Spatial-Temporal Analysis of Dissolved Metal Pollutants near an Unlined Municipal Landfill in a Semi-Arid Climate

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Abstract

Groundwater contamination in urban aquifers is a major concern in relation to domestic water uses and may lead to deterioration in surface water quality. Most modern sanitary landfills are designed and operated in a way that minimize impacts of leachate on groundwater. Engineered liners have not been installed in the key landfill cell at the Regina landfill, located in Saskatchewan, and there is field evidence of groundwater pollution due to the operation of the sanitary landfill. The City of Regina landfill is located on top of an urban aquifer and the City has implemented a comprehensive groundwater monitoring program. In this study, data was collected from 5 groups of wells located upstream and downstream of the landfill in 2012 and 2015. Statistical analysis shows that in 2015, Manganese (Mn), Uranium (U), Arsenic (As) and Iron (Fe) exceeded the Saskatchewan drinking water quality standard limits. However, in 2012 other than the aforementioned heavy metals, Cadmium (Cd) concentration was also above the guideline. Mn concentration increased from 2012 to 2015 while As and Fe decreased in the same period. Statistically significant positive correlations were observed between Al, B and As; Ni and Cd; Ca and U and Mn; Zn and Cu in 2015. Two linear regression models are developed for pH and dissolved Mn in groundwater.

Keywords: Urban aquifer; Dissolved metals; Groundwater pollution; Correlation matrix; Regression analysis, Semi-arid climate

Introduction

Permanent disposal in sanitary landfills is one of the most common solid waste treatment methods in Canada [1,2]. The major concern with the use of landfill technology is the generation of landfill gas and leachate during anaerobic decomposition of buried organics. Due to the abundance of closed and active landfills, Canadian studies on modelling of landfill gas and leachate are not uncommon [3-5]. Landfill leachate, if not properly managed, can percolate through soil formations and contaminate groundwater [6-8]. Specifically, elevated levels of heavy metal concentrations are toxic to many living organisms and is a major threat to groundwater quality [9-12]. Groundwater is the main water source in Canada. In Saskatchewan, about 45% of the population rely on groundwater for municipal, domestic and rural use [13,14]. As such, residents of Saskatchewan are especially vulnerable to groundwater contamination due to landfill leachate. The 60ha unlined landfill cell in the City of Regina, the capital city of Saskatchewan, has been in service since 1961. The City implemented a groundwater monitoring program at the proximity of the site. However, frequent missing data and irregular sampling frequency make interpretation of data difficult

[3,4,15]. In a previous study conducted by Fallah et al. [5] three groundwater quality indices such as heavy metal evaluation index, heavy metal pollution index and degree of contamination were investigated using geostatistical analysis and ordinary kriging. In this study, correlation analysis and multiple linear regression (MLR) were applied to investigate the parameters and to explore the interactions between them. Most groundwater studies focus on waste disposal sites or landfills with liner or some form of groundwater protection mechanism [16-18], however, there is a lack of literature on the operation of an unlined landfill on groundwater quality.

Dissolved metals in groundwater

The influence of dissolved metals on surface and groundwater quality has been widely studied due to their practical importance. However, inconsistencies in results regarding the metal concentration ranges and the dominating species are not uncommon. Chofqi et al. [19] studied the impact of landfill leachate near a coastal aquifer and reported elevated concentration of Cd (0.015-0.025mg/L) and Pb (0.06-0.1mg/L). In a study by Mor

et al. [6], a total of 7 metals (Cd, Cr, Cu, Fe, Ni, Pb and Zn) were investigated, and they found that Fe concentration in groundwater samples was considerably higher than the regulated levels and no correlation was observed among the heavy metals. Jaskelevicius & Lynikiene [20] focused on heavy metals that can jeopardize public health (Fe, Cu, Ni, Zn, Pb, Mn, Cr) and found that Fe has an average concentration exceeding 200 times the admissible value in Lithuania. Bahaa-Eldin et al. [21] reported higher concentrations of Fe and Pb (0.97mg/l and 0.32mg/l, respectively) in a Malaysian study and found that the metals originated from a nearby landfill site. Bandara et al. [22] studied landfill co-disposal practices on groundwater quality in a suburban landfill in Sri Lanka and reported that Cd exceeded the local drinking water quality standard at 0.025 to 0.038mg/L.

Correlation analysis is typically conducted to reduce the data dimensions and to examine the interactions between metal pollutants. In a case study in Casablanca, Morocco, Smahi et al. [8] studied the temporal changes of groundwater contamination using correlation analysis. Fe, Zn, Pb, Al, Mn, Cu, Cd, Cr were identified as important metal pollutants. Smahi et al. [8] however observed no significant correlations between heavy metals. Based on metal availability and mobility in landfill leachate, El-Salam et al. [23] selected eight metals (Ni, Pb, Cu, Mn, Cr, Cd, Zn and Fe) to evaluate the groundwater quality near an Egyptian landfill using correlation analysis. Mn and Fe were the two elements that exceeded the local drinking water guideline, and statistically significant correlations among Zn, Mn and Fe were observed in their study with a confidence level of 95% or higher. In an Indian study, De et al. [24] found that groundwater was heavily polluted with Cd, Fe, Hg, Mn, Ni and Pb as a result of leachate originating from an adjacent 24.7 ha landfill. De et al. [24] found that Hg was significantly correlated with Ni, Cr, and Cu, and similar significant correlations were observed between Cu with Pb and Ni.

