Determination of Nitrate and Nitrite levels in Soil and Groundwater in Al Hassa Area, Saudi Arabia

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Abstract

The levels of nitrate (NO_3^-) and nitrite (NO_2^-) contents in soil and groundwater in Al Hassa area, Kingdom of Saudi Arabia were determined. Water samples were collected from shallow groundwater at about 3 meters, and deep groundwater of three aquifers at different depths: Neogene (25-150 m), Dammam (260 m) and Umm er Radhuma (450-480 m). Values of NO_3^- were higher than NO_2^- in both water and soil samples. Nitrate values were 5.9 and 273.4 ppm in both deep and shallow groundwater, respectively. Significant differences in NO_3^- contents were found between shallow groundwater and aquifer water samples and also between the three aquifers. However, water samples of soil which received organic farmyard manure fertilized (OM) had an NO_3^- content of (61.8 ppm) which is significantly less than that which received OM+ urea (104.3 ppm). Four types of nitrogen fertilized soil were employed, (virgin soil (V), organic farmyard manure (OM), urea (U) and organic farmyard manure + urea (OM+U)). Soil samples were collected from three depths (0-25, 25-50 and 50-75 cm). Data showed that, the concentrations of NO_3^- have increased in the following order: V<OM<U<OM+U. The mean values were 3.93, 21.9, 26.6 and 45.2 ppm for V, OM, U and OM+U soils, respectively. The highest value was recorded in the third depth of V, U, and OM+U soils.

Introduction:

Since there are no perennial streams to provide a dependable water supply, most of the water being used in Saudi Arabia is groundwater or desalinated sea water from the Red Sea or the Arabian Gulf. Pollutants significantly alter the quality of ground water and often create critical health hazards. Three aquifers (Neogene, Dammam and Umm er Radhuma) are the main source of ground water in Al-Hassa Oasis. Some of the water samples obtained from Neogene aquifer contain high concentrations of nitrate (Water Atlas of Saudi Arabia, 1984). Extensive use of inorganic and organic nitrogen fertilizers in agriculture was identified as an important source of NO_3^- pollution that contaminates both ground and surface water (O'Neill and Gordon, 1994; Kolenbrander, 1972). Some of the applied nitrate fertilizers may be leached down into the deeper aquifers supplying drinking water. It was reported that 20-60% of the applied nitrogen (N) is taken up by arable crops and 40-80% by grass (Parker, 1972). Applying excessive N fertilizer could result in excess quantities of NO_3^- which moves unreacted downward into soil profiles. Moreovere, amending phosphate fertilizer or organic manure with nitrogen fertilizer could significantly increase NO_3^- concentration in the soil system and the increased NO_3^- over needed plant will be moved downward by leaching (Guo et al., 2001).

The hazardous effect of nitrate and nitrite on human health is known, although, nitrate itself is not toxic, nitrite originating from the reduction of nitrate induces methahemoglobinemia in infants. Nitrous acid may dissociate into OH^- and NO^+, and the latter radical oxidizes Fe^{2+} of the hemoglobin to Fe^{3+}, altering the adsorption of O_2 (Mengel and Kirkby, 1972).

According to the EPA (1976), the critical level of NO_3^- concentration in drinking water is 45 mg NO_3^-/l for human, 111 mg NO_3^-/l for chickens. Health advisory levels for most livestock are much higher, 40 ppm (180 mg NO_3^-/l)., It has been reported in Eastern region of in Saudi Arabia that the levels of nitrate ranged between 0 to 66 ppm in Dammamm aquifer, 1 to 660 ppm Neogene aquifer and 0-195 ppm in Wasia aquifer (Water Atlas of Saudi Arabia, 1984)

The aim of this study was to determine the levels of NO_3^- and NO_2^- in groundwater of different aquifers and soil in relation to the use of urea and organic farmyard manure fertilizers and projecting the effect of these fertilizers on the environmental pollution in Al-Hassa region.

