Applied Geochemistry 96 (2018) 244–251

Contents lists available at ScienceDirect

Applied Geochemistry

journal homepage: www.elsevier.com/locate/apgeochem

Long-term metal and arsenic mobility between wetlands and lakes: Variable histories within the same floodplain

Cristiane Q. Surbeck[a,*], Gregg R. Davidson[b], Daniel G. Wren[c]

[a] Department of Civil Engineering, University of Mississippi, University, MS, 38677-1848, USA
[b] Department of Geology and Geological Engineering, University of Mississippi, University, MS, 38677-1848, USA
[c] USDA ARS National Sedimentation Laboratory, Oxford, MS, 38655, USA

ARTICLE INFO

Editorial handling by Prof. M. Kersten

Keywords:
Wetlands
Trace metals
Arsenic
Remobilization
Contaminants

ABSTRACT

Short-term studies of wetlands show that wetlands scavenge pollutants, thereby mitigating the pollution of downstream water bodies. However, there are few long-term studies that demonstrate this effect. This study provides a wealth of data demonstrating that there is variable long-term mobility of metals and arsenic in wetland-lake systems. More than 900 samples were analyzed from 14 soil and sediment cores collected from six wetland-lake systems in the same floodplain. Cores were analyzed for a suite of trace elements (As, Co, Cu, Ni, Pb, Zn are reported in this paper), organic matter content, clay content, and water content. Sediment age was estimated using ^{210}Pb and ^{137}Cs data, facilitating comparison of element concentrations in contemporaneous wetland and lake sediments. In all six systems, sediment-laden water passes through the wetland before settling into the lake. In three of the systems, element concentrations were consistently higher in lake sediments, indicating a history of remobilization of wetland elements and subsequent deposition in lake sediments. Results in the other three lakes were variable, documenting unique long-term histories of element mobility and sequestration in otherwise similar systems.

1. Introduction

Many short-term studies have documented the ability of riparian wetlands to scavenge sediments and contaminants (e.g. Kitchens et al., 1975; Cooper et al., 1987; Knight et al., 1987; Hupp et al., 1993; Mitsch and Gosselink, 2000; Ye et al., 2003; Birch et al., 2004). These and related investigations typically measure the concentration or mass of contaminants entering and exiting a riparian system to document the potential for a riparian zone to act as a chemical buffer between non-point sources of pollution and downstream environments. By their nature, these studies are limited to observations of short-term reductions in contaminant transport. Little is known about the long-term (multi-decadal) fate of nonlabile contaminants in these systems. This paper addresses the long-term deposition of contaminants in wetland-lake systems in the Delta region of Mississippi, a region heavily affected by toxic metals compared to neighboring regions (Dockery and Thompson, 2016), a result of agricultural pesticide application.

A few studies have suggested that scavenging of contaminants by a riparian wetland can be ephemeral. DeLaune et al. (1981) reported higher P concentrations in the sediments of a shallow inland lake in southern Louisiana relative to sediments from an adjoining upstream marsh. The difference was attributed to the uptake of P by wetland plants that later died and decomposed. Subsequent outwash of decomposing organic matter transported the P-laden organic matter to the vegetation-free lake, where it was buried and sequestered. Weis and Weis (2004) also reported that wetlands can be sources or sinks, depending on whether metals accumulate in leaves or roots of plants, respectively. Additional variables or processes that influence the fate and transport of trace elements in these environments include redox potential, pH, temperature, mineral dissolution/precipitation, adsorption, complexation, and the nature and concentration of dissolved organic carbon (Schiff et al., 1990, Nepf and Oldham, 1997, Rupp et al., 2010, Shaheen et al., 2016).

Results similar to DeLaune et al. (1981) were documented in Sky Lake, Mississippi, by Galicki et al. (2008), who measured P, Pb, and As preserved in lake and wetland cores. The mass of P found in lake sediments was often many times larger than the mass in contemporaneous wetland soils (hereafter, “sediment” and “soil” will be referred to collectively as “sediment”). Pb and As were also much higher in lake sediment cores relative to upstream wetland sediments deposited at the same time. Sky Lake is fully surrounded by wetlands, and stream discharge rapidly disperses through the wetland, eliminating bypass as an

* Corresponding author.
E-mail address: csurbeck@olemiss.edu (C.Q. Surbeck).

https://doi.org/10.1016/j.apgeochem.2018.07.002
Received 2 March 2018; Received in revised form 30 June 2018; Accepted 4 July 2018
Available online 05 July 2018
0883-2927/ © 2018 Published by Elsevier Ltd.

explanation for the absence of elevated Pb or As in the wetland sediments. Metals and As are often preferentially adsorbed to clay or organic matter, resulting in higher concentrations with higher clay or organic matter content (hereafter referred to as “organic content”), but the lake sediments did not contain elevated clay or organic matter relative to the wetland sediments.

