Estimation of annual pollutant loadings in two small catchments and examination of their differences caused by regional properties

Water Science & Technology **Vol 53 No 2 pp 33–44** © IWA Publishing 2006

S. Fujii*, M. Moriya, P. Songprasert* and H. Ihara*****

*Research Center for Environmental Quality Control, Kyoto University, 1-2, Yumihama, Otsu, Shiga 520-0811, Japan

** CTI Engineering Co., Ltd., 1-14-6, Uekizaki, Urawa-ku, Saitama, 3300-0071, Japan

***Department of Soils and Fertilizers, National Agricultural Research Center, 3-1-1 Kannondai, Tsukuba, 305-8666, Japan

Abstract A series of runoff surveys was conducted for more than one year in two small catchments of the Kamo River basin (75.4 km^2) and the Takano River basin (66.8 km^2) in Kyoto, Japan, which adjoin each other, and may have the same precipitation pattern. The investigation consisted of a high-frequency periodic survey, a long-term regular survey and a storm event survey. The survey results were compared with the regional properties of the basins, and the following results were obtained. (1) Pollutant loadings were successfully estimated as two portions of base discharge and storm events discharge from the survey results. (2) Estimated annual loading of the sites was 2.9–4.5, 1.3–1.8, 17–27, 1.3–2.2, 0.076–0.97 t/km^2/y, respectively for COD_{Mn}, DOC, SS, TN and TP. (3) 52–53% of the whole flow, which was caused by rainfall events, conveyed 81–87, 68–73, 92–95, 64–67, 76–81% of the whole loading, respectively for COD_{Mn}, DOC, SS, TN and TP. (4) Differences of regional properties in two basins cause different runoff patterns, but the differences in runoff patterns also depend on the rainfall patterns.
In general, a more urbanized basin receives early and strong influence of precipitation on the storm event runoff.

Keywords Annual loading; regional properties; regression model; storm events; water quality

Introduction

Runoff of pollutants is a complicated phenomenon receiving various kinds of influences. Although many factors affect the runoff process, the most important factors might be regional properties of the river basins and the pattern of rainfall. The former is a spatial factor depending on conditions of the upstream basins, and includes population, land use, soil types, geographical features, and so on. The latter is a meteorological factor, and includes duration of rainfall period, maximum precipitation intensity, preceding non-rain duration, and so on. Both factors never show the same pattern if either time or place is different, so that the runoff also shows a quite different pattern in each place and each time, resulting in difficulty understanding the phenomenon. To fix one of the two factors would be a good method for evaluation of the other factor's effects. In this study, two connected river basins were selected as study fields and a series of surveys were conducted during the period for more than one year including storm events. From these results, annual runoff loadings were estimated and compared with the regional properties of two basins.

Materials and methods

Study sites

Kamo River and Takano River were selected as study fields (Figure 1). Both rivers are located in the northern part of Kyoto City, Japan and flow southward, becoming one

doi: 10.2166/wst.2006.036

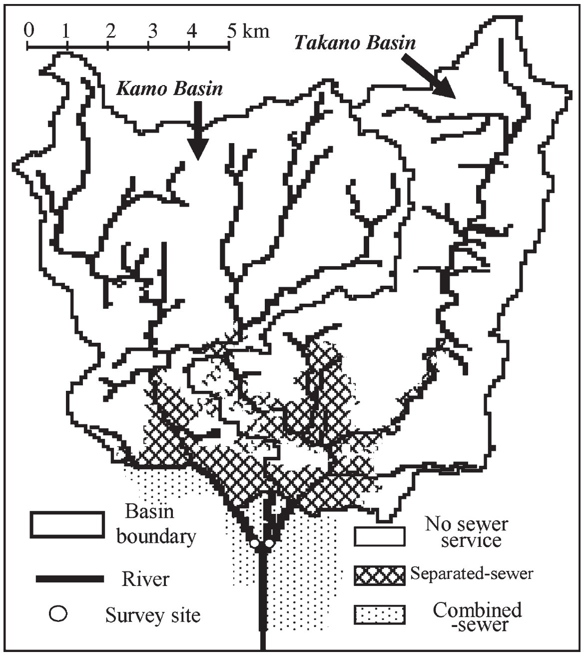

Figure 1 Research site boundaries and stream lines

stream. The observation site for each river was the point at about 20 m upstream from their juncture. Table 1 summarizes the characteristics of basins. To obtain these data, the area was divided into small rectangular cells of 3.0”SN (= 92.4 m) $*$ 4.5”EW (= 114 m) by the latitude and longitude, and flow directions of the cells were determined with position of streams and altitude of each cell (Songprasert *et al.*, 2003), so that the boundaries of both basins were identified. Several kinds of information were collected, and assigned to each cell, and the number of the cells corresponding to specified conditions was counted to calculate the area of the conditions.

