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Assessing impacts of cemeteries on water quality in an urban headwater watershed with mixed human-built infrastructure

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Abstract

Cemeteries are understudied integral components to urban watersheds, which provide ecosystem services but can also export nutrients, trace elements, and other contaminants to nearby water bodies. In this study, we focus on Meadowbrook Creek, an urban headwater stream in Syracuse, New York (USA), which has shown significant nitrate contributions from a local cemetery. We collected biweekly surface water samples over the course of 1 year from 2022 to 2023 for analysis of major and trace elemental concentrations including Na, Ca, Mg, K, F, Cl, sulfate, and nitrate. Here, we aim to assess the impact of various human infrastructures on urban stream water quality with a particular focus on the cemetery and nitrate. A comparison between the new dataset in this study and previously reported water chemistry data in Meadowbrook in 2012 suggests a decade-long impact of road salting and the cemetery on water quality particularly with respect to Na, Cl, and nitrate. Sulfate, Mg, Ca, and K are likely mainly geogenic. Stable nitrogen isotope data, the usage of concrete or steel vaults in the cemetery in the past 50 years, and the lack of correlation between nitrate and fluoride concentrations in stream water argue against burial decay products being a major source of nitrate to the stream. Instead, other nitrate sources that exist in the cemetery such as, fertilizer, decaying plant material, and wastewater, are more viable dominant nitrate sources. In addition, nitrate loading calculations indicate that the groundwater-connected reach, including the cemetery, acts as an annual net sink for nitrate despite the seasonally varying sink-source patterns.

KEYWORDS

burial practices, cemetery, ecosystem services, nitrate, nutrient, road salt, urban hydrology, urban stream

1 | INTRODUCTION

Urban green spaces, including cemeteries, provide important ecosystem services. Among these urban green spaces, cemeteries may also act as a source of contamination in urban streams. Cemeteries can potentially release nutrients, trace elements, bacteria, viruses, pharmaceuticals, and other contaminants to groundwater and soil, and subsequently surface water, due to the possible leaching of burial products (e.g., Brennan et al., 2018; Lautz et al., 2020) in addition to other human activities within the cemetery, for example, fertilizer. Burial products include bodies and other materials buried with them, such as embalming material and caskets. However, contamination

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from cemeteries is under-explored, partially due to their cultural and religious significance.

Unlike natural streams, urban streams generally have highly compromised or no riparian zones compared with streams in unimpacted natural areas, which if present, would facilitate the uptake and degradation of some pollutants (Ledford et al., 2016; Meyer et al., 2005). The presence of excessive pollutants and the absence of riparian zones make urban streams particularly vulnerable to anthropogenic pollution, including nutrients, major ions, and trace elements (Kaushal et al., 2008). Increased nutrient (e.g., nitrate) and major ion (e.g., sulfate, chloride, and sodium) inputs to urban streams are commonly attributable to road salt, fertilizers, sewage wastewater, and atmospheric deposition (Cañedo-Argüelles et al., 2013; Divers et al., 2014). Nitrate is of particular concern for water management due to its role as a limiting nutrient in many ecosystems. Increased nitrate loads can lead to algae blooms, which can be toxic and reduce dissolved oxygen, straining aquatic habitats. Chloride, another analyte of concern, which is dominant in road salt, can pose harm to urban infrastructure, aquatic life, and human health when present in water at elevated levels (Pieper et al., 2018; US EPA, 2022). Urban green spaces, including cemeteries, can help alleviate some water quality issues associated with urban infrastructure by increasing groundwater infiltration and serving as a riparian buffer.

Only a limited number of previous studies assessed the environmental impacts of urban cemeteries on water quality. For example, Brennan et al. (2018) found higher nutrient and trace metal concentrations in groundwater directly downgradient a cemetery in Lansing, Michigan, (USA). In addition, Lautz et al. (2020) examined concentration change in major ions such as nitrate, sulfate, sodium, and chloride upstream and downstream of St. Mary's Catholic Cemetery along Meadowbrook Creek, an urban creek in Syracuse, New York (USA), over the course of 1 year from 2012 to 2013. High frequency nitrate loading upstream and downstream of the cemetery from 2017 to 2019 was also previously estimated (Beltran et al., 2021). Lautz et al. (2020) observed an increase of 20–40 kg NO_3^-/day in stream water flowing through the cemetery in the months of June–September, based on which Lautz et al. hypothesized that St. Mary's Catholic Cemetery served as a significant nitrate source to Meadowbrook Creek. Based on the number of annual burials in St. Mary's Cemetery and the amount of nitrate in human burials, Lautz et al. estimated that up to 25%–50% of such nitrate increase could plausibly derive from burial decay products. However, the release of burial decay leachate to the stream has not been confirmed. Leaky urban infrastructure and fertilizers were also suggested as additional sources of nitrate in the Meadowbrook watershed overall, and nitrate concentrations in Meadowbrook are also controlled by seasonal and in-stream processes such as plant uptake, denitrification, and organic matter decomposition (Lautz et al., 2020; Ledford et al., 2017).

In this study, we focus on Meadowbrook Creek to assess the environmental impact of various urban infrastructure and activities (e.g., St. Mary's Catholic Cemetery and road salting), and geogenic sources on urban stream water quality with a particular focus on the role of the cemetery on nitrate. Meadowbrook Creek was selected due to the availability of rich historic water quality data at the site. Here, we assess the decadal change and seasonality of solute leachate released from the cemetery and other sources. We also evaluate the control of hydrologic connectivity and processes on nitrate loading in an urban stream. We hypothesize that the cemetery continues to be a seasonally varying, important source of nitrate to Meadowbrook Creek with an increase in cemetery-originated nitrate loading in the last decade due to further decay and continual addition of burial products. We also hypothesize that sodium and chloride concentrations have increased due to the continued use of road salt within the watershed.

2 | STUDY SITE

Meadowbrook Creek is an urban headwater stream located in Syracuse, New York (USA). The stream is sourced from a stormwater pond and flows east before discharging into Butternut Creek in Dewitt, New York. The stream is approximately 5.5 km long with a drainage area of 11.3 km^2 (Figure 1). Flow is perennial and it does not freeze over. Land use in Meadowbrook Creek watershed is primarily residential (62% of the drainage area) with the rest consisting of schools/religious institutions (10%), vacant land (9%), and cemeteries (9%) (Lautz et al., 2020).

The stream is much more urbanized in the upstream reach than in the downstream reach. Based on Ledford and Lautz (2015), the upstream watershed has 30% low impervious surface cover (ISC), >50% moderate ISC and 9% high ISC. The downstream watershed has 46% low ISC, 49% moderate ISC and 4% high ISC. Low, moderate and high ISC correspond to <20%, 20%–49% and >50% ISC. Much of the stream flows through the middle of Meadowbrook Drive, a heavily trafficked residential road. The upper 3.5 km of the stream is situated in this heavily urbanized area and flows through an artificially straightened and concrete-lined channel, which will be referred to as the 'disconnected reach' following Ledford and Lautz (2015) due to its limited connection to groundwater. In addition, this upstream portion is characterized by the absence of a riparian zone or floodplain, increased impervious surface area relative to the downstream portion, numerous culverts, and low baseflows. The lower 2 km of the stream, once again outlined by Ledford and Lautz (2015), has less artificial alteration and will be referred to as the 'connected reach' due to the strong connection to groundwater, that is, groundwater and stream water are free to interact through permeable surfaces in the stream channel, also known as the hyporheic zone. The downstream groundwater-connected reach is historically a gaining stream in the summer (June–October) and a losing stream in the winter (December–April). May and November function as transition months, where groundwater inputs and stream outputs to groundwater are approximately equal (Beltran et al., 2021; Lautz et al., 2020; Slosson et al., 2021). Meadowbrook watershed is underlain by the Cobleskill and Akron Members, Bertie Group, Camillus Shale, and the Syracuse and Rondout Formation (Belak, 1980; Muller, 1964). These bedrock units contain dolomite, gypsum, celestite, and halite, acting as

