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REVIEW

Methods for Estimating the Nitrogen Load on a Catchment Scale

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Abstract

A Windows-based system for estimating the potential nitrogen (N) outflow in a catchment has been developed. The system includes a conversion method for administrative-based data from national scale statistics, such as the agricultural census, into a catchment-based data by utilizing the geographic information system (GIS). The system calculates N loads based on the pollutant load unit (PLU) method in a catchment and converts them to potential N outflows by introducing the outflow coefficients. For the catchments of tributaries flowing into Lake Kasumigaura in central Japan, where non-point sources were predominant, the N loads and the potential N outflows from 1950 to 1995 were estimated. The actual N outflows from the tributaries were also estimated using the water quality monitoring data at the outlet of the tributary catchment and meteorological data. These potential N outflows and actual N outflows were compared. Results obtained were as follows. (i) Some estimates by the present method, such as the drainage area and cultivation areas of various crops in each tributary, well agreed with those estimated by other methods and published data, indicating that this method was capable of estimating catchment-based data from the administrative-based statistics. (ii) Changes in the potential outflows during the period from 1950 to 1995 reflected the changes in livestock production and methods of waste water treatments. (iii) Comparison between the N potential outflows and the actual N outflows suggested that about 47% of the generated N load which leached down to groundwater from the soil surface flowed out and that a time-lag between two outflows appeared. Thus, the developed system offered good possibilities of realistic quantitative analyses on the contribution of non-point sources to water quality on the scale of a catchment.

Discipline: Agricultural environment
Additional key words: GIS, N outflow coefficient, pollutant load unit, potential N outflow

Introduction

Nitrogen (N) is an essential element for crop production, but at the same time, excessively applied N to agricultural lands has a great potential of causing so called non-point pollutions[15] by flowing out to the groundwater system and eventually to the surface water. However, it is difficult to show how much of the N applied to agricultural lands in a catchment actually reaches aquatic environments, and when, because water and N movements in the soil involves complicated processes.

Currently, local governments in Japan have been adopting the simple "pollutant load unit" (PLU) method[13] to estimate and evaluate the effect of agricultural and human activities on water quality and to develop pollutant-control plans[4]. However, the method does not answer the questions of how much N derived from non-point sources enters the water and when[20], because those load units are usually determined using only outflow data independently of the amounts of N input on the soil surface; moreover, concerning environmental pollution, it neglects the importance of N removal activities in submerged soil such as rice paddies[5,24,32,33] and wetlands[38], which are a typical and widespread feature of monsoon Asia, as well as riparian zones[7] and the groundwater system[28]. The N removal activities in a rice paddy are considered as including N uptake by the rice plants, N accumulation in the soil and denitrification in the submerged reducing condition during the growing period[5,24,32,33]. Although these N removal activities in rice

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paddies are persistently expected, quantitative evaluation on a catchment scale has not been fully achieved yet, because of the lack of an appropriate tool.

Therefore, one of the requirements of an N outflow prediction tool should be an ability to consider the N removal activities in submerged soils and in other sites on the scale of a catchment. The present paper shows the very first part of developing such a model and it has actually turned out to be the most difficult part, since the development of N load generation methods on a catchment scale other than the simple PLU methods was essential.

For the estimation of N load generation, we decided to utilize national scale "qualitatively homogenous" information such as various censuses and statistics as much as possible in order to achieve minimal imbalance on the qualities of the calculations in different regions. There was, however, another hurdle to be overcome in utilizing such data for the calculation as catchment based values, since data in censuses and statistics are usually reported not on a catchment-basis but on an administrative-basis. Therefore, there was a need to develop a method of converting the administrative-based data to catchment-based data by utilizing the geographic information system (GIS). The GIS has been required for spatial fineness to realize the complicated land use distribution in Japan and has also been developed in order to utilize paper maps which were widely distributed from the governmental organization[8].

At the same time, a concept of "N outflow coefficient" is introduced in this paper to estimate "N potential outflow" as N load potentially released to the water systems such as groundwater beneath the soil and local drainage ditches and/or rivers. When an N outflow path is chosen for N load generated at each source, the N potential outflow is calculated as a product of the amount of the load and an appropriate outflow coefficient specifically established depending on the N outflow paths.

In this paper administrative-based data and GIS with other scientific evidences are used in an example of an application of the methods in some catchments within the Lake Kasumigaura watershed where rice paddies are commonly seen. We also discuss the features of fluctuations in potentials of N outflow from the catchments and the quantitative comparisons between the potentials and the actual N outflows at the outlet of the catchments.

