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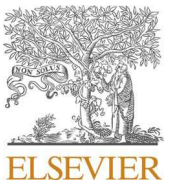

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Multi-decadal impacts of effluent loading on phosphorus sorption capacity in a restored wetland

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ABSTRACT

Natural wetlands are widely used and cost-effective systems for the passive remediation of phosphorus (P)-rich surface waters from various effluent sources. Yet the long-term biogeochemical impacts of effluent loading on wetland P retention capacity are unclear. Here, we had a unique opportunity to document the spatio-temporal evolution of sediment P sorption over a ~25-year period of constant municipal and industrial effluent loading, as part of a wetland restoration and wastewater treatment strategy in one of the largest restored wetlands in Canada. Sediment P sorption experiments across Frank Lake's three basins revealed a wide spatial variation in sorption capacity, closely linked to sediment geochemistry gradients (Ca, Fe, and Mn). Relative to a similar study ~25 years prior, P sorption capacity has become exhausted near the effluent inlet, but remarkably, remains elevated throughout the rest of the wetland. Compared to other prairie wetlands and global aquatic ecosystems, Frank Lake has a greater capacity overall to retain P through sediment sorption. Given the paucity of long-term (multi-decade) data on wetland response to effluent loading, we provide key insights into the dynamics of wetland P cycling in human-dominated watersheds.

1. Introduction

Environmental scientists and managers continue to grapple with problems linked to excess phosphorus (P) pollution (Bennett et al., 2001). One of the major sources of P pollution is wastewater effluent (Jarvie et al., 2006), which leads to costly environmental problems in downstream habitats including the proliferation of harmful algae (Hanson et al., 2020), shortage of dissolved oxygen (i.e., hypoxia, see Mallin and Cahoon, 2020) and population collapse of aerobic organisms (Mericas and Malone, 1984; Mishra et al., 2022). The use of natural wetlands to passively remediate surface water is a cost-effective method to reduce effluent P loads (Karjalainen et al., 2016) that has been in use for >50 years (Vymazal et al., 2021). Here, a natural wetland refers to an ecosystem essentially in its natural state (versus human-constructed wetlands), inundated with water and characterized by vegetation adapted to saturated soil conditions (Mitsch, 1992). The efficiency of natural wetlands in removing effluent P can reach 85% (Colares et al., 2020), mainly attributable to sediment sorption (Karjalainen et al., 2016). Sediment P-sorption ability, therefore, is a key indicator when evaluating wetland performance in P removal.

Over time, sustained loading of effluent alters wetland P-sorption capacity by modifying chemical (e.g., cation exchange capacity, dissolved oxygen concentration) and physical (e.g., hydraulic residence time) wetland features (Audet et al., 2020). On one hand, P-binding metal elements (such as Fe, Al and Ca) in effluent can modify the physical-chemical properties of wetland substrates via sedimentation and crystallization, providing additional sites for P sorption (Zhou et al., 2022). Elevated concentrations of Ca and Mg ions could also promote clay particle flocculation and reduce soil permeability, enabling P to bind more readily to wetland soil (Chibowski et al., 2011). Additionally, effluent salts increase solute content in wetlands, leading to more reducing conditions that stimulate organic matter burial and preservation, and greater P trapping in the sediment through polymerization (Mishra et al., 2022). On the other hand, increased wetland storage of effluent P can lower further sorption capacity and enhance P export (Haggard and Stoner, 2009; Yin et al., 2022). Salt-rich effluents can

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simultaneously cause P desorption, as excess sulfate and chloride compete with P for anion binding sites (Jin et al., 2013). Sulfide by-products can also react with oxidized iron in anoxic sediments, preventing iron-P complexation (Caraco et al., 1989; Heinrich et al., 2022). Additionally, effluent loading decreases the hydraulic residence time of wetlands, facilitating P desorption via sediment resuspension (Mendes et al., 2018). Thus, the impacts of effluent release on wetland P retention efficiency are difficult to predict, requiring detailed studies to elucidate the patterns and mechanisms underlying wetland P retention (Zhou et al., 2022). Yet Ury et al. (2023) highlight that most assessments of wetland P retention have focused on newly restored wetlands (0–3 years old). As a result, long term (multi-decade) estimates of wetland P retention efficiency remain uncertain.

Spatially, P sorption within a wetland can vary greatly, especially for those with multiple basins in sequence due to variable features including hydraulic residence time (Karjalainen et al., 2016), solute concentrations (Badiou et al., 2018), and soil characteristics (Kroetsch et al., 2011). Assessments of P sorption typically rely on data from a single point, or comparisons of inlet and outlet properties in small constructed wetlands (CWs), with relatively short residence time (Belmont et al., 2009; Zhou et al., 2022). Thus, predicting P sorption in multi-basin wetland complexes with variable environmental features remains a challenge (Karczmarczyk and Renman, 2011; Karjalainen et al., 2016), especially for wetlands receiving chemically-complex effluent.

Here we explored P sorption patterns and drivers in one of Canada's largest restored mineral-soil wetland complexes, Frank Lake, in the Prairie Pothole Region of Canada. This wetland complex has received inputs of agro-industrial and municipal effluent for >3 decades (Bogard et al., 2023). Despite expected exhaustion of P sorption in the wetland sediment layer (White et al., 1999), a mass balance showed the ecosystem retained over half the effluent P load >20 years post-flooding (Zhu et al., 2019). To explore the mechanisms underlying this observation, we collected sediment and aquatic geochemical data and conducted sediment P sorption incubations throughout the wetland. To facilitate a multi-decadal comparison of sorption capacity, our experiments were consistent with those of White et al. (1999). To frame our conclusions about the functioning of Frank Lake more broadly, we compared our findings to those from other Canadian prairie wetlands under distinct land use regimes (see 4.1 for more details).

