/l . In Fig.7, a broken line expresses the change during rain and a solid line that after rain. After the rain, the NO_3-N concentration of exuding water showed exponential decrease for about 8 hours. This exponential decrease might be characteristic to subsurface flow reflecting the decrease of NO_3-N stock in the soil which was continuously washed out by percolating water.

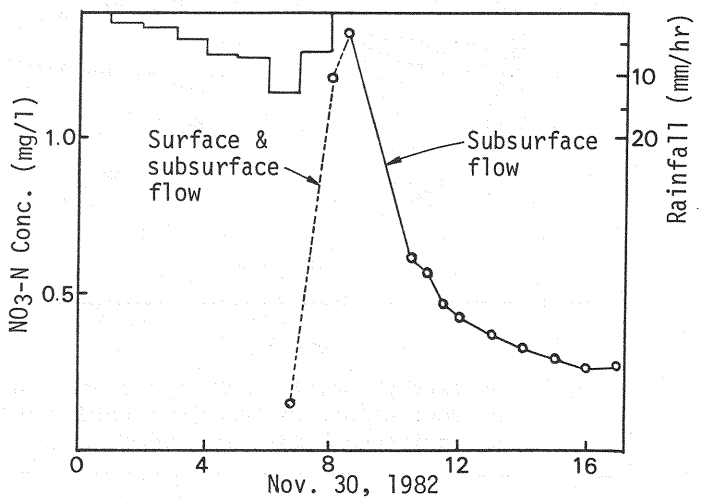

Fig.7 Temporal changes of NO_3-N concentration of water exuding from the slope covered by weeds.

To test the effect of wash-out and microbiological formation of NO_3^- in the soil, soil samples on the slope were collected and their NO_3-N contents were determined immediately after rain and 5 days later. The NO_3-N content was found to increase from 0.17 to 0.82 mg/kg-dry soil for 5 days, although sample variation was large. This indicates that NO_3^- is formed on clear days and washed out on rainy days. For the soil of bare sport ground, this change was only 0.07 to 0.12 mg/kg-dry soil , significantly smaller than that of the grass covered slope. This shows the importance of vegetation for NO_3^- formation.

6. NO_3^- in storm water

Storm water can be considered as a mixture of several flow components. Because the proportion of each component differs not only during a flood but also flood by flood depending on rainfall, basin characteristics and antecedent rainfalls, the NO_3-N concentration of total flow also differs intra and inter floods widely. This might reversely indicate that through examination of NO_3-N concentration changes of storm water, one could extract useful information about the change of the ratios of flow components. From this point of view, NO_3-N concentration and other water quality indices such as electric conductivity (E.C.) of storm water were examined in various rain events.

Figs.8.a-f show the rainfall, flow rate and the corresponding quality changes of river water at rains No.1 - 3 and 7 - 11. In Figs.8.a-d, the NO_3-N concentration shows the peaks several hours after the flow peak, while E.C. reaches to the minimal value at the same time of the flow peak. This observation has already been reported by Ebise et al. (1, 2 and 3) and explained as the result of time lag between the subsurface flow component and the surface flow component. A closer look at the figures reveals further implication.

