Sources and health risk assessments of nitrate in groundwater, West of Tahta area, Sohag, Egypt

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Introduction

Groundwater is an essential source of water for drinking, agricultural and industrial purposes in arid and semi-arid regions worldwide (Liu et al., 2010; Li, 2016). As a result of the rapid increase in population and urbanization, many environmental problems including groundwater pollution have been evolved (Burri et al., 2019; Shaharoona et al., 2019). Recently, many anthropogenic processes are responsible for groundwater contamination such as Ca, Fe, PO₄, NO₃, NH₄, SO₄ (Aiuppa et al., 2003; Redwan and Abdel Moneim, 2016a; Arslan, 2017; Burri et al., 2019) and heavy and trace metals such as Al, As, B, Ba, Fe, Co, Cd, Cr, Cu, Hg, Mo, Ni, Se, Si, V and Zn (Giammanco et al., 1998; Fu and Wang, 2011; Fu et al., 2014; Hosseini and Saremi, 2018; Kyere et al., 2018; Burri et al., 2019). Proper management of contaminants and identifying their possible sources is vital in arid and semi-arid regions (Qasemi et al., 2018b). Disposal sites of wastewater are considered great pollution sources and can create a serious threat to the environment through direct contact with the surface water and/or groundwater (McArthur et al., 2012). Wastewater contains many dissolved organic and inorganic matters, including heavy elements such as arsenic, copper, cadmium, chromium, zinc, lead, and pathogenic bacteria (Fu and Wang, 2011; Fu et al., 2014).

Among the different contaminants, nitrate is considered as a significant ion to limit water potability (Borzi et al., 2015). Nitrate concentration in water more than 10 mg/L may be toxic to infants and may lead to stomach cancer (Wu and Sun, 2016). According to the World Health Organization (WHO), very quick health problems can be produced through drinking nitrate-rich water. Nitrate values above 50 mg/L in drinking water (WHO, 2011) can be risky to human and animal lives (Sall and Vanclooster, 2009). Nitrate is considered a threat to the environment through direct contact with the surface water and/or groundwater (Burri et al., 2019). Nitrates degrade water quality, leading to the eutrophication and the spread of toxic algal blooms in water (Ji et al., 2013). High nitrate concentration in drinking water can raise the risk of infants' methemoglobinemia, diabetes, spontaneous abortions, cancer, thyroid gland problems, mutagenesis and teratogenesis (Fu et al., 2017). Environment Protection Agency (EPA) states that the nitrate concentration for drinking water should not exceed 45 mg/L (USEPA, 2013). Elevated nitrate or nitrite concentration showed adverse reproductive effects like abortions and many birth defects on animal (Manassaram et al., 2006).

Based on the International Agency for Research on Cancer regulations (IARC, 2010), nitrate and nitrite are treated as “probably carcinogenic to humans”. As a result, drinking water contaminated by the nitrate is a severe environmental health hazard that requires special attention by the decision-makers to reduce contamination (Qasemi et al., 2018a). Nitrate can be formed through natural fixation of nitrogen or human activities through sewage disposal, chemical fertilizers, landfills leachate, dairy products, and chemical industries, as along with organic matter nitrification processes by bacteria (Manassaram et al., 2006; Wu and Sun, 2016; Wang et al., 2018).

Few authors have investigated the groundwater potentiality and quality in the study area (Dawoud, 1997; Esam et al., 2012; Redwan and Abdel Moneim, 2016b) without giving the environmental hazards related to groundwater contamination. The aims of the current study were to (1) quantify the levels of nitrate in drinking water around the wastewater treatment plant and farmlands, west of Tahta city, Sohag,
Egypt and (2) compare the nitrate levels to the standards values of EPA, (3) determine human health impacts. The results obtained from this study regarding the nitrate sources and distribution will help the stakeholders and decision-makers for the current status and future water and/or wastewater improvement and management nationwide.