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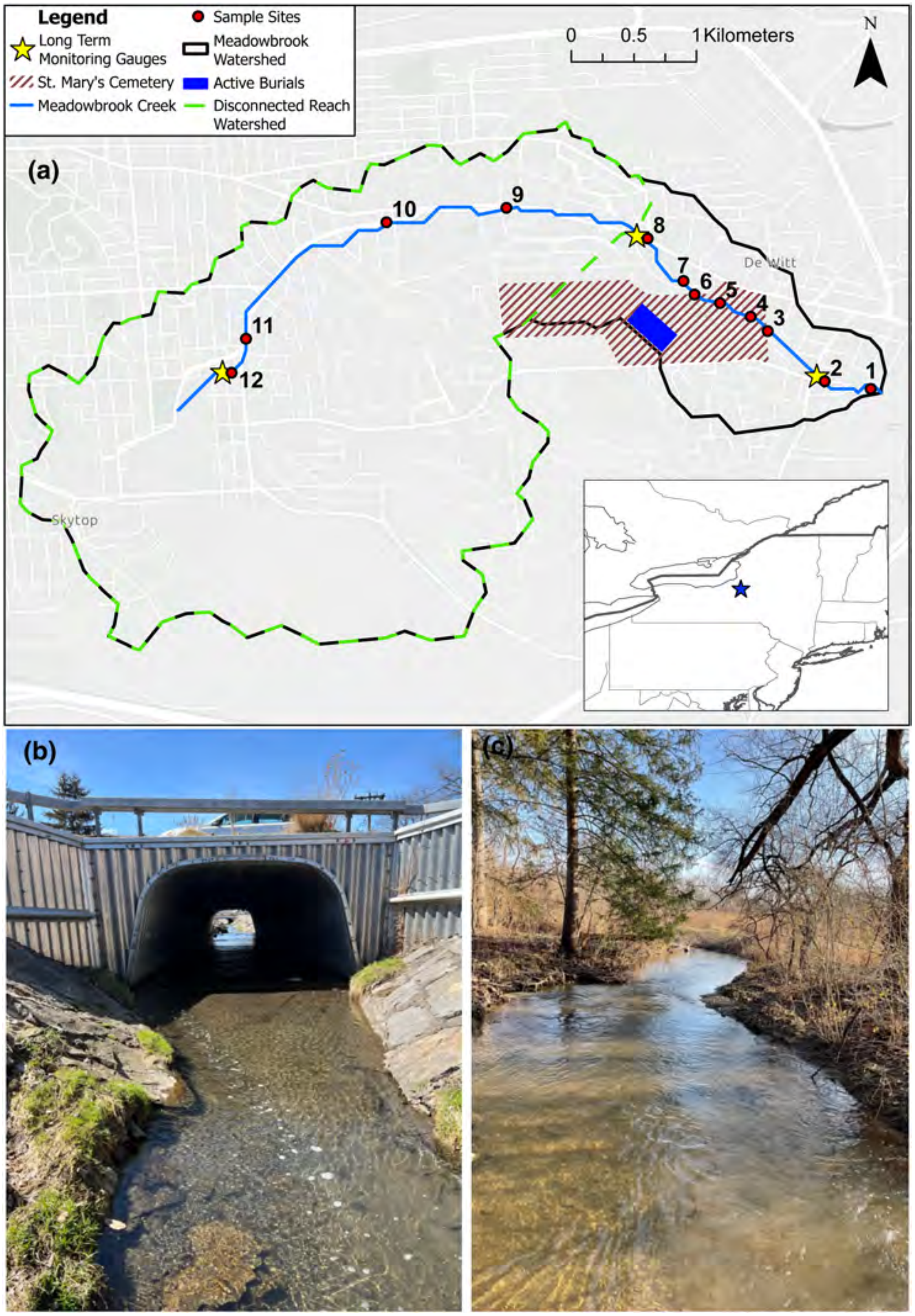

FIGURE 1 (a) The extent and relative position of Meadowbrook Creek watershed, cemetery boundaries, 12 sampling sites, long-term monitoring gauges, cemetery storm pond, and current active burial sites within the St. Mary's cemetery. (b, c) are field photos showing the typical environment and surroundings of the disconnected and connected reach, respectively.

geogenic sources of calcium, magnesium, sulfate, sodium, and chloride solutes in drainage waters of the basin.

St. Mary's Catholic Cemetery has an area of 0.85 km^2, through which over 500 m of Meadowbrook Creek flows. Slopes within the cemetery angle down towards Meadowbrook and range between 5% and 15%. Most burials within the last 10 years occur in sections outlined as 'Active Burials' (blue rectangle in Figure 1). The cemetery has been operational since the early 1900s and currently receives approximately 425 burials per year. Burials within the last 50 years at St. Mary's use concrete or steel vaults. Fertilizers are applied primarily at active burial sections 'a couple times per year', and elsewhere in the cemetery where needed. No irrigation is used and lawn clippings are not collected after mowing (T. Smith, personal communication, 9 May 2023).

3 | MATERIALS AND METHODS

3.1 | Stream water sampling and geochemical analysis

In this study, longitudinal synoptic stream sampling at 12 sites along the stream (Figure 1) was conducted approximately every 2 weeks for 1 year, starting in February 2022. Three sampling campaigns were

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TABLE 1 Site numbers, upstream distance from where Meadowbrook Creek flows into Butternut Creek, whether the stream channel is strongly connected to groundwater, and position of the site relative to St. Mary's Cemetery.

Site number	Distance upstream from the outlet (m)	Groundwater connection	Position relative to cemetery
1	0	Connected	Downstream
2	446	Connected	Downstream
3	973	Connected	Within
4	1100	Connected	Within
5	1262	Connected	Within
6	1504	Connected	Within
7	1647	Connected	Upstream
8	2064	Transition	Upstream
9	3645	Disconnected	Upstream
10	4208	Disconnected	Upstream
11	5034	Disconnected	Upstream
12	5564	Disconnected	Upstream

conducted during stormflows on 30 August 2022, 12 November 2022, and 20 January 2023. Stormflow collection occurred during, or immediately following (<6 h) significant rain events (>2 cm). Discharge values during these collection times were over an order of magnitude greater than pre-storm discharge values for the August and November storms, and four times higher than pre-storm values for the January storm. All sampling campaigns were completed within 2–3 h to ensure synoptic sampling. These 12 sampling sites were also sampled by Ledford and Lautz (2015), allowing the assessment of the decade-long impacts of the cemetery on surface water quality. Within the connected reach, two of the 12 sites are downstream of the cemetery, four sites within the cemetery and two are immediately upstream of the cemetery. The remaining four sites are further upstream in the disconnected reach (Table 1). At each site, two water samples were collected and filtered with 0.45 μm surfactant-free cellulose acetate membrane syringe filters. Samples were placed on ice until they were transported to the lab and refrigerated to preserve the water chemistry of the sample. One set of filtered samples was analyzed for anion concentrations including chloride (Cl), fluoride (F), nitrate, and sulfate. This analysis was conducted using a 930 Compact Flex Metrohm ion chromatographer at the Center for Environmental Systems Engineering Laboratories at Syracuse University. QA/QC in the ion chromatography was determined through continuing calibration verification (CCV) every 10 samples, with an accepted CCV value of 85%–115%. Samples on either side of a failing CCV were reanalyzed. The other set of filtered samples was acidified before being analyzed by a Perkin Elmer Avio 200 Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) at the Hydrogeochemistry and Environmental Data Sciences Laboratory at Syracuse University for dissolved sodium (Na), calcium (Ca), magnesium (Mg) and potassium (K) concentrations. Error in the ICP-OES result is determined as the relative standard deviation of three replicate measures, which vary from <1% to 10%. Below detection limit (BDL) measurements were reported at one-half the detection limit level for any statistical analyses. The summary of these geochemical results is listed in Table 2.

Following the year-long sampling campaigns, two more sets of water samples from downstream (sites 1, 2), within (sites 3, 4, 6), and upstream (sites 8, 12) of the cemetery were collected on 29 June 2023 and 3 July 2023 for nitrate isotopes using the denitrifier method (Weigand et al., 2016; Yu & Elliott, 2018) to further constrain the source of dissolved nitrate. Briefly, denitrifying bacteria lacking the nitrous oxide (N_2O) reductase enzyme (*Pseudomonas chlororaphos ssp. Aureofaciens*) are used to convert 20 nmol of NO_3^- into gaseous N_2O. The N_2O is then purified in a series of chemical traps, cryofocused, and finally analyzed for *m/z* 44, 45 and 46 on a continuous flow isotope ratio mass spectrometer (Elementar Isoprime precisION, Germany). International nitrate reference materials USGS34 and USGS35 and an internal working standard gravimetrically prepared from USGS32 and USGS34 were used to calibrate the isotope measurements. The analytical precision for $\delta^{15}N$ and $\delta^{18}O$ determinations are ±0.1‰ and ±0.3‰, respectively.

