RESEARCH ARTICLE

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Characteristics and Disposal Options of Sludges from an Oil Refinery Wastewater Treatment Plant in Iran, 2013

Mehdi Ahmadi[a,b], Zahra Tamimi[c], Nemat Jaafarzadeh[a,b], Pari Teymouri[d,e], Rohangiz Maleki[a,b]

[a]Environmental Technologies Research Center, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran.
[b]Department of Environmental Health Engineering, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran.
[c]Department of Environmental Sciences, Ahvaz Science and Research Branch, Islamic Azad University, Ahvaz, Iran.
[d]Environmental Health Research Center, Kurdistan University of Medical Sciences, Sanandaj, Iran.
[e]Department of Environmental Health Engineering, Kurdistan University of Medical Sciences, Sanandaj, Iran

***Correspondence** should be addressed to Ms. Pari Teymouri, **Email**: Pari.Teymouri@yahoo.com

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Background & Aims of the Study: Industrial wastewater sludges must be disposed in a safe way because they have hazardous effects on the human and environment. The aim of this study is to investigate the physicochemical characteristics and disposal options of sludges from oil-water separator (OWS) and dissolved air flotation (DAF) clarifier of a Refinery wastewater treatment plant.

Materials & Methods: Sludge samples were collected in grab sampling manner, in 6 month (April-September 2013) in order to be analyzed for their physicochemical characteristics. Kolmogorov-Smirnov Z, independent t-test, Mann-Whitney U test, one sample t-test and Wilcoxon signed rank test were used for statistical analysis. Canadian Soil Quality Guidelines (CSQG) and Florida Department of Environmental Protection Soil Cleanup Target Levels (FDEPSCTLs) were used to discuss the disposal fate of the generated sludge.

Results: As, Cd, Cu, Pb and Se were not detected in the studied sludge. As compared with CSQG, the investigated sludge were polluted for residential/parkland, agricultural, commercial and industrial applications, because they contained high concentrations of Cr, Ni and Zn. Also, according to FDEPSCTLs, the studied sludges were not suitable for residential and non-residential applications due to their high Al and Ni contents. DAF sludge had a high Zn concentration for residential application, too.

Conclusions: present sludge management in the studied plant needs to be revised because metals' concentrations are above the international standards and guidelines.

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Background

Worldwide increase in environmental concerns has led to its inclusion in concepts such as sustainable development, in which ecological, economic and social aspects of environmental issues are considered (1). Sludges are unwanted and unenviable residues produced in wastewater treatment process (2). Sludges need to be handled and disposed in a carefully controlled manner because they contain pollutants such as heavy metals with negative effects on human and environment, e.g. nonbiodegradability and bioaccumulation (3). Therefore, countries adopted strict regulations on environmental issues of sludges from industrial wastewater treatment processes (4). Such sludges can be managed in various ways. For example they can be used for manufacturing of construction materials (5,6) or in agricultural lands (7,8). They also can apply as wastewater treatment reagent (9) or sludge dewatering reagent (10). Finally, they can be land filled. Some advantages (from economic and environmental aspects) and some limitations, (complexity of the methods and pollutants problems in the sludges) have been introduced for the mentioned sludge management methods. Therefore, to select the most appropriate method, it is necessary to know the sludge characteristics.

Sludge generated in refineries from water–oil separators is a complex mixture of compounds, which many of them are carcinogenic, toxic and mutagenic and have been classified as primary environmental pollutants by US Environmental Protection Agency (11).

Aims of the study:

Present study aimed to determine the physical and chemical properties of sludges from oil-water separator (OWS) and dissolved air flotation (DAF) clarifier of a refinery plant in southwestern Iran. A comparison was made between the heavy metal concentrations in the studied sludges and Canadian Soil Quality Guidelines (CSQG) for the Protection of Environmental and Human Health (12) and Florida Department of Environmental Protection Soil Cleanup Target Levels (FDEPSCTLs) (13). Finally, some of disposal methods were discussed.