Materials and Methods

Study Area Description

The study area is situated in the desert zone west of Tahta city, west of the Nile, to the north of Sohag (about 400 km from Cairo) (Fig. 1). Many agricultural activities, new industrial zone and urban centers on the newly reclaimed areas can be recognized. The study area is surrounding from the west by the Eocene limestone plateau dissected by several W-E direction drainage basins with a general slope towards the agricultural areas and the River Nile. The study area belongs to the arid zone of Egypt, characterized by a dry climate with an average temperature and annual rainfall of about 23.8-24.0°C and 0.70-0.75 mm, respectively. The west Tahta wastewater treatment plant has a 35,000 m$^3$/day design capacity (Fig. 1), provided for about 400,000 citizens living in the Tahta city. The recycled water from the plant is used for irrigating casuarina trees for timber and firewood in an area of 4.5 km$^2$. The increasing amounts of wastewater reach the treatment plant that exceeds the design capacity by about 10.1 m$^3$/day, develop wastewater ponds with a potential risk of groundwater contamination. Septic tanks and pit latrines are common onsite sanitation facilities developed in most of the villages due to the lack of wastewater treatment facilities.

Geology

The sedimentary succession of the study area ranges in age from Lower Eocene to Recent as follows (Fig. 2):

a. Thebes Formation (Lower Eocene) (Said, 1960): massive to laminated limestone with flint and marl with *Nummulites* sp. and planktonic foraminifera.

b. Muneinha Formation (Early Pliocene) (Issawi et al., 1978): thick beds of quartz and clay (montmorillonite and kaolinite) of estuarine and fluvial origin. It forms the base of the water bearing formations in the Nile Valley (Fig. 2).

c. Qena Formation (Early Pleistocene) (Said, 1981): quartzitic sand and gravel deposited in braided/ and low-moderate meandering streams. It contains heavy minerals of metamorphic origin come by the river from the Sudanese Red Sea hills (Omer, 1996). This formation forms the main water bearing zone in the study area.

d. Kom Ombo Formation (Early Pleistocene- Middle Pleistocene)
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(Issawi and Hinnawi, 1980): alluvial and fluvial facies gravels and sands of igneous and metamorphic origins.

e. Ghawanim Formation (Middle Pleistocene) (Omer, 1996): cross-bedded fluvial sand and gravels with conglomerate and quartzitic sandstone bands and lenses formed by the Egyptian and Ethiopian water.

f. Dandara Formation (Late Middle Pleistocene) (Said, 1975): poorly sorted sands ranging from coarse to very fine grain sizes with sediments from Ethiopia and followed by sediments from equatorial Africa. Recent flash flood wadi deposits in the area deposits are distributed on the surface of the old sediments throughout study area (Fig. 2). These deposits include alluvial deposits which composed mainly of mud and silts with sandstone intercalations and fine sand grains.

**Hydrogeologic Characteristics**

The groundwater aquifer is represented by the Qena Formation (Abdel Moneim, 1999) that is overlain by recent sediments (mud and silts with sand and sandstone intercalations) under unconfined conditions (Fig. 2) and towards the east changed into semi-confined aquifer of the area underlying the floodplain Nile sediments (fine sand and silt with low vertical and horizontal permeability, < 15.6 m/day). The aquifer thickness varies from 20 to 80 meters, and increases towards the east (i.e. the Nile cultivated lands). Groundwater levels in the study area vary from 45.5 m in the northern part and 53 m in the southern part of the area. The groundwater hydraulic gradient is generally from the west to east having value ranging between 0.0006 and 0.0016 with an average of about 0.008. Recharge process of the Quaternary aquifer is from excess irrigation water seepage from the cultivated areas and irrigation canals and post-Eocene rainwater stored in the Pleistocene aquifer. The main groundwater flow direction is to the northwest following the topography of the floodplain and discharge. The second flow direction is west - east towards the River Nile as a result of canals recharge.

**Sampling and Analysis**

Sampling in 23 farm wells around the west of Tahta wastewater treatment plant and farmlands was collected to measure groundwater nitrate contamination. No samples were collected far away from the treatment plant in the old agricultural lands due to the presence of a residential community with private pit latrines. The agricultural crops in the study area are mainly wheat, corn, alfalfa, barley and onion. Field sampling was carried out during June 2016 using 300 pre-cleaned polyethylene plastic bottles. After pumping out water from the wells for about 3-4 min to remove any stagnant water in the pipes, the bottles were cleaned three times with the sampled water before filling it with the representative samples to exclude any external contamination. The groundwater samples were transported to the laboratory to be directly analyzed for nitrate, sulfate, and chloride concentrations using Dionex ion chromatography (IC). Carbonates were measured using titration method (Stumm and Morgan, 2012). The bacteriological analysis was carried out using the multiple tube fermentation technique. Methods precision was evaluated using calibrated standard solutions. Blanks and triplicated standard samples were applied for quality control and quality assurance (QC/QA). The sampling process and analytical techniques used were based on standard methods for the testing of Water and Wastewater (APHA, 2012).

