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Investigating Refinery Wastewater Impacts on Groundwater Sources – A Case Study of Pars Oil Refinery

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Abstract

Pars Oil Refinery, situated 22 km away from metropolitan Tehran, is one of the most important motor oil refineries in the country. Currently, the refinery is composed of various sections including the Furfural, Separation, Storage, Blending, Can-making, Barrel-making and Dyeing, Loading and Unloading, Antifreeze, Grease-making, Boiler, Repair, and Laboratory Units. In the study area, because of such reasons as the high groundwater table the company has failed to launch the biological section of its wastewater treatment plant. In addition, with the possibility of effluents from residents' sanitary wastewater as well as those from agriculture lands penetrating the soil and ending up in the underground water of the studied area, the present research was conducted on such an environmental dilemma. In order to investigate the hypothesis of effluent penetration into underground water, four stations were selected from among water wells existing throughout the area, and then the parameters of pH, chemical oxygen demand (COD), biological oxygen demand (BOD), nitrate, colour, nitrite, electrical conductivity (EC), sulfate, dissolved solids, ammonia, furfural, heavy metals, and total petroleum hydrocarbon (TPH) were analyzed. It is noteworthy that the analyses were conducted on water samples seasonally from December 2006 to May 2009. Afterwards, through comparison of the results obtained from the forenamed tests and using standard values, the research hypothesis was confirmed. In other words, effluents do penetrate the wells and pollute the underground water.

Keywords: Oil refinery, Water resources, Effluent, Furfural.

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چکیده

پالایشگاه نفت پارس در فاصله ۲۲ کیلومتری شهر تهران در نوع خود یکی از مهم‌ترین پالایشگاه‌های روغن موتور کشور محسوب می‌شود. این پالایشگاه، در حال حاضر از واحدهای فورفورال، واحد تفکیک، واحد مخازن، واحد بلندینگ، واحد قوطی سازی، واحد بشکه سازی و رنگ زنی، قسمت تخلیه بار و بارگیری، واحد ضدیخ، واحد گریس سازی، واحد دیگ‌های بخار و واحد تعمیرات و واحد آزمایشگاه تشکیل شده است. در محدوده مورد مطالعه دلایلی نظیر بالا بودن سطح آب‌های زیرزمینی، شکست کمپانی در راه اندازی بخش بیولوژیک سیستم تصفیه پساب و همچنین امکان ورود پساب‌های بهداشتی خانگی و نیز آب‌های کشاورزی که امکان نفوذ به درون خاک و راه‌یابی به آب‌های زیرزمینی در محدوده مورد مطالعه دارند، این تحقیقات در همین شرایط زیست‌محیطی صورت پذیرفت. برای بررسی این فرض که پساب‌ها به آب‌های زیرزمینی راه خواهند یافت، چهار ایستگاه روی چاه‌های آبی انتخاب شد که در تمام منطقه مورد مطالعه وجود داشتند. سپس پارامترهایی مانند pH، COD، BOD، نیترات، رنگ، نیتریت، رسانایی الکتریکی، سولفات، جامدات محلول، آمونیاک، فورفورال، فلزات سنگین و هیدروکربن‌های نفتی (TPH) اندازه‌گیری شدند. لازم به ذکر است که این اندازه‌گیری‌ها به صورت فصلی انجام گرفته و از دسامبر ۲۰۰۶ تا می ۲۰۰۹ ادامه پیدا کرده است. سپس با مقایسه نتایج حاصل از تست‌های مورد اشاره و استفاده از مقادیر استاندارد مربوط به این پارامترها، فرضیه مورد نظر اثبات گردید. به عبارت دیگر، جریان‌های پساب وارد چاه‌های آب شده و آب‌های زیرزمینی را آلوده می‌نماید.

کلمات کلیدی: پالایشگاه روغن، منابع آب، فورفورال، مواد زائد.

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Introduction

Use of resources such as oil and gas is a very important issue. As is evident to everyone, water and soil are vital components of life on Earth (Echenfelder, 2000). Therefore, such critical elements need to be managed so that additional pollutants are reduced to a minimum (Adames and Jenson, 1999). Due to the high economic importance of refineries, their environmental effects should be carefully assessed (Jenkins, 2001). The effluents of oil refineries contain various chemicals at differing concentrations (Lian, 2005). It is impossible to generalize the exact composition of such effluents because of factors such as the refinery itself as well as the kinds of units in operation at any specific time. Chemicals in the effluents include ammonia, sulphides, phenol, and hydrocarbons. Toxicity tests confirmed the toxicity of most refinery effluents in varying extents (Chin and Kaya, 2001).