Galicki et al. (2008) offered a hypothesis similar to the process described by DeLaune et al. (1981), where elements in the wetland are mobilized into the lake via decomposing litter. It was further suggested that following contaminant adsorption onto wetland organic litter, periodic aeration of the wetland enhances decomposition of organic material with subsequent remobilization of associated contaminants (DeLaune and Smith, 1985). As water levels rise and fall, some of the decomposition products are carried out to downstream water bodies. Repeated cycles of inundation and aeration eventually flush out most of what was initially scavenged. In lake sediments, redox conditions can be perpetually reducing (Howeler, 1972), where deposition may result in permanent sequestration and preservation of the elevated chemical influx.

The motivation for the current study was to determine if the apparent remobilization of trace metals and arsenic observed in Sky Lake was normative in riparian wetlands. Six oxbow wetland-lake systems (including Sky Lake) were selected from the same geographic region on the floodplain of the Lower Mississippi River, and remobilization was assessed by examining the long-term element concentrations in lake and wetland sediment cores. The results show a complex and variable history of trace metal and arsenic transport in otherwise similar systems.

2. Materials and methods

Over 900 sediment samples were collected from six wetland-lake systems: Beasley, Hampton, Moon, Roundaway, Sky, and Washington (Fig. 1). All lakes are oxbow cutoff lakes located in the intensively cultivated agricultural region of northwest Mississippi, and all lie on the ancestral floodplain of the Mississippi River. The lakes are variably surrounded by forested wetlands, with bald cypress as the dominant canopy species. Where Fig. 2 shows a thin wetland border, trees are typically only growing along the slope leading into the lake and in the shallow water at the lake edge.

2.1. Sampling, element analysis, and sediment dating

Sediment cores that were 1–3 m in length were collected, and most

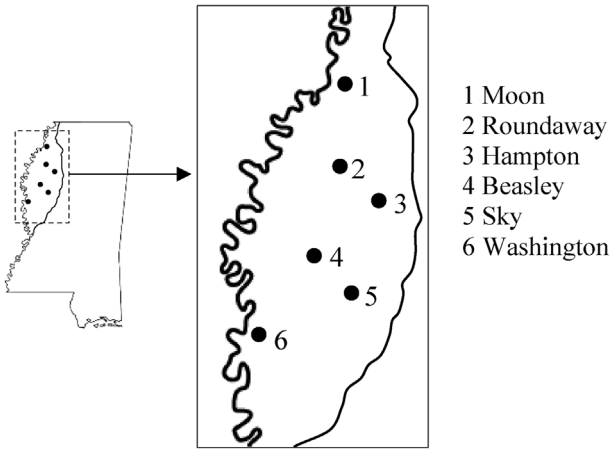

Fig. 1. Wetland-lake system locations. The state of Mississippi (USA) is shown with the agricultural Delta Region outlined and enlarged. The Delta Region is the portion of the Mississippi River floodplain inside the state boundary. The Mississippi River runs along the west side of the state.

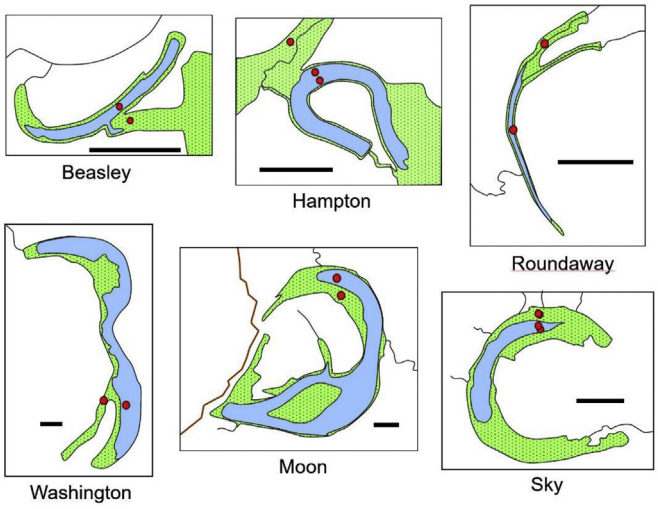

Fig. 2. Locations of core samples in each wetland-lake system. The extent of forested wetlands is represented by a green/stippled pattern. Surrounding land is agricultural at all six lakes. Horizontal bars denote a 1 km scale. The brown solid line west of Moon Lake is the Mississippi River levee. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

cores were sectioned and analyzed for a suite of trace elements in 1 cm increments. The core sampling locations are shown in Fig. 2, where the wetland sample locations were consistently upstream of the lake samples. To assess the reproducibility of measured element concentrations, two sets of duplicate cores were collected and analyzed from Sky and Hampton lakes.

Arsenic in sediment was analyzed using Induced Neutron Activation (INA). The remaining elements were analyzed using Inductively Coupled Plasma Mass Spectrometry (ICP-MS) following total digestion.

