



RESEARCH ARTICLE

SLUDGE CHARACTERIZATION OF KHARTOUM PETROLEUM REFINING WASTEWATER TREATMENT PLANT- KHARTOUM-SUDAN

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Manuscript Info

Manuscript History

Received: 19 May 2021
Final Accepted: 20 June 2021
Published: July 2021

Key words:-

Petroleum Sludge, Sludge Disposal, Toxic Waste, Wastewater Treatment Plant

Abstract

Clearance of sludge has made a major task in latest periods. In the current study sludge from KPRWTP were investigated concerning physicochemical characteristics & disposal options. Organic content of petroleum sludge & clay soil was $0.23 \pm 0.09\%$ and $0.186 \pm 0.1\%$, respectively. Metals average concentration: Chromium: 130.95 ± 27.32 mg/kg; Arsenic: 0.899 ± 0.434 mg/kg; lead: 47.83 ± 21.44 mg/kg; Cadmium: 18.33 ± 9.04 mg/kg; Copper: 209 ± 31.18 mg/kg; Zinc: 631 ± 159.57 mg/kg; Nickel: 6.6 ± 2.44 mg/kg; cobalt: 35.54 ± 10.08 mg/kg; manganese: 35.54 ± 10.08 mg/kg; iron: 501 ± 5490.04 mg/kg; titanium: 108.3 ± 18.33 mg/kg, and zirconium: 2.54 ± 0.246 mg/kg respectively. Sludge was made up of particles to some extent of bigger portion compared to soil (**sludge**: clay and silt 58.9%, sand 41.1%; **soil**: clay and silt 76%, sand 34%). Core constituents of petroleum sludge were Fe_2O_3 (38.45%), SiO_2 (8.55%), CaO (30.35%), MgO (0.85%). According to standards of (NJDEPSCC) As, Cd, Cu, Pb, Ni, & Zn have significantly lower concentration in sludge comparing guideline for residential and nonresidential utilization. On the other hand, FDEPSCCL showed Cu and Zn were only metals in sludge significantly have high concentration regarding guideline for residential applications only. The study confirmed that heavy metals under investigation were significantly lesser than quantities controlled by (TCLP) of (RCRA), with exception for chromium content.

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Introduction:-

The oilfield sludge is composed of organic materials and heavy metals with different scope of concentrations as stated by the American petroleum institute (API)(API,1989). Latest studies reported a great content of metals in produced sludge from refineries of petroleum stated that Chromium (480mg/kg), Nickel (480 mg/kg), lead (565 mg/kg), Copper (500 mg/kg), Zinc (1,299 mg/kg), Iron (60,200 mg/kg) (Da Rocha et al., 2010; Roldán et al., 2012; Marín et al., 2006; Admon et al., 2001).

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Disposed of petroleum sludge to the surroundings, generate risk associated with physicochemical characteristics of the adjacent environment, causing alteration in the morphological properties (Robertson et al., 2007). Thus, subsequent deficiency in vegetation growth and receiving nutrient uptake was noticed (Al-Mutairi et al., 2008).

Generation of huge quantities of sludge from the wastewater treatment, had the capacity to pollute soil, groundwater, & surface water and cause a risk to the natural resources and environment as a whole, because it contains detrimental level of contaminants including heavy metals (Stylianou et al., 2007). Disposal of metals is a great worry due to their being non-decomposable and their affinity to bioaccumulation. Thus, they can influence the health of living things and the quality of environment (Nair et al., 2008).

However, there are different technologies that concentrated sludge, and then decrease the bulks that we finally must dispose of. Moreover, sludge can be handled & stabilized, which can impart a low, but none-the-less marketable value to this waste. These technologies and practices do indeed constitute pollution prevention and waste minimization programs within water treatment plant operations. This driving us to a group of technologies that emphasis on: (A) thickening of sludge, (B) stabilization of sludge, (C) treatment and disposal of sludge. Therefore, sludge treatment is one of the most vital tasks in management of wastewater (Fytili and Zabaniotou, 2008). There are numerous techniques for treatment of sludge, each with benefits and disadvantages such as utilization of land (Shirani et al., 2010; Shomar et al., 2004), production of construction materials (Vieira et al., 2006; Silva et al., 2007), reagent of wastewater processing (Huanget al., 2011), dewatering of sludge (Ning et al., 2013), and landfills. To choose the appropriate method, it is significant the knowledge of the sludge properties. The aim of this work is to evaluate physicochemical sludge properties of a petroleum refining wastewater treatment plant (PRWWTP) in Northern part of Khartoum-Sudan.