In Fig.8.c, the 16 mm rainfall did not cause a remarkable peak of NO_3-N

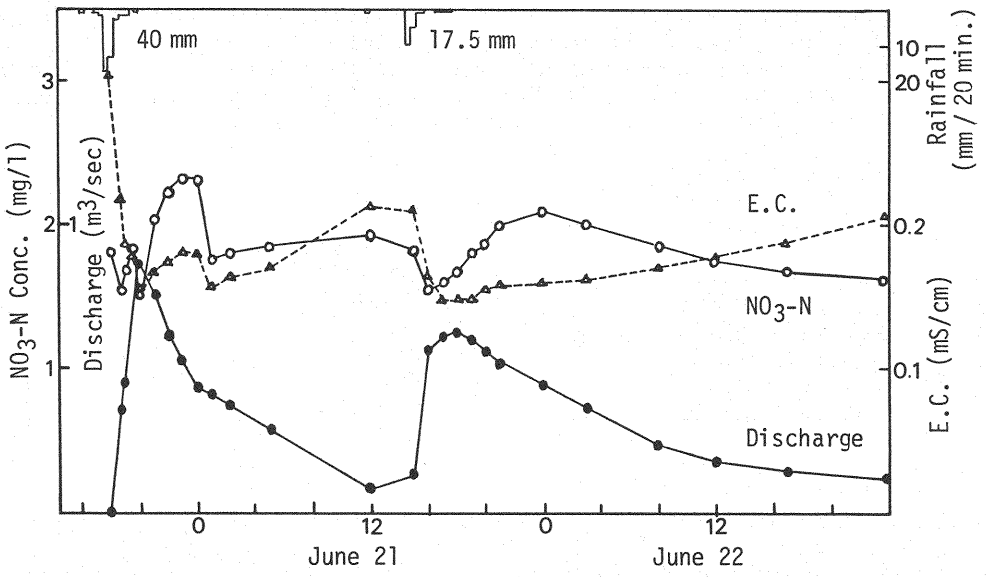

Fig.8.a Hyetograph, hydrograph and corresponding changes of NO_3-N concentration, and electric conductivity of river water at Kamiigawa Bridge at Rain No.1 and 2.

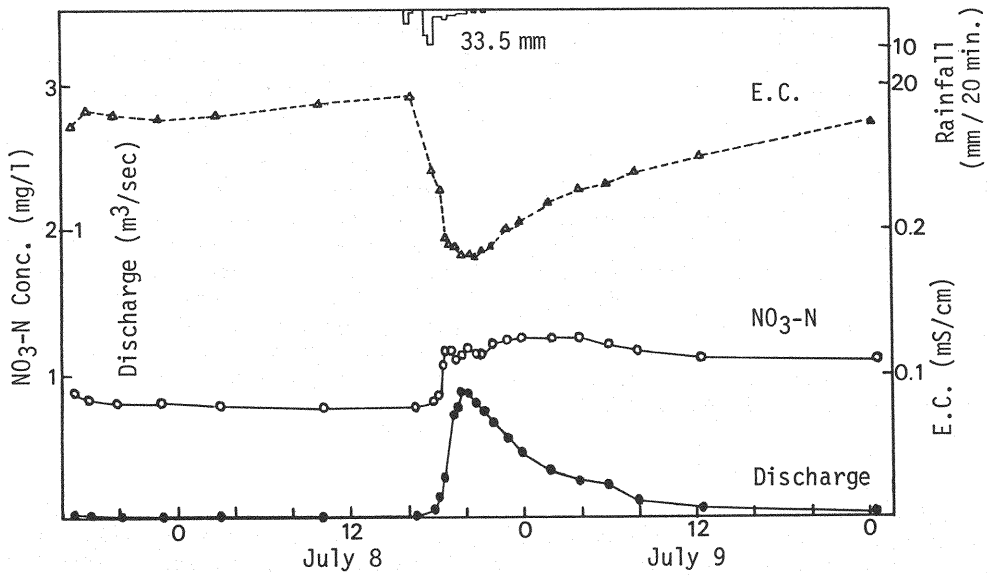

Fig.8.b Hyetograph, hydrograph and corresponding changes of NO_3-N concentration, and electric conductivity of river water at Kamiigawa Bridge at Rain No.3.

concentration, and the time lag between the NO_3^- and flow peaks was also ambiguous as compared with similar rainfalls such as the 17.5 mm rainfall of Fig.8.a and the 16 mm rainfall of Fig.8.d. Ambiguous peak and time lag were observed also in the 33.5 mm rainfall of Fig.8.b. Both rains in Figs.8.b and c had little antecedent rainfall and weak average intensity. It seems that the intensive rainfall and large antecedent rainfall create the clear NO_3^- peaks and the distinct time delay of NO3-N concentration from the flow peak. This is probably because such rain generates intensive surface runoff which dilutes the NO_3^- rich subsurface flow near the flow peak.

