Biogeochemistry (2011) 102:45–58
DOI 10.1007/s10533-010-9420-y

Spatial and temporal variation in phosphorus budgets for 24 watersheds in the Lake Erie and Lake Michigan basins

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Received: 19 October 2009 / Accepted: 17 February 2010 / Published online: 8 March 2010
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Abstract We estimated net anthropogenic phosphorus inputs (NAPI) to 18 Lake Michigan (LM) and 6 Lake Erie (LE) watersheds for 1974, 1978, 1982, 1987, and 1992. NAPI quantifies all anthropogenic inputs of P (fertilizer use, atmospheric deposition, and detergents) as well as trade of P in food and feed, which can be a net input or output. Fertilizer was the dominant input overall, varying by three orders of magnitude among the 24 watersheds, but detergent was the largest input in the most urbanized watershed. NAPI increased in relation to area of disturbed land ($R^2 = 0.90$) and decreased with forested and wetland area ($R^2 = 0.90$). Export of P by rivers varied with NAPI, especially for the 18 watersheds of LM ($R^2 = 0.93$), whereas the relationship was more variable among the six LE watersheds ($R^2 = 0.59$). On average, rivers of the LE watersheds exported about 10% of NAPI, whereas LM watersheds exported 5% of estimated NAPI. A comparison of our results with others as well as nitrogen (N) budgets suggests that fractional export of P may vary regionally, as has been reported for N, and the proportion of P inputs exported by rivers appears lower than comparable findings with N.

Keywords Phosphorus · NAPI · P export · River · Watershed · Nutrient budget · Nutrient loading · Lake Michigan · Lake Erie

Abbreviations

LE	Lake Erie
LM	Lake Michigan
NANI	Net anthropogenic nitrogen input
NAPI	Net anthropogenic phosphorus input
NLCD	National Land Cover Database
RMSE	Root mean squared error

Electronic supplementary material The online version of this article (doi:10.1007/s10533-010-9420-y) contains supplementary material, which is available to authorized users.

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Introduction

Eutrophication of the Great Lakes, the world's largest surface freshwater system, attracted great public attention in the 1970s as Lake Erie experienced hypolimnetic anoxia and other water quality problems associated with phosphorus (P) enrichment

(Rosa and Burns 1987). The 1972 Clean Water Act brought about reductions in point source loadings of P that dramatically lowered annual P loads and reversed many of the effects of cultural eutrophication. Despite apparent early success at reversing eutrophication since the 1960s, periodic hypoxia (dissolved oxygen <2 mg l^{-1}) in the hypolimnion of the central basin of Lake Erie has reemerged as a potential hazard to ecosystem health (Burns et al. 2005). In addition, toxic algal blooms have become more common and persistent in Lake Erie and other lower Great Lakes (Dyble et al. 2008; Ouellette et al. 2006). As a consequence, attention has re-focused on P loading and cycling within the Great Lakes basin.

Nutrient budgets that account for all inputs and outputs to a watershed or other land unit provide an estimate of net nutrient additions, which in turn are an indicator of potential pollution (Russell et al. 2008). Net inputs of new or anthropogenic N or P have been found to correlate strongly with land use, agricultural inputs, and human population density in a number of different regions (Boyer et al. 2002; Goolsby et al. 1999; Howarth et al. 1996; Russell et al. 2008). Further, the amount of N exported by rivers appears to be an approximately linear function of net anthropogenic N inputs (NANI), estimated to be 25% of inputs in studies conducted in watersheds of the northeastern U.S. (Boyer et al. 2002), 21% for all watersheds draining into Lake Michigan (Han and Allan 2008), but only 9% for 12 watersheds of the southeastern U.S. (Schaefer and Alber 2007), suggesting that fractional export (the proportion of NANI exported by rivers) may vary by region.

Analysis of net nutrient inputs and their relationship to river export has proven useful for identification of primary nutrient sources (Boyer et al. 2002), to predict present or future river export (Han et al. 2009; Howarth et al. 2006; McIsaac et al. 2001), and as an overall measure of anthropogenic nutrient loading (Howarth et al. 1996; Russell et al. 2008). N budgets have received greater attention, but P budgets also have been constructed for a variety of systems (Baker and Richards 2002; Borbor-Cordova et al. 2006; Bosch and Allan 2008; David and Gentry 2000; Goolsby et al. 1999; Russell et al. 2008). These previous efforts have several limitations, including little consistency in terms of included fluxes and how system boundaries are defined (Bosch and Allan 2008) and recognized inaccuracies due to omitted fluxes (Baker and Richards 2002; Bosch and Allan 2008). There have been few attempts to predict river P export based on budgeted inputs; however, the recent estimation of net anthropogenic phosphorus inputs (NAPI) to counties and watersheds of the Chesapeake Bay (Russell et al. 2008) marks the first comprehensive effort to determine P inputs and their relation to human-influenced sources and riverine TP export. As the phosphorus version of the widely used N budgeting method, NAPI includes all important anthropogenic P inputs and has the potential to bring consistency to future budgeting efforts.

In the present study we utilize the NAPI approach to construct P budgets for 24 watersheds in the Lake Michigan (LM) and Lake Erie (LE) basins (Fig. 1) over 5 agricultural census years from 1974 to 1992 to quantify the extent of spatial and temporal variation in NAPI and its primary sources. We evaluate the relationship between NAPI and riverine TP export to determine whether NAPI can be used to predict riverine P export, and how the relationship might vary with regional conditions. Land use, climate, and surficial geology were considered as potential drivers for observed NAPI patterns and river P exports. We conclude with a synthesis of NANI and NAPI work to date, focusing on differences in dominant individual NAPI and NANI fluxes and the changing relationship between nutrient inputs and river export for N and P.

