

## ASSESSMENT OF NITRATES AND THEIR HEALTH IMPACT IN QUATERNARY GROUNDWATER AQUIFER OF SOHAG GOVERNORATE, EGYPT

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### Abstract

The present study is destined to measure the nitrate levels of groundwater in Sohag Governorate aiming to determine their possible sources and evaluate their environmental and health impact. 17% of the analyzed samples have  $\text{NO}_3^-$  levels go over the MAL. Potentially harmful levels of  $\text{NO}_3^-$  exceeding nearly four times higher than the maximum allowable limit (MAL) for drinking water (45 mg/l) were reported. About 32% of the examined samples displaying nitrate levels more than 13 mg/l (pollution indicator). Nitrate is converted to nitrite in the infant's stomach causing transformation of hemoglobin (the oxygen carrier) to methemoglobin that can't carry oxygen. Therefore, oxygen starvation might occur causing serious disorders or even death (methemoglobinemia). Moreover, nitrate is a precursor in the formation of the carcinogenic *N*-nitroso compounds (NOC). Also, the augmentation of nitrate in groundwater is of vital concern because they indicate the groundwater pollution by anthropogenic contaminants (chemical and/or biological). Quantification of the distribution pattern of the environmental diseases (e.g. methemoglobinemia, cancer, kidney failure, neurological disorders) in Sohag Governorate and its interrelation with the drinking water pollution is strongly recommended; the convenient regulations can be then taken.

### Introduction

Nitrogen, an essential element in living matter, it is a major compound of proteins and is essential for Food and fiber production, it appears in our environment in several different forms. Nitrate is the major form of nitrogen it is present in natural waters only in small quantities. Although nitrate contamination of water can result from several natural and human activity-related sources, the on-site wastewater disposal is considered to be a significant contributor. Most of the nitrogen in excrete derives from the urine. The nitrogen forms in urine are highly soluble, and when mixed with water they are not easily liberated. Therefore, the major part of nitrogen will remain in wastewater. Once the conditions become anaerobic, the organic nitrogen is converted to ammonium ( $\text{NH}_4^+$ ) which is the most common nitrogen form reaching the wastewater disposal sites. There, ammonium is subjected to the process of nitrification to be transformed to nitrite ( $\text{NO}_2^-$ ) and then into nitrate ( $\text{NO}_3^-$ ). Nitrate is highly soluble in water and poorly retained in the soil; it can be thus transported long distances through the soil to reach the water sources. The leaching potential of  $\text{NO}_3^-$  within the soil system depends on many factors, of which the soil texture is most important (Smith and Cassel, 1991).

The present study is intended to measure the levels of nitrate ( $\text{NO}_3^-$ ) in the groundwater of Sohag Governorate (Fig. 1), aiming to determine their possible sources and discuss their environmental and health effects. Sohag governorate occupies a part from the Nile valley in Upper Egypt extending about 125 Km. It is bordered from the north by Assuit Governorate and from the south by Qena Governorate. Sohag Governorate is the 10<sup>th</sup> largest population in Egypt, with a total population of slightly less than four millions. About 78% of these numbers live in the rural communities; the remaining 22% are urban dwellers.

