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SYNTHESIS AND EMERGING IDEAS

Nitrogen budget and gaseous nitrogen loss in a tropical agricultural watershed

Natsuki Yoshikawa · Sho Shiozawa · Ardiansyah

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Abstract Although agricultural systems in tropical monsoon Asia play a central role in the global nitrogen (N) cycle, details of the N cycle in this region on a watershed scale remain unclear. This study quantified the N budget in a tropical watershed of 221 km^2 on Java Island, where paddy fields cover 28% of the land, by conducting field surveys. The amount of net biochemical gaseous N loss to the atmosphere (X_{GB}), which is generally difficult to determine, was calculated as the residual of the N balance. Assuming that NH_3 volatilization balances deposition, and hence subtracting NH_4–N from the N import with atmospheric deposition, the average total import and export of N per year was found to be 46.5 kg ha^{-1} year^{-1} over the watershed. Of this, 71% was imported as fertilizer (M_F) and 29% with atmospheric deposition (M_{AD}). On the export side, 42% was lost as X_{GB}, 37% with incineration of rice residues and wood fuel (X_{GI}), 13% with river discharge (X_D) and 9% with rice surplus export (X_R). A large portion of X_{GB}, and consequently, a small portion of X_D could be explained by the high rate of denitrification resulting from the high temperature and humid climate, and are thought to be common features of tropical watersheds where paddy fields are found.

Keywords Nitrogen budget · Tropical watershed · Gaseous nitrogen loss · Denitrification · NH_3 volatilization

N. Yoshikawa (✉)
Research Center for Natural Hazards and Disaster Recovery, Niigata University, 8050 Ikarashi 2-no-cho, Niigata 950-2181, Japan
e-mail: natsuky@agr.niigata-u.ac.jp

S. Shiozawa
Department of Biological and Environmental Engineering, The University of Tokyo, Tokyo, Bunkyo-ku, Tokyo 113-8657, Japan

Ardiansyah
Graduate School of Agricultural and Life Sciences, The University of Tokyo, 1-1-1 Yayoi, Bunkyo-ku, Tokyo 113-8657, Japan

Introduction

Human activities have considerably altered the global cycle of nitrogen (N) through artificial N fixation and mobilization. The most substantial human-induced perturbation of the N-cycle is attributed to agricultural activities, with exponentially increasing production and application of fertilizer (Schlesinger 1992; Gundersen et al. 1994; Vitousek et al. 1997; Downing et al. 1999; Galloway et al. 1995).

Agricultural systems in tropical monsoon Asia play a central role in the global N cycle. Since the green revolution of the 1970s, this region has been experiencing the most rapid agricultural modernization in the world. N fertilizer consumption has increased approximately 8-fold since 1970, and today this area represents 23% of the world consumption (FAOSTAT 2005). Combined with the recent increase in synthetic fertilizer use, the tropical climatic regime experiences N cycling at a higher rate than in temperate regions; high temperatures and moisture result in a high turnover of biomass and high microbial activity, leading to fast decomposition of organic matter and fast transformation of N, and high N exchange with the atmosphere. Abundant precipitation also contributes to the rapid N cycle through hydrological pathways. Despite its importance, however, the N cycle of tropical monsoon Asia has so far been treated with great uncertainty in studies concerned with tracing global N cycles. This is in part due to the limited number of studies conducted in tropical areas, and a lack of statistical and other informative data (Matthews 1994 in the context of atmospheric N emission; Bouwman and Boumans 2003 in the context of hydrological N transport).

Various studies and models have attempted to predict the N load of rivers and lakes by dynamically calculating the N load from each type of land use with given parameters (e.g. Beaujouan et al. 2001; Payaudeau 2001; Kato et al. 2002). Generally, it is not difficult to accurately estimate or predict N input and output due to human activities (fertilizer application, harvest, etc.); however, N loss to the atmosphere due to natural biochemical processes is difficult to determine. Although N output from rivers is easy to calculate from the river discharge and N concentration, no-one knows how much N is lost to the atmosphere, which may depend largely on the climate and land use practices. If the amount of N loss to the atmosphere due to natural processes is significant, it will also have a major effect on determining the N load of river water. In general, measurements of the actual N loss to the atmosphere at a watershed scale can only be realized by accurately estimating all the N input and output components in the N cycle in a watershed and determining the residual between the two, in the case where changes in N stock can be neglected when compared to the amounts of input and output components.

Various studies have conducted N budget analysis at the watershed level in temperate regions such as European countries and the USA, where N contamination of water has long been a social issue. However, some of these studies focus solely on the water quality of rivers, without considering N losses from the watershed (e.g. Smith 1977). Moreover, many studies acknowledge N losses but are not able to determine the detailed components, and thus fail to quantify gaseous N loss (e.g. Roberts 1987; Bechmann et al. 1998; Vuorenmaa 2002; Mcisaak et al. 2004). A number of other studies have attempted to estimate or guesstimate the flux of gaseous N loss (mainly through denitrification) using literature values (e.g. Houston et al. 1981; Freifelder et al. 1998; Kronvang et al. 1999; Quynh et al. 2005). However, the main causes for the failure in estimating the gaseous N loss in these studies resides in the nature of the watersheds or basins. Three major sinks of N are often identified, namely soil and groundwater storage, biomass storage, and gaseous emission to the atmosphere, but are not successfully separated.

The objective of this paper is to quantify the N budget in a tropical watershed lined with paddy fields, focusing specifically on the contribution of gaseous N loss to the total N export. Factors of N import and export were individually quantified using various methods and available data: river water sampling and analysis, intensive interviews with farmers, local meteorological and hydrological data, local and national statistical data, and analysis of satellite remote sensing images. Among the factors for N export, gaseous N loss is difficult to quantify on a watershed scale, though it is expected to be significant. In earlier studies on watershed N budgets, large uncertainties in gaseous N loss remain due to the lack of information on climatic, ecological and agricultural variables (Matthews 1994; van Breemen et al. 2002) as well as the complicated nature of N transfer within the watershed among each contributor of N. In this study, although the quantity of gaseous N loss was calculated as the residual between N import and export, we attempted to quantify this value by obtaining accurate values of all the other components in the N-budget. The study watershed, Cidanau watershed, Banten province, Indonesia, is suitable for such a study in that its hydrosphere and biosphere are completely segmented and isolated by the outer rim of a caldera, the geological formation of the watershed. Therefore, N exchange between the surrounding area and inside the watershed is limited. These factors made calculation of the N budget of the whole watershed simple, and hence minimized uncertainties, although the N cycle within the watershed is complicated and difficult to specify quantitatively.

