Estimation of particulate nutrient load using turbidity meter

K. Yamamoto* and T. Suetsugi**

*Ariake Sea Research Project, Saga University, Honjo 1, Saga-city, Saga 840-8502, Japan
(E-mail: *ko1yama@attglobal.net*)

**National Institute for Land and Infrastructure Management, 1 Asahi, Tsukuba, Ibaraki 305-0804, Japan
(E-mail: *suetsugi-t92fv@nilim.go.jp*)

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Abstract The "Nutrient Load Hysteresis Coefficient" was proposed to evaluate the hysteresis of the nutrient loads to flow rate quantitatively. This could classify the runoff patterns of nutrient load into 15 patterns. Linear relationships between the turbidity and the concentrations of particulate nutrients were observed. It was clarified that the linearity was caused by the influence of the particle size on turbidity output and accumulation of nutrients on smaller particles (diameter $< 23\,\mu m$). The *L-Q-Turb* method, which is a new method for the estimation of runoff loads of nutrients using a regression curve between the turbidity and the concentrations of particulate nutrients, was developed. This method could raise the precision of the estimation of nutrient loads even if they had strong hysteresis to flow rate. For example, as for the runoff load of total phosphorus load on flood events in a total of eight cases, the averaged error of estimation of total phosphorus load by the *L-Q-Turb* method was 11%, whereas the averaged estimation error by the regression curve between flow rate and nutrient load was 28%.

Keywords *L-Q* equation; *L-Q*-Turbidity method; nutrient load; turbidity

Introduction

To control and model water quality in lakes and other closed water areas in relation to eutrophication, it is important to accurately estimate the total input of nutrients and other chemical components from their basin. It is important that nutrient loads from the nonpoint sources are often discharged in the storm period and occupy over 50% of total discharge for a year (Tachibana *et al.*, 2001). For measuring the nutrient loads in the storm period, it is often the case that water samples are collected over 24 times per day. Therefore, it is almost impossible to measure the discharge of nutrients from the basin in every storm. That is why the relationships of nutrient load and flow rate such as the *L-Q* regression curve were developed and adapted to many rivers. However, the *L-Q* regression curve has difficulty in estimating the nutrient loads when they do not depend on the river flow rate. For example, rapid increase of nutrient loads by the suspension of mud by human activity or inflow of turbid water in ordinary water level period cannot be estimated by the *L-Q* equation. At present, *in situ* automatic analyzers of nutrient concentration are developed and released by manufacturers. It is ideal to measure the nutrient concentration for hours, but it is difficult to install them in all rivers due to its cost. The major reason for the fluctuation of total nutrient concentrations in storm periods is the fluctuation of particulate nutrient concentration. Therefore, if we could measure the particulate nutrient concentrations by any method, the estimation of total nutrient load would be more accurate. We studied measuring particulate nutrient concentrations by turbidity meter and presented a new estimation method for nutrient loads.

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Methods

Study field, observed period and chemical analysis

We studied nine time series of storm discharge in six rivers in Japan (Table 1). Flow rate and turbidity were measured *in situ*. We collected water samples and analyzed particle size distribution by laser scattered particle analyzer (SALD-3000, Shimadzu Co., Ltd.), Total Nitrogen (TN), Dissolved Nitrogen (DN), nitrate-nitrogen (NO_3^--N), nitrite-nitrogen (NO_2^--N), ammonium-nitrogen (NH_4^+-N), Total Phosphorus (TP), Dissolved Total Phosphorus (DP) and Dissolved Reactive Phosphorus (DRP) by auto analyzer (TRAACS, BRAN + LUEBBE) and Total Organic Carbon (TOC) and Dissolved Total Organic Carbon (DOC) by TOC meter (TOC-5000A, Shimadzu Co., Ltd.). Particulate components were calculated by the difference between the concentrations of total components and dissolved components.