Literature review suggests that landfill leachate is a common subsurface metal pollutant source and the relative concentrations of the metals are highly site-specific. Correlations among metals are only observed in some studies. MLR has been commonly applied in many recent surface and groundwater studies to analyze the relationships between various groundwater parameters and contaminant concentrations [25,26]. MLR models have the flexibility to apply many independent variables [27] and results are easy to understand and interpret [28,29]. Ahsan et al. [30] applied MLR for modeling As in groundwater using 113 wells in Bangladesh. Boy-Roura et al. [31] used MLR to evaluate aquifer vulnerability to nitrate pollution in Spain and to assess the relationships between the parameters. Pan et al. [15] used both regression techniques and machine learning approach to model the total dissolved solids in an urban aquifer and concluded that regression techniques such as MLR are more advantageous.

In this study, both correlation analysis and MLR are used to assess the dissolved heavy metals in groundwater near an unlined

landfill. Groundwater data in the years 2012 and 2015 are selected based on the availability of data to investigate temporal changes. The objectives of this study are to

(i)   investigate the groundwater pollution using eleven common heavy metals and to evaluate temporal changes during a 3-year period in a semi-arid climate,

(ii)   assess the correlation between these heavy metals in ground water wells in 2015 using correlation analysis, and

(iii)   generate empirical equations on selected parameters using linear regression. This study gives insight to dissolved metal contamination of a shallow aquifer with heavy metals near an unlined landfill. The present study is a continuation of a comprehensive study on the impact of an unlined municipal landfill to an urban shallow aquifer [3,5,15], with main focus on dissolved metal contamination near an unlined landfill.

Site description

The Regina Landfill is located northeast of the city and overlies both the Condie and Regina aquifers [32,33], and is the sole municipal landfill in the surrounding area (Figure 1). The operation of the primary landfill cell began in 1961 with a total footprint of 100ha. In 2011, the 60-ha primary cell (unlined) reached its full capacity. The landfill has expended in recent years with an additional 80 years of life expectancy [33]. The landfill has accepted a wide variety of materials containing construction and demolition, rubbish, recyclable concrete, fill dirt, residential recyclables, asphalt [34]. Over 380,000 tonnes of municipal solid waste is disposed of annually. Some industrial wastes are disposed of at the Regina landfill in the past and may contribute to the concentration of metal pollutants [20].

The Regina landfill site is located in a semi-arid zone and the precipitation is low (Joshua 2014) [5]. The geological formation at the study area is complex and is surrounded by a number of aquifers. The Condie, Regina, and Zehner aquifers overlap each other, as shown in Figure 2. The Condie aquifer is shallow and vertical migration of pollutants is possible from one aquifer to another. Near the landfill, the Condie aquifer is generally 8-10 m below ground surface and the regional groundwater flow direction is generally to the southwest. A description on the site geologic and flow regime can be found in Pan et al. [3,15] and Fallah et al. [5] (Figure 1 & 2).

Material and Method

Groundwater samples and monitoring wells

Sampling was done by environmental services at the City of Regina and the samples were delivered to a third party for analysis [36]. Groundwater samples considered in this study were collected from 27 monitoring wells (Table 1). The monitoring wells were classified into 5 groups located both upstream and downstream of the landfill, as shown in Figure 1. It should be

noted that no data is available for five wells (ID 30, 35, 62, 64, and 65) in 2012. In other words, the number of monitoring wells in the year 2012 and 2015 are 22 and 27, respectively. Therefore, in total 517 water samples are considered in this study. Information on sampling process has been previously reported by Pan et al. [3,15] and to avoid duplication has not been repeated here (Table 1).

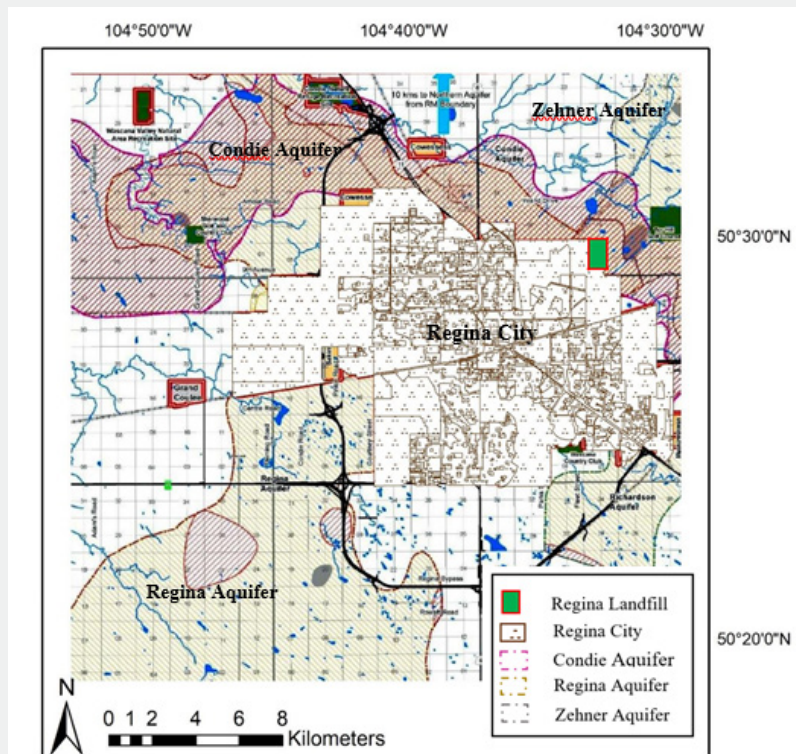**Figure 1:** Regina landfill and monitoring wells [33].

Figure 2: Regina area aquifers [35].

Table 1: Geospatial distribution of monitoring wells at the City of Regina landfill.

