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SOIL SCIENCE

Effect of Crude Oil Spillage on Chemical Properties of Soils of Moran and Duliajan Oil Fields of Assam

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ABSTRACT

Crude oil spillage is a regular phenomenon in the oil drilling sites and due to raining and flooding the spilled oil spread to the nearby cultivated field causing soil pollution and considerable reduction in the crop yield. Therefore, present investigation aimed to study the effect of crude oil spillage on chemical properties of soils of two major oil fields in Assam, India *viz.* MFN oil field, Moran and Kathalguri, HYR, Duliajan oil field. Five soil samples were collected in triplicates at an interval of 50 m from three directions (E, W& S) from the spilled areas upto 200 m horizontally and one sample from each direction was collected from adjacent unpolluted areas *i.e.* beyond 200 m as control. Standard analytical procedures were followed to determine soil pH, EC, organic carbon, total petroleum hydrocarbon, available nutrients, and exchangeable elements. Results revealed pH of the soils remarkably dropped towards spillage point making the soil strongly acidic in nature whereas the organic carbon content increased near the point of spillage in both oil fields. The total petroleum hydrocarbon (TPH) was recorded maximum at spillage point in both Moran and Duliajan oil fields and was negligible beyond 200m. The available N and P_2O_5 were low near the spillage point and substantially increased with distance. Conversely, available K_2O, exchangeable Ca^{2+} and Mg^{2+} significantly increased near the spilled point.

Highlights

- Chemical properties of agricultural soil nearby the oil fields were highly affected by presence of crude oil and detrimentally effected soil fertility status making it unfit for cultivation.

Keywords: Crude oil, hydrocarbon, acidic, organic carbon, available nutrient

Oil exploration is an age old activity taking place in wider areas of Assam since the pre independence period and it holds a unique position regarding the production and commercialization of petroleum oil. Assam is the third largest producer of petroleum and natural gas in the country and more than 15 per cent of the country's reserves are found in the Assam-Arakan Basin in Northeast India. Oil exploration involves activities like drilling and refining including production, transportation and refining of crude. These activities resulted in frequent accidental leakage in the nearby areas causing serious environmental pollution (Rowell 1977). Occurance of activities like oil well blow out, tanker accidents, accidental rupture of pipelines and routine clean-up operations which leads to oil spillage (Chang *et al.* 1996). Gogoi *et al.* (2003) mentioned that the problem is more serious in areas of heavy rainfall and flooding causing oil spillage induced soil pollution. Crude oil is a naturally occurring, non renewable resource unrefined petroleum product composed of a complex mixture of about 200 or more hydrocarbon and other organic materials. It is refined to produce usable products like gasoline, diesel, kerosene, asphalt and various other forms of petrochemicals. They are highly persistent in the environment due to the presence of large and complex molecules that composed the crude oil. They are toxic in nature and exhibit significant health risks to human (Hentati *et al.* 2013). Crude oil contamination beyond 3% concentration was reported to cause deleterious effects on soil biota and crop growth by alterating various soil properties and depleting soil fertility (Abii and Nwosu 2009; Osuji *et al.* 2005). Spilled crude-oil is generally denser than water and the organic hydrocarbon clogs soil pores thus, reduces and restricts permeability and expel water and air depriving plant roots the much needed water and air (Brian 1977; Essian and John 2010). Therefore, the present study was carried out with the aim of studying the effect of spilled crude oil from oil fields to nearby agricultural land on the chemical properties and fertility status of soil.

MATERIALS AND METHODS

Location of the study area

The present investigation was conducted in two oil fields of Assam namely, MFN oil field, Piyoli Nagar, Moran ($27^0$11′44″ N lat; $94^0$54′23″ E long) and Kathalguri, HYR, Duliajan (27^0 30′55″ N lat; $95^0$52′25″) of Dibrugarh and Tinsukia district, respectively, where extensive oil exploration activities are carried out by Oil India Limited (OIL). The areas experience climate with a maximum temperature of 38^0 and minimum temperature of 6^0, annual rainfall between 1800-2500 mm and relative humidity of 78%.