Figure 2. Cross-sectional lithological view and potential NO$_3^-$ pollution pathways in the study area.
**Statistical Analysis**

Statistical analyses of the groundwater hydrochemistry data were performed on normalized data using the statistiXL software (www.statistixl.com/) in order to quantify the inter-correlation between the different variables. The Kaiser criterion is used to set the maximal extracted factors numbers with eigenvalues higher than 1 (Lawrence and Upchurch, 1982; Costello and Osborne, 2005). The principal component analysis (PCA) loading values >0.75, 0.75-0.5, and <0.5 indicate a strong, moderate and weak respectively.

**Human Health Risk Assessment**

Human health risk assessment is “the process to estimate the nature and probability of unfavorable effects to humans due to chemicals exposure in a contaminated environment, now or in the future” (Ruiz et al., 2013). Health risk assessments could play a key role in health promotion and disease prevention both at an individual and a population level (Orlando and Wu, 2018). Human health risks are classified as non-carcinogenic (induced by non-carcinogens) risk and carcinogenic (induced by carcinogens) risk (Keramati et al., 2018; Miri et al., 2017). The chronic daily intake (CDI), hazard quotient (HQ) of single contaminant and the health risk index (HI) for numerous contaminants are key parameters that can be used for the quantification of health risks (Keramati et al., 2018). In the current study, we quantified the non-carcinogenic risk as a result of nitrate intake in drinking water for three age groups, adults, children, and infants based on the non-carcinogenic hazard quotient model proposed by the United States Environmental Protection Agency (USEPA, 2013). The direct ingestion, skin contact, and inhalation are the viable routes of contaminants exposure in water (Liang et al., 2016; Wang et al., 2011).

The CDI (chronic daily intake) (mg/kg/day) nitrate level in water can be computed based on the following equation (Liang et al., 2013; Miri et al., 2018):

$$ CDI = \frac{C_w \times D_I \times E_F \times E_P}{B_W \times A_T} $$  

(1)

Where CDI is the average daily dose (mg/kg/day), Cw is the contaminant concentration in drinking water (mg/L), DI is human daily water intake (L/d), EF is the frequency of exposure (day/year), EP is the mean exposure duration (years), BW is the mean weight of the body (kg), and AT is the mean time (day). The different parameters used for the health risk assessment are given in Table 1.

The non-carcinogenic hazard quotients (HQ) risks are the ratio between the potential exposure to a contaminant and the level at which no negative effects are expected. The non-carcinogenic HQ risk can be quantified by the following equation (Lim et al., 2008; Radfard et al., 2018):

$$ HQ = \frac{CDI}{RfD} $$  

(2)

Where RfD is the reference nitrate dosage in mg/kg/days which is equal to 1.6 mg/kg/days based on the USEPA (2013).

According to the USEPA regulations, HQ<1 represent unlikely harmful non-carcinogenic health impact by nitrate to the individual, whereas HQ≥1 denote a harmful impacts may occur (Huang et al., 2018; Jafari et al., 2018).

**Results and Discussion**

**Concentrations and Origin of Nitrate in Wells**

With a growing population, many countries like Egypt need adequate quantities of acceptable quality water for sustaining, human well-being, livelihoods and socio-economic development. Pollution is becoming one of the main threats to the availability of water resources. Improper management due to lack of financial limits and recommendations from the scientific communities in developing countries lead to severe problems in drinking water quality that needs detailed understanding to protect groundwater resource. Egypt is characterized by arid climate; water resources in many areas are scarce and depend mainly on the groundwater. Many factors are affecting the groundwater aquifers due to the overexploitation, extensive fertilizers and improper treated wastewater disposal (Shamruk et al., 2001; Ahmed and Ali, 2011; Redwan et al., 2016).

