Running head: Land-Use Legacy Mechanisms

Title: The land-use legacy effect: Towards a mechanistic understanding of time-lagged water quality responses to land use/cover

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Abstract

Numerous studies have linked land use/land cover (LULC) to aquatic ecosystem responses, however only a few have included the dynamics of changing LULC in their analysis. In this study, we explicitly recognize changing LULC by linking mechanistic groundwater flow and travel time models to a historical time series of LULC, creating a land-use legacy map. We then illustrate the utility of legacy maps to explore relationships between dynamic LULC and lake water chemistry. We tested two main concepts about mechanisms linking LULC and lake water chemistry: groundwater pathways are an important mechanism driving legacy effects; and, LULC over multiple spatial scales is more closely related to lake chemistry than LULC over a single spatial scale. We applied statistical models to twelve water chemistry variables, ranging from nutrients to relatively conservative ions, to better understand the roles of biogeochemical reactivity and solubility on connections between LULC and aquatic ecosystem response. Our study illustrates how different areas can have long groundwater pathways that represent different LULC than what can be seen on the landscape today. These groundwater pathways delay the arrival of nutrients and other water quality constituents, thus creating a legacy of historic land uses that eventually reaches surface water. We find that: 1) several water chemistry variables are best fit by legacy LULC while others have a stronger link to current LULC, and 2) single spatial scales of LULC analysis performed worse for most variables. Our novel combination of temporal and spatial scales was the best overall model fit for most variables, including SRP where this model explained 54% of the variation. We show that it is important to explicitly account for temporal and spatial context when linking LULC to ecosystem response.

Keywords: groundwater delivery, riparian dynamics, landscape change, MODFLOW, limnology, nutrients.

1. INTRODUCTION

Land use/land cover (LULC) has been linked to aquatic ecosystem responses, including prominent shifts in trophic status after urbanization or expansion of agriculture. A common approach to investigating aquatic ecosystem responses to changes in LULC involves correlating measured LULC within a specified area to measures of ecosystem condition(s). Several approaches have been used to define and delineate LULC contributing zone(s), including fixed-distance (Strayer et al. 2003, King et al. 2005, Martin & Soranno 2006) and flow distance buffers around a sampling station (Brazner et al. 2007), fixed-distance buffers around a stream (Hunsaker and Levine 1995, Van Sickle et al. 2004, Floyd et al. 2009) with varying distance upstream of a station (Sponseller et al. 2001, Frimpong et al. 2005), and different variations on "watersheds" (Soranno et al. 1996, Hollister et al. 2008). While each approach has had success, there remains uncertainty about how to best represent LULC contributing zones.

Regardless of spatial scale, these approaches assume that solutes originating from the landscape are delivered to the ecosystem at the same time LULC is measured. This is a reasonable assumption for delivery mechanisms that work over short time scales, such as overland flow which involves travel times on the order of days (Burcher 2009). However, groundwater delivery often takes decades to over a century (Pint et al. 2003, Pijanowski et al. 2007). Therefore, water delivered via groundwater likely retains significant characteristics from previous LULCs. In systems where groundwater provides a significant source of water, this temporal mismatch can obscure relationships between LULCs and ecosystem responses. For example, Martin et al. (2011) demonstrated that nested linear models including current LULC along with a series of historical LULC maps better described current lake water chemistry in a groundwater-dominated system. However, their correlative statistical approach could not distinguish amongst the three dominant historical LULC legacy mechanisms: soil storage and overland transport, groundwater delivery, and internal sediment storage and recycling.

Several studies have highlighted the importance of representing groundwater geochemistry processes (e.g., Kratz et al. 1997, Riera et al. 2000, Wayland et al. 2003) and transport delays in models of aquatic ecosystem response (Baker et al. 2006, Fraterrigo and Downing 2008, Kelly et al. 2008), yet only a few have linked the dynamics of changing LULC to a mechanistic understanding of flow paths and travel times (Boutt et al. 2001, Wayland et al. 2002). Pijanowski et al. (2007) coupled modelled historical LULC with a groundwater travel time model to produce a temporally-adjusted mosaic of LULC coined a “land-use legacy map”. They found large discrepancies between current and legacy LULC maps in their study watershed. For example, there was only a 22% agreement in spatial placement of urban LULC type between current and legacy maps. Additionally, 11% of their study area that was currently in human dominated LULCs (i.e., urban and agriculture) was assigned to forested cover type in the legacy map. A legacy map of LULC should thus improve models of ecosystem response by better representing the delayed influence of LULCs associated with slow groundwater pathways.

In addition to the effects of delivery modes on travel times for water to reach surface water ecosystems, chemical constituents are subjected to biogeochemical processing prior to delivery. For example, many human activities can increase the loading of Na^+ and Cl^- ions (e.g., road salting) in a watershed, but these ions are relatively conservative and subjected to little processing once within the aquatic ecosystem. In contrast, nutrients such as P and N are processed extensively once they enter aquatic ecosystems, further complicating their pathway from LULC sources and their eventual delivery to surface waters. Thus without explicitly considering landscape factors that relate to transport processing, the role of legacy pathways may be obscured.

Our goal in this study is to further untangle relationships between LULCs and ecosystem responses by explicitly including temporal scale in our representation of the landscape contributing zone. Specifically, we integrate groundwater travel times with historical LULCs to create a spatially explicit land-use legacy map. We also explore how the contributions of the riparian zone and the watershed might be combined to more completely reflect how water and solutes are delivered to ecosystems; specifically, do models including LULC at both the watershed and riparian scales outperform those at either scale individually? We also consider relationships between water chemistry constituents and LULC within the context of different transport mechanisms and biogeochemical processing across constituents. We use lakes as an example ecosystem to illustrate the utility of this approach, although it is generalizable across other ecosystem types.

We tested two hypotheses associated with mechanistic linkages between LULC and lake water chemistry:

1) Legacy LULC represented through delays in groundwater delivery is a strong driver of current lake water chemistry, and

2) A combination of watershed and riparian LULCs are important to lake water chemistry across a range of constituents.

To test these hypotheses, we apply a process-based groundwater travel time model in conjunction with a time series of spatial LULC data to create a land-use legacy map. We then construct a suite of models using multiple linear regression of twelve water chemistry variables, ranging from nutrients to relatively conservative ions, to better understand the mechanisms linking current and historical LULCs to lake water chemistry. This approach also allows us to examine the roles that biogeochemical reactivity and solubility play in connecting LULCs and aquatic ecosystems. Our a priori expectations were that: 1) chemicals with high solubility (e.g., NO_3) will have stronger relationships with legacy land uses via groundwater flow paths than those with low solubility (e.g., TP), which are affected more by surficial transport processes; 2) chemicals with low reactivity (e.g., relatively conservative ions) will have stronger links to groundwater flow paths than those that are more biogeochemically reactive (e.g., nitrogen and phosphorus).

2. METHODS

2.2 Study Area

This study was conducted in the Huron River Watershed (HRW) in Michigan (Fig. 1), where population growth exceeded 10% between 2000-2010. The 2,359 km^2 watershed has undergone extensive LULC change over the past century, dominated by agriculture in the early 1930's shifting to a primarily suburban society by the 1990's (Hay-Chmielewski et al. 1995, Rutledge and Lepczyk 2002, Martin et al. 2011). Groundwater is the primary source of water to surface water features in this region and the watershed has a high density of lakes that cover a range of historical LULC trajectories (Martin 2010).

2.3 Water Chemistry and Land Use Data

Surface water samples (at about 1m depth) were collected from the deepest locations within 35 lakes during spring 2008 overturn. Samples collected during a mixing event, such as spring overturn, reduces sample bias from within lake spatial variability that occurs during stratified periods and thus samples are more representative of the lake as a whole. Lake size ranged from 0.05 to 2.6 km^2. Samples intended for analysis of nutrients and other dissolved ions were filtered in the field using Millipore membrane filters with 0.45µm pore size and stored on ice during transport. Whole water samples were used for analysis of total nitrogen (TN) and total phosphorus (TP) and were frozen in the field.

