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Research Article

Risk Assessment of Recycled and Treated Water Polluted with Nitrate in Suburb Population

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Abstract
Groundwater is one of the sources used for drinking water. During the treatment process of nitrate, the efficiency of its removal should be carefully considered. This study was conducted to investigate the health risk of nitrates present in the treated wastewater of an oil facility mixed with well water for reuse in a residential area. To assess the risk of treated wastewater, the amount of nitrate in wastewater was sampled once every two weeks for a year and finally, 25 samples (collected in triplicates) were analyzed. Also for the microbial quality of water, the heterotrophic plate count was carried out. The average measured nitrate concentration in the samples is equal to 63.3 mg/L and the maximum measured concentration is equal to 134.7 mg/L. The nitrate risk assessment method was selected based on EPA standards. The results showed that the level of non-cancerous risk of Hazard Quotient (HQ) in 84% of the samples for infants was above one (HQ ≥ 1) and consumption of this water can be unsafe. But for the rest of the group, the water was safe to use even for a long time. For the Heterotroph Plate Count (HPC), the results showed that in one sample the number of bacteria was above the standard. Sensitivity should be considered in the efficiency of nitrate removal in the treatment plant system for the use of infants or another source of water should be used for consumption.
Keywords: Drinking water, nitrate, Risk assessment, non-carcinogenic.

Introduction

Water is an essential element for the life and survival of living organisms and one of the most important natural resources in the world. With the development of urbanization and industrialization of societies, water consumption is also increasing rapidly (Hamdi. 2022a; Derakhshan et al. 2018). However, the limitation of good quality water resources has caused the lack of water to be an important factor in the economic growth of societies. In addition to the limited water resources available in the world, the problems related to water pollution have also become an important environmental challenge today (Rajab Beigy et al. 2018; Mofradnia et al. 2019). Hence, in many developing countries, the availability of safe and sanitary water is a major concern Hamedi et al. (2022).

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Nitrate, nitrite, and fluoride are among the common inorganic pollutants of underground water sources that affect the non-carcinogenic health risk in the consuming population (Golaki et al. 2022). One of the most common chemical pollutants in water sources in most parts of the world is nitrate (Samaei. 2022), and if its presence in water sources exceeds the permissible limit, it can restrict the use of these sources (Moloantoa et al, 2022).

The most important sources of nitrate entering the groundwater include the disposal of wastewater in absorption wells and the discharge of wastewater from urban sewage treatment plants into the environment (Rezaei et al. 2019), the excessive use of chemical fertilizers and pesticides in agriculture (Mateo-Sagasta et al. 2017), and leachate leakage in sanitary landfills (Niu et al. 2022). In addition, one of the important challenges in the quality of groundwater is the existence of various industrial facilities in the suburbs of big cities and the leakage of their pollutants into these water sources.

All over the world, there are restrictions on the consumption of drinking water containing nitrates. The World Health Organization, the European Union, as well as the Iranian Environmental Protection Organization, have set the maximum permissible nitrate in drinking at 50 mg/L, and the American Environmental Protection Organization has determined 10 mg/L of nitrate (Darvishmotevalli et al. 2019). The permissible amount of nitrate in drinking water for the Environmental Protection Agency (EPA) is reported as 45 mg/L (Schullehner, et al. 2017).

Consuming water with a high concentration of nitrates causes systemic problems in humans, especially in children and infants (due to the higher ratio of water volume to their body weight). As a result of the activities of bacteria in the digestive system under anaerobic conditions, nitrate is reduced to nitrite (Ward et al. 2018). The absorption of nitrite by the blood causes a disturbance in the formation of hemoglobin, the production of methemoglobin, and a decrease in the oxygen-carrying capacity of red blood cells, and ultimately causes blue baby syndrome (methemoglobinemia) and even the death of infants (Golaki, 2022; Ward et al., 2018). But the effectiveness and side effects of drinking water containing nitrates are different for infants, children, and adults (Qasemi, 2018; Ward et al., 2018). The most likely ways for children to be exposed are through contaminated drinking water and food containing preservatives, such as cooked meats and sausages (Kumar and Puri 2012).