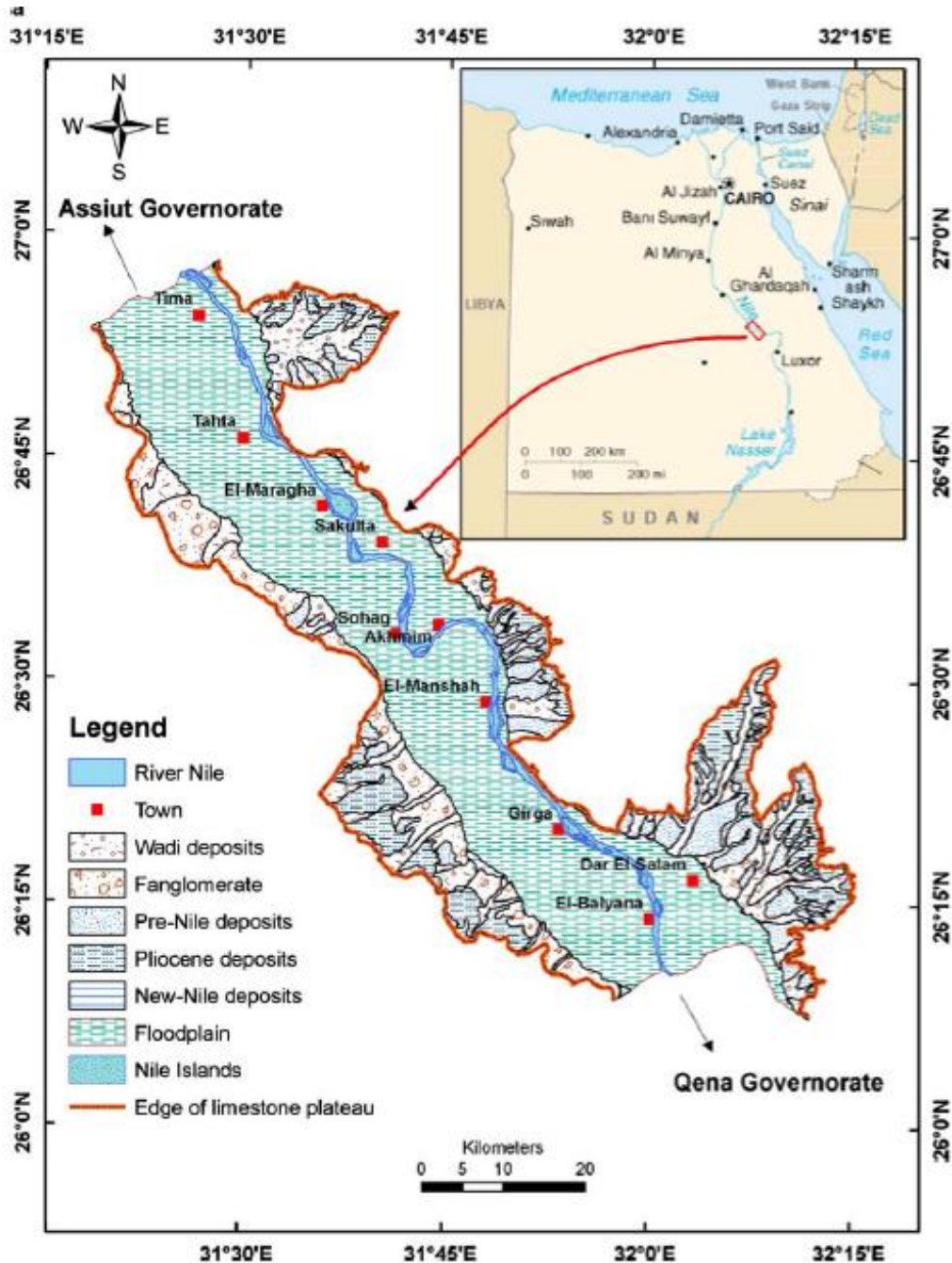


Fig. (1): Simplified geological map of Sohag governorate (TEGPC and CONOCO 1987, Ahmed 2011).

### Hydrological setting

The Quaternary aquifer in Sohag Governorate composed of Pliocene clay (Muniha Formation) which plays as the base of the aquifer, the Pliocene clay characterized by its impermeability and it confine the aquifer. It is overlying by Pleistocene fluvial sediments in the cultivated lands. The fluvial sediments is composed mainly from gravel and sands with clay lenses intercalations. These Pleistocene sediments are covered by the floodplain sediments. The floodplain sediments are formed of recent sediments which composed of semi-permeable silt bed and play as the semi-confined bed to the aquifer. It acts the top layer of the aquifer. Thus the aquifer which present in the cultivated lands is known as the semi-confined aquifer (Fig. 2).