Samples were collected and analyzed for ^{210}Pb and ^{137}Cs to estimate sedimentation rates and establish approximate age of deposition. Concentrations of trace metals were then compared between wetland and lake sediments with the same approximate age of deposition. The Supplementary Data file shows the element concentrations, depths, and deposition dates from 14 cores. Details of the sediment dating process are reported in Davidson et al. (2004), Walker et al. (2007), Wren et al. (2008), and Wren et al. (2016). Core sample thickness and depth were normalized to mean water content in order to account for compaction in lower levels and high water content in upper levels of the cores (Martin and Rice, 1981). Recently deposited sediments with high water content are often compressed because additional sediment accumulation displaces water from pores. The thickness of older strata may be lower than when the sediments were originally deposited. Normalizing by water content accounts for this difference and is important for valid comparisons between recent and older sedimentation rates. To determine if elevated concentrations of trace elements were due to higher clay or organic content, selected depths were analyzed for particle size and clay content by a combination of sieve, hydrometer, and laser particle size analysis, and organic content by loss on ignition (data included in the Supplementary Data file). Data for Sky Lake reported here were previously published in Galicki (2002) and Galicki et al. (2008). All other data are reported in this paper for the first time.

2.2. Statistical analysis

Statistical analyses were conducted to check normality of the data distribution and to compare means to determine if there were significant differences between wetland and lake sediment characteristics. All statistical analyses were conducted using SPSS Statistics V25 software (IBM Corporation, Armonk, NY, USA). To determine normality,

element concentrations, separated by wetland and lake for the range of years when both were available, were evaluated in histograms, tests for kurtosis and skewness, and the Kolmogorov-Smirnov and Shapiro-Wilk tests. These tests were accomplished using the Explore function in SPSS, where a significance of $p \leq 0.05$ indicated the data were not normal and therefore non-parametric. To compare whether concentrations from the wetlands and the lakes were statistically different, comparison tests were conducted for independent variables. When the data were normally distributed, a *t*-test was performed. For non-normally distributed data, the non-parametric Mann-Whitney U and Kolmogorov-Smirnov tests were performed. For all three comparison tests, a significance of $p \leq 0.05$ indicated that the means of the wetland and lake core samples were statistically different. Where duplicate cores were collected, the same statistical analyses were performed. In this case, the desired result was $p > 0.05$, indicating no statistically significant difference between duplicate cores, and therefore, reproducible results. All statistical tests followed the procedures described in Field (2005).

3. Results and discussion

The data from wetland and lake sediment cores, combined with time of deposition, show high variability in element concentrations. Results from several wetland-lake systems support the hypothesis that elements are remobilized from wetlands to downstream lakes, rather than being permanently sequestered in wetlands. Others suggest a more variable or complex history.

3.1. Reproducibility

To test the reproducibility of the data, the duplicate cores from Hampton and Sky Lakes were analyzed for selected metals along with their sediment deposition dates. Elevated concentrations at specific deposition times are not random, as shown in Fig. 3. Spikes in Co, Ni, and Pb occur at the same zones in the two duplicate lake cores. Arsenic was also analyzed in the duplicate core for Sky Lake, though there are no peaks in the concentrations. In the case of Co, Ni, and Pb shown in Fig. 3, the Independent Mann-Whitney and Independent Kolmogorov-Smirnov test results show that the duplicate data from the two lakes are not different ($p > 0.054$). In Sky Lake, sediment As concentrations between the duplicate cores were significantly different ($p < 0.01$), suggesting high sampling variability. Despite this variability, the range of As concentrations of 12–32 mg/kg in the duplicate lake cores is consistently higher than the range of concentrations of 4–9 mg/kg in the wetland core. This shows that even though the duplicate As concentrations were not statistically similar, both lake cores have higher concentrations than the wetland core. This reproducibility study demonstrates that a single core from each site should be sufficient to characterize the general trace-element profile with depth and time in the surrounding lake or wetland.

3.2. General trends in concentrations

General trends are reported in Table 1 and Fig. 4, where element concentrations since approximately 1890 from all six wetland-lake systems are shown. In Table 1, p values of ≤0.05 (in parentheses) indicate statistically significant differences between wetland and adjacent lake samples. Asterisks mark cases where $p > 0.05$, but differences are nonetheless observed in graphs of concentration vs time of deposition.

Some elements are consistently found at higher concentrations in lake than in wetland sediments. This trend is especially true for As and Co in four of the systems, with Roundaway and Moon being the exceptions (Table 1, Fig. 4). For the Beasley, Hampton, and Sky wetland-lake systems, element concentrations were generally higher in lake sediments, and peaks in element concentrations in lake sediments are often missing in contemporaneous wetland sediments. This history indicates that if contaminants are being initially scavenged by wetlands, they are being remobilized and released to downstream lakes over the long-term.

