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Key Points:
- Key scaling relationships for lake CO_2 concentrations vary geographically
- There are contiguous regions of similar scaling relationships
- Regional variation in lake CO_2 regulation reflects climate and land-use gradients

Supporting Information:
- Supporting Information S1
- Figure S1

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Continental-scale variation in controls of summer CO_2 in United States lakes

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Abstract Understanding the broad-scale response of lake CO_2 dynamics to global change is challenging because the relative importance of different controls of surface water CO_2 is not known across broad geographic extents. Using geostatistical analyses of 1080 lakes in the conterminous United States, we found that lake partial pressure of CO_2 (pCO_2) was controlled by different chemical and biological factors related to inputs and losses of CO_2 along climate, topography, geomorphology, and land use gradients. Despite weak spatial patterns in pCO_2 across the study extent, there were strong regional patterns in the pCO_2 driver-response relationships, i.e., in pCO_2 "regulation." Because relationships between lake CO_2 and its predictors varied spatially, global models performed poorly in explaining the variability in CO_2 for U.S. lakes. The geographically varying driver-response relationships of lake pCO_2 reflected major landscape gradients across the study extent and pointed to the importance of regional-scale variation in pCO_2 regulation. These results indicate a higher level of organization for these physically disconnected systems than previously thought and suggest that changes in climate and land use could induce shifts in the main pathways that determine the role of lakes as sources and sinks of atmospheric CO_2.

Plain Language Summary In this study we show that changes in climate and terrestrial landscapes could affect which are the main mechanisms responsible for the widespread emissions of CO_2 by lakes. Although mechanisms such as aquatic primary production, respiration by microorganisms, or terrestrial loadings of carbon have been studied extensively, their relative importance across broad geographic extents with different climate or land use remains unknown. Based on an analysis of 1080 lakes distributed across the continental U.S., we show that lake CO_2 dynamics depend on the climate and landscape context where these lakes are found, such as precipitation, elevation, percent agriculture, or wetlands in the lakes catchments. We observed a widespread effect of in-lake primary production, while the color of water, which has often been identified as one of the main controls of lake CO_2 in northern lakes, was important in only a small fraction of the lakes studied. Our results show that controls on lake CO2 dynamics vary geographically and that considering that variation will be important for creating accurate global carbon models.

1. Introduction

Inland waters emit and store globally significant amounts of carbon, despite the fact that they only cover about 3% of Earth's nonglaciated land surface [*Cole et al.*, 2007; *Tranvik et al.*, 2009; *Raymond et al.*, 2013, *Verpoorter et al.*, 2014]. Human-driven changes in temperature and atmospheric deposition, which impact ice cover and organic carbon and nutrient loadings to inland waters, have been argued to increase both CO_2 emission and organic carbon storage in aquatic ecosystems [*Downing et al.*, 2008; *Lapierre et al.*, 2013; *Regnier et al.*, 2013; *Anderson et al.*, 2014]. The same drivers, however, may lead to a decrease in CO_2 emissions from lakes as a result of organic carbon burial in lake sediments and increasing pH [*Balmer and Downing*, 2011; *Finlay et al.*, 2015]. These contrasting findings emphasize the absence of consistent and predictable patterns of aquatic carbon cycling along climate and land use gradients at broad spatial extents, which limits prediction of the changing contribution of lakes to continental carbon cycling in response to global change.

Understanding the broad-scale controls of lake CO_2 regulation is limited by a lack of knowledge about how the relationships between lake CO_2 and its main drivers vary geographically. Past studies have gathered large amounts of data on lake pCO_2 and optimized predictive linear models either at intraregional or global scales [e.g., *Sobek et al.*, 2005; *Marotta et al.*, 2009; *Lapierre and del Giorgio*, 2012], but these scales of study and this statistical approach may miss the scale at which much of the variability and interactions with landscape occur. Studies aimed at understanding carbon cycling and performed in different individual regions often yield contradictory findings on the relationships between lake partial pressure of CO_2 (pCO_2) and in-lake controls, such as the concentrations of dissolved organic carbon (DOC) and chlorophyll *a* (Chl *a*) that dictate the rates of production and uptake of CO_2 [*Sobek et al.*, 2003; *Webster et al.*, 2008; *Finlay et al.*, 2009; *Roehm et al.*, 2009; *Lapierre and del Giorgio*, 2012]. This suggests that approaches that have previously been successful may not apply when evaluating global carbon cycling. Moreover, the recent studies that have explicitly addressed cross-regional variation in limnological properties either focused on other elements or relied on predefined geographical units—or ecological regions—that may not capture the environmental gradients relevant for aquatic carbon cycling [*Fergus et al.*, 2011; *McDonald et al.*, 2013; *Seekell et al.*, 2014a; *Soranno et al.*, 2015]. Therefore, there is a need for novel frameworks and approaches to explore how lake pCO_2 driver-response relationships vary freely over space, across the broad climate and landscape gradients that may modulate the response of pCO_2 to different drivers.