In contrast, the long and intensive rainfalls brought by typhoons resulted in

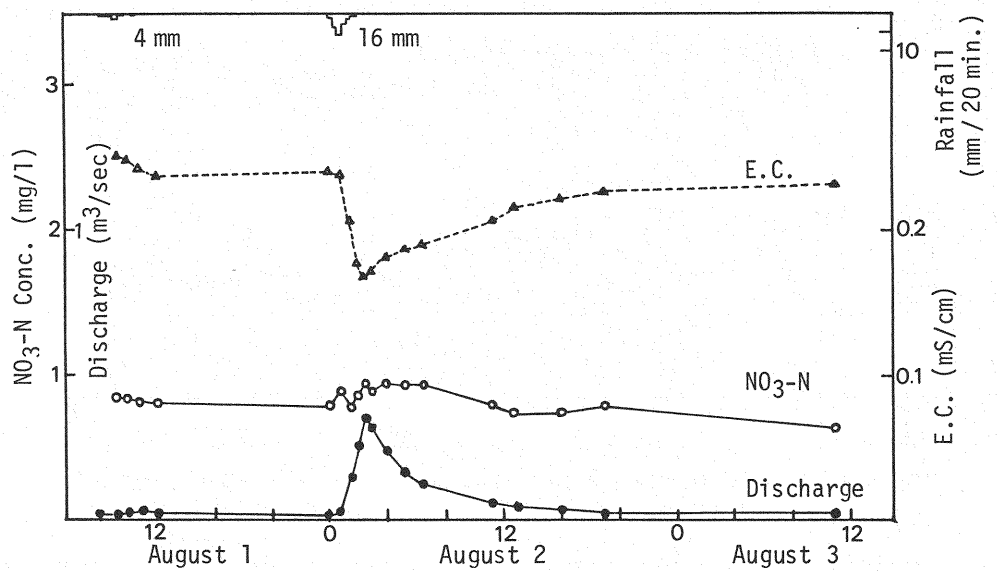

Fig.8.c Hyetograph, hydrograph and corresponding changes of NO_3-N concentration, and electric conductivity of river water at Kamiigawa Bridge at Rain No.7.

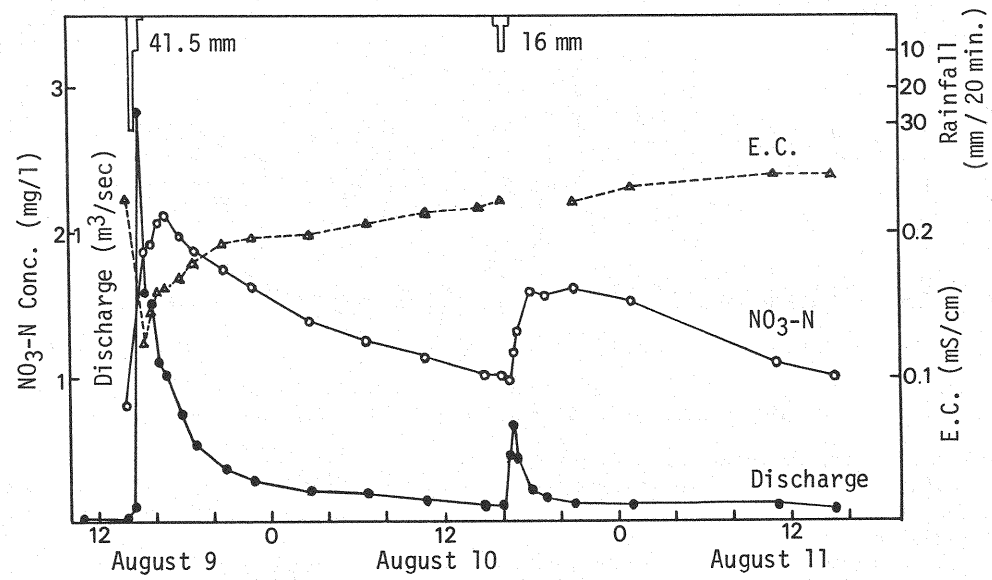

Fig.8.d Hyetograph, hydrograph and corresponding changes of NO_3-N concentration, and electric conductivity of river water at Kamiigawa Bridge at Rain No.8 and 9.