As shown in the table, forest is major land use, and occupies 89 and 80%, respectively for the Kamo River and Takano River basins. Both basins have residential areas, and most of them receive sewer service. Combined sewer service areas were excluded from the catchment calculation because almost all water produced there is conveyed to the outside, but separated sewer areas were included. These areas are shown in Figure 1.

Table 1 Properties of Kamo and Takano Basins

Area, km^2 (%)		Kamo River	Takano River
Land use	Total	75.4	66.8
	Forest	67.2 (89.1)	53.2 (79.6)
	Agriculture	1.5 (2.0)	3.0 (4.5)
	Residence	5.7 (7.6)	9.5 (14.2)
	Others	1.0 (0.5)	1.1 (14.2)
	Sewer service	6.4 (8.5)	10.5 (15.7)
Population, ca (%)	Total	41,300 [548]	62,700 [939]
	Non-sewer service	3,300 (7.9)	5,300 (8.5)
	Sewer service	38,000 (92.0)	57,000 (90.9)
Mean gradient (m/km)		364	320
Mean flow distance, km		16.9	14.1

Area and population in combined sewer service are excluded from the above statistics [population density, ca/km^2]

Percentages of residence are 5.7% for the Kamo River basin and 14.2% for the Takano River basin, and population densities are 548 and 939 ca/km^2, respectively. In general, the Takano is more urbanized than the Kamo.

Observation methods

The survey started from September 23, 2002. During the first 80 days, sampling and measurement were conducted every other day, and continued at intervals of 10 days until December 10, 2003. The survey is still ongoing with an extended interval of one month. The sampling time for these observations was fixed at 8:00. Table 2 summarizes these investigations. The observed items are flow rate, environmental conditions, particulate pollutants, organic matter, nutrients, anionic ions and metals. In addition to these regular observations, investigation of storm events was conducted six times as shown in Table 3. Those storm event surveys had 2–50 mm rainfall with the maximum precipitation intensity ranging from 1 to 12 mm/h. Duration of each survey ranged from 28 hours to 12 days. The number of samplings ranged from 5 to 28 times. The same items were measured in these surveys as in the regular observations.

Results and Discussion

Annual variations of flow rate and water quality

Figure 2 shows time-series variations of flow rate and some water quality indices in both sites from September 23, 2002 to December 10, 2003. Precipitation data used in the study were obtained from AmeDas (Automatic meteorological data acquisition system), and hourly data of Kyoto City observation station, which is located near the basins (Japan Meteorological Business Support Center, 2004). The figure shows summed values of hourly data in 6 hours. During the period, the basins received rainfall of 1999 mm in 281 days out of 444 days. Maximum rainfall was 28.5 mm in an hour, 53.5 mm in 6 hours, and 115 mm in a day. When one storm event is considered as a series of rainfalls with their intervals less than 12 hours, 114 storm events happened with 16.0 mm of the average precipitation in 11.6 hours of the average duration.

The flow rate fluctuated in the range of 0.16–18 m^3/s (Kamo River) and 0.076–14 m^3/s (Takano River). The pattern in each river consisted of a basic gentle change and several sudden sharp peaks. These peaks always occurred at storm events, but some storm events did not express such peaks. The durations of these peaks were usually less than one day, so that the regular sampling cannot explain all of the storm events even if it was conducted with the interval of two days. Kamo River and Takano River had very similar flow rates, but differences of about 30% were observed at the peaks, which occurred during storm events. This may be explained by the difference in land properties in both catchments.

