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The nitrate export in subtropical mountainous catchment: implication for land use change impact

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Abstract

Agricultural activity is the dominant factor affecting water quality and nitrate export, which causes eutrophication and episodic acidification in downstream water bodies (e.g., reservoirs, lakes, and coastal zones). However, in subtropical mountainous areas such environmental impact due to the land use change was rarely documented. In this study, we investigated 16 sub-catchments during 2007 and 2008 in the Chi-Chia-Wan catchment where is the sole habitat for the endemic species, Formosan landlocked salmon (*Oncorhynchus masou formosanus*).

The results revealed that the NO_3-N concentration in pristine catchments varied from 0.144 to 0.151 mg/L without significant seasonal variation. This concentration was comparable with other forestry catchments around the world. However, the annual nitrate export was around 375.3–677.1 kg/km^2/yr, much higher than other catchments due to the greater amount of rainfall. This is an important baseline for comparisons with other climate areas. As for the impact of agricultural activities, the catchments with some human disturbance, ∼5.2% of the catchment area, might yield 5947.2 kg N/km^2/yr – over 10-times higher than that of pristine catchment. Such high export caused by such a low level of disturbance might indicate that subtropical mountainous area is highly sensitive to agricultural activities. As for the land-use effect on nitrate yield, the forestry land might yield 488.5 ± 325.1 kg/km^2/yr and the vegetable farm could yield 298 465.4 ± 3347.2 kg/km^2/yr – 1000-times greater than the forestry. The estimated nitrate yields for land use classes were a crucial basis and useful for the land manager to assess the possible impacts (e.g., non-point source pollution evaluation and the recovery of land expropriation).

1 Introduction

High nitrate export from diffuse sources is a major cause of eutrophication and episodic acidification for the terrestrial, stream and coastal ecosystems (Vitousek et al., 1997;

Crimo and McDonnell, 1997; Creed and Band, 1998; Galloway et al., 2003; Meader and Goldstein, 2003; Wellington and Driscoll, 2004), including groundwater, rivers, lakes and coastal zones (Burns et al., 1998; Boesch et al., 2001; Rabalais, 2002; De Vries et al., 2003). In most cases, this significant portion of nitrogen export from catchments is due to non-point source fertilizer runoff. As the name indicates, such kind of runoff is relatively difficult to identify owing to the spatial and temporal heterogeneity of hydrological processes and agricultural activities (D'Arcy and Carignan, 1997; Herlihy et al., 1998; Hooke, 1999; Johnson et al., 2000). Meanwhile, this non-point source pollution is regional-dependent due to different environmental settings and agricultural behaviors. The stream nitrogen export has been proved to be highly correlated to the proportion of agricultural land in a catchment (Howarth et al., 2002). Many other previous studies, such as Poor and McDonnell (2007) in Oregon, USA; Zhang et al. (2009) in Han River, a major tributary of the Yangtze River in China; Ohrui and Mitchell (1998), Ogawa et al. (2006) and Tanaka and Suzuki (2009) in Japanese forested watersheds; Walton and Hunter (2009) in the Great Barrier Reef in Australia; and Rode et al. (2009) in Central Germany, has also addressed the issue. Dumont et al. (2005) estimated the global dissolved inorganic nitrogen (DIN) export and pointed out the low-latitude area and intensive agricultural area might be the hotspots for DIN export. However, the observations of nitrogen fluxes in low-latitude ecosystems are not well documented and understood (Kao et al., 2004). The comprehensive observations of stream nitrate export in low-latitude can aid in estimating nitrogen cycling in regional and global scale, evaluating land management, and other relevant applications.

Southern Asian countries in the tropical and subtropical latitude are characterized by rugged mountainous watersheds, heavy precipitation, plentiful biodiversity, and high population. Among them, Taiwan is a subtropical mountainous island with the elevation ranging from 0 to 4000 m a.s.l. The annual rainfall is around 2400 mm with 3–5 typhoon invasions from June to October. The stream discharge during the wet season occupies around 70–85% of the annual discharge. It is also recognized that this island ecosystem is relatively vulnerable in such environmental settings. Besides, slopeland

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development, which is usually regarded as a convenient way to support the society, degraded land severely. From the past two decades, the government made great efforts, such as delineation of protected area (e.g., national park) and agricultural land expropriation, to protect the environment. Such significant land use change provided a good opportunity to investigate the influence of land expropriation on stream nitrate export in the subtropical mountainous watersheds.

This study attempted to investigate the characteristics of nitrate concentrations and fluxes in subtropical mountains, and then compared to other countries. Meanwhile, we also estimated the nitrate yield of each land use type. To accurately estimate the nitrate export, the high-frequency (~3 h) sampling scheme was conducted during the typhoon events. This study can not only advance the understanding of the nitrate export in subtropical zone, which is important for the nitrogen budget study in regional and global scale, but also provide the useful baseline for land management.

2 Material and methods

2.1 Study site

The Chi-Chia-Wan watershed located in Central Taiwan drains area of 105 km^2, and it comprises rugged mountainous terrain with elevations ranging from 1131 to 3882 m a.s.l. Two major tributaries, Yi-Ka-Wan Creek and You-Sheng Creek with drainage areas of 74 and 31 km^2, converge into Chi-Chia-Wan Creek. Two discharge gauges measuring the discharge of Chi-Chia-Wan and You-Sheng Creek, and three precipitation gauges maintained by Taiwan Power Company are deployed in this area (Fig. 1a). The annual precipitation is around 2462 mm, which is around 3 times of the global mean. And around 75% of annual rainfall rains in the wet season (May to October), which tropical cyclone is the main contributor. The average daily discharge in Chi-Chia-Wan and You-Sheng Creek are 7.94 and 2.41 m^3/s, and are 11.80 and 4.07 m^3/s in the wet season, respectively. The mean air temperature is 15.8 °C with the 4 °C and 23 °C in January and July, respectively (Huang et al., 2006).

As for land use pattern, forest composed of Pinaceae (*Pinus taiwanensis*), Juglandaceae (*Juglans cathayensis*), Aceraceae (*Acer serrulatum*) and Betulaceae (*Alnus formosana*) covers 87% of the whole watershed (above C1 in Fig. 1a). The other land use types, including grass, bare land, orchard, active vegetable farms and inactive vegetable farms, occupy the rest 13% watershed area. Unlike the land use patterns in America and Europe, those cultivated land all occupy the riparian zones as Fig. 1b shows. This special characteristic can be usually found in the mountainous area, where alluvial terrace is more suitable for cultivation. To restore the watershed to conserve the endangered Formosan landlocked salmon (*Oncorhynchus masou formosanus*), Taiwan government began to expropriate the cultivated farms starting from vegetable farms along You-Sheng Creek. Here in this paper, the expropriated vegetable farms were categorized as inactive vegetation. On the other hand, active vegetation and orchard stand for currently-cultivated farms. To explore the influence of land use patterns on nitrate yield, the entire watershed was divided into 16 subwatersheds. The outlet of each subwatershed, which was also the water samples from, is illustrated in Fig. 1. From the pristine (C7, T1, and Y6) to the intensively-cultivated subwatersheds (C5), the delineation of subwatershed represented different levels of human disturbance. Since the Chi-Chia-Wan watershed is small, atmospheric deposition can be rationally assumed homogeneous watershed-wide. Hence, the different level of human disturbance, particularly indicated nitrogen input from fertilization here, could be taken as the only one parameter controlling nitrate concentration in the creek. The basic landscape characteristics of those sampling sites were shown in Table 1, and the detailed descriptions about the land use patterns in each subwatershed were shown in Table 2.