Methods

Study area

We selected 18 LM and 6 LE watersheds on the basis of availability of river water quality data, and a range of watershed land use, geological and climatic characteristics, and agricultural management practices (Table 1). The 18 LM watersheds comprise 74% of the entire LM basin, and the 6 LE watersheds cover 37% of the entire LE basin. We delineated the boundaries of the 24 watersheds upstream of the lower-most USGS gauging station (Table 1) using 30-m National Elevation Dataset (Fig. 1). Based on the 1992 National Land Cover Database (NLCD) (MRLC 1995), agricultural land dominates (>70%) LE watersheds, whereas land use in LM watersheds varies widely, exhibiting a general south to north trend from highly agricultural (50–80%) to primarily

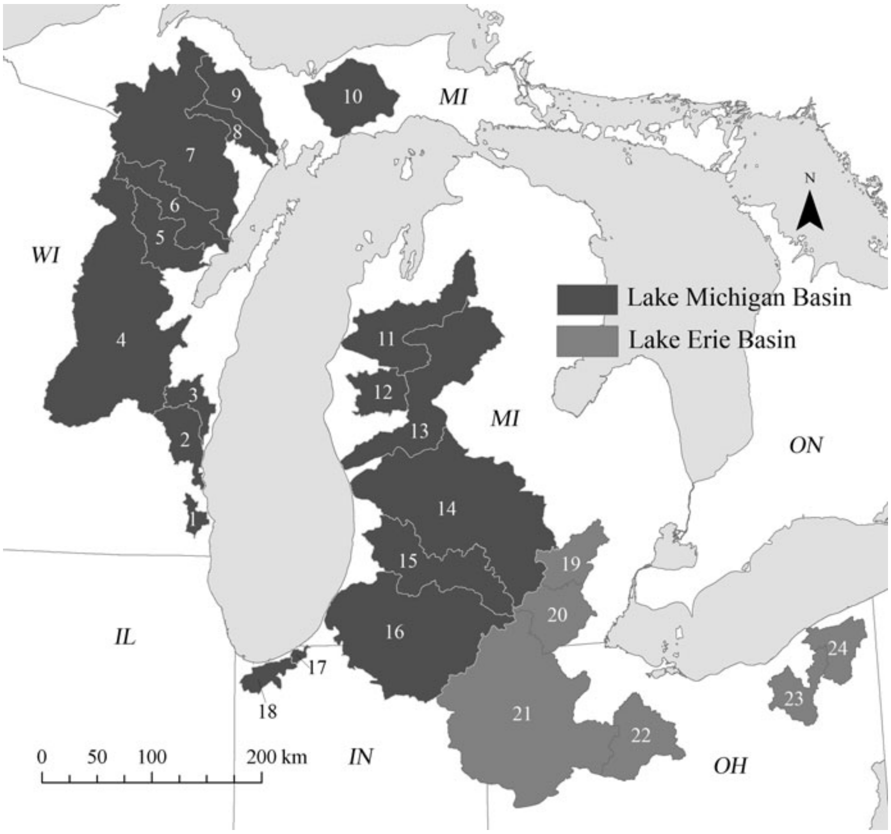

Fig. 1 Locations and areas of the 18 Lake Michigan watersheds and 6 Lake Erie watersheds as defined by furthest downstream USGS gauging station in each case. A legend for ID numbers can be found in Table 1

forested (>70%), with more urbanized watersheds (>18% urban) located in the south and southwestern regions of the basin (Table 1).

The 18 LM watersheds are characterized by unconsolidated surficial deposits of mostly loamy till and sand and gravel (Fullerton et al. 2004) (Table 1). In contrast, most of the LE watershed soils are dominated by clayey till. Thus the LM watersheds and the Huron watershed draining to LE have relatively permeable soils in comparison to the five remaining LE watersheds, consistent with differences in stream flow flashiness throughout the region (Baker and Richards 2002).

Monthly and annual precipitation data were obtained from the PRISM historical climate GIS data set (4 km × 4 km) for 5 years (1974, 1978, 1982, 1987, and 1992) (Daly and Gibson 2002), and grid values within each watershed were averaged using ArcGIS 9.0. Mean annual precipitation over the study period ranged from 780 mm/yr in a northern LM watershed to 970 mm/yr in the southern part of the LM basin, to 1040 mm/yr in the Grand River, Ohio, showing a strong spatial pattern toward greater precipitation along the north-to-south axis (Supplementary Material, Table S1). Mean annual water yield was calculated from USGS gage station data and varied by a factor of 2 during the study from the driest northern watersheds of the LM basin to the wetter southern watersheds of the LE basin.

We aggregated county-level agricultural statistics obtained from the USDA Census of Agriculture (USBC 1977, 1980, 1983, 1989, 1995) and conservation tillage data from the Conservation Technology Information Center (CTIC 2009) to the watershed scale based on agricultural land uses from the NLCD. Major crops of LM watersheds are corn (∼37% of total cropland), alfalfa (∼27%) and soybean (∼14%); LE watersheds differ to a degree, consisting of corn (∼37%), soybean (∼28%), and wheat (∼11%). In 1992, about 34 and 38% of all crop fields in the 18 LM and 6 LE watersheds, respectively, were planted using conservation tillage, a reduced-cultivation method that maintains residues from the past year's crop on 30% or more of the soil surface. However, the heavily cultivated southeastern LM watersheds implemented conservation tillage to a

Table 1 General characteristics of the 18 Lake Michigan and 6 Lake Erie watersheds

