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CURRENT EVIDENCE

Mud in the city: Effects of freshwater salinization on inland urban wetland nitrogen and phosphorus availability and export

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Scientific Significance Statement

Human-caused salinization of freshwater ecosystems is a well-known threat. Wetlands in cities are vulnerable to salinization from road salt, wastewater, weathering infrastructure, and other sources. Although we protect, conserve, restore, and build wetlands in cities to remove polluting nutrients, especially nitrogen and phosphorus, salt might fundamentally change wetland soils, microbes, and plants. Our current understanding of how salinization influences wetland sediment nutrient processing is based on studies that either mimic coastal saltwater intrusion or add only sodium chloride. Urban freshwater salinization is far more variable in ionic composition, ionic strength, and temporal loading than either of these scenarios. By reviewing the results of published experiments, we demonstrate that together, the effects of freshwater salinization diminish the capacity of wetlands to remove polluting nutrients.

Abstract

Salinization and eutrophication are nearly ubiquitous in watersheds with human activity. Despite the known impacts of the freshwater salinization syndrome (FSS) to organisms, we demonstrate a pronounced knowledge gap on how FSS alters wetland biogeochemistry. Most experiments assessing FSS and biogeochemistry pertain to coastal saltwater intrusion. The few inland wetland studies mostly add salt as sodium chloride. Sodium chloride alone does not reflect the ionic composition of inland salinization, which derives from heterogeneous sources, producing spatially and temporally variable ionic mixtures. We develop mechanistic hypotheses for how elevated ionic strength and changing ionic composition alter urban wetland sediment biogeochemistry, with the prediction that FSS diminishes nutrient removal capacity via a suite of related direct and indirect processes. We propose that future efforts specifically investigate inland urban wetlands, a category of wetland heavily relied on for its biogeochemical processing ability that is likely to be among the most impacted by salinization.

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Additional Supporting Information may be found in the online version of this article.

Anthropogenic freshwater salinization is ubiquitous (Cañedo-Argüelles Iglesias 2020; Kaushal et al. 2021). Increasing salt concentrations, enhanced variability, and changing ionic composition impact freshwater organisms (Hintz and Relyea 2019) and alter geochemical dynamics (Chambers et al. 2016). Salinization and its effects, also known as the freshwater salinization syndrome (FSS) (Kaushal et al. 2018, 2019, 2021), have been documented in diverse freshwater ecosystems including lakes (Dugan et al. 2017a), streams (Kaushal et al. 2005, 2018; Reisinger et al. 2019), rivers (Cañedo-Argüelles et al. 2013), coastal wetlands (Tully et al. 2019), and groundwater (Cassanelli and Robbins 2013; Kelly et al. 2018; Jamshidi et al. 2020). An emergent body of research on FSS has focused on streams and rivers at catchment scales (Kaushal et al. 2017, 2018; Stets et al. 2018; Schulz and Cañedo-Argüelles 2019; Kaushal et al. 2021). In contrast, FSS is understudied in wetlands, particularly noncoastal inland wetlands. Although several studies indicate that FSS impacts inland freshwater wetlands (Wilcox 1986; Pugh et al. 1996; Herbert et al. 2015; Rhodes and Guswa 2016), most published research on wetland salinization has focused on coastal wetlands experiencing salinization associated with sea level rise (Herbert et al. 2015; Tully et al. 2019) and arid region wetlands (Jolly et al. 2008). Inland wetlands, especially urban ecosystems like roadside ditches, stormwater ponds, and accidental wetlands (Palta et al. 2017; Clifford and Heffernan 2018; Sinclair et al. 2020; Tatariw et al. 2021), are often on the salinization "front lines," directly receiving anthropogenic salt loads (Van Meter et al. 2011; Hill and Sadowski 2016; Liang et al. 2017). Although relied upon, and sometimes specifically designed and constructed, for nutrient removal (Zedler 2003; Vymazal 2007), salinization may constrain urban wetland biogeochemical function through a combination of interrelated mechanisms.

Salinizing urban wetlands are ubiquitous in landscapes with human presence

Although human development has led to widespread global wetland loss and degradation, freshwater wetlands remain ubiquitous in many cities (van Asselen et al. 2013; Steele and Heffernan 2014). Three defining wetland features—hydrology, soils, and vegetation—are highly altered in urban settings (Palta and Stander 2020). Age, history, and varying degrees of active management create a complex mosaic of legacies and trajectories among urban ecosystems (Palta and Stander 2020). Many relict wetlands have not been drained or developed during known history, but are embedded in human-dominated landscapes where cities have "grown up" around them (Palta and Stander 2020). Restored and rehabilitated sites may contain legacies of contamination from periods of disturbance or draining for land reclamation (Griffin and Dahl 2016; Ravit et al. 2017). Constructed wetlands for water treatment, habitat mitigation, and other goals may contain a range of natural to human disturbed and created substrates (Vymazal 2007). Accidental wetlands form and persist on disturbed lands without intentional human construction or management (Palta et al. 2017; Maas et al. 2021).

Direct human intervention is a defining feature of most urban wetlands. Remnant, rehabilitated, and constructed wetlands may be subject to frequent and intensive management including imposed water-level regimes and invasive species control (Sinclair et al. 2020). In urban wetlands, current interventions, landscape drivers, and legacies of past use and impacts will shape salinization regimes and biogeochemical nitrogen (N) and phosphorus (P) cycling. While salinization-impacted biogeochemical processes are at play in salinizing wetlands regardless of salt source and specific land use, we argue that the impact of salinization on urban wetland biogeochemistry merits special consideration.

Understudied urban wetland biogeochemistry and salinization

Urban wetlands are generally understudied and often overlooked (Palta and Stander 2020). Few, if any, urban freshwater wetlands have been monitored at temporal resolutions (i.e., decadal scales) matching well-established long-term monitoring of urban streams, rivers, and lakes, in part because many urban wetlands are recently built or formed novel ecosystems (Palta et al. 2017; Clifford and Heffernan 2018; Maas et al. 2021). This lack of long-term data limits our ability to observe changing salt concentrations and leads us to rely on long-term monitoring of other aquatic ecosystems combined with shorter-term case studies to assess salinization in urban wetlands.

Excess salt drastically changes the geochemistry and biology of remnant wetlands (Wilcox 1986; Pugh et al. 1996; Rhodes and Guswa 2016) and shapes the structure and function of intentionally created urban wetlands (Van Meter et al. 2011; Hill and Sadowski 2016; Lam et al. 2020). The symptoms of FSS in wetlands are similar to those of lakes, streams, and rivers. Most freshwater wetlands are characterized by specific conductance values less than about 1000 μS cm^{-1} and chloride (Cl^-) concentrations less than 100 mg L^{-1} (Herbert et al. 2015), but salinizing urban wetlands commonly exhibit specific conductance values up to and over 25,000 μS cm^{-1} and Cl^- concentrations in the thousands of mg L^{-1} (Sanzo and Hecnar 2006; Van Meter et al. 2011; Hill and Sadowski 2016). Acutely salinized wetlands, such as urban stormwater ponds designed to intercept stormwater runoff, can contain salt concentrations exceeding those of ocean water, particularly in the deep waters with density stratification (Lam et al. 2020). Salinization symptoms are detected in urban wetland soils, which have higher pH and electrical conductivity than nonurban forested wetlands (Larson et al. 2016) and contain plant communities characterized by halophytic vegetation (Skultety and Matthews 2017, 2018).