The study watershed has attracted much attention since Yi-Ka-Wan Creek is now the only habitat for Formosan landlocked salmon in the world. Although the direct link between water quality and salmon survival remained unsolved, Taiwan government tried to recover aquatic water quality via watershed restoration. It provided us a very good chance to quantitatively understand the effects of land use changes on nitrate yield, particularly the improvement resulting from the expropriation. The results of this research will be an important reference for watershed management.

2.2 Nitrate monitoring and measurement

To identify the effects of land use changes on nitrate yield, 16 sampling sites were set up in the Chi-Chia-Wan catchment, which 9 sites in Yi-Ka-Wan Creek and 7 sites in You-Sheng Creek, intensively covering the entire watershed spatially. Water quality data, including regular samples, monthly samples, and event samples, have been analyzed in those sampling sites from 2007. Regular samples, which were regularly taken twice per week, were executed at C1, Y1, and K1 three sites. Monthly samples were implemented at the other sites, which at least one sample for each month. Event samples from two typhoon events, which were taken on 16–19 August 2007 (Sepat) and 4–7 October 2007 (Krosa) at the sampling interval of 3 h, were supplemented to understand nitrate behavior at C2, C7, Y1, and K1 four sites. The intensive sampling scheme could represent the temporal variations in this area.

To analyze NO_3, water samples taken from stream were immediately filtered through GF/F filters (0.7 mm) and the filtrate was quick-frozen in liquid nitrogen for NO_3 analysis. Nitrate was determined by ion chromatography (IC) using either a Dionex ICS-90 or 1500 instrument using the method of Welch et al. (1996) with detection limit of 0.01 mg/L. Nitrate was the major form of nitrogen export since the concentration of nitrite and ammonium were both below the detection limit.

2.3 Ungauged catchment discharge

Discharge is essential to estimate elemental flux, which is the product of elemental concentration and discharge. However, only two discharge gauges were installed among our 16 sampling sites; therefore modeling approach was needed to estimate discharge at those ungauged sites. In this study, a modified 3-layer TOPMODEL proposed by Huang et al. (2009) was applied. The historical reported discharge data in 2001–2005 at C1 (Chi-Chia-Wan Creek) and Y1 (You-Sheng Creek) were used to calibrate the model, and it was validated using the discharge in 2007–2008. The efficiency coefficient (EC) proposed by Nash and Sutcliffe (1970) was used to quantify the overall deviation between the simulated and observed hydrographs. While the observation and simulation perfectly matched, EC equaled to 1. The $\mathrm{EC}_{\log}$, the logarithmic Nash-Sutcliffe efficient, was also widely used to quantify the similarity in low flow condition (e.g., Güntner et al., 1999; De Smedt et al., 2000). The EC and $\mathrm{EC}_{\log}$ were presented as follows:

$$\mathrm{EC} = 1 - \frac{\sum_{i=1}^{N} (Q_{\mathrm{s},i} - Q_{\mathrm{o},i})^2}{\sum_{i=1}^{N} (Q_{\mathrm{o},i} - \overline{Q}_{\mathrm{o}})^2} \tag{1}$$

$$\mathrm{EC}_{\log} = 1 - \frac{\sum_{i=1}^{N} (\log(Q_{\mathrm{s},i}) - \log(Q_{\mathrm{o},i}))^2}{\sum_{i=1}^{N} (\log(Q_{\mathrm{o},i}) - \overline{\log(Q_{\mathrm{o}})})^2} \tag{2}$$

In Eqs. (1) and (2), $Q_{\mathrm{o},i}$ is the observed discharge at day i, $Q_{\mathrm{s},i}$ is the simulated discharge, and N is the total number of time steps during the evaluated period. Based on the two measures, the promising parameter set can be retrieved. The two calibrated parameter sets for C1 and Y1 were then applied to the sub-catchments in order to estimate the discharge for those ungauged catchments.

2.4 Nitrate-N flux/yield estimation

Based on the nitrate concentration measurements and ungauged catchment discharge estimations, $\mathrm{NO_3}$-N fluxes for each site were estimated. However, there was not a simple and easy way to estimate flux accurately due to several factors, such as sampling frequency, hydrological behaviors, and constitute characteristics (Lee et al., 2007). Therefore, three commonly used methods, namely, global mean, flow weighted, and rating curve method, were used in this study to estimate annual flux. The global mean method, as its name indicates, multiplies the average concentration of all samples by the total discharge. This method is the simplest one and does not yet take the hydrological responses into account. The following equation demonstrated this method (Birgand et al., 2010).

$$\mathrm{FLUX} = m\frac{\sum_{i=1}^{n} C_i}{n} \times Q_\mathrm{t} \tag{3}$$

Where FLUX is annual NO_3-N load (kg/yr); C_i is the NO_3-N concentration of water sample (mg/L); Q_t is the annual total discharge (m^3/yr); and n is the number of water samples; m is the conversion factor to convert the calculated values into specific unit (kg/yr). Considering the hydrological responses, the flow weighted method, which weights the concentration by discharge, was widely used as Eq. (4) shows. Where Q_i (m^3/day) is the discharge on the sampling day.

$$\mathrm{FLUX} = m\frac{\sum_{i=1}^{n} C_i Q_i}{\sum_{i=1}^{n} Q_i} \times Q_\mathrm{t} \tag{4}$$

The rating curve method, which regresses the observed concentration and discharge by power function, $C{=}aQ^b$, is recognized as an inspector to reveal the hydrological dynamics of transport. The parameter b denotes the hydrological dynamics. The larger b value (>1.0) indicates the strong enhancement. By contrast, the small b value denotes the dilution effect because the concentration decreases with the increase of Q.

Where Q_j (m^3/s) in Eq. (5) is the daily water discharge rate; C_j (mg/L) is an estimated constituent concentration on the j-th day; T (days) is number of days in a year.

$$\mathrm{FLUX} = m\sum_{j=1}^{T} Q_j C_J = m\sum_{j=1}^{T} aQ_j^{b+1} \tag{5}$$

Because of the complexity of hydrological dynamics and the limitation of observations, the three mentioned methods are not easy to justify. To avoid the arbitrary estimates, all the methods were applied in this study to estimate the NO_3-N fluxes.