Basin	ID	Watershed	State	USGS gauging station	Area (km^2)	Major surficial deposits (%)	Pop. density (km^{-2})	Land use (%)		
								Agr	For	Urb
Lake Michigan	1	Root	WI	04087240	510	C(100)	565	76.7	3.1	19.0
	2	Milwaukee	WI	04087000	1,818	L(68)	117	73.9	7.9	12.2
	3	Sheboygan	WI	04086000	1,106	L(54), C(38)	69	82.0	7.2	2.5
	4	Fox	WI	04084500	15,825	L(37), C(32), SG(27)	32	51.1	27.2	2.4
	5	Oconto	WI	04071000	2,543	L(46), SG(36)	10	27.5	52.1	0.7
	6	Peshtigo	WI	04069500	2,797	L(48), SG(51)	9	20.7	54.7	0.9
	7	Menominee	WI, MI	04066003	10,541	L(68), SG(31)	7	7.1	73.1	0.7
	8	Ford	MI	04059500	1,165	L(90)	3	7.1	53.5	0.2
	9	Escanaba	MI	04059000	2,253	L(67), SG(20)	7	5.4	66.7	1.1
	10	Manistique	MI	04056500	883	SG(62), P(19)	3	5.0	49.5	0.3
	11	Manistee	MI	04126000	4,343	SG(65), L(35)	8	18.3	73.1	1.0
	12	Pere-Marquette	MI	04122500	1,764	SG(55), L(42)	9	17.6	71.2	0.7
	13	Muskegon	MI	04122000	6,941	SG(51), L(36)	18	33.6	47.7	2.8
	14	Grand	MI	04119000	14,292	L(64), SG(26)	85	75.4	13.9	5.5
	15	Kalamazoo	MI	04108500	5,164	L(50), SG(48)	84	75.1	12.6	6.1
	16	St. Joseph	MI, IN	04101500	12,095	L(59), SG(29)	67	80.4	9.3	5.5
	17	Trail Creek	IN	04095300	153	SG(48), C(40)	237	50.0	27.7	18.0
	18	Burns Ditch	IN	04093500	857	C(88)	284	63.7	13.3	21.7
	Lake Michigan basin: area-weighted average					L(46), SG(39), C(12)	50	48.7	34.5	3.6
Lake Erie	19	Huron	MI	04174500	2,343	SG(49), L(41)	222	49.1	27.8	8.9
	20	Raisin	MI,OH	04176500	2,740	C(58), SG(28)	59	79.4	14.1	1.8
	21	Maumee	IN, OH	04193500	16,376	C(84)	50	87.8	7.5	2.6
	22	Sandusky	OH	04198000	3,217	C(89)	37	88.3	9.3	1.3
	23	Cuyahoga	OH	04208000	1,823	C(35), SG(27), L(23)	360	31.0	41.6	17.3
	24	Grand	OH	04212100	1,825	C(94)	62	38.8	44.2	2.0
	Lake Erie Basin: area-weighted average					C(74), SG(11), L(11)	96	77.0	14.6	3.8

Surficial deposit abbreviations are as follows: clay (C), loam (L), sand & gravel (SG), peat (P), and sand (S). Land use classes include row-crop, hayland, and pasture agriculture (Agr), forested (For), and industrial and residential urban (Urb)

greater extent (>50% of combined land area) than the mostly agricultural LE watersheds (e.g. Maumee: 40%; Sandusky: 37%).

Phosphorus budgets

P budgets were developed for each of the 24 watersheds using the Net Anthropogenic Phosphorus Inputs (NAPI) approach (Russell et al. 2008). NAPI is estimated at the watershed scale, considering only newly transported P from outside sources, and excluding internally recycled components such as animal and human P excretion. NAPI estimates the difference between anthropogenic inputs (fertilizer use, atmospheric deposition, imports of P in food and feed, and detergents) and outputs (export of P in food and feed). Our methods (Fig. 2) are similar to those of Russell et al. (2008), although some specific calculations differ including those for net import of P in food and feed, in which we use life cycle-adjusted animal population estimates as developed by Han and Allan (2008) for N budgets, and in the estimation of P detergents.

County-level P fertilizer application for years 1974–1982, 1987, and 1992 were obtained from the USGS Branch of Systems Analysis (Alexander and Smith 1990), USGS Water Resources Division (Battaglin 1994), and USGS National Water-Quality Assessment Program (Ruddy et al. 2006), respectively. County-level fertilizer estimates were aggregated to

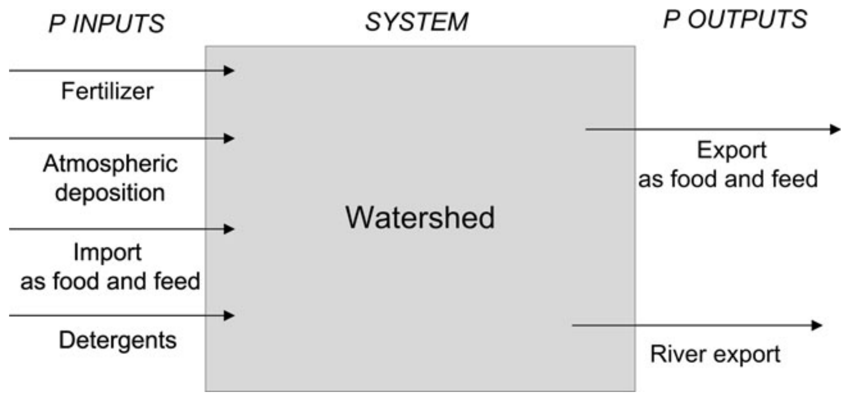

Fig. 2 Conceptual diagram for phosphorus budget including inputs, system, and outputs

the watershed level using area-weighted sums based on the proportion of each county lying within each watershed. Annual atmospheric deposition of TP was obtained for 1974 and 1978 from PLUARG (PLAURG 1977, 1978) and for the years 1982–1992 from the International Joint Commission.

Net trade of human food and animal feed is a flexible flux that can be a source (if net balance is positive) or a loss (if net balance is negative). Net trade of P in food and feed assumes that human and animal food and feed requirements are met by local agricultural production and by imports, and can be estimated by subtracting human and animal consumption from crop and animal production. Any surplus production is assumed to be exported. Crop P production was estimated using a combination of estimates of the P content of major crops (Supplementary Material, Table S2) and county-level crop production data from the Census of Agriculture from 1974 to 1992 (USBC 1977, 1980, 1983, 1989, 1995). Animal P production was estimated by sales data combined with weight of slaughtered livestock (USDA/NASS 2006a, b) and the P content of their edible portion (USDA 2005) (Supplementary Material, Table S3). Human consumption of P was estimated using the yearly population estimates (USBC 1982, 1990) multiplied by human consumption (0.6 kg-P capita^{-1} yr^{-1}) (USDA 2006) plus waste disposal (0.04 kg-P capita^{-1} yr^{-1}) (USEPA 1980) rates. Livestock P consumption was calculated from estimates of average annual populations of livestock (Han and Allan 2008; Kellogg et al. 2000), and P requirements per animal for each livestock group for the corresponding live weight in each year, as recommended by the National Research Council (NRC 1984, 1985, 1998, 2001).

