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Identification of the Potential Sources of Nitrate in Groundwater from Shallow Aquifer in Wadi Qudaid, Western Part of Saudi Arabia

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Abstract. An investigation to identify the potential contamination of groundwater from on-site domestic wastewater systems blasted in the alluvial deposits in Wadi Qudaid basin, west of Saudi Arabia, was conducted during April, 2010. Groundwater samples were collected at a variety of depths and distances along the groundwater flow path from the cesspool. Impacts of on-site systems on the shallow aquifer are evidenced by the elevated concentrations of NO_3^- and Cl. The chemical analyses results of the groundwater samples collected from private domestic wells in a residential site, show that the NO_3^- concentration in the groundwater exceeded the maximum contaminant level (MCL) of 45 mg/l. It ranges from 43 to 193 mg/l with an average of about 77.0 mg/l which is greater than the average background (19.0 mg/l) of the NO_3^- concentration in the undeveloped region within the wadi basin itself. The high NO_3^- content is a widespread pollutant that possesses a serious threat to the public health. Nitrate contamination is generally observed in close proximity to potential point waste sources. The dominant groundwater movement in the area is the major factor enhancing the groundwater deterioration by nitrate that leached from on-site wastewater disposal systems. Chloride-Nitrate and (Sodium + Nitrate) - Chloride and other relationships have been used to differentiate the potential sources of Cl^-. None of Faecal coliforms were detected in the groundwater samples even in samples with the highest NO_3^- concentrations, implying that the depth to the groundwater body was sufficient for bacterial die-off.

Keywords. Shallow aquifer, groundwater contamination, nitrate concentration

Introduction

The shallow aquifer in Wadi Qudaid basin in the western province of Saudi Arabia is considered the principal source of water supplies to the towns and villages. During the past two decades a rapid growth of population and development in these areas has led to concern about potential water quality impacts, because of the absence of municipal sewer services.

Disposal of sewage in the residential areas have been accomplished almost exclusively through the use of conventional on-site sewage systems. A traditional on-site sewage system consists of a cesspool and a subsurface absorption system. The cesspool is a shallow pit with different lengths and widths, and average depth of about 3 meters. It is poorly designed and often built by bricks. Under ideal conditions, the effluent is assimilated and treated within the top soil immediately below and adjacent to the cesspool. No regulations are implemented for setback distance and lot sizes requirements and its design and/or installation, to ensure that the vertical separation between the bottom of the cesspool and the water table is large enough so that unsaturated conditions will be maintained even during wet seasons.

Several investigations in the literature discussed the fate and movement of chemical constituents of septic/cesspool effluent into shallow groundwater (*e.g.* Bleifuss *et al.,* 2005; Yates, 1985; Scandura and Sobsey 1997; Whitehead and Geary. 2000; Geary, and Whitehead., (2001); and Xuan Xu, 2007). Generally, most concern appears to be related to NO_3^- and bacterial contamination of aquifers because of possible health problems from NO_3^-. Elevated concentrations of nitrate in drinking water are a cause for concern. Recently, the investigations carried out in similar condition of Wadi Qudaid (Alyamani, 2007; Saudi Geological Survey, 2004; and Sharaf, *et al*., 2004) shown that many of the domestic water supply wells extract waters with high NO_3^- concentration (100 mg/l) which is greater than the acceptable maximum contamination level (MCL) of 45 mg/l (10 mg/l of nitrate-nitrogen)

(EPA, 2004), while the WHO recommended maximum limit for nitrate concentration in drinking waters is 50 mg/l NO^{-}_{3} , equivalent to 11.3 mg /l as NO^{-}_{3} -N , (WHO, 2000). However, in the previous studies carried out in the country, the high NO_3^- contents of the groundwater were commonly linked to the nitrogen-based fertilizers used, with no particular attention given to the possible groundwater pollution by on-site sewage disposal systems used in these regions.