Materials and methods

1. Study site

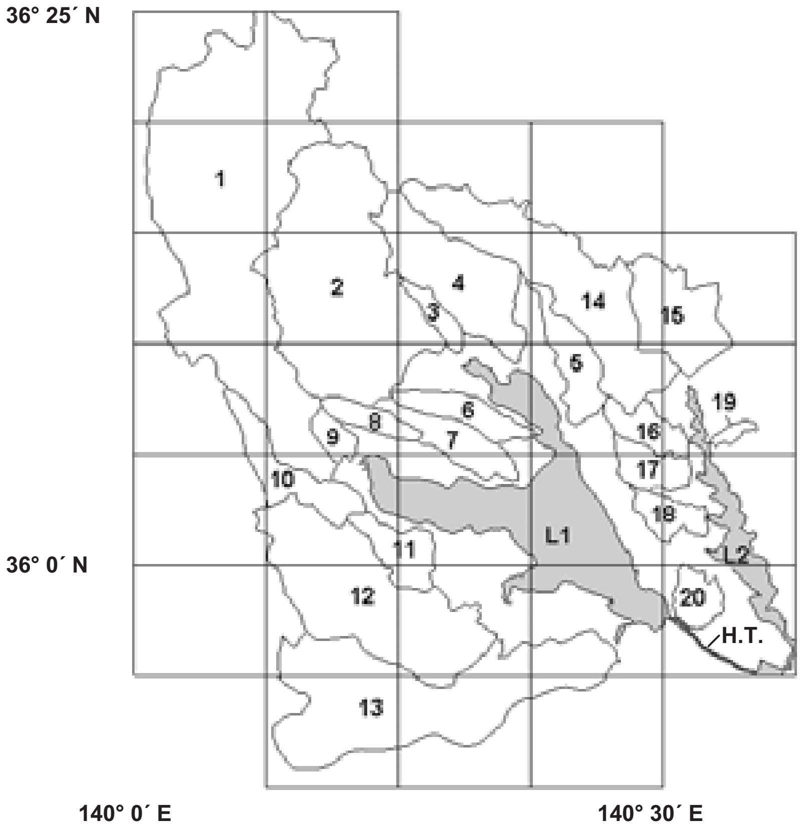
Lake Kasumigaura is the second largest lake in Japan with a surface area of about 220 km^2. The area of the watershed is about 2,157 km^2 [17]. The lake is located in central Japan at about 36° N and 140.4° E (Fig. 1). The land around the lake forms part of a plateau and is rather flat. About 20.5% of this area is periodically flooded for rice and lotus cultivation[16] mainly spreading in the low-lying areas of land. Some features of the typical tributary catchments within the watershed are listed in Table 1, indicating the predominance of non-point sources such as forest and agricultural lands as well as the higher activity in livestock production.

2. Preparation of digital map data

The geographic information system (GIS) and related programs operating on a Windows personal computer (PC) were developed using Borland C++ for Windows[1]. The GIS enabled basic bitmap file operations and overlay analysis by assigning various attributes to each pixel. The attributes included land use type, the names of a catchment and the old administrative district (AD) where the pixel belonged.

The details of the data manipulation are as follows. Bitmap files in different subjects were developed by digital scanning of outline images on maps. Elevation and land use data were scanned from land use maps (published between 1976 and 1983) and/or topographical maps (published in 1995 and 1996) of 1 to 25,000 scale[8]. The old AD data were scanned from old topographical maps of 1 to 50,000 scale[8], which had been developed around 1950. The borders of the catchments were scanned from maps of the river system survey (published in 1980) with a 1 to 50,000 scale[22]. The sewer distribution area borders were scanned from the sewer construction plan maps[27] (published in 1991). Then the bitmap files, each equivalent to a 1 to 25,000 scale map, were independently converted to rectangular shaped images so that each digital map consisted of 705 and 844 pixels for the north-south and east-west directions, respectively. One pixel in the resulting digital maps represented a square shaped area of land about 13 × 13 m^2 in size at 36° N. Lines on the digital maps were then corrected so that each line consisted of one pixel in width and all gaps between lines were appropriately bridged.

Attributes of land use type including the information of sewer distribution area, the names of the catchment and old AD were then assigned to each pixel by overlaying digital maps of each subject in the same extent. Three dimensional cross tabulations were developed based on the attributes of pixels so that the numbers of pixels of A (land use type) in B (old AD) within C (catchment) were sorted out.



Lake Kasumigaura consists of the Nishiura (L1) and the Kitaura (L2) with the Hitachi-Tone River (H.T.) in between. Its tributary catchments in this study include the rivers Sakura (1), Koise (2), Sanno (3), Sonobe (4), Kajinashi (5), Hishiki (6), Ichinose (7), Sakai (8), Shin (9), Hanamuro (10), Seimei (11), Ono (12), and Shintone (13) in the watershed of Nishiura; the rivers Tomoe (14), Hokota (15), Takeda (16), Yamada (17), Kura (18), and Taiyo (19) in the watershed of Kitaura; and the Yorokoshi River (20) which flows into the Hitachi-Tone River.

Fig. 1. Location of the study site

Table 1. Some features of tributary catchments in the Lake Kasumigaura watershed

Tributary	Drainage area (km^2)	Population[a)] (capita, head)				Land use[b)] (%)					Catchment No.[c)]
		Human	Cattle	Swine	Poultry	Forest, grassland	Rice paddy	Upland field, orchard	Urban area	Others	
Sakai	14.9	11,118	174	641	553	25.3	12.4	30.3	24.4	7.5	8
Sanno	10.8	24,256	153	1,091	5,953	14.3	14.5	31.5	33.5	6.1	3
Yamada	19.3	3,521	77	9,325	220	30.3	21.8	40.2	6.7	0.9	17
Takeda	19.6	3,723	130	1,403	16,780	36.4	16.6	38.5	7.0	1.6	16
Kajinashi	30.6	6,995	168	2,910	9,999	28.6	17.9	31.6	7.2	14.8	5
Yorokoshi	16.7	4,268	144	253	2,369	40.3	29.5	18.0	7.4	4.9	20

a): Population in 1995.
b): Land use distribution data based on maps developed between 1976 and 1983.
c): Number of catchments in Fig. 1.

