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- Skewness index quantifies spatial distribution within watersheds

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- Figure S1

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An index to characterize the spatial distribution of land use within watersheds and implications for river network nutrient removal and export

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Abstract The spatial distribution of land use and associated nutrient inputs may influence the efficacy of in-stream nutrient removal; however, the effect of source location on N removal and watershed N export has not been quantified. We present the skewness index, a metric to quantify the spatial distribution of land use within watersheds. Using this index and a river network nitrogen removal model, we quantified the effect of varying the location of developed land use within two watersheds on nutrient removal and export. The quantity and location of developed land use as well as runoff affected nitrogen removal and export. Because river network nitrogen removal is bypassed when sources are skewed toward the watershed mouth, varying the location of land use alone can double aquatic nitrogen removal. Nutrient sources skewed toward the distant headwaters maximized in-stream removal which in turn can reduce watershed export.

1. Introduction

Human activity has accelerated the nitrogen (N) cycle and enriched the landscape with reactive N [Galloway *et al.*, 2008]. As a consequence, most coastal rivers and bays in the U.S. are degraded by nutrient pollution [Howarth *et al.*, 2002], resulting in eutrophication [Cloern, 2001] which has negative ecological effects, such as algal blooms, anoxic zones, and loss of habitat, as well as associated economic impacts like declining property value and tourism [Pretty *et al.*, 2003; Dodds *et al.*, 2009]. The effect of human land use on water quality, nutrient enrichment, and export from watersheds is well established [Beaulac and Reckhow, 1982; Carpenter *et al.*, 1998; Foley *et al.*, 2005]. Of particular concern are nonpoint nutrient sources associated with human land uses such as urban/suburban and agricultural areas that are a major source of nutrients contributing to eutrophication [Carpenter *et al.*, 1998].

Studies considering the effects of land use on watershed nutrient export typically consider the aggregate measure of total area in a watershed and pay little attention to the spatial distribution of this land use [e.g. Cronan *et al.*, 1999; Goodridge and Melack, 2012]. Percent watershed impervious cover is also commonly used as an indicator of environmental degradation from urbanization [Arnold and Gibbons, 1996]. However, the spatial distribution of landscape patches and the spatial scale at which land use is considered can be important [Strayer *et al.*, 2003; King *et al.*, 2005]. Some studies have shown that land use in the riparian corridor [Sponseller *et al.*, 2001; Strayer *et al.*, 2003; Van Sickle and Johnson, 2008] or subcatchment [Allan *et al.*, 1997] can better predict in-stream conditions. King *et al.* [2005] found that the Euclidean distance of land use to a sampling point improved predictions of nitrate concentration in some streams. Though some have found that land use at the whole watershed scale may best predict nitrate flux [Strayer *et al.*, 2003], the distribution of land use patches within the watershed and watershed size may affect nutrient export due to cumulative in-stream processes.

N fluxes to coastal zones are a function of N inputs to the watershed, terrestrial retention, and N removal in transit through the river network. In-stream processing in river networks can remove substantial proportion of N flux controlling the magnitude and timing of N export [Bernhardt *et al.*, 2005; Mulholland *et al.*, 2008; Wollheim *et al.*, 2008a; Lin *et al.*, 2015]. Model simulations suggest that water residence time affects nutrient removal [Wollheim *et al.*, 2006]; therefore, landscape position of nutrient sources and their associated flow path distance may affect the potential for nutrient processing before reaching the ocean [Seitzinger *et al.*, 2002; Alexander *et al.*, 2002]. However, it is challenging to summarize complex spatial information regarding the distribution of nutrient sources within watersheds. Perhaps, because of this, the potential effect of source location on N removal and watershed N export has not been quantified.

Our goal for this study is to evaluate the sensitivity of N processing and export in river networks to watershed land use distribution. We first propose a metric to summarize the distribution of land use within watersheds in a river network context that may help to better understand N removal potential. We then evaluate the export and aquatic processing of N for two coastal watersheds in New England under current and scenarios of land use distribution patterns. We hypothesized that the location of land use within watersheds would control the opportunity for in-stream N removal, as denitrification, by regulating the time N inputs spent in the river network and therefore have a large effect on N removal and, to a lesser extent, N export.