Collection of Soil Samples

Five soil samples were collected in triplicates at an interval of 50 m from the spilled point upto 200 m in three directions *viz.* East, West and South. One sample from each direction was collected from adjacent unpolluted areas *i.e.* beyond 200 m as unpolluted control. A total of 36 samples (15 from 3 directions of crude oil spilled areas of each oil field and 6 from adjacent unpolluted areas of each oil field) were collected from a depth of 0-15 cm with the help of core sampler and carried in polythene bags. The soil samples collected were air dried, grinded and passed through 2 mm sieve and stored in polythene bags for laboratory analysis.

Laboratory analysis

The pH and electrical conductivity of soil was determined by 1:2.5 soil water suspension (Jackson 1973). Organic carbon content was determined by Walkley and Black wet digestion method as described by Jackson (1973), available nitrogen by alkaline potassium permanganate method (Subbiah and Asija 1956), available phosphorous by Bray and Kurtz No. I method (Jackson 1973), available potassium was determined flame-photometrically as outlined by Jackson (1973) and exchangeable calcium and magnesium by versenate titration method (Richards 1954). TPH was determined using method described by Baruah *et al.* (2016). Total petroleum hydrocarbon (TPH) content of the soil samples was estimated by Soxhlet apparatus. Petroleum benzene (boiling point 400–600 °C) was considered as an extracted solvent. Extracted solution was poured into a 50-ml beaker and evaporated to dryness in a rotary evaporator. TPH present in the soil samples was estimated by taking the weight of the beaker containing extracted solvents of samples till the weights become constant. Then, the value of the TPH was calculated by the following formula:

$$ppm = X - Y \times 10000 / W$$

where,

X = final weight of the beaker and oil and grease,

Y = initial weight of the beaker, and

W = sample weight

RESULTS AND DISCUSSION

pH

The soils of Moran and Duliajan oil fields exhibited strong to medium acidic nature and the pH ranged from 4.8 to 5.4 and 5.0 to 5.5, respectively (Table 1). The lowest soil pH was found at a distance 0 m at the spillage point in all the directions. The soil acidity declined with increase in distance from the spillage point and a maximum pH value was recorded in the unpolluted soils beyond 200 m.

Table 1: Chemical properties of soils of Moran and Duliajan oil fields

Distance (m)	pH		EC (dS m^{-1})		Organic carbon (g kg^{-1})		Total petroleum hydrocarbon (mg g^{-1})	
	Moran	Duliajan	Moran	Duliajan	Moran	Duliajan	Moran	Duliajan
East Direction								
0	4.9	5.0	0.06	0.05	27.4	26.3	8.5	11.6
50	5.1	5.1	0.06	0.06	23.5	22.3	8.6	9.3
100	5.2	5.3	0.08	0.07	20.9	19.3	4.0	8.1
150	5.3	5.4	0.09	0.09	16.4	15.81	3.2	4.6
200	5.4	5.4	0.10	0.11	9.3	9.7	1.1	1.2
>200	5.4	5.5	0.10	0.11	7.1	7.5	0.0	0.0
West Direction								
0	4.8	5.0	0.05	0.04	25.1	24.7	14.5	8.9
50	5.0	5.0	0.06	0.05	21.4	20.7	11.3	7.9
100	5.1	5.2	0.07	0.07	18.9	19.7	8.8	4.7
150	5.2	5.3	0.08	0.09	14.6	14.5	5.5	2.3
200	5.4	5.4	0.10	0.10	8.8	8.7	1.2	1.2
>200	5.4	5.5	0.10	0.11	7.2	7.4	0.0	0.0
South Direction								
0	4.9	5.0	0.05	0.05	26.7	25.5	10.7	9.6
50	5.0	5.3	0.05	0.05	22.4	21.4	10.5	9.1
100	5.1	5.3	0.07	0.07	19.5	18.2	7.4	7.2
150	5.2	5.4	0.09	0.08	15.5	14.8	4.2	4.5
200	5.4	5.5	0.10	0.09	9.7	9.6	1.2	1.2
>200	5.4	5.5	0.10	0.10	7.1	7.5	0.0	0.0
	SEM±	**LSD(5%)**	**SEM±**	**LSD(5%)**	**SEM±**	**LSD(5%)**	**SEM±**	**LSD(5%)**
Location	0.1	0.1	0.001	NS	0.32	NS	0.03	0.23
Distance	0.2	0.3	0.001	0.004	0.35	0.71	0.20	1.14
Direction	0.1	NS	0.001	0.003	0.34	1.00	0.13	0.93
Location × distance	NS	NS	NS	NS	NS	NS	0.13	0.26
Location × direction	NS	NS	NS	NS	NS	NS	0.26	0.76
Distance × direction	NS	NS	NS	NS	NS	NS	0.05	0.09
Location × distance × direction	NS	NS	NS	NS	NS	NS	0.05	0.09