Materials & Methods

Study site:

The studied refinery wastewater treatment plant locates in southwestern of Iran. It has been formed of three physical-chemical, biological and sludge thickening units. This study considered the sludges from primary treatment (oil-water separator (OWS) and dissolved air flotation (DAF) clarifier) (Figure 1)

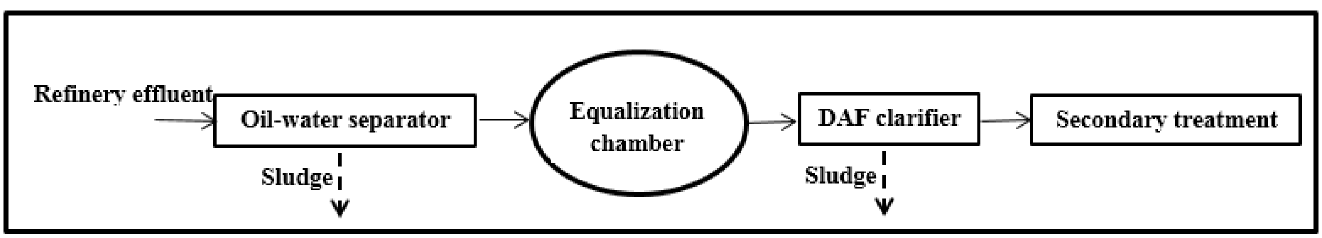

Figure 1) Primary treatment of the studied refinery wastewater treatment plant

Sludge from oil-water separation is mixed with clean soil in the ration of 3 to 1. Then, it is discharged on land surrounding the city. Sludge generated in DAF unit, after thickening, is used as a fertilizer supplement. The studied sludges are disposed as the mentioned manners, without any analysis on their components and any consideration of public health and environmental sustenance.

Sampling:

To analyze chemical characteristics, 9 slurry sludge samples were collected, in grab sampling manner, in 6 month (April-September 2013). 6 out of 9 slurry samples were filtered. Filtered sludges were analyzed for their heavy metal contents. Standard Methods for the Examination of Water and Waste water were applied for samples transportation and storage (14).

Sample preparation and digestion for heavy metal analysis:

After filtering and separation of liquid phase, 1 g of sludge (on the filter) was incinerated at 500˚C. The resulted ash digested with a mixture of concentrated HCl and HNO_3 on a hot plate. Then, the cooled suspension was filtered through an ashless filter paper and the filtrate was diluted to a volume of 100 ml with deionized water.

Analytical methods and instruments:

Some physicochemical characteristics including Total solids (TS), total suspended solids (TSS), total dissolved solids (TDS) (dried at 103–105 °C), fixed and volatile solids (ignited at 550 °C), pH (pH meter HACH, USA), conductivity (conductivity meter HACH, USA), turbidity (Nephelometric method, HACH 2100 AN, USA) and total organic carbon (TOC) (TOC-Analyzer, Analytik Jena, Germany) were determined. Al, As, Co, Cr, Fe, Mn, Ni, Se and Na contents in the sludge samples were measured by an atomic absorption spectroscopy (analytikajena model Contra A 3W, Germany). Cd, Cu, Pb and Zn concentrations were determined by a PolarographyVoltammetry (797 VA Computrace, Switzerland), and Ca and Mg in the sludge samples were determined using titration method.

Metals content were determined using both Standard Methods for the Examination of Water and Waste water and ASTM (14,15) and other parameters were measured according to the former (14). All chemicals used in this study were of analytical grade and Merck Chemical Company (Germany) products.

Statistical analysis:

Normality of the data was assessed using Kolmogorov-Smirnov Z analysis. Then, independent t-test (for normal data) and Mann-Whitney U test (for non- normal data) were used to compare the data of two sludges. One sample t-test (for normal data) and Wilcoxon signed rank test (for non- normal) were applied to compare metals concentration with the standard values for soil clean-up levels of CSQG and FDEPSCTLs because there are not such standards, regulations, or legal restrictions available in Iran.

Results

Physicochemicalcharacteristicsofsludge samples:

Table1 shows the result of physicochemical characteristics and statistical results of both OWS and DAF sludges. The results of Kolmogorov-Smirnov Z test showed that distributions of all parameters were non-normal. Thus, Mann-Whitney U test were used for them.

