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Effects of mountain agriculture on nutrient cycling at upstream watersheds

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Abstract

The expansion of agriculture to rugged mountains can exacerbate negative impacts of agriculture activities on ecosystem function. In this study, we monitored streamwater chemistry of four watersheds with varying proportions of agricultural lands (0.4, 3, 17, 22 %) and rainfall chemistry of two of the four watersheds at Feitsui Reservoir Watershed in northern Taiwan to examine the effects of agriculture on watershed nutrient cycling. We found that the greater the proportions of agricultural lands, the higher the ion concentrations, which is evident for fertilizer-associated ions (NO_3^-, K^+) but not for ions that are rich in soils (SO_4^{2-}, Ca^{2+}, Mg^{2+}), suggesting that agriculture enriched fertilizer-associated nutrients in streamwater. The watershed with the highest proportion of agricultural lands had higher concentrations of ions in rainfall and lower nutrient retention capacity (i.e. higher output–input ratio of ions) compared to the relatively pristine watershed, suggesting that agriculture can influence atmospheric deposition of nutrients and a system's ability to retain nutrients. Furthermore, we found that a forested watershed downstream of agricultural activities can dilute the concentrations of fertilizer-associated ions (NO_3^-, K^+) in streamwater by more than 70 %, indicating that specific landscape configurations help mitigate nutrient enrichment to aquatic systems. We estimated that agricultural lands at our study site contributed approximately 400 kg ha^{-1} yr^{-1} of NO_3-N and 260 kg ha^{-1} yr^{-1} of PO_4-P output via streamwater, an order of magnitude greater than previously reported around the globe and can only be matched by areas under intense fertilizer use. Furthermore, we re-constructed watershed nutrient fluxes to show that excessive leaching of N and P, and additional loss of N to the atmosphere via volatilization and denitrification, can occur under intense fertilizer use. In summary, this study demonstrated the pervasive impacts of agriculture activities, especially excessive fertilization, on ecosystem nutrient cycling at mountain watersheds.

1   Introduction

Agriculture expansion is taking place in some of the most rugged mountains in the world, including the Hindu-Kush Himalaya (Brown and Shrestha, 2000; Tulachan, 2001), India and China (Jodha et al., 1992), and the Andes (Sarmiento and Frolich, 2002). However, we are only beginning to understand the impacts of mountain agriculture on ecosystem services (Gao and Dong, 2003; Gordon et al., 2010). For example, it is well established that watershed nutrient cycling is tightly linked to land uses, and conversion of natural forests to agricultural lands causes nutrient enrichment, especially N and P, in streamwater (Omernik, 1976; Johnes, 1996; Tilman et al., 2001; Murty et al., 2002; Allan, 2004; Uriarte et al., 2011; Evans et al., 2014). Furthermore, such impacts are likely exacerbated by steep slopes and high precipitations, making mountain agriculture in the tropics and subtropics especially damaging to ecosystem function. Yet, empirical studies in tropical or subtropical mountain watersheds are very limited (Downing et al., 1999).

In addition to nutrient output in streamwater, cultivation and fertilization from agriculture have the potential to affect atmospheric deposition of nutrients (i.e., nutrient input via wet and dry deposition). Fine particles suspended from exposed lands and volatilized gases such as NH_3 from manure are scavenged by precipitation (van Breemen et al., 1982), which can then be deposited back to the watersheds. However, in contrast to the large number of reports on streamwater chemistry, very few studies have examined the effects of land use on precipitation chemistry.

Landscape configuration could potentially mitigate the negative effects of agriculture on watershed nutrient cycling. A study at Hubbard Brook Experimental Forest demonstrated that watershed-level responses were most sensitive to spatial scale at which much of the variation in element fluxes occurred in the first 10–20 ha surrounding the drainage area (Johnson et al., 2000). Such understanding has led to the common practice of establishing riparian buffer zone as a way to remove pollutants and prevent nutrients from entering streamwater (Muscutt et al., 1993). Without scarifying socioeco-

nomic benefits of agriculture, landscape configuration of mountain watersheds can be more carefully planned to mitigate negative impacts of agriculture on nutrient cycling.

Here we examined the effects of mountain agriculture on watershed nutrient cycling at Feitsui Reservoir Watershed (FRW) in subtropical Taiwan. We first compared streamwater chemistry across four watersheds within FRW, two with substantial agricultural land use and two primarily covered with natural forests. To illustrate the additional effects of agriculture on atmospheric deposition of nutrients and its role in watershed nutrient retention, we focused on the pair of watersheds with the highest and lowest agricultural covers, and compared their rainfall chemistry in relation to streamwater chemistry and watershed nutrient retention. The FRW is characterized with high rainfall (> 3000 mm; Taipei Feitsui Reservoir Administration), steep slopes (on average 42 %) and heavy use of fertilizers typical for tea plantations (T. C. Lin, personal communication with WenShan Branch, Tea Research and Extension Station, 2015). Many studies have demonstrated substantial nutrient efflux and sediment production from surrounding tea plantations to the reservoir over the last two decades (Chang and Wen, 1997; Lu et al., 1999; Kuo and Lee, 2004; Li and Yeh, 2004; Hsieh and Yang, 2006, 2007; Zehetner et al., 2008; Chiueh et al., 2011; Wu and Kuo, 2012), making FRW an ideal site for examining agriculture effects on watershed nutrient cycling.