If comparisons were made based on mass loading of elements rather than the concentration, the observed differences would be the same or magnified, because the rate of sedimentation accumulation in the lake environments is generally equal to or higher than the rate in the wetlands. (A higher rate of sedimentation means a thicker sediment layer, and a higher concentration in a thicker sediment layer means an even higher total element mass.)

The Lake Washington system was unique in that element concentrations in the lake were higher than in the wetland until sometime between 1967 and 1977. After that period, concentrations in the lake

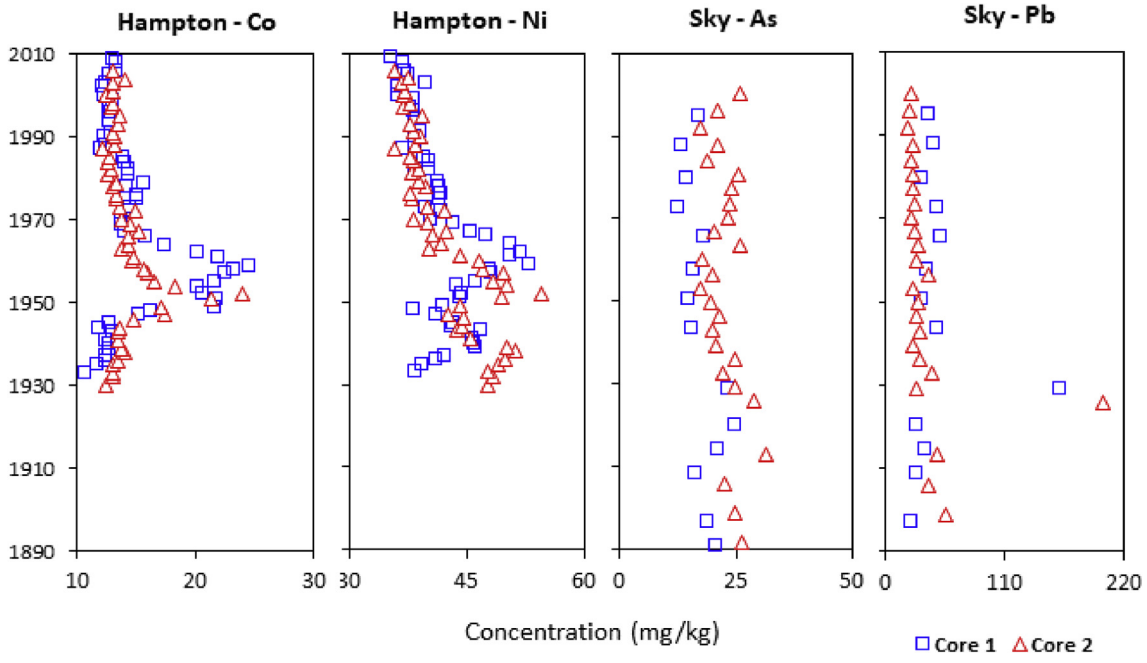

Fig. 3. Trace element concentrations in duplicate cores from the open-water environments of Hampton and Sky Lakes.

Table 1
Element concentrations in lake versus wetland sediment profile, 1890 to 2010. p-values of ≤0.05 indicate statistically significant differences between wetland and lake element concentrations. Shaded cells denote that the element concentrations in lakes are significantly higher than in wetlands.

	Beasley Lake	Hampton Lake	Moon Lake	Roundaway Lake	Sky Lake	Lake Washington
Arsenic	Higher in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Lower in lake before 1970, then higher ($p = 0.18$*)	Not significantly different ($p = 0.70$)	Higher in lake ($p < 0.01$)	Higher in lake, but peak high for wetland in 1971 ($p < 0.01$)
Cobalt	Higher in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Higher in lake ($p = 0.03$)	Lower in lake ($p < 0.01$)	Higher in lake ($p< 0.01$)	Higher in lake before 1967, then similar ($p < 0.01$)
Copper	Higher in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Lower in lake ($p < 0.01$)	Lower in lake before 1963, then similar ($p = 0.01$)	Higher in lake before 1977, then lower ($p = 0.10$*)
Lead	Not significantly different ($p = 0.38$)	Lower in lake before 1975, then similar ($p = 0.01$)	Lower in lake ($p < 0.01$)	Lower in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Higher in lake before 1971, then lower ($p = 0.32$*)
Nickel	Higher in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Not significantly different ($p = 0.50$)	Not significantly different ($p = 0.08$)	Higher in lake ($p < 0.01$)	Higher in lake before 1967, then similar ($p < 0.01$)
Zinc	Not significantly different ($p = 0.87$)	Higher in lake ($p < 0.01$)	Lower in lake ($p =< 0.01$)	Lower in lake ($p < 0.01$)	Higher in lake ($p < 0.01$)	Higher in lake before 1968, then similar, then lower after 1990 ($p = 0.75$*)

* Clear differences observed between wetland and open water element concentration in spite of $p > 0.05$

were either similar or lower than in the wetland. This behavior may coincide with reduced application of agricultural chemicals in the surrounding drainage area, though the higher concentrations in the lake existed well before agrichemicals were in widespread use. The changes, especially the reduction in metal concentrations, could also be related to changes in waste input from local industries. The results from the Washington system emphasize that variability in the history of delivery, mobilization, and retention of trace elements exists, not only between wetland-lake systems, but potentially within the same system at different times.