Group	Monitoring Wells	Comments
Group 1	67, 69, 70 and 78	Background Monitoring Wells. These monitoring wells are located upstream of the landfill. This group of wells is assumed to be out of the landfill zone of influence. The groundwater is flowing from north-east to southwest.
Group 2	84, 35, 45, and 118	These wells are located on the east side of the Landfill and inside the disposal area.
Group 3	112, 103, 104 and 114	They are located at the southwest corner of the disposal site, immediate downgradient of the landfill.
Group 4	81, 85, 71, 28, 23, 26, 30, and 32	They are located along the west side of the disposal site, immediate downgradient of the landfill.
Group 5	87, 86, 65, 64, 42, 43 and 62	Furthest away from the disposal site, located at the far west.

Statistical analysis and water quality standard

Statistical analysis on the concentration of eleven heavy metals: Aluminum (Al), Arsenic (As), Boron (B), Cadmium (Cd), Calcium (Ca), Nickel (Ni), Copper (Cu), Manganese (Mn), Zinc (Zn), Iron (Fe) and Uranium (U) was conducted in 2012 and 2015. No data was available for U in 2012. These heavy metals were chosen based on the literature review and the availability of ground water quality data from City of Regina. Maximum (Max), minimum (Min), median and the interquartile ranges of parameter (Q1 and Q3) were also identified. The criteria of contamination in monitoring wells are based on Saskatchewan's drinking water quality standard [37].

Correlation matrix and MLR

The 2015 well dataset is used to construct the correlation matrix to explore the potential interactions of metal pollutants in the subsurface environment. A coefficient of 0.75 to 1.00 indicates stronger correlation between the parameters, a coefficient ranges from 0.50 to 0.74 suggests a moderate correlation [38-40]. A p-value less than 0.05 was used to verify the significances of results [23,24]. Multiple linear regression (MLR) is defined with a dependent and two or more independent parameters and is described by Equation 1:

$$y = \beta_0 + \beta_i x_i + \beta_{i+1} x_{i+1} + \ldots\ldots + \beta_k x_k + \varepsilon \qquad (1)$$

Where:

y: dependent variable

x_i (i = 1, 2,.., k): independent variables

βi (i = 0, 1, 2,.., k): regression coefficients

ε: residual error

Prior to developing MLR equations, assumptions should be checked to ensure that MLR model can be applied to the dataset. The first assumption is that the dependent parameter should have a normal distribution and thus in this study, the distribution of the selected dependent parameters is evaluated using the two Kolmogorov-Smirnov (K-S) and Shapiro-Wilk tests. In the present study, correlation and MLR analyses were conducted using SPSS (v. 25) and a confidence interval of 95% was used [41,42]. The null hypothesis is that there is no normal distribution and no statistical differences between the dataset. Therefore, if the significance of the K-S and Shapiro-Wilk tests are higher than 0.05 (confidence interval of 95%), this means the data has a statistical normal distribution and the null hypothesis is failed to reject. The second step is to identify the multicollinearity problem, hence, the correlation coefficients between independent parameters obtained from the correlation analysis were compared to the critical Pearson correlation coefficient (R_{crit}) defined as presented by Equation 2 [43,44]. There is no multicollinearity if the absolute value of coefficient is greater than the R_{crit}.

$$R_{crit} = \frac{t_{crit}}{\sqrt{df + t_{crit}^2}} \qquad (2)$$
and

$$df = n - k \qquad (3)$$

where df is the analysis degree of freedom, n is the number of datasets, k presents the number of variables.

Using a two-tailed test with significance of 0.05, the R_{crit} value calculated in this study was 0.284. Another assumption in MLR is that the error or the residuals have a normal distribution and the residuals should hold the homoscedasticity assumption (e.g. the variance of the errors is constant [45]. All these assumptions should be satisfied in order to have a statistically valid MLP equation. Multiple regression is performed between dependent and independent parameters with a minimum confidence interval of 95% [41,42]. The validity of each regression model is verified using ANOVA test, with a p value less than 0.05.

Results and Discussion

Dissolved metals

Heavy metal concentration data for 2012 and 2015 are presented in Tables A1 & A2, respectively. Among the eleven metals considered in this study, Mn, U, As and Fe concentrations are noticeably higher than the local drinking water standards and they are separately discussed below.

Manganese

Figure 3 shows the concentrations of groundwater metal pollutants in 2015 at the study site. In this figure, the triangle symbol of the box plot represents the median value. The bottom and top edges of the box represent the first and third quantile (25th and 75th percentile) of each group. Moreover, the lower and upper level of the vertical solid line shows the minimum and maximum concentrations of the heavy metal in each group, respectively. Among the four metal pollutants considered in this

study, Manganese (Mn) was the only pollutant that was well over the horizontal dash line, representing the Saskatchewan drinking water guideline of 0.05mg/L, in all monitoring wells in 2015 (Figure 3a). Larger variations of Mn concentrations are observed in group 2. Group 1 (background) has the lowest mean concentration at about 1mg/L, as well as the lowest data variability. The median Mn concentration in the background group is 20 times higher than the standard. According to Mor et al. [6], municipal landfill leachate may cause Mn and Fe to be more soluble in groundwater.
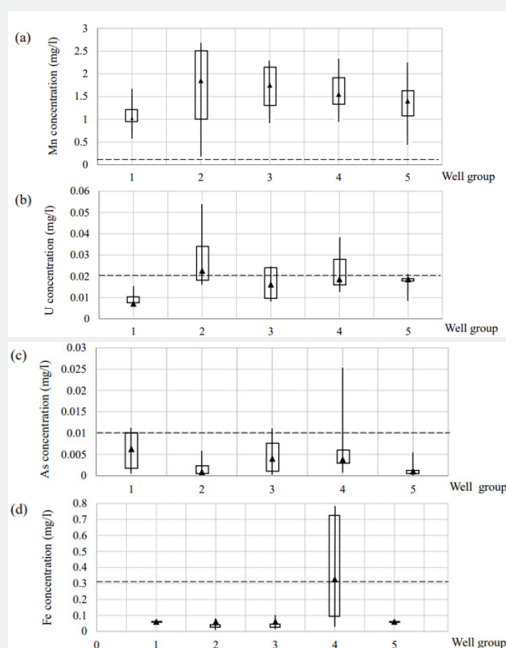

Figure 3: Concentrations of groundwater metal pollutants in 2015 at the study site for (a) Mn, (b) U, (c) As, and (d) Fe. Dash line presents Saskatchewan drinking water standard limit.