We hypothesize that agriculture will increase nutrient output in streamwater (H_1) as well as atmospheric input of nutrients through rainfall (H_2), and ultimately, it will have a negative effect on nutrient retention (H_3). Our specific predictions are:

1. Watersheds with substantial agricultural lands have higher concentrations and fluxes of fertilizer-associated ions in the streamwater than forested watersheds (H_1).

2. Watersheds with substantial agricultural lands have higher concentrations and fluxes of fertilizer-associated ions in the rainfall than forested watersheds (H_2).

3. Watersheds with substantial agricultural lands have lower nutrient retention than forested watersheds (H_3).

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In addition, we explored: (1) the role of landscape configuration in mitigating agriculture effects by quantifying the dilution effects of a downstream forested watershed on the upstream watersheds with substantial agricultural activities; and (2) the N and P dynamics associated with agriculture by quantifying the differences in their fluxes between a forested watershed (background values) and a nearby watershed with substantial agriculture activities.

2 Materials and methods

2.1 Study site

The Feitsui Reservoir Watershed (FRW) is located in Peishi Creek of northern Taiwan, with a drainage area of 303 km^2. The elevation of FRW ranges from 45 to 1127 m, with a mean slope of 42 % (Fig. 1). The underlying geology of the FRW region is mainly argillite and slate with sandstone interbeds, and the soils are mostly Entisols and Inceptisols with high silt contents (Zehetner et al., 2008).

Annual precipitation is high and has high spatial variability ranging from 3500 mm in the southwest portion of FRW to 5100 mm in the northwest during 2001–2010 (J. C. Huang, unpublished data). The vegetation is primarily composed of secondary-growth, mixed broad-leaf forests dominated by Fagaceae and Lauraceae (Chen, 1993). Approximately 16 % of the FRW is agricultural lands with tea plantations covering an area of 1200 ha, or 25 % of all agricultural lands (Chang and Wen, 1997; Chou et al., 2007). Due to the high market value of tea, fertilizers and pesticides are heavily applied to tea plantations. In 1986 the FRW was designated as a water resource protection area, followed by the construction of Feitsui Reservoir in 1987. Today, the reservoir provides drinking water to the six million people in Taipei Metropolitan. The forests in the FRW have been protected (no cutting, thinning or converting to agricultural use) since 1986. Therefore, current agriculture activities are limited to private lands with a pre-existing agriculture use.

2.2   Sampling regime

Four watersheds of FRW (A1, A2, F1, F2; Fig. 1) with varying proportions of agricultural cover (22 % in A1, 17 % in A2, 2.9 % in F1, 0.4 % in F2; Table 1) were included in this study. Weekly samples of streamwater were collected from all four watersheds.
5   Natural forest remains the most dominant land cover type for all four watersheds (68 % in A1, 76 % in A2, 93 % in F1, 99 % in F2; Table 1), making agricultural activities the primary contributor to the differences in landscape across the four watersheds. In addition, weekly samples of rainwater were collected from the two watersheds with the lowest (F2) and highest proportions of agricultural lands (A1). A1, A2 and F2 are small
10   watersheds (< 3 ha) drained by first order streams whereas F1 is a much larger watershed (86 ha) drained by a second order stream that drains through A1 and A2 (Fig. 1). We collected weekly rainfall and streamwater samples every Tuesday from September 2011 to August 2013. Rainfall samples were collected using a 20 cm diameter polyethylene (PE) bucket, from which a 600 mL subsample was taken and placed into
15   a PE bottle for transportation back to the laboratory. Streamwater samples were collected by diving a PE bucket into the stream, and similar to rainfall sampling, a 600 mL subsample was also taken and placed into a PE bottle for transportation back to the laboratory.

2.3   Water chemistry

20   All samples were transported back to the laboratory within 24 h. Conductivity and pH of the water samples were always measured on the same day of collection. The samples were filtered with 0.45-μm filter paper. Major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}, NH_4^+) and anions (Cl^-, SO_4^{2-}, NO_3^-) were analyzed by ion chromatography on filtered samples using Dionex ICS 1000 and DX 120 (Thermo Fisher Scientific Inc. Sunnyvale, CA,
25   USA). PO_4^{3-} was measured using standard vitamin C-molybdenum blue method with the detection limit of 0.01 μM (APHA, 2005). Prior to chemical analysis samples were stored at 4 °C without any preserves.

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Data on rainfall and streamflow quantity of the watersheds were estimated from the nearest rain gauges and discharge gauges that are maintained by the Central Weather Bureau and Water Resource Agency of Taiwan, respectively. Three rain gauges and two discharge gauges were used. The distance between a watershed and its nearest rain gauges was 1.0–8.5 km, and that between a watershed and its nearest discharge gauges was 3.0–5.0 km. The rainfall of a watershed was directly assigned to the values registered at the nearest rain gauges. The streamflow of a watershed was estimated by area ratio method.