In the present study, Wadi Qudaid basin, where residential areas lie on shallow aquifer, was selected to identify any impacts arising from on-site wastewater disposal systems used in relation to groundwater quality.

Description of the Study Area

During the last 20 years, the residential areas have grown rapidly (Fig. 1). The wadi lies entirely within basement complex of the Arabian Shield. Geologically, three stratigraphic units were recognized by Ramsay (1986): These are: Quaternary deposits, Tertiary rocks and Precambrian layered rocks. The Quaternary deposits almost found along the main wadi course. These deposits cover extensive areas of the region particularly the down stream part. These sediments range in size from coarse sand to pebble and gravel that serve as a permeable conduit for the percolation of surface water into the aquifers. The Tertiary basalt flows which consist of gray, medium to coarse grained alkali olivine basalt. The Precambrian layered rocks are predominantly lavas and associated volcaniclastic rocks, ranging from basalts to rhyolites, slightly metamorphosed. The main wadi channel has a gentle slope towards the southwest direction. Clay and silt have also been observed in different places of the main course.

Over the study area, the rainfall is irregular and has torrential nature. The annual average rainfall is about 110 mm. The rainfall season is from December to May. December and January receive nearly about 70% of the annual rainfall. During the rest of the year, rainfall is limited to isolated events. The general recharge source of the aquifer has been commonly attributed to infrequent runoff events, which infiltrate to the water table promptly by a process much enhanced through the zone of aeration. The water table is shallow and varies between 10 to 15 m from the wadi floor. The wadi channel is topographically lower than the surrounding areas by nearly 1.5 m. Most of the existing wells have been dug into the bed rock and almost entirely penetrate the weathered and fractured zones. The major groundwater is pumped from the surficial deposits overlying the bed rock. Most of the drilled wells are placed along and the side of the main wadi channel. The most common type of well construction is the large diameter well, with an average diameter of 1.5 meters. The thickness of the fractured and weathered zones varies from 2 to 4 m. The groundwater is under unconfined condition and the groundwater flow is from northeast to southwest (Fig.1).

