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Effects of different N sources on riverine DIN export and retention in subtropical high-standing island, Taiwan

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Abstract

Increases in nitrogen (N) availability and mobility resulting from anthropogenic activities has substantially altered N cycle both locally and globally. Taiwan characterized by the subtropical montane landscape with abundant rainfall, downwind to the most rapidly industrializing east coast of China can be a demonstration site for extreme high N input and riverine DIN (dissolved inorganic N) export. We used 49 watersheds classified into low-, moderate-, and highly-disturbed categories based on population density to illustrate their differences in nitrogen inputs through atmospheric N deposition, synthetic fertilizers and human emission and DIN export ratios. Our results showed that the island-wide average riverine DIN export is $\sim 3800\,\mathrm{kg\,N\,km^{-2}\,yr^{-1}}$, approximately 18-fold higher than the global average mostly due to the large input of synthetic fertilizers. The average riverine DIN export ratio is 0.30–0.51, which is much higher than the average of 0.20–0.25 of large rivers around the world indicating excessive N input relative to ecosystem demand or retention capacity. The low-disturbed watersheds, despite of high N input, only export 0.06–0.18 of the input so were well buffered to changes in input quantity suggesting high efficiency of nitrogen usage or high N retention capacity of the less disturbed watersheds. The high retention capacity probably is due to the effective uptake by secondary forests in the watersheds. The moderate-disturbed watersheds show a linear increase of output with increases in total N inputs and a mean DIN export ratio of 0.20 to 0.31. The main difference in land use between low and moderately disturbed watershed is the relative proportions of agricultural land and forests, not the built-up lands. Thus, their greater DIN export quantity could be attributed to N fertilizers used in the agricultural lands. The greater export ratios also imply that agricultural lands have lower proportional N retention capacity and that reforestation could be an effective land management practice to reduce riverine DIN export. The export ratio of the highly-disturbed watersheds is 0.42–0.53, which is very high and suggests that much of the N input is transported downstream and the need of improvement in wastewater treatment capacity or sewerage systems. The increases in riverine DIN

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export ratio along with the gradient of human disturbance indicates a gradient in N saturation in subtropical Taiwan. Our results help to understand factors controlling riverine DIN export and provide a sound basis for N emissions/pollution control.

1 Introduction

Rapid increases of population and food demand inevitably hasten the emission of anthropogenic nitrogen (N) via N-rich sewage, fertilizer runoff, and burning of fossil fuels leading to 10-fold increase in anthropogenic N emission since the 1860 s (Galloway et al., 2004). A substantial proportion of the increased N input is transported to river systems. Observed global riverine DIN export varies from 0.60 to 2200 $kg N km^{-2} yr^{-1}$ (He et al., 2011) while modeled DIN export varies from 0.0004 to 5217 $kg N km^{-2} yr^{-1}$ (Dumont et al., 2005). The more than 6-order variation reflects large spatial differences and complexity of N input, retention and transport processes (Howarth, 1998; Smith et al., 2005; Seitzinger et al., 2005). The recent high N input may have exceeded the limit of the earth system as a safe operating space for humanity (Rockström et al., 2009). Unfortunately, the increasing trend of N emission is likely to continue in the near future, especially in Asia (Ohara et al., 2007). The elevated anthropogenic N has increased the availability and mobility of N and as such altered N cycle from local to global scales (Vitousek et al., 1997; Gruber and Galloway, 2008). High quantity of N fertilizers has been shown to degrade soil fertility, water quality, and create hypoxic zones in coastal areas (Fitzpatrick et al., 2004; Brown et al., 2009; Tu, 2009; Jiang and Yan, 2010). Because N is a common limiting nutrient element in many ecosystems, availability of N is an important factor characterizing biodiversity and ecosystem function (Aber, 1989, 1998). Anthropogenic N enrichment has resulted in decline in ecosystem function and biodiversity at a global scale (Townsend et al., 2009; Hutchison and Henry, 2010).

In freshwater systems, eutrophication is a common consequence of the increased availability of dissolved inorganic N (DIN includes NO_2, NO_3, and NH_4), aside from

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increased phosphorus levels (Conley et al., 2009). Currently, most empirical studies of DIN export were conducted at a geographically limited group of drainage basins in the developed temperate region (Smith et al., 2005; Galloway et al., 2008). In contrast, N deposition and DIN export in subtropical and tropical regions such as eastern Asia characterized by high population, intensive agriculture system, and rapid industrialization is not well documented. Because industrialization in East Asia is among the most rapid in the world, the impact of enhanced anthropogenic N emission on watershed nutrient cycling is expected to get worse in near future. Thus, understanding DIN concentration and export in this region is critical for a thorough understanding of global watershed nutrient cycling in relation to anthropogenic N emission.

Increases in population, agriculture activities, and atmospheric N deposition are the well-recognized indicators of elevated riverine DIN export. Levels of proportional riverine DIN export, theoretically less than the total input, is determined by N retention and transfer processes within a watershed. Thus, the riverine DIN export ratio, defined as the riverine DIN export over the total inputs, can be used to evaluate the nutrient-cycling function of a watershed. Quantification of N inputs and outputs of river systems with different levels of human disturbance (e.g., Groffman et al., 2004) provides scientific evidences of how human activities affect N cycling at a scale relevant to land management and a basis for sound land management to improve water quality. Previous studies indicated that ~ 20–25 % of the N inputs is transported by river systems and the rest is retained or denitrified in large rivers in North America and Europe (Howarth et al., 2006). Recent studies show that riverine N export will increase 10–27 % in the next 3 decades due to the increase of N inputs (Bouwman et al., 2013), while riverine N export ratio will increase 10–35 % due to increase of rainfall which may shorten the water residence time (Howarth et al., 2006). Compared to the temperate region, we have very limited knowledge on riverine N export ratios in relation to human activities in tropical and subtropical regions regardless of their very rapid increases in N emission.

Taiwan is a 36 000 km^2 subtropical island characterized by rugged topography, high rainfall, high industrialization and a very large population (23 million) so that like many

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countries in East Asia the emission of N and export of DIN in Taiwan is expected to be very high. In addition, with the rich background information on various forms of N inputs, topography and land use intensity, it is an ideal site to assess the relative contributions of major emission sources to total N input and the effects of physical environmental characteristics and human impacts on DIN export. Therefore, this study aims to (1) characterize the riverine DIN export in subtropical watersheds in Taiwan and (2) explore the relationship between land use and DIN export in Taiwanese rivers compared to other rivers of the world.