Diverse salt sources

Salinizing urban wetlands receive ions from diverse anthropogenic sources (Supporting Information Table S1) (Griffith 2017; Overbo et al. 2021), leading to complex salinization regimes characterized by both elevated salt concentrations and a variable suite of ionic compositions (Fig. 1). De-icing salts (mainly NaCl but including $CaCl_2$, $MgCl_2$, and brine products) (Wilcox 1986; Pugh et al. 1996; Foos 2003; Hill and Sadowski 2016; Rhodes and Guswa 2016) and coastal saltwater intrusion (Tully et al. 2019) are perhaps the most conspicuous salinization drivers. However, heterogeneous pathways can deliver myriad salts to wetlands. Pervasive salinization occurs even in warmer regions that do not generally require de-icing, including the southern United States (Texas) and in the tropics (Puerto Rico) (Steele and Aitkenhead-Peterson 2011; Potter et al. 2014). Wastewater is a substantial salt source (Overbo et al. 2021), whether directly from treatment plant effluent (Steele and Aitkenhead-Peterson 2011; Kerr 2017; Liang et al. 2017), leaky wastewater infrastructure and septic systems (Potter et al. 2014; Hill and Sadowski 2016; Hoghooghi et al. 2016), or treated and reclaimed wastewater used for irrigation (Toor and Lusk 2011; US EPA 2012). The weathering of cement and rock-based infrastructure and anthropogenic "technosol" soils generates Ca^{2+}, Mg^{2+}, SO_4^{2-}, HCO_3^- ions (Davies et al. 2010; Séré et al. 2010; Chambers et al. 2016) because raw materials mined for use in construction and then incorporated as artifacts in urban soils include calcite, gypsum, dolomite, and other calcareous minerals and rocks (Lehmann and Stahr 2007; Chambers et al. 2016). Salts are one component of novel anthropogenic "chemical cocktails" (Kaushal et al. 2019) which also include elevated nutrients (Hobbie et al. 2017; Yang and Toor 2017; Iverson et al. 2018), metals (Davis et al. 2001; Schuler and Relyea 2018), and synthetic chemicals (Bernhardt et al. 2017), all of which interact with one another, resulting in uncertain consequences for nutrient removal functions. The diverse salt sources to urban wetlands result in complex and unpredictable salinization scenarios, which are largely shaped by the identity and nature of hydrologic inputs to wetlands.

Urban wetland hydrology

The hydrologic structure, connectivity, and watershed location of wetlands determine salt sources and transport, shaping salinization regimes (Figs. 1, 2). Within wetlands, water-level fluctuations and flow regimes shape the mixing, concentrations, and processing of salt ions (Fig. 2). Hydrology also drives within-wetland N and P biogeochemical processes (Reddy and DeLaune 2008; Mitsch and Gosselink 2015). Ultimately, wetland hydrologic regime (including human management decisions) determines the timing, magnitude, and ionic composition of both salt and nutrient exports. In cities, artificial drainage networks, impervious surfaces, engineered water systems, and microclimatic effects of urban development drastically alter every component of the hydrologic cycle at both local and catchment scales (McGrane 2016). Many features of urban hydrology, particularly in humid climates, enhance the temporal variability of salt loading, concentrations, and ionic composition.

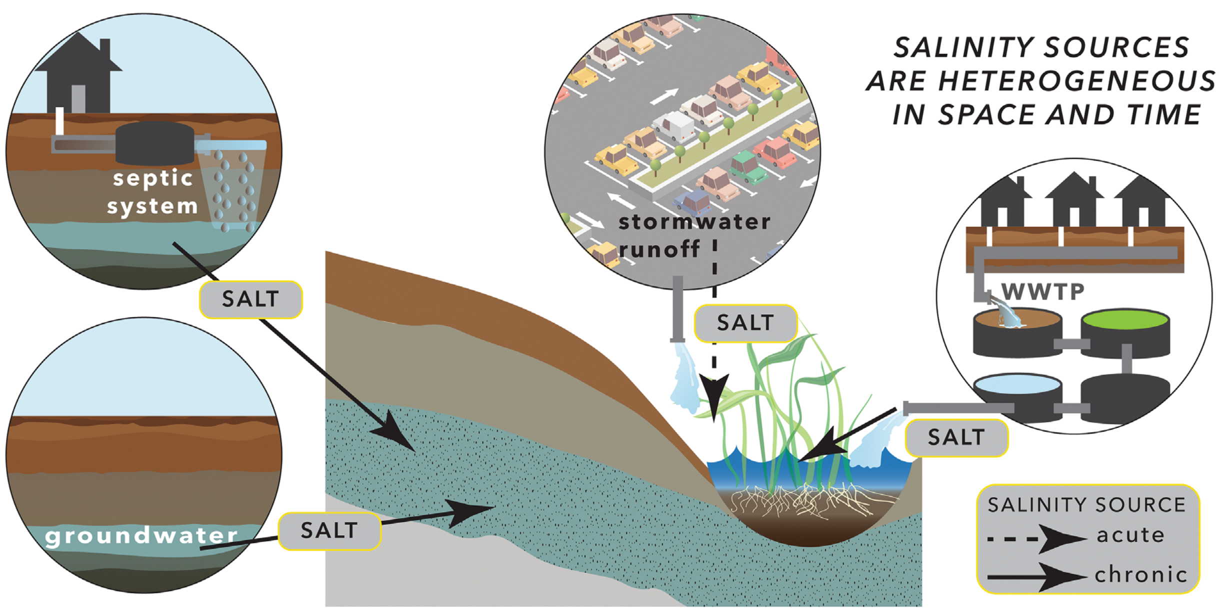

Fig. 1. Diverse salt sources. Inland urban freshwater wetlands experience salinization from diverse sources (e.g., septic, groundwater, wastewater, stormwater) resulting in complex and unpredictable salinization regimes, as characterized by the intensity and timing of acute (dashed arrow) and chronic (solid arrow) salt loads as well as the altered ionic composition imposed by mixtures of salts from a variety of sources.