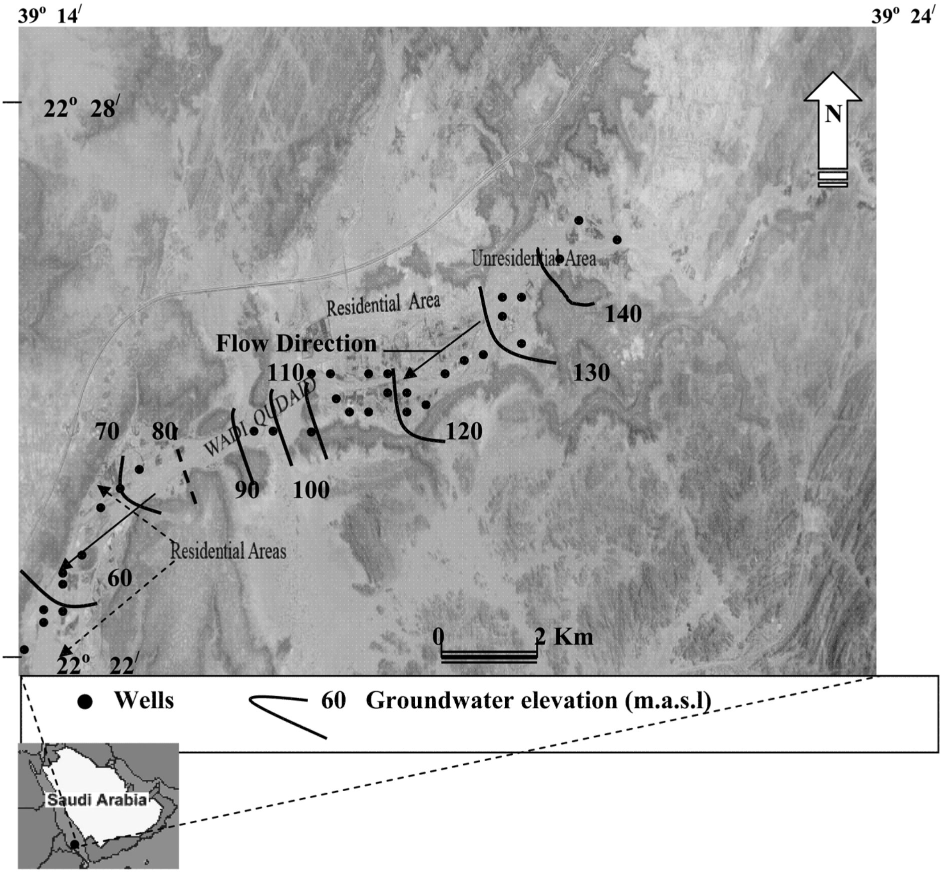


Fig. 1. Location map of the study area.

Methodology and Sampling Collection

During April, 2010, a total of 34 groundwater samples were collected from private domestic water supply wells in the wadi (Fig.1). For comparison purpose, the background of groundwater chemistry was determined in 7 samples (Table 1) that were collected from wells in the uppermost part of the wadi, which least likely to be impacted by human activity (Fig.1). Groundwater temperature and pH were measured in situ. All the groundwater samples were analyzed for some major ions including calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^{+}), sulfate (SO_4^{2-}), bicarbonate (HCO_3^{-}), chloride (Cl^{-}), inorganic nutrients involved (nitrate (NO_3^{-}), nitrite (NO_2^{-}), ammonia (NH_4^{+}) as nitrogen and phosphorus PO_4^{3-}), and trace elements including lead (Pb), Cadmium (Cd), manganese (Mn), zinc (Zn), and iron (Fe). Each well was pumped for at least 5 minutes prior to sampling. These analyses were carried out in Saudi Geological Survey (SGS) Laboratories,. These elements were analyzed using the standard methods (APH/AWWA/WPCF, 1989) and ICP-Optical Emission Spectrometer, Optima 2000 DV. In the laboratory, the groundwater samples were biologically analyzed for total coliform, fecal coliform and fecal streptococci, dissolved oxygen (DO) and biological oxygen demand (BOD). The results of these chemical analyses are given in Table 1.

Results and Discussion

Groundwater Chemistry

The chemical composition results of the groundwater in the study area were summarized in (Table 1). It commonly shows that the groundwater mineralization in the residential area is generally elevated from that observed in the background in unresidential area, with nitrate and chloride concentrations being significantly elevated. The Cl^{-} concentrations in the residential area range from 413 to 1218 mg/l, with an average of about 765 mg/ l, which is twice the average reference of undeveloped area (346 mg/ l). The NO_3^{-} concentrations in residential area increased from 43 to 193 mg/l with an average of about 77 mg/l. which is 3 times higher than the average background concentrations (24.7 mg/l). The concentration of SO_4^{2-} in water varies from 235 to 805 mg/l with an average of about 714 mg/ l which is higher than that observed in unresidential area. The high concentrations of $SO_4{}^{2-}$ are probably due to the oxidation of pyrite (FeS_2), which is a very common accessory mineral in the country rocks (Moore and Al-Rehili, 1989) and/or the secondary mineral of gypsum in the form of evaporate salts as indicated by XRD analysis. The other major constituents such as Na^{+}, Ca^{2+} and Mg^{2+} and HCO_3^{-} are considerably above the background concentrations.

Table 1. The chemical analyses results of the groundwater samples.