2 Methods

2.1 Population density and land use

There are over 23 million people living in Taiwan with area of 36 000 km^2 giving a population density of ~ 640 cap km^{-2} (Fig. 1a). Most people live in the western plains, particularly in cities. For example, over 5 million people settle in the biggest metropolitan area, Taipei City, and the population density reaches 20 000 cap km^{-2} there. Population density from county-based census is converted to county-based population density which would be clipped by the watershed polygons afterward to compute population density for each watershed. Since the human-associated wastewater is a main source of riverine N export, the per capita N loading is essential and is estimated by GDP. The average Taiwan GDP during 2002–2012 is ~ 30 000 US dollars, using GDP-based estimation proposed by Van Drecht et al. (2009), the per capita human N emission is estimated to be ~ 6.42 kg N cap^{-1} yr^{-1}.

Forest is the main land cover type of the island and agricultural land mainly distributed on the west plains with some scattering on the hillsides. The agriculture in Taiwan is featured by an intensive cropping system, which can produce two to five harvests a year (two batches per year for paddy rice) due to intense sunlight, high precipitation and temperature. The total arable land area is ~ 8550 km^2, in which paddy

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fields account for 4445 km^2 mostly in the western plain (Fig. 1b). Fodder and sweet maize are planted on the plains from central to southern Taiwan and sugar canes are cultivated in the southern coastal plain (Council of agriculture, executive yuan, Taiwan, www.coa.gov.tw). The remaining arable lands (4106 km^2) spread over the hills, usually along the roads or streams. Still some arable lands distribute in uplands with tea, orchard, vegetable, and bamboo as the main crops. The tea fields and bamboo fields account for approximately 190 and 300 km^2, respectively. Agriculture production in Taiwan largely depends on immense inputs of synthetic fertilizers and crop protection measures. For examples, 500 and 420 $kg\,N\,ha^{-1}$ of N fertilizers are commonly applied for bamboo shoot and tea fields per year and paddy requires 246 $kg\,N\,ha^{-1}$ of N fertilizers annually (Table 1). Other crops, such as maize, sugar cane, watermelon, banana, and pineapple, also need more than 100 $kg\,N\,ha^{-1}$ of N fertilizers per year (Table 1). The intensive fertilizer application is not unique to Taiwan as it is also common in other East Asian countries. For example, the synthetic fertilizer applied in Lake Dianchi in China can be up to 150–200 $kg\,N\,ha^{-1}$ (Gao et al., 2014).

2.2 Basic watershed characteristics

A total of 49 watersheds distributing throughout the island were used for estimates of watershed DIN export in this study (Fig. 1c). The watersheds were classified into 3 levels of human disturbance: low disturbed, moderately disturbed, and highly disturbed based on population density of < 20, 20–200 and > 200 $cap\,km^{-2}$. There are 16, 15, and 18 watersheds in the low disturbed, moderately disturbed, and highly disturbed categories, respectively. The watershed environmental settings of the three categories are given in Table 2. The drainage areas of most of the watersheds are less than 1000 km^2. The average slopes and the drainage densities of the three categories vary from 66 to 29 % and 1.4 to 1.6 km^{-1} (Table 2). The mean annual runoff of the 49 watersheds is between 2200 and 2500 mm.

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Forests dominate the landscape covering more than 50 % in all the watersheds and the proportion of forest cover decreases sharply from 0.87 to 0.51 from low to highly disturbed watersheds (Table 3). Contrarily, the proportion of built-up land increases from 0.03 to 0.11 and agricultural land increases from 0.11 to 0.38 from low to highly disturbed watersheds, respectively (Table 3). Regarding crops grown, grain and orchards are the dominant crops in low disturbed watersheds. For moderately disturbed watersheds, orchard and bamboo are the main agricultural products. Orchard and paddy account for a considerable fraction in highly disturbed watersheds (Table 3). Unlike other watersheds in temperate regions where agricultural lands may comprise more than half of the drainage area, agriculture in Taiwan and other mountainous region is constrained by the rugged landscape.

2.3 N deposition

Data of atmospheric N deposition, synthetic fertilizer use, and human emission was collected and compiled for the determination of the N inputs and the computation of riverine N export ratios. Note that the emission from livestock is excluded from the analysis because it is not a primary one and most of the livestock farms are located in the central to southern coastal zones, not in the mountainous region where the studied watersheds are located.

For atmospheric N deposition, two modeling studies estimated a global average of 145.3 and 282.9 $kg\,N\,km^{-2}\,yr^{-1}$ for 1993 and 2050, respectively (Lelieveld and Dentener, 2000; Jeuken et al., 2001). Based on the two studies the N deposition in East Asia surges from $\sim 1600\,kg\,N\,km^{-2}\,yr^{-1}$ in 1993 to $\sim 3800\,kg\,N\,km^{-2}\,yr^{-1}$ in 2050 with a cross-year average of 2700 $kg\,N\,km^{-2}\,yr^{-1}$ (Fig. 2). Our local N deposition data were based on national-wide acid rain monitoring network with a total of 11 sites initiated by Taiwan EPA (Environmental Protection Administration) since 2002 (Fig. 2). Rainfall samples were collected on daily basis for determination of pH, conductivity, and concentrations of major cations and anions. From data of the 11 sites a map of island-

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wide N deposition was developed using the inverse distance weighted method with an exponent of 2.

From the 2007–2013 island-wide monitoring data the annual mean wet N deposition, including NO_3 and NH_4, is 1515 $kg\,N\,km^{-2}\,yr^{-1}$ with NH_4-N accounting for 59 % of the total deposition. Dry N deposition was estimated to be 23–50 % of wet N deposition (unpublished data from Lin, N.H.) and thus a factor of 0.4 is used to estimate the dry N deposition. The total N deposition was computed as the summation of wet and dry deposition as presented in Fig. 2 and has an island-wide mean of ∼ 2121 kg $kg\,N\,km^{-2}\,yr^{-1}$. The consistent N deposition from global modelling and local observations illustrate that high deposition is one of the features in this region.

2.4 Stream water level and DIN monitoring

The Water Resources Agency (WRA, http://www.wra.gov.tw/) and Environmental Protection Administration (http://www.epa.gov.tw) of Taiwan are in charge of monitoring water quantity and quality, respectively. Streamflow and DIN data from 49 water level monitoring stations (Fig. 1c) in which both water level and water quality data are available during 2002–2012 are used in this study. The water level stations are widely deployed in bridge piers and record water levels automatically. Because typhoon storms often re-shape the channel geometry, the stream cross sections are measured two or three times per year to ensure the applicability of the rating curve for streamflow estimation.