2.4 Statistical analysis

We used the general linear model with repeated measurements to compare monthly concentration and flux of ions in streamwater among the four watersheds (F1, F2, A1, A2), followed by LSD post-hoc comparisons. NH_4^+ was excluded from streamwater analysis due to its low concentration. We used one-tail paired t test to examine if monthly ion concentration (volume weighted from weekly samples) and flux in rainfall were higher at the watershed with higher agricultural land cover (A1) than the more pristine watershed (F2). One-tail paired t test was also used to compare the ratio of monthly nutrient output via streamflow and input via rainfall between A1 and F2 with a priori assumption of higher nutrient retention capacity (lower ratio) of F2 than A1. All statistical analysis was conducted using SPSS 22 (IBM Corporation, New York).

3 Results

3.1 Streamwater chemistry

The concentrations of all analyzed ions in streamwater differed significantly among the four watersheds (Fig. 2a). A1, the watershed with the highest proportion covered by agricultural lands, had significantly higher concentrations of all ions except H^+ and PO_4^{3-} than the other thee watersheds (Fig. 2a). In contrast, F2, the watershed with the

lowest proportion covered by agricultural lands and steepest slopes, had the lowest concentrations of H^+, Na^+, K^+, Cl^- and NO_3^- yet the second highest concentrations for ions rich in soils and soil solution including Ca^{2+}, Mg^{2+}, and SO_4^{2-} (Fig. 3a).

Similar to ion concentration, the fluxes of all ions differed significantly among watersheds (Fig. 2b). A1 had the largest fluxes of K^+, Ca^{2+}, Mg^{2+}, NO_3^- and SO_4^{2-} among the four watersheds and F2 had the smallest fluxes of H^+, Na^+, K^+, Mg^{2+}, Cl^-, and NO_3^- among the four watersheds. PO_4^{3-} flux was significantly larger at A1 and A2, which were not different from each other, than F1 and F2, which were also not different from each other (Fig. 2b). Although the fluxes of Na^+ and Cl^- differed significantly among A1, A2 and F1, these differences were considerably smaller than the difference between the three watersheds and F2 (Fig. 2b).

3.2 Rainfall chemistry

Five of the 10 measured ions had significant or marginally significant higher concentrations in A1 than F2 (H^+, Na^+, Cl^-, NO_3^-, $p < 0.05$; NH_4^+, $p = 0.067$; Fig. 3a). Furthermore, seven of the 10 measured ions had significant or marginally significant higher fluxes in A1 than F2 (H^+, Ca^{2+}, Cl^-, $p < 0.05$; Na^+, Mg^{2+}, NH_4^+, NO_3^-, $p < 0.1$; Fig. 3b).

3.3 Output–input ratio

Annual output–input ratio for watersheds A1 and F2, an indication of ecosystem retention or loss of ions, was larger than unity for all analyzed ions except H^+ and NO_3^-, indicating prevalent element loss in both watersheds. Monthly output–input ratios for all analyzed ions were consistently lower at F2 than A1 ($p < 0.05$). The differences in output–input ratio between A1 and F2 were larger for H^+ and the three ions that are important ingredients of fertilizers (NO_3^-, K^+, and PO_4^{3-}) than other ions on both monthly (not shown) and annual bases (Fig. 4). Notably, annual output–input ratio of NO_3^- in F2 not only was lower than A1, it was in fact below unity, indicating no element loss.

4 Discussion

4.1 Streamwater chemistry

The watershed with the highest proportion of agricultural lands (A1) had the highest concentrations and fluxes of most ions in streamwater, suggesting the role of agriculture on increasing nutrient output. Furthermore, the fact that output of fertilizer-associated ions (NO_3^- and K^+) perfectly matched to the proportion of agricultural lands across the four watersheds (i.e. rank of proportion of agricultural lands from high to low: A1, A2, F1, F2; rank of ion concentration and flux from high to low: A1, A2, F1, F2) strongly indicates the effects of agriculture on streamwater chemistry and supports our H_1.

However, streamwater chemistry is driven by complex processes beyond a single factor of land use. For example, P is also an important component of fertilizers, but unlike NO_3^- and K^+, concentration of PO_4^{3-} at F2 was not significantly different from A1 and A2, and all were significantly higher than F1. Greater erosion and leaching associated with the steeper slopes of F2 may have matched the effect of fertilization, and led F2 to have a PO_4^{3-} concentration as high as A1 and A2. To further illustrate this topographic effect, we compared streamwater chemistry between the two forested watersheds (F1 and F2), removing the potential confounding effect of land use. The steeper F2 (48 %), indeed had a higher PO_4^{3-} concentration than less steep F1 (39 %), despite that F2 has a higher natural forest cover than F1. Soil erosion is arguably the greatest concern to most P mitigation programs because concentration of P on surfaces of soil particles is often orders of magnitude greater than in soil solution (Sharpley et al., 2002; Kleinman et al., 2011). Therefore, it is not surprising that topology may be a more important driver for riverine P than land use at our study site. The enhanced erosion/leaching associated with steeper slope at F2 may also explain why F2 had the second highest concentration of SO_4^{2-}, Ca^{2+}, and Mg^{2+}, ions that are abundant in soils. It is worth noting that due to the lower rainfall at F2 from topographic shading, element fluxes are

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lowered, which likely led to the similar fluxes of PO_4^{3-}, SO_4^{2-} and Ca^{2+} between the two forested watersheds, and even the lowered Mg^{2+} flux at F2 compared to F1.

