

## **Hydrogeochemical Analysis and Spatial Distribution of Groundwater Quality in Roundhill Landfill Vicinity of South Africa**

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# Hydrogeochemical Analysis and Spatial Distribution of Groundwater Quality in Roundhill Landfill Vicinity of South Africa

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**ABSTRACT:** Groundwater is the most reliable resource for consumptive uses worldwide, but it is vulnerable to anthropogenic pollution in this post-industrialization era. Pollution of the resource may result from anthropogenic activities; hence, analysing the effects of leachate on groundwater is imperative. This study assessed the spatial distribution of physicochemical parameters of groundwater in Roundhill landfill vicinity of South Africa and conducted their hydrogeochemical analysis. Water samples were collected from 3 boreholes in the landfill surroundings and analysed for selected physicochemical characteristics. Spatial distribution of these parameters showed dominant pollution by  $Mn^{2+}$ ,  $Fe^{2+}$ , and  $NH_4^+$ , which surpassed prescribed allowable limits of the country in most of the study area. Possibilities of simple dissolution and ion mixing were deduced from the Durov diagram. Magnesium carbonate, sodium chloride, and mixed faces of groundwater were dominant in boreholes 1 to 3, respectively. The dominance of  $Ca^{2+}$ ,  $Cl^-$ ,  $Mg^{2+}$ , and  $NH_4^+$  ions in some boreholes suggested anthropogenic pollution. Landfill leachate was associated with groundwater pollution in the study area.

**KEYWORDS:** Contamination, hydrogeochemical, groundwater, landfill leachate, pollution, spatial distribution

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## Introduction

Groundwater is the largest and most essential resource of potable water worldwide and more than 1.5 million people depend on it daily.<sup>1</sup> In sub-Saharan Africa including South Africa, the resource is deemed as the most reliable to meet the ever-increasing water demands.<sup>2</sup> However, groundwater's vulnerability to contamination has become a growing concern. Groundwater pollution mainly results from the rise of urbanization and industrialization although it can be geogenic. The pollution varies temporally and spatially. These developments prompt the generation of solid waste, its indiscriminate dumping, and mismanagement in landfills and open dump yards. Landfilling is a common solid waste management approach worldwide although such facilities are pre-established pollution threats to both surface water and groundwater through leachate infiltration in capped and uncapped landfills.<sup>3</sup>

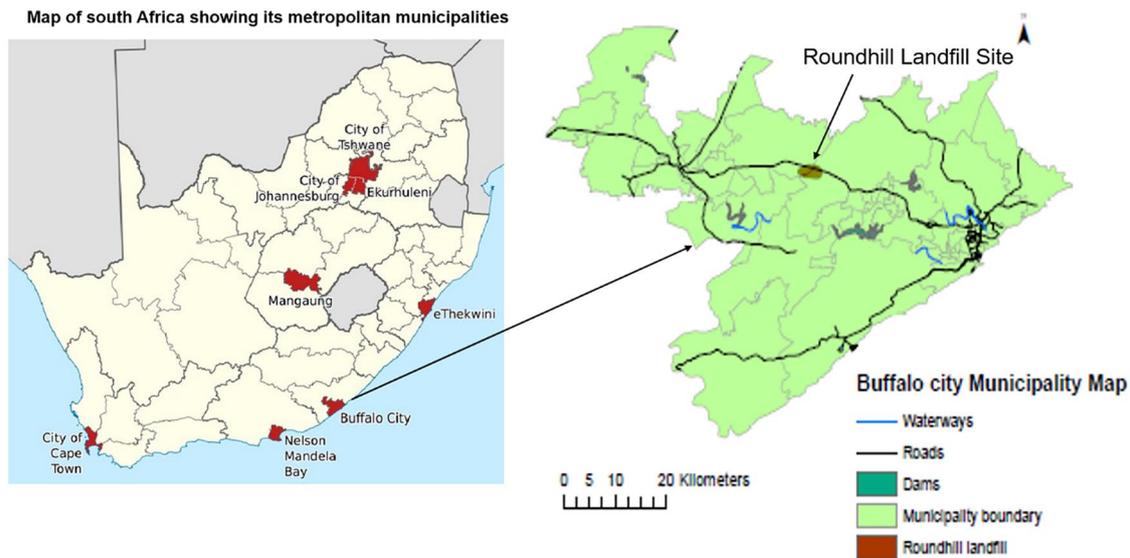
A number of studies evaluating the effects of leachate on groundwater resources have been done and resultant findings have differed due to the site-specific nature of aquifers. One such study reported minimal impact although its contamination by bacteria was reported.<sup>4</sup> Another physicochemical parameter analysis of groundwater during post-monsoon and pre-monsoon seasons in 3 Indian landfill vicinities deemed the resource unsuitable for consumption.<sup>5</sup> In South Africa's Mahikeng area, groundwater showed minimal pollution and assayed physicochemical parameters were within South

African National Standards (SANS)<sup>6</sup> permissible limits with exception of turbidity and *Escherichia coli*.<sup>1</sup> Borehole drinking water in Mpumalanga and Northwest provinces of South Africa was contaminated with leachate and required pre-treatment before consumption.<sup>7</sup>

