

## ***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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W. Zhang This paper has two merits: The first one is to provide information about one of the highly populated watersheds in the world, with a tremendous level of N contamination. The fact that about 30%-50% of human-induced N would be ended in rivers. That is extremely interesting, given that different findings have been achieved when we compared with similar research in other watersheds. The second one is that the authors can enhance our understanding of factors controlling riverine N exports through the comparison among different watershed groups. Overall, I enjoyed reading this paper and believe that it will make a nice contribution to Biogeosciences.

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Reply: We appreciate that the reviewer recognized the merits of our study. Indeed, it's very surprising that the low-disturbed (semi-pristine) watersheds can retain most of the nitrogen, even when the atmospheric N input reaches as high as 2000 kg-N km<sup>-2</sup>yr<sup>-1</sup>. For moderately-disturbed (agricultural-dominated) watersheds, the intensive fertilizer application which is common in Southeast Asia is very likely a challenging issue in the near future. The good relationship between agricultural area and DIN export, comparing with the other watersheds in the world, clearly indicates that the non-point source pollution is more severe in this than other regions. The extremely high riverine DIN export and the high NH<sub>4</sub>:NO<sub>3</sub> ratio in the highly-disturbed watersheds indicate that the limited wastewater treatment and incomplete sewer drainage system cannot effectively remove DIN from the water and thus pose the potential risk of eutrophication.

However, the methodology used in the study is a little bit different from previous studies, although the discussion is sufficient and the conclusion is noteworthy. In this study, atmospheric N deposition, fertilizer N and human emission was summed as total N input. But this method could be subjected to high error. The commonly used method is NANI methodology, which was proposed by Howarth et al., (1996). NANI has been widely accepted as almost complete inventory for calculating human-induced N. NANI sums N contributions from atmospheric deposition, fertilizer application, agricultural biological fixation, and net import/export of N in food and feed to a watershed. To me, I think your N inventory is incomplete, and hence calculated N input could be underestimated. If N accounting method of this paper is quite different from other studies, how much confidence do we have with the extremely high value of 30%-50%? I would not prefer to argue whether your methodology is suitable or not, but more discusses on the method should be guaranteed. Below, I provide some suggestions for your further consideration: (1) I cannot quite understand why you exclude N inputs of biological N fixation, food and feed imports and/or livestock excretion (usually, livestock N excretion was incorporated with the estimate of food & feed imports). Can you explain more on this? I can give you more evidence for your further consideration: (a) As you have mentioned in the paper, many of these watersheds are dominated by forestland and/or

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cropland. The land cover type is a little bit similar to northeastern U.S.A.. Boyer et al. (2002) had addressed that biological N fixation could be as high as 30% of total N inputs. However, one should be cautious because of the high uncertainties in the estimate of biological N fixation (Sobota et al., 2013). But this really implies that biological N fixation cannot be just omitted. (b) As a curious idea, I have checked the imported food from other countries in Taiwan. High amount of food (e.g., more than 1 million tons of wheat) was imported annually. In part, this number addresses that N inputs through this source should be significant. You can refer: <http://faostat3.fao.org/browse/T/TP/E>. More evidence could be seen in other similar watersheds. For example, in Huai River Basin of P.R. China (Zhang et al., 2015), which is also highly populated watershed, about 70% of land cover in this watershed is cropland. Even so, this watershed was still relied on food and feed import. Hence, I believe food and feed imported N may be also significant in Taiwan. (c) You should mention more on why you exclude feed N (i.e., livestock excretion N). The number is expected to be very small? Can you provide more evidence?

Reply: We are glad to receive the professional comments raised by the reviewer, which would certainly help us to improve the discussion and strengthen our conclusion. In this review report, the reviewer proposed to use the NANI approach (or model) to complete the N budget, particularly for biological N fixation and net import/export of N in food and feed. First of all, there are many modeling approaches focusing on the N budget at watershed scale, such as NANI, GLOBAL\_NEWs, and SPRROW etc. Modeling the N budget is the next step in our study in this subject. However, here we focused on the data compilation and investigation of factors controlling DIN export in the subtropical mountainous region which should be highlighted in global syntheses. For N inputs, it is well recognized that atmospheric N deposition, natural biological N fixation, fertilizer application/agricultural biological fixation, human emission, and livestock excretion are the main N sources. Basically, NANI sums human emission and livestock excretion in the calculation of net import/export of N in food and feed. Therefore, the reviewer suggested us to elaborate the biological N fixation, net import/export of N in food and

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feed, respectively. Below, we addressed the 3 points raised by reviewer.