Study Area

The studied Khartoum Refinery Company limited (KRC) located at Al Gaili town, about 80 Km, from Khartoum the Capital of Sudan, 12 Km from river Nile, 6 km from Altafhi high way, and beside the rail way. Wastewater of petroleum industry from the investigate area has been collected and pretreated, the treated water will be discharge into the oxidation pond and KRC Parks. The generated sludge has been collected from the tank of sedimentation and dumped outside the study area. The key treatment technology that been used is Cyclic Activated Sludge System (CASS) which is consist of the following stages.

Pre-treatment stage (Grid):

It's a mechanical stage for blocking the suspended substances in sewage it put before wastewater collection point and adjustment pond to hold up the big solid in order to avoid the bad influence to following treatment process.

Adjustment Stage:

It is a pond used for the adjusting the un-regular discharge and water quality of wastewater collection by setting the balance of water quality, and make the wastewater enter biological treatment stage with design capacity.

Biological treatment:

The biological treatment process of the system will be performed by Cyclic Activated Sludge System ponds, which have different function of primary sedimentation, aeration, secondary sedimentation all as one unit. It is an intermittent reaction system. In this pond active sludge process is running repeatedly between aeration and un-aeration period, the bio-chemical reaction and sludge water separation process will be also finished in the pond.

Materials and Methods:-

Sampling

Samples of sludge petroleum were obtained from refining plant of treatment wastewater (PRWWTP) in Northern part of Khartoum-Sudan (Fig. 1), 20 sludge samples were collected on weekly base in period from June –November 2014 from secondary sedimentation tank of the Cyclic Activated Sludge System ponds process. Standard of procedures for Water and Wastewater Examination was applied during transporting and storage of samples (APHA, 2005).

Characterization of sludge

Samples of petroleum sludge were obtained from refining wastewater treatment plant of Khartoum Refinery Company limited (KRC), secondary sedimentation tank (unit 3), Khartoum, Sudan. This Company hires a unit of chemical and biological treatment and utilize CaCO_3 and Fe_2SO_4 as coagulant. Sludge moisture and organic contents were determined, pH (781 pH meter, Metrohm, Switzerland), electrical conductivity (712 Conduct meter, Metrohm, Switzerland), were determined by APHA method (APHA, 2012). Regarding the heavy metal determination, five grams from sieved ground dried sludge sample was reacted with a blend of concentrated HCL and HNO_3 (1:3) for one day (24 hours) on a hot plate, then to make a 500 ml solution a volume of distilled water (500 ml) was boiled with the sample for two and half hours. Lastly, the obtained mixture was passed through 0.45 filter paper then the collected filtrate was used to evaluate the content of Chromium, Arsenic, Lead, Cadmium, Nickel, copper, and Zinc by using Atomic Absorption Spectrophotometer (AAS) (Shimadzu AA 6800) (Raposo et al., 2009; Juel et al., 2015; Saha and Hossain, 2011).

Statistical analysis

All samples analyses were repeated three times. The means and standard deviation ($\pm$ SD) were done according to SAS program (SAS, 1989). The different parameters of heavy metals and soil solution composition were explored using correlation.