Roundhill landfill in Eastern Cape, South Africa, reported operational challenges such as shortage of cover material leading to exposed waste, inadequately lined air-spaces that alter the landfill's slopes, illegal dumping, and waste picking.<sup>8</sup> These challenges enhance leachate generation and its unmanaged flow, which increases its risk of contamination to surrounding resources. According to Stroebel et al.,<sup>9</sup> groundwater is the most reliable and accepted source of water in the area. As such, incidents of leachate overflow and seepage from the faulty collection system in the landfill could lead to groundwater pollution.<sup>10</sup> Buffalo city metropolitan municipality, where the landfill is situated, sought for specialist services to improve infrastructure and reduce potential pollution risk in the facility.<sup>10</sup> Although these corrective measures are in place, leachate characterization, functionality, and effects on the surrounding groundwater resources remain scantily understood and a possible pollution threat. In addition, spatial distribution of physicochemical parameters in the area is non-delineated. This study aimed at determining the spatial distribution of groundwater physicochemical characteristics and its hydro-geochemistry.



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**Figure 1.** Study area and the location of Roundhill landfill.

## Materials and Methods

### Study area and sampling

Roundhill landfill is found in Buffalo city metropolitan municipality of Eastern Cape, South Africa (latitude 32°53'13.66"S and longitude 27°37'26.20"E; Figure 1). The facility is located near a national road and was previously used as natural grassland. It borders active croplands and dairy farms in the northeast and southeast, respectively. It is a general large landfill, which receives more than 500 tonnes of waste daily and is found in an area vulnerable to leachate generation due to positive water balance. The landfill facility began operations in 2006 and serves Buffalo city municipality and the southeast coast parts of the province.<sup>10</sup> The site is located between Nahoon and Buffalo river catchments and its geology is characterized by dykes and dolerite sheets dating to Jurassic age as well as Beaufort sediment of the Karoo family.<sup>9</sup> Area groundwater flows to the southeast and northeast and is classified as a minor aquifer with low groundwater yields of 1 L/s in most boreholes and a deep depth of >40 m. The area has clayey soils.<sup>9</sup> To evaluate the effect of leachate on groundwater, samples were collected in 3 monitoring boreholes near the landfill facility (Figure 2). A handheld GARMIN GPS-60 receiver was used to locate the sampling points whose coordinates were georeferenced and documented as shown in Table 1. This GPS technology enhanced the precision of data obtained and integrated it into the geographical information system. Samples were collected in plastic containers, acidified with 1% (v/v) nitric acid, and stored at 4°C for analysis.

### Determination of physicochemical and microbial parameters in water

**Physical parameters.** Physical parameters considered in this study were colour, turbidity, and total suspended solids (TSS).



**Figure 2.** Distribution of monitoring boreholes in the relation to the landfill proximity. BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3.

Colour was determined using a handheld tristimulus colorimeter and resultant values were converted to a colour scale in the Platinum–Cobalt (Pt-Co) system.<sup>11</sup> Turbidity was determined using nephelometry documented using standard methods,<sup>12</sup> and readings were in nephelometric turbidity units (NTU). Total suspended solids were determined using gravimetric method, where 100 mL of sample was shaken and filtered through a pre-weighed dry filter paper. Collected suspended residue and filter paper were oven-dried at 105°C to constant mass then cooled. Total suspended solids were then determined using equation (1)

$$\text{TSS in mg/L} = \frac{\text{Weight of filter paper and dried residue} - \text{Weight of filter paper}}{\text{Sample volume}} \quad (1)$$

**Chemical parameters.** Various chemical parameters including pH, total dissolved solids (TDS), electric conductivity (EC),

**Table 1.** Location of sampling sites.

WATER SOURCE	ABBREVIATION	LOCATION
Borehole 1	BH 1	32°53'29"S 27°37'27"E
Borehole 2	BH 2	32°53'26"S 27°37'19"E
Borehole 3	BH 3	32°53'24"S 27°37'32"E

major cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{Fe}^{2+}$ ,  $\text{K}^+$ , and  $\text{Mn}^{2+}$ ), and anions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{F}^-$ ) were assessed. pH was measured using a digital metre that had been pre-calibrated using pH buffers. The metre was inserted in water samples before recording the readings from a screen. Total dissolved solids and EC levels in groundwater were determined using a conductivity metre pre-calibrated using distilled water. Major cations were determined using flame atomic absorption spectrophotometry (FAAS) equipment and EPA method 700B.<sup>13</sup> The method involved aqua regia digestion of samples in nitric acid using a hot plate followed by sample cooling and dilution. Background standards were prepared and used to correct elemental interferences during analysis and ensure high recoveries. Argentometry was used to determine  $\text{Cl}^-$  while  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were determined by turbidimetry and ultraviolet spectrophotometry, respectively.<sup>14</sup> Calorimetry was used to determine  $\text{F}^-$  and  $\text{NH}_4^+$  in water.<sup>14</sup>

