Received: 30 June 2022 | Accepted: 20 September 2022

DOI: 10.1002/jeq2.20416

Journal of Environmental Quality

TECHNICAL REPORT

Stormwater drives seasonal geochemical processes beneath an infiltration basin

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Assigned to Associate Editor Peter Kopittke.

Abstract

Deicing salt is an important component of road safety during winter storms. Stormwater infiltration best management practices aim to prevent the salt from polluting streams and waterways, but this may shift pollutants to groundwater resources. In response to limited field studies investigating groundwater quality impacts caused by input of salt from stormwater infiltration best management practices, we monitored water levels and quality of groundwater at various depths in an unconfined aquifer around a stormwater infiltration basin using in situ sensors coupled with grab sampling. Our observations revealed differences in groundwater chemistry with depth in the aquifer and processes that were driven by the seasonal changes in the chemistry of stormwater (salt-impacted in winter and fresh in non-winter) recharging the aquifer. Water–matrix interactions in the vadose zone beneath the basin affected the transport of sodium (Na) into groundwater following non-winter recharge. Sodium movement through the aquifer was delayed relative to chloride (Cl), indicating a longer residence time of Na in the vadose zone. Radium (Ra) concentrations were correlated with Cl concentrations, suggesting salt-impacted recharge caused desorption of Ra into groundwater because of increased salinity. Stormwater-influenced groundwater followed a preferential flow path due to heterogeneity of the aquifer materials, and water chemistry varied with time and location along the flow path. These results highlight the importance of well screen length, placement and depth, and frequency of observations when designing a monitoring network.

1 | INTRODUCTION

The process of polluted roadway stormwater flowing from the edge of the pavement to the ground and entering the subsurface via infiltration is straightforward. However, modern transportation drainage infrastructure often transports stormwater hundreds of meters from the roadway before entering the subsurface (Brumley et al., 2018; Cassanelli & Robbins, 2013; Fischer et al., 2003). Stormwater runoff from roadways and parking lots is often conveyed to stormwater management areas, many of which are designed with the intent of flood control and protection of aquatic life and habitat, prioritizing infiltration to the subsurface over runoff into a surface water body (Andres et al., 2018a; Beckingham et al., 2019; Pitt et al., 1999). Stormwater recharge to groundwater can lead to contamination of aquifers and groundwater resources (Brumley et al., 2018; Burgis et al., 2020; Granato et al., 1995).

Climate change and dry pavement policies are causing increased reliance on deicer application, requiring a better

Abbreviations: BEX, base exchange index; d-excess, deuterium excess; PTC, pressure-temperature-specific conductance; SC, specific conductance; TDS, total dissolved solids.

understanding of how the deicer components move through the environment (Daley et al., 2009; Stirpe et al., 2017). Chloride (Cl) and sodium (Na) concentrations in surface waters and groundwater have been increasing in recent years, a portion of which is attributed to deicer application (Bhide et al., 2021; Kaushal et al., 2021; Panno et al., 2006). Regional-scale salt mass balance studies have revealed that up to 77% of deicer salt applied to road surfaces is retained in the subsurface and does not leave the watershed by the following winter season (Howard & Haynes, 1993; Kelly et al., 2012; Meriano et al., 2009; Novotny, 2009). Increased travel time from roadway to stream may reduce impacts on surface water quality; however, it may take hundreds of years for deicer components to flush through the groundwater system (Bester et al., 2006; Qian et al., 2020). Because of adsorption and other geochemical interactions within soil and the aquifer matrix, sodium residence time in the watershed is longer than that of chloride (Bhide et al., 2021; Snodgrass et al., 2017). Sodium retention can negatively affect soil structure and reduce infiltration, increasing stormwater residence time in a detention pond or basin (Fujimaki et al., 2006; Stirpe et al., 2017). Aquifer heterogeneity complicates prediction of when a river or stream may be most affected by an influx of sodium from baseflow contribution.

In multilayered aquifer systems, such as the North Atlantic Coastal Plain, most confined aquifers receive recharge from a water table aquifer (Masterson et al., 2015), and deeper aquifers may be susceptible to contamination from anthropogenic activities and surficial processes (Ayotte et al., 2011; Bester et al., 2006; Carleton, 2010). Heterogeneity in the hydraulic and geochemical properties of surficial material can affect the transport and retention of deicer components into deeper groundwater (Kelly et al., 2018; Morales & Oswald, 2020; Novotny, 2009). Well design can promote artificial connections between aquifer layers, causing dilution of contaminants that are more prevalent in one portion of the subsurface (Ayotte et al., 2011; Bester et al., 2006; Church & Granato, 1996), encouraging migration of contaminants between aquifers (Church & Granato, 1996; Landon et al., 2008; Strohmeier, 2008), or introducing new contaminants that have been mobilized as a result of mixing between waters of varied chemistry (Ayotte et al., 2011).

The chemistry of recharge water will influence groundwater quality. Roadside studies have observed increasing aquifer salinity, measured as either Cl, Na, or conductivity, as deicer-impacted runoff migrates through the vadose zone and recharges the water table (Baraza & Hasenmueller, 2021; Daley et al., 2009; Herb et al., 2017; Rivett et al., 2016; Robinson & Hasenmueller, 2017). Seasonal trends in stormwater quality will affect water–matrix reactions in shallow aquifers, including mobilization of metals (Amrhein et al., 1992; Granato et al., 1995) and other contaminants such as radionuclides (Andres & McQuiggan, 2019; Bolton, 2000; McNaboe et al., 2017). Studies that have combined deicer impacts with stormwater management practices have observed salt storage in shallow soil (Burgis et al., 2020; Lam et al., 2020; Snodgrass et al., 2017; Wilde, 1994). However, there have been limited field investigations that combine the effects of deicer salt on groundwater as a result of stormwater-related infrastructure and fate and transport after the deicer-impacted stormwater recharges the aquifer (Job, 2021).

