Anonymous Referee #1

Received and published: 3 June 2015

Overall comments, Although the impact of agriculture on river water quality has been well studied around world, the case study in a subtropical tea plantation is still limited at this point. I recognize, therefore, this manuscript provided valuable information on N input-output for mitigation of anthropogenic N loss to river system from the watershed covered by tea plantation. However, I found some inadequate discussions and structures to be revised as mentioned below.

Major comments

Comment 1. Impact of agriculture on rainfall chemistry. Although rain water chemistry indicated some significant or marginal differences of concentrations and fluxes between A1 and F2, it would be still doubtful that this differences are certainly caused by fertilizer from the surrounding agricultural area. When the spatial variability of rainfall chemistry in a landscape scale is taken accounted, the simple analysis of the difference in Figure 3 is not enough to confirm your discussion. Regarding with the ion flux in rainwater, you should also discuss the difference of rainwater amount between two sites (A1>F2, described in Line 27 of Page 4793). I would sat that you need more site replication of rainfall observation and further evidences to discuss the impact of agriculture on rain water chemistry onsite. I recommend deleting the all discussion on the impact of agriculture on rainwater from the manuscripts.

We agree that additional site replication on rainfall data would strengthen our discussion on the effect of agriculture on rainfall chemistry. However, we found that concentrations of all ions analyzed were higher at the watershed with high tea plantation than the one that was almost completely forested. Although the significant differences in Na and Cl could be related to spatial variation of oceanic influences, the significantly higher concentrations of N and K provide good indication of agriculture influences. The effect of N from agriculture activities (fertilization) on atmospheric deposition has been reported (e.g., van Breemen et al., 1982). To avoid over-statement from only one-pair of watersheds, we modified the language in the text to be more conservative. Despite the somewhat limited spatial replication, this manuscript provides one of the first data sets pointing to the potential effects of agriculture on rainfall chemistry from the region, amongst the rich literature on how agriculture affects watershed nutrient cycling through streamwater chemistry.

van Breemen N., Burrough P. A., Velthorst E. J., van Dobben H. F., de Wit T., Ridder T. B., and Reijnders H. F. R.: Soil acidification from atmospheric ammonium sulfate in

forest canopy throughfall, Nature, 299, 548-550.

Comment 2. Data presentation and method description The budget analysis is very important for your discussion (Figure 5). The text of the methods and assumption for Figure 5 (from page 4797 line 24 to page 4798 line 19) should be described in the methods section, “2 Material and methods”. The information of fertilizer application (page 4798 line 8) should be described in the “2.1 Study site”. Furthermore, data explanation of the input and output budget in Figure 5 should be described in the result section, “3 Results”.
We moved the assumptions and other details about the calculations to “Materials and Methods”. The information of fertilizer application was added to the “Study site” with more details provided in the “Materials and Methods”. We also described the N and P fluxes more explicitly in the “Results”. With the restructuring and added information we believe that the discussion is now clearer and easier to follow.

Comment 3. Tea plantation Although tea plantation is one of the dominant agriculture activities around the study site, this is not a representative of all agriculture as a whole. The uniqueness of this study would be “tea plantation” as an agricultural land use with much fertilizer than other crop. Therefore, I recommend revising the manuscript title from “mountain agriculture” to “mountain tea plantation” to inform this case study correctly. Also, abstract, discussion and conclusion need to convert “agriculture” to “tea plantation”.
We agree that using “tea plantation” instead of the more general “agriculture” is a good way to highlight the uniqueness of this study and to draw readers’ attention to a rarely studied agricultural system. Therefore, we have made the language changes accordingly throughout the manuscript title and its text.

Editorial comments
Comment 4. Figure 2 and 3; Please indicate the meaning of “X100” or “”X10” in the caption correctly.
Following the comment made by another reviewer, Figures 2 and 3 are replaced by Tables 2 and 3 and there is no X100 or X10 in the revised manuscript.

Comment 5. Figure 3; Table presentation would be much valuable for these data with the water flux data rather than figure.
We replaced Figure 2 and Figure 3 with Table 2 and Table 3.

Comment 6. Figure 5; Explain which figures are N and P in the caption. In the figure, difference of N output was 101 (=106 – 5.6) between two sites, while the manuscript indicate 90 (page 4796 line 17). Which is correct?
After considering the comments made by all reviewers we decided to make the figure only for N. As for the numbers, we have double checked the calculations and put the numbers into a table for clarification (Table 4).

Comment 7. You often use “topology” in the text. It might be “topography”.
We replaced “topology” with “topography” throughout the manuscript.

Comment 8. I couldn’t understand the meaning of “should A1 has 100% agriculture lands" (page 4796 line 20). Reword it
The sentence is changed to “Scaling up from 22% of tea plantation cover to 100%, the stream N and P outputs from A1 could reach as high as 450 kg ha-1 yr-1 and 2.5 kg ha-1 yr-1, respectively.”.

Anonymous Referee #2

The authors present a study based on two years of weekly stream water and precipitation chemistry measurements made in mountainous Taiwanese catchments. The authors have collected an interesting and potentially very useful data set but I have a number of serious reservations that I hope can be addressed.

The authors present a study that potentially has broad scientific significance and will be of interest to a wide audience working on catchment-scale element cycling and eutrophication problems.

In its present form, it is hard to judge the scientific quality of the manuscript. Specifically, not enough information is provided about the methods used to calculate element fluxes.

The overall presentation quality is fair but I believe that could be improved to good or excellent if the authors are able to incorporate my suggestions below.

The authors report concentrations and fluxes from streams in four catchments and two precipitation monitoring stations. Agricultural land use ranges from almost none to approximately 22%. As there are three headwater catchments and one mesoscale catchment (see Figure 1), I wonder if this paper might be better focused only on the headwater catchments.
One of our main findings is that a forested watershed downstream from a watershed with substantial agricultural cover has much lower concentrations of nutrients (by more than 70% for nitrate). The result also demonstrate that the inclusion of the large watershed, F1, revealed that monitoring in large watersheds maybe insufficient, particularly in agriculture-intensive region with heavy fertilizer application. Although, the strong dilution and landscape configuration can efficiently remove the nutrient, the heavily pollution may occur in tributaries.

The quality of the written English is generally very good, with a few exceptions. For example, p 4787, l. 28, “scarifying” is used in a very unusual manner and p 4790 l 27, the authors probably meant “without any preservatives”. I suggest that, if the paper is eventually accepted, the authors retain the services of a professional English language editor to ensure that all word choices are appropriate.
We changed “scarifying” to “sacrificing” and “without any preserves” to “without any preservatives”. We carefully checked the English throughout the manuscript and had an Ecology professor, Dr. Craig E. Martin, at the University to proofread the manuscript.

I am a little confused about the overall purpose of this paper. The authors note that agriculture is increasing in rugged mountain landscapes, yet it seems that any increase in agricultural area is forbidden in the reservoir catchment where they are working. Furthermore, the authors note a lack of published research on agriculture in montane areas, yet present numerous citations to earlier work in the study basin. I believe there is relevance to the work presented here as I think we do need to know more about the possible effects of agricultural intensification in humid montane environments but I would like the authors to clarify their focus.

It is correct that no additional increases in agricultural area is allowed at the study site. However existing agricultural area "is still having an impact at the study site", and throughout many (sub)tropical mountain areas around the world, agricultural area is expending.

It is also correct that quite a few studies have examined nutrient efflux and sediment output to the Feitsui Reservoir. We modified and added the following statements to clarify our focus.

"Yet, to our knowledge none examined both the effects of spatial configuration of agriculture lands on nutrient export and the effects of agriculture on atmospheric deposition. The FRW is rare among (sub)tropical mountain watersheds in that the effects of agriculture on its streamwater quality have been intensively studied. With the addition of this study, we believe that the FRW can serve as a classic case illustrating the effects of agriculture on nutrient cycling in watersheds with rugged topography and high precipitation, which can be very informative to other less-studied (sub)tropical mountain watersheds."

The authors do not provide enough information to assess the credibility of their flux estimates. They note that precipitation and streamflow were obtained from the Central Weather Bureau and Water Resource Agency of Taiwan (p 4791, l. 1-5) but provide insufficient information to interpret flux calculations. Specifically, I would like to see additional figures which present (i) monthly precipitation for each study month from the three rain gauges (mm/month) and (ii) monthly runoff (mm/month) from the two discharge gauges.

We moved the description of how we constructed nutrient fluxes from the Discussion to the Methods, and added more details to clarify our calculations of nutrient fluxes. We also added a figure (as a supplemental) to show the monthly precipitation and runoff (Fig. S1) as suggested by the reviewer.

I would also like more information about how the flux calculations were performed. I assume for the precipitation fluxes, weekly values were estimated by multiplying the concentration in a precipitation sample by the depth of precipitation over the previous week and then aggregating to monthly or annual scales. There are a number of different ways in which stream fluxes could have been estimated and I would really like this to be clarified. Such clarification is especially important given the extremely high fluxes reported by the authors. If the flux numbers are correct, they are really quite remarkable. The information on mean streamwater and rainfall chemistry is interesting but I do not think worth two of the four figures in the paper. This information could be summarized in a table and plots provided of concentrations and fluxes over time.