2. Methods

2.1. Site description

The Frank Lake wetland complex (FL) in southern Alberta, Canada (Fig. 1), is a productive wetland that supports more than 190 bird species (White and Bayley, 1999; Zhu et al., 2019). FL has a total area of 10.1 km^2 across three basins (Basin 1, 5.1 km^2; Basin 2, 3.6 km^2; Basin 3, 1.4 km^2, see Zhou et al. (2023)). It is shallow, with a mean depth of 0.67 m (Basins 1 and 2) and 0.3 m (Basin 3) (White and Bayley, 2001). FL is ice-covered typically from November to April (~5 months). Sustained regional drought caused the wetland to dry in the 1980s, prompting water-level restoration through the diversion of wastewater into the wetland (White and Bayley, 1999; White et al., 1999). A pipeline was installed in 1989 to divert municipal (town of High River) and agro-industrial (Cargill Foods beef processing plant) effluent into the western side of Basin 1, to Basins 2 then 3, and finally discharging into the downstream Little Bow River (Fig. 1). The volume of annual effluent input is ~3.47 million m^3, and this P rich effluent is the near-exclusive (~90–100%) hydrologic source maintaining water levels in the wetland (Zhou et al., 2023). Open-water habitat dominates Basins 1 and 2, while Basin 3 is only inundated under wet conditions, remaining largely dry with wet meadow vegetation dominating the basin. Due to the long history of intense nutrient loading (approximately 90% of which originates from effluent input) FL is hypereutrophic, with P concentrations typically >2 mg L^{-1} (at three outlets, as reported by Zhu et al., 2019). The residence time of FL is 0.8 years calculated as the ratio between annual average lake volume and outflow discharge (Zhu et al., 2019). The semi-arid climate of the region leads to a high evaporation rate in the wetland (annual average of 862 mm), nearly twice as much as mean annual precipitation (450 mm, see Zhou et al., 2023). For more details about the wetland ecosystem, hydrological conditions and bathymetric characteristics, readers are referred to previous studies (Bogard et al., 2023; Flanagan et al., 2022; White and Bayley, 1999; White et al., 1999;

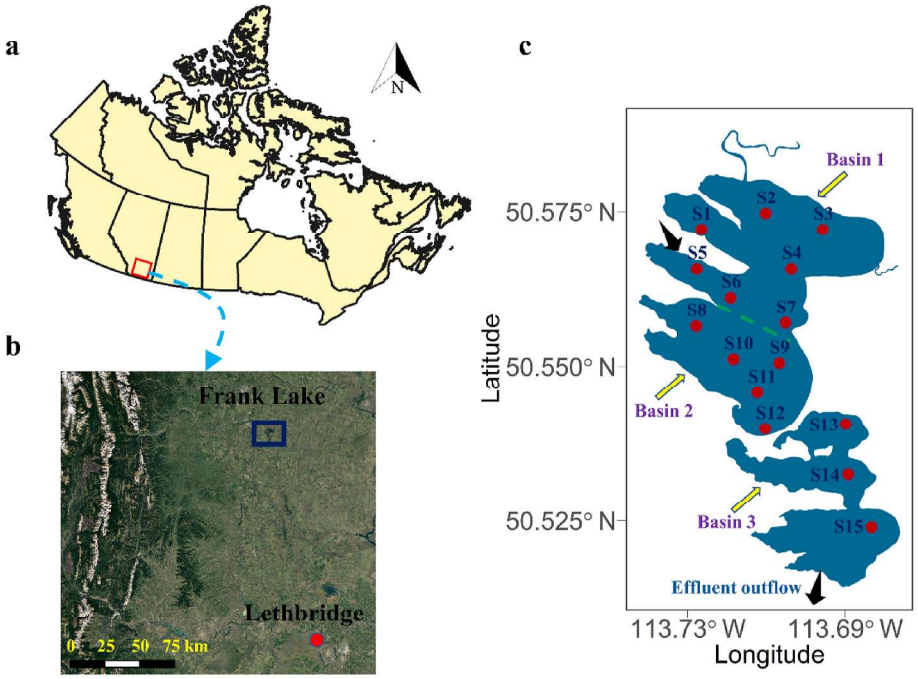

Fig. 1. (a) Location of Frank Lake in Alberta, Canada; (b) Terrain feature of Frank Lake and the region nearby; (c) Sampling sites for Frank Lake Basin 1, 2 and 3, with the black arrows indicating the effluent inlet and wetland outlet locations. Here, panel c shows the maximum extent of flooded land, which is much smaller during drought conditions.

Zhou et al., 2023).

2.2. Field sampling

During ice-free months, we took monthly surface water samples (at a depth of ~0.25 m) between August 2021 and October 2023, from the head of the pipe transporting effluent into Basin 1, and at the outlets of the 3 basins (Fig. 1), with samples from Basin 3 outlet only taken when water was present. Whole water samples were filtered through 0.45 μm high-capacity capsule filters into acid washed containers for further laboratory analyses. From August 2022 on, SRP concentrations in survey samples were quantified at the Element Laboratory (Calgary), following the APHA standard method (Rice et al., 2012), and lab-based filtration of samples.

In the fall of 2021, we conducted an extensive sediment and soil sampling campaign. Surface (10 cm) sediment was collected from all flooded locations using a 5 cm diameter plexiglass coring tube. Four samples were typically collected from one location, combined into one clean plastic bag and homogenized, then sealed and transported to the lab on ice in the dark. The four sub-samples for each location were from different horizontal (but nearby) sites, instead of vertical samples within one core. Near-shore samples were collected on foot, while open-water samples were collected by boat. Non-flooded samples in Basin 3 were collected by digging a ~20 × 20 cm areal surface soil sample with a spade, collecting the top 10 cm of the profile, and transporting the soil to the lab as above.

2.3. Laboratory analyses

Sediment and soil geochemical properties were characterized at the Natural Resources Analytical Lab at the University of Alberta following standard methods. Samples were dried and weighed (wet and dry weight). Extractions were done by equilibrating 2.5 g of dried sediment sample with 25 mL of Mehlich 3 (M3) solution for 5 min and filtering via Whatman No. 40 filter paper. This extraction method was used to facilitate comparisons with past results on prairie wetland sediment P sorption and relationships with sediment and soil properties (Badiou et al., 2018). From the extracts, concentrations of dissolved metals, S, and P were analyzed using inductively coupled plasma-optical emission spectroscopy (ICP-OES) using a Thermo iCAP6300 Duo, following a modified EPA 6010d method. Certified standards were used for calibration. Total organic N and carbon (C) content in dried and weighed solids were quantified using a Thermo Flanagan et al., 2022 Organic Elemental Analyzer (EA).

Water samples were taken during the extensive sediment survey, and samples from the sorption experiments were all analyzed for SRP. The molybdenum blue method was applied to quantify SRP in filtered water samples (0.45 μm), either using a continuous flow analyzer (Xylem-O/I Analytical FS3700) or, for a small subset of surface water samples (sites S2, S4, S6), using spectrophotometric analysis. In all cases, standard curves were generated and known standards were used to calibrate SRP concentrations.