4.2 Rainfall chemistry

We confirmed that agriculture activities can influence watershed nutrient cycling via atmospheric deposition in our study site (H_2). We found higher concentrations and fluxes of NO_3^- and NH_4^+ in rainfall at A1, a watershed with 22 % of agricultural lands, compared to F2, the watershed almost entirely covered by natural forests. Ammonium sulfate, urea and calcium ammonium nitrate [$5Ca(NO_3)_2 \cdot NH_4NO_3 \cdot 10H_2O$] that contain high quantity of NO_3^- and NH_4^+ are commonly used N-fertilizers in Taiwan (Huang, 1994). Therefore, in tea plantations at FRW, substantial suspension and volatilization of ammonium sulfate, urea and calcium ammonium nitrate have likely contributed to the high concentrations and fluxes of NO_3^- and NH_4^+ in rainfall at A1. The lack of significantly higher concentrations of PO_4^{3-} and K^+ at A1 than F2 in rainfall possibly reflects the low mobility of PO_4^{3-} and smaller quantity of P in fertilizers compared to N.

Once in the atmosphere, aerosols/chemicals can be transported to other locations but most will deposit on surrounding ecosystems. In central Taiwan, high NH_4^+ concentration in precipitation at a high elevation forest (2000 m) was attributed to mountain agriculture 10 km away (Ding et al., 2011). With the predicted expansion of agriculture to the mountains both in Taiwan and many other regions (Jodha et al., 1992; Brown and Shrestha, 2000; Tulachan, 2001), even pristine ecosystems are not free from the impacts (e.g. acidification and eutrophication associated with H^+ and NO_3^-) of agriculture activities.

Because Taiwan is a small island, sea salt aerosols are important components of rainfall (Lin et al., 2000). The distance to the coast, specifically, has been used to explain variation of Na^+ and Cl^- concentrations in precipitation among four sites in central Taiwan (Ding et al., 2011). The higher concentration and flux of Na^+ and Cl^-, and to a lesser degree Mg^{2+}, at A1 than F2 likely reflects such oceanic influences. The

watersheds receive winter rains, along with sea salt aerosols, from the north/northeast coasts (Northeast Monsoon). While A1 is located on the windward side, F2 is on the leeward side. Therefore, a substantial proportion of the sea salt aerosols may have been intercepted before they can reach F2. Although summer rains move from the opposite direction, the watersheds are relatively far from the west/southwest coasts (> 60 km), making summer rains less important to input of sea salt aerosols to the watersheds.

In contrast to Na^+ and Cl^-, the differences in topographic position and distance to the ocean between A1 and F2 seemed to have limited effect on SO_4^{2-} deposition. Many studies reported significant contribution of long-range transport of S and N from eastern China to Taiwan via Northeast Monsoon (Lin et al., 2005; Junker et al., 2009). Because A1 is on the windward side of Northeast Monsoon it may experience higher input of pollutants from long range transport than F2, which is on the leeward side. The lack of significant differences in SO_4^{2-} between the two watersheds suggest that the two watersheds are too close to show differential influences of pollutants that are transported from sources several hundred kilometers away.

4.3 Output–input ratio

The larger output–input ratio of all ions at A1 than F2 confirmed our prediction that agriculture activities lower ecosystem nutrient retention capacity (H_3). The multiple canopy layer, rich epiphytes, and lush ground vegetation in natural forests all contribute to the absorption of nutrients and reduce the velocity of rainfall, thereby reduce the leaching of nutrients (Jordan, 1980; Nadkarni, 1984; Chandrashekara and Ramakrishnan, 1994). Due to the high rainfall and very rough topography in many tropical and subtropical mountain watersheds the potential of nutrient deficiency caused by leaching is high. Thus, maintaining high nutrient retention capacity by natural vegetation cover is critical for maintaining site fertility and should be of high management priority.

4.4 Landscape configuration and streamwater chemistry

The large differences in NO_3^- concentration and flux between F1 and A1, A2 highlights the role of landscape configuration on streamwater chemistry. Both A1 and A2 are sub-watersheds of F1; however, the influence of agricultural lands on A1 and A2 largely dissipated as water enters into forested F1. Specifically, the concentrations of NO_3^- and K^+ were 70 % lower at F1 than at A1 and A2. Comparing to the difference in concentration and flux of NO_3^- between F1 and F2 (< 30 %), that between F1 and A1, A2 is striking (> 300 %; Fig. 3). Thus, by constraining agriculture activities away from the main stream and maintaining natural cover of its watershed, the impact of agriculture on nutrient enrichment could be reduced. Our result confirmed the importance of landscape configuration on streamwater chemistry (Dillon and Molot, 1997; Johnson et al., 1997; Palmer et al., 2004).

4.5 N and P output from agriculture

Given the proximity in location and similarity in topology between A1 and F1, their differences in nutrient output should largely reflect the effects of land conversion from natural forest (F1) to agricultural lands (A1). By subtracting nutrient output at F1 from A1, we estimated N and P output from agriculture at A1 to be approximately 90 and 47 $kg\,ha^{-1}\,yr^{-1}$, respectively. Factoring in the 22 % of agricultural lands at A1, the contribution from agricultural lands would have been 400 and 260 $kg\,ha^{-1}\,yr^{-1}$ for N and P, respectively, should A1 has 100 % agricultural lands.