Standard Methods (APHA 1998) were used for spectrophotometric analyses of ammonium (NH_4, 4500-NH3-G), nitrate (NO_3, 4500-NO3-E), soluble reactive phosphorus (SRP, 4500-P.E.), and TP (4500-P.E and 4500-N.C). Calcium (Ca), magnesium (Mg), potassium (K), silica (SiO_2) and sodium (Na) were measured by flame atomic absorption spectrophotometry. Chloride (Cl) and sulfate (SO_4) were measured using Dionex membrane-suppression ion chromatography. Total nitrogen concentrations were analyzed using the 2nd derivative of the absorbance curve at 224 nm following persulfate digestion (Crumpton et al. 1992, Bachmann and Canfield 1996).

We used biogeochemical reactivity and solubility to organize water chemistry variables into *a priori* groups (Table 1). Phosphorus and nitrogen were the most reactive variables in our study. We analyzed both particulate (TN and TP) and dissolved forms (NO_3, NH_4, and SRP) of these variables, with the dissolved forms having higher reactivity than particulate forms. Ca, Si, and SO_4 were grouped as reactive ions due to their role as micronutrients in metabolism for macrophytes, diatoms, and bacteria (Wetzel 2001). Ions that play a much smaller role in the metabolism of aquatic organisms (i.e., Cl, K, Na, and Mg) and are therefore influenced more strongly by external supply than internal conversions were classified as conservative (Wetzel 2001).

A multi-temporal GIS database of LULC data was constructed for eight time steps (circa 1800, 1880, 1938, 1955, 1968, 1978, 1995, and 2006; Fig. 2). Pre-settlement LULC data (circa 1800) was developed by the Michigan Department of Natural Resources based on General Land Office surveyors original descriptions. The majority of our LULC data was digitized from aerial photography (1938, 1955, 1968, 1978, and 1995). Digitized LULC polygons for 1978 from the Michigan Resource Information System (MIRIS) served as a base for digitizing all other aerial photo derived data (1938, 1955, 1968, and 1995), following procedures outlined in Rutledge (2001). Photos were scanned (150 dpi), registered and rectified to the 1978 data using county roads. Land use/cover polygons were then digitized and classified based on a modified version of the Anderson et al. (1976) LULC classification scheme at 30 m resolution.

In addition to these observation-based LULC datasets, we created two datasets to approximate changes spanning time periods without available (1800-1938) or comparable data (1995-2008). To span the 100+ years between the pre-settlement LULC and the 1938 LULC, we created a mid-point LULC dataset (1880) by reclassifying all urban and agricultural areas in the 1938 dataset to open/grassland land covers. We assume that the widespread logging and subsistence farming of the area bore closer resemblance to the modern open land/grassland land cover class than to modern urban and industrialized agriculture land uses. To span the 13 years between the 1995 LULC dataset and 2008 (when our water samples were collected), we modified our 1995 LULC dataset following patterns of urban and forest expansion seen in the 2001 and 2006 National Land Cover Datasets (NLCD). NLCD is derived from satellite estimates of LULC, thus to compare this with the aerial imagery based estimates used for the majority of our LULC periods we carefully constrained the allowable changes. Our 2006 LULC map accounts for urban and forest growth under the following constraints: 1) new urban areas were incorporated if they were within one pixel (30 m) of a road, and not classified as water or wetland in 1995; and 2) new forest areas were incorporated if they were classified as open or agriculture in the 1995 dataset as this approximates succession following agricultural abandonment. All data were compiled into a multi-temporal GIS database for analysis.

2.4 Groundwater Travel Time Modeling

We estimated groundwater travel times following procedures similar to Pijanowski et al. (2007), following a two-step procedure, where: 1) a groundwater flow model is constructed which provides water table elevations across the region, and 2) groundwater travel times are estimated using GIS. While this method allows for directly simulating travel times with the groundwater flow model, we have chosen to estimate travel times using the water table map for computational efficiency. Note also that this allows the method to be applied in regions where a groundwater flow model is not available, but maps of hydraulic conductivity and water table elevations are available.

We built our MODFLOW (Harbaugh et al. 2000) groundwater flow model using the Groundwater Modeling System preprocessor (BYU 1994) to facilitate the discretization of geospatial input. A digital version of the Farrand and Bell (1982) Quaternary Geology map with polygons of surficial geology zones was obtained from the Michigan Geographic Data Library. The area within the HRGW has variable surficial geology types, but the region is dominated by glacial outwash and end moraine deposits (Martin, 2010). The model domain consisted of 180,000 cells, approximately 109 x 109 m in size, in one layer. The geometry of the aquifer bottom was interpolated from bedrock elevations (D. Lusch, Michigan State University, personal communication). We used surface elevations from the USGS 1/3rd arc second National Elevation Dataset (NED, resolution ~10m).

We also used the NED surface elevations to develop stream coverages using procedures and tools in ArcInfo 8.3 (ESRI, Inc.): FILL SINKS, FLOW DIRECTION, and FLOW ACCUMULATION. The location of stream cells were defined using a threshold of 15,000 cells using the STREAM DEFINITION tool, which produced a stream network comparable to known hydrography features. Stream cells were combined with cells along lake edges to create a complete representation of hydrography features within the model.

Recharge was estimated using measured flows from the Ypsilanti USGS gauge on the Huron River (#4174800) for 1974-1994. Low flow values for each year in this period of record were averaged and divided by the drainage area, providing the regional recharge estimate of 49 cm/yr. This is approximately 50% of the mean annual precipitation measured in the nearby city of Ann Arbor.

Static water levels from 15,581 wells within the study region recorded in the Michigan Department of Environmental Quality Statewide Groundwater Database were used to interpolate water table elevations across our model domain (HRW plus 1 km buffer). We filtered this dataset to only include wells with the following characteristics: 1) located within the model boundary; 2) reported surface elevation within 2m of NED surface elevation at that location; and 3) casing deeper than the reported static water level. This groundwater surface was then used to delineate a groundwatershed for all river cells upstream of Ypsilanti, as all of the study lakes were located in this region. We use the phrase *groundwatershed* to parallel how *watershed* is used for surface water drainage. Therefore, the groundwatershed represents the area contributing groundwater to a particular point (e.g., a given study lake). The modeled groundwatershed, which we refer to as the Huron River GroundWatershed (HRGW), was then used as the model boundary for the MODFLOW groundwater model. The interpolated static water levels were used as an initial condition in the model.

A digital version of the Farrand and Bell (1982) Quaternary Geology map with polygons of surficial geology zones was obtained from the Michigan Geographic Data Library. The area within the HRGW has variable surficial geology types, but the region is dominated by glacial outwash and end moraine deposits (Martin, 2010).

Hydraulic conductivity (*K*) values were optimized using PEST 10.0 (Doherty 2004), which automatically adjusts *K* values to minimize residuals between simulated and observed groundwater levels. Surficial geology types were grouped during this process to reduce the number of parameters while maintaining low residual errors between observed and simulated groundwater levels. Following *K* optimization, simulated groundwater elevations were similar to observations from the wells database with a R^2 of 0.9 (Martin, 2010). Although a small number of wells had high residual water levels, they were surrounded by other wells with groundwater elevations similar to modeled values. We tested the influence of such outliers on the parameter estimates by removing them from the estimation process and found that they had little effect on the estimated parameters; they were thus kept in the well dataset.

Groundwater flow velocities and two-dimensional travel times were calculated based on Darcy's Law: $Q = -KAi$ where, Q is discharge in m^3/day, K is hydraulic conductivity in m/day optimized in the hydraulic conductivity model, A is the cross sectional area of groundwater flow in m^2, and i is the hydraulic gradient calculated from the simulated water table elevations. Groundwater flow velocity (v, m/day) was calculated as $v = Q/(A * n)$, where n is porosity of the aquifer. Travel times were then calculated by integrating the ratio of flow length to flow velocity along modeled groundwater flow paths, using FLOWDIRECTION and FLOWLENGTH tools in ArcGIS.