Our goal was to evaluate deicer impact on groundwater beneath a managed stormwater recharge site by identifying, through field monitoring and synthesis of previous investigations and project-collected data, water–matrix reactions and the potential drivers of those reactions. Seasonal groundwater sampling provided snapshots of water chemistry changes, and in situ automated sensors allowed us to track movement of groundwater and deicing components through time and space. Well clusters screened at various aquifer depths allowed us to investigate different portions of the aquifer with different permeability and hydraulic conductivity. Our results enhance existing conceptual models of stormwater infiltration by considering heterogeneities in hydraulic and geochemical properties within an aquifer and along the flow path.

Core Ideas

- Stormwater infiltration affects groundwater recharge chemistry and water–aquifer matrix interactions.
- Cl and Na were retained in the vadose zone beneath the basin with lag-time between their respective releases.
- Cl caused desorption of Ra and mobilization into groundwater.
- Evaporation occurred between stormwater inflow and infiltration to the water table.
- Stormwater recharge-influenced groundwater preferentially moved through higher-permeability layers.

2 | METHODS

2.1 | Study site

The study site is a state-managed infiltration basin (the basin) located near Middletown, DE (Figure 1). The basin was constructed in 2018 with a footprint of just over 2,900 m^2 and manages stormwater runoff from portions of a four-lane limited-access highway and a two-lane road. Stormwater enters the basin from two 0.61 m (24-in)-diameter concrete inlet pipes located in the northeast and southeast

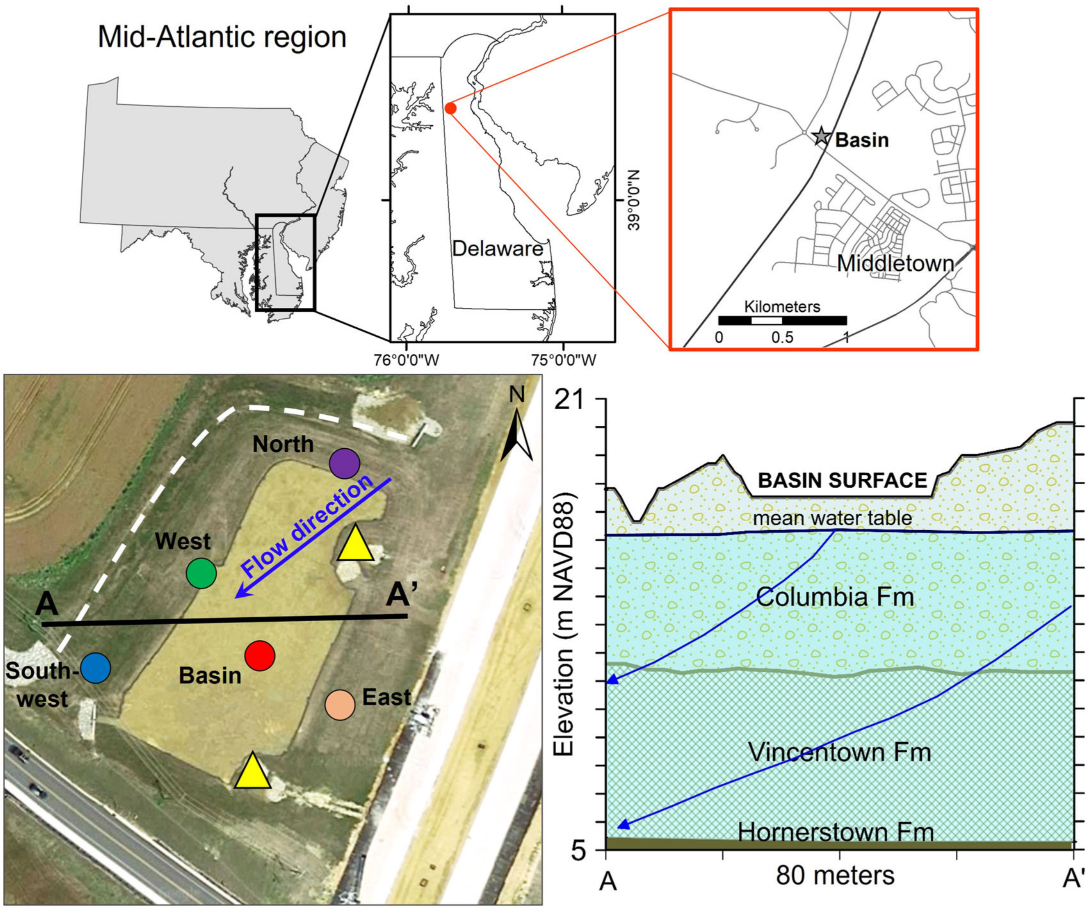

FIGURE 1 Regional overview map and site location map in Middletown, DE. Site plan shows the general site area and locations of monitoring well clusters at the basin. The well clusters are circles and are labeled with the cluster identifier. Yellow triangles denote the locations of stormwater inlets. Dashed white line shows the location of the perimeter ditch. Blue lines on the site map and cross-section show the direction of groundwater flow. Cross-section on the bottom right shows the mean water table and geological formations (Fm) along the line from A to A′

corners of the basin. Flow from the inlets is routed through stone-covered forebays prior to entering the basin. Outflow from the basin is controlled by a concrete v-notch weir with the crest set 0.31 m above the basin surface. A 1.38 m-high berm surrounds the infiltration basin. A perimeter drainage ditch, with bottom elevation below the basin surface, runs along the northern and western sides of the berm and routes basin outflow and runoff from an area outside of the basin catchment to a swale draining to the west.

On average, Middletown, DE, receives 100 to 130 cm of annual precipitation (NCEI, n.d.), with approximately 50 cm of winter snowfall occurring from December to early March (DEOS, 2020). During this project, snowfall ranged from trace amounts in 2019–2020 to 27.9 cm in 2020–2021 (DEOS, 2020). Delaware's Department of Transportation uses a combination of NaCl rock salt and brine to treat roadways in the state (DelDOT, personal communication, 7 July 2021). In this work, both rock salt and brine will be referred to as simply road "deicer."