Instruments

A turbidity meter (ATU 5-8M, Alec Electronics Co., Ltd.) was installed in the river. We set the turbidity meter 50 cm above the riverbed. The type of the turbidity meter was that of measuring backscatter of infrared (wavelength = 880 nm in air, and 660 nm in water) from suspended solid. A portable turbidity meter (AAQ-1183, Alec Electronics Co., Ltd.) was also deployed to measure the turbidity of the collected water. To measure the turbidity of the collected water, water samples were filled into the matte-black bucket.

Nutrient Load Hysteresis Coefficient (NLHC)

In the storm period, the loads of chemical components fluctuate with flow rate. In many cases, the load of chemical components increases with the increase of flow rate.

It is recognized that particulate component loads often loop in the plot of flow rate and load. Higher concentration of particulate components was often observed in the phase of increasing rather than decreasing, although their flow rates were the same. Figure 1 shows the dependence and hysteresis of the nutrient load to the flow rate. Though NO_3^--N load could be expressed as a one-valued function of flow rate, particulate phosphorus load could not. Therefore, we proposed the hysteresis coefficient of load of chemical components, namely Nutrient Load Hysteresis Coefficient (NLHC), to evaluate the hysteresis of the nutrient load to flow rate as follows.

When

$$\sum_{k=1}^{n-1}(L_k + L_{k+1})(Q_{k+1} - Q_k) \geq 0 \tag{1}$$

then the hysteresis coefficient H is defined as

$$H = \frac{\sum_{k=1}^{n-1}(L_k + L_{k+1})(Q_{k+1} - Q_k)}{\sum_{k=1}^{M-1}(L_k + L_{k+1})(Q_{k+1} - Q_k)} \tag{2}$$

and when

$$\sum_{k=1}^{n-1}(L_k + L_{k+1})(Q_{k+1} - Q_k) < 0 \tag{3}$$

then the hysteresis coefficient H is defined as

$$H = -\frac{\sum_{k=1}^{n-1}(L_k + L_{k+1})(Q_{k+1} - Q_k)}{\sum_{k=M}^{n-1}(L_k + L_{k+1})(Q_{k+1} - Q_k)} \tag{4}$$

Table 1 Sampling stations and sampling periods

Sampling station	River	Watershed (km^2)	Land use	Sampling period	Rainfall (mm)
28.1 KP	Hinuma Riv.	190	Paddy field, upland field	12–13 Sep. 2002	21
				30 Sep.–2 Oct. 2002	80
Kataniwa-A	Kataniwa Riv.	1.1	Forest	10–11 July 2002	105
				30 Sep.–2 Oct. 2002	80
Kataniwa-B	Kataniwa Riv.	3.4	Forest, quarry	10–11 July 2002	105
				30 Sep.–2 Oct. 2002	80
Inadasawa	Inadasawa Riv.	4.5	Paddy field, quarry	30 Sep.–2 Oct. 2002	80
Maekawa	Hinuma Mae Riv.	80	Paddy field, upland field	30 Sep.–2 Oct. 2002	80
Yotsugi	Shira Riv.	480	Paddy field, upland field, forest	24–25 June 2002	127

where L_k expresses the load of the chemical component of sampling number k, Q_k expresses the flow rate of the river at the sample k, and n means the total number of samples + 1 (Figure 2).

The value of the absolute H means the magnitude of the hysteresis, which is the fraction of S_1 to $(S_1 + S_2)$ in Figure 3. The maximum value of the H is one. Positive H means that the increasing rate of the load exceeds that of flow rate of the river, and the decreasing rate of the load exceeds that of flow rate, and clockwise rotation as shown in Figure 3. On the other hand, negative H means that the decreasing rate of the load exceeds that of flow rate of the river, and decreasing rate of the load exceeds that of flow rate, and anticlockwise rotation as shown in Figure 3. If absolute H is nearly equal to zero, it means that there is less hysteresis about flow rate and load. To calculate H, the curve needs to be closed. Therefore, L_1 and Q_1 are given as L_n and Q_n for the sake of simplicity.

Results and discussion

Concentration of chemical components in the storm period

Flow weighted averaged concentrations of chemical components in river water are shown in Table 2. SS concentration was up to 1,000 mg/L at the river whose basins have quarries. Over 80% of TOC, 90% of TP, and 30% of TN were particulate. Therefore, accurate estimation of concentration of particulate components will lead to accurate estimation of total discharged load.