Table 2 Outline of regular sampling

Period	Frequency
2002/09/23 ~ 2002/12/12	every 2 days
2002/12/12 ~ 2003/12/10	every 10 days
2003/12/10 ~ (continue)	every month
Observed Items	
Flow rate, Temperature, pH, DO, SS, VSS	
COD_{Mn}, D-COD_{Mn}, TN, DN, TP, DP, DOC	
IC, SiO_2, NH_4-N, NO_2-N, NO_3-N, PO_4-P	
SO_4^{-2}, Cl^-, D-Metals (Na, Mg, Ca, Fe, etc)	

Table 3 Outline of storm events investigation

Storm event	Sampling			Precipitation		Preceding conditions	
	Start	End	No.	Total mm	*1mm/h	*2hrs	*3mm
Se1	02/10/19 11 h	10/20 15 h	9	4.5	1.5	91	1.0
Se2	02/11/07 21 h	11/08 12 h	5	2.0	1.0	139	24.0
Se3	03/07/21 03 h	07/22 12 h	6	5.0	2.0	53	0.5
Se4	03/07/23 09 h	07/28 08 h	14	33.0	12.0	48	5.5
Se5	03/11/05 16 h	11/08 08 h	15	12.0	4.5	47	13.0
Se6	03/11/28 21 h	12/10 08 h	28	53.5	8.5	82	51.5

*1 Maximum precipitation intensity
*2 Duration from the end of previous rainfall to the storm event
*3 Cumulative rainfall during preceding 7 days

Concentrations of SS, TN and TP were shown as examples of water quality variations. These items also had a pattern consisting of a basic gentle change and several sharp sudden peaks. SS was the item that had quite high peaks. In regular sampling, 90% of SS concentrations were less than 7 mg/L (Kamo River), or 5 mg/L (Takano River), but the concentrations at storm events suddenly increased to the level of 20–100 mg/L. On the other hand, TN received less significant effects of storm events, and was less fluctuated. The coefficient of variance (standard deviation / mean) in regular samples was only 31% (Takano), or 39% (Kamo) for TN, but 135% (Kamo), or 204% (Takano) for SS.

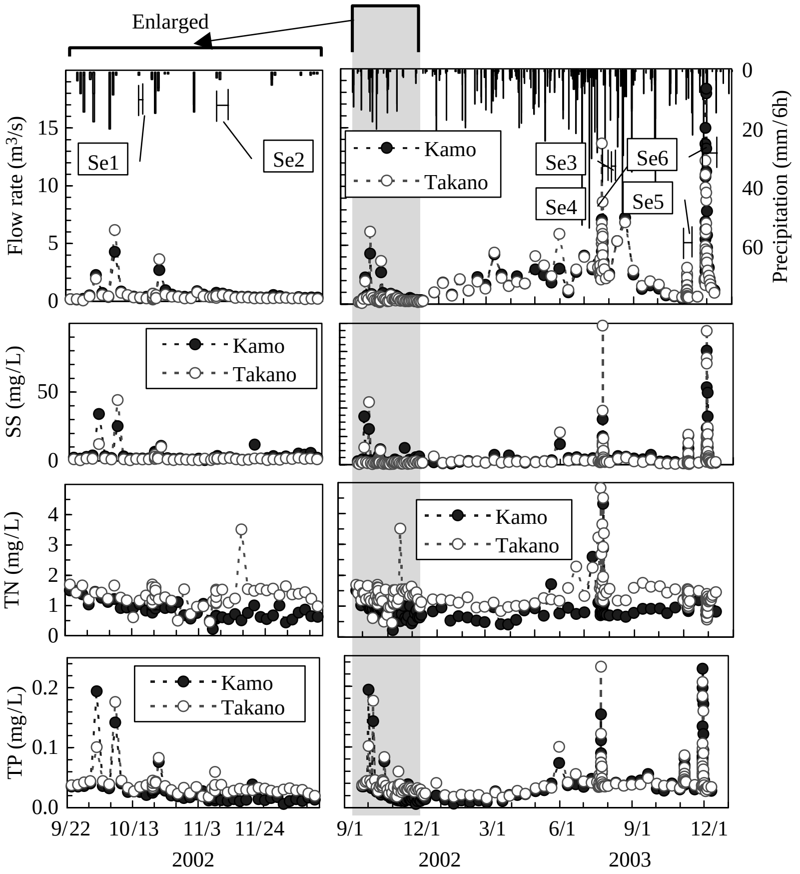

Figure 2 Fluctuations of flow rate, SS, TN and TP

Concentration differences of both rivers were, however, more obvious in TN, and Takano River showed 1.5 times the concentration of that in Kamo River. TP made an intermediate pattern between SS and TN.