2. Methods

We quantified land use distribution within watersheds in terms of skewness toward or away from the river mouth. Because there was no established metric, we developed an index to summarize the spatial distribution of land uses within watersheds considering the surface water flow path distance from potential sources to the watershed mouth. This skewness index (SI) is

$$SI_{lu} = \frac{lu \text{ weighted mean flow path distance}}{\text{unweighted mean flow path distance}} \quad (1)$$

The unweighted mean flow path distance is the average distance traveled by water through the hydrologic network. We calculated land use weighted mean flow path distance for grid cell i to grid cell n as

$$\frac{\sum_{i=1}^n lu * FD}{\sum_{i=1}^n lu} \quad (2)$$

where lu is the proportion of each grid cell in the watershed occupied by a given land use type and FD is the flow path distance from that grid cell along the river network to the watershed mouth. To calculate SI values, we used land use data from the 2006 National Land Cover Database (NLCD) [Fry *et al.*, 2011]. A SI value of 1 represents no skewness in the distribution of land use within the watershed while $SI < 1$ represents skewness of land use and associated N sources toward the river mouth, and $SI > 1$ represents skewness toward the most distant headwaters.

We modeled export and in-stream processing of dissolved inorganic N (DIN) using the N removal model within the Framework for Aquatic Modeling in the Earth System (FrAMES) [Wollheim *et al.*, 2008a, Wollheim *et al.*, 2008b, Stewart *et al.*, 2011]. FrAMES is a spatially distributed, grid-based model of river network hydrology and biogeochemical processes that operates on a daily time step. Modeled N removal, as denitrification, is based on measured denitrification rates from the Lotic Intersite Nitrogen eXperiments 2 [Mulholland *et al.*, 2008] and varies with water temperature and ambient DIN concentrations in each grid cell (equations (3)–(5)) [Wollheim *et al.*, 2008b].

Denitrification uptake velocity ($m d^{-1}$) at the reference temperature of 20°C ($Vf_{den-Ref}$) was calculated for every grid cell

$$Vf_{den-Ref} = 10^{(den_int + \log_{10}(DIN_{conc}) * den_slope)} * 864 \quad (3)$$

where den_int and den_slope are -2.975 and -0.493 , respectively [Mulholland *et al.*, 2008], DIN_{conc} is the concentration of DIN in microgram per liter, and 864 is a conversion factor. Denitrification uptake velocity in the grid cell is then calculated as

$$Vf_{den} = Vf_{den-Ref} * Q10^{((waterT - 20) / 10)} \quad (4)$$

where $water T$ is the water temperature (°C) in the grid cell and $Q10 = 2$. Total DIN removal (mass/mass) in each grid cell is calculated as

$$R = 1.0 - e^{(-1.0 * Vf_{den}/HL)} \quad (5)$$

where $HL = \text{discharge}/(\text{width} * \text{length})$. Simulated discharge ($m^3 s^{-1}$), width (m), and length and water temperature (m) for each grid cell are provided by FrAMES and are described in Stewart *et al.* [2013].

We used the DIN loading function developed for the Ipswich watershed [Wollheim *et al.*, 2008b] in both watersheds. This loading function uses grid cell percent human land use, defined as the sum of residential, industrial, and agricultural, to predict runoff DIN concentration as a function of runoff quantity [Wollheim *et al.*, 2008b].

Therefore, we aggregated the developed (residential and industrial) and agricultural (crop and hay/pasture) land use categories from the NLCD data to quantify human land use for each grid cell. Though lumping agriculture with urban/suburban land uses would not be appropriate in areas where agriculture is intensive, it is an acceptable simplification where agricultural land represents a relatively small contribution to nutrient loading compared to developed land use and agriculture primarily represents low-intensity uses such as pastures and hay fields. In addition, in the Lamprey watershed, we added a waste water treatment facility to DIN loading. The waste water facility is a point source input to the grid cell where it is located. We assumed that daily DIN input from this facility to the river is the annual DIN mass/365 (Annual data: *Piscataqua Region Estuaries Partnership* [2012]).