Relating lake CO_2 to landscape properties is challenging, however, because of a scale mismatch between the temporal variability in lake carbon cycling and in the geographical variables that are routinely available at broad spatial extents. As a consequence, there is typically no direct relationship between geographic variables and lake concentrations of CO_2 [*Lapierre and del Giorgio*, 2012; *Sepulveda-Jauregui et al.*, 2015]. This is in part because the effect of static, long-term averages of geographic properties on lake pCO_2 is transmitted to the freshwater carbon cycle indirectly through drivers that respond to climate and landscape signals and that are correlated with in-lake processes [*Lapierre et al.*, 2015]. Here we used a two-step statistical approach to (1) understand continental-scale variation in within-lake regulation of surface water pCO_2 (using geographically weighted regression and cluster analysis) then (2) establish the regional-scale landscape and climate contexts leading to geographic differences in within-lake pCO_2 regulation.

2. Methods

2.1. Data Sources

Water chemistry and landscape data were obtained from the U.S. Environmental Protection Agency (EPA) National Lake Assessment [*U.S. Environmental Protection Agency* (*USEPA*), 2009], which used a probability-based survey across adapted Level II Ecoregions [*Omernik*, 1987] consisting of similar land use, topography, climate, and natural vegetation. The data include both natural lakes and artificial water bodies (ranging from small private impoundments to large multipurpose reservoirs); all are larger than 4 ha and deeper than 1 m. A total of 1157 individual sites was sampled between June and September of 2007; 1080 sites had data for all of the variables used in this study. We calculated the mean value for sites that were sampled more than once.

Water sampling and geographical analyses were performed based on standard chemistry and geographic information systems procedures, respectively (see *USEPA* [2009] for details). We used the data provided by EPA, except for the partial pressure of CO_2 (pCO_2, in μatm), which we calculated from the estimated CO_2 concentration based on pH, calculated alkalinity, and temperature and then transformed into partial pressure based on Henry's law [*Stumm and Morgan*, 1996]. We used "calculated alkalinity" based on major ion chemistry to avoid biases associated with the contributions of organic acids to alkalinity measurements made by Gran titration [*Wetzel and Likens*, 2000; *Hunt et al.*, 2011; *Waller et al.*, 2012; *Abril et al.*, 2015]. Although pH can be difficult to measure accurately in poorly buffered waters with low pH values, all lakes in the study had pH $>=$ 5 (Table 1), minimizing this as a potential source of uncertainty in our analysis [e.g., *Seekell et al.*, 2014b; *Seekell and Gudasz*, 2016].

There is a lack of data on the direct processes involved in the production and losses of CO_2 in lakes and in the loadings of carbon and nutrients from land to water at broad scales. This issue relates to trade-offs between the spatial extent of studies and the logistical difficulties in routinely performing the measurements. Because the monitoring programs that collect data at broad spatial extents have traditionally been developed to address questions (e.g., eutrophication and acidification) that are not directly related to the aquatic carbon cycle, researchers have to rely on surrogate variables that inform on aquatic C cycling processes. Therefore,

Table 1. Descriptive Statistics of the Main Variables Used in the Analyses

Variable	Units	Mean	Median	2.5–97.5 Percentiles
pCO_2	μatm	1,465.2	588.3	6.4–10,553
Chl *a*	μgL^{-1}	24.6	7.4	0.6–192.4
Color	PCU	15.1	11.0	0–50
Alkalinity	μequivalent L^{-1}	2,479.1	1,691.1	49.5–12,587.3
TN:TP	unitless	76.3	46.8	6.0–311.9
TN	$\mu g\ L^{-1}$	1,002.7	562.3	83.8–4,466.8
TP	$\mu g\ L^{-1}$	96.5	24.0	1.0–616.6
Secchi	m	2.2	1.4	0.2–8.2
pH		8.1	8.2	6.2–9.6
Elevation	m	618.9	338.8	9.1–2,657.9
Agriculture	% of catchment	20.2	8.1	0–76.6
MAP	mm y^{-1}	853.0	863.0	273–1,493
Wetlands	% of catchment	4.9	1.5	0–26.8

we used a series of well-known proxies of CO_2 production and loss processes to explain variation in lake pCO_2. These include (i) DOC and colored DOC (a tracer of terrestrial DOC) [see *Lapierre et al.*, 2013] (hereafter referred to as "Color") as proxies of CO_2 production from DOC processing, (ii) nutrients (total N (TN) and total P (TP)) and Chl *a* as proxies of CO_2 consumption from pelagic primary production [*Balmer and Downing*, 2011], and (iii) total alkalinity (based on Gran titration, which comprises carbonate alkalinity and organic acids and tAlk) as a proxy of lake buffering status, which is closely related to watershed characteristics and has been associated with inorganic carbon originating from catchment soils [*Striegl et al.*, 2000; *Humborg et al.*, 2010; *McDonald et al.*, 2013]. Once we established spatially varying responses of pCO_2 to proxies, we related them to broad-scale landscape and climate properties (e.g., lake and landscape morphometry, land cover, land use, and climate). Our approach is based on the assumption that the landscape features (e.g., climate, land use, and elevation) influence the proxies (Color, Chl *a*, tAlk, and nutrients), which themselves affect the different mechanisms of pCO_2 control in lakes [*Lapierre et al.*, 2015; *Sepulveda-Jauregui et al.*, 2015].