The pollution of groundwater, surface water, and air by hazardous and toxic chemicals is considered a major problem of the industrialized world (Rajasulochana, 2009). The handling of large volumes of materials as part of the refining process has caused spills and, as a consequence, led to widespread contamination. Leakages from underground storage tanks and other storage areas on refinery sites have resulted in the contamination of soil and groundwater (Steven, 1996). The distribution of copper, chromium, lead, and zinc in water and sediments was studied in Skeleton Creek, Oklahoma, following conditions of high discharge and after an oil refinery cleanup operation (Harold, 1976). The cytotoxic effects of refinery wastewater collected

from different stations on the Kizilirmak River affecting *Vicia faba L.* root tip cells were evaluated by Wake in 2004 and the heavy metal concentrations in the water samples were determined using an Atomic Absorption Spectrophotometer (AAS) (Wake, 2004). A serious threat posed by oil-related pollution is the impact it has on underground water. In the event of oil spills, effluent discharge, or acid rain, the pollution seeps into the ground and mixes with the underground water (Legborsisaro, 2007). An additional pollutant from the refinery which will impose long-term environmental impacts on underground and surface waters is outgoing sewage (John, 1998). Over a 12-year period (1987–1998), the ground water, treated wastewater, and soil at the agricultural farm on the premises of the Mathura Oil Refinery were monitored for various physical-chemistry characteristics, (Saeedi and Ahmad, 2002).

The importance of the aforementioned problems prompted the authors of the current study to research the Pars Oil Refinery system which experiences some of these conditions, such as the residential environment of the refinery surroundings, wide gardens, and farms. It is necessary to mention that no research has yet been done regarding the industrial wastewater impacts of Pars Oil Refinery on the surrounding environment. Since other industrial units are not active and throughout the study area there are only residential homes and, farther on, agricultural lands, the current study was aimed at determining the impacts of the wastewater output of Pars Oil Refinery on underground water resources which supply drinking water for the residents.

In this study, a literature review was initially conducted to determine the importance of the issue around the world. Reducing the consumption of raw materials and energy, increasing the efficiency in energy consumption, and protecting water and soil resources can be pointed to as the most important goals of this project. By performing a literature review of internal research, a civil engineering master's thesis (Esfehani, 1997) was considered an appropriate basis for this respect. Esfehani had examined the quality of the untreated wastewater at Shahid Hasheminejad Gas Refinery. Consequently, the most important pollutants of this refinery were recognized as hydrogen sulfide, oil material, and diethanolamine. In this study, qualitative and quantitative surveys of wastewater were performed during normal operation and under annual maintenance conditions. The capability of physical-chemical and biological treatments of sanitary and industrial wastewaters collected in the pilot was studied. Finally, using pilot investigations, the wastewater treatment system for the gas refinery was designed.

Materials and Methods

The Study Area

Ghods City is situated at a latitude of 35°43' N and a longitude of 51°07' E. It is regarded as one of the Shahryar ensemble of cities in Tehran: Shahryar, covering an area of about 1,329 km², is located west and southwest of Tehran. It extends from the North to the Tehran-Karaj highway, from the East to Tehran, from the South to the Tehran-Saveh highway, and from the West, it leads to Karaj. Ghods City, with an area of 1,800

ha, is located on the northeastern edge of Shahryar, near the major Tehran-Karaj highway. Its average altitude is 1170 m above sea level, and its distance from Tehran is approximately 18 km.

Identification of Pars Oil Refinery

Pars Oil Company was founded in 1958 and promptly established a refinery in Ghods City, which is located 22 km from Tehran. The refinery first delivered its oil-products to the market in 1963. Pars Oil Company's current products include engine, industrial, and marine oils, lubricants, paraffin, anti-freeze, and brake fluid. The company has produced marine oils since 1994 in collaboration with Shell International Company.

The refinery's sewage is classified into three major categories, each having its own separate channels:

- A) Waste containing high soluble salts (High TDS) which includes the cooling towers' underwater boiler and the sewage from resin renovation with a salt solution.
- B) Solvent sewage containing furfural solvent, methyl ethyl ketone, and toluene.
- C) Oil sewage containing the sewage from unit washing, restaurant sewage, and rain water.

