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# Appraisal of surface-groundwater anthropogenic indicators and associated human health risk in El Sharqia Governorate, Egypt

Abdel Hameed M. El-Aassar<sup>a</sup>, Rasha A. Hussien<sup>b</sup>, Faten A. Mohamed<sup>b</sup>, Selda Oterkus<sup>c</sup> and Erkan Oterkus<sup>c,\*</sup>

<sup>a</sup> Egypt Desalination Research Center of Excellence (EDRC) and Hydrogeochemistry Department, Desert Research Center (DRC), Cairo, Egypt

<sup>b</sup> Nuclear and Radiological Safety Research Center, Egyptian Atomic Energy Authority (EAEA), Cairo, Egypt

<sup>c</sup> PeriDynamics Research Centre (PDRC), University of Strathclyde, Glasgow, United Kingdom

\*Corresponding author. E-mail: erkan.oterkus@strath.ac.uk

#### ABSTRACT

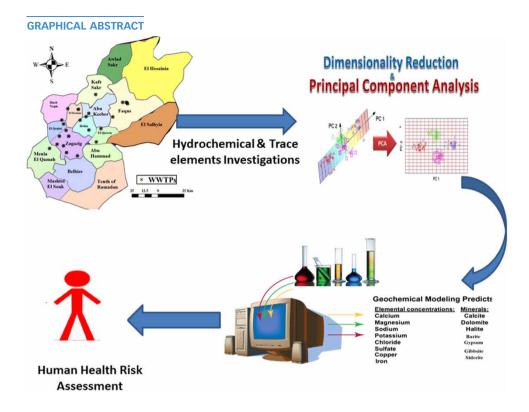
The aim of this study was to integrate hydrogeochemistry with a multivariate statistical approach to understand the various processes that control the evolution/contamination of water resources in El Sharqia Governorate, Egypt with a particular emphasis on direct/indirect risks to human health. To achieve this, a representative collection of 21 groundwater and 35 drainage samples was taken and examined for physical, chemical, and trace element measurements. Results indicated that in shallow groundwater and drainage water samples, the relative abundance of major cations is  $Na^+ > Mg^{2+} > Ca^{2+} > K^+$ , and for anions it is  $HCO_3 > Cl^- > SO_4^{2-}$  (on a molar basis). Natural processes involving the dissolution/precipitation of some minerals and other processes including leaching of solid waste, overuse of agricultural fertilizers application, and high loads of discharged sewage water are responsible for the evolution of water resources in El Sharqia Governorate. Ammonia, nitrate, biological oxygen demand (BOD), phosphate, turbidity, iron, manganese, lead, and aluminum concentrations were found to be higher than the limits set by internatio2nal drinking water regulations. The health risk index (HRI) values for children were found to be higher than these for adults when the water resources are used for drinking purposes, which poses a risk to human health.

Key words: Egypt, El Sharqia Governorate, geochemical modeling, human health risk, hydrochemistry, modeling mixing process

# HIGHLIGHTS

- An integration of hydrogeochemistry with a multivariate statistical approach was applied.
- Various processes that control the evolution/contamination of water resources in El Sharqia Governorate, Egypt were investigated.
- A particular emphasis on direct/indirect risks to human health was given.
- A risk to human health was found.
- HRI values for children were higher than those for adults when used for drinking purposes.

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# **1. INTRODUCTION**

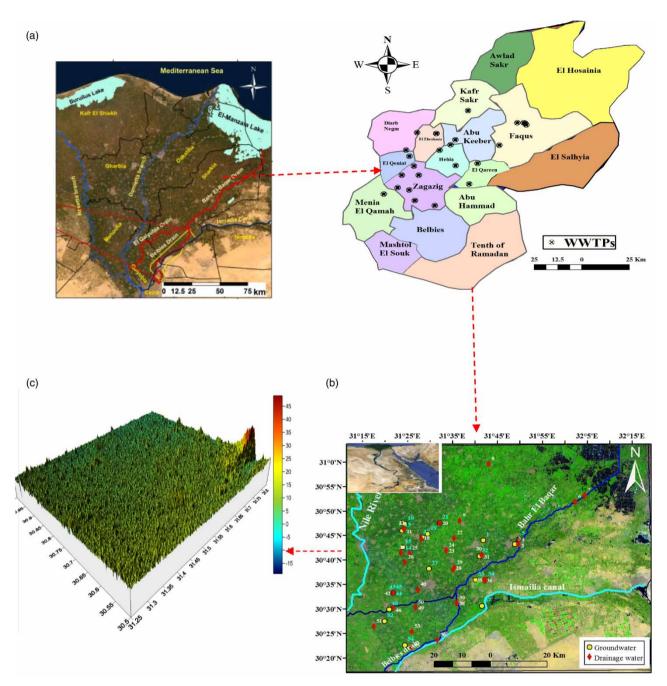
Water is an essential part of our life; however, freshwater makes up a tiny portion of only 0.01% of the entire amount of water on the earth's surface (Amini et al. 2016; Moghaddama et al. 2018) and the vast bulk of the water on the earth's surface is saline or salt water (with an average salinity of 35%). Increasing human population, rapid urbanization rate, and industrial expansion led to an increment of water necessities all over the world, especially in developing countries. Water resources can be surfaces such as lakes, canals, or groundwater that are buried under the ground surface in geological formations (aquifers). In general, two main constraining factors defining water suitability for different agricultural, domestic, and industrial purposes are water availability or quantity and quality. Relying on surface water alone is not sufficient to meet these demands because of its limitations and inadequacy in most arid and semi-arid areas of developing countries. Consequently, reliance on groundwater resources has dramatically increased in these regions (Zhou et al. 2020). However, human activities have negative impacts on groundwater quality. Several anthropogenic pollution sources were found to be responsible for deterioration of water resources in developing countries such as industrial wastewater discharges, use of fertilizers and pesticides, wastewater irrigation, and atmospheric transportation that have led to the accumulation of heavy metals in the soils worldwide, especially in developing countries. The risks to human health caused by exposure to heavy metals present should not be disregarded in light of the aforementioned issues. It is crucial to assess the level of heavy metal pollution in soil and agricultural regions as well as the potential health risks caused by these toxicants. Non-point source pollution from discharging wastewater is the greatest threat to the sustainable use of water resources (surface and groundwater) in megacities. Excess untreated wastewater from villages and rural areas in the majority of developing countries is frequently discharged directly into water pathways, while household, commercial, and industrial effluents, as well as raw, untreated sewage are frequently dumped into surface and groundwater sources. Rainstorms eventually wash the wastewater into the water bodies or it percolates there. Stagnating wastewater in open lagoons and on the sides of the road frequently serves as a mosquito-breeding ground and a haven for various germs and viruses. Additionally, harmful substances like oil and grease, insecticides, ammonia, and heavy metals are present in wastewater pools. When point source pollution is reduced in many countries (even if wastewater treatment plants (WWTPs) begin to reach their capacity limits), climate (global) change impacts could increase pollution due to urban or agricultural run-off. In recent years, groundwater investigations were paying attention to assessing and understanding the hydrochemical characteristics and water quality using several effective tools, including geochemical modeling, Geographical Information System (GIS), statistical approaches (multivariate statistical analysis), and water quality indices. Many factors control groundwater hydrochemistry including dissolution, precipitation, ion-exchange, sorption-desorption together with residence time along the flow path. A number of geochemical models were developed for tracing groundwater evolution along the flow path through inverse modeling or studying different mixing processes affecting the quality of water resources in the studied areas. The use of geochemical models is increasing in addressing groundwater quality problems involving geochemistry (Slimani et al. 2015; Berihu et al. 2017; Liu et al. 2020). Assessment of the contamination risk of water resources to human health is a useful technique for determining the potential negative consequences of environmental pollutants. This method has been used extensively to estimate the heavy metal contamination of drinking water and soils (Liu et al. 2016; Fakhri et al. 2018; Kamani et al. 2018; Keramati et al. 2018a, 2018b; Rezaei et al. 2018; Youssef et al. 2018). Recently, many studies have focused on this field (Sohrabi et al. 2016; Fakhri et al. 2018; Kamani et al. 2018; Keramati et al. 2018a, 2018b; Rezaei et al. 2018; Youssef et al. 2018) and this technique has been widely utilized to determine the level of heavy metal contamination of soils and drinking water (Liu et al. 2016, Fakhri et al. 2018). The main objectives of the present study is an integration of hydrochemical and multivariate statistical investigations to evaluate major and minor nutrients including chemical and trace elements measurements (ammonia, nitrate, phosphate, biological oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon (TOC), oil, and grease) to identify anthropogenic indicators for surface and groundwater resources. There are also geochemical modeling applications, especially for mixing processes that affect the chemical composition and water quality of El Sharqia Governorate. Finally, we assess potential human health risk hazards of trace elements for different age groups of people including children and adults who probably rely on these water resources for different purposes.