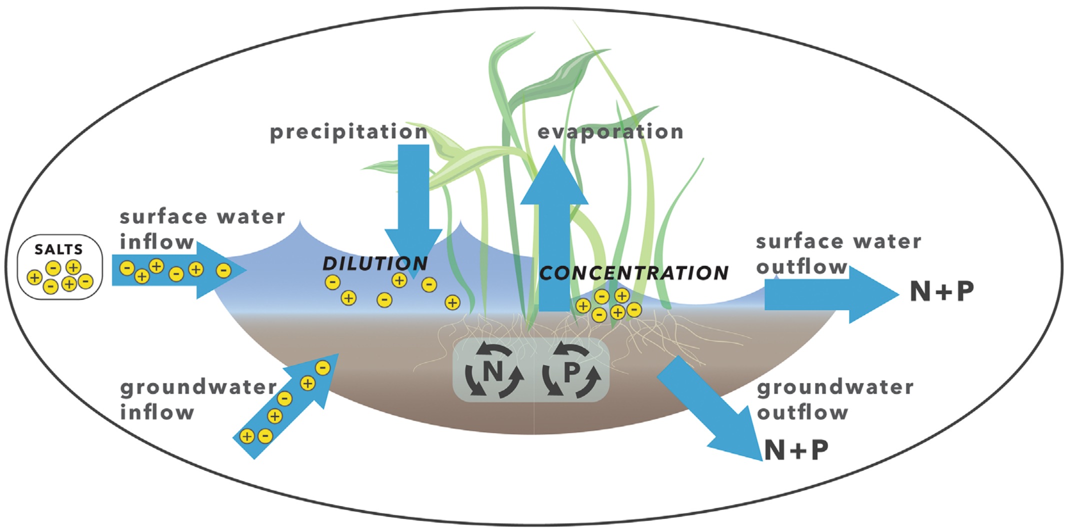

Fig. 2. Within wetland systems, hydrology shapes both salinization regime and nutrient inputs as well as internal system processing. Wetland geomorphology interacts with hydrology to shape patterns of salt dilution and concentration, and biogeochemical processing within wetlands influences salt and is influenced by salt. Salt concentrations and ionic compositions interact with wetland sediment nutrient biogeochemistry to influence within wetland nutrient availability and salt and nutrient exports.

Runoff and baseflow

In mesic and humid climates, highly connected impervious surfaces and artificial drainage limit soil infiltration, increasing surface runoff and peak flows (Paul and Meyer 2001), which creates flashier conditions in wetlands that receive surface water runoff, piped inflows and/or contributions from urban streams (O'Driscoll et al. 2010). When runoff, pipes, and streams carry anthropogenic salts, this hydrologic flashiness can coincide with "ionic flashiness" (Blaszczak et al. 2019), causing not only elevated salt concentrations but greater variability in concentrations and ionic composition. Despite the common story that urbanization leads to increased flashiness, more recent work in arid regions has revealed climate and context dependence (Hale et al. 2016; McPhillips et al. 2019; Li et al. 2020). In mesic and humid climates like the eastern United States, urban wetlands tend to be drier than their less urban counterparts (Ehrenfeld et al. 2003) and experience flashier inputs (Smith and Smith 2015). In contrast, urban aquatic systems in arid regions like Phoenix, Arizona (USA) are less flashy and less variable than their nonurban counterparts due to the buffering of stormwater detention, engineered drainage systems, and leaky infrastructure (McPhillips et al. 2019). These drivers may cause salinizing wetlands in arid regions to hold water more frequently than they might have otherwise, and the nature of this "urban baseflow" will influence salinization regimes (Bonneau et al. 2018; Kaushal et al. 2021).

Groundwater–surface water interactions

Natural and altered interactions between groundwater and surface water at catchment and wetland scales influence freshwater wetland salinization both through subsurface salt, and through effects on hydrology. Impervious surfaces, sealed soils, and piped artificial drainage generally decrease infiltration and groundwater recharge (Price 2011; McGrane 2016), although engineered infrastructure, particularly aging and leaking infrastructure, can interact with subsurface flow paths (O'Driscoll et al. 2010; Kaushal and Belt 2012; McGrane 2016). In arid regions, the widespread use of stormwater retention basins and other infiltration features may enhance catchment-scale groundwater recharge (McPhillips et al. 2019). In remnant wetlands and constructed features that promote infiltration, urban wetlands can be important sites for groundwater recharge (McLaughlin and Cohen 2013), but may contribute to subsurface groundwater salinization if these wetlands are accumulating salt from upstream sources including stormwater runoff (Ostendorf et al. 2009; Snodgrass et al. 2017). In some constructed, impacted, and natural wetland systems, infiltration is intentionally or unintentionally prevented by installed linings, compacted soils from construction activities, or naturally impermeable substrates like clay layers. In these wetlands with minimal or no groundwater exchange and infiltration, accumulated salt is exported downstream during overflow events (Barbier et al. 2018). Small, hydrologically isolated urban wetlands with impermeable layers (Barbier et al. 2018; Maas et al. 2021) have a high propensity for drought-induced water-level drawdowns and associated evaporative concentration of salt ions (Nielsen and Brock 2009; Steele and Heffernan 2014; Siddig et al. 2020).

Precipitation and evapotranspiration

Urban environments shape microclimates, including urban heat island effects, which alters precipitation and evaporation regimes. Although an active area of research, urbanization generally seems to enhance the amount and intensity of local precipitation (McGrane 2016). Depending on antecedent conditions, precipitation events may mobilize and transport salt loads to wetlands, contributing to long-term salt accumulation, or may temporarily dilute salt in places and at times where and when salt inputs are low (Barbier et al. 2018). Replacing vegetated land with impervious surfaces, sealed soils, and artificial drainage networks may decrease water losses through evapotranspiration at catchment scales (Rose and Peters 2001; McGrane 2016; Li et al. 2020), contributing to increased storm flows, stream flows, and flashiness. However, within open, vegetated urban wetlands and stormwater features designed to enhance evapotranspiration, evapotranspiration may enhance water losses, contribute to lower water levels, and create longer dry periods (McLaughlin and Cohen 2013). As climate change continues to intensify both precipitation events and droughts, these effects may be compounded in urban environments where microclimate and vegetation changes are acute. Increasingly variable hydrologic regimes from the combined effects of climate change and expanding urbanization will coincide with increasingly variable salinization regimes and less predictable impacts to the many interacting processes that create net ecosystem nutrient removal function.

Geomorphology: Urban wetland size, shape, and structure

The shape, size, and structure of urban aquatic ecosystems, including wetlands, are different from those in undeveloped and nonurban land uses (Steele and Heffernan 2014). Unique urban wetland shapes may influence the nature of, and biogeochemical responses to, wetland salinization. Wetlands in urban areas are often more fragmented and smaller, on average, than in nonurban areas (Steele and Heffernan 2014; Van Meter and Basu 2015), but with increased perimeter to area ratios (Aguilera et al. 2020). Higher perimeter to area ratios are indicative of greater connectivity of upland and wetland areas, and of a larger shallow portion of the wetland that supports the water-level fluctuations and wet–dry cycles; these in turn establish redox regimes that shift between anoxic and oxic conditions (Hefting et al. 2004; Van Meter and Basu 2015). Although smaller wetlands may be plagued by higher salt concentrations due to less dilution, the concomitant increase in perimeter to area ratio may facilitate higher biogeochemical processing rates related to wet–dry cycles, such as denitrification, in urban wetlands than in nonurban wetlands (Van Meter and Basu 2015; Cheng and Basu 2017). The relatively greater proportion of shallow areas in urban than nonurban wetlands may also amplify drying-and-rewetting event-driven mobilization of nutrients (Kinsman-Costello et al. 2016), accumulated salts, and oxidation products (e.g., nitrate, NO_3^-, and sulfate, SO_4^{2-}) from sediments. Little to no research exists that investigates potential tradeoffs between wetland size, perimeter complexity, salinization regime, and nutrient biogeochemistry. This information could be critical, however, in informing wetland design to minimize salinization impacts and maximize nutrient removal services.