Variations of flow rate and water quality in storm events

Figure 3 shows variations of the flow rate and some water quality items in three storm events. In this figure, the axis scales of flow rate and SS were changed in three events because the range of fluctuation was quite different. Flow rate was very sensitive to the rainfall, and the influences were observed even in the case of 2 mm precipitation (Se2). Flow rate increased very sharply when rainfall started, but a few hours of delay were always observed. In the case of heavy rain (Se6, 53.5 mm), obvious effects still remained a few days after the rainfall stopped. SS showed a similar pattern, but the change was more obvious than flow rate. The concentration increased to 10–100 times within a few hours, and returned back to the previous level within one day if the rainfall ceased. This pattern was also observed in COD_{Mn}, D-COD_{Mn}, DOC, VSS, TP, DP and D-Al.

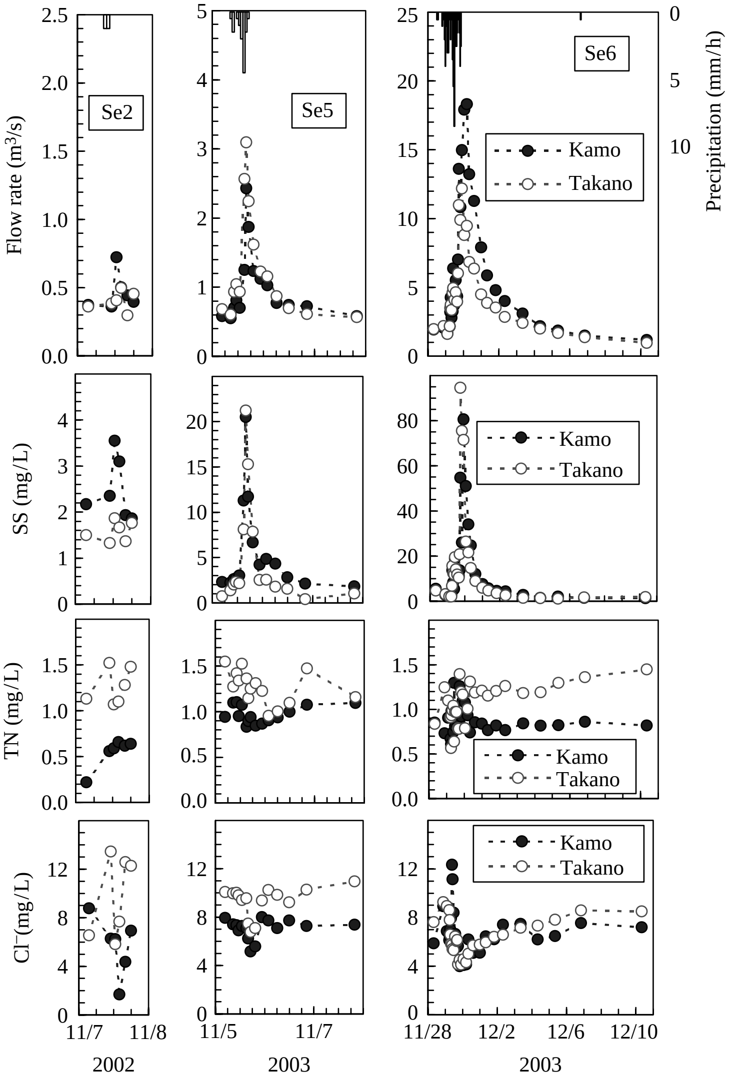

Figure 3 Fluctuations of flow rate, SS, TN and chloride during storm events

The items related to organic matter and particulate substances seem to have a similar pattern. In case of particulate substances, this pattern can be explained by washout of deposited substances, which are accumulated on riverbeds or land surface during dry weather. The increase of discharge by storm events might promote the rinse of such deposits, resulting in the increase of organic matter (D-COD$_{\mathrm{Mn}}$, DOC) during storm events.

On the other hand, ionic species, such as chloride, showed a different pattern. They decreased with the increase of flow rate and returned back to the usual level when the flow rate was recovered. These items were IC, Cl$^-$, SO$_4^{2-}$-S, D-Na, D-Ca, D-Mg and D-Sr. These items seem to be rapidly leached from the watershed without being trapped on riverbeds or the land surface, receiving the effect of dilution of rainwater directly. TN, NO$_2^-$-N, NH$_4^+$-N, SiO$_2$-Si and K had patterns different from the above two. These items might have other discharge processes or receive both effects.