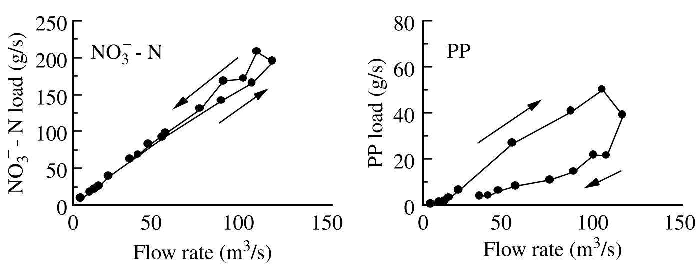

Figure 1 Relationships between nutrient loads L and flow rate Q, observed in the storm period, 1–2 Oct., 2002 at 28.1 KP in the Hinuma River, NO_3^--N (left), particulate phosphorus (right)

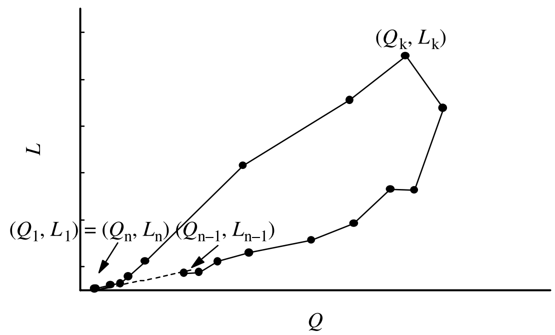

Figure 2 Example of the relationships between flow rate Q and nutrient load L; for the sake of simplicity, Q_1 and L_1 are given as Q_n and L_n

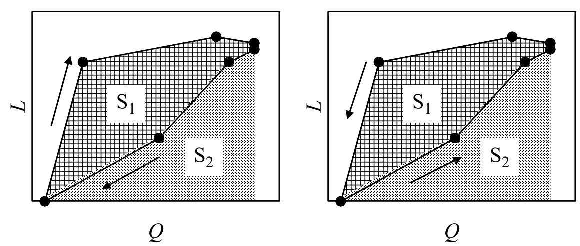

Figure 3 Two patterns of the fluctuation of nutrient load L with flow rate Q; clockwise rotation type (left) and anticlockwise rotation type (right)

Classification of characteristics of discharged nutrient loads in the storm period

Modeling of the discharged load L by the power function of flow rate Q is widely used, as below

$$L = CQ^n \tag{5}$$

where C and n are constants. It is recognized that if $n > 1$, concentration rises with the flow rate (Increasing type; I), and if $n < 1$, concentration drops with flow rate (Decreasing type; D). If value of n is nearly equal to 1, it means that the concentration is constant irrespective of flow rate (Constant type; C). Moreover, using the NLHC, we could classify the patterns of nutrient discharged load into 15 patterns as shown in Table 3. The results of the classification of the characteristics of fluctuation of the discharged load are shown in Table 4. Turbidity, SS, PN, PP, TP, TOC and POC loads have large value of n and H, and are classified into I^{++}. It is clear that particulate components have quite strong hysteresis and their concentrations rise with increasing flow rate. DP load also has strong hysteresis despite the concentration tending to be constant. The groups of + or ++ are the groups of the components which are supplied from the watershed immediately with increasing flow rate. Though DP has a strong hysteresis, the proportion of it in TP is negligible in the storm period. On the other hand, inorganic nitrogen compounds and DOC loads have less hysteresis and can be modeled with the function of the flow rate. It is clear that the load of the components classified into I^{++} group can hardly be modeled with flow rate.

