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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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The study by Huang et al. quantifies DIN fluxes from Taiwanese watersheds across a range of land use, population, and Nitrogen input rates. The results are compared extensively to other world watersheds reported in the literature. The watersheds of Taiwan have greater precipitation and N input rates than is typical in global syntheses, so studying these watersheds is a good rationale for expanding the global response surface for looking at N retention capacity. They find that watershed N retention declines with increasing N loading (or impact), and that much of the decline is due to increasing NH4 exports. The Taiwanese watersheds have a much higher proportion

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of ammonium export than other world watersheds, especially in the impacted watersheds. Overall this is a good paper, but I am unsure that the export flux estimates are robust. Whereas the ammonium concentrations were estimated monthly, nitrate concentrations were only measured 4x per year. This is very infrequent, and may miss many of the storm events, when concentrations are often quite dynamic compared to base flow. This is particularly true in impacted watersheds (both urban and agricultural). I assume many of the nitrate measurements were collected during relatively lower flows since these are more frequent. If concentrations dilute during storms (as is common for DIN in many agricultural systems), this would be an overestimate of N exports. It will be impossible to address this with the data in hand. However, this must be evaluated (e.g. what is the mean flow during sample periods compared to annual mean flow? What are flow weighted concentrations?), and then discussed for each N form. Perhaps there are some estimates of storm event nutrient samples that can be used to discuss this issue as well. The limited nutrient sampling may also contribute to the patterns in NH4:NO3 in Taiwan compared to other world rivers. Error in this ratio is likely to be greatest in more disturbed watersheds due to dynamic flow and concentration patterns. This also needs some discussion. Further DIN alone does not represent the N export budget. DON and PON are likely also important, the latter particularly in Taiwan with high flows, large storm events, and steep slopes. These are not considered at all. Part of the difference might be due to the relative importance of organic vs. inorganic forms across watersheds. Discussion about how this may influence the results is also needed. The method for estimating fertilizer and human waste loading to each watershed is not described. While N deposition estimates are likely fairly robust, these other loads are not as easy to obtain, and so should be included. It is especially a problem when scaling to watershed boundaries, which likely differ in scale from where the data to estimate loads comes from. I also think there needs to be more discussion on why export ratio is higher in impacted watersheds. Discussion (e.g. 16412.8-11) doesn't discuss why change in population or land cover result in lower retention rates. Why are these watersheds at more advances stage of N excess. Mechanism of in-

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creasing rainfall would also affect low impact watersheds, so this alone not a reason. Is population the best indicator to classify watershed impact? It seems ag land cover would be better, given the sensitivity to this. The writing overall is very understandable, but the text still needs a good edit.

Reply: We are grateful for the professional comments raised by the reviewer, which made us think more deeply about our study. Through taking the comments into our revision we believe that our revised manuscript is more focused and more readable. In summary, there are 5 concerns in this review report. They are: (1) is sampling frequency sufficient to estimate the 'representative' flux, particularly for nitrate; (2) The role of DON and PN in Taiwanese rivers; (3) Adding clear descriptions of the estimation of fertilizer and human waste loading to each watershed; (4) Explain why the export ratio is higher in impacted watersheds; (5) why population is chosen to classify watersheds. Below is our point-to-point reply.

Comment (1): Sampling frequency is 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. We appreciated the reviewer's suggestion for comparing mean flow during sample periods with annual mean flow and for the use of the flow weighted concentrations to evaluate the validity of our sampling frequency. In the revised manuscript, we added this in a supplementary table. 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 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 NH4: NO3 from weighted concentration of 6 sub-watersheds with weekly sampling scheme and the pattern was similar to our figure 6 (Lee et al., 2014). Thus, the high ratio of NH4: NO3 in the highly-disturbed watersheds we found in this study should be real. 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 (2): The reviewer asked us to describe the role of DON and PN in Taiwanese rivers. 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 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 uM and 32.51 uM, 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 uM (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.

Comment (3): 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. Secondary, we used the individual watershed polygon to clip the land cover layer (map) and then multiple the individual crop area within

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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 (4): The high export ratio for the highly disturbed watershed is indeed striking and there are two possible causes. First, most of the high DIN runoff from urban entering streams may be due to the limited wastewater treatment and incomplete sewer drainage system which cannot effectively handle the large quantity of DIN emitted by the large population. For treatment facilities in Taiwan (http://sewagework.cpami.gov.tw/), the average N removal efficiency is ∼50% and the installation rate in Taipei and Kaohsiung (the largest two cities in Taiwan) are ∼70 and 30% of the household. Extremely high NH4 concentration, over 300 uM, in the urban drainage systems (Lee et al., 2014) indicating the inadequateness of the water treatment systems. This is likely also the reason for the very high NH4 concentration in downstream or highly-disturbed watersheds. Second, the farms close to the urban are more heavily fertilized than those in the mountains. For example, most paddy fields are located in the downstream and most of them were very heavily fertilized compared to the farms in the mountainous region. Thus, considering the urban locality, N excess is more likely to occur in the downstream watersheds.

Comment (5): Both population and agriculture land cover are good for classifying the watershed impacts. Agriculture land cover under similar environmental settings and fertilizer practices is a good indicator for non-point pollution as we showed in Figure 4. However, our results (see Fig. 5) clearly show that the changes in riverine DIN export is more closely related to population density than to agriculture land cover at broader scales (as reported in some global syntheses). Thus, we chose to use population for classification watershed impacts.

