

Interactive comment on “Effects of different N sources on riverine DIN export and retention in subtropical high-standing island, Taiwan” by J.-C. Huang et al.

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Received and published: 14 January 2016

This paper addresses an important topic of growing concern and interest, the fate of N in watersheds in southeast Asia. The paper is well organized, but needs further editing for English usage. In general the discussion does not provide enough information for the reader (or the authors) to make an informed interpretation of the differences in DIN export among the watershed types. For example, there is no discussion of the sources of wastewater from highly developed lands, are there modern wastewater treatment facilities? Are there areas of septic systems? Are there areas with no treatment of wastewater? What types of wastewater treatment are used and how much do they

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vary among the watersheds? While I understand the authors are presenting a large scale analysis of N input and export from a number of watersheds across Taiwan, a more thorough investigation of the three broad N sources presented would improve the paper. The different types of agriculture are presented with estimates of N fertilizer use, although it is unclear how those estimates are converted into N input estimates on a watershed scale. A similar investigation of the potential N inputs from the highly populated areas seems appropriate and would improve the discussion. In addition, it would be useful to include a short discussion of the forest history in these watersheds. Are the forests in the moderately and highly disturbed watersheds similar to those in the low disturbed watersheds? If so it would be interesting to note that even with the same retention capacity the moderately and highly disturbed watersheds have completely overwhelmed that capacity. DIN export is estimated based on very few samples which may be one of the reasons the results from this study do not agree with early studies. While the author's explanation of why these results differ may also be true it is very difficult to make the determination based on export calculated with quarterly nitrate samples and monthly ammonium samples. I suspect storm runoff plays an important role in N transport in these watersheds, were any of the samples collected during storms? How well did the sampling strategy capture the range in flow conditions? Furthermore, there is no discussion of organic nitrogen. While the focus of the paper is DIN, I suspect DON is a large contributor to total N export in this region. In general the paper is lacking information for many important components of N inputs and exports and as a result it is difficult to evaluate the accuracy of the conclusions. Reply: It's good to receive the constructive comments which certainly help to improve this study and elevate the scientific merit in global syntheses of N export. We summarize the comments into five main points: (1) addressing the sources of the wastewater from the highly impacted watersheds, (2) the estimation of N inputs in relation to agriculture and population, (3) adding a short discussion of the forest history, (4) the effects of sampling frequency and storm event in flux estimation. (5) the proportion of DON in the total riverine N export in the watersheds. Below are our point-by-point reply.

Comment (1): In Taiwan, the septic system is a basic infrastructure island-wide, even in the mountainous tribes and villages. For treatment facilities, according to the thorough investigation in 2010 (<http://sewagework.cpami.gov.tw/>), there are total 49 wastewater treatment centers with daily capacity of ~ 2.3 million tons in Taiwan. Most of them are secondary treatments that remove 80% NH_4 but NO_3 removal is not required. Thus, the average N removal efficiency is $\sim 50\%$. Although only 20% of NH_4 left in the water after treatment, our previous studies still found extremely high NH_4 concentration in the urban drainage systems (Lee et al., 2014), due to the low installation rate of the sewage system. The installation rate in Taipei and Kaohsiung (the largest two cities in Taiwan) are ~ 70 and 30% of the household. Therefore, the NH_4 concentration over 300 μM is commonly found in the ditches close to the streams in downstream. The distribution of the treatment centers and their treatment capacity are shown in supplementary Fig. 1. As expected, most of the treatment centers are located in the cities with only a few in the mountainous region. We added these descriptions when we classified the watersheds.

Lee, T.Y., Shih, Y.T., Huang, J.C., Kao, S.J., Shiah, F.K., Liu, K.K. (2014) Speciation and dynamics of dissolved inorganic nitrogen export in the Danshui River, Taiwan, *Biogeosciences*, doi:10.5194/bg-11-5307-2014.

Comment (2): We estimated N export from fertilizer and human waste to each watershed in the revision. First, we established a look-up table with crop type and fertilizer amount, as partly shown in Table 1. Second, we used the individual watershed polygon to clip the land cover layer (map) and then multiple the individual crop area within each watershed with fertilizer amount to estimate the fertilizers used in each watershed. Similarly, we calculated the population density for the each county from census and used watershed polygon to clip the population density layer. The population density multiplying the county area within the watershed would give the population density for the individual watershed.

Comment (3): Approximately 58% of the land area of Taiwan is covered by forests.

