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RESEARCH ARTICLE

An integrated multivariate statistical approach for the evaluation of spatial variations in groundwater quality near an unlined landfill

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Abstract

Groundwater is a major resource for water supply in Canada, and 43 of 68 Saskatchewan municipalities rely on groundwater or combined groundwater and surface water sources. The Regina landfill is built on top of the Condie aquifer, without an engineered liner. Missing data and inconsistent sampling make a traditional groundwater assessment difficult. An integrated statistical approach using principle component analysis, correlation analysis, ion plots, and multiple linear regression is used to study groundwater contamination at the Regina landfill. Geological locations of the water samples were explicitly considered. The abundance of cations in the groundwater was $Ca^{2+} > Mg^{2+} > Na^{+} > K^{+} > Mn^{2+}$; and for anions $SO_4^{2-} > HCO_3^{-} > Cl^{-}$. Correlation analysis and ion plots pointed to gypsum and halite dissolution being the main factors affecting groundwater chemistry. Principal component analysis yielded three principal components, responsible for 80.7% of the total variance. For all monitoring well groups, the sodium absorption ratio was generally less than one. The variation in the ratio from monitoring well groups suggests possible groundwater contamination from landfill operation. Wilcox diagrams indicate groundwater near the landfill is unsuitable for irrigation. A two-step multiple linear regression was used to develop a model for total hardness prediction.

Keywords Hydrochemical analysis · Groundwater quality · Principal component analysis · Multiple linear regression · Drinking and irrigation suitability

Introduction

Groundwater is the predominant water source for consumption, domestic services, industry, manufacturing, agriculture, and almost all aspects of human life (Nagaraju et al. 2014; Spanos et al. 2015; Machiwal and Jha 2015) in many populated areas around the globe. In many Canadian municipalities, groundwater is the primary water source. For example, 43 out of 68 municipalities in Saskatchewan use groundwater or combined groundwater and surface water (Rutherford 2004). This source services about 45% of the population for washing, farming, and other domestic use in Saskatchewan (Government of Canada 2013; Canadian Municipal Water Consortium 2015; Natural Resources Canada 2017; Environment and Climate Change Canada 2017), indicating the significance of groundwater resources. The only municipal solid waste (MSW) disposal site in the Regina area is located on top of the Condie aquifer, located in Regina, Saskatchewan, which is a concern as a contamination source (City of Regina 2016).

The impacts on groundwater quality from landfill operations are widely studied in Canada and abroad (Bakis and Tuncan 2011; Van Stempvoort et al. 2011; Talalaj 2014; Han et al. 2016; Pan et al. 2017; Pan and Ng 2018). Unlike other studies on urban aquifers (Bakis and Tuncan 2011; Greis et al. 2012), the Condie aquifer examined in this study warrants a systematic statistical approach in groundwater quality assessment due to its geographical, climatic, and technical complexities. These complexities include (i) the majority of Regina MSW landfill site was built directly on native soil, a calcium-rich montmorillonite clay (Barbour and Fredlund 1989), on top of the Condie aquifer in 1960s without an engineered liner (City of Regina 2016; Bruce et al. 2017; Richter and Ng 2017; Vu et al. 2017; Bruce et al. 2018); (ii) an elevated level of salt concentrations in groundwater, probably due to the

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extensive use of road salts common in cold climates (Marsalek 2003), and (iii) missing data and inconsistent sampling time in the annual groundwater monitoring programs. As such, an integrated approach using various statistical techniques was required to assess the groundwater quality of the Condie aquifer near the landfill (Pan et al. 2017; Pan and Ng 2018).

A wide range of indices and plots are being proposed and applied in water quality studies. Talalaj (2014) proposed a new index known as the landfill water pollution index (LWPI) to quantify the overall water quality near a landfill site in Poland. Hassen et al. (2016) evaluated the groundwater quality with respect to drinking and agricultural purposes using indicators such as residual sodium carbonate (RSC), water quality index (WQI), and several chemical and isotopic fingerprints. Machiwal and Jha (2015) used box-whisker plots to establish linkages between the quality of groundwater and rainfalls. Nagaraju et al. (2014) and Gu et al. (2015) successfully used Piper plots to demonstrate the inter-relationships of various ions qualitatively. Nagaraju et al. (2014) and Hassen et al. (2016) adopted the Wilcox diagram to determine the salinity and alkalinity hazards of water and to evaluate the suitability for irrigation in India and Tunisia, respectively. In this study, Wilcox diagram and boxplot are adopted to better illustrate the water quality categories of samples, and the concentrations of water quality indicators SAR and TH.

In the literature, most groundwater quality studies ignore the spatial distribution of groundwater samples and water quality data in the vicinity of a site is analyzed collectively (Maiti et al. 2013; Machiwal and Jha 2015; Viswanath et al. 2015; Hassen et al. 2016). Unlike previous studies, the present work explicitly considers the geographical locations of the monitoring wells and the landfill footprint with respect to the regional groundwater flow pattern. Distinct groups of monitoring wells were identified, and data in the groups was integrated into various plots and compared.

Given the complexities and the site-specific conditions of the study area, an integrated statistical approach using correlation analysis, principal component analysis, and ion plots is warranted. Spatial information is modeled by strategically grouping the monitoring wells with respect to their geographical locations and the groundwater flow pattern to reveal the potential impacts from the operation of the unlined landfill. Unusual values of TH are observed at the site and a two-step multiple linear regression (MLR) is developed and verified. The objectives of this study are to (i) investigate the ion correlations of the samples and identify mineral dissolution/precipitation processes given the complexities of the site; (ii) evaluate the groundwater quality for irrigation and drinking suitability purposes using sodium adsorption ratio (SAR) and total hardness (TH) respectively; and (iii) identify the influencing ions and their inter-relationships on TH and propose a MLR prediction model. This study presents some of the first attempts at groundwater assessments for an active landfill with unlined cells using an integrated statistical and spatial approach.

