

## Investigation of Some Qualitative Characteristics of Sludge Produced in Hamadan Wastewater Treatment Plant and Its Application in Agricultural Lands



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Published online June 29 2019



#### Abstract

In this study, qualitative characteristics of the sludge produced in Hamadan wastewater treatment plant and the feasibility of its application in agricultural lands were investigated. Samples were taken from the primary and secondary sludge and indices such as VS/TS, PH, TKN, COD, TKN, SOUR, Na, Ca, SAR, heavy metals and biological properties (the mean fecal coliform count and the number of parasitic eggs) were measured. The results were compared with the United States Environmental Protection Agency (US EPA) standards (40 FCR-503). The results showed that the VS/TS ratio of the raw sludge was 0.8, 0.55, and 0.55 and that of the secondary sludge was 0.65, 0.28, and 0.32 for fall, winter, and summer, respectively. The average concentration of COD, TKN, SOUR, Na, and Ca for the initial sludge was 51283, 107, 0.50, 609, and 952 and for secondary sludge, it was 35595, 81, 4.90, 306, and 493 mg/L, respectively. The MPN for primary and secondary sludge was determined to be  $19.83 \times 10^6$  and  $186 \times 10^6$ , respectively and the average number of parasite eggs in primary and secondary sludge was 7.05 and 7.2 in 4 g of dry solids. Entamoeba coli had the highest number of parasite. The results of this study showed that the highest concentration of heavy metals in the sludge was 21396 mg/kg. The values obtained for the above-mentioned indices and heavy metals were in standards range. The results of this study show that none of the primary and secondary sludge samples has been properly stabilized. The values obtained for biological characteristics of sludge were not in the standard range. Therefore, it was revealed that the use of sludge for agriculture needed more stabilization.

**Keywords:** Sludge characteristics, Sludge discharge, Sludge management, Sludge application, Heavy metal

Received December 24, 2018; Revised May 25, 2019; Accepted June 2, 2019

## 1. Introduction

In the treatment process of various types of wastewater, millions of tons of sludge are annually produced worldwide (1). Uncontrolled sludge contains various types of contaminants including bacteria, viruses, and parasites, as well as heavy metals and toxic organic compounds that can threaten the health of humans and other creatures through contaminating the agricultural products and the environment. Therefore, it is necessary to consider the rules and regulations for the disposal and reuse of sludge (2). The United States Environmental Protection Agency (US EPA)promulgated 40 CFR Part 503 which was published in 1993. It included the standards for sewage sludge applied to the land, placed on a surface disposal site, or fired in a sewage sludge incinerator (3). Sludge treatment and stabilization process account for about 40%-50% of total wastewater treatment costs. However, the reuse of treated sludge with the observance of the health aspects can offset some of the costs (4).

Sludge management includes pre-treatment, minimization, stabilization, burning, composting and sanitation. By taking a look at the history of the use and management of sludge in industrialized countries, it can be seen that the utilization and economic use of sludge for different purposes such as agricultural applications have developed in most of these countries. According to Table 1, in some European countries and the United States, more than 40% to 60% of sludge is used in agricultural

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Country	Dry Sludge		Sludge Managem	nent Methods (%)	
	Production (t/a)	Agriculture	Burning	Disposed	Other Methods
Spain	23 000	61	-	10	29
Denmark	150000	43	28	29	-
Sweden	180000	60	-	40	-
Netherlands	180000	53	10	29	8
Australia	250000	28	37	35	-
Switzerland	250000	50	20	30	-
Portugal	300000	80	-	12	8
Italy	800 000	34	11	55	-
France	900 000	27	20	53	-
Britain	1 500 000	51	5	16	28
Germany	2750000	25	10	65	-
USA	7 690 000	45	3	21	30

Table 1. Produced Sludge Volume and the Contribution of Each of the Common Methods to Sludge Management in Some Countries in 2012 (4,5)

Table 2. Study Variables

Parameters	Unit	Measurement Method
рН	-	Paper pH meter with glass electrode (4500-H)
Nitrogen	mg/L	Total Kjeldahl Nitrogen (4500-N A)
COD	mg/L	Closed, 5220 D
Calcium ion	meq/L	Titration (EDTA) (3500-Ca D)
Sodium ion	meq/L	Flame Publishing (FT728)
SOUR	mg/L	BOD Bottle and DO Drop (1683)
VS	mg/L	Furnace (2540 A)
TS	mg/L	Furnace (2540 B)
Egg parasites	MPN/100ml	Formalin ether concentration technique (1623)
MPN (TC)	MPN/100ml	Lactose (9 tubes) (1680)
E. coli (FC)	MPN/100ml	Lactose (9 tubes) (1680)

land (5).