In response to this and the previous comments, we added more details to our description on the *calculations of the fluxes. Specifically in the revised manuscript we stated that "Weekly element fluxes through rainfall and streamflow of A1 and F2 were derived by multiplying weekly concentrations by weekly rainfall/streamflow. Monthly fluxes were accumulated from weekly fluxes, and when a weekly sample spanned over two months, it was divided into the two months in proportion to the rainfall/streamflow quantity."*

To properly display the values of all elements on one figure, we expressed some elements in 10 or 100 base units (e.g., 100 base unit for P). Unfortunately, when we were double-checking the flux values, we found a calculation mistake. The values for P output fluxes were taken from the values in 100 base unit rather than the original values. This is the reason for the much higher P outputs compared to inputs and we fixed this mistake in the revision. In addition, following the reviewer's suggestions, we now summarized streamwater and rainfall chemistry in a table and added two figures (now figures 2 and 3) to illustrate the temporal pattern of nutrient concentrations. We did not add a figure of monthly nutrient flux because it did not provide much information.

I am quite skeptical about the authors' assertion that erosion could have been responsible for higher phosphate concentrations in the F2 catchment (p 4793). I have no doubt that steeper slopes will, all other things being equal, have greater rates of erosion. Higher rates of erosion might explain higher concentrations of particulate phosphorus. However, the authors report dissolved phosphate concentrations from filtered samples. If higher rates of erosion will in fact lead to higher phosphate concentrations, the authors need to do a better job of explaining and justifying this phenomenon.
We agree that erosion is one of the main drivers for increased particulate phosphorus. However, many studies have shown that watersheds with high erosions can lead to increased dissolved phosphate concentrations as well (see examples listed below). To strengthen our discussion on the potential link between erosion on phosphate concentrations, we added several new citations (Gaynor and Findlay, 1995; Turtola and Jaakkola, 1995; Liu et al., 2006; Chang et al., 2008; Lee et al., 2013) in the revised manuscript, particularly the studies in Taiwan as they are highly relevant to current study.

- *Chang, C.H., Wen, C.G., Huang, C.H., Chang, S.P., Lee, C.S.: Nonpoint source pollution loading from an undisturbed tropic forest area. Environmental Monitoring and Assessment, 146: 113-126, 2008.*
- *Gaynor, J. D., and Findlay, W. I. Soil and phosphorus loss from conservation and conventional tillage in corn production. Journal of environmental quality, 24(4), 734-741, 1995.*
- *Lee, T.Y., Huang, J.C.*, Kao, S.J., Tung, C.P. (2013) Temporal variation of nitrate and phosphate transport in headwater catchments: the hydrological controls and land use alteration, Biogeosciences, 10 (4): 2617-2632, doi: 10.5194/bg-10-2617-2013.*
- *Liu, W.C., Chen, H.H., Hsieh, W.H., Chang, C.H.: Linking watershed and eutrophication*

modeling for the Shihmen Reservoir, Taiwan. Water Science and Technology, 54: 39-46, 2006.

- *Turtola, E., and Jaakkola, A. Loss of phosphorus by surface runoff and leaching from a heavy clay soil under barley and grass ley in Finland. Acta Agriculturae Scandinavica B-Plant Soil Sciences, 45(3), 159-165, 1995.*

I am also quite skeptical about the authors' proposed link between agricultural land use in the catchment and precipitation chemistry. Volatilization of ammonia from livestock (or perhaps fertilizer) is a well documented phenomenon. The authors report elevated levels of ammonium sulfate, urea and calcium ammonium sulfate (p 4794, l 9-11) in precipitation. I am quite concerned that what the authors are actually reporting is contamination of their precipitation samples. Did they weight the filters before and after to rule out presence of large amounts of particulates in the precipitation samples?
As the reviewer correctly pointed out, volatilization of ammonia from livestock (or fertilizers) is indeed well documented. In our analysis all samples were filtered through 0.45-μm filter papers, therefore it is unlikely that large amounts of particulates from volatilization of ammonia would present in the filtered samples.

The flux numbers are very difficult to understand. Please consider a table which presents fertilizer inputs, atmospheric inputs, harvest outputs and runoff losses for agriculture and forest land cover types in each study catchment. I read the paper several times and could not work out the numbers to my satisfaction.
We added more details in the revised manuscript to clarify our calculations of fluxes of N and P. We also added a table (Table 4) of the values of fluxes to make it easier to follow the numbers and calculations.

Finally, I would like to thank the authors for the opportunity to review this thought provoking paper (is it a positive term?). I hope that they will find my comments useful as I believe they have a potentially important contribution to our understanding of human impacts on water quality.
We thank the reviewer for the very thorough review which gave us an opportunity to thoroughly think through the focus and structure of our manuscript. We believe that the revised manuscript is a much more focused and better structured and hope that it contributes to our understanding of human impacts on mountain watershed nutrient cycling.

Anonymous Referee #3

Although the authors have compiled an interesting dataset and ask interesting questions regarding the impact of agriculture on watershed nutrient cycling, the manuscript is significantly weakened by the lack of a coherent and detailed presentation of watershed-scale mass balance data. In particular, all discussion of input-output ratios in Section 3.3 is presented out of context, with no discussion of how inputs and out-puts are calculated. To make the paper suitable for publication, the authors must work to provide a clear accounting of relevant nutrient inputs and outputs, provided details regarding their methodology in the methods section, and then place their discussion in the context of this quantitative analysis.
In the revision we added substantially more information on how we did the calculations of element fluxes. We removed the comparisons of output-input ratio between agriculture and forested watersheds for all elements except N. This is because the inputs for elements other than N and phosphorus (P) are not well understood, and the output of P lacks information on particulate P. Therefore, only N allows for a clear accounting. By providing more details in flux calculations and limiting the analyses to elements with the potential for clear accounting, we believe that the inferences made in the revised manuscript are greatly strengthened from the earlier draft.

Other issues to address:
p. 4786, line 15 Is it a novel finding that a dilution effect would occur when a nonagricultural watershed with low nitrate concentrations is present downstream from a high nitrate-yielding agricultural catchment? You may want to qualify this discussion, saying “As expected: : :”
Yes, we have modified the sentence to say “As expected…”

p. 4786, lines 18-20 Your estimate of nitrate contributed from agricultural land (400 kg/ha-y) is indeed high, but so are the fertilizer N input values that you give later in the paper. You may find it useful to place this analysis in terms of a NANI-style analysis (see Howarth et al. 2011, Hong et al. 2013, Boyer et al. 2002), where you analyze the relationship between riverine N output and your calculated net N inputs.
The suggestion raised by the reviewer is also what we want to present in this study. Indeed, the mass balance approach, like NANI, is a good way to understanding the nitrogen cycling in the watershed. In this revision, we added an equation to quantitatively describe the inputs and outputs. Furthermore, we added information on riverine nitrogen output ratios and discussed how they might reveal the characteristics of nitrogen retention in our mountainous watersheds with tea plantations.

p. 4787, line 6 Can you clarify your point about the impact of agriculture on ecosystem services?

Because we did not examine ecosystem services in this study per se, we removed this sentence to give more room to address other issues raised by the reviewers.

p. 4787, lines 10-11 You say that the impacts of agriculture are "likely" exacerbated by steep slopes and high precipitation, but give no references. Can you give more context and literature support to your hypothesis that the impacts of agriculture have unique impacts on the surrounding ecosystem in a mountain environment?

We rephrased this sentence with additional citations as follows: "The impacts are likely exacerbated by steep slopes and high precipitation as residence time is reduced and leaching potential increased under such conditions (Brouwer and Powell 1998; Tokuchi et al., 1999)."

0. 4787, line 18 You make the point here about the impact of nutrients on atmospheric deposition. In general, your treatment of the atmospheric deposition portion of the analysis in the paper is weaker than the analysis of stream data. You make the point, for example, that most NH3, for example, is "scavenged by precipitation" and then redeposited. If what is going up (NH4+) simply comes down again, what do we actually learn from this analysis of the deposition data?

Although what goes up will eventually come down, different chemicals come down at different distances, which has management implications at larger spatial scale. For example, SO_x *and* NO_x *can travel much greater distances than* NH_4^+ *which mostly deposited in adjacent ecosystems.*

p. 4788, lines 11-12 You refer to heavy use of fertilizers, but don't give a range. The input values are crucial here, so it is important to give real numbers.

We added the numbers of fertilizer inputs (425 to 2373 kg N ha^{-1} yr^{-1} *and 99 to 551 kg P* ha^{-1} yr^{-1}*) and added a new citation (Water Resources Agency 2010).*

p. 4788, line 21 You say that agriculture will have a negative effect on nutrient retention, but how are you defining this retention? Are you talking about a percent retention or absolute magnitude? And for N, for example, what constitutes retention? If N is being denitrified, is this included in the retention term?

Here we focus on the retention of nutrients especially N relative (proportional) to inputs so it is a percent retention (or retention ratio). In our calculation all input that stayed within the watershed (i.e., not lost through streamflow and harvest or emitted to the atmosphere) was considered retained within the watershed. Although denitrification was not included in the calculation (as we did not have the data) we considered denitrification as a component of output (because denitrified nitrogen oxides can be transported to other locations) and *discussed its influences on N retention ratio in the revised manuscript.*

p. 4789, line 20 Again, you say fertilizers are heavily applied, but don't give values. Please quantify.