Freshwater salinization syndrome signatures

The magnitude, variability, timing, and chemical composition of salinization across spatial and temporal scales are all ecologically relevant features of wetland freshwater salinization and can be collectively considered as a “FSS signature.” Wetland FSS signatures differ across ecosystems and among regions due to differences in salt sources, climate, geology, and anthropogenic setting (e.g., infrastructure age and type, wetland management regime). Wetlands receiving inputs from urban impervious surfaces and artificial drainage networks will experience not only higher average salt concentrations, but more variable salt concentrations and ionic compositions (Blaszczak et al. 2019) than less urban systems. The salinization regime of a “frontline” wetland directly receiving surface runoff from roads and parking lots will be distinctly different from that of a groundwater or river-fed wetland with a larger drainage area that may integrate salinization from multiple sources. Run-off fed wetlands with relatively small drainage areas may experience more temporally variable, ionically “flashy” loadings in contrast to wetlands with larger drainage areas integrating flow from multiple sources. Storm runoff directly entering wetlands will likely contain the full suite of cations and anions directly contributed from nearby salt sources like road salt (e.g., Na^+ and Cl^-), whereas systems that integrate multiple sources from longer flow paths may receive salt loads dominated by Cl^- and displaced soil cations like Ca^{2+}, as Na^+ cations may sorb to soils along flowpaths.

Many urban wetlands likely experience chronically elevated salt concentrations due to consistent input from stable sources like wastewater treatment effluent and from salinizing groundwater and subsurface input (Foos 2003; Ledford et al. 2016). Chronic, consistent sources may be punctuated by acute inputs of high salt concentrations during salt-mobilizing storm events, evaporative concentration due to drought (Siddig et al. 2020), and/or ice formation concentration in shallow wetlands (Dugan et al. 2017b). Conversely, during times and in places where storm events do not mobilize salts, systems with chronically elevated salinity may experience dilution pulses during storms, also contributing to the temporal heterogeneity in salt concentrations. Seasonal and interannual variability may alter wetland salt loads, processing, storage, and export (Rhodes and Guswa 2016), which will result in observed seasonal variability in ionic composition and concentrations in inflows, surface water, pore waters, and outflows.

Within wetlands, the persistence and magnitude of salt stress will vary along dominant flow paths, as wetland sediments near inflows will experience the most salt exposure, with exposure declining as salt interacts with and is stored in sediments as water moves through the wetland interior to the outflow. Plant and microbial community structure will reflect these within-wetland salinity gradients, such that communities nearest a major inflow may have lower biomass if new salt exposure harms vulnerable plants and/or more salt-tolerant species assemblages. The broad range of salinization regimes that urban wetlands likely experience provides opportunities to examine how salinization alters fundamental biogeochemical processes by investigating the full range of changes to salt concentration, variability, and ionic composition. In short, salinizing urban wetlands are ripe for mechanistically informative and management-relevant investigations into how freshwater salinization influences biogeochemistry.

Salinization effects on nutrient cycling

Salinization alters environmental chemistry through both the collective effects of increased concentrations and the specific effects of individual ions, particularly sodium (Na^+) and SO_4^{2-}. The interrelated chemical effects that accompany salinization (Kaushal et al. 2018a; Tully et al. 2019) include increased ionic strength, sodicity, alkalinization, and sulfidization, which in combination shape biogeochemical processes. Ionic strength measures the effect of electrical attractions and repulsions between all cations and anions in a solution (Stumm and Morgan 1996). Ionic composition influences ionic strength because divalent cations like Ca^{2+} contribute four times as much to ionic strength as monovalent ions like Na^+. Ionic strength establishes the osmotic conditions that organisms must cope with (Volkmar et al. 1998; Griffith 2017) and is one determinant of sorption–desorption equilibria and ion exchange between soils and solutions (Barrow et al. 1980a, 1980b; Seitzinger 1991; Rysgaard et al. 1999).

Sodicity measures the amount of Na^+ relative to other cations (Rengasamy and Olsson 1991; Wong et al. 2010; Steele and Aitkenhead-Peterson 2013). Salinity enhances aggregation of soil particles, whereas sodicity causes particle dispersion that ultimately reduces soil water infiltration capacity (Wong et al. 2010; Litalien and Zeeb 2020). Elevated soil Na^+ can cause the dispersion and transport of colloids and associated metals and nutrients (Norrström and Bergstedt 2001). Although the influence of sodicity has been thoroughly investigated in agricultural soils (Rengasamy and Olsson 1991; Sumner 1993), less work has investigated how sodicity influences flooded soils and sediments. Changes to soil physical properties due to salinization and sodicity may be particularly conspicuous in wetland soils that, by nature or design, experience prolonged dry periods without inundation. Elevated Na^+ can increase alkalinity and pH as ion exchange depletes soils of base cations (Norrström and Bergstedt 2001; Kaushal et al. 2013).

Alkalinization occurs as concentrations of salts with strong bases increase, especially from human-accelerated weathering of carbonate mineral-based materials like concrete (Kaushal et al. 2020), increasing the acid-neutralizing capacity of urban freshwaters. Alkalinization and pH influence biogeochemical processes through indirect and direct effects, including the toxicity of contaminants (Kinsman-Costello et al. 2015), adsorption processes and sorption–desorption equilibrium (Stumm and Morgan 1996; Gustafsson et al. 2012; Ranjbar and Jalali 2013), sediment–surface water nutrient exchange (Seitzinger 1991; Huang et al. 2005), and carbon cycling (Ardón et al. 2016). The effect of increasing alkalinity on phosphate sorption depends on soil composition (Huang et al. 2005). Higher pH and alkalinity diminishes the strength of phosphate sorption to iron (oxyhydr)oxides, but enhances co-precipitation with and sorption to calcium carbonate minerals (Huang et al. 2005). Among N cycling processes, nitrification is particularly sensitive to changes in alkalinity and pH. If carbonate alkalinity decreases, nitrification can stop completely, limiting the supply of NO_3^- (Biesterfeld et al. 2003). However, in most urban systems weathering of concrete is a source of carbonate and urban waters are more alkaline (Kaushal et al. 2020), which supports nitrification.