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Prior to 1980s, forests are under extensive exploitation island-wide, only forests in very high elevations with very limited accessibility were not heavily deforested. The large scale deforestation led to serious consequences in soil and water conservation including many catastrophic landslides and debris flows. Then, due to public pressure, deforestation largely stopped in late 1980 and was entirely prohibited since 1991. Many low- to mid-elevation forests are undergoing secondary growth. In general, the NPP (net primary productivity) and nitrogen uptake in temperate forest vary from 1000-4000 g-DM m⁻²yr⁻¹ and 5000-25000 kg-N km⁻²yr⁻¹ (5-25 g-N m⁻²yr⁻¹) with good relationship (Finzi et al., 2007). Since the NPP in Taiwan forest is ~8000 g-DM m⁻²yr⁻¹ and the forest is still growing, the N uptake in subtropical should be high and thus likely responsible for N retention, particularly in the low-disturbed watersheds. We added the above description in the discussion section for clarification.

Finzi, A.C., Norby, R.J., Calfapietra, C., Gallet-Budynek, A., Gielen, B., Holmes, W.E., Hoosbeek, M.R., Iversen, C.M., Jackson, R.B., Kubiske, M.E., Ledford, J., Liberloo, M., Oren, R., Polle, A., Pritchard, S., Zak, D.R., Schlesinger, W.H., Ceulemans, R.: Increases in nitrogen uptake rather than nitrogen-use efficiency support higher rates of temperate forest productivity under elevated CO₂, PNAS vol 104(35): 14014-14019, doi:10.1073/pnas.0706518104.

Comment (4): This comment also raised by another reviewer. Sampling frequency is indeed an important issue for calculating flux, particularly for small mountainous rivers in which the storm discharge variation can surge to 2 or 3 orders of magnitude, compared to the low flows. In fact, we addressed this issue in our previous studies in several mountainous headwater catchments and a nested watershed in central and northern Taiwan (Huang et al., 2012; Lee et al., 2013; Lee et al., 2014; Lin et al., 2015; Shih et al., revised). In those studies, we even did some high-frequency sampling works (every three hours) during typhoons. We found that the relationship between nitrate concentration and streamflow varied from hydrological enhancement to dilution with the urbanization gradient, but most watersheds showed hydrological control over

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nitrate loading. It means that the nitrate loading could be well estimated by streamflow (based on 3- or 7-day sampling frequency). We also found that the proportion of nitrate loading during storm events is approximately similar with the proportion of streamflow on an annual base (Lee et al., 2013). Moreover, in our previous study in Danshuei River (one of the largest rivers in Taiwan), we calculated the ratios of NH_4 : NO_3 from weighted concentration of 6 sub-watersheds with weekly sampling scheme and the pattern was similar to our figure 6 (Lee et al., 2014). However, we agreed that the quarterly sampling per year is not ideal for nitrate loading and we addressed the point (by adding the information described in this paragraph) in the revised result and discussion.

Huang, J.C.*, Lee, T.Y., Kao, S.J., Hsu, S.C., Lin, H.J., Peng, T.R. (2012) Land use effect and hydrological control on nitrate yield in subtropical mountainous watersheds, *Hydrology and Earth Systems Sciences*, 16 (3): 699-714, doi:10.5194/hess-16-699-2012. 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. Lee, T.Y., Shih, Y.T., Huang, J.C., Kao, S.J., Shiah, F.K., Liu, K.K. (2014) Speciation and dynamics of dissolved inorganic nitrogen export in the Danshui River, Taiwan, *Biogeosciences*, doi:10.5194/bg-11-5307-2014. Lin, T.C., Shaner, P.-J. L., Wang, L.-J., Shih, Y.-T., Wang, C.-P., Huang, G.-H., Huang, J.C.* (2015) Effects of mountain tea plantations on nutrient cycling at upstream watersheds, *Hydrology and Earth System Sciences*, 19, 4493-4504, doi: 10.5194/hess-19-4493-2015.

Comment (5): A similar comment also raised by another reviewer who asked us to describe the role of DON and PN in Taiwanese rivers. Below is the response of this comment. First, as our title indicates that this study focused on the riverine DIN export not a comprehensive N budget. We focused on DIN because it is an important indicator of water quality and comprises the majority of total riverine nitrogen in both Taiwan and the world (Galloway et al., 2004; McCrackin et al., 2014). In our previous works

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and some unpublished data sets, we found that DON accounts for less than 20% of the total dissolved nitrogen in many highly-disturbed watersheds (Lee et al., 2014) and in upstream watersheds, it is less than 5% due to very low DOM in the lotic streams. Because the proportion of DON is not significant for total riverine N, we focused on DIN in this study. For PN, storms transport a large quantity of sediments during high flows in the steep landscape. We agreed that the sediments should contain considerable PN, but the majority of the sediments come from landslides which are not directly caused by human activities (Huang et al., 2012). In fact, we have some observations along a tributary in Danshuei River, which is featured by the urbanization gradient from upstream to downstream. The observations showed that the PN concentration in upstream and downstream are ~ 4.67 μM and 32.51 μM , respectively which is less than 10% of DIN in normal flow regime. We did a PN sampling during a rainstorm along a river system and found that PN concentration in downstream sites reached ~ 60.28 μM (Huang unpublished data). However most storms only lasted one to a few days. Thus, our focus on riverine DIN export associated with human activities should not lead to a biased understanding of patterns of overall nitrogen export. Yet, we agreed that the role of DON and PN would be a good next step in our study of N cycling in Taiwan. We included this response with the cited references in the “Discussion” section of our revised manuscript.