2.5 Legacy LULC Map

The legacy map was created by combining simulated groundwater travel times, as described above, with interpreted historical LULC categories for each model cell. The calculated travel times (Fig. 3 – step 1) were categorized based on the mid-point between LULC time steps, adjusting to accommodate for the 2 years between our sampling (2008) and the closest available LULC time step (2006). These reclassified travel times (Fig. 3 – step 2) were then combined with the LULC maps for the particular years to produce a legacy LULC map (Fig. 3 – step 3), which is thus a spatially explicit representation of the historic LULC that corresponds to groundwater delivery times. Finally, we compared the legacy LULCs to the 2006 LULCs, highlighting areas where the two differ (Fig. 3 – step 4).

2.6 Statistical Analyses

The relationships between lake water chemistry variables and both current and legacy LULC were analyzed using multiple linear regression. We used two equations to construct five multiple linear regression models, each designed to analyze specific spatial or temporal aspects of solute transport into lakes.

First, we regressed each chemistry variable against the current proportional cover of the five LULC classes within either the study lake groundwatersheds or within the 50m riparian zone only. Groundwatersheds are used for this analysis rather than surface watersheds because groundwater flow provides the dominant transport pathway in this region. The equation for this model was:

$$y_i = \beta_0 + \beta_{urb}x_{urb} + \beta_{ag}x_{ag} + \beta_{open}x_{open} + \beta_{for}x_{for} + \beta_{wet}x_{wet} + \varepsilon_i \qquad (1)$$

where, y_i is the value of water chemistry i; β_0 is the constant (y-axis intercept); β_p is the regression coefficient for x_p (the proportional cover of LULC *p*: *urb* = urban, *ag* = agriculture, *open* = open, *for* = forest, and *wet* = wetlands); and is the error associated with the model of . This equation thus represents a model where current LULC drives water chemistry. We then applied the same model structure using the legacy LULC, to represent the situation where groundwater is the primary driver of lake water chemistry, and delays in delivery time are explicitly considered.

In our next suite of models, we separated LULC in the groundwatershed from LULC in the 50m riparian zone (i.e. LULC within the riparian zone was masked from the groundwatershed). This builds from the basic equation presented in (1) to allow the impact of LULC in the riparian zone to differ in its impact from LULC in non-riparian portions of the groundwatershed. Although we did not include specific mechanisms for these potential differences, delivery and processing of solutes in riparian zones often operate differently than in the rest of the groundwatershed. Travel time within riparian areas are short and thus were only associated with current LULCs. These regression equations have five LULC types measured over two spatial extents, resulting in ten regression parameters, as follows:

$$y_i = \beta_0 + [\beta_{urb}x_{urb} + \cdots + \beta_{wet}x_{wet}]_{GW-SHED} +$$
$$[\beta_{urb}x_{urb} + \cdots + \beta_{wet}x_{wet}]_{RIPARIAN} + \varepsilon_i \qquad (2)$$

where the bracketed terms followed by "$_{GW\text{-}SHED}$" represent the groundwatershed scale; and "$_{RIPARIAN}$" is measured over the 50 m river buffer. Equation 2 describes the compound effect of overland flow from riparian zones along with the broader groundwatershed influence whereas Equation 1 describes the effects of riparian zones or the groundwatershed in isolation.

Comparing among models with different number of parameters, such as we have with Equations 1 vs. 2, requires careful evaluation of explanatory power and benefits from the evaluation of multiple model fit criteria. We use the coefficient of determination (R^2), Akaike Information Criteria (AIC), and AIC weights to compare the explanatory power of each model (Burnham and Anderson 2002). AIC directly incorporates model fit as the residual sum of squares and was chosen to objectively select amongst models with varying numbers of parameters. Additionally, the structure of our regression models includes LULC proportions that are not independent of each other. As a result of this multicollinearity, we do not interpret the regression coefficients. However, multicollinearity does not prevent the use of the model as a whole or interpretation of the associated R^2 or AIC values (Burnham and Anderson 2002).

3. RESULTS

3.2 Groundwater travel time

Modeled groundwater travel times in the HRGW ranged from <1 yr to >500 years, encompassing each of the eight time classes of available LULC data (Fig. 4). Just over 30% of the HRGW has simulated groundwater travel times between 0-8 years. Approximately 5% of the study area had simulated travel times longer than a century, mostly in the central and northeastern portions of the study area. The distribution of groundwater travel times within the sampled lake groundwatersheds was very similar to those across the HRGW as a whole: the largest difference being sampled lakes had 9% more area represented by the shortest travel time category (data not shown).

3.3 Legacy LULC

Urban and agriculture LULCs dominated the study area in both 2006 and in the legacy map (Fig. 5). However, 2006 LULC had roughly 10% more urban and 6% less agriculture than the legacy map. There were also small differences between open and forest categories. Overall, 14% of the study area had a different LULC in the legacy map than in the 2006 map. The largest difference in legacy LULC between the HRGW and the lake groundwatersheds was for agriculture, as sampled lakes had 13% less agriculture (data not shown).

The riparian zones in this study area were dominated by natural landscape features (>60% water, wetland, and forest combined). However, as is the case in many populated areas, urban LULCs comprised a majority of the remaining riparian zone (>25%).

3.4 Single Scale Regression Results

Models based solely on current groundwatershed LULCs varied widely in their explanatory power, depending on the water chemistry parameters considered. For more conservative parameters, the models typically account for greater than 60% of the variance in water chemistry, while models of more reactive variables did not perform as well. Overall, explanatory power was lowest for the most biogeochemically reactive variables, and generally increased as reactivity decreased (Fig. 6). Phosphorus (TP and SRP) and nitrogen (TN, NO_3, and NH_4) had the lowest R^2 values in these models, approximately 0.2 to 0.3. Models for the reactive ions (Si, Ca, SO_4) generally had an R^2 between 0.4 and 0.6, while the conservative ions (Na, Cl) had the highest explanatory power, greater than 0.7.

Models incorporating legacy LULC at the groundwatershed scale had similar R^2 values (within 2%) to models based solely on current LULC for all but four variables: NH_4, Si, Na, and

Cl (Fig. 6A, Supplementary material). Adding this temporal representation of LULC improved the model for NH_4 and Si by about 5% relative to current LULC. Notably, current LULC models explained 17% and 13% more variation in Na, and Cl, respectively, than their legacy counterparts.

Models spatially restricted to riparian LULC had the highest explanatory power for TP, SRP, and TN; whereas current and legacy groundwatershed models had much higher R^2 values for Ca, Mg, Na, and Cl (Fig. 6A). This indicates that in addition to temporal aspects of biogeochemical transport and processing, riparian LULC plays a particularly important role for nutrients.

3.5 Dual Scale Regression Results

Building on this result, regressions that combine both groundwatershed and riparian-zone LULC (Fig. 6B; based on Equation 2), produced greater explanatory power than models developed for single spatial scales in isolation (Fig. 6A). The groundwatershed summaries incorporated either current or legacy LULC, whereas current LULC was used exclusively for the riparian summaries to represent quicker transport pathways. These models allow regression coefficients for LULC types in riparian areas to differ from those of the groundwatershed. Models including legacy LULC had higher R^2 for TN, NH_4, and SO_4; whereas current LULC models had higher R^2 for Mg, Na, and Cl (Fig. 6B). The remaining six water chemistry variables did not have substantial differences in R^2 between the legacy and current LULC models.

As expected, increasing the number of parameters in the regression models produces a higher R^2; to address this effect we used AIC weights to compare results across the two different model structures in our analyses (Fig. 6, Supplementary Material). For the 12 modeled water chemistry variables, only 2 models using a single spatial scale (either groundwatershed or riparian zone) were strongly highlighted using AIC weights (TN and NO_3), whereas combined spatial scales (groundwatershed plus riparian) were AIC-selected for all 10 remaining water chemistry variables (Fig. 6 and Supplementary material). Of these 10 constituents, models with legacy LULC provided the best fit for TP, NH_4, SO_4, and K; whereas current LULC provided the best fits to SRP, Cl, Mg, and Na. Results for Ca and Si indicate a nearly balanced split between current and legacy LULC (Supplementary material).