We calculated flow path distances and the skewness index using RiverGIS (version 2.1), a river network based geographic information system developed at University of New Hampshire which allows for flow path analysis of hydrological data sets [Vorosmarty *et al.*, 1998]. We used a stream topological network of gridded river channels with a spatial resolution of 15 s latitude by 15 s longitude (approximately 500 m) for the Ipswich and Lamprey river networks. To calculate the skewness index in large Northeast U.S. watersheds, we used a river network with spatial resolution of 3 min latitude by 3 min longitude (approximately 4.75 km) [Stewart *et al.*, 2013]. Both networks were derived from Hydrological data and maps based on Shuttle Elevation Derivatives at multiple Scales (<http://hydrosheds.cr.usgs.gov>) using the regridding algorithm described in Fekete *et al.* [2001] and verified against the National Hydrography Dataset (nhd.usgs.gov).

We assessed the potential effect of land use distribution on watershed export and river network removal of dissolved inorganic N (DIN) by varying the location of land use within two watersheds to generate a range of SI. We used two watersheds of similar size with differing amounts of developed land use (Figure 1). The Lamprey River watershed is located in southeast New Hampshire draining to the Great Bay estuary with 9% and 5% of the area developed and agricultural, respectively (Table 1). The Ipswich River watershed located in northeast Massachusetts has much more developed land use due to its proximity to the city of Boston with 32% of the watershed area and developed and also 5% of the area classified as agricultural (Table 1).

To quantify the effect on river network DIN removal and watershed export of one aspect of land use distribution within watersheds, the skewness toward or away from watershed outlet, we aggregated human land use in the two test watersheds and varied the location of developed grid cells to generate contrasting SI values (Figure 1). We aimed to produce a SI of 1 (land use centered within the watershed) and the largest and smallest SI values possible given watershed template and human land use area. To accentuate skewness and generate a wider range of SI values, we aggregated human land use into the least possible number of grid cells while maintain the same total area of human land use. Though this aggregation allows to best test the effect of varying SI, this aggregation also makes the results of these scenarios not realistic and not comparable to model output using actual land use distribution because land use intensity affects DIN loading. So that DIN input is equal for each scenario, we used evenly distributed runoff set to generate annual average discharge (from 2000 to 2010). To illustrate variability of effect of land use skewness on river network DIN removal with changing discharge, we also reran the model for double and half the annual discharge.

To gain a broader understanding of how land use distribution varies in the Northeast U.S., we conducted a regional analysis of the distribution of land use within large watersheds. We considered the James, Susquehanna, Delaware, Hudson, Connecticut, Merrimack, and Penobscot watersheds which cover a large area of the region, encompass several urban centers, and represent a range of land use intensity (Table 2). We calculated SI for developed and agricultural land use in each watershed.

3. Results and Discussion

The amount, intensity, and location of land use within watersheds affected nitrogen export. Because it has more than twice as much developed land use, the Ipswich watershed exported more DIN than the Lamprey under actual land use distribution (Table 1). Concentrating developed land use for the scenarios also increased watershed DIN export regardless of where in the watershed the developed land use occurred (Table 1 and Figure 2). However, the location of land use within a watershed can also affect in-stream DIN removal (Figure 3) and watershed DIN export (Figure 2).