Sulfidization is the least considered impact of freshwater salinization in inland ecosystems due to a focus on NaCl and other major road salt ions, and the assumption that sulfur cycling is unimportant in traditionally low-SO_4^{2-}, noncoastal ecosystems (Duan and Kaushal 2015; Haq et al. 2018). We use "sulfidization" to describe when sulfide and other forms of sulfur accumulate in freshwater ecosystems, especially in anoxic soils and sediments, due to elevated SO_4^{2-} loads. Emerging evidence demonstrates that SO_4^{2-} concentrations are elevated in inland urban freshwater ecosystems along with other salt ions (Moore et al. 2017; Reisinger et al. 2019), although to a more variable and different degree than in coastal ecosystems experiencing marine salt intrusion (Tully et al. 2019). Although SO_4^{2-} makes up a relatively small proportion of the ionic strength of noncoastal salinizing solutions, it can have an inordinately influential biogeochemical effect when conditions in organic-rich, low-oxygen wetland sediments promote its reduction to sulfide (Baldwin and Mitchell 2012; Kinsman-Costello et al. 2015). Hydrogen sulfide is directly toxic to organisms (Kinsman-Costello et al. 2015) and inhibits microbial enzymes that mediate N transformations (Brunet and Garcia-Gil 1996) and is also geochemically reactive. Sulfide binds strongly with soil Fe and other metals, which then can diminish soil P storage by "stealing" iron oxide sorption capacity (Roden and Edmonds 1997).

Over time, sulfidization and cation exchange in chronically salinized systems create a cumulative geochemical "memory" of past salinization in wetland sediments (Johnson et al. 2019), which may have important implications for wetland biogeochemical functions when sediment conditions rapidly change, such as during and after extreme drying and reflooding. In this situation, oxidation of previously stored reduced sulfide when sediments are drying may lead to pulsed release of SO_4^{2-}, acid, and other oxidation products upon re-wetting (Kinsman-Costello et al. 2014, 2016). The combined chemical effects of freshwater salinization drastically alter not only the biotic structure of ecosystems through direct effects on organisms, but also N processing and P storage functions through both direct geochemical and indirect biological processes.

Knowledge gap: Biogeochemical freshwater salinization experiments inadequately reflect urban wetland salinization regimes

A robust body of evidence demonstrating the organismal and ecosystem-level impacts of freshwater salinization has emerged, but gaps limit our understanding of the biogeochemistry of salinization in urban wetlands. To demonstrate and confirm this knowledge gap, we reviewed studies that report the results of freshwater salinization experiments in aquatic or semi-aquatic soils and sediments in which salt concentrations were experimentally elevated in field or lab settings and indices of biogeochemical processes related to N and/or P availability and net export were measured. Although this review is not exhaustive, repeated literature searches with a variety of keyword combinations (including combinations of "freshwater salinization," "salinization," "wetlands," "soils," and "sediments") and citation tracking give us confidence that our assessment adequately reflects the state of the literature at the time of publication. Our primary focus is on N and P cycling, so we only include studies directly relevant to N and/or P cycling. We noted whether each study pertained to inland or coastal wetlands and the suite of salt ions used to experimentally elevate salt concentrations (i.e., marine salts, NaCl only, or other combinations, Supporting Information Table S2).

We identified 34 peer-reviewed papers that report the effects of soil and/or aquatic sediment salinization on N and/or P biogeochemistry. The studies tend to either examine the full suite of ions at oceanic ratios in the context of coastal systems, or focus only on NaCl as a reflection of road salt (Table 1; Supporting Information Table S1). Of these, most (24) pertain to coastal systems in the context of saltwater intrusion (Table 1). Experiments with marine salts use actual sea water or artificial sea water salt mixtures (Supporting Information Table S1) including Instant Ocean (Marton et al. 2012) and TropicMarin (van Dijk et al. 2015) with fixed ionic ratios reflective of oceanic water. Fewer studies (12) report biogeochemical salinization experiments in inland wetlands, where most salinization is ascribed to road salt and usually only NaCl is used as a salt source. Six of the studies from inland settings explicitly concern urban land use and/or impacts of roads (Norrström and Bergstedt 2001; Hale and Groffman 2006; Kim and Koretsky 2013; Duan and Kaushal 2015; Craig and Zhu 2018; Haq et al. 2018). In three studies, salt sources in addition to or distinct from NaCl are used, either in combinations or as individual treatments of alternate road salts like $MgCl_2$ and $CaCl_2$ (Beltman et al. 2000; Kim and Koretsky 2013; Craig and Zhu 2018). Only two studies of inland wetland salinization incorporated SO_4^{2-} in experimental salt treatments (Beltman et al. 2000; Donato et al. 2020). Neither sea salt nor NaCl alone adequately reflect the variable nature of inland urban freshwater wetland salinization regimes, and our ability to design and manage wetlands for nutrient removal services is limited by this knowledge gap.

Table 1. Quantities of published studies reporting the effects on experimental salinization on nitrogen and/or phosphorus biogeochemical processes in the context of freshwater coastal wetlands experiencing or vulnerable to sea level rise ("Coastal") or noncoastal, inland, often urban wetlands experiencing or vulnerable to freshwater salinization ("Inland"). In 34 peer-reviewed papers, one reported results of both Marine Suite and NaCl Only salts (Donato et al. 2020), thus 35 total studies are enumerated.

	Experimental salt ions			
Study system	Marine suite	NaCl only	Other	Total
Coastal	21	3	0	24
Inland	1	7	3	11

Biogeochemical effects of urban wetland freshwater salinization

To develop synthetic hypotheses that predict how N and P biogeochemical processes in wetland sediments are altered as urban wetlands salinize, we first review the existing literature on how salinization influences sediment N cycling processes, sediment P cycling processes, and biologically mediated processes that influence the cycling of those nutrients. Finally, we integrate these results with fundamental biogeochemical principles to create synthetic summary hypotheses.

Effects of salinization on sediment N cycling processes

Salinization alters ecosystem N flux rates and pool sizes (Zhou et al. 2017) through a variety of processes (Fig. 3). Microbially mediated redox transformations dominate wetland sediment N cycling, but other biotic and geochemical processes also contribute. Through ionic exchange processes, Na^+ and other salt cations can cause desorption of cationic ammonium (NH_4^+) from soils and sediments (Seitzinger 1991; Rysgaard et al. 1999). This desorption can enhance rates of sediment NH_4^+ release from both coastal and inland freshwater sediments (Gardner et al. 1991; Baldwin et al. 2006; Weston et al. 2006; Jun et al. 2013) and ultimately lead to net ecosystem release of reactive N (Ardón et al. 2013) (Fig. 3).