2.4. Sediment P-sorption experiments

The P sorption experiment was conducted following methods of Badiou et al. (2018). Sediment samples were homogenized, after removing visible plants and stones. Subsets of 1.0 g (weighed with a precision scale) were added to seven 50 mL Pyrex vials each containing one of the following concentrations of KH_2PO_4 solution: 0.2, 0.5, 1.0, 5.0, 10.0, or 20.0 mg P L^{-1}, plus one random duplicate of one of the concentrations. To ensure equilibration, vials were placed on an end-over-end LabquakeR Thermolyne set to 8 rotations per minute and held at a constant temperature of ~20 °C for 24h. Vials were then filtered on pre-weighed Whatman GF/C filters and the filtrate was analyzed within the day for SRP (see above). To determine the dry weight of the sediment sample, filters were dried in a desiccator for 24h, placed in an oven at 100° for 24h, then re-weighed.

2.5. Quantification and comparison of P-sorption capability in 1995 and 2021

We calculated the total amount of P sorbed in FL sediment (S) using the equation (Kang et al., 2009):

$$S = S_0 + S' \tag{1}$$

where S_0 is the amount of P originally sorbed (mg kg^{-1}) and S' is added P sorbed (mg kg^{-1}) in the sediment (Fig. S1). Here, the S' was calculated by:

$$S' = (C_0 V - C_t V) / M \tag{2}$$

where C_0 is the initial concentration of P added (mg L^{-1}), C_t the P concentration in the equilibrium solution (mg L^{-1}), V the volume of solution, and M the dry mass of sediment. We quantified S_0 by fitting the Freundlich isotherm equation with an exponent of 1/3 following Badiou et al. (2018) and references therein:

$$S' = k \times C_t^{1/3} - S_0 \tag{3}$$

where k is a dimensionless number illustrating the slope of the sorption curve.

After testing the appropriateness of three distinct fitting methods for isotherm construction (One- and Two-surface Langmuir Isotherm, and Langmuir-Freundlich models), we identified that the latter two were not appropriate for these data (fully described in Text S1 and Fig. S2). Thus, we fit the One-Surface Langmuir Isotherm model (OSL, equation (4)):

$$S = \frac{S_{max} K C_t}{1 + K C_t} \tag{4}$$

where S_{max} indicates the maximum P sorption capacity by the sediment (i.e., the predicted sorption potential, mg kg^{-1}), and K the Langmuir binding-strength coefficient (L mg^{-1}). By fitting the linear regression model between C_t and S, we also quantified two other important parameters (EPC_0 and PEBC) related to P sorption potentials. Here, EPC_0 is the equilibrium aqueous-phase P concentration where there is no net exchange of P between sediment and water, with the value determined as the intercept of the regression line with the x-axis (Sui and Thompson, 2000). PEBC is the P equilibrium buffering capacity indicating the sediment's ability to sorb additional P, determined as the slope of the regression line (Hongthanat et al., 2016). Following the methods of Badiou et al. (2018) values of C_t, corresponding to the four lowest initial P concentrations (from 0 to 1 mg L^{-1}), were used in calculations.

For select locations in Basin 1 we compared the results calculated above with those from 1995 (i.e., 1 year after the complete filling of Frank Lake by the late 1993), to elucidate its long-term evolution of P sorption capability. On 15–19th June 1995, nineteen sediment cores from Basin 1 were collected, of which the total P content was analyzed following Mayer (1981). Surface sediment samples were used to conduct P-sorption experiments following a similar approach to our methods (White et al. (1999)), with the surface water amended to the P concentrations of 25, 50, 75, 100, 200, 300, 400, and 500 μg P l^{-1}. The authors quantified P sorbed to the sediment according to Eq. (2) (see Fig. 4 in White et al., 1999). We then digitized their results via Origin 2023b (OriginPro, 2023) and determined parameters including S_0 and PEBC, by using the same methods as described above. Moreover, we compared our results on P-sorption capability with other Canadian prairie wetlands, expanding the scope of our research beyond the Frank Lake case study (see section 4.1 for more information).

2.6. Statistical analyses

All data processing was performed using R version 4.2.2 (Team, 2022), with packages listed in the supporting information (Text S2). The Inverse Distance Weighing (IDW) interpolation algorithm was used to interpolate spatial distribution of P sorption and buffering parameters. IDW assigns weights to known data points based on their distances from the interpolation point, with closer points having more influence on the interpolated value. This distance-based weighting method is well-suited to handling irregular basin boundaries (like at Frank Lake), reducing spatial uncertainties by incorporating the specific ecological and geographical context of the basin directly into the interpolation process (Khouni et al., 2021). For more detailed information about the IDW interpolation, readers are referred to Lu and Wong (2008) and Getis (2009). Principal components analysis (PCA) was used to characterize general relationships between sediment geochemistry parameters. We used Pearson correlation analysis and multiple linear regression models (MLR) to explore how sediment chemical properties related to P sorption metrics, and applied Akaike Information Criterion (AIC) to guide MLR model selection. Boxplots are shown using default settings in R, where the midpoint is the median value within a group, ends of boxes represent the 1st and 3rd quartiles, whiskers extend to the minimum and maximum points with the length <1.5 times the interquartile range, and points outside are outliers. The Kruskal–Wallis test was used to assess inter-group differences, and if the null hypothesis was rejected ($p < 0.05$) then the Dunn's post hoc test was used to identify differences between each group.

3. Results

3.1. Sediment chemical properties in Frank Lake

Concentrations of major P-binding elements in sediment extractions were lower in Basin 3 than other basins (Table S1). The mean concentrations of extractable Fe were >1000 mg kg^{-1} in Basins 1 and 2, three times higher than in Basin 3. Similar differences were seen for concentrations of Ca and Mn (Fig. S3). Concentrations of P in sediment in Basin 1, especially the effluent-receiving area (S5-S7) were up to 2000 mg kg^{-1}, with values decreasing through Basins 2 and 3. In contrast, mean concentrations of Cu in Basin 3 were 9.4 mg kg^{-1}, slightly greater than in other basins (Table S1). Light metal (i.e., Mg and Na) and S content was more uniform across basins (Kruskal-Wallis; $p = 0.48$, 0.42 and 0.47, respectively). For Zn, concentrations were lower in Basin 2 but comparable between Basin 1 and 3 (Dunn's test: $p = 0.75$) The PCA summarized these sediment differences well. The first three PCs explained 85.9% of variation in chemical parameters (PC1: 52.7%, PC2: 21.1% and PC3: 12.1%). Sites in Basins 1 and 2 largely overlapped in the PCA with more similar geochemical features than for Basin 3 (Fig. S4). The exception was S12 (southern edge of Basin 2), which had a higher PC2 score, and higher concentrations of Mn, Ca and Fe. The sites in Basin 3 were unique, with PC2 scores between −4 and −2, associated with lower content of P-binding elements (Fig. S4).