Assessing the pollution of water bodies from non-point sources is a comprehensive and time-consuming task. Many factors, such as topography, deforestation, urbanization and hydrodynamics, might alter the water chemistry (Rose and Peters, 2009; Swank et al., 2001). The land use pattern and its proportion of the watershed are the dominant factors. Incorporating the land use data, the NO_3-N yield of each land use class can be estimated quantitatively. In this study, it is assumed that the annual export of NO_3-N for each kind of land use can be represented as a constant k ($kg/km^2/yr$), NO_3-N yield factor. The annual flux for each sub-catchment is the summation of the product of yield factors and the corresponding land use areas. Unlike the export coefficient method in PLOAD model (EPA, USA, 2001), the yield factors in this study were derived through inverse calculation based on observed flux. In PLOAD model, users look up the yield factors from the lookup table to calculate loads. The flux for each sub-catchment can be estimated as Eq. (6) shows.

$$\mathrm{FLUX} = \sum_{i=1}^{n} k_i \times A_i \tag{6}$$

Where FLUX is the annual load (kg/yr), k_i ($kg/km^2/yr$) and A_i (km^2) is the yield factor and the area for land use i. In this study, there are 16 sampling sties representing NO_3-N export from 16 sub-catchments, which all of them can be described as Eq. (6). Besides, each sub-catchment has at most 8 different land use classes. Hence, NO_3-N yield factor can be easily obtained from the 16 algebraic equations with 8 unknowns. However, to consider the uncertainties from the assumptions, the trial and error procedure was applied to estimate the yield factor using RMSE (root mean square error) as a performance measure. 300 000 yield factor combinations (each combination includes 8 yield factors) were generated to estimate NO_3-N loads at the sub-catchment outlets for 2007 and 2008, separately. Only the combinations could minimize the 2007 and 2008 RMSE simultaneously were selected. Therefore, the selected yield factors, which can reduce the annual variation and the landscape heterogeneity effect (e.g., land use locations), are reliable. This yield factor, similar to land cover indicator, is the most important component for the assessment of the pollution of water bodies from non-point sources (Munafo et al., 2005; Lane et al., 2006).

3 Results and discussion

3.1 Discharge simulation

After calibration, the simulated discharge in C1 and Y1 promisingly agreed with the measured discharge. The performance measures of EC and EC_{log} were 0.89 and 0.90 for Chi-Chia-Wan Creek; 0.83 and 0.84 for You-Sheng Creek. The two calibrated parameter sets were applied to the 2007–2008 discharge data for validation. Similarly, the two parameter sets also performed the simulation applicably with the performance measures of 0.84 and 0.89 for C1 (Chi-Chia-Wan) and 0.81 and 0.83 for Y1 (You-Sheng), as Fig. 2 shows. In C1, both the peak flow and low flow could be accurately simulated. In Y1, the peak flow could also be accurately estimated but the simulated low flow was a little bit higher than the observed one. This inconsistent outcome might be a result of the irrigation of some illegal plantation in Y3 and Y7. Nevertheless, the comparable results between the calibration and validation provided the reliable simulations and showed the confidence in these parameter sets, which were the basis for estimating the ungauged sub-catchments.

3.2 NO_3-N concentration pattern

The time series variation of NO_3-N concentration and simulated discharge (including two high-frequency sampling during typhoon periods) in Y1 and K1 were shown in Fig. 3. The NO_3-N concentration patterns in the two catchments were quite different. In Y1, we found the decreasing trend of NO_3-N concentration (from 0.35 to 1.5 mg/L) during the study period. The decrease of NO_3-N concentration could be attributed to the reduction of agricultural activities since 2005. Moreover, a distinct notification was the highest peak of nitrate concentration, which always occurred during the first flush flood in each year. It might indicate that the first typhoon could flush out the most accumulated nitrate near surface. After that, the nitrate concentrations were not much higher than normal condition even during the typhoon period, because of the limited nitrate supply near the surface or insufficient accumulation. By contrast, K1 revealed an opposite pattern, which was regarded as a pristine catchment (depending on the land use pattern; see Fig. 1 or Table 2). General speaking, the nitrate concentration was around $0.1 \sim 0.4$ mg/L, and the seasonal variation was also insignificant. Compared with Y1, the nitrate concentrations were much lower and the nitrate peak also only occurred in the first rainstorm. However, in contrast to Y1, the nitrate concentrations during the typhoon period were around 0.2 mg/L, which was a little bit higher compared to the normal condition. For nitrate concentration variation, the transport dynamics among nitrogen stock accumulation, rainstorm magnitude, and the rainstorm sequence might need further investigation to reveal the nitrogen removal in forestry ecosystems.

The NO_3-N concentrations of the 16 sites were shown in Table 3 and Fig. 4. Each site had at least 29 samples. We found that the NO_3-N concentration in You-Sheng catchment (Y1 to Y7) were around 0.4 mg/L in the dry season, excluding the high concentration ~ 1.8 mg/L in Y3 and ~ 1.0 mg/L in Y7 due to fewer illegally planted vegetables. Relatively higher NO_3-N concentration in Y2 and Y1 was attributed to the diffusion effect. The concentrations during the wet season were a little higher than those in the dry seasons. In You-Sheng catchment, the coefficients of variation were around 0.24

to 0.87, and no significant season difference could be determined. Moreover, the nitrate concentrations in the You-Sheng catchment increased with the discharge increase (Fig. 4).

In the Yi-Ka-Wan catchment, the sub-catchments can be classified into three distinct types according to the NO_3-N concentration and the land use pattern. T1, C7 and K1 belonged to the first type, pristine catchment. The NO_3-N concentrations were 0.11 ∼0.18 mg/L and 0.10∼0.17 mg/L in dry and wet season, respectively. Such concentration was comparable with the other pristine catchment around the world (Dumont et al., 2005). In terms of land use patterns, the C6, C4, C3, C2, and C1 were the second type, moderately-cultivated catchment. In those catchments, the NO_3-N concentrations were around 0.65 and 0.71 mg/L in dry and wet season, respectively. Those catchments were characterized by a few agricultural activities. C5, the intensively-cultivated catchment, had the highest nitrate concentration. The NO_3-N concentrations were 15.45 and 10.97 mg/L in both the dry and wet seasons, respectively. The relatively low nitrate concentration in the wet season might indicate the nitrogen leaching effect because the nitrate stored in groundwater storage was diluted by the surface or near-surface runoff in the wet season. For the relationship between NO_3-N concentration and discharge, the responses in Yi-Ka-Wan sub-catchments were different from those in You-Sheng ones. The nitrate concentration in Yi-Ka-Wan seemed not to significantly increase with the corresponding discharge increase, except C5. No correlation between nitrate concentration and discharge might reveal a pristine condition. The strong positive correlation between nitrate concentration and discharge in You-Sheng catchment could demonstrate the effect of the fertilizer runoff and agricultural activities. However, whether for the Yi-Ka-Wan or You-Sheng catchment in both dry and wet season, the nitrate concentrations measured in the outlet of each sub-catchment were all highly-positively correlated with the proportion of cultivated land in each sub-catchment.

