Title: **Watershed buffering of legacy phosphorus pressure at a regional scale: A comparison across space and time**

Shortened Title: **Watershed phosphorus buffering**

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Abstract

Phosphorus (P) plays a crucial role in both agricultural productivity and water quality. There has been growing recognition of the importance of 'legacy' P (that which has accumulated in watersheds over time), for understanding contemporary water quality outcomes; however, little is known about how different watersheds respond to cumulative P pressures. The "buffering capacity" concept describes the ability of watersheds to attenuate P loading to surface waters by retaining cumulative P inputs over time. To explore the role of various watershed characteristics in buffering capacity, we compared Net Anthropogenic P Input (NAPI) estimates to riverine total P flux across a thirty-year time span (1981-2011) in 16 large watersheds in southern Quebec, Canada. We used this historic P data to calculate indices describing long- and short-term buffering for these watersheds. We then examined the correlation between this buffering capacity and a set of key geochemical, hydrological, landscape, and socio-ecological variables that were hypothesized to influence P buffering dynamics. Both long- and short-term buffering metrics were most strongly correlated with hydrologic characteristics, indicating that watershed hydrology may be the most prominent characteristic in P buffering within watersheds. However, we found that considering estimates of long-term P accumulation along with biophysical characteristics of the watershed (including hydrology) predicts water quality better (R^2=0.69) than either factor would alone (R^2=0.35). Our findings provide a step towards improving models of watersheds' unique relationships to P pressure and can help guide management of historically agricultural landscapes with considerable amounts of legacy P.

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Manuscript Highlights

- Watershed buffering is a concept used to explore how watersheds respond to legacy P pressure
- Hydrology and landscape features play major role in watershed buffering
- Combining P data and watershed characteristics gives a comprehensive picture of watershed buffering

Key Words

Phosphorus, watershed, agriculture, historical legacy, land use, hydrology, soil P, nutrient budgets

Introduction

The application of mineral phosphorus (P) fertilizers derived from non-renewable phosphate rock to agricultural lands has led to massive changes in the P cycle at both local and global scales (Bennett and others, 2001; Childers and others, 2011). While P management varies from region to region, many agricultural watersheds in North America have become enriched with P due to long periods in which P inputs via fertilisers and manures exceeded the amount of P removed through crop harvest (Hansen and others, 2002; Jarvie and others, 2013). This accumulated P is largely stored in watershed soils, where it is commonly described as 'legacy P' referring both to its long history and its implications for delayed recovery of eutrophied waterbodies (Sharpley and others, 2013; Rowe and others, 2016). The slow release of legacy P from watershed soils and sediments is a significant source of P inputs to surface waters (King and others, 2017; Motew and others, 2017), and causes persistent water quality issues in some lakes (Carpenter, 2005).

A watershed's ability to retain added P in its soils and sediments can be described as its 'buffering capacity', which can be considered as a key watershed-based regulating service for restricting the export of P from soilscapes to waterbodies and minimising the water quality impacts of agriculture (Doody and others, 2016). This regulating service can be compromised if mounting pressure from long-term P inputs goes unnoticed. An understanding of watershed buffering capacity, the watershed characteristics that determine it, and how it can be managed through interventions, therefore provides an opportunity to maximise P retention and reduce eutrophication impacts to an acceptable level (Carpenter, 2005; Rowe and others, 2016).

Watershed buffering capacity is a product of the diverse properties of the landscape, and is dynamic through space and time (Doody and others, 2016): *geochemical characteristics*, such as soil type or texture (Vaz and others, 1993; McDowell and others, 2003; Kleinman and others, 2011); *hydrological characteristics*, such as water yield and residence time (Reed and Carpenter, 2002; Borbor-Cordova and others, 2006); and *landscape characteristics* of the watershed, such as the composition and configuration of land use (Fraterrigo and Downing, 2008; Qiu and Turner, 2015) or rates of biological activity. In addition, buffering capacity is also impacted by socio-economic land management decisions, such as riparian zone maintenance, artificial drainage, the nature and timing of nutrient applications, as well as the amount and configuration of above-ground biomass (Osborne and Kovacic, 1993; Reed and Carpenter, 2002; Gentry and others, 2007). These diverse biophysical and social characteristics are likely to govern the quantity of P that watersheds can retain, and may therefore be useful in predicting how much excess P a watershed will retain and for how long. This is important to ensure that tipping points for accelerated leakage of P do not occur, and it can also help prioritise land use planning to avoid fundamental conflicts between P pressures driven by food production and water quality (Gordon and others, 2008; Doody and others, 2016).

Although past research has elucidated some of the key factors relevant to watershed buffering capacity (such as Fraterrigo and Downing (2008) whose findings highlight the relative importance of soil type and land cover), uncertainty remains regarding the specific contribution of different watershed characteristics and how they influence the degree of buffering in agricultural watersheds (Doody and others, 2016). A greater understanding of how buffering capacity is conferred on watersheds can help identify the vulnerability of watersheds to P pressure arising from changes in land use, including agricultural intensification and urbanization, and help in determining how this vulnerability might be managed for optimum delivery of ecosystem services.

We assessed the buffering capacity of a diverse set of watersheds in southern Quebec, Canada, and investigated the watershed characteristics most closely associated with buffering capacity

(Figure 1). Using historic and contemporary anthropogenic P input data (Goyette and others, 2016), we assessed both long- and short-term processes of P retention in the landscape using a novel "buffering capacity" index. We hypothesized that watersheds have innate long- and short-term buffering capacities based on their geochemical, hydrological and landscape characteristics, and that human activities and management act in addition to the three primary drivers considered in this study (Doody and others, 2016). We also hypothesized that watershed characteristics would have either an equal or greater impact on riverine P flux than legacy P levels. We further hypothesized that watershed characteristics and processes that promote short-term buffering would be different to those that enable long-term buffering.

Study Site

The sixteen watersheds examined in this study fall within the greater Saint Lawrence River Basin in Southern Quebec (QC), Canada (at 46°7'N, 72°42'W). These watersheds are home to approximately 4 million people and drain a total land area of ~73,000 km^2 (Figure 2). The dominant historical land cover was temperate forest; however, agricultural expansion starting around the end of the 19^{th} Century reduced forest cover, with more rapid agricultural intensification during the 1970s (MacDonald and Bennett, 2009). The predominant contemporary land uses include intensive row-crops, swine and dairy production, forage cropping, nature recreation, maple syrup production, as well as residential and urban development (Environment and Development, 2001; MacDonald and Bennett, 2009; Raudsepp-Hearne and others, 2010). This region contains predominantly acidic sandy/loamy soils, with extensive use of artificial drainage systems to remove ground and surface water from arable land (Jutras, 1967). Multiple studies have estimated high levels of soil test P enrichment in the region's agricultural soils, primarily related to historic P inputs from agriculture (MacDonald and Bennett, 2009; van Bochove and others, 2012; Goyette and others, 2016). The most heavily cultivated catchments are the Richelieu, Yamaska and Nicolet (>45 percent of total area cultivated); these three watersheds also have some of the highest levels of P input. The most populated watersheds are Richelieu and St. Francois. The most forested watersheds are Rouge and Lievre; and the watersheds with highest soil clay content are Richelieu and Nord, which are also the two watersheds with the highest estimated percentage of drained agricultural land.

Methods

To quantify the degree of buffering capacity in each watershed, we compiled and analysed long-term data on cumulative P inputs (1901-2011), together with data on historic river flows, and water quality (1981-2011) in each watershed from various sources. There were no regular measurements of river flow or water quality available prior to 1979 to link P input pressure to riverine P flux. We then compared two indicators of watershed buffering capacity (a long-term buffering index (LBI) and a short-term buffering index (SBI)) with various landscape characteristics to determine the major factors that mediate P buffering at the watershed scale.

Cumulative P Input data

We compiled P input data from Goyette and others (2016), who calculated net anthropogenic P input (NAPI) for all river watersheds draining directly to the Saint Lawrence River on a decadal or bi-decadal basis between the years 1901 and 2011. The NAPI values correspond to the difference between P imports to and exports from a watershed due to human activities, and thus indicate the net imports of 'new' anthropogenic P in each watershed during each of the study years (Figure 3). NAPI inputs include the P embedded in fertilizer imports, as well as P imported as food, animal feed, and detergents; P outputs in NAPI include the P embedded in exported crops and animal products (Russell and others, 2008). Using components of the NAPI data, we also calculated net P inputs to just cropland soils (arable and grass) based on the difference between the P embedded in inputs to croplands in each given year (fertilizer and animal feed) and the P embedded in exports from croplands in each given year (crops and animal products). Because NAPI data only exists for 10- or 5-year time steps, we interpolated between time steps to estimate annual values throughout the century. These estimates of watershed P accumulation do not include the cumulative P losses to surface waters that occurred prior to the start of river monitoring, or during the water quality monitoring period 1981-2011. Based on the 1981-2011 period, riverine P losses represented 1-60 percent of annual NAPI P inputs.

Water quality data

Monthly riverine P concentration data and daily water flow data between the years 1979 to 2011 were collected by two Quebec environmental monitoring agencies: *Ministère du Développement durable, de l'Environnement et de la Lutte contre les changements climatiques* (MDDELCC) and *Centre d'expertise hydrique du Québec* (CEHQ). To convert P concentration (mg/L) to riverine flux (kg yr^{-1}), we calculated the annual flow-weighted P concentrations (FWPC) and then estimated annual P loading based on the FWPC and water discharge using the U.S. Army Corps of Engineers' FLUX32 software (Walker, 1999). The software uses a jackknife approach to calculate variance. This variance is then used to weight data points in the resulting load calculation. We calculated annual P flux values for each of the 16 watersheds for seven 5-year time steps between the years 1981 and 2011 to correspond with the time scale of the NAPI data. Flux calculations were divided by the total watershed area to determine the kilograms of P flux per area per year of study (kg km^{-2} yr^{-1}).