3.2. Spatial distribution of P sorption and geochemical correlates

Our experiments revealed spatial heterogeneity of P-sorption metrics (Fig. 2). The mean value of PEBC was 1224 and 1306 L kg^{-1} for Basins 1 and 2, respectively, but only 166 L kg^{-1} for Basin 3 (Fig. 2; Table S2). Similarly, mean S_{max} values decreased from 1974 mg kg^{-1} (Basin 1) and 2117 mg kg^{-1} (Basin 2) to 408 mg kg^{-1} (Basin 3). Mean EPC_0 values were higher in Basin 1 (0.31 mg L^{-1}) than Basins 2 (0.10 mg L^{-1}) and 3 (0.16 mg L^{-1}) (Fig. 2; Table S2). Values of S_{max} (3454.8 mg kg^{-1}) were greatest at the effluent-receiving point S7 indicating, in theory, the maximum P-sorption at this site. At site S7, however, P originally sorbed in the sediment reached 1502 mg kg^{-1}, double the average value of the entire wetland (Fig. 2; Table S2). The corresponding EPC_0 at S7, accordingly, rose to 0.96 mg L^{-1}, while remaining below 0.35 mg L^{-1} for other sites.

Spatial gradients of P sorption were strongly predicted by sediment geochemical patterns (Fig. 3). Both S_0 and S_{max} were positively correlated with each other, as well as the sediment content of major P-binding elements (e.g., Ca, Fe and Mn; r = 0.6 to 0.9). Values of EPC_0 correlated with sediment N content (percentage dry mass, $p = 0.004$, r = 0.7) and Cu ($p = 0.002$, r = 0.7) (Fig. 3). Values of PEBC were only correlated

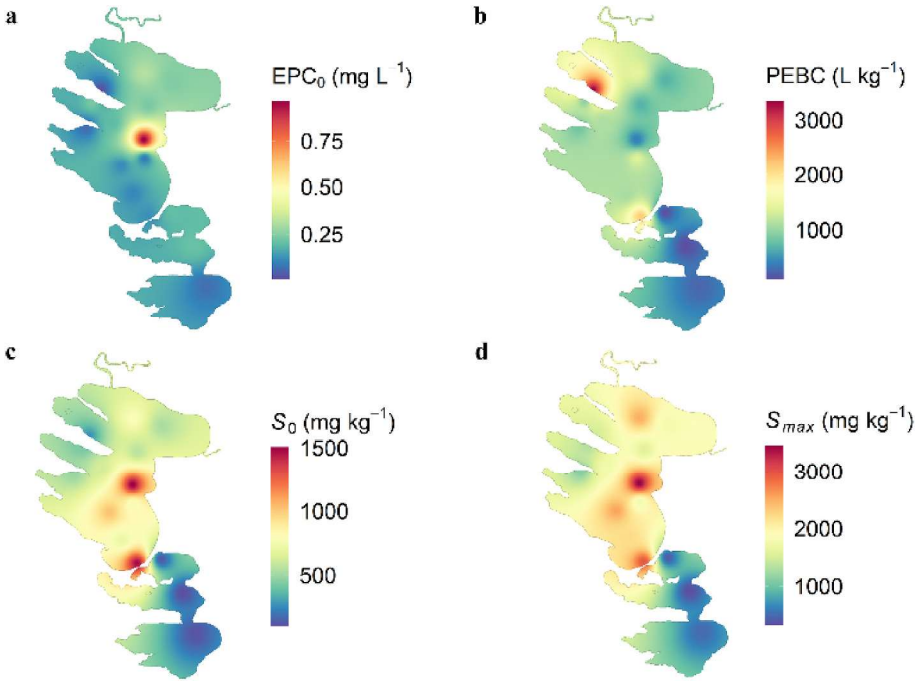

Fig. 2. Spatial distribution of P sorption and buffering parameters (panels a to d show EPC_0, PEBC, S_0 *and* S_{max}, respectively). See Table S2 and methods for parameter definitions.

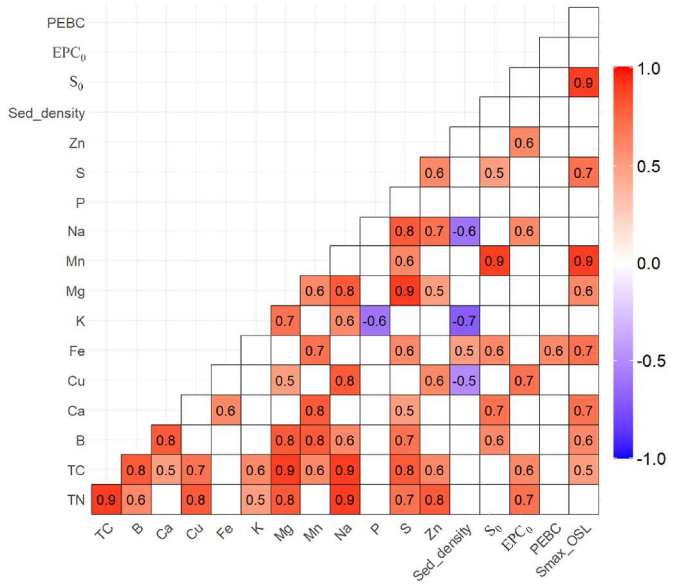

Fig. 3. Correlation diagram indicating the relationship between sediment chemical properties and P sorption metrics in Frank Lake. Correlations with p values > 0.05 are considered insignificant (coefficient values left blank).

with Fe ($p = 0.009$, r = 0.6) (Fig. 3). To best predict spatial patterns in Fig. 2, MLR models were generated, and those with the lowest AIC scores are reported ($R^2 > 0.75$ for all; Table S3 for model details). All models had AIC values lower than those for associated null models (Table S3). Our results confirmed the important role of sediment Fe, Ca and Mn content in predicting S_0 and S_{max}, though the collinearity of key sediment properties (e.g., Fe and Mn; Fig. S4) led to only one such parameter being selected in final MLR models (Mn selected in MLR for S_{max}; Table S3). Comparison of simulated and observed results showed the points were evenly distributed around the identity line (Fig. S5), suggesting models contained no systematic error. The availability of metals including Fe and Mn, as well as Ca ions appear to be important in sustaining the decades-long P sorption capacity in Frank Lake.