2.2. Statistical Analyses

Most studies of aquatic C cycling use multiple linear regression (or analog) analyses, perhaps influenced by the "predictive limnology" tradition that was highly effective for studies of Chl *a* versus TP relationships that have slopes that do not vary substantially geographically [*Peters*, 1986]. However, for carbocentric limnology, these relationships fit poorly and there is evidence that it may be due to spatially varying relationships [*Sobek et al.*, 2005; *Prairie*, 2008; *Lapierre and del Giorgio*, 2012]. To test this idea, we first fit a—spatially fixed—multiple linear regression between pCO_2 and all chemical, climate, land use/land cover, and landscape and lake morphometry variables available in the NLA data (data available at https://www.epa.gov/national-aquatic-resource-surveys/data-national-aquatic-resource-surveys) using forward selection and JMP 10 software (SAS institute, Cary, NC). We then fit geographically weighted regression (GWR) [*Fotheringham et al.*, 2002] using SAM 4.0 [*Rangel et al.*, 2010] to assess the spatially varying response of pCO_2 to a range of potential proxies. Running GWR computationally expensive, hence we used the MLR forward selection to inform on the variables explaining the most variation in lake *p*CO2 to be included in spatially varying models. Data for all analyses were transformed to meet normality and homoscedasticity assumptions prior to analysis. We tested several relevant climate, landscape, and lake properties and selected the most parsimonious model based on r^2, number of predictors, and biogeochemical interpretation (supporting information Table S1). In cases where model fit was similar, we prioritized the model with the lowest number of parameters to facilitate interpretation.

We modified the approach used by [*Seekell et al.*, 2014a, 2014ab] that assessed the spatially varying regulation of DOC in Swedish lakes. Briefly, GWR estimates model parameters for every lake and its closest neighbors through a moving window that includes 10 to 15% of the total number of lakes in the data set (in order to minimize AIC); lakes receive decreasing weight in calibrating the local regressions when they are farther from the center of the moving window. We further ran GWR models on random subsets containing 75% of the full data set to assess the sensitivity of the observed patterns to outliers and irregular spatial coverage. The performance of the spatially varying GWR and spatially fixed MLR models was compared from coefficient of correlation and AIC. Equal performance of both types of models implies that global models adequately capture the spatial variation in lake pCO_2. Alternatively, better performance of GWR models implies poor predictive power of global models and spatially varying drivers-response relationships in lake pCO_2.

We visualized the spatial variation in the relationship between pCO_2 and the proxies by mapping the significant *t* values (slope of the regression divided by standard error of the estimate, $p < 0.05$) from the most parsimonious GWR model using ArcGIS v.10.1. Proxies are lake-level indicators of pCO_2 regulation; if the relationship between pCO_2 and proxies varies spatially, landscape and climate contexts are probably important in determining which factors dominate the regulation. We then used cascade *k*-means cluster analyses (Vegan R package) [*Oksanen et al.*, 2013] on the *t* values obtained from the GWR to identify the optimal number of spatial clusters that describe lake CO_2 driver-response relationships. Briefly, these analyses identify the number of clusters that optimizes ratio of the sum of squares within versus among the clusters. If the optimal number of clusters is 1 or there are multiple noncontiguous clusters, driver-response relationships are either homogenous or vary unpredictably over space. The formation of multiple, mostly contiguous clusters suggests spatial structure in the CO_2 driver-response relationships and indicate regions of similar CO_2 regulation.

Finally, we performed classification and regression tree (CART) analysis (using JMP 10) to predict cluster membership. This process helps elucidate the regional-scale context that leads to geographic differences in within-lake pCO_2 regulation. In other words, we used all the climate, land cover/land use, and landscape and lake morphometry variables available in the NLA data set to predict cluster membership, not individual lake pCO_2 values. We used a semiautomated forward selection, whereby the best predictor variable entered the model first and was not allowed to reenter the model subsequently. Although this approach was less efficient for predicting cluster membership compared to an unconstrained selection, it met our goal of helping to understand the effects of different predictor variables along climate, land cover, and land use gradients.

3. Results

3.1. Spatial Patterns in Lake pCO_2 Driver-Response Relationships

The environmental properties of the lakes varied over 2 orders of magnitude (Table 1). The range (6.4 to 10 553 μatm (2.5–97.5 percentiles)), mean (1466 μatm), and median (588 μatm) of pCO_2 are within the range of reported value in other broad-scale studies [*Cole et al.*, 1994; *Sobek et al.*, 2005; *Lapierre et al.*, 2013] and indicate that most lakes (65%) were supersatured and emitted CO_2 to the atmosphere.