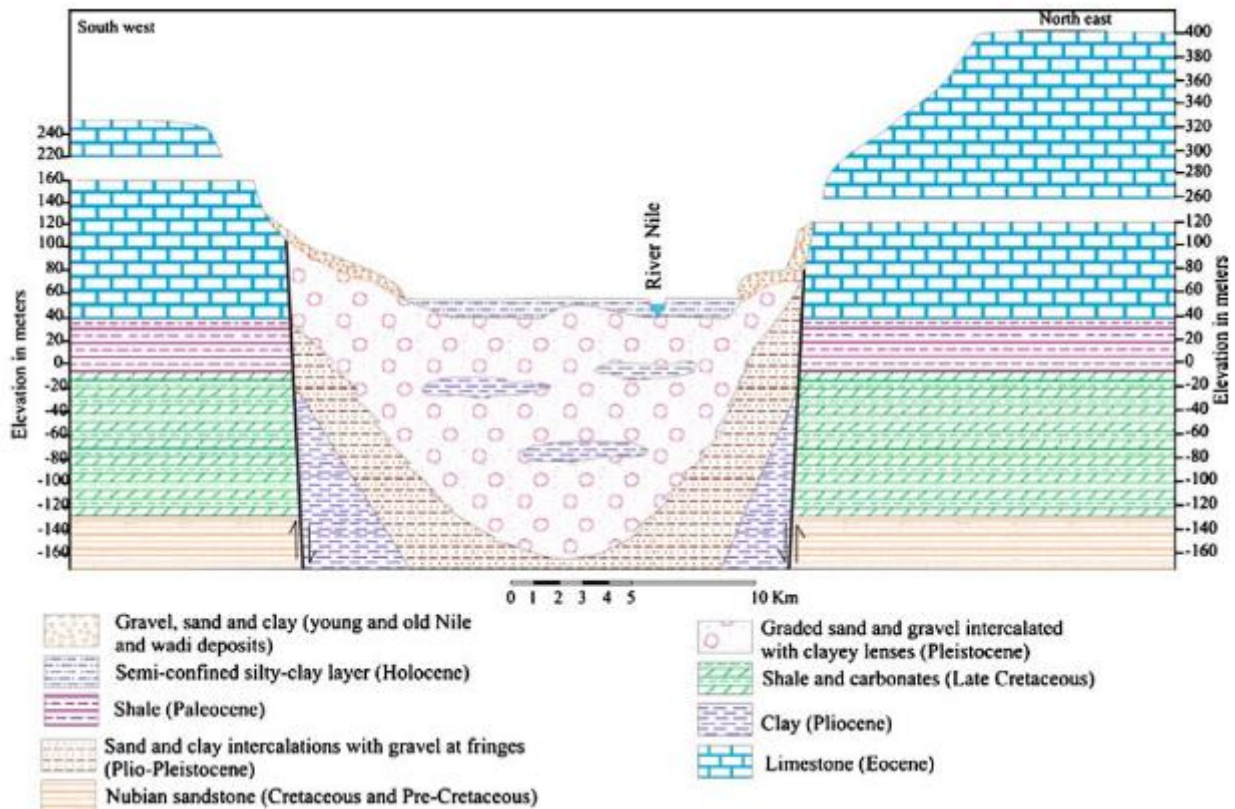


Fig. 2: Hydrological section crossing the Nile Valley showing the Quaternary aquifer (RIGW 1990, Ahmed 2011).

The aquifer system is recharged by the infiltration of the surplus of the irrigation water that leaks beneath the cultivated lands vertically and seeps in the direction of the reclaimed desert lands horizontally, seepage from irrigation canals such as west Tahta and west Girga projects in the reclaimed desert lands. These canals play an important position in the pattern of water table of the aquifer and get its water from the dominant Nile water. And it is important to mention that these canals play a very important role in recharging its surrounding aquifer. While the seepage from the Nile water to the aquifer is a secondary source for recharge. Connate water that deviates during the rainstorm ears in the desert areas forms other means of groundwater source in the study area (Omer and Abdel Moneim 2001). The most important form of groundwater discharge in the study area is that pumped from the public municipal and private wells that extracted for different uses. And the evapotranspiration processes considered as the secondary discharge.

### Methodology

Determination of the source of  $\text{NO}_3^-$  and the factors controlling their distribution pattern in the groundwater in Sohag governorate is the main objective of the present study. The study is concerned with the distribution pattern of  $\text{NO}_3^-$  in the groundwater in the study area and also to monitor their spatial variability. Groundwater samples (858 samples) representing the different eco-regions were collected and analyzed for their contents of nitrate (Fig. 3).

The nitrate content ( $\text{NO}_3^-$ ) in the groundwater samples was spectrophotometrically estimated applying the sodium salicylate method (DEWAS, 1980).