Study area

The study was carried out in Cidanau watershed, situated on the northwestern tip of Java island (6°8′ S–6°17′ S latitude, 105°52′ E–106°03′ E longitude), Indonesia. Western Java has a tropical monsoon climate characterized by two pronounced seasons, dry and rainy. The dry season extends roughly from April to mid-October, and the rainy season from mid-October to March. The annual mean atmospheric temperature in Cidanau watershed is 26.5°C, and does not vary significantly throughout the year. The mean annual precipitation during 1995–2004 was approximately 2,600 mm fluctuating between 1,000 and 3,900 mm.

The study watershed is a 221 km^2 drainage basin with forest (58%), paddy fields (28%) swamp forest (5%), swamp (4%) and residential area (5%) (Fig. 1).

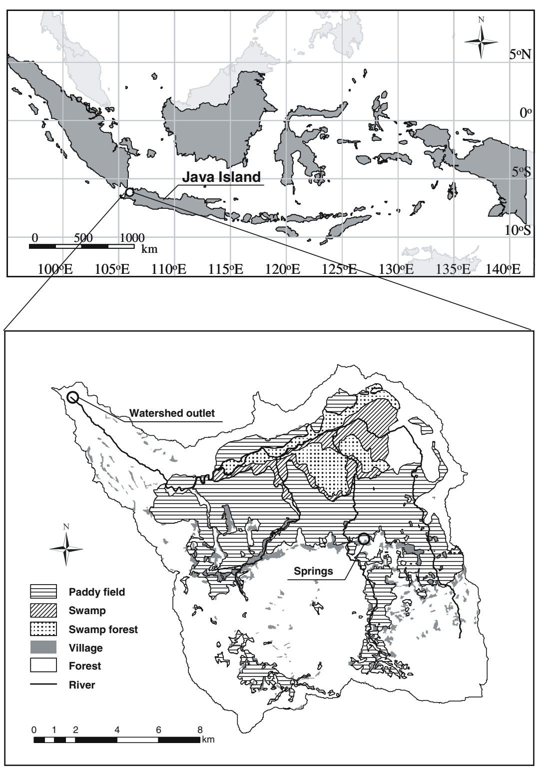

Fig. 1 Map of the Cidanau watershed

Satellite remote sensing analysis indicated that these land use proportions have not been subjected to significant changes from logging or the development of agricultural fields since the early 1970s (Tsuyuki et al. 2003).

Cultivated fields in the area consist predominantly of rainfed paddy fields with a plot-to-plot irrigation system. Although there are some minor irrigation canals, beneficiaries of such facilities are rather limited. The average yield per crop in this watershed, 3.2 ton ha^{-1} $crop^{-1}$ based on our survey, is lower than that of West Java where the average is 5.3 ton ha^{-1} $crop^{-1}$ (Makarim 2000); however, this low yield is compensated for by the high crop intensity, roughly 200% (Yoshikawa et al. 2006), owing to the availability of water.

The watershed was originally a caldera of the Danau volcanic complex, bounded by a caldera rim and adjacent high volcanic terrain. Because of this unique topography, not only the hydrosphere but also the biosphere is well segmented and isolated, thus restricting transportation of agricultural products across the boundary.

The Cidanau River, which functions to drain the entire watershed, flows out from the caldera through a waterfall in a narrow valley located in the outer rim of the caldera, which then flows into the Sunda Strait. Four major tributaries join the Cidanau River, of which two stemming rivers, the Ciomas and Cisuwarna, flow through the Rawa Danau swamp area. There is an intake weir owned by PT. Krakatau Tirta Industri (hereafter referred to as KTI), the local water supply company, located near the mouth of the Cidanau River, from which runoff discharge is observed on a daily basis. KTI distributes water to all industries in Cilegon city as well as domestically to Serang and Cilegon cities.

Materials and methods

N budget model for the Cidanau watershed

In general, mass conservation of N in any system is described by

$$\Delta S = M - X \tag{1}$$

where M is the mass of total N import into the system during a specified time period, X is the mass of total

N export out of the system, and ΔS is the change in the mass of N within the system during the same period. The N budget model quantifies specific factors that compose the flow terms of M and X, and storage changes of ΔS in the watershed. According to our field survey, we considered a N budget model for the study watershed that lists the pathways of N import into and export from the watershed, as presented in Fig. 2.

Total N import into the watershed (M) consisted of two factors, chemical fertilizer input (M_F) and atmospheric deposition (M_{AD}) consisting of wet deposition (M_{WD}) and dry deposition (M_{DD}). In the study watershed, chemical fertilizer is applied in the form of urea only for rice growing in paddy fields, while crops in upland fields received very little fertilizer. Therefore, M_F could be determined from the total area of cultivated paddy fields in a year in the watershed and the amount of fertilizer input per unit area of paddy field per crop cultivation.

For the term of atmospheric deposition (M_{AD}), the value of wet deposition (M_{WD}) was determined as the product of the total nitrogen (TN) concentration and the amount of precipitation in a year, excluding NH_4–N content and including nitrate and organic N content, from the result of rainfall water analysis. NH_4^+ content in rainfall was excluded from M_{WD} because we assumed that all NH_4–N content in rain is returned as NH_3 volatized from the watershed; hence, it should not be counted as N input when determining the N budget in the whole watershed. This point will be discussed later.

The measurement of dry deposition (M_{DD}) was not conducted in this study. However, we attempted to specify its value using the proportion of wet and dry N depositions observed in West Java cited from the literature.

Total N export (X), on the other hand, was considered to occur in four forms, namely riverine transport (X_D), export of rice produced in the watershed (X_R), gaseous N loss attributed to incineration of crop residues (X_{GI}), and net gaseous N loss attributed to biochemical processes (X_{GB}).

X_D was calculated from the product of the TN concentration of river water and discharge at the watershed outlet. X_R is N export represented by the sale of rice outside the watershed. Rice production is more than self-sufficient in this watershed, and rice surplus to that consumed by the residents is exported.

Two terms represent gaseous N loss to the atmosphere, X_{GI} and X_{GB}. X_{GI} results from the incineration of biomass or anthropogenic processes. Most farmers burn crop residues such as rice straw and hull as waste from paddy fields and rice mills. The N content of these residues is released into the atmosphere through combustion processes mostly as N oxide (NO) and N dioxide (NO_2), often referred to as NO_x. Incineration of woodfuel is another process included in X_{GI}.