Two stream gauges measuring high-frequency (every 15 min) specific conductance, temperature, and water stage were installed along Meadowbrook in 2017 (Slosson et al., 2021) approximately 500 m upstream and downstream of the cemetery, respectively (Figure 1). Rating curves were established by Slosson et al. (2021) to convert water stage to discharge. These rating curves were not recalibrated in this study, which might introduce some errors. However, these monitoring sites have been frequently visited and maintained to clear the stream channel. Historical data were downloaded from the literature (Beltran, 2021; Ledford & Lautz, 2023).

The comparisons in median concentrations were based on Wilcoxon Rank-Sum tests at the significance level of $p = 0.05$.

3.2 | Stream nitrate load modeling

Nitrate load modeling was completed at the previously mentioned upstream and downstream gauging sites using the United States Geological Survey (USGS) LOADEST model (Runkel et al., 2004) in R via

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TABLE 2 Analytes examined and their method detection limits, number of samples ADL, minimum, maximum, median, and mean values, water quality guidelines, and water quality guideline type. Guideline types correspond with EPA Chronic Aquatic Life Standards (AL), New York State Aquatic Life (NYS AL), New York State Drinking Water (NYS DW) and EPA Drinking Water Secondary Guidelines (DW Secondary), (US EPA, 2018, 2022; Zambrano & Stoner, 1998). When calculating mean and median values, BDL measurements were replaced with half the detection limit. BDL represents below detection limit.

Analyte	Method detection limit (mg/L)	Number of samples	# of samples above detection limit	Minimum (mg/L)	Maximum (mg/L)	Median (mg/L)	Mean (mg/L)	Water quality guideline or standard (mg/L)	Water quality standard or guideline type
Calcium	0.0105	288	288	11.7874	404.3207	236.6042	230.8942	–	–
Chloride	0.0200	288	288	113.5078	843.4924	286.0777	331.3885	230.0000	AL
Fluoride	0.0080	288	286	BDL	1.8731	0.2413	0.3595	1.5000	NYS AL
Magnesium	0.0064	288	288	5.1006	60.4528	36.0656	35.8015	35.0000	NYS DW
Nitrate	0.3100	288	264	BDL	14.9007	2.2759	2.8447	10.0000	NYS DW
Potassium	0.0200	288	288	0.3435	9.5068	2.6601	2.6079	–	–
Sodium	0.0220	288	288	50.2073	469.5093	154.3634	180.2311	60.0000	DW Secondary
Sulfate	0.0870	288	288	128.5279	929.5845	482.1932	481.0655	250.0000	DW Secondary

TABLE 3 Optimal values for model selection criterion and coefficient values for the upstream and downstream LOADEST models.

Site	AIC	R^2	Bias percentage	a_0	a_1	a_2
Upstream	79.59	0.6848	23.25	4.2683	1.3828	−0.6137
Downstream	25.35	0.7307	−0.67	3.8951	0.6349	−0.286

Abbreviation: AIC, Akaike information criterion.

the package 'rloadest'. The natural logarithm of both discharge and concentration measurements were calculated and combined to develop a model that estimates nitrate loads based on high-resolution discharge data from 1 February 2022 to 31 January 2023 to encapsulate the entirety of the field sampling in this study.

This model estimates loads using the Adjusted Maximum Likelihood Estimation (AMLE) and can include a variety of variables including seasonality, temperature, and specific conductance, but primarily uses discharge. The load models chosen for these analyses were selected based on three factors: the Akaike Information Criterion (AIC) score, *r*-squared value, and bias percentage. Each of these values is shown in Table 3. The lower the AIC score and the higher the *r*-squared value, the better the model performance. Models with bias percentage above 25% or below −25% are generally not recommended.

For both sites selected for nitrate load estimation modeling (upstream and downstream of the cemetery), Model 2 (Runkel et al., 2004) reported the lowest AIC, highest *r*-squared, and lowest bias percentage. Model 2 is outlined by the formula:

$$\ln(\text{load}) = a_0 + a_1 \ln(Q) + a_2 \ln(Q)^2, \quad (1)$$

where a_0, a_1 and a_2 are coefficients to be optimized in the model fitting and development procedures. Optimal coefficients are shown in Table 3. Q is discharge corrected for collinearity by subtracting the center of ln(streamflow) from ln(streamflow). Models that accounted for other variables such as time, specific conductance, and temperature were developed and evaluated, but not used due to poorer performance. Model residuals were normally distributed and all coefficients were statistically significant at the confidence level of $p = 0.05$ for both models. All modeled nitrate concentrations for both upstream and downstream models were within the range of grab sample nitrate concentrations collected from the sites (BDL-8.29 mg/L for upstream and 0.68–6.52 mg/L for downstream). The storm events sampled occurred on the highest discharge days over the sampling period for both the downstream and upstream sites (12 November and 20 January, respectively). The difference in nitrate loads between sites upstream and downstream of the cemetery was calculated to determine estimated nitrate loading from the cemetery from 1 February 2022 to 31 January 2023.

4 | RESULTS AND DISCUSSION

4.1 | Varied surface water-groundwater interactions under baseflow condition

Meadowbrook discharge varied seasonally in 2022–2023. In summer (mid-May through mid-October), the connected reach between the two gauges acted as a gaining stream with the downstream gauge recording higher baseflows than the upstream gauge (Figure 2a), while the connected reach was a losing stream from mid-October through mid-May.

Such discharge patterns in the connected reach are likely regulated by precipitation, evapotranspiration (ET) and surface water-groundwater interaction through the hyporheic zone. The seasonality in discharge is hypothesized to be primarily driven by the variability in

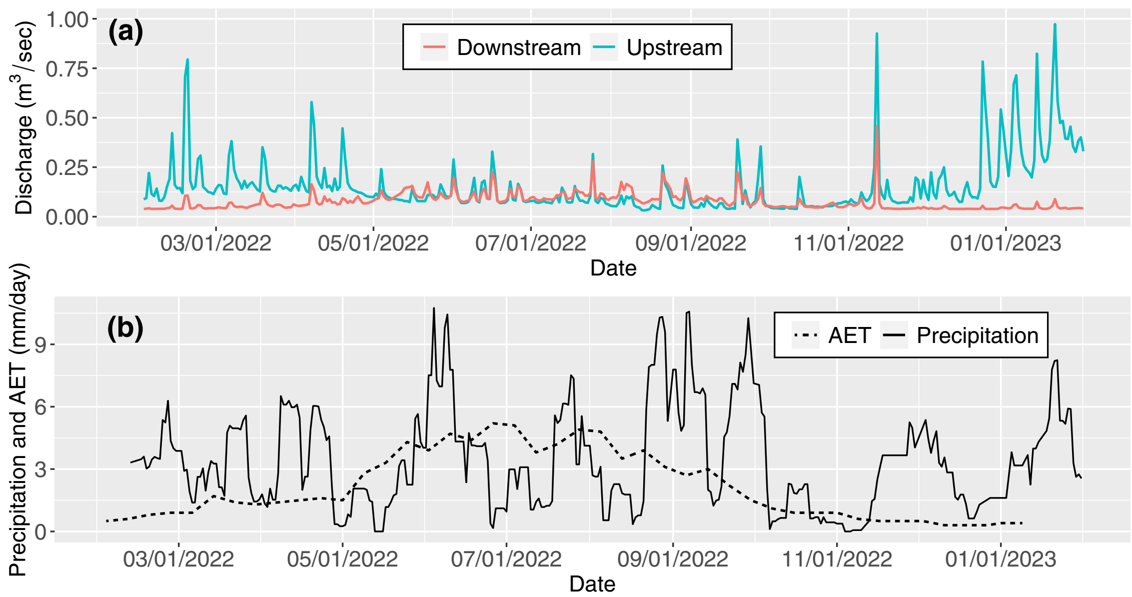

FIGURE 2 Daily averaged discharge values upstream and downstream of the cemetery reach (a) and daily actual evapotranspiration (ET) and eight-day moving average of precipitation (b). Actual ET data are sourced from eight-day satellite-based remote sensing data (Running et al., 2017). Precipitation data (Menne, Durre, Korzeniewski, et al., 2012; Menne, Durre, Vose, et al., 2012) is smoothed using an 8-day moving average to match.