# 2. STUDY AREA DESCRIPTION

The Governorate of El Sharqia under study is situated in the northern Nile Delta of Egypt, approximately between longitudes 31° 15' and 32° 15' E and latitudes 30° 20' and 31° N with a 4,922 km<sup>2</sup> territory, it overtakes Al-Behaira Governorate as the second-largest governorate in the Nile delta region. According to the Egyptian Central Agency for Public Mobilization and Statistics, its population exceeded 7 million inhabitants per capita in 2017 and is projected to reach 10 million inhabitants per capita in 2032. The Eastern Delta region, Egypt, was taken as a pilot area that suffers from surface and groundwater deterioration due to excessive use of treated/untreated sewage effluent for irrigation purposes causing many human health hazards as shown in Figure 1(a). The study area is currently served by a sewerage system including integrated WWTPs as shown in Figure 1(b). The combined primary and secondary treated wastewater, with agricultural and industrial wastes, are discharged to some drainage channels combined with the untreated excess flow that is disposed of directly to the nearby main drains and then to Manzala Lake. Please note that drainage water in the study area implies streams that contain the excess of irrigated water seepage from the surrounding agricultural fields. Several studies on soil productivity potentials, hydrochemical and groundwater quality evaluation (Embaby et al. 2014; Mansour 2020), seasonal variations in the microbiological and physiochemical characteristics of municipal wastewater (Mahgoub et al. 2015), the effects of on-site sewage disposal on groundwater at Minia Al-Qamh, (Atwa et al. 2015), sedimentological and hydrochemical studies of groundwater (Mabrouk et al. 2016), hydrogeological investigations of the quaternary aquifer (El-Sayed et al. 2011), and soil productivity potentials (Rashed 2016) were all conducted in the El Sharqia Governorate. The values of ammonia, nitrate, BOD, phosphate, turbidity, iron, manganese, lead, and other contaminants were found to exceed the limits of drinking water international standards.

#### 3. GEOLOGICAL AND HYDROLOGICAL SETTINGS

Deposits from the quaternary and tertiary acquifers make up the majority of the eastern Nile Delta as shown in Figure 2. The Nile sediments that occasionally have sands blown into them define the quaternary deposits. The sand, clay, and gravel content of the sediments vary both laterally and vertically. Uncomfortably, such sediments are present on the older units of Pleistocene tertiary. Early, Middle, and Late Pleistocene are used to categorize ancient deltaic deposits. Loose sands with cherty pebbles are a good representation of early Pleistocene deposits. They get thicker in the north, reaching a maximum thickness of roughly 900 m in El Matariya, and get thinner in the east. As a result, the deposits from the Middle Pleistocene are referred to as Mit Ghamr Formation. Sands and gravel with sporadic, discontinuous clay lenses serve as their



**Figure 1** | (a) Map of the study area with drainage and groundwater sample locations; (b) location of WWTPs in El Sharqia Governorate; and (c) the digital elevation model (DEM).

representation. The sandy aquifer of the eastern Nile Delta is densely covered in erratic paleo-topography, including submerged Gizera sands and abandoned canals. Fluvial sediments serve as a representation of the top portions. Fluvial sediments form the higher portions. The Mit Ghamr formation in general and these sands, in particular, constitute significant parameters in the restriction of pollution of this aquifer. Sand and clay intercalations, in certain places covered by a thin, hard, crusty, sandy limestone deposit, define the Late Pleistocene.

As a result, clay was hydrogeologically partitioned from fluviatile deposits (river sands), fine to medium sands with thin intercalations of clay and silt, with a maximum thickness of around 30 m. This reflects the floodplain deposits of the former Damietta Branch. It symbolizes the upper, unconfined aquifer with intercalated clay. In the western regions, the

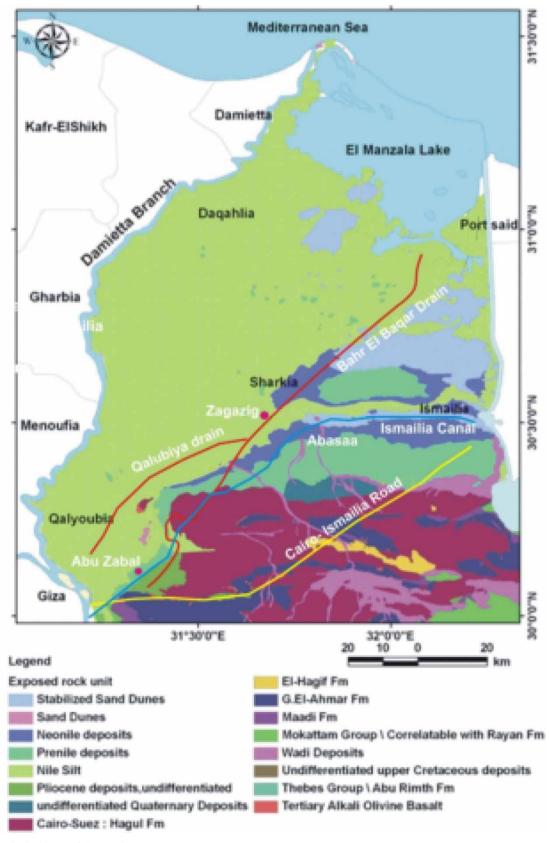


Figure 2 | Geological map of the study area.

ratio of sand to clay can exceed 50%, while it declines in the east and north. This series' alternation of coarse and fine particles supports the idea that these sediments date from the Late Pleistocene.

# 4. MATERIALS AND METHODS

# 4.1. Sample collection and analysis

Fieldwork was conducted in 2021 by collecting a representative set of 35 drainage and 21 groundwater samples. These water samples were collected in 500-mL pre-washed polyethylene bottles with de-ionized water. The samples were kept at 4 °C in the laboratory to prevent microbial changes in water chemistry. The samples were geo-referenced using GPS (Trimble, Juno S-3 model). Samples were analyzed at the hydrogeochemistry department of Desert Research Center (DRC) according to the methods adopted by the United States Geological Survey (Rainwater & Thatcher 1960; Fishman & Friedman 1989) and the American Society for Testing and Materials (ASTM 2002). The chosen wells have a depth range of 8-13 m below the surface. Major cation (Ca, Mg, Na, and K) and anion (Cl, SO<sub>4</sub>, and HCO<sub>3</sub>) concentrations were measured as chemical parameters. At the time of the sampling, all wells in El Sharqia Governorate were in operation (Figure 1). Using conventional EDTA, total hardness (TH) of CaCO<sub>3</sub> and Ca<sup>2+</sup> was examined. Mg<sup>2+</sup> was determined. A flame photometer was used to measure the levels of Na<sup>+</sup> and K<sup>+</sup>. By titrating with HCl, the total alkalinity, CaCO<sub>3</sub>, CO<sub>3</sub><sup>2-</sup>, and HCO<sub>3</sub><sup>-</sup> were calculated. Using the conventional Hg (NO<sub>3</sub>)<sub>2</sub> titration, Cl was determined. A UV/Visible spectrophotometer was used to analyze  $SO_4^2$  and NO<sub>3</sub>. Trace element contents (Al<sup>3+</sup>, Cd<sup>2+</sup>, Cr<sup>3+</sup>, Cu<sup>+</sup>, Fe<sup>2+</sup>, Mo<sup>2+</sup>, Mn<sup>2+</sup>, Ni<sup>2+</sup>, Pb<sup>2+</sup>, V<sup>5+</sup>, and Zn<sup>2+</sup>) of the water samples were determined using inductively coupled argon plasma (ICP). The obtained chemical data are expressed in milligrams per litre (mg/l). Some parameters including the depth to water, total well depth, pH, temperature, EC, CO<sub>2</sub><sup>-</sup>, HCO<sub>3</sub><sup>-</sup>, TOC, COD, and NO<sub>3</sub><sup>-</sup> were determined in situ using pH, EC meter, 3510 Jenway – UK, for CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup> titrimetrically against sulfuric acid by neutralization and measure of TOC, COD, and NO<sub>3</sub><sup>-</sup> by using compact photometer PF-12Plus (Macherey-Nagel GmbH & Co. KG, filter photometer). The units of measurement for all parameters are mg/l and meg/l. The charge balance between the difference of cations and anions (expressed in meq/l) divided by their sum was used to evaluate the quality of the data:

$$\frac{\sum (\text{Cations} - \text{Anions})}{\sum (\text{Cations} + \text{Anions})} \times 100 \tag{1}$$

# 4.2. Geochemical modeling

In the domains of hydrogeology and geochemistry, the saturation index is a crucial geochemical statistic that is frequently helpful for determining the presence of several common minerals in the groundwater system (Deutsch 1997). The following equation (Lloyd & Heathcote 1985) was used in this investigation to determine saturation indices (SIs):

$$SI = \frac{\log(IAP)}{K_s(T)} \tag{2}$$

where  $K_s$  (T) is the equilibrium constant of the reaction taken into consideration at the sample temperature, and IAP is the relevant ion activity product, which may be computed by multiplying the ion activity coefficient *I* and the composition concentration mi. SI equals zero when the groundwater is fully saturated with certain minerals; positive SI values indicate oversaturation, and negative SI values indicate undersaturation (Appelo & Postma 1994; Drever 1997). The two or more endmembers that participate in the mixing process inside the aquifer and contribute to recharge are identified using a simulation of the mixing process. Finally, the modeled composition of the mixture is compared to the actual composition of water found at the quaternary aquifer.