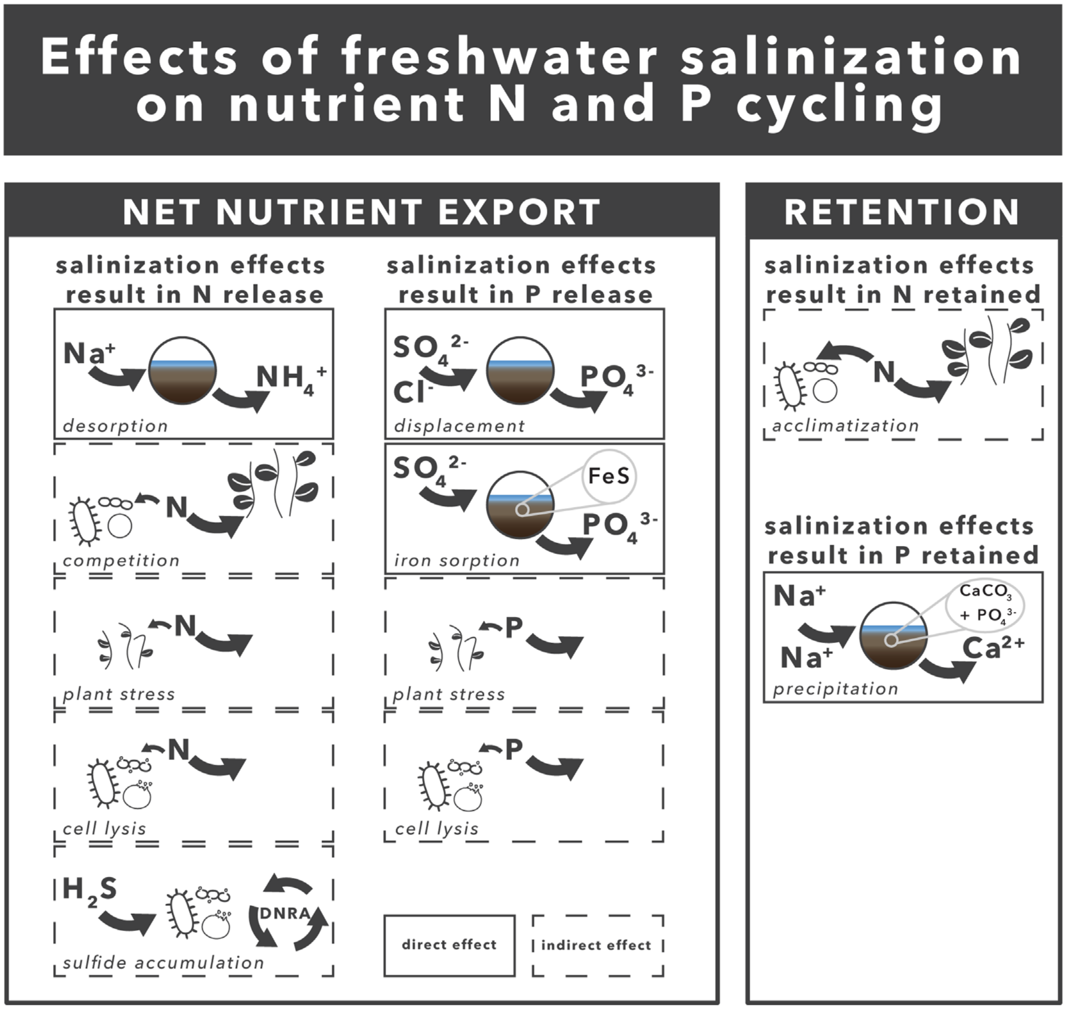

Fig. 3. Illustrations of the interacting direct and indirect ways by which freshwater salinization influences nitrogen (N) and phosphorus (P) cycling processes in wetland sediments. Of the identified processes, the majority (9 of 11) are predicted to lead to net higher nutrient availability and export, whereas only two processes are predicted to lead to improved wetland nutrient retention.

Salt-driven NH_4^+ desorption may change ecosystem N removal rates by altering the outcome of plant-microbe competition for N. Plants may deplete inorganic soil N through uptake of salt-displaced NH_4^+ (Boudsocq et al. 2012) before the NH_4^+ can be microbially oxidized via nitrification to NO_3^-. When plants outcompete microbes for NH_4^+ in this way, nitrification and coupled nitrification–denitrification are limited, ultimately diminishing permanent N removal (Fig. 3). Although plants remove reactive N and store organic N in their tissue, organic N is remineralized to NH_4^+ upon plant senescence, and thus plant storage does not permanently remove N from ecosystems (Lee et al. 2009).

Although some effects of salinization on N cycling processes, like NH_4^+ release, may occur soon after the addition of salt water, a shift in dominant pathways may take longer after sediment properties change and microbial communities acclimate (Neubauer et al. 2019). Wetland sediments typically support high rates of permanent removal of reactive N (NO_3^- and NH_4^+) via the anaerobic microbial process of denitrification and more rarely, anaerobic NH_4^+ oxidation (annamox) (Lee et al. 2009). The direct effects of salinization on microbially mediated N transformations depend on whether past salt exposure has assembled a salt-tolerant microbial community (Rath and Rousk 2015). In systems with a history of exposure (e.g., roadside soils), microbial communities are distinct from those in unsalinized systems (Lancaster et al. 2016), but often maintain N processing function. In urban and roadside systems with salinization histories of decades or more, salt additions often do not change indices of N cycling rates including denitrification enzyme activity (Hale and Groffman 2006), denitrification rates (Lancaster et al. 2016), net N mineralization, and nitrification (Craig and Zhu 2018). In salt-naive sediments, on the other hand, enhanced salinity can diminish denitrification (Seo et al. 2008; Lancaster et al. 2016), likely due to osmotic stress (Yan et al. 2015) and ion-specific toxicity (Macêdo et al. 2019) to microbes (Fig. 3).

Sulfidization may alter NO_3^- removal by promoting dissimilatory NO_3^- reduction to NH_4^+ (DNRA). In DNRA, NO_3^--N is transformed to NH_4^+ rather than N_2 gas, and therefore remains biochemically reactive in the system (An and Gardner 2002). A shift from denitrification to DNRA is observed along gradients from low to high salinity in estuaries (Giblin et al. 2010), presumably due to sulfide accumulation (Weston et al. 2006) which inhibits denitrification enzyme activity (Brunet and Garcia-Gil 1996). Some chemolithotrophic microbes directly link the sulfur and N cycles by oxidizing sulfide as they reduce NO_3^--N to NH_4^+ in DNRA (Brunet and Garcia-Gil 1996). While the importance of DNRA tends to increase with increasing salinity in estuarine ecosystems, denitrification can still dominate N cycling in highly saline environments (Smyth et al. 2013). Free sulfide alters the composition of N-transforming microbial communities (Brunet and Garcia-Gil 1996; Murphy et al. 2020) and also inhibits nitrification (Joye and Hollibaugh 1995), limiting NO_3^- supply. Toxic sulfide sometimes decreases plant productivity (Kinsman-Costello et al. 2015), which may indirectly influence the balance of N-removal pathways by altering the quality and quantity of organic carbon inputs relative to NO_3^-. Generally, at low C : NO_3^- ratios (which may emerge as plant productivity declines due to sulfide toxicity), denitrification dominates, while at high C : NO_3^- ratios DNRA tends to dominate (Tiedje 1988; Hardison et al. 2015).

Effects of salinization on sediment P cycling processes

Wetland sediments store P through sedimentation and burial of P-containing particles (Noe et al. 2019). A proportionally small, but ecologically critical, fraction of the inorganic phosphate (PO_4^{3-}) ions interact with soil minerals through dynamic geochemical associations, in particular sorption to iron oxides and co-precipitation with calcium carbonates (Noe et al. 2019). Changes in soil chemistry alter interactions between PO_4^{3-} and sediments, enhancing or diminishing the bioavailability and transport potential of sediment-stored P.