4. DISCUSSION

Legacy effects have been shown to be important in terrestrial (Foster et al. 1998, 2003, Chauvat et al. 2007), stream (Harding et al. 1998, McTammany et al. 2007), and lake (Martin et al. 2011) ecosystems. These studies used a correlational approach, relying solely on statistical relations to infer legacy effects. Our study moves beyond the purely correlational approach by combining temporal and spatial changes in LULC with a mechanistic model of groundwater flow paths to create a process-inferred representation of legacy LULC. We show how this legacy map can help connect changes in LULC to important ecosystem characteristics, such as water chemistry in lakes. We also offer a more complete analysis of the relationships between LULC and ecosystem dynamics by allowing LULC within riparian zones to act independently from LULC in groundwatersheds. This mechanistic approach is generalizable across ecosystem types and will likely increase the accuracy and prediction ability in other studies linking LULC with ecosystem responses. Model results would likely benefit by incorporating additional mechanisms known to influence groundwater biogeochemistry and internal lake ecosystem dynamics.

4.1 Temporal Scale: Land-use legacy Effects

Incorporating legacy LULCs in our models improved their fit to observed data, as supported by AIC weights, for many of the water chemistry variables examined. Legacy models outperformed current models for TP, NH_4, SO_4, and K; whereas SRP, TN, Cl, Mg, and Na were better modelled by current LULC. Notably, adding legacy LULC increased explanatory power for NH_4 in comparison to current LULC models, especially when spatial scales were combined. This pattern is consistent with characteristics of delayed transport of NH_4 through saturated soils and deep groundwater pathways, which have been observed in areas of similar underlying geology (Bohkle et al. 2006). Quinlan et al. (2006) found that as groundwater increasingly dominates lake inflows, ammonia concentrations increase relative to nitrate. It is possible that biological nitrogen fixation from decades of historical agriculture in our study area created an influx of ammonia to the groundwater system, comparable to the contaminant plume studied in Bohkle et al (2006), which was observed to be travelling at a much slower rate than groundwater velocities suggest. On the other hand, nitrate moves through groundwater 5-10 times faster than ammonia (Fronczyk et al. 2016), which may explain why both legacy and current NO_3 models performed similarly and no one model was clearly selected by AIC.

Though clear differences between legacy and current groundwatershed LULC models were apparent, our results suggest that the hydrogeology of the study area is dominated by relatively short groundwater travel times, and as such, was an area where groundwater legacies might be less likely to strongly influence the current water chemistry of lakes. Rapid changes in LULC occurred in the HRW between 1938 and 1968 (Martin et al. 2011), a period represented by groundwater ages from only 17% of the groundwatershed (Fig. 4). Changes in LULC largely stabilized during the time represented by the majority of the groundwater ages, showing only small declines in agriculture (2%) and increases in urban (5%) area. Legacy effects through groundwater delivery would be expected to be more apparent in: 1) areas where groundwater travel times are long enough to reach back further in time, or 2) areas where LULC is undergoing rapid conversions within areas of shorter delivery times.

4.2 Spatial Scale: Whole Groundwatershed vs. Riparian Zone

In addition to legacy effects through delays in groundwater travel times, biogeochemical processing in riparian zones (e.g., denitrification, uptake, sorption) also affects relationships between LULC and the chemistry of surface water bodies. Riparian LULCs have been shown to be important for both surface and subsurface transport of materials to streams and lakes (Peterjohn and Correll 1984, Gregory et al. 1991, Groffman et al. 2002, 2004), and have been recognized for reducing nonpoint source pollution (Gregory et al. 1991, Groffman et al. 2002, Craig et al. 2008). We show that some lake water chemistry variables were predicted as well at both catchment and riparian scales (NO_3, SO_4, Si, and K), while riparian LULC had higher explanatory power than groundwatershed LULC for other constituents (TP, SRP, and TN).

Phosphorus (TP and SRP) was best modelled by combined LULC at both the whole groundwatershed and riparian scales. Overland flow is a dominant transport mechanism for particulate forms of phosphorus (Banner et al. 2009, Hoffman et al. 2009b). Because particulates are limited in transport distance, LULC in close proximity (i.e. riparian zones) can deliver more TP to nearby ecosystems. Our results further support the strong relationship between phosphorus and proximal LULC (Johnston et al. 1990, Weller et al. 1996), but also add to the growing interest in the variable effects of subsurface pathways (Fraterrigo and Downing 2008). Notably, our results with the combined groundwatershed and riparian model show a 34 percent increase in explanatory power for SRP over the highest single scale model (riparian buffer). The combined scale models here also increased explanatory power by 8% and 32% for TP and SRP, respectively, from our previous study where current watershed LULC had been selected as the best model (Martin at el. 2011). These results further support that phosphorus is transported via distributed overland and groundwater pathways (Fraterrigo and Downing 2008, Dupas et al. 2015).

Nitrogen can also be transported long distances via groundwater pathways. In groundwater dominated regions, nitrogen is primarily transported in dissolved forms through subsurface pathways (Peterjohn and Correll 1984, Walsh and Kunapo 2009). Here we find that riparian zones are important controls on nitrogen concentrations in lakes, but do not dominate for either NO_3 or NH_4 likely due to long subsurface transport pathways. Single-scale models were selected by AIC weights for TN and NO_3 (riparian-only and all three models, respectively), while model fits for NH_4 benefitted substantially when both spatial scales were combined. This could be due to the highly reactive nature of NH_4 and dependence on specific geochemical conditions along flow pathways, including delayed transport relative to the bulk groundwater flow velocity.

Our study also provides a comparison across water chemistry variables not often included in studies relating LULC to aquatic ecosystem dynamics. We only found three other studies reporting results linking conservative ions to landscape features (Ryszkowski et al. 1999, Boutt et al. 2001, Wayland et al. 2002). In a study of biogeochemical barriers within agriculture fields, termed shelterbelts, Ryszkowski et al. (1999) show the utility of using Ca and Mg as "conservative" tracers for comparison with nutrient concentrations under trees that have been planted as windbreaks. Our results relating relatively conservative ions to catchment and riparian LULC show that diffuse transport of these chemicals through groundwater pathways overwhelms a weaker relationship found with riparian zones. Future studies of landscape connections to aquatic ecosystems can benefit from the use of naturally occurring "conservative" ions as tracers.

To date, there have been numerous methods proposed to incorporate riparian zones, which mainly focus on defining the most appropriate spatial extent of measurement (Baker et al. 2006, van Sickle and Johnson 2008). In contrast, our emphasis was to model the impact of the riparian zone simultaneously with temporal descriptions of LULC over the larger catchment scale. Our results support the early sentiment of Gregory et al. (1991) who say:

> "The importance of riparian zones far exceeds their minor proportion of the land base because of their prominent location within the landscape and the intricate linkages between terrestrial and aquatic ecosystems."

We specifically built these models hierarchically by adding riparian processes to the model of GW legacies in an attempt to statistically define the important role of riparian zones.

5. Conclusions

This study provides the first quantitative assessment of the role of groundwater-generated land use legacies on water quality in lakes. We move beyond place-dependent correlations (i.e. landscape position) and implement a generalizable, mechanistic modeling framework. Land-use legacy is a powerful concept to link historical changes on the landscape to current water chemistry via a wide range of groundwater travel times. Critically, managers should be aware of the consequences of these legacies when setting expectations related to water quality goals. As such, changes on the landscape made today may not fully impact water quality for decades to come.