precipitation and ET throughout the year. High ET in the summer (Figure 2b) likely depletes the storm pond (consistent with field observations) via evaporation and reduces the amount of water flowing into the pond, subsequently depleting the disconnected reach. Drier soils due to higher ET in the summer could also reduce flows as drier soils soak up more precipitation, resulting in less runoff to the channel. In the winter, lower ET rates lead to higher pond levels and thus higher discharge within the disconnected reach. Wetter (or frozen) soils in fall and winter would lead to more runoff to the channel. In the connected reach, the variability in the groundwater table and stream water level throughout the year can further lead to dynamic surface water-groundwater flow patterns. In particular, frozen ground and snow-dominated precipitation in the winter can reduce groundwater recharge, likely contributing to a lower water table, as suggested by Hyman-Rabeler and Loheide (2023) for other northern US states (i.e., Illinois, Michigan, and Wisconsin) in a climate zone similar to Syracuse. However, further research and data is warranted to test this hypothesis. Following snowmelt and the ground thawing, the water table receives more recharge and recovers, leading to more groundwater discharge to the stream in the connected reach.

4.2 | Sources of Na, Ca, Mg, K, F, Cl, and sulfate in Meadowbrook watershed where various human-built infrastructure overlaps

Number of samples (total and above detection limit) and basic statistics of each geochemical analyte are listed in Table 2 along with corresponding water quality standards. Water quality standards were based primarily on aquatic life standards. If no aquatic life standards were available, drinking water standards were listed. Nitrate concentrations, sources, and loads in Meadowbrook are discussed in depth in Section 4.3.

A Spearman correlation analysis was conducted to examine if any pairs of analytes share common sources (Figure 3). Spearman correlation analysis was chosen over Pearson correlation because Spearman analysis can handle censored data (e.g., BDL values). Dissolved sulfate in Meadowbrook is mainly derived from gypsum in the underlying bedrock units. K, Ca, and Mg concentrations all correlate with sulfate concentration suggesting a common geogenic source for these analytes. Unlike Mg and K, Ca and sulfate both exhibit a strong seasonality, which might reflect seasonally varied hydrological controls on the release and transport of different analytes into Meadowbrook Creek (Figure 4). Wilcoxon rank sum tests show that annual median values of Ca, sulfate, and K are statistically significantly higher in 2012 than in 2022 for the entire stream ($p < 0.01$, $W = 24\,044$, 29 281 and 20 883, respectively, and $n = 314$) (Figure 5). This might be due to the dilution effect as evidenced by the lower precipitation in 2012 (90 cm) than in 2022 (113 cm) (Menne, Durre, Korzeniewski, et al., 2012; Menne, Durre, Vose, et al., 2012). Concentration-discharge (C-Q) relationships were assessed at baseflow for the eight major water ions collected from 2017 to 2023 (Beltran et al., 2021 and this study) to further assess the solute source as well as the hydrological controls on solute release into Meadowbrook. Baseflow for data collected during 2022–2023 is defined as samples collected outside of the previously identified storm samples. Baseflow for data collected in 2017–2019 was determined as the lower 90th percentile as individual storm events were not identified in the data. This

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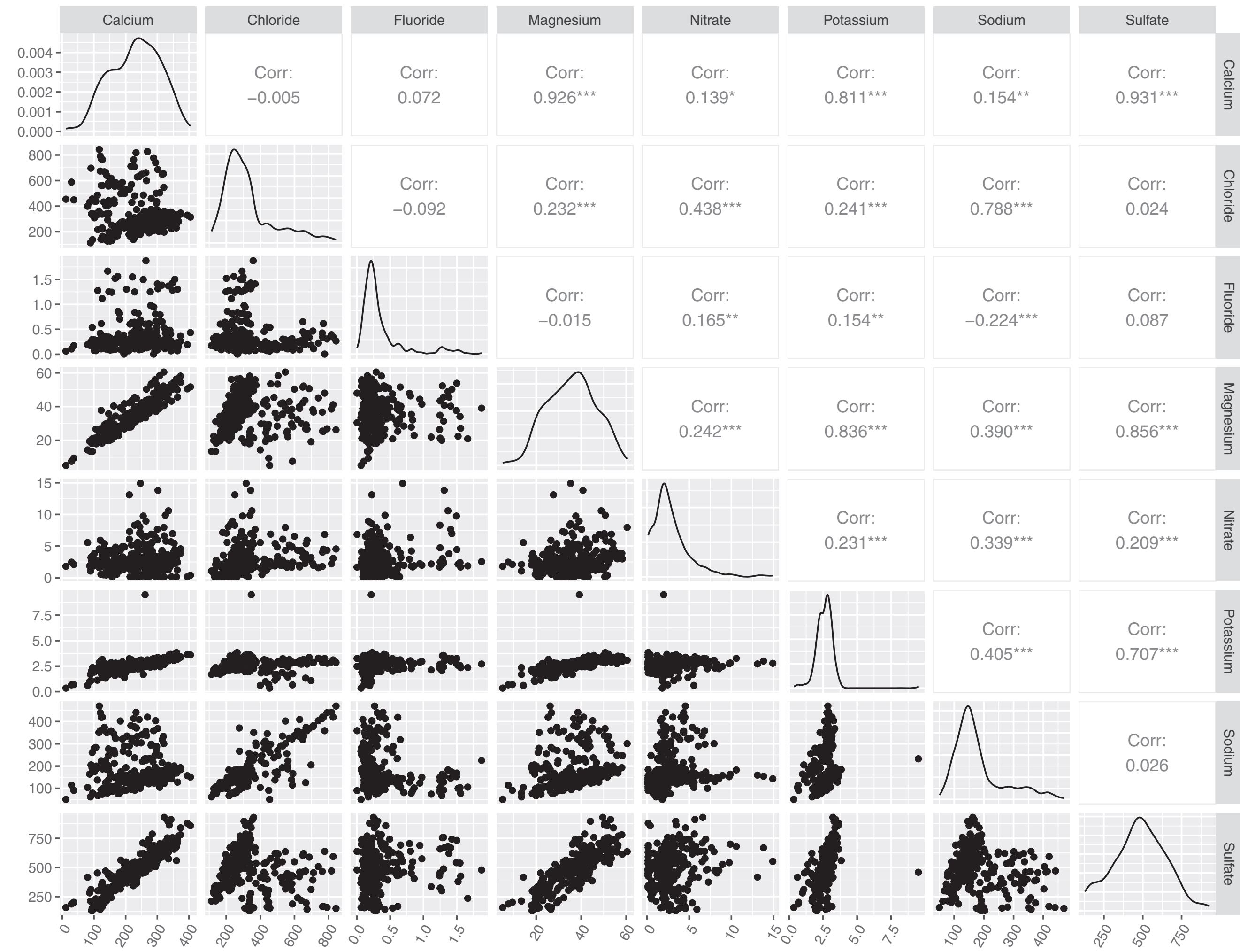

FIGURE 3 Correlation matrix and scatterplots for all analytes. Spearman correlation coefficient is shown in each box. "***" indicates a significance level at 0.001, "**" at 0.01, "*" at 0.05, and "." at 0.1.

resulted in similar values of discharge to the 2022–2023 baseflows. Such relationships were evaluated based on site (upstream vs. downstream of the cemetery in the connected reach) and season (whether the connected reach was gaining or losing). C-Q relationships are shown in Figure 6. Ca, Mg, and sulfate all show dilution patterns—decreased concentration at higher discharge. This again supports a common geogenic source for these three analytes. Unlike these three analytes, K shows dilution at both sites during the gaining season, and no change at either site during the losing season, likely reflecting different flow paths, contributing source material and/or hyporheic zone dynamics such as plant uptake, where K and other analytes are hypothesized to differ in riparian zones in part due to vegetation (Ledesma et al., 2013).