3.3 NO_3-N flux/yield at the catchment outlets

The NO_3-N flux versus discharge was illustrated in Fig. 5. Generally, the NO_3-N flux was highly and positively correlated to discharge among all sub-catchments (Table 4). In power function fitting, we discovered the R^2 values ranged from 0.59–0.89. The coefficient *a*, indicating the flux amount, was extremely high, as 468.33 in C5. By contrast, the pristine catchments, like C7, T1, and K1 had the lower *a* values. The coefficient, *b*, reveals the enhancement and dilution effect. For most of the 16 sites, the NO_3-N flux seemed to increase with the increasing discharge ($b>1$). However, the distinct dilution effect could be found in the Y1, Y2, and C5 sub-catchment ($b<1$) indicating the significant effect of nitrate leaching to groundwater. Therefore, the NO_3-N flux was diluted as the discharge increased. Moreover, the specific NO_3-N fluxes (normalized by drainage area) ranged from 0.1 to 100 kg/day/km^2. For the pristine catchments, such as T1, C7, and K1 (Fig. 5h,i,m), the NO_3-N fluxes were about 0.1 kg/day/km^2 as the discharge was low. As in the high flow period, the specific NO_3-N fluxes were fewer over 100 kg/day/km^2. For moderately-cultivated catchments, like Y7, Y3, C5 (Fig. 5a,e,k), the specific NO_3-N fluxes ranged from 0.1 to 100 kg/day/km^2. We could infer that the specific NO_3-N flux would exceed 100 kg/day/km^2 as flooding. The 2007 and 2008 NO_3-N fluxes derived from the three methods (global mean, flow weighted, and rating curve) were illustrated in Table 5. Generally, the annual NO_3-N fluxes ranged from 393.4–42 920.1 kg/km^2/yr, corresponding with the agricultural area. For pristine catchments (including T1, C7, and K1), the annual NO_3-N export was around 393.4–657.7 kg/km^2/yr. For moderately-cultivated catchments, the annual export was around 1568.5–6716.7 kg/km^2/yr. With the intensive agricultural activities, like C5, the NO_3-N flux would surge over 40 000 kg/km^2/yr. The agricultural activities enhanced the NO_3-N over 50–100 times. Compared to the other catchments around the world, the NO_3-N export was around 58.0–582.8 kg/km^2/yr from pristine to intensive agricultural catchments in USA. In Germany, the pristine catchment exported NO_3-N around 400 kg/km^2/yr, but the catchments with larger arable area exported 1850–4120 kg/km^2/yr. As for Australia, the average export from the pristine catchments was around 284 kg/km^2/yr under the relatively high annual rainfall. And the small anthropogenic land would cause NO_3-N export as high as 2304 kg/km^2/yr due to the high annual rainfall. For subtropical mountainous areas, the nitrate concentration in this study was comparable with other catchments around the world. However, over 5–8 times rainfall or discharge caused relatively high nitrate export. Another possible explanation of the nitrate export in forest catchments resulted from the high wet deposition of nitrate (Kao et al., 2004). Under the similar environmental setting between Kao's study and our study catchment, the differences in NO_3-N export between the sub-catchments were significant to identify the over fertilization applications. The relationship between sub-catchment NO_3-N yield (both in 2007 and 2008) and area percentage of cultivated land within the sub-catchment was shown in Fig. 6. Although there were three methods applied to estimate NO_3-N flux, the average NO_3-N flux (of three estimated values) in 2007 and 2008 for each sub-catchment were so consistent that the uncertainty of flux estimation due to sampling frequency and estimation method may be minimized (Table 5).

3.4 NO_3-N yield from land use classes

The effects or contributions from each land use class were then quantitatively determined. In this study, there were 16 constraints (16 sites) to solve 8 unknowns (yield factors of 8 land use classes) implying the results should be relatively robust and reliable. For each year, the optimal combination of 8 land use NO_3-N yields was obtained. The reliable simulations for each site compared with observations were shown in Fig. 7. The subtle differences between the 2007 and 2008 optimal simulations demonstrated the representativeness of the solutions (8 land use NO_3-N yields).

Therefore, the posterior distributions of yield factors for land use class can be determined (Gupta et al., 1998). The optimized NO_3-N yield factors for each land use were: 488.5±25.1 kg/km^2/yr for primary forest; 888.5±25.1 kg/km^2/yr for mixed forest; 1311.6±29.3 kg/km^2/yr for second forest; 3930.8±41.8 kg/km^2/yr for grass; 2123.2±58.6 kg/km^2/yr for bareland; 5423.1±58.6 kg/km^2/yr for orchard; 298 465.4±3347.2 kg/km^2/yr for active vegetation; and 85 308.2±418.4 kg/km^2/yr for inactive vegetation (Table 7). As expected, the forests had the lowest yield. The orchard, active vegetation, and inactive vegetation, which all of them involved fertilization, had the highest yield. The bare land and grass had the medium value.

As for the impacts of agricultural activities, the orchard yielded 5423±58.6 kg/km^2/yr, which was over 10 times higher than the forest, and could be regarded as one of the nitrate export sources. Active vegetation's yield factor was even higher, which was around 298 465.4±3347.2 kg/km^2/yr. We were not surprised at such extremely high value. For the moderately-cultivated catchments, agricultural land occupied only ∼1–2%, the nitrate concentration increased 10 fold higher than the pristine catchment's. If the NO_3-N yield from the forest in the cultivated catchment remained the same in the pristine one, the agricultural land should yield 500–1000 times more than the forestry land to reach the nitrate concentration level. The major source of this high yield resulted from over fertilization. In this region, most vegetable farms located in the relatively gentle area, such as fluvial terraces. Those areas were composed by gravels without fertile soil and fine grains. To maintain the production of vegetables, the farmer always used excess fertilizers. The previous research (Peng et al., 2006, 2007) addressed that farmers in this area customarily fertilized 300 000 kg urea ($(NH_2)_2CO$) per km^2 annually on vegetable farms, which equaled to 138 000 kg-N/km^2/yr. This amount was comparable to the estimated NO_3-N yield from the active vegetation and evident. Surprisingly, the NO_3-N yield from the inactive vegetation was still high after being expropriated for three years. Although the yield factor has dropped to one-fourth of the active vegetation's, which was around 85 308.2±418 kg/km^2/yr. The results implied the excess fertilizer, exceeding the demand of the vegetation, remained in the soil and then gradually released into the stream or leached to groundwater. The farm expropriation could certainly reduce the pollution but it would be time-consuming. The high NO_3-N yield from the inactive vegetation was the witness.

Land managers are concerned with land use policy and planning to improve water quality. The nitrate yield factors estimated by this study provided an important guideline. The possible impact of any new development can be considered in advance. By contrast, the possible land recovery can also be evaluated. This study is valuable for land managers who attempt to evaluate the land use changes and assess the environmental impacts.

4 Conclusions

Land use, particularly the proportion of agricultural land, was regarded as a significant factor dominating the nitrate export. And nitrate is a major cause of eutrophication and episodic acidification. Owing to the different environmental settings and agricultural activities, land use planning and management should base on the local background in the regional scale, such as understanding the possible contribution of each land use class to assess environmental impacts. In this study, we intensively investigated 16 sites in Chi-Chia-Wan catchment, which is the sole habitat for the endemic species, the Formosan landlocked salmon (*Oncorhynchus masou formosanus*).