different patterns of NO_3-N concentration changes as shown in Figs.8.e and f. Fig.8.e is the case of typhoon on August 15 - 16, 1983. The gradual decrease of NO_3-N concentration is more prominent in e than in f, accompanying several humps at flow peaks. Because the stock of NO_3^- in the soil is subject to wash-out by percolating water, the increase of NO_3-N at rain is larger after a long absence from former rains, and small with consecutive rainfalls. In Fig.8.f of the typhoon on September 27 - 28, 1983, the NO_3-N concentration increased when the first low peak of flow came along and did not change much afterwards until the second higher flow peak arrived. At the second peak, the rain caused the surface flow that contained a large amount of suspended soil particles as seen in the SS change. This suspended soil particles would have released much NO_3^-, so that the high NO_3-N

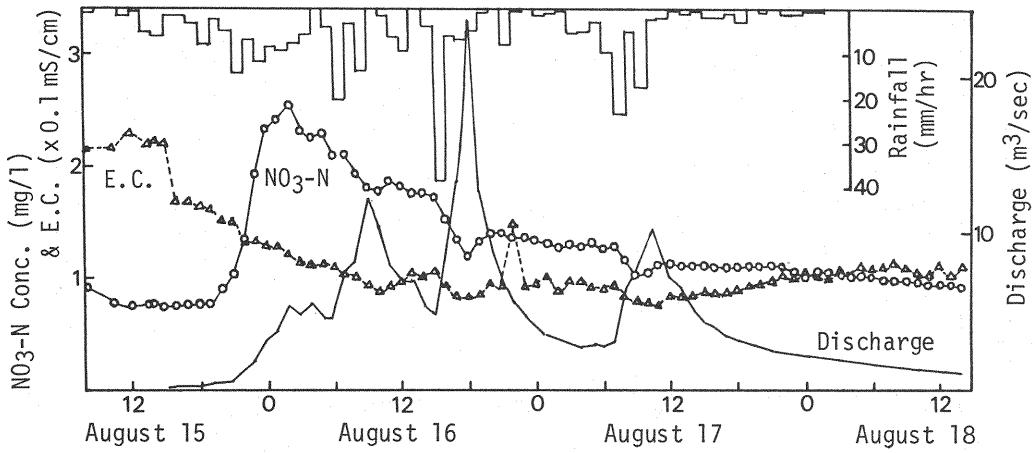

Fig.8.e Hyetograph, hydrograph and corresponding changes of NO_3-N concentration, and electric conductivity of river water at Kamiigawa Bridge at Rain No.10 caused by typhoon.

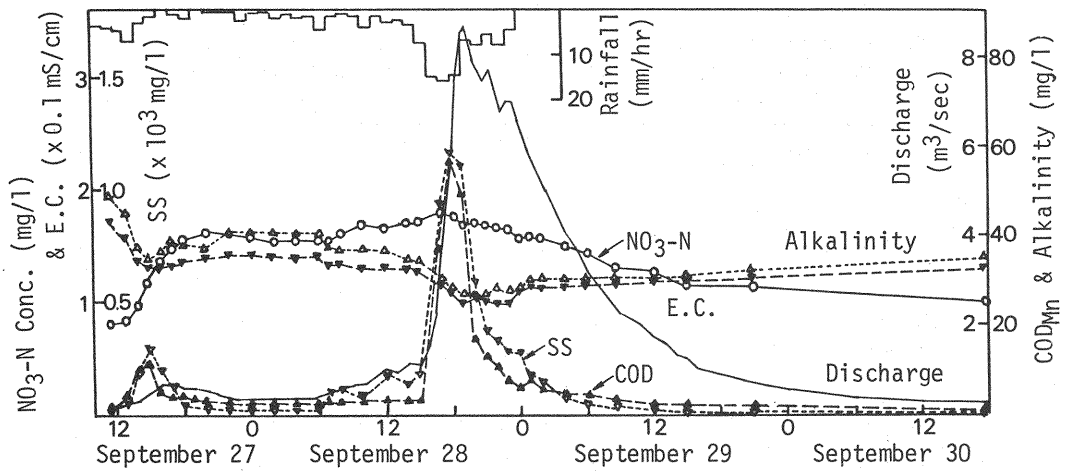

Fig.8.f Hyetograph, hydrograph and corresponding changes of river water quality at Kamiigawa Bridge at Rain No.11 caused by typhoon.

concentration was observed in this peak.