Another study evaluated the risks of carcinogenesis related to exposure to nitrates in drinking water in Iran (Darvishmotevalli et al. 2019). The obtained results indicated that the drinking water reserves of the cities of Tehran, Mashhad (Khorasan Razavi), Zahedan (Sistan and Baluchistan), Shiraz (Pers), Qom, Ardabil, and Ahwaz (Khuzestan) have nitrate concentration higher than the recommended limit by the World Health Organization and Institute of Standard and Industrial Research of Iran (ISIRI).

In northeastern Iran fifty-eight samples of groundwater were collected from wells and springs and analyzed by Qasemi et al. (2018). According to the obtained results, visible changes in the nitrate concentration were observed with the change in sampling location. Also, the results of the health risk assessment including HQ values for 41% of children and infants were above the safety level, which indicates the sensitivity and high health risk for infants and children. Similarly, Radfar et al (2018) evaluated the health risk of nitrates in drinking water in rural areas of Khash. The results showed that the concentration of nitrate in drinking water varied from 6 to 35 mg/L (average 16.083 mg/L) and the average HQ values in 4% of the samples in the age groups of infants, children, teenagers, and adults are above the standard. Golki et al (2022) analyzed the health risk and spatial distribution of nitrate, nitrite, fluoride, and coliform pollutants in the drinking water sources of Kazeroon, Iran (Golaki et al. 2022). In this study, 25 groundwater wells were investigated. The maximum concentration of nitrate was reported as 25.5 mg/L and the average concentration of the samples was 13.5 mg/L. HQ coefficient values of nitrate pollutants were below one in all age groups. Finally, the results show a higher effect of nitrate compared to nitrite and fluoride in the health risk of age groups.

The health risk assessment related to fluoride, nitrate, and nitrite pollutants in drinking water in Sanandaj city, Iran was examined. The number of samples collected in the studied area was 106 samples. The maximum concentration of nitrate reported in this study was 80 mg/L and the average CDI of nitrate in men, women, and children was 0.4258, 0.5110, and 1.1454, respectively. According to the results, all three studied groups were exposed to the risks of nitrate contact, and HQ was higher than one in each of the three groups, which indicated the need for preventive and corrective measures in the drinking water system of the region (Rezaei et al. 2019).

The water quality has to be suitable for drinking and other usage for household chores, according to WHO. For analyzing the microbial quality of drinking water, the heterotroph bacteria of the water have to be monitored. Heterotrophic Plate Count (HPC) is an indicator of the presence of bacteria in water (Falcone-Dias and Farache Filho 2013). The standard level of heterotrophic bacteria in drinking water is 500 CFU/ml. The amount of bacteria above this level is not safe for drinking. Studies show that even in waters with a little amount of organic substance the growth of heterotrophs is possible (Yari et al. 2018).

Research on the quality of drinking water in Sari, Iran showed that 25 % of samples were HPC positive and 12 % of the samples were above the standard level (Gholami-Borujeni et al. 2021).

Studies show nitrate is one of the main challenges of drinking water quality in Iranian cities, and the necessity of systematic and periodic reviews to ensure the quality status of pollutants and the level of risk caused by them to the health of citizens is essential. The purpose of the study was to investigate the nitrate pollution risk and microbial quality of semi-industrial water effluent treatment plant on the health of the nearby residents in the suburbs of a city. Few studies had demonstrated the treatment plant process and the use of a mixture of well water (groundwater sources) and the sanitary sewage effluent of the oil industry adjacent to the residential settlement.