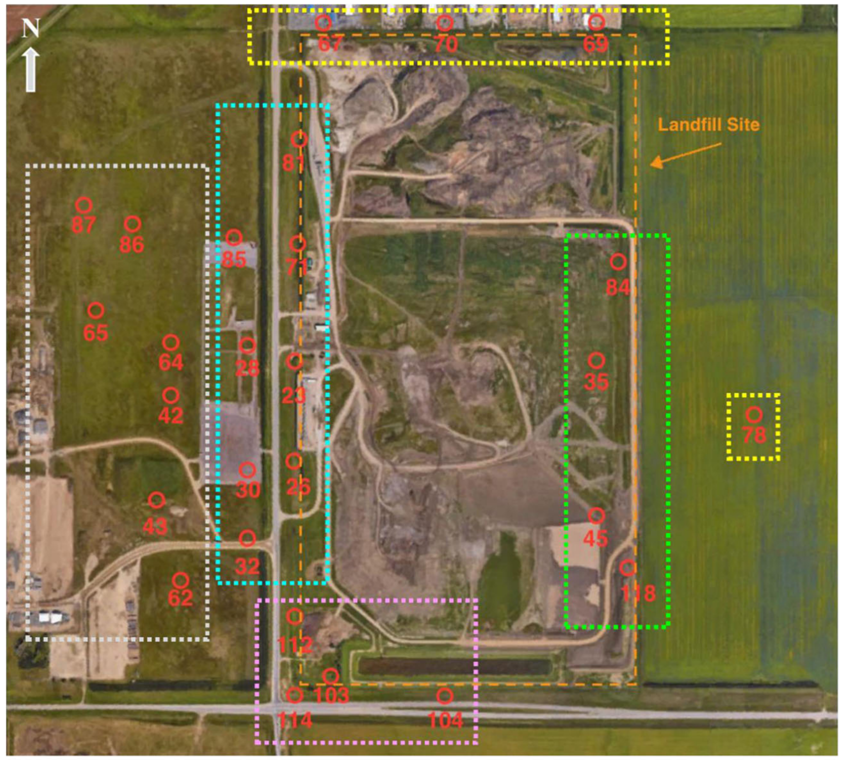
Materials and methods

Study area and site description

Located in Regina, Saskatchewan, a semi-arid region of the Canadian prairies, the Condie aquifer is one of two major aquifers near the city, providing water to the city for anthropogenic uses and industrial activities (City of Regina 2013). According to the City of Regina (2013), the groundwater quality in the Condie aquifer is affected by the lithology, geochemical processes, water-rock interactions, mineralization, and the surrounding environment such as the unlined landfill (City of Regina 2016). The Condie aquifer is located in part, under the city's sole municipal solid waste disposal facility. The Regina landfill was built in the 1960s, when engineered liners were not installed in the landfill cells (City of Regina 2016). The stratigraphy of the landfill site consists of 0.5 to 4 m of topsoil and lacustrine clay, overlying the Condie Formation, Battleford and Floral Formation's till, and Upper Floral Formation Sand and Gravel Unit (Maathuis and Van der Kamp 1986). The groundwater level is roughly eight to ten meters below the ground surface, with regional groundwater flow towards the west and northwest. A general description on the hydrogeological conditions of the site is provided by a previous study (Pan et al. in press) and is not repeated here. The majority of recharge occurs from west and northwest of the landfill, and the hydraulic conductivity in the aquifer varies from 1.4×10^{-6} m/s to as high as 2.3×10^{-3} m/s, depending on the saturated thickness and effective grain size of the soil (City of Regina 2016). A variety of materials are accepted at the landfill, including municipal waste, shingles, asphalts, concrete, fill dirt, and other materials (City of Regina 2018). The unlined old landfill cell is located at the North side of the landfill (Fig. 1).

Data source and uncertainties

Over the years, there have been concerns regarding potential groundwater contamination from landfill leachate (City of Regina 2016). The City of Regina began its groundwater monitoring program at the study area in the 1970s. Monitoring wells were installed gradually in the vicinity of the landfill. Groundwater samples were sent to a third party laboratory for analysis. Data used in this study is obtained from a series of Groundwater Monitoring



(yellow)	Background Monitoring Wells: 67, 69, 70, 78
(green)	East Monitoring Wells: 35, 45, 84, 118
(pink)	South Monitoring Wells: 103, 104, 112, 114
(cyan)	West Monitoring Wells: 23, 26, 28, 30, 32, 71, 81, 85
(grey)	Far West Monitoring Wells: 42, 43, 62, 64, 65, 86, 87

Fig. 1 Regina landfill site and groundwater monitoring wells (derived from Google Maps 2018)

Program Reports published by the City of Regina (City of Regina 2012, 2013, 2014, 2015, 2016). In 2013 and 2014, only limited data was reported due to well maintenance, well drying or decommission (City of Regina 2014, 2015). Only 12 and 9 monitoring well data were reported in 2013 and 2014, respectively. Given the objectives of the study, only data in 2011, 2012, and 2015 are included. In any given year, a data coverage greater than 93.5% is attained. Sample collection was conducted biannually (2011, 2015) or annually (2012).

Some of the monitoring wells were decommissioned or built during the study period and were thus ignored. As such, only 27 monitoring wells were included in the present study (ID# 23, 26, 28, 30, 32, 35, 42, 43, 45, 62, 64, 65, 67, 69, 70, 71, 78, 81, 84, 85, 86, 87, 103, 104, 112, 114, and 118), as shown in Fig. 1. In order to investigate the potential impacts from the unlined landfill, the 27 selected monitoring wells were categorized into five groups according to their locations: Background group—monitoring wells 67, 69, 70, and 78; East group—monitoring wells 35, 45, 84, and 118; South group—monitoring wells 103, 104, 112, and 114; West group—monitoring wells 23, 26, 28, 30, 32, 71, 81, and 85; and Far West group—monitoring wells 42, 43, 62, 64, 65, 86, and 87. The background group is located upstream outside the footprint of the landfill. The East group is located within the landfill area along the East landfill boundary. The South and West groups are located immediately downstream of the landfill. The far west group is located furthest away from the landfill downstream.

Parameters and indicators

Selection of target parameters

Wide varieties of hydrochemical and physical parameters of the samples were reported by the City, including pH, salts, trace metals, and volatile organic compounds. In the present study, a total of 14 parameters are selected, including eight trace metals: arsenic (As), calcium (Ca), magnesium (Mg), manganese (Mn), potassium (K), sodium (Na), and uranium (U), as well as seven ionic species and groundwater parameters: bicarbonate (HCO_3^-), chloride (Cl^-), sulfate (SO_4^{2-}), total dissolved solids (TDS), total hardness (TH), pH, and electric conductivity (EC). The 14 chemical and physical parameters are carefully selected based on their magnitudes, ranges, data availability, and health and safety concerns (Saskatchewan Ministry of Environment 2016; Health Canada 2017).

Water salinity and irrigation use

Sodium ions can be released from groundwater-rock interactions, the use of sodium-rich fertilizer and road salts, and many other processes (Maiti et al. 2013). High salinity in groundwater, however, reduces its usefulness as irrigation water, and may also impact the health of human receptors (Vineis et al. 2011; Maiti et al. 2013; Talukder et al. 2016). Water-use-efficiency decreases as the salinity increases in agricultural water and causes adverse impacts such as reducing crop root water uptake (Wang et al. 2017). Also, soil-salt interactions may disperse negatively charged clay particles and destabilize the soil structure, leading to yield loss (Ishaku et al. 2011). High sodium bicarbonate concentrations, for instance, can cause dissolution of organic fertilizers, weaken the physical properties of soil, and render the soil unsuitable for growing plants (Foster et al. 2008; Hassen et al. 2016; Pan et al. 2017). Salinity is an important indicator when assessing the irrigation suitability of water. In this paper, sodium adsorption ratio (SAR) is used to represent and measure salinity, which is defined as (Karanth 1987):

$$\mathrm{SAR} = \frac{\mathrm{Na}^{+}}{\sqrt{\frac{\mathrm{Ca}^{2+} + \mathrm{Mg}^{2+}}{2}}} \quad (1)$$

where all the parameters are in milliequivalents per liter (meq/L).