The most parsimonious spatially fixed multiple linear regression (MLR) model that fit all of the lake data poorly explained the variability in lake pCO_2 ($\log pCO_2 = 0.30 \log\text{Color} - 0.56 \log\text{Chl } a - 0.54 \log\text{Secchi}$, $r^2 = 0.10$, $n = 1080$, $p < 0.001$). The most parsimonious spatially varying GWR model, in turn, explained 31% of the variation in pCO_2 across the lakes. This model performed much better than its ordinary least squares analog (185 AIC improvement; see supporting information Table S1), with an almost threefold improvement in variation explained, despite the much higher number of effective parameters [see *Fotheringham et al.*, 2002]. This model included, in decreasing order of contribution, Chl *a*, Color, and tAlk as predictor variables. Concentrations of DOC and nutrients did not meaningfully improve model performance ($r^2 = 0.34$ when those three predictors were added, i.e., a 0.03 improvement). Interestingly, a GWR model with only randomized predictors explained as much variation in lake pCO_2 as the most parsimonious MLR. This result suggests significant but weak spatial structure in pCO_2 that is captured by splitting the study extent into smaller groups of nearby sites and attributing an average pCO_2 value to these lakes. We further ran GWR models on random subsets containing 75% of the complete data set and found no major difference in the overall pattern when compared to the full data set (supporting information Figure S1), suggesting that the patterns are robust. No variable related to climate, net primary productivity in the catchment, land cover, land use, or lake morphometry was significant when Chl *a*, Color, and tAlk were included as predictors in the best GWR model, suggesting that these landscape context variables do not directly control lake surface water pCO_2—rather, pCO_2 at broad scales is driven by proxies representing within-lake processes.

There were distinct spatial patterns in the relationship between lake pCO_2 and its drivers, as shown by the lake-specific *t* values (slope divided by standard deviation; see section 2). Color and tAlk had positive effects on lake pCO_2, when significant (Figures 1a and 1b), while Chl *a* had a negative effect on pCO_2, when significant (Figure 1c). Chl *a* had the most widespread effect, having a significant effect on pCO_2 in 68% of the lakes. Most of the lakes where tAlk had a significant effect on pCO_2 also had a significant effect of Chl *a*, whereas there were several lakes in central southern U.S. and western U.S. where Chl *a* was the only significant predictor of lake pCO_2 (Figure 1).There was almost no spatial overlap in the effect of Color and tAlk on lake pCO_2. The effect of Color was most apparent in midwest U.S. lakes, central U.S., and southeastern U.S. TAlk