Detergent P for laundry, dish-washing, and industrial use was assumed to be a new input imported from outside each watershed because the principal deposits of phosphate (PO_4) rock, the source material of commercial P products, occur outside the Great Lakes region (USGS 2000). We estimated P import for PO_4-based laundry detergent use from county-level human population multiplied by annual per capita detergent use (6.67 kg capita^{-1} yr^{-1}) and the historical P content of laundry detergent, including changes resulting from a ban on P in detergents that went into effect after 1972 in Michigan and Indiana, 1979 in Wisconsin and 1988 in Ohio (Chapra 1980; Litke 1999). Taking into account an increase in the proportion of households equipped with automatic dishwashers (25% in 1974, 50% in 1987, 90% in 1992) and a decrease in the frequency of dishwasher use over time (7 times/week in 1974 to 5.6 times/week in 1992) (Brenner 1987; Ligman et al. 1974), per capita P dishwashing detergent use was assumed to be 0.33, 0.31, 0.5, 0.74, and 0.5 kg-P capita^{-1} yr^{-1} for the five census years from 1974 to 1992. We obtained estimates of total P loads from industrial or commercial facilities for each watershed from the Great Lakes Water Quality Board (GLWQB 1980, 1981, 1983, 1985, 1987, 1989, 1992). Since atmospheric deposition of P is sufficiently small relative to other inputs of P to watersheds (less than 1% of total P input) (Baker and Richards 2002), many studies ignored this term in their P input calculations (David and Gentry 2000; Goolsby et al. 1999; Russell et al. 2008).

Animal P manure is considered to be an internal cycling term rather than a new P input in NAPI budgets, but this flux is also estimated in this paper for comparison purposes. The methods and assumptions described by Kellogg et al. (2000) were used to estimate the P content of manure produced by various types of livestock (Supplementary Material, Table S4).

Riverine TP concentration and fluxes

For the 18 LM and 6 LE watersheds we estimated annual river TP export loads for the five census years from 1974 to 1992 based on approximately monthly TP concentrations (USEPA 2007) and daily discharges (USGS 2008) using the USGS Estimator Regression Model (Cohn et al. 1989; Richards 1998). In addition, daily TP concentrations were summed to estimate annual TP loads for 5 LE tributaries for various time periods including the Raisin for 1992, Maumee for 1978–1992, Sandusky for 1978–1992, Cuyahoga for 1982–1992, and Grand for 1992 (NCWQR 2008).

Results

Phosphorus budgets

For all census years from 1974 to 1992, fertilizer was the largest individual source of P for most of the 24 watersheds of Lake Michigan and Lake Erie (Table 2). However, net import of P in detergents was the largest source in the urban Cuyahoga watershed. The amount of P fertilizer applied varied greatly across the 24 watersheds, ranging from 10 kg-P km^{-2} yr^{-1} in the Escanaba watershed to 1,858 kg-P km^{-2} yr^{-1} in the Sandusky watershed. This variation was strongly correlated with acreage of harvested corn plus wheat ($R^2 = 0.95$) as a percent of each watershed, and less strongly with percent total

Table 2 Complete NAPI budgets for each watershed averaged across 5 agricultural census years (1974, 1978, 1982, 1987, 1992)

Key	Watershed name	Atmos. Dep.	Fert.	Net import in food	Net import in feed	Detergents	NAPI	Manure	Crop export
1	Root	6	648	254	−137	415	1186	194	459
2	Milwaukee	6	793	−78	120	92	934	558	605
3	Sheboygan	6	1130	−189	146	62	1156	678	708
4	Fox	6	574	−119	62	25	548	391	449
5	Oconto	6	277	−72	62	8	280	236	225
6	Peshtigo	6	136	−25	33	6	155	97	88
7	Menominee	6	42	−7	14	6	61	39	36
8	Ford	6	88	−4	20	8	117	42	36
9	Escanaba	6	10	6	6	9	36	9	7
10	Manistique	6	14	1	6	2	28	8	7
11	Manistee	6	85	−4	23	9	119	44	41
12	Pere-Marquette	6	329	−14	49	10	380	88	96
13	Muskegon	6	184	−29	56	9	226	125	122
14	Grand (MI)	6	786	−41	42	55	848	234	425
15	Kalamazoo	6	704	−36	93	45	812	200	391
16	St. Joseph	6	1068	−68	15	35	1056	243	662
17	Trail Creek	6	1323	−30	−465	35	869	165	802
18	Burns Ditch	6	1626	89	−426	142	1438	87	637
Avg. area-weighted (LM)		6	501	−50	38	31	558	204	314
19	Huron	14	548	128	−46	182	826	121	269
20	Raisin	14	1203	−42	−262	45	959	160	623
21	Maumee	14	1460	−90	−315	43	1112	154	875
22	Sandusky	14	1858	−89	−533	42	1291	116	908
23	Cuyahoga	15	244	194	25	648	1125	94	124
24	Grand (OH)	15	336	13	85	117	566	176	154
Avg. area-weighted (LE)		6	1310	−42	−265	99	1112	145	710

Manure and crop export fluxes are included here for reference but are not included in budgets. All units in kg-P km^{-2} yr^{-1}

cropland ($R^2 = 0.71$). Fertilizer P inputs declined after 1982, although this trend was not statistically significant ($R^2 = 0.57$, $p = 0.14$).

Average import of detergent P ranged from <10 kg-P km^{-2} yr^{-1} in forested LM watersheds to 648 kg-P km^{-2} yr^{-1} in the urbanized Cuyahoga watershed in the LE basin (Table 2). Detergent P inputs showed a positive relationship with urban area ($R^2 = 0.61$, $p < 0.0001$). For all watersheds, no statistically significant temporal trends were apparent in P imported in detergents from 1974 to 1992. However, all Lake Erie watersheds in Ohio experienced a sharp drop in P detergent use after 1988 following enactment of the ban on P-based detergents. Atmospheric P inputs to watersheds were negligible compared with other sources (Table 2).