The microbiology of groundwater was assessed using 2 measures: total coliforms (TC) and *E. coli*, which are faecal coliforms. The 2 parameters were determined using EPA method 1604.<sup>15</sup> In the method, 100 mL of sampled water was filtered through a cellulose ester membrane filter of 47 mm and 0.45  $\mu\text{m}$  pore size that does not allow bacteria filtration. The filter was placed in MI agar in a plate and incubated for 24 hours at 35°C. Resultant colonies were inspected and blue and white colonies resulting due to ultraviolet light exposure were counted as TC. The blue colonies resulting from the breakdown of Indoxyl- $\beta$ -D-glucuronide (IBDG) by  $\beta$ -glucuronidase enzyme in the media and fluorescent light were considered as *E. coli*.<sup>15</sup>

### Hydrogeochemical analysis of groundwater

Acquired physicochemical data were graphically analysed using Piper, Durov, and Schoeller diagrams that were plotted using Aquachem v3.7 software. The Piper diagram modified by Handa<sup>16</sup> consists of 2 separate triangular representations of cations and anions and a diamond shape of combined ions that are used to explain hydrogeochemical faces of water samples. This tri-linear diagram depicts water chemical aspects definitively and have been used in many studies.<sup>17,18</sup> A Durov<sup>19</sup> diagram was used to explain the chemical characteristics of groundwater by displaying dominant ions as percentage milliequivalents in 2 ternary graphs representing ions and summing up to 100%.

This approach has been used in several hydrogeochemical analyses.<sup>17,18</sup> Schoeller<sup>20</sup> diagram was used to assess variations in concentrations and relations of assayed parameters.

### Spatial representation of physicochemical parameters

Spatial interpolation in this study was used to produce continuous information using the 3 distinct borehole locations. Inverse distance weighted (IDW) method which interpolates using known values of a specified area was used. In the method, values near the prediction location influenced interpolated values more than those far away; hence, more weight was assigned to the former and weight decrease was directly proportional to distance.<sup>21</sup> Geostatistical relationships of known borehole points used Arc-GIS 10.3 software to interpolate physicochemical parameters of groundwater in the study area and represent their distribution in maps.