Calculating Short- and Long-term Buffering Capacity

We estimated the Short-term Buffering Index (SBI) for each watershed by determining the percentage of the annual net P input that was retained by a watershed in each study year. Short-term retention was calculated as the difference between annual watershed net anthropogenic P inputs and annual P flux into the river for the corresponding year. In that sense, this metric is very similar to (inverse of) rates of fractional P export from NAPI that have been explored in previous studies (Hong and others, 2012; Goyette and others, 2016). The SBI value is an indication of the short-term trapping of P within the watershed. A high SBI value would indicate a greater retention of *recently-applied* P. To resolve the magnitude of difference between these two values, we calculated the relative degree of retention using the log-scaled accumulated P and riverine TP flux values:

$$SBI = (1 - \frac{\log(Riverine\ TP\ flux\ (kg \cdot km^{-2} \cdot yr^{-1}))}{\log(annual\ NAPI\ (kg \cdot km^{-2} \cdot yr^{-1}))})$$

We estimated the Long-term Buffering Index (LBI) for each watershed by determining the percentage of historically accumulated P that was being retained by a watershed in each study year. Historically accumulated P was estimated from cumulative net anthropogenic P inputs to the watersheds (between 1901 and the study year). The LBI value is an indication of the *capacity* of the watershed to absorb net P pressure and implies that some watersheds retain higher levels of accumulated P than others before releasing this P to a downstream water body. High LBI therefore signifies a large watershed buffering capacity meaning that more P is retained in the watershed and therefore has a greater concentration of P per unit of watershed area. The LBI was calculated as the difference between cumulative watershed net anthropogenic P inputs (between 1901 and the study year) and P flux to the river in that study year:

$$LBI = (1 - \frac{\log(Riverine\ TP\ flux\ (kg \cdot km^{-2} \cdot yr^{-1}))}{\log(cumulative\ NAPI\ (kg \cdot km^{-2} \cdot yr^{-1}))})$$

Watershed data

We determined watershed boundaries and collected geochemical, hydrological, landscape, and socio-ecological spatially explicit information for the 16 watersheds from a variety of sources (Table 1). This information was used to investigate which watershed characteristics are most closely associated with long- and short-term P buffering capacity.

Watershed Boundaries. We obtained Level 7 watershed boundaries from HydroSHEDS (Lehner and Grill, 2013), corresponding to watersheds roughly between 1000 to 7000 km^2, because it matched most closely with the scale of the NAPI data (Goyette and others, 2016). In two of our study watersheds (Lievre and Saint-Francois), sub-watersheds representing “upper” and “lower” halves were combined so as to correspond with the NAPI data.

Geochemical Characteristics. We used soil P data from Beaudet and others (2003), which were based on ~110,000 soil samples collected between the years 1995 and 2011 (kg P ha^{-1}; Mehlich-3 derived, taken at a 20-cm depth). We aggregated municipal-level P data and partitioned it to the corresponding watershed. Soil characteristics, such as texture and pH, were summarized for each watershed to determine dominant soil qualities. Data was obtained by the Soil Landscapes of Canada database, version 3.2 (Soil Landscapes of Canada Working Group, 2010).

Hydrological Characteristics. To calculate water yield for watersheds, we obtained streamflow data (m^3/sec daily throughout the study years) at the outflow point of each watershed. We calculated a simple average annual streamflow and divided this by the area of the watershed to determine the water yield per km^2, sometimes referred to as specific runoff (Behrendt and Opitz, 1999). We calculated the baseflow index for each watershed through time using the web-based hydrograph analysis tool (WHAT) developed by Lim and others (2005). This tool interprets streamflow data using signal analysis to separate high and low frequencies within the data. High frequency waves have been associated with direct runoff, and low frequency waves have been associated with baseflow (Eckhardt, 2005). We calculated mean percent slope of the total watershed area, as well as the sinuosity of the major rivers within each of the watersheds in ArcGIS v. 10.1 (ESRI, 2012).

Landscape characteristics. To determine the degree of vegetative cover for each watershed, we calculated the normalized difference vegetation index (NDVI) for each study year between 1981 and 2011 (Weier and Herring, 2000). Higher NDVI values indicate greater vegetative cover. Landsat pixels with <10 percent cloud cover were aggregated in the years preceding each study year to create composite cloud-free satellite images. We repeated the NDVI calculation for the 100-meter buffer regions around rivers in each watershed as an indication of riparian vegetative cover.

To determine the land use/land cover of watersheds, we consolidated 145 land use classes into eight broad categories to determine the land use trends of the area; this data was made available from the *Quebec Ministère du Développement durable, de l'Environnement et de la Lutte contre les changements climatiques* (Bissonnette and Lavoie, 2015). In a rasterized format, we calculated percent land cover types in each watershed. We repeated percent land cover for the 100-meter buffer regions around major rivers in each watershed to get an indication of riparian land use.

To calculate metrics of landscape configuration, we used FRAGSTATS 4.0 (McGarigal and others, 2012). We selected a set of landscape configuration metrics that were ecologically relevant to P transport mechanisms, including fragmentation indices (patch density [PD] and edge density [ED]), and connectivity indices (patch cohesion [COHESION] and probability of adjacency [PLADJ]), as well as an interspersion index, known as contagion [CONTAG] (McGarigal and others, 2012; Qiu and Turner, 2015).

Socio-ecological Characteristics. We calculated watershed population density by partitioning municipality-scaled population density values to watershed boundaries. To calculate average field size in each watershed, we calculated the size of each farm in the study area using a polygon map of all of QC farms available from *La Financiere agricole Quebec*. To account for the average market capital value of farms (dollars per farm) for each watershed by dividing the total market value (TMV) of all of the farms in each census subdivision by the total number of farms, and then we partitioned TMV to watershed boundaries; this data was available from the Quebec Agricultural Census.

Sub-surface drainage is important because it involves the piping of water from agricultural fields directly into water bodies, which is a potential pathway of P mobilization in agricultural watersheds in this region (Gentry and others, 2007). However, no complete dataset exists on the presence and location of sub-surface drainage in Quebec. We therefore estimated the presence of drained land based on a combination of land use and soil type data, assuming that subsurface draining occurs where row crops were grown on areas of poorly drained soil (as indicated by the Soil Landscapes of Canada database) (Sugg, 2007). Specifically, we overlaid spatial agricultural land use and soils data from the Government of Quebec (*La Financière agricole du Québec,* www.fadq.qc.ca) in ArcGIS and calculated the percent area in each watershed where row cropping was happening in very poor (VP), poor (P), or imperfectly drained (I) soils.

Data Analysis

We used non-parametric Spearman's correlation coefficients to estimate the relationship between buffering indicators and various watershed characteristics. We used a scatterplot matrix to account for multi-collinearity among variables. We also performed linear regressions between watershed characteristics, P accumulation values, and riverine P flux values to determine whether landscape characteristics or P accumulation had more explanatory power over riverine P flux. All statistical analysis was performed using R v. 3.3.1 (R Core Team, 2016).

Results

P accumulation and riverine P flux

All of the study watersheds have been accumulating P since the beginning of the 20th century. By 2011, long-term cumulative net P inputs (total P accumulation since 1901) in the study watersheds ranged from ~6,000 (Lievre) to ~100,000 kg P km^{-2} (Yamaska), with an average of ~29,000 kg P km^{-2} accumulated across the entire study area (Figure 3a). In the last 30 years, some watersheds, such as Yamaska, and L'Assomption, experienced higher rates of P accumulation compared to earlier in the century, whereas others, such as Lievre and Petite Nation, showed lower rates of P accumulation over time (Figure 3a). Cumulative P accumulation values represent the total amounts of P pressure these watersheds have to buffer to prevent any P loss to water. The gap between total NAPI and net cropland P input (i.e. cropland surplus) represents the P imported to watersheds embedded in food imports for human consumption and in detergents. The watersheds with the largest gap between cropland P surpluses and total NAPI include Yamaska and Richelieu (average 485 kg P km^{-2} yr^{-1} and 361 kg P km^{-2} yr^{-1}, respectively; Figure 3b). There was a significant correlation between soil P levels and cumulative P accumulation values since 1901, both for accumulated total P (Spearman's rho, $\rho=0.68$, $p<0.00$) and for estimated values for accumulated cropland P (Spearman's rho, $\rho=0.61$, $p=0.01$).

In general, between the years 1981 and 2011, annual net P inputs (NAPI) exceeded riverine P flux values, as represented by the gap between the red and green lines on Figure 3b. This gap is an estimate of the amount of anthropogenic P that is retained by each watershed for a given year and varied widely between watersheds. Annual inputs exceeded riverine P flux by as much as 2094 kg P km^{-2} (Yamaska, 1986) for a year in which the Yamaska watershed retained approximately 97 percent of anthropogenic P inputs, to as low as -5 kg P km^{-2} (Petite Nation, 2006) for a year in which there was a net loss of P from the Petite Nation watershed through crop exports. On average, the watershed with the largest gap between annual NAPI and riverine P flux (and thus the most P retention) was Yamaska (1753 kg P km^{-2} yr^{-1}; 96 percent retention), and the watershed with the smallest gap was Lievre (43 kg P km^{-2} yr^{-1}; 69 percent retention).