Road construction has disturbed the topography and shallow subsurface around the basin through regrading, excavation, and compaction of materials. Shallow subsurface geological formations at the site are the Columbia, Vincentown, and Hornerstown Formations (Ramsey, 2005) (Figure 1). At the field site, sandy and moderately permeable materials of the Vincentown Formation are in direct contact with the Columbia Formation, and these geologic units function as an unconfined aquifer known as the Columbia aquifer. At the site, aquifer matrix material is predominantly quartz with no calcite (Ramsey, 2005). Where the Vincentown is separated from surficial material by a confining layer, the Vincentown hosts the Rancocas aquifer (Woodruff, 1992). The Hornerstown Formation functions as a confining unit at this site and forms the base of the unconfined aquifer. The Vincentown and Hornerstown Formations contain glauconite, which is associated with elevated uranium concentrations that can be a source for radon and radium in groundwater (Woodruff et al., 1992).

2.2 | Groundwater and stormwater monitoring

The groundwater-monitoring network at the basin consists of four clusters of monitoring wells located around the infiltration basin and one cluster in the middle of the basin (Supplemental Table S1). Each cluster consists of four wells that screen various depth intervals (Supplemental Figures S1 and S2). Each well was assigned to a depth group—shallow, mid-shallow, mid-deep, or deep. Surface water infrastructure consisted of post-mounted polyvinyl chloride stilling wells located in the inlet forebays and at the basin-side of the outlet weir structure.

From mid-May 2019 to mid-February 2022, each monitoring well and surface water stilling well were equipped with a pressure-temperature-specific conductance (PTC) sensor and set to continuously record at 15-min intervals. PTC sensors were periodically downloaded and calibrated according to manufacturer specifications throughout the project period. An additional PTC was installed periodically in the perimeter ditch to determine if groundwater that recharged through the basin discharged to the ditch. Manual groundwater levels were measured with an electric tape using methods described in Andres et al. (2018b). Slug tests were performed on monitoring wells to determine hydraulic conductivity. Procedural and analysis methods followed those published by Butler (1997) and Bouwer & Rice (1976).

2.3 | Water chemistry sampling

We collected 20 discrete stormwater samples following winter and non-winter runoff events (Supplemental Table S2). During some winter runoff events, multiple samples were collected several hours or days apart. Five rounds of groundwater samples were collected from project monitoring wells to monitor seasonal variability: fall (November) 2019, summer (July) 2020, winter (February–March) 2021, summer (June) 2021, and fall (November) 2021. Groundwater and stormwater sampling procedures are detailed in the Supplemental Material. Water was filtered in the field with a dedicated 0.45-micron capsule filter for water quality samples and with a 30-cm-diameter 0.45-micron cellulose acetate filter for large volume (20-L) radium isotope samples.

Stormwater samples were analyzed at the Delaware Department of Natural Resources and Environmental Control Environmental Laboratory for Cl and Br. Groundwater and select stormwater samples were analyzed at the environmental laboratory for a suite of common ions and trace metals (Supplemental Table S3). Temperature, pH, specific conductance (SC), and dissolved oxygen were measured in the field using a multi-parameter meter. Split samples were collected from monitoring wells and surface waters and submitted to the University of Delaware Environmental Isotope Geochemistry Laboratory for analysis of stable isotopes ^2H and ^{18}O in H_2O using a Los Gatos Research model LWIA-24 liquid water isotope analyzer. Stable isotope results were reported as delta (δ) values relative to Vienna Standard Mean Ocean Water (VSMOW) in units of per mil (‰). Radium isotopes ^{226}Ra and ^{228}Ra were analyzed by gamma spectrometry; details of the sampling and analysis are in the Supplemental Material.

2.4 | Statistical methods and classifications used to assess water quality

To determine whether groundwater was influenced by non-winter or winter stormwater, we used modified base exchange index (BEX) (Stuyfzand, 1999, 2008) values to classify the water as freshening, salinizing or neutral during sampling events. The BEX relies on the principle that Na is preferentially adsorbed onto soil particles as salinized water is introduced into the aquifer (groundwater is undersaturated in Na and oversaturated in Ca with respect to Cl, K, and Mg). However, the equation assumes a seawater influence by adding a multiplier to the Cl component that accounts for the ratio of (Na + K +Mg)/Cl in average seawater. Since the salt input to the basin system at our site is not seawater, we eliminated this constant from the Stuyfzand equation for non-dolomitic aquifers to account for our site:

$$\text{BEX} = \text{Na} + \text{K} + \text{Mg} - \text{Cl}\,(\text{meq/L})$$

When classifying BEX values, we used the established threshold of $+/-\ (0.5 + 0.02 \times \text{Cl})$ to account for natural dissolution of silicate aquifer material and analytical differences in the chemistry results (Stuyfzand, 2008). A negative BEX value below the threshold indicates the groundwater is salinizing and influenced by winter recharge from the basin. A positive BEX value above the threshold indicates the groundwater is freshening and influenced by non-winter recharge. Values of BEX at or near 0 indicate neutral groundwater.

The Spearman rank correlation coefficient function in R (R Core Team, 2019) was used to assess correlation between concentrations of Cl, Na, and BEX classifications. Significance was determined using a 95% confidence interval threshold (p value $< = .05$).

3 | RESULTS

3.1 | Site hydrogeology and water budget

Throughout the project period, there was little to no overflow (<1%) from the outlet weir (McQuiggan & Andres, 2021). Groundwater flow is highly influenced by aquifer stratigraphy. Hydraulic conductivity is heterogeneous, ranging from 1.1 to 35.3 m d^{-1} (arithmetic mean of 8.8 m d^{-1}, geometric mean of 5.4 m d^{-1}, and geometric standard deviation of 2.7 m d^{-1}) (Supplemental Table S1). Hydraulic conductivity of the Columbia Formation had the largest range (1.9 to 35.3 m d^{-1}), and the Vincentown and Hornerstown Formations had a smaller range (2.9 to 15.1 m d^{-1}).

