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Impact of oil refinery wastes on water and soil quality: A case study

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ABSTRACT

Bongaigaon Refinery, Assam in India is in the business of refining crude oil since 1974. The effluent of the refinery drains into the Tunia river, which flows through agricultural lands. The present study investigates the pollution level of the Tunia river water as well as the soil of the nearby agricultural lands. Water samples were analyzed for p^H, EC, TDS, TSS, SAR, sulphate, phosphate and heavy metals Cr, Ni, Mn, Zn, & Cu. Soil samples were analyzed for p^H, EC, Na^+, K^+, sulphate, phosphate and Cr, Ni, Mn, Zn, and Cu. Contamination levels of Cr, Ni, Mn and Cu in the river water were under the permissible limit set by FEPA, except Zn, which was in alarming level. However, the values for Cr, Mn and Cu were much higher than the limit set by FAO. The study also reveals the enrichment of Cr, Mn, Ni and Cu in the agricultural soil irrigated by the Tunia river water.

KEYWORDS

Cr;
Mn;
Ni;
Zn;
Cu;
Refinery effluent.

INTRODUCTION

Environmental degradation due to rapid urbanization and industrialization has emerged as a great threat to all developing nations. Industries are discharging various pollutants into the atmosphere everyday polluting the air, water and soil environment. Among various industries, the petroleum industry is one of the major contributors of different contaminants to the environment. The refineries yield large quantity of effluent in the form of waste water. Due to the ineffectiveness of purification systems, wastewaters may become seriously dangerous, leading to the accumulation of toxic products in the receiving water bodies as well as in the soil system[7, 21]. The wastewater released by the refineries are characterized by the presence of large quantities of polycyclic and aromatic hydrocarbons, phenols, metal derivatives, surface-active substances, sulphides, naphthylenic acids and other chemicals. Although, some effluents contain considerable amount of nutrients which may be beneficial for plants, the major environmental problems arises from the refinery effluent are acidification of soil, high contents of heavy metals and sulphate and also the presence of organic contaminants such as polycyclic aromatic hydrocarbons in the soil[33]. These compounds are highly toxic due to their genotoxic, mutagenic and carcinogenic potential. While such contaminant penetrates into the soil, it disturbs the structure of the soil and modifies its physico-chemical properties. It was reported that various physico-chemical properties of soil like P^H, conductivity, redox potential, water holding capacity, organic carbon, sulphate, phosphate, fluoride, sodium, potassium show considerable variation when irrigated with refinery effluent[17, 29, 38].

Several investigations have shown the harmful effects of long term use of refinery wastewater on agricultural soil[34, 37]. It was reported that crops grown in these polluted soils accumulate heavy metals to such an extent that it causes health hazards to human beings and animals[12, 27, 28]. The use of refinery effluent for raising agricultural crops is bound to aggravate trace metal contamination in near future[1, 2, 18]. These observations were also supported by[16, 20, 24], who showed the adverse effect of accumulation of trace metal in the soil. In health researches also it has been established that soil which accumulates heavy metals are dangerous for health. High concentrations of these metals in the soil can be the factors of people's death or poisoning[3]. Most of heavy metals (like mercury (Hg), plumbum (Pb), cadmium (Cd), chromium (Cr), copper (Cu), nickel (Ni), zinc (Zn), cobalt (Co), vanadium (V), molybdenum (Mo), beryllium (Be), uranium (U), strontium (Sr), arsenic (As) and other have shown all or at least several negative effects on health. Their effect may be carcinogenic, mutagenic, teratogenic, neurotoxical[4].

The aim of the present study was to assess the impact of refinery effluent on the receiving water bodies as well as on the agricultural soil irrigated by this water.

MATERIALS & METHODS

Site description

The area chosen for the study is in the vicinity of Indian Oil Corporation Ltd. (Bongaigaon Refinery); formerly known as Bongaigaon Refinery and Petrochemical Limited (BRPL), Dhaligaon, Assam in India. The plants of Bongaigaon Refinery are located by the side of National Highway No. 31C at latitude 26.47^{0}N and longitude 90.57^{0}E. Bongaigaon Refinery is in the business of refining crude oil for production of petroleum fuels and other value-added petrochemicals like xylene, DMT etc. since 1974. The refinery has a capacity to process 2.35 million tones of crude oil per annum. It produces various types of domestics, industrial and automotive fuels.