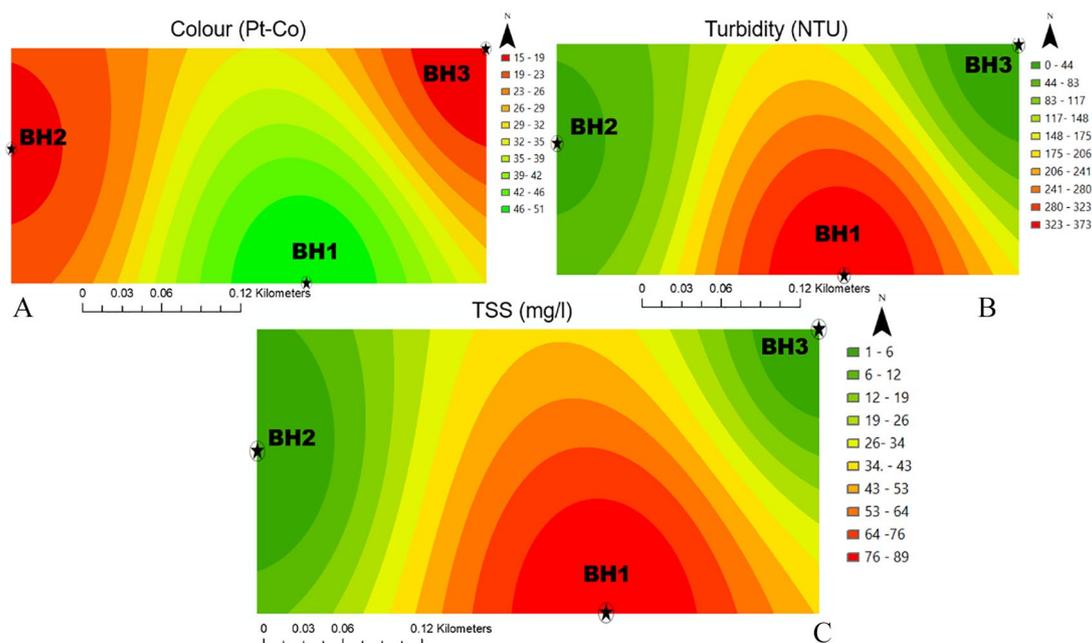
## Results and Discussion

### Physical parameters

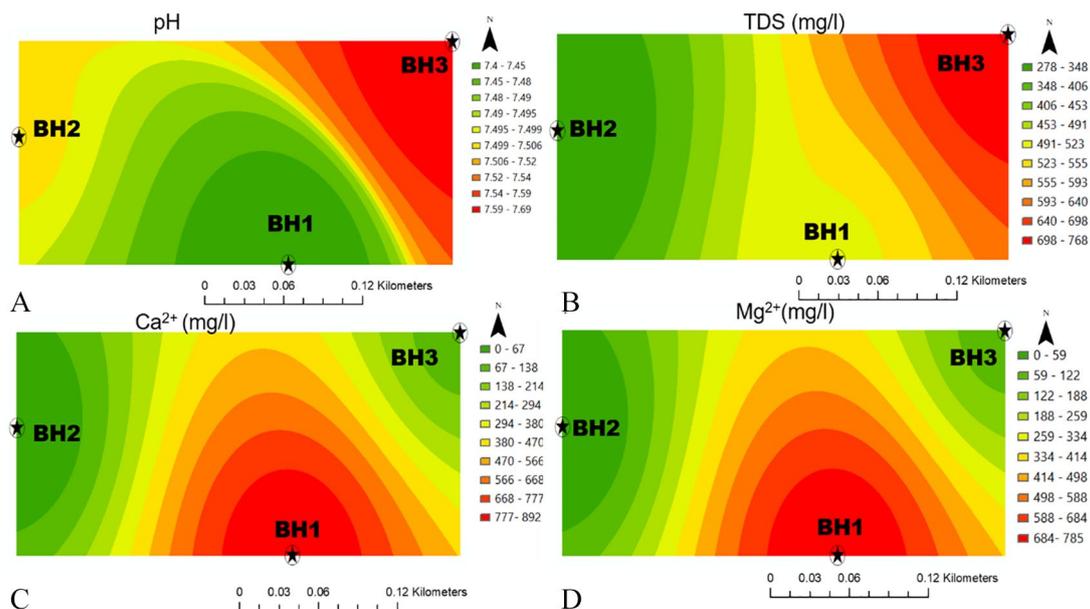
Physical parameters assayed in groundwater and their spatial distribution were as shown in Figure 3. The colour of water ranged from 15 to 51 Pt-Co units (Figure 3A). Areas around all boreholes were above the permissible requirements of  $\leq 15$  Pt-Co units. High Pt-Co units physically indicate pollution.<sup>21,22</sup> Turbidity levels ranged from 0 to 373 NTU and the highest levels were recorded in borehole 1 (BH 1; Figure 3B). Most parts of the study area had NTU levels  $>1$ , which was beyond SANS permissible limits and the results corresponded to unsightly appearance of groundwater, which could be due to the presence of suspended solids such as sediments, algae, coloured inorganic, and organic matter in the boreholes.<sup>14</sup> The levels of non-filterable residue of water, TSS ranged between 1 and 89 mg/L, and BH 1 reported elevated levels compared with the others (Figure 3C). High TSS levels probably corresponded to the presence of foreign materials including bacteria and inorganic matter. In Niger Delta wells, elevated TSS and turbidity levels were attributable to municipal solid waste leachate from area dumpsites.<sup>14</sup>

### Chemical Parameters

Chemical characteristics of groundwater in the study area and their spatial distribution was as shown in Figures 4 to 7. Figure 4 showed the distribution of pH, TDS,  $\text{Ca}^{2+}$ , and  $\text{Mg}^{2+}$ . pH levels in all the boreholes ranged between 7.4 and 7.69 reflecting neutral to slightly alkaline waters that were within SANS permissible limits of  $\geq 5$  to  $\leq 9.7$ . Total dissolved solids levels were between 278 and 768 mg/L, and borehole 3 (BH 3) had the highest levels. At the 3 sampling points, values were within the  $\leq 1200$  mg/L limit by SANS but inferred to saline water that contained anions and cations possibly from microorganisms and heavy metals in groundwater from leachate. A study



**Figure 3.** Physical parameters of groundwater representing (A) colour, (B) turbidity, and (C) TSS. BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3; NTU, nephelometric turbidity units; Pt-Co, Platinum–Cobalt; TSS, total suspended solids.