#### 4.3. Human health risk assessment

The phrase 'exceedance level,' which refers to a unitless notion, is used to describe the extent to which various PTMs (potential toxic metals) exceed their respective World Health Organization (WHO) permitted limits and can be expressed as:

$$Exceedance \ level = \frac{Concentration \ of \ a \ quality \ parameter}{WHO \ acceptable \ limit}$$

In the studied area, both anthropogenic and natural factors contributed to the contamination of the majority of the groundwater. Additional analysis, including chronic daily intake (CDI) and health risk index (HRI) calculations, should be made to determine how much human health risk exposure is there (HRI). The consumption rate of element concentrations and the kind of toxicity are the key determinants of chronic health risk indices related to water consumption.

# 4.3.1. Chronic daily intake

The majority of PTMs enter the body through a number of different routes, including ingestion, inhalation, and cutaneous exposure. The most prominent route for PTMs to reach the human body is by oral intake. According to the equation modified by Khan *et al.* (2013a) and Khan, Shahnaz *et al.* (2013b), the CDI of PTMs by drinking groundwater was computed as follows:

$$CDI = CPTMs \times \left(\frac{DIPTMs}{bw}\right)$$
(4)

In this formula, CPTMs stand for PTM concentrations, DIPTMs for daily water intake and bw for body weight. For adults and children, respectively, the projected daily groundwater consumption rates were 2 and 1 l/day, while the assumed average body weights were 70 and 20 kg, respectively.

#### 4.3.2. Health risk index

The HRI of PTMs for humans is calculated by substituting the values of PTMs in the following equation as (USEPA 2005; Shah *et al.* 2012):

$$HRI = \frac{CDI}{RfD}$$
(5)

RfD stands for the reference dose. Based on the estimated results, it was stated that the HRI values less than 1 revealed that there was no risk to humans posed by the ingestion of groundwater through drinking. The United States Environmental Protection Agency (USEPA 2005) defined the RfD as 'an estimate of a daily oral exposure to the human population that is likely to be without an appreciable risk of deleterious effects during a lifetime.' HRI levels greater than 1, however, indicate a risk to human health.

#### 4.3.3. Carcinogenic analysis

Carcinogenic analysis of the probable cancer risks due to exposure to a specific dose of heavy metal in drinking water can be computed using the ILCR (Sultana *et al.* 2017). The ILCR is defined as the incremental probability of a person developing any type of cancer over a lifetime as a result of 24 h/day exposure to a given daily amount of a carcinogenic element for 70 years. The following equation (Equation (6)) was commonly used for the calculation of the lifetime cancer risk:

$$ILCR = CDI.CSF$$

(6)

where CSF is the cancer slope factor and is defined as the risk generated by a lifetime average amount of 1 mg/kg/day of carcinogen chemical and is contaminant specific. The permissible limits are considered to be  $10^{-6}$  and  $<10^{-4}$  for a single carcinogenic element and multi-element carcinogens (Tepanosyan *et al.* 2017).

# 5. RESULTS AND DISCUSSION

#### 5.1. Descriptive statistics of hydrochemical parameters

Table 1 shows the descriptive statistics of the physicochemical characteristics and trace element results of drainage and groundwater collected in El Sharqia Governorate, with minimum, maximum, and average values for all parameters in the quaternary aquifer compared with guideline values set by the Environmental Protection Agency (USEPA 2009) and WHO (2017) guidelines for drinking water purposes. These results indicate the following:

• The average pH of groundwater samples was 7.58, with values ranging from 7.06 to 8.19. The pH ranged from 6.54 to 7.6, with an average value of 7.08, for drainage water samples. The results showed circumstances that ranged from being slightly acidic to slightly alkaline, which may be related to pollution issues.

	Element	Min.	Max.	Average	SD		Min.	Max.	Average	SD	WHO (2017)
Groundwater	Na(meq/l)	1.30	10.00	4.86	50.36	Drainage	3.04	12.61	6.43	49.99	8.696
	K (meq/l)	0.05	1.45	0.20	11.79	water	0.18	1.28	0.5	7.907	0.256
	Mg (meq/l)	1.01	6.86	3.40	22.16		0.81	7.57	3.73	20.14	2.500
	$Ca \ (meq/l)$	0.81	8.98	3.43	38.29		1.61	8.3	3.45	23.72	3.750
	CO <sub>3</sub> (meq/l)	0.00	2.24	0.85	20.24		0	0.98	0.23	8.29	
	HCO <sub>3</sub> (meq/l)	2.24	7.42	4.73	93.7		2.1	10.92	5.7	121.79	4.918
	$SO_4 (meq/l)$	0.12	5.20	2.29	83.02		1.04	6.66	3.19	65.29	10.417
	Cl (meq/l)	0.68	12.23	4.18	114.45		2.33	10.96	5.21	63.75	7.042
	$NH4^+$ (mg/l)	1.96	54.88	13.32	13.6		3.92	41.16	22.83	9.31	0.5
	NO <sub>2</sub> (mg/l)	0	2.17	0.2883	0.5		0.014	0.925	0.221	13.8	
	$NO_3$ (mg/l)	1.96	37.24	17.44	10.18		0	56.84	21.9	0.178	45
	$PO_4 (mg/l)$	0.01	0.61	0.295	0.156		0.023	0.502	0.2488	0.11	
	F (mg/l)	2.4	16	8.64	5.39		1.6	41.6	13.82	10.5	
	BOD (mg/l)	0.00083	0.031	0.0108	0.008		0.00083	0.0366	0.0158	0.0103	
	COD (mg/l)	0	140	33.04	39.27		0	240	67.56	50.66	
	TOC (mg/l)	0	0.0381	0.0098	0.008		0	0.0902	0.01137	0.0153	
	Turbidity (NTU)	0.36	11.95	4.011	3.42		1.09	52.2	18.81	13.18	
	TSS	0	30	11.91	10.9		2	668	71.02	114.7	
	Oil and grease	0	161	56.04	43.11		0	160	65.27	44.43	
	pН	7.06	8.19	7.58	0.279		6.54	7.6	7.08	0.277	
	EC (µS/cm)	363	2,268	1,124	520		872	2,083	1,373	283.75	
	TDS (mg/l)	214	1,227	650	293		421	1,401	796	199.34	1,000
	Ag (mg/l)	0.006	0.054	0.030			0.009	0.093	0.033		,
	Al (mg/l)	0.023	0.776	0.231	0.18		0.028	5.703	0.654	0.96	
	B (mg/l)	0.008	0.192	0.072			0.030	0.189	0.084		
	Ba (mg/l)	0.019	0.286	0.139	0.08		0.003	0.165	0.074	0.04	
	Cd (mg/l)	0.003	0.026	0.010	0.006		0.001	0.040	0.012	0.008	
	Co (mg/l)	0.002	0.041	0.016	0.01		0.002	0.036	0.015	0.009	
	Cr (mg/l)	0.027	0.051	0.041	0.01		0.011	0.043	0.025	0.009	
	Cu (mg/l)	0.008	0.216	0.084	0.05		0.014	0.202	0.070	0.05	
	Fe (mg/l)	0.035	1.762	0.609	0.53		0.020	5.622	1.099	1.08	
	Mn (mg/l)	0.019	2.273	0.601			0.003	0.376	0.222		
	Mo (mg/l)	0.004	0.106	0.055			-0.044	0.117	0.055		
	Ni (mg/l)	0.002	0.056	0.033	0.01		0.005	0.059	0.030	0.014	
	Pb $(mg/l)$	0.041	0.160	0.117	0.06		0.009	0.266	0.094	0.066	
	Si (mg/l)	5.253	20.390	11.046			0.151	9.789	5.889		
	Sr $(mg/l)$	0.110	1.748	0.710	0.42		0.023	8.370	1.176	1.47	
	V (mg/l)	0.018	0.117	0.056	J		0.012	0.203	0.059		
	Zn (mg/l)	0.002	0.570	0.151	0.13		-0.005	0.524	0.095	0.09	