Groundwater elevations ranged from 15.5 to 17.5 m above the North American Vertical datum of 1988 (0.2 to 2.2 m below the basin surface) and responded to storm events within a day (Supplemental Figure S5). Groundwater at the site flowed from the northeast to the southwest. Groundwater head elevations between wells at each cluster indicate predominantly lateral flow with the exception of potential vertical movement beneath the basin and west well clusters following heavy precipitation events. Larger head differences were observed between clusters following runoff-generating precipitation events and were attributed to water table mounding beneath the basin as well as hydrostatic loading.

3.2 | Stable isotopes (^{2}H and ^{18}O)

Stable isotope ratios of water (δ^{2}H and δ^{18}O values) were within the expected range of precipitation and river water for the Mid-Atlantic region (Kendall & Coplen, 2001) (Supplemental Tables S6 and S7). The δ^{2}H values ranged from −21.7 to −61.8‰ in groundwater samples and −36.2 to −68.3‰ in stormwater samples. The δ^{18}O values ranged from −4.2 to −8.3‰ in groundwater samples and −5.6 to −11.4‰ in stormwater samples. Samples were grouped by sample date and assigned to a winter or a non-winter group (Figure 2). Winter stormwater had lower δ^{2}H and δ^{18}O values than non-winter stormwater, consistent with current seasonal precipitation trends in the Mid-Atlantic (Dansgaard, 1964; Kendall & Coplen, 2001). Groundwater δ^{2}H and δ^{18}O values were generally similar to those of non-winter stormwater, indicating greater amounts of recharge during the summer months. Samples collected from downgradient wells were isotopically heavier than those collected from upgradient wells, consistent with enhanced summer recharge by infiltration within the basin.

Most samples plotted above the Global Meteoric Water Line (GMWL) (Craig, 1961) (Figure 2). Deuterium excess (d-excess) is the measured deviation from the GMWL with respect to δ^{2}H (Dansgaard, 1964). Deuterium excess in groundwater samples ranged from 4.4 to 18.3‰ (mean 13.1‰) and in stormwater samples from 8.7 to 23.2‰ (mean 18.6‰). Winter stormwater plots were within a range that is consistent with colder-weather precipitation in a humid climate (Jasechko, 2019). Shallow groundwater beneath the infiltration basin had more variable d-excess (greater spread) and smaller d-excess values, in particular those collected during summer months, indicating water that may have been subject to evaporation under non-equilibrium conditions before recharging the aquifer (Jasechko, 2019).

3.3 | Stormwater and groundwater chemistry

The relationship between field-measured SC and lab-measured Cl in groundwater and stormwater was defined using linear regression (Supplemental Figures S3 and S4). We calculated Cl directly from PTC-measured SC, using SC as a proxy for tracking the movement of deicer-impacted stormwater. Specific conductance data are particularly effective in tracking salinized groundwater (Daley et al., 2009; Galella et al., 2021; Moore et al., 2020; Snodgrass et al., 2017), especially when coupled with simultaneous head measurements (Figure 3).

Statistics of Cl concentrations in stormwater are calculated from the continuous sensor measurements. Mean stormwater Cl concentrations were higher in winter (December–February, 930 mg L^{-1}) and spring (March–May, 355 mg L^{-1}) than summer (June–August, 140 mg L^{-1}) and fall (September–November, 87 mg L^{-1}). Stormwater during the 2019–2020 mild winter (trace snow) had less Cl (mean Cl concentration 179 mg L^{-1}) than during the 2020–2021 average winter (mean Cl concentration 1,682 mg L^{-1}). Winter stormwater had higher concentrations of all major ions, except for HCO_3, than non-winter stormwater. Water in the perimeter ditch had consistently low (<200 µS cm^{-1}) SC, confirming that salty groundwater from the basin was not discharging to the ditch.

Statistics of groundwater major ion results broken down by location (well cluster) and depth grouping are shown on Supplemental Tables S4 and S5. Four groups were observed, corresponding with a condition or process occurring at the site: (Group 1) groundwater upgradient of the basin, (Group 2) groundwater from deeper wells completed in the Vincentown Formation, (Group 3) salinizing groundwater, and (Group 4) freshening groundwater (Figure 4). These groups, identified in Figure 4, are generally related to a water type (i.e. Na-Cl) based on the dominant ions in the samples.

Both winter and non-winter stormwater were clearly affected by salt (Na-Cl type). In contrast, upgradient, non-stormwater-impacted groundwater found mid-aquifer in the upgradient north and east well clusters (Group 1) had concentrations of Cl generally below 50 mg L^{-1} and total dissolved solids (TDS) concentrations below 100 mg L^{-1} (mixed Ca-Mg-SO_4/Cl and Ca-Mg-HCO_3 type). This is consistent with historical Columbia aquifer chemistry data from the area (Bachman & Ferrari, 1995). Overall site groundwater was slightly acidic to neutral and oxic. Groundwater at these background locations and depths had neutral BEX indices, with the