We found the nitrate concentration in pristine catchments was around 0.099–0.178 mg/L, which is comparable with the other forestry catchment in the world. However, the annual NO_3-N export was much higher, which was around 393.4–657.7 kg/km^2/yr due to the larger rainfall. As for the impact of agricultural activities, we found that the catchments with some human disturbance, ~5.2%, yielded 5947.2 kg-N/km^2/yr, which were over 10-times higher than usual. This is an important baseline for comparing with other climate areas around the world. It also reveals that the subtropical mountainous catchment is relatively sensitive in responding to the smaller land use change with such intensive fertilization.

Based on the intensive observations, MO_3-N yield factor for each land use was estimated. The forestry land yielded 488.5±25.1 kg/km^2/yr and the vegetable farm yielded 298 465.4±3347.2 kg/km^2/yr, which was 1000 times higher than the forestry. Even the inactive vegetable farm, where farmers stopped fertilizing, yielded 85 308.2±418 kg/km^2/yr. It implied the groundwater contamination is difficult to removal. The estimated nitrate yield for each land use class offers a useful basis for land managers to assess the possible impact of land use changes.

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Table 1. The landscape characteristics of the sampling sites.

Site ID	Drainage area (km^2)	Average slope (degree)	Average elevation (m)	Max. flow length (km)	Stream type[*]
Y7	4.34	55.8	2336	5.18	M
Y6	2.07	53.6	2208	3.11	T
Y5	4.43	60.0	2287	4.06	T
Y4	2.81	57.1	2310	3.35	T
Y3	23.28	56.5	2216	11.46	M
Y2	2.99	60.7	2259	3.92	M
Y1	30.92	56.8	2182	15.48	M
T1	11.31	72.3	2560	6.12	T
C7	25.36	76.9	2883	10.49	M
C6	39.75	74.1	2735	11.93	M
C5	1.59	57.9	2204	3.04	T
C4	48.66	71.4	2634	14.21	M
K1	21.05	74.4	2577	10.80	T
C3	71.91	71.8	2596	16.32	M
C2	74.03	71.4	2581	17.09	M
C1	105.01	67.1	2463	17.13	M

[*] M means the sampling sites located in main streams and T indicated the sites in tributaries. The line separated the You-Sheng and Yi-Ka-Wan subcatchment.

Table 2. The land use composition (%) of these sampling sites.

Site ID	Natural forest	Mixed forest	Secondary forest	Grass	Bare land	Orchard	Active vegetation	Inactive vegetation
Y7	8.9	1.9	85.0	1.5	1.3	0.0	0.0	3.0
Y6	0.0	10.3	88.5	0.0	0.9	0.0	0.0	0.6
Y5	4.1	20.4	67.2	0.5	2.2	5.1	0.0	1.4
Y4	0.9	24.9	71.2	0.8	1.0	0.4	0.0	0.7
Y3	2.9	21.8	66.4	0.9	1.9	2.9	0.0	4.1
Y2	0.2	23.1	70.5	1.1	1.8	2.6	0.0	0.1
Y1	2.6	23.5	62.9	0.8	1.9	5.2	0.0	3.7
T1	67.8	2.2	26.8	2.7	0.4	0.0	0.0	0.0
C7	76.6	3.5	4.2	11.7	3.5	0.0	0.0	0.0
C6	66.9	4.4	15.0	10.1	2.4	0.0	0.7	0.0
C5	1.1	46.1	9.7	17.2	1.2	11.6	13.1	0.0
C4	54.9	11.6	17.4	10.7	2.2	1.3	1.3	0.0
K1	47.8	22.5	19.7	7.6	2.2	0.1	0.0	0.0
C3	50.7	15.2	19.2	10.1	2.2	1.2	0.9	0.0
C2	47.4	15.9	21.8	9.7	2.0	1.7	1.0	0.0
C1	35.7	17.8	32.7	7.4	2.1	2.5	1.4	0.5

Table 3. The sampling number, mean and coefficient variation of NO_3-N concentration in the 16 sites.

Site ID	Sample number	Dry season (mg/L)		Wet season (mg/L)	
		Mean	C.V.	Mean	C.V.
Y7	30	1.034	0.41	0.883	0.87
Y6	29	0.377	0.24	0.497	0.43
Y5	30	0.404	0.39	0.619	0.55
Y4	29	0.447	0.42	0.646	0.61
Y3	29	1.800	0.45	1.876	0.67
Y2	29	1.075	0.45	1.104	0.40
Y1	213 (44)	1.093	0.71	1.447	0.42
T1	27	0.178	0.58	0.169	0.22
C7	60 (41)	0.113	0.34	0.099	0.30
C6	30	0.436	0.38	0.587	0.40
C5	30	15.445	0.22	10.972	0.19
C4	30	0.910	0.28	1.003	0.29
K1	217 (43)	0.163	0.95	0.165	0.42
C3	30	0.504	0.36	0.456	0.28
C2	29 (44)	0.673	0.24	0.689	0.32
C1	218	0.723	0.32	0.824	0.40

The parentheses indicate the typhoon sample number

Table 4. The annual NO_3-N flux estimator for the 16 sites and the R^2 values.

Site ID	Power fitting, $F = aQ^b$:	R^2
Y7	$33.17\,Q^{1.34}$	0.73
Y6	$32.6\,Q^{1.22}$	0.89
Y5	$19.37\,Q^{1.40}$	0.88
Y4	$30.74\,Q^{1.28}$	0.84
Y3	$6.94\,Q^{1.34}$	0.75
Y2	$22.04\,Q^{0.91}$	0.59
Y1	$3.00\,Q^{0.97}$	0.75
T1	$1.13\,Q^{1.14}$	0.66
C7	$0.35\,Q^{1.04}$	0.68
C6	$0.74\,Q^{1.43}$	0.79
C5	$468.33\,Q^{0.84}$	0.81
C4	$1.35\,Q^{1.21}$	0.86
K1	$0.48\,Q^{1.44}$	0.75
C3	$0.49\,Q^{1.08}$	0.74
C2	$0.62\,Q^{1.16}$	0.87
C1	$0.41\,Q^{1.21}$	0.88

Table 5. The specific NO_3-N flux among the 16 sites.