Estimation of particulate components by turbidity

Effect of the particle size distribution on output of the sensor of the turbidity meter. The type of the turbidity meter we used is measuring the strength of the backscatter of light

Table 2 Flow weighted averaged concentration of chemical components in river water in the storm period in Table 1

St. Unit	Period	*n*	Turb. mg/L	SS mg/L	PN mg/L	DN mg/L	NO_3^--N mg/L	NO_2^--N µg/L	NH_4^+-N µg/L	PP µg/L	DP µg/L	DRP µg/L	POC mg/L	DOC mg/L
28.1 KP	Sep.02	12	390	419	1.24	2.07	1.85	1	11	491	19	16	11.6	1.4
	Oct. 02	19	273	258	0.68	1.93	1.76	2	2	247	19	18	7.9	2.2
Kataniwa-A	Jul. 02	8	359	392	1.11	1.17	0.90	1	1	179	1	1	33.3	1.6
	Oct. 02	16	98	90	0.47	1.17	1.08	1	1	52	1	1	6.6	1.8
Kataniwa-B	Jul. 02	17	813	1,498	1.05	0.92	0.81	1	1	479	1	1	19.5	0.6
	Oct. 02	15	640	1,188	0.62	1.00	0.96	1	1	287	3	1	9.9	1.0
Inadasawa	Oct. 02	12	1,614	2,444	1.18	0.54	0.43	3	3	311	2	1	26.5	2.0
Maekawa	Oct. 02	15	299	377	1.43	4.69	4.28	5	5	549	43	39	16.1	6.7
Yotsugi	Jun. 02	27	156	202	0.74	1.43	1.27	2	2	444	45	37	7.2	0.7

Table 3 Classification of runoff load of nutrients L by modeling with $L = CQ^n$ and Nutrient Load Hysteresis Coefficient H

	$n < 0.9$	$0.9 \le n \le 1.1$	$1.1 < n$
$0.25 < H$	D^{++}	C^{++}	I^{++}
$0.1 < H \le 0.25$	D^{+}	C^{+}	I^{+}
$-0.1 \le H \le 0.1$	D	C	I
$-0.25 < H \le -0.1$	D^{-}	C^{-}	I^{-}
$H < -0.25$	D^{-}	C^{-}	I^{-}

(wavelength = 880 nm in air). Therefore, the output of the turbidity meter is affected by the particle size distribution of the SS in water. Turbidity (*Turb.*) and SS concentration can be expressed as

$$Turb = a \cdot SS^b \tag{6}$$

where a and b are constants. The output of the turbidity is mainly controlled by the concentration of the fine fraction ($d < 20\,\mu m$) of the SS (Yokoyama, 2002). The fraction of coarse sand does not affect the output of the turbidity meter. If we use the same particle size of SS, b in Eq. (6) will be close to 1 if the fine particle dominates in SS.

Relationship between turbidity and particulate nutrient concentration. In general, the correlation coefficient of SS concentration and turbidity is high and SS concentration can be estimated by turbidity meter with proper calibration. We found that the correlation coefficients of particulate nutrients and POC were also high. An example of the relationships between particulate nutrient concentrations and turbidity is shown in Figure 4. The relationships between particulate nutrient concentrations and SS concentration are also shown in Figure 4. PN concentration and PP concentration could be expressed as the power functions of the SS concentration and turbidity as follows:

$$C = \alpha \cdot Turb^m \tag{7}$$

$$C = \beta \cdot SSC^n \tag{8}$$

Table 4 Results of classification of runoff load of nutrients L by modeling with power number n in the $L = CQ^n$ and Nutrient Load Hysteresis Coefficient H of the time series in Table 1