This study analyzes a suite of 12 water chemistry constituents across a range of biogeochemical reactivity and solubility to better understand the role of spatial LULC influences (i.e. riparian vs. groundwatershed) versus temporal influences (i.e. current vs. legacy). We find that models built solely on groundwatershed land use (either current or legacy) can account for most of the variability of lake water chemistry for conservative constituents. Whereas land cover alone describes far less variability of the more biogeochemically reactive constituents, indicating other mechanisms not controlled by land use play an important role. If the effects of riparian zone LULC are incorporated along with groundwatershed LULC, model explanatory power increases substantially. These combined models were selected as the most supported models according to AIC weights for 10 out of the 12 constituents.

Models incorporating legacy land use were selected for TP, NH_4, SO_4, and K, and performed as well as current LULC models for NO3, Si, and Ca. The importance of land-use legacy is likely to be much more substantial in watersheds with longer groundwater travel times and more land use change than our pilot study area, the Huron River Watershed. We anticipate, however, that the combined-scale modeling results are broadly applicable elsewhere, as these relationships are dictated more by biogeochemical reactivity occurring at timescales much shorter than legacy transport.

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Table 1. Characteristics of the study lakes including water chemistry variables, categorized into a priori groups. Minimum, maximum, mean, and coefficient of variation are shown for each variable. Phosphorus species include total phosphorus (TP), and soluble reactive phosphorus (SRP). Nitrogen species include total nitrogen (TN), nitrate (NO_3), and ammonia (NH_4).

Reactive ions include calcium (Ca), silica (SiO_2), and sulfate (SO_4). Relatively conservative ions include chloride (Cl), potassium (K), magnesium (Mg), and sodium (Na).

Lake characteristic	Minimum	Maximum	Mean	CV
Phosphorus				
TP (µg/L)	5.4	45.0	24.1	0.37
SRP (µg/L)	0.33	3.45	1.15	0.56
Nitrogen				
TN (mg/L)	0.53	1.85	0.94	0.30
NO_3 (mg/L)	0.00	1.03	0.20	1.20
NH_4 (µg/L)	3	143	36	0.96
Reactive Ions				
Ca (mg/L)	16	92	52	0.38
SiO_2 (mg/L)	0.00	5.61	1.53	0.88
SO_4 (mg/L)	1.9	88.4	26.1	0.83
Conservative Ions				
Cl (mg/L)	2	242	66	0.90
K (mg/L)	0.58	7.53	2.09	0.57
Mg (mg/L)	3.9	24.7	15.4	0.26
Na (mg/L)	0.9	77.3	24.0	0.88

Figure 1. Map of the Huron River Watershed within the lower peninsula of Michigan, including detailed hydrography features and the cities of Ann Arbor and Ypsilanti. Lakes sampled for this study are indicated by black dots.

Figure 2. Map of land use/cover (LULC) from pre-settlement, 1880, 1938, 1955, 1968, 1978, 1996, and 2006 within the Huron River Watershed.

Figure 3. Steps to create a land-use legacy map. Step 1: The simulated groundwater travel times in years were calculated for each model cell downgradient to a discharge point (e.g. river) using Darcy's Law. Step 2: Groundwater travel time year was reclassified to match times with LULC data (2006, 1995, 1978, 1968, 1955, 1938, 1880 or Pre-settlement). Step 3: LULC type for each cell was assigned using time categories from step 2. Step 4: LULC from 2006, masking areas where LULC is the same between the legacy and 2006 maps.

Figure 4. Groundwater travel times reclassified to represent the 8 time steps of land cover data. Time classes span from midpoint to midpoint around a specified year. Percent of each groundwater travel time category within the Huron River groundwater groundwatershed (HRGW) are shown in the pie chart.

Figure 5. Land use/cover within the model area showing A) 2006 and B) legacy land use/cover, and C) areas with different land use/cover types between the 2006 and the legacy land use/cover.

Figure 6. Coefficients of determination (R^2) from regression models of 12 water chemistry variables using A) Equation 1 or B) Equation 2 with current and legacy LULC characterized over the groundwatershed (**GW**) and/or the riparian zone (**Rip.**). Best-fit models selected by AIC weights are shown with heavy black squares for each water chemistry variable. NOTE: chemistry variables are arranged from most reactive (phosphorus) to least reactive (conservative ions). Lines connecting categorical data markers have been added to aid interpretation.

Figure 1.

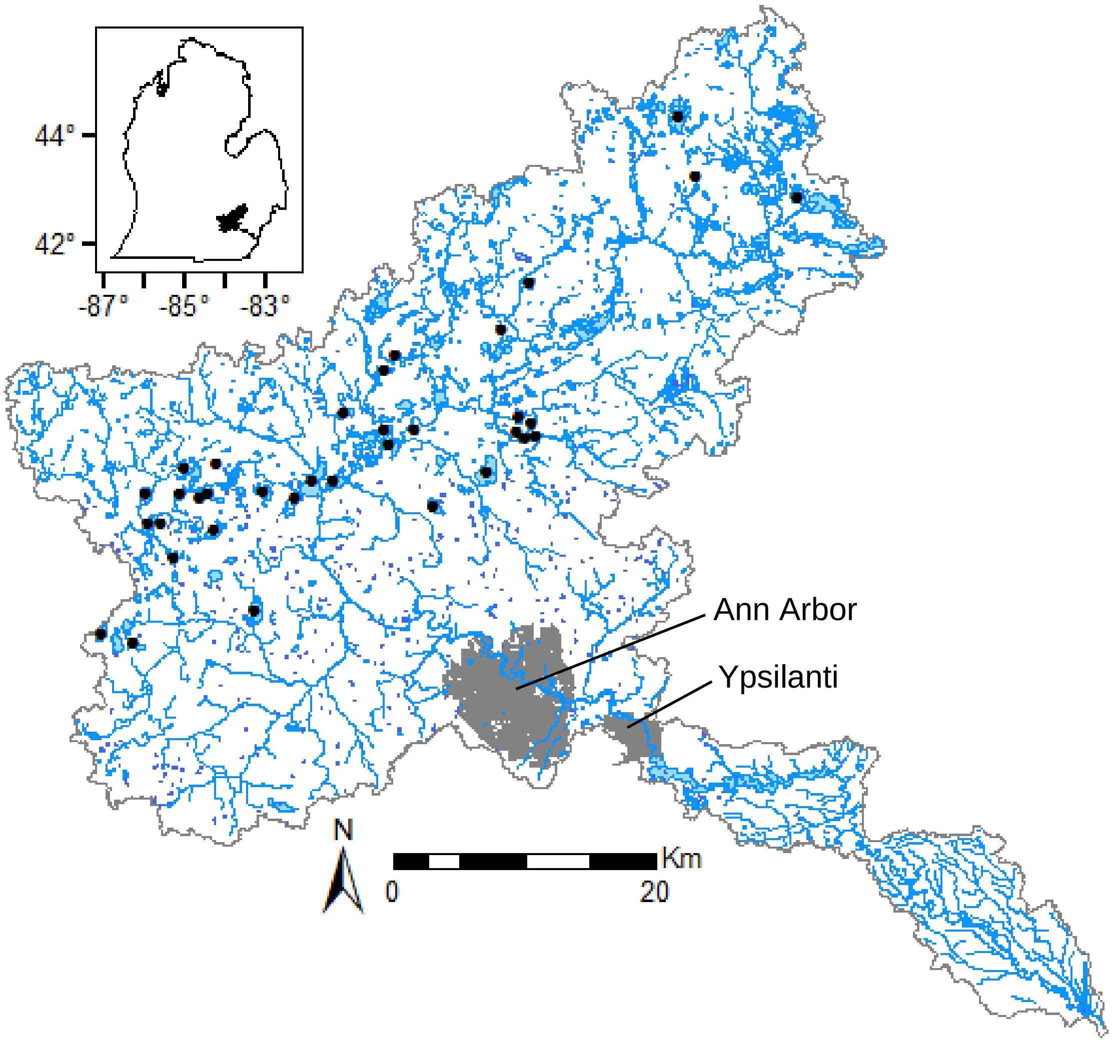


Figure 2.