Site ID	Annual specific nitrate yield (kg/km^2/yr)							
	2007				2008			
	Rating curve	Global mean	Flow weighted	average	Rating curve	Global mean	Flow weighted	average
Y7	4704.8	2480.5	3979.2	3721.5	4692.2	2594.4	4161.8	3816.1
Y6	2099.5	1136.9	1469.2	1568.5	1989.7	1189.1	1586.3	1588.4
Y5	3284.5	1291.9	1902.3	2159.6	3060.0	1351.2	2080.9	2164.0
Y4	3019.7	1343.6	2007.7	2123.7	2770.6	1405.3	2099.8	2091.9
Y3	8075.3	4728.5	7139.3	6647.7	7737.6	4945.5	7466.9	6716.7
Y2	1968.2	2816.4	2480.5	2421.7	2110.9	2945.7	2594.4	2550.3
Y1	2576.8	3307.4	3229.9	3038.0	2720.5	3459.1	3378.1	3185.9
T1	630.7	606.3	642.0	626.3	615.8	577.0	610.9	601.2
C7	396.1	392.0	392.0	393.4	379.6	373.0	373.0	375.2
C6	2585.4	1817.4	2173.7	2192.2	2591.3	1729.4	2068.5	2129.7
C5	36 146.3	47 607.6	45 006.3	42 920.1	33 621.7	45 304.0	42 828.5	40 584.7
C4	4719.3	3385.3	3741.6	3948.7	4670.2	3221.5	3560.6	3817.4
K1	761.4	534.5	677.1	657.7	764.4	508.7	644.3	639.1
C3	1856.5	1710.5	1746.1	1771.0	1792.3	1627.7	1661.6	1693.9
C2	3150.4	2423.1	2601.3	2724.9	3088.1	2305.9	2475.4	2623.2
C1	3774.7	2736.7	3296.2	3269.2	3735.4	2604.3	3136.7	3158.8

Table 6. The NO_3-N annual yield observed in this study and others around the world.

Region	Site number	Runoff (mm/yr)	Anthropogenic land (%)	NO_3-N export ($kg/km^2/yr$)	Citation
Northeastern US	8	591	9.81	132.9	Boyer et al. (2002)
Northeastern US	8	500	32.85	582.8	Boyer et al. (2002)
California, USA	11	421	3.2	58.0	Sobota et al. (2009)
California, USA	12	235	24.4	170.9	Sobota et al. (2009)
Central Germany	3	117	66.2	1850–4120	Rode et al. (2009)
Germany	2	257–538	<1.0	336–493	Langusch et al. (2002)
Northeastern Australia	13	965	0.4	284	Hunter et al. (2008)
Northeastern Australia	3	1960	1.0	2304	Hunter et al. (2008)
Northern Taiwan	4	2100	<0.1	660	Kao et al. (2004)
Northern Taiwan	4	2100	>1.0	2550	Kao et al. (2004)
Central Taiwan	3	3300	<0.1	548.3	This study
Central Taiwan	13	3300	5.2	5947.2	This study

Table 7. The NO_3-N annual yield for the land use class.

	NO_3-N annual yield (kg/km^2/yr)			
Landuse class	Average	Standard deviation	Maximum	Minimum
Natural forest	488.5	25.1	524.9	420.7
Mixed forest	888.5	25.1	924.9	820.7
Secondary forest	1311.6	29.3	1354.1	1232.5
Grass	3930.8	41.8	3991.5	3817.9
Bare land	2123.1	58.6	2208.2	1965.1
Orchard	5423.1	58.6	5508.2	5265.1
Active vegetation	298 465.4	3347.2	303 323.5	289 431.4
Inactive vegetation	85 308.2	418.4	85 915.4	84 178.9

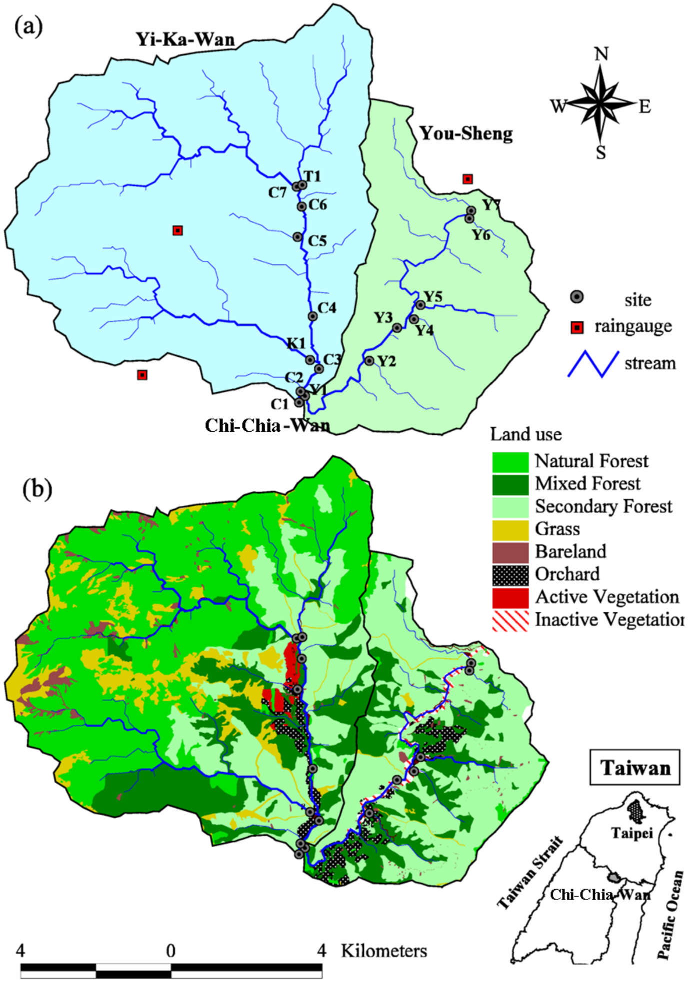

Fig. 1. Landscape **(a)** and land use pattern **(b)** in the study area.

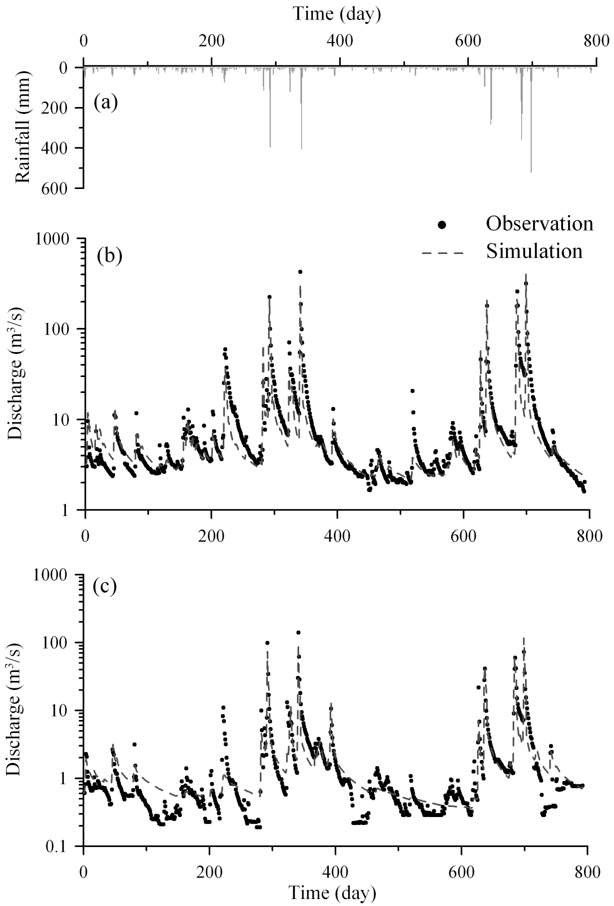

Fig. 2. The observed and simulated discharge from 1 November 2006 to 31 October 2008. The rainfall hyetograph was shown in plot **(a)**. The observed and simulated discharge in C1 and Y1 were shown in plot **(b)** and **(c)**, respectively. The black dot meant the observation and the gray dash curve was the simulation.