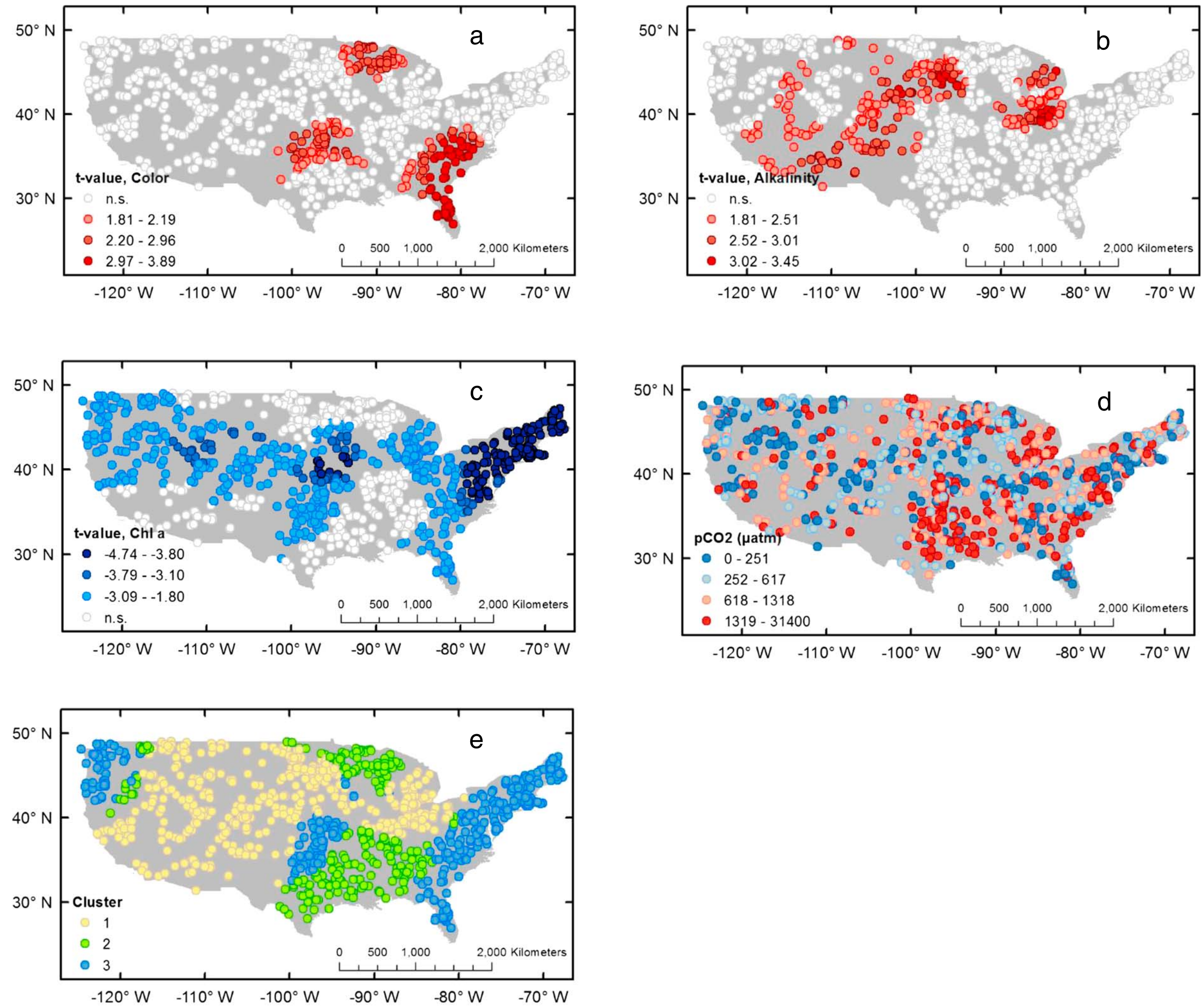

Figure 1. The spatially varying relationships of (a) Color, (b) alkalinity, and (c) Chl *a* with pCO_2 in U.S. lakes. Colored dots on Figures 1a–1c represent significant relationship between the proxy and pCO_2 based on geographically weighted regressions. Colors indicate classes of *t* values (slope of the regression divided by standard error of the estimate), with red denoting a positive effect, blue denoting a negative effect, and white denoting no statistically significant effect on pCO_2. Despite an absence of spatial pattern in (d) pCO_2, the spatial patterns in lake pCO_2 driver-response relationships translated into the formation of (e) spatial clusters of pCO_2 "regulation." Clusters include lakes with comparable response of pCO_2 to Chl *a*, Color, and tAlk (see Table 2). Note that the map displays the boundary of U.S. territories, not just the land area.

was a significant predictor of lake pCO_2 on the southern and eastern edge of the rocky mountains and in the great lakes region.

The spatially varying pCO_2 driver-response relationships (Figure 1) translated into the formation of three spatial clusters (Figure 1e); these clusters represent geographic regions characterized by similar relationships between pCO_2 and Chl *a*, Color, and tAlk. Cluster 1 was the most widespread and included lakes that were predominantly correlated with Chl *a* and tAlk; lakes in this cluster had the lowest median pCO_2, Chl *a*, and Color values but the highest tAlk values (Table 2). Cluster 2 included lakes from central U.S. where pCO_2 was predominantly correlated with Chl *a* and Color but had little effect of tAlk, plus a group of lakes in northwestern U.S. that were mainly correlated with Chl *a*. Lakes in this cluster had the highest median pCO_2 and Chl *a* values and intermediate Color and tAlk values (Table 2). Finally, Cluster 3 included mostly lakes in the eastern U.S., plus a group of lakes in the central and northwestern parts of the country (Figure 1e); this cluster included lakes where the effect of Chl *a* and Color overlapped but where there was rarely a significant effect of tAlk on pCO_2. Lakes in this cluster had intermediate median pCO_2, Chl *a*, and Color values and the lowest tAlk values (Table 2). Together, these results highlight the strong spatial patterns in pCO_2 regulation, as illustrated by regional patterns in CO_2 driver-response relationships (Figure 1e).

Table 2. Mean *t* Value (Slope of the Regression Divided by Standard Error of the Estimate) for Each Proxy, by Cluster[a]

Cluster	*t*-Chl *a*	*t*-Color	*t*-tAlk	pCO_2	Chl *a*	Color	tAlk
1	−2.11	0.23	1.76	508	6.9	10	2789
2	−1.10	1.63	−0.40	730	8.8	12	728
3	−3.52	2.14	−0.44	608	7.1	12	549

[a]n = 525, 246, and 376 for clusters 1, 2 and 3, respectively. Clusters were formed by *k*-means analysis and correspond to groups of lakes with a similar relationship between lake pCO_2 and proxy variables. We also show median values of pCO_2 (μatm), Chl *a* (μgL^{-1}), Color (PCU), and tAlk (μequivalent L^{-1}) in each cluster.

3.2. Climate, Landscape, and Land Use Effects on Lake pCO_2 Driver-Response Relationships

The spatial clusters in the pCO_2 driver-response relationships aligned well with climate and landscape features across the study area (Figure 2). Although the classification and regression tree (CART) did not create homogenous groups of lake cluster membership (i.e., the "leaves" of the classification tree contain lakes from more than one cluster; see bottom row of Figure 2), there was a clear pattern in the proportion of lakes belonging to each cluster along gradients of mean annual precipitation (MAP), lake elevation, and percent agriculture and wetlands in the catchment (Figure 2). No other lake or landscape variables had significant effects on cluster membership once the above predictors were included. Mean annual precipitation was most associated with spatial patterns in the response of pCO_2 to drivers; there were only a few lakes from Cluster 3 found in landscapes with lower than 990 mm y^{-1}, and almost no lakes from Cluster 1 were found in landscapes with higher than 990 mm y^{-1}. While Chl *a* appeared to have an effect on pCO_2 in all areas, its effect was stronger (i.e., higher t-value, meaning steeper slope, lower slope standard deviation, or both; Table 2) in wetter landscapes.