LOCATION	STATISTIC	NO_2 (mg/l)	PO_4 (mg/l)	NH_4 (mg/l)	Fe (ppb)	Zn (ppb)	Mn (ppb)	Pb (ppb)	Cd (ppb)	Al (ppb)	DO (mg/l)	BOD
UNRESIDENTIAL AREA (= 7 samples)	MAX		1.73			485.0	3.61	0.32	0.19	20.8	0.72	0.17
	MIN	<0.03	0.13	<0.04	<0.10	0.21	0.11	0.09	0.06	11.4	0.19	0.0
	MEAN		0.05			75.0	0.54	0.14	0.12	13.5	0.36	0.08
RESIDENTIAL AREA (= 27 samples)	MAX					385.0	4.61	0.45	0.34	23.8	3.23	0.64
	MIN	<0.03	<0.09	<0.04	<0.10	0.29	0.12	0.10	0.10	12.4	0.45	0.0
	MEAN					99.0	0.68	0.21	0.15	16.5	1.62	0.23

LOCATION	STATISTIC	Ca (mg/l)	Mg (mg/l)	Na (mg/l)	K (mg/l)	Cl (mg/l)	HCO_3 (mg/l)	NO_3 (mg/l)	SO_4 (mg/l)	PH	EC (us/cm)
UNRESIDENTIAL AREA (= 7 samples)	MAX	204	206	322	7.2	393	282	30	280	7.93	2720
	MIN	73.1	37	240	0.9	295	126	19	265	7.21	1270
	MEAN	208	137	413	3.5	346	207	24.7	514	7.55	1090
RESIDENTIAL AREA (= 27 samples)	MAX	341	301	1108	15.5	1218	266	193	805	7.91	4902
	MIN	142	70	245	0.3	413	163	43	235	7.04	1765
	MEAN	256	157	562	5.1	765	210	77	714	7.46	2960

Concentrations of NH4+ and NO2 are rather low and their values are almost less than 0.04 and 0.03 mg/l. The maximum concentration of PO_4^+ is 1.73 mg/l, while the average is 0.05 mg/l. In the residential area, the Pb concentrations range between 0.10 and 0.45 ppb, with an average of 0.21 ppb. The groundwater was oxygenated where the dissolved oxygen (DO) in residential area ranged between 0.45 and 3.23 mg/l, with an average of about 1.62 mg/l, whereas, the average (DO) in unresidential area is about 0.36 mg/l, which is lower from that observed in the developed area. The biological oxygen demands (BOD) in residential area vary from zero to 0.64 mg/l and average of about 0.23 mg/l. Most pristine waters will have a 5-day, BOD below 1 mg/l. moderately polluted may have a BOD value in the range of 2 to 8 mg/l. (Clair *et al*., 2003).

Coliform bacteria were not detected, even in samples with the highest NO_3^- concentrations, suggesting that substantial residence time of these waters in fractured and weathered rocks was sufficient for bacterial die-off (Geary and Whitehead, 2001).

Groundwater Movement

The general flow pattern of the groundwater movement is from northeast to southwest direction. Four major ions, namely; Ca, Na, Cl, and SO_4 their concentrations are increased towards the downstream part and to some extent follow the direction of the groundwater flow direction Fig. 1, whereas NO_3, Mg, HCO_3 and K contents were randomly distributed and have not reflected the general flow pattern of the groundwater. Such situation when occurs may indicate that local processes influence their concentrations.

Sources of Nitrate (NO_3) in the Groundwater

Generally, the major sources of nitrate include fertilizer, animal manure, sewage and atmospheric deposition. In addition, effluent from cesspools is enriched with ammonium ion (NH_4^+). Ammonium ion can be preferably absorbed onto sediments surrounding a cesspool, and under oxidizing conditions ammonium ion (NH_4^+) is converted to nitrate ion (NO_3^-) according to following equations:

$$NH_4 + + 1.5\ O_2 \leftrightarrow NO_2 - + H_2O + 2\ H^+ \quad (1)$$

$$NO_2 - + 0.5\ O_2 \leftrightarrow NO_3 \quad (2)$$

$NO_3^{\cdot}$ is quite soluble and completely mobile when it is dissolved. Within the study area, most common sources of nitrate are natural and artificial fertilizers as well as the subsurface disposal of human waste. The natural fertilizer consists of animal waste, whereas the artificial fertilizer depends on nitrogen-based fertilizers.