For biological N fixation (BNF): There are two types of BNF: one is the agricultural BNF and the other is the natural BNF. As Boyer et al (2002) concluded the total BNF can contribute around 29% of the total N input ( $\sim 3088$  kg N km<sup>-2</sup> yr<sup>-1</sup>) in northeastern US with 24% from agricultural BNF and 5% from natural BNF. It means that the natural BNF in the northeastern US only contributes  $\sim 167$  kg N km<sup>-2</sup> yr<sup>-1</sup> which is quite low, compared to the total N input. In contrast, the reviewer's study found that the natural BNF contributes as high as 1500-2200 kg N km<sup>-2</sup> yr<sup>-1</sup>, but it only accounted for only 7% of the total N input. Although the proportions of the natural BNFs (5% and 7%) in the two regions are similar, the absolute values ( $\sim 167$  and 1500-2200 kg N km<sup>-2</sup> yr<sup>-1</sup>) varies larger than 10-fold. We think this is the reviewer's main concern. In global syntheses, natural BNF in forest ecosystems varies from 1600 in temperate region to 2500 kg N km<sup>-2</sup> yr<sup>-1</sup> in tropical region (Cleveland et al., 1999). However, the latest study showed that the BNF in tropical or subtropical zone is not significant. Most natural BNF in tropical forest may be less than 600 kg-N km<sup>2</sup> yr<sup>-1</sup>. It means that the previous studies may overestimate the natural BNF by more than 5 times (Sullivan et al., 2014). Using this new estimate, the natural BNF accounted for less than 5% of the total input in our study so that contribution from natural BNF was not included in our analysis. Agricultural BNF could be a significant N source; for example of Alfalfa (*Medicago sativa* L.), can fix more than 21800 kg-N km<sup>2</sup> yr<sup>-1</sup> (Mclsaac et al., 2002). In fact, most studies have found that the nitrogen from agricultural BNF is comparable with that from the fertilizers. So far, the data quality of agricultural BNF in Taiwan is fragmentary and we cannot confidently compile the data set for each crop. In order to avoid introducing more uncertainties, we assumed that the agricultural BNF can be replaced by the amount of synthetic fertilizers in our original calculation. We revised the statements in the corresponding text in section of methodology to clarify our calculation.

Sullivan, B.W., Smith, W.K., Townsend, A.R., Nasto, M.K., Reed, S.C., Chazdon, R.L., Cleveland, C.C., Spatially robust estimates of biological nitrogen (N) fixation imply sub-

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stantial human alteration of the tropical N cycle. PNAS, vol. 111(22): 8101-8106, 2014.

For Net N import/export: As the reviewer pointed out, food import/export is an important source for calculating N export in NANI approach. Taking net food flow into account is a good way for estimating human emission in country scale. However, it is very difficult to re-allocate or track the imported foods at to individual watersheds (Swaney et al., 2015). Moreover, food amount is a good surrogate of N export when mainly considering human physiologic responses, but is likely an incomplete estimate for export from human activities (e.g. vehicle combustion and energy consumption). The GDP-based approach proposed by Van Drecht et al. (2009) is also a reasonable alternative surrogate of human activities. We addressed this issue in the section of methodology for clarification.

Swaney, D.P., Hong, B., Selvam, A.P., Howarth, R.W., Ramesh, R., Purvaja, R.: Net anthropogenic nitrogen inputs and nitrogen fluxes from India watersheds: An initial assessment, Journal of Marine Systems 141: 45-58, 2015.

For Livestock excretion: We agreed that livestock excretion N is an important source in the temperate region where the cattle and swine are the main sources of protein, because per capita N excretion from livestock excretion is very high, particularly for cattle. Below, we listed the number of cattle, cow, and swine in Taiwan, US, and China to show why we did not include the livestock in this study (Table 1). From the table it is clear that the number and density of cattle and cows in Taiwan are much lower than in the other two countries. Due to the very small number and low density, we did not include this source of N in our calculation. Although the density of swine are much higher than the other two countries, it is important to note that  $\sim 2/3$  of the island is mountainous region that have very limited number of swine. Most pig farms are located in the west plain and regarded as a point source. Therefore, they are requested to set sewer system to treat wastewater before entering stream. Because most of the studied watersheds are in the mountainous region, the export from livestock excretion should

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be minimum and so is not included in our analysis. We addressed this point in the Method section and added the table in supplementary in the revision.

Additional Comment (2) About the analysis on the impacts of N inputs on DIN, some individual research on nitrate or ammonia could be helpful. I listed some of them for your consideration (please see below).

Reply: We appreciated the reviewer sharing the papers. We added them in our references and used them in the discussion section to clarify the speciation of the DIN in the three categories.

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/12/C9016/2016/bgd-12-C9016-2016-supplement.pdf>

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

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