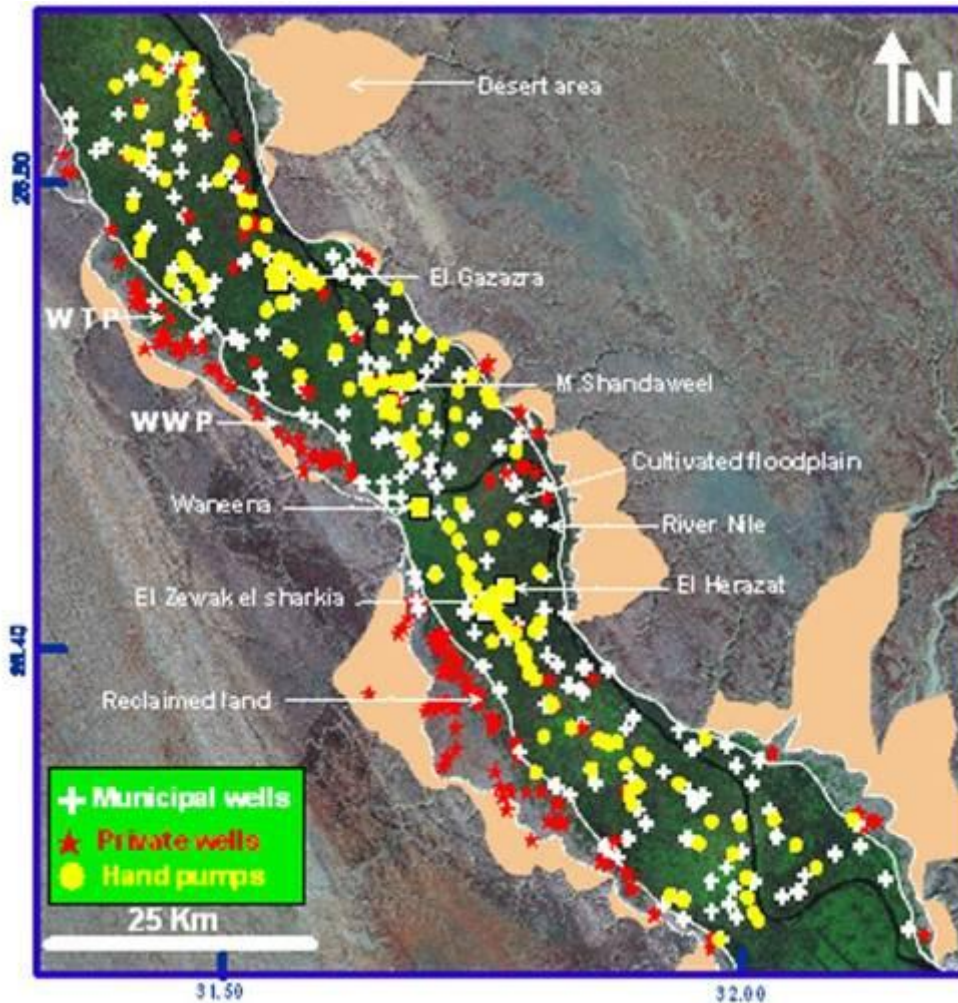


Fig. 3: Map of the Sohag Governorate showing the groundwater samples sites. (WTP: West Tahta Project- WWP: West Waste Project).

## Results and Discussion

Groundwater samples of the Pliocene aquifer show a wide range of nitrate content fluctuating from 0.37 mg/l to 111 mg/l with a mean value of 11.4 mg/l. The majority of samples (of 20 samples) possesses the nitrate content fluctuating within the non-outlier range (<17.8 mg/l). Only one sample displays markedly elevated  $\text{NO}_3^-$  concentration (111 mg/l). The general low content of  $\text{NO}_3^-$  in the Pliocene aquifer confirms its limited vulnerability. The elevated concentration in the mentioned wells implies that the Pliocene aquifer is partly connected with nearby Pleistocene unconfined aquifer, where the nitrogen fertilization is the main source.

Regarding the unconfined aquifer, the measured nitrate of the analyzed groundwater samples (143) have a wide range varying from about 0.002 mg/l to 162 mg/l with an average of 71.7 mg/l.

The statistical analysis (Table 1) shows that 25% of the examined water samples display obviously low  $\text{NO}_3^-$  content varying from 0.002 mg/l to 18.2 mg/l (lower quartile). On the other hand, 25% of the samples have markedly elevated level of  $\text{NO}_3^-$  ranging from 119 mg/l (upper quartile) to 162 mg/l. In addition, the interquartile range (IQR= 101 mg/l) is characteristically high. So, the extremely wide range of  $\text{NO}_3^-$  in the Pleistocene unconfined aquifer is confirmed.

Reviewing the spatial distribution pattern of  $\text{NO}_3^-$  throughout the unconfined aquifer, the following notes can be extracted:

The majority of samples displaying elevated levels of  $\text{NO}_3^-$  are pumped from:

a- Wells located in the older reclaimed lands, reflecting the effect of the intensive long-term nitrogen fertilization.

b- Wells located downflow from the West Sohag Wastewater Disposal Project suggesting that wastewater is confirmly penetrating the permeable vadoze zone to reach the groundwater underneath.

The markedly elevated levels of  $\text{NO}_3^-$  in the unconfined aquifer in general and the mentioned locations in particular confirm the concept that this aquifer is highly vulnerable. Most samples showing depleted levels of  $\text{NO}_3^-$  are extracted from wells located along the border with the low vulnerable semiconfined aquifer.

The nitrate concentration in groundwater samples from the semiconfined aquifer (mean= 11.5 mg/l) is significantly lower than that reported in the unconfined aquifer (mean= 71.7 mg/l); the matter which is statistically confirmed ( $p= 0.000$  at 95% confidence level). The lower level of  $\text{NO}_3^-$  in the semiconfined aquifer relative to the unconfined aquifer although the longer time of nitrogen fertilization is controlled by the aquifer vulnerability which is higher in the unconfined aquifer.