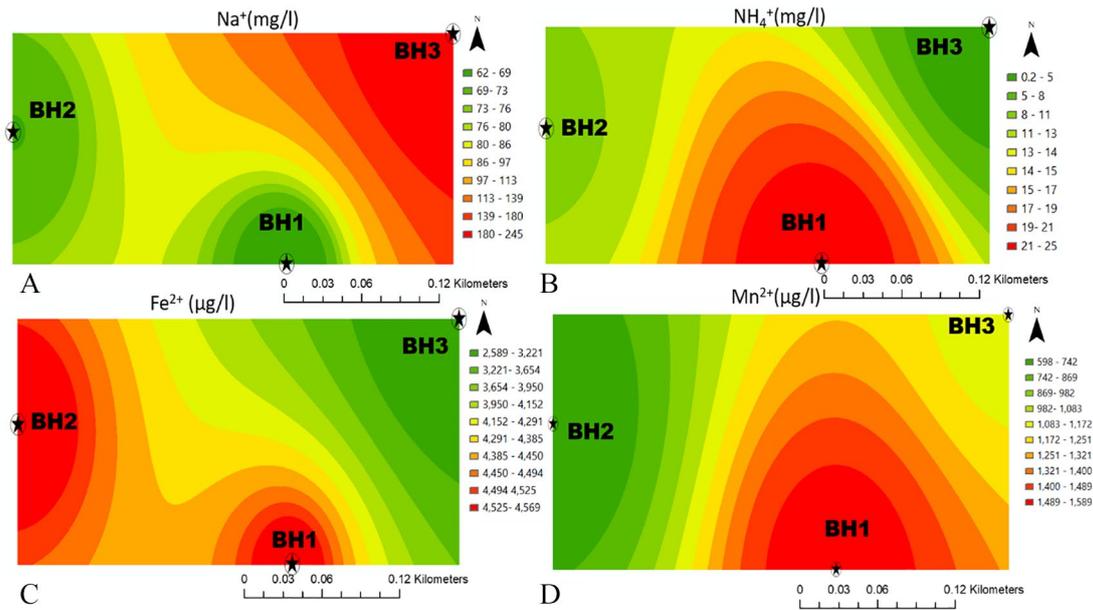


**Figure 4.** Chemical parameters of groundwater in the study area representing (A) pH, (B) TDS, (C)  $\text{Ca}^{2+}$ , and (D)  $\text{Mg}^{2+}$ . BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3; TDS, total dissolved solids.

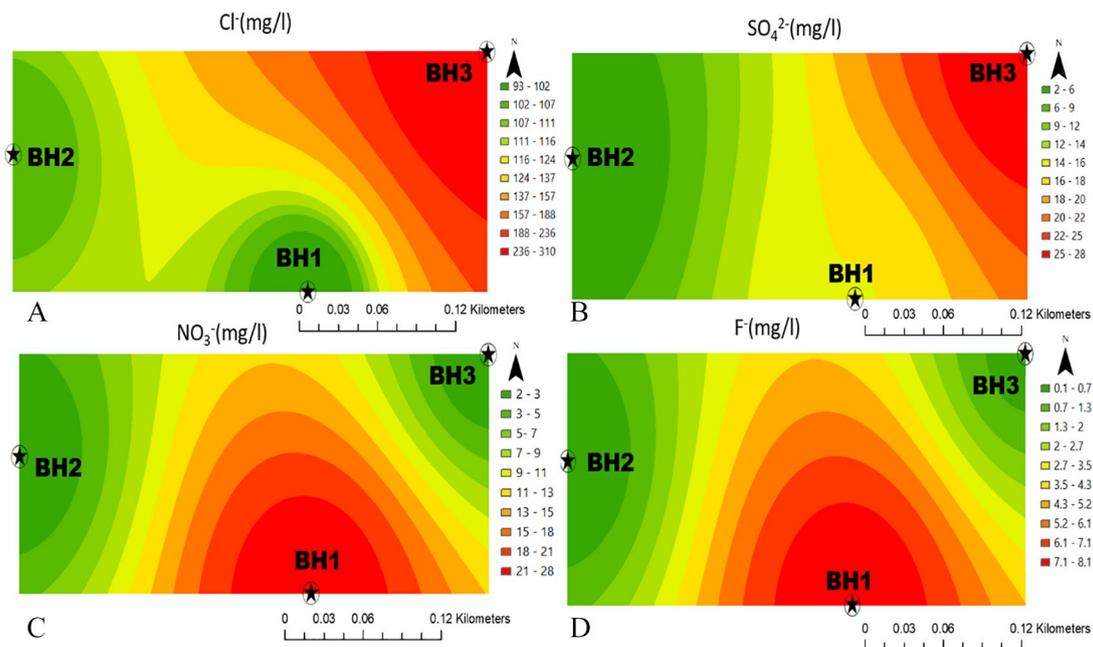
in Thiruvananthapuram district of India reported high TDS levels in groundwater and attributed the trend to the water's eutrophic nature due to major cations from waste dumping in the vicinity.<sup>23</sup> Unscientific disposal of solid wastes in Warri, Agbor, and Agbarho areas of Delta state, Nigeria, leading to leachate runoff was attributed to the high TDS levels in area groundwater.<sup>14</sup>

Calcium and magnesium levels (Figure 4C and D) were used to assess groundwater quality as they are associated with its hardness. Their levels ranged from 0 to 892 and 0 to 785 mg/L

in respective order and BH 1 had the highest levels in both cases. Most of the study area levels were beyond the documented permissible levels of 75 mg/L for  $\text{Ca}^{2+}$  and 30 mg/L for  $\text{Mg}^{2+}$ .<sup>24</sup> Elevated levels pointed to very hard groundwater possibly because of polyvalent metallic ions' seepage from contaminated soils, solid waste, and sedimentary rocks of the vicinity. According to Barakat et al,<sup>25</sup> elevated levels of  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  in groundwater resulted from leaching of their  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{CO}_3^{2-}$  salts whose sources were from solid waste dumping and lithogenic origin in a case study at Piedmont, Morocco.