It is noteworthy that sewage, with high concentrations of soluble salts which are free of organic material contaminations, is directly channelled outside the refinery. Solvent and oil wastewater are initially mixed together in API ponds and these materials are driven by paddles moving on the surface and floor of the API.

Oily light materials are accumulated in cement ponds and pumped to the related reservoir on the boiler tank farm. Other parts are

accumulated in sludge ponds near the API and transferred to sand filters by means of pumps. The water then returns from the sand filter to the west API, and the sludge which accumulates in the sand filter is discharged at regular intervals. At this stage, free oil is separated from the sewage. A DAF system is used to separate emulsified oil. After crossing the API pond, the effluent is sent to balancing tanks by pumps. After reaching uniformity in the balancing tanks, the effluent is directed to the DAF system by effluent pumps. The light materials which accumulate on the surface are collected by a Piral scoop and returned to the API pool. The purified water is then discharged to the refinery output water channel.

The Research Workflow

In this research, the quality and quantity of the sewage flowing out from different treatment sections of Pars Oil Refinery were examined. The refinery sewage can be generally categorized into different groups, each of which flows through a separate channel. Sewage with a high concentration of soluble minerals includes the under-waters of the cooling towers and boilers, while sewage from the reclamation of resins via sodium chloride contains no organic substances, and is directly discharged. This also includes solvents sewage (furfural) and greasy sewage (resulting from washing the units, restaurant sewage, and rain). Two stations were selected on the open-top channel and two potable water wells on the transfer path were sampled as seen in Figure 1. The sampling sites were determined on the basis of the sewage path, the groundwater table, and the probability of sewage penetration

into the soil and groundwater in the vicinity of the residential and agricultural land. The parameters of pH, COD, BOD, nitrate, colour, nitrite, conductivity, sulphate, soluble solids, ammonia, furfural, heavy metals, and total petroleum hydrocarbons (TPH) in the water samples were tested on a seasonal basis (once every each three months) from December 2005 till May 2007. To conduct the above-mentioned tests, the sewage was pumped in polymer containers through four stages every half hour.

After sampling, the aforementioned tests were performed once at half hourly intervals and the sewage was pumped in four stages in polymer containers in the compound. In order to test the furfural, the sewage was pumped into glass containers sealed with paraffin. The samples were transported to the laboratory at a temperature of -2°C. All the containers were washed with distilled water before and after sampling.

It should be mentioned that guidelines in "Standard Method" (1992) were applied to the forenamed tests (see: Table 1). Furthermore, the densities of TPH were measured according to the method described by Moupam (1999). SPECTRA AA-200 and Gas Chromatography (GC) were applied to measure heavy metals and TPH, respectively. The pH of the samples was determined using a Horoba pH meter. To examine the changing trends at the four sampling stations, some comparative graphs were used for the pollutant parameters including COD, BOD, nitrate, colour, ammoniac nitrogen, and heavy metals such as Zn, Cd, Pb and Fe (EPA, 2002). The aforementioned tests were carried out once an hour during four phases in a compound pattern.