Results and Discussions:-

Sludge characterization

Table 1 displays metal content & physicochemical characteristics of the sample from petroleum sludge and soil of Khartoum Refinery Company. The petroleum sludge and clay soil moisture content were $75 \pm 8.34\%$ & $15.06 \pm 3.56\%$, respectively. Organic content of both sludge and soil were $0.23 \pm 0.09\%$ and $0.186 \pm 0.1\%$, respectively. Most oilfield waters and refinery have pH between 4.0 and 8.0 while in Sudan where this study was carried it fluctuated from 7.0 to 8.0 (Ali et al., 2017). It is well known that calcium and iron mixtures solubility is approximately high dependent on pH of the medium, and the tendency of water to form scales is reduced at lower levels of pH, therefore, the calcium and iron precipitation in pipelines of Sudanese oil industry is predicted to be in high level (Nuha, 2013). The investigated sludge pH was slightly alkaline (8.65 ± 0.45). Electrical conductivity of the investigated petroleum sludge was high compared with other studies reported by some researchers (4, 15, and 16). It is well known that high electrical conductivity means that high salinity which create condition of incompatibility of sludge for application in land. Average concentration of the metals were: Chromium: 130.95 ± 27.32 mg/kg; Arsenic: 0.899 ± 0.434 mg/kg; lead: 47.83 ± 21.44 mg/kg; Cadmium: 18.33 ± 9.04 mg/kg; Copper: 209 ± 31.18 mg/kg; Zinc: 631 ± 159.57 mg/kg; Nickel: 6.6 ± 2.44 mg/kg; cobalt: 35.54 ± 10.08 mg/kg; manganese: 35.54 ± 10.08 mg/kg; iron: 501 ± 5490.04 mg/kg; titanium: 108.3 ± 18.33 mg/kg, and zirconium: 2.54 ± 0.246 mg/kg respectively (Table 1). Also this table shows the sequence of metals investigated in the petroleum sludge as: Zinc > Iron > Copper > Chromium > Titanium > lead > Cobalt > Cadmium > Manganese > Nickel > Zirconium > Arsenic, we noticed that the content of iron in the sludge approximately was high this attributed to the utilization of ferric chloride (FeCl_3) and ferrous sulphate (FeSO_4) during wastewater treatment processes as coagulant-flocculants and co-oxidation elements respectively. Sludge composition may differ from company to company reliant on the types of treatment procedures involved and chemicals used. Nevertheless, these metals had been the prevalent ingredient in the sludge of petroleum as reported in other research work. Previous studies showed that Chromium, Cadmium, and lead contents were high compared to the content of Nickel, Zinc, and Copper which were low and this due to the characteristics of the produced sludge (Wang et al., 2005).

Petroleum sludge and soil exhibit distribution of particle size and chemical composition as a ratio of available oxides are explained respectively in **Tables 2 and 3**. Sludge of petroleum was made up of elements atoms of a slightly bigger bulk portion compared to the soil (**sludge**: clay and silt 58.9%, sand 41.1%; **soil**: clay and silt 76%, sand 34%). The core constituents of sludge of petroleum were Fe_2O_3 (38.45%), SiO_2 (8.55%), CaO (30.35%), MgO (0.85%). The soil components (clay) that are used for manufacturing of brick was made up of comparable constituents that exist in the sludge of petroleum but in altered amounts. Consequently soil (clay) can be somewhat substituted by sludge of petroleum as raw substances for manufacture of construction materials.

Applications of sludge of wastewater treatment plant

Application of sludge in Industrial, Residential, and Nonresidential utilizations:

Wastewater treatment plant generated Enormous quantity of sludge daily, which is upturn the worries over their disposal and related costs. Table 4 explain the arithmetic outcomes of the judgment of the metals examined with the criteria resolved by New Jersey Department of Environmental Protection Soil Cleanup Criteria (NJDEPSCC) (NJDEP, 1999), and Florida Department of Environmental Protection Soil Cleanup Target Levels (FDEPSCCTLs) (FLDEP, 2006). According to standards of (NJDEPSCC) As, Cd, Cu, Pb, Ni, and Zn have significantly lower concentration in sludge with regards to guideline assigned for residential and nonresidential land usages. On the other hand chromium represent the exception metals since was significantly have higher concentration of Cr guideline assigned for residential and nonresidential land usage. In case of using this metals for industrial application both copper and zinc were significantly higher than the assigned guidelines for industrial utilization so were restricted for this application (Table 4).

On the other hand comparing our study results with Florida Department of Environmental Protection Soil Cleanup Target Levels (FDEPSCCTLs) it showed that Cu and Zn were only metals in sludge that significantly have high concentration regarding guideline assigned for residential applications only. Other metal factors have lower contents related with FDEPCTLs guidelines for direct contact of soil in both nonresidential and industrial utilizations. Therefore, all other investigated metals were suitable for utilization for nonresidential and industrial application since they have concentration significantly lower than that guidelines assigned for application.