Thick layers of sedimentary rock exist in Southern Saskatchewan [46] and they may contribute to the elevated Mn level. According to the British Columbia groundwater association [47], elevated Mn and Fe may also be the result of weathering from minerals and rocks. Buamah et al. [48] found similar results in a site near the gold belt zone of Ghana and concluded that Manganese occurrence in aquifers might be originated from dissolution of compounds such as MnO_2.

Groups 2 and 3 have higher median Mn concentrations than other groups (Figure 3a), probably due to the fact that the wells in these groups are mostly located within the disposal area (Figure 1). Elevated Mn in groundwater was found to be related to the operation of a landfill in an Egyptian study [23]. The results from this study suggest that the landfill operation may negatively impact the groundwater quality, at least in terms of the Mn concentration.

Figure 4 shows the temporal changes of the metal concentration during the study period. The mean Mn concentrations in 2012 and 2015 are 1.35mg/L and 1.47mg/L, respectively. The concentration

of Mn generally increased during the 3-year period, as illustrated in Figure 4a. As discussed, data of five wells (ID 30, 35, 62, 64, and 65) are not available in 2012, therefore, temporal comparisons are not possible, and the data are omitted. All monitoring wells in group 2 show noticeable increases (14-33%) in Manganese concentration during the study period. Mn concentration in well 104 in group 3 increased from 0.78mg/L to 2.1mg/L, representing the highest percentage increase of 170%. A similar trend is observed in Fe concentration (Figure 4c), but not in As concentration (Figure 4b), as discussed in the following sections. Well 87 in Group 5 observed the largest reduction in Mn from 1.4 to 0.55mg/L or about 61%. Well 87 is located on the left-hand side of the landfill (Figure 1) and could be considered as spatially isolated from the landfill operation (Figure 3 & 4).

In 2015, about 27% of well samples had a higher Uranium concentration than the local drinking water standard (0.02mg/L). The highest concentration of U was observed in group 2 (monitoring well 118) with a concentration of 0.054mg/L,

about 2.7 times higher than the standard (Figure 3b). Group 1 (background) again had the lowest median U concentration. However, unlike other groups, Group 2 is the only group with a median U concentration higher than the standard of 0.02mg/L.

Group 5 is located furthest from the landfill (Figure 1), and less variabilities in data are observed (Figure 3b). The 2012 Uranium data is not available, and temporal analysis is not possible.
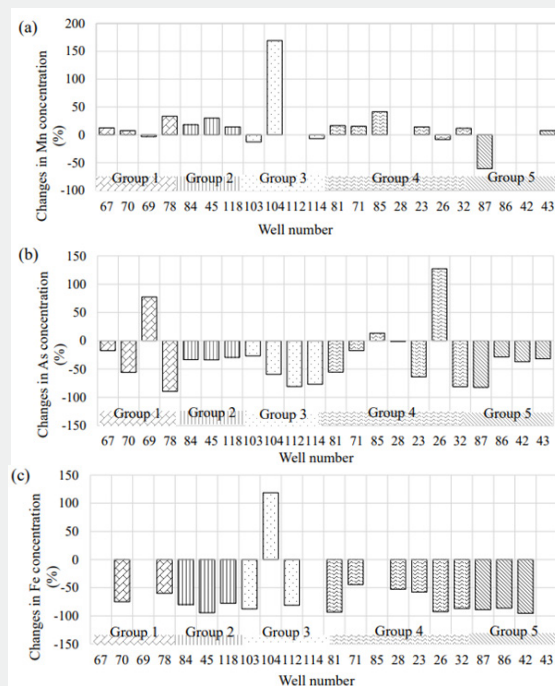

Figure 4: Temporal analysis of selected metal pollutants in 2012 and 2015 at the study site for (a) Mn, (b) As, and (c) Fe.

The 2012 and 2015 average groundwater pH were 7.75 and 7.77, respectively. Uranium is highly adsorbed by sediments when pH is between 6 to 10 [49]. In shallow aquifers, the presence of U is mainly related to weathering of uranium-bearing tills that release dissolved uranium to the groundwater [50], and field evidence has been reported in many parts of Canada such as southern Alberta [50], southeastern Manitoba [51], and northern Ontario [52]. According to Ranville et al. (2007), this phenomenon is observed in the glacial till deposits in Saskatchewan. The increases in the dissolved U concentrations in groundwater may be related to till oxidation, and surficial weathering of tills might be a more important contributor of U contamination in Condie aquifer in this study. However, direct evidence is not available to validate this claim.

Arsenic

As shown in Figure 3c, median As concentration in all wells are below the standard at 0.01mg/L. Group 1 (background) and group 2 monitoring wells have the highest and lowest median concentrations in 2015, respectively (Figure 3c). Unlike other metal pollutants considered in this study, the variability of As concentration is considerably higher in the background group (Group 1). It also appears that the operation of a 60ha unlined landfill has no apparent effect on As concentration in groundwater.

According to Buchhammer et al. [10], As in groundwater may originate from chemical rock weathering and soil erosion. Local geology and precipitation rates also impact pH and As concentration in groundwater [11,53].