Net imports of P in food and feed varied widely across watersheds (Table 2). The amount of P consumed by local livestock (i.e., animal P consumption) was greatest in the agricultural Sheboygan (831 kg-P km^{-2} yr^{-1}), followed by the Milwaukee (705 kg-P km^{-2} yr^{-1}) and St. Joseph (638 kg-P km^{-2} yr^{-1}) watersheds, and these three watersheds had much higher animal P consumption than the ten other primarily agricultural watersheds of LM and LE, which ranged from 141 to 484 kg-P km^{-2} yr^{-1}. Watersheds where more than ~40% of the land area was cultivated in major crops such as corn, soybean, and wheat exported surplus crop P in excess of animal P demand. Import of P in human food was largest in urbanized watersheds including the Root, Huron, and Cuyahoga. Across the five census years, forested watersheds imported relatively small amounts of P as food and feed (<50 kg-P km^{-2} yr^{-1}).

Summing all net inputs of P (atmospheric deposition, fertilizer, net import of food and feed, and detergents), NAPI for the 18 LM watersheds averaged over 1974–1992 ranged from 28 kg-P km^{-2} yr^{-1} (Manistique) in a northern forested region to 1,438 kg-P km^{-2} yr^{-1} (Burns Ditch) in a southern LM agricultural and urbanized watershed (Table 2). Average annual NAPI to six watersheds of LE basin ranged from 566 kg-P km^{-2} yr^{-1} in the least disturbed watershed (Grand, Ohio) to 1,291 kg-P km^{-2} yr^{-1} in a highly agricultural watershed (Sandusky). Net P inputs to each watershed correlated with land use, showing strong relationships with area in agriculture (Fig. 3a, $R^2 = 0.81$) and with urbanized and agricultural lands (Fig. 3b, $R^2 =$ 0.90), and a negative relationship with forested land and wetland (Fig. 3c, $R^2 = 0.90$). There was a weaker but statistically significant positive correlation with urban land alone ($R^2 = 0.40$, $p = 0.001$).

NAPI declined over the study period (linear regression between NAPI and year, $p = 0.005$), showing strong downward trends for a majority of

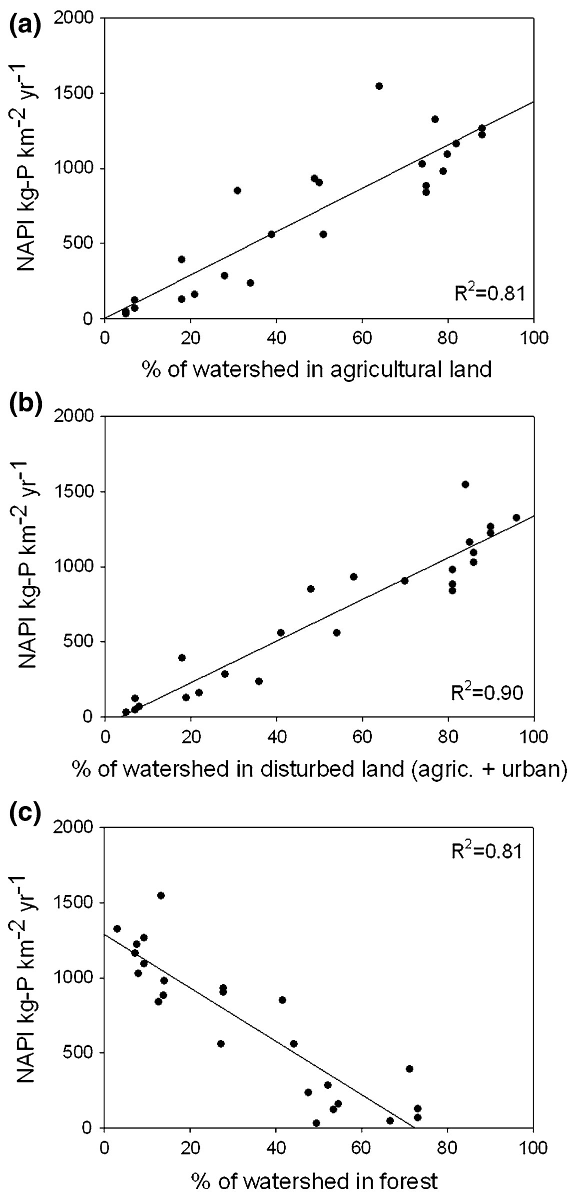

Fig. 3 Phosphorus inputs to the 24 LM and LE watersheds are strongly related to land use, showing a positive relationship with land in agriculture (**a**), a positive relationship with land in agriculture and urban use combined (**b**), and a negative relationship with forested land (**c**)

the disturbed watersheds of LM basin and five LE watersheds, with R^2 values generally in the range of 0.8. This can be attributed to declines in P fertilizer application in agricultural watersheds as well as in P detergent use within urbanized watersheds. However, NAPI was unchanged over time for all forested LM watersheds and the most forested LE watershed (Grand in Ohio); in addition, three of the more disturbed LM watersheds (Milwaukee, Sheboygan, St. Joseph) showed no temporal change.

Riverine TP exports

Annual riverine export of phosphorus varied widely across watersheds and years (Fig. 4). For the 18 LM watersheds annual TP export ranged from less than 6 kg-P km^{-2} yr^{-1} in the forested Ford to 80 kg-P km^{-2} yr^{-1} in the urbanized and intensively cultivated Burns Ditch (Fig. 4a). TP exports were much higher for the six LE watersheds, ranging from 30 kg-P km^{-2} yr^{-1} in the Huron to 159 kg-P km^{-2} yr^{-1} in the urbanized Cuyahoga (Fig. 4b).

Phosphorus export was nonlinearly and positively related to disturbed area (agriculture plus urban), explaining 67% of variation in log-transformed river TP exports when all watersheds and years are pooled. Annual water discharge explained temporal variation in river TP exports between years for only the Milwaukee, Sandusky, Escanaba, and Maumee watersheds ($p < 0.05$), and there was no significant relationship between water discharge and spatial variation in river TP exports among the 24 watersheds ($p > 0.15$ for all years). Similarly, mean annual precipitation was not significantly related to temporal variation in river TP exports for all watersheds and spatial variation in river exports across 24 watersheds for most of years except for 1992 ($R^2 = 0.51$, $p < 0.0001$).

In contrast to the downward temporal trend in NAPI, river TP exports showed no temporal trend in the six LE tributaries or in the majority of LM rivers across the five census years from 1974 to 1992. However, downward trends in river TP exports were evident for six LM tributaries (Sheboygan, Ford, Escanaba, Kalamazoo, St. Joseph, Burns Ditch) over 1974–1992 (based on a linear regression between river TP export and year for individual rivers, $p < 0.05$).