Response of flow rate and water quality to rainfall

As discussed in the previous section, flow rate increased with some time lag after rainfall started. This point was analyzed with mutual correlation analysis between precipitation and flow rate. Figure 4 shows the results of Takano River, which showed a more obvious relationship between precipitation and flow rate. In this analysis, a leaner interpolation was used to make the flow rate a continuous function from discrete measured data. As shown in the figure, obvious correlation peaks were observed in most of the storm events with the time lag of 1–4 hours. The correlation peaks in Kamo River weekly appeared with slightly longer time lag. Water quality items related to organic matter and particulate substances also showed a similar pattern.

On the other hand, the decreasing pattern after these peaks was different between flow rate and water quality items. Figure 5 shows variation of flow rate and SS in Takano River at Se6. SS decreased with an exponential pattern very rapidly. The time needed to become one tenth was about a quarter of that for flow rate.

Estimation of pollutant loading during storm events

As discussed in the previous section, data on regular sampling could hardly reflect storm events, so that an estimation method for the effects of storm events is required to evaluate annual pollutant loading from a watershed. Then, we adopted a regression equation model to estimate loading from the rainfall data (Ebise, 2004). Figure 6 shows its conceptual chart. This model has the following assumptions:

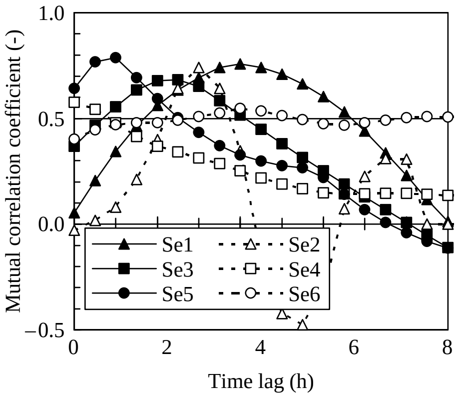

Figure 4 Mutual correlation analysis for delay of flow discharge in Takano River

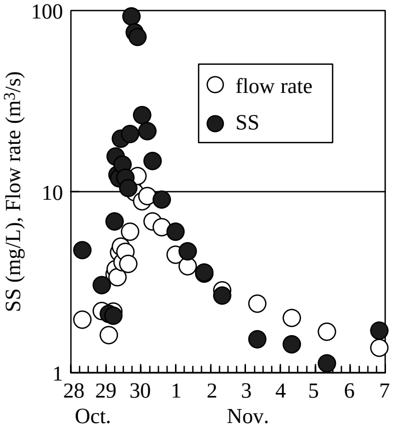

Figure 5 Change of flow rate and SS in Se6 (Takano River)

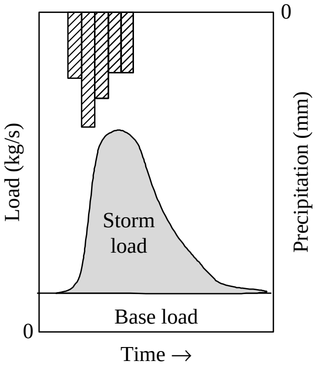

Figure 6 Concept of load structure

(1) Discharge loading consists of a base and a storm portion.
(2) The base loading part is never affected by storm events, and is assumed to be constant.
(3) The storm loading is an increment part that increases after storm events.
(4) The whole loading in one storm event is a function of the whole flow quantity, and is given in the following equation:

$$\sum_{i}^{N} Ls_i/A = a \times \left(\sum_{i}^{N} Q_i/A \right)^n \tag{1}$$

where Q_i and Ls_i are flow quantity and loading during $t_{i\text{-}1}$ to t_i (t_0: start time, t_N: end time), A is the area of the basin and a and n are parameters.

(1) The flow quantity is considered as a function of total precipitation amount, and is given in the following equation:

$$\sum_{i}^{N} Q_i/A = b \times \left(\sum_{i}^{N} R_i/A \right)^m \tag{2}$$

where R_i is precipitation amount during t_{i-1} to t_i.

For the practical calculation of the loading in the two basins, we used the following procedures.

(1) The period from January 1, 2003 to December 31, 2003 was applied for the calculation of annual loading.

(2) Fifteen out of 34 regular samplings were used for the base loading calculation. The samples were regarded as the data that scarcely receive the effects of any storm events.

(3) A series of rainfalls with their intervals less than 12 hours was considered as one storm event. Such storm events were counted 100 times in the period.