3. Estimation of population and sewer use population within an old administrative district

Out of the information used in the calculation of N loads of domestic life, data of total population[30] and sewer distribution rates[11] were only available on the AD basis of the published date and the values on the old AD basis were not available except for 1950. As it was critical to have data on total population and sewer use population in the extent of an old AD, which was identical to that of AD in 1950, those two values had to be converted into the old AD basis in order that data processing in 1960 and later years were compatible with other N sources based on the agricultural census[31] in which all data were compiled on the old AD basis. The conversion procedure was as follows.

The rates of population change for the categories of an old AD[30] (*a*, *b*, *c* and *d*) were computed by solving the following equation on the assumption that the population in an AD of the concerning year was determined both by the populations of 1950 in old ADs which were comprised in the AD and by the rates '*a*, *b*, *c* and *d*' with 'λ' being zero.

$$\begin{bmatrix} x_{1A} & x_{1B} & x_{1C} & x_{1D} \\ x_{2A} & x_{2B} & x_{2C} & x_{2D} \\ x_{3A} & x_{3B} & x_{3C} & x_{3D} \\ \cdots & \cdots & \cdots & \cdots \end{bmatrix} \begin{pmatrix} a \\ b \\ c \\ d \end{pmatrix} + \lambda = \begin{pmatrix} y_1 \\ y_2 \\ y_3 \\ \cdots \end{pmatrix}$$

where

$x_{ij} = \sum P_{ijk}$,

P_{ijk} is population of 1950 in an old AD[30] which was categorized in *j* and comprised in the AD*i* of the concerning year,

i is number of the current AD,

j is category of old ADs[30]; '*A*' as urban area, '*B*' as agricultural area on plain land, '*C*' as agricultural area between plain and mountainous land, and '*D*' as agricultural area in mountainous land;

k is number of old ADs in the same category, *j*, which is comprised in the AD*i* of the concerning year,

a, b, c and d are rates of population change for each category, *j*, for the concerning year, and

y_i is population in AD*i* of the concerning year[30].

Then the resulting '*a*' was corrected by comparing the real data, y_1, with the calculation result, $ax_{1A}+bx_{1B}+cx_{1C}+dx_{1D}$. The correction value of '*a*' (a') was computed as follows:

$$a' = \frac{y_1}{ax_{1A} + bx_{1B} + cx_{1C} + dx_{1D}}$$

The population in the old AD in the concerning year 'x'' was estimated as in the following equation, depending on the category:

x_i' = either of $a' \times a \times x_{iA}$, $b \times x_{iB}$, $c \times x_{iC}$, or $d \times x_{iD}$

Following the calculation above, the sewer use population within the catchment was calculated. Firstly, a list of the current AD within a catchment and another list of the old AD covering each current AD within the catchment were developed. Then the sewer use population in a current AD[11,27] (*nPW*) was written as follows:

$$nPW = R \times nPT$$

where *R* is ratio of sewer use population[11] of the current AD, and *nPT* is total population in the current AD[30].

Next step was determining numbers of pixels consisting of residential area ($nPixT_i$) and sewer distributing area ($nPixWT_i$) in each old AD using the GIS; noting that the symbol '*i*' here represented the number of old AD in the current AD. Those values were decided by overlapping the land use map and the old AD border map. Then values k_i and r_i were calculated for each old AD using these numbers:

$$k_i = nPixT_i / nPT_i$$

$$r_i = nPixWT_i / nPixT_i$$

where nPT_i is total population in the old AD estimated above.

Then the estimated total population in the current AD (nPW') was calculated in the following equation:

$$nPW' = \sum nPW_i = \sum (k_i \times f1 \times r_i \times nPixT_i)$$

where $f1 = R/r$ and $r = \sum nPixWT_i / \sum nPixT_i$.

The sewer use population (nPW_i) and the non-use population ($nNPW_i$) in an old AD were calculated as follows:

$$nPW_i = k_i \times f2 \times f1 \times r_i \times nPixT_i$$

where $f2 = nPW/nPW'$, and

$$nNPW_i = nPT_i - nPW_i .$$

Using the results, populations in the unit pixel for sewer distributing area ($nUPW_i$) and in non-distributing area ($nUNPW_i$) were calculated for an old AD as follows:

$$nUPW_i = nPW_i / nPixWT_i$$

$$nUNPW_i = nNPW_i / (nPixT_i - nPixWT_i)$$

Then a list of populations in the unit pixel for sewer distributing and non-distributing areas was developed for all old ADs after calculations above were made for all current ADs within the catchment. Finally the sewer use and non-use populations within the catchment were calculated as the total sum of these populations in each old AD calculated based both on the number of pixels of sewer distributing and non-distributing area within the catchment and on populations in the unit pixel for each old AD as calculated above according to the number of pixels of sewer distributing area and non-distributing in all old ADs within the catchment area.

It should be noted that the sewer distribution data were based on the maps of the sewer construction plan[27] published in 1991 and the values $nUPW_i$ and $nUNPW_i$ included a certain level of errors in the estimation.

4. Conversion of old AD based data into catchment based data

All variables in the census of agriculture[31] were recorded on an old AD basis. The population was estimated on an old AD basis as mentioned in the first half of the previous section. Then, those variables were converted to the catchment based data by referring distributions of land use attributes of inner and outer parts of the catchment within an old AD. In order to do this, reasonable relationships between each old AD based variable with one or more land use attribute(s) were established such as total population (an old AD based variable) with residential area (a land use attribute), cultivation area of potatoes with upland field, and number of cows with grass land and livestock barn, for example. Values of the old AD based variables were then split into in and out of the catchment proportionally based on the numbers of pixels of relating land use attribute(s) inner and outer of the same extent.