Specific comments 16400-21 Most watershed N retention is not in rivers, but in the terrestrial part of the watershed. Reply: Yes. The sentence '. . .or denitrified in large rivers. . ..' is not clear. We changed 'in large rivers' to 'in the large river basins'.

16402.26. What is mean annual precip? Reply: We added the mean annual precipitation "between 2000-4000 mm/yr" to the sentence.

16403. 12. Change to N loading (deposition usually refers to atmospheric only). Reply: Corrected it accordingly.

16406.3. NH4 retention capacity is really also nitrification capacity, i.e. NH4 converted to NO3 and exported in this form Reply: The inference, 'indicating the saturation of NH4 retention capacity at highly disturbed watersheds' maybe one step too far. We rephrased it to 'indicating that most of the NH4 may come from domestic wastewater'.

16407.24 Runoff cannot be only factor controlling DIN export, because Taiwan N exports seem to be disproportionately high, perhaps due to differences in inputs mentioned. Reply: The word, 'predominantly', is too strong. We changed it to 'plays an import role on DIN export'.

16408.19-24. I don't understand the example. Wouldn't 100% rice field in N.A. be the same as âĹij66% rice cover in Taiwan? Reply: Because rice is harvested twice a year with vegetables in between so that the amount of N fertilizers applied is at least 418 kg ha-1 yr-1 as listed in FAO (2002). In North America rice is harvested once a year with the application of N fertilizers at 135 kg ha-1 yr-1 so that 100% rice cover in North America would be ∼32% rice cover in Taiwan. We corrected it (32% instead of 10%).

16409.2 Why compare NH4 flux to global NO3 flux? This sentence is unclear. I think you are saying the NH4 is much more predominant in Taiwan watersheds compared to the rest of the worlds rivers. Please rephrase. Reply: Thanks for the correction, we rephrased it accordingly.

16409.9 NH4 is not volatilized (NH3 is). Once dissolved in water NH4 is not volatilized. Note that NH4 is oxidized to NO3, so something in Taiwan seems to be limiting this process. Reply: Thanks for the correction, we removed “or volatilized” from the sentence.

16409.19. Sentence is contradictory. If not very low, then DO demand is not high. Nitrification can still occur at lower DO levels than what is low in Taiwan. Reply: The sentence, “the high DO demand. . .”, may not be not clear. We rephrased to, “the relatively low DO implies that nitrification maybe substantial in the highly-disturbed streams.”

16409.21-30. If residence time is low, then removal cannot explain low NH4 in the low and moderately disturbed watersheds. Explanations need to be consistent. Reply: We removed the sentence, ‘Water residence time in Taiwan. . ...’ and added the following sentence, ‘Because the highly-disturbed watersheds were characterized by dense population, the domestic wastewater with abundant NH4 due to the limited treatment facilities and incomplete sewer drainage system could be the main source of riverine DIN.’ The last sentence in this paragraph was also modified to ‘the high human emission and the insufficient transformation result in the dominance of NH4 downstream.’

16411.2 Change irrelevant to “Small compared to. . ..” Reply: Changed accordingly.

16411.24. But the N retention capacity of regrowing forest is finite. Once mature, they will be N saturated, and become leakier, according to Aber et al. N saturation hypothesis. Please add some discussion of this. Reply: We agree that once a forest matures it would have limited capacity to take up nitrogen. The N saturation hypothesis proposed by Aber (1989, 1998) emphasizes on the gradual N saturation when a forest is exposed to high N deposition it is not so much about forest age although age certainly matters. In fact, the N saturation hypothesis implies that even mature forest could benefit from N addition when first exposed to N addition and this N fertilization effect has been an important issue in global change ecology. The current concern is that the effect of increased carbon dioxide on enhancing forest growth is largely dependent on nitrogen availability. To avoid the confusion of N saturation and age-dependent nutrient uptake, we modified the sentence to “If forest growth is indeed key to the low DIN export and export ratio, then reforestation could be an effective management practice to reduce DIN export and therefore reduce the risk of downstream eutrophication, especially if the trees are regularly harvested before the forest matures and the ability to take up nutrients declines."

16413.16. Mention of dry year comes out of the blue here. No mention of climate variability earlier, and I don' think the statement is true. During dry year, nutrient fluxes from watersheds will also be much lower, whereas residence time of coastal systems will not change as much since they may be tide dominated. Reply: We eliminated the statement, '…especially in dry years when….'. Besides, we excluded the tidal channel when we selected the watersheds so that tidal effect is minimal in the selected watersheds. This information is added to our revised manuscript.

Figure1. No legend for populations. Reply: We added the legend.

Table 5 does not match the presentation of results in section 3.2 (for fertilizer and human emission) or totals in 3.2. Puts doubt into other results. Reply: What we showed in section 3.2 are the N input amount per percent of fertilizer and human emission. The fertilizer amounts are 2596, 4190, and 10596 kg-N/km-2 yr-1 (from Table 5) and the percentages of agricultural land (from Table 3) are 11, 16, and 38% for the low-, moderately-, and highly-disturbed watersheds, respectively. Thus, the fertilizer amounts in each percent of agricultural land are 236, 262, and 279 kg-N km-2 yr-1 for the low-, moderately-, and highly-disturbed watersheds. We did the similar way to human emission.

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

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