Lin, C.H. (2015) Research on biogeochemical condition in Danshuei River midstream and downstream with observation and 1-D advection-diffusion-reaction model simulation (Master dissertation). Graduate Institute of Hydrological and Oceanic Science, National Central University, Zhongli District, Taoyuan, Taiwan. McCrackin, M.L., Harrison, J.A., Compton, J.E. (2014) Factors influencing export of dissolved inorganic nitrogen by major rivers: A new, seasonal, spatially explicit, global model, *Global Biogeochemical Cycles*, 28, doi:10.1002/2013GB004723. Galloway, J.N. et al. (2004) Nitrogen cycles: Past, present, and future. *Biogeochemistry*, 70(2): 153-226.

Specific Comments: Abstract: The abstract summarizes the paper well, but needs

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further editing. Line 15: the term “buffered” is not the best choice because of its association with acid-base chemistry, however the whole sentence needs editing – I suggest “The low-disturbance watersheds had high N retention capacity with an export ratio of 0.06~0.18 despite high N input.” Reply: Thanks for the revised sentence. We changed accordingly.

Line 22: a less general term than “built-up lands” would be helpful here: residential, urban, industrial - something that gives the reader a better sense of the type of development. Reply: We added “(e.g., residential, urban, industrial)” in parenthesis following the built-up lands for clarification.

Line 28: I am not sure what is meant by sewerage systems, does it mean septic systems or a specific type of wastewater treatment facility or is it referring to a type of industrial wastewater treatment? Reply: We replaced the “wastewater treatment capacity or sewerage systems” with “the treatment efficiency of N removal and a more complete installation of household septic”.

Introduction: The Introduction provides a good review of pertinent literature. Page 16400 Line 12: I suggest “sources” or “drivers” rather than “indicators” Reply: We used “sources” to replace the “indicators”.

Page 16400 Line 12: “Levels of proportional riverine DIN export” I think this refers to DIN export ratio? I suggest “N retention and transfer processes control the amount of DIN exported from a watershed, typically less than the total watershed N input. Reply: Changed accordingly.

Page 16400 Line 15: Replace “over” with “divided by” Reply: Changed accordingly.

Methods Page 16402 Lines 10-12: In Table 1 the authors indicate that the fertilizer application rates are prescribed by the COA (please spell out all abbreviations). This should be indicated in the text rather than describing the amounts as crop “needs”. Reply: Rephrased and put the formal name of COA (Council of Agriculture, Taiwan) in

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the text.

Page 16404 Lines 5-6: Please provide more information about how dry deposition was estimated. Citing unpublished data is inadequate. We need to know how these percentages were derived. If the method and data used are described they will no longer be unpublished and can be cited in the future. Reply: We added it accordingly.

Page 16404 Lines 18-20: How were rating curves developed, there is no mention of discharge measurements related to the stream stage measurements? The cross-section geometry and stage height does not produce a discharge. Reply: We rephrased this sentence to: 'The WRA crew measured the stream cross sections two or three times per year. Based on the cross section, the flow velocity of each sub-cross section was measured and then integrated to obtain the discharge several times. The observed discharge with the water levels can develop the rating curve for estimating the discharge via the monitoring water level.'

Page 16404 Lines 24-25: Stating that laboratories followed standard operating procedures is inadequate. There are only 3 analytical types to describe and they should be described briefly. Quarterly sampling is a very coarse basis for N export calculations. Please describe the limitations of the dataset and the possible error introduced from using such infrequent sampling. Reply: We revised the comment based on the main comment (4) that we mentioned earlier.

Results and Discussion Page 16406 Lines 23-25: Aren't there other options besides population density control, what about better wastewater treatment facilities? Reply: We agreed with the reviewer and added wastewater treatment into the sentence.

Page 16407 Lines 20-24: Given that runoff is the primary multiplier in export calculations it is not surprising that export and runoff are strongly related. Reply: In general, this comment is right when the relationship between concentration and discharge is coherent enough. The word, "predominantly", is a little too strong. We replaced it to "plays an important role on DIN export".