In Japan, due to lack of land, burning is used more than other disposal methods (5,6). In Iran, sludge of municipal wastewater treatment plants is often used for fertilization of agricultural fields and gardens (6). However, studies by Farzadkia (7) and Farzadkia and Taherkhani (8) in different cities of the country indicate that there are no standards for sludge disposal or desired costs in previous years.

Therefore, the aim of this study was to determine the physico-chemical and biological properties and the concentration of heavy metals in the sludge of municipal wastewater treatment plant in Hamadan, compare the results with global standards, and to assess the feasibility of its use for agricultural purposes.

## 2. Materials and Methods

### 2.1. Sampling

This descriptive-analytic study was carried out on sludge of urban wastewater treatment plant in Hamadan in 2014 during summer, fall, and winter. Currently, municipal wastewater treatment plant in Hamadan covers a population of 250 000 people, equivalent to 55 000 cubic meters per day. The treatment of the municipal wastewater is mainly carried out through aerobic/anoxic method. In this study, the parameters of TS, VS, pH, N, Na, Ca, COD, SOUR, MPN, *E. coli*, eggs of parasites, and heavy metals were evaluated in primary and secondary sludge. The sampling was carried out in fall, winter, and summer, according to the methods of analysis mentioned in Table 2, based on the *Standard Methods for the Examination of Water and Wastewater*.

The results were compared with US EPA standards for the use or disposal of sewage sludge (40 CFR 503) and other valid standards.

In this study, the samples were also examined in terms of the presence of heavy metals (11 heavy metals).

All chemicals used in this study including mercury sulfate, silver sulfate, potassium dichromate, sulfuric acid, potassium hydrogen phthalate, filter paper, potassium sulfate or selenium sulfate, boric acid, methylene blue dye, methylene red dye were provided from Merck Company, Germany.

## 2.2. Analysis

For the analysis and measurement of heavy metals, the ICP-OES (Inductively coupled plasma optical emission spectroscopy) device manufactured by Australia (VistaPro, Vari) was used.

### 3. Results and Discussion

The results are discussed in four aspects to evaluate the quality of sludge produced in the treatment plant and to compare it with standards for use in agriculture.

#### 3.1. Investigation of Physical Quality of Wastewater Sludge

The average total and volatile solid concentrations of the samples in the three seasons of summer, fall, and winter are presented in Table 3. According to Table 3, the VS/TS

Table 3. TS and VS Values of Primary and Secondary Sludge (mg/L)

Type of Sludge	Itom	Summer	Fall	Winter	Average	Average
Type of Studge	ittem				(mg/L)	(%)
Primary sludge	TS	1115	1020	1660	1265	100
	VS	620	820	920	786.7	62.2
Sacandan, dudaa	TS	1165	400	380	648.3	100
Secondary sludge	VS	380	260	110	250	38.6

ratio for the primary sludge are 0.8, 0.55 and 0.55 and for secondary sludge, are 0.65, 0.28, and 0.32 for fall, winter, and summer, respectively. Based on these results, it was indicated that the sludge was not sufficiently stabilized in the treatment plant (the maximum allowable level is 0.6) (9) and if it is added to the soil, the desired results would not be achieved. In a study conducted on the physical and chemical properties of the sludge of Gaza treatment plant, El-Nahhal et al found that the application of sludge to the soil may alter its physical and chemical properties (10). In another study, Bahremand et al studied the effect of the sewage sludge on the physical properties of the soil after adding it to the soil at four levels of 0, 25, 50 and 100 tons/ha in Najafabad, Isfahan province. The results of this study showed that adding the sludge to soil has a favorable effect on the physical properties of the soil (11).