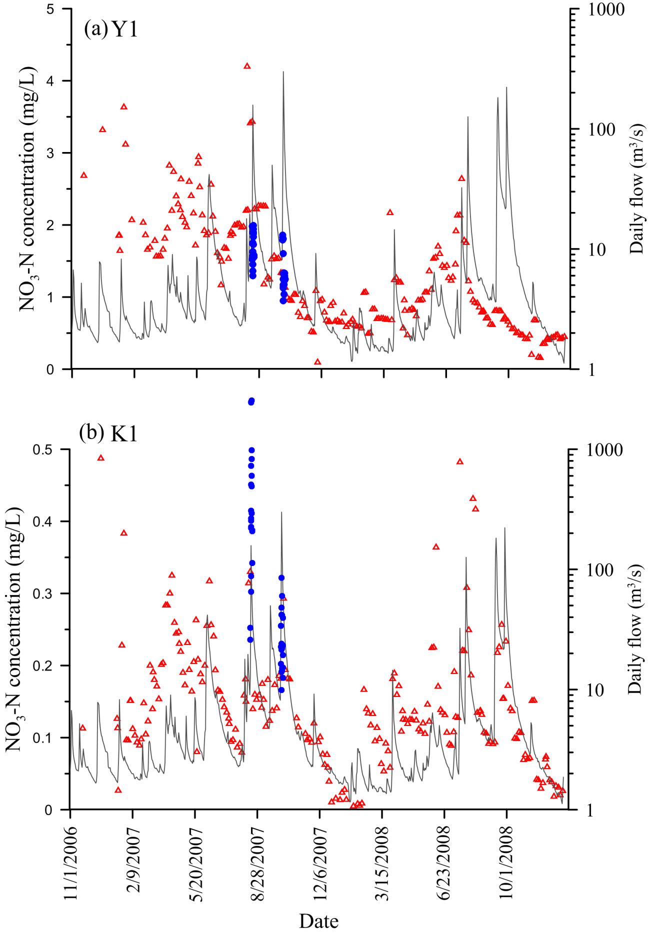

Fig. 3. The observed NO$_3$-N concentration and the simulated discharge. The blue circles were samples taken during the typhoon periods at a frequency of 3 h.

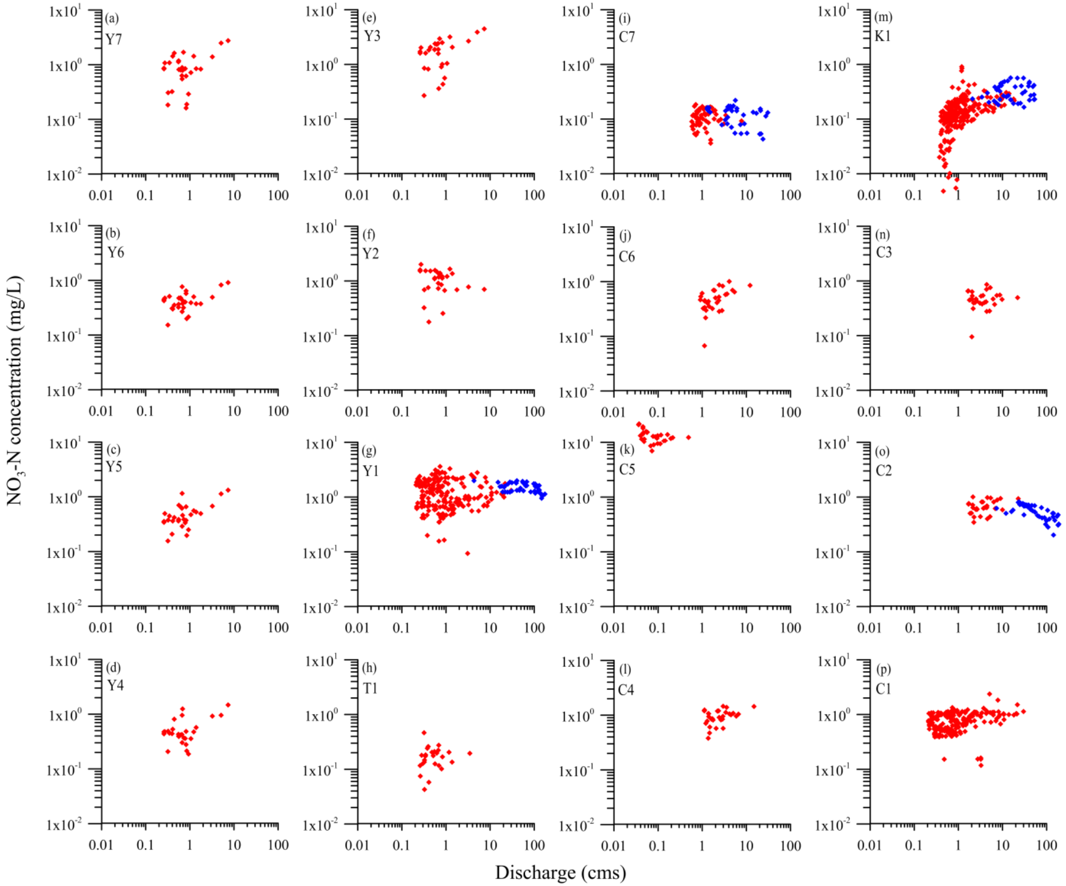

Fig. 4. NO_3-N concentration versus discharge among the 16 sites. The blue circles indicate the typhoon samples.