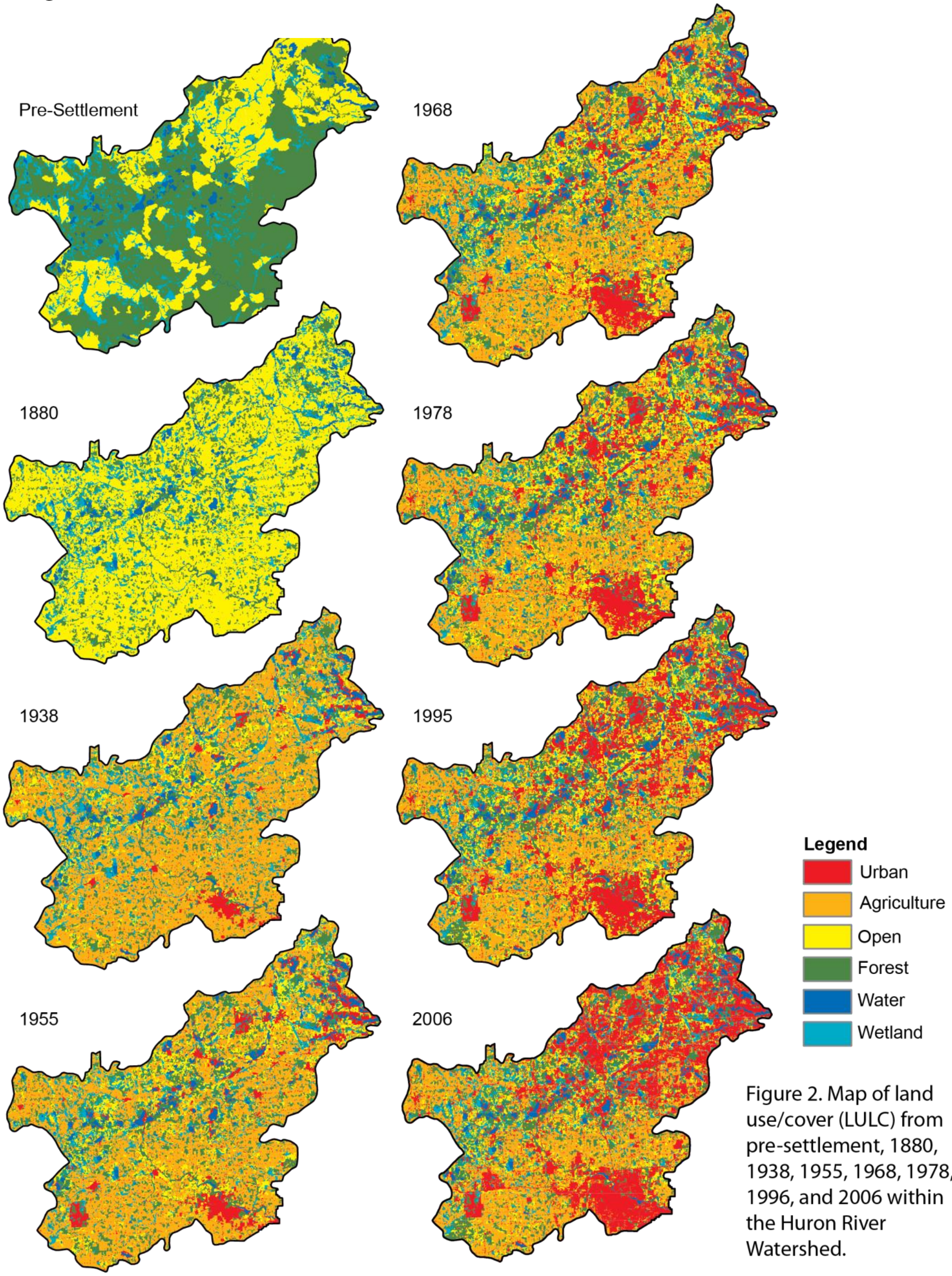


Figure 2. Map of land use/cover (LULC) from pre-settlement, 1880, 1938, 1955, 1968, 1978, 1996, and 2006 within the Huron River Watershed.

Figure 3.

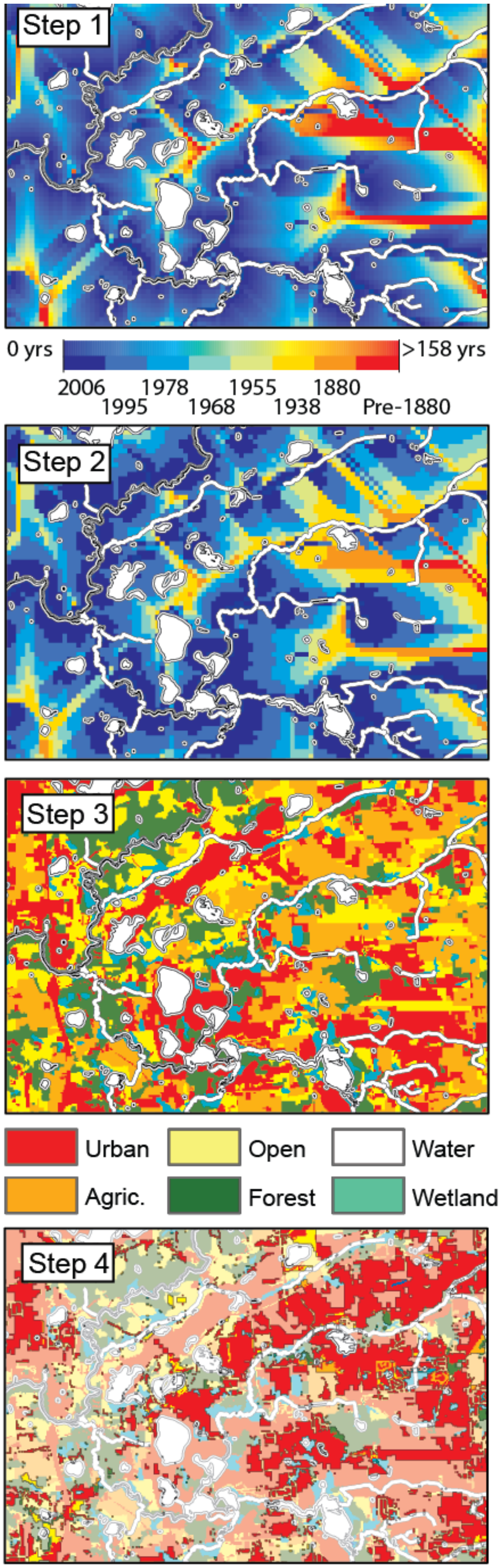
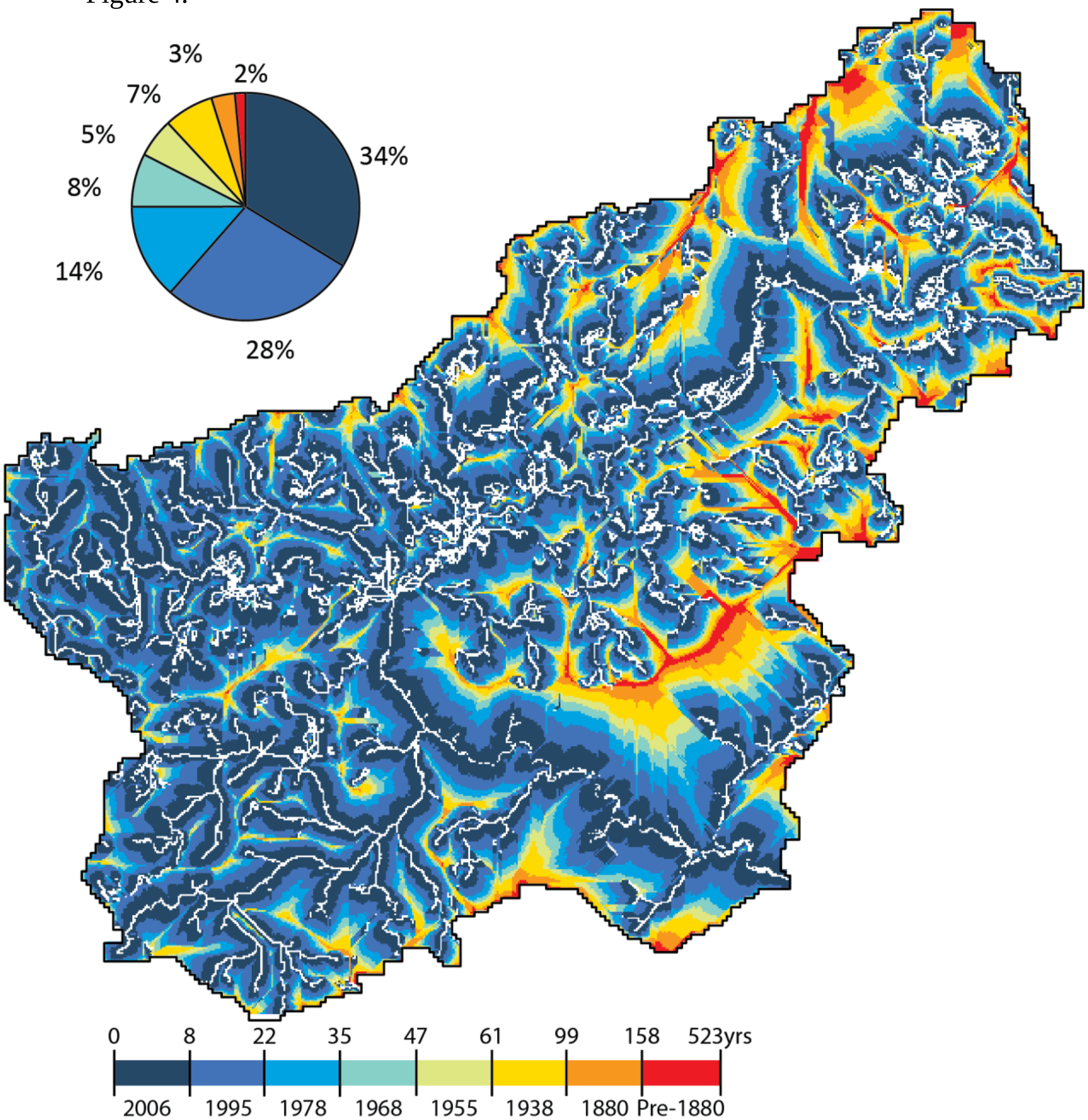