3.3. Temporal evolution of P sorption in Frank Lake

The capacity of P retention in Basin 1 has decreased since 1995 (Fig. 4), as indicated by comparison of our results with those from White et al. (1999). At a concentration of added P of 0.2 mg L^{-1} (i.e., C_0 in Eq. (2)), mean sediment sorption in effluent-receiving areas (sites S5-S7) decreased from 346 (1995) to −35 mg kg^{-1} (in 2021, see Fig. 4a). The lowest value in 2021 was −100 mg kg^{-1} (at S7), indicating a shift from past P sink to source in 2021. For the other sites in Basin 1 (S1-S4), the amount of P sorbed was 7 mg kg^{-1} in 2021, versus 1113 mg kg^{-1} in 1995. We observed similar trends when $C_0 = 0.5$ mg L^{-1}, for which mean P sorbed at the effluent-receiving area decreased from 1042 to −6 mg kg^{-1}, and from 1692 mg kg^{-1} to 24 mg kg^{-1} (from 1995 to 2021) at the other sites of Basin 1 (Fig. 4b).

The P buffering capacity in Basin 1 was markedly higher in 1995 than 2021 (t-test: $p = 0.013$; Fig. 4c). For the effluent-receiving area, the average PEBC in 1995 (2134 L kg^{-1}) was more than double that in 2021 (816 L kg^{-1}) and as low as 390 L kg^{-1} at S7. Similarly, PEBC at sites S1-S4 decreased from 3436 L kg^{-1} (1995) to 1531 L kg^{-1} (2021). Conversely, the amount of P originally sorbed in the sediment (S_0) increased through time, from 136 mg kg^{-1} (sites S1-S4) and 209 mg kg^{-1} (S5-S7) in 1995, to 535 and 851 mg kg^{-1}, respectively, in 2021 (Fig. 4d). Additionally, the S_0 within Basin 1 varied greatly in 2021 with the standard error (SE) of 330 mg kg^{-1} (S1-S4) and 106 mg kg^{-1} (S5-S7), which were much higher than in 1995 (15 mg kg^{-1} and 23 mg kg^{-1}, respectively). Despite this clear decrease in sediment P sorption capability since 1995, our results indicated that the Frank Lake wetland in Basin 1 currently remains a P sink, largely due to the high SRP concentrations in the overlying surface water (Fig. 5) that greatly exceed EPC_0 values (normally <0.3 mg L^{-1}).

4. Discussion

Here, we documented key aspects of P cycling in Frank Lake, one of the largest restored wetland complexes in Canada, restored in 1989 as part of a major conservation effort (White and Bayley, 1999). We had a unique opportunity to elucidate sediment P sorption trends over a ~25-year period during which Frank Lake underwent agro-industrial and municipal effluent loading. In this field, the vast majority of related work is done using one time point, with no temporal dimensions to their studies. Thus, incorporating two time points spanning multiple decades in our study is highly valuable and deliberate. Our results indicate that relative to a similar study conducted in 1995, P sorption capacity has now become exhausted near the effluent inlet, but remarkably, remains elevated throughout the rest of the wetland. Additionally, the wetland remains an overall net P sink (Zhu et al., 2019) at current concentrations of surface water SRP (Fig. 5). Aquatic habitats of Basins 1 and 2 had far higher P sorption capacity (Fig. 2) than the wet meadow habitat in Basin 3 that is exposed to the atmosphere in drought conditions and low water levels. Sorption capacity was strongly dependent on gradients of sediment content of multiple P-binding elements including Ca, Fe and Mn (Fig. 3), which were present at high enough concentrations to sustain P sorption in most wetland locations (Fig. S3). We present a unique documentation of a successful wetland restoration project that has provided stakeholders with decades of sediment P retention and water quality treatment.

4.1. Frank Lake versus other prairie and global aquatic ecosystems

We compared our findings for Frank Lake to other Canadian prairie wetlands under distinct land use regimes using published (Badiou et al., 2018) and unpublished datasets (Fig. 6). These included drained and intact wetlands located in the Broughton's Creek (BC) watershed, and the freshwater coastal wetlands within Delta Marsh (DM) on the south shore of Lake Manitoba (Fig. 6b and c). The wetlands each had a maximum depth <3m and shared similar climate characteristics with Frank Lake. Yet concentrations of TP in the surface water of these wetlands were comparatively low (0.11 mg L^{-1} to 1.19 mg L^{-1}), as none were used to treat wastewater. Following methods outlined in sections 2.3 and 2.4, we estimated the specific P sorption capacity of sediments.

Compared to many other aquatic ecosystems, Frank Lake has a greater capacity overall to retain P through sediment sorption. Frank Lake had a mean S_{max} of 1709 mg kg^{-1}, consistent with inland coastal wetlands within Delta Marsh (mean $S_{max} = 1476$ mg kg^{-1}; $p = 0.58$), but greater than intact prairie wetlands (mean $S_{max} = 1018$ mg kg^{-1}; $p = 0.053$) and drained prairie wetlands (mean $S_{max} = 470$ mg kg^{-1}; $p = 4.7 \times 10^{-7}$) (Fig. 6d–Table S4). The values of maximum P sorption capacity reported here were much higher than those reported for the sediments of shallow polymictic lakes (Zhou et al., 2005), marine wetlands (Wang and Li, 2010) and littoral zones in stratified lakes (Carmignani and Roy, 2017). Higher S_{max} values for the sediments of prairie wetlands studied here may be attributed to their unique geochemical properties that include elevated organic matter content (up to 20% dry wt. on average, data not shown), which can facilitate complexation of P with Mg and Fe (Yang et al., 2019). Further, these wetlands often have greater coverage of emergent aquatic plants (e.g., hard-stem bulrush and *Typha* spp.) that enhance P sorption potential of sediments (Bhomia and Reddy, 2018). By contrast, our S_{max} results were lower than those for some deep pelagic sediments in stratified lakes (e.g., Little Rock Lake (Detenbeck and Brezonik, 1991), or Lake Čertovo jezero (Borovec and Hejzlar, 2001)). This discrepancy could be due to the fact that deep lake benthic zones may undergo accumulation of more amorphous Fe and finer grained