Fig. 5 shows the concentrations of four metals in Hampton Lake, which allows for detailed exploration of trends for one lake. In lake sediments, Co, Cu, and Ni have significant peaks at the same deposition time, suggesting that all may have a similar source. It may be that each occurred as impurities in agricultural chemicals applied around this lake before 1960. Another common concentration peak in the lake occurs around 1940, for Cu, Ni, and Zn.

Wetland bypass would not be a reason for the elevated Hampton Lake concentrations. Elements had to pass through the wetland to reach the lake, but the concentrations in the wetland are lower and lack the peaks observed in the lake. In this wetland, a low sedimentation rate caused each sample increment to represent a long period of time, resulting in muted peaks; therefore, short-term concentration spikes are not readily apparent. Elements such as Co, however, were significantly elevated for more than a decade in lake sediments (1950s). If the wetland sediments served as a sink for contaminants, a contemporaneous increase in the wetland sediments should be evident, even with multi-year sampling.

At the Sky Lake site, samples from approximately 1900 to 2000 indicate the As, Co, Pb, Ni, and Zn concentrations were always higher in the lake than in the upstream wetland (Fig. 4). Between 1920 and 1929, concentrations of As, Co, and Pb spiked in the lake but not in the wetland (Fig. 6). Similar to Hampton Lake, water must pass through the wetland before reaching the lake, so wetland bypass is not a reason for the peaks in the lake core concentrations.

3.3. Organic matter and clay

Organic content and clay fraction were measured for five of the lakes in core increments with high and low metal concentration zones in order to investigate possible relationships with element concentrations. Organic content measurements were also made for Sky Lake during earlier work published in Galicki et al. (2008). Fig. 7 shows that elevated metal concentrations in these systems are not due to elevated fractions of organic material or clay. For example, in the Beasley system, core samples with high and low Cu concentrations do not have concomitant changes in organic matter or clay content. A similar situation is found in the Hampton system for Ni. In the Sky system,

