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Direct groundwater discharge and vulnerability to hidden nutrient loads along the Great Lakes coast of the United States

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ABSTRACT

Direct groundwater discharge delivers nutrients from land and lakebed sediments to the Great Lakes, which impacts lake water quality. Broad spatial distributions of discharging groundwater are often difficult to measure directly. We present high resolution estimates of direct groundwater discharge across 43% of the Great Lakes coastline based on a water budget approach that uses hydroclimatic models and high-resolution hydrographic data available within the United States. We also integrate land use data to identify coastal areas vulnerable to high groundwater-borne nutrient loads. Estimated rates of direct groundwater discharge along the Great Lakes coast are highly variable, but generally are greatest for Lake Erie and Lake Michigan. Almost one-third of Lake Erie's United States coastline is vulnerable to groundwater sources of nutrients. To assess uncertainties and limitations in our vulnerability analysis, a vulnerable site along Lake Erie was selected for detailed field measurements of direct groundwater discharge rates and nutrient fluxes. Measured discharge rates were significantly lower than water budget-based estimates (354 ± 25 $m^3\,y^{-1}\,m^{-1}$ compared to 588 ± 181 $m^3\,y^{-1}\,m^{-1}$). Dissolved phosphorous concentrations in the lakebed were elevated compared to onshore groundwater, while nitrate concentrations were lower, indicative of a highly reactive sediment-water interface. Some of the measured phosphorus may be locally sourced from desorption of legacy P or mineralization of organic matter in the lakebed, which our vulnerability framework does not include. Much of the land-derived nitrogen may be transformed along groundwater flow paths prior to discharge. While model-based estimates of direct groundwater discharge and vulnerability to nutrient loading are important for managing Great Lakes water quality, direct field observations remain essential for quantifying fluxes.

1. Introduction

Large algal blooms pose severe problems for lake ecosystems and the coastal communities that surround them (Davis, 1969; Glibert et al., 2005; Backer and McGillicuddy, 2006; Michalak et al., 2013). The sheer biomass of algal blooms can stress ecosystems. Algae deplete oxygen leading to hypoxia and anoxia, and algal mats block sunlight from reaching plants below the surface, reducing stability of plant life in the water column (Anderson et al., 2002; Glibert et al., 2005). Some algae also produce toxins like microcystin and anatoxin that kill fish and harm humans (Landsberg, 2002; Glibert et al., 2005; Backer and McGillicuddy, 2006).

Algal blooms frequently cover large areas of the Great Lakes, which prompted the creation of the Great Lakes Water Quality Agreement between Canada and the United States in the 1960s. The agreement was designed to improve water quality by reducing nutrient loading (Backer and McGillicuddy, 2006; Stow et al., 2015; Dolan, 1993). After an initial reduction in algal blooms and increase in oxygen levels throughout the Great Lakes (Makarewicz, 1993; Makarewicz et al., 1999), conditions began to deteriorate again by the mid-1990s. In 2011, Lake Erie experienced the most extensive algal bloom in recorded history (Burns et al., 2005; Bridgeman et al., 2013; Michalak et al., 2013)

The persistence of algal blooms has been attributed to anthropogenic additions of phosphorous (P) and nitrogen (N) (Glibert et al., 2005). It is important to understand how these nutrients are delivered in order to accurately address water quality issues in the Great Lakes (Matisoff et al., 2016). Nutrients can be transported by rivers or direct groundwater discharge (groundwater

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that is not intercepted by rivers but rather discharges along the coast). Nutrient loading from rivers can be estimated using measured discharge rates (obtained from river gauging) and nutrient concentrations (Quilbé et al., 2006). However, direct groundwater discharge, i.e. the outflow of terrestrially-derived water across a lakebed, is difficult to quantify. Nutrient concentrations in discharging groundwater are also difficult to ascertain because the sediment-water interface is often a reactive zone that alters concentrations of discharging nutrients (Lewandowski et al., 2015; Robinson, 2015). As a result, accurate measurements of nutrient fluxes to the coast are sparse, and direct groundwater discharge is often overlooked as a source of nutrients that stimulates algal blooms (Kilroy and Coxon, 2005; Lewandowski et al., 2015; Rosenberry et al., 2015).

Direct groundwater discharge occurs wherever hydraulic head in the onshore aquifer is elevated above the lake water table (Grannemann et al., 2000; Robinson, 2015). Most studies of direct groundwater discharge to the Great Lakes have focused on Lake Michigan. Overall, direct groundwater may contribute 1–12% of total water inflow to Lake Michigan (Grannemann et al., 2000; Hoaglund et al., 2002; Robinson, 2015). These rates have generally been estimated using groundwater models or measured hydraulic heads and Darcy's law, which require assumptions about aquifer properties (Grannemann et al., 2000). Direct measurements of groundwater seepage are sparse. In one study, Cherkauer and Hensel (1986) used both groundwater models and direct measurements to calculate rates of 153.4 and 87.7 $m^3\,y^{-1}\,m^{-1}$, respectively at Mequon, Wisconsin.

Even less is known about nutrient loads associated with direct groundwater discharge to the Great Lakes (Robinson, 2015). Sources of groundwater-borne nutrients to the Great Lakes include agricultural areas, septic systems, leaky infrastructure, and landfills along coastal catchments (Robinson, 2015). In other lakes, water-budget calculations and field observations have been used to quantify nutrient loading (Meinikmann et al., 2013; Meinikmann et al., 2015). Here, we use a water-budget approach to generate new estimates of direct groundwater discharge to the Great Lakes coast of the United States. We then use these estimates to identify areas that may be prone to high nutrient loads from groundwater. Next, we examine a vulnerable location on the Lake Erie coast, where we measured direct groundwater discharge rates and nutrient fluxes. Using insights from field observations, we evaluate strengths and limitations of our vulnerability assessment approach. We show that both large-scale model-based estimates and site-specific field observations are essential for constraining groundwater fluxes and potential nutrient loads to the Great Lakes coast.

2. Methods

2.1. Water budget

We estimated direct groundwater discharge to the Great Lakes using a water-budget approach that has been previously used to quantify fresh submarine groundwater discharge to oceans (Zektser and Loaiciga, 1993; Destouni et al., 2008; Sawyer et al., 2016) and is capable of resolving high-resolution continental-scale discharge rates. Briefly, we identified coastal areas of land that fall outside the contributing catchment areas of rivers and streams (Fig. 1, inset). All runoff in these coastal catchments flows directly and exclusively to the coast. If groundwater divides coincide with topographic boundaries, then groundwater in these catchments also flows exclusively to the coast, and we can consider coastal catchments as recharge zones for direct groundwater discharge to the coast (Fig. 1). For each coastal recharge zone, we assume that the annual recharge volume equals the annual volume of direct groundwater discharge (Sawyer et al., 2016). Groundwater extraction is considered negligible. Because we include no net groundwater import from upland catchments (Schaller and Fan, 2009a), our assumptions about recharge areas are most appropriate for the shallow unconfined aquifer. The method may neglect a significant component of direct groundwater discharge from confined aquifers that recharge farther inland.