X_{GB}, on the other hand, represents net gaseous N loss due to biochemical processes, and may be composed of two biological processes: denitrification, which positively contributes to X_{GB}, and biological N fixation, which negatively contributes to X_{GB}. X_{GB} is

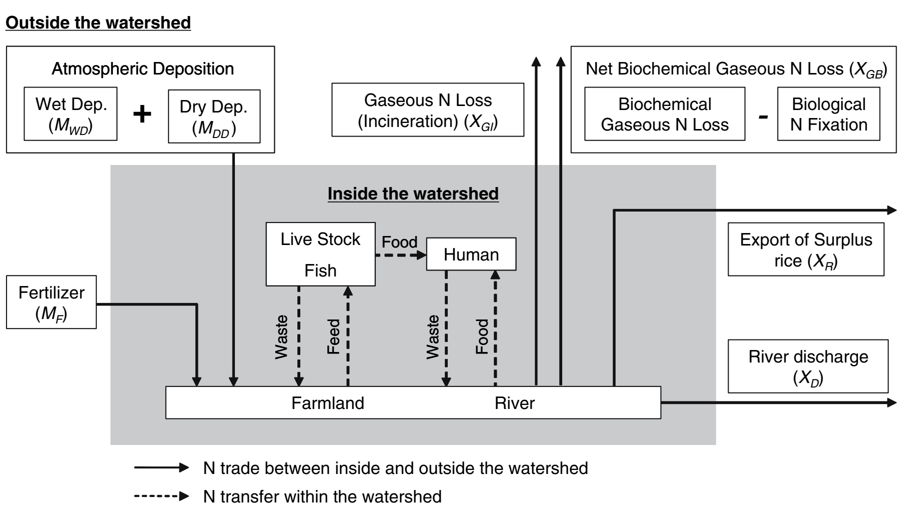

Fig. 2 The N budget model of the study watershed

a net value of N_2 gas exchange between the watershed and atmosphere and was not directly measured but determined as the residual between the sum of N import and measurable N export terms.

N transport and transformation occurring inside the watershed through human activity is very complicated, as shown in Fig. 2, with several pathways of N transfer involving humans, livestock/fish, and farmland/river. About 100,000 people residing inside the watershed rely on the area for food and subsequently exhaust N, as do the thousands of cattle, goats, and chickens. It is essentially very difficult to comprehend all these N transfers since no reliable statistical data for calculating them exist. Nevertheless, the N budget model could be simplified here owing to the characteristics of the study watershed, i.e. when solely considering the trade between the watershed and the outside world. That is, according to our survey, this watershed is basically self-sufficient with respect to food and feed. Moreover, rice production is more than enough to feed the entire population of the watershed, and hence no rice is imported. Furthermore, livestock including cattle, chickens, sheep and goats are grazed in and around villages, and no commercial feed is used. Feed for fish in fish breeding ponds is also provided from within the watershed making use of locally produced crop residues. Consequently, all crop and animal products produced in the watershed, except for the fractional rice export, are consumed within the watershed, and therefore, have no effect on the N balance of the area. As a result, human N transport between the watershed and the outside world occurs only in the form of fertilizer import, rice surplus export and incineration of crop residues and wood fuel.

When conditions of the watershed that affect M or X (population, fertilizer application, land use/cover, etc.) have become almost constant for a considerable time period, N flow reaches an almost steady state. That is, ΔS in Eq. 1 becomes much smaller than the flow terms of M and X in the long-term, with M and X being almost proportional to time while ΔS can only fluctuate seasonally. Thus, M and X become balanced and Eq. 1 is reduced to

$$M = X \tag{2}$$

While we maintain that Eq. 2 is the basic condition of the watershed, evaluation of ΔS will be discussed later.

Field survey and data for calculating N-budget

Six field surveys were conducted between 2003 and 2005. All consisted of collecting river water samples, measuring discharge rates at each sampling point, collecting ground truth data for satellite remote sensing analysis, and interviewing randomly selected farmers with regard to the acreage of their paddy field holding, the quantity of N fertilizer application, and treatment of rice residues.

River water samples were obtained from a total of 87 locations throughout the watershed on different days with various water discharge rates. Total N was determined using a Shimadu Total N Analyzer, and NO_3^- NO_2^- and NH_4^+ concentrations using a Hitachi ion chromatograph in our laboratory.

All annual data for the N budget model were consistently calculated based on the water year defined as the 12-month period from September 1 through August 31 of the following year, since typically the lowest stable river water flows occur between August and September, in mid dry season.

Daily precipitation data between 1995 and 2004 at rain stations in and around the watershed were obtained from the Water Resources Management Agency (BPSDA), Serang office, Banten province, Indonesia. Representative daily precipitation over the entire watershed was then calculated using data from the four rain stations closest to the watershed, and weight-averaged by applying the Thiessen polygon method. The annual bulk precipitation of the watershed in a certain water year t (P_t) is represented by the sum of daily precipitation in year t multiplied by the watershed acreage (here: 220 km^2).

We used the Cidanau River daily flow data for 1995 through 2004 water years measured by KTI at the pumping station, which is located at the outlet of the watershed (Fig. 1), to determine daily and annual water volumes for the watershed.

Gross acreage of paddy field under cultivation (GPA) is a crucial measure for calculating the N budget because it is necessary for determining M_F, X_R and X_{GI}. However, in the present study, we were not able to obtain reliable statistical data. Estimating GPA is not easy in tropical regions where temperatures are high and constant all year round, and rice can be sown and grown throughout the year, thus making triple cropping feasible as long as water is available. In fact, GPA in the watershed varies widely within a year and among years depending on the variability of water. Yoshikawa and Shiozawa (2006) made an estimation of the cultivated paddy acreage in the study watershed using eight satellite images acquired between 1991 and 2004. They subsequently found a strong linear correlation between the estimated value and preceding cumulative precipitation for 90 days up until the day of the estimation (Fig. 3). By applying the obtained relationship, the average cultivated paddy field acreage was calculated from daily precipitation data for 1995 through 2004.

The amount of N fertilizer input per unit area (F) and unit rice yield (y) were obtained through interviews with 307 farmers residing in the watershed. Average results were then calculated by weighting the findings with the size of the land holding of each farmer, which varied widely from 0.06 to 5 ha. The weighted average of N fertilizer input was found to be 144 kg ha^{-1} crop^{-1} and that of yield to be 3.2 ton ha^{-1} crop^{-1}. Ratios of incineration treatment of rice straw were also obtained through interviews with 103 farmers and averaged in the same way. The resulting ratio of incineration was 86% in the dry season and 21% in the rainy season. To determine rice consumption per capita, we employed the national average value of Indonesia, 149 kg year^{-1}, obtained from the literature (Hossain 2005).