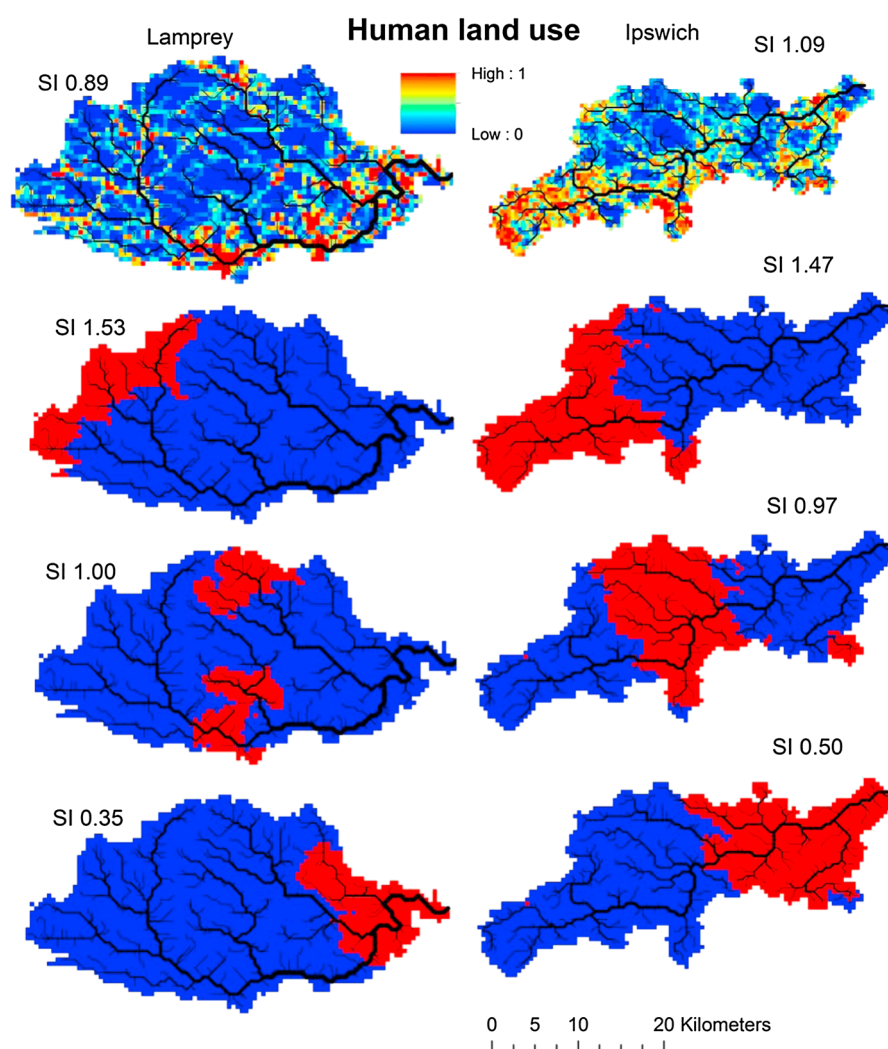


Figure 1. The (left) Lamprey and (right) Ipswich watersheds showing actual distribution of human land use (proportion developed + agricultural) and the three scenarios for which human land use was concentrated and located to generate a range of skewness indices from skewed toward the headwaters, centered, and skewed toward the watershed outlet.

Actual land use distribution in the Lamprey has a SI of 0.89 indicating that human land use is skewed toward the watershed outlet while the SI of 1.09 in the Ipswich indicates skewness toward the headwaters (Figure 1). Even though a SI greater than 1 in the Ipswich watersheds indicates more potential for in-stream DIN removal than in the Lamprey, increased DIN loading from the greater amount of developed land use still results in larger DIN export from the Ipswich watershed compared to the Lamprey (Table 1).

In the Lamprey watershed, we generated SI values ranging between 0.35 and 1.53, but in the Ipswich the range was more constrained between 0.50 and 1.47 due to a larger area of the watershed being developed

Table 1. Characteristics of the Study Watersheds^a

Watershed	Area (km ²)	Population Density (km ⁻²)	Area Developed (%)	Skewness	Measured DIN Export (kg km ⁻² yr ⁻¹)	Modeled DIN Export (kg km ⁻² yr ⁻¹)
Lamprey	474	72	14	089	77	71
Ipswich	400	310	37	1.09	171	185

^aArea developed and land use skewness represent both developed and agricultural land use categories (5% of each watershed is agricultural land with the remaining developed area being residential/commercial). Population density is calculated from 2010 census data. Both measured and modeled DIN export values are median for 2009–2009. Lamprey-measured DIN export from Daley *et al.* [2010] and Ipswich-measured DIN export from Morse and Wollheim [2014].