Na and Cl concentrations are correlated showing statistically significantly higher values in winter (losing season) than in summer (gaining season) ($p < 0.01$, $W = 1873$ and 2388, respectively, and $n = 155$) (Figure 4). This pattern suggests a common source, i.e., road salt application that is prevailing in the Meadowbrook watershed in wintertime, which is consistent with what Ledford and Lautz (2015) observed in Meadowbrook in 2012. The connected reach likely releases stored road salt throughout the gaining season (i.e., summer) that has accumulated during the losing season. This phenomenon was proposed by Ledford and Lautz (2015) for data in 2012–2013 and is still observed in 2022–2023. Cl routinely exhibits concentrations higher than aquatic life guidelines of 260 mg/L, warranting better urban water management to safeguard aquatic species from elevated chloride levels in urban water bodies. Na and Cl concentrations display no statistically significant difference in median between 2012 and 2022 ($p = 0.197$ and 0.233, $W = 40\,506$ and 40 902, respectively, and $n = 314$). Predominantly sourced in the watershed as road salt, Na and Cl had higher instantaneous peaks in 2012 potentially due to the higher snowfall amounts than in 2022 (272 cm compared to 165 cm, Menne, Durre, Korzeniewski, et al., 2012; Menne, Durre, Vose, et al., 2012) likely resulting in more road salt being used. However, these higher instantaneous peaks might also be due to the low temporal resolution of sampling resulting in 2022–2023 samples that missed the highest peaks in 2022–2023. It is worth noting that the much lower snowfall in 2022 did not yield statistically significantly different

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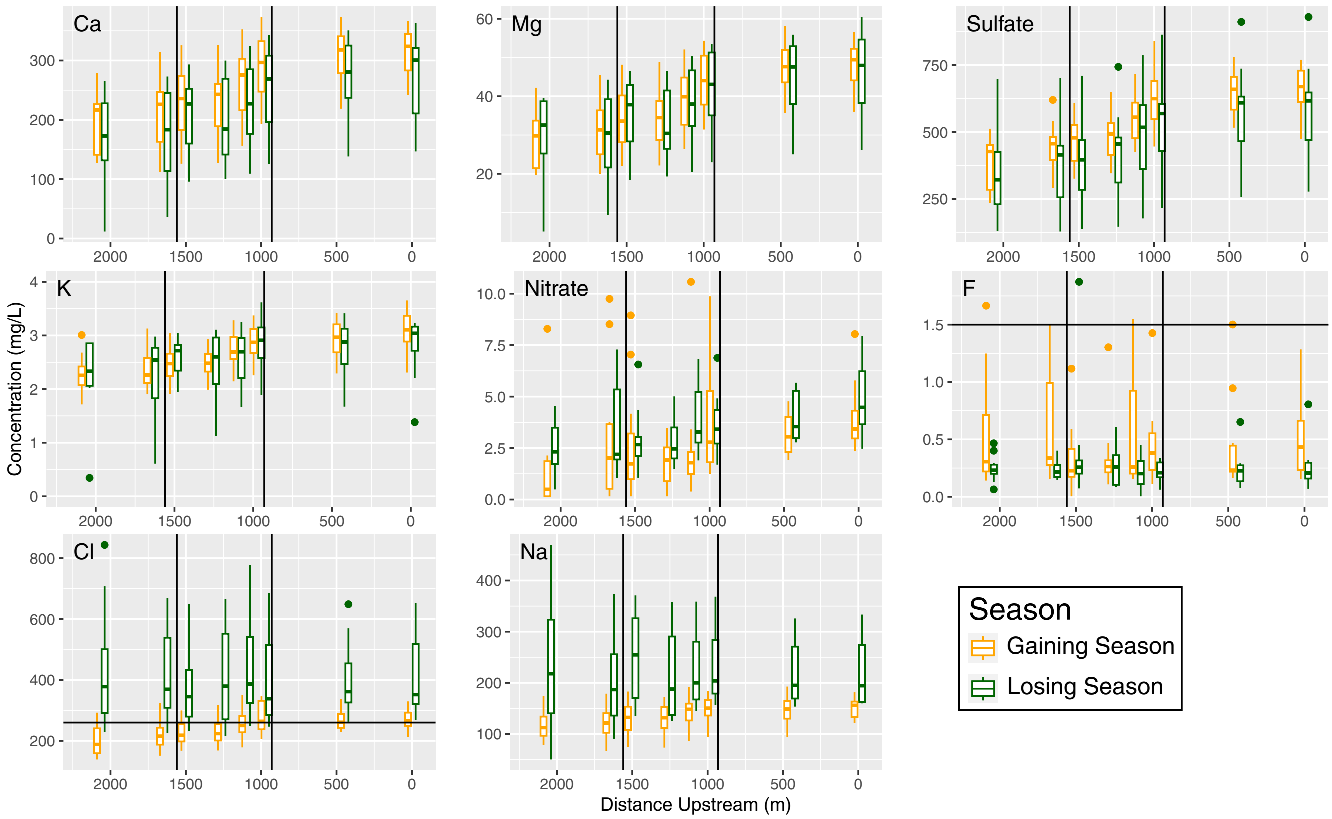

FIGURE 4 Boxplots showing the concentration distribution of Ca, Mg, sulfate, K, nitrate, F, Cl, and Na of samples collected from eight sites within the connected reach in Meadowbrook, with green and orange boxes representing losing and gaining seasons, respectively. Vertical black lines indicate the upstream and downstream boundaries of the cemetery. Horizontal black lines denote aquatic life water quality standards.

lower median Na and Cl concentrations than in 2012 (Figure 5). This corroborates with the findings from the literature (Kaushal et al., 2005, 2023; Kelly et al., 2008; Ledford et al., 2016) that while seasonal application of road salt can impact salt-related instantaneous peak analyte concentrations, legacy buildup of road salt has a lasting impact on the watershed as it does in much of the northeastern USA. C-Q plots for Na and Cl show that as the flows increase in the winter at the upstream site, where road density—and therefore road salting—is higher, road salt will undergo greater flushing from the watershed into the stream, increasing concentration. At the downstream site, concentrations remain stable at higher flows, likely due to the lower density of roads and the tendency to store road salt in the stream banks (Ledford et al., 2016). This flushing phenomenon does not happen in the summer as there is limited road salt within the watershed to flush out at baseflow. The flat slopes occurring in Figure 6 indicate that there is a consistent supply of Na and Cl in the groundwater in the gaining season, contributing to baseflows. This is likely from road salt being used within the watershed over the course of decades (Kaushal et al., 2005; Kelly et al., 2008).

Fluoride (F) shows a strong seasonality, reporting statistically significantly higher values in the gaining season ($p < 0.01$, $W = 13\,526$, $n = 155$). We do not observe significant spatial variation within and downstream of the cemetery compared to upstream of the cemetery ($p = 0.276$, $W = 36\,048$, $n = 165, 71$, respectively) (Figure 4). Unlike this study, Lautz et al. (2020) showed that F concentrations significantly decreased within the cemetery in 2012 and hypothesized that longitudinal fluoride variation along Meadowbrook is due to a lack of leaking sewer and water infrastructure within the cemetery. In both 2012 and 2022, F concentration in Meadowbrook Creek failed to increase through the cemetery, likely excluding the cemetery (including burial decay products) as a primary source of F in Meadowbrook (see Section 4.3 for details). Fluoride data exhibits C-Q patterns with a slope not significantly different from zero, except for upstream during the losing season where it undergoes slight dilution at higher baseflows. Such slopes demonstrate that fluoride is not diluted at higher flows. This indicates a source that varies with the flow, potentially including geogenic sources, fertilizers, and/or road salt (Ahmad et al., 2022; Granato, 1996).