Table 1 | Descriptive statistics of physiochemical characteristics and trace element results of collected water samples

- The fact that drainage water had higher EC and TDS levels than groundwater may be attributable to anthropogenic pollution inputs. With an average of 1,373  $\mu$ S/cm, the EC of drainage water ranged from 872 to 2,083  $\mu$ S/cm. The average EC in groundwater samples was 1,124  $\mu$ S/cm, with values ranging from 363 to 2,268  $\mu$ S/cm. 19% of the collected samples had TDS values above the WHO (2017) standard limits for drinking water, which varied from 214 to 1,226 mg/l. TDS concentrations in drainage water samples ranged from 422 to 1,401 mg/l, exceeding WHO (2017) guidelines for drinking purposes by 12% (Table 1). In accordance with WHO salinity classifications from 2017, about 81% of groundwater samples and 89% of drainage water were categorized as fresh water, while 19 and 11%, respectively, were classified as somewhat saline.
- The relative abundance of major cations in shallow groundwater is  $Na^+ > Mg^{2+} > Ca^{2+} > K^+$  (on a molar basis) and  $HCO_3^- > Cl^- > SO_4^{2-}$  for anions by 43 and 38%, respectively. For drainage water samples, the predominant cations are  $Na^+ > Mg^{2+} > Ca^{2+} > K^+$  (on a molar basis) and  $HCO_3^- > Cl^- > SO_4^{2-}$  for anions by 51 and 57%, respectively.

- However, the maximum  $Ca^{2+}$  and  $Mg^{2+}$  concentrations in groundwater samples, 180 and 83 mg/l, respectively, are greater than WHO (2004) guideline limits by 38 and 62%, respectively. With the exception of sample number 54 (Na = 230 mg/l), the maximum sodium concentration values are 230 mg/l within the established limits. However, groundwater samples have potassium concentrations of 57 mg/l, which are 14% higher than WHO (2004) limits. The maximum salt and potassium values in drainage water are 12 and 97%, respectively, above WHO guideline limits. HCO<sub>3</sub><sup>-</sup> ions present in shallow groundwater samples may be due to the dissolution of carbonate rocks, soils, and atmospheric carbon dioxide.
- The first dominating anion in the study area is the bicarbonate ion. The lacustrine deposits present in the quaternary aquifer may be the source of the majority of Cl<sup>-</sup> in the groundwater. Sedimentary rocks like gypsum (CaSO<sub>4</sub>.2H<sub>2</sub>O) and anhydrite (CaSO<sub>4</sub>), which permit active dissolving, leaching, and ion-exchange processes, are the most prevalent and significant occurrences of sulfate ions in the researched area. The breakdown of organic matter in the soil and the addition of leachable sulphate in fertilizers used in intensively farmed areas are two additional sources of sulphate addition to groundwater.

#### 5.2. Assessment of pollution indicators in collected water samples

The environmental impacts of anthropogenic pollution on the general water resources quality in the study area will be evaluated according to the variations of ammonia, nitrate, phosphate, BOD, OD, TOC oil, and grease with spatial distribution as shown in Figures 4 and 5 and Table 1 as follows:

- The breakdown of organic substances containing nitrogen can produce ammonia in water. The main causes of ammonia contamination in the environment are the discharge of agricultural, industrial, and sewage effluent. Ammonia levels in the water are a sign of potential bacterial, sewage, and animal waste pollution. The tested groundwater and drainage water samples have ammonia concentrations that range from 1.96 to 54.88 mg/l and from 3.94 to 41.16 mg/l, respectively. All of these samples are above the NH<sup>+</sup><sub>4</sub> drinking water maximum suggested level of 0.5 mg/l. While nitrate is a naturally occurring substance in the environment, nitrite is rarely seen in large concentrations. However, nitrate can reduce to nitrite, and in reducing environment, toxicological effects could manifest. Agricultural practices, wastewater transporting human and animal excrement, and other factors can create nitrate contamination even though nitrate may be present in the environment naturally (Boyacioglu 2007; WHO 2011). Nitrate levels in the drainage water samples and groundwater samples that were examined ranged from 0 to 56.84 mg/l and 1.96 to 37.24 mg/l, respectively. Most drainage water samples are above the acceptable maximum limit (WHO 2017).
- Anthropogenic sources are responsible for high  $NO_3^-$ ,  $PO_4^{3-}$ , and  $Cl^-$  concentrations as illustrated in Figure 3 for the relationships of  $NO_3$  vs.  $PO_4$  and  $NO_3$  vs. Cl in (mg/l), respectively, in both drainage and groundwater samples.
- Dissolution/precipitation of organic materials and the presence of some metallic compounds like iron, manganese, and chromium, and the discharge of industrial effluents into surface water are just a few of the variables that can affect the turbidity and color of water systems. Color changes that result from these factors could potentially signal a dangerous issue (WHO 2011). Organic and/or inorganic substances that are suspended or colloidal in the water can also produce turbidity. Turbidity can be a symptom of potential microbial contamination because bacteria can cling to particles (TSE 1998; Cude

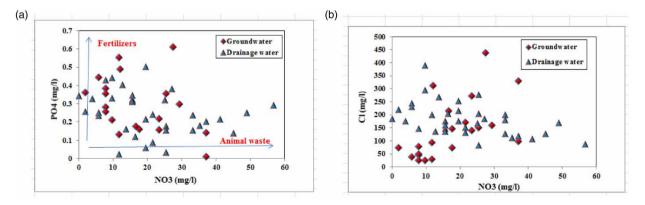


Figure 3 | Relationships of (a)  $NO_3$  vs.  $PO_4$  and (b)  $NO_3$  vs. Cl in (mg/l).

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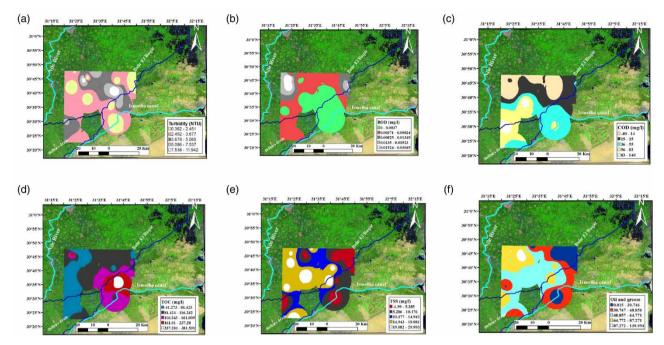


Figure 4 | Spatial distribution of (a) turbidity; (b) BOD; (c) COD; (d) TOC; (e) TSS; and (f) oil and grease for groundwater.

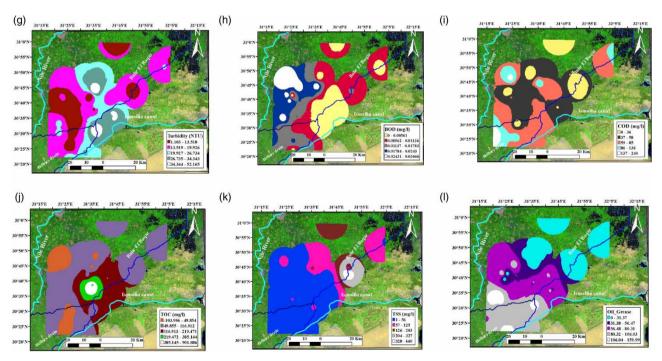


Figure 5 | Spatial distribution of (g) turbidity; (h) BOD; (i) COD; (j) TOC; (k) TSS; and (l) oil and grease for drainage water.

2001; WHO 2011; Akkoyunlu & Akiner 2012). The groundwater's mean value is below the allowable limit, according to the results for color and turbidity. As seen in Table 1 and Figures 4 and 5 drainage water has issues with turbidity, color, and/or clarity.

• The water body has been contaminated by both oxidizable organic and inorganic contaminants, according to the COD and TOC readings (Mohamed *et al.* 2015). The mean COD of groundwater and drainage water is 33.04 and 67.6 respectively.