Wilcox diagrams are used to examine the suitability of water for irrigation uses and to classify water into different suitability levels (Wilcox 1955; Nagaraju et al. 2014). Two parameters are required to plot Wilcox diagrams: electric conductivity (EC) and sodium percentage (SP). Electric conductivity refers to the capacity of groundwater to conduct electricity. High electric conductivity represents a high concentration of dissolved metal cations and metal salts. Sodium percentage (SP) is used to signify the relative concentrations of sodium and potassium among other metal cations. Sodium percentage is defined by Wilcox (1955) as:

$$\mathrm{Na}\ (\%) = \frac{\mathrm{Na}^{+} + \mathrm{K}^{+}}{\mathrm{Na}^{+} + \mathrm{K}^{+} + \mathrm{Ca}^{2+} + \mathrm{Mg}^{2+}} \times 100\% \quad (2)$$

all parameters are in meq/L.

Total hardness and water quality for drinking purposes

Total hardness (TH) is typically expressed as equivalent concentrations of calcium carbonate ($CaCO_3$). Although there is no evidence showing immediate adverse health effects, hard water causes scaling and damages water supply infrastructure, affects the taste of water, and reduces the ability to produce foam (Ishaku et al. 2011). Health Canada classifies water by its calcium carbonate concentration in four levels: soft—0 to 60 mg/L; medium hard—60 to 120 mg/L; hard—120 to 180 mg/L; and very hard—greater than 180 mg/L (Health Canada 1979). Boxplots are used in this study to examine the scattering and the skewness of the TH data with respect to the spatial distributions of the wells.

An integrated spatial and statistical approach

Correlation coefficients, ion exchange processes

The relative concentrations of the ionic species in groundwater reveal the chemical properties of the aquifer, the natural environment, and other nearby industrial activities. The correlation coefficient (CC) is used to identify the correlation between major cations and anions. A coefficient $> |0.75|$ is regarded as "strongly correlated" (Selvakumar et al. 2017) and is adopted in this study.

By plotting major cations such as calcium, magnesium, and sodium against bicarbonate, sulfate, or chloride, and by studying the data distribution patterns with ratio lines (such as the 1:1 ratio line), various ion exchange, dissolution, and mineralization processes in the aquifer can be identified and studied statistically.

Principal components and loadings

In this study, PCA is used not only as a dimensional reduction technique but also as a tool to study the association among variables. The analysis extracts eigenvalues from the original set, to form new principal components (PC) which are orthogonal, and therefore unrelated, to each other (Ravikumar and Somashekar 2017; Abou Zakhem et al. 2017). Each PC explains part of the total variance, and typically only a few PCs are required to explain the majority of the variance. Only PCs with eigenvalues greater than one are considered significant in this study (Cattell and Jaspers 1967). Loadings larger than 0.5 (Abou Zakhem et al. 2017; Selvakumar et al. 2017) are grouped together to ensure a significant correlation of the variables within a given group. Varimax rotation and Kaiser Normalization are conducted using MATLAB (v. R2016a) and SPSS (v. 25).

Multiple linear regression

Multiple linear regression is a technique to model the relationship between two or more independent variables and a dependent variable, by generating a linear equation (Bingham and Fry 2010a, b). A dual-step MLR model is developed for TH using SPSS (v. 25). In the first regression, all 13 parameters were selected as independent variables and a confidence interval of 95% was applied (Civelekoglu et al. 2007; Viswanath et al. 2015). Only statistically significant parameters in the first regression ($p < 0.05$) were used as independent variables for the second regression to build the final TH model.

Results and discussion

Hydrochemical and physical parameters

Table 1 compares the parameters with their respective guideline values. On average, the abundance of ion concentrations in this study is in the order of $Ca^{2+} > Mg^{2+} > Na^{+} > K^{+} > Mn^{2+}$ for cations, and $SO_4^{2-} > HCO_3^{-} > Cl^{-}$ for anions. The observed trends from 2011, 2012, and 2015 were consistent with previous findings at the Regina landfill site when only 2015 data were considered (Pan et al. 2017; Pan and Ng 2018). No significant changes in groundwater chemical composition are identified at the study area, at least from 2011 to 2015. Unlike other cations, the average Mg^{2+} concentration (3.7 mg/L) is noticeably higher than the guideline value. It is also interesting to note that the standard deviation of the Na^{+} concentration is relatively high (60.3 mg/L). The mean values of SO_4^{2-}, TDS, and TH all exceeded the guideline values.

Correlation of parameters

Correlation coefficients (CC) of the parameters are presented in Table 2 to investigate the possible soil-water interactions. Correlation coefficients greater than |0.75| are regarded as "strongly correlated" (Selvakumar et al. 2017) and are shown in bold. For instance, sulfate is strongly and positively correlated with calcium and magnesium (CC = + 0.90 and + 0.85, respectively), it is, therefore, logical to assume the possible dissolution of gypsum ($CaSO_4 \cdot 2H_2O$). Moreover, bicarbonate and calcium are mildly correlated (CC = + 0.55), suggesting that calcite may not be the sole source of calcium. As reported by Barbour and Fredlund (1989), Regina clay in the area is predominantly Ca-rich montmorillonite. The strong correlation between sodium and chloride (CC = + 0.93) suggests halite dissolution may be one of the major chemical reactions affecting water chemistry near the landfill site. Similar results were reported in other aquifers (Reddy 2013; Nagaraju et al. 2014; Hassen et al. 2016). Calcium and magnesium also have a strong correlation (CC = + 0.94), suggesting the possible dissolution of dolomite ($CaMg(CO_3)_2$). Similar to other studies (Khanna 2015; Abou Zakhem et al. 2017), both calcium and magnesium have strong and positive correlations with TDS and TH (all CC > + 0.94). Correlation analysis results support the use of Ca^{2+} and Mg^{2+} to model TDS and TH.

Soil-water interactions

The relative ionic strength of major cations and anions are plotted in order to study ion exchange and chemical dissolution processes. A plot of calcium versus sulfate (Fig. 2) shows a linear relationship ($R^2 = 0.91$), indicating the possible dissolution of gypsum ($CaSO_4 \cdot 2H_2O$) or anhydrite ($CaSO_4$). However, over 80% of the data points are located below the

Table 1 Magnitude and variability of the target parameters and the guideline values

Tested items	Canadian guidelines/standards	All monitoring wells			
Trace metals		Max	Mean	Min	STD
Arsenic	0.01	0.028	0.006	0.0003	0.006
Calcium	No value[a]	500	319	100	106
Magnesium	No value[a]	220	116	32	47
Manganese	0.05 (0.1[b])	190	3.7	0.06	21.1
Potassium	No value[a]	49	13	5	8.4
Sodium	200	360	62	16	60.3
Uranium	0.02	0.054	0.016	0.0054	0.0
General parameters					
Bicarbonate	No value[a]	850	415	290	100
Chloride	250	560	62	1.3	99
Sulfate	250	1800	995	170	420
Total dissolved solids	1000	3400	1779	500	693
Tot. hardness $CaCO_3$	500	2000	1277	390	451
Lab pH	6.5–8.5	8.2	7.8	7.27	0.2
Lab conductivity (μs/cm)	No value[a]	4800	2262.9	750	825.9