Material and Methods

Area of Study

The studied treatment plant is located in the suburbs of a city, where two types of incoming water sources (sanitary sewage from the nearby oil industry and underground well water) are treated in the treatment plant. Some operational conditions of the plants include a 90 cubic meters per day flow rate of sanitary wastewater into the treatment plant whereas the flow rate pumped from the groundwater well is 170 cubic meters per day. the main operating units used in the refinery process were:1. Water, sludge, and oil separation unit. 2. Neutralization pond (injection of 10% sulfuric acid), pH regulation, and aeration. 3. Unit for removal of oil emulsion and sludge removal. 4. Garbage collector and settling pond. 5. Biological aeration tank. 6. Aerobic digester tank and sludge dewatering package. 7. Chlorination unit.

The residential settlement adjacent to the refinery has a population of around 2000 people, and in this study, the analyzed statistical sample is 10% of the total population (Ahmad, 2021; Butu et al., 2020), equivalent to 200 people.

The information from the statistical sample was collected and used during field visits and conducting interviews and completing the questionnaire. It is worth mentioning that due to providing information on the quality results of treated water and wastewater and nitrate risk assessment and the heterotrophic analysis, for the neighboring residential area and the possibility of some misinterpretation of the results of the study, the location, and name of the treatment plant is not mentioned.

Nitrate risk assessment

This component investigated the nitrates risk assessment in the treated wastewater containing a mixture of groundwater and an oil industry sanitary wastewater. Firstly, the amount of final nitrate in the effluent of the treatment plant was measured, and assuming its use as a source of drinking water for the nearby residential area, a health risk assessment was made for the target statistical population. According to the EPA standard, human health risk assessment consists of four main steps namely risk identification, dose-response evaluation, exposure assessment, and risk assessment (Rezaei et al. 2019), adopted in this study to calculate and identify the health risk of nitrate abundance in treated water.

Sampling and measuring of nitrate in the effluent

To measure the concentration of nitrates, sampling was done monthly from the outlet of the refinery treatment plant for one year. Sampling and analysis were done using the standard method of water and wastewater 4500-$NO3^-$. The samples were transferred to the laboratory in plastic containers at a temperature of 4°C and within 24 hours the amount of nitrate was measured by a spectrophotometer at a wavelength of 220 nm (Moeini and Azhdarpoor (2021).

Nitrate health risk assessment model

With the increase of pollutants in water and the potential impact on consumer health, a health risk assessment was carried out. The method used to perform the health risk assessment was based on the non-carcinogenic risk factor model proposed by the United States Environmental Protection Agency (USEPA 2013) (Adimalla, 2020; Qasemi et al., 2018). The prediction of nitrate pollutant risk in the drinking water of a town with a population of about 2000 people was investigated. The amount of non-carcinogenic was calculated for the age group of infants (less than two years old), children (two to ten years old), adolescents (ten to eighteen years old), and adults (over eighteen years old). The evaluation criteria assess the non-carcinogenic risk of nitrates in water. In order to evaluate the risk assessment results, the weight of 200 people was measured. In this sample, 17 people were under 2 years old; 29 people were between 2 and 10 years old, 42 people were between 10 and 18 years old and 112 people were over 18 years old.

To calculate the Chronic Daily Intake (CDI) coefficient, the necessary parameters for four age groups are specified in Table 1. The amount of daily water consumption is calculated from the average weight of people. The number of years exposed to nitrates is also considered based on the average age of people (Moazeni et al. 2014).

Table 1. Nitrate risk assessment parameters and values are taken from the statistical sample of the residential settlement under study

Parameter	Unit	Infant (less than 2 years)	Child (2 to 10 years)	Teenager (10 to 18 years old)	Adult (above 18 years)
DI	L/day	0.61	1.25	1.58	1.95
BW	kg	7.9	16.4	39.8	77.4
RfD	mg/kg.day	1.6	1.6	1.6	1.6
EP	year	1	6	14	35

