



# Apportioning riverine DIN load to export coefficients of land uses in an urbanized watershed



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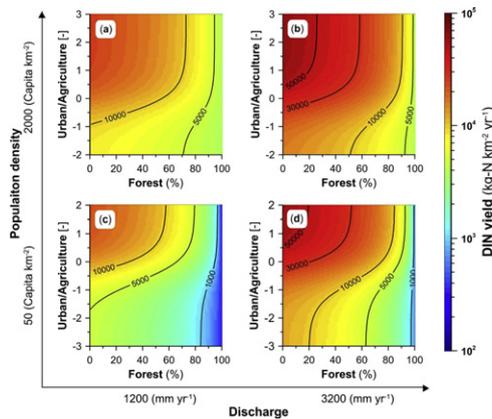
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## HIGHLIGHTS

- High forestry export in Taiwan is attributed to high rainfall and atmospheric N deposition.
- Both area and export coefficients are essential for assessing the riverine DIN export.
- Inclusion of urban export coefficient and treatment capacity can complete the human-associated wastewater.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The apportionment of riverine dissolved inorganic nitrogen (DIN) load to individual land use on a watershed scale demands the support of accurate DIN load estimation and differentiation of point and non-point sources, but both of them are rarely quantitatively determined in small montane watersheds. We introduced the Danshui River watershed of Taiwan, a mountainous urbanized watershed, to determine the export coefficients via a reverse Monte Carlo approach from riverine DIN load. The results showed that the dynamics of N fluctuation determines the load estimation method and sampling frequency. On a monthly sampling frequency basis, the average load estimation of the methods (GM, FW, and LI) outperformed that of individual method. Export coefficient analysis showed that the forest DIN yield of  $521.5 \text{ kg-N km}^{-2} \text{ yr}^{-1}$  was  $\sim 2.7$ -fold higher than the global riverine DIN yield (mainly from temperate large rivers with various land use compositions). Such a high yield was attributable to high rainfall and atmospheric N deposition. The export coefficient of agriculture was disproportionately larger than forest suggesting that a small replacement of forest to agriculture could lead to considerable change of DIN load. The analysis of differentiation between point and non-point sources showed that the untreated wastewater (non-point source), accounting for  $\sim 93\%$  of the total human-associated wastewater, resulted in a high export coefficient of urban. The inclusion of the treated and untreated wastewater completes the N budget of wastewater. The export coefficient approach serves well to assess the riverine DIN load and to improve the understanding of N cascade.

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## 1. Introduction

Huge amount of additive and reactive nitrogen (e.g., fertilizer, excrement and vehicle combustion) lost to the environment has resulted in water pollution and health issues (McKenzie and Townsend, 2007), such as eutrophication, estuary and coastal hypoxia (Downing et al., 1999; Parr and Mason, 2003) and cyanosis (low-oxygenated blood). This issue has fueled environmental concerns about the impact of broad-scale changes in land use on riverine N exports (e.g., Jones et al., 2001), particularly for regions stressed by growing population and agricultural activities, like Southeastern Asia and India. DIN (dissolved inorganic nitrogen, including  $\text{NO}_2$ ,  $\text{NO}_3$ , and  $\text{NH}_4$ ) often comprising the largest fraction of total riverine N (Galloway et al., 2004) is the most discussed N species. Thus, a large number of water quality models based on different theoretical frameworks, from simple export coefficient models, such as PLOAD (US EPA, 2001), and multiple regression models (Smith et al., 2005; Lee et al., 2014), such as SPARROW (Alexander et al., 2002), to complex mechanistic models, such as SWAT (Arnold et al., 1998), were proposed. However, those modeling works rely strongly on accurate DIN load estimations, reliable export coefficients of different land uses and the differentiation of point and non-point sources. Moreover, considerable regional variations also dampen the modeling works in global N synthesis (Howarth et al., 1996).

Taiwan, characterized by the high-standing montane landscape with abundant rainfall, has approximately 23 million people settled on it. Intensive agriculture systems widely spreading in the plain and hill areas are essential for supporting the packed population. In this environmental setting, the averaged riverine DIN yield of  $\sim 3800 \text{ kg-N km}^{-2} \text{ yr}^{-1}$  is approximately 18 times of the global average (Huang et al., 2016). However, the DIN concentration in Taiwan forest is comparable with the worldwide forest watersheds indicating disproportional export coefficients of forest, as well as other land uses. Besides, the downstream riverine DIN yield can reach over  $10,000 \text{ kg-N km}^{-2} \text{ yr}^{-1}$  showing the dominance of human emission in N loads (Lee et al., 2014). The importance of the DIN export in Southeastern Asia and the relation between agriculture and population were documented recently (Van Drecht et al., 2009; Lee et al., 2014); nevertheless, the quantitative determination of the export coefficients and the pathways of human emissions are rarely investigated.