Step 1
0 yrs
>158 yrs
2006
1995
1978
1968
1955
1938
1880
Pre-1880
Step 2
Step 3
Urban
Open
Water
Agric.
Forest
Wetland
Step 4

Figure 4.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Pre-1880

Figure 5.

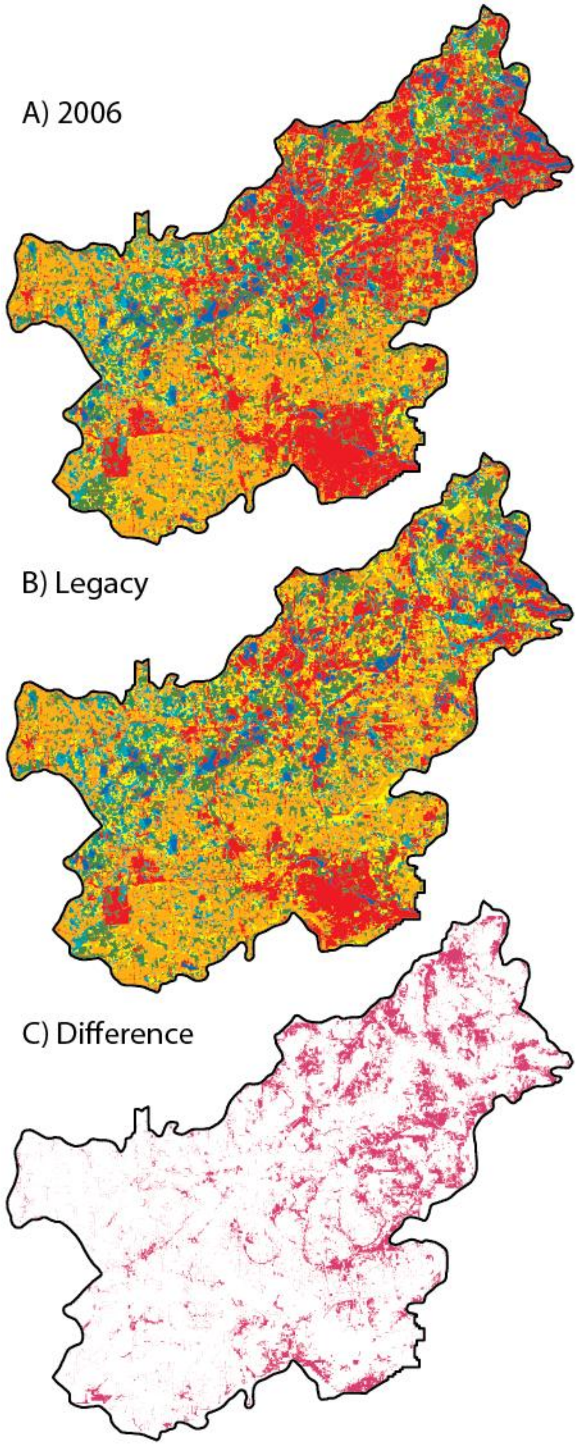
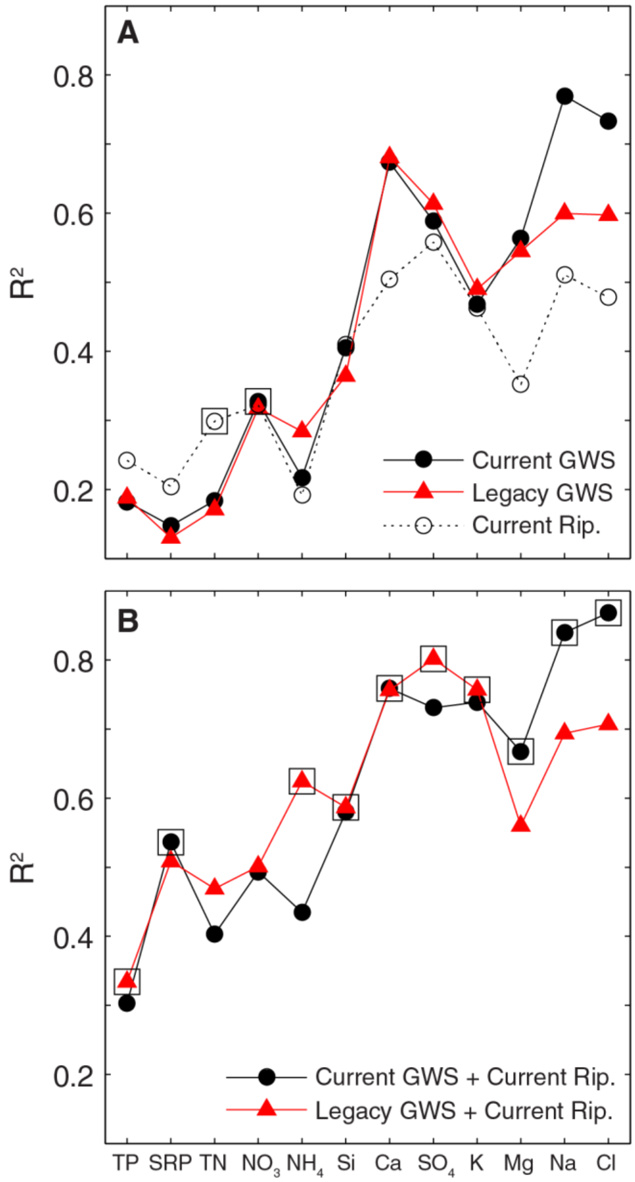


Figure 6.