Figures 4 and 5 reflect the degraded groundwater quality in this zone as a result of continuous sewage disposal in the study area especially around the main drains (Bahr El Baqr and Belbies). Poor water quality is indicated by high BOD values, which are associated with waste discharges that contain increased microbial activity brought on by the degradation of organic materials, as well as organic and nutritional content. In all of the wells assessed during the monitoring, BOD values were less than 5.0 mg O<sub>2</sub> L<sup>-1</sup>.

- For drainage and groundwater samples, it was found that some trace elements, including Al, Fe, Mn, Pb, Si, and Sr, exceeded WHO limits for drinking purposes.
- The concentrations of iron in the analyzed groundwater and drainage water samples vary from 0.03 to 1.76 mg/l and from 0.02 to 5.62 mg/l, respectively. All these samples exceed the maximum recommended limit for Fe<sup>3+</sup> in drinking water (0.3 mg/l) where various iron salts are used as coagulating agents in water-treatment plants and cast iron, steel, galvanized iron pipes are used for water distribution.
- The concentrations of  $Mn^{2+}$  in drainage and groundwater vary from 0.002 to 0.376 and 0.018 to 2.27 mg/l, respectively. Most of these samples exceed the maximum recommended limit for  $Mn^{2+}$  in drinking water which may be due to the discharge of industrial effluents from the steel and iron factories. Lead was one of the first non-ferrous metals used by man. It has been used in many industrial applications such as batteries and cable sheeting. Lead does not appear to be an essential element for life for any organism. It is less toxic to plants than mercury and copper, with adverse effects being noted at concentration levels between 100 and 5,000 µg/l.
- Lead is toxic to humans. It substitutes calcium in bone and accumulates in it. Lead poisoning is manifested by anemia, kidney disease, and disturbances of the central nervous system. Lead poisoning suffered in childhood may cause mental retardation and convulsions in later life. The limit of lead in drinking water is 0.01 mg/l (WHO 2011). Lead concentrations varied between 0.04–0.159 mg/l and 0.009–0.266 mg/l in groundwater and drainage water, respectively. The results suggested that most of the mean values of trace elements in drainage water were higher than those in groundwater. It could manifest that surface water exhibited relatively more pollution problems.

# 5.3. Geochemical modeling

Using the PHREEQC software, a geochemical model was created for potential hydrochemical reactions along the flow routes (Parkhurst & Appelo 1999). Groundwater solutes may be produced by three geochemical processes: evaporation, carbonate dissolution/precipitation, and silicate weathering (Garrels & MacKenzie 1971). Table 2 shows statistical outcomes of the saturation index of drainage and groundwater samples using PHREEQC. The findings showed that several minerals and gases, such

Minerals			Min.	Max.	Average		Min.	Max.	Average
Calcite	CaCO <sub>3</sub>	Groundwater	-0.493	0.848	0.29	Drainage water	-5.818	0.277	-0.45
Dolomite	CaMg(CO <sub>3</sub> ) <sub>2</sub>		0.04	2.773	1.67		-10.302	1.758	0.22
Cerussite	PbCO <sub>3</sub>		-0.905	-0.37	-0.56		-1.714	-0.297	-0.85
Smithsonite	ZnCO <sub>3</sub>		-3.122	-0.182	-1.40		-8.729	-1.146	-2.46
Strontianite	SrCO <sub>3</sub>		-2.225	-0.581	-1.32		-7.546	-0.1	-2.13
Witherite	BaCO <sub>3</sub>		-3.81	-1.872	-2.87		-9.18	-0.045	-3.64
Siderite	FeCO <sub>3</sub>		-0.757	0.749	-0.18		-6.369	0.79	-0.68
Rhodochrosite	MnCO <sub>3</sub>		-1.769	0.794	-0.22		-7.461	0.004	- 1.23
Anhydrite	$CaSO_4$		-3.697	-1.385	-2.23		-7.171	-1.278	-2.19
Gypsum	CaSO <sub>4</sub>		-3.407	-1.096	-1.94		-5.323	0.221	-1.71
Barite	BaSO <sub>4</sub>		-1.333	0.757	0.10		-7.859	0.767	-0.45
Celesite	$SrSO_4$		-4.08	-1.621	-2.51		-11.747	-1.357	-2.93
Halite	NaCl		-7.603	-5.834	-6.63		-6.834	3.917	-5.46
Gibbsite	Al(OH) <sub>3</sub>		1.837	4.272	3.17		2.323	4.642	3.67

Table 2 | Statistical results of the saturation index for the collected drainage and groundwater samples

as cerussite, smithsonite, strontianite, witherite, anhydrite, gypsum, celesite, and halite, were undersaturated in the drainage and groundwater samples of the quaternary aquifer, requiring dissolution to bring them into equilibrium. Minerals/gases like calcite, dolomite, siderite, rhodochrosite, barite, and gibbsite have a propensity to precipitate in drainage and groundwater samples.

# 5.4. Multivariate statistical analyses

With support for a wide range of factors, SPSS version 22.0 software was used to perform mathematical and statistical computations on the data (Matiatos *et al.* 2014).

# 5.4.1. Factor analysis

A statistical method for examining the correlations between numerous variables is factor analysis. This strategy entails minimizing information loss while condensing the data from a large number of original variables into a more manageable set of uncorrelated main components (factors) (Hair *et al.* 1992; Abu Salem *et al.* 2017; El Alfy *et al.* 2018).

# 5.4.2. Principal component analysis

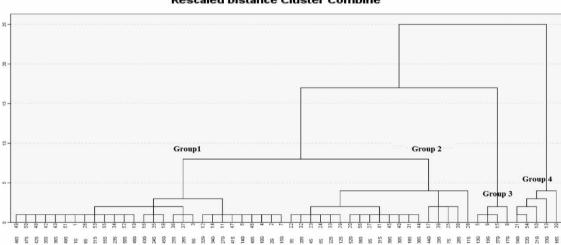
Principal component analysis (PCA) was done to determine the mechanisms controlling ion concentrations. The factor loadings of each original variable were investigated after extracting factors with eigenvalues greater than 1 (Kaiser 1960). Eight distinct components that account for the majority of the variability were identified using eigenvalues and varimax rotation. According to Table 3, the overall variance for the water samples was around 73.17%. Statistically, the first dominant factor (F1) is responsible for 20.79% of the variance in the data. Strongly positive loadings on EC, TDS, hardness, Na, SO<sub>4</sub>, Cl, and

	PC1	PC2	PC3	PC4	PC5	PC6	PC7	PC8
pН	- 0.600	- 0.163	- 0.129	-0.035	0.515	0.063	- 0.091	0.052
EC	0.948	-0.076	-0.063	-0.054	0.135	-0.090	0.053	0.015
TDS	0.967	-0.122	-0.034	-0.038	0.126	-0.081	-0.002	0.003
Hardness	-0.259	-0.126	0.209	0.703	0.206	-0.243	0.136	-0.017
Na	0.864	-0.192	0.089	-0.080	0.132	0.076	0.016	-0.013
$SO_4$	0.862	-0.018	-0.080	0.159	0.117	0.155	-0.051	0.046
Cl	0.877	-0.041	-0.030	-0.037	0.075	- 0.260	0.150	-0.065
Al	0.036	0.660	-0.293	0.262	-0.051	0.270	-0.239	-0.181
Cd	-0.104	0.234	0.715	-0.251	-0.166	0.142	0.119	-0.193
Co	0.007	0.354	-0.081	-0.539	0.225	0.330	0.224	0.089
Cr	0.044	0.524	-0.306	0.191	0.351	-0.063	0.065	-0.371
Fe	0.180	0.592	0.557	0.239	-0.077	0.097	-0.161	-0.002
Ni	0.170	-0.023	-0.125	0.039	-0.468	-0.306	-0.363	-0.294
Pb	-0.164	-0.098	-0.029	0.145	-0.234	-0.030	0.733	-0.397
Sr	-0.047	0.363	-0.081	0.080	0.149	-0.435	0.128	0.498
NO <sub>2</sub>	0.071	-0.253	0.011	0.643	0.055	0.299	0.233	0.317
NO <sub>3</sub>	-0.023	0.670	-0.050	-0.032	0.432	-0.071	0.200	0.015
$NH_4$	0.238	0.398	0.022	-0.071	-0.477	-0.015	0.385	0.098
TSS	0.163	0.329	0.828	0.017	0.139	0.045	-0.105	0.130
COD	0.184	0.570	-0.409	0.251	-0.116	0.353	-0.075	-0.037
TOC	0.158	-0.211	029	0.261	-0.388	0.490	-0.005	0.228
S	-0.016	0.409	-0.012	0.212	-0.310	-0.506	-0.071	0.138
Oil-grease	-0.110	0.272	-0.338	-0.320	-0.323	-0.042	0.075	0.394
% of Variance	20.795	12.500	9.061	7.725	7.330	6.055	5.048	4.659
Cumulative %	20.795	33.296	42.356	50.081	57.411	63.465	68.513	73.172