The Regina landfill is located in a semi-arid area, and mass exchange between As and the geological materials may be mainly controlled by the presence of Fe and Al mineral oxides and hydroxides phases [54]. According to Welch [55], dissolved organic matters can be another factor in increasing As and should be investigated in future studies. The mean value of Arsenic concentration slightly decreased from 0.0073mg/L in 2012 to 0.0051mg/L in 2015. With the exceptions of well 26, 69, and 85, Arsenic concentration generally decreased during the study period (Figure 4b). Group 3 is located immediately downstream of the landfill and a constant decreasing As trend is observed. It is interesting to note that increase in groundwater concentration in well 26 is only observed in As, but not Mn and Fe.

Iron

About 14.8% of well samples were over the Saskatchewan drinking water guideline for Iron in 2015, and all of them originated from group 4 wells (Figure 3d). With the exception of group 4, all results are precise with minimal of variability. It is not

clear why significantly larger Fe variability is observed in group 4. Group 4 contains more wells (Figure 1), but larger variability is not observed with respect to other metal contaminants (Figure 3a-3c). Buamah et al. [48] reported that the source of Fe in groundwater may be due to the dissolution of iron (II) and the reduction of iron oxyhydroxides.

A closer look at the data suggest that the mean Fe concentrations were substantially higher in 2012. The mean concentration of Fe shows significant decrease from 1.2mg/L in 2012 to 0.16mg/L in 2015. Except well 104 in Group 3, reductions in Fe are observed in all wells.

Correlation matrix

Table 2: Correlation matrix between various groundwater parameters in 2015.

	pH	Al	U	Fe	Zn	Mn	Cu	Ni	Ca	Cd	As	B
pH	1											
Al	-0.15	1										
U	-0.32	0.23	1									
Fe	-0.37	0.19	0.27	1								
Zn	-0.02	0.03	-0.01	0.02	1							
Mn	-0.36	-0.12	0.42*	0.21	0.02	1						
Cu	0.32	0.11	0.2	-0.26	**0.56****	-0.06	1					
Ni	-0.12	0.15	0.43*	-0.18	0.06	0.1	0.36	1				
Ca	**-0.54****	0.07	**0.65****	0.25	0.08	0.44*	0.1	0.36	1			
Cd	-0.06	-0.04	-0.01	-0.09	-0.08	0.08	-0.03	**0.69****	0.06	1		
As	-0.04	**0.78****	-0.02	0.31	-0.02	-0.08	0.01	-0.17	-0.24	-0.12	1	
B	-0.36	**0.87****	0.31	0.22	0.05	0.09	0.04	0.18	0.23	-0.04	**0.68****	1

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

A total of 11 metal pollutants and groundwater pH were studied using correlation analysis. The resulting correlation matrix is shown in Table 2, with statistically significant results bolded. The results indicate that Al is strongly correlated with As and B (r = 0.78 and 0.87, respectively). Ni is moderately correlated with Cd (r = 0.69), and Cu is moderately correlated with Zn (r = 0.56), with p < 0.01 in this study. These correlations may be originated from the improper disposal of consumer electronics and batteries. B also showed a significant correlation with As (r = 0.68, p < 0.01). The correlation between B and As in groundwater has also been noticed in other studies [54,56]. Ca and U show a moderate correlation with r value of 0.65 (p < 0.01) in this study (Table 2).

In 2015, the pH in well waters measured from 7.43 to 8.17 with an average value of 7.77. Heavy metal solubility in groundwater decreases with alkalinity [9]. This was also observed in Jaskelevicius & Lynikiene [20] where all heavy metal concentrations were lower in samples with higher pH, and a significant negative correlation was observed. With exception of Cu, groundwater pH is negatively correlated with all metals. The correlations between pH, Ca and Fe were also observed in different studies [6,54], as summarized in Table 3. Huang et al. [57], reported that a strong negative correlation (r = -0.95, p < 0.01) existed between Fe and pH (Table 3). In this study, the correlation

between Fe and pH is, however, weak and insignificant (r = 0.37, p > 0.05). In the current study, no significant correlation between Mn and pH is observed. In the studies by Buamah et al. [48] & Huang et al. [57], moderate negative correlations were observed between pH and Mn (r = - 0.69 and r = -0.64, respectively, p < 0.01) in groundwater with pH below 6.5. Insignificant correlations of pH with Mn were also reported in a study by Rao [58], who suggested that Mn and Fe released easily from host rocks and metal organic compounds under acidic conditions. Comparisons of results are tabulated in Table 3. Various studies suggested that pH, Ca and Mn may impact U mobility in groundwater [49,59-61] due to the water rock interactions and microbial activity result in increases in carbonate alkalinity and Ca in groundwater wells. The relationship between As and pH in groundwater has been documented by Welch [55]. A weak and insignificant correlation between As and pH are however observed in this study (Table 3). The results suggest the complexity of shallow aquifer and the importance of using site-specific data in geological studies (Table 3).

Equations from MLR

In this study, regression analysis was conducted on the parameters identified in correlation matrix using trial and error. The sets of significant correlated parameters identified

using correlation analysis were Ca, U, pH and Mn; Al, As and B. According to the K-S test, the p value of the Ca, U, Al, As and B were ranged from 0.0001 to 0.028 all greater than 0.05. Therefore, the normality assumption was violated, and it was realized that these parameters are not suitable for developing MLR models. Using the K-S test, only pH and Mn were normally distributed with p value 0.200 for both, hence, Mn was selected as dependent parameter while Ca, U and pH were independent factors with correlation coefficients of 0.54, 0.42 and 0.36, respectively. No significant collinearity exist between predictor parameters since correlation coefficients were greater than $R_{crit} = 0.284$. From the regression analysis, the model had R2 and adjusted R2 of 0.247 and 0.148, respectively. ANOVA test showed the model was not statistically significant with p = 0.084 greater than 0.05. In the MLR equation, the coefficients of Ca, U and pH were 0.001, 14.222 and -0.571 with p value 0.514, 0.320 and 0.403 greater than 0.05 and were not statistically significant.