The HQ and CDI approaches were used to assess the noncarcinogenic health effects of nitrate, in drinking water. To determine the exposure to nitrate in drinking water, CDI (mg/kg/day) was first calculated using equation 1:

$$CDI = \frac{C_W \times DI \times EF \times EP}{BW \times AT} \qquad (1)$$

Where Cw is the chemical concentration in water (mg/L). DI is the average Daily Intake rate of water (l/d). EF and EP are Exposure Frequency (days/year) and Exposure Period (year), respectively. BW and AT are the Body Weight (kg) and Average Time (days), respectively (Su et al.2013). Table 1 shows the constant coefficients used in calculating CDI. To estimate the significant difference in exposure and final potential for noncarcinogenic health effects caused by nitrate concentration in water, the HQ was calculated using equation 2:

$$HQ = \frac{CDI}{RfD} \qquad (2)$$

Where the RfD is the reference dose (mg/kg/day). An HQ value of more than 1 (HQ > 1) will show a significant adverse noncarcinogenic health risk. Based on the USEPA, RfD used in this study for nitrate is 1.6 mg/kg/day (Alif Adham and Shaharuddin, 2014; Darvishmotevalli et al., 2019; EPA, 2002; Qasemi et al., 2018).

Heterotroph bacterial count

One of the methods used for microbial monitoring of drinking water quality is HPC. In this method, the cultivable heterotrophic bacteria are cultured and incubated and colony count is carried out. To analyze the bacterial quality of the water for use as drinking water for the suburban the sampling of the input and output of the wastewater and treated wastewater was done once every month for one year and 24 samples were collected and analyzed. The samples were preserved at 4ºC and transferred to the laboratory. HPC test was done using R_2A agar medium and the plates were incubated at 35ºC for 48 hours Yari et al. (2018).

Results and discussion

Investigating and assessing the risk of pollutants in water is very important for human life and human health. The possibility of pollutant transfer through the release of industrial pollutants to underground wells in its vicinity can pose risks to the health of the consumers. In this study, the amount of nitrate in the effluent of the treatment plant transferred to the well was evaluated every two weeks for one year. In total, the amount of nitrate in 25 samples (in triplicates) was measured. A risk assessment was carried out in order to reuse treated wastewater as drinking water for a nearby suburban population. The suburb in question had a population of 2000 people, and in this research 200 people were assumed. The amount of nitrate measured from the effluent of the refinery once every two weeks and for one year was sampled, analyzed, and reported.

The results showed that 58% of the samples have nitrate above the permissible limit. The highest amount of nitrate is 134.7 mg/L and the lowest amount is 9.9 mg/L. In Figure 1, the results of calculating the CDI coefficient for four different age groups are shown.