Relating NAPI to river export

Spatial relationship between NAPI and river export across watersheds

For the 18 rivers draining into LM, NAPI consistently accounted for a high fraction of spatial variation in

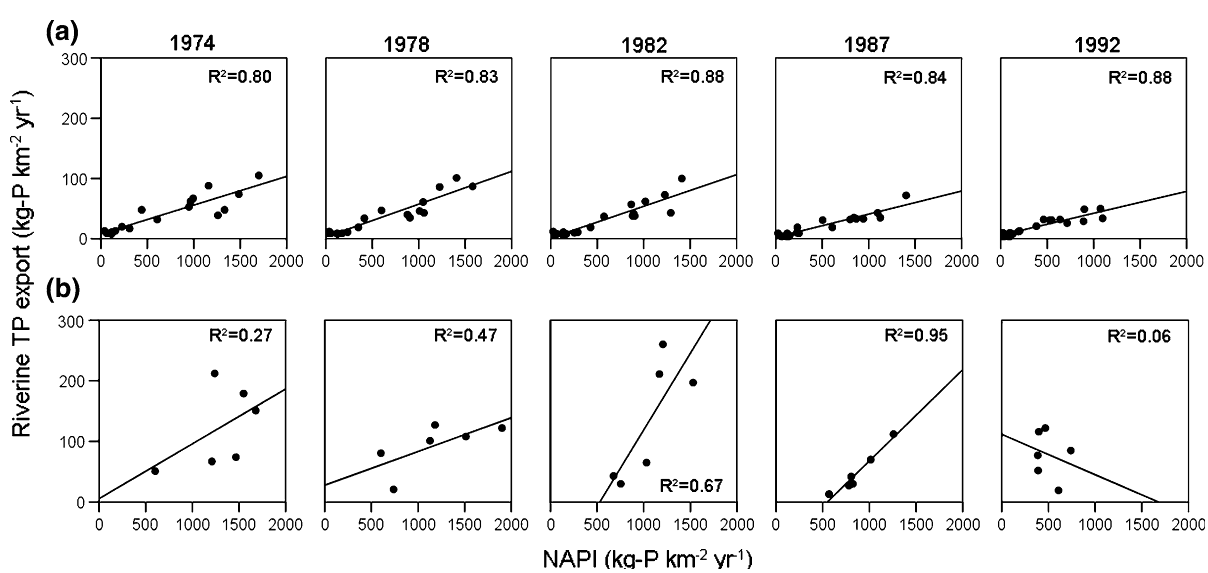

Fig. 4 Relationship between NAPI and river total phosphorus (TP) export for Lake Michigan (**a**) and Lake Erie (**b**) watersheds across the 5 agricultural census years

riverine TP exports for each of the 5 agricultural census years (Fig. 4). Average annual data from the 18 LM watersheds resulted in a strong linear relationship between NAPI and river TP exports ($R^2 = 0.93$). In addition, the linear regression of P export with NAPI had higher precision as measured by root mean squared error (RMSE = 11 kg-P km^{-2} yr^{-1}) compared to individual P input terms (RMSE range = 15 (fertilizer) − 25 (net import of P for food and feed) kg-P km^{-2} yr^{-1}). Among individual P input terms, fertilizer ($R^2 = 0.62$–0.82, $p < 0.01$) and net import of detergents ($R^2 = 0.25$–0.45, $p < 0.05$) were significantly related to river P export; however, net import of P in food and feed was not ($R^2 = 0.03$–0.2, $p > 0.05$).

In contrast to LM watersheds, spatial relationships between river TP exports and NAPI for the six LE watersheds were less consistent across years ($R^2 = 0.06$ to 0.95, Fig. 4). Only the driest year (1987) showed a strong positive correlation ($R^2 = 0.95$, $p < 0.01$). In addition, no individual P inputs were correlated with river TP exports from the six LE watersheds.

Slopes of the regression between NAPI and riverine TP exports including all 18 LM watersheds varied significantly by census year, ranging from 0.036 in 1992 ($p < 0.0001$) to 0.054 in 1974 ($p < 0.0001$) (Fig. 4), indicating that on average, 3.6–5.4% of P inputs to watersheds are exported by rivers. LE watersheds, in contrast, exported a greater fraction of their P inputs over time, ranging from 5.6% in 1978 ($p = 0.133$) to 25.3% in 1982 ($p = 0.053$). However, the LE slope coefficients were not statistically significant except for 1987, when 15% of P was exported ($p < 0.001$).

After averaging across years, a strong predictive relationship was evident between P inputs to watersheds and river TP export for the 18 watersheds of the LM basin ($R^2 = 0.93$, River TP export = 0.048 × NAPI + 3.69, $p < 0.01$). The relationship for the six LE watersheds was more variable ($R^2 = 0.59$, River TP export = 0.164 × NAPI − 65.32, $p = 0.49$) and had a negative intercept. However, neither intercept was statistically different from zero ($p = 0.133$ for LM, $p = 0.49$ for LE). We then suppressed intercepts, as was also done by Russell et al. (2008) in a similar study of the Chesapeake Bay region, in order to compare fractional P export (i.e. proportion of NAPI exported by rivers) across regions and studies.