The watershed population was obtained from the local office of Badan Pusat Statistik (BPS-national statistics bureau) for 1996–1998, and 2002. For the

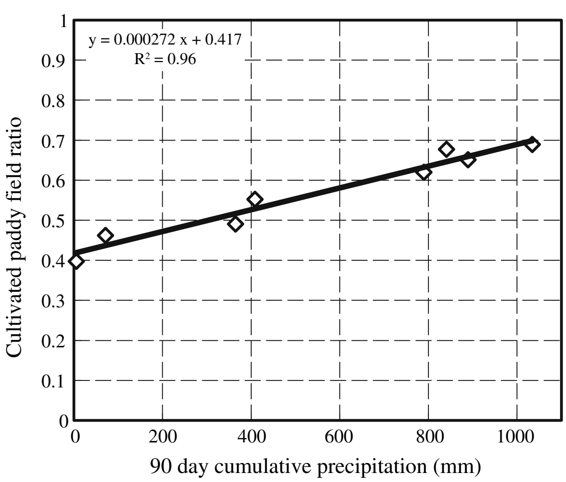

Fig. 3 Correlation between 90-day cumulative precipitation and the cultivated paddy ratio of the paddy field area in the watershed

missing years, the population was interpolated assuming constant growth. Accordingly, the watershed population was 103,000 in 1996 and 114,700 in 2004 (population density of approximately 520 people km^{-2}, which is a typical density in rural areas in Java, the most populated island in the world). Since population growth is relatively slow in this area (approximately 1% per annum), we used the average population over a 10-year period (111,200 people).

Method for calculating N-budget components of N import

N import through fertilizer application

To calculate M_F, as well as X_R and X_{GI}, we first estimated the annual GPA by applying the regression function obtained by Yoshikawa and Shiozawa (2006) from the analysis of satellite images, as mentioned earlier. The areal ratio of cultivated paddy fields to total paddy fields on a day i is expressed as a function of the 90-day cumulative precipitation prior to day i (Fig. 3):

$$f(P_{90i}) = 0.000272P_{90i} + 0.417, \quad f(P_{90i}) \leq 1 \tag{3}$$

where $f(P_{90i})$ is the ratio of cultivated paddy fields to total paddy fields on arbitrary day i, and P_{90i} is the preceding 90-day cumulative precipitation. This equation indicates that 2.7% of additional paddy field area is cultivated with every 100 mm increase in cumulative rainfall, while roughly 42% of the paddy field area is cultivated even with a preceding 90-day cumulative precipitation of zero. Using the value of $f(P_{90i})$, annual gross cultivated paddy acreage in water year t (GPA_t) is calculated as

$$GPA_t = A \cdot \frac{1}{120} \sum_{i=1}^{365} f(P_{90i}), \quad f(P_{90i}) \leq 1 \tag{4}$$

where A is the total paddy field area (ha) and 120 is an approximate rice growing duration in days.

Accordingly, M_F was calculated by multiplying the GPA by the average amount of N fertilizer application per ha per crop (F).

N import through atmospheric deposition

The amount of N imported from wet deposition (M_{WD}) was calculated by multiplying the average annual bulk precipitation (P) of the watershed by the sum of the average NO_3–N and organic N concentrations of rainfall samples, or TN minus the NH_4^+ component contained in rainfall samples assuming that all the NH_3 volatilized from the watershed is redeposited onto the watershed.

To estimate the value of dry deposition (M_{DD}), we employed a proportion of wet and dry N deposition cited from the literature. Fortunately, Gillett et al. (1999) have estimated dry deposition at two sites (Jakarta and Bogor) located in West Java, of which the Bogor site has a similar wet N deposition value to that we measured, a similar climate (average annual precipitation of 3,000 mm) to our study area and similar land use as their observatory station is surrounded by paddy field. According to their study, wet deposition of NO_3^-–N was estimated to be 12.7 kg N ha^{-1} year^{-1} (NO_3^-: 49 meq m^{-2} year^{-1}), and dry deposition of NO_2–N to be 2.8 kg N ha^{-1} y^{-1} (NO_2^-: 20 meq m^{-2} year^{-1}) and HNO_3 to be 1.1 kg N ha^{-1} year^{-1} (HNO_3^-: 8 meq m^{-2} year^{-1}). The proportion of dry deposition was 24% in the total atmospheric deposition. Another experimental study, conducted in a rural area in Japan during the warm and rainy season, also demonstrates a similar proportion of 26% (Hayashi et al.). From these studies, it is plausible that the dry N deposition in our study site contributes approximately one-quarter to the total atmospheric N deposition. Although there is very little information on organic N in dry deposition, we assumed that the same proportion of organic N is deposited.

Components of N export

N export through river discharge

River water samples were collected at the outlet of the watershed five times with different discharges. Although daily river discharge data were available, TN concentration data were limited to those measured at the time of our field surveys. Since the TN concentration of river water is likely to vary depending on the river discharge, we related the sampled TN concentrations with the river discharge rate on the day the samples were obtained, as shown in Fig. 4, to estimate the daily TN concentration from the daily discharge; the TN concentration decreases with increasing discharge. The annual N load exported through river discharge (X_D) at the outlet of the watershed was then calculated by multiplying the daily river discharge (Q_i) on day i by the TN concentration of day i, which was estimated from the daily river discharge using the regression function ($TN_{OUTi}(Q_i)$) in Fig. 4:

$$X_D = \sum Q_i \cdot TN_{OUTi}(Q_i) \tag{5}$$

The resulting average TN concentration ($X_D/\sum Q_i$) was 0.44 mg l^{-1}. Although the number of water samples collected from the outlet for calculation of the TN concentration was limited, we believe that this average TN concentration is sufficiently reliable. Instead of conducting frequent field surveys, we chose to conduct intensive and extensive sampling of 199 river samples from 87 locations throughout the watershed during six surveys conducted in both dry and rainy seasons, including immediately after the start of the latter. The average TN concentration was 0.46 mg l^{-1} with a minimum value of 0.04 mg l^{-1} and maximum of 1.12 mg l^{-1} (Fig. 5). More than 70% of the samples had a TN concentration between 0.2 and 0.6 mg l^{-1}.