Coastal recharge zones were delineated using high-resolution hydrographic data for the Great Lakes Region of the United States obtained from the National Hydrography Dataset, NHDPlus (McKay et al., 2012). First, coastlines were extracted from the polyline data of rivers, streams, and coasts. A total of 3008 coastal segments were identified with an average length of 2.2 km and a standard deviation of 2.9 km. The coastlines were grouped by reach code, since arbitrarily small coastal segments often share the same reach code, and reach codes tend to divide areas of coastline between rivers and streams. Reach codes are 14-digit integers that combine the 8-digit Hydrologic Unit Code of the U.S. Geological Survey (Seaber et al., 1987) and a unique 6-digit arbitrary number together forming a unique identifier for each reach of NHDPlus. The association by reach code resulted in 1619 coastal segments with average length of 4.1 km and standard deviation of 6.7 km. Next, the recharge zone for each coastline was extracted using the common NHDPlus integer identifier relating polylines and polygons. The average recharge zone area is 4.3 km^2 and the standard deviation is 11.2 km^2. None of the coastal recharge zones contain any streams by design. The NHDPlus data is only available for the United States coast (Fig. 1). Smaller bodies of water within the Great Lakes system, such as St. Clair Lake, are also not included in the dataset. Our analysis therefore spans 43% of the Great Lakes coast. Note that while hydrographic datasets exist for the Canadian coastline of the Great Lakes (Lehner et al., 2008), differences in spatial resolution of the underlying topography stand in the way of consistent direct groundwater discharge estimates across scales (Destouni et al., 2008).

Recharge rates were derived from hydroclimatic reconstructions using the second phase of NASA's North American Land Data Assimilation System, NLDAS2 (Xia et al., 2012). Non-infiltrating runoff was excluded since it discharges to the coast as overland flow. For each coastal recharge zone, volumetric recharge ($m^3\,y^{-1}$) was calculated by extracting the infiltrating runoff value nearest to the recharge zone centroid and multiplying by area. Direct groundwater discharge per unit length of coast ($m^3\,y^{-1}\,m^{-1}$) was obtained by dividing the volumetric recharge rate by shoreline length (m) for each coastal recharge zone.

2.2. Vulnerability

We identified coastlines vulnerable to groundwater-borne nutrient inputs using the method of Sawyer et al. (2016) that is based on two criteria: direct groundwater discharge rates and land use within recharge zones. Our conceptual model is that groundwater-borne nutrient inputs are greater when a large volume of water discharges to the coast from highly impacted recharge zones. Specifically, we define segments of the coast as vulnerable when two criteria are met: 1) the percentage of agricultural and developed land-cover within the associated recharge zone is above average (37%), and 2) the groundwater flux is above the average value for the United States Great Lakes coast (381 $m^3\,y^{-1}\,m^{-1}$). In each coastal recharge zone, we determined the vulnerable fraction of developed and agricultural land from the National Land Cover Database (NLCD). This includes developed open spaces, developed low, medium, and high intensity areas, pastures, and cultivated crops.

Our vulnerability analysis does not predict nutrient loads, but instead estimates the likelihood of higher loading rates now and

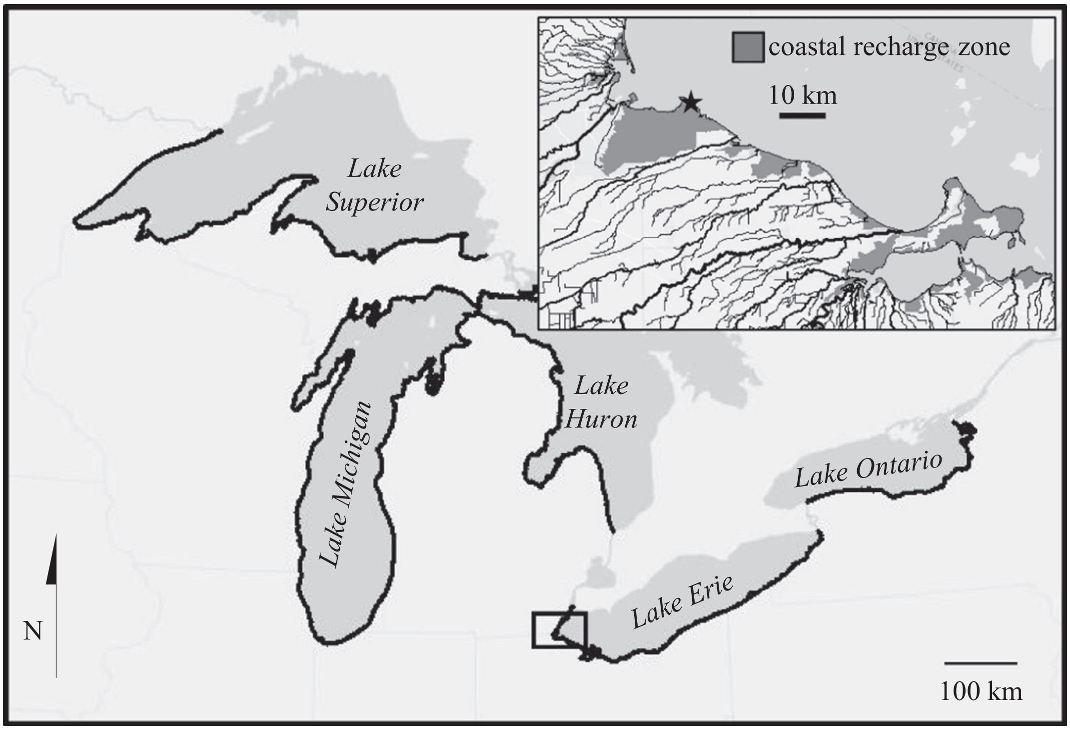

Fig. 1. Location of the study site (star, inset). Coastal recharge zones contribute water directly to the coast instead of streams. NHDPlus data is only available for the US coastline (bold lines).

in the future. Biogeochemical transformations can occur along groundwater flow paths and influence nutrient fluxes at the sediment-water interface (Duff and Triska, 1990; Hill, 1996; Kroeger and Charette, 2008). Also, it can take decades for nutrients in recharging water to reach the coast (Meals et al., 2010). The vulnerability analysis is useful for identifying areas that should be considered for measurements or monitoring.

2.3. Field site

We selected a vulnerable beach on Lake Erie for focused measurements of direct groundwater discharge rates and groundwater-borne nutrient fluxes. Due to the heterogeneous nature of the Great Lakes coast, this single field site is not necessarily representative of much of the coast and also provides insufficient data to validate the entire water budget and vulnerability analysis. However, it serves as a platform for considering methodological improvements. The water budget analysis revealed many vulnerable beaches along Lake Erie, but we selected this site for its easy access and logistical advantages. The site (41 41′57.62″ N, 83 °19′32.95″ W) is located in the lake's western basin at Cedar Point National Wildlife Refuge (Fig. 1). It lies on a gradually sloping beach, allowing for installation of seepage meters without scuba gear. The beach is part of a spit separating Lake Erie from a highly vegetated marsh to the south. Land use within the recharge zone is mixed. The land immediately adjacent to the beach is marsh and forest, but much of the area is agricultural, and the city of Toledo lies only 18 km southwest. The Maumee River discharges to Lake Erie approximately 12 km west of the study site. The surficial geology generally consists of recent sand deposits overlying glacial till and lake shale (Fuller, 1996).