[a] "No value" represents health-based guideline not available for drinking water (WHO, 2011)

[b] Value from WHO guidelines (WHO 2011). Other values are adopted from Health Canada (2017)

Table 2 Correlation coefficient matrix of the 14 target parameters

	As	Ca	Mg	Mn	K	Na	U	HCO_3	Cl	SO_4	TDS	TH	pH	EC
As	1													
Ca	− 0.24	1												
Mg	− 0.11	*0.94*	1											
Mn	0.05	0.20	0.19	1										
K	0.18	0.02	0.12	0.01	1									
Na	0.30	0.42	0.59	− 0.02	0.38	1								
U	0.07	0.67	0.71	0.08	0.16	0.60	1							
HCO_3	0.23	0.55	*0.75*	0.01	0.24	*0.79*	0.66	1						
Cl	0.19	0.41	0.57	− 0.03	0.34	*0.93*	0.50	*0.82*	1					
SO_4	− 0.16	*0.90*	*0.85*	0.05	0.04	0.46	0.71	0.49	0.36	1				
TDS	− 0.10	*0.95*	*0.96*	0.08	0.13	0.64	*0.76*	0.72	0.61	*0.90*	1			
TH	− 0.19	*0.96*	*0.94*	0.19	0.06	0.48	0.69	0.59	0.46	*0.86*	*0.94*	1		
pH	0.05	− 0.56	− 0.55	− 0.12	0.20	− 0.31	− 0.43	− 0.42	− 0.29	− 0.51	− 0.60	− 0.57	1	
EC	− 0.04	*0.88*	*0.94*	0.03	0.17	*0.75*	0.73	*0.79*	0.73	*0.84*	*0.97*	*0.88*	− 0.58	1

Note: Strong correlations with coefficient > |0.75| are shown in italics

TDS total dissolved solids

TH total hardness

EC electrical conductivity

1:1 ratio line, and the slope of the best-fit line is less than unity. The sulfate concentration is generally higher than the calcium concentration, with excess sulfate possibly originating from other sources. This is especially true in higher concentration ranges ($Ca^{2+} > 10$ meq/L). A plot of calcium and magnesium versus sulfate (Fig. 3) shows a linear pattern ($R^2 = 0.91$) with a best-fit line slope of 1.01, suggesting possible dissolution of kieserite (main component: $MgSO_4 \cdot H_2O$). Most data points

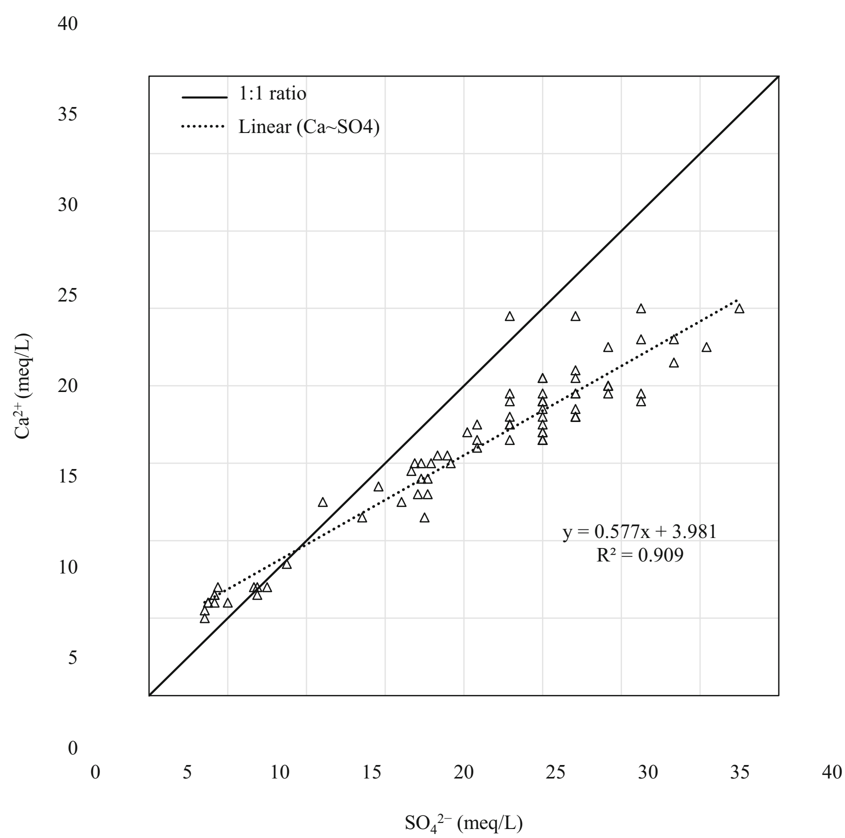
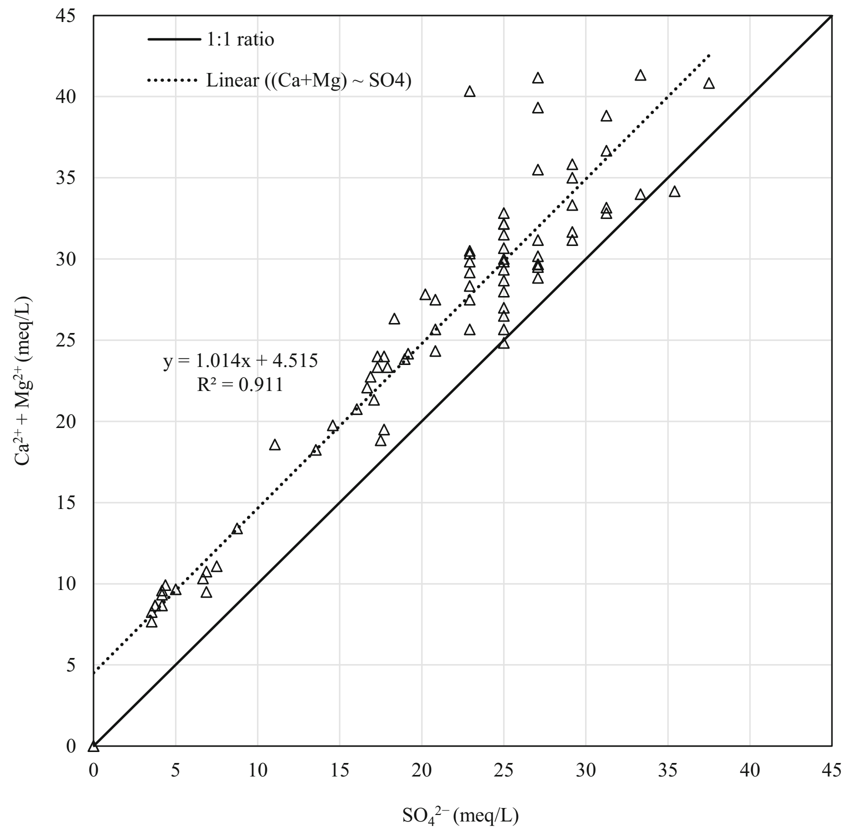