## 3.2. The Investigation of Chemical Quality of Wastewater Sludge

The values of the chemical properties of the sludge in different seasons are presented in Table 4. According to Table 4, the mean nitrogen concentration (in the form of TKN) in primary and secondary sludge was determined to be 107.28 and 81 mg/L, respectively, and the mean sodium concentration was determined to be 609.16 and 305.66 mg/L, respectively. The amount of potassium in sludge is usually low (between 0.22 and 2.64% of its dry weight). Therefore, the nutritional value of the sludge is due to its phosphorus and nitrogen content (12). In a study conducted on the sludge produced in the water treatment plant by Rosana et al. (2004), the nitrogen and phosphorus concentrations were found to be 19.9 and 54 mg/L (13). Moreover, Torabian et al reported that the mean nitrogen, phosphorus and potassium contents were 5403, 2625, 874 mg/kg dry sludge, respectively (14). Moreover, in another study, Bina et al reported the mean carbon content of 50-80% (15). Recently, the EPA has focused on nitrogen concentration in produced sludge (4). The groundwater is rich in phosphorus and nitrogen due to the passage of waters through the sludge. Therefore, limiting the use of sludge has been extensively considered to prevent the phenomenon of eutrophication. In developing countries, a number of reports have been published that indicated the severe pollution of shallow wells through sludge wells. Problems caused by phosphorus arise when the soil becomes saturated with phosphorus because phosphorus has less movement in soil and it is quickly absorbed by

particles of soil or organic matter (4). A study by Taheri et al also confirms the above-mentioned results (16). The average SOUR in aerobic stabilized sludge is about 2 mg  $O_2/g$  VS.h (9). As shown in Table 4, the average SOUR for the sludge stabilized in this study is 4.9 mg  $O_2/g$ VS.h which suggests the lack of stabilization of sludge in aerobic conditions. It seems that sludge has not been well established in this regard (17). Typically, the pH range of sludge in wastewater treatment plants is 6-9. According to Table 4, the average pH for primary sludge is 5.64 and it is 5.82 for secondary sludge. The pH values obtained in the study (the pH below 7) and the nature of the sludge indicate the absence of sludge stabilization (17). This pH range disturbs the growth and proliferation of microorganisms, as well as the treatment and stabilization of the sludge. The use of sewage sludge significantly reduces the pH of the soil which leads to an increase in the potential for the absorption of heavy metals and their movement in the soil profile. Therefore, measures should be taken in the long-term applications of this sludge to increase the soil buffering capacity. Scientists have proposed different sets of characteristics that affect soil quality and determined the soil quality index based on the total set of characteristics affecting soil quality. In addition, a limited number of soil properties, which provide a better representation of soil quality, have been suggested as a set of minimum properties that affect soil quality (18). Ions of Na<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup> are the dominant exchange ions in the soils of arid regions. The sodium adsorption ratio (SAR) has been mentioned as an effective

lable 4. Chemical Properties of Sewage Sludge in Hamadan (mg	in Hamadan (mg/	Sludge in	Sewage	of	Properties	hemical	4. (	<b>Fable</b>
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Type of sludge	Solid part	Summer	Fall	Winter
	рН	5.75	5.7	5.49
Primary sludge	COD	33150	41500	79200
	TKN	79.35	155	87.5
	SOUR	0.48	0.56	0.48
	Na	415.5	762	650
	Ca	1031	1102	723
	рН	5.6	5.9	5.96
Secondary sludge	COD	33150	35035	38600
	TKN	50.5	140	52.5
	SOUR	5.3	5.2	4.21
	Na	201	396	320
	Ca	520.5	556	402

Table 5. The Amount of Sodium, Calcium and Magnesium

	Unit	Summer	Autumn	Winter
Na <sup>+</sup>	Primary sludge (meq/L)	18.7	33.13	28.26
	Secondary sludge (meq/L)	8.74	17.21	13.91
C 21	Primary sludge (meq/L)	51.55	55.1	36.15
Ca	Secondary sludge (meq/L)	26.02	27.8	20.1
14.21	Primary (meq/L)	623.25	516.5	728.58
IVIg2+	Secondary sludge (meq/L)	529.58	756.25	223.16

indicator of the suitability of water for use in agricultural irrigation (19). In the study of the effect of sludge on agronomic soils, the final sodium absorption ratio of the soil solution was considered. Table 5 shows the amount of calcium, sodium, and magnesium in the sludge (mg/L).

The SAR in three seasons of summer, fall, and winter for primary sludge was 1.01, 1.96 and 1.44 mmol/L and for secondary sludge, it was 0.497, 0.869 and 1.26 mmol/L, respectively. The results show that the highest SAR calculated for the initial sludge was observed in the fall, and the lowest SAR for secondary sludge was reported in summer. The SAR reported for secondary sludge in the summer and fall was less than 1, but it reached more than 1 in winter. This is due to a 75% decrease in the magnesium content of this sludge in winter. However, this sodium absorption ratio is much lower than the standard for grouping of agricultural waters based on SAR. Therefore, the application of this sludge is allowed in agricultural soils considering the sodium adsorption ratio.