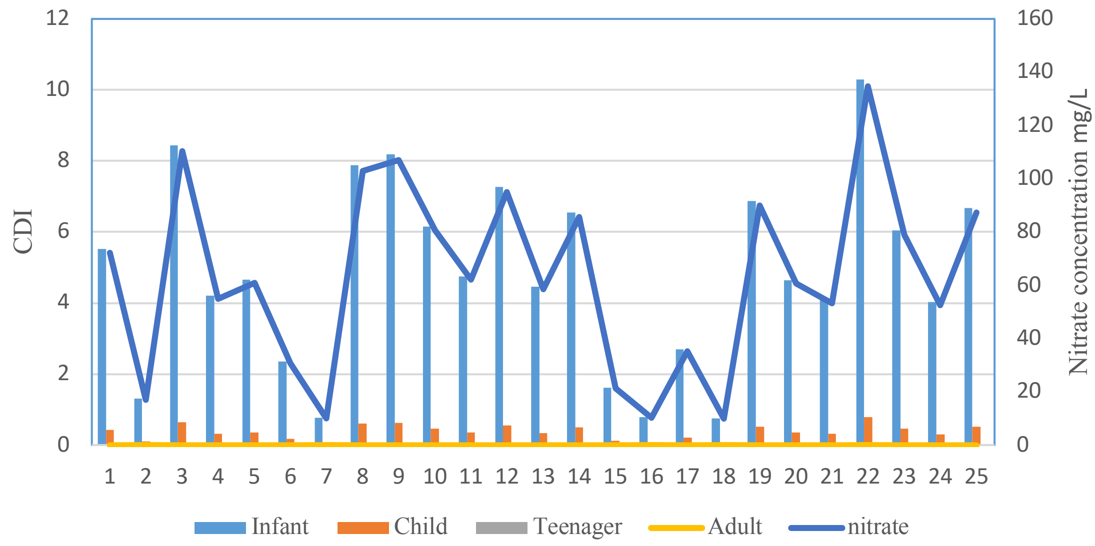

Figure 1. CDI coefficient according to the EPA standard

To compute the non-carcinogenic risk factor and the impact of the pollutant on human health, the HQ factor has been calculated, which is shown in Figure 2 for four age groups.

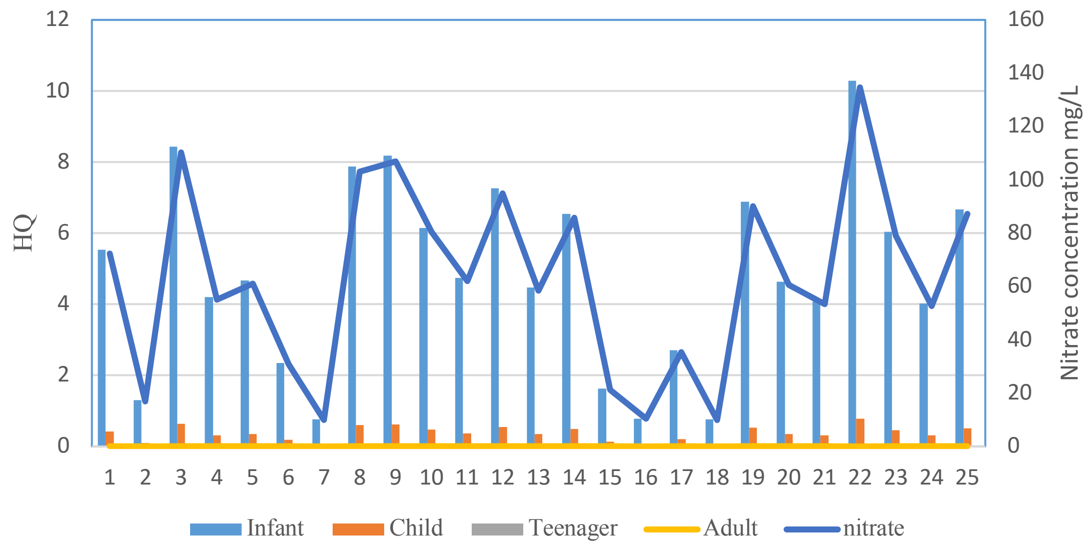

Figure 2. HQ amount for the four age groups

As it is known, most of the risk level is for the newborn age group. Due to the low weight of infants, they are more at risk compared to others (Huang et al. 2017). An increase in the amount of nitrate has a direct effect on the amount of HQ (Fallahzadeh RA et al. 2019). Due to the difference in the physiology and tolerance of pollutants, these age groups (infants, children, teenagers, and adults) show different effects. The difference in pollutant effects depends on the factors in the calculation formula (Zhai et al. 2017). The results obtained from the studies of Yari et al. in Iranshahr showed that the amount of HQ for infants is higher than for other ages (Shalyari et al. 2019). Children are at higher risk than adults ingesting pollutants such as nitrates and being exposed to pollutants per unit of body weight (Chen et al. 2016). Although most of the statistical samples of nitrate concentration in purified water for drinking consumption in the intended residential complex were higher than the standard, the HQ coefficient obtained for the age groups of children, youth, and adults was less than one, and only infants were at high risk.

Of course, it should be noted that the limits of concentration outside the standard have not had many intensities and the worst-case limit is double the standard limit.