For water quality, the sampling crews of WRA and EPA take the water samples on a monthly basis. Temperature, pH and electrical conductivity (EC) are measured in situ and other parameters including dissolved oxygen, cations, anions, *Escherichia coli* (*E. coli*), and selected heavy metals, are determined followed the standard operation procedure in laboratories. For DIN species, the ammonium was measured monthly, while nitrate and nitrite were analyzed every three months. Based on the monthly and seasonal sampling frequency, the simple approach of a global mean which multiples the average of DIN concentrations by stream discharge was applied.

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3 Results and discussions

3.1 Riverine DIN concentration and flux

The island-wide mean DIN concentration and flux calculated from the 49 watersheds are 148.3 µM and 3800 $kg\,N\,km^{-2}\,yr^{-1}$, respectively. However, they vary considerably among the three categories of watersheds with different levels of human impacts. The mean annual riverine DIN concentrations were 29.9, 56.9, 329.4 µM and the corresponding exports were 909.2, 1815.8, and 8020.3 $kg\,N\,km^{-2}\,yr^{-1}$ for low-, moderately- and highly-disturbed watersheds, respectively (Fig. 3). For N species, mean NO_3 concentration and export increased from 23.8 to 99.6 µM and 724.7 to 3243 $kg\,N\,km^{-2}\,yr^{-1}$ from low to highly disturbed watersheds (Table 4). Similarly, the NH_4 concentration and export increased from 5.6 to 214.2 µM and 169 to 4343 $kg\,N\,km^{-2}\,yr^{-1}$ from low- to highly-disturbed watersheds, respectively (Table 4). The nitrite was not shown since it only accounted for a very small fraction (< 0.05) of DIN. The patterns and levels of DIN concentrations and fluxes of the 49 watersheds were consistent with results reported for over 20 sub-catchments within 2 river networks in northern and central Taiwan (Huang et al., 2012; Lee et al., 2014). Note that NO_3 was the dominant species for low- and moderately disturbed watersheds, but NH_4 was the dominant species and accounted for more than 50 % of annual DIN flux for highly-disturbed watersheds.

Both NO_3 and NH_4 exports were significantly correlated to population density ($r^2 = 0.19$ for NO_3 and 0.18 for NH_4, both p values < 0.01) and proportion of agricultural land cover ($r^2 = 0.34$ for NO_3 and 0.50 for NH_4, both p values < 0.01) (Fig. 4). The intensive cropping system and dense population likely contribute to the 10-fold greater DIN export at the highly disturbed watersheds relative to the low disturbed watersheds. While the relationship between population density and NO_3 and NH_4 export is very similar, NH_4 increased more dramatically than NO_3 with increases in relative agricultural land cover (Fig. 4). Among the three categories of watersheds, NO_3 export increase gradually with increases in both agricultural land cover and population density. By contrast,

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the NH_4 export does not change much from low to moderately disturbed watersheds (Fig. 4), but amplified dramatically with increases in both agricultural land cover and population density in highly disturbed watersheds indicating the saturation of NH_4 retention capacity at highly disturbed watersheds (Fig. 4).

3.2 Total N input

Mean of the total annul N input increases from 4900 kg N km^{-2} yr^{-1} for low disturbed watersheds, 6600 kg N km^{-2} yr^{-1} for moderately disturbed watersheds, to 16 700 kg N km^{-2} yr^{-1} for highly disturbed watershed (Table 5). Because atmospheric N deposition is in a small range in all watershed categories (2000–2200 kg N km^{-2} yr^{-1}), it could not cause the increases in total N deposition from low to highly disturbed watersheds. Instead the increases mainly originate from fertilizer applications and human emissions (Table 5). Assuming synthetic fertilizers are applied only in agriculture lands, the input of N from fertilizers in each percent of agricultural land (a total of 11, 16 and 38 % in low, moderately, and highly disturbed watersheds) increased only slightly from 236 kg N km^{-2} yr^{-1}, in low disturbed watersheds, to 254 kg N km^{-2} yr^{-1} in moderately disturbed watershed to 279 kg N km^{-2} yr^{-1} in highly disturbed watersheds. In contrast assuming human emissions were entirely originating from built-up lands the input of N from human emission of each percent of constructed land increased dramatically from 24 kg N km^{-2} yr^{-1} in low disturbed watersheds, 118 kg N km^{-2} yr^{-1} in moderately disturbed watersheds to 349 kg N km^{-2} yr^{-1} in highly disturbed watersheds. While N input per unit agricultural land is relative constant, input per unit built-up land increased dramatically as the relative area of built-up land and increase indicating disproportional increase in population density as the proportion of build-up land increase. The result illustrates that population density control may be one of the most effective control measures of N emission and the resultant DIN export.

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3.3 River DIN exports in Taiwan and the world

Since 1960's global N flux from land to ocean has been elevated by 2-fold (Howarth et al., 2002). Regionally, it has increased ∼ 4-fold in the Mississippi River basin, 8-fold in northeastern US and has been raised more than 10-fold in the rivers draining into the North Sea (Howarth et al., 2002). Dumont et al. (2005) reported that the global DIN export to coastal waters is ∼ 25 $\mathrm{Tg\,N\,yr^{-1}}$ with more than 60 % stemming from anthropogenic sources. Giving the global land of 120 million $\mathrm{km^2}$, the average DIN export per unit land area is 208 $\mathrm{kg\,N\,km^{-2}\,yr^{-1}}$. Comparing to this global picture, the level of riverine DIN export in Taiwan, ∼ 3800 $\mathrm{kg\,N\,km^{-2}\,yr^{-1}}$, is extraordinarily high as it is 18-fold higher than the global average. Generally, the riverine N export is strongly dependent on climatic factors and human activities; increasing from temperate to tropical climates and from pristine to disturbed watersheds (Howarth, 1998; Smith et al., 2005; Seitzinger et al., 2005). For example, the riverine N export is 76 $\mathrm{kg\,N\,km^{-2}\,yr^{-1}}$ for the watershed with sparse population in northern Canada, but reaches 1450 $\mathrm{kg\,N\,km^{-2}\,yr^{-1}}$ for the developed watersheds of the North Sea (Howarth et al., 1996).

He et al. (2011) reported a positive relationship between DIN export and runoff, population density and proportion of agricultural land cover at the global scale. Adding Taiwan to the global pictures reveals interesting phenomena (Fig. 5). Globally the DIN export increases with increases in runoff and Taiwan is clearly on the very high end of both runoff and DIN export. The global pattern exists regardless of the large differences in the efficiency of wastewater treatment among regions and among countries of the same region suggesting that despite of the large influences of N inputs from wastewater treatment, agriculture and industrial inputs, runoff is predominantly controlling DIN export.