5. Calculation of N load and N potential outflow

In the system, N loads derived from human activities such as domestic life, agriculture and animal husbandry, and from precipitation were calculated by using the modified PLU method shown below, and N potential outflows as amounts of N potentially discharged to the environment on a catchment basis were estimated by considering the N outflow coefficients.

The N loads were calculated on a catchment basis basically every 5 years since 1950 except for 1955 when the issue of the census of agriculture was skipped. The calculation was also made on an annual basis in order for the calculation results to be compatible with information in the census of agriculture[31].

(1) Calculation of N loads from sources

a. Precipitation

N loads from precipitation were estimated by the product of total amount of precipitation in a year and N concentration. The amount of precipitation for the whole catchment was represented by the measurement at the weather station[39] within or near by the catchment. When the data were not available for years of analysis, available data in the nearest year was substituted. 0.008 kg N/mm/ha of N concentration in precipitation was used for the calculation, which was derived from published data[36].

b. Domestic life

N loads from domestic life were divided into two categories; human waste and gray water. Since they were routed differently before being processed and discharged to the environment, they had to be handled as different sources.

The pollutant load units for N from published data[15] were used; 9.0 (g N/capita/day) for human waste and 1.7 (same unit) for gray water.

b-1. Human waste

Three major pathways of human waste transport and treatment were (i) through sewers, (ii) night soil collection, and (iii) treatment in household water treatment facilities (HWTFs) or septic tanks (noting that there are two types of HWTFs; a single type processes only human waste and a combined type processes both human waste and gray water). The sewer use population was estimated by the procedure mentioned previously and the ratios of population using each treatment were estimated from published data[21], and in all cases the N load generation was calculated as a product of pollutant load unit and the population adopting each treatment.

The N routed through sewers was not counted as a load to the soil surface in a catchment because it was transported through sewer pipes from the toilet to the treatment facility directly without being discarded in between. However, as N in treated water was discharged from the treatment facility, the N load should be counted in the catchment where the outlet of the treated water existed.

Most parts of N derived from night soil collection were considered to be treated in a treatment facility and the remainder was discarded into soil without treatments. The ratios of those and distribution of single and combined HWTFs were estimated from published data[21].

b-2. Gray water

Gray water was also differently processed; (i) through sewers or processed in a combined HWTF, which were handled in the same manner as in the previous section, (ii) discarded into house lots, (iii) discarded into agricultural lands, and (iv) discharged into water courses including irrigation-drainage canals, drainage canals and rivers. The ratios of population adopting each treatment were estimated from published data[21]. In each case, the N load generation was calculated in the same way as the case of human waste.

N discharge rates at the treatments[15] were set to 0.68, 0.01, 0.71, 0.58 and 1.00 for N transported through the sewer, collected as night soil and treated in a treatment facility, processed in a single HWTF, processed in a combined HWTF, and discharged to water courses, respectively. Those rates were multiplied by the corresponding N load generation calculated above in order to estimate N loads.

c. Agriculture

N load by fertilizer application was considered as that by agricultural activities. Standard application rates of fertilizer[25] were assumed as the amounts of fertilizer use on each crop type (kg N/ha). The cultivation areas for each crop type (ha) were derived based on the census of agriculture[31] as explained previously. The N loads from fertilizer use for each crop type were calculated as a product of the standard application rate and the cultivation area for the crop.

d. Animal husbandry

The following pollutant load units for livestock waste[9,15] were used; 33.7 (kg N/head/y) and 29.2 for cattle dung and urine, 5.59 and 2.74 for swine dung and urine, and 0.45 for chicken droppings, respectively. Livestock population in a catchment was calculated using data in the census of agriculture[31], except for the broiler chicken population that was estimated from the number of shipped heads in the census[31] by multiplying by 0.2. N load generation was calculated as a product of the population and the corresponding pollutant load unit.

It was assumed that the livestock waste was applied to agricultural land as liquefied slurry or farmyard manure[29]. As it was known that a significant amount of N is lost through volatilization of ammonia gas and through denitrification during storage and the composting process, and just after application of manure and urine to soil[23,26], the N load generation was multiplied by volatilizing coefficients in order to estimate actual N loads on the soil surface as follows: 0.5 for cattle and swine urine[26], 0.23 for cattle dung, 0.149 for swine dung, and 0.447 for chicken droppings[23]. As the information on livestock waste distribution was limited, it was assumed that all livestock waste was consumed within the same catchment of the production.

(2) Calculation of N potential outflow

N load from each source was converted to N potential outflow by being multiplied by N outflow coefficients. N potential outflows on agricultural land were calculated by N loads for a crop type as the amount of fertilizer application being multiplied by the leaching rate for the crop[15,34], except that zero was set as the leaching rate for paddy rice. N potential outflows for other N sources, such as animal waste, precipitation and some parts of N derived from domestic life, on soils including agricultural land, forest land, housing lots and others were calculated as a product of the loads and the outflow coefficient of soil, 0.45[15,18]. On the other hand, N outflow coefficients for N loads that were not applied on soil but discharged to water courses out of those derived from domestic life were set to 1.0.