Materials and Methods:

Study Area:

Al-Hassa oasis is occupies some 60 km^2 in land of the Arabian Gulf coast between 25 5$^-$ and 25 40$^-$ N Latitude and 49 10$^-$ and 49 55$^-$ E Longitude, and covers an area of approximately 20000 ha. The water of Al-Hassa oasis discharges from a karistified neogene aquifer which is supplied by Umm-er-Raduma formation. This stratum occurs at a depth of some 280 m with a thickness of approximately 320 m (Al-Sayari and Zotl, 1978). There are 32 main springs in Al- Hassa oasis from Neogene, Dammam and Umm-er- Raduma aquifers.

Water sampling and analysis:

Water samples were collected from groundwater at 3 meters depth and from the three aquifers (Neogene, Dammam and Umm-er- Raduma).
Two locations, which consist of several farms, were selected to collect groundwater samples. Location I received farmyard manure (OM) where location II, treated with organic farmyard manure and urea (OM+U). Water samples were then taken from shallow groundwater (approximately 3 meter), Neogene, Dammam, and Umm-er- Raduma.

To study the impact of nitrogen fertilizer type on the content of NO_3^- and NO_2^- in collected groundwater samples, split plot design was used. The main plot was the location and sub main plot was the depth.. Seven samples (7 wells/aquifer) as replicates were collected from each depth. Some chemical properties of used organic manure (average of 10 samples) were determined according to Black (1965). The % values of N, P, K, Ca, Mg, and S were 0.49, 0.20, 0.53, 0.71, 0.18 and 0.09, respectively. Whereas the concentrations values of Zn, Fe and Mn were 6.2, 7.6 and 5.3 ppm, respectively.

Aquifer water samples were collected from the pump outlets of the wells, while shallow groundwater samples were collected from soil profile (1m width X 2m length X about 3m deep). Samples were filtered through a Whatman No. 42 filter paper in polyethylene bottles that had been washed with diluted HCl and thoroughly rinsed with less than 2 uS/dm water. Bottles were titely capped, and stored in ice bags for analysis. The chemical analysis was carried out according to Black (1965). The concentrations of NO_3^- and NO_2^- in collected ground water samples were determined out by Norman et al. (1985) method.

Soil sampling and analysis:

One hundred and twenty soil samples were collected from different fields at Al Hassa area from three depths (0-25, 25-50 and 50-75 cm) to study the effect of organic manure and urea fertilizer on the nitrate contents in soils. Soil samples were grouped according to the source of nitrogen fertilizer into four sets: no fertilizer (virgin soil), organic manure, urea and urea + organic manure. Samples were air dried, ground, sieved to less than 2 mm diameter and analyzed for total N and C according to the methods of Black (1965). Dried soil was shaken with 2 M KCl using 1:10 soil: solution ratio and filtered through Whatman filter paper No. 42. Soil extracts were analyzed for NO_3^- by Norman et al. (1985) method.

Data were statistically analyzed according to Gomes and Gomes (1984). Analysis of variance was carried out and LSD test was used to compare the means.

Results and Discussion

1) General chemical analysis of water samples

The results of chemical analysis of water samples from the two locations at shallow groundwater and the three aquifers of different depth are depicted in Tables 1 and 2. Data demonstrated that electrical conductivity (EC) values in location I decreased with the depth up to Dammam aquifer and then increased in UER aquifer. While, the EC values decreased with depth of the sample source in the location II. Data revealed that salt concentrations in location II were generally higher than that in location I .This may be related to the release of salts into soil solution under the effect of organic matter and urea fertilizers. However, pH values were similar for the different depths of both locations. For major ions, data summarized in Table 1 showed that the concentration of cations of location I decreased in the following order $Na^{+} > Ca^{2+} > Mg^{2+}$ for the samples of upper ground water and Dammam aquifer while in the Neogene and URE aquifers, Mg^{2+} values were greater than Ca^{2+} . Whereas, for anions the order was $Cl^{-} > HCO_3^{-} > SO_4^{2-}$ in the Neogene, Dammam and URE aquifers. But in upper ground water the order was $SO_4^{2-} > Cl^{-} > HCO_3^{-}$.