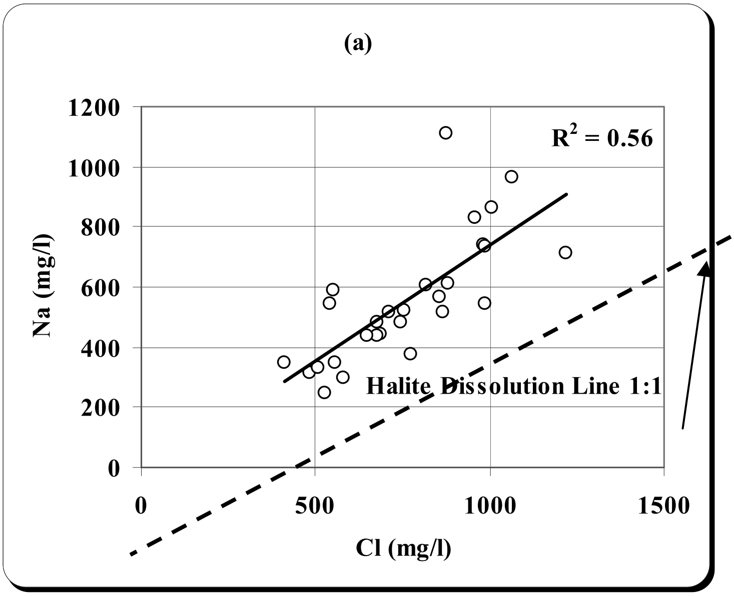
However, within the Wadi Qudaid basin these two major sources of NO_3 might be expected. These are agricultural activities and wastewater that infiltrate from on-site systems in the residential areas, in which chemical and natural fertilizers are not used intensively, because in many cultivation areas that are scattered along the main course of the wadi are merely palm-tree farms. Therefore, the wastewater that infiltrates from on-site systems in the residential areas to groundwater is the only source of high NO_3 concentrations. This conclusion might be substantial since the high NO_3 is only observed in the residential areas, Table 1.

Generally, in an unsewered residential area and shallow groundwater, both NO_3^- and Cl are the most significant contaminants associated with domestic wastewater (EPA, 2004). Chloride (Cl^-) is a good indicator parameter of sewage impacts because it is not subjected to adsorption, ion exchange, or oxidation-reduction "redox" reactions. To differentiate the potential source(s) of NO_3^- and Cl^- in the groundwater of the study area; the plots of Na^+ *vs* Cl^-, NO_3^- *vs* Cl , $NO_3^- + Na^+$ *vs* Cl^- and dissolved oxygen (DO) *vs* NO_3^- are shown in Fig. 2 and 3.

The relationship between Na and Cl ions was given (Fig. 2a), which might be practically utilized to identify their concentrations due to evaporation processes (Eugster and Jones, 1979). It shows a low correlation with ($R^2 = 0.56$). The data points lie very close to the halite dissolution line (1:1), indicating that halite is probably the sole source of these two ions which resulted from the evaporation processes. However, halite is not the only potential source for Cl^- ion. The observed deviation of the data points from the dissolution line may have resulted from an excess of Cl^- ions that entered the solution. It is more likely that the Cl ions entered the groundwater by wastewater infiltrated from on-site systems used in the area. On the other hand, the relationship between NO_3^- and Cl^- is shown on Fig. 2b It indicates that Cl almost increases linearly with increasing NO_3^- with correlation coefficient (R^2) of about 0.40. Fig. 3a shows that neither Na^+ nor NO_3^- is by themselves sufficient to account for the Cl^- in the groundwater. But together these two elements do balance to some extent (R^2=0.70) the concentration trend of Cl^-. The contribution of NH_4^+ to the overall nitrate concentrations in the groundwater can be depicted from the positive relationship between NO_3^- and dissolved oxygen (DO) (Fig. 3b). It demonstrates that the NH_4^+ concentrations in the groundwater may be lost due to microbial conversion to nitrate (nitrification process) (Eqs.1 and 2). Two significant factors may account for the movement of contaminants from on-site wastewater systems to the groundwater. Firstly, the local groundwater flow system almost takes place throughout areas, (Fig.1) where the domestic wells are closed to surrounded by areas where cesspools are present; secondly, the wadi channel is topographically lower than the surrounding areas by nearly 1.5 m.