Theoretically, the export coefficients, reversely determined by monitoring riverine loads from watersheds, represent average conditions of land uses (Bowes et al., 2008; Shrestha et al., 2008). Riverine substance load which fluctuates dynamically with flow regimes and biogeochemical processes has induced different methods for estimation (e.g., Birgand et al., 2010, 2011). The bias in load estimation associated with flow regimes and sampling frequencies should be determined for constraining the uncertainties (Lee et al., 2009; Huang et al., 2012). Even with an accurate load estimation, the proportion of point and non-point sources remains questionable. Conventionally, the point sources, accounting for 40% of the total pollutant to the ocean, globally (Seitzinger et al., 2005), were identified through considering the connection ratio and treatment efficiency of sewerage systems straightforwardly, but the identification lacked validation. Recently, the non-point sources in urban areas from the untreated wastewater was found important (Groffman et al., 2004; Fitzpatrick et al., 2004; Brown et al., 2009) underscoring the determination of non-point sources in urban areas can provide an independent validation in pollutant delivery.

In this study, the riverine DIN loads from 16 sub-watersheds in Danshui River of Taiwan supplemented with population census and land use were introduced. The objectives were to (a) evaluate the accuracy and precision of riverine DIN loads through different sampling frequencies and methods, (b) determine the export coefficients of land uses through Monte Carlo approach, and (c) estimate the proportion of the point and non-point sources from human-associated wastewater. This synthetic framework provided the first-order assessment of riverine DIN load change in the environment with dense population and

intensive agriculture. With the knowledge of regional export coefficients of specific N sources, the consequences of additive N inputs can be better understood.

## 2. Materials and methods

### 2.1. Characteristics of the study area

The Danshui River, one of the main rivers in Taiwan, has a drainage area of  $2726 \text{ km}^2$  and flows through Taipei City, which has an area of  $272 \text{ km}^2$  and 6.8 million inhabitants. The average population density in the Danshui River watershed and Taipei is  $\sim 2500$  and  $\sim 25,000 \text{ capita km}^{-2}$ , respectively. The Danshui River has three major tributaries from north to south (Fig. 1), namely, the Keelung (K), Sindian (S), and Dahan (D) Rivers, which merge in Taipei City. The annual rainfall and its wet season ratio were  $3750 \text{ mm}$  and  $54\%$  for (K),  $4236 \text{ mm}$  and  $65\%$  for (S), and  $2742 \text{ mm}$  and  $70\%$  for (D) during 2000–2010 with seasonal variation increasing southward. The north-easterly (October to February) of the East Asian Monsoon brings considerable precipitation to northern Taiwan, thus balancing the warm and wet summer and resulting in an indistinct seasonality in the Keelung River watershed.

NLSC (National Land Surveying and Mapping Center) compiled aerial photos, satellite imageries, and field surveys to identify 103 land use types for the entire island and then released the comprehensive land use dataset in 2006 (NLSC – National Land Surveying and Mapping Center, 2008). In this study, the 103 land use types were reclassified into six main categories: forest, urban, cropland, orchard, paddy, and water body. The forest category includes natural forest, secondary forest, bamboo, and shrubs. The agricultural land use includes three categories: paddy (0.5% of the whole Danshui basin), cropland (2.5%), and orchards (2.3%, mainly on hillsides of 100–1200 m a.s.l.). Human-made facilities, such as houses/business buildings (7.0%), roads (3.7%),

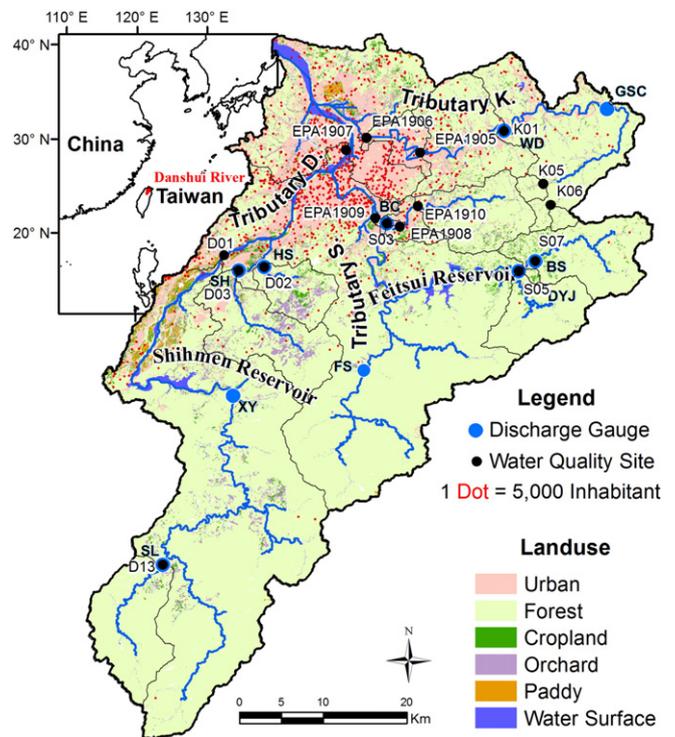


Fig. 1. Landscape of the Danshui River watershed, including land use pattern, population (red dots), and sampling sites. A total of 6.8 million inhabitants live in Taipei City. The blue and black dots represent the discharge gauge and water sampling sites, respectively.

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