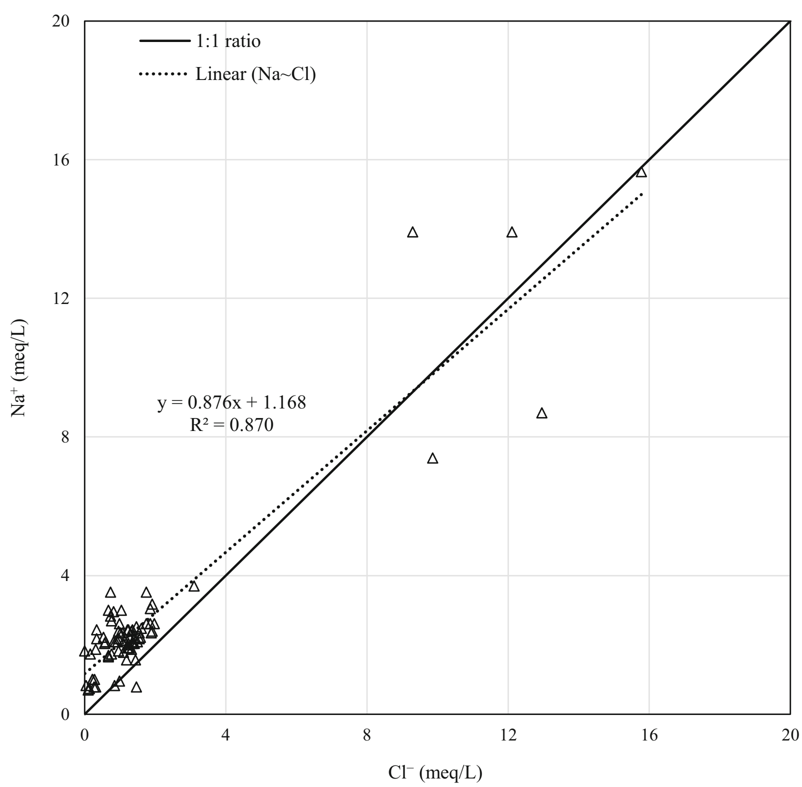
Fig. 2 Ion plot of calcium versus sulfate

Fig. 3 Ion plot of calcium and magnesium versus sulfate



are located on top of the 1:1 line, and with more data scattering in a higher concentration range ($Ca^{2+} + Mg^{2+} > 35$ meq/L), it suggests a more complicated mineralization process (Hassen et al. 2016). Figure 4 shows the concentration of sodium with

Fig. 4 Ion plot of sodium versus chloride



respect to chloride. A slightly weaker linear trend ($R^2 = 0.87$, slope = 0.88) is observed, probably due to the absence of mid-range data. More data points are located in the lower range due to the relatively low concentration in most samples. Data scattering is more pronounced in higher concentration ranges (with Na^+ or $Cl^- > 4$ meq/L). More data points are located on the top of the 1:1 ratio line, suggesting multiple sources of sodium. Results suggest halite dissolution may be the main source of salinity; however, the use of road salts in the region (Marsalek 2003) may be another reason for salinity. More work is required to confirm the exact source or combination of sources. Plots of Ca^{2+} + Mg^{2+} against HCO_3^- + SO_4^{2-} in Fig. 5 also shows a linear pattern ($R^2 = 0.95$) which is close to 1:1 line (slope = 0.91), suggesting the possible dissolution of dolomite, gypsum, and calcite (Hassen et al. 2016). More data points are located below the 1:1 ratio line, indicating an excessive concentration of bicarbonate and sulfate over calcium and magnesium, which may lead to ion exchange.

Principal component analysis

Principal component analysis is often implemented for water sample classification, cluster feature identification, as well as sample groups' hydrochemical property comparisons (Lucas and Jauzein 2008; Villegas et al. 2013). Depending on the variables and specifications of the study areas, normally three to four principal components are identified after conducting PCA. For instance, Han et al. (2014) identified three PCA groups on groundwater quality in Zhoukou, China, and found that landfill leachate had a statistically greater impact on groundwater quality in winter seasons. Jiang et al. (2015) used PCA to reduce the number of parameters from 22 to 4 and studied the concentration of arsenic in groundwater by applying various statistical techniques. In this study, a total of three principal components are obtained from the 14 input parameters (Table 1). Table 3 provides the coefficients of parameters for each component in descending order.

The first principal component (PC1) takes 49.6% of total variance, suggesting that nearly half of the set can be statistically represented by PC1 alone. The second principal component (PC2) contains five parameters and explains 23.5% of the total variance. The third principal component (PC3), which contains only one parameter (Mn^{2+}), explains 7.6% of the total variance. These three principal components explain 80.7% of the total variance, providing adequate representation of the set (Hu et al. 2013; Jiang et al. 2015; Hassen et al. 2016).

PC1 comprises eight parameters, however, the parameters that are strongly correlated (coefficient > |0.75|) include three types of parameters: (i): physical parameters (electric conductivity, total dissolved solids, and total hardness); (ii) major metallic cations (calcium and magnesium); and (iii) the anion