N export through rice sales

N contained in exported rice (X_R) was calculated as

$$X_R = (Y_R - C_R)0.07/5.95 \tag{6}$$

where Y_R is the amount of rice produced, C_R is rice consumption by the residents of the watershed,

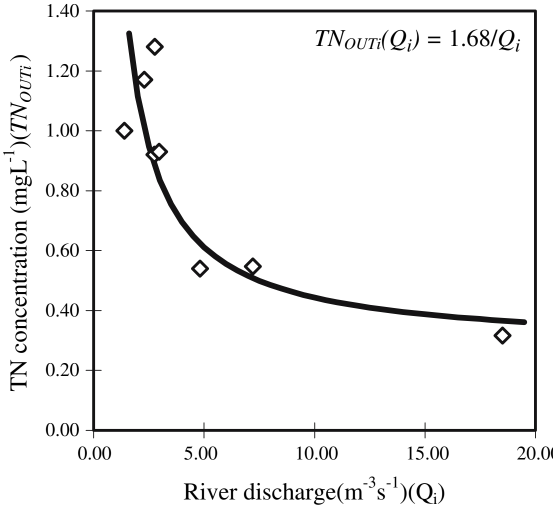

Fig. 4 Relationship between river discharge and the TN concentration at KTI pumping station. Points represent the observed data and the curve the regression function

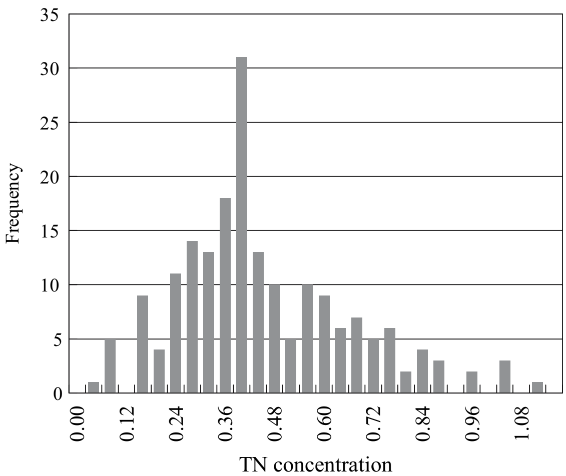

Fig. 5 TN concentrations of the river water samples (199 samples collected at 84 locations)

0.07 (g/g) is the approximate protein content of milled rice, and 5.95 is the protein/N conversion factor (FAO/WHO 1973). Y_R was found as a product of *GPA* and the rice yield of the watershed (y), and C_R as a product of the watershed population and the average consumption of rice per capita in Indonesia (c).

N loss due to incineration of crop residues and wood fuel

Most farmers in the watershed burn rice straw to ease tillage and control rice stem rot following the harvest of rice during the dry season. However, they plough the residues back into the paddled paddy fields or leave them on levies during the rainy season. According to interviews with 103 randomly selected farmers, 85% of the rice straw is burned during the dry season and 21% during the rainy season. All the rice hulls are also burned regardless of the season. Hull weight represents approximately 16% of milled rice, and its N content ranges from 0.43% to 0.55% with an average of 0.49% at 14% moisture (Juliano et al. 1985).

The N harvest index (*NHI*), which is defined as the ratio of the N content in grains to the total N content of the plant above ground level, ranges from 53% to 60% with a mean value of 56% for tropical rice (Ying et al. 1998). In other words, approximately 44% of the N in an entire plant is contained in rice straw. The proportion of gross rice production in the dry season and rainy season was calculated to be approximately 50% for each between 1996 and 2004. The dry season is one month longer than the rainy season but cultivation intensity is higher in the latter. Therefore, N losses through incineration of rice straw (X_{RS}) and rice hulls (X_{RH}) were calculated as

$$X_{RS} = (Y_R \cdot 0.07/5.95) \cdot \frac{(1 - NHI)}{NHI} \cdot \frac{(IR_D + IR_R)}{2} \tag{7}$$

$$X_{RH} = Y_{UR} \cdot 0.16 \cdot 0.0049 \tag{8}$$

where Y_R and Y_{UR} are the total production of milled rice and that of unhulled rice, respectively, *NHI* is the N harvest index, IR_D and IR_R are the ratios of rice straw incineration in the dry and wet seasons, respectively, and 0.0049 (g/g) is the N content of rice hull.

Another component of N loss due to incineration is domestic incineration of wood fuel. In rural areas in Indonesia, people still use wood fuel for cooking, and in the watershed, the wood fuel they burn is collected from nearby local villages. According to the results of an extensive survey of rural West Java by Soesastro (1984), wood fuel consumption per capita (c_{WF}) was estimated to be 679 kg year^{-1} (1.86 kg day^{-1}) on average. Since the typical N content of wood is 0.2–0.3% with an average of 0.25% (Ludwig et al. 2003), annual gaseous N loss through wood fuel consumption in the study watershed was calculated as

$$X_{WF} = 0.0025 c_{WF} \cdot POP \tag{9}$$

Thus, the total gaseous N loss through human activity relating to incineration was determined as the sum of three components: X_{RS}, X_{RH} and X_{WF}.

N export through net gaseous loss attributed to biochemical processes

The net gaseous N loss due to natural biochemical processes, X_{GB}, was obtained as the residual of all other components of N import and N export that were quantitatively identified. This was calculated as the difference between total N import (M) and the sum of N export through river discharge (X_D), surplus rice sales (X_R), and gaseous N loss attributed to residue incineration (X_{GI}).

Result

Table 1 shows the values of all variables used for calculating the watershed N budget, and Fig. 6 demonstrates the N flow per ha per year, which was obtained by dividing the N import and export values in Table 1 by the area of the watershed (22,100 ha). The values depending on river discharge data or rainfall data are presented as 9-year averages for the 1995–2004 water years.