We added the following information in the revision:

"… the amount of fertilizer used is assumed to be close to 786 kg N ha-1 yr-1 and 171 kg P ha-1 yr-1, the values taken from a case study in which the management practices (e.g., applications of fertilizers and pesticides, time and yield of harvests) were carefully recorded by a farmer in the same region as the current study (Tsai and Tsai, 2008). Although only one farmer was involved in the case study, the values are consistent to those reported by FAO (2002) and very close to the mean values across 10 tea plantations in our study area (743 kg N ha-1 yr-1 ranging from 425 to 2373 kg N ha^{-1} yr^{-1}, and 179 kg P ha^{-1} yr^{-1} ranging from 99 to 550 kg P ha^{-1} yr^{-1}; Water Resources Agency 2010)."

p. 4791, line 13 You say that you use a paired t-test to establish whether there is a statistically significant difference between watersheds. Is there a reason that you used a paired test? It seems that an unpaired t-test might be more appropriate here.

Because samples were taken weekly in the two locations with close spatial proximity, their precipitation data were likely influenced by the same temporal patterns in the air masses. Therefore, we used a paired t-test to address the shared temporal pattern between the two locations. Nonetheless based on the comment we also did un-paired tests and the results (statistical significance) were basically the same although the p values changed.

p. 4792, section 3.3 This section regarding output-input ratios needs more explanation. You are reporting results regarding these ratios, but there is nothing here that tells the reader how you have obtained these results. You should consider adding a section to Materials and Methods which details the methodology for whatever calculations you have done.

We removed the output-input ratios for all elements except N in the revised manuscript. For N, we added more details to the Method to clarify the calculations.

Line 4796, section 4.5 Your calculations here need to be better explained. You say that you subtract nutrient output at F1 from that at A1 to estimate the N and P output from agriculture at A1, but the logic behind this isn't clear. It would be helpful here to have a table of your calculated inputs and outputs for each watershed.

We rephrased the sentence to clarify the logic behind our calculations:

"Because the proportion of agriculture cover was very low at F2 (i.e., 0.38%), and the resulting fertilizer input and harvest output were small and already accounted for (Table 4), we treated F2 as a background and attributed the differences between A1 and F2 to

agriculture activities." In the revised manuscript we used A1 and F2 for the comparison because we believe this is a more direct comparison as we do not have precipitation data of F1. We also provided a table of inputs and outputs of N and P for A1 and F2 (Table 4).

Line 4800, lines7-10 In your conclusion you describe the effects of agriculture on these mountain watersheds, and conclude that "agriculture activities have a more pervasive impact on watershed nutrient cycling than previously recognized." There is a very large body of literature detailing the impacts of agriculture on watershed nutrient cycling, so this statement seems unwarranted.
We rephrased the first conclusion to: "Agriculture and forested watersheds in tropical/subtropical mountains could have distinct patterns of nutrient cycling. Even a moderate proportion of tea plantation cover (17-22%) in mountain watersheds, when in combination with steep slopes and high precipitation, could lead to much higher ion concentrations in both streamwater (nutrient output) and rainwater (nutrient input), and much lower N retention ratio at watershed scale. Thus, mountain watersheds may be particularly vulnerable to agriculture expansion."

p. 4800, lines 19-21 You report that agricultural lands in your study are contributing 400 kg/ha-y N, and comment on the uniqueness of the finding. You don't, however, provide the context for these outputs (very high fertilizer inputs, >700 kg /ha-y). You also don't fully explain how you can estimate rates of P output (260 kg/ha-y) that are more than three times the estimated inputs (75 kg/ha-y). Again, spending more time on developing and explaining your nutrient mass balance for the watershed would strengthen your conclusions. This may also involve explaining the unique fertilizer requirements of a tea plantation and the typically very large fertilizer inputs.
We added more details on the derivation of nutrient fluxes in the Method. We also added information on the effect of high fertilizer input on streamwater element output in the discussion. The fluxes of P was not correctly calculated in our original manuscript. After the recalculation the output was not nearly as large so much of the discussion on P was removed in the revised manuscript. We added information about over-fertilization on tea plantations by citing a study in Japan (Oh et al. 2006).
Oh, K., Kato, T., Li, Z. P., Li, F. Y.: Environmental problems from tea cultivation in Japan and a control measure using calcium cyanamide, Pedosphere, 16(6), 770-777, 2006.

Although the paper is generally well written, there are some problems with grammar that should be corrected. Some examples include: "high precipitations" (p. 4787, line 10) "scarifying socioeconomic benefits" (p. 4787, line 25) "is characterized with high rainfall" (p. 4788, line 10) "without any preserves" (p. 4790, line 27)

We changed these to “is characterized by high precipitation”, “sacrificing socioeconomic benefits: and “without any preservatives”. We also carefully checked through the manuscript for language issues and had an ecologist, Dr. Craig E. Martin, at the University of Kansas proofread the manuscript.

Effects of mountain tea plantation 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 and rainfall chemistry of mountain watersheds at Feitsui Reservoir Watershed in northern Taiwan to examine the effects of agriculture on watershed nutrient cycling. We found that the greater the proportion of tea plantation cover, the higher the concentrations of fertilizer-associated ions (NO_3^-, K^+) in stramwater of the four mountain watersheds examined; on the other hand, the concentrations of the ions that are rich in soils (SO_4^{2-}, Ca^{2+}, Mg^{2+}) did not increase with the proportion of tea plantation cover, suggesting that agriculture enriched fertilizer-associated nutrients in streamwater. Of the two watersheds for which rainfall chemistry was available, the one with higher proportion of tea plantation cover had higher concentrations of ions in rainfall and retained less nitrogen in proportion to input compared to the more pristine watershed, suggesting that agriculture can influence atmospheric deposition of nutrients and a system's ability to retain nutrients. As expected, we found that a forested watershed downstream of agricultural activities can dilute the concentrations of NO_3^- in streamwater by more than 70%, indicating that such a landscape configuration helps mitigate nutrient enrichment in aquatic systems even for watersheds with steep topography. We estimated that tea plantation at our study site contributed approximately 450 kg ha^{-1} yr^{-1} of NO_3-N via streamwater, an order of magnitude greater than previously reported for agriculture lands around the globe and can only be matched by areas under intense fertilizer use. Furthermore, we constructed watershed N fluxes to show that excessive leaching of N, and additional loss 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.

Key Words: mountain agriculture, tea plantation, nutrient cycling, nitrogen retention ratio, output-input ratio, Feitsui Reservoir, fertilization

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, China (Johda et al., 1992) and the Andes (Sarmiento and Frolich, 2002). It is well established that watershed nutrient cycling is tightly linked to land use, 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). The impacts are likely exacerbated by steep slopes and high precipitation as residence time is reduced and leaching potential increased under such conditions (Brouwer and Powell 1998; Tokuchi et al., 1999). Thus, mountain agriculture in the tropics and subtropics characterized with high precipitation is likely to have substantial negative impact on ecosystem function. Yet, empirical studies in tropical or subtropical mountain watersheds are very limited.

In addition to nutrient output in streamwater, cultivation and fertilization on agriculture lands could 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, few studies of watershed nutrient cycling have examined the effects of land use on precipitation chemistry.

Proper 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 approximately 10–20 ha surrounding the drainage area, where much of the variation in element fluxes occurred (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 (reviewed by Muscutt et al., 2001). Through proper landscape configuration, negative impacts of agriculture on nutrient cycling of mountain watersheds may also be reduced without sacrificing socioeconomic benefits of agriculture. However, what constitutes a proper landscape configuration is likely to vary with climate and topography.

Here we examined the effects of mountain agriculture, mainly tea plantation, 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 assess the 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 tea plantation covers, and compared their rainfall chemistry in relation to streamwater chemistry. The FRW is characterized with high rainfall (> 3000 mm; Taipei Feitsui Reservoir Administration), steep slopes (on average 42%) and heavy use of fertilizers in tea plantation (425 to 2373 kg N ha^{-1} yr^{-1} and 99 to 551 kg P ha^{-1} yr^{-1}; Water Resources Agency 2010, see Methods for details). Many studies have demonstrated substantial nutrient efflux and sediment production from surrounding tea plantation to the reservoir over the past 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). Yet, to our knowledge none examined both the effects of spatial configuration of agriculture lands on nutrient export and the effects of agriculture on atmospheric deposition. The FRW is rare among (sub)tropical mountain watersheds in that the effects of agriculture on its streamwater quality have been intensively studied. With the addition of this study, we believe that the FRW can serve as a classic case illustrating the effects of agriculture on nutrient cycling in watersheds with rugged topography and high precipitation, which can be very informative to other less-studied (sub)tropical mountain watersheds.

We hypothesized that agriculture would increase nutrient output in streamwater (H_1) as well as atmospheric input of nutrients through rainfall (H_2). We also hypothesized that through the disruption of natural vegetation, agriculture would increase nutrient leaching and decrease retention ratio of essential nutrient elements (*H_3*). Our specific predictions are:

1. Watersheds with higher proportion of tea plantation cover have higher concentrations and fluxes of fertilizer-associated ions in the streamwater than forested watersheds (*H_1*).
2. Watersheds with higher proportion of tea plantation cover have higher concentrations and fluxes of fertilizer-associated ions in the rainfall than forested watersheds (*H_2*).
3. Watersheds with higher proportion of tea plantation cover have lower nitrogen retention ratio (in proportion to input) than forested watersheds (*H_3*).

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

2. Materials and Methods

2.1 Study site

The FRW is located along the Peishi Creek of northern Taiwan, with a drainage area of 303 km^2. The elevation of the FRW ranges from 45 m 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 spatially varied, ranging from 3500 mm in the southwest portion of the 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 plantation covering an area of 1200 ha, or 25% of all agricultural lands (Chang and Wen, 1997; Chou et al., 2007). In 1986 the FRW was designated as a water resource protection area, followed by the construction of the 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 which is still having an impact at the study site.