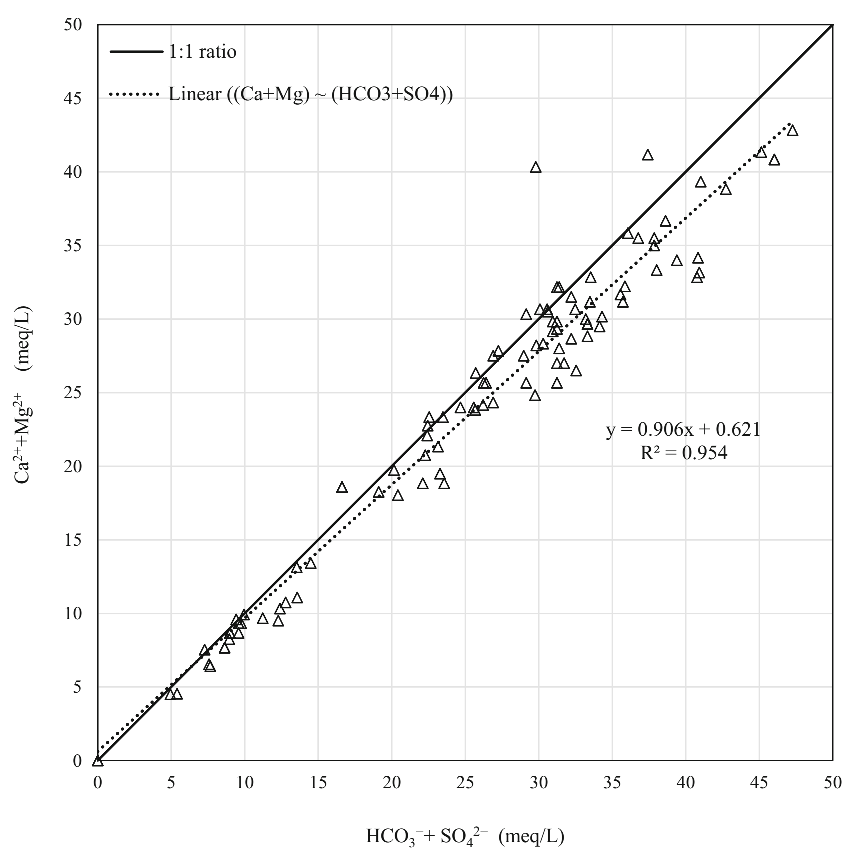


Fig. 5 Ion plot of calcium and magnesium versus bicarbonate and sulfate

Table 3 Rotated component matrix and principal components

Parameters	Principle component		
	1	2	3
Ca	*0.973*	0.073	0.053
TH	*0.967*	0.157	0.060
SO_4	*0.965*	0.079	− 0.075
TDS	*0.938*	0.322	− 0.036
Mg	*0.926*	0.298	0.066
EC	*0.870*	0.459	− 0.082
U	*0.686*	0.408	0.045
pH	*− 0.661*	− 0.044	− 0.114
Na	0.398	*0.857*	− 0.068
Cl	0.372	*0.832*	− 0.110
HCO_3	*0.542*	*0.727*	− 0.015
K	0.052	*0.608*	− 0.024
As	− 0.335	*0.595*	0.362
Mn	0.156	− 0.071	*0.935*

Note: Only coefficients > \|0.5\| are considered significant and italicized

sulfate, which can be combined with calcium and magnesium to form highly dissolvable salts and minerals, including gypsum ($CaSO_4·2H_2O$), anhydrite ($CaSO_4$), and cranswickite ($MgSO_4·4H_2O$). PC2 includes mainly two types of parameters: (1) metallic cations such as sodium and potassium, and (2) major anions such as chloride and bicarbonate. Similar to PC1, the cations and anions in PC2 can easily form highly dissolvable salts and minerals include sylvite (KCl), nahcolite ($NaHCO_3$), and halite (NaCl) in a normal range of pH. This finding is consistent with the results from the correlation matrix (Table 2). PC3 contains only one parameter, manganese, suggesting the possibility of multiple sources of manganese near the site. Some highly dissolvable salts such as gypsum, anhydrite, and halite are also found in other groundwater studies (Hassen et al. 2016).

The results from the correlation matrix, ion plots, and PCA provide an integrated assessment on the hydrochemical properties of the aquifer. Minerals such as gypsum and halite dissolution are identified as the dominant process which may affect the groundwater chemistry.

Salinity hazard and irrigation suitability

In this study, Sodium Absorption Ratio (SAR) is used to evaluate the salinity of water samples during the study period. Figure 6 compares the SAR with respect to different spatial groups. In Fig. 6, the central bar of the boxplot indicates the median, the circle symbol represents the mean value, and the bottom and top edges of the box represent the first and third quantile (25th and 75th percentile, denoted as Q1 and Q3, respectively) of each group. Whiskers outside of the box represent the maximum and minimum values.

It is clear from Fig. 6 that the Background group, located immediately upstream of the landfill, shows the lowest mean and median SAR values. The mean and median values are approximately equal and the data points are narrowly spread with a normal distribution. A similar data spread pattern is observed in the Far West group, located downstream furthest away from the landfill site. The magnitude of SAR in this group is comparable to the Background group (median = 0.51 mg/L), with a narrowly spread data set (Q1 = 0.50 mg/L, Q3 = 0.59 mg/L).

The East group has a slightly higher median SAR value than the Background group, with a reasonable spread of data. The South group (median = 0.685 mg/L, Q1 = 0.58 mg/L, Q3 = 0.79 mg/L) and West group (median = 0.677 mg/L, Q1 = 0.56 mg/L, Q3 = 0.86 mg/L) are both located immediate downstream of the landfill, and have the highest median values and data spread among the groups. The mean SAR value of the West Group is noticeably greater than the respective median value, indicating the data set is heavily skewed. It is believed that a number of overestimates might be responsible for the upward skew of the West group, although no evidence is available to verify this claim. With the exception of the West group, insignificant variability exists in SAR values at a given group and between groups. For all groups, the mean SAR value is generally close to, or less than, unity. Although the SAR values are found to be negligible near the site, differences among the groups suggest possible groundwater contamination from the operation of the unlined landfill.

Classification of water based on sodium percentage can be represented by a Wilcox diagram (Nagaraju et al. 2014; Hassen et al. 2016). As shown in Fig. 7, most water samples fell between "Good to Permissible" (17.7%) and "Doubtful to Unsuitable" (62.0%) zones. One "unsuitable" sample is observed due to the excessive sodium percentage and EC. In total, 50 out of 79 samples (63.2%) exceeded the permissible EC of 2000 ms/cm. Results from the Wilcox diagram indicate that groundwater in the Condie aquifer is generally not suitable for direct irrigation due to high salinity. Filtration and dilution of groundwater are recommended to reduce water salinity for irrigation purposes.

Total hardness and drinking suitability

Total hardness is a measurement of the concentration of calcium and magnesium ions, expressed in terms of calcium carbonate concentration (Selvakumar et al. 2017). Similar to the SAR results, box plots (Fig. 8) are used to study the data distribution from different spatial groups due to the skew of the data. In Fig. 8, the central line of the box plot and the circle symbol represent the median and mean value, respectively. The bottom and top edges of the box represent the first and