Interestingly, few sites in the study of global N export (He et al., 2011) have annual runoff greater than 1 $\mathrm{m\,yr^{-1}}$, while the runoff of all the 49 watersheds in Taiwan is, on average, 2 $\mathrm{m\,yr^{-1}}$. High runoff is not limited to Taiwan many tropical and subtropical forested watersheds show a similar pattern. For example, the forested watersheds in

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north Australia also have abundant runoff and high DIN export (Hunter et al., 2008). Compared to the global scale the variation of runoff within Taiwan (1.5–2.5 m) is relatively small, but DIN export of the 49 watersheds varies largely even at the global scale (from ∼900 to ∼8000 $kg\,N\,km^{-2}\,yr^{-1}$) (Fig. 5). Yet, even 900 $kg\,N\,km^{-2}\,yr^{-1}$ is on the high end of the global picture. Thus, adding Taiwan to the figure of global N export pattern fills the gap for regions with both high runoff and high DIN export.

Among the three factors, population density, agriculture activity and runoff, only population density shows a consistent positive relationship with DIN export at both the global scale and our island scale (Fig. 5). Runoff is a good predictor only at the global scale and agriculture is a good predictor only at the island scale (Fig. 5). The lack of predictability on DIN export in Taiwan based on runoff might be due to the limited variation in runoff relative to the 10-fold differences in DIN export (Fig. 5). The lack of predictability of agriculture land cover on DIN export at the global scale may seem surprising because it is a well-recognized non-point source of DIN. The differences in the farming practices among regions could be an important factor leading to the lack of a significant positive relationship between agricultural land cover and DIN export. In general, fertilizers are applied at much larger quantities in regions with intensive farming systems such as China, Taiwan, and Thailand than in extensive farming systems such as North America, Europe and Australia. For example, in rice fields N is applied at approximately 418 $kg\,ha^{-1}\,yr^{-1}$ in Taiwan (∼200 $kg\,ha^{-1}$ for a harvest; FAO 2002), 194 $kg\,ha^{-1}\,yr^{-1}$ in China but only 135 $kg\,ha^{-1}\,yr^{-1}$ in California, USA (Yan et al., 2003). As a result of such large differences, DIN export of a watershed with 100 % of rice field in North America may not be different from the export of a watershed with 10 % of rice field in East Asia. As a result of the differences in farming systems, the relationship between DIN export and agricultural land cover is likely comparable only in regions with similar management practices (Huang et al., 2012). For global assessments, the amount of N fertilizers coupling with the proportion of agricultural land cover would be necessary for predicting riverine DIN export in relation to agriculture activities.

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Adding the 49 watersheds in Taiwan to the global picture in He et al. (2011) shows that NH_4 approximately accounts for 10 to 100 % of nitrate and nitrite in other large rivers around the world as well as the low- and moderately disturbed watersheds in Taiwan (Fig. 6). While only one of the 49 watersheds has a NH_4 to NO_3 + NO_2 ratio less than 0.1, approximately 50 % (16/34) of the rivers in He et al. (2011) had a ratio less than 0.1. Moreover, none of the rivers in He et al. (2011) had a ratio greater and 1.0 but 10 of the highly disturbed watersheds in Taiwan has a ratio greater than 1.0 and one of them even reaches 10. Globally NO_3 is the predominant species of DIN export, partly because NO_3 is more stable than NH_4, which is easily oxidized or volatilized. For example, NH_4 volatilization or suspension from agricultural systems in the US accounts for the same order of magnitude as NO_3 leaching into streams (Howarth et al., 1996; Sutton et al., 2000). The oxidization and volatilization are expected to be greater in tropical climates due to warmer temperatures. Besides, the NH_4 may quickly decline in stream systems due to rapid uptake and transformation (Alexander et al., 2000; Peterson et al., 2001). The dominance of NH_4 in DIN export for the highly-disturbed watersheds in Taiwan might be due to high NH_4 input and low dissolved oxygen (DO) levels limiting NH_4 oxidization. The average DO is $\sim 6.8\,mg\,L^{-1}$ for those watersheds with NH_4 to NO_3 + NO_2 ratio greater than 1, and $\sim 8.4\,mg\,L^{-1}$ for the remaining watersheds. The $\sim 6.8\,mg\,L^{-1}$ of DO in those watersheds, in fact, is not very low implying the high DO demand in this region.

Using ^{15}N isotopic analysis Peng et al. (2012) found very limited NH_4 removal in upstream and suggested that NH_4 export from these watersheds to streams is very limited and this explains the very low NH_4 content in the low and moderately disturbed watersheds. In contrast, our highly disturbed watersheds have high NH_4 concentrations and fluxes (Figs. 3 and 4). Water residence time in Taiwan is, in general, very short due to the small drainage area ($< 1000\,km^2$) and steep slopes ($> 30\,\%$) so that is unfavorable for NH_4 removal (Halbfaß et al., 2010) because ammonia oxidization and/or assimilation is minimized. In such lotic environment, some biogeochemical processes such as nitrification may play some role in NH_4 transformation (Venohr et al., 2005), but likely

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insufficient for transforming most of the abundant NH_4 in highly disturbed watersheds. In other words, the high human emission and the short water residence time result in the dominance of NH_4 downstream.

3.4 Riverine N export ratio

Although both total N input and output increase from low to highly disturbed watersheds, the increases are greater for output (from 575 to 15 689 kg N km^{-2} yr^{-1}) than input (from 3805 to 26 668 kg N km^{-2} yr^{-1}) (Table 5). As a result the measured DIN export ratio increases from 0.18 (ranging from 0.05 to 0.20) for the low disturbed watershed, 0.27 (0.11 to 0.38) for the moderately disturbed watersheds, to 0.42 (0.21 to 0.73) for the highly disturbed watersheds, with an average of 0.30 (Table 5).The highly disturbed watersheds have greater DIN export both in absolute quantity (per unit area) and relative to N input (i.e., export ratio) than less disturbed watersheds. Simple linear regression models indicated that the slope between riverine DIN export and N input among the three categories were 0.06, 0.20, and 0.53 with R^2 of 0.03, 0.62, and 0.51 for low to highly disturbed watersheds, respectively (Fig. 7c–e). The regression models support that with per unit increases in N input the rise in DIN export was greater at more disturbed watersheds than less disturbed watersheds. Increases of DIN output in absolute quantity could be at least partially explained by greater N inputs, but other factors must have contributed to the greater DIN export ratios at more disturbed watersheds.

Although there is an overall increases in DIN export ratio with increases in N input, such a relationship does not exist for the low disturbed watersheds in which DIN export ratio is not significantly controlled by N input (Fig. 7b). The capacity of plants to take up N for growth and the amount of N that can be hold in soil (e.g., NH_4 in the inter-layer space of Vermiculite and Montmorillonite and N in soil organic matters) greatly determine ecosystem N retention capacity. If N input is less than the retention capacity, then DIN export may be irrelevant to N input because most of the additional N input may be retained within the watershed until it exceeds the retention capacity.