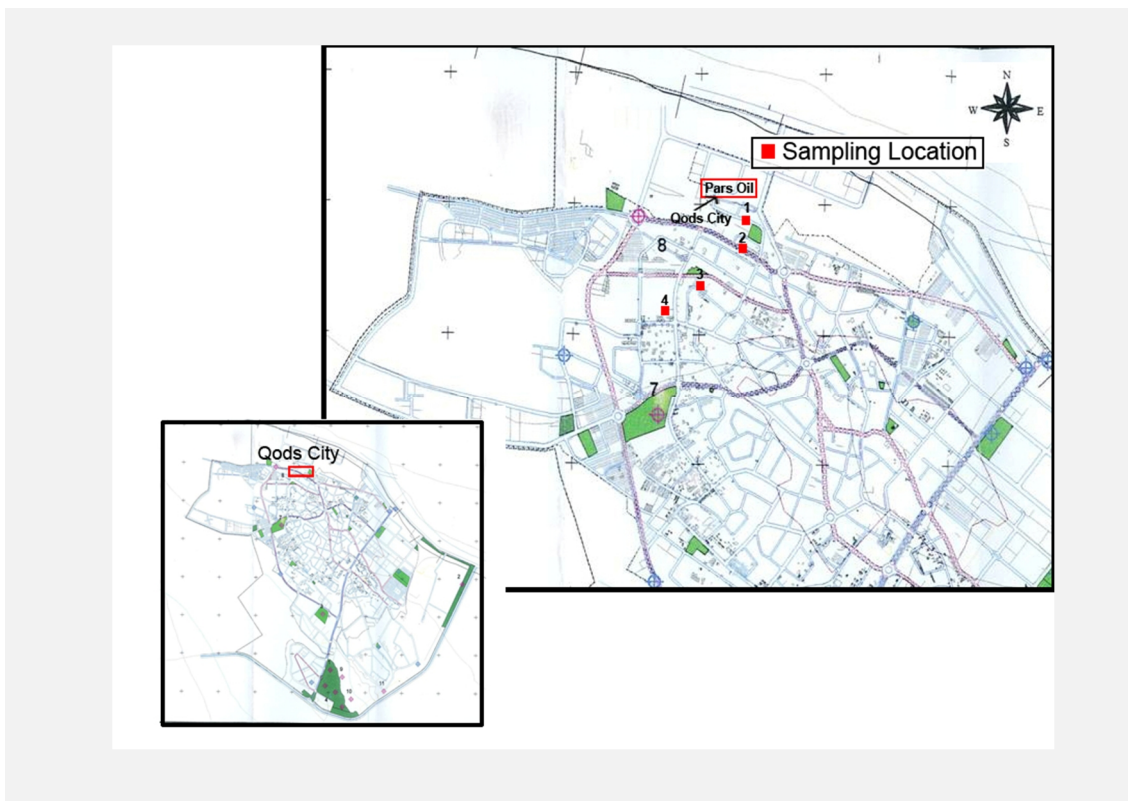


Figure 1. Sampling location from surface and ground water sources (GIS map).

Extraction and Measurement of Heavy Metals from Water Samples

First, 250 ml of the sample was taken and, after the addition of 15-20 ml nitric acid, was placed on a hot plate and heated. During heating, the soluble was not allowed to boil or pour out of the plate. Heating the soluble continued until its volume reduced to 15-20 ml concentrate and a clear sample was obtained. In the case of a failure to obtain a clear sample, about 5 ml of acid were added to the soluble and heating continued until the required transparency was achieved. The resulting sample was poured by distilled water into a 50 ml flask. The lid of the container was closed (in this way, the sample can be kept longer in the refrigerator) and it was used for heavy metals analysis.

Table 1. Standard methods for tests (Greenberg, 1992).

Test	Unit	Standard Method
pH	-	ASTM D1293
TDS	mg/L	Standard.Method2540C
SO_4^{2-}	mg/L	Standard.Method4500B
NO_3^-	mg/L	ASTM D419
NO_2^-	mg/L	ASTM D1254
COD	mg/L	Standard.Method5220B
BOD	mg/L	Standard.Method5210B
Zn	mg/L	Standard. Method 3111B
Cad	mg/L	Standard. Method 3111B
Fe	mg/L	Standard. Method 3111B
Pb	mg/L	Standard.Method5210B
Color	Pt-CO Hazen	Standard.Method2120B
Ammonia Nitrogen	mg/L	ASTM D 1426-03

Measurement of Heavy Metals

In this experiment, the concentration of heavy metals in the digested water samples was measured. The device used for determining the concentration of heavy metals was a SPECTRAA-200 atomic absorb device which determines the concentration of heavy metals to ppb level.

In this section, the calibration curves of the devices are presented to measure, in samples injected into the device, to approve the device performance.

Determination of petroleum hydrocarbons (TPH)

In this experiment, the concentration of total petroleum hydrocarbons (TPH) was measured by liquid to liquid extraction and gas chromatography (GC) methods based on the instructions provided by Moopam (1999).

After sampling and extracting the petroleum hydrocarbons compounds from the samples, a gas chromatograph device (GC-14A SHIMDZU Co.) was used in order to determine the TPH concentration.

The thermal-temporal programme of gas chromatograph (GC) used in this study to measure the concentration of petroleum hydrocarbons is presented in Table 2 (according to Moopam, 1999).