Freshwater sediments tend to have greater PO_4^{3-} sorption capacity and lower pore water PO_4^{3-} concentrations than brackish and saline sediments (Sundareshwar and Morris 1999; Jordan et al. 2008), in part because freshwater sediments tend to contain more sorptive metal oxides, but also because of how ionic strength and pH influence sorption (Barrow et al. 1980a, 1980b; Stumm and Morgan 1996). Salt anions like Cl^- and SO_4^{2-} compete with anionic PO_4^{3-} for sorption sites (Fig. 3). At the higher pH of saline environments, the net surface charge of iron oxy(hydr)oxides tends to be more negative, which electrostatically discourages PO_4^{3-} anion sorption (Stumm and Morgan 1996). Thus, the increases in pH and alkalinity associated with the FSS may also contribute to lower sediment PO_4^{3-} sorption capacities. When salinization includes SO_4^{2-} and drives sulfidization, sulfide and Fe interact strongly, diminishing sediment Fe binding capacity for PO_4^{3-} (Roden and Edmonds 1997) and leading to less sediment P storage and/or release of previously stored P from sediments (Lamers et al. 2002; Weston et al. 2006) (Fig. 3). In combination, the geochemical effects of salinization should decrease wetland sediment PO_4^{3-} sorption capacity.

Despite predictions that the increased ionic strength associated with salinization should weaken PO_4^{3-} sorption, experiments have yielded conflicting results. Some find that greater salinity accompanies greater P availability and less PO_4^{3-} sorption than expected (Beltman et al. 2000; Weston et al. 2010; Williams et al. 2014; Herbert et al. 2015; Haq et al. 2018; Steinmuller and Chambers 2018) while others find that salinity causes lower P availability and less release of P from sediments (Baldwin et al. 2006; Jun et al. 2013; van Diggelen et al. 2014; van Dijk et al. 2015, 2019). Although increases in ionic strength may diminish PO_4^{3-} sorption, concomitant changes also influence sediment P retention. Specifically, Na^+-driven displacement of soil Ca^{2+} (Norrström and Bergstedt 2001) along with increasing pH may enhance complexation of PO_4^{3-} with calcium carbonate precipitates, resulting in lower P availability and higher P retention (Fig. 3) (Jun et al. 2013; van Dijk et al. 2015, 2019). The net effect of salinization on geochemical P retention will depend not only on the degree of salinization and the ionic composition of salinizing water (especially the relative importance of SO_4^{2-}), but also on sediment characteristics (e.g., mineral content and sorption properties) and P loads, as PO_4^{3-} sorption is an equilibrium process.

Indirect effects on N and P cycling: Community and organismal impacts

Microbes

Salinization influences microbial physiology, biological processes, community composition, and diversity through osmotic stress and ion-specific toxicity, but the resulting effects on biogeochemistry depend on whether microbes acclimate to saltier conditions. Rapid and acute osmotic stress, as when rainwater rapidly dilutes salinized soil water, can cause microbial cell lysis and sudden release of stored inorganic and labile organic forms of N and P (Turner and Haygarth 2001; Schimel et al. 2007; Wood 2015) (Fig. 3). Sudden pulses of salt shift community structure and alter biogeochemical functions more than long-term salinity changes (Chambers et al. 2013; Mansour et al. 2018; Steinmuller and Chambers 2018) (Fig. 3). When long-term salinity change results in stable, salt-tolerant microbial communities, microbially mediated biogeochemical functions like denitrification may continue at rates similar to pre- or un-salinized sediments (Allison and Martiny 2008; Graham et al. 2016).

Plants

Salinization effects on plant communities cascade to biogeochemical implications. Effects on plants include osmotic stress, ion toxicity, and nutrient deficiencies linked to the displacement and leaching of cations by Na^+ (Forman and Alexander 1998; Findlay and Kelly 2011; Tiwari and Rachlin 2018). At very high concentrations, as are often found within a few meters of roads, plants suffer direct injury (Lumis et al. 1976), sometimes creating "salt-burned" areas devoid of vegetation (Scott and Davison 1982). Further from roads, or under chronically salinized conditions, salinization alters plant species composition. Invasive, salinization-tolerant species, such as *Phragmites australis* and *Typha angustifolia*, can invade and expand (Wilcox 1986; Panno et al. 1999; Brisson et al. 2010). Maritime halophytes have invaded far inland along major roads in response to salinization (Scott and Davison 1982; Brauer and Geber 2002; Fekete et al. 2018; Skultety and Matthews 2018), forming novel salt-tolerant plant communities in roadside wetlands (Skultety and Matthews 2018). Invasion by dense stands of clonal species such as *P. australis* may lead to increased plant biomass, sediment accumulation and nutrient uptake in a wetland, supporting greater plant nutrient uptake and reduced leaching.

Salinization-driven changes to plant communities indirectly affect nutrient cycling by interfering with plant carbon inputs to soils or via changes in plant nutrient uptake, resulting in altered nutrient storage in soils and plant biomass (Yuckin and Rooney 2019). Salinity limits plant biomass, even under favorable nutrient conditions (Smart and Barko 1980). Even halophytic plants adapted to salt marsh conditions grow better at lower salinities (Mendelssohn and Morris 2000). Decreased N and P storage due to diminished plant biomass may be the most important plant-mediated influence on N and P cycling and export from salinized wetlands. Following an acute salizination event, plant biomass should decrease, leading to decreased organic matter input to soil, decreased plant nutrient uptake, and increased leaching, leading to greater nutrient availability and/or export (Fig. 3). However, N content can be greater in some halophytic plants due to the use of nonprotein forms of N for osmotic balance (Hassall 2014), potentially counter-acting some of the lost nutrient storage due to lost plant biomass. A meta-analysis of 33 experimental salinization observations in coastal wetlands found an 18% increase in plant biomass N content (Zhou et al. 2017). Ultimately, the combination of changes to total plant biomass as well as changes in plant tissue-specific nutrient content will contribute to net changes in ecosystem nutrient removal function.

Animals

Salinization has both direct and indirect effects on aquatic fauna, which in turn mediate nutrient cycling (Covich et al. 1999; Vanni 2002). High salt can be toxic to animals, but sensitivity is highly species-specific (Griffith 2017; Hintz and Relyea 2019). Although concentrations high enough to cause direct mortality appear to be rare under field conditions (Blasius and Merritt 2002; Findlay and Kelly 2011), salinization causes changes in species composition and trophic interactions that sometimes decrease diversity (Petranka and Doyle 2010; Morgan et al. 2012; Hintz et al. 2017). As ion concentrations increase beyond the isotonic point of an organism, the organism must expend additional energy for osmoregulation (Griffith 2017), thus decreasing energy available for other functions such as feeding and reproduction (Venâncio et al. 2018; Entrekin et al. 2019), leading to nonlethal effects (Entrekin et al. 2019). For example, a decrease in plant productivity and the deposition of salt-enriched leaves might decrease detritivore consumption and productivity (Entrekin et al. 2019), slowing nutrient mineralization rates. Animals also mediate biogeochemistry through their influence on physical, chemical, and structural ecosystem features. For example, bioturbators burrow in sediments, enhancing water movement across the sediment water interface through active and passive bioirrigation and altering sediment–surface water nutrient exchange (Mermillod-Blondin and Rosenberg 2006; Meysman et al. 2006). The effects of salinization on bioturbating ecosystem engineers remain largely unexplored. Despite demonstrated impacts of salinization on animal species, communities, and trophic interactions, the potential indirect impacts on wetland nutrient cycling, mediated through impacts on biogeochemically relevant animals and their actions, remains unexplored.