Table 2. Land Use Skewness in Large Northeast U.S. Watersheds

Watershed	% Land Use		Skewness Index	
	Developed	Agriculture	Developed	Agriculture
Connecticut	10	7	0.67	0.93
Delaware	20	19	0.61	0.86
Hudson	11	15	0.64	0.98
James	10	14	0.75	0.99
Merrimack	17	5	0.69	0.86
Penobscot	2	2	0.56	0.60
Susquehanna	8	26	0.81	0.94

(Figure 1). Land use skewed toward the headwaters resulted in the highest local DIN concentration due to lower dilution capacity (data not shown), but twofold greater in-stream DIN removal at the network scale compared to land use skewed toward the watershed outlet (Figure 3). Land use skewed toward the headwaters results in higher DIN flux (H mix line) throughout the river network (Figure 2). Because land use area is the same across scenarios regardless of SI, DIN export without in-stream removal processing (conservative mixing, “mix” solid lines) is identical (Figure 2). However, the location of land use, as well as the amount of

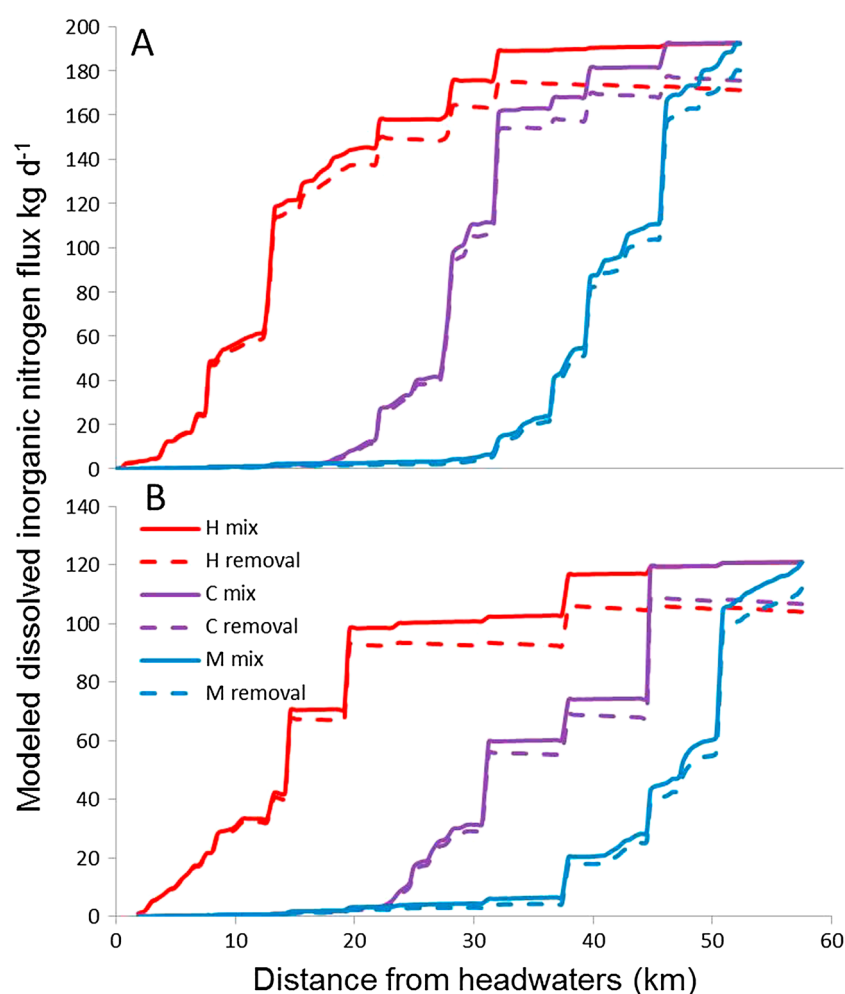


Figure 2. Modeled DIN flux watershed profiles with land use skewed toward the headwaters (H, red), centered (C, purple), and toward the watershed mouth (M, blue) for the (a) Ipswich and (b) Lamprey watersheds. Solid lines represent modeled conservative mixing of DIN (mix), and the dashed line shows modeled DIN concentration with in-stream processing (removal).