Similar increase in soil acidity of crude oil affected soils of Duliajan oil field was also reported by Basumatary *et al.* (2012). The presence of more amount of crude oil near the drilling point led to the production of acidic intermediates such as phenolic acid, organic acid, esters and fatty acid in the polluted soils through the microbial metabolism in soil (Odu 1981) which might increase the soil acidity. The decrease in pH values of soils with increasing oil content is evident from the negative correlation existing between pH and TPH content of the soil (r = -0.639**; Table 3). Similar results were also reported by several authors (Oyem and Oyem 2013; Basumatary *et al.* 2012; Barua *et al.* 2011). It was also observed that the pH of soils varied significantly in different directions. West direction of Moran oil field was found slightly more acidic as compared to the other two sides while Duliajan oil field exhibited significantly higher pH value in east direction compared to other two directions which might be due to gradient effect.

Electrical conductivity

The absolute values of EC in the study areas were very low and were significantly lower in the crude oil polluted soils of Moran and Duliajan oil fields. The values ranged from 0.05 to 0.10 dS m^{-1} in Moran soils and from 0.04 to 0.11 dS m^{-1} in Duliajan soils (Table 1). The variations in the EC values with respect to different directions were found to be significant at 5% level of probability. Moran soil recorded the lowest EC value in the west direction followed by south and east. While east direction reported the lowest EC value in the Duliajan oil field. Osuji and Nwoye (2007) explained the possible reason for such variation in EC values were not likely because of direct influence of spillage since organic compounds like crude oil cannot conduct electrical current very well. However, the anoxic biodegradation mechanism through direct dehydrogenation allowed the anaerobic metabolism of hydrocarbons in the presence of an electron acceptor such as nitrate ion, which may be partially responsible for the observed differences in EC. Talukdar (2014) also reported that electrical conductivity decreases linearly with the increase in the percentage of crude oil contamination. Mitchell (1976) elucidated that the dielectric constants of certain liquids such as oil, waste chemicals etc. are different from that of normal water. The dielectric constant of soil system changes when crude oil is mixed with it. This change might have caused the change in electrical conductivity of crude oil contaminated soils.

Organic carbon

Organic carbon content of soils was maximum at spillage point *i.e.* 0 m (27.4 g kg^{-1}) in Moran oil field and it decreased gradually with increase in distance from drilling point. The lowest values were reported in unpolluted soils beyond 200 m (7.1 g kg^{-1}). In case of Duliajan oil field, organic carbon content in soils ranged from 7.4 to 26.3 g kg^{-1}. Basumatary *et al.* (2012) reported similar increase in organic carbon content of crude oil polluted soils of Duliajan. It was observed that the organic carbon content in the spillage point (0 m) increased by 74% in Moran oil field and 71.8% in Duliajan oil field *vis-a-vis* unpolluted soils. This increase in organic carbon content as influenced by TPH content was evident from the significant positive correlation (r = 0.912**) existed between them (Table 3). This increment in the organic carbon content of soil might be due to several reasons. Firstly, hydrocarbons as the name indicated are compounds containing chiefly carbon and hydrogen atoms. About 50 to 98 percent of the total weight of crude oil consists of thousands of individual compounds of hydrocarbons (Irwin et al. 1998). Hydrocarbon contaminated sites receive continuous inputs of carbon which could have been converted to soil organic carbon and thus, enhanced the level of organic carbon and organic matter in the affected sites. Similar findings were reported by Benka-Coker and Ekundayo (1995), Ekundayo and Obuekwe (1997), Shukry (2013) who noted increment of organic carbon content of oil polluted soils in Southern Nigeria. Secondly, after volatilization and degradation processes of the low and highly degradable Hydrocarbon molecules of the crude oil samples respectively, the high molecular hydrocarbons of the crude oil get distributed on the soil surface which added to the organic carbon content of the soils. Thirdly, the presence of crude oil on the soil medium created an anaerobic condition within the medium which led to the death of most of the aerobic micro organisms that formed part of the organic substance and thus, added to the organic matter content of the soil. Also, Nudelman *et al.* (2002) stated that sorbed decayed substances got distributed and formed covalent bounds with the soil organic matter by oxidative coupling.