To examine the flow rate together with water quality, the NO_3-N load and the total ion load calculated from E.C. value were plotted against the flow rate in the logarithmic paper as seen in Figs.9.a , b, Figs.10.a , b and Figs.11.a , b. For the typical single rain, the load-flow relationship is more straightforward for the total ion than for NO_3-N as shown in Figs.9.a and b. However, this is not clear for two sequential rains as seen in Figs.10.a and b. For heavy and long-duration rains like the one brought by typhoon, the relationship with NO_3-N load is complexed as compared with that with the total ion load as seen in Figs.11.a and b.

To compare characteristics of contaminants in water as tracers of flow components, SS, COD_{Mn} and alkalinity were also determined at Rain No.11. Results are shown in Fig.8.f. SS and COD_{Mn} reached to the maximum at early stage of runoff. They seem mainly brought by surface flow. But they are not suitable as quantitative tracers of surface flow because the amount of small particles which determine SS and COD_{Mn} are influenced not only by the volume of discharge but also by the velocity of flow. Alkalinity showed the pattern similar to E.C. Alkalinity is known to be mainly dependent on CO_3^{2-} and HCO_3^- which are rich in groundwater, and is considered as a more comprehensive index of chemical quality of water than E.C. But alkalinity changes in a short time after sampling so that it may not be reliable as a tracer.

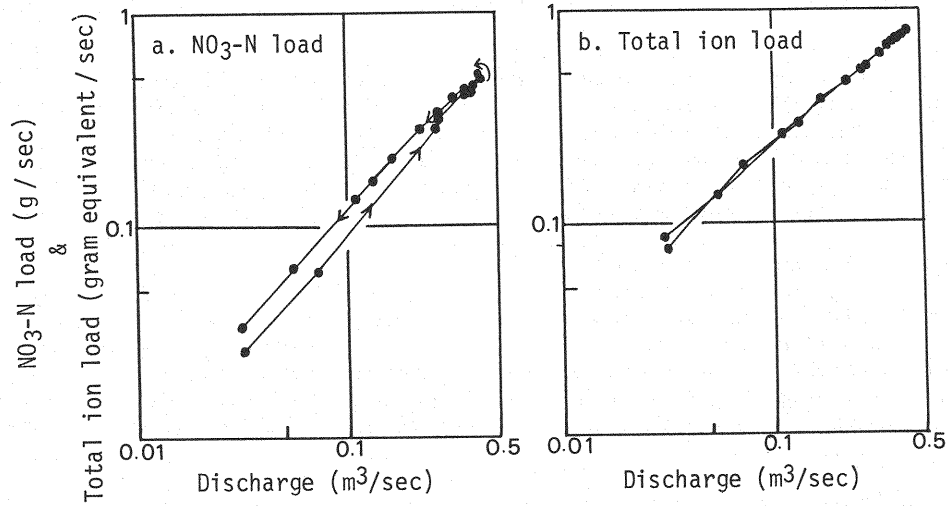

Fig.9 NO_3-N and total ion load vs. discharge at Rain No.3.