# 3.3. The Investigation of Biological Properties of Wastewater Sludge

The study of the biological properties of sludge is important in assessing the sludge reuse in agriculture as a fertilizer (20). In the present study, the mean fecal coliforms count for the primary sludge was  $19.83 \times 10^6$ and for the secondary sludge, it was  $186 \times 10^6$  (Figs. 1 and 2). The results show that the raw and secondary sludge produced in this wastewater treatment plant during the three seasons (summer, fall, and winter) was dramatically different from the Class A regulations and it was also significantly different from the Class B of the US EPA regulations. Hence, the disposal or reuse of this sludge is totally inconsistent with the principles of environmental protection and has a high level of health risk. In addition, the average probable number of parasite in 4 g of dry solids of raw and secondary sludge was 7.7 and 2.7, respectively. Only total and fecal coliforms counts are sufficient to determine the biological quality of the sludge and there is no need for other microbiological assays. The importance of parasite has led to testing total parasite for all samples (21). The highest number of parasite was found for Entamoeba coli (cyst) (Table 6). Additionally, the highest number of parasite was recorded in summer for raw sludge. In the studies of Mesdaghinia et al (22), Takdastan et al (23), and Rahmani et al (2), which focused on the biological characteristics of the sludge produced in the treatment plant, it was indicated that the sludge produced in studied treatment plants did not meet the standards and should be used with limitations. Naji Rad et al conducted a study to measure the populations of coliforms in the sludge samples of three different sewage sludge treatment plants including Shoush, Ekbatan and Shahrak-e-Gharb and to compare the results with the standards. They concluded that these sludge samples were categorized as class B and there would be limitations in their applications (1). Based on all studies conducted in this field, none of the sludge of wastewater treatment plants in the country meets Class A standards (40 CFR Part 503). This goes against the fact that the present proposal also confirms this result. Shokouhi et al (24) reported that the sludge obtained from the stabilization process was appropriate for use in agriculture and met biological index of class A standards (40 CFR Part 503). Autothermal thermophilic aerobic digestion (ATAD) is an aerobic method for digestion of the sludge of treated wastewater. The ATAD does not require any external power source and its performance in removing organic compounds from the sludge is acceptable. Moreover, the output product is in accordance with the Class A standards of EPA (24).

## 3.4. The Concentration of Heavy Metals in Sludge

Concerns about heavy metal pollution are due to three important properties of these materials, including non-biodegradability, toxicity to soil organisms, and persistence in soil (1). In 2007, Banaras Hindu University (India) published an article on the global application of sludge in agriculture, which emphasized the limited use of sludge due to the accumulation of metals in soil and vegetables, and suggested that plant growth is limited in sludge-amended soils (25). As shown in Table 7, the average concentrations of these elements in the samples studied did not exceed the EPA standards (16) and the highest concentrations of heavy metals belonged to aluminum and magnesium. Therefore, the use of this sludge, in terms of these elements, is permissible for all sludge applications. The EPA standard for lead concentration is 300 mg/kg, and its mean concentration was 103 mg/ kg for both types of sludge studied in the present study.

Table 6. Number of parasite in the Sludge of Wastewater Treatment Plant in Hamadan in Different Seasons

Democión ferre	11.5	Summer		The first week of fall		The second week of fall	
Parasite type	Unit	Primary	Secondary	Primary	Secondary	Primary	Secondary
Entamoeba coli (cyst)	N/10 cc	21	15	10	4	10	8
Giardia SP (cyst)	N/10cc	5	0	2	0	5	0
Oxyuridae SP (ova)	N/10 cc	1	0	0	0	0	0
Nematoda SP (larva)	N/10cc	15	0	8	3	14	0
Nematoda SP (ova)	N/10 cc	6	3	0	0	0	3
Blastocystis hominis (cyst)	N/10 cc	10	5	10	5	10	4

	Primary, Summer	Secondary, Summer	Primary, Fall	Secondary, Fall	Primary, Winter	Secondary, Winter	Average
Al	16418	26896	22154	15340	18609	28956	21396
Ba	193	206	178	161	156	198	182
Ca	29987	45856	38156	28871	39874	51425	39028
Cr	31	44	36	28	39	46	37
Cu	155	147	120	131	140	105	133
Fe	17790	25823	22167	16096	8508	26717	19516
Mg	7479	7111	6198	9075	8743	2678	6880
Pb	60	80	102	48	81	49	70
Sr	954	1336	1135	815	967	1329	2179
Zn	574	615	520	557	858	534	610
Cd	5	5	5	4	4	4	4.5