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This is probably the case for our low disturbed watersheds in which the riverine DIN export is ~ 900 $kg\,N\,km^{-2}\,yr^{-1}$ and seems irrelevant to total input ranging from 2000 to 8000 $kg\,N\,km^{-2}\,yr^{-1}$.

The high DIN export ratios in alpine watersheds of the Front Range of Colorado with low atmospheric deposition in comparison to other regions where watersheds retain more N regardless of higher N input can be used to illustrate the role of biological uptake (Campbell et al., 2000). The N export ratio in alpine watersheds in Colorado ranged from 0.55 to 0.71 although atmospheric deposition was only 320–550 $kg\,N\,km^{-2}\,yr^{-1}$ (Campbell et al., 2000) and by contrast the N export ratio for Baltimore LTER sites was less than 0.1 although the input of 1120 $kg\,N\,km^{-2}\,yr^{-1}$ was much higher (Groffman et al., 2004; Kaushal et al., 2008). It has been suggested that the mature alpine forests in Colorado may have shown symptoms of advanced stages of nitrogen excess so that the forests have very limited capacity to retain N in spite of the low atmospheric N deposition.

Our low disturbed watersheds, which mostly distributed in the mountain ranges are characterized by high atmospheric N input and riverine DIN export ratios (0.18) slightly less than those of large rivers around the world with 0.2–0.25 (Howarth et al., 2006). The low disturbed watersheds are predominately covered by natural forests which experienced intensive exploitation for several decades until 1991, when a strict law to protect natural forests was enforced. It is likely that the secondary forests are still capable of taking up large amounts of N leading to the low DIN export ratio and the lack of a close relationship between N input and output. If forest growth is indeed key to the low DIN export and export ratio, then reforestation could be an effective management practice to reduce DIN export and therefore reduce the risk of downstream eutrophication.

The moderately and highly disturbed watershed show a linear relationship between DIN export and total N input with a slop of 0.2 for moderately disturbed watersheds and 0.53 for highly disturbed watersheds (Fig. 7d and e). The positive linear relationships suggest that the watersheds are unable to retain most of the increased N input and

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the likeliness of N excess (Liu et al., 2010). The greater export ratios in moderately disturbed watersheds than low disturbed watersheds most likely relate to its greater agricultural land cover (16 %) compared to low disturbed watersheds (11 %) (While the proportion of developed area is similar between the two categories of watersheds) and suggest that agricultural lands have lower N retention capacity than forests. Although agricultural lands only cover a small proportion of the watersheds, they have a high impact on riverine DIN export. Thus, concentrating management efforts on agricultural lands could have major effects for the reduction on DIN export. The even greater riverine DIN export ratios of the highly-disturbed watersheds than the moderately disturbed watersheds likely result from greater population density on the constructed lands and greater agriculture intensity at the highly disturbed watersheds.

The considerable variation of export ratio 0.21–0.73 may reflect the different treatment efficiencies in sewage systems and cropping systems among watersheds. In addition, the very high DIN export ratios in highly disturbed watersheds indicate that the N input is transported by riverine systems very efficiently so that eutrophication threat is even greater downstreams. The high export ratios also indicate that the highly disturbed watersheds are probably at more advanced stages of N excess and that the current removal processes (e.g., denitrification, vegetation uptake) cannot compensate for the increasing N additions (Downing et al., 1999). Additionally, recent studies report increases in rainfall intensity in Taiwan as a result of recent climate change. (Huang et al., 2014; Liu et al., 2009). Such increases in rainfall intensity would lead to shortened water residence time and therefore further enhance riverine DIN export. Improving the sewage systems and the efficiency of N fertilizer application is of particular importance for mitigating riverine N export in Taiwan and other regions with high population densities and intensive cropping systems.

4 Concluding remarks

We characterized riverine DIN export of subtropical watersheds in Taiwan using a total of 49 watersheds classified as low-, moderately-, and highly-disturbed based on population densities (from < 20 to $> 200\,\text{cap}\,\text{km}^{-2}$). The average riverine DIN export is $\sim 3800\,\text{kg}\,\text{N}\,\text{km}^{-2}\,\text{yr}^{-1}$, approximately 18 times higher than the global average and is attributable to the high runoff, atmospheric N deposition, intensive crop systems, and high population densities. Since the runoff varied only two fold and atmospheric deposition varied less than 30 %, the ~ 10 times greater DIN export of highly disturbed watershed ($8000\,\text{kg}\,\text{N}\,\text{km}^{-2}\,\text{yr}^{-1}$), compared to low disturbed watersheds ($900\,\text{kg}\,\text{N}\,\text{km}^{-2}\,\text{yr}^{-1}$) likely resulted from differences in inputs from agricultural lands, total human emission and watershed N retention capacity. Nitrate is the dominant N species for large rivers around the world and our low and moderately disturbed watersheds, whereas NH_4 is the dominant species in the highly-disturbed watersheds possibly due to high human emission and the short water residence time, which is unfavorable for ammonia removal. The high N export to downstream system poses risk of eutrophication especially in dry years when water residence time is longer.

Our averaged riverine DIN export ratio, 0.30–0.51, is higher than those of other rivers with average values of approximately 0.2–0.25. Even the low-disturbed watersheds have very high atmospheric N input ($2200\,\text{kg}\,\text{N}\,\text{km}^{-2}\,\text{yr}^{-1}$). However, they only export 0.06–0.18 of the N input implying that the watersheds have high N use efficiency and/or retention capacity possibly due to the growing secondary forests. The moderately disturbed watersheds export 0.20–0.27 of the total input ranging from 3330–$15\,315\,\text{kg}\,\text{N}\,\text{km}^{-2}\,\text{yr}^{-1}$. The linear increase of output with total input indicates the degradation of nitrogen retention capacity and symptoms of nitrogen excess. The riverine DIN export ratios of the highly-disturbed watersheds are 0.42–0.53, which can be inferred to synergistic effects of population density and agriculture. The high N export ratios indicate that the additive N input is transported very efficiently by riverine systems and that the systems may be in advanced stages of N excess. The results also indicate that current removal processes (e.g., denitrification, vegetation uptake) cannot effectively reduce riverine N export and measures such as major improvements in sewage system are required to reduce N export.

Acknowledgements. This study was sponsored by NSC Taiwan grants (NSC 103-2116-M-002-020, 102-2923-M-002-001-MY3, 103-2621-M-002-016, 103-2621-M-003-003-) and the project ECATA (grant number I 1396) funded by FWF (the Austrian Science Fund).