Total Petroleum Hydrocarbon

The TPH content of both the soils was found to be higher in the vicinity of oil drilling point and decreased significantly with increase in distance. The lowest TPH content was observed in soils at a distance 200m and no traces of TPH was observed beyond 200 m This decrease TPH content with increase in distance from the drilling point might be due to the declined flow rate of runoff water carrying crude oil with distance and thus more amounts got deposited near the drilling point *vis-a-vis* distant places. The reason for decreased runoff flows possibly due to the increased density and viscosity of the water caused by the presence of much denser and viscous crude oil. Declining diffusivity of hydrocarbon with distance might be another important factor of horizontal movement of crude oil since affinity forces on soil domain play significant role in mobility of the later. According to McGill *et al.* (1981), the viscosity of the hydrocarbon governed the extent of physical movement of PHC in the soil profile. Moreover, TPH content of soils also varied significantly with direction. The maximum TPH content were reported in the west direction in Moran soils and in case of Duliajan soils it was found to be more in east direction. This might be due to increase in slope of the respective area of either direction.

Available Nitrogen

Amount of available nitrogen content varied significantly with distance from the spillage point

Table 2: Available nutrients (kg ha^{-1}) and Exchangeable cations [cmol (p+) kg^{-1}] contents of soils of Moran and Duliajan oil fields

Distance (m)	Available nutrients						Exchangeable cations			
	N		P_2O_5		K_2O		Ca^{2+}		Mg^{2+}	
	Moran	Duliajan	Moran	Duliajan	Moran	Duliajan	Moran	Duliajan	Moran	Duliajan
East direction										
0	226.6	229.6	16.5	17.5	222.7	218.7	1.06	1.08	0.58	0.62
50	251.4	259.4	17.9	18.1	218.6	217.2	1.03	1.05	0.55	0.56
100	272.5	275.3	20.7	19.9	212.7	211.9	0.98	0.99	0.52	0.51
150	279.2	281.3	22.8	21.6	210.1	204.4	0.91	0.94	0.50	0.47
200	280.8	284.8	25. 6	24.7	194.3	193.0	0.86	0.88	0.47	0.43
>200	282.3	286.2	29.7	27.6	190.6	195.1	0.84	0.86	0.42	0.41
West Direction										
0	221.7	224.9	15.5	17.7	227.5	222.8	1.21	1.17	0.62	0.65
50	250.4	257.3	16.7	18.5	223.7	220.8	1.12	1.08	0.58	0.61
100	265.6	270.4	18.3	20.6	218.4	216.3	1.09	1.02	0.54	0.56
150	272.3	275.6	20.7	21.9	215.6	210.6	0.94	0.90	0.47	0.51
200	278.2	281.2	24.5	23.7	196.2	197.8	0.88	0.87	0.42	0.46
>200	280.6	283.2	29.5	26.7	191.6	194.0	0.85	0.82	0.41	0.42
South Direction										
0	223.6	227.1	15.9	17.9	224.4	220.5	1.11	1.07	0.65	0.63
50	252.6	255.6	16.8	19.6	222.3	235.8	1.05	1.02	0.61	0.59
100	268.6	273.1	19.6	21.6	219.6	214.6	0.98	0.98	0.57	0.54
150	276.6	277.6	21.5	23.2	212.9	206.1	0.93	0.95	0.52	0.48
200	281.9	283.9	24.7	25.5	196.0	195.8	0.87	0.87	0.48	0.44
>200	285.6	284.6	29.5	29.9	192.5	194.8	0.83	0.85	0.43	0.40
	SEM±	**LSD(5%)**	**SEM±**	**LSD (5%)**	**SEM±**	**LSD (5%)**	**SEM±**	**LSD(5%)**	**SEM±**	**LSD(5%)**
Location	3.5	NS	0.4	0.7	3.0	NS	0.01	NS	0.01	NS
Direction	3.8	NS	0.4	NS	3.3	NS	0.01	0.03	0.01	NS
Distance	3.8	11.0	0.4	1.2	3.3	9.5	0.02	0.04	0.01	0.03