2.3.1. Seepage meter measurements

Lee-type seepage meters were constructed from the ends of steel drums with an internal diameter of 57 cm (Lee, 1977) and deployed on September 5, 2015. Fifteen seepage meters were arranged along three shore-perpendicular transects: Transect 1, Transect 2, and Transect 3 (Fig. 2). Seepage meters were spaced approximately 5 m apart, in water depths ranging from 0.6 to 1.5 meters. The distance between each transect was approximately 200 m. In order to resolve shore-parallel variation in seepage rates, additional seepage meters were deployed at even intervals between transects. A nest of four seepage meters, spaced 1 m apart, was also installed to measure small scale-heterogeneity in seepage rates (Fig. 2).

After installation, seepage meters were allowed to equilibrate for twenty-four hours with open valves prior to taking measurements. Plastic autoclaved collection bags were prefilled with 1.89 liters of lake water and weighed. The bags were attached to seepage meters for approximately two hours, after which they were removed and weighed again. Seepage rates were calculated from the difference between the initial and final water mass per length of sample time. Two rounds of measurements were made on the same day, and the rates were averaged. Weather conditions were generally calm and waves were minimal during both rounds of measurements. Based on the precision of our scale, the precision of our seepage rates was ±0.24 cm d^{-1}. This precision was propagated as a proxy for error when we calculated volumetric fluxes of direct groundwater discharge at the site.

2.3.2. Concentrations and nutrient fluxes

To compute nutrient flux to the lake, pore water was sampled next to each seepage meter at a depth of 25 cm below the sediment-water interface. Samples were obtained by suction using a syringe attached to a steel tube (0.5 cm inner diameter and 0.6 cm outer diameter with a screened interval of 3.5 cm). One tubing volume (∼24 ml) was discarded before sample collection.

For comparison, water samples were also collected from the lake, nearby marsh, and onshore aquifer. Temporary piezometers were installed to sample onshore groundwater. The piezometers were constructed of 4.5 cm outer-diameter PVC and screened through the water table. The piezometers were fully purged with

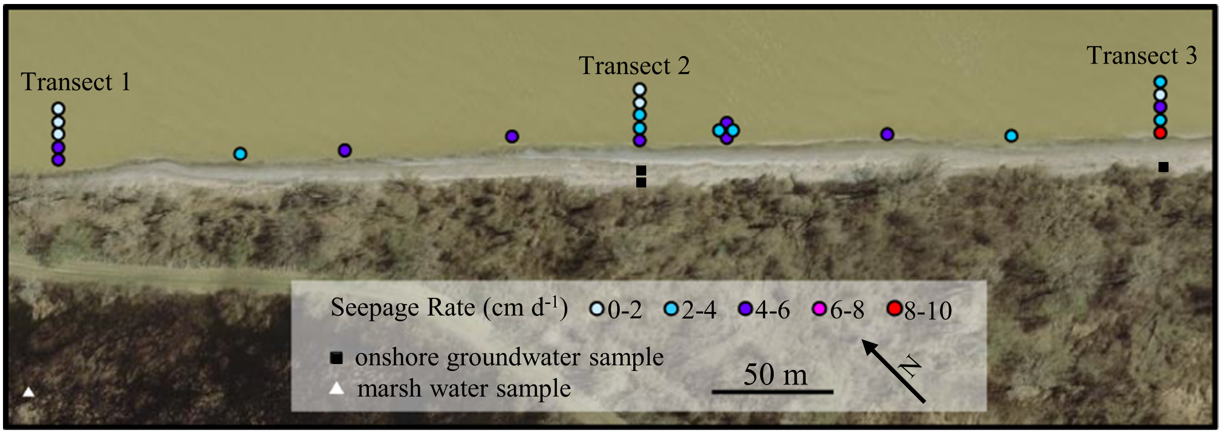

Fig. 2. Average seepage rates of round 1 and round 2 (circles). Locations of onshore groundwater and marsh water samples are also shown with squares and triangles respectively.

a peristaltic pump and then sampled. All water samples were filtered (0.45 μm), immediately placed on ice, and transferred to a freezer within 12 h of collection. $NO_2^- + NO_3^-$-N and NH_4^+-N were measured using a Skalar flow-injection nutrient analyzer, and their summed concentrations are reported as dissolved inorganic nitrogen (DIN). Major anions, including phosphate (PO_4^{3-}), were measured using ion chromatography. Detection limits for NO_2^- + NO_3-N, NH_4^+-N and PO_4^{3-} were 0.014, 0.0026 and 0.019 mg L^{-1}, respectively.

Nutrient fluxes were calculated as the product of the concentration at each seepage meter and the seepage rate. Where the concentration was below detection, we calculated fluxes using the detection limit.

3. Great Lakes analysis

3.1. Patterns and rates of direct groundwater discharge

The total annual volume of direct groundwater discharge to the Great Lakes (U.S. portion only) is 2.54 $km^3\,y^{-1}$. The average volumetric flux per unit length of shoreline is 381 $m^3\,y^{-1}\,m^{-1}$. Lake Erie and Lake Michigan have the highest average fluxes (477 $m^3\,y^{-1}\,m^{-1}$ and 410 $m^3\,y^{-1}\,m^{-1}$, respectively). Average flux is lowest to Lake Ontario (308 $m^3\,y^{-1}\,m^{-1}$) (Table 1). Direct groundwater discharge rates vary spatially along the coast of individual lakes (Fig. 3). Regionally, high discharge rates are concentrated on the south-central and southeastern coasts of Lake Erie, as well as the northeastern and southwestern coasts of Lake Michigan. Other areas of high discharge include the southeastern coast of Lake Superior, northwestern coast of Lake Huron, and eastern coast of Lake Ontario.

Onshore infiltration (percolation of surface water to the subsurface) is a key control on the pattern of direct groundwater discharge that reflects both climate and geology. Infiltration is controlled by precipitation and the capacity of the land surface to accept water. Much of the Great Lakes shoreline is bounded by glacial till and outwash deposits, which can vary widely in permeability. Coastal areas along the Western Lake Erie Basin (west of Cleveland) generally consist of less permeable silty glacial till. This area corresponds with lower discharge rates on the map (Fig. 3). East of Cleveland, tills are sandier (Fullerton et al., 1991). Also, bedrock bluffs 15–20 m high make up most of the shoreline east of Cleveland (Morang et al., 2011) and may allow greater head gradients to develop near the coast. The water budget analysis correspondingly shows high discharge rates along the eastern portion of Lake Erie (Fig. 3). Over small areas, permeable zones such as spits where glacial sands have been reworked may allow for locally elevated infiltration and groundwater discharge rates. Our field site is located on one such spit.

The geometry of coastal recharge zones is another key factor influencing patterns of direct groundwater discharge. Although Lake Ontario has the highest infiltration rate among the Great Lakes, its average volumetric flux of groundwater per length of shoreline is lowest. Its recharge zones do not stretch as far inland and therefore contribute less groundwater to the coast. Long, narrow recharge zones convey more groundwater per unit length coastline.