Table 3 | Loadings of the experimental variables on the significant principal component analysis (PCA)

pH are characteristics of this component. pH has a substantially negative loading. This element, which might also be called the salinity factor, reveals the impact of lithogenesis on the groundwater. The influence of sewage contamination can be seen in the second factor (F2), which accounts for 10.02% of the overall variation. It exhibits high positive loadings on Al and COD as well as moderately positive loadings on nitrate and Cr. Additionally, the third factor (F3) exhibits substantial positive loadings on Fe and TSS, indicating anthropogenic input for these elements, and it describes 9.72% of the overall variance. A strong positive loading on Na, a moderately negative loading on Co, oil, and grasses, and a moderately positive loading on NO<sub>2</sub> are all displayed by the fourth factor (F4), which accounts for 7.98% of the overall variation. With a substantial positive loading on Ni and Zn, the fifth factor (F5), which explains 8% of the overall variance, demonstrates anthropogenic impacts on groundwater. The final factor (F6), which contributes 7% of the overall variation, exhibits a high negative loading on Cd and a moderately positive loading on Cr (Table 3).

One method for identifying various classes and groupings within the studied data is hierarchical cluster analysis (HCA), with the outcomes shown as a dendrogram (Davis 1986). According to the proximity of the water quality measures, the



Dendrogram using Average Linkage (Between Groups) Rescaled Distance Cluster Combine

Figure 6 | Q-mode cluster analysis dendrogram for 56 samples and 23 chemical variables.

Table 4 | Parameter values of the four principal water groups

	Group 1			Group 2			Group 3			Group 4		
	Min.	Max.	Average									
pН	6.54	8	7.35	6.63	7.6	7.05	7.23	8.19	7.705	7	7.6	7.269
EC (µS/cm)	788	1,275	1,055	1,339	1,815	1,532	363	648	506	1,999	2,268	2,099
TDS (mg/l)	422	810	605	773	1,051	898	214	338	284	1,088	1,401	1,224
Ca (mg/l)	36.36	101	61.37	32.3	166	77.03	16.16	36.36	26	78.9	180	111
Mg (mg/l)	9.8	72	37.65	12.63	78.53	48.23	12	27	18.5	58	92	77
Na (mg/l)	30	150	102	120	220	170.8	32	86	59.33	150	290	213
K (mg/l)	2	30	13.3	5	31	17.47	2	8	4.667	6	57	29
CO <sub>3</sub> (mg/l)	0	67.2	16.8	0	29.4	5.8	0	42	18.2	8.4	50.4	28.8
HCO <sub>3</sub> (mg/l)	128	504	306	153	614	348	137	324	226	205	666	427
SO <sub>4</sub> (mg/l)	30	210	100.6	80	320	189	6	30	17.3	170	300	236
Cl (mg/l)	24.29	185	120	145.7	330	222	24.29	44	34	214	437	322

analyzed groundwater samples have been divided into four primary clusters: 1, 2, 3, and 4 as shown in (Figure 6). Total dissolved solids (TDS) and ionic composition, which are governed by hydrogeochemical and physicochemical circumstances, form the basis for the clustering of these samples.

# 5.5. Modeling of mixing groups

The clustering procedure was carried out by the Ward's linkage approach using the Euclidean distance as a measure of sample similarity, which was based on the clustering Q-technique, in which similarity associations among water samples were explored. Figure 6 shows the dendrogram's findings. Based on the dendrogram classification, four preliminary groupings were chosen. With an average value for each metric shown in Table 4, each group represents a hydrochemical facies.

Two triangles, one for cations and the other for anions, as well as a diamond-shaped region for both cations and anions are included in the Piper trilinear diagram (Piper 1994). Major cations ( $Ca^{+2}$ ,  $Mg^{+2}$ , and  $Na^+ + K^+$ ) and major anions ( $Cl^-$ ,  $SO_4$  and  $HCO_3^-$ ), as determined by chemical analysis, are plotted in the diamond shape (stated as meq/l). Figure 7 shows the outcomes of the four groups' chemical analyses. Except for a few examples of group 2 (Na–Cl) and group 3 (Mg–HCO<sub>3</sub>) water types, the majority of the groups are made up of mixed water types.

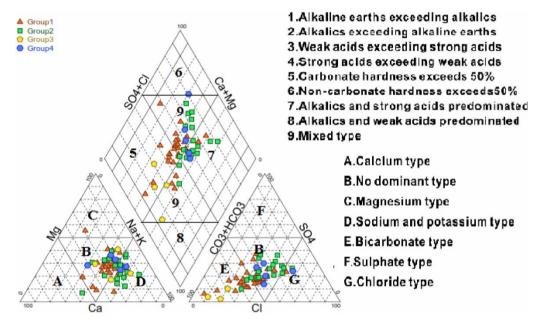


Figure 7 | Piper diagram for water samples.

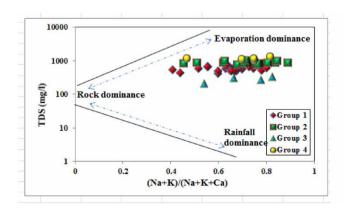
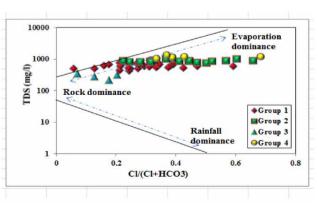


Figure 8 | Gibbs plot for the four principal water groups.



Mixing groups	HCO <sub>3</sub>	Ca	CI	к	Mg	Na	SO4
90% Group 1: 10% Group 2	0.004036	0.000897	0.001028	0.000059	0.000415	0.001697	0.000365
10% Group 1: 90% Group 2	0.003726	0.000816	0.003769	0.000120	0.000508	0.004831	0.000781
80% Group 1: 20% Group 2	0.003997	0.000887	0.001371	0.000067	0.000427	0.002089	0.000417
20% Group 1: 80% Group 2	0.003765	0.000827	0.003427	0.000113	0.000497	0.004439	0.000729
70% Group 1: 30% Group 2	0.003958	0.000877	0.001713	0.000074	0.000438	0.002480	0.000469
30% Group 1: 70% Group 2	0.003803	0.000837	0.003084	0.000105	0.000485	0.004047	0.000677
50% Group 1: 50% Group 2	0.003881	0.000857	0.002399	0.000090	0.000462	0.003264	0.000573
90% Group 2: 10% Group 3	0.003625	0.000766	0.003769	0.000120	0.000517	0.004840	0.000756
10% Group 2: 90% Group 3	0.003130	0.000444	0.001028	0.000059	0.000496	0.001775	0.000140
80% Group 2: 20% Group 3	0.003563	0.000726	0.003427	0.000113	0.000515	0.004457	0.000679
20% Group 2: 80% Group 3	0.003192	0.000484	0.001371	0.000067	0.000499	0.002158	0.000217
70% Group 2: 30% Group 3	0.003501	0.000685	0.003084	0.000105	0.000512	0.004073	0.000602
30% Group 2: 70% Group 3	0.003254	0.000524	0.001713	0.000074	0.000502	0.002541	0.000294
50% Group 2: 50% Group 3	0.003377	0.000605	0.002399	0.000090	0.000507	0.003307	0.000448
90% Group 3: 10% Group 4	0.003160	0.000560	0.001221	0.000061	0.000683	0.001906	0.000233
10% Group 3: 90% Group 4	0.003903	0.001814	0.005506	0.000143	0.002198	0.006017	0.001600
80% Group 3: 20% Group 4	0.003253	0.000717	0.001757	0.000072	0.000873	0.002420	0.000404
20% Group 3: 80% Group 4	0.003810	0.001657	0.004970	0.000133	0.002009	0.005503	0.001429
70% Group 3: 30% Group 4	0.003346	0.000873	0.002292	0.000082	0.001062	0.002934	0.000575
30% Group 3: 70% Group 4	0.003717	0.001500	0.004435	0.000123	0.001820	0.004989	0.001259
50% Group 3: 50% Group 4	0.003532	0.001187	0.003363	0.000102	0.001441	0.003961	0.000917
90% Group 1: 10% Group 4	0.004066	0.001014	0.001221	0.000061	0.000602	0.001828	0.000458
10% Group 1: 90% Group 4	0.004003	0.001864	0.005506	0.000143	0.002189	0.006008	0.001625
80% Group 1: 20% Group 4	0.004059	0.001120	0.001757	0.000072	0.000800	0.002350	0.000604
20% Group 1: 80% Group 4	0.004011	0.001758	0.004970	0.000133	0.001991	0.005485	0.001479
70% Group 1: 30% Group 4	0.004051	0.001226	0.002292	0.000082	0.000999	0.002873	0.000750
30% Group 1: 70% Group 4	0.004019	0.001651	0.004435	0.000123	0.001792	0.004963	0.001334
50% Group 1: 50% Group 4	0.004035	0.001439	0.003363	0.000102	0.001395	0.003918	0.001042
90% Group 2: 10% Group 4	0.004463	0.000923	0.004305	0.000131	0.000707	0.005353	0.000927
10% Group 2: 90% Group 4	0.004047	0.001854	0.005849	0.000151	0.002201	0.006400	0.001677
80% Group 2: 20% Group 4	0.004411	0.001039	0.004498	0.000133	0.000893	0.005484	0.001021
20% Group 2: 80% Group 4	0.004099	0.001738	0.005656	0.000149	0.002014	0.006269	0.001584
70% Group 2: 30% Group 4	0.004359	0.001156	0.004691	0.000136	0.001080	0.005615	0.001115
30% Group 2: 70% Group 4	0.004151	0.001621	0.005463	0.000146	0.001827	0.006138	0.001490
50% Group 2: 50% Group 4	0.004255	0.001388	0.005077	0.000141	0.001454	0.005876	0.001302
90% Group 1: 10% Group 3	0.003918	0.000857	0.000685	0.000051	0.000412	0.001314	0.000287
10% Group 1: 90% Group 3	0.002666	0.000454	0.000685	0.000051	0.000485	0.001384	0.000087
80% Group 1: 20% Group 3	0.003761	0.000807	0.000685	0.000051	0.000421	0.001323	0.000262
20% Group 1: 80% Group 3	0.002823	0.000504	0.000685	0.000051	0.000476	0.001375	0.000113
70% Group 1: 30% Group 3	0.003605	0.000756	0.000685	0.000051	0.000430	0.001331	0.000237
30% Group 1: 70% Group 3	0.002979	0.000555	0.000685	0.000051	0.000467	0.001366	0.000137
50% Group 1: 50% Group 3	0.003292	0.000655	0.000685	0.000051	0.000448	0.001349	0.000187