On the import side, N fertilizer application (M_F) constitutes a major contributing factor at 33.1 kgN ha^{-1} year^{-1} or approximately 71% of the total N import into the watershed. The remainder can be explained by atmospheric deposition (M_{AD}), contributing to 13.4 kgN ha^{-1} year^{-1} or approximately 29%, of which wet deposition (M_{WD}) and dry deposition (M_{DD}) are 10.7 kgN ha^{-1} year^{-1} (23%) and 2.7 kgN ha^{-1} year^{-1} (6%) respectively. In total, 46.5 kgN ha^{-1} year^{-1} is imported into the watershed. On the export side, N export through surplus rice sales (X_R) and river discharge (X_D) were found to be relatively small at 4.0 kgN ha^{-1} year^{-1} and 5.9 kgN ha^{-1} year^{-1} or 9% and 13% of the total

Table 1 The variables used in the analysis

Variable	Description	Value
M	Total N import	1,028 ton N year^{-1}
M_F	N import through fertilizer application	731 ton N year^{-1}
M_{AD}	N import through atmospheric deposition	297 ton N year^{-1}
M_{WD}	N import through wet deposition	237ton N year^{-1}
M_{DD}	N import through dry depostion	59 ton N year^{-1}
X	Total N export	1,028 ton N year^{-1}
X_D	N export through river discharge	129 ton N year^{-1}
X_R	N export through rice sales	89 ton N year^{-1}
X_{GI}	N export through gaseous loss (incineration)	376 ton N year^{-1}
X_{RS}	N export through gaseous loss (incineration of rice straw)	178 ton N y^{-1}
X_{RH}	N export through gaseous loss (incineration of rice hulls)	29 ton N year^{-1}
X_{WF}	N export through gaseous loss (incineration of wood fuel)	170 ton N year^{-1}
X_{GB}	Net N export through gaseous loss (biochemical)	434 ton N year^{-1}
A	Paddy field area in the watershed	6,070 ha
GPA	Annual gross cultivated paddy acreage	10,980 ha
P	Annual average bulk precipitation	2,660 mm
P_{90i}	90-day cumulative precipitation prior to day i	0–1,870 mm
F	Average N fertilizer application per ha per crop	66.2 kgN ha^{-1} crop^{-1}
TN_P	Average TN concentration of precipitation	0.75 mg l^{-1}
TN_{OUT}	TN concentration of river water at the watershed outlet	0.32–1.28 mg l^{-1}
Q_i	River discharge at the watershed outlet	0.7–79.1 m^3 s^{-1}
C_R	Consumption of rice by residents (milled rice)	16,570 ton year^{-1}
C	Consumption of rice per capita (milled rice)	149 kg person^{-1} year^{-1}
POP	Population of the watershed	111,200 persons
Y_R	Annual average production of rice (milled rice)	23,990 ton year^{-1}
Y_{UR}	Annual average production of rice (unhulled rice)	35,800 ton year^{-1}
Y	Rice yield	3.52 ton ha^{-1} crop^{-1}
NHI	Nitrogen harvest index	0.56
IR_D	Ratio of incineration of rice straw in the dry season	0.85
IR_R	Ratio of incineration of rice straw in the rainy season	0.21
c_{WF}	Wood fuel consumption per capita	679 kg person^{-1} year^{-1}

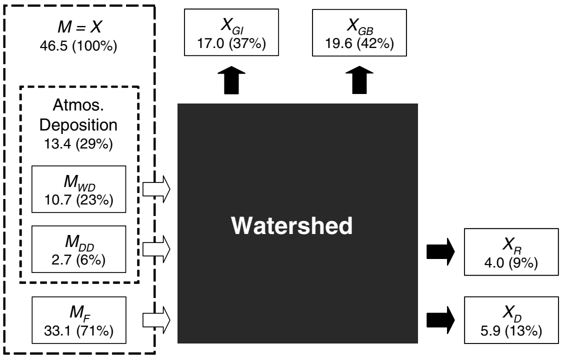

Fig. 6 Results of N flow into/out of the study area per unit area of the watershed. Unit is kgN ha^{-1} year^{-1} representing the average of the entire watershed (221 km^2), and values in % represent the percentage of total input. (M, total import; X, total export; M_{WD}, import with wet deposition; M_{DD}, import with dry deposition; M_F, import with fertilizer; X_R, export through rice sales; X_D, export through river discharge; X_{GI}, export through incineration; X_{GB}, export through net biochemical gaseous loss; obtained as the residual

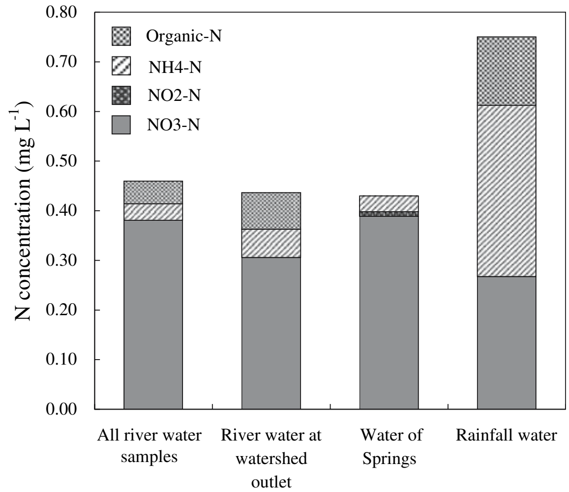

Fig. 7 Average TN concentrations and the different components. The average value of river water at the watershed outlet was calculated on the basis of the function shown in Fig. 4. Other values were obtained by averaging all water samples in each category

N import, respectively. On the other hand, the gaseous N loss attributed to incineration (X_{GI}) was responsible for a greater fraction of the total N export at 17.0 kgN ha^{-1} year^{-1} or 37%, of which crop residue incineration (X_{RS} + X_{RH}) accounted for 9.3 kgN ha^{-1} year^{-1} or 20%, and wood fuel incineration (X_{WF}) for 7.7 kgN ha^{-1} year^{-1} or 17%. The residual is represented by net gaseous N loss attributed to biochemical processes (X_{GB}), which accounted for an outstandingly high fraction of the total N export, 19.6 kgN ha^{-1} year^{-1}, constituting 42% of all the N exports out of the watershed.

Discussion

An important observation derived from the sampled water in the present study was the fact that the average TN concentration at the outlet of the watershed (TN_{OUT}) (0.44 mgN l^{-1}) was lower than that of rainfall (TN_P) (0.75 mgN l^{-1}) (Fig. 7). Since the total discharge of water outflow from the watershed is 45% less than the amount of total rainfall due to evapotranspiration, the lower TN concentration of the river water indicates that the total amount of N that outflows from the watershed with river water is lower than the N input with rainfall. Consequently, N export through river discharge was found to be only 13% of the total N import. This proportion of N export through river discharge is smaller than the results of a study in the northeastern USA (van Breemen et al. 2002), where N export through river discharge represented an average of 20% of the total N input, despite the much larger precipitation and river discharge of our study watershed.