Water extraction is not included in our analysis, and direct groundwater discharge rates are likely overestimated in areas with substantial drawdown. For example, withdrawal due to municipal pumping in Chicago and Milwaukee reduces the amount of groundwater discharge along the southwestern coast of Lake Michigan (Feinstein et al., 2010).

3.2. Comparison with other models and methods

The total volumetric rate of direct groundwater discharge to Lake Michigan (0.97 $km^3\,y^{-1}$) is comparable to results from a

Table 1
Water budget statistics for the Great Lakes.

Lake	Coastline Analyzed (km)	Coastline Analyzed (%)	Infiltration (cm y^{-1})	Volumetric Flow Rate ($km^3\,y^{-1}$)	Average Discharge Rate ($m^3\,y^{-1}\,m^{-1}$)	Vulnerable Coastline (%)
Lake Erie	895	65	42	0.42	477	31
Lake Huron	1313	24	31	0.44	337	6
Lake Michigan	2355	88	37	0.97	410	21
Lake Ontario	530	45	45	0.16	308	22
Lake Superior	1576	33	35	0.54	345	0.7
Great Lakes	6669	43	36	2.54	381	15

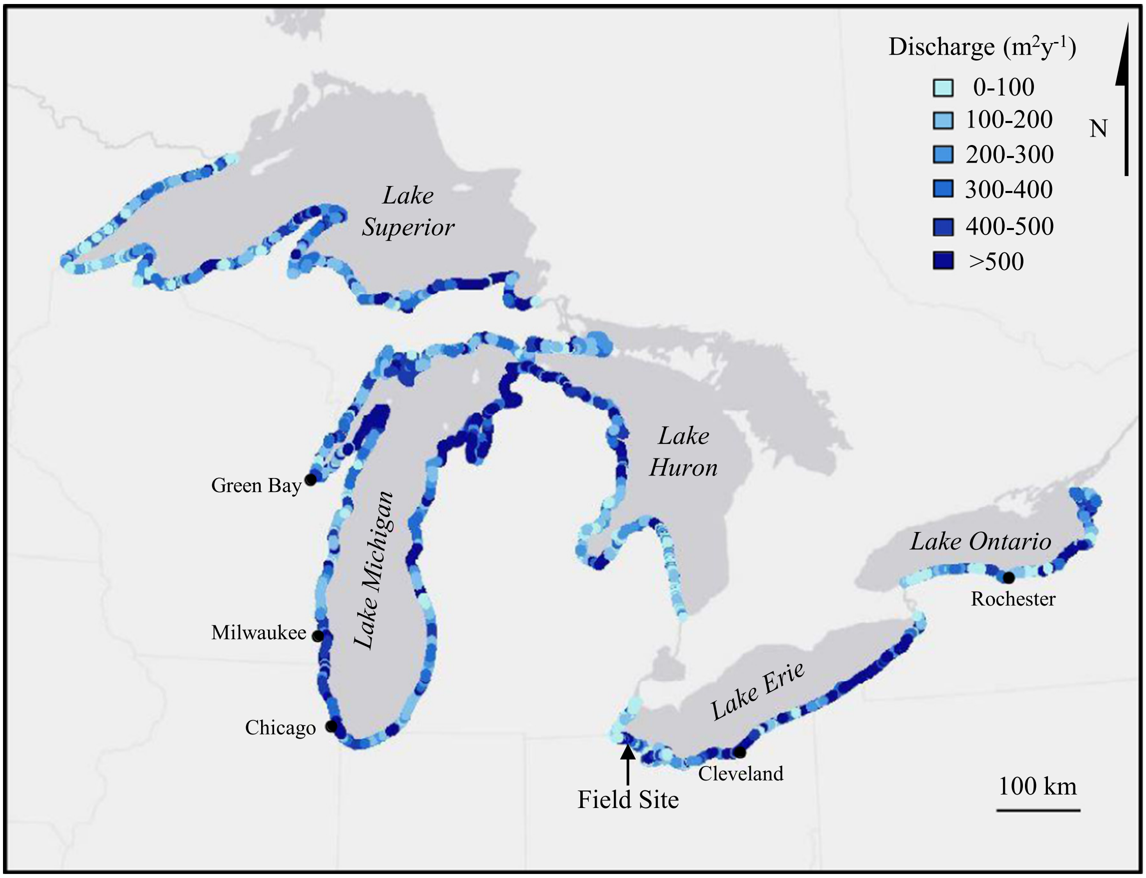

Fig. 3. Direct ground water discharge to the Great Lakes. Discharge rates are high (>500 $m^2 y^{-1}$) along several reaches of coastline; most notably, east and central sections of Lake Erie, northeast Lake Michigan southeast Lake Superior and the southern coast of northwest Lake Huron.

groundwater modeling study by Feinstein et al. (2010). They reported a total volumetric discharge rate of only 0.24 $km^3 y^{-1}$ but suggested that the rate may be up to 3.8 times greater, based on grid sensitivity studies. Both the Lake Michigan model and our water budget analysis predict high discharge rates along the northeast coast of Lake Michigan as well as low discharge rates near Green Bay (compare Fig. 3 and Fig. 72E-in Feinstein et al. (2010)). However, the Lake Michigan model predicts low discharge rates between Milwaukee and Chicago due to municipal groundwater pumping, where our water budget approach incorrectly predicts high rates. Furthermore, a direct comparison of our estimates against the groundwater model for the entire Lake Michigan coast shows no correlation (Fig. 4). A comparison of studies for five other specific sites suggests agreement to within an order of magnitude (Fig. 4). These comparisons underscore the large uncertainties in attempting to estimate direct groundwater discharge rates at any given location.

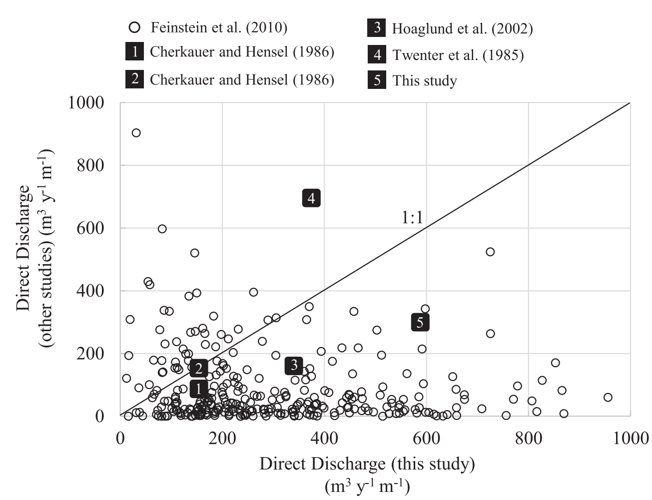

Fig. 4. Direct groundwater discharge to Great Lakes from water budget analysis in this study compared to results from a groundwater model for the Lake Michigan basin (Feinstein et al., 2010), seepage meter and groundwater model, respectively at Mequon, Wisconsin (Cherkauer and Hensel, 1986), groundwater model at Saginaw, Michigan (Hoaglund et al., 2002), groundwater model at East Bay Township (Twenter et al., 1985) and seepage meter observations (this study).