2.2 Sampling regime

Four watersheds of the FRW (A1, A2, F1, F2; Fig. 1) with varying proportions of tea plantation cover (22% in A1, 17% in A2, 2.9% in F1, 0.4% in F2; Table 1) were included in this study. Agriculture of other crops consists only small proportions of the watersheds (<1%) so it was not included in Table 1. Natural forests are the most dominant land cover for all four watersheds (68% in A1, 76% in A2, 93% in F1, 99% in F2; Table 1), making tea plantation the primary contributor to the differences in landscape across the four watersheds. Weekly samples of streamwater were collected from all 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 watersheds (< 3 km^2) drained by first order streams whereas F1 is a much larger watershed (86 km^2) drained by a third order stream that drains through A1 and A2 (Fig. 1). We collected weekly rainfall and streamwater samples every Tuesday from September 2012 to August 2014. Rainfall samples were collected using a 20-cm diameter polyethylene (PE) bucket, from which a 600-mL subsample was taken and placed into 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 taken and placed into a PE bottle for transportation back to the laboratory.

2.3 Water chemistry

All samples were transported back to the laboratory within 24 hours. Conductivity and pH of the water samples were measured on the same day of collection. The samples were filtered through 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, 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 preservatives.

Data on rainfall and streamflow quantity of the watersheds were estimated from the rain gauges and discharge gauges maintained by the Central Weather Bureau and Water Resource Agency of Taiwan, respectively. 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 weekly and monthly rainfall of a watershed was directly assigned to the values registered at the nearest rain gauge (i.e., COA530 for A1 and COA540 for F2, Fig. 1 and Fig. S1a). The weekly and monthly streamflow of a watershed was estimated by the area ratio method in which the streamflow was assigned to the values registered at the nearest discharge gauge (i.e., 1140H099 for A1, A2, and F1, and 1140H097 for F2, Fig. S1b) and then adjusted by the area ratio of the studied watershed relative to the watershed where the discharge gauge was located. The validity of this method has been confirmed for several watersheds in Taiwan (Huang et al., 2012; Lee et al., 2014).

2.4 Element fluxes

Weekly element fluxes through rainfall and streamflow of A1 and F2 were derived by multiplying weekly concentrations by weekly rainfall/streamflow. Monthly fluxes were accumulated from weekly fluxes, and when a weekly sample spanned over two months, it was divided into the two months in proportion to the rainfall/streamflow quantity.

In order to provide a more comprehensive understanding on how mountain agriculture affects watershed nutrient cycling, we constructed and compared N and P fluxes for watersheds with the highest (A1) and lowest (F2) tea plantation cover. We made three assumptions in the calculation of watershed nutrient fluxes. First, we assumed the input from dry deposition is 28% of that from precipitation for both watersheds. This value was based on a study using Na^+ ratio method at the Fushan Experimental Forest (Lin et al., 2000), a natural hardwood forest 17 km south of the FRW. Second, the amount of fertilizer used is assumed to be close to 786 kg N ha^{-1} yr^{-1} and 171 kg P ha^{-1} yr^{-1}, the values taken from a case study in which the management practices (e.g., applications of fertilizers and pesticides, time and yield of harvests) were carefully recorded by a farmer in the same region as the current study (Tsai and Tsai, 2008). Although only one farmer was involved in the case study, the values are consistent to those reported by FAO (2002) and very close to the mean values across 10 tea plantations in our study area (743 kg N ha-1 yr-1 ranging from 425 to 2373 kg N ha^{-1} yr^{-1}, and 179 kg P ha^{-1} yr^{-1} ranging from 99 to 550 kg P ha^{-1} yr^{-1}; Water Resources Agency 2010). Adjusting for the proportion of agriculture lands (22.1%, 0.38%), the amount of fertilizers used in A1 was estimated to be 173.7 kg N ha^{-1} yr^{-1} and 37.8 kg P ha^{-1} yr^{-1}, and that in F2 to be 3 kg N ha^{-1} yr^{-1} and 0.6 kg P ha^{-1} yr^{-1}. There was very little change in biomass of tea plantation after 10 years because tea plants are regularly trimmed with the litter left in the field to maintain the same height optimal for harvest. Thus, our third 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 and the amount of P removed (7.35 kg ha^{-1} yr^{-1}) was calculated using the median of P:N ratios (0.065) reported for tea trees in Taiwan (Tsai and Tsai, 2008). After adjusting for the proportion of tea plantation cover, A1 was estimated to have 25.0 kg N ha^{-1} yr^{-1} and 1.6 kg P ha^{-1} yr^{-1} removed through harvest, and F2 to have 0.43 kg N ha^{-1} yr^{-1} and 0.03 kg P ha^{-1} yr^{-1} removed through harvest. Using the following mass balance model, we constructed fluxes of N and P of the two watersheds:

$$\text{Ratio}_{ret} = 1 - \frac{OUT_{riv} + OUT_{harv}}{IN_{dep} + IN_{fer} + IN_{fix}} \qquad \text{Eq. 1}$$

Here, $\text{Ratio}_{\text{ret}}$ indicates the ratio of N input to the watershed that was retained within the watershed. The OUT_{riv} and OUT_{harv} are the riverine N export and harvest, respectively.

The IN_{dep}, IN_{fer}, and IN_{fix} indicate the atmospheric deposition, fertilizer application, and biological fixation. Note that the biologic fixation term was not used for P calculation. Since the tea plantation does not use leguminous crop as fertilizers and the biological fixation in tropical forest is known to be less than 10 kg N ha^{-1} yr^{-1} (Sullivan et al. 2014), the IN_{fix} is assumed to be between 0 and 10 kg N ha^{-1} yr^{-1}. We did not include the loss through denitrification and volatilization within tea field in the calculation of N retention ratio because we did not have good estimates. However, the effects of such uncertainties and omissions on estimating N retention ratio were discussed. We did not calculate the retention ratio for P because the majority of P in watersheds was in particulate forms (Smith et al., 1991) that were not analyzed in our study.

2.5 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). All statistical analysis was conducted using SPSS 22.0 (IBM Corporation, New York).

3. Results

3.1 Streamwater chemistry

The concentrations of all analyzed ions in streamwater differed significantly among the four watersheds (Table 2). A1, the watershed with the highest proportion covered by tea plantation, had significantly higher concentrations of all ions except H^+ than the other three watersheds (Table 2, Fig. 2). In contrast, F2, the watershed with the lowest proportion covered by tea plantations, had the lowest concentrations of H^+, Na^+, K^+, Cl^- and NO_3^-. Furthermore, it is worth noting that F2, the watershed with the steepest slopes, had the second highest concentrations of ions rich in soils and soil solution, including Ca^{2+}, Mg^{2+} and SO_4^{2-} (Table 2, Fig. 2).

Similar to ion concentration, the fluxes of all ions differed significantly among watersheds (Table 2). A1 had the largest fluxes of K^+, Ca^{2+}, Mg^{2+}, NO_3^- and SO_4^{2-} and F2 had the smallest fluxes of H^+, Na^+, K^+, Mg^{2+}, Cl^-, and NO_3^- (Table 2). 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 (Table 2). Although the fluxes of Na^+ and Cl^- differed significantly among A1, A2 and F1, these differences were considerably smaller than the differences between the three watersheds and F2 (Table 2).

3.2 Rainfall chemistry

Five of the 10 measured ions had significant ($p < 0.05$) or marginally significant ($p < 0.1$) higher concentrations in A1 than F2 (H^+, Na^+, Cl^-, NO_3^-, $p < 0.05$; NH_4^+, $p = 0.067$; Table 3, Fig. 3). 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^-, $p < 0.1$; Table 3).

3.3 N and P fluxes

Because the proportion of agriculture cover was very low at F2 (i.e., 0.38%), and the resulting fertilizer input and harvest output were small and already accounted for (Table 4), we treated F2 as a background and attributed the differences between A1 and F2 to agriculture activities. We estimated that stream N and P outputs from the tea plantation at A1 to be approximately 105.7 kg ha^{-1} yr^{-1} and 5.6 kg ha^{-1} yr^{-1}, respectively (Table 4). Scaling up from 22% of tea plantation cover to 100%, the stream N and P outputs from A1 could reach as high as 450 kg ha^{-1} yr^{-1} and 2.5 kg ha^{-1} yr^{-1}, respectively.

From our mass balance construction of element fluxes, N input exceeded output at both watersheds (Table 4, Fig. 4). At A1, 35% of the N input (69 kg ha^{-1} yr^{-1}) to the watershed was retained (Table 3 and Fig. 4). At F2, 72% of the N input (15 kg N ha^{-1} yr^{-1}) was retained (Table 4 and Fig. 4). For P, the output through streamflow (2.6 kg ha^{-1} yr^{-1}) was smaller than the input through atmospheric deposition (3.6kg ha^{-1} yr^{-1}) at F2. At A1, the output of P through streamflow and harvest (5.8 kg ha^{-1} yr^{-1}) was greater than the input through atmospheric deposition (4.6 kg ha^{-1} yr^{-1}), but when fertilization was taken into account, the total output of PO_4^{3-}-P was trivial relative to total P input (42.4 kg ha^{-1} yr^{-1}) (Table 4).