Computation Methods

Accuracy was investigated by repeatable tests and standard solutions at different concentrations. Accordingly, to achieve greater certainty, all calibration documentation of the laboratory equipment was surveyed before performing any test. To evaluate laboratory performance and

analyse accuracy, six duplicate samples were isolated from each station. At each station, eight duplicate samples were taken in separate containers from each stage. The analytical error of the equipment was evaluated by two graphical and computational methods by considering the results of duplicate samples and their comparative evaluations with the results of the main samples. For the graphical method, a control chart known as the Thomson Chart designed by Thomson- Hobart (1976) was used. The horizontal axis of this logarithmic chart represents the average obtained from two series of measurements, and its vertical axis refers to the difference between them. To check the analysis accuracy, the chart presents two criteria: Loxodrome 10% and 1% error.

Table 2. Thermal-temporal program of gas chromatograph used in this study to measure the concentration of petroleum hydrocarbons.

Variable under study	Quantity
Type of Detector	Flame Ionized Detector(FID)
Detector Temp (° C)	330
Injector Temp (° C)	250
Column Initial Temp (° C)	60
Column Initial Time (min)	2
Column Final Temp (° C)	270
Temp Rate	10 ° C /min
Column Final Time (min)	22
Column Program Rate (°C/min)	10
Stop Time (min)	21
Detector	1
Range of Detector	2
Polarity of Detector	2
Hydrogen Flow (kg/m ³)	0/60
Air Flow (kg/m ³)	0/40
Nitrogen Flow (kg/cm ³)	50
Injection Sample Volume (µL)	10

As regards the current research, all charts showed an error rate of less than 10%, so the accuracy was approved. In the computation method for information of relative error after diffraction computation (variance) of duplicate samples, the reliability coefficient related to analysis at a 95% level of trust was calculated and then relative error was obtained.

Results

The typical results of quantitative analysis of PH, COD, BOD, nitrate, nitrite, colour, conductivity, ammoniac nitrogen, heavy metals, and TPH are presented in Tables 3 and 4 and Figures 2 and 3. These results indicate that, in some cases, the amount of pollutants in the sampled regional water is higher than standard amounts for potable water offered by WHO (Chin and Wong, 2001).

The comparative graphs show the procedure for changes in the parameters including COD, BOD, nitrate, colour, ammonia, and heavy metals at the four sampling stations.

Based on the above points, examination of the quantities achieved indicates no wide fluctuation for the procedure of variations for the measured parameters during the four seasons. Therefore, weather changes and the amount of rain had no significant effect on the results obtained; however, the measured parameters show a meaningful varied procedure at the sampling stations according to their distance from the polluting sources.

The results clearly indicate that the quantities achieved are a function of the distance between the sampling stations and the outlet canals of the refinery sewage.

Table 3. Typical results of the analysis of water taken from sampling stations.

No	Parameter	unit	Station 1	Standard deviation	Station 2	Standard deviation	Station 3	Standard deviation	Station 4	Standard deviation
			mean		mean		mean		mean	
1	Fe	mg/L	0.417	0.21188	0.3625	0.20189	0.344	0.1374	0.645	0.2265
2	Zn	mg/L	0.34	0.15979	0.3225	0.14127	0.3025	0.1646	0.5025	0.05377
3	Pb	mg/L	0.032	0.02609	0.03075	0.02708	0.0287	0.02243	0.062	0.1849
4	Cd	mg/L	0.011	0.00351	0.014	0.01304	0.0137	0.01565	0.0377	0.00866

Table 4. Typical results of the analysis of water taken from sampling stations.

No	Parameter	unit	Station 1	Standard deviation	Station 2	Standard deviation	Station 3	Standard deviation	Station 4	Standard deviation
			1		2		3		4	
1	TDS	mg/L	1466.6	77.8	1236.3	55.4	1.95	99.8	1178	86.5544
2	S ⁻²	mg/L	4.6666	1.50444	3.9	0.95394	2.63333	0.64291	3.53333	1.70392
3	SO ₄ ⁻²	mg/L	354	89.40358	200.666	62.9629	184	65.88627	206	69.93568
4	COD	mg/L	176	42.57934	111.666	8.32666	91	17.34935	87.6666	12.01388
5	BOD	mg/L	1.1	12.28821	68.3333	16.19671	54.6666	10.78579	87	19.97498
6	NO ₃ ⁻	mg/L	6.9666	0.30551	6.06667	0.37859	5.43333	0.75719	5.03333	1.00664
7	NH ₃	mg/L	3.2666	2.12211	2.76667	1.74738	2.9	0.17321	6.1	0.51962
8	COLOR	Pt-Co Hazen	30	5.56776	17.3333	3.51188	15.3333	1.52753	16.5	5.4467