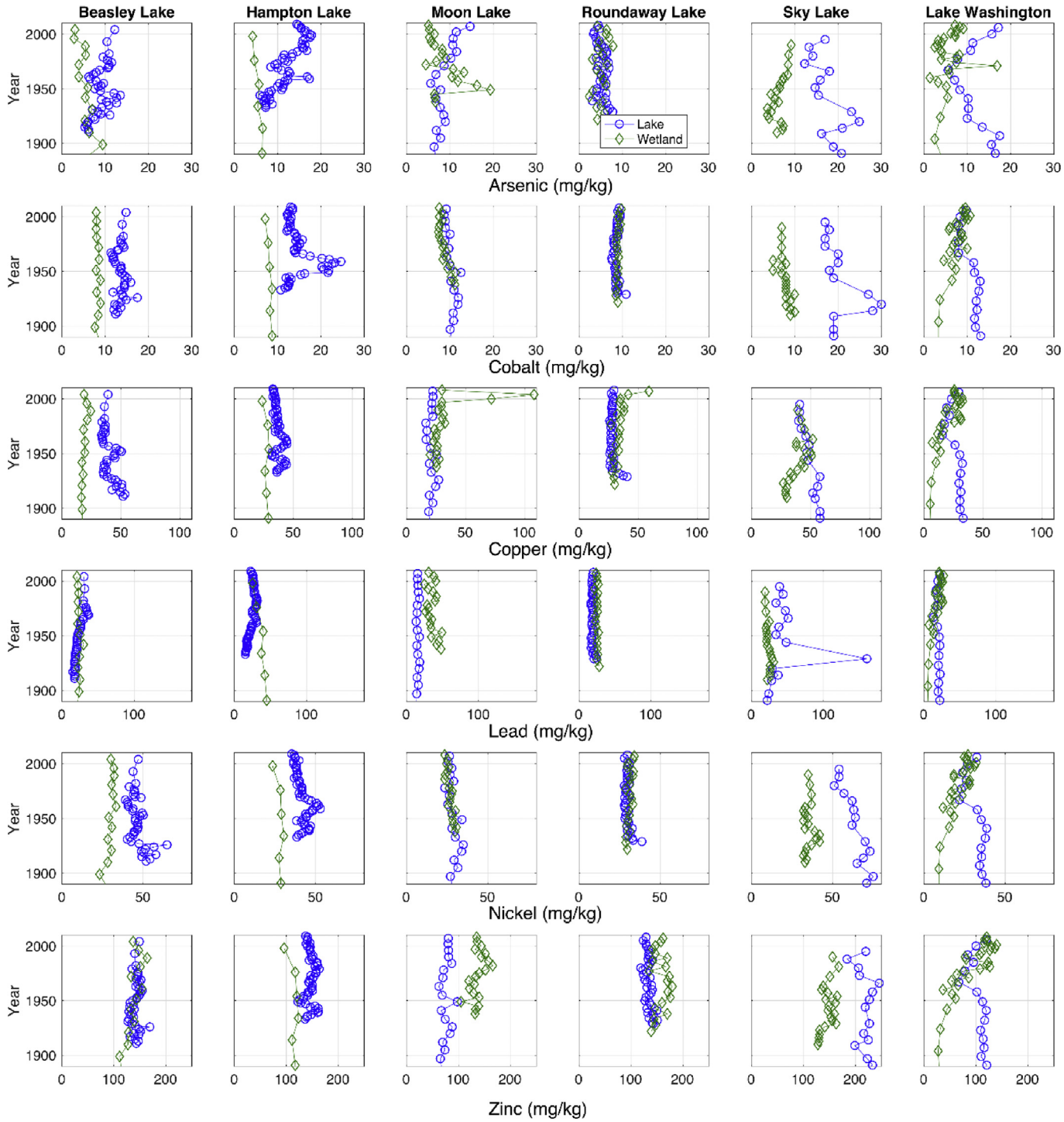

Fig. 4. Element concentrations in contemporaneous sediments in six different wetland-lake systems. Lower sampling density for wetland cores was due to lower sediment accumulation rates (greater number of years represented in each sample). Detection limits in mg/kg are 0.1 for As, 0.1 for Co, 0.01 for Cu, 0.01 for Pb, 0.1 for Ni, and 0.1 for Zn. Analytical uncertainty (1σ) is approximately ± 10% for all.

although Pb is consistently higher in the lake core and has a peak in 1929, organic content concentrations are similar in the lake and wetland cores. Clay content is higher in the wetland than in the lake around 1930, despite the Pb peak in the lake, showing a lack of association between elevated Pb concentration and clay content. In the Washington wetland sediment, varying clay content is also not associated with varying Co concentrations.

3.4. Further interpretation of patterns in element mobility

A counter-argument to our explanation that wetlands are releasing contaminants into lakes, would be that sediment and elements in runoff may have bypassed the wetlands during high flow events, depositing directly into the lakes. However, Hampton and Sky lakes are entirely surrounded by wetlands (Fig. 2), with runoff fanning out and slowing upon entry into the wetlands. High-flow events are not channelized through the wetlands. Further, at Sky Lake, the wetland core was

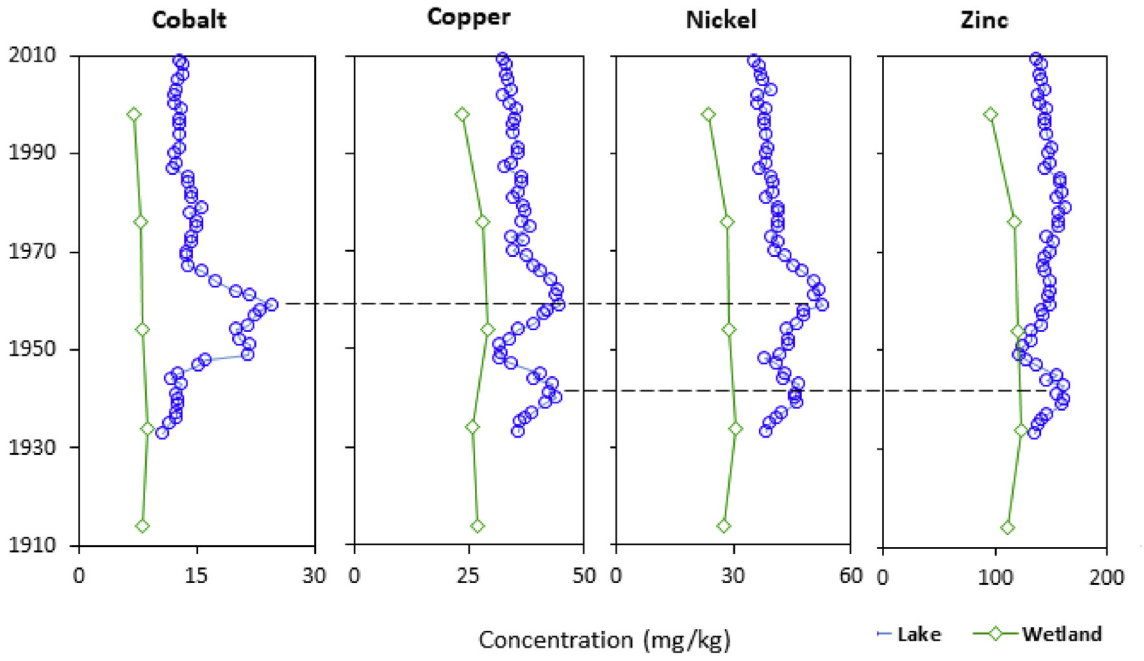

Fig. 5. Concentrations of Cobalt (Co), Copper (Cu), Nickel (Ni), and Zinc (Zn) in sediment cores of the Hampton wetland-lake system.

collected immediately adjacent to where a stream discharges into the wetland, so it was not possible for elements to bypass the wetland without significant exposure to wetland litter. At all other wetland-lake systems, the sampling locations were selected so that the lake cores were similarly downstream of the wetland; therefore, wetland bypass is not an adequate explanation for elevated element concentrations in the lake sediments.