The total N potential outflow in the catchment was calculated as the sum of the potentials of all sources, except for the potential of N transported through the sewer because the loads should have been released at the site of the sewer treatment facility which usually existed outside the catchment.

6. Estimation of actual N outflow at the monitoring point in a river

Annual N outflow in a river was estimated using national scale water quality monitoring data such as the National Water Quality Monitoring Data Base[10], and meteorological data such as AMeDAS (Automated Meteorological Data Acquisition System[39]). The annual N outflow in a river (kg N/y) was calculated as a product of annual volume of water flow (Q, 10^3 m^3/y) and annual average concentration of total N (mg N/L). Annual volume of water flow was estimated as in the equation:

$$Q = (P - Ep) \times A$$

where

P is annual precipitation (mm/y),
Ep is annual evapotranspiration (mm/y), and
A is catchment area (km^2).

Annual precipitation was calculated using data of AMeDAS and annual evapotranspiration was calculated from daily evapotranspiration estimated by the Hamon method[3]. Annual average concentration of total N in the river water was computed as a mean of measurements of total N concentration throughout the year.

Results and discussion

1. Verification of adequacy in estimated catchment-based variables

Some of the features of the 6 tributary catchments within the Lake Kasumigaura watershed estimated from the developed methods are shown in Table 1. To verify the outputs of our method, we compared the estimates by the present method with those by other methods and published data (Fig. 2).

First, total drainage areas of 18 tributary catchments were compared with the values listed in the original watershed border map[22] (Fig. 2, a). The correlation coefficient (r) of two sets of drainage areas was 0.998 ($n = 18$, $p < 0.001$) and the slope of the regression line was almost 1.0, suggesting that the calculation by the present method successfully reproduced the drainage areas of their original maps. Secondly, concerning the estimates of cultivation areas[19] and livestock productions[14], total cultivation areas for all crops in 12 catchments in 1970 by the present method were compared with the estimates of Moriizumi et al.[19] and were shown to be closely related with a significant correlation coefficient: $r = 0.994$ ($n = 12$, $p < 0.001$), and with slopes of the regression line being nearly 1.0 (0.953) (Fig. 2, b-1). Although some of the crop types showed lower fittings compared to the 1:1 lines and the estimated number of head of swine in one catchment had to be omitted as an outlier (Fig. 2, c-2), the correlation coefficient of each variable was relatively high with the lowest ($r = 0.805$, $p < 0.01$) in cultivation areas of tubers and roots (Fig. 2, b-3) and the highest (0.997, $p < 0.001$) in areas of paddy rice (Fig. 2, b-2). Thirdly, total populations in 14 catchments in 1970 estimated by the developed method were compared with the corresponding populations by Hayashi and Shio using statistics on a grid square basis[6] (Fig. 2, d). The correlation coefficient was 0.983 with 0.931 as the slope of the regression line, suggesting significantly high correlations between the two sets of estimates ($p < 0.001$).

Thus, most of the outputs by the methods showed good correlations with slopes of the regression lines being nearly 1.0, when compared with the estimates by different methods. Unfortunately, we did not verify the adequacy of the estimate in sewer use population in a catchment, since there seemed to be no other trustworthy report referring to it. However, we concluded that the developed methods have sufficient capability of computing reasonable estimates in a drainage area and other variables within a catchment.

In addition, another advantage of the methods should be emphasized and that is utilizing national scale statistics enables estimations to be made of the same quality throughout Japan because of the uniform and good availability of the data.

However, it should be noted that during the series of analysis, the same GIS map data which were developed based on the printed maps published between the 1970's and 1990's were used except for a set of maps on old administrative district borders which were published in the 1950's. Although especially distribution of land use might have changed through the years of analysis among catchments, we used the same land use maps published between 1976 and 1983. This might have led to a certain level of errors in estimations of variables because their calculations were based on distribution of land use in and