The effluents of Bongaigaon Refinery refinery drains into a small rivulet the Tunia nala originating from the refinery complex and ultimately drains into river Tunia in the southern side of the refinery. This river passes through the agricultural land mainly rice field of several villages like Dhaligaon, Kukurmari, Dolaigaon, Bhakharibhitha and Mulagaon located along its way. The water of this river is used by the farmers for raising crops. Though Bongaigaon Refinery has been taking various measures to control the pollution, an investigation showed that the quality and productivity of the crops grown in these areas are not satisfactory. It can be presumed that crops grown in these soils may accumulate heavy metals and various organic and inorganic contaminants to such an extent that it can causes health hazards to human beings and animals.

The present study had been conducted in two phases.

a. Whether Tunia river water is suitable for irrigation or not
b. Whether the water of this river affects the nearby agricultural soil.

Sample collection

Five water samples of Tunia River, from different locations were collected in clean 2 litre polyethylene containers. The collected samples were analyzed for p^H, electrical conductivity (EC), redox potential (RP), phosphate, sulphate, sodium adsorption ratio (SAR), and also for heavy metals like Cr, Ni, Mn, Zn, and Cu.

Soil samples were collected from the paddy fields from both sides (four from each side) of the Tunia River. Eight different locations and two different depths (0-25cm and 25-50cm) had been randomly identified to collect those samples. Samples were air dried and sieved through a 2-mm sieve prior to analysis. The processed soil samples were then analyzed for p^H, electrical conductivity (EC), redox potential (RP), organic carbon content (OC), phosphate, sulphate, exchangeable cations like Na^+ and K^+ and also for the above mentioned heavy metals.

All the results obtained from the above analysis were compared with a soil sample (control C) taken from a virgin area where the affect of the effluent does not come in contact.

EXPERIMENTAL

Soil pH and electrical conductivity was measured in water using a soil-to-water dispersion ratio of 1:5. Organic matter (OM) content in the soil was determined with the Walkley–Black method of dichromate acid oxidation of C[8]. Concentration of sulphate, phosphate were analysed by standard methods in UV-Visible spectrophotometer[36] while exchangeable cations Na^+ and K^+ were analyzed by Flame Photometer. For analysis of the heavy metals, one gram of the soil sample was digested with a mixture of HNO_3, HClO4 and HCL in 10:4:1 ratio. The presence of metals were then determined with Atomic Absorption Spectrophotometer (Shimadzu AA 7000)

The water samples were analyzed for various physico-chemical properties according to[5]. Concentrations of heavy metals were determined in AAS after digesting with conc., HNO_3.

RESULTS AND DISCUSSION

Effect of effluent on river water

Physicochemical parameters and some metal/non-metal ions concentration in the Tunia River water receiving refinery effluents were investigated and compared with FEPA (Federal of Environment Protection Agency)and FAO (Food and agriculture Organization) standards for discharge of effluent in the surface water (TABLE 1).

TABLE 1 : Physicochemical parameters and some metal/ non-metal ions concentration in water

Parameter	Mean	Sd	FEPA	FAO
p^H	5.70	0.20	6-9	6.5-8.0
EC	0.167	0.01	NS	<0.7
TDS	141.6	16.22	2000	<450
TSS	22.8	8.78	30	NS
SAR	9.03	0.46	NS	<3.0
SO_4	197.95	24.43	50	NS
PO_3	BDL	------	5.0	NS
Cr	0.609	0.0007	1.0	0.10
Ni	0.185	0.003	1.0	0.20
Mn	1.15	0.645	5.0	0.20
Zn	0.952	0.620	1.0	2.00
Cu	0.449	0.023	1.0	0.20

EC in ms/cm, TDS, TSS, SO4, PO_3 & heavy metals are in mg/l

The average of pH obtained from the study was 5.7. The implication from this study is that the water of the river is acidic in nature. The value was out of the range set by FAO and FEPA. The means of EC, TDS, TSS, were within the limits set by FEPA as well as FAO for the discharge of wastewater into surface water. However, the mean content of sulphate was much higher than the limit set by the agencies. High concentrations of sulphate can be problematic as they are capable of being reduced to hydrogen sulphide, a toxic, foul-smelling gas. A field survey also reveals that foul smell emanates from the river and small fishes of the river smell chemicals after frying or roasting which is unfit for human consumption. Sodium hazard of irrigation water can be well understood by knowing the SAR values. Sodium Adsorption Ratio (SAR) is an estimate of the degree to which sodium will be adsorbed by the soil. It is used to evaluate the suitability of water for irrigation. High value of SAR means that sodium in the water may replace calcium and magnesium ions in the soil, potentially causing damage to the soil structure[23] and plant growth. SAR is calculated from the following formula[32].