4.3 | Nitrate in Meadowbrook watershed with a focus on St. Mary's cemetery

Nitrate median concentrations are statistically significantly higher in the winter (compared to summer) ($p < 0.01$, $W = 7014$, $n = 155$) and increasing within the cemetery consistent with previous years (Lautz et al., 2020; Ledford et al., 2017). Higher nitrate in the winter might be due to lower denitrification rates and less plant uptake. Nitrate

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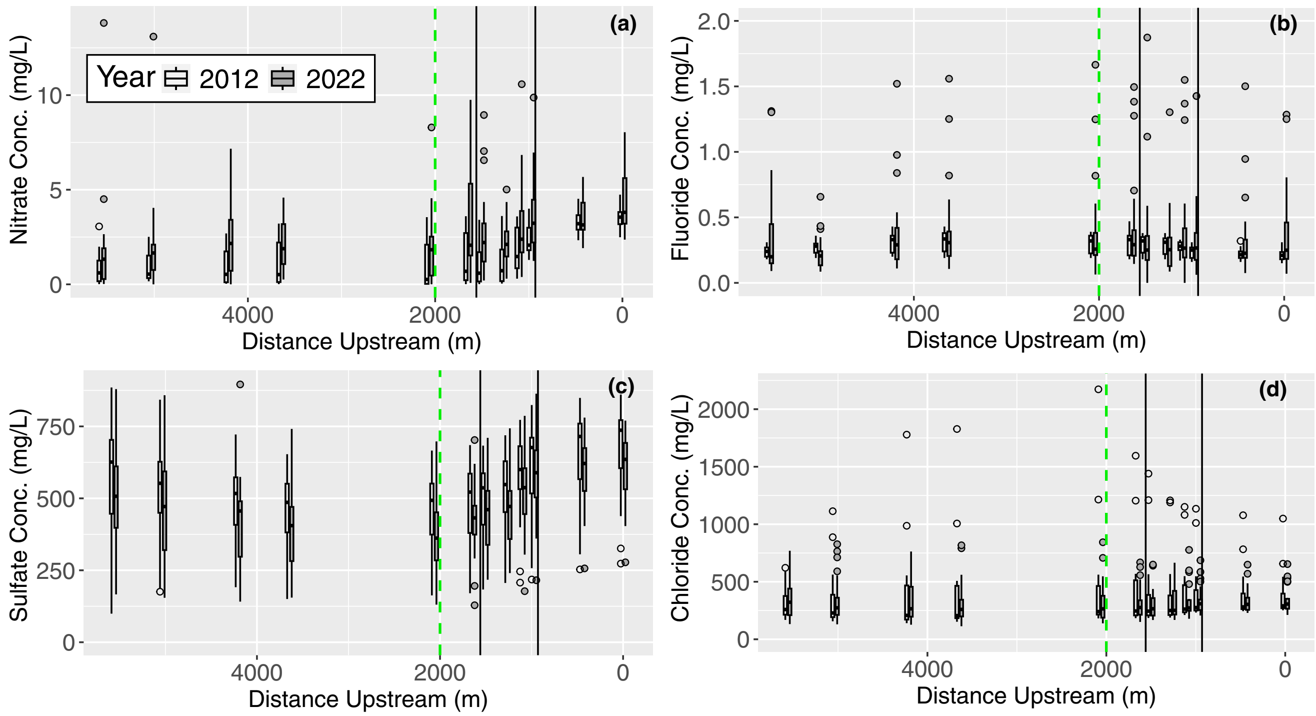

FIGURE 5 Boxplots showing the concentration distribution of (a) nitrate, (b) fluoride, (c) sulfate, and (d) chloride within Meadowbrook, with hollow (left) and filled (right) black boxes representing the 2012 and 2022 sample years, respectively. Vertical black lines indicate the upstream and downstream boundaries of the cemetery, and the dashed green lines indicate the boundary between the upstream disconnected reach and the downstream connected reach. Magnesium, calcium, and potassium follow the same patterns as sulfate. Sodium follows the same patterns as chloride.

concentration increases from upstream to within/downstream of the cemetery (Figure 4), suggest that the cemetery is releasing nitrate to Meadowbrook Creek, which is also consistent with previous research (e.g., Lautz et al., 2020). Nitrate within the watershed and within the cemetery has increased since 2012 ($p < 0.01$, $W = 49\,387$, $n = 314$). One change in cemetery management relative to 2012 is that fertilizer is now used on cemetery grounds. This was not the case in prior years (Lautz et al., 2020). The C-Q plot of nitrate exhibits flushing patterns at the upstream site in the losing season. Less nitrate is taken up in-stream in the winter, allowing nitrate to accumulate in the surface water as higher flows flush nitrate out of the watershed. Nitrate in the upstream site could be sourced from fertilizers, wastewater, organic inputs such as lawn trimmings, or wet deposition. This C-Q pattern switches at the downstream site in the losing season to dilution at higher baseflows. Gaining season slopes are not statistically significantly different from zero, consistent with previous studies indicating that nitrate in urban watersheds follows negative or insignificant trends in the C-Q plot (Balerna et al., 2021; Duncan et al., 2017). In particular, Duncan et al. (2017) showed that annual precipitation can lead to either positive or negative nitrate C-Q relationships in small (<15 km^2) urban watersheds, with overall multi-year trends being balanced out. Balerna et al. (2021) did not offer an explanation but showed that small urban watersheds over the course of multiple years had insignificant nitrate C-Q relationships.

In Figure 7, nitrate δ^{15}N and δ^{18}O values of water samples collected from downstream (sites 1, 2), within (sites 3, 4, 6), and upstream (sites 8, 12) of the cemetery are plotted along with typical isotopic ratios of five common nitrate sources in urban streams: atmosphere-derived nitrate, ammonium-based fertilizer, nitrate-based fertilizer, soil nitrate, and human/animal waste. The typical δ^{15}N value of human bone is generally greater than 10‰ (O'Connell et al., 2012). δ^{15}N values of all collected Meadowbrook water samples are no more than 7.6‰, likely excluding burial decay products as a major nitrate source (Figure 7). δ^{15}N and δ^{18}O values of nitrate in Meadowbrook increase and decrease downstream, respectively. This suggests a mixing of at least two endmembers, with one characterized by high δ^{18}O and low δ^{15}N and the other with low δ^{18}O and high δ^{15}N. Meadowbrook Creek originates from a stormwater retention pond, which allows ample time for surface water to equilibrate with the atmosphere. In addition, δ^{15}N (and δ^{18}O) values of all samples are within or higher (and within or lower) than that of nitrate fertilizer. Thus, atmosphere-derived nitrate and/or nitrate fertilizer are deemed likely to be the high δ^{18}O/low δ^{15}N endmember. As water flows downstream, nitrate derived from other sources, for example, soil nitrate, more fertilizers, and human/animal waste, are released into Meadowbrook, which brings down nitrate δ^{18}O and elevates nitrate δ^{15}N values. At Meadowbrook, baseflow water samples at two stream sites (i.e., at the end of disconnected reach and the end of connected