The per-hectare output of N and P reported here is extraordinary high compared to those reported for many agriculture watersheds around the globe. For example, a study from Baltimore Ecosystem Study reported an annual output of NO_3-N at 13–20 $kg\,ha^{-1}\,yr^{-1}$ for a 7.8 ha watershed that is completely covered by agricultural lands and has gentle slopes (Groffman et al., 2004). In four watersheds with 30–40 % covered by row crops, and fertilizer application at 50–70 $kg\,ha^{-1}\,yr^{-1}$ N and 7–16 $kg\,ha^{-1}\,yr^{-1}$ P in southeastern coastal plain of the United States, nutrient output through stream-

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flow was $< 6 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$ for N and $< 3 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$ for P (Lowrance et al., 1985). In the Great Barrier Reef, Australia, total output via streamflow was approximately $5 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$ for NO_3-N and $0.3 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$ for dissolved P from a watershed with 29 % of the land covered by pasture and 14 % by crop lands (Hunter and Walton, 2008).

High N and P output from agricultural lands is probably common in Taiwan and other regions under intensive agriculture (Huang et al., 2012). In the Dansheui River of northeastern Taiwan, output of dissolved inorganic N ranged from 3 to $100 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$ from relatively pristine headwaters covered mostly by natural forests to populous estuary (Lee et al., 2014; Shih et al., 2015). In humid southeastern China, N output from a watershed with 17.5 % agricultural lands, steep slopes (the watershed has a mean slope of 21 % of which the site is located in the particularly hilly upstream region), and heavy application of N fertilizers (300–$1000 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$), reached $73 \, \mathrm{kg\,ha^{-1}\,yr^{-1}}$ (Chen et al., 2008), approximately the same magnitude as those reported here. Although the small headwater streams remove nitrogen efficiently (Peterson et al., 2001), Our study clearly demonstrated that high application of fertilizers in regions with high rainfall and very steep slopes could lead to extremely high output of N and P and, therefore, eutrophication risk of downstream watersheds.

4.6 Watershed nutrient fluxes

In order to provide a more comprehensive understanding on how mountain agriculture affects watershed nutrient cycling, we re-constructed nutrient fluxes for A1 and F1 (Fig. 5). We used F1 as the background watershed with minimum agriculture activities to contrast it against A1 for reasons previously stated (e.g., close proximity, similar topology). We made four assumptions in the calculation of watershed nutrient fluxes. First, we assumed similar atmospheric deposition between F1 and F2 given that they are both forested, less than 10 km apart, and share a similar precipitation amount (F1: 7600 mm, F2: 7140 mm during the two sampling years; Central Weather Bureau of

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Taiwan). Although this may underestimate atmospheric deposition at F1 given that it is more affected by agriculture activities (from A1 and A2) than F2 the resulting bias should be minimum due to the proximity of F1 and F2. Second, we assumed similar proportional contribution of dry deposition relative to precipitation between the two watersheds and Fushan Experimental Forest (approximately 28 % using Na^+ ratio method, Lin et al., 2000), a natural hardwood forest 17 km south of FRW. Third, we assumed the amount of fertilizer used is similar to that reported in a case study done in the same region as the current study (786 kg N ha^{-1} yr^{-1} and 75 kg P ha^{-1} yr^{-1}) (Tsai and Tsai, 2008). There was very little change in biomass of tea plantations after 10 yearsbecause tea plants are regularly trimmed with the litter left in the field to maintain the same height optimal for harvest. Thus, our fourth assumption is that N and P lost due to uptake by tea trees is equivalent to N and P in harvested tea leaves. The amount of N removed through tea harvest (113 kg ha^{-1} yr^{-1}) was taken from the same case study (Tsai and Tsai, 2008) and the amount of P (1.6 kg ha^{-1} yr^{-1}) removed was calculated using the medium of N : P ratios (15.4) reported for tea trees in Taiwan (Tsai and Tsai, 2008). Using simple mass balance (i.e., input from atmospheric deposition and fertilizer equals output via streamwater, harvest and volatilization/suspension), we then estimated the amount of N fertilizers that was volatilized/suspended from A1 and the retention rates of N and P in A1 and F1.

If only output through streamwater and input through atmospheric deposition are considered, the retention ratio (1 – output/input) of N would be approximately 47 % for A1 and 68 % for F1. Based on a recent synthesis (Sullivan et al., 2014) biological N fixation (BNF) in tropical forests is not as high as previously reported and, on average, is slightly less than 10 kg ha^{-1} yr^{-1} for secondary forests. Thus, adding BNF into N input could increase N retention ratio in F1. The high retention ratio of F1 suggests that the secondary natural forest is probably still growing. In contrast, because N fertilizers were applied at rates that are one order of magnitude greater than BNF and high N fertilization is known to negatively affect BNF (Sanginga et al., 1989; Fuentes-Ramírez et al., 1999), adding BNF to nutrient input has little effects on N retention at A1.

The N retention rates of A1 and F1, however, do not take the loss through volatilization and denitrification into account. Because it rains very frequently at FRW, soil moisture is likely high throughout the year, and consequently, N loss through denitrification could be substantial. In addition, because fertilizers are applied in solid form so that volatilization of NH_3 could also be high. Thus, if both denitrification and volatilization are taken into calculation the N retention rate at A1 would be even lower. The return of N back to the atmosphere through denitrification and volatilization helps explain the higher atmospheric N deposition at A1 than F2. The low retention rate and the resulting high leaching loss of N at A1, which is approximately 20 times the levels of F1, impose a major threat to streamwater and reservoir eutrophication.