The effect of land cover and land use became apparent once the major effects of precipitation and elevation were taken into account (lower levels of the CART analysis, Figure 2). In particular, there were higher proportions of lakes where pCO_2 was primarily influenced by tAlk and Chl *a* in landscapes with low MAP and high percent agriculture in the catchment ($>$ 15.5) and higher proportions of lakes with a strong effect of Chl *a* and Color on pCO_2 in landscapes with high MAP and percent wetlands $<$0.2 in the catchment (Figure 2 and Table 2). Furthermore, although the mean *t* value of Color (Table 2) is lower for Cluster 3 than for Cluster 2, there was a higher proportion of lakes with a significant effect of Color in Cluster 3 (Figure 1a versus Figure 1e). Most of the latter lakes were found in landscapes with lower percent agriculture and higher percent wetlands in their catchment, representative of the pristine temperate and boreal lakes from which much of the knowledge on aquatic carbon budgets is derived.

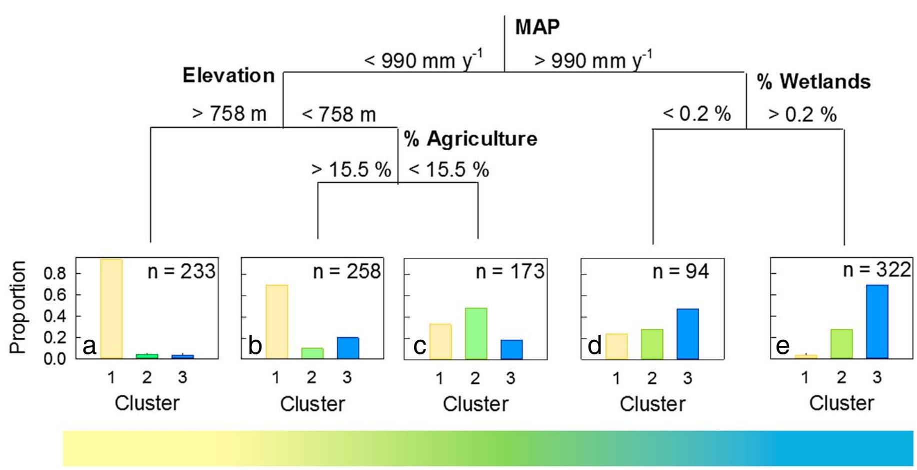

Figure 2. Aligning pCO_2 driver-response relationships with climate and landscape features, using classification and regression tree (CART). The climate and landscape properties did not form homogenous groups in terms of cluster membership (overall r^2 = 0.34) but formed a clear gradient in the proportions of lakes belonging to the different clusters based on the pCO_2 driver-response relationships along gradients of mean annual precipitation (MAP), elevation, and percent agriculture and wetlands in the catchment. The bottom colored line illustrates the shift from the dominance of lakes belonging to cluster 1 to cluster 2 to cluster 3 from the left to the right.

4. Discussion

4.1. Spatially Varying Regulation of Lake *p*CO_2

We identified broad-scale patterns in the response of lake *p*CO_2 to proxies of the production and loss of CO_2 across the contiguous United States. Although intuitive, the unidirectional effect of each of these individual proxies for such a large and heterogeneous study extent contrasts with individual studies that have found that *p*CO_2 may be negatively related to DOC [*Finlay et al*., 2009; *Balmer and Downing*, 2011] or positively related to Chl *a* and nutrients [*Kortelainen et al*., 2006; *Webster et al*., 2008]. The authors of the latter studies did not interpret these patterns as causal; however, it is possible that these variables covaried with *p*CO_2, and *p*CO_2 itself was mainly driven by other local factors. The negative effect of Chl *a* on *p*CO_2 is consistent with its known association with high pelagic and benthic primary production [*Brylinsky and Mann*, 1973; *Morin et al*., 1999] and thus CO_2 uptake that exceeds CO_2 production by respiration of primary producers. The positive relationship between lake *p*CO_2 and tAlk is coherent with previous studies that have identified land-derived inorganic carbon as an important control of lake *p*CO_2 in lakes and rivers [*Striegl et al*., 2000; *Stets et al*., 2009; *Humborg et al*., 2010; *McDonald et al*., 2013]. The positive effect of Color and tAlk in some areas of the U.S. is consistent with their role as indicators of inorganic and organic carbon terrestrial inputs that directly or indirectly (via DOC processing) contribute to CO_2 supersaturation in lakes and rivers [*Humborg et al*., 2010; *Lapierre et al*., 2013; *Sobek et al*., 2003]. The effect of Chl *a*, Color, and tAlk *p*CO_2 is presumably similar in every lake, at least in terms of direction, in this and previous studies, but a key result from this study is that the relative importance of any given pathway appears to vary along geographical and environmental gradients.