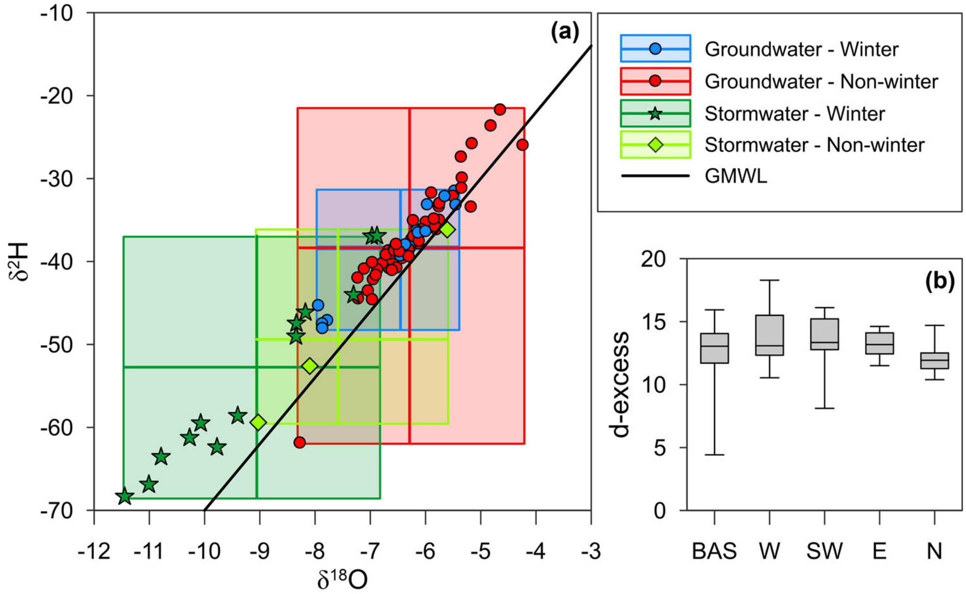

FIGURE 2 (a) Stable isotope δ^2H and δ^{18}O values in groundwater and stormwater samples, separated by winter and non-winter sample events. Shaded boxes are surrounding each sample grouping and the north–south axes for each grouping box represent the mean δ^{18}O value for the group and the east–west axes represent the mean δ^2H value for the group. (b) Deuterium excess (d-excess) for groundwater samples by well cluster (BAS, basin; E, east; N, north; SW, southwest; W, west; GMWL, Global Meteoric Water Line)

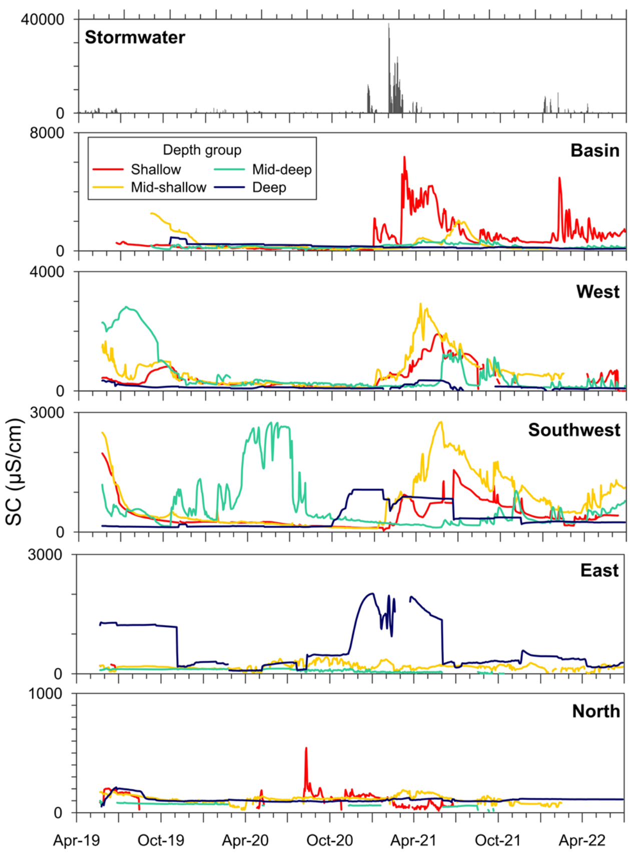

FIGURE 3 Specific conductance (SC) measurements from each well cluster and stormwater inflow. Each plot is color-coded by monitoring well depth intervals

exception of two samples that were slightly freshening (BEX value positive, yet <1) (Figure 4, Supplemental Table S8). These two samples, collected during non-winter events, had the lowest values in the freshening category (0.58 and 0.60), indicating minimal exchange reactions.

Deeper groundwater (Group 2) plotted between upgradient groundwater and freshening downgradient groundwater (Figure 4). While overall TDS (72 to 383 mg L^{-1}, median 104 mg L^{-1}) and Na concentrations (2.63 to 21.70 mg L^{-1}, median 5.05 mg L^{-1}) were low, the Na proportion in deeper groundwater was greater than that in shallower, upgradient groundwater. pH ranged from acidic to basic (4.36 to 8.15, median 5.34). Dissolved oxygen, while mainly oxic, was generally lower than groundwater in the shallower depth groups (median 5.40 mg L^{-1} compared with 7.37 mg L^{-1}), as was TDS (median 104 mg L^{-1} compared with 160 mg L^{-1}) and Ca (median 5.32 mg L^{-1} compared with 8.94 mg L^{-1}). Elevated nitrate concentrations (>0.4 mg L^{-1}), an indicator of pollution from agricultural sources, in deep groundwater were inversely proportional to iron concentrations, which agrees with previous studies in this area (Bachman & Ferrari, 1995).

Salinizing shallow groundwater beneath the basin had the highest concentrations of Cl, Na, Ca, and TDS and SC measurements out of all project groundwater samples. Maximum values of these parameters were measured during the winter sampling event. Both salinizing (Group 3, Na-Cl type) and freshening (Group 4, Na-Cl-HCO_3 type) groundwater were observed in shallow and mid-aquifer groundwater beneath and downgradient of the infiltration basin. Molar ratios of