Across all the watersheds, higher cumulative P tends to be associated with higher annual riverine P flux, and this cross-watershed relationship is consistent over time (Figure 4). Indeed, riverine P export tended to increase at a faster rate in relation to the total accumulated P after 2001 (i.e. steeper gradients) suggesting that the different watersheds might be gradually losing more and more P. Among the watersheds, there was a significant positive correlation between the average NAPI value within the basin and the average riverine P flux values from 1981-2011 (Spearman's rho, $\rho=0.76$, $p<0.00$). However, each individual watershed showed distinct and generally opposite dynamics over time, wherein annual riverine P flux almost always decreased as cumulative P accumulation increased after 1981 (Figure 5). The trend was similar when comparing riverine P flux and cumulative cropland P values. This apparent contradictory pattern of P mobility may be attributable to changes in climate, different long-term and short-term P retention dynamics, and changes in regulatory policies and practice in the region over the study period.

Buffering capacity estimates

The values and spatial variation of LBI and SBI were not consistent across the watersheds (Table 2; Figure 6). The average LBI value across the study period (1981-2011) was 0.63 (s.d. 0.06) with a relatively narrow min-max range of 0.15. The watersheds with the highest average LBI values over the study period were Richelieu, Rouge, Petite Nation, and Lievre. The watersheds with the lowest average LBIs included Loup, Etchemin, and Sainte-Anne. The LBI values increased through time in a majority of watersheds because riverine fluxes generally decreased, even as cumulative net P inputs increased (Figure 5). The average SBI value was 0.34 (s.d. 0.12) with a min-max range of 0.2. Unlike LBI, there was no temporal trend among these values. The watersheds with the highest average SBI values were St. Francois, Yamaska and Richelieu. The watersheds with the lowest average SBI values were Nord, Sainte-Anne, and Petite Nation. LBI and SBI values were not significantly related to each other (Spearman's rho, ρ=0.32, p=0.22) suggesting they represent differing aspects of watershed buffering capacity. The watersheds that deviated most strongly between LBI and SBI were Petite Nation, Nord, Lievre, Ste-Anne, and Jacque Cartier (Figure 6).

Watershed characteristics

The sixteen watersheds showed a diverse range of geochemical, hydrological, landscape, and socio-ecological characteristics that might be used to explain differences in watershed buffering capacity and riverine P flux (Table 2).

This region is generally dominated by acidic, sandy soils and soil pH tended to increase on the more clayey soils (Spearman's rho, ρ=0.47, p=0.06), Table S1. Highest clay content in the region was 41 percent in Richelieu. Soil test P values were highest in the historically agricultural watersheds and were positively correlated with percent agricultural land cover (Spearman's rho, ρ =0.84, p<0.001). The highest average soil test P value was 204 kg ha^{-1} in Yamaska, which is considered high for agricultural production (Beaulieu and others, 2006).

The watersheds also varied in their hydrological characteristics. Watershed percent slope values were relatively low in this region, mostly under 20 percent. Watersheds with low mean percent slope tended to have lower water yield (Spearman's rho, ρ=0.45, p=0.08), Table S1. Baseflow index, an indication of the relative contribution of groundwater to the water flow within the watershed, also varied across the watersheds (from 0.59 to 0.99); watersheds with higher baseflow were generally flatter and had lower water yield.

Watershed landscape composition and configuration variables were strongly correlated, as watersheds consisted mostly of agriculture and forested land cover (Table S2). However, only two of the watersheds have a majority of agricultural land cover, Richelieu (68 percent) and Yamaska (55 percent). Most of the watersheds have relatively small amounts of developed (built-up) land (10 percent or less), and developed land area was strongly correlated with population density (Spearman's rho, ρ=0.86, p=<0.001). Tile-drained agricultural land is quite common in the Quebec landscape; in about half of the watersheds, we estimated that upwards of 80 percent of agricultural croplands contain artificial drainage (with a range of 0 and 35 percent of total watershed area).

Role of watershed characteristics in explaining buffering indices and riverine TP flux

The geochemistry, hydrology, and landscape configuration of the watersheds explained a significant but variably weak proportion of our two buffering capacity indicators (Figures 7a and 7b). LBI was significantly ($p<0.05$) correlated only to baseflow index, whilst SBI was significantly influenced only by watershed size, slope, and market capital, which is linked to the proportion of the watershed in productive agriculture (Tables S1 and S2). The near significance but weak ($\rho<0.30$) relationships obtained with many other watershed characteristics suggests buffering capacity is a complex metric that integrates numerous watershed processes over space and time. The following paragraphs summarize the key correlation results (i.e., where $\rho>0.30$ and $p=<0.01$).

For geochemical variables, soil test P was moderately positively correlated with SBI (Spearman's rho, $\rho=0.49$, $p=0.06$; Figure 7a), which is likely an indication of the high levels of recent P application in these watersheds. However, soil test P did not influence LBI ($\rho=-0.34$, $p=0.20$) probably because this routinely measured fraction of soil P represents a very small part of the total legacy soil P which will have accumulated in these soils (Rowe and others, 2016).

SBI and LBI both tended to decrease as water yield increased ($\rho=-0.44$, $p=0.09$ and $\rho=-0.49$, $p=0.05$, respectively) and similar negative relationships were observed with slope because of the positive link between slope and water yield. This suggests that hydrological residence time plays a key role in P buffering with low P retention associated with faster flows. However, in stark contrast to LBI ($\rho=0.63$, $p=0.01$), SBI was not influenced at all by baseflow index (Figure 7a) which suggests that this is a hydrological dynamic that mainly impacts long-term P retention. This may be because of the longer time it takes for P to infiltrate groundwater than surface water. All of the watersheds showed a greater contribution from groundwater relative to surface water, such that baseflow indices might be expected to have less influence on SBI.

Both the LBI and SBI were positively correlated with watershed size (ρ=0.46, *p*=0.07 and ρ=0.56, *p*=0.02, respectively; Figure 7a), suggesting that larger watersheds have greater opportunities to retain P inputs both in the short-term and long-term. The buffering indicators also had some correlations with socio-ecological watershed characteristics. The SBI values were positively correlated with "market capital", which is a measure of the relative wealth of farms in the landscape (ρ=0.52, *p*=0.04; Figure 7a). This correlation suggests that landscapes containing farmers with more assets and capabilities can collectively "buffer" P in the short term through the adoption of best land management practices. Counter to our expectations, the LBI and SBI values were not influenced by the percentage of cropland with agricultural tile-drainage (ρ=-0.32 *p*=0.23; Figure 7a).

Landscape composition and configuration variables were also correlated with buffering indicators. SBI values were positively correlated with percent agricultural cover (ρ=0.54, *p*=0.02; Figure 7b), likely because agricultural lands are the sites of a majority of anthropogenic P inputs. For configuration variables, we found the SBI values to be positively correlated with agricultural edge density (ED) (ρ=0.55, *p*=0.03; Figure 7b), which perhaps signifies that dispersed agricultural land can decrease the intensity of P runoff and increase the landscapes ability to retain mobilized P in the short-term. The SBI values were negatively correlated with the Forest Probability of Adjacency (PLADJ) indicator (ρ=-0.57, *p*=0.02; Figure 7b), offering some support that landscapes with higher levels of forest connectivity have greater abilities to buffer P pressure. The SBI also had a negative correlation with the contagion index (ρ=-0.45, p=0.08; Figure 7b) which is an indication of the homogeneity of land use types on the landscape.

Of the watershed variables, riverine TP flux was significantly ($p < 0.05$) positively correlated to soil test P (ρ=0.57, p=0.02) and negatively correlated to baseflow index (ρ=-0.64, p=0.01), Table S1. Watersheds with a higher proportion of crop available soil P and surface runoff were therefore exporting the most P. Riverine TP flux was also highly negatively correlated to LBI (ρ=-0.79, p<0.01) but not to SBI (Table S1). Multiple regression analysis indicated that a model accounting for both cumulative P and baseflow index values was the strongest overall predictor of riverine TP flux ($R^2 = 0.685$, $p<0.01$ Table 3). Therefore, while watershed P enrichment (either as soil test P or cumulative legacy P) and watershed hydrology (baseflow index) were individually helpful to understand potential impacts on water quality arising from anthropogenic inputs of P, their combined influence provided a more comprehensive indicator of watershed vulnerability to P leakage (as additive terms).

Discussion

The variation in accumulated legacy P since 1901 between watersheds was very large and reflected the different patterns of land use change, historic P inputs and demography across the study catchments. This long-term P accumulation was driven by fertiliser and feed inputs into agriculture (Fig. 3b), and although this legacy P was not strongly reflected in current soil test P values, our analysis nevertheless found it of critical importance for water quality in this region of southern Quebec (MacDonald and Bennett, 2009; Goyette and others, 2016). Watersheds with relatively higher levels of accumulated legacy P had significantly poorer water quality as measured by riverine TP export and flow-weighted P concentrations. However, despite the overall increase in legacy P stored in the region over the study period, TP export declined in almost every watershed over the thirty-year period from 1981 to 2011. This apparent contradiction is likely due to a number of regulatory policies aimed at improving water quality which were implemented in Quebec during this period: for example, improved sewage treatment infrastructure, wastewater treatment plant upgrades, and legislation intended to reduce excess P use and runoff from farms (Painchaud, 1997; Montpetit and Coleman, 1999; Mailhot and others, 2002; Boutin, 2005). This implies that while some environmental policies and infrastructure upgrades can improve water quality through time, watersheds with the highest magnitudes of legacy P pressure will continue to face persistent water quality problems. In a recent analysis of watershed P inputs and riverine P export across the USA, annual P inputs and runoff explained ca. 50 percent of the variance in riverine P flux (Metson and others, 2017).

We also observed that, while anthropogenic P accumulation continued to mount over the study period, the *rate* of P accumulation (relative levels of annual NAPI P surpluses) decreased during the study period. This may be in response to the environmental policies mentioned earlier and matches general trends in nutrient use efficiencies observed in North America during this time (Dobermann and Cassman, 2002; Vitousek and others, 2009; Goyette and others, 2016). While nutrient management policies have been partially successful at decreasing NAPI in the region, as long as watershed P continues to mount, the stores of legacy P continue to pose a long-term risk to water bodies (Carpenter, 2005). Extensive time-lags are common between interventions to improve water quality through agricultural management practices and measureable outcomes in aquatic ecosystems in general, but these can be particularly long in the case of P (Meals and others, 2010).