Table 7. Average Concentrations of Heavy Metals in Sludge in Different Seasons (mg/kg)

The standard value for cadmium concentration is 39 mg/ kg, and it was observed to be 4.5 mg/kg. The standards for nickel and chromium concentrations are 420 and 1200 mg/kg, respectively, and its value for chromium was observed to be 37.33 mg/kg. Aluminum, iron, and manganese contents are not specified in the standards of sludge for its use in agriculture or disposal. However, due to the high concentrations of these elements in comparison with other metals and their related problems, consideration should be given to eliminating the high concentrations of these toxic heavy metals from the environment and the waters or their use for various purposes. Houdaji et al (26) conducted a study on the cress, lettuce, and spinach cultivated in soils fertilized by sewage sludge and reported that the amount of leads in leaves of cress significantly increased compared to control plants. The results of a study conducted by Ghamari and Danesh (12) indicated that the application of sewage sludge to the soil has led to a significant increase in the concentration of lead and cadmium in the plant. Rahmani et al (2) concluded that all heavy metal elements studied in the sludge were in acceptable level, except for arsenic, and the use of sludge is permissible. Wong et al (27) conducted a study on Chinese cabbage cultivated in the soils modified with sludge showed very low amounts of heavy metals (except for Arsenic), which were less than the acceptable standards. Naji Rad et al (1) concluded that

350 300 250 200 150 100 50 0 Summer Autumn Winter



Fig. 1. Average Fecal Coliform Count in Different Seasons.

due to the presence of the high amounts of heavy metals, especially zinc and copper, none of the sludge samples have the quality to be applied to agricultural land without restriction.

## 4. Conclusion

In this study, some physical, biological and chemical properties of sludge produced in Hamadan wastewater treatment plant were investigated. The results showed that if this sludge is used for agricultural purposes and surface disposal, more measures should be taken to stabilize and improve its quality. The results of this study showed that the VS/TS ratio for raw sludge was 0.8, 0.55, and 0.55 and for secondary sludge, it was determined to be 0.65, 0.28 and 0.32 in autumn, winter and summer, respectively, which indicated that neither type of sludge was stabilized in the treatment plant. Moreover, the investigation of pH values (mean pH of 5.64 for primary sludge and 5.82 for secondary sludge) showed that neither type of sludge was sufficiently stabilized in this treatment plant. The mean concentration measured for COD, TKN, SOUR for primary sludge was reported to be 51283, 107, 0.50, while it was determined to be 35595, 81, and 4.90 mg/L for secondary sludge, respectively, which illustrate that neither type of sludge was stabilized. Based on the results, the values obtained for Na, Ca and SAR were 952 mg/L, 609 mg/L and 1.5 for primary sludge and



Fig. 2. Average Total Coliform Count of Sludge in Different Seasons.

were 306 mg/L, 493 mg/L and 0.87 for secondary sludge, respectively. Since these values did not exceed the standard levels, the application of this sludge is permissible. On the other hand, the results of biological characteristics of primary and secondary sludge of Hamadan wastewater treatment plant determined by the average fecal coliforms count and the mean most probable number of parasitic eggs in each sludge revealed that the MPN for primary and secondary sludge was determined to be  $19.83 \times 10^6$ and  $186 \times 10^6$ , respectively and the average number of parasite eggs in raw and secondary sludge was 7.05 and 2.7 in 4 g of dry solids, respectively. Furthermore, the highest number of parasitic eggs belonged to Entamoeba *histolytica* (cyst), indicating that these characteristics were significantly different from Class A regulations and were also significantly different from Class B regulations (US EPA). The use and reuse of studied sludge are inconsistent with the principles of environmental protection and has many health hazards. Therefore, it is recommended that appropriate sludge stabilization system should be provided to improve the quality of both primary and secondary sludge produced. The evaluation of the results of this study showed that the highest concentration of heavy metals in the sludge belonged to aluminum (21396 mg/kg) and the lowest concentration was observed for cadmium (4.5 mg/ kg). According to the results obtained from investigation of the heavy metals, the average concentration of these elements in primary and secondary sludge produced in this treatment plant did not exceed EPA standards, therefore, the application of this type of sludge for all purposes discussed is permissible in terms of these elements.

#### **Conflict of Interest Disclosures**

The authors declare that there is no conflict of interests.

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