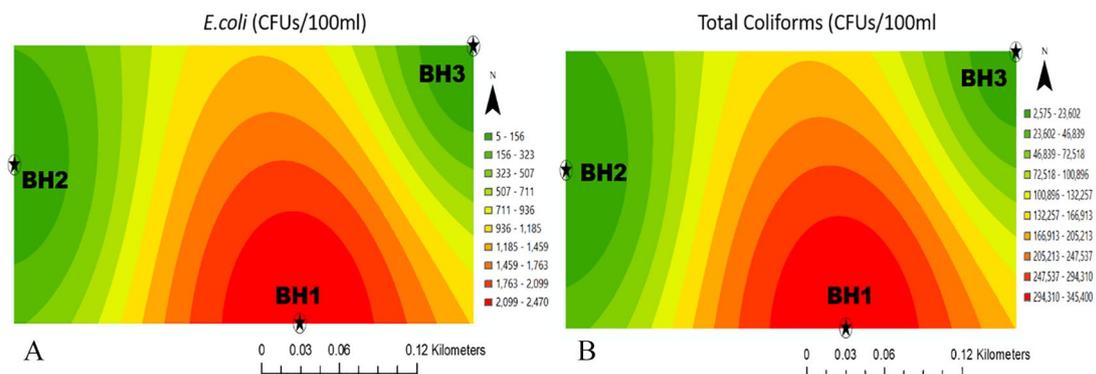
**Figure 5.** Chemical parameters of groundwater representing (A)  $\text{Na}^+$ , (B)  $\text{NH}_4^+$ , (C)  $\text{Fe}^{2+}$ , and (D)  $\text{Mn}^{2+}$ . BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3.



**Figure 6.** Chemical parameters of groundwater in the study area representing (A)  $\text{Cl}^-$ , (B)  $\text{SO}_4^{2-}$ , (C)  $\text{NO}_3^-$ , and (D)  $\text{F}^-$ . BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3.

Ammonium levels of groundwater ranged from 0.2 to 25 mg/L (Figure 5B) and BH 1 areas had the highest levels. Most of the region had levels  $>1.5$  mg/L, which is the SANS permissible limit. Elevated levels possibly indicated the leaching of nitrogen-rich water from landfill leachate and the decay of organic matter in the vicinity. Elevated levels of  $\text{NH}_4^+$  in groundwater of El-Arab landfill vicinity of Egypt were attributed to leachate contamination.<sup>4</sup> Spatial distribution of  $\text{Na}^+$ ,  $\text{Fe}^{2+}$ , and  $\text{Mn}^{2+}$  concentrations are shown in Figure 5A to D, respectively. Levels of  $\text{Na}^+$  ranged from 62 to 245 mg/L and

elevated levels beyond the SANS 200 mg/L limits were areas near BH 3. Levels of  $\text{Fe}^{2+}$  and  $\text{Mn}^{2+}$  were beyond the allowable limits of 300 and 100  $\mu\text{g/L}$ , respectively, in the entire study area. The 3 elements depicted area groundwater to be of poor aesthetic appearance and possibly corresponded to their natural origin in addition to anthropogenic sources through the leaking of landfill leachate. According to Sidibe et al,<sup>26</sup>  $\text{Fe}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{Na}^+$  levels are elevated in groundwater due to weathering of rocks and minerals that contain the elements or the generation of leachate from dumped solid waste.



**Figure 7.** Chemical parameters in sampled groundwater representing (A) *Escherichia coli* and (B) total coliforms. BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3; CFU, colony forming units.

Figure 6 showed the spatial distribution of  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ , and  $\text{F}^-$  ions in sampled groundwater. Chloride levels ranged from 93 to 310 mg/L and were beyond the SANS allowable limits of  $\leq 300$  mg/L in areas near BH 3. Levels of  $\text{F}^-$  ranged between 0.1 and 8.1 mg/L and most regions of the study area were beyond the  $\leq 1.5$  mg/L allowable limits by SANS. Although  $\text{Cl}^-$  and  $\text{F}^-$  occur in groundwater naturally, observed elevated levels could be attributable to anthropogenic sources such as landfill leachate seepage considering that the study area is a natural grassland. In major cities of Indochina Peninsular countries, anthropogenic practices such as poor solid waste and wastewater management were blamed for the elevated levels of  $\text{Cl}^-$  in groundwater.<sup>27</sup> Fluoride contamination of groundwater was associated with disposal of solid wastes such as ceramics in open dumpsites.<sup>4,14,23</sup>

Nutrient composition of groundwater was assessed using  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  (Figure 6B and C). Nitrates which are important in the development and growth of algae ranged between 2 and 28 mg/L in the study area. Regions near BH 1 had elevated levels beyond SANS recommendations of  $\leq 11$  mg/L while sulphate concentrations in the region were within the permissible limits of  $\leq 250$  mg/L. Elevated  $\text{NO}_3^-$  levels could be attributable to the oxidizing effect of landfill leachate, which results to water nitrification where ammonia is oxidized to nitrates while sulphates are reduced, hence the lower concentrations of the latter. A similar trend was observed in a Croatian catchment located near a landfill whereby elevated levels of  $\text{NO}_3^-$  in groundwater were attributable to leachate's redox capacity on soils and the vadose zone during infiltration.<sup>28</sup> Similarly, high nutrient content in groundwater was an indicator of anthropogenic-based pollution from landfill leachate in Egypt.<sup>4</sup>