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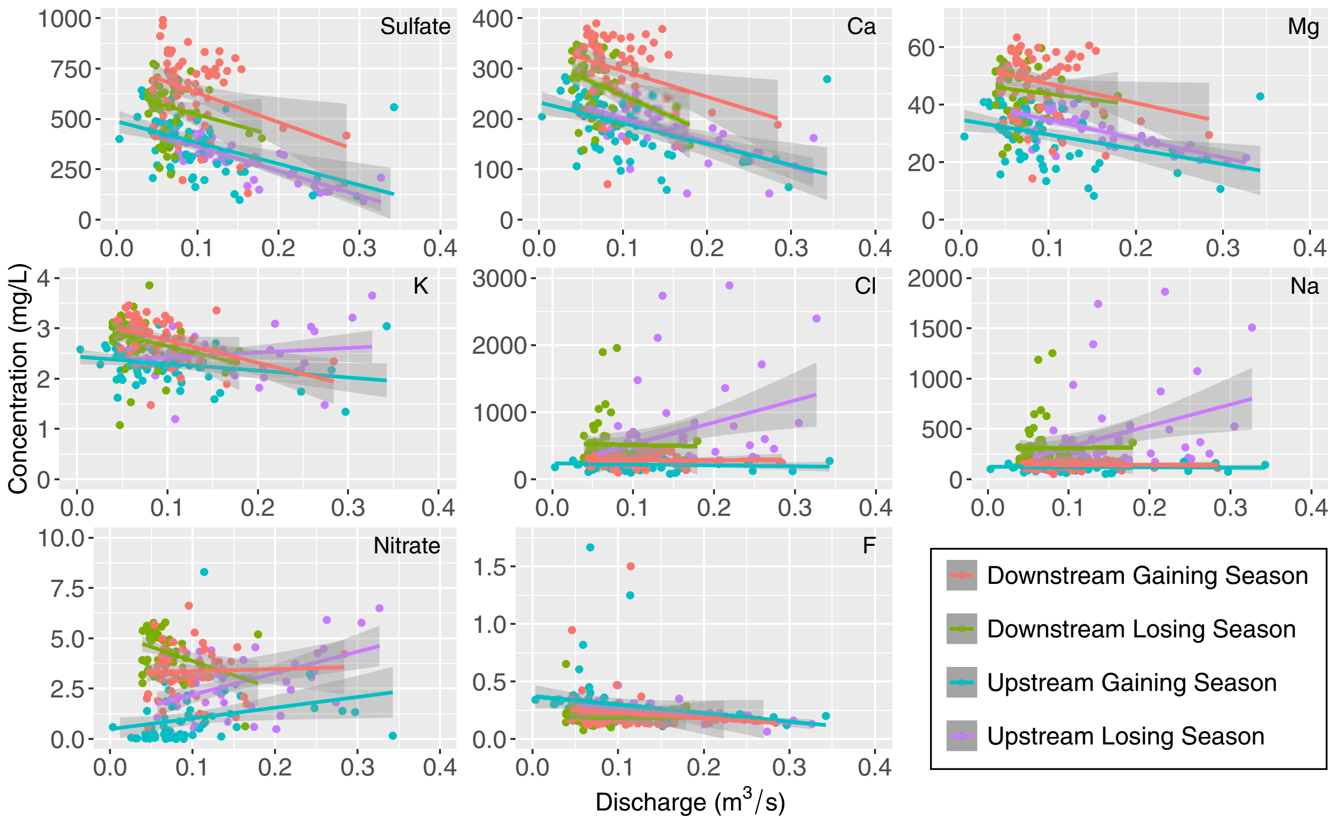

FIGURE 6 Concentration-discharge plots for sulfate, Ca, Mg, K, Cl, Na, nitrate, and F at the baseflow condition. Regression lines with 95% confidence intervals are shown for four scenarios: upstream or downstream of the cemetery reach during the gaining or losing season.

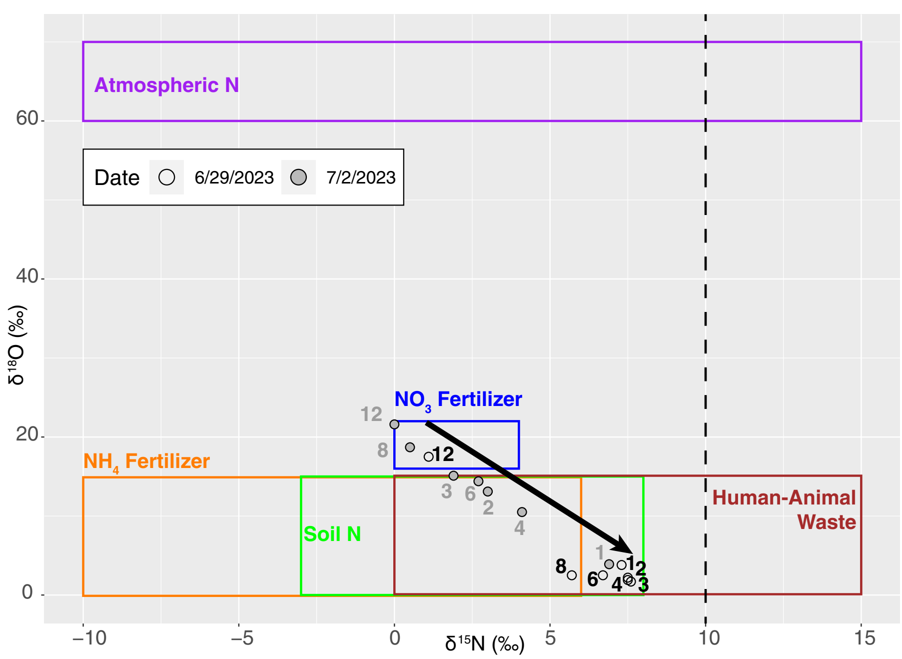

FIGURE 7 $\delta^{18}O$ and $\delta^{15}N$ values of samples taken on 29 June 2023 and 2 July 2023 in Meadowbrook. Coloured rectangles represent typical isotope signatures from various sources (Kendall, 1998). The black arrow represents the general sample location, pointing in the direction from upstream to downstream. The general pattern is that upstream samples are higher in $\delta^{18}O$ and lower in $\delta^{15}N$, which decreases and increases, respectively, as moving downstream. The vertical black dash line represents typical human bone $\delta^{15}N$ values (O'Connell et al., 2012).

reach) were analyzed for nitrogen and oxygen isotopes of nitrate in 2012–2013 (Ledford et al., 2017). Based on the isotope data, Ledford et al. (2017) concluded that baseflow nitrate in Meadowbrook Creek might be mainly sourced from the mixing of wastewater and groundwater. They also acknowledged that such baseflow samples might not reflect the contribution from sources related to overland flow and storm events (e.g., fertilizer). Many water samples collected in 2023 present similar nitrate isotope data as those in 2012–2013,

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while most samples that are further upstream from the two sites visited in 2012–2013 show much higher $\delta^{18}O$/lower $\delta^{15}N$ values approaching the nitrate fertilizer zone. In particular, in 2012–2013, $\delta^{15}N$ values ranged from 1.9‰–12.6‰, slightly higher than the range of 0‰–7.6‰ found in 2023. $\delta^{18}O$ values in 2012–2013 ranged from −9.0‰–8.0‰, much lower than the 1.7‰–21.6‰ found in 2023. Higher $\delta^{18}O$ and lower $\delta^{15}N$ at the same sites between different years suggest a potentially stronger influence of atmospheric and nitrate-based fertilizer in 2023 than in 2012–2013. The occurrence of $\delta^{15}N$ over 10‰ in 2012–2013 indicates that nitrate isotopes derived from human bones could be present in the water, although these values also fall within wastewater $\delta^{15}N$ values. These observations also highlight the importance of longitudinal synoptic sampling, preferred at high frequency (e.g., weekly and bi-weekly) in stream-related research.

The preliminary results from Lautz et al. (2020) suggested that up to 25%–50% of such nitrate leachate from the cemetery could be potentially sourced from burial decay products. In this study, we have compiled multiple lines of evidence which together suggest that burial decay products are likely not a major source of nitrate to Meadowbrook. First, burial vaults have been required in St. Mary's since the 1970s (T. Smith, personal communication, 9 May 2023). Therefore, the usage of vaults in St. Mary's within the last 50 years could likely prevent burial decay products from freely leaching out and into the nearby stream water. Similarly, Saba et al. (2023) reported that a cemetery with vaults showed no burial decay product impact on nearby groundwater quality, including nitrate concentrations. In addition, fluoride (F), which accumulates in human teeth and bones, can be released into the environment as part of the burial decay products (Dent et al., 2004). F concentration in Meadowbrook Creek either declines in 2012 or stays constant in 2022 as it flows through the cemetery in the gaining season, arguing against the presence of major impacts of burial decay products on Meadowbrook water quality. Third, the isotopic composition of Meadowbrook nitrate shows that the nitrate is more likely to be dominantly sourced from atmospheric nitrate, fertilizers, and wastewater.