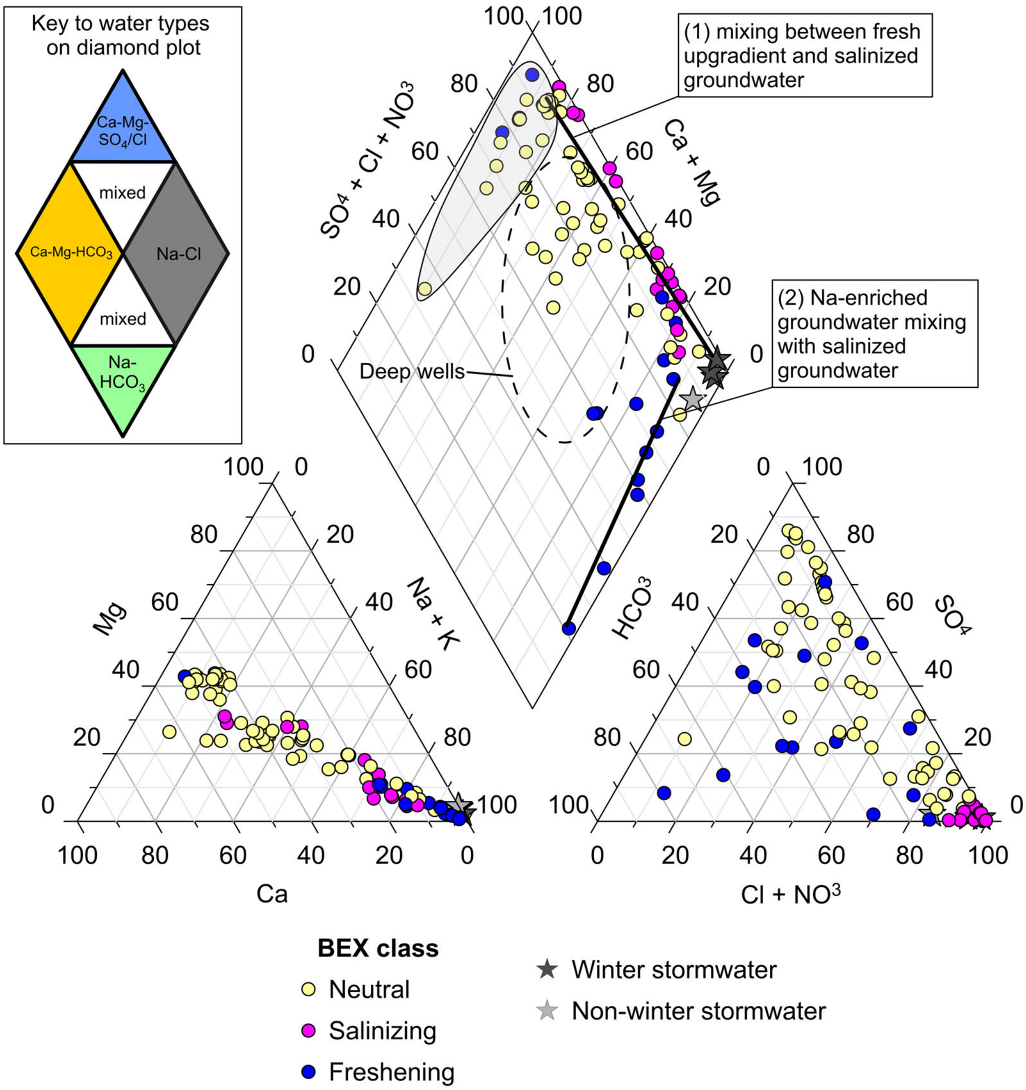

FIGURE 4 Piper diagram showing groundwater and stormwater samples. Symbols are color-coded to their base exchange index (BEX) classification. Shaded polygon represents site groundwater upgradient of the infiltration basin and the dashed line indicates groundwater from wells in the deep depth group. Two mixing lines (A and B) correspond with the salinization and freshening of groundwater that has been influenced by stormwater recharge from the infiltration basin. The key in the top left corner indicates the dominant ions in the sample (i.e., water type)

Na/Cl were positively correlated to the BEX value (ρ = .66, p value < .05), with positive BEX values that indicate freshening groundwater, associated with higher Na/Cl ratios and negative BEX values that indicate salinizing groundwater, associated with lower Na/Cl ratios. Stormwater-impacted groundwater showed a spatially salinizing progression from upgradient (Ca-Mg-Cl) to downgradient to Na-Cl, along mixing line A in Figure 4. Mixing line B (Figure 4) shows a temporal freshening progression from Na-Cl to Na-Cl-HCO_3 that occurs as fresher spring and summer recharge flushes salinized water through the aquifer. During each sampling event, groundwater in at least one well at each of the basin, west, and southwest clusters was salinizing (Figure 3). The depth at which salinizing groundwater was observed varied by sampling event. The highest concentrations of Ca (>30 mg L^{-1}) were in salinizing groundwater, and the highest concentrations of bicarbonate (>50 mg L^{-1}) were in freshening groundwater.

3.4 | Radium isotopes in groundwater

The activities of Ra isotopes ranged from 0.04 to 1.83 pCi L^{-1} (^{226}Ra) and 0.05 to 3.09 pCi L^{-1} (^{228}Ra). The combined Ra activity (^{226}Ra + ^{228}Ra) in all samples was below the 5 pCi L^{-1} maximum contaminant level for drinking water (USEPA, 2000). Combined activity exhibited a fairly strong positive correlation with Cl (R^2 = .66) in the well clusters southwest and west of the stormwater basin (Figure 5, Supplemental Table S6).

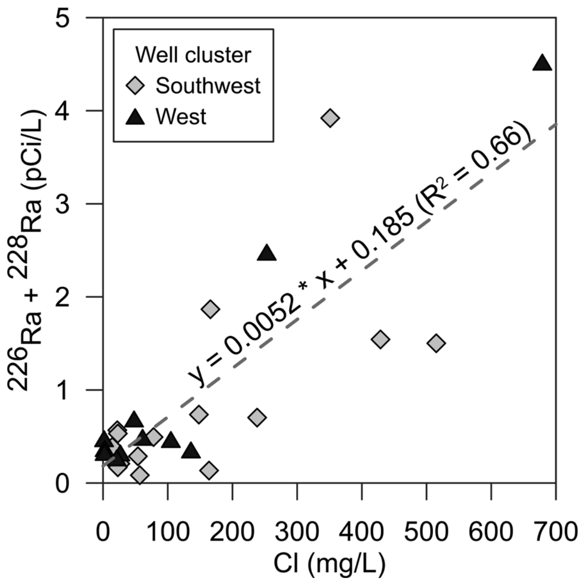

FIGURE 5 Correlation between total Ra (^{226}Ra + ^{228}Ra) and Cl concentration in groundwater from the west and southwest clusters

4 | DISCUSSION

Although sensor-derived time-series data clearly illustrate the movement of saline water under the study site, data document only what was occurring at discrete points within the aquifer. Hydraulic conductivity testing indicated that the aquifer was highly heterogeneous; the connectivity of high-permeability layers and the paths of affected groundwater are not precisely known.