Components	n				H				Classification
	Max.	Av.	Min.	S.D.	Max.	Av.	Min.	S.D.	
Turbidity	2.13	1.67	1.18	0.25	0.86	0.46	0.00	0.29	I^{++}
SS	2.29	1.83	1.05	0.34	0.79	0.51	0.11	0.27	I^{++}
NH_4^+-N	1.33	1.05	1.00	0.10	0.61	−0.03	−0.38	0.28	C
NO_2^--N	1.61	1.16	1.00	0.22	0.27	−0.02	−0.66	0.28	I
NO_3^--N	1.37	1.04	0.88	0.21	0.13	−0.12	−0.30	0.15	C^{-}
DN	1.33	1.04	0.87	0.18	0.14	−0.10	−0.28	0.14	C
TN	1.40	1.20	1.06	0.11	0.31	0.17	0.02	0.10	I^{+}
PN	2.07	1.57	1.04	0.28	0.70	0.43	−0.01	0.27	I^{++}
DRP	1.19	0.89	0.21	0.28	0.73	0.16	−0.05	0.25	D^{+}
DP	1.38	0.91	0.35	0.30	0.96	0.33	−0.16	0.38	C^{++}
TP	1.91	1.51	0.91	0.26	0.77	0.51	0.07	0.26	I^{++}
PP	1.94	1.59	0.92	0.30	0.77	0.50	−0.04	0.29	I^{++}
TOC	2.04	1.62	1.04	0.29	0.81	0.34	−0.40	0.42	I^{++}
DOC	1.67	1.14	0.95	0.21	0.66	−0.01	−0.80	0.54	I
POC	2.24	1.75	1.06	0.32	0.84	0.39	−0.40	0.44	I^{++}

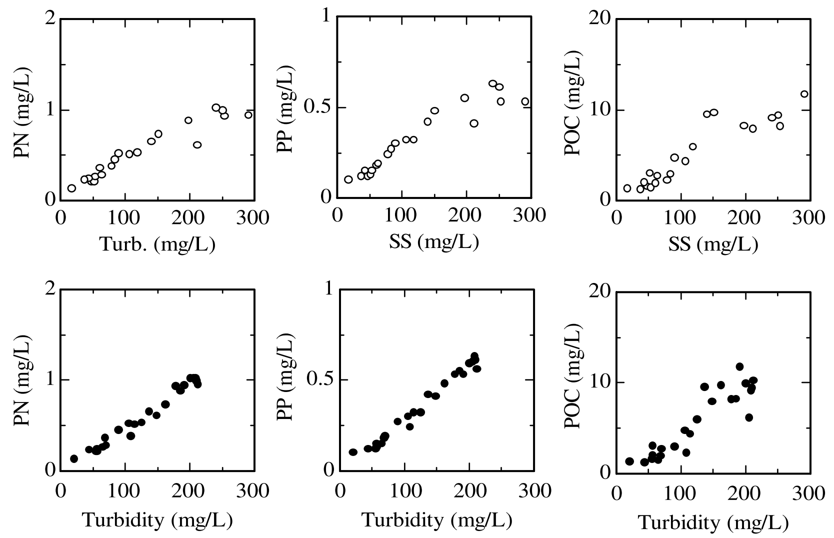

Figure 4 The relationships between turbidity, SS and particulate components on 24–25 June, 2002, at the Yotsugi Bridge of the Shira River

where C is concentration of particulate components, *SSC* is SS concentration, and *Turb* is turbidity. α, β, m, and n are constants. Averaged n and m for SS, PN, PP, POC are shown in Table 5. The values of the power coefficients of the n are lower than the values of m and 1. This means that higher concentration of SS leads to lower values of PN/SS, PP/SS, POC/SS. On the other hand, the values of m are more close to 1. Particularly, PP concentration can be expressed by the linear function of turbidity. The correlation coefficient of PN and POC is lower than that of PP.

Particulate distribution and the fraction of particulate nutrients. Particulate phosphorus exists as a component of the body of organisms and organic debris, or it is adsorbed on the surface of minerals or hydroxides. In the case of being adsorbed on the surface of minerals, it is thought that the amount of the adsorbed phosphorus increases with the specific surface area. Therefore, it is assumed that warse materials contained lower concentration. Typical particle size distribution of SS in the river water, which was collected from the surface water of the river in the storm period, is shown in Figure 5. More than 90% of the total volume of SS did not exceed the particle size of 100 μm.