In general, the difference between N import and N export represents the N stock change within a system during the period imports and exports are accounted for. If environmental settings of the watershed that control the N budget have not changed greatly for a long period, the N stock of the whole watershed, i.e. that in biomass, soil and groundwater, can be assumed to be in a steady state; that is, assuming that N stock change is negligible compared with the amount of N import and export in the long-term, then N import and export will become balanced, or $M = X$.

With respect to the biomass stock in the watershed, forests and swamp forest, which occupy 58% and 5% of the land cover, respectively, are responsible for the largest percentage. The land use and cover in the study watershed have not been appreciably altered due to deforestation or afforestation for at least 30 years according to satellite analysis (Tsuyuki et al. 2003), and the areal extent of forest and swamp forest has remained constant. Consequently, these forests can be regarded as basically pristine "old-growth" forests. Therefore, decomposition of biomass balances production and the amount of biomass stored in the forest is constant if there is no recent significant change in N input.

However, it is likely that biomass production has recently started to exceed its decomposition and the amount of biomass stored in the forests is currently increasing. The reason is that the TN concentration in rainfall water and from dry deposition of ammonia within the forests must have increased owing to application of chemical fertilizer in surrounding paddy fields and the recent increase in NO_x in the atmosphere due to air pollution. The rate of atmospheric N deposition in the watershed is estimated to be 23.0 kg ha^{-1} year^{-1} (including NH_3), which might have been less in the past. The significant increase in N input in the watershed must have started when chemical fertilizer was introduced (40–50 years ago), but this input may have become mostly constant for at least the last 20 years.

In tropical forest, where decomposition of organic mater in soil is fast due to the high temperature, organic mater in soil is mostly stored on the ground surface. N supply with litterfall, the primary input to the forest floor, is estimated to be 60–220 kg ha^{-1} year^{-1} in lowland tropical forest (Vitousek et al. 1986). On the other hand, basically the same rates of decomposition of litter on the forest soil surface and organic matter in soil should occur, resulting in the mineralization of organic N. The mineralized N is dissolved in soil water and taken up by roots to again become biomass of living trees. This N cycling in soil in tropical forests is much more rapid than in forests in a temperate climate (Vitousek et al. 1986; Swift et al. 1989). Therefore, even though a considerable amount of N is stored in the soil biomass, it is likely that the biomass and N content in soil had become a steady state that corresponds to the increased N input within a short period in the past, considering the high rate of biochemical N cycling in soil in tropical forest.

However, biomass of living trees may need a much longer time to reach steady state after the increase in N input, and therefore it is important to discuss. Typically, biomass stock of living trees in a pristine tropical rain forest is estimated to be roughly 400 tons ha^{-1} (Cannell 1982). Given the 0.25% of N in such biomass, N stock is calculated to be 1,000 kgN ha^{-1}. Therefore, the forest biomass per unit area of the watershed on a whole is approximately 630 kgN ha^{-1} year^{-1} (63% of the land cover). Supposing that the forest biomass is currently increasing by a rate of 10% during 30 years, the annual increment of N in the forests is estimated to be about 2 kgN ha^{-1} (630 kgN ha^{-1} year^{-1} times 10% divided by 30 years). This quantity is still sufficiently small in terms of the residual in the analysis conducted above, 19.6 kgN ha^{-1} year^{-1}. Moreover, the presumption of a 10% increase in biomass stock during 30 years may be higher than reality for the trees of the "old-growth" forest. Thus, N stock change in biomass in these forests is likely to be negligible in terms of explaining the residual amount. However, the actual rate of N stock change in forest is uncertain.

Let us examine the changes in the amounts of inorganic N stored in soil, water and soil solid in an absorbed form in the forest compartment of the watershed. Annual average downward water flux in the bottom of the root zone (annual precipitation minus evapotranspiration) is about 1,200 mm year^{-1} while the infiltration flux at the soil surface is about 2,600 mm year^{-1} (annual precipitation). If we assume the volumetric water content of the root zone is 0.5 and that N exists only within soil water, then the 1,200 mm year^{-1} water flux can displace N in a 2.4 m thick soil layer in 1 year. Here we assume a large capacity of absorption, for example, ten times of N can change in whole soil including the absorbed form of N (mainly NH_4^+) with the change in N concentration in the soil water (a retardation factor of 10 is assumed for solute movement in soil). Even in this example, N stored in the 2.4 m layer is displaced and reaches a steady state within 10 years. This calculation indicates that inorganic N stored in forest soil is mostly in steady state owing to the large downward water flux.

On the soil surface in forest, there is a layer of accumulated organic matter where the rate of decomposition of the organic matter must be high because of the high temperature in a tropical climate, and therefore the rate of oxygen consumption is high. This condition should cause the subsoil (near the surface) to deoxidize and a suitable condition for denitrification exist. NH_4^+ and NH_3 deposited from the atmosphere or N mineralized from an organic form may turn to NO_3^- (nitrification) in the surface layer, with NO_3^- that move to the deoxidized subsoil may become N_2 gas (denitrification) by dissolved organic carbon being consumed. Thus, TN concentration in soil water will be reduced near the surface, where there is abundant organic carbon because of denitrification, and most of the N remaining in soil water will be uptaken through roots during infiltrating through the root zone. We assumed that the amount of atmospheric N deposition approximately balances the net biochemical N loss to the atmosphere within a forest; the amount of N leached from the root zone with soil water flow is negligible, which can be confirmed from the low TN concentration of streams measured in the forest where no residential area exists upstream.

The swamp forest, however, needs to be discussed separately because decomposition of organic matter on ground is much slower than for the other forests, and a peat layer 3.6 m thick on average is formed. The rate of organic carbon accumulation on the surface of the swamp forest in the study site is estimated to be 12 mgC cm^{-2} year^{-1} (1200 kgC ha^{-1} year^{-1}), and N accumulation is estimated to be 40 kg ha^{-1} year^{-1} according to a C/N ratio of 30 for the peat core (Tareq et al. 2004, 2005). However, the rate of increase in accumulated C and N in the swamp forest is not certain (it is unknown if the amount of peat is increasing or in steady state) because the rate of decomposition over the 3.6 m thick peat layer is not known. Even if a possible maximum annual N accumulation rate of 40 kg ha^{-1} is assumed, it contributes only 2 kg ha^{-1} year^{-1} to the N stock increase of the watershed average since the swamp forest occupies 5% of the watershed.