Table 1) Physicochemical characteristics and statistical results of slurry and cake sludge

Parameter/unit	sludge	number of Samples	Mean±SD	Median	P_{Valu}
VSS (%W)	OWS	9	69.15±9.53	65.45	<0.000
	DAF	9	92.82±2.87	92.47	
FSS(%W)	OWS	9	30.85±7.18	34.06	<0.000
	DAF	9	7.18±2.86	7.53	
TSS (mg/l)	OWS	9	17920.67±7589.48	17900.00	0.605
	DAF	9	23518.89±2.86	65.45	
TDS (mg/l)	OWS	9	6332.33±4627.85	5330.00	0.796
	DAF	9	6088.33±4429.01	4360.00	
EC (µs/cm)	OWS	9	5057.78±2795.76	4220.00	0.863
	DAF	9	4870.00±2735.36	4040.00	
pH	OWS	9	7.70±0.52	7.59	0.605
	DAF	9	7.38±0.29	7.40	

*P-value<0.05 is considered as significant

VSS and FSS in the DAF sludge were significantly higher and lower than OWS sludge, respectively. TSS, TDS, EC and pH variations in both sludges were not significant compared to each other (p>0.05).

Metal content of the sludge samples:

Table 2 represents the metal content of the studied filtered sludges. According to Table 2, no As, Cd, Cu, Pb and Se were detected in them. The orders of the studied metals in both sludges were as follows:

OWS: Fe>Al>Ca>Ni>Zn>Mg>Mn>Na>Cr>Co
DAF: Al>Fe>Ni>Zn>Ca>Mg>Mn>Cr>Na>Co

Table 2) Metal content of filtered sludge

Metal	sludge	number of Samples	Mean±SD	Median	P_{Value}
As	OWS	6	N.D	N.D	-
	DAF	6	N.D	N.D	
Al	OWS	6	15616.08±6294.89	13387.44	0.650^M
	DAF	6	94770.95±75930.92	66993.83	
Ca	OWS	6	629.67±264.30	629.00	0.530^T
	DAF	6	720.56±331.69	562.00	
Cd	OWS	6	N.D	N.D	-
	DAF	6	N.D	N.D	
Co	OWS	6	10.51±0.94	10.50	0.015^T
	DAF	5	7.678079±1.91	6.93	
Cr	OWS	6	52.98±20.70	56.84	0.002^M
	DAF	6	246.63±260.15	130.20	
Cu	OWS	6	N.D	N.D	-
	DAF	6	N.D	N.D	
Fe	OWS	6	28390.98±2700.12	27650.98	0.385^T
	DAF	6	39446.28±29659.78	46294.24	
Mg	OWS	6	354.33±136.40	324.00	0.950^T
	DAF	6	358.44±138.62	352.00	
Mn	OWS	6	327.96±59.63	317.40	0.396^T
	DAF	6	267.91±150.39	255.87	
Na	OWS	6	251.23±137.44	287.55	0.605^M
	DAF	6	196.08±120.28	147.32	
Ni	OWS	6	374.31±215.29	290.65	0.065^M
	DAF	6	6177.79±4792.23	7350.87	
Pb	OWS	6	N.D	N.D	-
	DAF	6	N.D	N.D	
Se	OWS	6	N.D	N.D	-
	DAF	6			
Zn	OWS	6	367.90±30.66	369.12	0.002^M
	DAF	6	955.66±34.95	956.08	

Unit of all metal content is mg/Kg
*P-value<0.05 is considered as significant
M and T stands for Mann Whitney U test and T-Test, respectively

Fe in the OWS and Al in the DAF sludges had the highest concentration amongst the studied metals. Ni had the first ranks amongst the heavy metals and Co was the one with the lowest concentration. Comparing the metals' concentrations, it was found that Co in OWS sludge was significantly higher than DAF sludge (P<0.05). On the other hand, Cr and Zn were the metals with significantly higher concentration in DAF sludge than OWS sludge (P<0.05).

Comparing the studied heavy metals with the guidelines determined by CSQG and FDEPSCTLs showed that according to CSQG, Cr, Ni and Zn concentrations in the studied DAF sludge, and Ni and Zn in OWS sludge were significantly higher than the permitted values for residential/ parkland, agricultural, commercial and industrial applications (Table 3). Al and Ni concentrations were also significantly higher than FDEPSCTLs standards for residential and nonresidential applications. Table 3 also represents that compared to FDEPSCTLs, Zn concentrations in the sludge from DAF unit were significantly beyond the standards for residential applications, which restricts sludge's use for such applications.