4.1 | Geological controls on groundwater quality

Hydrochemistry of the aquifer at the study site varies with depth and location with respect to the basin. Upgradient wells of all depths show minimal impact of recharging salty groundwater (Figures 3 and 4). The impact of stormwater recharge on deeper formation material beneath and downgradient of the basin is dependent on aquifer heterogeneity and connectivity. Heterogeneous permeability at the site resulted in complex mixing patterns throughout the aquifer. This is consistent with the larger-scale results of Morales & Oswald (2020), who found that stormwater movement within a watershed is influenced by geology, and the results of additional site-scale investigations at their site indicated that anthropogenically influenced changes to a flow regime can cause mixing between groundwater bodies having distinct hydrochemistry (Ayotte et al., 2011; Szabo et al., 2005).

Sensor data show that deicer-impacted stormwater generally did not migrate into deeper portions of the aquifer at the site (Figure 3). The BEX classifications of deeper groundwater were generally neutral because of lack of connectivity between the shallow and deeper portions of the aquifer. However, deep groundwater at the east and basin clusters was freshening during fall 2019. We believe this is the result of non-deicer-impacted stormwater entering the basin and recharging groundwater following a period of low recharge and a 1.2 m decline in water levels. During this period of water level decline at our site, volumetric soil moisture content was low, dropping to 0.15 (median for the project duration was 0.29; DEOS, 2020), and precipitation caused mounding of the water table below the basin resulting in lateral groundwater movement. We attribute the greater proportion and lack of a seasonal pattern in elevated Na occurrences in deeper groundwater to natural groundwater–matrix reactions. Source material for the elevated Na and trace metals is glauconite in the Vincentown Formation, and Na is exchanged for Ca in groundwater. Elevated Na from ion exchange reactions between groundwater and glauconitic sediment has been historically documented in this unit in Delaware and Maryland (known as Aquia in Maryland) (Drummond, 1998).

4.2 | Stormwater quality influences ion exchange and Ra mobilization

Shallow groundwater depleted in Na as a result of Na retention in the vadose zone following deicer-impacted infiltration has been well documented (Granato et al., 1995; Robinson et al., 2017). Sun et al. (2012) found that groundwater Na/Cl molar ratios <1 can be an effective indicator of Na retention during infiltration. Shallow groundwater beneath the basin had a Na/Cl ratio of <1 during winter and >1 during summer and fall, revealing evidence of Na retention and release (Supplemental Figure S6). Deicer-impacted runoff infiltrates through the basin; some Na is retained in the vadose zone, and water that reaches the water table is elevated in Cl but depleted in Na (low-Na/high-Cl). This water then migrates along a preferred flow path, mixing with existing groundwater. Throughout the summer and fall, low-Na/low-to-moderate-Cl stormwater "flushes" Na from the vadose zone. This results in high-Na/low-to-moderate-Cl, non-winter stormwater (or stormwater that was not deicer-impacted) recharging the water table beneath the basin. These findings are consistent with previous field studies of the vadose zone and shallow groundwater (Burgis et al., 2020; Baraza & Hasenmueller, 2021).

Our conceptual model of seasonal water–matrix processes is illustrated by the mixing lines on Figure 4. The Na-depleted/Cl-enriched groundwater corresponds with a BEX classification of salinizing—native groundwater mixing with deicer-impacted stormwater (mixing line A). The Na-enriched/Cl-depleted groundwater corresponds with a BEX classification of freshening (mixing line B). For many instances of freshening groundwater, recharging stormwater was mixing with previously salinized groundwater. We show that mixing of these two water types with existing aquifer water can be tracked, and the magnitude of mixing evaluated, using BEX classifications and sensor-based continuous monitoring.

The timing of Cl and Na movement through the vadose zone to groundwater was dependent on the amount of deicer applied during the winter and timing and magnitude of subsequent precipitation. For example, after winter deicer application, winter recharge transports Cl into shallow groundwater beneath the basin, and Na is retained in the vadose zone beneath the basin. However, if there is less recharge in following months, we expect there will be less flushing of Na to the water table, creating a greater time lag between Cl and Na movement. More frequent groundwater sampling for Na is needed to determine the inter-seasonal lag time. Cl in shallow groundwater beneath the basin in 2021 did not return to pre-winter conditions before the first salt application of the following 2021–2022 winter (Figure 3). Grab shallow basin groundwater samples show that Na release persisted into the following winter season (Supplemental Figure S6). This shows that the effects of deicer can compound and persist in groundwater for multiple seasons to years, a finding similar to observations by Robinson et al. (2017) and Snodgrass et al. (2017). Without accounting for Na retention, large-scale salt budgets, which typically use Cl as a proxy for salt, will underestimate transport time of deicer through a watershed.

The correlations between combined Ra activity, ^{228}Ra/^{226}Ra, and Cl concentration in the southwest and west well clusters indicate that salinized recharge mobilizes Ra downflow of the basin (Figure 5). Given there is poor correlation between Ra and geologic unit at the site, there likely are Ra sources in both units. These correlations most likely reflect Ra desorption by competitive ion exchange with Na, and chloride complex formation (Lindsey et al., 2021). These processes would preferentially mobilize ^{228}Ra where it is enriched by alpha-recoil in the adsorbed and mobile pool of Ra in shallow aquifers (Krishnaswami et al., 1982; Luo et al., 2000). Additional evidence for the role of NaCl-rich stormwater in mobilizing Ra is the negative correlation of change in Ra activity between sampling events with δ^{18}O, indicating that water recharged by winter precipitation is associated with deicer and Ra enrichment in the aquifer (Figure 6).