The importance of landscape characteristics for short- and long-term buffering capacity

Our results highlight how buffering capacity is the result of a combined dynamic between both the degree and nature of legacy P accumulation as well as the watershed characteristics that impact the transport of P across the landscape. Researchers and managers can have a more comprehensive picture of P buffering and landscape vulnerability by investigating both legacy P indicators as well as the biophysical characteristics, such as the baseflow index, that mediate the release and transport of legacy P.

Buffering is a process that operates on different time scales, from long-term legacy P storage in soils to the short-term landscape trapping of more recently applied P. In our analysis, indices representing long- and short-term buffering (SBI and LBI) bore no significant correlation to each other, suggesting that different landscape characteristics may be mediating these two buffering processes. The difference between LBI and SBI values also provided additional insights into the patterns of P accumulation in each watershed. In some watersheds, such as Lievre and Petite Nation, the LBI was much higher than the SBI. Such watersheds have a history of P accumulation in the landscape but have seen substantial reductions in NAPI levels in the last 30 years. A greater proportion of the riverine P flux from these watersheds is from historic accumulation, so longer-term landscape processes likely dominate the buffering dynamic in these watersheds, where baseflow contributes the majority of the river flow and the baseflow index was strongly correlated with the LBI. In contrast, we generally found that SBI correlated more strongly with watershed characteristics from all four categories of buffering, which suggests that short-term buffering dynamics are more strongly mediated by the landscape characteristics considered in this analysis.

The fact that the two indicators had, in general, the same relationships with the key hydrological variable of specific runoff, and related physical characteristics such as watershed size and mean percent slope, shows that innate hydrological characteristics may be the most powerful drivers of both short-term and long-term P retention processes (Metson and others, 2017). The story is less clear for land use/land cover, both in terms of composition and configuration. Our analysis suggest landscape connectivity and heterogeneity are important factors in buffering, but high degrees of co-linearity among composition and configuration metrics make it difficult to identify the most important indicators of P retention among these variables. However, since land use/land cover is relatively more flexible in terms of watershed management (relative to, for example, topography), a more in-depth study of land use/ land cover impacts on watershed P retention would be worthwhile. Knowledge about which landscape characteristics mediate the relationship between reduced P pressure and water quality outcomes can also help us to manage our expectations concerning the mitigation actions and the likelihood of time lags in response (Meals and others, 2010; Sharpley and others, 2013).

Conclusions

New insights into release and transport processes of legacy P within watersheds, as well as a greater knowledge of the landscape characteristics that determine a watershed's specific buffering capacity, could help to target landscape management to help reduce the vulnerability of surface waters to P pressures. Locally, such information could help to inform specific ways in which land managers can increase a watershed's resilience to P pressure and decrease the vulnerability of water systems (Jarvie and others, 2013). Regionally, such information can be used to identify which watersheds are more or less vulnerable to increased P pressure arising from agricultural development or urbanization. These insights can also aid in the prioritization of land use and the ability to decide on whether land should be cultivated or spared for conservation (Doody and others, 2016).

A watershed's capacity to buffer the effects of stored P has significant impacts on people and ecosystems in agricultural watersheds. P buffering processes both represent the short-term protection of water quality degradation as well as the long-term vulnerability of water systems to legacy P runoff. The use of buffering in socio-ecological research could aid in the development of holistic watershed management which recognizes non-linear, long-term ecological dynamics, and sees long-term P retention as both a critical threat, as well as an opportunity for the reclamation of an essential resource.

Acknowledgements

We thank Helen Jarvie and Donnacha Doody for comments on previous drafts. We thank Guillaume Larocque at the Québec Centre for Biodiversity Science for technical assistance and support. This research was funded by the Natural Science and Engineering Research Council (NSERC) Discovery Grants (DG) to Elena Bennett and the Economics for the Anthropocene (E4A) program.

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Table Legends:

Table 1. Sources of spatial data

Table 2. Indicators of P buffering capacity in the 16 study watersheds

Table 3. Biophysical predictors of riverine TP flux

Table 1

Component of Buffering capacity	Indicator of P buffering capacity and unit	Spatial Resolution	Temporal Resolution	Data Source
Soil	Percent clay	~ 1 km	Single value (~2001)	Soil landscapes of Canada
	pH (measured in calcium chloride)	~ 1 km	Single value (~2001)	Soil landscapes of Canada
	Soil P levels (kg/ha)	QC Municipalities	Single value (~2001)	MAPAQ
Hydrological	Watershed size (km^2)	15 sec	Single value	HydroSHEDS, HydroBASINS with lakes - level 7
	Percent slope	3 sec	Single value	HydroSHEDS, void-filled elevation
	Water yield (m^3/km^2/year)	NA	Daily values (1979-2011)	CEHQ
	Baseflow index	NA	Annual values (1979-2011)	CEHQ
	Sinuosity ratio	15 sec	Single value	HydroSHEDS, river network
Landscape and socio-ecological	Vegetative cover (NDVI)	~30 m	Annual values (1985-2011)	Landsat remote sensing
	% Land use classes	-30 m	Single value (~2010)	MDDELCC
	Landscape configuration metrics	-30 m	Single value (~2010)	MDDELCC
	Population density	QC Municipalities	Bi-decadal census years (1981-2011)	Quebec population census
	Average field size (km^2)	~.5 m	Single value (~2008)	La Financière agricole Québec
	Average market capital ($/farm)	QC Municipalities	Bi-decadal census years (1981-2011)	Quebec agricultural census
	% Agricultural tile drainage	~1 km	Single value ~2011	Soil landscapes of Canada La Financière agricole Québec
P flux	NAPI	QC Municipalities	Decadal and bi-decadal census years (1981-2011)	Goyette et al. (2016)
	Riverine P flux	NA	~Monthly values (1979-2011) ~Daily values (1979-2011)	MDDEFP-BQMA (concentration) CEHQ (flow)

Table 2

		Soil Indicators				Hydrological Indicators					Landscape and Socio-ecological Indicators				P flux		
Watershed	Soil test P (kg/ha)	% clay	pH	Mean % slope	Mean H_2O yield ($m^3/km^2/sec$)	Baseflow index	Sinuosity index	Basin area (km^2)	% Agriculture	% Forest	% Wetlands	% agriculture tile-drained	Population density (pop/ha)	Average NAPI ($kg/km^2/yr$)	Average TP riverine flux ($kg/km^2/yr$)	Buffering index long	Buffering index short
Batiscan	69.26	16.50	5.23	11.55	.021	0.783	0.813	4732.56	5.83	77.91	6.82	48.23	52.25	126.37	25.34	0.643	0.333
Becancour	126.52	8.54	5.10	9.05	.022	0.665	0.816	2698.66	33.01	52.16	7.22	89.27	154.25	576.29	71.53	0.596	0.345
Chaudiere	121.35	13.29	4.83	5.56	.020	0.612	0.850	6604.29	22.01	64.13	9.25	23.64	96.43	553.37	46.13	0.628	0.399
Etchemin	161.45	15.14	4.81	20.87	.031	0.633	0.819	1547.08	30.69	55.54	6.27	25.13	132.38	771.51	84.74	0.598	0.343
Jacque-Cartier	155.48	10.92	5.08	18.08	.031	0.738	0.848	3362.49	10.72	70.64	4.69	29.3	186.59	112.46	29.80	0.618	0.295
L'Assomption	178.60	25.28	5.70	11.63	.019	0.737	0.815	5114.49	22.69	60.57	4.04	84.91	161.00	586.29	54.74	0.598	0.370
Lievre	80.34	15.90	5.40	9.41	.026	0.995	0.834	9564.14	2.82	79.64	8.51	0.56	29.41	62.62	19.27	0.665	0.269
Loup	156.61	30.65	5.49	9.79	.018	0.744	0.826	4002.81	24.02	63.36	4.89	83.74	56.80	368.39	43.54	0.590	0.355
Nicolet	136.78	10.13	5.13	9.51	.022	0.585	0.827	3377.9	45.08	44.33	4.22	89.27	117.09	733.89	75.14	0.598	0.354
Nord	119.89	38.58	5.55	8.46	.022	0.727	0.823	2284.68	13.35	67.94	7.9	91.08	119.27	102.23	39.62	0.603	0.199
Petite Nation	102.42	10.95	5.40	10.08	.017	0.85	0.840	2240.17	7.36	75.58	7.16	16.46	23.40	65.52	16.38	0.681	0.234
Richelieu	160.57	41.01	5.86	6.08	.018	0.901	0.791	3568.09	68.16	16.27	3.74	91.79	306.13	692.11	19.14	0.725	0.556
Rouge	76.76	4.74	5.55	10.70	.020	0.799	0.828	5610.44	2.19	80.92	8.93	0	25.95	102.23	18.35	0.691	0.378
Ste. Anne	91.87	7.87	5.09	12.69	.032	0.704	0.851	2759.37	10.23	75.84	4.43	59.68	51.23	155.28	57.31	0.575	0.217
St. Francois	123.51	15.94	4.98	6.30	.025	0.691	0.859	10426.3	21.6	60.85	13.5	32.32	103.71	410.78	40.19	0.645	0.400
Yamaska	204.11	20.69	5.37	5.04	.012	0.853	0.824	4944.05	55.09	33.83	5.54	85.93	122.57	1828.83	75.91	0.623	0.436

Table 3.