Surprisingly, from our construction of N fluxes, the loss of N through annual harvest at A1 approximately equals the annual atmospheric deposition ($26\,\mathrm{kg\,ha^{-1}\,yr^{-1}}$), of which only a small portion should come from fertilizer (atmospheric N deposition at F2 is only 8 kg lower than A1, suggesting $< 8\,\mathrm{kg}$ of atmospheric N deposition could potentially come from fertilizer). In other words, to maintain the current harvest, not much N fertilization is actually required and most of the currently excessive fertilization is lost either through hydrological process (i.e. leaching) to streams and the Feitsui Reservoir or returned to the atmosphere, both of which could have negative environmental impacts.

Although both N and P are important components of fertilizers their fates within an ecosystem are very different. Due to the lack of gaseous form, P can only be lost to the atmosphere through suspension of fine particles. Therefore, leaching through streamwater becomes the main pathway of P loss from an ecosystem. Surprisingly the output of P through streamwater is more than the total input at A1. Agriculture activities often disrupt soil stability and increase soil erosion. Excessive P leaching caused by agriculture activities at A1 likely contributed to the greater P output than input. Such process could gradually degrade ecosystem fertility, particularly for lowland Taiwan where soils are likely P limited (Lee et al., 2013). Although P fertilizers are applied at a much lower rate than N, on an input to demand (amount harvested through tea

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leaves) ratio, they are even more excessively applied compared to N (input to demand ratios of P and N are 23 and 7 respectively). Our re-construction of N and P fluxes clearly showed that both N and P are applied at rates that are neither ecologically nor economically sound, and such excessive fertilization may cause fundamental changes in watershed nutrient cycling (Fig. 5).

5 Conclusions

1. Mountain watersheds with moderate proportion of agricultural lands (17–22 % of agricultural lands) had much higher concentrations of most ions in both streamwater (nutrient output) and rainwater (nutrient input) than forested watersheds. Furthermore, they also showed lower nutrient retention capacity than forested watersheds. The result indicates that agriculture activities have a more pervasive impact on watershed nutrient cycling than previously recognized and that mountain watersheds are vulnerable to agriculture expansion.

2. Topographic control is also important in nutrient output of mountain watersheds, particularly for ions that are rich in soils, such as SO_4^{2-}, Ca^{2+} and Mg^{2+}.

3. Proper spatial configuration of agricultural lands in mountain watersheds can mitigate the impact of agriculture on NO_3^- output by 70 %, thus reducing the risk of eutrophication for streams and lakes.

4. We produced conservative estimates of the contribution of agricultural lands to N and P output in streamwater for one of our watersheds (i.e. A1) at approximately 400 and 260 $kg\,ha^{-1}\,yr^{-1}$ respectively. These estimates exceeded previous reports around the globe, and can only be matched in magnitude by one study in China where fertilizers were excessively applied.

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5. The re-constructed element fluxes for the watersheds indicate excessive leaching of N and P, and additional loss of N to the atmosphere via volatilization and denitrification, which likely resulted from excessive fertilizer use.

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Table 1. Basic information of the studied watersheds.