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Page 16407: It is not at all surprising that there is not a strong relation between Agriculture and DIN export at a global scale. All agriculture is not created equal in terms of N export and as the authors point out total runoff and runoff ratios vary widely across the globe. Reply: We wonder what the reviewer really meant 16408 L12-14 in which we agreed with the reviewer and “may seem surprising’ to ‘could be expected”.

Page 16408 Line 3: “. . .even at the global scale. . .” should be “. . .even when compared to the global scale. . .” Reply: Rephrased accordingly.

Page 16409 Line 7: Typo at the end of the line, should read “greater than” not “greater and” Reply: We checked this sentence and it is correct.

Page 16410 Lines 2-3: Again this discussion would benefit from a more thorough description of the types of human emissions in these watersheds. Are these mainly from wastewater treatment plants? Are there septic systems? Raw sewage inputs? Reply: We re-wrote this paragraph according to the reply of main comment (1).

Page 16412 Line 3: What about the type of agriculture? There is more bamboo and paddy in the moderately disturbed watersheds as well. Reply: Yes, the crop types are different among regions. Most paddy applied fertilizers heavily is in the highly-disturbed watersheds. We added this description in the revised text for clarification.

Page 16412 Lines 7-8: To what types of land management are the authors referring? Changes in land use? Fertilizer application rates? Erosion control? Are there some types of land management that could provide more benefit than others? Reply: Yes, there are many land management practices which can reduce the DIN export but we do not have empirical evidences to prefer any particular practice to others. Therefore, we added “(e.g., erosion control, precision fertilization)” following “management efforts”.

Page 16412 Line 10: What are constructed lands? This sentence is hard to follow, perhaps it can be restructured for better clarity. Reply: To make it clear we added “(buildings, houses, roads etc)” in parenthesis following constructed land.

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Page 16412 Lines 23-25: While this statement is likely true the study presents no data regarding the sewage systems or N fertilizer application methods to support this conclusion. Reply: We rephrased this paragraph according to the main comment 1. Please see our response to the comment.

Concluding remarks Page 16413: “Since the runoff varied only two fold and atmospheric deposition varied less than 30%, the ~ 10 times greater DIN export of highly disturbed watershed (8000 kgNkm⁻² yr⁻¹), compared to low disturbed watersheds (900 kgNkm⁻² yr⁻¹) likely resulted from differences in inputs from agricultural lands, total human emission and watershed N retention capacity.” With some additional work this study could provide more quantitative conclusions relating the relative contributions of agriculture, human emissions, and DIN retention. Reply: We agree that with additional data and work it is possible to provide more quantitative conclusions. The additional data and work is not minor and could be a study by itself. We think the results in this study deliver important information at its current state.

Page 16413 Lines 15-16: Problems with coastal and downstream eutrophication are not discussed in the paper. If there are such problems they should be discussed in the introduction. Reply: Coastal eutrophication is common in places with high N (and P) input and our results do show very high N export from downstream watersheds. Thus, we think this remark is a legitimate one.

Page 16413 Lines 17-28: While I do not disagree with the final conclusions of the paper, they would be more convincing with a more detailed investigation of the human inputs. What specific sewage treatment methods need improving? How would the authors expect such improvements to compare to reductions in fertilizer application rates? Given the plausible reductions in both of those inputs what impact could be expected given the extremely high rates of DIN export? Reply: While we think the improving methods provided by the reviewer are all great measures we do not feel that we have enough detail data to prefer one to another. Thus, rather than proposing any specific improving method, we highlight the consequences of such high DIN export and

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the needs for improving N removal.

Tables: Table 2: If these values are means for all watersheds in each disturbance category that should be indicated in the caption. Reply: We addressed the mean values in the caption.

Table 3: A more explicit description of developed land use is required. Built-up is too general. Please break down the percentages of residential, urban, and industrial land use or some other similar breakdown. Reply: Because we did not distinguish different types of build-up in our analysis so that we think it is not necessary to break it down to different types but we do have data that could be added as a supplemental table.

Figures: All figures are very well-drafted and easy to read. Figures 1 and 2: The maps are particularly well drafted. Reply: Thank you.

Figure 3. Please include the time period for which the means apply. Reply: We put the time period in the revised figure 3.

Interactive comment on Biogeosciences Discuss., 12, 16397, 2015.

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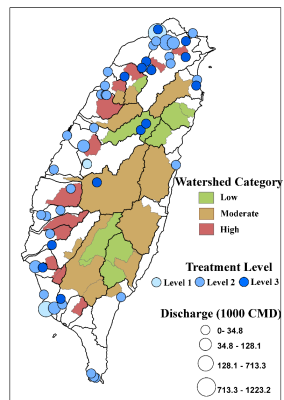
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Supplementary Fig. 1 The distribution of water treatment centers in Taiwan.

Fig. 1. supplementary figure 1

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