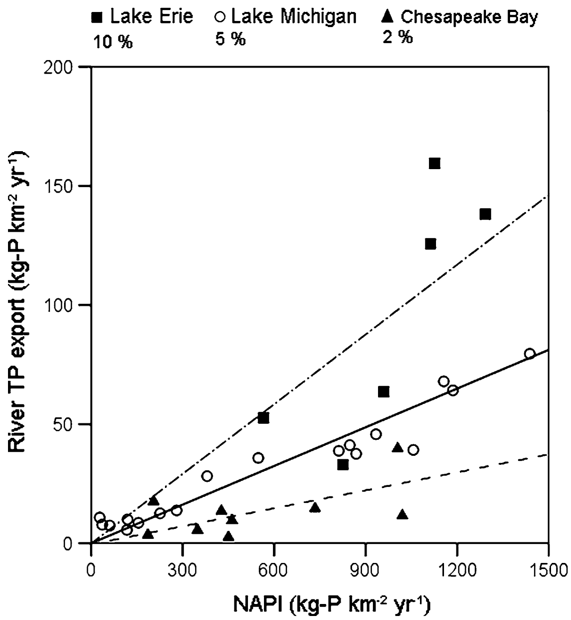

Fig. 5 Regression relationship between river total phosphorus export and NAPI for individual watersheds of the Lake Erie, Lake Michigan, and Chesapeake Bay basins. Lake Erie and Lake Michigan data points are from the present study and represent averages for each watershed across the 5 agricultural census years (1974, 1978, 1982, 1987, 1992). Chesapeake Bay data points come from Russell et al. (2008) and represent 9 primarily forested watersheds. A linear regression was used to fit the data for the three basins without intercepts to allow comparison of fractional delivery among regions

This analysis indicates that approximately 5% of P inputs on average are exported from watersheds of Lake Michigan, compared with ~10% for Lake Erie and 2% for Chesapeake Bay watersheds (Fig. 5).

Temporal relationship between NAPI and river export within a watershed

Neither NAPI nor any individual P input terms explained a significant fraction of the temporal variation in river TP exports across the five census years for most LM and LE watersheds (Supplementary Material, Fig. S1). However, the Burns Ditch ($R^2 = 0.84$, $p < 0.05$) was an exception, showing a high positive correlation across years between NAPI and P export. For most of LM watersheds, this fractional export did not widely vary by year; however fractional export varied substantially across years in LE watersheds, especially the Maumee and Sandusky. In addition, for these two rivers temporal variation in fractional delivery of NAPI was explained well by annual precipitation in the Maumee ($R^2 = 0.995$, $p < 0.001$) and Sandusky ($R^2 = 0.995$, $p < 0.001$).

Discussion

Estimation of net anthropogenic phosphorus inputs to watersheds of the Lake Michigan and Lake Erie basins during 1974–1992 demonstrates much variation in NAPI fluxes, river P loads, and the relationship between NAPI and river P export. Fertilizer application was the dominant P input overall, varying by three orders of magnitude across the 24 watersheds in the study. NAPI increased in relation to area of disturbed land area ($R^2 = 0.90$) and decreased with forested and wetland area ($R^2 = 0.90$), as expected since anthropogenic land use leads to more P inputs and more natural land area indicates less anthropogenic land use. River TP loads varied less than NAPI estimates over the study period, but river TP loads were also positively related to disturbed land ($R^2 = 0.67$), though non-linearly.

NAPI declined over time in the majority of disturbed study watersheds, due primarily to a reduction in P fertilizer use, especially in agricultural watersheds, as well as a decline in detergent P use in urbanized watersheds. However, declines in river export of P over 1974–1992 were observed in only six LM watersheds, and there was not a consistent temporal relationship between P export and NAPI across all watersheds. In addition, no consistent relationships were observed between river TP loads and water discharge or precipitation across all watersheds and years. The 5-year spacing of agricultural census data as well as availability of river export data limits our ability to explore temporal trends. Regional and national studies of river P concentrations have reported more downward than upward trends from 1975 to 1994 (Alexander and Smith 2006) and no significant trend over 1993–2003 (Sprague and Lorenz 2009), with regional and local exceptions in both directions. Thus the relatively weak temporal relationships found in this study may be due to the limited availability of temporal data as well as weak overall trends in the concentrations and fluxes of riverine nutrients for the time period evaluated. It is also possible that legacy sediments and associated P from historical land or stream channel alterations may still be impacting riverine P loads to some extent and irrespective of current conditions.

Different land use practices and human activities strongly influence NAPI estimates and individual fluxes, underlying differences observed between LM and LE watersheds. The average fertilizer application rate in LE watersheds was nearly three times greater than LM watersheds (Table 2), which is attributable to more intensive row-crop agriculture in the LE basin (Table 1). In contrast, manure application rates were higher on average in LM basin (Table 2), which had more livestock relative to LE watersheds. The strong positive correlation between NAPI estimates and disturbed land cover can be explained by greater fertilizer inputs in agricultural areas and greater detergent inputs in urban areas. The strong negative correlation between NAPI and natural area is due to both the absence of P inputs in these areas and possibly the presence of P storage processes as well.

Other studies support our individual input and NAPI results. Previous P budgeting work has found fertilizer application rates to range from 101 to 1550 kg-P km^{-2} yr^{-1} in the Midwestern U.S. (Baker and Richards 2002; Bosch and Allan 2008; David and Gentry 2000; Goolsby et al. 1999), thus the range of 10–1858 kg-P km^{-2} yr^{-1} from the present study seems reasonable given the wide variation in land use practices in LM and LE watersheds. Prior studies also confirm the dominant contribution of fertilizer inputs in watershed P budgets. Using values from previous P budgeting studies in the Midwest and recalculating net inputs similar to the NAPI approach, we find that the present study's range for NAPI from 28 to 1438 kg-P km^{-2} yr^{-1} is consistent with a literature range of 14–956 kg-P km^{-2} yr^{-1} (Baker and Richards 2002; Bosch and Allan 2008; David and Gentry 2000; Goolsby et al. 1999). NAPI estimates for the Chesapeake Bay region of the U.S. reported values from 2 to 7846 kg-P km^{-2} yr^{-1} among 266 counties in the study region, with an average value of 452 kg-P km^{-2} yr^{-1} (Russell et al. 2008). Their higher values of NAPI relative to ours likely are due to the use of much smaller study units (counties), which may capture greater small-scale variation, as well as the inclusion of highly urbanized counties with high population densities such as Philadelphia. In the present study and all previous work, NAPI has been found to be smallest in forested areas, much higher in agriculturally dominated areas, and highest in concentrated urban areas. In addition, such differences in NAPI estimates between the two studies may be derived from difference in the methods and parameters to estimate livestock P consumption. Results from Russell et al. (2008) are higher especially in high milk cow production areas due to their higher rates of livestock P consumption for milk cows compared to ours (Table S4).