Groundwater pumped from the shallow zone exhibit a wide range of  $\text{NO}_3^-$  fluctuating from 0.001 mg/l to 157 mg/l with a mean value of 17.8 mg/l. Statistically 50% of the samples are extremely depleted in  $\text{NO}_3^-$  being vary from 0.001 mg/l to 5.3 mg/l (median). On the other hand, 25% of these samples shows marked elevated  $\text{NO}_3^-$  content ranging from 23.5 mg/l (upper quartile) to 157 mg/l. Moreover, anomalous levels of  $\text{NO}_3^-$  ( $> 56.8$  mg/l) were reported to be considered as outlier values. Most of samples showing elevated levels of  $\text{NO}_3^-$  are located in the rural residential communities indicating the effect of the domestic wastewater as a significant source of nitrogen compounds. On the other hand, some water samples possessing high  $\text{NO}_3^-$  concentrations are extracted by hand-pumps situated in the cultivated floodplain reflecting the effect of the nitrogen fertilization. The nitrate level in groundwater samples from the medium zone is significantly lower than that reported in the shallow zone ( $p= 0.02$  at 95% significant level). Also, nitrate of the medium zone is more uniform than the shallow zone (Table 1).

Table (1): Descriptive statistics of nitrate (mg/l) in the Quaternary aquifer in Sohag area.

Aquifer		Min	Max	Mea n	5%	10 %	L.q	Media n	U.q	90%	95%	C.V	
Pliocene aquifer		0.3 7	110. 0	11.3	0.4 2	0.68	1.30	2.82	5.97	33.5	82.6	230. 0	
Pleistocene aquifer	Unconfined	0.0 0	162. 0	71.7	1.3 5	6.32	18.2 0	78.30	119.0 0	131. 0	137. 0	69.0	
	Semiconfined	Shallow zone	0.0 0	157. 0	17.7	0.3 9	0.64	1.26	5.33	23.50	49.6	87.1	153. 0
		Mediu m zone	0.4 5	57.1	8.82	0.7 7	0.89	1.74	4.91	10.40	22.1	33.4	128. 0
		Deeper zone	0.0 0	92.1	4.19	0.0 0	0.01	0.65	1.77	4.12	10.6	16.6	191. 0

Min.= minimum; Max.= maximum; L.q= lower quartile; U.q= upper quartile; C.V= coefficient of variation; 5%, 10%, 90%, 95% = percentiles.

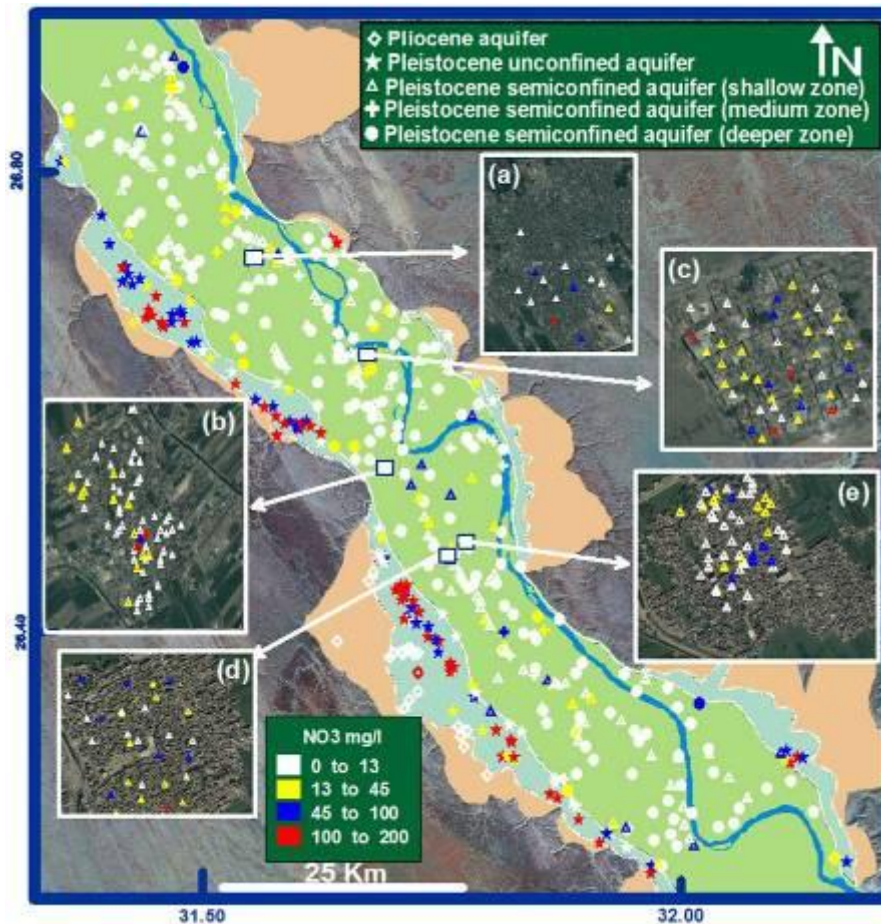


Fig. 4: Map showing the distribution pattern of nitrate in the groundwater aquifer throughout Sohag Governorate.