4. Discussion

4.1 Streamwater chemistry

The watershed with the highest proportion of tea plantation cover (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 the output of fertilizer-associated ions (NO_3^- and K^+) matched perfectly to the proportion of tea plantation cover across the four watersheds (i.e., the rank of the proportion of tea plantation cover from high to low: A1, A2, F1, F2; rank of ion concentration and flux from high to low: A1, A2, F1, F2) strongly supports the effects of agriculture on streamwater chemistry (H_1).

However, streamwater chemistry is affected 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. Erosion is known to enhance leaching loss of PO_4^{3-} (Gaynor and Findlay, 1995; Turtola and Jaakkola, 1995; Liu et al., 2006; Chang et al., 2008; Lee et al., 2013). The 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 the less steep F1 (39%) (Fig. 2, Table 2), despite that F2 has a higher proportion of natural forest cover. Soil erosion is arguably the greatest concern to most P mitigation programs because the concentration of P on surfaces of soil particles is often orders of magnitude greater than that in soil solution (Sharpley et al., 2002; Kleinman et al., 2011). Therefore, it is not surprising that topography 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+}, the ions that are abundant in soils.

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 tea plantation cover, compared to F2, the watershed almost entirely covered by natural forests. Ammonium sulfate, urea and calcium ammonium nitrate [$5Ca(NO_3)_2.NH_4NO_3.10H_2O$] 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 likely contributed to the high concentrations and fluxes of NO_3^- and NH_4^+ in rainfall at A1. On the other hand, the concentrations of PO_4^{3-} and K^+ in rainfall were not higher at A1 compared to F2, which may be explained by the low mobility of PO_4^{3-} and smaller quantity of P and K in fertilizers.

Once in the atmosphere, aerosols/chemicals can be transported to other locations but most of them will deposit to nearby ecosystems. In central Taiwan, high NH_4^+ concentration in precipitation at a high elevation forest (2,000 m) was attributed to mountain agriculture that occurred 10 km away (Ding et al., 2011). With the predicted expansion of agriculture to the mountains both in Taiwan and many other regions (Johda 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 concentrations and fluxes of Na^+ and Cl^-, and to a lesser degree Mg^{2+}, at A1 than F2 likely reflected 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 the 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 Landscape configuration and streamwater chemistry

The large differences in NO_3^- concentration and flux between F1 and A1, A2 highlight the role of landscape configuration on streamwater chemistry. Both A1 and A2 are sub-watersheds of F1; however, the influence of tea plantation on A1 and A2 largely dissipated as water entered into forested F1. Specifically, the concentration of NO_3^- was 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. 2). 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

The per-hectare output of N from tea plantation reported here (450 kg ha^{-1} yr^{-1}) 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). For the four watersheds that were 30-40% covered by row crops and received fertilization at 50-70 kg ha^{-1} yr^{-1} N in southeastern coastal plain of the United States, nutrient output through streamflow was < 6 kg N ha^{-1} yr^{-1} (Lowrance et al., 1985). In the Great Barrier Reef, Australia, total output via streamflow was approximately 5 kg ha^{-1} yr^{-1} for NO_3-N from a watershed with 29% of the land covered by pasture and 14% by crop lands (Hunter and Walton, 2008).

High N output from agricultural lands is probably common in Taiwan and other regions under intensive fertilizer use. It has been reported that over-fertilization is common in Japan, Korea and Taiwan, and despite an estimated 23-63% over-fertilization, the use of fertilizers is still increasing in the region (Ahmed, 1996). In the Dansheui River of northeastern Taiwan, the output of dissolved inorganic N ranged from 3 kg ha^{-1} yr^{-1} in relatively pristine headwaters covered mostly by natural forests to 100 kg ha^{-1} yr^{-1} in 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% and the site is located in the hilly upstream region), and very heavy application of N fertilizers (300-1000 kg ha^{-1} yr^{-1}), reached 73 kg ha^{-1} yr^{-1} (Chen et al., 2008), approximately the same magnitude as those reported here. Our study clearly demonstrated that high application of fertilizers in regions with high rainfall and steep slopes could lead to extremely high output of N and, therefore, eutrophication risk for downstream watersheds. The misconception that heavy fertilization leads to high economic profit has resulted in the popular practice of heavy fertilization in tea plantations, commonly at a level similar to or higher than that in our study site (740 kg N ha^{-1} yr^{-1}). For example, conventional N fertilization in tea plantation is approximately 1100 kg ha^{-1} yr^{-1} in Japan, which is more than twice the suggested amount with same tea yield (Oh et al., 2006).

In contrast to N, most of the P fertilizers was retained within the watershed or transported in particulate form so that dissolved P only accounts for a small proportion of the input. In most agriculture watersheds, the majority (>90%) of P leaves the watersheds in particulate form (Smith et al., 1991) and the loss in dissolved form (i.e., PO_4^{3-}) through runoff is relatively minor (Brady and Weil 1999). Thus, while the dissolved form of P could respond to land use changes, a complete P budget at watershed scale still requires reliable estimates on the particulate P.

4.6 Watershed N fluxes

The 72% N retention at F2 is likely an underestimate because the input from biological N fixation (BNF) was not included in the calculation. Based on a recent synthesis (Sullivan et al. 2014), 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 to N input could increase N retention ratio at F2 (assuming a BNF of 10 kg ha^{-1} yr^{-1}, the N retention ratio at F2 would increase from 72% to 81%). The high N retention ratio of F2 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 at A1, 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 ratio at A1 (assuming a BNF of 10 kg ha^{-1} yr^{-1}, the N retention ratio at A1 would increase from 35% to 37%).

In addition to BNF, the calculation of N retention ratio did not take into account the loss through volatilization and denitrification. Because it rains frequently at the 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 account, the N retention ratio 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 ratio and the resulting high leaching loss of N at A1, impose a major threat to the streamwater quality that could lead to reservoir eutrophication.

Surprisingly, from our construction of the N fluxes, the loss of N through annual harvest (25 kg ha^{-1} yr^{-1}) at A1 approximately equals the annual atmospheric deposition (26 kg ha^{-1} yr^{-1}), of which only a small portion should have come from fertilizers (atmospheric N deposition at F2 is only 8 kg lower than A1, suggesting that less than 8 kg of atmospheric N deposition could potentially come from fertilizers). In other words, to maintain the current harvest, not much N fertilization is actually required, and most of the 173.7 kg N ha^{-1} yr^{-1} from fertilization are simply lost through hydrological process (i.e., leaching) to the streams and the Feitsui Reservoir and/or returned to the atmosphere, both of which could have negative environmental impacts. Our construction of the element fluxes clearly showed that the N fertilizers are applied at rates that are neither ecologically nor economically sound, and such excessive fertilization may cause fundamental changes in watershed nutrient cycling (Fig. 4).

5. Conclusions

1. Agriculture and forested watersheds in tropical/subtropical mountains could have distinct patterns of nutrient cycling. Even a moderate proportion of tea plantation cover (17-22%) in mountain watersheds, when in combination with steep slopes and high precipitation, could lead to much higher ion concentrations in both streamwater (nutrient output) and rainwater (nutrient input), and much lower N retention ratio at watershed scale. Thus, mountain watersheds may be particularly vulnerable to agriculture expansion.
2. Topographic control is important in nutrient leaching from 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. The contribution of tea plantation to the N output in streamwater for one of the studied watersheds (i.e., A1) is estimated at approximately 450 kg N ha^{-1} yr^{-1}. This level of fertilization exceeds previous reports around the globe, and can only be matched in magnitude by one study in China where fertilizers were excessively applied.

5. The conservative construction of the N fluxes for the watersheds indicates over-fertilization at one of the studied watersheds (i.e., A1), which likely resulted in leaching of N and additional loss of N to the atmosphere via volatilization and denitrification.

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Table 1. Basic information of the studied watersheds.

	A1	A2	F1	F2
Area (km^2)	2.92	1.36	86.04	0.67
Slop (%)	39.3	34.8	38.7	48.1
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

Table 2. Mean (± 1 standard error) monthly ion concentration (volume-weighted from weekly samples) and flux of streamflow.

A1, A2, F1 and F2 denote the four watersheds; diff: post-hoc comparisons among the four watersheds with different letters indicating statistical differences ($p < 0.05$).

Ion	Concentration (μeq L^{-1})					Flux (meq m^{-2} mo^{-1})				
	A1	A2	F1	F2	diff	A1	A2	F1	F2	diff
H^+	0.96±0.006	1.22±0.006	0.91±0.007	0.76±0.004	a,b,a,c	0.030±0.001	0.038±0.001	0.036±0.001	0.016±0.004	a,b,ab,c
Na^+	266±4.88	254±3.65	233±4.45	231±4.10	a,b,c,c	76.4±1.74	73.0±1.70	80.1±1.68	46.7±0.90	a,b,ab,c
K^+	282±0.87	213±6.27	125±0.49	108±3.63	a,b,c,d	8.24±0.20	6.14±0.14	4.27±0.50	2.19±0.36	a,b,c,d
Ca^{2+}	306±7.49	193±5.41	170±7.34	273±8.04	a,b,c,d	87.0±1.92	54.1±1.17	55.8±1.02	54.4±1.00	a,b,b,b
Mg^{2+}	255±5.10	188±4.25	148±4.72	206±5.78	a,b,c,d	72.5±1.62	52.8±1.15	49.2±0.94	41.0±0.74	a,b,b,c
Cl^-	199±4.00	182±3.06	178±4.76	145±2.55	a,b,b,c	59.2±1.51	53.2±1.34	62.8±1.49	29.8±0.64	a,b,a,c
NO_3^-	209±5.31	158±2.80	28.3±0.76	16.1±0.95	a,b,c,d	62.9±1.63	46.8±1.19	10.2±0.25	3.32±0.078	a,b,c,d
SO_4^{2-}	212±6.29	123±3.96	116±3.96	183±6.45	a,b,c,d	59.2±1.30	33.9±0.74	39.1±0.78	35.7±0.66	a,b,b,b
PO_4^{2-}	1.50±0.182	1.38±0.174	0.72±0.114	1.29±0.026	a,b,b,a	1.14±0.0030	1.08±0.0054	0.69±0.028	0.69±0.0030	a,a,b,b

Table 3. Mean (± 1 standard error) monthly ion concentration (volume-weighted from weekly samples) and flux of rainfall.