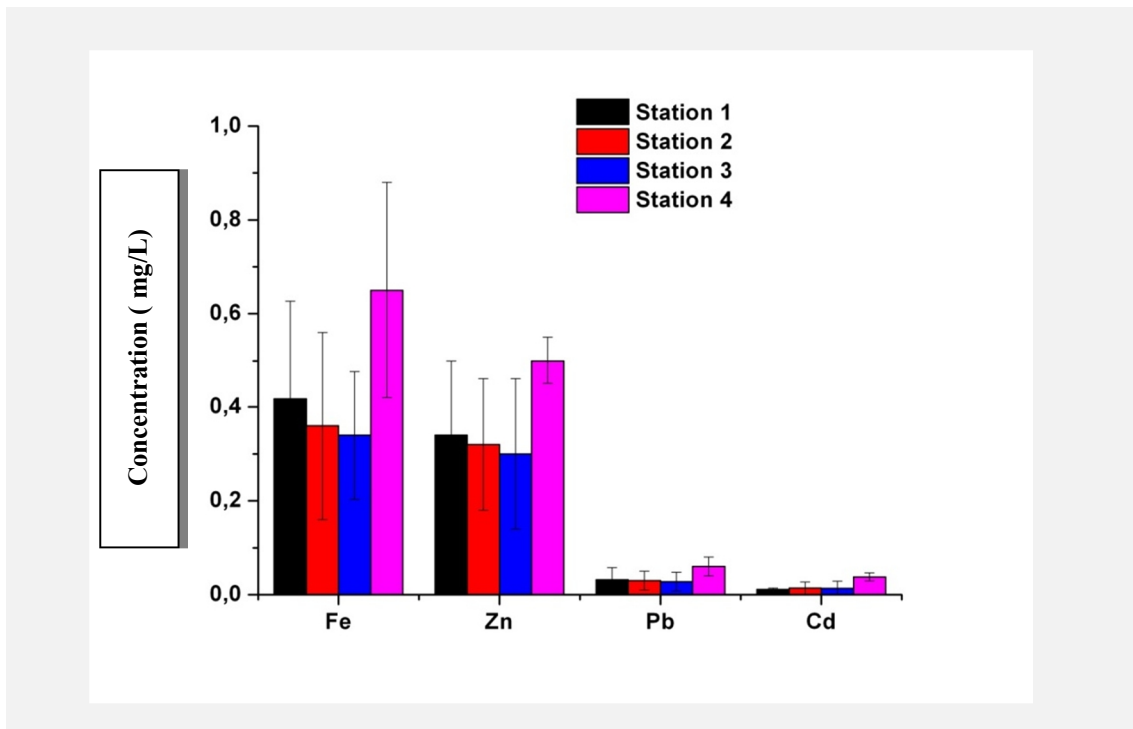


Figure 2. Typical results of the analysis of water taken from sampling stations.