Figure 4 represents a conceptual pollutant transport model in the groundwater aquifer from the cesspool areas to the groundwater storage in the study area. The impact of cesspool on the groundwater contamination can be traced from the maximum values of NO_3^- (193 mg/l) and Cl^- (1218 mg/l) was recorded in a well close to the lager residential area. On the other hand, under the prevailing groundwater flow direction, pollution of the groundwater by NO_3^- continues further downstream. Therefore great concern should be paid to contaminants flow to other areas particularly along downgrdient flow (Fig.1).

Questions must be posed as to the adequacy under the new (EPA, 2000) regulations of the minimum thickness requirements (between 0.9-1.2m) of soil to prevent bacteria and viruses from entering groundwater. It must be kept in mind that the EPA manual requires 1.2 m of unsaturated subsoil beneath the invert of the percolation trench. In many parts of the area studied, the water table is within.10 to 15 m below the surface particularly in the wet weather, where the thickness of unsaturated zone is small. This figure therefore must be taken into consideration with many other factors mentioned above and not used on its own.



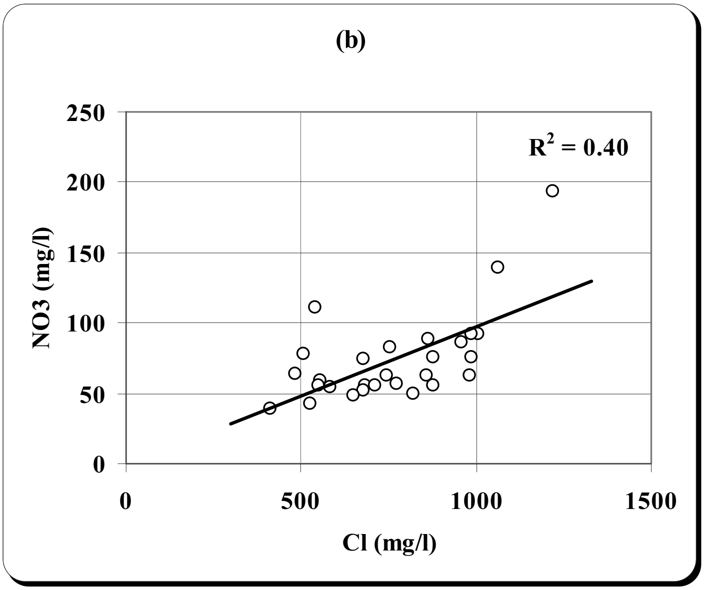
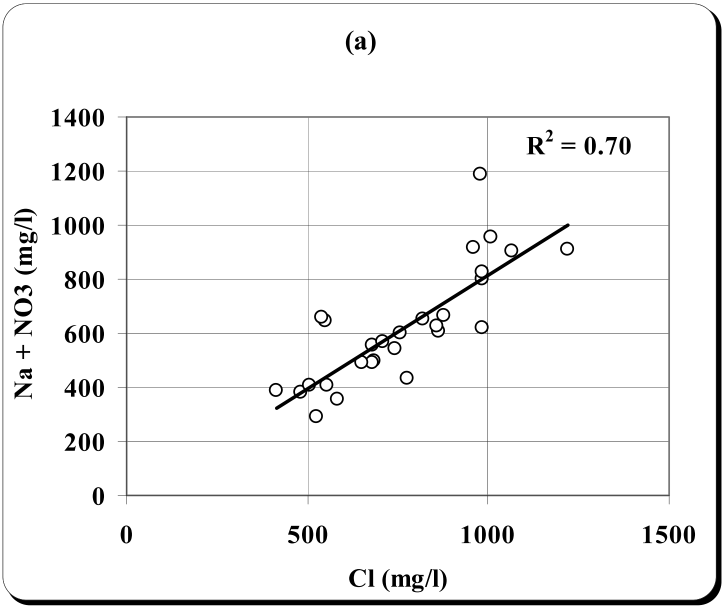