$$\mathbf{SAR} = \frac{\mathbf{Na^+}}{\sqrt{\frac{(\mathbf{Ca^{2+}} + \mathbf{Mg^{2+}})}{\mathbf{2}}}}$$

The SAR values for most of the analyzed water samples were beyond the permissible limit set by FAO. The sodium hazard is a function of both SAR and Salinity. TABLE 2 shows the interpretation criteria for salinity and SAR for water quality for irrigation by FAO.

TABLE 2 : Guidelines for interpretation of water quality for irrigation

Infiltration	None	Slight to moderate moderat moderate	Severe
SAR = 0 - 3 and EC	> 0.7	0.7 - 0.2	< 0.2
3 -6	> 1.2	1.2 - 0.3	< 0.3
6-12	> 1.9	1.9 - 0.5	< 0.5
12-20	> 2.9	2.9 - 1.3	< 1.3
20-40	> 5.0	5.0 - 2.9	< 2.9

From the study, the mean EC value for the analyzed samples was found to be 0.176 whereas mean SAR value was 9.03. Thus according to the above guidelines (TABLE 2) it may be concluded that Tunia river water has severe restriction for irrigation purpose.

The mean concentrations of heavy metals Cr, Ni, Mn, Zn and Cu in the river water were 0.609, 0.185, 1.15, 0.952 and 0.449 mg/l respectively. All these values were under the permissible limit set by FEPA except Zn, which was in alarming level (TABLE 1). However, the values for Cr, Mn and Cu were much higher than the limit set by FAO. The mean value for Zn was under the permissible limit and Ni was in alarming level. However, long term continuous use of this water for irrigation purpose will definitely worsen the soil condition and productivity of the crops. Additionally, crops grown in these areas may accumulate heavy metals which ultimately enter in the food chain.

Effect of effluent receiving water on soil

Physicochemical parameter

The physicochemical properties and the heavy metal content of the nearby agricultural soil irrigated by Tunia river water were estimated. The concentrations of the most significant physicochemical parameters of the soil are shown in TABLE 3.

TABLE 3 : Concentration of various physicochemical parameters of the soil

S.No.	p^H	EC	OC	OM	Na	K	SO_3	PO_4
R1	4.10	0.008	1.89	3.26	6.50	0.50	115.88	BDL
R2	4.38	0.007	0.78	1.34	6.25	0.25	78.55	BDL
R3	4.14	0.009	1.38	2.38	8.50	3.50	167.81	BDL
R4	4.17	0.019	2.16	3.72	17.25	13.50	126.55	BDL
R5	3.96	0.020	1.47	2.53	23.00	17.50	193.02	BDL
R6	4.15	0.011	1.98	3.41	16.00	10.50	389.47	BDL
R7	4.37	0.010	0.66	1.14	14.75	18.75	521.61	BDL
R8	4.42	0.015	0.69	1.19	17.00	9.25	99.31	BDL
L1	4.07	0.020	1.86	3.21	15.00	18.00	282.41	BDL
L2	4.20	0.019	1.38	2.38	15.00	13.00	219.45	BDL
L3	4.41	0.010	1.29	2.22	8.75	3.00	173.20	BDL
L4	4.90	0.012	0.69	1.19	13.25	8.00	273.65	BDL
L5	4.17	0.013	1.56	2.69	13.50	3.50	420.48	BDL
L6	4.22	0.030	1.62	2.79	15.25	9.50	391.62	BDL
L7	3.95	0.032	0.63	1.08	15.75	10.25	483.45	BDL
L8	4.51	0.012	0.81	1.39	12.50	8.75	376.12	BDL
C	5.93	0.002	0.56	0.96	9.50	5.25	25.5	BDL

Right side of the river, L: Left side of the river; R1, R3, R5, R7 and L1, L3, L5, L7: (0-25 cm depth); R2, R4, R6, R8 and L2, L4, L6, L8: (25-50 cm depth); EC in ms/cm, Na & K in ppm, SO_4 & PO_3 in mg/kg, OC & OM in %; BDL: Below detection limit

Measurement of soil pH and electrical conductivity (EC) parameters provides valuable information for assessing soil condition for plant growth, nutrient cycling and biological activity. Soil pH is important because it influences the availability and plant uptake of micronutrients including heavy metals[19]. The average value of soil p^H from the right side of the river was 4.21 (from 3.96 to 4.42) and from the left side were 4.30 (from 3.95 to 4.51) with slight increase with depth. This indicates that the soil from both sides of the river were acidic in nature. It was observed that the values were lower than the 'C' value (5.93). This indicates that the use of the effluent receiving water had affected the soil p^H. Low pH can prevent root respiration and uptake of water and nutrients. It can also have deleterious effect on the soil microorganisms[30].