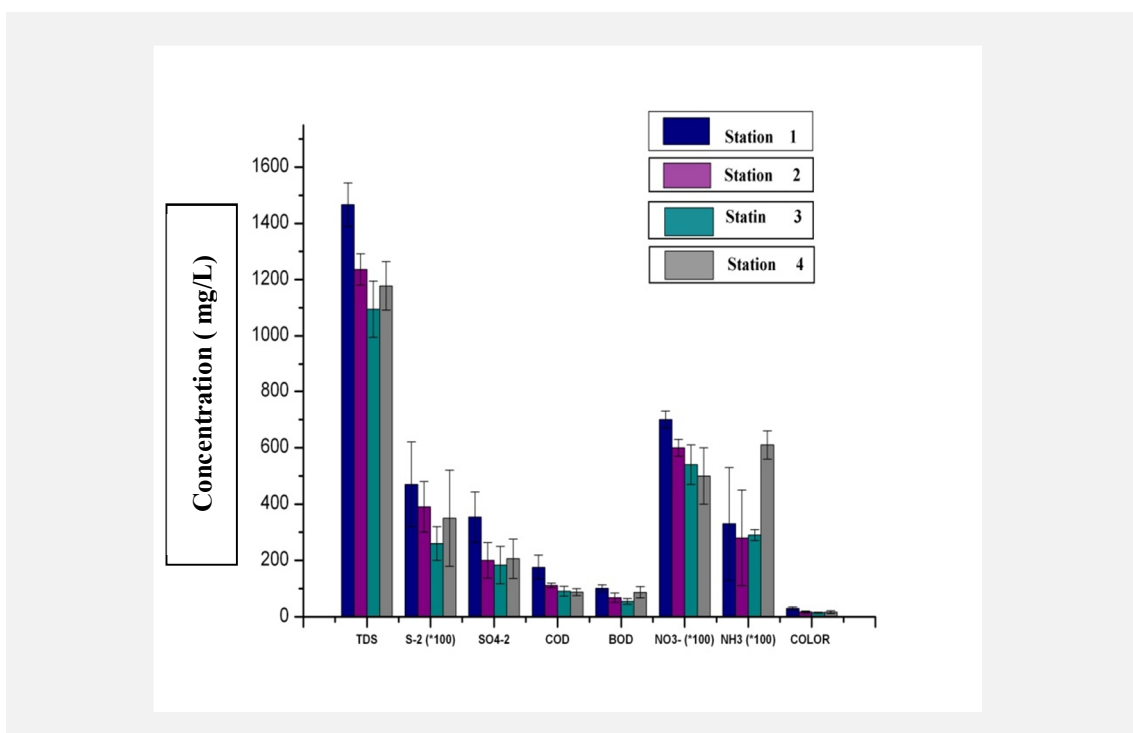


Figure 3. Typical results of the analysis of water taken from sampling stations.

Discussion

Taking into consideration the data obtained from sample analysis as well as the dispersion and situation of the wells, the following points can be concluded:

1. The amounts for the parameters, including electrical conductivity, soluble solids, calcium, magnesium, and hardness, were identical at all sampling stations. However, the range of the calcium and magnesium in a research conducted by Uzoekwa (2011) were variable (34.74 ± 0.09 - 13.45 ± 0.33 and 2.74 ± 0.18 - 1.80 ± 0.06 , respectively). Also, the amount of EC was equal to 1150.41 ± 0.01 – 151.50 ± 0.71 in Uzoekwa's research (2011). Identical results were obtained in this study, indicating the uniformity of the research site.
2. The amounts for BOD, COD, and ammoniac nitrogen at the sampling stations were very high. The other stations, however, passed the standard limit with a high difference. Otokunefor (2005) achieved the same results for BOD and COD. Considering the sporadic plan of the sampling stations and their proximity to the sewage transfer path, the increase in the above-mentioned parameters is a function of distance from the refinery's untreated sewage. It should be mentioned that the sampling stations are influenced by wastewater from residential areas, as well. The average amount of nitrate was high at all stations, which indicates that the effect of wastewater from residential areas is influenced by the outgoing sewage from the refinery.
3. The amounts for the measured heavy metals Zn, Cd, Pb, and Fe at stations 1, 2, and 3 did not have a wide fluctuation, however, the measured amounts of heavy metals at station 4 showed a high level of increase in comparison with the other stations. Such an increase occurred due to the proximity of station 4 to farmlands. Station 4 is influenced by the consumed pesticides and poisons. The findings of Otokunefor (2005), however, revealed that Pb and Cd were low at all sampling stations.
4. Furfural measured about 5 (mg/L) at all stations. Although the seepage of this poisonous and dangerous substance into the water resources was limited, the amount is still considerable; thus it is considered to be an indicator of untreated refinery sewage. The amount of this element at stations which are far from the sewage transfer path confirms that it flows into the receiving areas without being treated.
5. The TPH of the samples was evaluated qualitatively; therefore the existence of TPH in the samples was not verified, and the related report was very concise. According to the results achieved at stations 1, 2, and 3 (apart from station 4 which is close to farmlands), a small amount of TPH (equal to 0.003mg /lit) was reported (due to the closeness to the canal outlet). Like the other examined parameters, it confirms the seepage of refinery sewage into regional underground water resources.

Ultimately, analysis of the data gathered shows that the variations among the measured parameters are not considerable within different seasons. Changes in climatic factors and the amount of rain did not have any considerable effect on the analytical results of different samples.

It is hoped that with the commissioning of the refinery's biological treatment unit, the aforementioned difficulties will be solved as soon as possible.

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