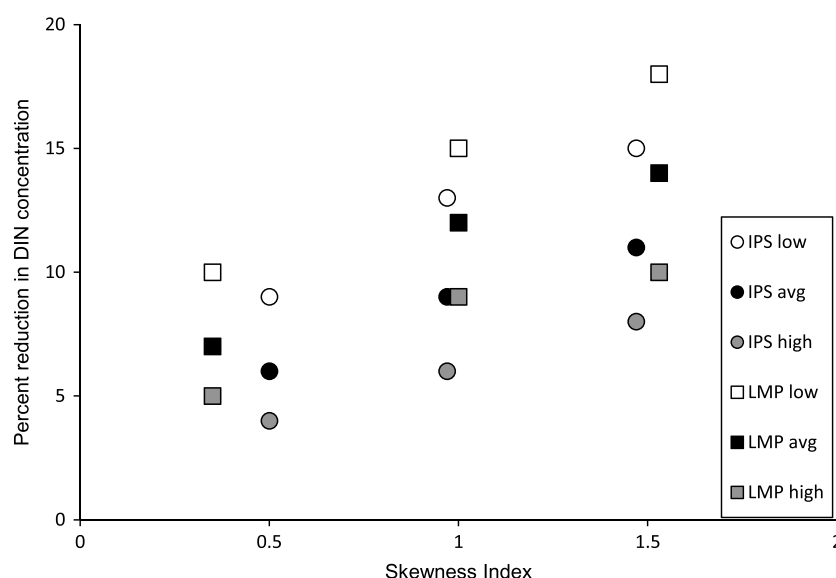


Figure 3. The percent of dissolved inorganic nitrogen (DIN) concentration that is removed by in-stream processing with varying location of human land use within the Ipswich and Lamprey watersheds. The skewness index on the x axis indicates if land use is skewed toward the headwaters (>1), centered (1), or skewed toward the watershed outlet (<1). Black symbols represent results of simulations at annual average discharge, open symbols half of annual average discharge (low), and gray circles double annual average discharge (high) in each watershed.

land use, and discharge affected in-stream removal of DIN (Figure 3). For the Lamprey watershed, at average annual discharge, in-stream processes reduced DIN concentration by 7, 12, and 14% when SI was 0.35, 1.00, and 1.53, respectively (Figure 3). In the Ipswich watershed, in-stream removal of DIN reduced concentrations by 6, 9, and 11% when SI was 0.50, 0.97, and 1.47, respectively (Figure 3). Increasing discharge reduced in-stream DIN removal (Figure 3). For similar SI, DIN removal was greater in the Lamprey compared to the Ipswich (Figure 3) due to lower DIN concentrations resulting from less DIN input from a smaller area of human land use (Figure 1 and Table 1). Nutrient uptake becomes less efficient as ambient nutrient concentrations increase [Mulholland *et al.*, 2008]; therefore, increased loading can reduce the efficiency of in-stream nutrient removal. At average annual discharge, land use skewed toward the headwaters results in a reduction in DIN export of 8 and 9 kg d^{-1} in the Lamprey and Ipswich, respectively, compared to the same land use skewed toward the watershed outlet (Figure 2).