Table (1)

Chemical contents, pH and EC of water samples collected from location I*.

Properties	Upper Ground water	Deep ground water		
Aquifer name	-	Neogene	Dammam	URE**
Depth (m)	3	25-150	260	450-480
No. of samples	7	7	7	7
pH	7.3-8.1	6.9-7.9	7.7-7.8	7.7-7.8
EC (ds/m)	5.2-8.0	2.3-3.9	0.5-1.0	0.7-3.1
Soluble cations and anion		(mmole l^{-1})		
Ca^{2+}	4.8-32.1	2.6-4.3	1.5-4.0	1.5-6.3
Mg^{2+}	4.3-14.5	4.0-7.0	1.0-3.3	2.0-4.6
Na^{+}	18.3-67.6	15.8-23.1	2.0-2.6	3.0-19.7
SO_4^{2-}	7.1-39.8	1.8-16.5	1.2-1.4	1.0-10.2
Cl^{-}	3.8-57.0	6.8-22.5	1.7-2.0	1.0 - 5.6
HCO_3^{-}	1.0-3.1	3.8-5.4	1.7-2.4	2.4- 4.4

*Crops were fertilized with farmyard manure only.
**URE: Umm er Radhuma aquifer.

Table (2)

Chemical contents, pH and EC of water samples collected from location II*

Properties	Upper Ground water	Deep ground water		
Aquifer name	-	Neogene	Dammam	URE**
Depth (m)	3	80	250	450
No. of samples	7	1	2	2
pH	6.7-8.6	7.5	7.2-7.4	7.2-7.3
EC (ds/m)	5.1-18.5	2.8	1.9-2.1	1.3-1.7
Soluble cations and anion		(mmole l^{-1})		
Ca^{2+}	15.0-50.7	7.4	2.3-2.5	1.4-3.0
Mg^{2+}	4.5-26.9	4.8	0.3-2.4	1.4-2.0
Na^{+}	10.3-125.7	15.7	8.9-15.3	1.5-12.2
SO_4^{2-}	5.2-75.6	13.1	5.2-7.1	3.9-4.3
Cl^{-}	17.5-90.0	16.0	7.0-8.0	5.2-8.8
HCO_3^{-}	1.3-3.0	0.1	0.1	0.1-0.3

*Crops were fertilized with farmyard manure + urea.

**URE: Umm er Radhuma aquifer.

2) Effect of organic manure and N fertilizer on NO_3^- content in ground water :

Table 3 shows the levels of NO_3^- in groundwater (shallow groundwater (SGW), Neogene, Dammam and Umm-er- Raduma aquifers) in the two locations, which received two different sources of N fertilizer (OM and OM+U, respectively). The results revealed that, NO_3^- concentration varied from 2.7 to 213.1 ppm for location I, while in location II, the level of NO_3^- ranged between 9 and 333.7 ppm. Nitrate concentration was found to be aquifer and fertilizer dependent. However, higher value of nitrate was found in location II (Table 3). Moreover, the highest concentration of NO_3^- was detected in SGW samples in both locations. The mean of the concentration of NO_3^- for location I and II at the SGW ranged between 213.1to333.7 ppm, respectively. However, the lowest NO_3^- concentrations were recorded at deep groundwater (depth: 120-420 m) where, the mean values of NO_3^- concentration were 2.7 and q ppm in locations I and II, respectively. These results indicate that groundwater of soil fertilized with urea and organic manure was more polluted with NO_3^- than that fertilized with organic manure alone.

Table (3)

Nitrate levels in ground water from different aquifers in location I and II treated with two types of fertilizers.