	A1	A2	F1	F2
Area (ha)	2.92	1.36	86.04	0.67
Slop (%)	39.3	34.8	38.7	48.1
Aspect land use (%)				
Natural forest	68.0	75.5	93.5	99.2
Agriculture	22.1	17.1	2.87	0.38
Road	3.61	2.96	0.77	0.00
Building	1.54	1.31	0.35	0.00
Water body	0.69	0.19	1.12	0.00
Others	4.11	2.96	1.44	0.38
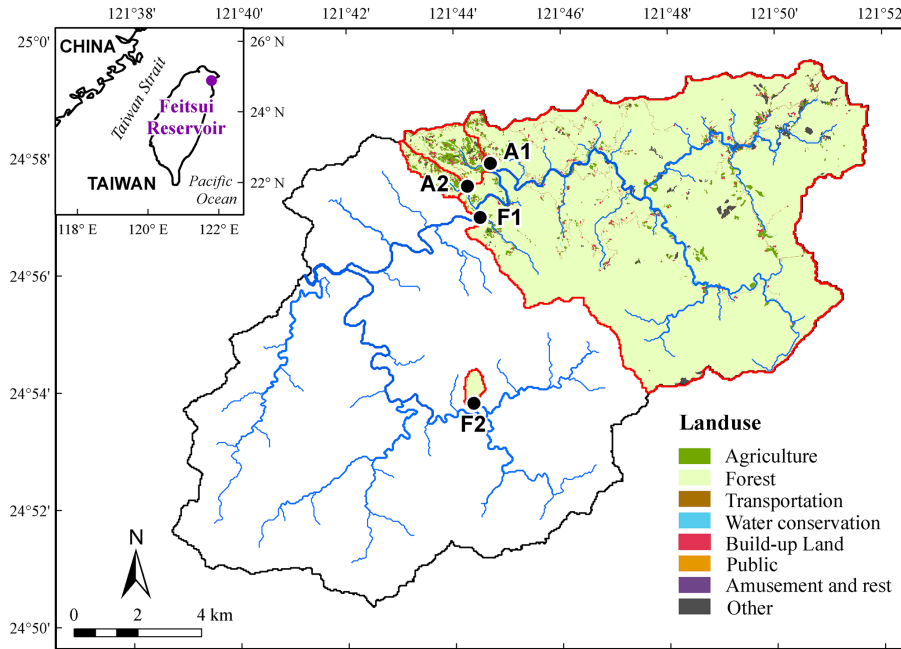
Figure 1. Location and land use distribution of the studied watersheds.
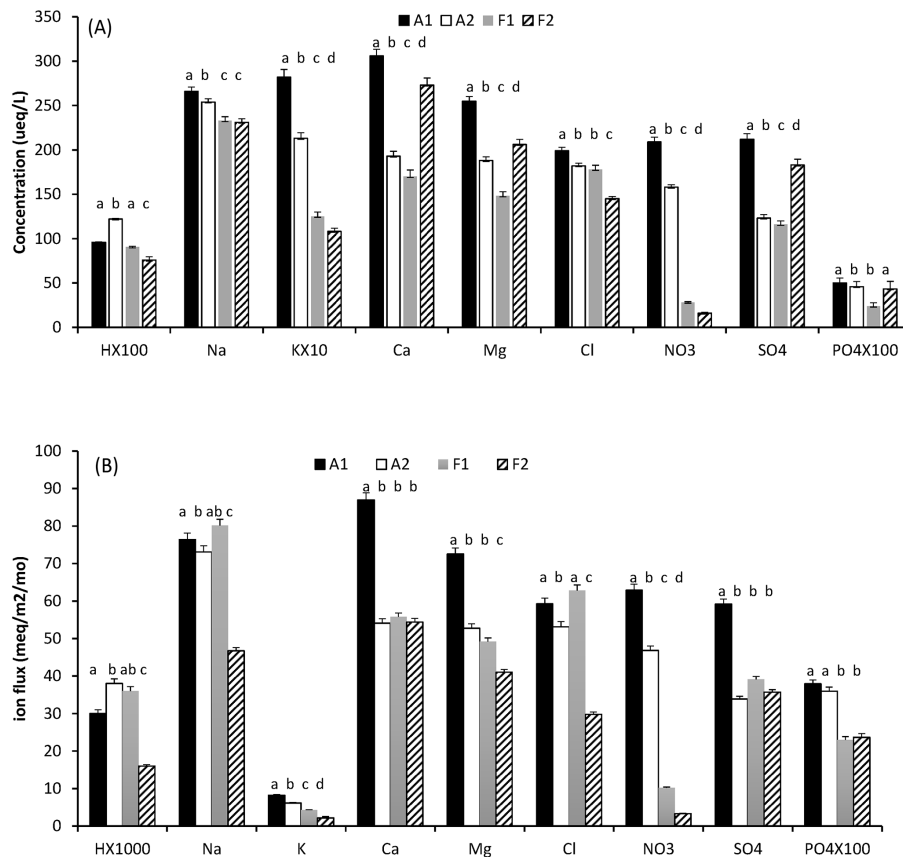
Figure 2. Mean monthly ion concentration (volume-weighted from weekly samples) **(a)** and flux **(b)** of streamwater of watersheds A1, A2, F1, and F2. Two ions sharing no common letter are significantly different ($p < 0.05$). Error bar: 1 standard error.
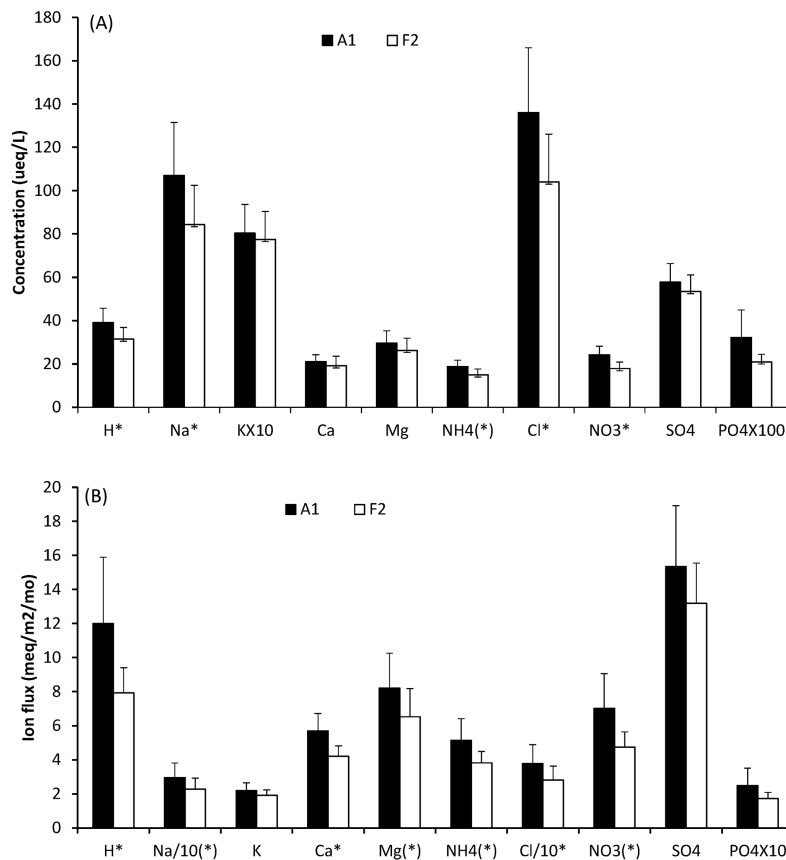
Figure 3. Mean monthly ion concentration (volume-weighted from weekly samples) **(a)** and flux **(b)** of rainfall of watersheds A1 and F2. ∗: significant difference between the two watershed ($p < 0.05$); (∗): marginally significant difference between the two watersheds ($0.05 < p < 0.10$). Error bar: 1 standard error.
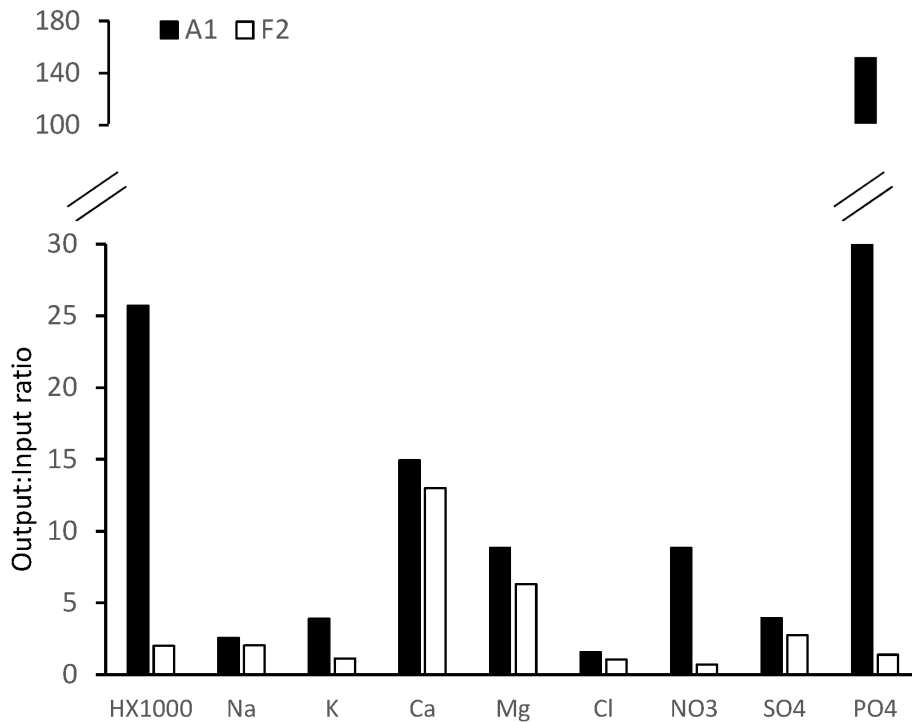
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Figure 4. Annual output–input ratios of ions of watersheds A1 and F2.