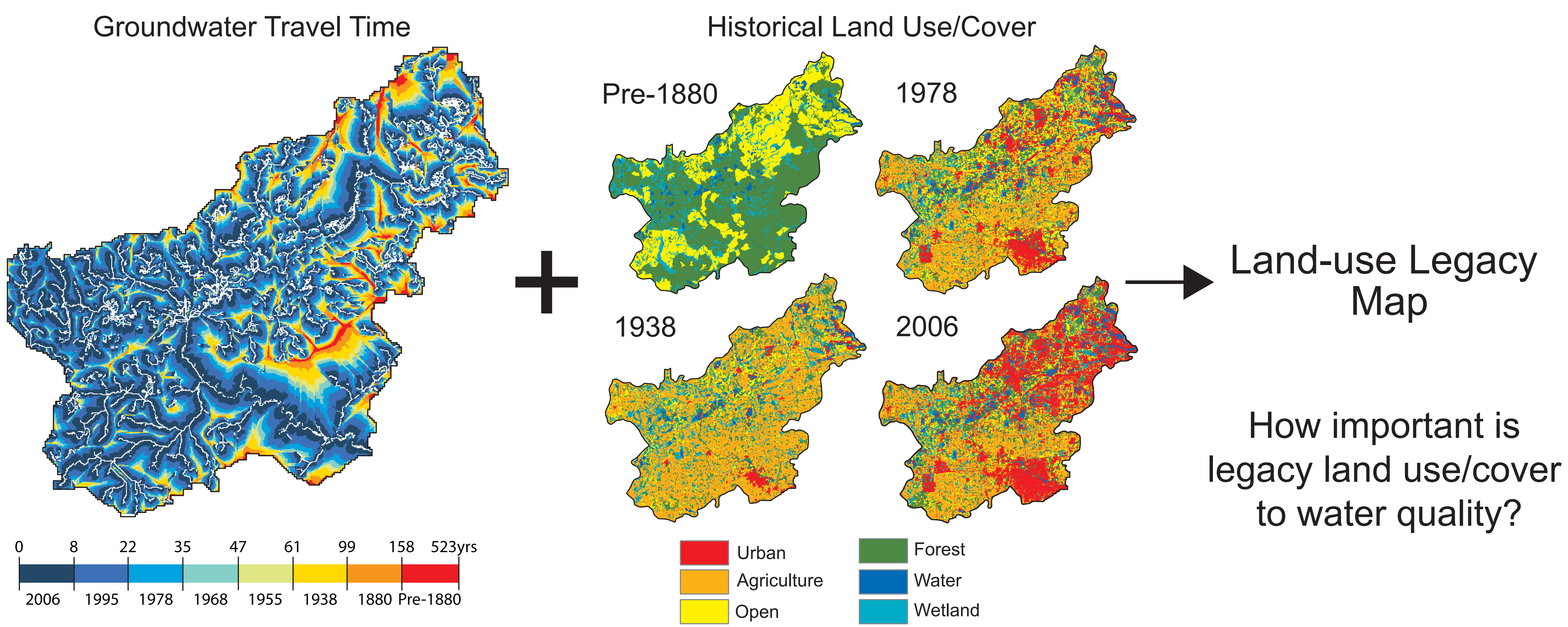

A
0.8
0.6
0.4
0.2
R²
Current GWS
Legacy GWS
Current Rip.
B
0.8
0.6
0.4
0.2
R²
Current GWS + Current Rip.
Legacy GWS + Current Rip.
TP
SRP
TN
NO3
NH4
Si
Ca
SO4
K
Mg
Na
Cl

SUPPLEMENTARY MATERIAL

Goodness of fit measures used to construct Figure 6: R^2, Akaike Information Criteria (AIC) and AIC weights. The Bold values in the table indicate the best selection based on the criteria at the top of the column below. Italicized cells indicate where AIC weights are too close to indicate strong model preference.

Model		R^2	AIC	AIC weight
TP	Current Groundwatershed	0.18	378	0
	Legacy Groundwatershed	0.19	378	0
	Current Riparian Buffer	0.24	374	0
	Legacy Groundwatershed + Current Riparian Buffer	**0.33**	255	**68**
	Current Groundwatershed + Current Riparian Buffer	0.30	257	32
SRP	Current Groundwatershed	0.15	104	0
	Legacy Groundwatershed	0.13	105	0
	Current Riparian Buffer	0.20	101	0
	Legacy Groundwatershed + Current Riparian Buffer	0.51	64	27
	Current Groundwatershed + Current Riparian Buffer	**0.54**	62	**73**
TN	Current Groundwatershed	0.18	9.3	2
	Legacy Groundwatershed	0.17	10.1	1
	Current Riparian Buffer	0.30	1.6	**96**
	Legacy Groundwatershed + Current Riparian Buffer	**0.47**	14	0
	Current Groundwatershed + Current Riparian Buffer	0.40	18	0
NO_3	Current Groundwatershed	0.33	-23	***40***
	Legacy Groundwatershed	0.32	-22	*28*
	Current Riparian Buffer	0.32	-22	*33*
	Legacy Groundwatershed + Current Riparian Buffer	**0.50**	-1	0
	Current Groundwatershed + Current Riparian Buffer	0.49	-0.7	0
NH_4	Current Groundwatershed	0.22	502	0
	Legacy Groundwatershed	0.28	498	0
	Current Riparian Buffer	0.19	504	0
	Legacy Groundwatershed + Current Riparian Buffer	**0.62**	323	**100**
	Current Groundwatershed + Current Riparian Buffer	0.43	337	0
Si	Current Groundwatershed	0.41	167	0
	Legacy Groundwatershed	0.36	170	0
	Current Riparian Buffer	0.41	166	0
	Legacy Groundwatershed + Current Riparian Buffer	**0.59**	110	***57***
	Current Groundwatershed + Current Riparian Buffer	0.58	111	*43*
Ca	Current Groundwatershed	0.67	393	0
	Legacy Groundwatershed	0.68	392	0
	Current Riparian Buffer	0.50	414	0
	Legacy Groundwatershed + Current Riparian Buffer	0.76	273	*45*

	Current Groundwatershed + Current Riparian Buffer	**0.76**	273	***55***
SO_4	Current Groundwatershed	0.59	414	0
	Legacy Groundwatershed	0.61	411	0
	Current Riparian Buffer	0.56	418	0
	Legacy Groundwatershed + Current Riparian Buffer	**0.80**	274	**99**
	Current Groundwatershed + Current Riparian Buffer	0.73	284	1
Cl	Current Groundwatershed	0.73	529	0
	Legacy Groundwatershed	0.60	550	0
	Current Riparian Buffer	0.48	563	0
	Legacy Groundwatershed + Current Riparian Buffer	0.71	356	0
	Current Groundwatershed + Current Riparian Buffer	**0.87**	329	**100**
K	Current Groundwatershed	0.47	132	0
	Legacy Groundwatershed	0.49	130	0
	Current Riparian Buffer	0.46	132	0
	Legacy Groundwatershed + Current Riparian Buffer	**0.76**	83	**77**
	Current Groundwatershed + Current Riparian Buffer	0.74	86	23
Mg	Current Groundwatershed	0.56	257	0
	Legacy Groundwatershed	0.54	260	0
	Current Riparian Buffer	0.35	278	0
	Legacy Groundwatershed + Current Riparian Buffer	0.56	181	1
	Current Groundwatershed + Current Riparian Buffer	**0.67**	171	**99**
Na	Current Groundwatershed	0.77	409	0
	Legacy Groundwatershed	0.60	437	0
	Current Riparian Buffer	0.51	447	0
	Legacy Groundwatershed + Current Riparian Buffer	0.69	287	0
	Current Groundwatershed + Current Riparian Buffer	**0.84**	265	**100**


Groundwater Travel Time
Historical Land Use/Cover
Pre-1880
1978
1938
2006
0 8 22 35 47 61 99 158 523yrs
2006 1995 1978 1968 1955 1938 1880 Pre-1880
Urban
Agriculture
Open
Forest
Water
Wetland
Land-use Legacy Map
How important is legacy land use/cover to water quality?