Bypass is more likely at systems such as Washington if substantial inflows enter from the eastern edge where it lacks a perimeter wetland. At Roundaway, where direct runoff from fields can reach the lake with minimal residence time in a wetland, wetland and lake concentration chemical profiles are essentially the same. The circumstances at Washington are more complex, where element concentrations were consistently higher in the lake until the 1960s or 1970s, followed by a dramatic change to essentially equal concentrations for several elements (Co, Cu, Pb, Ni, Zn) in the lake and wetland. The Washington system emphasizes that the history of trace elment delivery, mobilization, and sequestration not only varies between otherwise similar lake-wetland systems but can even vary significantly over time in the same system. These results agree with the conclusions of Kadlec and Wallace (2008) in reviewing studies of treatment wetlands, where it was found that results could not always be extrapolated from one wetland to another, even if wetlands appear to be similar.

4. Conclusions

Based on over 900 sediment core samples for six different wetland-lake systems in the same floodplain, it can be concluded that each

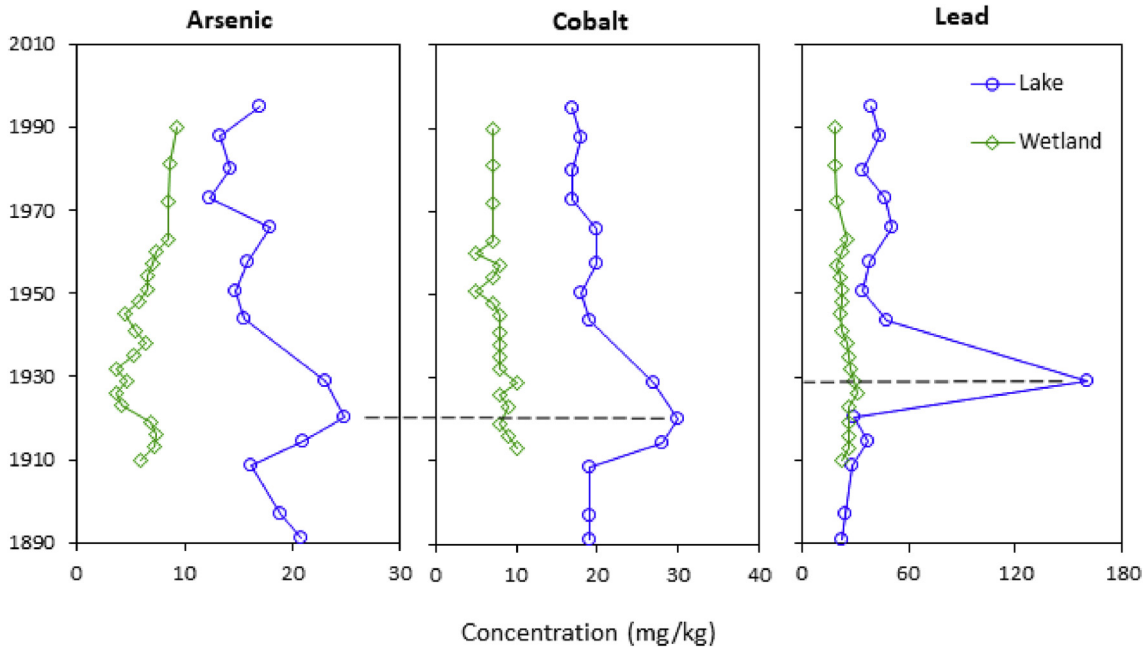

Fig. 6. Concentrations of Arsenic (As), Cobalt (Co), and Lead (Pb) in sediment cores of the Sky wetland-lake system.