Table 3) Statistical results of comparing the investigated heavy metals with the standards determined by CSQG and FDEPSCTLs

Heavy metal	Sludge type	CSQG								FDEPSCTLs soil direct exposure			
		Residential/ parkland		Agricultural		Commercial		Industrial		Residential		Non Residential	
		Standard (mg kg^{-1})	P_V^*	Standard (mg kg^{-1})	P_V^*	Standard (mg kg^{-1})	P_V^*	Standard (mg kg^{-1})	P_V^*	Standard (mg kg^{-1})	P_V^*	Standard (mg kg^{-1})	P_V^*
Al	OWS	-	-	-	-	-	-	-	-	80000	0.028$^{\downarrow W}$	-	-
	DAF		-		-	-	-	-	-		0.654$^{\uparrow O}$		-
Co	OWS	40	<0.000$^{\downarrow O}$	50	<0.000$^{\downarrow O}$	300	<0.000$^{\downarrow O}$	300	<0.000$^{\downarrow O}$	-	-	-	-
	DAF		<0.000$^{\downarrow O}$		<0.000$^{\downarrow O}$		<0.000$^{\downarrow O}$		<0.0001^{O}		-		-
Cr	OWS	64	0.249$^{\downarrow O}$	64	0.249$^{\downarrow O}$	87	0.010$^{\downarrow O}$	87	0.010$^{\downarrow O}$	210	<0.000$^{\downarrow O}$	470	0.075$^{\downarrow W}$
	DAF		0.028$^{\uparrow W}$		0.472$^{\uparrow W}$		0.028$^{\uparrow W}$		0.028$^{\uparrow W}$		0.463$^{\downarrow W}$		<0.000$^{\downarrow O}$
Fe	OWS	-	-	-	-	-	-	-	-	53000	<0.000$^{\downarrow O}$	-	-
	DAF		-		-		-		-		0.314$^{\downarrow O}$		-
Mn	OWS	-	-	-	-	-	-	-	-	3500	<0.000$^{\downarrow O}$	43000	<0.000$^{\downarrow O}$
	DAF		-		-		-		-		<0.0001^{O}		<0.000$^{\downarrow O}$
Ni	OWS	50	0.024$^{\uparrow O}$	50	0.024$^{\uparrow O}$	50	0.024$^{\uparrow O}$	50	0.024$^{\uparrow O}$	340	0.712$^{\uparrow O}$	35000	<0.000$^{\downarrow O}$
	DAF		0.026$^{\uparrow O}$		0.026$^{\uparrow O}$		0.026$^{\uparrow O}$		0.026$^{\uparrow O}$		0.031$^{\uparrow O}$		<0.000$^{\downarrow O}$
Zn	OWS	200	<0.000$^{\uparrow O}$	200	<0.000$^{\uparrow O}$	360	0.556$^{\uparrow O}$	360	0.556$^{\uparrow O}$	440	0.002$^{\downarrow O}$	11000	<0.000$^{\downarrow O}$
	DAF		<0.000$^{\uparrow O}$		<0.000$^{\uparrow O}$		<0.000$^{\uparrow O}$		<0.000$^{\uparrow O}$		<0.000$^{\uparrow O}$		<0.000$^{\downarrow O}$

*P-value<0.05 is considered as significant.
$^{\uparrow}$ and $^{\downarrow}$ indicate that measured amounts are higher and lower than standards, respectively.
O and W are p-values from one sample t-test and Wilcoxon signed rank test, respectively.

Discussion

Physicochemical characteristics and metal content of sludge samples:

Compared to TSS, TDS, EC and pH in sludges from a still mill wastewater treatment plant (16) and a petrochemical wastewater treatment plant (17), TSS in the studied sludges was higher; TDS was higher than sludge from still mill sludge (16) and lower than petrochemical sludge (17). Also, pH in this study was almost in the same range and lower than its range in sludges from still mill (16) and petrochemical treatment plants (17), respectively. The EC amount for studied DAF and OWS sludges were similar and higher than that of reported by Ahmadi et al. (16), for a still mill treatment plant sludge, respectively.

Karamalidis and Voudrias (18) in their study investigated a mixture of different kinds of oily sludge, such as API type gravity separator sludge and DAF sludge. Comparing our results with those of Karamalidis and Voudrias it was observed that in our studied sludge Cu, Fe, Pb and Zn were lower, and Co, Cr and Ni were higher than theirs. Since sludges had different nature and origins, such dissimilarity in the results can be expected.