Salinity is a soil property referring to the amount of soluble salt in the soil. Lower the value of EC, lower is the accumulation of salt in the soil. Moreover, electrical conductivity is proportional to the sum of cations and anions. Univalent cations such as Na^+ are more mobile than multivalent ions such as Ca^{2+} and Al^{3+}. Similarly, univalent anions such as Cl^- are more mobile than multivalent ions such as SO_4^{2-} and CO_3^{2-}, which are in turn more mobile than charged humic substances. Thus, sample with Na^+ and Cl^- as its dominant dissolved species will have a higher conductivity than one dominated by Ca^{2+} and SO_4^{2-} (www.gtk.fi/publ/foregsatlas/text/EC.pdf). By agricultural standards, soils with an EC greater than 4 ms/cm are considered saline[25]. The average value of soil EC from the right side of the river was 0.012 ms/cm (from 0.007 ms/cm to 0.020 ms/cm) and from the left side was 0.018 ms/cm (from 0.010 ms/cm to 0.032 ms/cm). The value for the control soil was 0.002 ms/cm. The data for EC reveals that the soils from both sides of the river were nonsaline. However salinity slightly increases in the left side of the river.

The average value for soil Na from the right side of the river was 13.65 ppm (from 6.25 to 23.00 ppm) and from the left side was 13.62 ppm (from 8.75 to 15.75 ppm). The average value of soil K from the right side of the river was 9.22 ppm (from 0.25 to 18.75 ppm) and from the left side was 9.25 ppm (from 3.00 to 18.00 ppm). The 'C' value for Na was 9.50 ppm and for K was 5.25 ppm. Soils with an accumulation of exchangeable sodium were often characterized by poor tilth and low permeability making them unfavorable for plant growth. The study indicates that both exchangeable Na and K were greater than the 'C' values except the locations R1, R2, R3 and L3. It was also revealed from the study that the soil contains more sodium than potassium.

The values for organic carbon (OC) from both sides of the river ranged from 0.063% to 2.16 % and organic matter from 1.08% to 3.72%. The 'C' values for both the parameter were 0.56% and 0.96% respectively. This indicates that both sides of the river were contaminated by organic carbon, which may deposit in the soil as a result of irrigated water from the Tunia River.

The sulphate content from the right side of the river ranged from 78.55 mg/kg to 521.61 mg/kg and from the left side ranged from 173.20 mg/kg to 483.45 mg/kg whereas the value for the control soil was 25.5 mg/kg. This indicates that the soil from both sides of the river were rich in sulphate. High Sulphur content in the Tunia river water, receiving refinery effluent, which was used for irrigation, was responsible for the high sulphate content in the soil. In the study area, deposition of sulphur was seen over the leaves and grains of the rice crops and also on the bank of the river. If the sulphate continues to accumulate in the soil, the same may create problems in near future[14, 24, 26].

Phosphorous is an essential nutrient element needed for plant growth. The phosphorous absorbed by plants exists in soil as inorganic orthophosphate ions, viz, $H_2PO_4^-$, HPO_4^{-2}, and PO_4^{-3}. However the phosphate content of the study area as well as the in the effluent was below the detection limit which indicates the low fertility of the soil.

Heavy metals

The mean concentrations of Chromium (Cr), Nickel (Ni), Manganese (Mn), Zinc (Zn), and Copper (Cu) in the soil solution were reported in TABLE 4.

These heavy metal concentrations in the soils irrigated by the Tunia river water receiving Refinery effluent may be compared with the FAO threshold values (Source: Wastewater quality guidelines for agricultural use series title: FAO irrigation and drainage papers - 47 1992 T0551/E) for crop production (Figure 1-5). The first observation to make from a comparison of the irrigated soils and control sites were that study area soils were naturally high in some of these heavy metal trace elements. These results were observed significantly in case of Cr Mn, and Cu. In case of Ni the control site soils were below detection limit. It is only in respect of Zn; the concentrations of the sample sites as well as the control site were under the permissible limit set by FAO for crop production. The somewhat high heavy metal trace element content of the soils may be due to their colluvial-cum-alluvial origin and the imperfectly drained ground conditions experienced during periods of heavy rainfall[12].