and a minimum 221.7 kg ha^{-1} was recorded at 0 m in Moran soils and gradually increased with increase in distance from the spillage point (Table 2). The highest values were found in the unpolluted soils beyond 200 m (282.3 kg ha^{-1}). Similar results were obtained in the soils of Duliajan oil field. The values in the soils ranged from 224.9 kg ha^{-1} to 286.2 kg ha^{-1}. It was also observed that there was a decrease of about 21.0% and 21.4% in the nitrogen content of the crude oil polluted soils of Moran and Duliajan oil field, respectively. Presence of crude oil in the spilled soils consisted of high amount of carbonaceous compound which resulted in an imbalance in the C:N ratio at the spilled site compared to the control site which might caused nitrogen deficiency in oil soaked soil and retarded the growth of bacteria that utilizes carbon as a food source. (Atlas and Bartha 2005). Similar result was earlier reported by Jobson *et al.* (1974) stated that the C:N ratio greater than 17:1 in soils resulted in net immobilization of the nutrients by microbes leading to loss of soil fertility. Secondly, the increment in denitrifying bacterial population might cause temporal immobilization of this nutrient by the microbes (Shukry *et al.* 2013). Odu *et al.* (1985) explained that oil degrading or hydrocarbon utilizing microbes such as *Azotobacter spp*. normally became more abundant in the crude oil polluted soils while nitrifying bacteria such as *Nitrosomonas spp*. reduced in number. This might reduced the NO_3-N in crude oil contaminated soils and thus the available nitrogen content showed lower values with increased crude oil concentration in soils. Similar findings were also reported by Osuji and Nwoye (2007).

Available phosphorus

Availability of phosphorus in crude oil affected areas showed similar trend as that of available nitrogen. The available phosphorus content of Moran and Duliajan oil fields were presented in Table 3. The amount of available phosphorus decreased significantly with increased proximity

Table 3: Correlation coefficients of various chemical and available nutrients of soils of Moran and Duliajan oil fields

	pH	EC ($dS\ m^{-1}$)	OC ($g\ kg^{-1}$)	TPH ($mg\ g^{-1}$)	Aval.N ($kg\ ha^{-1}$)	Aval.P_2O_5 ($kg\ ha^{-1}$)	Aval.K_2O ($kg\ ha^{-1}$)	Ca (c mol (p+) kg^{-1})	Mg (c mol (p+) kg^{-1})
pH	1								
EC ($dS\ m^{-1}$)	0.612**	1							
OC ($g\ kg^{-1}$)	-0.666**	-0.891**	1						
TPH ($mg\ g^{-1}$)	-0.639**	-0.909**	0.912**	1					
Aval.N ($kg\ ha^{-1}$)	0.292*	0.519**	-0.518**	-0.558**	1				
Aval.P_2O_5 ($kg\ ha^{-1}$)	0.566**	0.757**	-0.795**	-0.810**	0.484**	1			
Aval.K_2O ($kg\ ha^{-1}$)	-0.460**	-0.664**	0.702**	0.687**	-0.198*	-0.545**	1		
Ca^{2+} (c mol (p+) kg^{-1})	-0.623**	-0.857**	0.835**	0.871**	-0.405**	-0.742**	0.670**	1	
Mg^{2+} (c mol (p+) kg^{-1})	-0.571**	-0.799**	0.836**	0.808**	-0.479**	-0.682**	0.673**	0.761**	1