Continued deicer input to the infiltration basin may, over time, deplete the Ra source. However, as chloride is chemically conservative, continued deicer impact will increase salinity in the aquifer, as has been observed in some local water supply systems (Andres & McQuiggan 2019). Further, as several studies have noted the association between increased salinity and elevated Ra in groundwater in the region (Lindsey et al., 2021; Bolton, 2000; Andres & McQuiggan, 2019), we suspect that there will be increased incidence of Ra contamination of water supply wells. This may become a significant concern where new development constructs infiltration basins in and around agricultural areas, as phosphate-based fertilizer may be a source material for Ra mobilization to groundwater (Szabo et al., 1997). Additional research is needed to better understand the mechanisms of Ra release and readsorption.

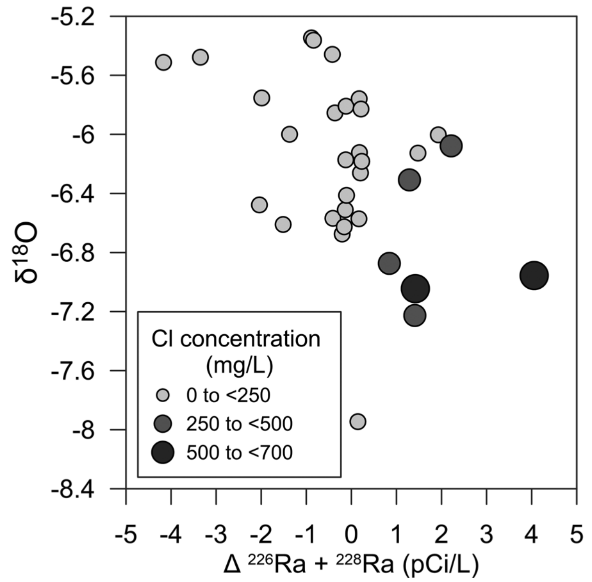

FIGURE 6 Comparison of δ^{18}O and change in total Ra (^{226}Ra + ^{228}Ra) activity in groundwater between sampling events. Only samples with +/−0.10 pCi L^{-1} change are shown. Symbol size and shading correspond with Cl concentration

4.3 | Surface physical processes impact quality of stormwater recharge

A difference in d-excess between stormwater and shallow groundwater indicated that evaporative isotopic fractionation occurred in the basin between stormwater inflow and recharge to the water table, especially during the summer and fall. This is consistent with observations and evapotranspiration estimates made at nearby meteorological stations (DEOS, 2020). This contrasts with the results of others (Baraza & Hasenmueller, 2021; Beak et al., 2020), who found no evidence of evaporative fractionation occurring during infiltration. Sites with best management practices that use engineered materials to reduce stormwater detention aboveground and increase infiltration rates, such as the permeable pavement and infiltration gallery of Beak et al. (2020), may experience less evaporation both in the basin and in the vadose zone than sites that do not use materials that promote infiltration. Additionally, sites with a shallow water table or a rapid pathway from surface to groundwater, such as the karst site of Baraza & Hasenmueller (2021), will also experience less evaporation in the vadose zone and more rapid mixing of stormwater and existing groundwater.

Direct observations indicate that evaporation is occurring before infiltration. During March–July 2020, the basin contained standing water for day-to-week stretches. Reduced infiltration rates at the surface are suspected after visual inspection of the basin surface revealed a hard, polygonal-cracked crust that is commonly associated with sediment structure degradation from elevated Na (Fujimaki et al., 2006). Evaporation of deicer-impacted stormwater will concentrate the contaminants in recharge water. Groundwater observations reflect limited dilution by subsurface mixing and dispersion, placing value on the importance of avoiding evaporation prior to infiltration to the water table.

5 | CONCLUSIONS

Groundwater chemistry results indicate Cl transport into and through the shallow aquifer beneath the basin as a result of winter deicer application to roadways. Chloride preferentially flowed through high-permeability layers. Climate variables drove temporary changes in groundwater flow pattern and solute transport to deeper portions of the aquifer. Sodium retention in the vadose zone beneath the basin occurred following deicer-impacted stormwater recharge. As freshened stormwater infiltrated through the basin during non-winter months, stored Na was released to groundwater. Sodium-depleted and enriched groundwater followed the same subsurface pathways as Cl-impacted groundwater. Deicer-impacted recharge did not dilute throughout the aquifer thickness, emphasizing the importance of management practices that reduce application. If the basin is not recharging properly, evaporation can concentrate salt in the stormwater. Alternating NaCl with other deicers, like $CaCl$, may balance out the effects of Na retention. Radium was mobilized with increasing Cl in groundwater; however, this did not necessarily correspond with elevated Cl concentrations, suggesting that, once mobilized, other geochemical factors can prevent readsorption onto aquifer solids.

ACKNOWLEDGMENTS

Funding for this project was provided by the Delaware Department of Transportation. Amanda Stevens and Sara Esposito of Delaware's Department of Transportation and Emily Whiting were integral in coordinating site access as well as providing information on road salt application and site construction. A. Roros was partially funded by a University of Delaware Water Resources Center fellowship. The authors thank P. Steve McCreary for assistance with monitoring well installation, infrastructure fabrication and set-up, Shawn Cava and Sheng Yao for their assistance with field work, and Linnea Heraty for analyzing the stable isotopes at the EIGL. We acknowledge Jarrod Miller, Charles Burgis, Paul Imhoff, and Derya Akpinar for their helpful conversations.

AUTHOR CONTRIBUTIONS

Rachel McQuiggan: Conceptualization; Data curation; Formal analysis; Investigation; Validation; Visualization; Writing-original draft. A. Scott Andres: Conceptualization; Funding acquisition; Investigation; Supervision; Writing-review & editing. Andreanna Roros: Data curation; Formal analysis; Investigation. Neil C. Sturchio: Investigation; Resources; Writing-review & editing.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

How to cite this article: McQuiggan, R., Andres, A. S., Roros, A., & Sturchio, N. C. (2022). Stormwater drives seasonal geochemical processes beneath an infiltration basin. *Journal of Environmental Quality*, 1–13. https://doi.org/10.1002/jeq2.20416