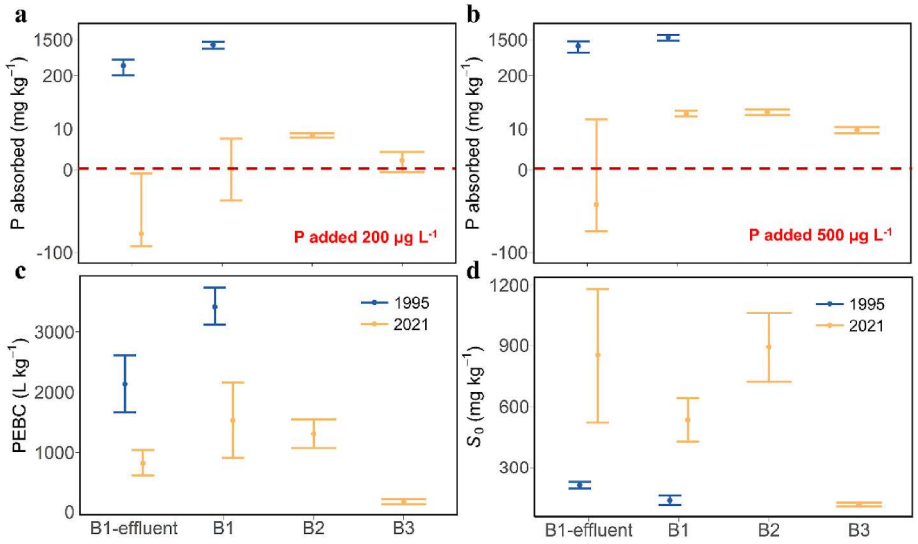

Fig. 4. Dynamics of P sorption in Frank Lake (mean and SE) before and after long-term input of municipal effluent (1995 vs. 2021). Data in 1995 are from White et al. (1999). Here B1-effluent indicates the sampling sites S5 to S7, and B1 from S1 to S4. Figures a and b show the amount of added P sorbed in sediment (i.e., S' in equation. (1)) under the experimental SRP concentration in surface water of 200 μg L^{-1} and 500 μg L^{-1}, respectively, and the red dashed lines are at P absorbed = 0 indicating the boundary between P source and sink. See Table S2 and methods for parameter definitions.

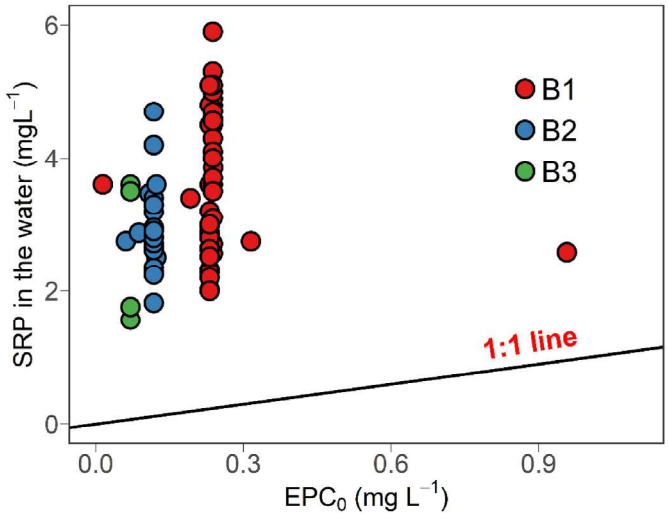

Fig. 5. Comparison of ambient surface water SRP concentration in Frank Lake (monthly samples from 2021 to 2023) versus EPC$_0$ values of corresponding basins. See Table S2 and methods for parameter definitions.

sediments with more surface area for P sorption (Detenbeck and Brezonik, 1991). Overall, prairie wetlands appear to provide substantial P retention capacity on the landscape relative to other global aquatic habitats, and this service can persist for decades, even with intense P loading (in Frank Lake), or with restoration following heavy land use (Badiou et al., 2018, Fig. 6).

While prairie wetland sediments have a great capacity for P retention, after >25 years of P loading this function can become saturated, as seen in Frank Lake. In 2021, EPC$_0$ values were elevated, averaging 0.21 mg L^{-1} (median of 0.19 mg L^{-1}), statistically similar to values for drained prairie wetlands (p = 0.57; Fig. 6d). These values were far higher than published values for shallow temperate zone lakes (Cyr et al., 2009), or median values (0.05 mg L^{-1}) from a global stream meta-analysis (Simpson et al., 2021). This indicates that for Frank Lake, the concentration of water column SRP required to sustain net sediment P sorption exceeds many aquatic ecosystems worldwide but is consistent with other drained prairie wetlands. A similar conclusion applies to S_0, the amount of P initially sorbed in sediments prior to experimental manipulation. Mean S_0 in Frank Lake in 2021 (633 mg kg^{-1}) was significantly greater ($p = 1.65 \times 10^{-10}$) than for other prairie wetland categories (55–84 mg kg^{-1}; Fig. 6d–Table S4). In contrast, if only considering the wet meadow sites of Frank Lake (S13-S15), which were exposed to the atmosphere and had little interaction with effluent, the mean S_0 in 2021 for these habitats was much lower (116 mg kg^{-1}; Fig. 6d–Table S2) and similar to both other prairie wetlands (Fig. 6d) and values for Basin 1 in fall 1995 (Fig. 4d). This suggests that habitat within Frank Lake still exists that has the capacity to sorb excess quantities of P, yet hydrologic conditions are needed that may enhance soil-water interactions in such locations. Ultimately, Frank Lake may be a model for the trajectory of change in prairie wetland sediment P storage under sustained (multi-decadal) loading.