Regression	**Equation**	**R^2 value**
Log TP flux ~ P accumulation	$y = 26.8 + .001\ x_1$	0.382
Log TP flux ~ Baseflow Index	$y = 144 – 133\ x_2$	0.359
Log TP flux ~ P accumulation + Baseflow Index	$y = 116 + .001\ x_1 – 117\ x_2$	0.685

Figure Legends:

Figure 1. Conceptual model illustrating the buffering capacity concept and hypotheses underlying this study. Various geochemical, hydrological, landscape, and social factors could interact to mediate the relationship between P pressure and ecological impact by affecting buffering capacity. Therefore, the watershed's ultimate capacity to buffer increasing P pressure is dictated by a combination of innate landscape characteristics, cumulative P pressure due to continuous P inputs, and human management.

Figure 1. Map of the study area. The study area comprises 16 large watersheds located in the Province of Quebec, Canada. The study area surrounds the City of Montreal in the west, and Quebec City, in the east. The dominant land covers in these watersheds include agriculture, forest and developed (built-up) land. All watersheds drain into the Saint Lawrence River, which culminates in the Lower Saint Lawrence Estuary and Gulf of Saint Lawrence to the east (see inset map).

Figure 3a. Summary of P accumulation across the 16 watersheds showing varying levels of cumulative P accumulation over the last century (1901-2011). These values should be interpreted as proxies for long-term P accumulation in each watershed because they do not account for riverine P export over time. The vertical dotted lines denote the 30-year time span of the riverine TP flux data (1981-2011).

Figure 3b. Summary of P flows across the 16 watersheds show varying levels of total net-P input (NAPI), net cropland-P input only (fertilizer and animal feed inputs minus crop outputs), as well as riverine P flux over the 35-year study period. The 'gap' between the NAPI (red) and riverine P flux (green) lines represents the amount of P that has accumulated within the total watershed area in each year. The ‘gap’ between the net cropland P inputs (purple) and riverine P flux (green) lines represents the amount of P that has accumulated within watershed croplands in each year.

Figure 4. The consistent relationship between the TP accumulation and TP flux in each study year across all watersheds for 7 study years between 1981 and 2011. TP accumulation refers to cumulative P inputs since 1901 up to the study year in each watershed.

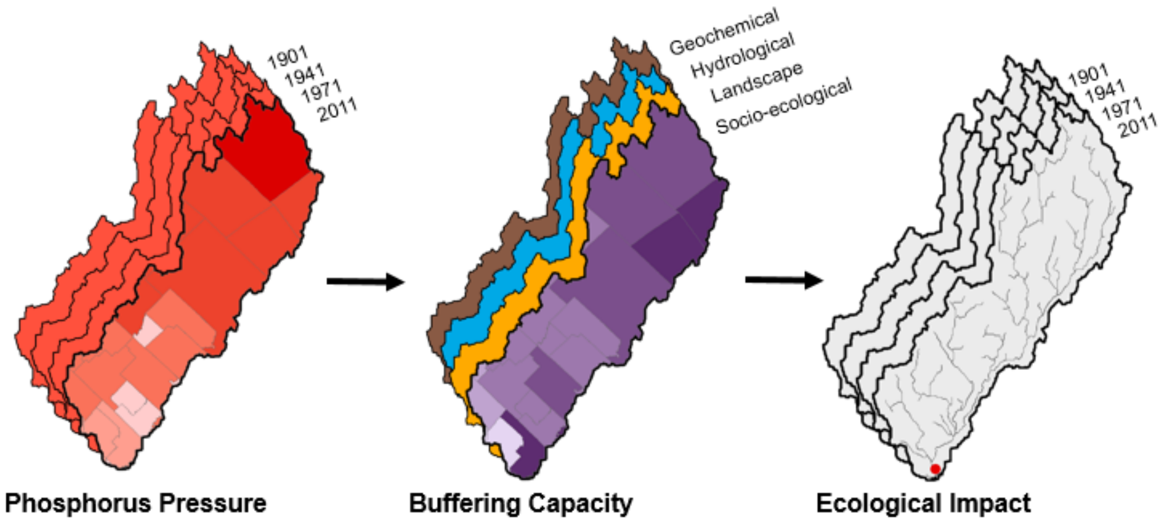
Figure 5. The variable relationship between annual TP accumulation and riverine TP flux for individual watersheds over time. Cumulative P accumulation since 1901 and annual riverine P flux for individual watersheds through time can be compared to the cross-watershed relationships shown in Figure 4. Several watersheds show a decrease in riverine P flux despite continued P accumulation.

Figure 6: Maps of average buffering values in study watersheds Average P buffering capacity estimates for each of the study watersheds using the (A) Long-term Buffering Index, LBI and (B) Short-term Buffering index, SBI.

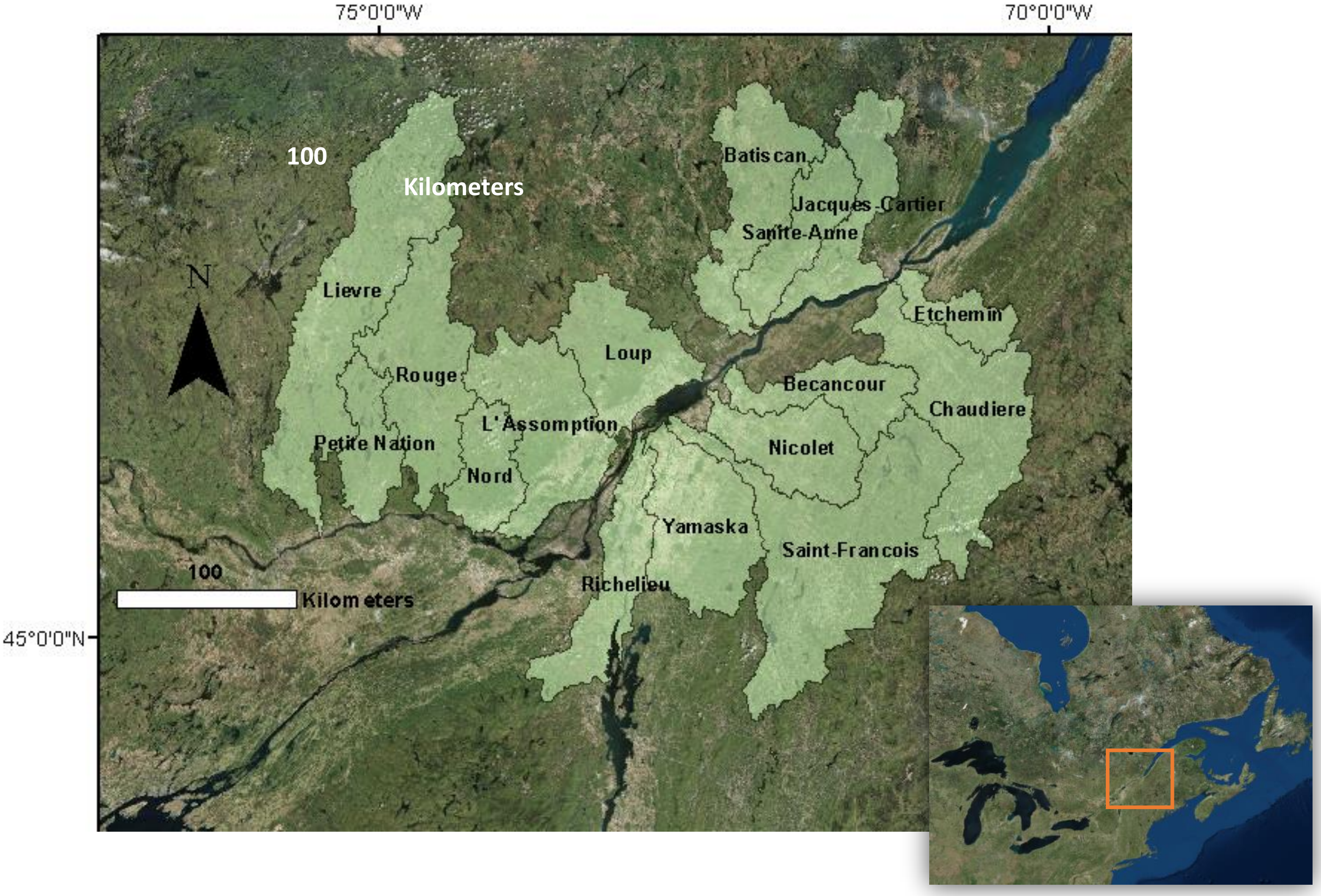
Figure 7a: Scatterplots showing the relationship between select geochemical, hydrological, and socio-ecological watershed characteristics compared to long- and short-term buffering index variables. The r-values displayed represent Spearman correlation coefficients (rho, ρ).

Figure 7b: Buffering indices and landscape characteristics. These graphs show the relationships between long- and short-term buffering index variables and select landscape composition and configuration characteristics. The r-values displayed represent Spearman correlation coefficients (ρ).