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Table 1. Crop area and yield and estimated quantities of fertilizers used in Taiwan (from FAO, 2002)[a].

Crop	Harvested area (ha)	Average yield ($kg\,ha^{-1}$)	Est. quantities of fertilizers used[b] (ton)		
			N	P_2O_5	K_2O
Paddy and rice	353 065	10 646	41 793	15 314	16 462
Maize (fodder and sweet)	34 854	11 970	3485	2440	1742
Groundnut	26 495	2535	539	1192	1060
Sugar cane	30 657	81 337	6438	1073	2146
Tea	19 142	1103	8040	2680	2680
Bamboo shoot	29 819	12 030	14 910	3727	5069
Watermelon	19 720	19 685	3550	2366	3944
Banana	8961	23 718	2216	1090	4433
Pineapple	7340	47 474	2422	440	1321
Mango	18 700	11 065	449	299	673
Litchi	10 530	9385	316	211	284

[a] Only the primary crops are listed.
[b] The fertilizer amount are suggested by COA which surveys the crop plantation and declares the suggested values for farmers.

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Table 2. Watershed environmental settings of the three categories of watersheds.

category	number	N deposition ($kg\,N\,km^{-2}$)	Area (km^2)	Runoff (mm)	Avg. slope (%)	DD[a] (km^{-1})	Pop. Den. (cap km^{-2})
Low disturbed	16	2223.5 (369.3)	592.1 (486.2)	2253.3 (554.0)	66.3 (5.6)	1.35 (0.05)	11.4 (4.2)
Moderately disturbed	15	2033.3 (458.4)	1030.6 (912.6)	2419.4 (638.1)	55.3 (7.6)	1.40 (0.10)	55.2 (22.2)
Highly disturbed	18	2200.5 (351.9)	378.7 (461.9)	2268.6 (541.5)	29.1 (14.0)	1.63 (0.18)	598.2 (585.8)

[a] DD: Drainage density defined as the total stream length over drainage area, the unit is (km^{-1}).
Parentheses indicate the standard deviation of the individual category.

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Table 3. Relative land cover composition of the three categories of watersheds.

category	Forest	Built-up	Agriculture				
			total	paddy	crops	orchard	bamboo
Low disturbed	0.87 (0.06)	0.03 (0.01)	0.11 (0.08)	0.0 N/A	0.05 (0.06)	0.05 (0.03)	0.01 (0.01)
Moderately disturbed	0.81 (0.11)	0.03 (0.01)	0.16 (0.10)	0.02 (0.03)	0.05 (0.06)	0.08 (0.08)	0.02 (0.04)
Highly disturbed	0.51 (0.22)	0.11 (0.05)	0.38 (0.19)	0.06 (0.08)	0.09 (0.06)	0.23 (0.13)	0.08 (0.11)

Parentheses indicate the standard deviation of the individual category.
N/A: No paddy in the low-disturbed watersheds.

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Table 4. Mean annual NO_3, NH_4 and DIN concentrations and riverine fluxes of the three categories of watersheds between 2002 and 2012.

category	NO_3		NH_4		DIN	
	conc.	flux	conc.	flux	conc.	flux
Low disturbed	23.8 (9.9)	724.7 (242.7)	5.6 (3.2)	168.9 (86.8)	29.9 (11.0)	909.2 (285.0)
Moderately disturbed	50.2 (31.1)	1601.7 (854.2)	5.7 (2.8)	181.9 (87.9)	56.9 (31.5)	1815.8 (854.0)
Highly disturbed	99.6 (42.9)	3243.1 (1579.0)	214.2 (204.5)	4343.3 (3702.5)	329.4 (215.6)	8020.3 (4204.7)

Parentheses indicate the standard deviation of the individual category.
Unit for concentration and flux are µM and $kg\,N\,km^{-2}$, respectively.

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Table 5. Nitrogen inputs, riverine output and riverine export ratios of the three categories of watersheds.

	Low-disturbed	Moderate-disturbed	Highly-disturbed	All watersheds
Atmospheric deposition	2223.5	2033.3	2200.5	2156.8
Synthetic fertilizer	2596.0	4190.1	10 594.8	6022.3
[a]Human emission	73.43	354.1	3840.4	1543.1
Total input	4892.9	6577.5	16 635.7	9722.2
Riverine DIN output	909.2	1815.8	8020.3	3800.0
[b]Averaged export ratio	0.18	0.27	0.42	0.30
[c]Regressive export ratio	0.06	0.20	0.53	0.51

Unit for inputs and output is $kg\,N\,km^{-2}\,yr^{-1}$.
[a] Human emission is calculated from per capita emission using GDP-based estimation proposed by Van Drecht et al. (2009).
[b] Averaged export ratio is the arithmetical mean of the export ratios in the category.
[c] Regressive export ratio indicates the regressive slope of DIN output with DIN input in the category.

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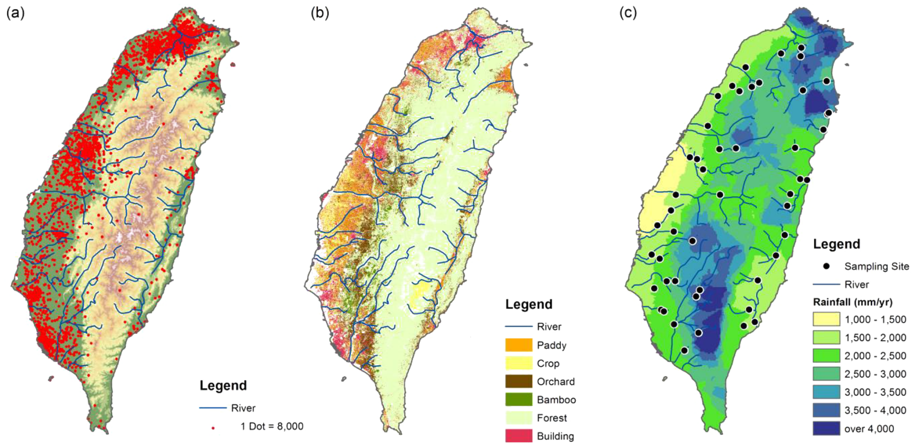

Figure 1. Three maps of Taiwan to show the population density **(a)**, land cover **(b)**, and mean annual rainfall **(c)**. The annual rainfall dataset is provided from water resources agency during 2002–2012. Population dataset is derived from county census from Ministry of Interior. Land cover dataset is distributed by Ministry of Interior in 2006.