Fig. 2. Correlation diagrams showing the relationship between (a) Na *vs* Cl. and (b) NO_3 *vs* Cl.



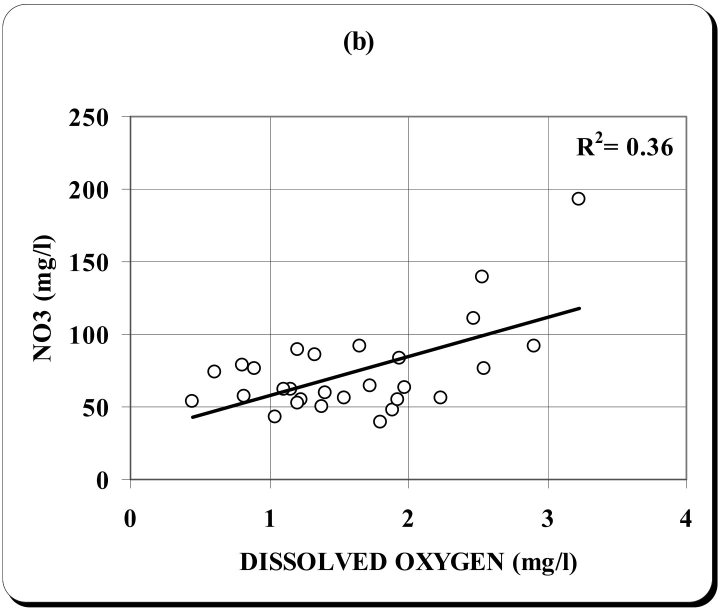


Fig. 3. Correlation diagrams showing the relationship between (a) Na+NO_3 *vs* Cl; and (b) NO_3 *vs* dissolved oxygen (DO).

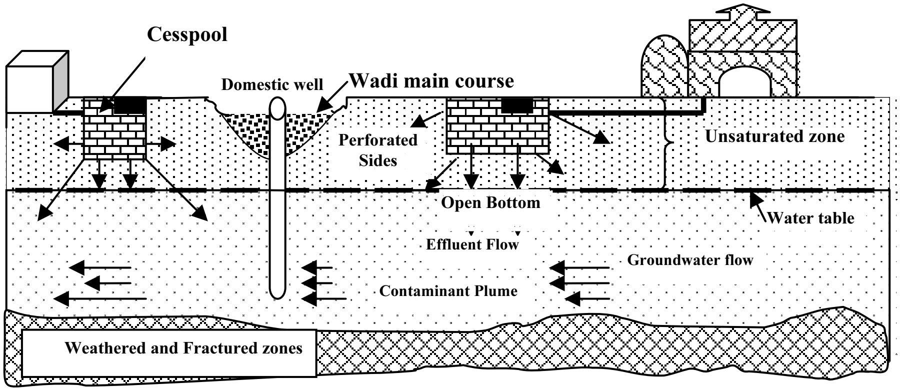


Fig. 4. Conceptual model of contaminant flow from the cesspool to the groundwater body in the study area (not scaled).