4.2. Factors controlling spatiotemporal variations of P-sorption

We found a wide range in P sorption patterns throughout Frank Lake linked to gradients of sediment geochemistry and hydrologic variability. The maximum P sorbed in Basin 3 was 20% of that in Basins 1 & 2 (Fig. 2, Table S2). This is not surprising, as sediments in Basin 3 are mainly composed of light and coarse-grained sediment, with the density ranging between 0.6 and 0.8 g cm^{-3}. Dense organic rich sediments and fine clays dominated in Basin 1 & 2 (density >1 g cm^{-3}; Fig. S6). These finer particles have higher specific surface area, which provides more surface for P sorption (Liang et al., 2019). Sediments in Basin 3 are often exposed to the atmosphere (e.g., under current drought conditions), leading to more porous and desiccated sediment. Further, gradients in sediment properties and sorption capacity could be partly attributed to different hydraulic residence time (HRT) within each basin. During wet seasons in which all basins were inundated, outflow discharge in the three basins was quite similar to each other (between 0.20 m^{3}s^{-1} and 0.25 m^{3}s^{-1}, see Zhu et al. (2019)) while the volume in Basins 1 & 2 is > 4 times greater than in Basin 3. This resulted in a lower approximate HRT in Basin 3 (21 days) than Basins 1 and 2 (185 and 111 days), at least under previous periods of elevated water level and discharge (Zhou et al., 2023). Lower HRT tends to reduce sinking rates and sediment accumulation of P-binding elements from the water column (Liang et al., 2019), corresponding well with our findings (e.g., lower concentration of Fe and Mn in Basin 3 soils compared to the other two basins). It should be noted that lower HRT can also suppress P sedimentation from the

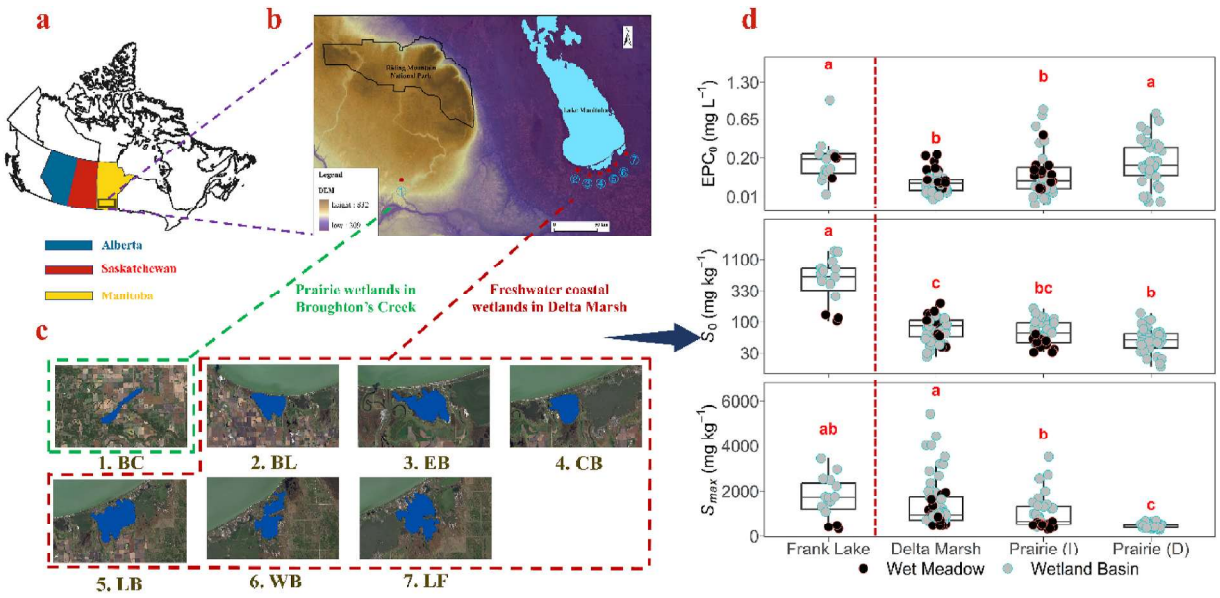

Fig. 6. (a) Location of the three prairie provinces in Canada; (b) Topographic map of Manitoba; (c) The shape of prairie (including intact (I) and drained (D)) wetlands in Broughton's Creek (BC) and freshwater coastal wetlands in Delta March (DM) involved in the study: Broughton's Creek (BC), Big Lake (BL), Cadham Bay (CB), Eaglesnest Bay (EB), Lake Francis (LF), Lyttle Bay (LB) and Waterhen Bay (WB); (d) Comparison of P sorption parameters between Frank Lake and the other prairie wetlands for: EPC0 (top), S_0 (middle) and S_{max} (bottom). Points with two different colors indicate the results for wet meadow and the main submerged basin, respectively in each wetland. Variables sharing the same letter above denote no significant difference at $p < 0.05$ (Dunn's test) . Here, all the sampling sites within drained wetlands of Broughton's Creek are in the wetland basin.

surface water, possibly contributing to less P originally adsorbed in Basin 3. The S_0 was positively related with S_{max}, accordingly, with the correlation coefficient up to 0.9 in FL ($p < 0.01$, Fig. 3), which indicated the effluent input plays a dual role in modifying P-sorption capability of the wetland and the effect largely depends on its chemical constitution (i.e., balance between the sedimentation of P and P-binding elements). Overall, our results help to clarify how variation in sediment physical-chemical properties influences the P sorption capacity of restored wetlands and provide key insights into mechanisms underlying P cycling in human-dominated watersheds.

The sediment-specific or inter-basin differences within Frank Lake that shape P sorption patterns represent proximal controls that operate within the broader environmental and management factors ultimately regulating P sorption capacity in Frank Lake. Roughly half (9.8 tons yr^{-1}) the P transported through Frank Lake is retained (Zhu et al., 2019). This is the most important driver of declining sediment P retention capacity. Long-term effluent input can also alter soil structure in the wetland by reducing soil porosity, saturating binding sites, and thereby decreasing potential P sorption capacity (Amador and Loomis, 2020). The extent to which such changes in soil physical structure have occurred in Frank Lake and are contributing to P sorption patterns are unclear, but could be explored in future work using different methods such as Scanning Electron Microscopy (SEM) combined with Energy Dispersive X-ray Spectroscopy (EDS) (Kaplan et al., 2016). Additionally, owing to the continuous effluent input and surface drainage, Frank Lake has been transformed from an endorheic system with no outflow (pre-1995) to a flow-through wetland connected to the river network in non-drought years (Zhu et al., 2019; Zhou et al., 2023). In the North American prairies, sediments in intact wetlands are often anoxic, with decreased soil organic matter (SOM) decomposition rates (Badiou et al., 2018; Tangen et al., 2015). Increased SOM preservation directly enriches the sediment P pool and enhances P buffering (Yang et al., 2019), while enhancing the concentration of P-binding elements like Mg, Al and Fe, through the formation of strong organo-mineral associations and polyvalent cation bridges (Zhao et al., 2023). Conversely, wetlands like Frank Lake with increased outflow tend to have more aerobic habitat that alters sediment redox, enhances SOM mineralization, and limits P complexation with Mg and Fe (Lützow et al., 2006). This is especially the case for Basin 3 where variability between wet and dry conditions further increases SOM mineralization (Schönbrunner et al., 2012). Additionally, outflow promotes the export of soluble ferrous Fe, further decreasing Fe availability for P binding (Badiou et al., 2018). Moreover, increasing salt (Cl^- and SO_4^{2-}) content resulting from effluent release and evaporation in downstream basins (Denny, 2024; Zhu et al., 2019) can directly compete with P for binding sites on oxidized iron, thereby further reducing P sorption capacity in Frank Lake.