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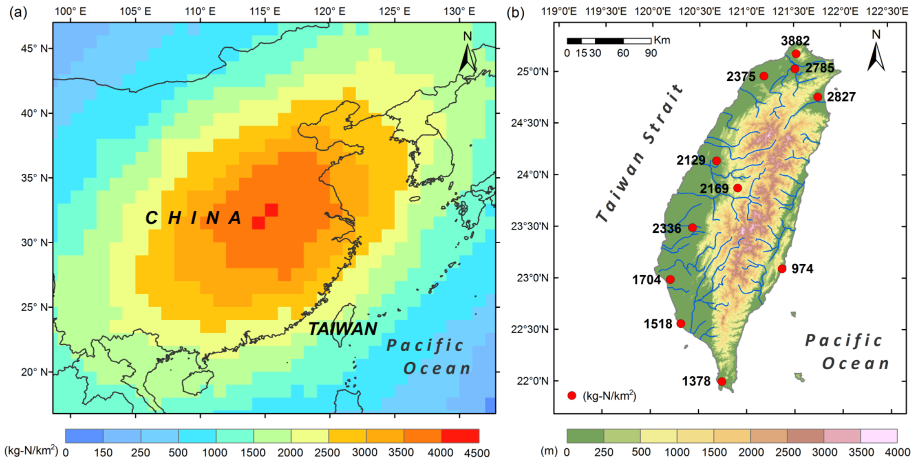

Figure 2. Atmospheric N deposition in eastern Asia (dataset: from ORNL DAAC, NASA, http://daac.ornl.gov) **(a)**. This dataset was generated using a global three-dimensional chemistry-transport model (Lelieveld and Dentener, 2000; Jeuken et al., 2001 re-sampled to 1° × 1°). The observational long-term N deposition (including NO_3 and NH_4) in Taiwan (Lin NH, unpublished data) **(b)**. The observed wet deposition is used to estimate the dry deposition by an empirical factor of 0.4 based on (Lin NH, unpublished data).

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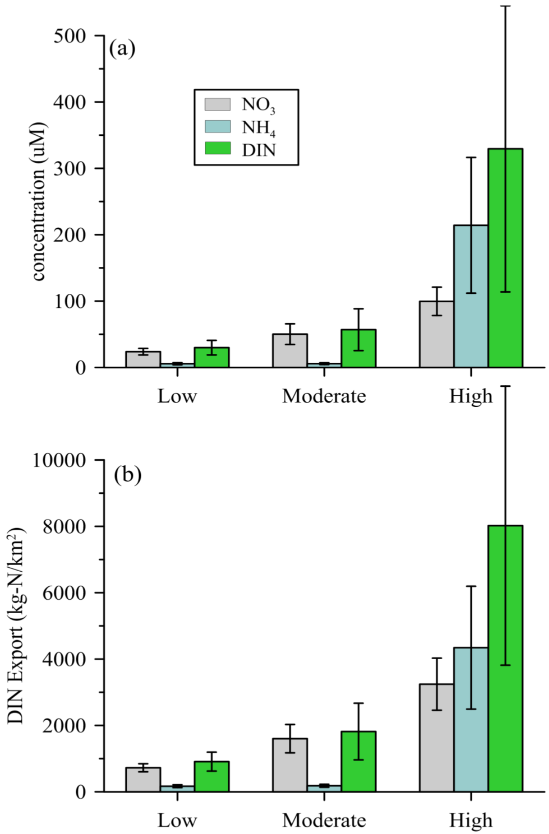

Figure 3. Mean riverine DIN concentration **(a)** and annual export **(b)** of the three categories of watershed classes. The gray, sky blue, and green bars represent the NO_3, NH_4, and DIN of the three classes. Standard deviation for each bar is given as well.

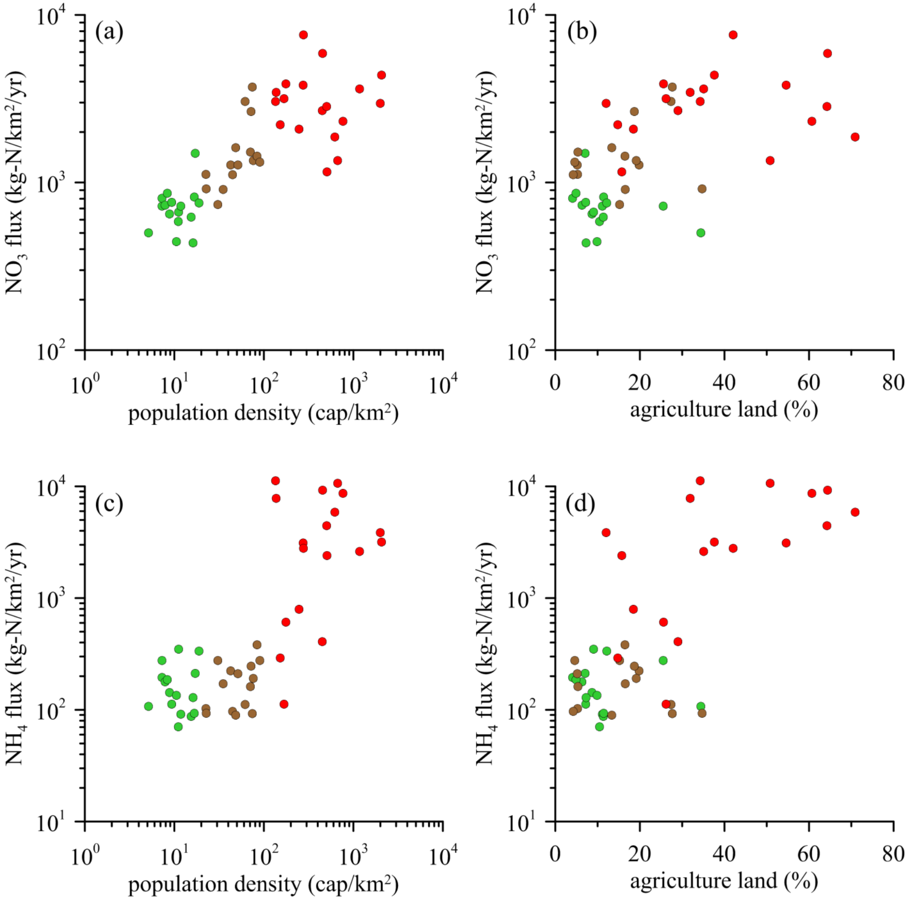

Figure 4. Scatter plot of NO_3 and NH_4 export associated with population density **(a)** and **(b)**, and relative agricultural land cover **(c)** and **(d)**. The green, brown, and red dots show the low, moderate, and highly disturbed watersheds, respectively. The *r* squared values for **(a)**, **(b)**, **(c)** and **(d)** are 0.19, 0.34, 0.18, and 0.50 with *p* values < 0.01, respectively.