(4) Discharge increments were calculated for six storm events (Table 3) from the special observation data and the base flow rate. These increments were accumulated during the period of each storm event. The resultant cumulative value was regressed with its cumulative precipitation amount, and the parameter values for the equation (Eq−2) were determined.

(5) Similarly, the increments of loading were calculated, and the cumulative value was regressed with the cumulative flow rate. The parameter values for the equation (1) were determined.

Figure 7 illustrates the relationship between precipitation and discharge in both rivers in a log−log chart. Cumulative discharge had a linear relationship to the whole precipitation in a logarithmic scale, and the correlation coefficient of them was 0.973 for Kamo River and 0.968 for Takano River. This means the regression equations shown in the figure can be available for the estimation of flow rate in other storm events. The slope value was obviously higher than unity, being 1.67 for Kamo, and 1.77 for Takano. This means that stronger precipitation yields higher discharge flow. For example, percentage of rainfall appearing as the storm flow is estimated to be only 3% at the precipitation of 2 mm, but to be more than 40% at that of 50 mm.

Similarly, the relationship between cumulative discharge and cumulative loading is shown in Figure 8, using COD_{Mn} as an example. In case of COD_{Mn}, a high linearity was observed in a logarithmic scale yielding a high correlation coefficient (0.987, 0.988). The slope was slightly higher than unity, indicating that a bigger storm might cause much higher effects.

The relationship between discharge and loading was different among water quality items. Table 4 summarizes the characteristics of such a relationship with parameter values in equation 1 and correlation coefficient values. The values of n, which expresses

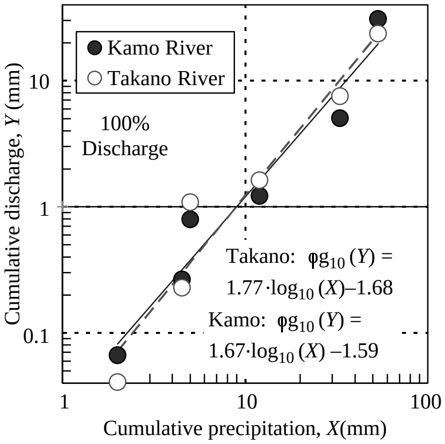

Figure 7 Relationship between precipitation and discharge

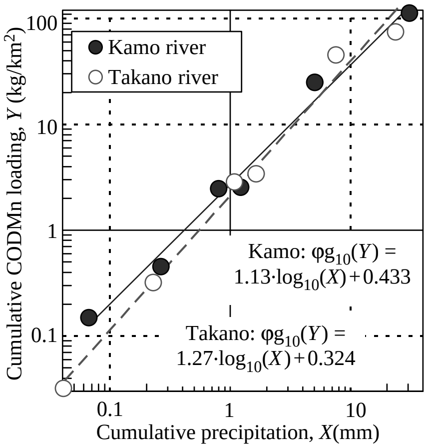

Figure 8 Relationship between precipitation and load

the magnitude of flow influence, were ranging from 0.77 to 1.4. The differences between water quality items were higher than those between the two rivers. The items having *n* more than 1 in both rivers were indices related to organic matter or particulate substrate such as COD_{Mn}, DOC, SS, and TP, while those having *n* less than 1 were indices related to ionic portions such as IC, Cl^- and Na. SS and VSS had very high values of *n* in the range of 1.3–1.4. This means if cumulative flow rate became 100 times, the loading