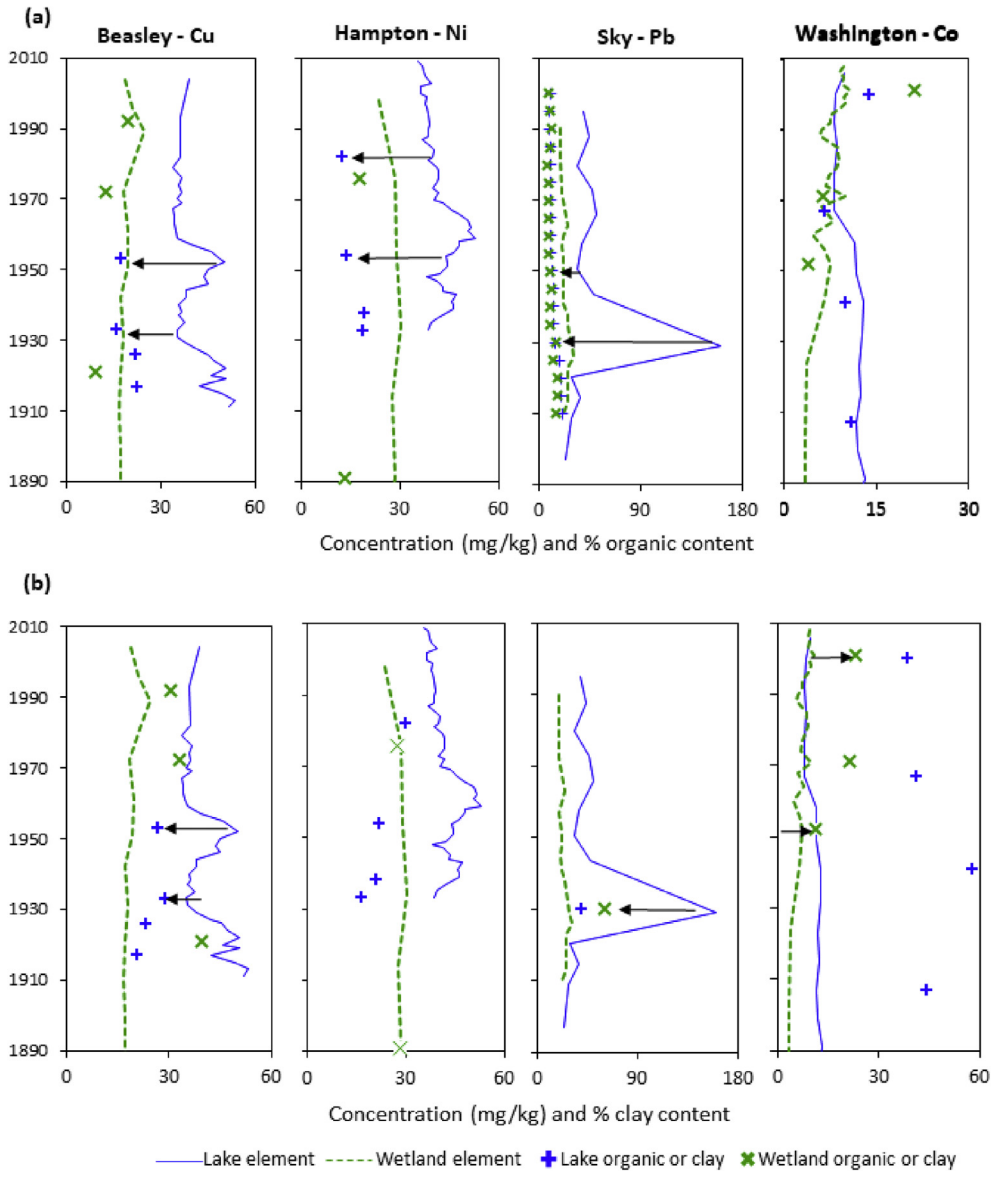

Fig. 7. (a) Organic content and (b) clay content measured at depths with high and low metal concentrations. Arrows draw attention to high or low element concentrations in lake sediments that are not uniquely associated with high or low organic or clay content.

system has a unique contaminant history in terms of timing, variety of flux, mobilization, and retention. Therefore, the long-term fate and transport of metals can be variable even in seemingly equivalent systems. Element concentrations in deposits in lakes are often higher than in the adjoining upstream wetlands, leading to the conclusion that, for these systems, riparian wetlands do not permanently sequester chemically persistent contaminants. Flushing of scavenged trace elements from these wetlands into the lakes appears likely. The results indicate that these wetlands should be considered as potential long-term *sources* of trace elements.

While this study shows the variable history of contaminants in wetland-lake systems, additional studies are needed to further elucidate the reasons for the different contaminant behaviors. Further laboratory simulations can allow the elimination of various complicating variables that cloud the interpretation of the data presented here. Such studies could incorporate the collection of data on microbial communities and their effect on the forms of contaminants, vegetation (in particular, whether metals accumulate in roots or leaves), types of sediments, biogeochemical cycles, and cycles of wetting and drying.

Acknowledgements

This study was partially funded by the Mississippi Water Resources Research Institute through a grant from the USGS. Appreciation is expressed to land owners for allowing access to field sites. Assistance with sample collection and processing was conducted by Steven Utroska, William Walker, and Glenn Gray. Dr. Matt Moore of the USDA-ARS National Sedimentation Laboratory provided valuable insights on agricultural pesticide use and effects on contaminant concentrations. Two anonymous reviewers provided feedback that led to the improvement of the manuscript.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.apgeochem.2018.07.002.

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