A1 and F2 denote the two watersheds; an asterisk * indicates a significant difference between the two watershed ($p < 0.05$); an asterisk inside a parenthesis (*) indicates a marginally significant difference between the two watersheds ($p < 0.1$).

Ion	Concentration (μeq L^{-1})		Flux (meq m^{-2} mo^{-1})	
	A1	F2	A1	F2
H^+	39±6.7	31±5.4*	12±3.9	7.9±1.5*
Na^+	107±24	84±18*	30±8.5	23±6.5(*)
K^+	8.0±1.3	7.8±1.3	2.2±0.45	1.9±0.32
Ca^{2+}	21±3.2	19±4.4	5.7±1.0	4.2±0.61*
Mg^{2+}	30±5.8	26±5.6	8.2±2.1	6.5±1.7(*)
NH_4^+	19±2.9	15±2.7(*)	5.1±1.3	3.8±0.67(*)
Cl^-	140±30	100±22*	38±11	28±8.2*
NO_3^-	24±3.9	18±3.0*	7.0±2.0	4.7±0.90(*)
SO_4^{2-}	58±8.6	53±7.7	15±3.6	13±2.4
PO_4^{3-}	0.96±0.03	0.63±0.03	0.75±0.30	0.51±0.12

Table 4. Inputs and outputs of nitrogen and phosphors of watersheds A1 and F2. See text for the assumptions made in the calculations of dry deposition, fertilization and harvest.

		Nitrogen (kg ha^{-1} yr^{-1})		Phosphors (kg ha^{-1} yr^{-1})	
		A1	F2	A1	F2
Input					
	Wet deposition	20.4	14.3	3.6	2.8
	Dry deposition	5.7	4.0	1.0	0.8
	Fertilization	173.7	3.0	37.8	0.6
	Total	199.8	21.3	42.4	4.2
output					
	Harvest	25.0	0.4	1.6	0.0
	Stream output*	105.7	5.6	4.2	2.6
	Total	130.7	6.0	5.8	2.6

*For stream output, only dissolved inorganic forms are considered.

Legends of figures

Figure 1. Location and land use distribution of the studied watersheds.

Figure 2. Monthly ion concentration (volume-weighted from weekly samples) of streamwater of watersheds A1, A2, F1, and F2.

Figure 3. Monthly ion concentration (volume-weighted from weekly samples) of rainfall of watersheds A1 and F2.

Figure 4. Schematic diagram of N fluxes of watersheds A1 and F2. A1 represents a watershed with 22% agricultural lands and 68% forests (a); F2 represents a watershed with 0.38% agricultural lands and 99% forests (b). Biological N fixation is not included in the diagram and its effects on N retention ratio is described in the Discussion.

Fig. S1. Monthly streamflow (A) and rainfall (B) and streamflow of discharge gauges and rain gauges used in the study.

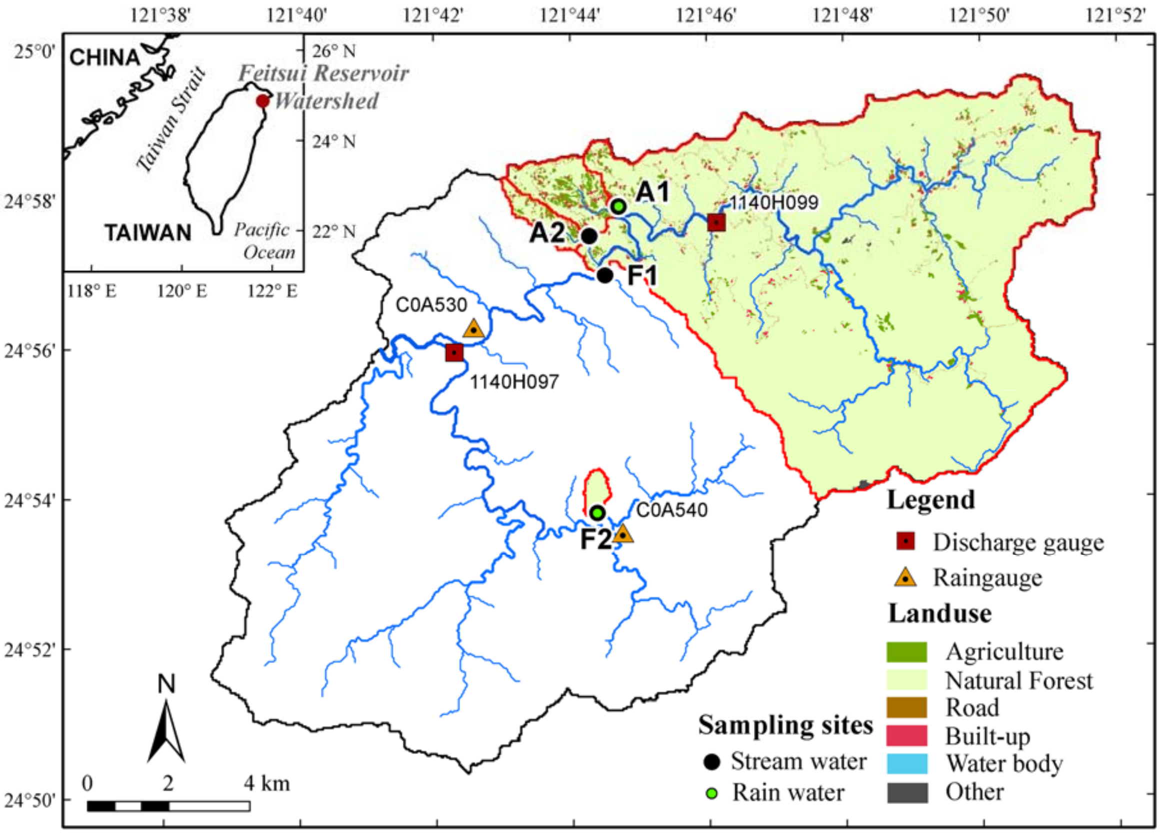
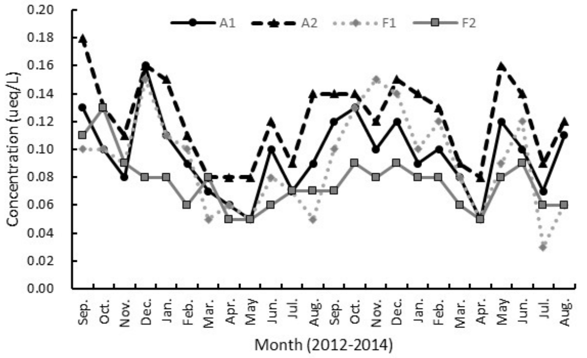
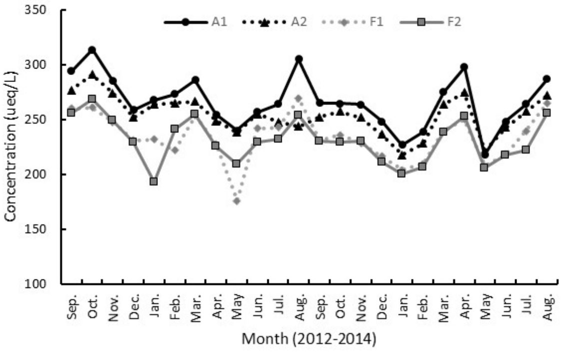


Figure 1. Location and land use distribution of the studied watersheds.