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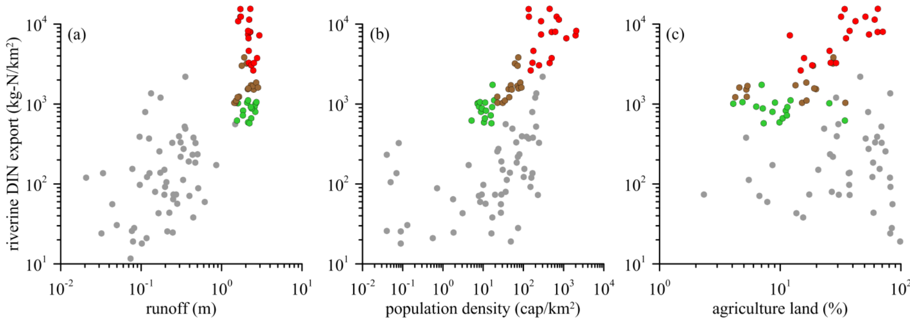

Figure 5. Scatter plot of riverine DIN export associated with runoff **(a)**, population density **(b)**, and relative agricultural land cover **(c)**. The gray dots represent the global river data retrieved from He et al. (2011). The green, brown, and red dots show the low, moderate, and highly disturbed watersheds, respectively.

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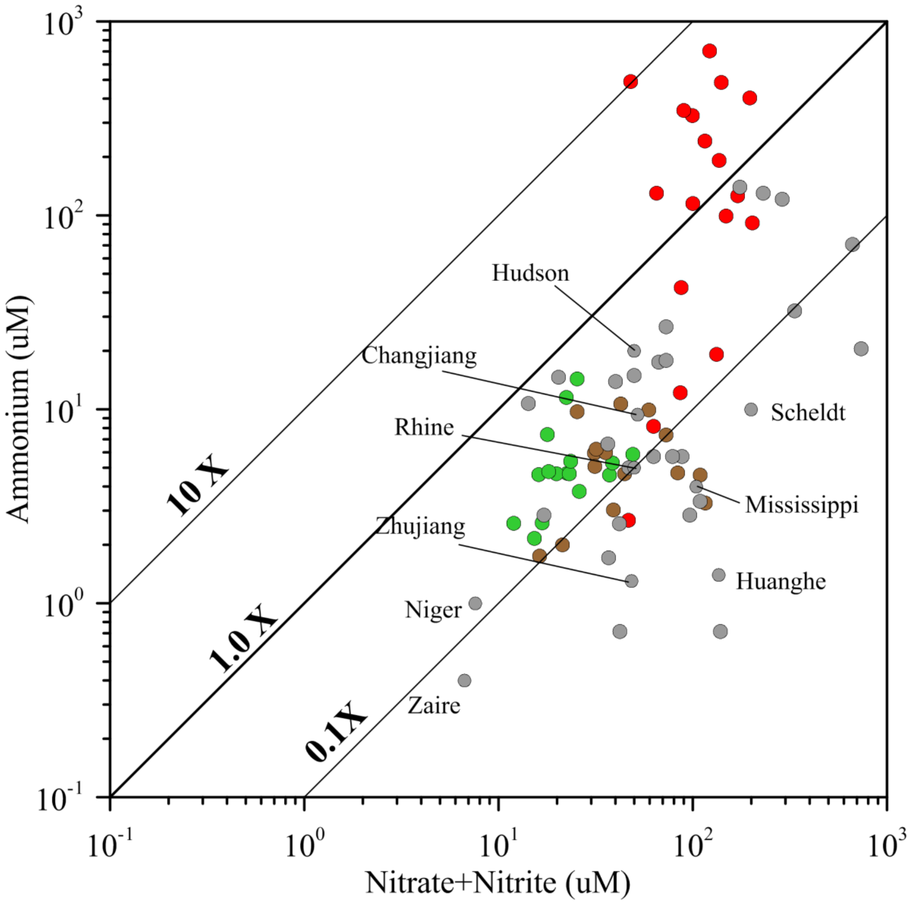

Figure 6. The DIN composition of large rivers around the world (He et al., 2011) and the 49 rivers in Taiwan. The green, brown, and red dots represent the rivers of the low-, moderate- and highly-disturbed watersheds.

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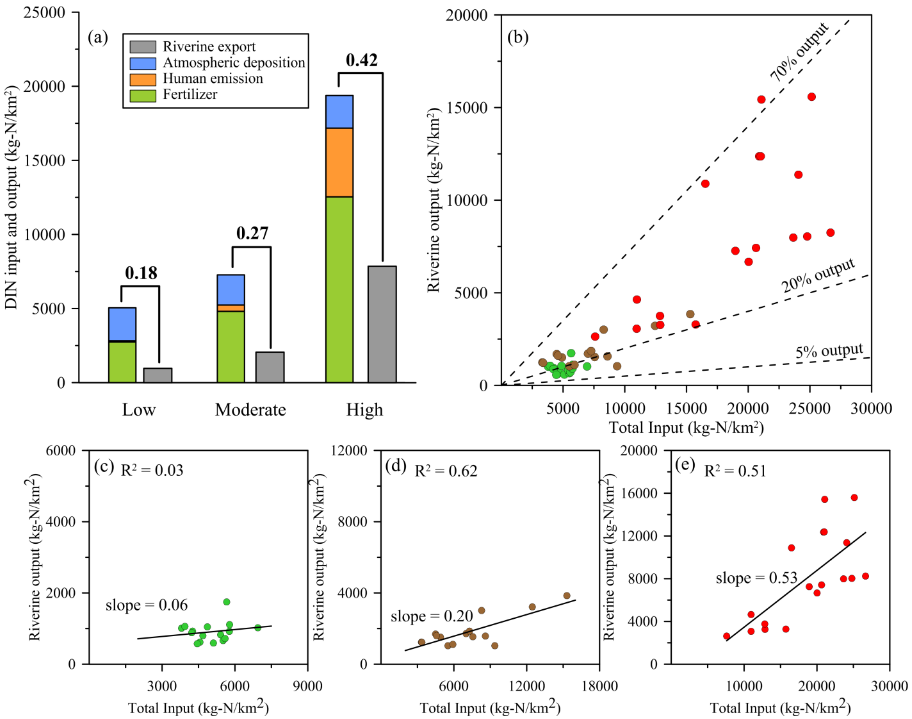

Figure 7. Nitrogen input and riverine output in the three categories of the watersheds **(a)**. Riverine DIN export against the total nitrogen input **(b)**. The regressive slope of riverine output for the low disturbed (green dots) **(c)**, moderate (brown dots) **(d)**, and highly disturbed (red dots) **(e)** watersheds.