Table 4 Parameter values of storm runoff equations and estimation of loadings

	Parameters of regression Equation[1]						Loading[2]	
	Kamo River			Takano River			Kamo River	Takano River
	a	*n*	R^2	*a*	*n*	R^2		
Flow	0.026	1.668	0.973	0.021	1.765	0.968	1,131 (51.7)	1,323 (53.2)
COD_{Mn}	2.70	1.128	0.987	2.11	1.270	0.988	2.93 (80.7)	4.46 (86.5)
D-COD_{Mn}	1.93	1.099	0.980	1.46	1.045	0.970	1.96 (78.5)	1.64 (72.8)
DOC	1.24	1.069	0.968	1.17	1.129	0.992	1.32 (68.1)	1.77 (72.5)
IC	6.88	0.852	0.984	5.16	0.984	0.995	6.15 (42.5)	7.59 (45.4)
SS	8.58	1.348	0.994	7.75	1.428	0.985	17.17 (92.0)	26.96 (95.7)
VSS	2.29	1.289	0.997	2.25	1.313	0.983	3.68 (93.4)	5.11 (94.6)
Cl^-	4.94	0.962	0.984	5.49	0.933	0.983	5.95 (43.3)	8.63 (36.0)
SO_4^{2-}-S	7.86	0.945	0.990	3.97	1.233	0.984	8.60 (45.4)	12.54 (48.8)
SiO_2-Si	3.32	1.104	0.996	3.36	0.940	0.991	3.91 (45.5)	4.34 (44.8)
Ca	8.66	0.811	0.899	6.34	1.000	0.980	8.98 (32.8)	11.77 (37.9)
Mg	1.33	0.782	0.834	1.24	0.842	0.925	1.35 (30.9)	1.88 (28.1)
Na	4.51	0.825	0.843	4.25	0.845	0.958	4.55 (34.9)	6.07 (30.1)
K	1.02	0.845	0.943	0.93	0.979	0.990	0.78 (48.4)	1.36 (45.2)
TN	0.74	1.210	0.894	1.22	1.149	0.818	1,275 (66.6)	2,247 (63.8)
DN	0.89	0.918	0.987	0.57	1.149	0.948	773 (52.6)	1,456 (46.4)
NO_3^--N	0.68	1.012	0.941	0.73	1.113	0.969	748 (54.8)	1,438 (51.9)
NO_2^--N	0.0046	0.772	0.994	0.0046	0.903	0.979	3.29 (42.7)	5.48 (43.4)
NH_4^+-N	0.026	1.278	0.766	0.035	1.081	0.801	45.7 (82.3)	40.5 (79.8)
TP	0.049	1.241	0.992	0.051	1.223	0.988	76.2 (81.8)	94.7 (81.0)
DP	0.026	1.234	0.944	0.035	1.073	0.977	42.8 (75.4)	47.5 (65.6)
D-Al	0.026	1.006	0.798	0.009	1.260	0.869	21.0 (73.6)	20.6 (74.4)
D-Fe	0.045	0.912	0.980	0.031	0.945	0.993	42.6 (53.2)	41.1 (51.1)
D-Sr	0.045	0.797	0.859	0.031	0.941	0.787	48.3 (30.7)	56.5 (31.9)

[1] Parameters in Eq-1 (in flow, $a \Rightarrow b$, $n \Rightarrow m$ in Eq-2), $Z = a \cdot Y^n$; $Y = b \cdot X^m$, *X*: cumulative precipitation (mm), *Y*: Cumulative discharge (mm, $= 10^3 m^3/km^2$), *X*: cumulative loading (km/km^2)

[2] Unit; Flow (mm) COD_{Mn} to K, ($t/km^2/y$), TN to D-Sr ($kg/km^2/y$), ()Storm events %

Table 5 Comparison of unit loading factors for pollutant discharge runoff in Japanese rivers

		This study		**Takashima *et al.*, 1995**		**Ebise, 1989**		**Ichiki *et al.*, 1999 (storm events)**				
		Kamo	**Takano**	**Ohbori**	**Ohtsu**	**San-oh**	**Koise**	**Tanjin**	**Yama-shina**	**Juzenji**	**Isasa**	**Houryu**
Area (km^2)		75.4	66.8	31.5	36.7	12.4	151.5	25.8	32.2	0.9	4.2	7.1
Land use (%)	Forest (%)	89	80	28	23	15	46					
	Agriculture (%)	2	5	16	29	46	44					
	Residence (%)	8	14	56	48	39	9	54	34	80	72	26
Population density (ca/km^2)		548	939	5,488	5,207	1,772	247	6,058	4,409	2,128	2,410	992
Precipitation (mm/y)		1,814	1,814	1,751	1,576	1,193	1,193	1,562	1,562	1,562	1,562	1,562
Flow rate (mm/y)		1,131	1,323	1,428	1,271	1,157	601					
Unit loading factor (kg/km^2/y)	COD_{Mn}	2,930	4,460	17,500	17,400	9,650	4,580	17,500	6,060	2,280	5,750	7,500
	TN	1,270	2,250	9,660	8,610	3,411	1,794		2,070	1,990	1,720	3,500
	TP	76	97	1964	998	399	125		230	480	310	410

would become 430–600 times. "*a*" is the parameter value indicating strength of the loading for each index, and gives discharge concentration at the total precipitation of 1 mm.