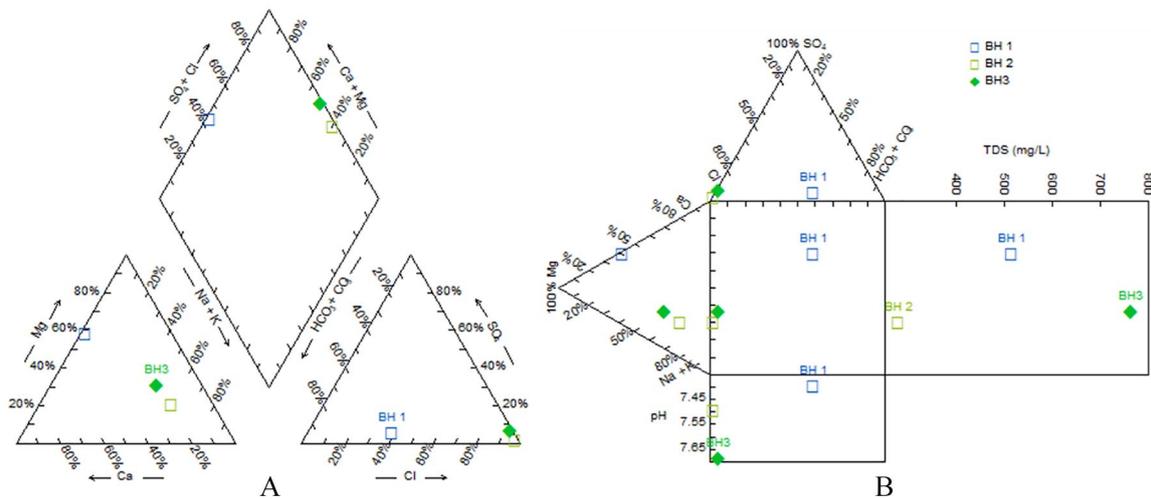
Results on the spatial distribution of faecal coliforms and total coliforms in the study area were as shown in Figure 7. All boreholes and the entire area had detectable *E. coli* colony forming units (CFUs). This was above the 0 CFU/100 mL recommendations by SANS and points to the presence of disease-causing organisms in the groundwater. A study by Wanda et al,<sup>7</sup> in groundwater of Northwest province, South Africa, detected *E. coli* and attributed it to the presence of

pathogens that rendered it harmful for consumption. *E. coli* could be an indication of sewage and leachate contamination in groundwater as established in 3 Polish landfill by Talalaj and Biedka.<sup>29</sup> Total coliforms were detected throughout the study area and their levels were beyond SANS  $< 10$  CFU/100 mL limit. This observation could be due to leachate contamination in groundwater and could indicate the presence of pathogens. In Pondicherry and Tamil Nadu states of India,<sup>30</sup> TC detected in groundwater was attributable to the entry of contaminated leachate from the vicinity and had disease-causing microorganisms.