Location[@]	Nitrate concentration (ppm) at Upper ground water and Aquifer				Mean[$]
	SGW[*]	**Neogene**	**Dammam**	**UER[*]**	
I	213.1	23	8.3	2.7	61.8
II	333.7	53.6	21.1	9.0	104.3
Mean[$$]	273.4	38.3	14.7	5.9	
Significance between	Level of significance		LSD at 0.05		
Location	**		18.2		
Aquifer	**		24.2		
Location X Interaction	**		15.1		

[@]Location I and II : location I and II received farmyard manure fertilizer and farmyard manure + Urea fertilizer, respectively.

[*]SGW and URE: upper ground water and Umm er Radhuma aquifer, respectively.

$: The whole mean values of nitrate concentration at location I and II.

$$: The whole mean values of nitrate concentration in upper ground water and aquifers.

The results of upper ground water agreed with that of O'Neill and Gorden (1994) and Pekarova and Pekar (1996). They reported that the decrease in nitrate concentration in surface water is mainly due to the decrease in mineral nitrogen fertilizer. Also, Chinkuyu et. al. (2000) found that the poultry manure at a rate of 168 kg N/ha resulted in the lower NO_3^- concentration in sub-surface drain water when compared with the application of commercial N fertilizer. In the same manner, Kryza and Stasko (2000) concluded that increasing nitrate concentration up to 12 mg NO_3^-/l was due to the extensive use of mineral fertilizer. The increasing NO_3^- content of shallow groundwater due to the use of mineral nitrate fertilizer increases the NO_3^- hazard as water pollutant, where NO_3^- concentration exceeded the level recommended by American Public Health Association (1976). In the case of aquifer waters of the two locations are usable without any risk.

3) Effect of organic manure and N fertilizer on NO_2^- content in ground water:

The data in Table 4 show NO_2^- concentration in groundwater of the two previous locations. The data revealed that NO_2^- level decreases with depth, e.g. the upper aquifer (Neogene) was more polluted than the deeper one (Dammam and Umm er Radhuma). Data also showed that there were variations in NO_2^- levels in the two locations. This may be due to the fertilizer type or other factors such as oxidation-reduction reactions in soil system. Furthermore, NO_2^- concentrations were less than that of NO_3^- in the two locations. These results were in agreement with results reported by Stuart et al., (1995), who concluded that the general lack of well water contamination could be the result of the nature of agricultural practices used in the region and/or the effect of the denitrification of nitrogen fertilizers.

Table (4)

Nitrite levels in ground water from different aquifers in location I and II treated with two types of fertilizers.

Location[@]	Nitrite concentration (ppm) at Upper ground water and Aquifer				**Mean[$]**
	SGW[*]	Neogene	Dammam	UER[*]	
I	1.6	0.14	0.14	0.06	0.46
II	1.9	0.23	0.23	0.08	0.59
Mean[$$]	1.75	0.19	0.07	0.07	
Significance between	Level of significance		LSD at 0.05		
Location	**		0.12		
Aquifer	**		0.06		
Location X Interaction	**		0.02		

[@]Location I and II: location I and II received farmyard manure fertilizer and farmyard manure + Urea fertilizer, respectively.
[*]SGW and URE: upper ground water and Umm er Radhuma aquifer, respectively.
$: The whole mean values of nitrate concentration at location I and II.
$$: The whole mean values of nitrate concentration in upper ground water and aquifers.

4) Relationship between NO_3^- levels and C/N ratio of different soils:

A) Unfertilized soil (virgin soil) :

Table 5 summarizes the average of NO_3^- concentrations and C/N ratios for different soils and depths. Data showed that NO_3^- concentrations were 2.5 and 5.1 ppm in the second (25-50 cm) and third layer (50-75 cm) of unfertilized soil, respectively. On the other hand, the values of C/N ratio ranged between 29.4 and 61.9, while, the values of C/N ratio decreased with increasing depth. Insignificant negative correlation coefficient (-0.08) was found between NO_3^- level and C/N ratio. The results may be due to the low values of total nitrogen and organic matter in virgin soil.