Another major observation that might be made is that, based on FAO (1985) recommended maximum levels of heavy metals for crop production, the Tunia river water and the irrigated soils from both sides of the river had higher values than the recommended levels of Cr (i.e. 0.10), Mn (i.e.0.20) and Cu (i.e.0.20), while the levels of Zn are lower than the maximum threshold values recommend for crop production. The levels of

TABLE 4 : Average concentration of Heavy metals in soil

Heavy metals	Mean (mg/l)	Control	Standard deviation	FAO threshold values (mg/l)	Remarks
Chromium	1.11	0.40	0.25	0.01	Not generally recognized as an essential growth element. Conservative limits recommended due to lack of knowledge on its toxicity to plants
Manganese	6.65	1.27	2.90	0.20	Toxic to a number of crops at few-tenths to a few mg/l, but usually only in acid soils.
Nickel	0.55	BDL	0.09	0.20	Toxic to a number of plants at 0.5 mg/l to 1.0 mg/l; reduced toxicity at neutral or alkaline pH
Copper	0.56	0.17	0.12	0.20	Toxic to a number of plants at 0.1 to 1.0 mg/l in nutrient solutions
Zinc	0.99	0.35	0.29	2.00	Toxic to many plants at widely varying concentrations; reduced toxicity at pH > 6.0 and in fine textured or organic soils.