Application of sludge in Building and construction utilization

Waste stabilization can be applied as a building substance providing that the substance owns the essential engineering characteristics and lacks lethal contaminants to an adequate level (Hassan et al., 2014; Rouf and Hossain, 2003). Treatment and stabilization of hazardous waste in building substances (concrete & brick) is by valorization which has been used in numerous conditions relatively effectively for the circumstances of tannery and sewage sludge (Hassan et al., 2014; Patel and Pandey, 2012; Cusidó and Cremades, 2012; Praveen et al., 2015; Chiang et al., 2009). However this practice of application of sludge in such substances does not essentially include interaction of chemicals among the waste produced & the substance, it has established to be an operational technique to decrease the possible risk of the waste by changing it into a less poisonous & movable however the activity may differ reliant on the sort of utilized waste product. Several research reports have revealed that sludge can be successfully steadied in building constituents for instance ceramic tiles, concrete, & other engineering tools (Montañés et al., 2014; Basegio et al., 2002; Giugliano and Paggi, 1985). The majority of these methods have not been upgraded at a level appropriate for treatment requests & industrial processing & are still possibly risky for the environment (Alibardi and Cossu, 2016).

The sludge of the current studied has a good potential for utilizing as a raw substance for manufacture of building materials. However, the present of heavy metals, which was above the required level, can limit its use for this determination. On other hand the Florida Department of Environmental Protection guidance provided a blending techniques, so that sludge can be blended with unpolluted clay soil to decrease the possible health risks from exposure to the sludge, providing that the subsequent blend was suitable for valuable utilization. To assess the suitable combined percentage (percentage of combined substance to sludge) for dropping the pollutants enclosed in the sludge. Florida Department of Environmental Protection (FDEP) endorses the following equivalence (FLDEP, 2006):

Ratio of blending = $\frac{A-B}{C}$ (1) (FLDEP, 2006).

B-C

Where,

A $\equiv$ amount of contaminant in the sludge, mg/kg,

B $\equiv$ target amount of the blended material, mg/kg,

C $\equiv$ amount of contaminant in the material used for blending, mg/kg.

Some of researchers found that small percentage of fine sludge from steel was useful to ceramic building out of the consideration of the content of heavy metals (Vieira et al., 2006). On the other hand, the technology of stabilization and solidification was used to improve the properties of the sludge from electroplating activities by producing substances which is beneficial for concrete block building (Silva et al., 2007).

Application of sludge in wastewater treatment plant

Utilization of sludge in the wastewater treatment plants should be well-thought-out under precaution given to investigation of nature of chemicals (compositions), sludge volume, and loading of solids which required to be taken in the accounts since it may disturb the capacity of wastewater solid items & rise the costs of operational and maintenance. Previous studies highlighted application of sludge as adsorbent materials for removal of contaminants, metals like phosphorus, and heavy metals (e.g., Pb and Cu) in the wastewater, in addition to that sludge give brilliant removal results when used as coagulant substances in refinery of wastewater from vegetable oil and can be comparable with aluminum sulfate and ferric chloride in its removal efficiencies (Basibuyuk and Kalat, 2004; Yang et al., 2006; Georgantas and Grigoropoulou, 2005; Wu CH et al., 2004). Also, there is some experience in applying sludge in removal of dyestuff substances from wastewater of both tannery and textile industries (Chu, 2001).

Application of sludge in Land use

Petroleum sludge has a serious environmental issue in view of its content of toxic substances including heavy metals (Mantiset al., 2005). The diverse content of sludge of petroleum formed at various handling plants and the differences between refineries requires understanding of the sludge chemical composition before the application on the land. Petroleum sludge properties rest on both the treatment process of wastewater and treatment of sludge. Commonly the sludge of petroleum is formed of heavy metals and organic materials with scope of thickness as stated by the American petroleum institute (API, 1989). Latest research works confirmed metal high content in sludge of petroleum from refineries was stated as Zinc (1,299 mg/kg), Iron (60,200 mg/kg), Copper (500 mg/kg), Chromium (480 mg/kg), Nickel (480 mg/kg), and lead (565 mg/kg) (Da Rocha et al., 2010; Roldán et al., 2012; Marín et al., 2006; Admon et al., 2001). Utilization of wastes in land require availability of organic and easily biodegradable materials to inhibit buildup of contaminants in the topsoil, it is essential to focus on the degree of contaminants in the sludge & the accumulation level of a contaminants spread over to the land. Commonly land uses normal techniques of waste use which can possibly improve the profile of soil (Pathak et al., 2009). Therefore only sludge with suggested level of content should be functional in land. Previously some researchers reported the potentiality of sludge from food industry in improving the productivity of soil by combined the vermin composting 30% cow manure (Yadav and Garg, 2009). They found that iron content was increased significantly in the final compost, while the contents of copper, manganese, and zinc in the mixture was in a decreasing rate (Yadav and Garg, 2009).