Estimation of annual loading

Based on the above parameter values, annual loadings in 2003 (from January 1 to December 31) were estimated in both rivers. The results are shown in Table 4. In that year, 100 storm events (a series of rainfalls with their intervals less than 12 hours) occurred, and 76 events out of 100 had precipitations of 2.0–53.5 mm, which was the interpolating range of the regression equation. Eighteen events happened in the extrapolating range less than 2 mm, but they must not cause any important estimation errors because the sum of their cumulative precipitation amounts is 13.5 mm, only 0.75% of the whole precipitation amount (= 1,810 mm). On the other hand, storm events in the outer extrapolating range (>53.5 mm) may have important influences on the estimation of annual loadings. Such storm events happened six times (58.5, 64.5, 67, 105.5, 123.5, 127 mm), and their sum became 30% of annual precipitation in 2003. The sum of the estimated discharge in the outer range occupied 51% (Kamo River) and 54% (Takano River) of the whole flows in 2003.

Estimated annual loading of the sites was 2.9–4.5, 1.3–1.8, 17–27, 1.3–2.2, 0.076–0.97 t/km^2/y, respectively for COD_{Mn}, DOC, SS, TN and TP. Comparison of both rivers indicates the Takano River basin has a structure to discharge more pollutants with increased flow rate, probably due to more urbanized land use. Organic matter (COD_{Mn}, SS) and nitrogenous substances (TN, DN and NO_3^--N) showed high differences of more than 1.5 in the ratios (Takano/Kamo). On the other hand, loadings of NH_4^+-N, D-Al and D-Fe, which may be related to soil components, were smaller in Takano River than in Kamo River although the specific flow rate in Takano River was 1.2 times that in Kamo River. The storm events caused 52 (Kamo) or 53 (Takano) % of the whole flow, and conveyed 81–87, 68–73, 92–95, 64–67, 76–81% of the whole loading, respectively for COD_{Mn}, DOC, SS, TN and TP.

These values (unit loading factors) were compared with previous studies (Table 5). Since land use pattern and population density are different, the values are also different. The values in the study are roughly half or less of the values in other studies. Our surveyed areas have less population with a majority of forest cover. Therefore, our values may be considered as rather background values for river basin runoff.

Conclusions

In this study, pollutant runoff patterns were investigated in two small catchments, the Kamo River basin and the Takano River basin, based on a series of surveys. The main results obtained are as follows.

(1) Pollutant loadings were successfully estimated as two portions of base discharge and storm events discharge from the survey results.

(2) Estimated annual loadings of the sites were 2.9–4.5, 1.3–1.8, 17–27, 1.3–2.2, 0.076–0.97 t/km^2/y, respectively for COD_{Mn}, DOC, SS, TN and TP.

(3) 52–53% of the whole flow which was caused by rainfall events conveyed 81–87, 68–73, 92–95, 64–67, 76–81% of the whole loading, respectively for COD_{Mn}, DOC, SS, TN and TP.

(4) Differences of regional properties in the two basins cause different runoff patterns, but the differences in runoff patterns also depend on the rainfall patterns. In general, a more urbanized basin receives an early and strong influence of precipitation on the storm event runoff.

References

Ebise, S. (1989). Amount and change of annual discharge loadings from different land-use areas. *Water Pollution Research*, **12**, 497–505.

Ebise, S. (2004). Nonpoint pollution source, in "Formulas, models and tables in environmental engineering" (edited by the Publication Task Group for Environmental Engineering), *Japan Society of Civil Engineers*, pp. 333–344.

Ichiki, A., Yamada, K., Nakatani, Y., Yamada, T. and Okubo, T. (1999). Comparison of pollutant runoff characteristics in several river basins. *Proc. of Environmental Engineering Study Forum*, **36**, 88–90.

Japan Meteorological Business Support Center (2004). Weather observation data in electrical data reading room, http://www.data.kishou.go.jp/.

Songprasert, P., Moriya, M., Fujii, S., Nagare, H., Ihara, H. and Shimizu, Y. (2003). Influence of basin properties on river pollution loading in fine weather. *CD Proceedings of IWA Asia-Pacific Regional Conference*, **9**, p.1Q5A04.

Takashima, E., *et al.* (1995). Characteristics of pollutant loadings in rivers to Teganuma Lake, and their estimation of annual loadings. *Jour. of Japan Society on Wat. Environ.*, **18**, 297–306.