The analyzed water samples from the medium zone have nitrate values ranging from 0.451 mg/l to 57.1 mg/l with an average value of 8.8 mg/l. However, the majority of samples (90%) show NO<sub>3</sub><sup>-</sup> content less than 22.1 mg/l (90% percentile) confirming the lower level of NO<sub>3</sub><sup>-</sup> compared with the shallow zone. Only two samples have elevated NO<sub>3</sub><sup>-</sup> content (>23.5 mg/l) to be considered as outlier values.

The groundwater samples pumping from the deeper zone display NO<sub>3</sub><sup>-</sup> values ranging from 0.00 mg/l to 92.1 with a mean value of 4.2 mg/l. The major part of the examined samples (90%) shows depleted levels of NO<sub>3</sub><sup>-</sup> (<10.6 mg/l). Although the general low content of NO<sub>3</sub><sup>-</sup> in the deeper zone, abnormal levels were reported in some samples to be considered outlier values. These samples are mostly situated in the rural residential areas reflecting the effect of the domestic wastewater.

It is clear that nitrate is getting lower downward in the semiconfined aquifer from the shallow (mean= 17.8 mg/l) through the medium (mean= 8.8 mg/l) and the deeper (mean= 4.2 mg/l) zones. This suggests that the anthropogenic activities adding NO<sub>3</sub><sup>-</sup> to the aquifer are more effective in the shallow zone and declined downward. In addition, the reducing conditions which convert nitrate into ammonia become more effective with depth.

### Health effects of nitrogen compounds and ground water quality

Although nitrate itself is considered to be of low toxicity, its transformation into other forms as nitrite (NO<sub>2</sub><sup>-</sup>) and N-nitroso compounds is responsible for many adverse health effects. Nitrate has been implicated in a number of health outcomes. These effects include methemoglobinemia, hypertension, increased infant mortality, central nervous system, birth defects, diabetes, respiratory tract infections, changes to the immune system and even cancer via

the bacterial production of N-nitroso compounds (Morton, 1971; Malberg et al., 1978; Dorsch et al. 1984; Kozliuk et al., 1989; Kostraba et al., 1992; Hill, 1999;). Upon these, methemoglobinemia is directly linked to high nitrate levels in drinking water. In the infant stomach, nitrate ( $\text{NO}_3^-$ ) is bacterially reduced into nitrite ( $\text{NO}_2^-$ ). In the blood stream, nitrite oxidizes the ferrous ion ( $\text{Fe}^{2+}$ ) of the hemoglobin into the ferric ion ( $\text{Fe}^{3+}$ ) to be transformed into methemoglobin. Hemoglobin is considered the carrier of oxygen and carbon dioxide in the body, whereas methemoglobin can't carry oxygen or carbon dioxide (Fecham et al., 1986). The inability of blood to effectively transport oxygen and carbon dioxide causes oxygen starvation and results in acute distress to the system causing the methemoglobinemia (or blue baby disease). Symptoms of methemoglobinemia include an unusual bluish gray or brownish gray skin color, irritability and excessive crying in children with moderate levels of MetHb, and drowsiness and lethargy at higher levels (Brunning-Fann and Kaneene 1993). Chocolate-colored blood is an indirect clue indicating the presence of elevated MetHb levels.

Nitrite derived from nitrate can react with amines and amides to form N-nitroso compounds, which may have carcinogenic properties (Van Maanen et al. 1996). In Colombia and Italy, high levels of nitrate in well waters are associated with an increased risk of gastric cancer (Cuello and Correa 1976; Gilli et al. 1984). In a cross sectional study in an area with a high incidence of gastric cancer in north eastern China, an association between high levels of nitrate in drinking water supplies and neoplastic changes in the stomach was observed (Xu et al. 1992). The association of stomach cancer and high nitrate levels were also reported by Boeing (1991). A link between cancer risk and endogenous nitrosation as a result of high intake of nitrate and/or nitrite and nitrosatable compounds is possible (Speijers et al., 1989; FAO/WHO, 1996, WHO, 2007). Depending on their work in the Netherlands, Van Maanen et al. (1996) reported that urinary excretion of N-nitrosamines was observed during high nitrate exposure. They concluded that drinking water contamination by nitrate may imply a genetic risk. However, no convincing evidence was found of an association between gastric cancer and the consumption of drinking water in which nitrate concentrations of up to 45 mg/l were present (WHO, 2007).

Possible relationships between nitrate intake and effects on the thyroid have also been studied, as it is known that nitrate competitively inhibits iodine uptake. In addition to effects of nitrate on the thyroid observed in animal studies and in livestock, epidemiological studies revealed indications for an antithyroid effect of nitrate in humans. The nitrate effect on thyroid function is strong if a nutritional iodine deficiency exists simultaneously (Höring et al., 1991; Höring, 1992). Höring & Schiller (1987), Sauerbrey & Andree (1988), and Van Maanen et al. (1994) found that inorganic nitrate in drinking water is a manifested factor of endemic goitre. Both the experimental and epidemiological studies give the impression that nitrate in drinking-water has a stronger effect on thyroid function than nitrate in food.