B) Fertilized soil with farmyard manure (FYM)

The NO_3^- concentration and C/N ratio at different depths of fertilized soil shown in Table 5 revealed that NO_3^- concentration and C/N ratio followed the same pattern observed above whereas, both values decreased with depth. The values varied from 15 to 28.2 ppm and from 26.7 to 28.4 for NO_3^- concentration and C/N ratio, respectively. The mean values of NO_3^- and C/N ratio were 21.9 ppm and 28.12, respectively. A positive highly significant correlation coefficient (0.924) was found between NO_3^- levels and C/N ratios which may be related to the presence of organic matter which controls the release and transformation of N fertilizer to NO_3^-. Similar result was obtained by Hutchings and martin (1994) in which the addition of organic manure to soil increased both NO_3^- accumulation and C/N ratio.

C) Urea fertilized soil

The results of NO_3^- concentration and C/N ratio at different depths of soil fertilized with urea are presented in Table 5. The data showed that NO_3^- level ranged between 15 ppm in the second depth and 32.9 ppm in the third layer, where the average level of NO_3^- was 26.6 ppm. The values of C/N ratios vary from 13.8 to 19.4. These values decreased with depth. The values were normal for increasing NO_3^- concentration in soil. The average of C/N ratio was 17.2. A negative correlation coefficient (-0.386) was found between NO_3^- levels and C/N ratios which may be an indication of urea dissociation to NO_3^- in soil system rather than the organic matter. This is in agreement with the finding of Hutchings and Martin (1994), where they confirm that the addition of nitrogen fertilizer ($NaNO_3$) in order to narrow the C/N ratio of straw tends to cause an accumulation of nitrate in soil.

D) Fertilized soil with farmyard manure + urea

Nitrate concentration at different depth of soil treated with organic farmyard manure and urea have the same pattern observed for virgin soil and that treated with urea fertilizer (Table 5). However, NO_3^- concentrations were 37.6 ppm in the second layer and 59.9 ppm in third layer. The low values of NO_3^- levels in the second layer may be due to the activity of plant roots and the high values in the third layer were from nitrate accumulation at this layer. On the other hand, data in Table 5 showed that C/N ratio ranged between 10.3 and 12.8. Whereas higher value was found in the surface layer (0-25) and lower value was in the second (25-50 cm) and third layer (50-75 cm). A negative significant correlation was found between NO_3^- levels and C/N ratios in soil treated with organic farmyard manure and urea fertilizer, indicating the effect of this type of fertilizing. And it is in line with results reported by Hutchings and Martin (1994) which showed that the addition of $NaNO_3$ with alfalfa roots and straw to soil increased NO_3^- accumulation and decreased C/N ratio. This could be resulting from either increasing nitrification or decreasing NO3- assimilation.

Table (5)

The mean nitrate concentration (NO_3^-, ppm) and C/N ratio at different depth (cm) of unfertilized soil (virgin soil), and fertilized soil (farmyard manure, urea and farmyard manure + urea) and correlation coefficient between NO_3^- concentration and C/N ratio.

Depth (cm)	Virgin soil		Fertilized soil with					
	Without fertilizer		Farmyard manure		Urea		Farmyard manure + urea	
	NO_3^- (ppm)	C/N ratio	NO_3^- (ppm)	C/N ratio	NO_3^- (ppm)	C/N ratio	NO_3^- (ppm)	C/N ratio
0-25	4.2	61.9	28.2	28.4	32.8	19.4	38.0	12.8
25-50	2.5	32.8	22.5	28.3	15.0	18.6	37.6	10.3
50-75	5.1	29.4	15.0	26.7	32.9	13.8	59.9	10.3
Average	3.93	41.8	21.9	28.1	26.6	17.2	45.2	11.2
Correlation coefficient	-0.08 (N.S)$		0.924**		-0.386(N.S)		-0.735*	

$: N. S. the correlation coefficient is not significant at 0.05 level.