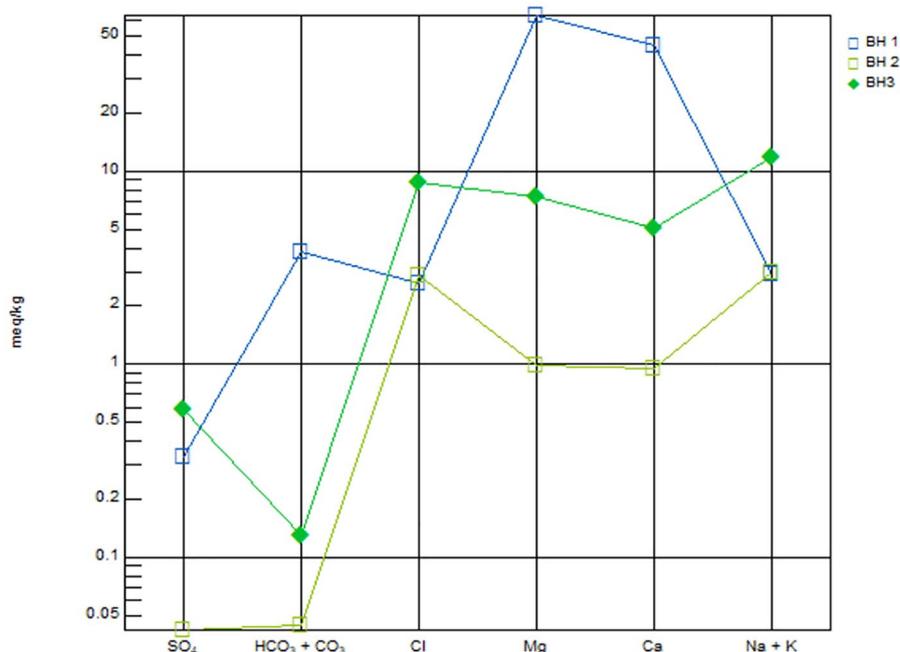
### Hydrochemical Faces of Groundwater

Piper and Durov diagrams were used to represent the hydrochemical faces of sampled water as showed in Figure 8. As Handa<sup>16</sup> explained, various faces were identified in the boreholes as shown in Figure 8A. Borehole 1 had Mg-type of water contrary to borehole 2 (BH 2) and BH 3 that had sodium plus potassium type. Chloride type of water was dominant in 2 boreholes with exception of BH 1 that had bicarbonate water. A combination of both cations and anions classified waters from BH 1 as magnesium bicarbonate, BH 2 was sodium chloride and BH 3 was mixed. A study in Karnataka state of India established that groundwater of the area was dominated with  $\text{Ca}^{2+}$ - $\text{Mg}^{2+}$ - $\text{HCO}_3^-$  ions in a hydro-chemical analysis using this approach.<sup>17</sup> Different ions were localized in various sampling sites. A research by Sugghosh et al<sup>18</sup> established that groundwater of Uttar Pradesh region of India had localized distribution of cations at various sampling sites using a Piper diagram. Sampled groundwater in BH 1 had high alkaline earth metals and was characterized by strong acids unlike BH 2 and BH 3 that had moderate amounts of these metals. These results agreed with a hydrogeochemical analysis of groundwater from SRLIS river basin (India), whereby alkaline earth metals were dominant.<sup>17</sup>

Results of the Durov diagram (Figure 8B) showed localized distribution of cations just as in the Piper diagram. Borehole 1 showed simple dissolution and mixing of cations while BH 2



**Figure 8.** Hydrogeochemistry of groundwater in the study area representing (A) Piper and (B) Durov diagram. BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3.



**Figure 9.** Schoeller presentation of chemical aspects of sampling sites. BH1 indicates borehole 1; BH2, borehole 2; BH3, borehole 3.

showed high dominance of Na<sup>+</sup> and Cl<sup>-</sup>, hence the possibility of reverse ion exchange. A similar trend was reported using this approach in a hydro-chemical analysis of groundwater in India whereby some sampling sites exhibited high dominance of Na<sup>+</sup> and Cl<sup>-</sup> ions while others had mixed ion dissolution where none of the ions were dominant.<sup>17</sup> The presence of anions Cl<sup>-</sup> as dominant anions localized in BH 2 to 3 could be associated with anthropogenic contamination. A similar trend was demonstrated using a Durov diagram in a hydrogeochemical evolution assessment of groundwater in India whereby Cl<sup>-</sup> dominated waters occurred.<sup>18</sup> The dominance of Na<sup>+</sup> and Mg<sup>2+</sup> in BH 2 to 3 and BH 1, respectively, compared with Ca<sup>2+</sup> is an indicator of foreign pollution to groundwater. According to Hussien and Faiyad,<sup>31</sup> natural groundwater has

high levels of Ca<sup>2+</sup> but the dominance of Na<sup>+</sup> and Mg<sup>2+</sup> cations results from dissolution of polluted rocks and soils.

Unique aspects of each sampling sites were presented using the Schoeller diagram as shown in Figure 9. Borehole 1 showed high concentrations of all assessed parameters with exception of Cl<sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, which corresponded to the highest pollution compared with BH 2 to 3. High levels of cations (Mg<sup>2+</sup> and Ca<sup>2+</sup>) corresponding to enriched water was evident in all boreholes. Groundwater quality assessment in Rafsanjan region of Iran reported high levels of cations using this approach and attributed it to pollution of urban water.<sup>32</sup> Shapes of Schoeller representations of sampling sites in this study were typical of polluted water as was established in a geochemical analysis of groundwater in Khorramabad, Iran, whereby peaks of ions

concentrations displayed using this approach were correlated to pollution.<sup>33</sup> Hoaghia et al<sup>34</sup> also noted that high peaks in Schoeller diagrams are geochemical signatures of pollution in a study that assessed contamination in groundwater of Sibiu county of Romania.

## Conclusions

Spatial representation of physicochemical parameters of groundwater in Roundhill landfill vicinity and its hydrogeochemical analysis made several revelations regarding its nature. First,  $\text{Fe}^{2+}$ ,  $\text{Mn}^{2+}$ , and  $\text{NH}_4^+$  that surpassed SANS allowable limits confirmed pollution of groundwater in most of the study area. Pollution was dominant in BH 1 whereby most parameters were elevated compared with other boreholes. A hydrogeochemical analysis using Durov diagram depicted reverse ion exchange in BH 2 to 3 due to  $\text{Na}^+$  and  $\text{Cl}^-$  dominance while BH 1 showed simple ion mixing and had no dominant ions. Magnesium carbonate water was deduced from BH 1 that was dominated with strong acids and alkaline earth metals using the Piper diagram while BH 2 had sodium chloride water. The dominance of cations ( $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ ) compared with anions in all boreholes suggested anthropogenic pollution. Roundhill landfill leachate is a probable cause of pollution in the area.

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## Author Contributions

JN undertook the groundwater sampling, analysis and prepared the manuscript. EO supervised the sampling, analysis of groundwater and corrected the draft manuscript.

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