WHO (1984) prescribed a drinking water standard of 45 mg/l for nitrate. The Egyptian maximum permissible limit (MPL) of nitrate is equivalent to the stated drinking water standard. Worth mentioning, the current drinking water standard and health advisory level of nitrate (45 mg/l) is based only on the non cancer health effects related to infantile methemoglobinemia (Kross et al., 1993). This means that, the drinking water standard for nitrate (45 mg/l) was set primarily to prevent the incident of infant cyanosis, or methemoglobinemia. The maximum permissible limit of ammonia in drinking water approved by the European Union (EU) is 0.5 mg/l  $\text{NH}_4^+$ ; the same limit was accepted in the Egyptian environmental regulations (EHCW, 2007).

Natural levels of nitrogen compounds in groundwater are usually very low; the augmentation of nitrogen compounds (nitrate and ammonia) in groundwater is thus considered as pollution indicator (WHO, 1993). So, elevated levels of nitrogen compounds are of concern not



only because of their potential adverse effects, but also because they imply reaching of anthropogenic contaminants to such water resource. Therefore, other dangerous chemical and biological pollutants are strongly expected to be present in such water source. However, concentrations of nitrates over than 13 mg/l are usually considered as indicative of anthropogenic pollution (Madison & Brunett 1985, Kross et al., 1993).

Worthwhile to mention that, the augmentation of nitrate and ammonia in groundwater is of concern not only because of their potential adverse effect, but also because they indicate the groundwater pollution and confirm reaching of anthropogenic contaminants to such water resource. Therefore, other chemical (organic and inorganic) and biological pollutants are strongly expected to be also present.

Therefore, other chemical (organic and inorganic) and biological pollutants are strongly expected to be also present.

Figures (5) show the distribution pattern of nitrate and ammonia throughout the groundwater aquifer in Sohag Governorate. Sites displaying nitrate and ammonia concentrations meeting and those exceeding the Egyptian maximum permissible limits (45 and 0.5 mg/l, respectively) are marked out.

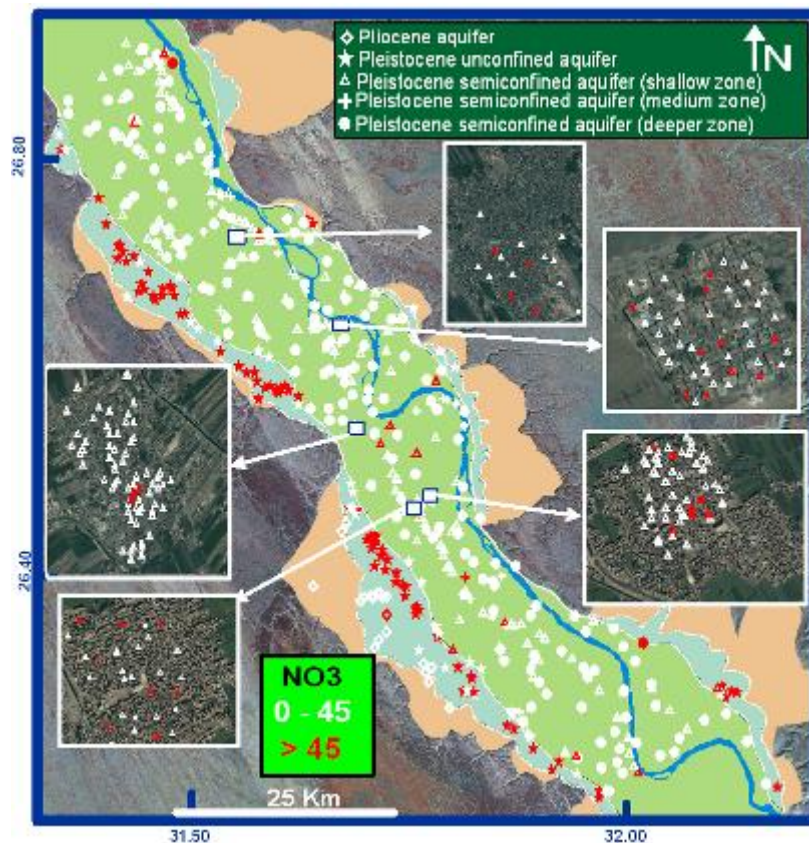


Fig. 5: Distribution pattern of nitrates in the groundwater aquifer throughout Sohag Governorate.