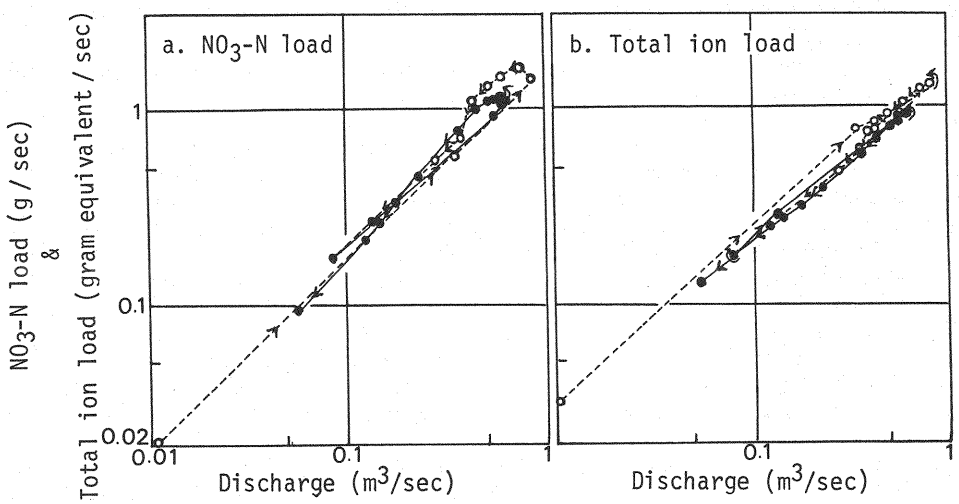

Fig.10 NO_3-N and total ion load vs. discharge at Rain No.2.

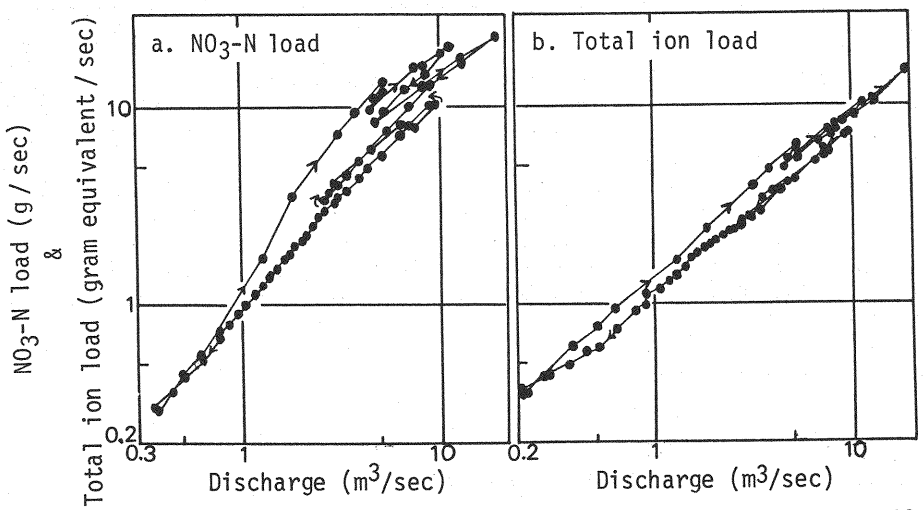

Fig.11 NO_3-N and total ion load vs. discharge at Rain No.10.

CONCLUSIONS

Major observation results are as follows:

1. The NO_3-N concentration of river water on clear days assumed the minimum value about 0.5 mg/l in August, gradually increased through autumn and winter to 0.9 mg/l . Then it rapidly increased in the farming season to the maximum value of 2.5 mg/l in June when paddy fields were flooded. Spatial change was conceived primarily as a result of human activities along the river, especially agricultural activities. After intensive rainfalls such as typhoons, the NO_3-N concentration in river water showed a prominent decrease probably because the NO_3-N stock in the soil was considerably exhausted by wash-out.

2. The NO_3-N concentration of spring waters was 0.87 mg/l on an average, higher than river water but found soon decrease as they trickled downward few hundred meters along the river. The reason is yet uncertain. NO_3-N in domestic well waters widely ranged depending on their running conditions and inner vegetation. The average NO_3-N concentration of waters of well-managed wells was found 2.3 mg/l , which was larger than that of storm runoff. This difference may be caused by the same reason as the difference between the spring and the river waters.