It is beyond the objective of this paper to present a comprehensive assessment of cumulative and comparative risk for the study area. The average concentration of NO_3^- is about 77.0 mg/l. This is almost twice above the risk value of the drinking water level (45 mg/l; as 10 mg/l of nitrate-nitrogen). Increasing NO_3^- concentrations will continue to increase as long as on-site systems contribute significantly to the risk value. Therefore, regulations for cesspool systems that are used in the area should be implemented.

Conclusion

In the investigated area, open dug wells are generally not considered as safe sources for drinking water, because they are very vulnerable to contamination. The widespread increases in nitrate concentrations in groundwater have been caused by an increase in the input of nitrogen into the environment as a result of human activities. The groundwater samples of the tested dug wells show contamination by nitrate. Nitrate represents the chemical of greatest concern in the groundwater under unsewered developments. In the residential area, the nitrate concentration in the groundwater ranges between 43 and 193 mg/l with an average of about 77.0 mg/l that exceeds the drinking water standard as well as the background concentration in unresidential area. Impacts from on-site systems on the shallow aquifer are highly witnessed as evidenced by the elevated concentrations of NO_3^- and Cl. Coliform bacteria have not been detected in the groundwater samples, which may be due to the substantial residence time of these waters in sediments which causes bacteria to die-off. Lot-size and setback distances of the cesspool are critical factor in determining the amount of natural attenuation that occurs between the location where cesspool effluents enter the aquifer, and the nearest down-gradient point of groundwater withdrawal.

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التعرف على المصدر (المصادر) المحتملة للنترات في المياه الجوفية للخزان المائي الضحل بوادي قديد، الجزء الغربي من المملكة العربية السعودية

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المستخلص. ركزت الدراسة الحالية على إمكانية التعرف على تلوث المياه الجوفية المحتمل في موقع تستخدم فيه أنظمة الصرف الصحي المنزلي التي أنشئت في الرواسب الوديانية بحوض وادي قديد، غرب المملكة العربية السعودية. خلال شهر نيسان ٢٠١٠ تم جمع عينات من المياه الجوفية في مجموعة متنوعة من أعماق ومسافات على طول مسار تدفق المياه الجوفية من أنظمة الصرف الصحي المنزلي. ويستدل تأثير أنظمة الصرف على المياه الجوفية الضحلة من ارتفاع تركيزعنصري النترات والكلورايد Cl, NO_3^- بالمياه. نتائج التحليل الكيميائي لعينات المياه الجوفية التي تم جمعها من الآبار الخاصة بالقرب من المناطق السكنية تبين أن تركيز NO_3^- في المياه الجوفية تجاوزت الحد الأقصى المسموح لمستوى الملوثات؛ ٤٥ ملجم/لتر. وهي تتراوح ٤٣–١٩٣ ملجم/لتر بمتوسط قدره حوالي ٧٧,٠ ملجم/لتر، والذي يزيد عن متوسط النترات (١٩,٠ ملجم/لتر) في المنطقة غير المأهولة بالسكان داخل حوض الوادي نفسه. ارتفاع تركيز محتوى NO_3^- منتشر على نطاق واسع مسببا تهديدا خطيرا

للصحة العامة. ويلاحظ عموما التلوث بالنترات على مقربة من مصادر الصرف الصحي. حركة المياه الجوفية السائدة في المنطقة هي العامل الرئيسي في تعزيز تدهور المياه الجوفية بارتفاع تركيز النترات التي تتسرب في المناطق التي توجد بها أنظمة التخلص من مياه الصرف الصحي. استخدمت علاقات كلوريد/نترات والصوديوم + النترات/الكلوريد وغيرها من العلاقات للتفريق بين المصادر المحتملة للكلور. لم يتم الكشف عن وجود البكتيريا البرازية في عينات من المياه الجوفية حتى في العينات مع تركيزات أعلى لـ NO_3^-، مما يعني ضمنا أن عمق المياه الجوفية كان كافيا لموتها.