Table 3: Correlation coefficients reported in different groundwater studies (2000-2019).

Reference	Location	Site Description	Contaminants	Parameters and Key Findings		
Welch [55]	United States	Alkaline aquifer, existence of iron oxide	As, presence of organic carbon and iron oxide	pH	As	weak
Smedley et al. [56]	Argentina	Silts and fine sands; cold, arid environment	As, F, NO_3-N, B, Mo, Se and U and high salinity	As	pH	r=0.46
				As	B	weak
Mor et al. [6]	India	Semi-arid environment, dry conditions associated with hot summers and cold winters, high conductivity values. pH range 7.02 to 7.85.	Cu, Fe and Zn	pH	Fe, Ca	r=-0.172
Mor et al. [6]	India	Wells are under the unlined landfill		No correlation		
Bhattacharya et al. [54]	Argentina	Semi-arid western part of Chaco Plain. Layers of gravel, sand, silt and clays, forming several sub-aquifers. Sediment contains Mn, Fe and Al oxides and hydroxides. Presence of organic ash. pH range: 6.4 - 9.3 (average of 7.6).	The mobility of As was assessed in the study not the heavy metals contamination	pH	Ca	r=-0.6
				As	Ca	r=-0.47
				As	B	r=0.76
				As	pH	r=0.59
Buamah et al. [48]	Ghana	Wells are located in the gold-belt zone of Ghana. Sulphide minerals especially arsenopyrite. pH range is 5.6-6.5.	Mn, Fe and As	As dependent	pH, Fe and Mn	r=-0.69
Giménez et al. [11]	Argentina	pH varies from 6.50 to 8.94 with the average of 7.54 (neutral to slightly alkaline).	As	Fe	As	r=0.17
				Mn	As	r=-0.07
Smahi et al. [8]	Morocco	Unlined landfill, bedrock of the landfill is formed of Cambrian and Ordovician marine sediments modified by the Hercynian orogenesis.	Organic matter	Heavy metal		No correlation
Huang et al. [57]	China	Subtropical monsoon climate, pH changes from 6.2 to 7.2	Fe and Mn	pH	Mn	r=-0.643
				Fe	Mn	r=0.602
				Zn	Cu	r=0.228
				pH	Fe	r=-0.95
Salam et al. [23]	Egypt	Wells under the municipal solid waste landfill, Unconfined aquifer, pH ranges from 7.2 to 7.9.	Organics, salts and heavy metals (Fe, Mn and Cd)	Mn	Zn, Fe	r=0.985, r=0.362
De et al. [24]	India	The soil strata of the city consist of clay, silt, various grades of sand gravels and pebbles. Hot and humid climate.	Fe, Cd, Hg, Mn, Ni and Pb	Hg	Cu, Ni	r=0.907, r=0.957
				Cu	Ni, Pb	r=0.900, r=0.944

Present study	Canada	Wells are located under the unlined. Regina landfill semi-arid zone with low precipitation. pH average is 7.7.	Mn, Fe and As	Al	B	r=0.87
				Al	As	r=0.78
				Zn	Cu	r=0.56
				pH	Fe	r=-0.41
				pH	Ca	r=-0.54
				B	As	r=0.68

After testing different parameters for constructing MLR, the simple linear regression model between pH and Ca was found to satisfy all assumptions. pH was normally distributed with p values of 0.2 and 0.7 (p > 0.05) based on K-S and Shapiro-Wilk test, respectively. In this model, the statistically significant R2 of 0.297 and adjusted R2 of 0.268 with p = 0.003 from ANOVA test were between dependent and explanatory parameters. Based on the homoscedasticity results from the Levene's test of equality of error variances, it was observed that there is no significant differences of the error variances with the p value of 0.051 and this condition is satisfied. The results from the normality tests of the residuals showed both of K-S and Shapiro-Wilk tests had the p value greater than 0.05 (p of 0.200 and 0.094, respectively). Therefore, there was no evidence of any significant deviation from normality for the residuals. It was found that the data normality and equality of variance were satisfied the conditions for performing linear regression between pH and Ca. Therefore, the equation for linear regression was defined as:

$$pH = -0.001Ca + 8.107 \qquad (4)$$

Equation 4 is valid for aquifer with slightly alkaline pH (7.43 < pH < 8.17), and Ca range of (120mg/L < Ca < 500mg/L). The magnitude of the coefficient of Ca is noticeably small, probably due to the narrow range of pH encountered in the Condie aquifer.

Linear regression analysis was also performed for Mn and U, while Mn is the dependent parameter. K-S and Shapiro-Wilk normality tests had the p values of 0.200 and 0.888, respectively, showing the normal distribution of Mn data. The model was statistically significant (p = 0.029) with weak R2 of 0.176 and adjusted R2 of 0.143. The results of the Levene's test (p = 0.228) revealed the constant variance of the error term, thus, the homoscedasticity assumption was satisfied. Using the K-S and Shapiro-Wilk tests (p value of 0.200 and 0.700, respectively) the normality of the residual error was also observed. The equation for Mn is valid for aquifers with Mn concentration between 0.18-2.6mg/L and U concentration between 0.0066 - 0.054 mg/L was provided as follow:

$$Mn = 24.536U + 1.005 \qquad (5)$$

As shown in Figure 5a & 5b these linear regression models can only accurately predict about 29.7% and 17.6% of the observed pH and Mn data, respectively. From the result, it can be concluded that the linear regression method is not helpful in accurately predicting and modeling heavy metals in Condie aquifer around the Regina landfill. The proposed equations may only be applicable to sites having similar hydrogeological conditions (Figure 5).
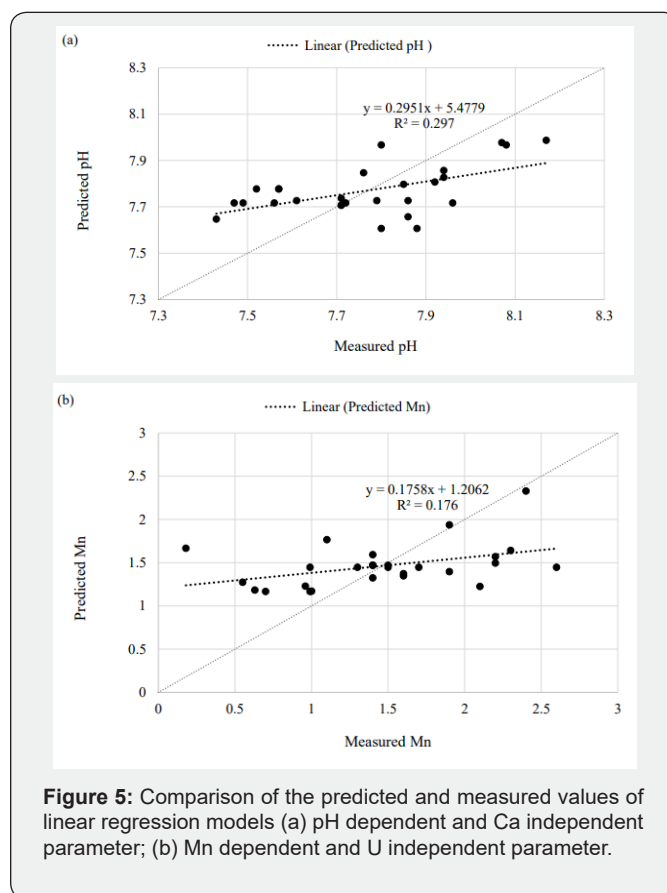

Figure 5: Comparison of the predicted and measured values of linear regression models (a) pH dependent and Ca independent parameter; (b) Mn dependent and U independent parameter.

Conclusion

Due to the existence of an unlined landfill in the City of Regina, and potential influence of landfill leachate on groundwater quality, pollution with heavy metals was assessed using correlation analysis and MLR. Statistical analysis showed that Mn concentrations moderately increased from 1.35 to 1.47mg/l over the study period. Mn concentration is however higher than local drinking water standards. The increases in Mn concentrations in group 2 and 4 monitoring wells may be the result of operations of the unlined landfill. According to the data from monitoring wells, the U contamination in groundwater was very low and might be related to groundwater pH and surficial weathering as reported

in a number of other Canadian studies. Both Mn and U had the highest mean concentration upstream of the landfill (group 2 monitoring wells). In 2015, Mn, U, As and Fe exceeded the Saskatchewan drinking water quality standard limits. However, in 2012, the heavy metals that exceeded the guideline were: Mn, As, Fe and Cd. Overall, the groundwater monitoring wells did not have severe contamination in 2015 except for Mn. Based on correlation analysis, Ni was positively correlated with Cd, Zn with Cu. In this study, it was attempted to generate MLR models using heavy metal parameters, however, among them only pH and Mn had a normal distribution, thus two linear models were provided for pH and Mn. The proposed equations provide statistically significant, simple estimates of key groundwater metal pollutants in semi-arid regions with similar hydrogeological conditions.

Appendix A

Table A1 & A2

Table A1: Concentration of heavy metals in 2012 (Note: Data is not available for five wells: ID 30, 35, 62, 64, and 65 in 2012).

	Monitor-ing wells (ID)	pH	Alumi-num (mg/l)	Arsenic (mg/l)	Boron (mg/l)	Cadmium (mg/l)	Calcium (mg/l)	Nickel (mg/l)	Copper (mg/l)	Manganese (mg/l)	Zinc (mg/l)	Iron (mg/l)	Ura-nium (mg/l)
	67	7.74	0.0032	0.012	0.068	0.037	110	0.00067	0.0034	0.88	0.004	0.06	-
	70	7.83	0.11	0.0059	0.086	0.06	130	0.00095	0.0004	0.93	0.008	0.24	-
	69	7.83	0.008	0.0062	0.096	0.053	120	0.0005	0.0007	0.65	0.012	0.06	-
Group 1	78	7.91	0.0087	0.0055	0.079	4.90E-05	300	0.011	0.0034	1.2	0.014	0.15	-
	84	7.68	0.0035	0.0018	0.087	0.16	360	0.0017	0.013	1.1	0.023	0.6	-
	35	-	-	-	-	-	-	-	-	-	-	-	-
	45	7.7	0.0032	0.0083	0.13	0.065	360	0.0019	0.0004	2	0.017	1.1	-
Group 2	118	7.58	0.0045	0.0007	0.1	0.45	430	0.025	0.014	2.1	0.015	0.27	-
	103	7.91	0.046	0.015	0.15	0.00011	170	0.00072	0.0032	1.1	0.012	0.49	-
	104	7.91	0.0094	0.016	0.21	0.00007	140	0.00079	0.0003	0.78	0.015	0.064	-
	112	7.77	0.006	0.0024	0.11	0.069	330	0.0049	0.005	2.2	0.016	0.32	-
Group 3	114	7.82	0.0035	0.0065	0.15	0.093	290	0.0035	0.006	1.5	0.013	0.06	-
	81	7.71	0.014	0.0074	0.059	0.064	270	0.0006	0.0079	1.2	0.01	0.94	-
	71	7.74	0.0011	0.0052	0.084	0.11	350	0.0015	0.0089	1.3	0.013	1.4	-
	85	7.85	0.0018	0.0007	0.095	0.13	350	0.014	0.0057	0.7	0.012	0.06	-
	28	7.6	0.0021	0.0084	0.45	0.00019	490	0.0072	0.0045		0.014	1.5	-
	23	7.67	0.0047	0.0069	0.14	0.00016	380	0.013	0.0073	1.4	0.083	0.33	-
	26	7.45	0.0025	0.011	1.2	0.24	460	0.012	0.015	1.2	0.049	5.1	-
	30	-	-	-	-	-	-	-	-	-	-	-	-
Group 4	32	7.61	0.011	0.028	0.12	0.14	400	0.0042	0.013	1.7	0.02	5.7	-
	87	7.81	0.001	0.0018	0.092	0.064	410	0.0019	0.0039	1.4	0.013	0.55	-
	86	7.8	0.0048	0.0014	0.071	0.055	410	0.0052	0.0083	1.5	0.01	0.44	-
	42	7.68	0.0023	0.0081	0.14	0.062	390	0.0011	0.0036	2.2	0.014	1.3	-
	64	-	-	-	-	-	-	-	-	-	-	-	-
	65	-	-	-	-	-	-	-	-	-	-	-	-
	43	7.78	0.0035	0.0006	0.12	0.27	310	0.012	0.0004	1.3	0.004	0.06	-
Group 5	62	-	-	-	-	-	-	-	-	-	-	-	-