 Table 5 | Modeled major ion concentrations between the four principal water groups (expressed in molality)

In order to distinguish between samples whose chemical composition is primarily changed by freshwater dilution, rock weathering or evaporation/precipitation, Gibbs (1970) devised a presentation. This is based on the relationship between TDS and the cationic and anionic sides' respective ratios of  $(Na/(Na + K) \text{ or } (Cl/(Cl + HCO_3))$ . The distribution of the four main water groups under study on Gibbs diagrams (Figure 8) illustrates how evaporation and rock/water interaction mechanisms dominate the salt composition.

The first group of water, group 1, has salinity ranging from 422 to 810 mg/l with an average value of 605 mg/l with abundance orders (meq/l) of Na<sup>+</sup> > Mg<sup>2+</sup> > Ca<sup>2+</sup> > K<sup>+</sup> and HCO<sub>3</sub><sup>-</sup> > Cl<sup>-</sup> > SO<sub>4</sub><sup>2-</sup> > CO<sub>3</sub><sup>2-</sup>. This water group is classified as HCO<sub>3</sub><sup>-</sup> alkaline earth water type. This is probably derived from carbonate precipitation. For group 2, the salinity of water samples varied from 773 to 1,051 mg/l with an average value of 898 mg/l, while for group 3, the salinity ranged from 214 to 338 mg/l with an average value of 284 mg/l (freshwater). For group 4 water samples, the cationic composition was dominated by Na<sup>+</sup> and Mg<sup>2+</sup> followed by Ca<sup>2+</sup> and K<sup>+</sup> and Cl<sup>-</sup> > HCO<sub>3</sub><sup>-</sup> > SO<sub>4</sub><sup>2-</sup> > CO<sub>3</sub><sup>2-</sup> and high salinity ranged from 1,088 to 1,401 mg/l with an average value of 1,224 mg/l. Applying mixing process simulation between the four principal water groups using PHREEQC software is to indicate precisely the routes of mixing sources within the quaternary aquifer in El Sharqia Governorate. The geochemical modeling results shown in Table 5 illustrate that simulation of a double-mixing process (20% Group 1):80% Group 4) leads to calcium contents similar to those of samples 24 and 36. Also, mixing of 10% Group

Table 6 | Non- carcinogenic risks to humans from drainage and groundwater samples.

	Groundwater					
	Children			Adult		
Sample	Min	Мах	Average	Min	Max	Average
Al	$9.753\times10^{-1}$	$7.571\times 10^1$	$1.497 \times 10^1$	$7.663\times10^{-1}$	$2.475\times 10^1$	7.921
Cd	$5.462\times 10^{-5}$	$2.060\times 10^{-3}$	$4.156\times10^{-4}$	$4.400\times10^{-5}$	$1.659\times 10^{-3}$	$\textbf{3.348}\times \textbf{10}^{-4}$
Co	$1.951\times 10^{-3}$	$8.017\times 10^{-2}$	$2.002\times10^{-2}$	$1.571\times 10^{-3}$	$6.459\times10^{-2}$	$1.613\times 10^{-2}$
Cr	$1.300\times10^{-1}$	$6.619\times10^{-1}$	$2.438\times10^{-1}$	$1.048\times10^{-1}$	$5.332\times10^{-1}$	$1.964\times10^{-1}$
Fe	$1.115\times10^{-3}$	$9.820\times10^{-2}$	$\textbf{2.818}\times \textbf{10}^{-2}$	$8.980\times10^{-4}$	$7.911\times10^{-2}$	$2.270\times 10^{-2}$
Ni	$\textbf{3.901} \times \textbf{10}^{-4}$	$1.092\times10^{-2}$	$2.489\times 10^{-3}$	$\textbf{3.143}\times \textbf{10}^{-4}$	$8.800\times 10^{-3}$	$2.005\times 10^{-3}$
Pb	0.000	1.730	$7.750\times10^{-1}$	$6.984\times10^{-2}$	1.393	$6.762\times 10^{-1}$
Sr	$7.179\times10^{-3}$	$1.137\times10^{-1}$	$4.617\times10^{-2}$	$5.783\times10^{-3}$	$9.156\times10^{-2}$	$\textbf{3.720}\times 10^{-2}$
Zn	$1.040\times10^{-4}$	$7.406\times10^{-2}$	$1.619\times10^{-2}$	$8.381\times 10^{-5}$	$5.966\times 10^{-2}$	$1.304\times 10^{-2}$
Cu	$8.541\times 10^{-3}$	$2.278\times10^{-1}$	$8.844\times 10^{-2}$	$6.880\times10^{-3}$	$1.835\times10^{-1}$	$7.124\times10^{-2}$
Ba	$3.609\times 10^{-3}$	$5.577\times10^{-2}$	$2.713\times10^{-2}$	$2.907\times 10^{-3}$	$4.493\times10^{-2}$	$2.186\times 10^{-2}$
	Drainage water					
Sample	Min	Мах	Average	Min	Мах	Average
Al	0.000	$5.562\times 10^2$	$4.771 \times 10^1$	$7.857\times10^{-1}$	$9.366\times 10^1$	$1.856\times 10^1$
Cd	$5.462\times10^{-5}$	$3.121\times 10^{-3}$	$5.319\times10^{-4}$	$4.400\times10^{-5}$	$2.514\times10^{-3}$	$4.364\times10^{-4}$
Co	$1.951\times 10^{-3}$	$6.944\times 10^{-2}$	$2.455\times10^{-2}$	$1.571\times 10^{-3}$	$5.594\times10^{-2}$	$1.977\times 10^{-2}$
Cr	$1.300\times 10^{-1}$	$5.553\times10^{-1}$	$1.939\times10^{-1}$	$1.048\times10^{-1}$	$4.473\times10^{-1}$	$1.562\times 10^{-1}$
Fe	$1.115\times 10^{-3}$	$3.133\times10^{-1}$	$5.312\times10^{-2}$	$8.980\times 10^{-4}$	$2.524\times10^{-1}$	$4.279\times 10^{-2}$
Ni	$3.901\times 10^{-4}$	$1.145\times10^{-2}$	$1.988\times 10^{-3}$	$\textbf{3.143}\times \textbf{10}^{-4}$	$9.224\times 10^{-3}$	$1.602\times 10^{-3}$
Pb	$8.670\times 10^{-2}$	2.886	$5.922\times10^{-1}$	$6.984\times10^{-2}$	2.325	$4.771\times10^{-1}$
Sr	$1.476\times10^{-3}$	$5.442\times10^{-1}$	$7.649\times10^{-2}$	$1.189\times10^{-3}$	$4.384\times10^{-1}$	$6.162\times 10^{-2}$
Zn	$\textbf{-7.022}\times10^{-4}$	$6.812\times10^{-2}$	$1.022\times 10^{-2}$	$\textbf{-5.657}\times10^{-4}$	$5.487\times10^{-2}$	$8.232\times 10^{-3}$
Cu	$6.327\times 10^{-3}$	$2.127\times10^{-1}$	$6.644\times 10^{-2}$	$5.097\times 10^{-3}$	$1.713\times10^{-1}$	$5.352\times 10^{-2}$
Ba	$6.242\times 10^{-4}$	$3.226\times 10^{-2}$	$1.434\times10^{-2}$	$5.029\times10^{-4}$	$2.599\times10^{-2}$	$1.155\times 10^{-2}$