In the current research work, as stated before, in comparison with the limits of international standards such as New Jersey Department of Environmental Protection Soil Cleanup Criteria (NJDEPSCC) and Florida Department of Environmental Protection Soil Cleanup Target Levels (FDEPCTLs) for residential & nonresidential for utilization in land, Chromium, copper and zinc contents in the investigated sludge was greater which create inappropriate situation for land use.

Sludge dumping

This the final step in waste management which is to dispose the sludge in the dumping allocated area. Since such sludge usually contain hazardous materials, which can generate severe environmental harms in groundwater, it is essential to protect the under part of landfill with the suitable impermeable substances, such as clay or plastic liner (UNEP, 2002). Leaching can be evaluated by the toxicity characteristic leaching procedure (TCLP) which stated that if the pollutant content in toxicity characteristic leaching method was above the recommended scheduled in the Land Disposal Restrictions (LDR) of Resource Conservation and Recovery Act (RCRA), then the sludge is categorized as hazardous (USEPA, 2011). Figure 3 shows that the heavy metals under investigation were significantly lesser than the quantities controlled by toxicity characteristic leaching procedure (TCLP) of Resource Conservation and Recovery Act (RCRA), with exception for chromium content that was expressively great in the investigated sludge. Therefore, the considered sludge under this research work must be disposed in a Resource Conservation and Recovery Act subtitle C class landfill, which indicates that current practice of dumping sludge should be abandoned. Khartoum refinery company in Sudan adopt currently practice of management of the studied petroleum wastewater sludge is to release it, in the form of dry solid materials, in lined evaporation ponds in the site. In such case of disposal method, sludge cannot add to landfill leachate, to cause groundwater pollution, so the leachate cannot interfere with the food chain and cause health risk. These lined evaporated ponds could be utilized as an economic and harmless technique for the liquid from industries and disposal of sludge in dry environments (Al Yaqout, 2003).