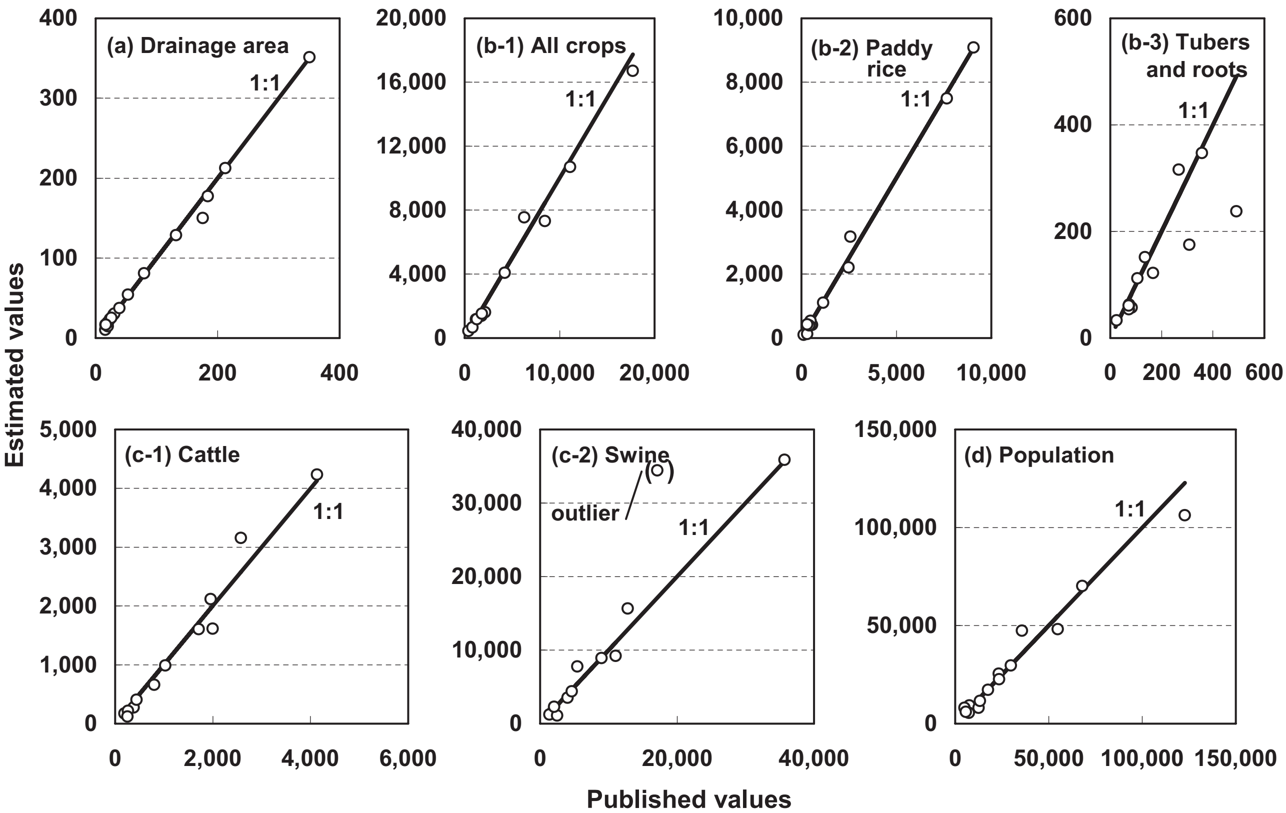


Fig. 2. Comparison of variables in tributary catchments within the Lake Kasumigaura watershed between published data and those estimated by the developed method

Solid line in each graph indicates a 1:1 relationship.
(a) Drainage area (km^2), n = 18, reference 22.
(b) Cultivation area (ha), n = 12, reference 19.
(c) Number of head of livestock (head), n = 12, reference 14.
(d) Population (capita), n = 14, reference 6.

out of a catchment. We did not verify the accuracies of those estimations on dates different from the date of the development of the maps, because of a lack of appropriate published data.

2. N potential outflows based upon the modified PLU method

Nitrogen potential outflows were calculated by the present modified PLU method. Fig. 3 shows the typical time courses in N outflows of 6 catchments out of 20 catchments. Table 1 also shows the characteristics of these catchments. In Fig. 3, the potential outflows from each source are shown as the differences of adjacent lines so that total potential outflows are shown as the uppermost line of each graph during the period between 1950 and 1995.

Fluctuations in total potential outflow seemed almost coincident with the increase and/or decrease of potential outflow from animal husbandry (e.g. Fig. 3, a, b, c & d) and/or domestic life (e.g. Fig. 3, e & f), while N potential outflow from other sources seemed relatively stable. The fluctuations in potential outflow from animal husbandry were mainly due to the increase and decrease in livestock population, while those from domestic life were due to the changes in methods of waste water treatment rather than population changes. For example, a slight increase in potential outflow of domestic life after 1980 (Fig. 3, a, b, c, d & e) reflected the increased distribution of HWTFs instead of night soil collection, resulting in increased discharges of treated water to local water ways, while significant reduction of N discharge was achieved by the higher sewer distribution rate in the catchment of Sakai River (92% in 1995, Fig. 3, f). Thus, the present modified PLU method was successfully able to picture changes in N load generation activities and N potential outflows in these catchments.