Summary: Synthetic hypotheses

Our literature review supports a hypothesis that collectively, the interacting effects of elevated ionic strength and sulfidization *diminish nutrient removal capacity in urban wetlands*, causing wetlands to be less effective at removing polluting N and P and, in some cases, leading to wetlands functioning as a source, rather than a sink, for N and P.

Increasing salt concentrations

Increased ionic strength impacts N and P removal through chemical and microbial processes and organismal salt stress. Conspicuous changes to P cycling occur due to direct impacts on geochemical processes, whereas changes to N cycling primarily occur through indirect impacts on microbial and plant-mediated processes. Elevated ionic strength diminishes sediment sorption capacity for NH_4^+ and PO_4^{3-}, leading to net nutrient release from sediments into surface waters, particularly under anoxic conditions and in response to transient high-ionic strength incidents. Osmotic stress and toxicity induced declines in plant biomass and community compositional changes indirectly influence ecosystem N and P cycling by altering assimilatory and dissimilatory uptake pathways, altering N and P storage within plant biomass, and changing mineralization rates. The enhanced variability in salt concentration and ionic strength experienced by many urban wetlands may exacerbate the effects of osmotic stress on wetland communities, constraining community composition to organisms that are tolerant of wide salinity ranges and variability.

Sulfidization

In freshwater wetlands, SO_4^{2-} is a biogeochemical "keystone ion." When elevated salts include high SO_4^{2-} concentrations, as they often do, sediment SO_4^{2-} reduction can lead to multiple cascading biogeochemical effects, the balance of which will depend on sediment biogeochemical characteristics (e.g., iron content), but which ultimately diminish nutrient removal capacity. Sediment–surface water PO_4^{3-} exchange rates are shaped by chronic salinization to the extent that sediment has been sulfidized and iron oxide sorption sites have been replaced by FeS. In low-iron sediments where available sulfide accumulates in excess of iron, sulfidization may also shift dominant microbial N-processing pathways away from denitrification to DNRA, resulting in less net removal of N as NO_3^- is converted to NH_4^+ and retained in the system or exported rather than denitrified to nitrogenous gases.

Research needs

Directly assessing the influence of salinization on wetland biogeochemistry and testing the above hypotheses will improve our ability to protect, restore, construct, and manage wetlands for nutrient removal, particularly in urban settings. The current general lack of comprehensive, multisite urban wetland investigations is severely limiting. Many individual wetland case studies demonstrate the potential effectiveness, but also variability, of urban wetland nutrient load reduction (Brown 1984; Wadzuk et al. 2010). Fewer broad-scale studies similar to those that monitor urban streams and rivers exist to inform urban wetland management. Deliberate, holistic monitoring of salinizing urban wetlands representing diverse climate regimes, anthropogenic settings, legacies, and management strategies will contribute to not only filling the specific knowledge gap reviewed here, but a general deficit in our understanding of these societally important yet persistently understudied ecosystems. In short, not only are we uncertain of the ultimate outcomes of salinization impacts to urban freshwater wetlands, but we also know little of how fundamental biogeochemical mechanisms play out in novel urban wetland ecosystems.

Ecosystem field studies: Comprehensively assessing urban wetland systems

Wetlands are integrated systems, and thus comprehensive monitoring of hydrology, water quality, soils, plant, and microbial community dynamics is required to understand how salt may be impacting the ability of wetlands to remove nutrients. Measuring loads, interior pools, and export rates of major cations, anions, and nutrient forms will support the characterization of the diverse FSS signatures that wetlands experience, and will indicate how salt loads and nutrient cycling processes interact. Monitoring across seasons and years over a variety of hydrologic conditions (i.e., baseflow, drought, storm events, etc.) will reveal potential "hot moments" and control points in salt loading and biogeochemical impacts including acute salt pulses from snowmelt events or drought-induced evaporative concentration. Studying ecosystems receiving a range of hydrologic inputs and at contrasting watershed positions will support our understanding of how variable mixtures of salt sources and loading patterns set the stage of freshwater wetland salinization. Assessing diverse wetlands, including novel accidental and constructed wetlands along with extant relict and restored wetlands, will inform how land use history and human-engineered management interventions interact to shape the biogeochemical outcomes of salinization. And as is the case in not only urban areas, but ecosystem ecology in all settings, human dimensions of urban wetland functioning cannot be overlooked, particularly given the direct and prominent nature of management interventions in settings with human occupancy.

Lab and field salt manipulations: Environmentally relevant ionic compositions

To elucidate the biogeochemical mechanisms by which elevated salt ions change nutrient mass balances, experimental salt additions to sediment-surface water microcosms and/or mesocosms using treatments that reflect realistic signatures are necessary. Experimental salt additions that represent neither the fixed ionic ratios of marine salt solutions nor solely NaCl will pose more complex, but more realistic and mechanistically informative experimental results. These mechanistic studies can be designed to disentangle the complex biogeochemical web of processes that salt may influence by distinguishing direct physicochemical effects from indirect biologically mediated effects. In particular, the "keystone" role of SO_4^{2-} when present as an ion in salinizing wetlands must be directly assessed for its potential cascading biogeochemical impacts to both N and P cycles. Experiments designed to assess the influence of the greater variability in ionic strength often observed in wetlands by comparing pulsed and/or fluctuating salt loads to stable elevated loading will also inform mechanistic understanding of salinization impacts. Finally, to fully understand urban wetland biogeochemistry, explicit investigation of the interactions among elevated salt, nutrients, and other contaminants making up the "chemical cocktails" that impact urban ecosystems (Kaushal et al. 2018, 2019) will be necessary.

Conclusion: We should study freshwater salinization in urban wetlands

The inherent heterogeneity of urban wetlands and their settings present both challenges and opportunities to understand the mechanisms by which salinization alters sediment biogeochemistry and the outcomes of salinization effects across diverse regimes. While long-term water quality data from streams and rivers provide an integrative picture of the legacies and trajectories of freshwater salinization at watershed scales, investigating salinization regimes in wetlands can provide an invaluable window into ecosystem-level ecological and biogeochemical impacts. Wetlands are diverse in structure, connectivity, and landscape position, and thus can represent the full range of salinization regimes in terms of rates of change of concentration, and seasonal and event-driven variability, along with a range of changes in ionic composition, all of which may have variable impacts on biogeochemical processes. Together, this diversity of salinization regimes may provide a rich area of study to mechanistically understand how salt alters soil and sediment biogeochemical processes. Improved understanding of salinization-impacted nutrient cycling will support improved predictions of how salinizing aquatic ecosystems function, inform management to enhance desired ecosystem services, and may provide evidence to justify wetland protection and conservation.

Data Availability Statement

No new data were collected for this Current Evidence article, which synthesizes past literature. No data sets have been submitted to a data repository. All synthesized information can be found in the article itself, supplemental information associated with this manuscript, and in the references we cite.

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