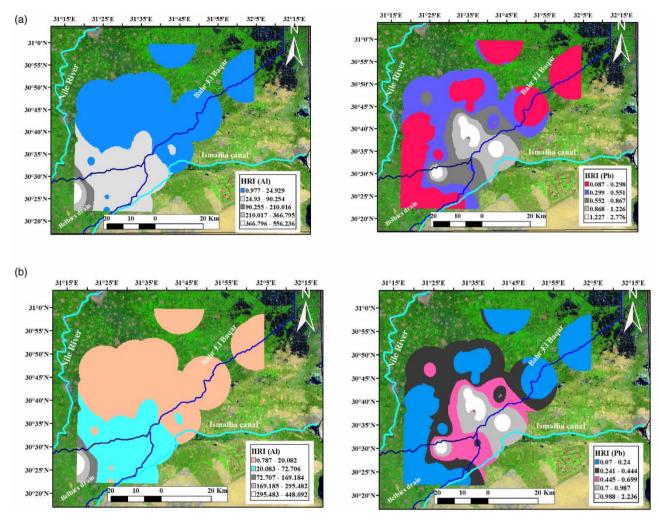


Figure 9 | (a) Spatial distribution of children HRI for Al and Pb of drainage water samples and (b) spatial distribution of adult Health Risk Index (HRI) for Al and Pb of drainage water samples.

1:90% Group 2 leads to chloride contents similar to those of samples 25, 35, 42, 46 and 49. Mixing of 90% Group 1:10% Group 2 gives sulphate contents similar to those of samples 28 and 53.

#### 6. POTENTIAL HUMAN HEALTH RISK ASSESSMENT

# 6.1. Non-carcinogenic analysis

Results of trace elements analysis are illustrated in Table 1, where trace elements in groundwater samples were ranked as follows: Fe > Sr > Al > Zn > Ba > Cu > Pb > Ni > Cr > Co > Cd and for drainage water samples were ranked as follows: Sr > Al > Fe > Zn > Pb > Cu > Ba > Ni > Cr > Cd > Co according to their concentrations in mg/l. Examining HRI values for different age groups of people including children and adults in the study area are shown in Table 6 with spatial distribution of HRI values for both drainage and groundwater samples in Figures 9 and 10.

Results of the HRI of collected water samples reveal that all samples have HRI < 1 for all trace elements except Al and Pb which have HRI > 1 for both children and adults in drainage and groundwater samples. Spatial distribution of HRI values in Figures 9 and 10 of drainage and groundwater samples revealed that drainage water has higher HRI values than groundwater samples, especially near the drains with higher values for children than adults confirming that children are more sensitive to the adverse health effects of metals that have non-carcinogenic risks (USEPA 2012), because children are most likely to have oral intake by hand and mouth (Kusin *et al.* 2018).

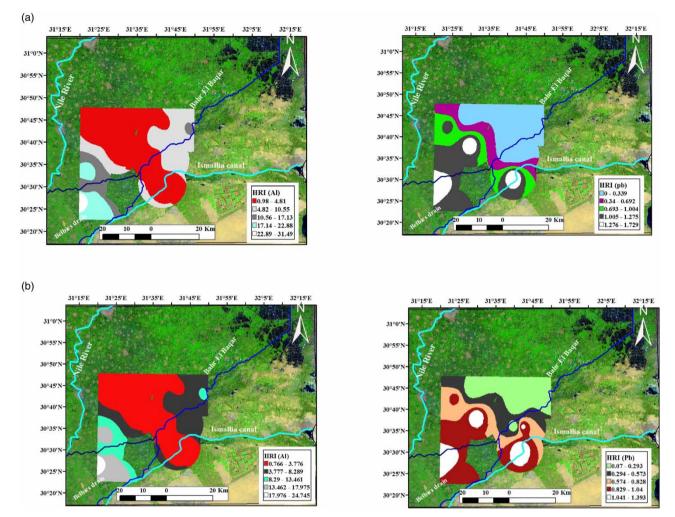


Figure 10 | (a) Spatial distribution of children HRI for Al and Pb of groundwater samples and (b) spatial distribution of adult Health Risk Index (HRI) for Al and Pb of groundwater samples.

# 6.2. Carcinogenic risk analysis

Heavy metals such as Cr, Ni and Pb can potentially enhance the risk of cancer in humans (Tani & Barrington 2005). Longterm exposure to low amounts of toxic elements could result in many types of cancer diseases. Using Cr, Ni, and Pb as carcinogens, the total exposure of the residents was calculated for groundwater samples used for drinking purposes as shown in Table 7.

For one heavy metal, an ILCR less than  $1 \times 10^{-6}$  is considered insignificant and the cancer risk can be neglected; while an ILCR above  $1 \times 10^{-4}$  is considered harmful and the cancer risk is troublesome. Results of Table 7 indicate that chromium

 Table 7 | The incremental lifetime cancer risk (ILCR) values of carcinogenic human health risks via total exposure (ingestion and dermal contact) to the drinking water of the study area for adults

	ILCR					
	Cr	Ni	Pb			
Min	$1.675\times 10^{-6}$	$6.865\times10^{-10}$	$1.389\times 10^{-7}$			
Max	$8.527\times 10^{-6}$	$1.922\times 10^{-8}$	$2.772\times 10^{-6}$			
Average	$3.141\times 10^{-6}$	$4.380\times10^{-9}$	$1.345\times 10^{-6}$			

and lead may have a chance of cancer risk, while nickel has the lowest chance for cancer risk from the contaminants to resident's people.

# 7. CONCLUSION

This study was accomplished to appraise the main factors controlling water resources evolution/pollution indicators emphasizing direct/indirect human health risks in El Sharqia Governorate, Egypt. Most of the collected groundwater samples were shallow with depths ranging from 8 to 13 m, which implies that these wells are more vulnerable to contamination. Salinity classifications show that about 81% of groundwater samples and 89% of drainage water were freshwater, while about 19 and 11% were classified as slightly saline, respectively. Groundwater quality in the study area was controlled by natural processes (involving dissolution/precipitation of minerals, cation exchange, and evaporation) or anthropogenic factors (including leaching of solid waste, overuse of agricultural fertilizers, high loads of discharged sewage water) responsible for water quality deterioration. It was found that ammonia, nitrate, BOD, phosphate, turbidity, iron, manganese, lead, and aluminum values exceeded the limit of drinking water international standards. A human health risk was identified in the case of Pb and Al with high HRI values for different age groups of people including children and adults exceeding unity. It seems that the aquifer in the study area is quite vulnerable to pollution.

# 8. RECOMMENDATIONS

An advanced sanitary drainage network must be designed; chemical and bacteriological analyses must be carried out periodically for surface and groundwater to ensure the suitability of water for different purposes, taking into consideration the different hydrological and soil parameters that affect the susceptibility of the aquifer to pollution.

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# **AUTHOR CONTRIBUTIONS**

All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by A.H.M.E.-A., R.A.H., F.A.M., S.O., and E.O. The first draft of the manuscript was written, revised, and previous versions of the manuscript were commented. All authors read and approved the final manuscript.'

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#### DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

# **CONFLICT OF INTEREST**

The authors declare there is no conflict.

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