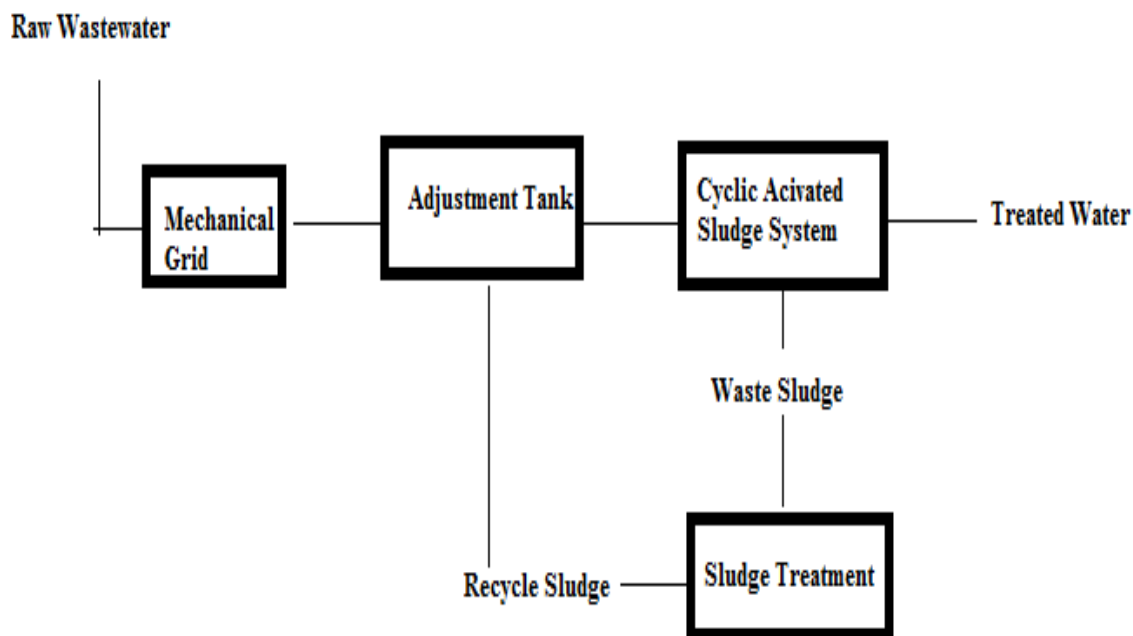


Figure 1:- Wastewater treatment plant.

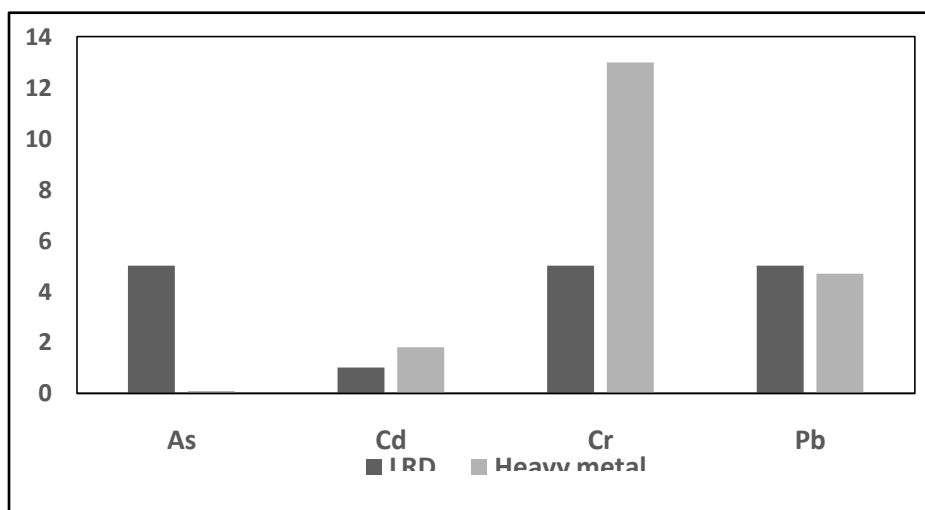


Figure 2:- Land Disposal Restrictions (LDR) of heavy metals of [Resource Conservation and Recovery Act \(RCRA\)](#).

Table 1:- Physicochemical properties samples of the sludge and soil of Khartoum petroleum Refinery.

Physicochemical Parameters	Petroleum sludge	Soil
pH	8.65±0.45	10.27±0.35
Electrical conductivity (EC) (µS/cm)	350.4±50.75	532.6±70.5
Moisture (%)	75 ± 8.34	15.06± 3.56
Organic content (%)	0.23 ±0.02	0.186±0.03
Ca, mg/kg	463.67±	-
Na (mg/kg)	1586.4±305.43	1642.5±345.43
K (mg/kg)	53.56±15.09	79.5±20.3
P (mg/kg)	0.24±0.09	0.40±0.10
Total metals contents (mg/kg)		
Cr	130.95±27.32	7.53±2.09

Pb	47.83±21.44	3.27±1.06
Cd	18.33±9.04	0.47±0.1
As	0.899±0.434	-
Co	35.54±10.08	5.5±2.11
Ni	6.6±2.44	12±4.33
Mn	20.5±0.75	184±30.22
Cu	209±31.18	180±28.33
Fe	501±5490.04	-
Ti	108.3±18.33	-
Zn	631±159.57	10±
Zr	2.54±0.246	-

Table 2:- Particle size distribution of sludge of petroleum and soil.

Particle size (µ)	Petroleum sludge (%)	Soil (%)
< 20	58.9	76
20-200	21.7	15
200-1000	10.3	9
1000-2000	05.4	-
>2mm	03.7	-

Table 3:- Chemical constituents of sludge of petroleum and soil (Dry weight, %).