Though it is well established that human population is concentrated in coastal areas [Crowell *et al.*, 2007], the distribution of land use within smaller coastal watersheds can vary as illustrated by the patterns in the Lamprey and Ipswich watersheds. To place the land use distribution of the Ipswich and Lamprey as well as the scenarios we generated in a regional context, we also calculated SI for developed and agricultural land use in large watersheds of the Northeastern US. In these watersheds, agricultural and developed land uses were strongly skewed toward the watershed mouth as indicated by $\text{SI} < 1$, though in some cases, such as the Hudson and the James, agriculture was minimally skewed (Table 2). SI for developed land use ranged from 0.81 in the Susquehanna to 0.56 in the Penobscot and SI for agricultural land use ranged from 0.99 in the James to 0.60 in the Penobscot (Table 2). In each case, the distribution of developed land use was more skewed toward the watershed mouth than was agricultural land use indicating that farmland is located in the hinterlands beyond cities (Table 2).

The SI characterizes the spatial distribution of land use within watersheds in a manner that indicates the average residence time of developed land use runoff in the river network and the potential for N removal. On average, developed land use had a SI of 0.68, indicating that flow path distances to the watershed mouth, and therefore residence times, of runoff from developed land is less than that of average runoff for each watershed. The average SI for agricultural land use was 0.88, suggesting that N inputs from agricultural land use have a greater opportunity to be processed within the river network compared to inputs from urban/suburban land use. Furthermore, the spatial distribution of human land use may help to explain

why some watersheds are more retentive of N inputs than others. For example, in the James River watershed, both developed and agricultural land are less skewed toward the watershed outlet than other watersheds (Table 2) and have lower proportions of riverine N export [Boyer *et al.*, 2002, Figure 6].

When there is a mismatch between the location of ecosystem services supply and demand, the utility of the ecosystem service can be limited [Bagstad *et al.*, 2013, Wollheim *et al.*, 2013]. The ecosystem service of N removal occurs throughout the river network [Hale *et al.*, 2014]. Headwater streams play an important role in controlling nitrogen export from watersheds [Peterson *et al.*, 2001]. However, larger rivers may also be important in processing N and regulating watershed N export [Wollheim *et al.*, 2006; Tank *et al.*, 2008; Stewart *et al.*, 2011; Hall *et al.*, 2013]. When sources of N are skewed toward the watershed mouth, potential N processing in the headwaters and midorder streams is bypassed, therefore limiting the utility of the ecosystem service. Nutrient processing in streams is strongly controlled by hydrology with in-stream nutrient processing declining as discharge increases and water residence time decreases [Royer *et al.*, 2004]. The spatial distribution of land use and discharge both affect river network N removal so that more DIN is removed when land use is skewed toward the distant headwaters, but the proportion of DIN removed is less with increasing discharge for all SI values (Figure 3). Thus, coastal watersheds where human land use is clustered near river mouths may be more vulnerable to eutrophication from land use change, especially during high flow periods.

Maximizing in-stream DIN removal by exploiting surface water flow path length and water residence time between DIN sources and watershed mouth may lead to reduced DIN export [Behrendt and Opitz, 2000, Alexander *et al.*, 2008; Green *et al.*, 2009] but would produce trade-offs in water quality within the river network. Concentrating N sources in the headwaters would result in higher nutrient concentrations throughout the river network (Figure 2). This would shift the impact from the coastal zone to degrading water quality throughout the river network. Reduced water quality would limit ecosystem services of the river systems such as providing drinking water. Elevated levels of nitrate in drinking water are a health hazard [Terblanche, 1991], and 66% of public water supply in the U.S. is withdrawn from surface waters [Kenny *et al.*, 2005]. Using ecosystem services should be done in a manner to maximize the utility of the environment without damaging the resource and considering trade-offs.

Our findings highlight that this ecosystem service of in-stream N removal is maximized when N sources are skewed toward the distant headwaters, especially during low flow periods, which in turn can reduce watershed N export. Therefore, the spatial distribution of land use rather than simply the total land use amount can influence ecosystem health, function, and services. Analytical approaches to evaluate and quantify the spatial distribution of land use, such as the SI, can be used to understand ecosystem current condition and future risk as well as inform management and development planning. We found that the skewness of N sources likely influences N processing and export on an annual scale, and it is likely to have the largest effect on DIN fluxes during warm low flow periods when the potential for in-stream processing is greatest. The SI could be used to prioritize subwatersheds and areas for nutrient runoff management.

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