Nitrate is unlikely to be sourced from burials prior to the 1970s based on the estimated residence time of groundwater and nitrogen and the isotopic signature of our data. Van Meter et al. (2017) showed that nitrogen older than 50 years accounts for approximately 33% and 5% of the nitrogen at the basin outlet for the Mississippi (3 000 000 km^2) and Susquehanna (70 000 km^2) rivers. Smaller rivers have faster biogeochemical cycling of nitrate (Basu et al., 2011), suggesting that nitrate residence time within small watersheds like Meadowbrook is shorter than 50 years. This suggests that very little, if any, nitrogen in Meadowbrook is the result of pre-1970s burials. It is also important to note that soil organic nitrogen released from pre-1970s might have accumulated in a 'protected pool' of soil organic nitrogen that has a residence time on the order of 1000 years (Van Meter et al., 2017). However, St. Mary's has only been operational since the early 1900s, likely eliminating this 'protected pool' as a source of nitrate. In addition, our nitrogen and oxygen isotope data points to limited denitrification signals. If long water (and thus nitrate) travel time was expected, we would expect to see coupled enrichment of nitrogen and oxygen isotopes due to subsurface denitrification (Yu et al., 2023), that is, N and O isotopes will be enriched following a 1:1 to 1:0.5 line, but that is not the case here (Figure 7). Therefore, we suggest that burial decay products are less likely to act as a major source of nitrate in Meadowbrook water. Instead, other nitrate sources in St. Mary's Cemetery, including fertilizers, grass clippings, leaf litter, atmospheric deposition, and wastewater, should be considered. Higher nitrate concentrations within the cemetery may also be exacerbated near the stream edge by the lack of dense vegetation that uptake nitrate, in contrast to other areas of the connected reach (Fennessy & Cronk, 1997). The operation manager of St. Mary's Cemetery shared that fertilizers were used 'a couple of times' per year in active zones, which confirms that fertilizers are a potential nitrate source in St. Mary's. However, we cannot definitively rule out burial products as a source of nitrate to Meadowbrook, due to the small sample size of isotope data collected in 2023 in this study, the possibility of vaults leaking, and the assumption that this system follows residence time patterns observed in previous literature.

Nitrate load modeling was completed for the two sites upstream and downstream of the cemetery to calculate cemetery-derived nitrate loads. Overall, St. Mary's Cemetery acts as a nitrate sink on an annual basis. From 1 February 2022 to 31 January 2023, the upstream site has an estimated load of 18 540 kg, and the downstream site has a load estimate of 9388 kg. There is seasonal variation, and the cemetery is a net source of nitrate during the summer and early fall, but overall load differences highlight the annual 50% drop in nitrate loads between upstream and downstream of the cemetery (Figure 8). These seasonal patterns generally correspond with Beltran et al. (2021), but 2022–2023 estimated nitrate load values are higher at the upstream site than the 2018 and 2019 water year values (13 700 and 14 100 kg, respectively). The downstream site discharged an estimated 9640 and 10 900 kg of nitrate in water years 2018 and 2019, respectively, which are slightly higher than the 2022–2023 value.

The active zone, where burials are occurring and where cemetery fertilizer is applied, makes up approximately 600 000 square feet of the cemetery. Assuming a standard lawn application rate of one pound of nitrate per 1000 square feet applied four times per year during late spring, summer, and early fall, this comes out to approximately 1100 kg of nitrate applied in the cemetery active zone. Assuming an uptake rate of 50% (Craswell, 2021) this results in 550 kg of nitrate loading due to fertilizer over 5 months, approximately 3.7 kg per day. Lautz et al. (2020) estimated a 20–40 kg nitrate per day increase between the upstream and downstream gauges during baseflow in summer months, which is similar to what we saw in 2022–2023. This means that fertilizer used in the active zone of the cemetery might only make up 10%–20% of the nitrate loading increase between the gauges, suggesting that other sources of nitrate contribute to nitrate loading increase.

Nitrate loading is driven primarily by patterns of surface-groundwater interaction, as loading corresponds to when the stream is a losing and gaining stream within the cemetery reach and is shown

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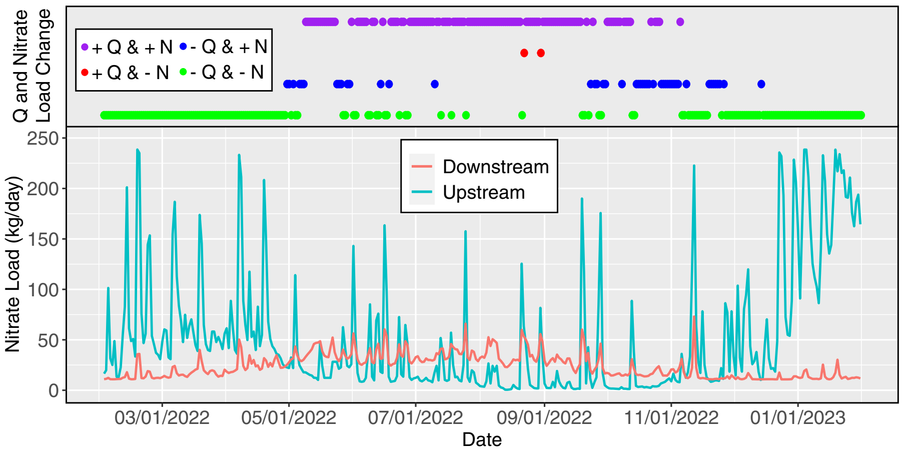

FIGURE 8 Daily nitrate loads estimated with LOADEST models for upstream and downstream gauges of the cemetery. Coloured dots are used to indicate if the nitrate load (denoted by 'N') and discharge (denoted by 'Q') are gaining ('+') or losing ('−') between the two sites.

in Figure 8, where we predominantly see the stream is losing both discharge and nitrate load or gaining both discharge and nitrate load on a given day. However, an exception to this general pattern is that Meadowbrook might experience a net gain of nitrate loads while also having a net loss of discharge throughout the cemetery, particularly in fall and spring (Figure 8). The fall and spring phenomena of a net loss in discharge but a net gain in nitrate loads are indicative of high nitrate concentrations in the groundwater within the cemetery. This reach likely experiences dynamic hyporheic exchange with a small net loss of stream water to groundwater during this time. However, the high-concentration cemetery groundwater carries enough nitrate relative to the stream water to offset the nitrate losses that are associated with a net loss of discharge and result in increased nitrate loads during these time periods. Jimenez-Fernandez et al. (2022) describes a headwater stream undergoing various hyporheic exchange patterns including both net gaining from groundwater input and net losing to groundwater input throughout the 500-meter reach studied. Meadowbrook likely undergoes a similar pattern throughout the cemetery and future research should identify more specific areas of the gaining and losing stream. Another possibility is that varying residence times within the hyporheic zone may lead to spatial and temporal changes in nitrification and denitrification patterns (e.g., Zarnetske et al., 2011). Although the cemetery reach can be a seasonal source of nitrate, it acts as an annual nitrate sink. Practices such as reducing fertilizer usage, cleaning up lawn clippings, and phytoremediation via increasing near-riparian vegetation may help turn the reach into a year-round sink.

5 | CONCLUSIONS

Meadowbrook Creek in Syracuse, New York (USA) provides a unique opportunity to examine anthropogenic impact and hydrological control on solute release and contributions to an urban stream in a setting with various types of human-built infrastructure (e.g., cemetery, residential, paved roads). Our bi-weekly longitudinal stream sampling and continuous discharge measurements focused in and around St. Mary's cemetery over a year demonstrated highly variable spatiotemporal hydrogeochemical patterns representing contributions from a wide variety of solute sources, primarily including the cemetery and road salting. In particular, geogenic solutes such as calcium and potassium undergo various spatial and temporal trends and may be driven by the variability in precipitation and seasonal surface water-groundwater interaction. Decadal trends from 2012 to 2022 indicate that sodium and chloride sourced from maintained use of road salt continue to be a detriment to aquatic life, although the cemetery reach has been shown it can help attenuate the problem by benefiting from the ecosystem functions provided by its riparian zones and connection to groundwater relative to more urbanized channels. Previous studies suggested that up to 25%–50% of the observed nitrate increase in Meadowbrook Creek along St. Mary's cemetery could plausibly derive from burial decay products. However, we suggest that burial decay products are unlikely to be a significant source for nitrate (and other analytes) in Meadowbrook due to the sealed burial vaults and other prolific nitrate sources within the watershed such as fertilizer, decaying plant matter, and wastewater, along with nitrogen isotopic analysis. Nitrate loading calculation indicates that the cemetery acts as an annual net sink for nitrate within the watershed, providing an ecosystem service for the urban stream.

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CONFLICT OF INTEREST STATEMENT

The authors declare no competing financial interest.

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DATA AVAILABILITY STATEMENT

Upon publication, datasets discussed in this work will be made publicly available with a data DOI via the CUAHSI HydroShare. During the manuscript review process, all datasets have been made available to the editors and reviewers via a HydroShare resource link: https://doi.org/10.4211/hs.bd53d08c08504c48a1273010c53d4580.

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