Nitrate in groundwater may result from sewage disposal, livestock, agricultural fertilizers, parks, lawns, gardens and natural occurring nitrogen sources (Nemčić-Jurec and Jazbec, 2017; Wei et al., 2017) (Fig. 3). The parameters $\text{Cl}^-$, $\text{SO}_4^{2-}$, $\text{Ca}^{2+}$ and particularly $\text{NO}_3^-$ are precisely the significant ions of nitrogen fertilizers which include ammonium chloride ($\text{NH}_4\text{Cl}$), ammonium sulfate ($\text{NH}_4\text{SO}_4$), ammonium nitrate ($\text{NH}_4\text{NO}_3$) and calcium nitrate ($\text{Ca} (\text{NO}_3)_2$) (Wei et al., 2017). NH$_4^+$ ions are difficultly preserved in soils, some of its ions are absorbed by plant roots and soils, and others are transformed into NO$_3^-$ by nitrification (Mekala and Nambi, 2016), as shown in Fig. 3.

The bacteriological impacts on groundwater samples showed a positive indication of the presence of fecal coliform bacteria (52% of samples) (Fig. 4) started from the disposal site and extended northeast about 2.0 km following the local topography, except for wells no. 1, 2,

| Risk exposure factors | Values for groups |
|-----------------------|-------------------|
| $C_w$                 | Adults (age>19)   |
|                       | Children (6>age>12)|
|                       | Infants (age<1)   |
|                       | Unit              |
| DI                    | 2                 |
|                       | 1.5               |
|                       | 0.8               |
|                       | mg/L              |
| $F$                   | 365               |
|                       | 365               |
|                       | 365               |
|                       | L/day             |
| EP                    | 40                |
|                       | 10                |
|                       | 1                 |
|                       | years             |
| BW                    | 70                |
|                       | 20                |
|                       | 10                |
|                       | kg                |
| AT                    | 14,600            |
|                       | 3650              |
|                       | 365               |
|                       | days              |
The nitrate concentrations showed wide spatial variability in farm wells (23 wells) of the studied area, with values going from 0.38 up to 59 mg/L with a mean concentration of 24.8 mg/L. In the current study, it is clearly recognized that all the investigated farm wells had nitrate values beneath the EPA standard for nitrate, except one site with a nitrate concentration of 59 mg/L (Figs. 4 and 5).

Cl⁻ is a good indicator of sewage contamination of groundwater as it is not dependent on the physical, chemical and microbiological processes (Fig. 6) (Xing and Liu, 2016; Wei et al., 2017; Meghdadi and Javar, 2018). Cl⁻ from chemical fertilizers commonly come with a pregnant increase in NO₃⁻ concentrations, whereas higher Cl⁻ concentrations

Figure 3. Scheme for nitrogen cycle and transformation (modified after Wei et al., 2017).

Figure 4. Aerial view and potential NO₃⁻ pollution sources. The signs +/− indicate the presence/non-presence of coliform bacteria.

(Figs. 4 and 5).

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with relatively low NO$_3^-$ often indicate domestic sewage and industrial wastewater as well as livestock effluent. As a result, the ratio of NO$_3^-$/Cl$^-$ (Fig. 6) can offer detailed information for the study of NO$_3^-$ sources in groundwater (Chen et al., 2009; Xing and Liu, 2016). Groundwater samples with high Cl$^-$ contents and low NO$_3^-$/Cl$^-$ ratios were primarily polluted by sewage from the wastewater plant or from livestock effluent. The highest ratios were around the wastewater treatment plant. Groundwater samples with high NO$_3^-$ and low Cl$^-$ concentrations may relate to the agricultural input. Figs. 1 and 4 also show the study area classified into the agriculture land, reclaimed land and the desert zone. By combining the distribution of NO$_3^-$ concentrations of groundwater samples, it can be concluded that the samples with high concentrations were mainly distributed toward the reclaimed and the agricultural land and desert zone around the wastewater treatment plant.