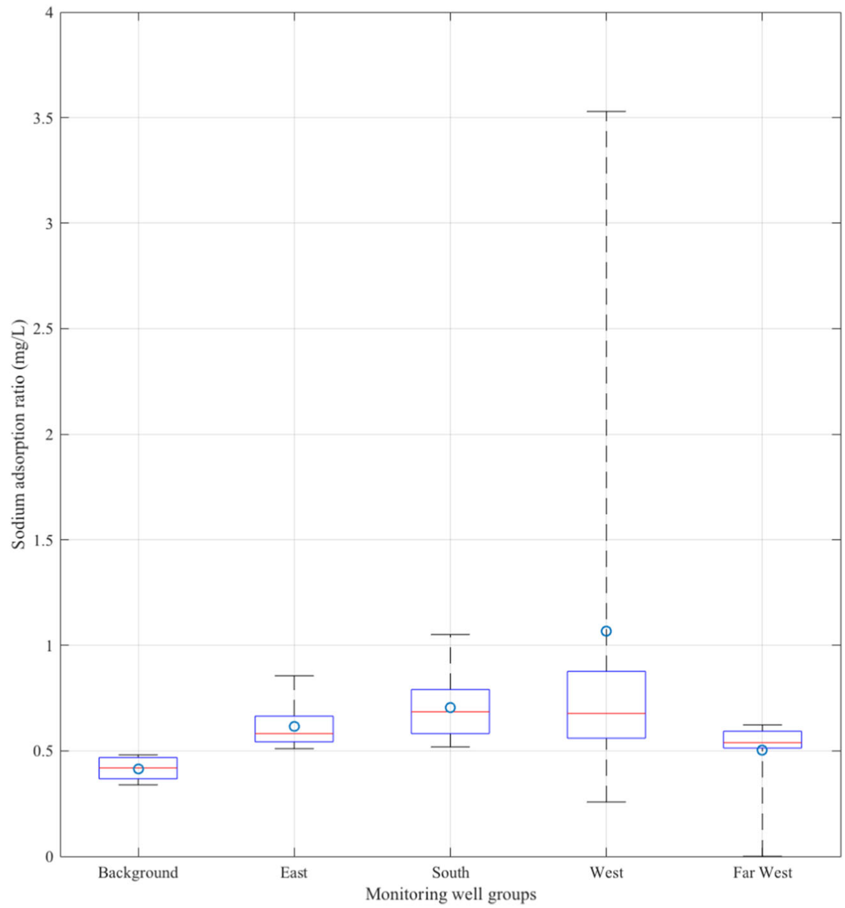


Fig. 6 Boxplot of SAR with respect to monitoring well spatial location

third quantile (25th and 75th percentile) of each group. Whiskers represent the maximum and minimum values. With the exception of the East and West groups, significantly different mean and median values are observed due to the skew in the distribution of the TH data. Unlike SAR data (Fig. 6), significant variability exists at a given group and between groups (Fig. 8).

The median TH concentration in the Background group is 455 mg/L and is less than the mean due to the skew of the data. An extreme TH value of 1500 mg/L was observed during the study period. Median values from all non-background groups, however, have exceeded the guideline value of $CaCO_3$ = 500 mg/L (Table 1). In East group, the median value of TH is 1600 mg/L, slightly higher than the West group and Far

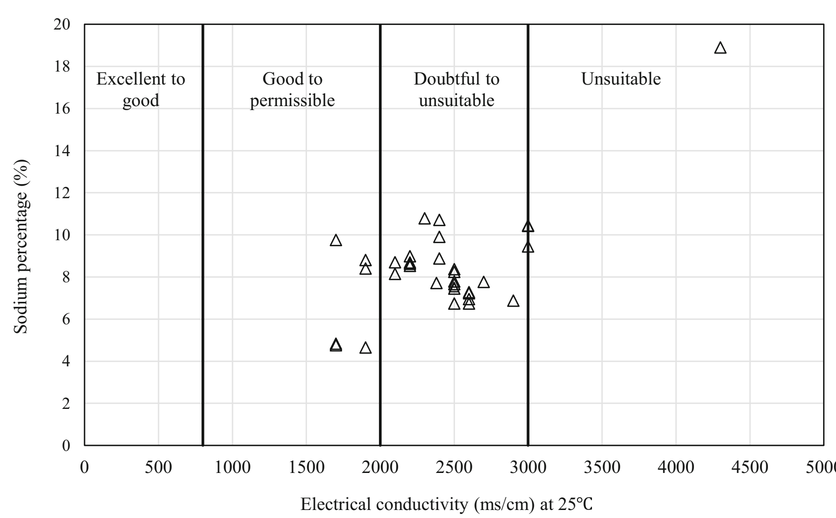


Fig. 7 Wilcox diagram for samples collected near the Regina landfill from 2011 to 2015

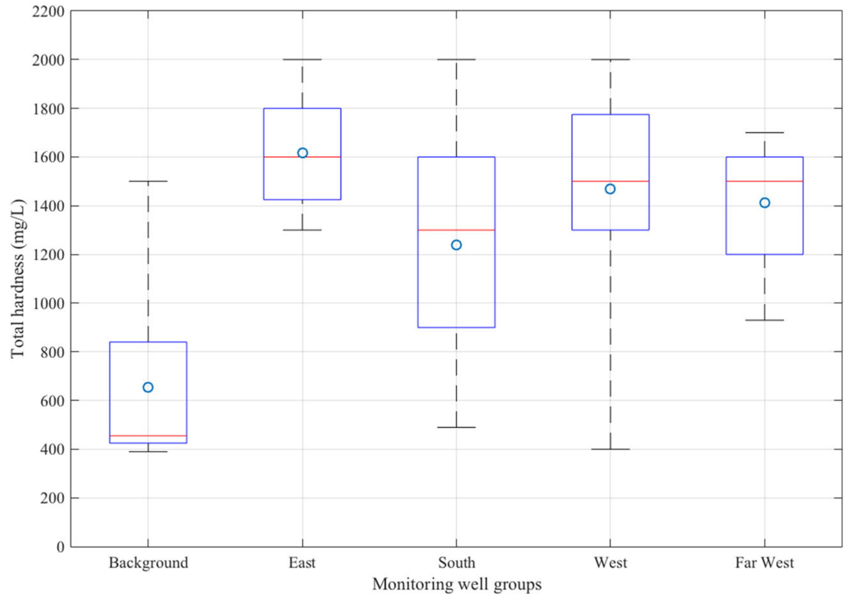


Fig. 8 Comparison of total hardness with respect to spatial locations from 2011 to 2015

West group (median of both groups = 1500 mg/L), and higher than the South group (median TH = 1300 mg/L). Both the South and West groups are located immediately downstream of the landfill. A significant spread of TH data is observed in the South group (Q1 = 900 mg/L, Q3 = 1600 mg/L), followed by the West group (Q1 = 1300 mg/L, Q3 = 1775 mg/L).

In comparison to other studies, both the magnitude and range of TH in the present study are significantly greater, likely due to the duration of the monitoring period (2011–2015) and other site-specific considerations such as the operation of the unlined landfill and the heavy use of road salts in winter seasons (City of Regina 2016). Missing data at the site complicates the analysis and poses further challenges to the modeling of the target parameter for conventional approaches. For example, TH concentrations at the present site (< 200 ha) ranged from 390 to 2000 mg/L with a standard deviation of 451 mg/L. For comparison, the average value of TH from 78 wells at an 18,000-ha Chinese site was 360 mg/L with a standard deviation of 147 mg/L (Gu et al. 2015), and the range of TH values from 30 wells at a 12,000-ha site in India was 9.3 to 180.2 mg/L (Viswanath et al. 2015).

In comparison to the guidelines, TH concentrations at the Regina landfill are also noticeably greater (Table 1). TH concentrations from non-background groups are at least 0.86 to 3 times greater than the guideline concentration. An MLR model on TH is developed to explore the elevated TH values and to investigate the influencing parameters and their respective chemical interactions.