Disposal options of the studied sludges:

Sludge quality requirements should consider sludge management methods, disposal and reuse practices (19). Following options have been suggested for sludge disposal and reuse:

Land use:

Solid waste management by land use method restricts disposal of hazardous or toxic wastes in upper soil zone, because they can accumulate in the soil. Therefore, to use this method, it is necessary to consider the pollutants contents in the sludge and their loading rate applying to the land. If pollutants' concentrations were within the recommended concentrations, then, sludge can be used on land (20).

In this study, as compared with CSQG standards for park land and agricultural use, Cr, Ni and Zn concentrations in the studied sludges were higher, making them unsuitable for land application. But, sludge from DAF unit, after thickening, is used as a fertilizer supplement, which means that its application should be limited. Shumar et al. (7) studied on sewer and industrial sludge samples from Gaza strip. They reported that samples were clean of heavy metals, except for Zn that in 85% of samples exceeded the standards for land use. Ahmadi et al, in 2013 and 2014 reported the exceeding of heavy metals in their studied sludges from the guidelines for land application (16,17).

Residential, commercial and Industrial uses:

Table 3 represents that Ni had concentrations significantly higher than CSQG and FDEPSCTLs for all kinds of applications. Compared with CSQG, Cr and Zn both have restrictions to be used for residential, commercial and industrial purposes. Zn also was significantly higher than FDEPSCTLs for residential applications. Some residential and non-residential applications of sludges have been discussed below:

Using as building and construction materials:

Resulted ash from sludge incineration, contains high amounts of Fe, Al, Ca and Si. Therefore, it can be used as a raw material for manufacturing of brick, tile, block and other construction materials (21). Metal content analysis (Table 2) showed that Fe, Al and Ca have the first to third rank in the studied OWS sludge. Al and Fe were the metals with the highest concentration in DAF sludge. Therefore, the studied sludges are potentially good to be used as a raw material for manufacturing of construction materials. But, it should be noted that their metal content is above the standard values and their application should be restricted. To solve this problem and reduce negative health effects, FDEP recommends blend ratio, (Eq. 1), to blend such sludges with a proper amount of uncontaminated soil, and the resulted mixture is appropriate for the purpose (13).

$$\text{Blend Ratio}=\frac{(A-B)}{(B-C)} \qquad (1)$$

Where, A = concentration of contaminant in the sludge, mg/kg, B = target concentration of the blended material, mg/kg, C=concentration of contaminant in the material used for blending, mg/kg.

Karamalidis et al (18), in their work investigated the release of Zn, Ni, Cu and CrO_4 2- as a function of pH from a cement based stabilized/solidified refinery oily sludge and its ash. Their results showed that 98% of good immobilization was obtained for metals of solidified ash at pH values greater than 6 and 93% of solidified oily sludge at pH values higher than 7. Although, they did not compare their results with any standard or guideline. In another study by Silva et al. (5), stabilized/ solidification technology was used to treat sludge from an electroplating industry. The resultant product was used as a raw material to build concrete block. The resultant block had a very low metal leachability and its solubility was low, too, indicating a low environmental impact.

Using in wastewater treatment:

Sludge can be used in waste water treatment process as it a coagulant agent in wastewater treatment (9) as an agent for sludge dewatering (10).

Other uses of sludge:

Oil sludge contains large amount of combustionables with high heating values, make it a useful recycling resource for energy by its converting to fuel oil. For example, Shie et al. (22) used oil sludge from refinery plant as a raw material for pyrolysis.

Landfilling:

Landfilling is the final option for those sludges with no reuse or recycling chance. Proper lining materials such as clay or plastic liners must be used for such landfills to protect contamination of ground waters by hazardous materials in sludges (23).

As mentioned before, sludge from oil-water separation in the studied refinery plant, is mixed with clean soil in the ration of 3 to 1 and discharged on land surrounding the city. Al Yagout (24) recommended that lined evaporation ponds are economic and safe in handling industrial liquid and sludge in arid climates.

Conclusion

In conclusion, present sludge management in the studied plant needs to be revised. It also needs to be said that there are some other parameters, including different organic and inorganic compounds, in CSQG and FDEPSCTLs than those investigated in this study. We suggest investigation of these other parameters in future studies.

Footnotes

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Conflict of Interest:

The authors declared no conflict of interest.

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