It was reported that the concentration of Particulate Reactive Phosphorus (PRP) in the fraction of SS from 8 μm to 44 μm was much higher than that of SS over 44 μm (Kawabe *et al.*, 1997). To clarify the accumulation of nutrients in the fine fractions, multiple linear regression analysis was applied. We set SS concentration in each fraction and POC concentration as explaining variables, and PN and PP concentrations as objective variables. The results of the analysis showed that the fine fraction of SS ($0\,\mu m < d < 23\,\mu m$) was correlated with PP at 1% significant level and partial correlation coefficient of 0.580 (Table 6). Concentration of PP was dependent on the concentration of the fine particle of SS. On the other hand, PN concentration was affected by POC concentration because PN

Table 5 Averaged power coefficients in Eqs. (7) and (8) for nine time series of the floods shown in Table 1

	Turb	SS	PN	PP	POC
m	–	1.21 (R = 0.963)	0.812 (R = 0.925)	0.994 (R = 0.966)	1.11 (R = 0.914)
n	0.798 (R = 0.963)	–	0.689 (R = 0.890)	0.867 (R = 0.946)	0.911 (R = 0.906)

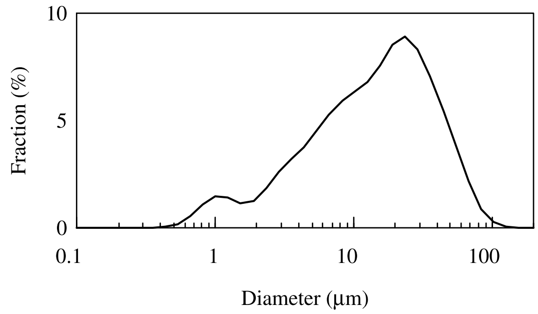

Figure 5 Volumetric particle size distribution of SS in river water in the storm period, Yotsugi Station in the Shira River, 25 June 2002

was found in the organic substances or debris of the organisms. We found that the accumulation of particulate nutrient concentration on fine particles and higher sensitivity of the turbidity meter for fine particles led to the linear relationship between turbidity and particulate nutrient concentration, particularly for phosphorus.

Estimation of runoff load of nutrients using turbidity meter (*L-Q-Turb* Method)

We proposed an advanced method of estimating runoff load of nutrients using turbidity in the L-Q-Turb method. Loads of dissolved nitrogen L_{DN} and phosphorus L_{DP} are modeled by an ordinary L-Q regression curve,

$$L_{DN} = C_{DN}Q^{n} \tag{9}$$

$$L_{DP} = C_{DP}Q^{p} \tag{10}$$

where Q is flow rate, and C_{DN}, C_{DP}, n and p are constants. Particulate nitrogen concentration, C_{PN} and particulate phosphorus concentration, C_{PP} are modeled by

$$C_{PN} = \alpha_{PN}Turb^{\beta} \tag{11}$$

$$C_{PP} = \alpha_{PP}Turb^{\gamma} \tag{12}$$

where $Turb$ is turbidity, α_{PN}, α_{PP}, β and γ are constants.

Table 6 Results of the multiple regression analysis of the river water of 28.1 KP station on 12–13 Sep., 2002 ($n = 12$) and 1–2 Oct., 2002 ($n = 17$)

	Explaining variable	PN	PP
Correlation coefficient	SS (0 μm < d < 23 μm)	0.935*	0.952**
	SS (23 μm < d < 54 μm)	0.842	0.851
	SS (54 μm < d < 103 μm)	0.464	0.461
	SSC (103 μm < d)	0.195	0.187
	POC	0.811*	0.735
Partial correlation coefficient	SSC (0 μm < d < 23 μm)	0.418	0.580**
	SSC (23 μm < d < 54 μm)	0.240	0.151
	SS (54 μm < d < 103 μm)	−0.297	−0.157
	SS (103 μm < d)	0.076	−0.029
	POC	0.498*	0.105
Standard partial regression coefficient	SS (0 μm < d < 23 μm)	0.524*	0.795**
	SS (23 μm < d < 54 μm)	0.370	0.225
	SS (54 μm < d < 103 μm)	−0.284	−0.157
	SS (103 μm < d)	0.033	−0.012
	POC	0.286	0.052
Coefficient of determination		0.891	0.887

**1% of significant level; *5% of significant level

Table 7 Comparison of the calculated cumulative discharged load of Total Nitrogen (upper) and Total Phosphorus (lower)