To evaluate the risk of nitrates in water, the non-carcinogenic coefficient was calculated for four age groups. The results showed that the value of the HQ coefficient for the infants in most samples is higher than the permissible limit. In the age group under 2 years, in 20 collected samples, the HQ value was more than one, which shows that nitrate in water can have non-carcinogenic effects for this age group. According to the obtained results, the highest

Amount of HQ for infants is 6.43 and the lowest amount is 0.47. For other age groups, this coefficient is less than one. The second highest amount of HQ belongs to children, due to their greater exposure to pollutants. The highest value and the lowest value for children are 0.49 and 0.03, respectively. Adolescents and adults are ranked third and fourth so their risk levels are very low. Table 2 is a comparison of the amount of nitrate from a few studies carried out in different places.

Table 2. The average amount of nitrate in current work and comparison with other works.

Place of sampling	Average of nitrate (mg/l)	Sample description	No of samples	Reference
-	63.28	well and oil industry effluent	25	-
Nirmal Province, India	40	well	30	Adimalla and Qian (2021)
Wanbei, China	24.01	well	11	Hu et al. (2021)
Tadla, Morocco	24.73±15.49	well	21	Barakat (2020)
Zhangjiakou, China	29.72	well	489	Wang et al. (2021)
Tehran	36.15±14.74	drinking water	148	Kalteh et al. (2022)
West of Tehran	0.02±4.6	drinking water	-	Haji Seyed Mohammad Shirazi et al. (2021)
Hormozgan	7.37±5.61	well	54	Mohammadpour et al. (2022)
Robat Karim	2.1 2.05	drinking water well	40 (well) 32(drinking water)	Panahi and Alavi Moghaddam (2012)

Heterotrophic plate count results

The results for the heterotrophic plate count for the assay of the quality of effluent were carried out for one month and the result is shown in table 3.

The results show that the number of heterotroph bacteria in most of the samples is below 500 CFU/ml. In 33 % of the samples, the heterotroph bacteria count was between 100-500 CFU/ml. Only one of the samples was above the standard level.

Table 3. Colony count of the heterotrophic bacteria.

No	Before treatment	After treatment
1	283	46
2	159	<30
3	168	<30
4	215	<30
5	420	145
6	360	75
7	652	510
8	490	230
9	540	367
10	138	<30
11	296	58
12	474	264

Conclusion

This study was conducted on the drinking water obtained from the treated water of an underground well in the vicinity of one of the oil facilities of a city. The health risk assessment of nitrates on the residents of the residential complex was studied. The findings showed that the nitrate concentration for the studied population is higher than the national (Iran) and international quality standards. The findings show that out of the four studied age groups exposed to nitrate (HQ > 1), only infants are at risk in most cases and the rest of the groups have a favorable condition. Even though the majority of samples of treated water for drinking consumption in the intended residential complex, nitrate concentration was higher than the standard, the HQ coefficient obtained for the age groups of children, teenagers, and adults was less than one.

Although the majority of statistical samples of nitrate concentration in treated water for drinking consumption in the intended residential complex were higher than the standard, the HQ coefficient obtained for children, adolescents, and adults was less than one. Probably, this issue can be due to the small distance of the measured nitrate concentrations compared to the standard limits, and the higher the intensity of nitrate concentration the higher the risk. Infants are more at risk due to consuming a higher amount of water and being exposed to pollutants per unit of body weight.

Finally, the performance of the studied treatment plant can be considered appropriate and the consumers of the treated water should consider consuming it for infants and should use alternative water sources with better quality for the infants. According to the results for other age groups (children, teenagers, and adults), the long-term use of this water will not have non-cancerous risks for the consumers, and its use will be safe for the people under study.

The result of HPC showed that in most of the samples analyzed after treatment, the amount of heterotroph bacteria was below the standard limit and only one of the samples was above 500 CFU/ml.

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