NAPI as a predictor of river P loads

For the 18 LM watersheds, NAPI explained 93% of spatial variation in river TP exports among the 18 LM watersheds, and 5% of anthropogenic P was exported from rivers. In contrast, net P inputs explained only 59% of variation in river TP export for the 6 LE watersheds, and the river export rate was higher at $\sim$10% (Fig. 5). It should be emphasized that these estimates are derived from averages over multiple census years; estimates for individual census years can vary considerably. For example, river P export as a fraction of NAPI varied across years from only 3.6 to 5.4% for LM watersheds, but from 5.6 to 25.3% for LE watersheds. Variation in river P exports and fractional P exports among years was strongly dependant on annual precipitation for two of the LE watersheds, the highly agricultural Maumee and Sandusky.

Differences in fractional export of P within and among regions may be attributable to a number of factors, including different land cover, soils and climate as well as agricultural practices. In addition to the likely influence of land cover differences between the LM and LE basins on NAPI fluxes, these two basins also differ markedly in their surficial geology, which may affect how well NAPI can serve as a predictor of P in river export. The LM watersheds have mostly loam, sand and gravel soils, whereas LE watersheds are dominated by clay soils (Table 1). The poorly drained soils of the LE basin are a possible explanation for our finding of a weaker relationship between NAPI and river TP export, as well as a higher fractional export. High clay content and poor drainage result in flashier river discharge (Baker and Richards 2002), which may enhance P export in particulate form. This geology also necessitates tile drainage, which in turn may favor greater export of dissolved P. In Illinois watersheds, Royer et al. (2006) found that dissolved reactive phosphorus loads were greater in streams with tile drainage, and also that high river discharges ($\geq$90th percentile) accounted for over 80% of annual P loads. Thus, it is reasonable to surmise that high river discharge events impact annual P loads more in these LE watersheds compared to the LM watersheds. These hydrologic impacts in the LE watersheds then likely mask relationships between NAPI and riverine P export.

For nine watersheds that make up 93% of the nontidal drainage of the Chesapeake Bay, Russell et al. (2008) estimated that net anthropogenic P inputs accounted for 34% of the spatial variation in riverine TP fluxes, and about 2% of NAPI was discharged from these watersheds. Russell et al. suggest that their low estimate of fractional exports may be due to the small scale of their watersheds, and thus less variation in land use patterns, agricultural production systems and human activities; overall the Chesapeake Bay watersheds were more forested and less agricultural than either LM or LE watersheds. In fact, the highest percentage of land in row crops was 29% in the Chesapeake watersheds, whereas ten of the 24 LM watersheds had >30% agricultural land (maximum of 82% in the Sheboygan), and LE watersheds ranged from 31 to 88% agriculture. The presence of more forest and less agricultural land may explain why Chesapeake Bay watersheds would retain a larger proportion of watershed P inputs compared to LM or LE watersheds.

Fractional P export was highly sensitive to interannual variation in precipitation for the two high-yielding and highly agricultural LE watersheds. Their fractional export varied from 5 to 29% (Maumee) and from 7 to 26% (Sandusky), in accord with variation in annual precipitation. Such higher fractional exports from the Maumee and Sandusky compared to other watersheds and high sensitivity of fractional exports to climate is likely to be driven by several factors. The integrated effects of the relatively high clay content of soils (Table 1), which slows infiltration of rainfall into the ground; the substantial buildup of P in clay soil due to long term net P accumulations from high fertilizer applications; and manure applied for intensive cultivation (Baker and Richards 2002; Calhoun et al. 2002) may all contribute to the flushing of high quantities of P attached to clay into rivers during high flow events (Baker 1993; Royer et al. 2006).

Comparison of NAPI with NANI

Similarities in the NANI and NAPI approaches and literature provide an interesting opportunity to compare patterns observed using both approaches. In previous NANI work, different sources have been shown to be important to overall fluxes (Boyer et al. 2002; Han and Allan 2008) depending on land cover characteristics of different watersheds. In forest-dominated watersheds, atmospheric deposition of N is the largest input in all cases (Boyer et al. 2002; Han and Allan 2008) whereas N imported as human food is the largest term in urbanized watersheds (Boyer et al. 2002). In agricultural-dominated watersheds, N fixation in cropland and fertilizer application are the most important N inputs (Boyer et al. 2002; Han and Allan 2008). This study and prior work (Russell et al. 2008) have found fertilizer consistently to be the largest P input to nearly all watersheds. Nitrogen budgets include an added layer of complexity with gaseous forms of N and much higher atmospheric deposition and re-volatilization rates relative to P. In addition, most of the N is exported in dissolved form, while most of the P is exported in particulate form. Thus, nutrient management strategies in targeted watersheds may be very different for N and P load reduction goals.

Despite these differences in budget terms, both NANI and NAPI have shown promise for predicting riverine nutrient export. Linear regression analyses have estimated TN river export to be about 20–25% of NANI in Northeastern U.S. and Lake Michigan (Boyer et al. 2002; Han and Allan 2008). However, an export estimate from the southeastern U.S. gave a lower value of 9% (Schaefer and Alber 2007), indicating that fractional N delivery may vary regionally. The present study and Russell et al. (2008) provide the first similar estimates for fractional P export at the regional scale, and place river TP export between 2 and 15% of NAPI (Fig. 5) with a range of correlation strengths ($R^2 = 0.34$–0.93). It appears then that rivers export a smaller proportion of watershed P inputs compared to N inputs. This pattern has been observed in previous research as well (Bosch and Allan 2008; David and Gentry 2000), and may reflect the capacity of P to adsorb readily onto soil particles.

The ability to predict riverine P or N export from anthropogenic loading estimates aids in the assessment of sources and strengthens the case for employing land use as a ready indicator of nutrient export to receiving waters. However, regional variation in input-export relationships is in need of further study to determine the range of fractional export estimates and the conditions that may result in this variation. In addition, to enhance confidence in our ability to forecast or hindcast nutrient export from loadings, when feasible it will be important to expand the timeframe over which temporal correlations are examined.

Acknowledgements We thank Dave Dolan for data and discussions regarding river P concentrations, Ashley Burtner for assistance with data management and Peter Richards and Donald Scavia for reviews of an earlier draft of the manuscript. This is contribution 09-007 of the Lake Erie Ecofore study funded by NOAA award NA07OAR432000.

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