Figure 1


1901
1941
1971
2011
Geochemical
Hydrological
Landscape
Socio-ecological
1901
1941
1971
2011
Phosphorus Pressure
Buffering Capacity
Ecological Impact

710 **Figure 2**



75°0'0"W
70°0'0"W
45°0'0"N
100
Kilometers
N
Lievre
Rouge
Petite Nation
Nord
L'Assomption
Loup
Batiscan
Jacques-Cartier
Sainte-Anne
Etchemin
Becancour
Chaudiere
Nicolet
Yamaska
Saint-Francois
Richelieu
100
Kilometers

725 **Figure 3a**

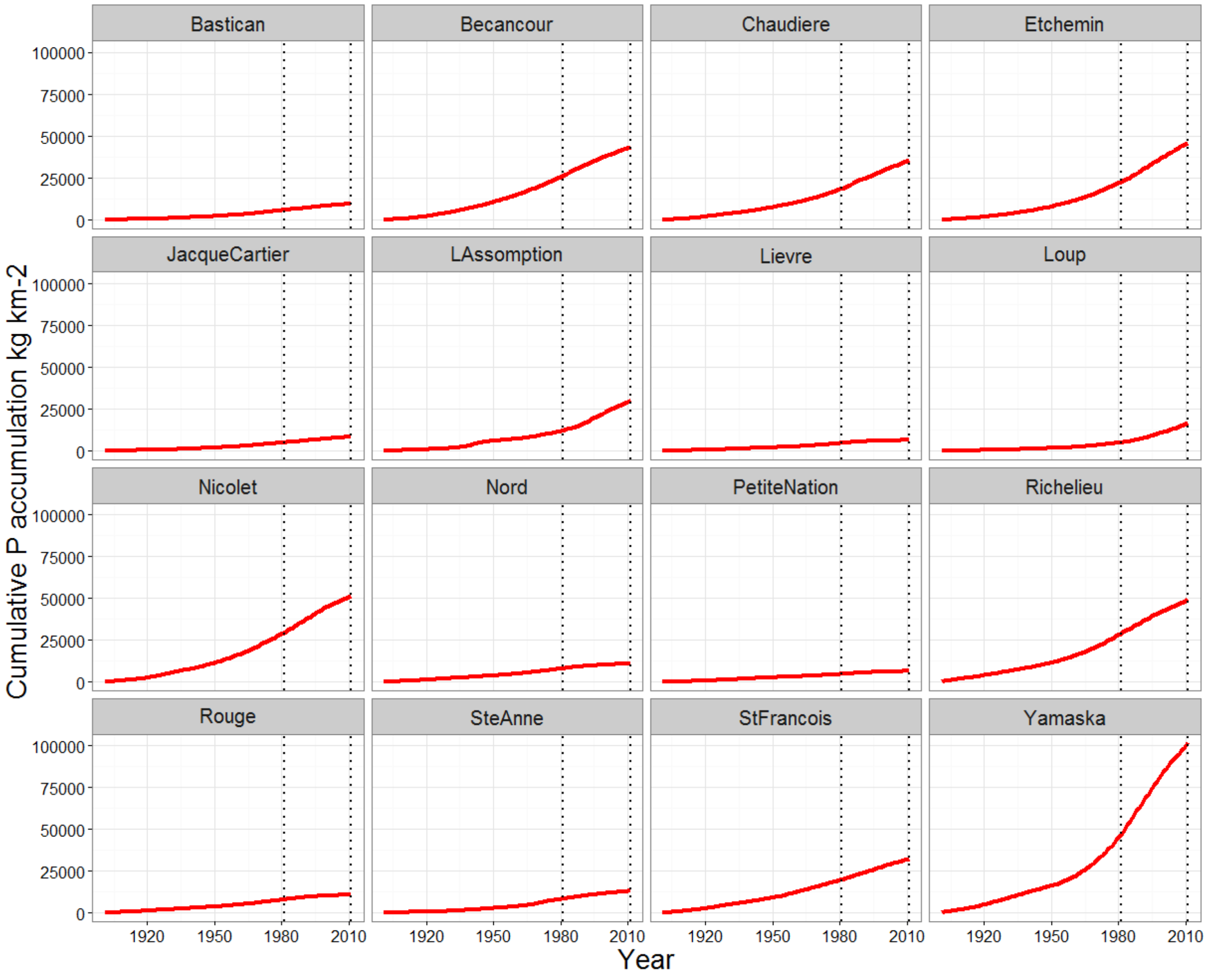
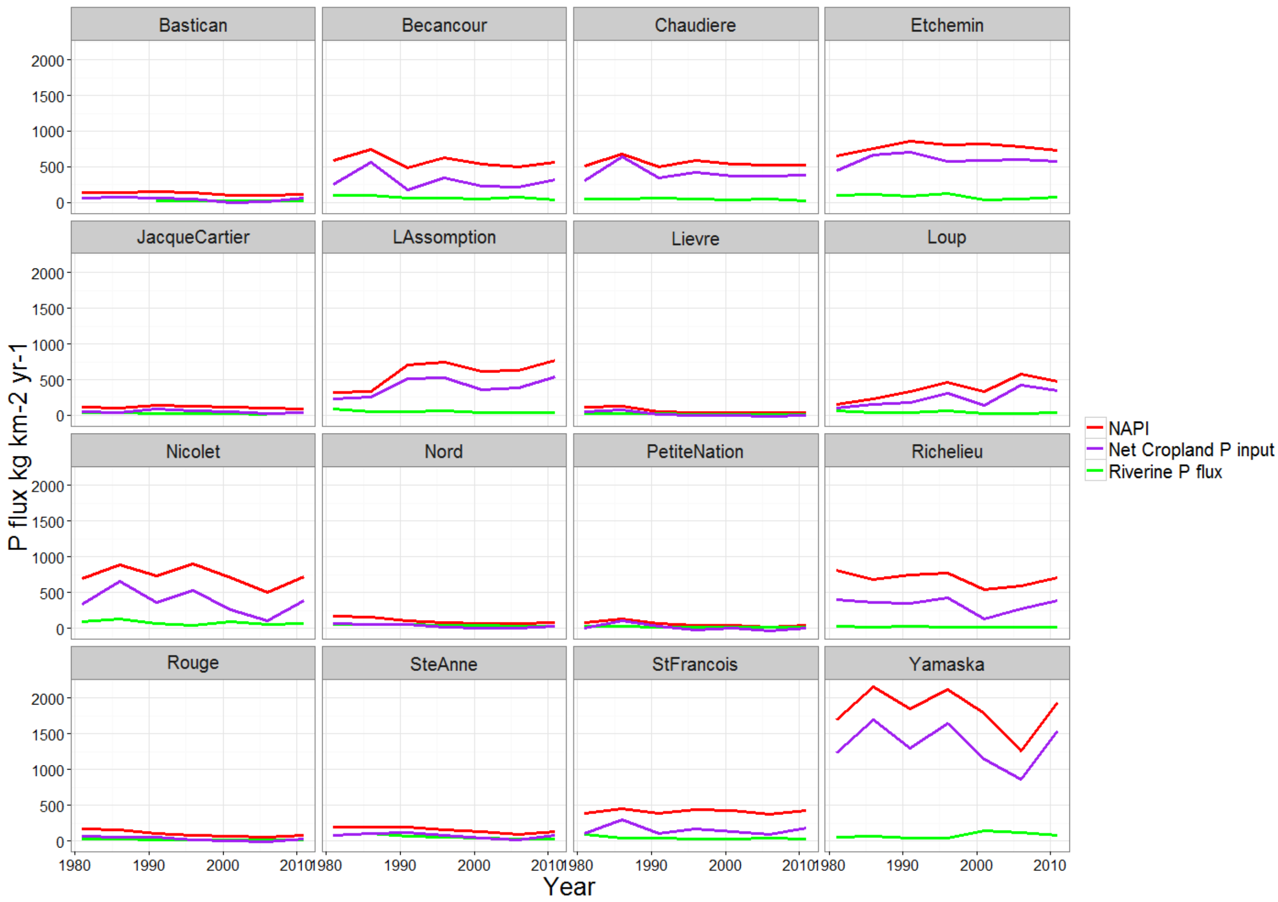


Figure 3b



Bastican
Becancour
Chaudiere
Etchemin
JacqueCartier
LAssomption
Lievre
Loup
Nicolet
Nord
PetiteNation
Richelieu
Rouge
SteAnne
StFrancois
Yamaska
P flux kg km-2 yr-1
Year
0
500
1000
1500
2000
1980
1990
2000
2010
NAPI
Net Cropland P input
Riverine P flux