Multiple linear regression and prediction model

Multiple linear regression is used in many recent groundwater assessment studies, which mostly focus on the prediction of water elevation or recharge modeling (Sahoo and Jha 2013; Mogaji et al. 2015; Ebrahimi and Rajaee 2017; Salem et al. 2017) or contaminant concentrations (Cho et al. 2011; Arora and Reddy 2014; Brix et al. 2017). The use of MLR on the prediction of physical groundwater parameters such as TH is very limited. A study in India (Viswanath et al. 2015) developed an MLR model for total dissolved solids using concentrations of seven ionic species, however, failed to verify the model accuracy using actual data. In the present study, a statistically significant MLR model of TH is developed, and the model is verified with observed data. A two-step multiple linear regression on TH is conducted and verified in this study. In the first regression, TH is selected as the dependent variable, and all 13 remaining parameters (As, Ca, Mg, Mn, K, Na, U, HCO_3, Cl, SO_4, TDS, pH, and EC) are used as independent variables. Similar to other groundwater studies, a confidence interval of at least 95% is used (Civelekoglu et al. 2007; Viswanath et al. 2015). Three ions are identified as significant factors in the regression model: Ca^{2+} ($p < 0.0001$), Mg^{2+} ($p < 0.0001$), and Cl^- ($p < 0.02$). The remaining ten parameters with p values greater than 0.05 are excluded. The significance of the model is verified using ANOVA, with a p value < 0.0001, and the fitted linear model adequately describes the data set with an adjusted coefficient of determination (R_{adj}^2) > 0.99. A second MLR is then conducted using Ca^{2+}, Mg^{2+}, and Cl^- as independent variables and TH as an independent variable. The significance of three selected parameters in the MLR model is all acceptable: Ca^{2+} with $p < 0.0001$, Mg^{2+} with $p < 0.0001$, and Cl^- with $p = 0.01$. The R_{adj}^2 of the three-parameter linear model is 0.995, with a p value < 0.0001 from the F test. The prediction equation of total hardness obtained by the dual-step MLR is:

$$\mathrm{TH} = 2.393 \times Ca^{2+} + 4.502 \times Mg^{2+} - 0.136 \times Cl^{-} - 1.793 \quad (3)$$

The equation is derived for ion ranges of Ca^{2+}: (100~500 mg/L); Mg^{2+}: (32~220 mg/L); Cl^{-}: (1.3~560 mg/L). All parameters are in mg/L. Unlike Ca^{2+} and Mg^{2+}, the coefficient of Cl^{-} is negative (− 0.136) for TH. This is probably due to the electrical attraction between the negatively charged Cl^{-} and the cations. Similar studies were conducted regarding the concentration of TDS and found that both calcium and chloride are correlated to TDS (Viswanath et al. 2015; Selvakumar et al. 2017).

Predicted TH using the MLR model are plotted against the observed values (Fig. 9). The fitted line (slope = 0.995) compares well to the 1:1 line (gray line). The adequacy of the linear model is confirmed with an $R^2 = 0.995$, for the range from 390 mg/L to 2000 mg/L. Predictions in extreme ranges (TH concentrations < 700 and > 1500 mg/L) are comparable with the mid-range values, demonstrating the robustness of the proposed linear model. Results suggest that the two-step MLR model adequately describes TH using data in this study.

Conclusion

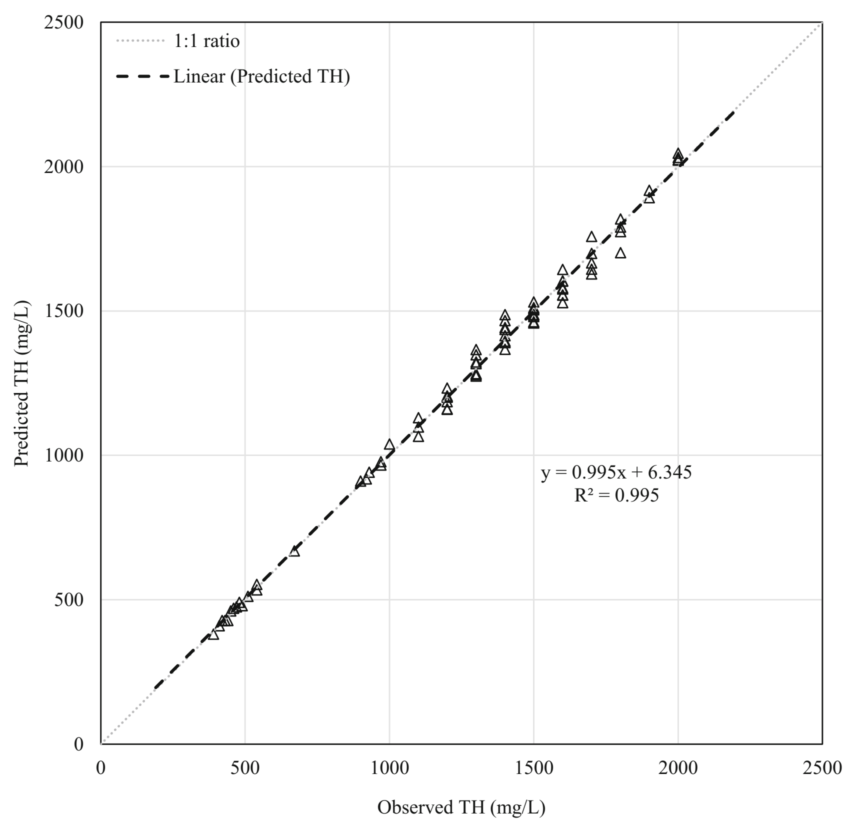
Hydrochemical groundwater properties of an urban aquifer were systematically assessed, using 3 years (2011, 2012, and 2015) of water samples from 27 monitoring wells near an unlined landfill site. The abundance of the cations are in the order of $Ca^{2+} > Mg^{2+} > Na^{+} > K^{+} > Mn^{2+,}$ and abundance of the anions are in the order of $SO_4^{2-} > HCO_3^{-} > Cl^{-}$.

A strong correlation exists between sulfate and calcium (CC = + 0.90), sodium and chloride (CC = + 0. 93), as well as sulfate and magnesium (CC = + 0.85). Calcium and magnesium are also strongly correlated (CC = + 0.94). Ion plots revealed the correlation of the ions and suggested the water chemistry of the aquifer is affected by several gypsum, halite, and dolomite dissolution processes. The principal component analysis identified three principal components, PC1, PC2, and PC3, which represents 49.6%, 23.5%, 7.6% of total variance, respectively. The three components adequately represent over 80.7% of total variance.

Over 63.2% of the samples fell into categories of “Doubtful to unsuitable” and “unsuitable” using the Wilcox diagram, indicating the unsuitability of the groundwater for irrigation purposes. With the exception of the West group, insignificant variability exists in SAR values at a given group and between groups. The box plots revealed the spread of the TH data, and they are generally skewed. TH from non-background groups is significantly greater than the drinking water guideline (500 mg/L), rendering the water unsafe for human consumption.

Results from a dual-step multiple linear regression model ($R^2 > 0.99$) suggested that calcium and magnesium are

Fig. 9 Comparison between the observed TH and the predicted TH using the proposed MLR model



positively correlated to the concentration of TH, whereas chloride is negatively correlated. The TH values of the regression model were verified by the actual data.

It is found that the integrated spatial multivariate statistical approach adopted in this study has provided a comprehensive assessment of the groundwater quality near an unlined landfill site.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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