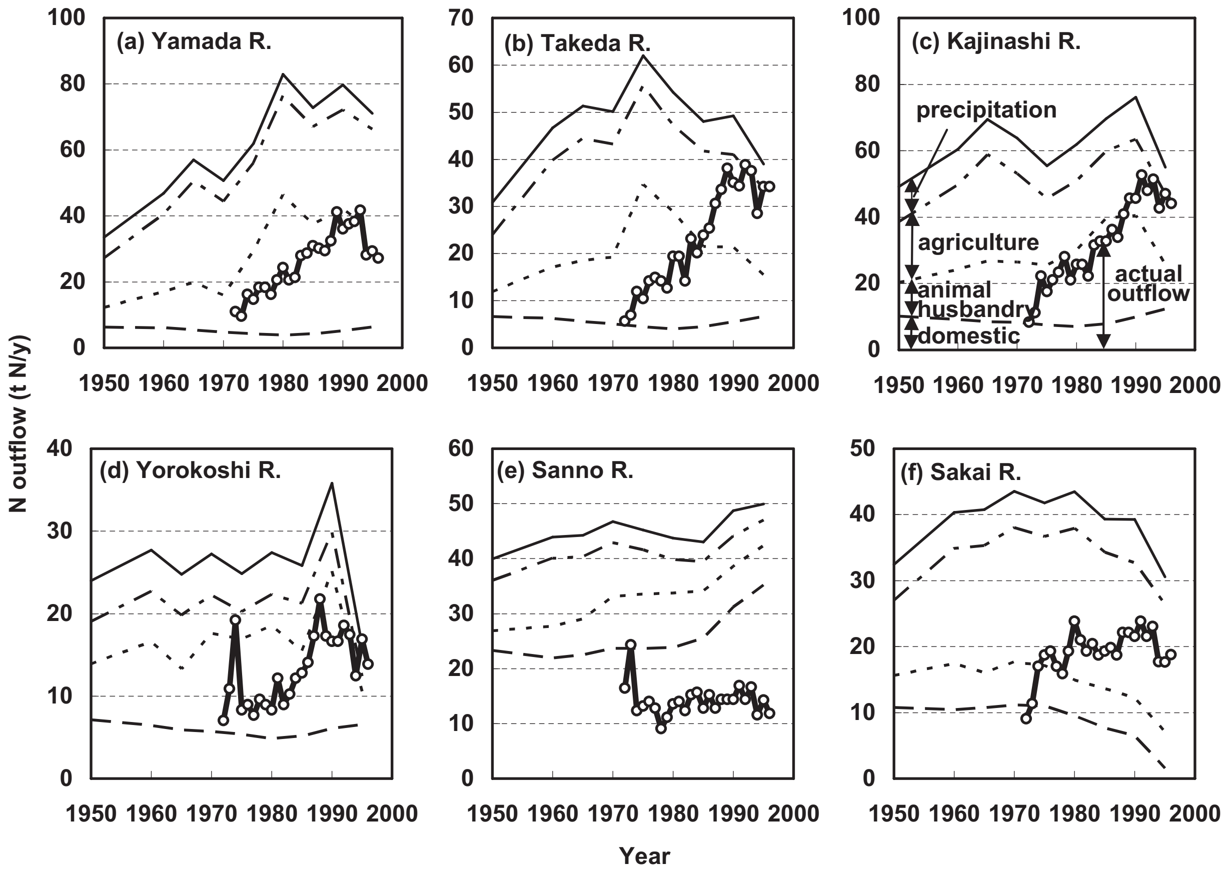


Fig. 3. N potential outflows in a catchment and actual outflows in the corresponding river

The differences between each line show the amounts of the potential outflows from each source: — — —, domestic life ; - - - -, animal husbandry ; — - —, agriculture ; ———, precipitation.
—o—: The actual outflows in the corresponding river.

3. Comparison between the total potential outflow and the actual outflow

The actual outflows were calculated after 1971 when the national water quality monitoring data[10] became available. These are shown as solid lines in Fig. 3. The most striking trend in Fig. 3 is the distinct differences between the total potential outflow and the actual outflow. It is known that not all N loaded in a catchment was recovered at the outlet because of several functions of a catchment; such as conversion of N to gaseous forms through denitrification and/or N storage in biomass, groundwater or soils of the landscape[2].

Out of 20 catchments processed, 11 catchments showed distinct peaks in both the total potential outflow and the actual outflow (e.g. Fig. 3, a, b, c & d), while either of the two peaks could not be detected in the rest of the 9 catchments (e.g. Fig. 3, e & f). In some out of the 11 catchments showing two distinct peaks, the peak of the total potential outflow tended to be followed by that of the actual outflow; i.e. a time-lag between the two peaks appeared (Fig. 3, a & b). Such a time-lag in the actual outflow in a catchment was also found in another study[37], and the involvement of N accumulation in soils was suggested for the time-lag[37]. Length of time-lags may depend on the storage site and its capacity in the soil-groundwater system in a catchment. This is further discussed in the following section.

Most catchments with no distinct peaks detected in either the total N potential outflow or the actual outflow showed wider and irregular fluctuations in the actual outflows (Fig. 3, e & f). In these catchments industrial area occupied more than 3% of the whole catchment area according to the land use distribution data. These were also characterized by higher urban area ratios in catchments of Sakai and Sanno Rivers (Table 1). As the water budget in the catchments may be affected by industry as well as the additional N load[33], the irregular fluctuations in the actual outflows might have been caused by industrial activities, which were not involved in the calculations in this study. This suggests that involvement of industry and other point sources in the estimation of water budget and N load may contribute to improve the accuracies of estimations in the actual outflow in catchments with higher industrial activities. For the other catchments with lower industrial area ratio, we think that the estimation of the actual outflow without counting the point sources can be reasonable because the average contribution of industry to the N load in the whole Kasumigaura watershed was reported to be very limited (1.9% in 1996[16]).

4. Length of time-lag in outflow

The dynamic behavior of N in soil, due to such factors as storage and loss through denitrification, has been suggested to be a possible cause of the delayed appearance of N in the actual N outflow[37] and the difference in the heights of the two peaks[2]. In order to make the relationships between these two outflows clearer, the amounts of N which flow directly into a water body or onto pavement were subtracted both from the total potential outflows and from the actual outflow. This corrected potential outflow can be considered as outflows through the soil surface. As a result, 13 out of 20 catchments showed peaks in both N outflows and the length of the time-lag varied between 0 and 25 years (Table 2). The delayed appearances of the peaks in the actual N outflows originating from N applied to the soil surface were obvious, but the lengths of the time-lag became somehow different from those in Fig. 3.