to the drilling point. The values were recorded minimum at a distance 0 m at the spillage point in crude oil polluted soils of Moran (15.5 kg ha^{-1}) and Duliajan (17.5 kg ha^{-1}). While the highest available phosphorus in soils of Moran (29.5 kg ha^{-1}) and Duliajan (29.9 kg ha^{-1}) oil fields were observed in the unpolluted soils beyond 200 m. The lower concentration of available phosphorus in crude oil contaminated soils might be due to the following reasons. Firstly, the increased concentration of TPH in soil increased the concentration of carbon which might affected the equilibrium of nutrients in soils. The C: P ratio in soil increased and the microbes that utilized TPH as a carbon source could utilize available phosphate as an alternate source of nutrient while they degrade the hydrocarbons (Jobson *et al.* 1974, Braddock et al.1997, Wang et al. 2009). Secondly, phosphate solubility is maximum at pH 6.5 (Riser-Robert 1998) and consequently lower pH values in crude oil polluted soils could lower the available phosphorus in soils. The presence of high concentration of iron and calcium in crude oil polluted soils could possibly another factor that bound the phosphate ion and reduced its availability in soils. The results in the present investigation are in accordance with the above perception. It is important to mention here that available phosphorus concentration decreased by 47% in Moran and by 41 % in Duliajan oil field and the trend was similar to that of available N.

Available potassium

Soil available potassium increased with increased crude oil concentration and decreased distance. Available potassium was maximum near the drilling point whereas its concentration decreased beyond 200 m in both Moran and Duliajan oil fields (Table 2). The K_2O content of Moran oil field ranged from 190.6 to 224.4 kg ha^{-1}. Duliajan soils recorded the highest available K_2O content (222.8 kg ha^{-1}) at 0 m distance from the drilling point and lowest of 194.0 kg ha^{-1} beyond 200 m. This increase in the K_2O concentration near the spillage point might be due to the deposition of more potassium drilled out of the deeper soil layers during the crude oil drilling operation and got deposited in the upper soil layer near the drilling point. Moreover, oil refineries use sodium and potassium salts in processing operations which find their way to the nearby water bodies along with the effluents and thus ionic concentration may build up resulting in more potassium in crude oil contaminated soil (Deka and Devi 1994; Konwar and Jha 2010). It was observed that the available potassium content didn't have any significant difference with respect to the direction of sampling. The interaction effect between distance and direction were not significant for both the Moran and Duliajan oil fields.

Exchangeable calcium and magnesium

The amount of exchangeable calcium and magnesium in the crude oil affected areas of Moran and Duliajan oil fields were presented in Table 2. Results revealed that the amount of exchangeable Ca^{2+} and Mg^{2+} were maximum at a distance 0 m from the drilling point. The values of exchangeable Ca^{2+} and Mg^{2+} in the soils of Moran oil field ranged from 0.8 to 1.2 cmol (p+) kg^{-1} and 0.4 to 0.6 cmol (p+) kg^{-1} respectively. The lowest exchangeable Ca^{2+} and Mg^{2+} value were reported in the soils beyond 200 m. Similar trend was followed in the soils of Duliajan oil field. The exch. Ca^{2+} and Mg^{2+} in the soils varied from 0.8 to 1.2 cmol (p+) kg^{-1} and 0.4 to 0.6 cmol (p+) kg^{-1} respectively. The exchangeable Ca^{2+} and Mg^{2+} in both the oil fields, however, varied insignificantly with the direction of sampling. Increase in exchangeable Ca^{2+} and Mg^{2+} like that of available K^{+} in the crude oil contaminated areas might be attributed to the deposition of soils from deeper layer which possibly contained higher amount of calcium and magnesium. At the same time higher precipitation prevalent in the study area might have washed out the calcium and magnesium thus lower values of Ca^{2+} and Mg^{2+} in the unpolluted soils. Similar results were also reported by Ogboghodo *et al.* (2004).

CONCLUSION

It can be concluded from the study that the total petroleum hydrocarbon content is comparatively higher in the vicinity of drilling side and gradually decrease with the increase in distance. The presence of TPH in soil significantly affected chemical properties and fertility status of soil resulted in increased soil acidity and decreased available soil nutrients. Therefore, steps should be taken in order to minimize the spillage of crude oil from the drilling sites in order to restore the agricultural ecosystem and minimize livelihood risks of the local farmers.

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