3.3. Vulnerability to groundwater-borne nutrient inputs

Of the 6669 km of United States Great Lakes shoreline, 15% is potentially vulnerable to groundwater-borne nutrients (Fig. 5) based on our criteria (Section 2.2). Lake Erie has the greatest proportion of vulnerable coastline (31%) (Table 1). Much of the area near Cleveland is particularly vulnerable, but localized zones occur throughout Lake Erie's coast. Lake Erie's high vulnerability is reflective of its high concentration of agricultural and developed land uses, combined with relatively high direct groundwater discharge rates. On average, 76% of Lake Erie's coastal recharge areas have an above-average fraction of contaminant-prone land use, compared to 28% for the remaining four Great Lakes combined. The average volumetric discharge rate to the United States Lake Erie coast is 477 $m^3 y^{-1} m^{-1}$, while the average volumetric discharge for the other four lakes is 350 $m^3 y^{-1} m^{-1}$ (28% lower). Vulnerable areas in the other Great Lakes include the southwestern coast of Lake Michigan from Milwaukee to Chicago and numerous

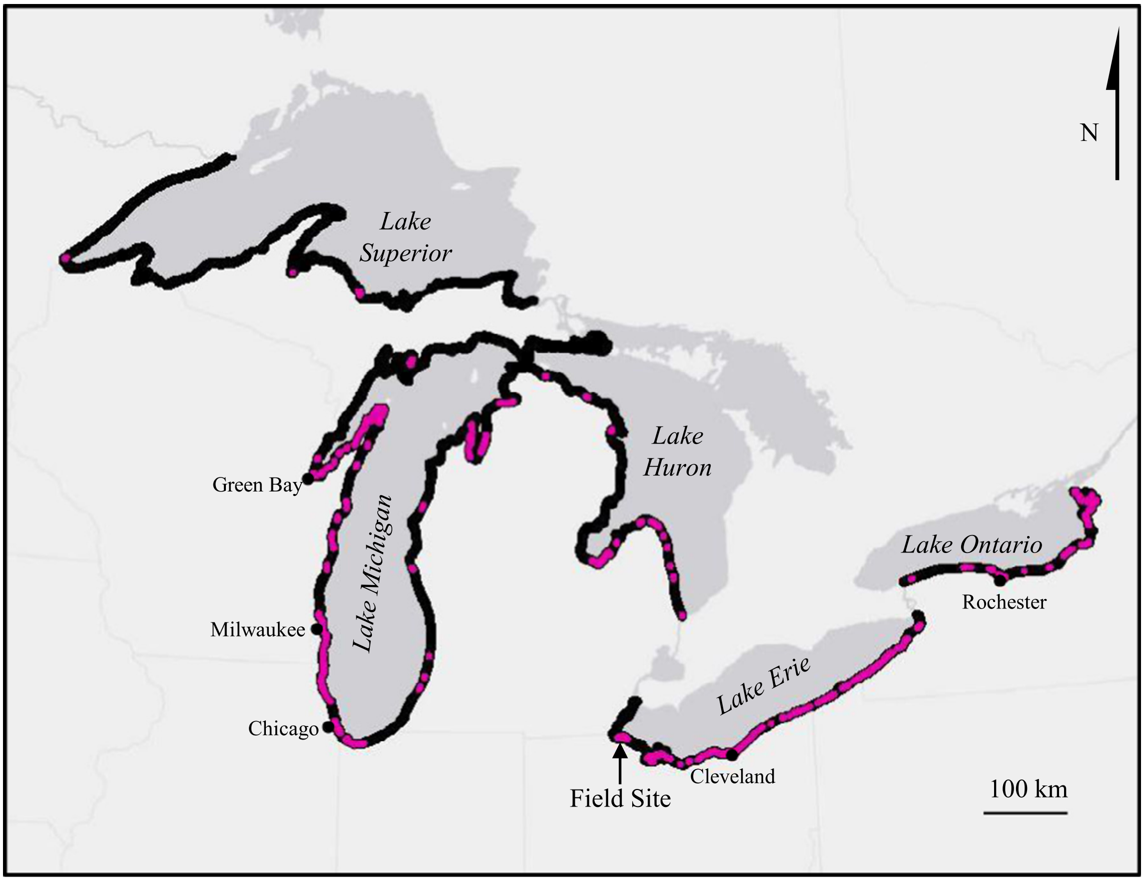

Fig. 5. Vulnerability of Great Lakes to contaminant inputs from groundwater (pink). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

portions of Lake Ontario. Less than 1% of the United States Lake Superior coastline is vulnerable, due to its low rate of direct groundwater discharge, agricultural activity, and development.

4. Field study of a vulnerable beach

4.1. Direct groundwater discharge

Seepage meter measurements indicate net discharge across the study site with a mean rate of 3.15 cm d^{-1} and range of 0.47–10.34 cm d^{-1} (Fig. 2). Net infiltration occurred at only one seepage meter during one round of sampling. Rates were heterogeneous over small spatial scales of meters, although the variation was modest compared to some similar studies (Schneider et al., 2005a; Shaw and Prepas, 1990; Toran et al., 2015). In the nest of four closely-spaced seepage meters, specific discharge rates ranged from 2.9 to 5.1 cm d^{-1}. Along transects, seepage meter rates showed similar trends with distance from the shoreline. Nearshore (0–10 m) rates averaged 8.0 cm d^{-1} and declined offshore (25–30 m) to approximately 1 cm d^{-1}. The offshore trends can be described with an exponential relationship (McBride and Pfannkuch, 1975) (Fig. 6). The best-fit relationship for the specific discharge rate, q [m d^{-1}], as a function of distance offshore, x [m], is:

$$q = 0.0784e^{-0.081x} \quad (1)$$

Along each 20-meter transect, the integrated groundwater flux (using Riemann sum) was 228, 232, and 320 $m^3\,y^{-1}\,m^{-1}$, respectively (average 260 ± 18 $m^3\,y^{-1}\,m^{-1}$). However, groundwater discharge should extend beyond these transects (Bokuniewicz, 1980; Burnett et al., 2006; Russoniello et al., 2013). Integrating Eq. (1), the total volumetric flux of groundwater per unit length of coast is 354 ± 25 $m^3\,y^{-1}\,m^{-1}$. For comparison, the water budget approach yields a greater estimate of 588 ± 181 $m^3\,y^{-1}\,m^{-1}$.