3. The NO_3-N concentration of rain water was highest at the beginning of rainfall and decreased as the rainfall continued, with superposition of oscillatory behavior reciprocal to the rainfall intensity. The range was from 0.0 to 0.3 mg/l lower than that of river water.

4. The NO_3-N concentration of non-infiltrated surface water from impervious area was found similar to that of rain water. The water running in the drain pipe from the roof top showed even lower concentration than the rain water.

5. The NO_3-N concentration of the subsurface runoff from 7 m slope covered with weeds gradually decreased in an exponential form after rain stopped. The NO_3-N content of the soil 5 days later was, however, found increased again by 0.65 mg/kg-dry soil from the level immediately after the rain. The increase was only 0.05 mg/kg-dry soil in the bare sport ground.

6. The NO_3-N concentration of single storm water usually showed its peak few hours later than the flow rate peak but this tendency largely depended on rainfall conditions and antecedent rainfalls. The NO_3-N peaks of storm waters by sequential rains were larger with more antecedent rainfalls. The NO_3-N concentration seems to reflect the subsurface flow rate change, while SS and COD_{Mn} reflect surface flow rate, and alkalinity and E.C. groundwater rate.

Those findings 1. through 6. clearly indicate that the high content of NO_3-N load of river water during floods is not brought by rain or surface discharge, but by the discharge once infiltrated to the ground and passed through the humus layer which is the sole source of NO_3^- supply and may coincide with so called A-horizon in pedology.

The discharge components are often classified into surface flow, subsurface flow and groundwater. The distinction of these components is not necessarily clear nor common among hydrologists. In this paper, the surface flow is defined as the water discharged to the river without experiencing significant contact with the humus soil on the thin layer of ground surface. It consists of the discharge from the roads, roofs, hard ground surface and the like. The subsurface flow or interflow is the discharge once infiltrated into the ground, but discharged before reaching to the aquifer, and the groundwater is the discharge from the aquifer.

Let those components be indicated by the suffixes S, I and G and the total discharge by T, then their NO_3-N concentrations (mg/l) are indicated by $C_T(t)$, $C_S(t)$, $C_I(t)$ and $C_G(t)$, respectively. Since the river discharge at time t, $Q_T(t)$ (m^3/sec), is the sum of three components, $Q_S(t)$, $Q_I(t)$ and $Q_G(t)$, the continuity relations of water mass and NO_3-N load are

$$Q_S(t) + Q_I(t) + Q_G(t) = Q_T(t) \tag{1}$$

$$C_S(t) \cdot Q_S(t) + C_I(t) \cdot Q_I(t) + C_G(t) \cdot Q_G(t) = C_T(t) \cdot Q_T(t) \tag{2}$$

Among the eight variables of these equations, only $Q_T(t)$ and $C_T(t)$ are observable but no others. The finding 4. suggests that $C_S(t)$ may be considered nearly equal to the average value of $C_R(t)$ of rain which is observable. But yet five variables are unknown as against two equations. Thus no straightforward method can separate and identify the three discharge components. Among various possible simplification procedures to get through this problem, the following two may be worth considering:

1) $C_I(t)$ may be exogenously given as a function of the initial condition of the ground surface, i.e., the condition of NO_3-N sources and a time dependent reducing factor such as $\exp(-\lambda t)$.

2) $Q_G(t)$ may be identified separately by different approaches such as using ^3H and other radio isotopes or other chemical factors like electric conductivity. The mathematical and statistical approaches may also be useful for this purpose.

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APPENDIX - NOTATION

The following symbols are used in this paper:

t = time;

$C_G(t)$ = NO_3-N concentration of groundwater at time t;

$C_I(t)$ = NO_3-N concentration of subsurface flow water at time t;

$C_R(t)$ = NO_3-N concentration of rain water at time t;

$C_S(t)$ = NO_3-N concentration of surface flow water at time t;

$C_T(t)$ = NO_3-N concentration of total flow water at time t;

$Q_G(t)$ = groundwater flow rate at time t;

$Q_I(t)$ = subsurface flow rate at time t;

$Q_S(t)$ = surface flow rate at time t; and

$Q_T(t)$ = total flow rate at time t.