Figure 4

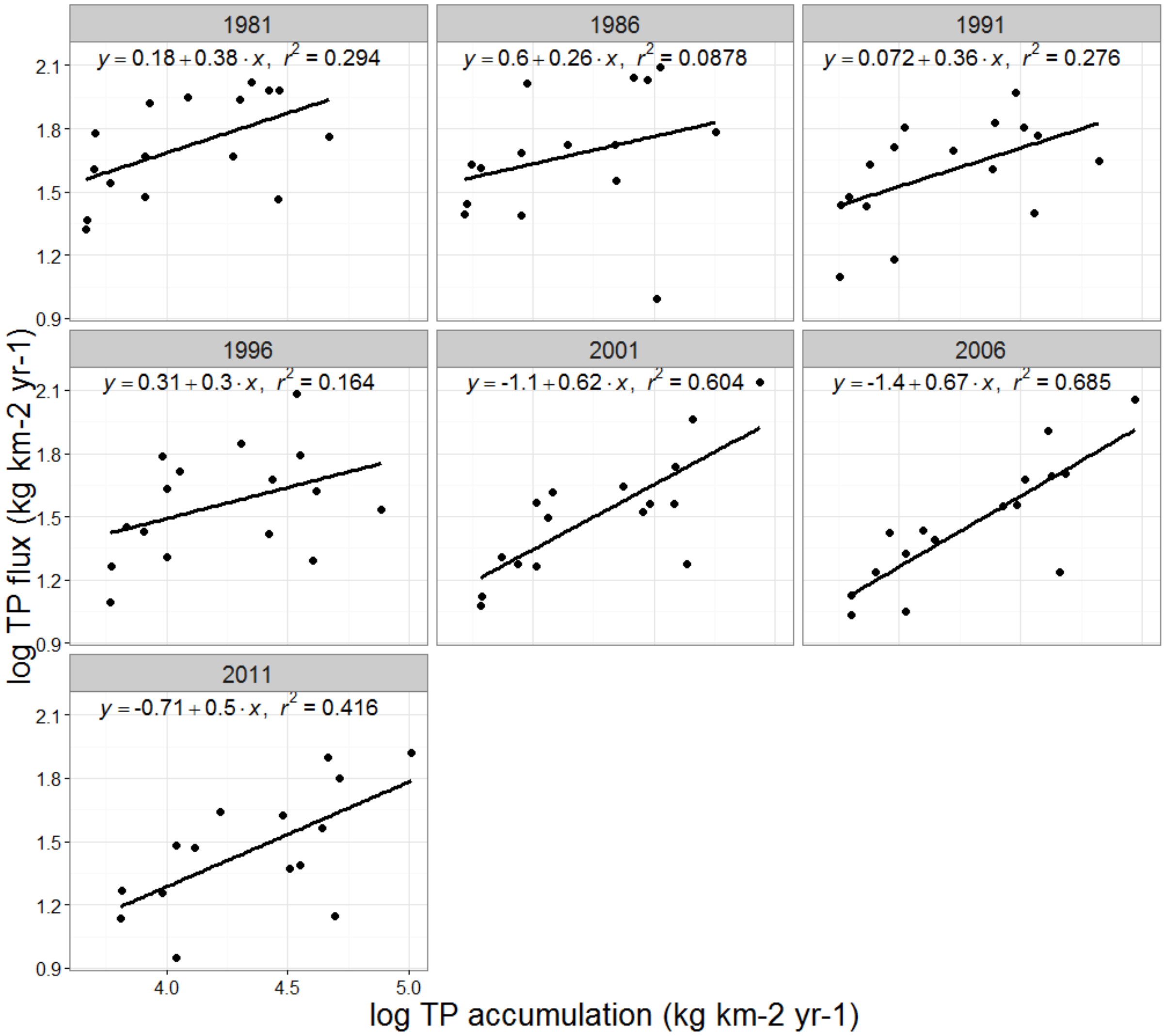

1981
y = 0.18 + 0.38 · x, r^2 = 0.294
1986
y = 0.6 + 0.26 · x, r^2 = 0.0878
1991
y = 0.072 + 0.36 · x, r^2 = 0.276
1996
y = 0.31 + 0.3 · x, r^2 = 0.164
2001
y = -1.1 + 0.62 · x, r^2 = 0.604
2006
y = -1.4 + 0.67 · x, r^2 = 0.685
2011
y = -0.71 + 0.5 · x, r^2 = 0.416
log TP flux (kg km-2 yr-1)
log TP accumulation (kg km-2 yr-1)
0.9
1.2
1.5
1.8
2.1
4.0
4.5
5.0

Figure 5

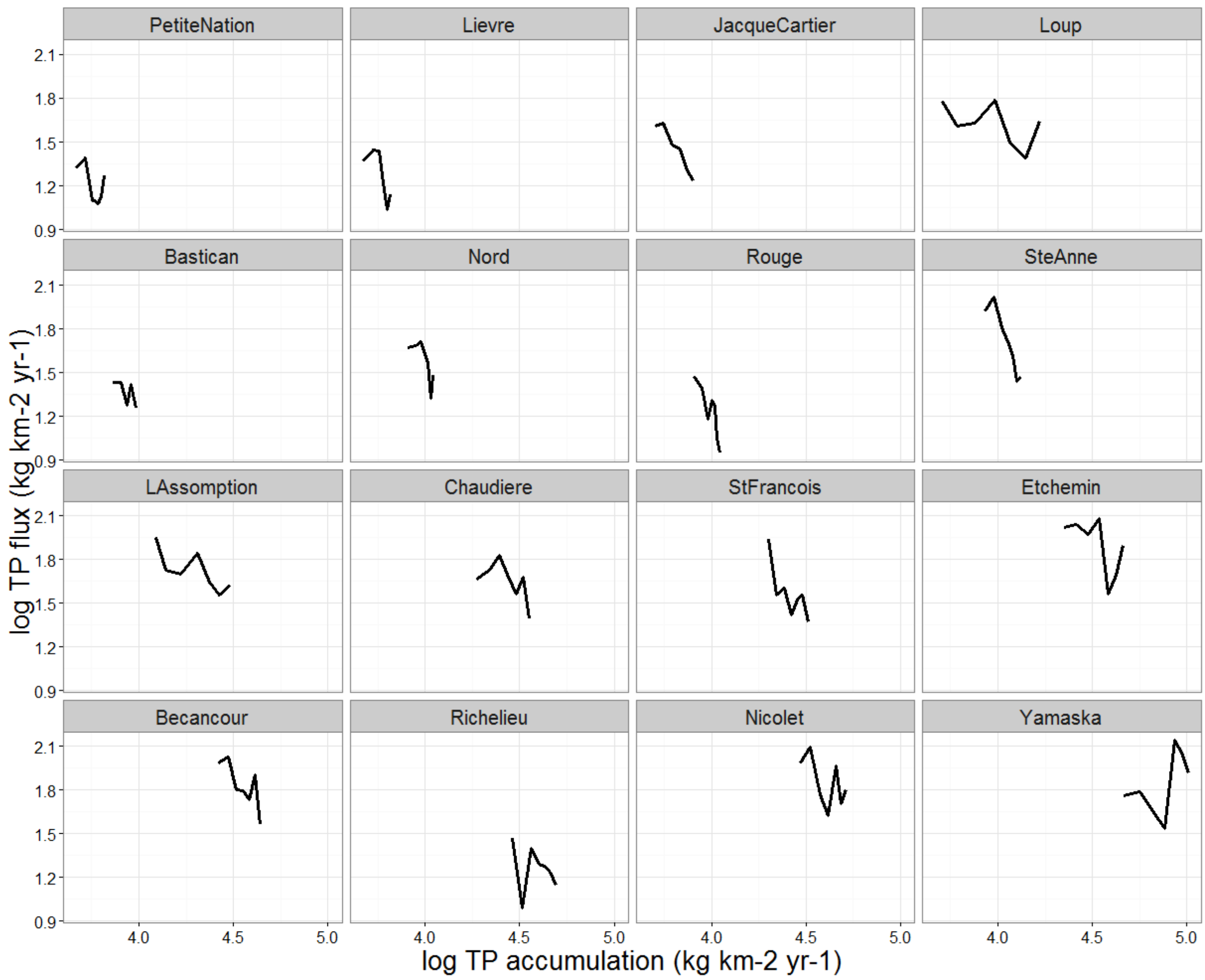
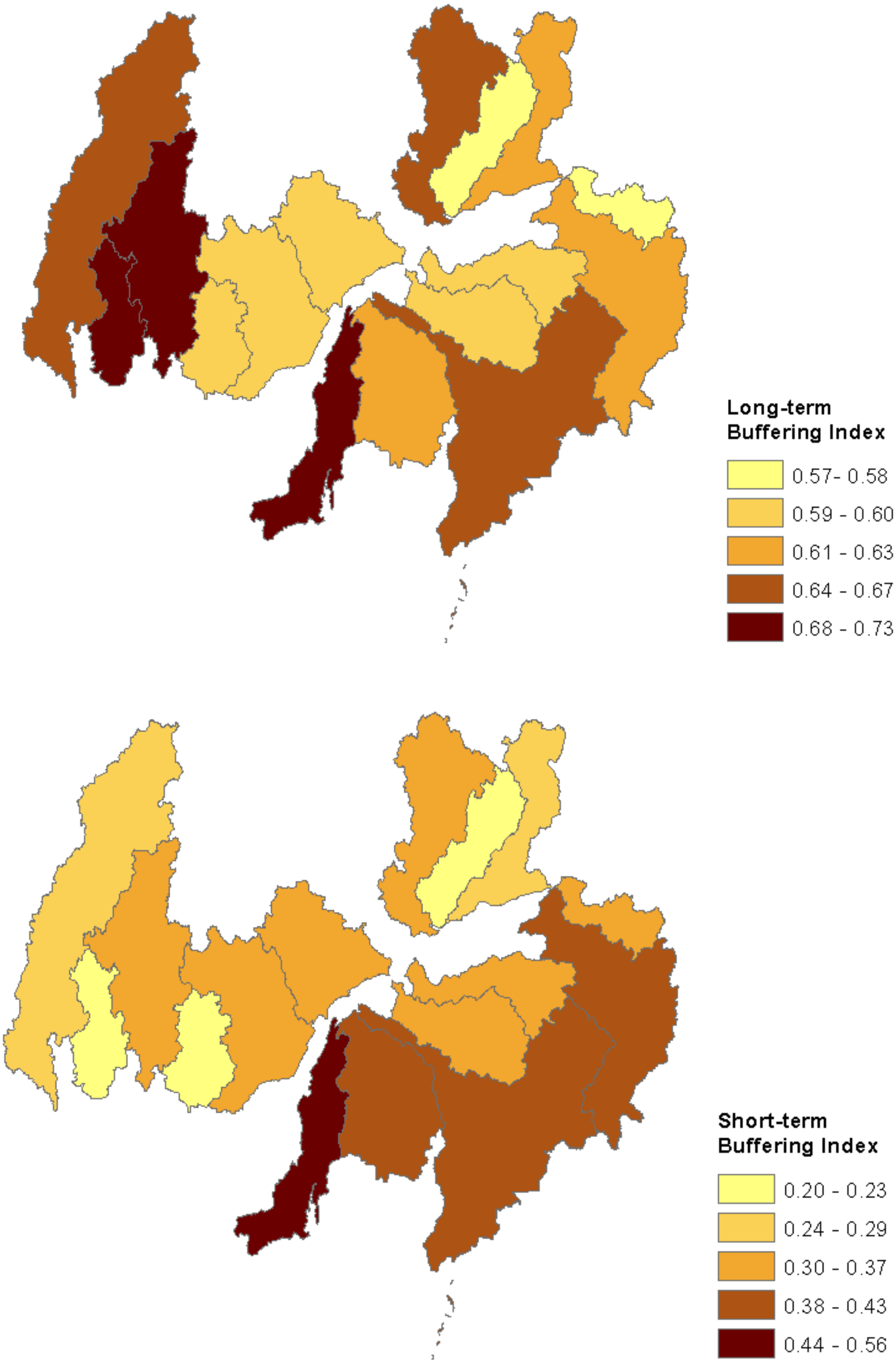