Geographic variation in lake *p*CO_2 regulation does not necessarily involve strong spatial patterns in raw *p*CO_2 values. Interestingly, a spatially varying GWR model with a random variable as the sole predictor explained as much variation in lake *p*CO_2 (r^2 = 0.11) as the best spatially fixed multiple linear regression model. The variation in this GWR model is entirely explained by the varying local intercept (slope coefficients are never significant), meaning that there is weak spatial autocorrelation in lake *p*CO_2 that is captured by subsampling the whole data set into groups of nearby lakes and attributing a mean *p*CO_2 (i.e., the intercept) to those lakes. Thus, there is as much unexplained spatial variation in lake *p*CO_2 as is explained by the best MLR model. It should be noted, however, that having four random predictors did not improve explained variation as compared to including just one predictor (supporting information Table S1), suggesting that most of the variation in the best GWR model (Figure 1) is explained by the indirect effects of the proxy variables included. Likewise, there is nearly no variance explained in four random response variables by the four predictors included in the best GWR model (r^2 ranging between 0.02 and 0.03, supporting information Table S1), showing that there is no inherent model structure responsible for the spatial patterns shown here in the regulation of lake *p*CO_2. There are thus significant but weak spatial patterns in lake *p*CO_2 across the study extent, and the presence of spatial patterns in the *p*CO_2 driver-response relationships emphasizes the need to consider spatial heterogeneity in lake *p*CO_2 regulation at broad spatial extents.

We identified spatial patterns in lake *p*CO_2 driver-response relationships across the conterminous U.S. through the study of well-known proxies related to the production or losses of CO_2 rather than the processes themselves [*Hanson et al*., 2003; *Humborg et al*., 2010; *Lapierre et al*., 2013; *McDonald et al*., 2013]. Although the causality of proxy-based relationships is less explicit than process-based relationships, they allow us to explore how local processes are reflected in broad-scale patterns of lake C cycling when detailed process measurements are not available, as is frequently the case in broad-scale studies. This approach allows for generalizations, whereas detailed local studies untangle complex mechanisms. The sampling scheme—which was limited to the summer season—may not allow us to draw conclusions on annual CO_2 dynamics for the sampled lakes. It may further have overemphasized the importance of Chl *a* as a widespread driver of lake *p*CO_2, but the effect of Chl *a* on *p*CO_2 was strong even in lakes from Cluster 1 where median Chl *a* concentrations were the lowest, suggesting that this may not be a summer-specific result. Nonetheless, limiting the temporal extent presumably reduced confounding temporal effects and allowed us to identify spatial patterns.

No climate, land cover/land use, landscape, or lake morphometry variable was a significant predictor of lake *p*CO_2 in either spatially variable fixed models when concentrations of Chl *a* or organic and inorganic carbon were included. These results are consistent with previous northern regional studies that have found no direct relationships between geographic variables and lake pCO$_2$ [*Lapierre and del Giorgio*, 2012; *Sepulveda-Jauregui et al.*, 2015], and they support the idea that the effect of static geographic properties on lake pCO$_2$ is transmitted to the freshwater carbon cycle indirectly through proxies that respond to climate and landscape signals that are correlated with local processes [*Lapierre et al.*, 2015]. Therefore, it appears that while proxies explain lake-to-lake variation in pCO$_2$ along local regression lines, the regional landscape and climate contexts explain the difference in the slope of those lines between regions.

4.2. Regional Climate and Landscape Constraints on the Regulation of Lake pCO$_2$

We found contrasting spatial patterns in the driver-response relationships of lake pCO$_2$ that aligned well with landscape and climate properties (Figure 2). In particular, the CART analysis provided a formal, quantitative framework to identify the main context variables responsible for grouping lakes with similar pCO$_2$ regulation. While pCO$_2$ was related to Chl *a* across the different clusters, the effect of other proxies was differentially expressed across the study extent. For example, mean annual precipitation (MAP) was the main context variable that clustered lakes with similar pCO$_2$ driver-response relationships; the effect of tAlk was the strongest in lakes situated in regions with MAP $<$990 mm y^{-1}, whereas regions with higher MAP tended to include lakes with pCO$_2$ that were more strongly correlated with Color. These results suggest that for lakes with stronger surface hydrological connections with their catchments, there is a similarly stronger role of internal processes, such as CO_2 production from DOC processing and CO_2 uptake from primary production. This result presumably relates to growing season length and watershed characteristics like slope and terrestrial net primary production. Moreover, given similar geology, higher precipitation could involve dilution and reaction rate limitation of chemical weathering, leading to relatively lower loadings of alkalinity. Therefore, the role of loadings of terrestrial organic carbon and nutrients in driving aquatic processes involved in the production or consumption of CO_2 appears relatively more important in wetter catchments [*Rantakari and Kortelainen*, 2005; *Butman and Raymond*, 2011].