Another possible sink for N is groundwater. The measured average TN concentration of all river water samples collected in the dry season (134 samples from 87 locations) was 0.46 mgN l^{-1}. This TN concentration should reflect the TN concentration in shallow groundwater since river discharges in the dry season are mostly recharged by shallow groundwater. In fact, the average TN concentration of a large spring located in the center of the watershed (Fig. 1) was 0.43 mgN l^{-1}, which is slightly lower than that of the river average. Since the flux of water seepage from shallow groundwater into deep groundwater is considered to be much smaller than the amount of river discharge, the amount of N carried with the water seepage into deep groundwater, resulting in N stock change, is thought to be negligibly small, if any, compared with the amount of N export with river discharge. Moreover, past water quality data (e.g. Kato et al. 2002) show that there is no tendency of increasing TN concentration in the river water at least over the past 5 years.

Given that no major sinks exist in the watershed, the residual between N import and export (X_{GB}) is considered to represent the net gaseous loss to the atmosphere through biochemical processes. Two independent biological processes of denitrification and biological N fixation are presented in the net value of X_{GB}. It is, however, difficult to estimate these two processes separately, and we believe separate estimations are not necessary for the N budget study; the important factor for the environment is the net value. Both denitrification and biological N fixation occur in paddy fields and forest, yet both significantly depend on N concentration; high TN concentration accelerates denitrification and reduces biological N fixation, resulting in a large net value of N emission to the atmosphere; low TN concentration, on the other hand, could result in an excess of biological N fixation (negative net value of X_{GB}). The large (positive) net value of X_{GB} indicates that denitrification is the dominant process of N loss from the watershed.

NH_3 volatilization and its deposition are other biochemical N exchanges between the watershed and atmosphere. However, as explained in the method section, we assumed that NH_3 volatilization balances its deposition on the watershed scale, and NH_3 deposition was not included in the term of atmospheric N deposition. Consequently, we consider that NH_3 volatilization is also not included in the residual value of the N budget. NH_3 volatilization commonly takes place when N fertilizer is applied in an organic form such as urea, and broadcasted on the surface of warm soil with a high water content (Hargrove et al. 1987; Clay et al. 1990; Schlesinger et al. 1992). NH_3 volatilization also occurs from human and animal waste. The rate of N loss through denitrification and NH_3 volatilization in paddy fields varies greatly depending on the method of fertilizer application. For example, according to field measurement experiments conducted in the Philippines by the IRRI, urea fertilizer broadcasted to transplanted flooded lowland paddy fields marks the largest total N loss (62%), whereby the dominant mechanism of loss was NH_3

volatilization (47%) and denitrification was relatively small (15%) (Buresh et al. 1990). NH_3 volatilization mostly occurs within one week of broadcasting urea (Fillery et al. 1986). Furthermore, the total N loss was smaller with the basal incorporation method (50%); the dominant mechanism being denitrification (33%) instead of NH_3 volatilization (17%). This suggests that substantial NH_3 volatilization occurs in paddy fields in the study watershed where urea fertilizer is customarily broadcast after transplantation of rice.

However, gaseous N generated by the two processes, NH_3 volatilization and denitrification, behaves differently. Volatilized NH_3 redeposits into places around the source of volatilization mostly within several hours or days (Howorth et al. 1996; Jordan et al. 1996; Boyer et al. 2002; Mcisaac 2004); it simply diffuses around the source. Mountain areas covered by forest in a watershed with surrounding paddy field areas may be a major sink of volatilized NH_3. The relatively high NH_4–N concentration (46% of the TN in the rain water samples) is a possible reason for redeposition of NH_3. Moreover, considering the inflow of NH_3 gas from neighboring areas outside the watershed as well as outflow from the watershed, net flow of NH_3 gas from the watershed to the outside should represent only a small portion of the NH_3 volatilization from paddy fields and residential areas. This is the reason why we assumed NH_3 volatilization balances with its deposition and did not include it in import nor export terms.

In contrast to NH_3 volatilization and deposition, denitrification emits N ultimately into the atmosphere (N fixation is not a serial process of denitrification but an independent process). Therefore, we regard the resulting value of X_{GB} as predominantly representing the amount of denitrification in excess to biological N fixation. This biological net N emission into the atmosphere is estimated to be more than three times the N export through river discharge.

High temperatures encourage denitrification, as well as most biochemical process involving soil microbes. The average atmospheric temperature in Indonesia is 26°C, which is roughly 11°C higher than the Japan average for example. Generally, a 10°C increase in temperature increases the rate of biochemical reactions within microorganisms 2- or 3-fold (Campbell et al. 1998), and the same is true for denitrification. Anaerobic conditions such as flooding of paddy fields also induce denitrification. High N loss may therefore be occurring within the watershed due to the high temperatures and high precipitation of the climate. The low TN concentration of the river water (about one quarter of rivers in paddy areas of Japan), in spite of considerable fertilizer input, is thought to be the result of a high rate of denitrification in the paddy fields and over the whole watershed. It is roughly estimated that 35% of the total net biological N loss within the watershed takes place in the paddy area and 65% in forest. The high rate of denitrification and low rate of N outflow with river water flow observed in the study area are thought to be common features of tropical paddy areas.

In the past, when no chemical fertilizer was applied and no air pollution due to industry or cars existed, biological N fixation must have been the major N input into the watershed and it exceeded denitrification (especially in paddy fields); the excess of N fixation might balance the sum of the other N export terms of river water flow, rice exports, and atmospheric N emission due to incineration of rice residue and wood fuel. However, nowadays, chemical fertilizer application and air pollution have made the watershed N-rich and the increased TN concentration over the watershed has decreased biological N fixation and increased both denitrification and N flow with river water. Apparently, denitrification has overwhelmed biological N fixation and become the dominant process of N loss from this agricultural watershed.

Conclusion

The major achievement of this study is that it clarified, for the first time, all components of the N cycle with good confidence in a tropical watershed in which the predominant land use is paddy fields. The finding of the dominance of net gaseous N emission in the N cycle is particularly worth noting. Biochemical net gaseous N emissions can be assessed as a residual of the N budget and, therefore, are reliable only when all other components of the N budget are accurately accounted for.

This study successfully quantified the net gaseous N loss. Consequently, it was clarified that the net gaseous N loss due to natural biological processes (denitrification) dominates more than 40% of the total

N export (and total N import), while N export with riverine transport represents only one-tenth of the total N export. Therefore, denitrification occurring in the watershed is thought to be the major factor determining the N load of river water. This outcome is mainly due to the environment of the watershed, which is characterized by a predominance of paddy fields, high atmospheric temperatures and abundant precipitation representative of the tropical climate.

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