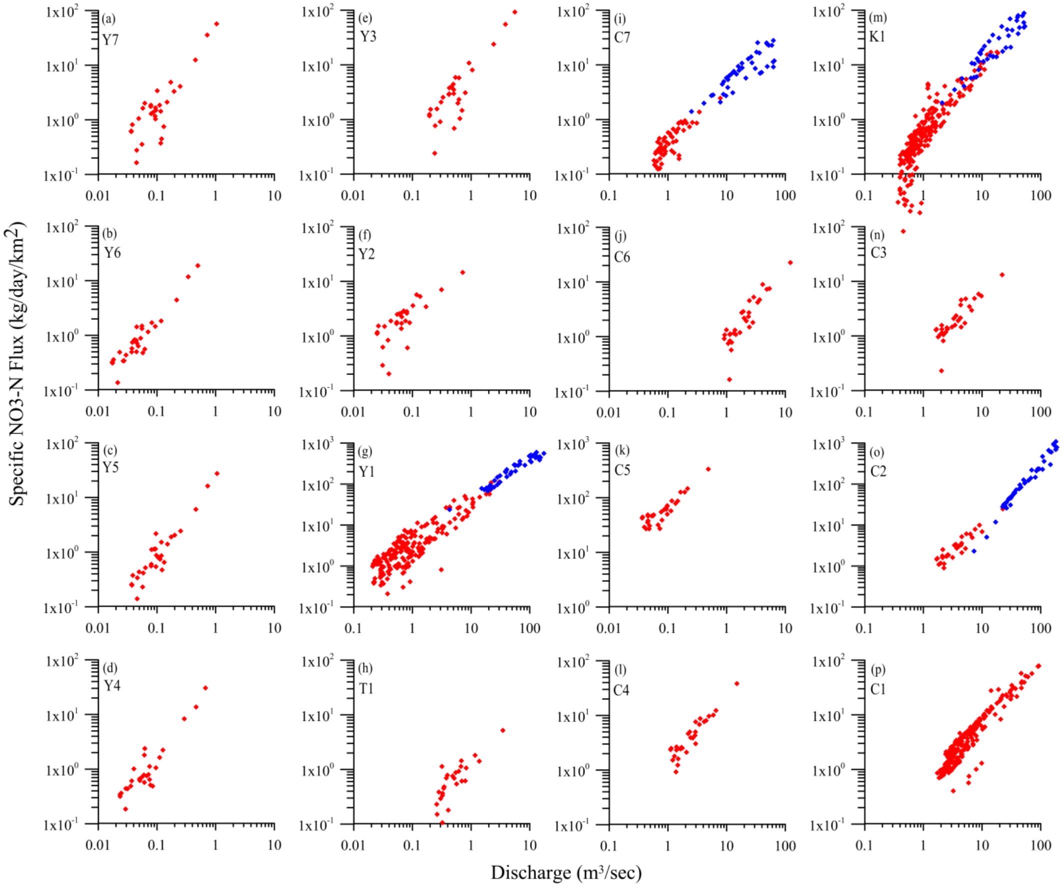

Fig. 5. The specific NO_3-N flux versus the discharge among the 16 sites and the blue circles are the typhoon samples.

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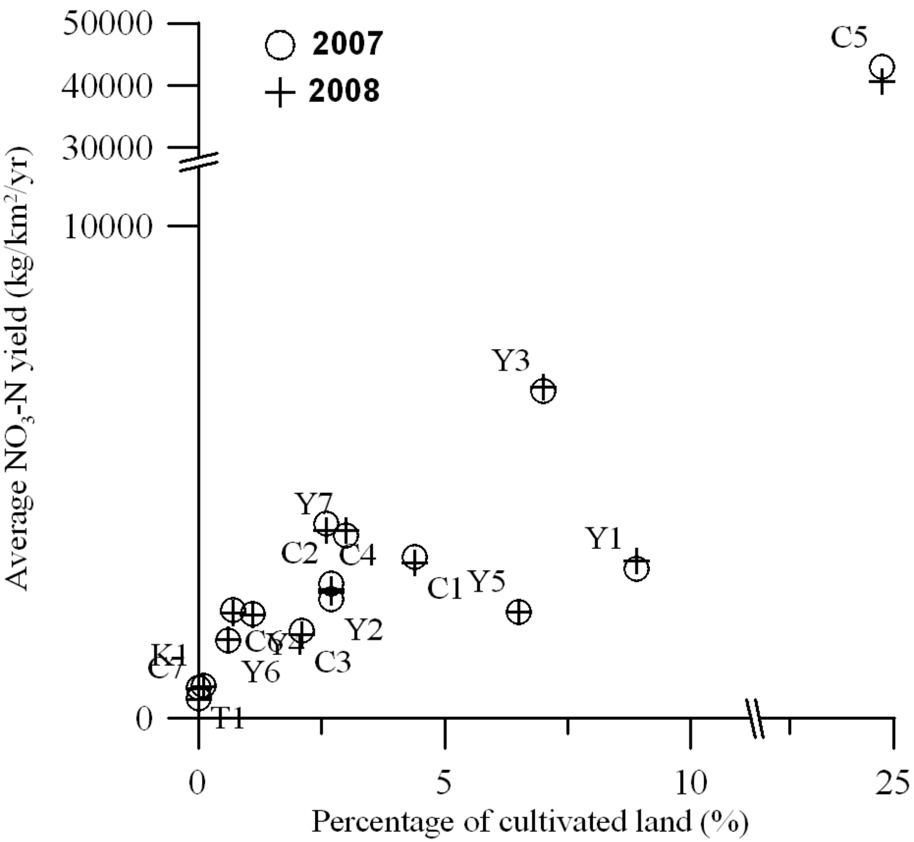

Fig. 6. The relation between the observed NO$_3$-N export and the percentage of cultivated land (%) for the 16 sites in 2007 and 2008.

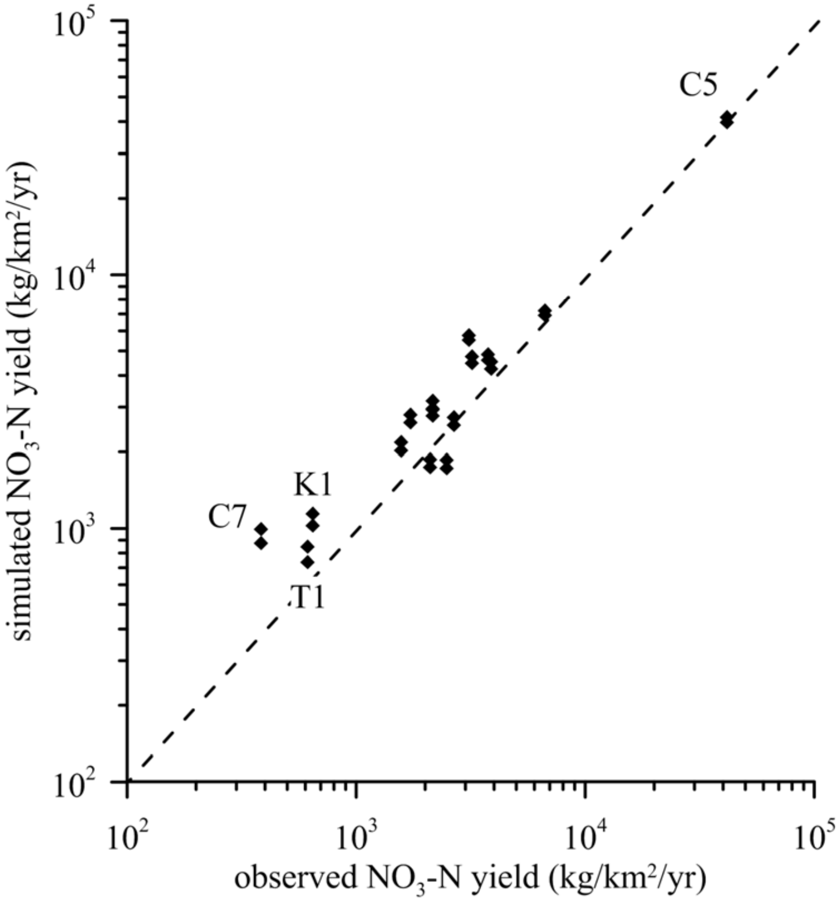

Fig. 7. The simulated and observed NO_3-N yield for the 16 sites. The simulated yields were derived from the optimal NO_3-N yield factor combination in 2007 and 2008, respectively. The dashed line indicated the 1:1 line.