PetiteNation
Lievre
JacqueCartier
Loup
Bastican
Nord
Rouge
SteAnne
LAssomption
Chaudiere
StFrancois
Etchemin
Becancour
Richelieu
Nicolet
Yamaska
log TP flux (kg km-2 yr-1)
log TP accumulation (kg km-2 yr-1)
0.9
1.2
1.5
1.8
2.1
4.0
4.5
5.0

Figure 6



Long-term
Buffering Index
0.57- 0.58
0.59 - 0.60
0.61 - 0.63
0.64 - 0.67
0.68 - 0.73
Short-term
Buffering Index
0.20 - 0.23
0.24 - 0.29
0.30 - 0.37
0.38 - 0.43
0.44 - 0.56

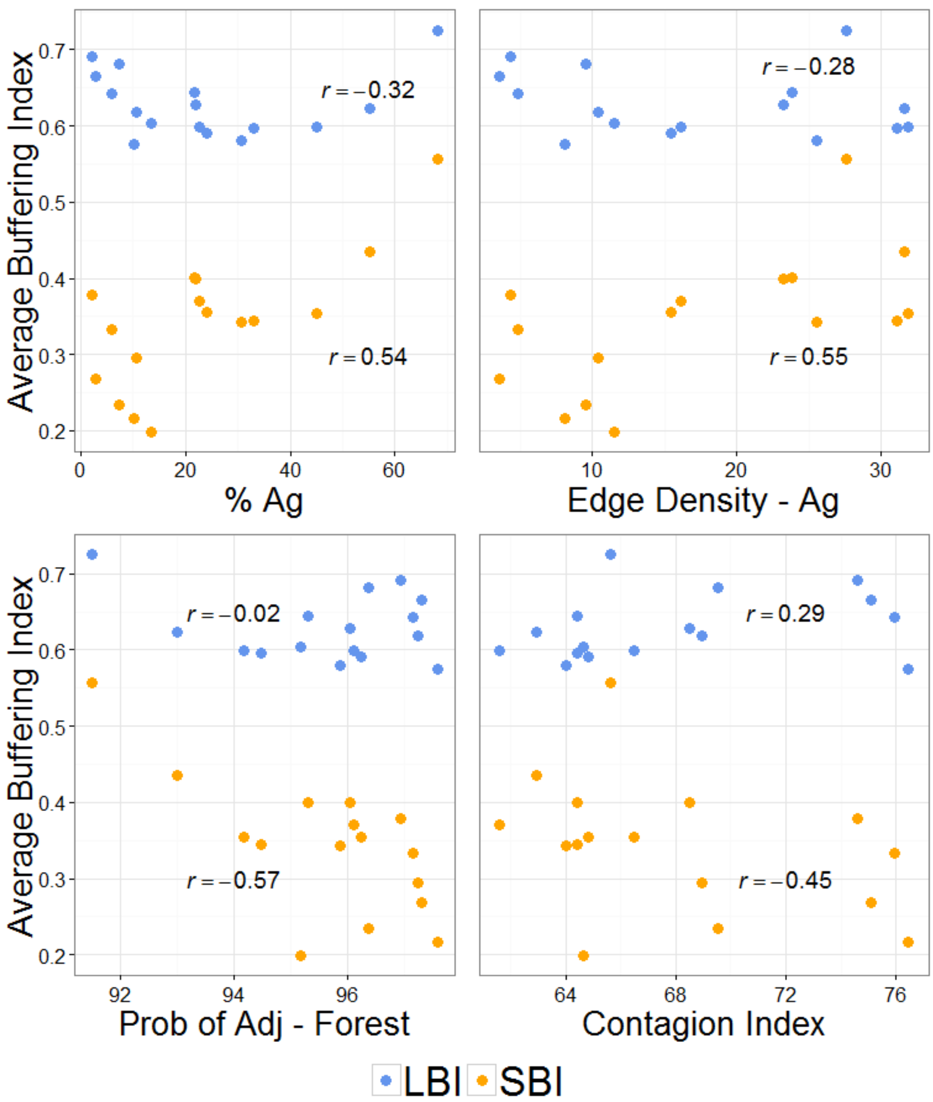
A

B

Figure 7a


Average Buffering Index
r = −0.34
r = 0.49
Ave. Soil P (kg P/ha)
r = 0.63
r = 0.05
Ave. Baseflow Index
r = −0.39
r = −0.52
Ave. Percent Slope
r = 0.46
r = 0.56
Watershed Size (km2)
r = −0.21
r = 0.52
Ave. Market Capital ($/farm)
r = −0.32
r = 0.16
% Tile Drained Ag Land
LBI
SBI

Figure 7b



800

Supplementary Information

Table S.1 Spearman correlations between P flux variables and geochemical, hydrological, and landscape watershed characteristics

Table S.2 Spearman correlations between P flux variables and landscape composition and configuration characteristics

Table S.1

	Buffer index long	Buffer index short	TP Flux	% Clay	pH	Soil P	% Slope	H2O yield	Baseflow index	sinuosity	Watershed size	NDVI	NDVI 100m	Pop density	Field size	Market capital	% tile drained
LBI	1																
SBI	0.32	1															
TP Flux	-0.79	0.14	1														
% Clay	0.15	0.25	-0.08	1													
pH	0.38	0.10	-0.49	0.47	1												
Soil P	-0.34	0.49	0.57	0.41	0.01	1											
% Slope	-0.39	-0.52	0.01	-0.37	-0.15	-0.11	1										
H2O yield	-0.44	-0.49	0.26	-0.45	-0.63	-0.25	0.45	1									
Baseflow index	0.63	0.05	-0.64	0.34	0.64	-0.12	-0.11	-0.48	1								
Sinuosity	0.04	-0.16	-0.09	-0.50	-0.49	-0.34	0.00	0.36	-0.20	1							
Watershed size	0.46	0.56	-0.19	0.14	0.10	-0.14	-0.39	-0.16	0.22	0.24	1						
NDVI	0.19	-0.37	-0.46	-0.11	0.29	-0.62	0.04	-0.10	0.16	0.37	0.14	1					
NDVI 100m	-0.11	-0.36	-0.11	-0.31	0.09	-0.44	-0.08	-0.06	-0.17	0.49	-0.15	0.79	1				
Pop density	-0.24	0.33	0.40	0.36	-0.01	0.77	-0.09	0.04	-0.21	-0.48	-0.20	-0.78	-0.66	1			
Field size	-0.07	0.25	0.16	0.58	0.50	0.32	-0.29	-0.39	0.23	-0.51	0.03	-0.25	-0.13	0.22	1		
Market capital	0.21	0.52	0.37	0.50	0.41	0.71	-0.32	-0.35	0.02	-0.52	-0.03	-0.42	-0.21	0.62	0.73	1	
% tile drained	-0.32	0.16	0.37	0.46	0.31	0.48	-0.34	-0.19	-0.15	-0.55	-0.29	-0.44	-0.14	0.63	0.75	0.79	1

*shaded boxes indicate p-values <0.05

Table S.2

	Buffer index long	Buffer index short	TP flux	% Ag	% Forest	% Devel oped	% Ag 100m	% Forest 100m	% devel oped 100m	% wetland	ED Ag	ED Devel oped	Cohesion Ag	Cohesion Developed	PD Forest	PD wetland	PLADJ forest	Contag
LBI	1																	
SBI	0.32	1																
TP Flux	-0.79	0.14	1															
% Ag	-0.32	0.54	0.62	1														
% Forest	0.24	-0.56	-0.59	-0.98	1													
% Developed	-0.07	0.38	0.27	0.66	-0.71	1												
% Ag 100m	-0.36	0.53	0.66	0.99	-0.96	0.61	1											
% Forest 100m	0.26	-0.54	-0.64	-0.96	0.96	-0.71	-0.96	1										
% Developed 100m	0.01	0.38	0.20	0.51	-0.58	0.94	0.45	-0.62	1									
% Wetlands	0.34	-0.04	-0.25	-0.48	0.44	-0.34	-0.52	0.41	-0.08	1								
ED Ag	-0.28	0.55	0.66	0.94	-0.96	0.63	0.92	-0.94	0.51	-0.29	1							
ED Developed	-0.08	0.39	0.24	0.63	-0.67	0.97	0.57	-0.65	0.93	-0.31	0.58	1						
Cohesion Ag	-0.18	0.39	0.55	0.74	-0.77	0.68	0.74	-0.79	0.55	-0.47	0.71	0.61	1					
Cohesion Developed	-0.06	0.35	0.24	0.60	-0.63	0.93	0.52	-0.62	0.84	-0.33	0.58	0.91	0.66	1				
PD Forest	-0.17	0.49	0.51	0.92	-0.96	0.80	0.89	-0.94	0.68	-0.40	0.92	0.76	0.81	0.76	1			
PD Wetland	0.20	-0.44	-0.59	-0.83	0.83	-0.59	-0.82	0.87	-0.44	0.36	-0.84	-0.46	-0.74	-0.59	-0.80	1		
PLADJ Forest	-0.02	-0.57	-0.38	-0.85	0.88	-0.70	-0.80	0.86	-0.64	0.14	-0.89	-0.68	-0.71	-0.65	-0.92	0.70	1	
Contagion	0.29	-0.45	-0.57	-0.72	0.77	-0.76	-0.68	0.74	-0.79	0.13	-0.73	-0.77	-0.60	-0.61	-0.76	0.47	0.71	1

*shaded boxes indicate p-values <0.05