This is indicated by using the principal component analysis (PCA) (Fig. 7). The first two eigenvectors and the loading plot of the varimax rotated component are depicted in Fig. 7. The first two PCs, 33.3% and 32.9%, covered more than 60 percent of the total variance in data. The first loading value showed the highest correlation between Cl$^-$, SO$_4^{2-}$ of 0.815 and 0.775 respectively. The concentration of SO$_4^{2-}$ is relatively limited compared with Cl$^-$ due to its small solubility (Wei et al., 2017). However, Cl$^-$ and SO$_4^{2-}$ typically increases in groundwater as a result of the application of chemical fertilizers (McQuillan, 2004; Mekala and Nambi, 2016). Other sources of Cl$^-$ include manure slurry contamination, e.g. from dairy farmlands (McQuillan, 2004) and leaching of marine Pliocene clay that form the base of the aquifer (Redwan and Abdel Moneim, 2016). In addition, SO$_4^{2-}$ ions can originate from gypsum and/or anhydrite dissolution. HCO$_3^-$ showed high negative loading that refers to the water–rock interaction due to carbonate-silicate rock dissolutions in the study area aquifer (Redwan and Abdel Moneim, 2016b). The high independence of Cl$^-$ on nitrate with a negative loading of nitrate (-0.051) indicate different sources in groundwater (Fig. 7). The second loading value of the highest positive value of NO$_3^-$, the negative NH$_4^+$ loading value of -0.831 could be described by the natural attenuation process of nitrate through denitrification process in groundwater contaminated by sewage effluents (Rivett et al., 2008). From the data it can be concluded that nitrate in the south-western parts around the wastewater treatment plant developed from the sewage contamination and the north-eastern and southern parts due to contamination from the agricultural activities and organic wastes (animal manure) (Fig. 4).
**Human Health Risk Assessment**

**Estimation of Chronic Daily Intake (CDI)**

So far, no investigation was carried out in the studied area to estimate the toxic load induced by the water contaminant, especially the nitrate non-carcinogenic impacts in groundwater. To quantify the health risk in the groundwater samples, nitrate was used to estimate the non-carcinogenic risk. Nitrate transformation into nitrite after ingestion (Swann, 1975) is the main cause of toxicity as nitrite can oxidize hemoglobin to methemoglobin, which cannot bind oxygen. If oxidation overthrows the protective reduction capacity of the cells, methemoglobin accumulation (methemoglobinemia) will exist (Jaffe, 1981). Drinking water represents about < 3–21% of the average adult intake of nitrate, other nitrate sources include vegetables and meat in the form of sodium nitrite (Jaffe, 1981). Also, nitric oxide can be oxidised in several dietary products into nitrate (Wogan et al., 1995). Higher dietary nitrate consumption increases the nitrate formation in case there is simultaneous exposure to nitro-satable drugs, which have nitrite substrate (Brender et al., 2004).

The CDI nitrate data and the calculated HQ values in groundwater from the investigated area via ingestion contact route were shown in Table 2 and Fig. 8, respectively. The CDI values for groundwater samples were ranged as 0.01–1.69 (mean 0.74), 0.03–4.43 (mean 1.94) and 0.03–4.72 (mean 2.07) mg/kg/day for adults, children, and infants, respectively. The significant exposure ways for nitrate in groundwater exist through ingestions.

**Estimation of Non-carcinogenic Risk Levels of Nitrate**

Hazard quotient (HQ) analysis model was used to appraise the non-carcinogenic risk in the groundwater of the study area based on the US Environmental Protection Agency methodology (USEPA, 2013). Adults, children and infants age groups were used to quantify the HQ due to ingesting the nitrate from groundwater.

The HQ values in the investigated area were in ranges of 0.01–1.05, 0.02–2.77 and 0.02–2.95 with mean values of 0.44, 1.16, and 1.24, for adults, children and infants, respectively. Generally, HQ values higher than 1 are considered of harmful health impacts, whereas values less than 1 are considered to be safe. The HQ values of 78% of infants, 70% children and 4% of adults were higher than the safe level (i.e., HQs >1), indicating that nitrate in groundwater would have serious health effects on these ages. Infants are obviously the most vulnerable age group to nitrate exposure.

Over the past years, various studies have used nitrate for groundwater contamination assessment. Qasemi et al. (2018a) identified in arid and semi-arid areas in villages of Azadshahr, northeastern Iran about 7% of the villages exceeds the EPA standard concentration for nitrate. In Prince Edward Island, Canada, 6% of domestic wells showed nitrate concentration higher than the WHO guideline (Savard, 2016); in Jinghui canal irrigation area of the loess region, northwest China, 50% of the groundwater samples crossed the drinking water standard for nitrate (Zhang et al., 2018).