The discrepancy between the two methods can be the result of several factors related to the design of our field campaign. First, heterogeneity in seepage rates occurs at a variety of scales, from meters to kilometers. Our field site spanned approximately 420 m of shoreline, or 7.6% of the segment of coastline over which the water budget was calculated. It is plausible that seepage at our study site was locally low compared to the broader area. Additional measurements are needed to understand how local rates at the study site compare with nearby areas within the same coastal recharge zone, but large-scale studies with many seepage meters are rare because they are labor intensive (Burnett et al., 2006). Second, seepage rates vary over annual and seasonal timescales (Michael et al., 2005; Schneider et al., 2005a). Field measurements were taken at the end of summer and would likely be lower than

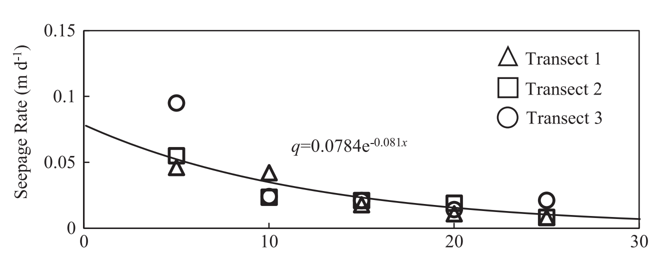

Fig. 6. Seepage rates decrease exponentially from shore at the study site.

the annual average (Michael et al., 2005). Third, and perhaps most importantly, we only deployed seepage meters at the beach and did not measure seepage in the marsh. Nearshore marshes likely intercept a significant portion of direct groundwater discharge. Some of this discharged groundwater may evaporate, but some may flow to the lake through connecting water bodies and should still be considered direct groundwater discharge. Our field-based estimate is likely low because we did not include direct groundwater discharge to the adjacent marsh.

Seepage meter measurements are also prone to errors and uncertainties. Seepage meter measurements may overestimate discharge in high energy environments due to velocity head variations (Libelo and MacIntyre, 1994; Schneider et al., 2005b; Rosenberry, 2008). We did not place our seepage meters in shelters to minimize velocity head effects (Libelo and MacIntyre, 1994; Rosenberry, 2008), but sampling was carried out over relatively calm conditions when currents and waves were minimal. Measured seepage rates can also be reduced by frictional energy losses associated with small diameter plumbing (Rosenberry and Morin, 2004). Our seepage meters employed relatively large-diameter plumbing connections (13 cm inner diameter).

Assumptions made in the water budget analysis may as well be a source of uncertainty in the study. For example, groundwater extraction may deduct from direct groundwater discharge or increase recharge due to lowering of the water table (Konikow and Kendy, 2005; Ferguson and Gleeson, 2012). Groundwater extraction is not accounted for in the NLDAS2 dataset, so we do not seek to include it in our water budget for consistency. However, depletion of groundwater due to extraction is minimal for the majority of the Great Lakes' recharge zones, with western Lake Michigan as an exception (Feinstein et al., 2010; Konikow, 2013). Thus, the assumption that extraction is negligible is valid for the majority of our analyzed areas, including our field site. We also assume that groundwater imports from upland catchments are negligible. However, the significance of flow contributions from upland catchments is difficult to measure (Schaller and Fan, 2009b). Regardless, both of these assumptions would tend to cause underestimation of direct groundwater discharge rates, yet our water budget estimate is greater than the rate from seepage meter measurements.

Despite the inherent uncertainties, both field-based and water budget-based methods have advantages. Unlike water budgets, seepage meters can be used to resolve temporal variability and small-scale spatial variability (Bokuniewicz, 1980; Michael et al., 2005; Russoniello et al., 2013). Meanwhile, water budgets provide a useful tool for examining regional trends in direct groundwater discharge to the Great Lakes system. Similar techniques have been used to map direct groundwater discharge at high resolution over the Baltic Sea and U.S. seaboards or at low resolution over the global oceans (Zektser and Loaiciga, 1993; Destouni et al., 2008). This approach using the NHDPlus data set allows for high-resolution continental-scale estimates over the United States. Because the resolution of the hydrography dataset influences the estimation of direct groundwater discharge, it is not straightforward to merge analyses across multiple hydrographic datasets from different countries. As the coverage of consistent hydrography datasets expands, the water-budget approach can be used to predict global distributions of direct groundwater discharge (Destouni et al., 2008).

4.2. Nutrient concentrations and fluxes

Dissolved phosphorous (DP) concentrations in lakebed pore water were elevated at some locations and averaged 0.12 mg L^{-1} (Fig. 7a, Fig. 8a). For comparison, DP concentrations in lake water, marsh water, and onshore groundwater were all below detection

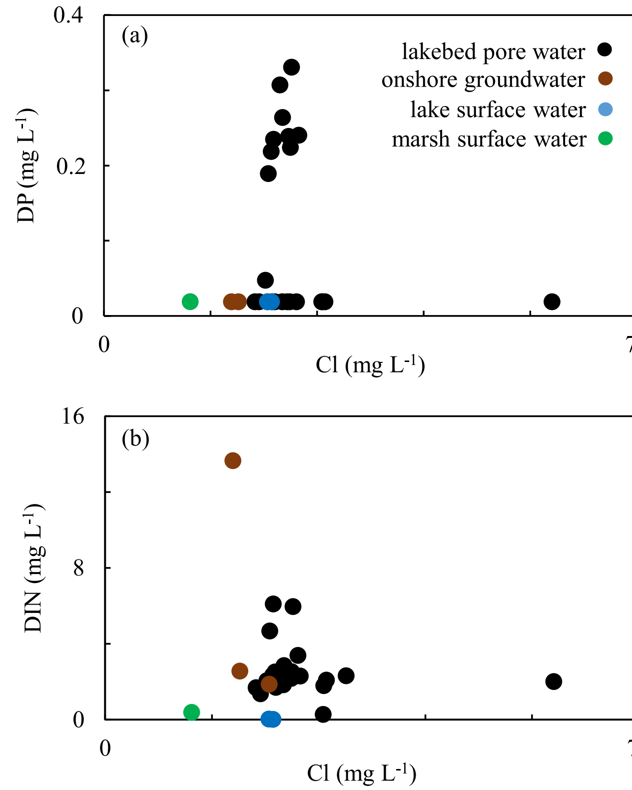

Fig. 7. Nutrient concentrations in water with respect to chloride samples from the Cedar Point field study. Lakebed pore water (black dots) is elevated in DP and low in DIN compared to lake surface water.

(0.019 mg L^{-1}) and reported as the detection limit (Fig. 7a, Table A.1). There were no discernible spatial trends in the locations of elevated lakebed DP (Fig. 8a) and no clear relation to ammonium, DIN, or groundwater fluxes (Table A.1). DP fluxes across the lakebed ranged from near zero, where concentrations were below detection, to 12.94 mg m^{-2} d^{-1}. Integrating along transects, the mass flux of DP per unit length of shoreline was 114.6, 39.7, and 105.0 mg m^{-1} d^{-1} for Transects 1 through 3, respectively (average of 86 ± 6.1 mg m^{-1} d^{-1}).

DIN concentrations averaged 2.57 mg L^{-1} in lakebed pore water (Fig. 8b), which mostly consisted of NH_4^+-N (Table A.1). DIN concentrations were low in lake and marsh waters (0.021 and 0.11 mg L^{-1}, respectively) (Fig. 7b). DIN concentrations in onshore groundwater were similar to or greater than lakebed pore water and averaged 6.0 mg L^{-1} (Fig. 7b). However, most of the onshore DIN consisted of NO_3-N (Table A.1). The source of DIN is unclear. Relatively pristine wetlands fringe the beach, but the broader recharge zone includes cultivated crop beyond the immediate vicinity of the study site. High DIN may be anthropogenic in origin if sourced from these distal agricultural areas, or may be produced locally from mineralization of organic matter followed by nitrification, or both. There was no clear relationship between DIN concentrations and groundwater fluxes (Fig. 8b). DIN fluxes across the lakebed ranged from 15.5 to 323 mg m^{-2} d^{-1}. The mass flux of DIN was 1340, 1300, and 3427 mg m^{-1} d^{-1} for Transects 1 through 3, respectively (average of 2022 ± 141.3 mg m^{-1} d^{-1}).