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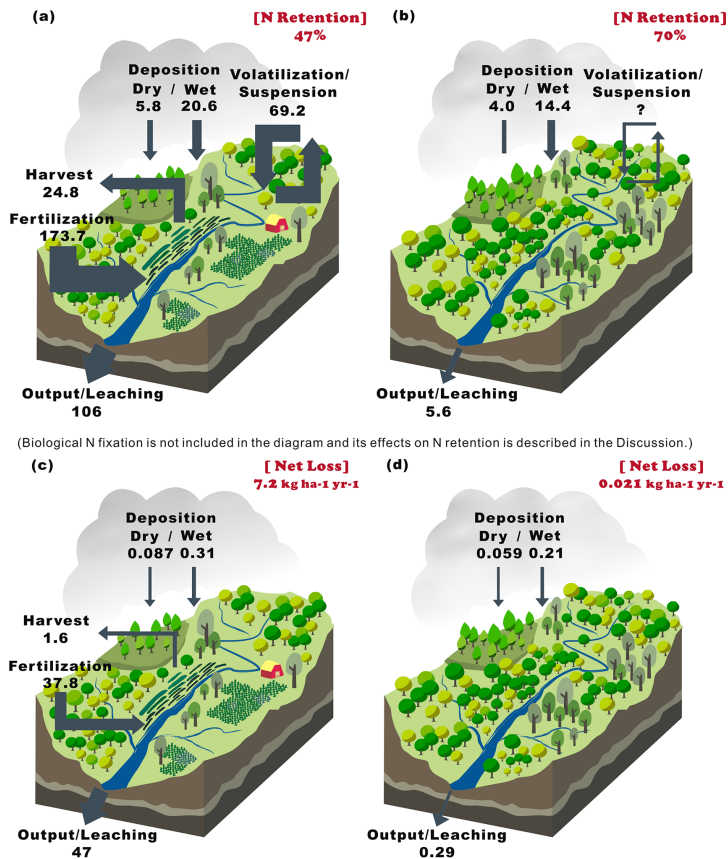
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Figure 5. Schematic diagram of N and P fluxes of watersheds A1 and F1. A1 represents a watershed with 22 % agricultural lands and 68 % forests **(a, c)**; F1 represents a watershed with 3 % agricultural lands and 94 % forests **(b, d)**. Biological N fixation is not included in the diagram and its effects on N retention is described in the Discussion.

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