Due to the increased population in developing countries, especially in arid and semi-arid areas, the excessive dependence on groundwater resources by local communities in villages for their daily water needs is enormously increased. Thereto, disposal activities and domestic sewage in the contaminated villages and intensive agrochemicals activities by using nitrogen fertilizers after the construction of the Aswan High dam and pesticides leached downward into

| S. No. | CDI adults (mg/kg/day) | CDI children (mg/kg/day) | CDI infants (mg/kg/day) |
|--------|------------------------|--------------------------|-------------------------|
| 1      | 0.91                   | 2.39                     | 2.55                    |
| 2      | 0.75                   | 1.97                     | 2.10                    |
| 3      | 0.65                   | 1.71                     | 1.83                    |
| 4      | 1.01                   | 2.64                     | 2.82                    |
| 5      | 1.01                   | 2.66                     | 2.83                    |
| 6      | 0.34                   | 0.90                     | 0.96                    |
| 7      | 0.57                   | 1.49                     | 1.58                    |
| 8      | 0.73                   | 1.92                     | 2.05                    |
| 9      | 0.57                   | 1.49                     | 1.59                    |
| 10     | 0.66                   | 1.73                     | 1.84                    |
| 11     | 0.50                   | 1.31                     | 1.39                    |
| 12     | 0.74                   | 1.95                     | 2.08                    |
| 13     | 0.64                   | 1.69                     | 1.80                    |
| 14     | 0.83                   | 2.18                     | 2.32                    |
| 15     | 0.89                   | 2.33                     | 2.48                    |
| 16     | 0.66                   | 1.73                     | 1.84                    |
| 17     | 1.06                   | 2.78                     | 2.96                    |
| 18     | 0.80                   | 2.10                     | 2.24                    |
| 19     | 0.57                   | 1.50                     | 1.60                    |
| 20     | 0.54                   | 1.43                     | 1.52                    |
| 21     | 1.69                   | 4.43                     | 4.72                    |
| 22     | 0.01                   | 0.03                     | 0.03                    |
| 23     | 0.89                   | 2.33                     | 2.48                    |

*Figure 8. Hazard quotient (HQ) values for nitrate in groundwater samples collected from the studied area.*
shallow groundwater by the infiltrated irrigation water, wastewater disposal activities and domestic sewage. The NO$_3^-$ from wastewater and agricultural fertilizers diffused slowly in the shallow groundwater system due to the low permeability of subsurface strata and scarcity of raining events. Due to the slow average rate of infiltration and the recent age of the wastewater treatment plant only the shallow groundwater aquifers will be affected and the deeper aquifers require long time to response to changes at the surface regarding wastewater disposal and agricultural practices. This is evidenced by Huang et al. (2013) that there was a time lag up to hundreds of years between land use changes and groundwater quality response, due to the buffering capacity of the relatively thick unsaturated zone. With time, the area of NO$_3^-$ pollution will expand in the shallow aquifers. Therefore, effective policies to prevent, remediate and monitor the non-point source contamination of nitrate are required. The current work needs collaboration from the medical science sectors to link the groundwater contamination to the actual community diseases due to groundwater contamination. Future research may involve continuous monitoring of groundwater that is vulnerable to various contaminations to provide useful assessment information to the authorities.

Conclusions

In the area West of Tahta, Sohag, Egypt, the nitrate concentrations were below the maximum permissible concentrations of 45 mg/L proposed by the USEPA, except in one site. Groundwater samples showed a positive indication of the presence of fecal coliform bacteria (52%) started from the disposal site and extended northeast following the local topography. The nitrate concentrations showed wide spatial variability in farm wells of the studied area, with values going from 0.38 up to 59 mg/L with a mean concentration of 24.8 mg/L. From the bacteriological, principal component, NO$_3^-$, NO$_2^-$/Cl$^-$ data evaluation, it can be concluded that nitrate in the south-western parts around the wastewater treatment plant developed from the sewage contamination and the north-eastern and southern parts due to contamination from the agricultural activities. The HQ values of 78% of infants, 70% children and 4% of adults were higher than the safety level (i.e., HQ$>1$), suggesting severe health effects on human health. Wastewater disposal activities and application of fertilizers are main the sources of nitrogen in waters in the area West of Tahta, which require proper management. It is prohibited to use untreated wastewater to irrigate the croplands in order to reduce the risks of harmful effects on health.

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