4.3. Nutrient transformation near the lakebed interface

Lakebeds are reactive interfaces that may serve as sources or sinks of nutrients to lake water (Frape and Patterson, 1981; LaBaugh et al., 1997; Kroeger and Charette, 2008). DP can be

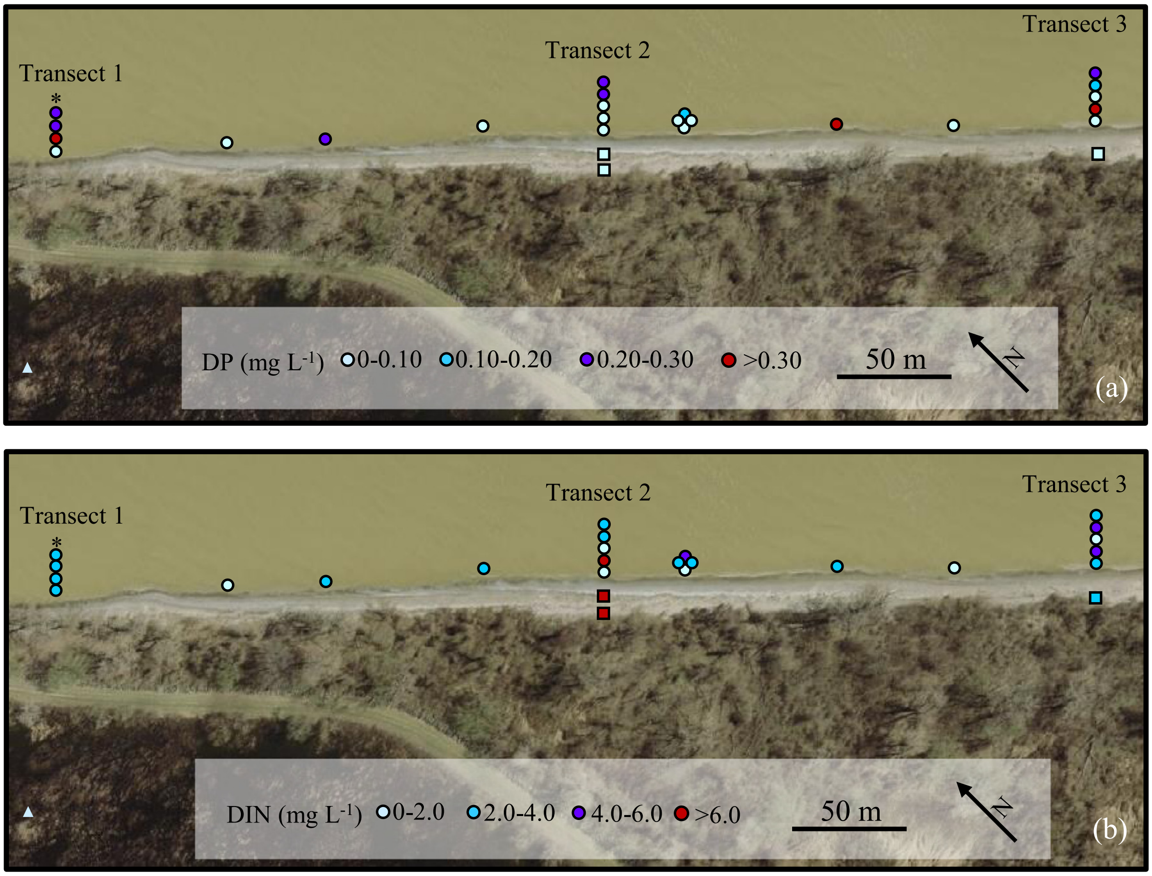

Fig. 8. (a) DP and DIN concentration at each seepage meter. Squares represent onshore wells and the triangle the marsh sample. Seepage meter #1 (asterisk) was not sampled.

sourced from sediments (mineralization of organic matter and desorption from autochthonous deposits), excreted by organisms, or derived from mixing with an unknown high DP water source. In contrast, uptake by plants and sorption to metal oxide minerals may reduce DP concentration (Robinson, 2015). At the study site, DP was negligible in onshore groundwater, lake surface water, and marsh samples, but present in most pore water samples, indicating a benthic source (likely organic matter mineralization and/or desorption of legacy P from mineral surfaces). We did not measure total phosphorous (TP) in lake water, but it is known to be elevated (Chaffin et al., 2011) due to inputs from contributing rivers (Baker et al., 2014). The Maumee River delivers a large sediment load from agricultural areas to Lake Erie, and its mouth is located near the study site. Some of the DP in lakebed pore water may be sourced from legacy P in deposits from the Maumee River (Green et al., 1978).

Reactions along shallow flow paths also influence nitrogen in groundwater (Duff and Triska, 1990; Hester et al., 2014). Nitrate, often the most common form of N in groundwater, is usually stable in aerobic zones, but is removed via denitrification in anoxic conditions or attenuated by microbial and plant uptake (Duff and Triska, 1990; Hill, 1996; Robinson, 2015). One of the onshore groundwater samples at Cedar Point was particularly high in nitrate (Fig. 7b), but the lower concentrations in lakebed porewater suggest that much of the nitrate may be removed before discharging to the lake. Dilution of high-nitrate groundwater with low-nitrate lake water or another low nitrate water source could also cause a decline in nitrate concentrations near the lakebed interface (Altman and Parizek, 1995; Speiran et al., 1998; Spruill, 2000). Using a conservative tracer like chloride, the change in nitrate due to reactions can be isolated from changes due to the mixing of lake water and groundwater, provided that there are no other water sources and the chloride concentrations in the lake water and groundwater are distinct. Unfortunately, chloride concentrations in some pore water samples exceed the concentrations in both lake water and groundwater end members, suggesting either an additional source of chloride is present, or the end-member concentrations vary over time (Fig. 7). The observed chloride concentrations (up to 63 mg L^{-1}) are similar to reported concentrations at another Lake Erie location (Haack et al., 2005). Because we cannot use chloride concentrations to isolate dilution and non-conservative removal of nitrate, we can only speculate that both processes likely contribute to low nitrate concentrations near the lakebed. Given the reducing conditions that are indicated by moderate phosphate and ammonium concentrations (Table A.1), it is likely that some denitrification is occurring. Depending on the rate of this removal process, nearshore sediments along Lake Erie may provide an important ecological service by reducing nitrate prior to discharge.