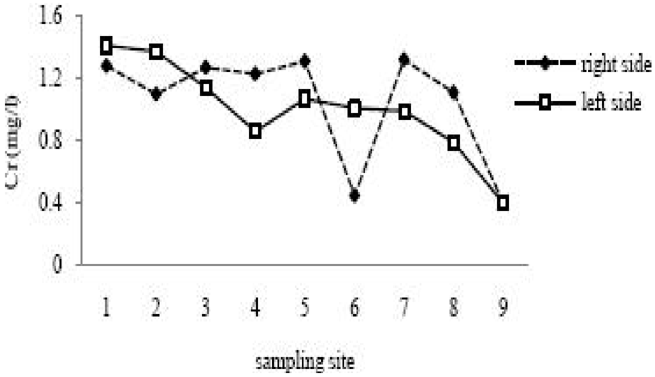

Figure 1 : Content of Chromium

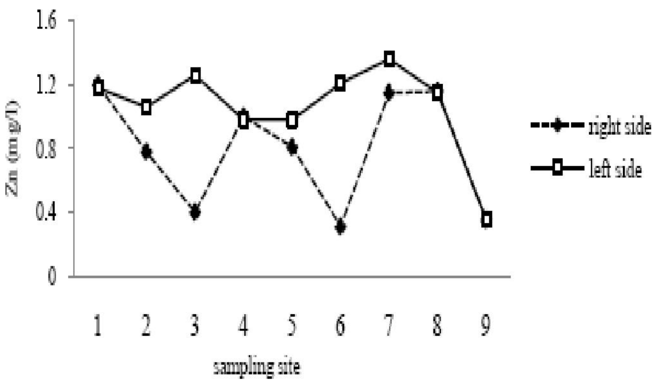

Figure 2 : Content of Zinc

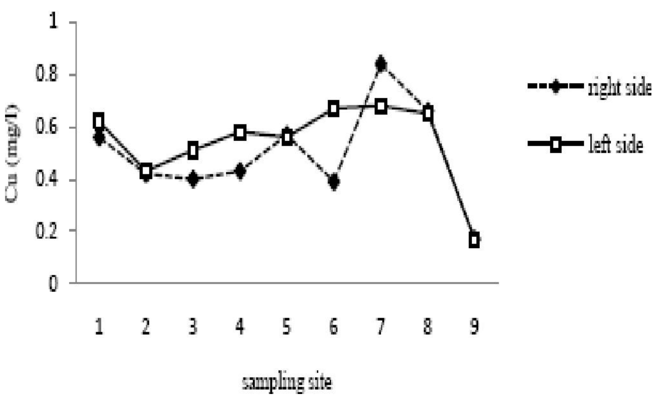

Figure 3 : Content of Copper

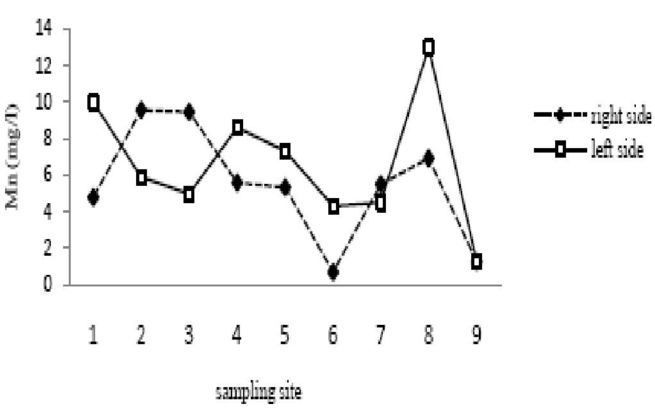

Figure 4 : Content of Manganese

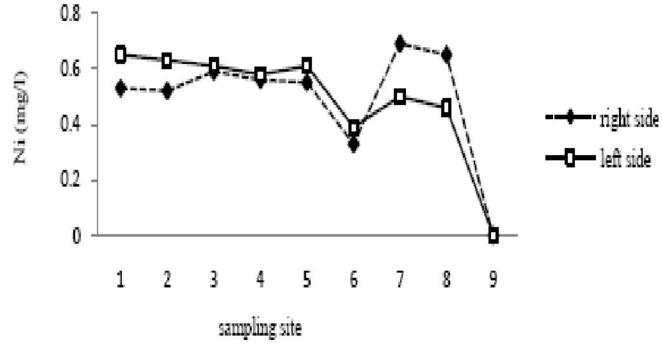

Figure 5 : Content of Nickel

Ni for the Tunia river water receiving refinery effluent were in alarming level while for the irrigated soil, the values were much higher than the threshold values recommended by FAO. It was also observed that irrigated water of the Tunia River had actually augmented the trace element content of the soils. According to[9] long-term use of sewage sludge, heavy metals can accumulate to phytotoxic levels and result in reduced plants growth and enhanced metal concentrations in plants, which consumed by animals, then enter the food chain. The Cr, Mn, Ni and Cu levels recorded in the soils are close to or higher than the toxic levels for crops; these levels can become problematic at low pH values.

Correlation study

TABLE 5 shows the correlation matrix between the soil heavy metal concentration levels of both sides of the river. Determination of the correlation between heavy metals in soils is very helpful for a better understanding of their spatial distribution. Although pollution resulted from single heavy metal is present in the environment, pollution from the combined heavy metal often occurs synchronously. The combined pollution caused by several heavy metals was not only present in water body receiving refinery effluent but also often occurred in irrigated soils[15]. It was reported by several investigators that that Cu, Zn, Pb and Cd were

TABLE 5 : Correlation matrix of soil heavy metal concentration levels

	Cr	Ni	Mn	Zn	Cu
Cr	1				
Ni	0.749**	1			
Mn	0,191	0.330	1		
Zn	0.325	0.323	0.099	1	
Cu	0.120	0.243	0.084	0.663**	1

** Correlation is significant at the 0.01 level (2- tailed) common anthropogenic and mutagenic elements in the urban environment and were well correlated[6, 10, 11, 31]. In the present study highest positive correlation was observed between Cr and Ni (0.749**) while Zn and Cu also shows significant positive corerelation (0.663**). Based on this study contamination of combined heavy metals in agricultural soils could be better analyzed.

CONCLUSION

The present investigation showed water of the Tunia River and the soil irrigated by this water were contaminated by the discharge of the refinery effluent. Pollution of the River water poses a serious threat to aquatic organisms and ultimately the entire ecosystem. Waste water irrigation led to the accumulation of heavy metals Cr, Mn and Cu in the soil and consequently will affect the agriculture. These effects are of particular importance for the farmers since these may reduce soil productivity, fertility and yield. Consumption of the food grown in these areas with elevated levels of heavy metals may lead to high level of body accumulation causing related health disorders. Correlation study indicates that pollution by combined heavy metals also occurred in the soil irrigated by the river water. Thus regular monitoring of heavy metal contamination in the Tunia river water and the soil irrigated by this water is necessary in order to reduce the health risk caused by taking the contaminated vegetables. Proper remedial measures should be applied to remediate already contaminated water and soil to promote safe cultivation for the benefit of the farmers with sustainable productivity.

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