Lakes situated in less humid landscapes were mainly related to concentrations of Chl *a* and tAlk (i.e., they were mainly from Cluster 1, see Figure 2, Table 2). Lakes from Cluster 1 had the lowest pCO$_2$ and Chl *a* values, but several-fold higher median tAlk values. These lakes were mainly found in either higher elevation landscapes (Figure 2a), or in lower elevation but highly agricultural landscapes (Figure 2b). Although we could not directly explore the role of surficial geology in our analyses, this result may be explained by higher elevation landscapes having less organic matter in soils that contribute to lake DOC and nutrients, such that for these lakes, CO_2 dynamics are dominated by terrestrial inputs of CO_2 [*Crawford et al.*, 2015] rather than in-lake biogeochemical processes. Likewise, lower elevation lakes in Cluster 1 tended to be in highly agricultural regions. The low pCO$_2$ values for those lakes are coherent with their negative correlation with Chl *a*, and might indicate external nutrient-driven uptake of CO_2 from primary production. The positive relationship with tAlk could be explained by farmers selecting well-buffered soils and/or by agricultural activity enhancing alkalinity and CO_2 export to rivers and lakes in these regions [*Raymond and Cole*, 2003; *Butman and Raymond*, 2011]. Furthermore, glaciation history or lake origin can affect lake pCO$_2$, but independent effects of such factors are hard to disentangle due to spatial overlap in their distribution. There is indeed an apparent shift in pCO$_2$ regulation across the glaciation line, where lakes shift from mostly natural to mostly reservoirs, but the spatial clusters in pCO$_2$ driver-response relationships (Figure 1e) do not match the distribution of lake origin [natural versus artificial impoundments, see *McDonald et al*. [2013]], and lake origin was not a significant factor in the CART model. This result does not mean that lakes and reservoirs behave the same way in terms of pCO$_2$ regulation, but this result suggests that human alteration of lake hydrology has relatively little effect on the broad-scale regulation of lake pCO$_2$ compared to other climate and landscape predictors, where other pathways are predominantly expressed.

Past studies have identified the overwhelming importance of terrestrial DOC in regulating lake pCO$_2$, but this evidence mostly originates from boreal and temperate zones with little human disturbance [*Sobek et al.*, 2005; *Cole et al.*, 2007; *Lapierre and del Giorgio*, 2012]. In the present study, we found a significant effect of Color on pCO$_2$ in Northern temperate lakes (Figure 1), in particular for those with high wetlands and low percent agriculture in their catchment (Figure 2). These lakes represent only 20% of the lakes analysed, however, suggesting that in a large proportion of the U.S. landscape, lake pCO$_2$ is predominantly regulated through pathways that do not involve processing of terrestrial organic carbon within lakes. High concentrations of colored DOC may limit primary production in boreal lakes, especially in the benthic environment, further decreasing the potential role of CO_2 update from algae as a main driver of *p*CO2 in northern landscapes [*Seekell et al*., 2015a, 2015b]. Therefore, our results suggest that the response of lake pCO_2 to proxies of the gains and losses of CO_2 in a large proportion of North America differs from what has been reported in other Northern landscapes, especially in low mean annual precipitation and human-impacted landscapes.

4.3. Regional-Scale Controls of pCO_2 in U.S. Lakes

The geographically varying driver-response relationships of lake pCO_2 point to the importance of regional-scale variation in pCO_2 regulation. Most limnological studies focus on explaining between-lake variation at either the within-region or global scales. However, our analyses show that there is important meso-scale variability, that this variability is manifested in heterogeneous scaling relationships, and that this variability is predictable based on climate and landscape characteristics. Improved understanding and prediction of the response of the aquatic carbon cycle to global change will benefit from considering this intermediate scale between the global models of concentrations and fluxes of CO_2 and the local, process-based patterns. Our results highlight how regionalization frameworks derived from lake driver-response relationships may adequately capture the spatial patterns in lake functioning at scales relevant for understanding the broad-scale patterns in lake carbon cycling.

Our findings challenge the applicability of global models for the prediction and understanding of lake CO_2 dynamics because the main pathways involved in the regulation of lake pCO_2 vary geographically across broad geographical, climate, and landscape gradients (Figures 1 and 2). For example, our results could suggest that increased mean annual precipitation may increase the relative importance of biological pathways regulating lake pCO_2 or that increasing agricultural and wetland coverage may increase loadings of terrestrial CO_2, in situ production of CO_2 from organic carbon processing, and an increase of CO_2 uptake from primary production [see *Heathcote et al*., 2015]. However, a key finding from our study is that lake pCO_2 response to changing climate and land use will not be uniform but will vary spatially depending on regional climate and landscape contexts; lakes lying in landscapes that are the closest to the climate, topography, and land use thresholds identified here (Figure 2) would likely be most susceptible to changes in lake pCO_2 regulation. Although the net effect of multiple environmental changes on lake pCO_2 remains unclear, our results show that climate and landscape contexts modulate the response of pCO_2 to different proxies. This, in turn, suggests that changing climate and land use have the potential to alter the dominant pathways that regulate lake pCO_2 across broad and heterogeneous landscapes, with implications for the role that lakes play as sources and sinks of atmospheric CO_2 for the global carbon budget.

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