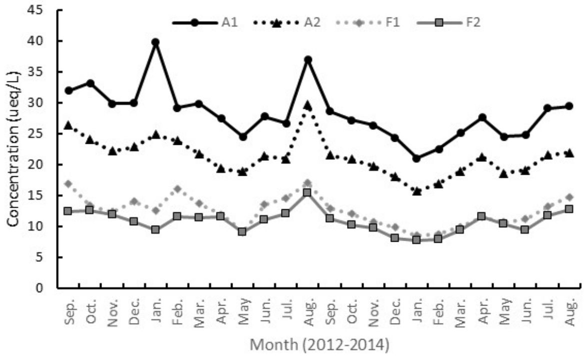
H^+



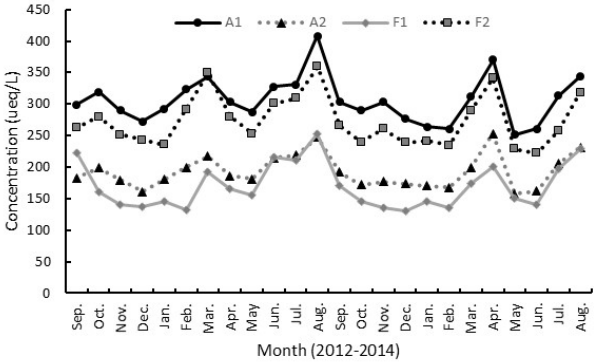
Na^+



K^+



Ca^{2+}



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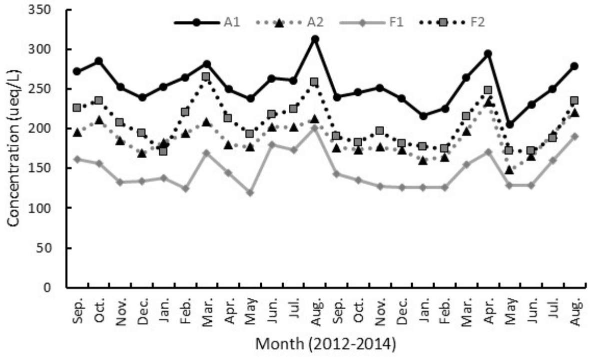
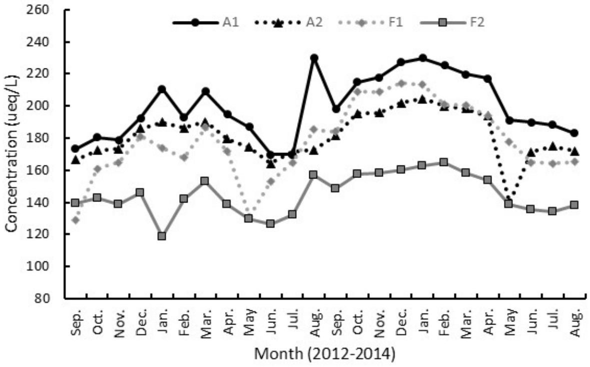
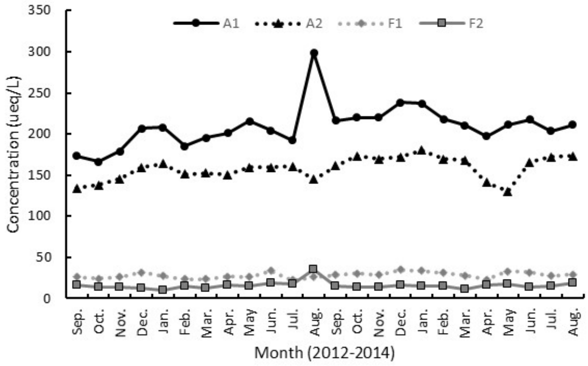


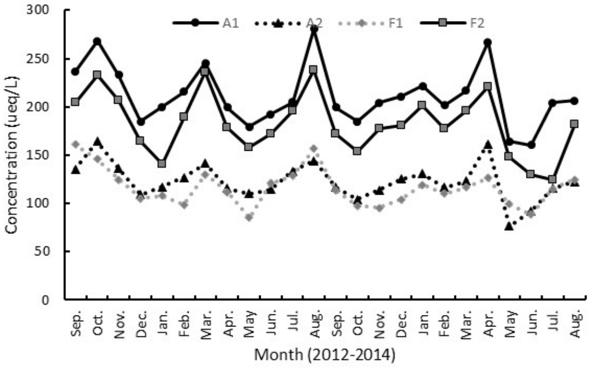
Cl^-



NO_3^-



SO_4^{2-}



PO_4^{3-}

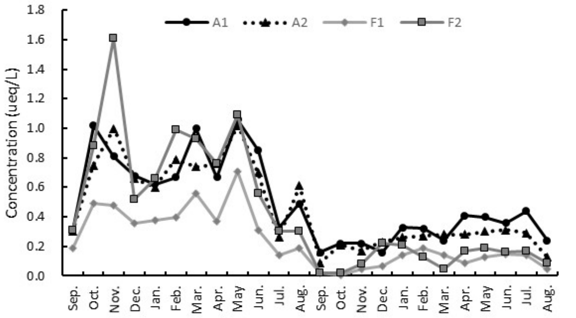
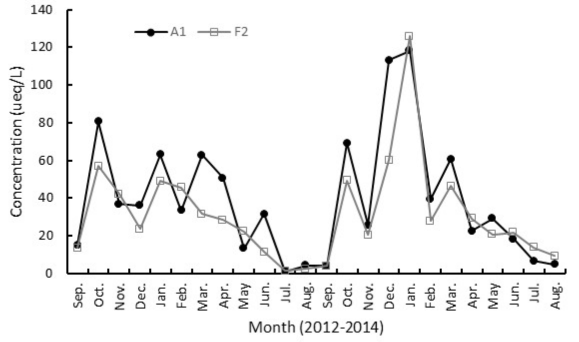
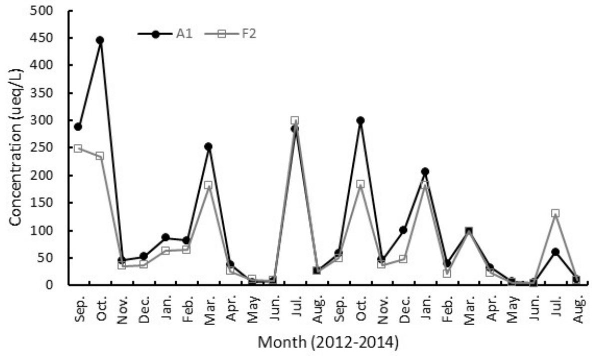


Figure 2. Monthly ion concentration (volume-weighted from weekly samples) of rainfall of watersheds A1 and F2.

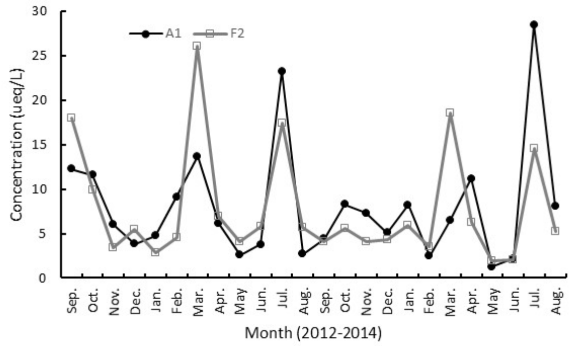
H^+



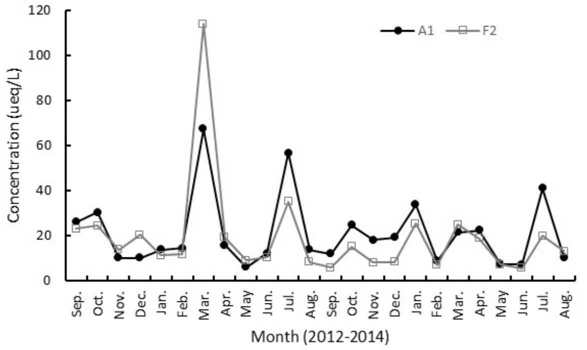
Na^+



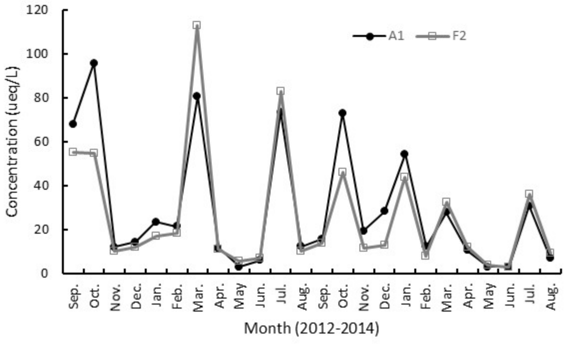
K^+



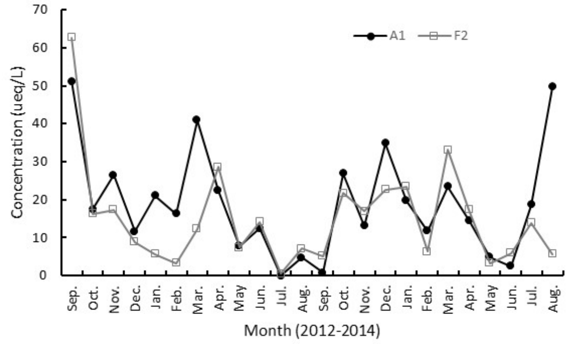
Ca^{2+}



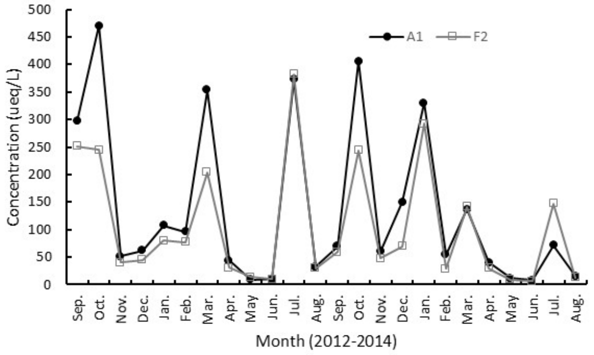
Mg^{2+}



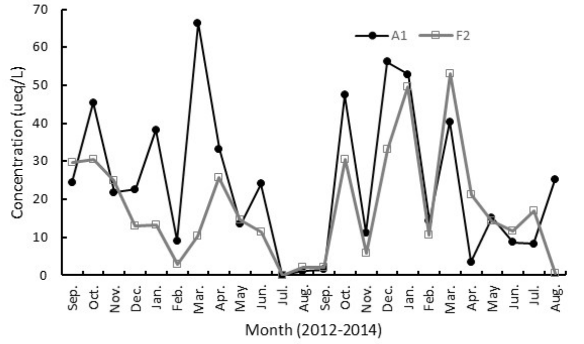
NH_4^+



Cl^-



NO_3^-



SO_4^{2-}

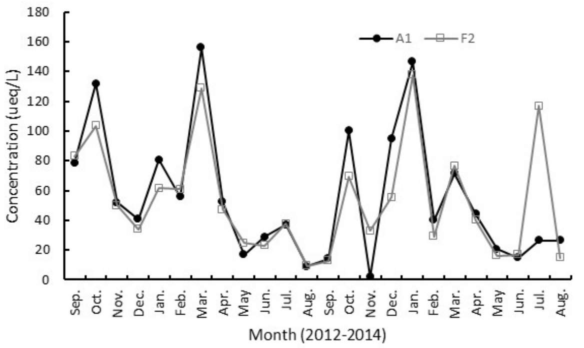


Figure 3. Monthly ion concentration (volume-weighted from weekly samples) of streamwater of watersheds A1, A2, F1, and F2.