Station	Storm period	Measured load (t)	L-Q Regression		L-Q-Turb Method	
			Load (t)	Error (%)	Load (t)	Error (%)
(a) Total Nitrogen						
28.1 KP	12–13 Sep.02	2.39	2.12	11.2	2.28	4.7
	1–2 Oct.02	15.1	14.9	1.1	15.1	0.0
Kataniwa-A	10–11 Jul.–02	0.126	0.105	16.9	0.134	5.9
	1–2 Oct.–02	0.166	0.179	8.0	0.158	4.7
Kataniwa-B	10–11 Jul.–02	0.311	0.297	4.6	0.284	9.4
	1–2 Oct.–02	0.269	0.286	6.2	0.259	3.9
Maekawa	1–2 Oct.–02	9.65	9.65	0.0	9.49	1.7
Yotsugi	24–25 Jun.–02	17.2	17.4	1.5	17.2	0.1
Averaged				6.2		3.8
(b) Total Phosphorus						
28.1 KP	12–13 Sep.02	375	154	58.9	330	13.9
	1–2 Oct.02	1,580	1,700	7.7	1,710	7.7
Kataniwa-A	10–11 Jul.–02	9.74	3.40	65.2	9.02	8.0
	1–2 Oct.–02	5.37	5.53	2.9	5.07	5.9
Kataniwa-B	10–11 Jul.–02	69.8	35.8	48.7	64.0	9.1
	1–2 Oct.–02	46.2	33.6	27.2	57.7	19.9
Maekawa	1–2 Oct.–02	845	792	6.3	760	11.2
Yotsugi	24–25 Jun.–02	3,500	3,400	2.8	3,100	12.9
Averaged				27.5		11.1

Load: cumulative discharged load. Error: root mean square of error which was calculated by $\sqrt{(L_M - L_E)^2/L_M{}^2}$, where L_M: measured cumulative load, and L_E: estimated cumulative load

As a consequence, time series of the total nitrogen load, L_{TN} (t) and total phosphorus load, L_{TP} (t) are calculated as follows:

$$L_{TN}(t) = C_{DN}Q(t)^n + \alpha_{PN}Turb(t)^{\beta}Q(t) \tag{13}$$

$$L_{TN}(t) = C_{DP}Q(t)^p + \alpha_{PP}Turb(t)^{\gamma}Q(t) \tag{14}$$

where $Turb(t)$ is the time series of turbidity and $Q(t)$ is the time series of flow rate.

Evaluation of estimation method of runoff load of nutrients

Total discharged nutrient loads were calculated by the following three methods to evaluate the accuracy of the estimation of the load of nutrients: (1) actual discharged nutrient loads from the measured flow rate and nutrient concentrations; (2) runoff load modeled by *L-Q* regression curve and calculated total discharged load from the time series of flow rate; (3) calculation by *L-Q-Turb* method. The results of the comparison are shown in Table 7. Root mean square of errors by the *L-Q-Turb* method was half of that by *L-Q* regression curve. *L-Q-Turb* method could improve the accuracy of the estimation of the runoff load of nutrients in flood periods.

Conclusions

To establish a new estimation method of the runoff loads of nutrients, the relationship of particulate nutrient concentrations and turbidity was studied. The main conclusions are as follows. (1) The "Nutrient Load Hysteresis Coefficient (NLHC)" was proposed to evaluate the hysteresis of the nutrient loads to flow rate quantitatively. As a result of the NLHC analysis combined with the power number of the L-Q regression curve, we could classify the runoff patterns into 15. It was clarified that the fluctuation of the particulate nutrient loads has strong hysteresis; therefore they can hardly be modeled with flow rate.

(2) Linear relationships between the concentrations of particulate nutrients and turbidity were observed. It was clarified that the linearity was caused by the influence of the particle size on turbidity output and accumulation of nutrients on the smaller particles (diameter $<23\,\mu m$). (3) The *L-Q-Tb* method, which is the new method for the estimation of the nutrient loads using regression curve between the turbidity and the concentrations of particulate nutrients, was developed. This method could raise the precision of estimation of nutrient loads even if they had strong hysteresis to flow rate.

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