Our water chemistry data highlight the importance of calculating nutrient fluxes with concentrations measured near the sediment-water interface. Using onshore groundwater end members to calculate nutrient fluxes is common practice but not an accurate reflection of potential fluxes at the sediment-water interface (Schuster et al., 2003). Nutrient chemistry can vary over short distances in the subsurface, particularly near contrasting sediment types or converging flow paths with different limiting reactants (Hill et al., 2000). For example, Gu et al. (2007) showed that a sharp oxidation-reduction gradient within the top 15 cm of sediment at Cobb Mill Creek, Virginia, may be responsible for up to 80% loss of nitrate near the sediment-water interface. Rapid changes in chemistry occur at our study site over tens of meters between onshore piezometers and lakebed sampling locations. Using groundwater end member concentrations to calculate nutrient flux at the study site results in average DP and DIN fluxes of 0.68 and 5300 mg m^{-1} d^{-1} (compared to 86 and 2023 m^{-1} d^{-1}), underestimating and overestimating DP and DIN fluxes, respectively.

4.4. Significance of direct groundwater discharge as a source of nutrients to Lake Erie

An important question is whether groundwater is a significant source of nutrients to Lake Erie. We lack distributed chemical data from around Lake Erie to estimate the total flux of groundwater-borne nutrients. However, if we assume nutrient concentrations in groundwater discharge zones around Lake Erie are similar to those at the field site (average DP and DIN concentrations of 0.12 and 2.57 mg L^{-1}, respectively), based on the average direct groundwater discharge rate (477 m^2 y^{-1}; Table 1), and a total coastline length of 1400 km, the total DP and DIN fluxes are 2.5 and 54.3 g s^{-1}, respectively. For comparison, these fluxes represent 13% of the DP load and 4% of the DIN load to Lake Erie by the Maumee River, which is a major source of nutrients to Lake Erie (Baker et al., 2014; Stow et al., 2015). We note that rivers also carry a large particulate P load, while most particulate P is likely filtered from discharging groundwater. The DP load is especially important because it is more bioavailable than particulate P (Sonzogni et al., 1982). In summary, groundwater is potentially a small but non-negligible source of nutrients to Lake Erie. Unlike discharge from a river, direct groundwater discharge is diffuse and exhibits high spatial heterogeneity. The complexity in quantifying direct groundwater discharge as a nonpoint source of contamination makes management difficult compared to point source loading.

Though small in magnitude, nutrient fluxes from groundwater can influence primary production because N:P ratios are often significantly greater than in river water (Howarth, 1988; Slomp and Van Cappellen, 2004). The Redfield ratio of 16:1 represents an optimal condition for primary production and phytoplankton development (Howarth, 1988). Higher N:P ratios may favor P limitation, and vice versa (Lapointe, 1997; Weiskel and Howes, 1992). Groundwater at our field site delivers a DIN:DP ratio of 147:1, far exceeding the Redfield ratio and contributing to P limitation. In comparison, a N:P ratio of 21:1 was reported in lake water within the western basin of Lake Erie near Maumee Bay during the 2008 algal bloom (Chaffin et al., 2011). Assuming field measurements are representative of average P and N concentrations, discharging groundwater is high in N and has the potential to exacerbate P limitation.

5. Considerations for assessing vulnerability

Our coastal vulnerability map is a useful tool for revealing reaches of shoreline that are susceptible to inconspicuous nutrient loads from direct groundwater discharge. High resolution patterns in vulnerability can be resolved over scales of kilometers. An added benefit is that vulnerability thresholds could be modified to fit different applications. For example, with data on mining activities, thresholds could be selected to map vulnerability to mining-derived contaminants.

Our method does not identify locations susceptible to mobilization of legacy P from lakebed sediments. Phosphorus-laden sediments in groundwater discharge zones may be an important source of DP to pore water and ultimately lake water, especially in areas like our field site near the mouth of the Maumee River. Furthermore, biogeochemical turnover of internal P in lakebed sediment may also contribute to P loads in the lake (Lewandowski et al., 2015). DP contributions from onshore groundwater may be negligible. DP is often relatively immobile in groundwater because it tends to react with cations to form a wide range of metal-complex formations, absorb to sediment and be taken up by plants (Holman et al., 2008; Robinson, 2015). However, growing evidence suggests that the mobility of DP in groundwater has been underappreciated (Crowe et al., 2004; Robertson et al., 2005; Holman et al., 2008; Simonds et al., 2008; Robinson, 2015), and groundwater may sometimes transport consequential amounts of P. Our approach for identifying vulnerable coastlines only considers nutrients from onshore groundwater. A more robust vulnerability assessment would consider both potential DP sources from onshore activities in the recharge zone and DP release from sediments in the discharge zone. Including this second source would require an improved understanding of: 1) distributions of legacy P and potential desorption rates, and 2) distributions of organic matter and potential mineralization rates along the Great Lakes coast.

Our method also has weaknesses in predicting susceptibility to nitrate loading. Zones with high agricultural and developed land uses may not necessitate high N loads. For example, high N in groundwater samples at Cedar Point were expected based on our vulnerability map. However, it appeared that N was attenuated near the lakebed discharge zone. N loading may not be high in vulnerable areas if the shallow aquifer provides an attenuation service.

Spatial heterogeneity and temporal variations also complicate vulnerability assessments. Nutrient leaching from the soil to the water table varies across fine scales that depend on soil type, fertilizer input, climatic conditions, vegetation type, and depth to root zones (Coulibaly and Burn, 2004; Lewandowski et al., 2015). Also, land use is heterogeneous and variable at resolutions finer than the average recharge zone. By design, all areas within a given recharge zone are assumed to have a uniform recharge rate and the entire coastline has a uniform discharge rate. It is likely that areas of high and low vulnerability occur within a given recharge zone due to heterogeneity in fluxes or point sources of nutrients such as leaky septic tanks. Nevertheless, this approach provides a good platform for planning more detailed measurements of nutrient loading via groundwater. Vulnerability maps cannot replace direct field measurements.

6. Conclusion

Water budgets are a simple but powerful tool for revealing patterns of direct groundwater discharge to the coast, which can be used to identify areas of risk for nutrient contamination from groundwater. Of the Great Lakes, Lake Erie has the highest flux of groundwater per unit length of shoreline (477 m^3 y^{-1} m^{-1}) along with the highest percentage of vulnerable shoreline (31%). Lake Superior is the least vulnerable to groundwater-borne contamination with only 1% of its coastline marked as vulnerable. In regions where drawdown due to pumping reduces groundwater flow to the coast, the water budget approach may overestimate direct groundwater discharge. However, this method may also underestimate discharge where regional confined aquifers convey groundwater to the coast. Currently, our approach for mapping vulnerability to groundwater-borne nutrient loads does not consider P sources from the mineralization of organic matter or desorption of legacy P in discharge zones. Vulnerability predictions could be improved with better estimates of P content and potential mobilization rates from lakebed sediments.

The Great Lakes provide drinking water for millions of people in the United States and Canada and serve important economical and recreational purposes. Maps of estimated coastal vulnerability for the Great Lakes are essential for effective management of this fresh water resource. However, there remains limited direct field measurements to validate our approach, and vulnerability maps cannot replace direct field observations of nutrient fluxes from groundwater. A clear need exists for new measurements of groundwater-borne nutrient fluxes to the Great Lakes and other recreational water bodies across a variety of coastal land use types and geologies.

Acknowledgments and data

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jhydrol.2017.09.001.

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