* and ** the correlation coefficients are significant at 0.05 and 0.01 levels, respectively.

Conclusion:

In general, the level of both nitrate and nitrite in shallow groundwater or nitrate in soil in the studied area, exceeded the recommended maximum residue limits, which indicates possible hazardous risk for public health, environment and soil organisms by the effect of these chemicals. Therefore, in order to control the accumulation of such chemicals, the widespread usage of urea and other industrial fertilizers should be restricted and properly managed. With regard to nitrate and nitrite levels in groundwater, still within the recommended limit in accordance with international standards.

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تقدير النترات والنتريتات في التربة والمياه الجوفية في منطقة الأحساء بالمملكة العربية السعودية

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الملخص:

تم تقدير تركيز أيونات النترات (NO_3^-) والنتريتات (NO_2^-) في التربة والمياه الجوفية في منطقة الأحساء في المملكة العربية السعودية وعلاقته بنوع التسميد النتروجيني. تم تجميع عينات المياه من مياه أرضية ضحلة (عمق ٣ أمتار) ومياه جوفية من ثلاث طبقات خازنة للمياه علي أعماق مختلفة: النيوجين (٢٥ – ١٥٠ متر)، الدمام (٢٦٠ متر) و أم الرضمة (٤٥٠- ٤٨٠ متر). أظهرت البيانات أن قيم النترات (NO_3^-) أعلي من القيم التريتات (NO_2^-) في كل من عينات الماء والتربة. في عينات المياه كانت قيم النترات أعلي في المياه الجوفية الضحلة أعلي من عينات المياه الجوفية العميقة حيث تراوحت قيم النترات بين ٥،٩ و ٢٧٣،٤ جزء في المليون. وكانت هناك فروق معنوية بين تركيز النترات بين المصادر الجوفية الضحلة و عينات المياه الجوفية من الثلاث طبقات وكذلك بين مياه الطبقات الثلاثة. كما وجد أن كمية النترات في مياه الترب التي سمدت بسماد عضوي (٦١،٨ جزء في المليون) أقل معنويا من تلك التي جمعت من التربة التي سمدت بكل من يوريا + سماد عضوي (١٠٤،٣ جزء في المليون). وقد جمعت عينات التربة من ثلاثة أعماق (٠- ٢٥، ٢٥- ٥٠، ٥٠- ٧٥ سنتمتر) من الترب المختلفة طبقا لنوع السماد النتروجيني وعدمه: تربة غير مسمدة (V) وتربة مسمدة بسماد المزرعة العضوي (OM) وتربة مسمدة بسماد اليوريا (U) وتربة مسمدة بكل من يوريا بالإضافة الي سماد المزرعة العضوي (OM+U). اتضح من النتائج أن قيم تركيزات النترات في الترب هو كالتالي: OM+U > U > OM >V. وكان متوسط قيم تركيزات النترات هو ٣،٩٣، ٢١،٩، ٢٦،٦، ٤٥،٢ جزء في المليون في OM+U ,U ,OM ,V علي التوالي. وقد لوحظ أن أعلي قيم هي في الطبقة الثالثة (٢٥- ٥٠ سم) من هذه الترب. بينما كانت قيم نسبة الكربون : النتروجين (C/N) ٤١،٨، ٢٨،١، ١١،٢ في ترب OM+U ,U ,OM ,V علي التوالي. وقد قلت قيم ال C/N مع العمق. بينما وجد علاقة أرتباط سالبة بين NO_3^- و ونسبة C/N في الترب التي تحت الدراسة. ومن جهة أخري وجد أن معامل ارتباط موجب ومعنوي بين قيم كلا من تركيز النترات ونسبة C/N في تربة OM.