Chemical components	Petroleum sludge (%)	Soil (%)
SiO ₂	8.55	55.98
Al ₂ O ₃	0.86	13.03
Fe ₂ O ₃	38.45	8.30
TiO ₂	0.65	1.54
CaO	30.35	4.37
MgO	0.85	1.77
K ₂ O	0.76	1.18
MnO	0.35	0.13

Table 4:- Statistics of the explored heavy metals with the criteria estimated by NJDEPSCC&FDEPSCCLs.

Heavy metal		New Jersey Department of Environmental Protection Soil Cleanup Criteria (NJDEPSCC) [26],						Florida Department of Environmental Protection Soil Cleanup Target Levels (FDEPSCCLs)[27],					
metal	mg/kg	Residential standards	P-value	Non-Residential standards	P-value	Ind	P-value	Residential standards	P-value	Non-Residential	P-value	Ind	p-value
Arsenic	0.899	20	<0.001a	20	<0.001a	12	<0.001a	2.1	<0.001a	12	<0.001a	12	<0.001a
Cadmium	18	39	<0.001a	100	<0.001a	22	<0.001a	82	<0.001a	1700	<0.001a	17x10 ²	<0.001a
Chromium	130	120	0.006b	20	0.159b	87	0.006b	210	<0.001a	470	<0.001a	470	<0.001a
Copper	209	600	<0.001a	600	<0.001a	91	<0.001b	150	0.077b	89000	<0.001a	89x10 ³	<0.001a
Lead	47	400	<0.001a	600	<0.001a	600	<0.001a	400	<0.001a	1400	<0.001a	14x10 ²	<0.001a
Nickel	6.6	250	<0.001a	2400	<0.001a	50	<0.001a	340	<0.001a	35000	<0.001a	35x10 ³	<0.001a

Zinc	631	1500	<0.00 01a	1500	<0.00 01a	3 6 0	0.008 b	440	0.045 b	11000	<0.00 01a	63x 10 ⁴	<0.00 01a
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P-value<0.05 is considered as significant.

(a)&(b) represent the calculated quantities which are lower and higher than the standards, respectively.

Conclusions:-

The current research work investigated the physicochemical characteristic of petroleum sludge derived from Khartoum Refining Wastewater Treatment Plant in Sudan. To recognize their performance in diverse condition with an objective to decide their appropriateness as a possible building solid for residential, nonresidential, and industries. The arrangement of the investigated metals in the sludge of petroleum was as follows: Zinc > Iron > Copper > Chromium > Titanium > lead > Cobalt > Cadmium > Manganese > Nickel > Zirconium > Arsenic, we noticed that the content of iron in the sludge approximately was high, this attributed to the utilization of ferric chloride (FeCl₃) and ferrous sulphate (FeSO₄) during wastewater treatment processes as coagulant-flocculants and co-oxidation elements respectively. The Petroleum sludge was made up of units of a slightly bigger bulk portion compare to the soil (sludge: clay and silt 58.9%, sand 41.1%; soil: clay and silt 76%, sand 34%). The core constituents of petroleum sludge were Fe₂O₃ (38.45%), SiO₂ (8.55%), CaO (30.35%), MgO (0.85%). According to standards of (NJDEPSCC) As, Cd, Cu, Pb, Ni, and Zn have significantly lower concentration in sludge with regards to guideline assigned for residential and nonresidential land utilization. On the other hand, comparing our study results with FDEPSCCLs it showed that Cu and Zn were only metals in sludge that significantly have high concentration regarding guideline assigned for residential applications only. The study also shows that the heavy metals under investigation were significantly lesser than the quantities controlled by (TCLP) of (RCRA), with exception for chromium content that was expressively great in the investigated sludge. Therefore, the considered sludge under this research work must be disposed in a subtitle C class landfill of Resource Conservation and Recovery Act, which indicates that present method of sludge dumping must be discarded.

Acknowledgement:-

The authors are thanks to Khartoum Refinery petrochemical company for support of this project.

Authors Contributions

1. M.H. ^(†) (Associate Professor) wrote the introduction and Results and discussions.
2. T. K. (Research Assistant) conducted experiment part.
3. A.M.A. (Assistant Professor) conducted data collection and wrote the experimental part.
4. G.A.M.O. (Professor) wrote development of sludge and revised the manuscript.
5. E.E.D. (Associate Professor) did statistical analysis and conclusions.

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