It was reported that the successive and/or heavy application of organic N to soil accelerated N accumulation in the soil more than no or less application of organic N[12]. However, the relationship between the length of the time-lag and the rates of organic N application on the soil surface, which were estimated as the amount of N derived from livestock waste divided by total area of agricultural land use in a catchment, was not clear. This estimation in the rates of organic N application may not be precise enough in that we did not count other options relating to the fate of livestock waste such as the methods of the treatment process. In addition, the inter-catchment distribution of livestock compost has been growing recently[29]. These suggested the importance of considering other factors concerning N re-distribution in terms of regional circulation of compost as well as the N dynamics in the soil-groundwater system.

5. Difference in N removal through the outflow process

In order to evaluate the possible N removal activities in the catchments, the amounts of the potential outflow through the soil surface and the actual outflow at both peaks (Table 2) are plotted in Fig. 4. The relationship was significantly linear ($r = 0.993$, $p < 0.001$, $n = 13$) and the slope was 0.471. This suggested that 47% of N potential outflow which had originated from N load through the soil surface, actually flowed out to the outlet of the catchment on the river, and the rest was considered to be removed somehow before reaching the outlet. This value was regarded as the "N reaching coefficient" (Table 2) and the difference from 100% was considered as the "N removal ratio" in a catchment.

Concerning the magnitude of the N removal ratio,

Table 2. Time-lags and N reaching coefficients in 13 catchments within the Lake Kasumigaura watershed based upon the estimations of the N load applied to the soil surface

Tributary	Catchment No.[a)]	Peak in potential outflow through soil		Peak in actual outflow through soil		Time-lag	N reaching coefficient
		Year[b)] (Y1)	Amount[c)] (t N/y, A1)	Year (Y2)	Amount (t N/y, A2)	(y, Y2−Y1)	(%, A2/A1×100)
Hishiki	6	1980	79.9	1990	31.0	10	38.7
Hokota	15	1990	220.2	1995	103.8	5	47.1
Ichinose	7	1980	98.3	1990	37.9	10	38.5
Kajinashi	5	1990	69.8	1990	39.3	0	56.3
Kura	18	1980	61.5	1990	39.0	10	63.4
Sakai	8	1970	41.7	1980	20.5	10	49.0
Sakura	1	1965	785.0	1980	384.3	15	49.0
Shin	9	1980	37.1	1980	14.6	0	39.3
Shintone	13	1960	287.6	1985	96.1	25	33.4
Takeda	16	1975	60.8	1990	31.7	15	52.1
Tomoe	14	1985	447.4	1995	211.6	10	47.3
Yorokoshi	20	1990	32.0	1990	12.8	0	40.1
Yamada	17	1980	81.5	1990	32.8	10	40.2

a): Number of catchments in Fig. 1.
b): Year the peak appeared.
c): Amount of N outflow at the peak.

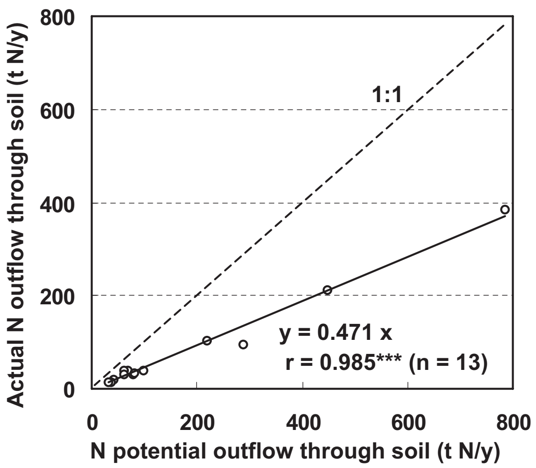


Fig. 4. Relationships between the amounts of N potential outflows and that of the corresponding actual N outflows at both peaks, originating from N applied to the soil surface

The X axis in the figure represents "N potential outflow originating from N applied to the soil surface", computed as a subtraction of a portion of N potential outflow which was discharged on water and pavement from the total sum of N potential outflows. The Y axis represents "actual N outflow originating from N applied to the soil surface", computed by a subtraction of a portion of N potential outflow which was discharged on water and pavement from the total actual outflow.

involvement of several functions was suggested as mentioned above[2], but it seemed difficult to separate contributions of individual functions to the whole N removal ratio. Among the N removal functions, denitrification activities were found to be significant in submerged soils such as wetlands[38], riparian zones[7], and also in groundwater[28]. In the study area, rice paddy soil, which was also known to remove N during the rice growing season through denitrification and plant uptake[5,24,32,33], was predominant (Table 1). Therefore, the relationship of the rice paddy area ratio was analyzed with the N reaching coefficient in order to assess the possible N removal activities in paddy soil. The relationship between them showed a weak and negative correlation ($r = -0.57$, $p < 0.05$, $n = 13$, data not shown), suggesting that the increase in rice paddy area tended to enhance the N removal from the catchments and that rice paddy fields may be one of the factors dominating the magnitude of the N removal ratio through denitrification and/or plant uptake. Further analyses suggested one of the important factors governing the rate of N removal is travel time of water through paddy or riparian areas[35] and an analysis including this aspect would help to explain the magnitude of N removal activity in the catchment. Also, other factors concerning denitrification in other sites as well as N storage in a catchment should be evaluated in further studies.

In conclusion, the present modified PLU method made possible an estimate of the potential outflow on the scale of a catchment. Quantitative analysis of the relationship between the potential and the actual N outflows reflected the characteristics of N outflows in various catchments around Lake Kasumigaura. Further study from different aspects and/or through comparison with more catchments in other regions may be of value to understand the fate of N loaded in a catchment.

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