Regarding the Pliocene aquifer, the majority (90%) of the analyzed groundwater samples display nitrate content (0.370 - 12.25 mg/l) that meet the Egyptian maximum permissible limit (MPL) of 45 mg/l. Accordingly, these samples are suitable for drinking and domestic purposes concerning their nitrate level. However, about 10% of the examined samples have  $\text{NO}_3^-$  values (54.8 to 110.5 mg/l) that markedly go beyond the MPL. About 35.7% of the groundwater samples from the unconfined Pleistocene aquifer possess nitrate values in the range 0.002 – 44.8 mg/l that correspond the Egyptian MPL and therefore they are suitable for domestic exploitation



with reference to their nitrate content. The other samples (about 64.3%) are incompatible for domestic uses concerning their nitrate levels being have  $\text{NO}_3^-$  values (45.8 – 161.9 mg/l) surpassing the stated limit. Moreover, nitrate levels exceeding nearly more than four times higher than the MAL were recorded in some wells ( $\approx 162$  mg/l).

With reference to the shallow zone of the Pleistocene semiconfined aquifer, about 87.5% of the considered groundwater samples are suitable for domestic purposes regarding their nitrate where they have  $\text{NO}_3^-$  contents (0.001 – 44.8 mg/l) meeting the Egyptian MPL. The rest samples (about 12.5%) are improper for domestic uses because of their elevated nitrate concentrations (46.1 - 157.0 mg/l) that go beyond the Egyptian MPL.

Concerning the medium zone of the Pleistocene semiconfined aquifer, the preponderance (98%) of the examined samples are suitable for domestic purposes regarding their nitrate content as they have  $\text{NO}_3^-$  levels (0.451 – 44.3 mg/l) meeting the Egyptian MPL. But only one sample is inappropriate for domestic employment as it has  $\text{NO}_3^-$  content (57.1 mg/l) exceeding the mentioned limit.

The vast majority of samples (about 99.3%) from the deeper zone of the Pleistocene semiconfined aquifer are safely drinkable regarding their nitrate content being have  $\text{NO}_3^-$  values (0.000– 38.9 mg/l) that are analogous to the Egyptian MPL. The rest two samples are unfit for domestic use as they have  $\text{NO}_3^-$  values (49.48 and 92.11 mg/l) greater than the stated MPL.

Considering the groundwater beneath the cultivated lands, water is abstracted from two three different zones: the shallow zone (private hand pumps <12 m depth), the medium zone (privet wells) and the deeper zone (municipal wells 30-60 m depth). In the shallow zone, the content of nitrogen compounds is generally higher than the medium and the deeper zones. About 12.5% of samples from the shallow zone have elevated levels of  $\text{NO}_3^-$  exceeding the MAL and the (EHCW, 2007). But in the medium zone, about 1.9% of the samples exceed this limit. And only two samples from the deeper zone have high concentration of  $\text{NO}_3^-$ .

The lower content of nitrate in the medium and deeper zones reflects their reduced amounts migrating to such deeper depths.

With regard to the samples beneath the sewage wastewater disposal sites, abnormal concentrations of nitrogen compounds were recorded . All samples possess  $\text{NO}_3^-$  contents surpassing, or very close to, the MAL and the (EHCW, 2007). The nitrogen compounds in the wastewater are derived mostly from the human activities. The nitrogen forms in urine are highly soluble and the major part is preserved in the wastewater. Once the conditions become anaerobic, the organic nitrogen is converted to  $\text{NH}_4^+$  which transformed into  $\text{NO}_3^-$  through the process of nitrification. The relative abundance of  $\text{NO}_3^-$  is controlled by many biological and chemical transformations.

With respect to the shallow, medium, deeper groundwater in the rural residential communities, elevated concentrations of both  $\text{NO}_3^-$  of samples displayed levels exceeding the MAL of  $\text{NO}_3^-$ . This abnormal content of nitrogen compounds is related to the widespread application of domestic waste cesspools in such communities.

### **Conclusion and recommendations**

The present study revealed that groundwater of Sohag Governorate frequently possesses elevated levels of  $\text{NO}_3^-$  exceeding the MAL and (EHCW, 2007) for drinking water. The nitrogen compounds in groundwater of Sohag are related to many factors which are mainly of anthropogenic source. Elevated levels of  $\text{NO}_3^-$  may be a factor responsible for the frequently incident methemoglobinemia (the blue baby disease).The World Health Organization cited

numerous cases of nitrite intoxication following ingestion of well water containing high levels of nitrate, almost 98% of which were associated with nitrate levels in the range of 44-88 ppm.

The drinking water in Sohag should be investigated for their content of nitrogen compounds. Water resources displaying elevated concentrations of  $\text{NO}_3^-$  should be eliminated and other sources must be employed. Stringent regulations should be taken to prevent domestic wastewater disposal into the shallow groundwater aquifer. The public wells should be at more depths to mitigate the problem of anthropogenic pollution (nitrate, ammonia, phosphate and micro-organisms).

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