Table A2: Concentration of heavy metals in 2015.

Monitoring wells (ID)	pH	Aluminum (mg/l)	Arsenic (mg/l)	Boron (mg/l)	Cadmium (mg/l)	Calcium (mg/l)	Nickel (mg/l)	Copper (mg/l)	Manganese (mg/l)	Zinc (mg/l)	Iron (mg/l)	Uranium (mg/l)
Group 1 67	8.07	0.003	0.0099	0.007	5.00E-05	130	5.00E-04	0.00035	0.99	0.0049	0.06	0.007
70	8.08	0.006	0.0026	0.008	2.00E-05	140	0.002	0.0005	1	0.0031	0.06	0.007
69	8.17	0.006	0.011	0.088	2.00E-05	120	1.00E-03	0.0038	0.63	0.003	0.06	0.007
78	7.86	0.003	0.00058	0.079	0.0004	380	0.012	0.0056	1.6	0.084	0.06	0.015
Group 2 84	7.56	0.003	0.0012	0.1	2.00E-05	390	0.002	0.00043	1.3	0.0042	0.12	0.018
35	7.8	0.003	0.00053	0.073	3.00E-05	500	0.007	0.0017	0.18	0.012	0.06	0.027
45	7.86	0.005	0.0055	0.14	0	450	0.004	0.0015	2.6	0.012	0.06	0.018
118	7.88	0.003	0.00048	0.12	0.0005	500	0.024	0.0032	2.4	0.013	0.06	0.054
Group 3 103	7.8	0.003	0.011	0.14	9.00E-05	140	0.002	0.0002	0.96	0.035	0.06	0.009
104	7.57	0.003	0.0065	0.17	5.00E-05	330	0.001	0.0002	2.1	0.0072	0.14	0.009
112	7.85	0.003	0.00046	0.12	5.00E-05	310	0.004	0.00072	2.2	0.0073	0.06	0.023
114	7.76	0.003	0.0015	0.11	5.00E-05	260	0.004	0.0007	1.4	0.003	0.06	0.024
Group 4 81	7.92	0.003	0.0033	0.071	3.00E-05	300	5.00E-04	0.00065	1.4	0.0064	0.063	0.013
71	7.96	0.005	0.0043	0.084	2.00E-05	390	0.001	0.0002	1.5	0.0079	0.78	0.019
85	7.57	0.003	0.00076	0.088	0.0001	330	0.017	0.0016	0.99	0.0034	0.06	0.018
28	7.49	0.003	0.0083	0.11	0	390	0.001	0.0003	1.9	0.014	0.71	0.016
23	7.52	0.003	0.0025	0.11	2.00E-05	330	0.002	0.0002	1.6	0.014	0.14	0.014
26	7.61	0.21	0.025	1.3	0.0002	380	0.013	0.0018	1.1	0.017	0.37	0.031
30	7.43	0.003	0.0033	0.72	0.0002	460	0.01	0.00054	2.3	0.016	0.28	0.026
Group 5 32	7.47	0.003	0.0053	0.12	6.00E-05	390	0.003	0.0003	1.9	0.022	0.75	0.038
87	7.79	0.003	0.00032	0.11	0.0001	380	0.013	0.00087	0.55	0.0059	0.06	0.011
86	7.72	0.003	0.001	0.08	2.00E-05	390	0.006	0.00053	1.5	0.0047	0.06	0.018
42	7.71	0.003	0.0051	0.15	4.00E-05	400	0.001	0.0006	2.2	0.0054	0.06	0.02
64	7.71	0.003	0.0011	0.12	0.21	370	0.035	0.00092	1.7	0.0076	0.06	0.018
65	7.71	0.003	0.0014	0.079	0.0002	400	0.004	0.00086	1.4	0.022	0.06	0.019
43	7.94	0.003	0.00043	0.13	0.0003	280	0.012	0.0013	1.4	0.012	0.06	0.019
62	7.94	0.003	0.0002	0.12	2.00E-05	250	5.00E-04	0.00053	0.7	0.03	0.06	0.007

Declarations

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Conflicts of interest/Competing interests

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

Availability of data and material

The data provided in appendix Table A1 & A2

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