In this study, P-sorption experiments were conducted within a single season, as our main objective was to explore spatial gradients and predictors of sorption across three basins of Frank Lake, and seasonal patterns were less of a focus. The sediment redox conditions and geochemical properties of the wetland remain relatively stable throughout the ice-free period (Bogard et al., 2023), which also stabilizes the P-sorption capacity of inundated sediments. We would expect that anoxic conditions in water during ice-covered months (Bogard et al., 2023) may lead to temporary shifts in redox properties that weaken P sorption capacity in Frank Lake. Yet during these periods, there is no outflow from the wetland, so any resuspension of bound P in the water would not enhance the loss of P from the system during winter months. Further, existing ecosystem mass balances (Zhu et al., 2019) capture such seasonality and show a strong capacity for P retention at the ecosystem scale throughout the year, agreeing with our overall findings that much of the wetland remains a P sink. Thus, we are confident that our experiment has not misrepresented ecosystem level functioning of the wetland. Finally, while we infer substantial decrease in sorption values for Basin 1 since the wetland was flooded (from ~10 to 100 fold between 1995 and 2021; Fig. 4), additional sediment incubation experiments in intermediate years would have strengthened our conclusion.

4.3. *Management implications for Frank Lake and other wetlands*

Under current management, future sustained input of P to Frank Lake will likely lead to further saturation of sediment P sorption capacity, as suggested by the declining temporal trend in sorption capacity

of Frank Lake Basin 1 sediments (Fig. 4) and elevated S_o values relative to other prairie wetlands (Fig. 6d). The average P assimilation capacity for natural wetlands in North America is ~1 g P m^{-2} yr^{-1}, above which wetland functioning becomes impaired (Richardson and Qian, 1999). Past work indicates that P loading to Frank Lake has surpassed this threshold (4.7 g P m^{-2} yr^{-1}, White and Bayley, 2001), and that the wetland may act as a P source for years to come, despite the fact that it currently sorbs additional P in sediments. The current sorption is largely driven by the high surface water SRP concentrations that exceed EPC_0, the threshold concentration at which sediments transition from P source to sink (Fig. 5). Taken together, these observations align with the prediction that sediment P retention in Frank Lake has declined in recent years, possibly supporting an increased P flux into the downstream Little Bow River (Zhu et al., 2019). The future source-sink balance in Frank Lake will ultimately depend on whether the areal extent of P desorption (e.g., as shown near the effluent outflow location) exceeds the sorption capacity of other remaining habitats beyond Basin 1.

From a management perspective, Frank Lake is an important case study. First, efforts to enhance or restore sediment P-sorption capacity may extend the ecosystem service of P removal. Yet the use of solid phase sorbents for P management such as La-modified bentonite (LMB) may not be effective due to its binding by dissolved organic carbon under oxic and polymictic conditions (Neweshy et al., 2022). Further, application of Al- or Fe salts to such a large open-water area may be cost prohibitive and have detrimental effects on biota in this Key Biodiversity Area (Key Biodiversity Areas Partnership, 2024). Second, an extreme quantity of P is now sorbed in the sediment of Frank Lake, and this legacy P reserve must be factored into watershed management decisions. Future efforts to reduce surface water P via source controls may reduce SRP to levels that shift sediments toward the release of P. Under drought conditions and low water levels, sediment P resuspension is unlikely to impact downstream river habitats due to the lack of wetland outflow (Zhou et al., 2023). This impact would be most pronounced when wet conditions persist and the wetland is connected to the downstream river. Third, as an extension of this last observation, P management strategies must consider the overarching controls on wetland functioning imposed by the regional climate. Sediment sorption/desorption processes respond to climatic changes (Eller et al., 2021; Jarvie et al., 2020) that modify wetland hydrodynamics (Karim et al., 2020). These factors must be considered when predicting future sorption capabilities of wetlands such as Frank Lake that have undergone historical nutrient loading, because the ecosystem services and financial implications associated with watershed P management are substantial (Smith et al., 2019).

5. Conclusions

Our results showed a wide spatial range in sorption capability throughout Frank Lake, one of the largest restored wetlands in Canada, which was successfully predicted by the long gradient of sediment content of major P-binding elements (e.g., Ca, Fe and Mn). Three decades of agro-industrial and municipal effluent loading has reduced the sediment P sorption capability in the first basin compared with 1995, indicated by decreasing P equilibrium buffering capacity (PEBC) and a greater amount of P sorbed in sediments (S_0). Surprisingly, the Frank Lake wetland remains a P sink due to high SRP concentrations in overlying surface water, plus the extensive sediment area in other basins that remain capable of sorbing P. The comparison of our findings to other Canadian prairie wetlands under distinct land use (drained, intact and freshwater coastal) regimes underscores that prairie wetlands have substantial P retention capacity on the landscape relative to other global aquatic habitats, but this function may become saturated after long-term P loading. Overall, Frank Lake can be regarded as a template illustrating the trajectory of change in prairie wetland sediment P storage under sustained (multi-decadal) loading.

CRediT authorship contribution statement

Chenxi Mi: Writing – review & editing, Writing – original draft. **Cynthia Soued:** Writing – original draft, Conceptualization. **Lauren E. Bortolotti:** Writing – review & editing, Conceptualization. **Pascal Badiou:** Writing – review & editing. **Bryan Page:** Writing – review & editing. **Mariya Denny:** Writing – review & editing, Visualization. **Matthew J. Bogard:** Writing – review & editing, Writing – original draft, Supervision, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2024.120256.

Data availability

All data are available in the Federated Research Data Repository (FRDR) at https://doi.org/10.20383/103.01083, except for the SRP measurements shown in Fig. 5, which are available upon request.

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