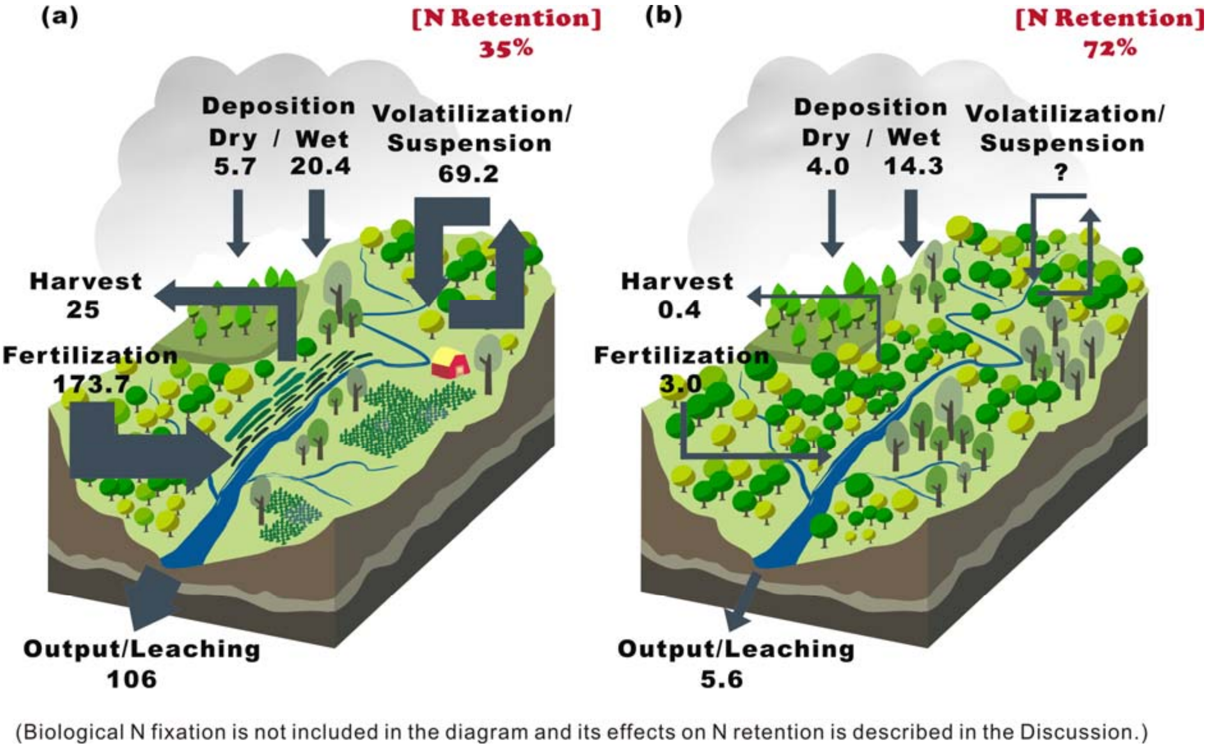
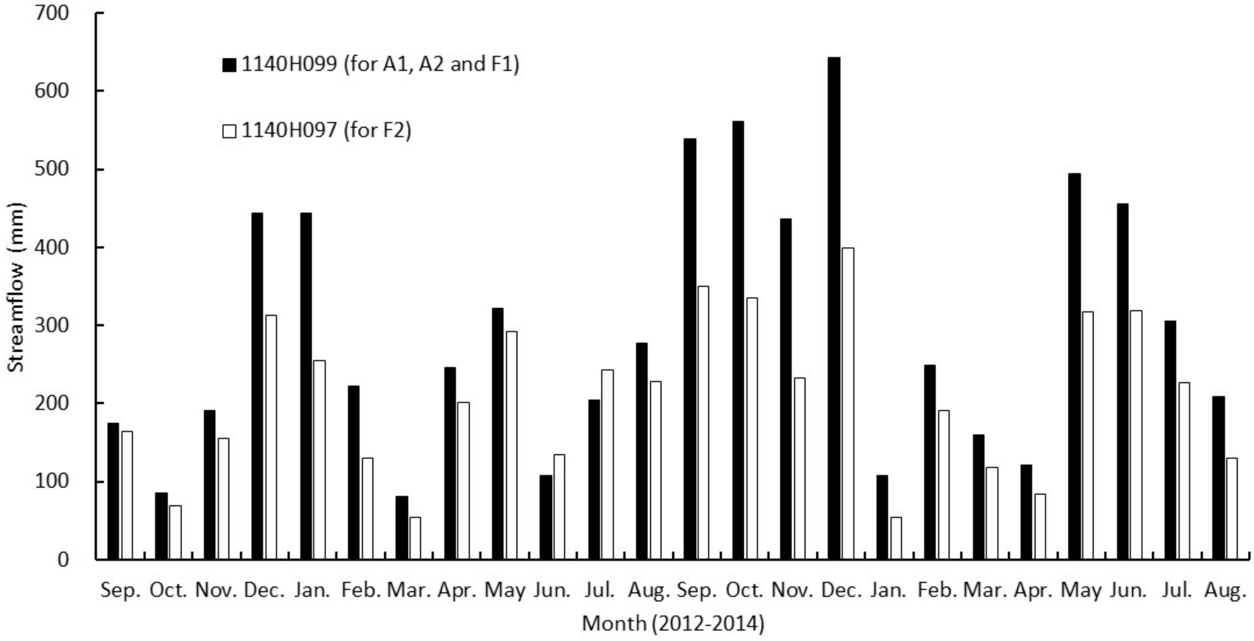


Figure 4. Schematic diagram of N fluxes of watersheds A1 and F2. A1 represents a watershed with 22% agricultural lands and 68% forests (a); F2 represents a watershed with 0.38% agricultural lands and 99% forests (b).

A



B

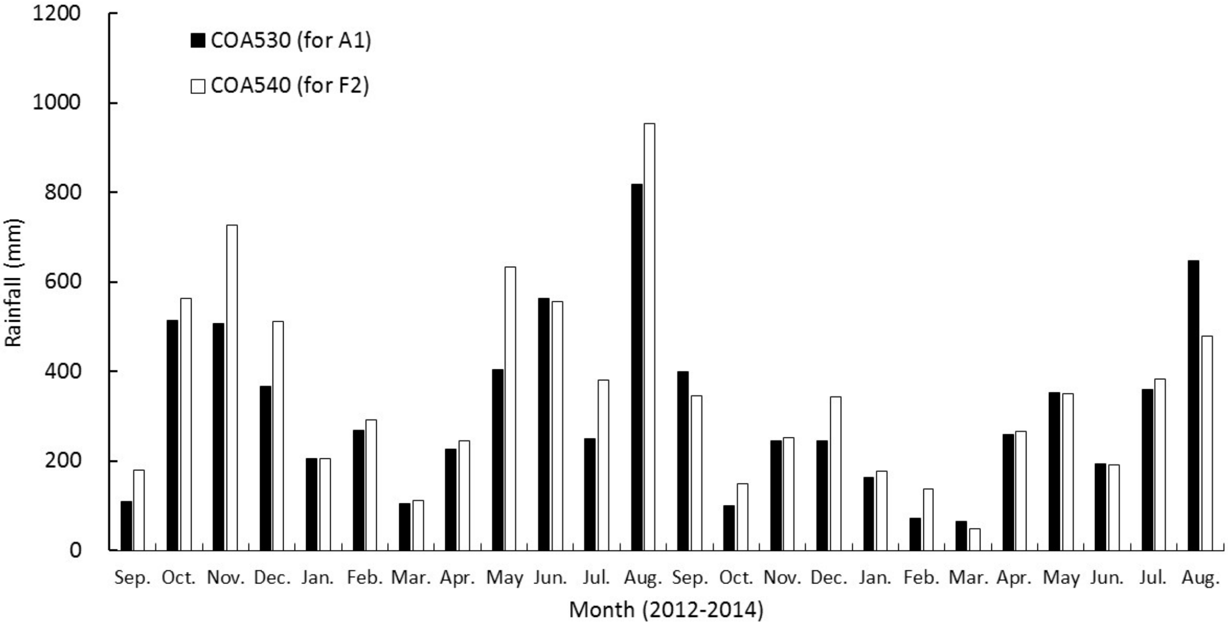


Fig. S1. Monthly streamflow (A) and rainfall (B) and streamflow of discharge gauges and rain gauges used in the study.