

## Use of Ionizing Radiation Technology for Treating Municipal Wastewater

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**Abstract:** In big cities, the cost of treating wastewater is increasing with more stringent environmental requirements. Ionizing radiation technology for treating municipal wastewater may be an alternative to reduce treatment costs. In this paper, laboratory tests were carried out using different doses of radiation to treat wastewater samples collected from the AL-Rustamia wastewater treatment plant in Baghdad city. According to the results, irradiation by gamma radiation with a dose ranging from 100 to 500 krad was efficient in reducing some of the physical contaminants. The organic contaminants were degraded and reduced to about 12% of their original concentrations. Generally, irradiation technology could effectively modify the characteristics of the wastewater to such levels that are compatible with Iraqi disposal standards. The results of the study also showed that, an experimental pilot plant study is required to optimize the cost of wastewater treatment through the use of this technology.

**Keywords:** Ionizing radiation, wastewater treatment

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### Introduction

The need for new technology in treating municipal wastewater is continuously developing. The newest technology should have advantages in minimizing the required cost for wastewater treatment.

In big cities, the sewerage works are costly compared to other municipalities' services. Then the cost should be minimized for sewerage works to mobilize this environmental service by municipalities. The construction of small efficient wastewater treatment plants for individual sections of the city will help in reducing the cost of sewerage works.

Technically, ionizing radiation is an advanced form of energy, available for societal use, but feared by many people as being an environmentally dangerous and health hazard. Radiation energy treatment for a clean water supply and wastewater treatment is currently under study in many countries. Greater application of this technology in the treatment of water, wastewater and sludge may help not only for the development of technical processes, but also on greater acceptance of the technology as engineering needs [1].

The typical costs for the treatment of wastewater by radiation are found to be favorably comparable with the other advanced wastewater treatment systems [2].

The objectives of this study are: To evaluate the applicability of ionizing radiation technology for the removal of biochemical oxygen demand (BOD) and fat, oil, and grease (FOG) from municipal wastewater; to assess the physical and biochemical characteristics of municipal wastewater before and after the irradiation; and to determine the optimum doses for irradiation of wastewater.

### Radiation Treatment

Radiation treatment may be defined as the application of ionizing radiation energy to produce a useful change in a material, such as disinfection. The amount of radiation energy absorbed in a material depends on both the chemical and physical state of the material and on the type and energy distribution of the radiation. The respective radiation dose units for biological comparison units are the rem and the sievert (sv) which indicate the physical dose unit multiplied by a radiation quality factor (QF).

The application of radiation energy to water and wastewater treatment needs to achieve a sufficient dose absorbed uniformly at a given flow rate and an economic yield. The involved factors include the type of radiation, its energy distribution, and the penetrability into the water stream, the geometric configuration of the radiation-water

interaction volume and the thickness of water normal to the radiation flux [1, 3].

There are many types of radiation such as gamma, beta, alpha, X-rays and UV. However, the type of radiation mostly used in the treatment of wastewater is gamma radiation. So in this study, gamma radiation was used in wastewater treatment. Gamma ray, emitted during the decay of radioactive atoms, which is electromagnetic radiation, has relatively high penetrating power. The intensity of a gamma ray source determines the exposure time for a given dosage. Some factors such as the source-to-water geometry and the presence of solids in the water affect the bulk density and irradiator design.

An important characteristic of gamma rays from either  $\text{Co}^{60}$  or  $\text{Cs}^{137}$  is that it is highly penetrating. For water, the half value and tenth value layer for  $\text{Co}^{60}$  gamma ray are 27 and 61 cm respectively. For  $\text{Cs}^{137}$  the half value and tenth value gamma ray are 24 and 58 cm, respectively. Therefore, it is clear that with an appropriate mixing to provide homogenous exposure, targets with thickness of 50-60 cm can be readily treated.

#### Biological Phase Treatment

Although the idea of using ionizing radiation for treatment of sewage waste was conceived almost five decades ago, it is only in recent years that there have been several reports on the potential role of radiation in the treatment of sewage and other wastewater. The rationale for utilizing radiation in treatment of sewage waste rests on number of documented facts [1]:

1. Radiation destroys microbial life and this property is considered particularly beneficial, since it will result in the inactivation of pathogenic micro flora;
2. Radiation is capable of altering the structure of organic molecules, thereby leading to a decrease in Biochemical Oxygen Demand ( $\text{BOD}_5$ ) and Chemical Oxygen Demand (COD).
3. Radiation can bring out physiochemical alteration in suspended solid, leading to the formation of more compact sludge and higher capacity for settling.

It is generally agreed that radiation is an exceptionally effective disinfection method. The sequence of events during radiation exposure includes three periods; latent period; demonstrable effects period, and recovery Period.

#### Responses of Microorganisms to Irradiation Used in Wastewater Treatment

The responses of microorganisms and parasites to a given dose can be altered in different ways. This is possible because the response depends on:

- physical factors (temperature and type of radiation);
- chemical factors (sensitizing and protecting agents);
- © Biological or physiological factors (growth phase and amount of DNA).

The effect of radiation on microorganisms and parasites can be modified not only by agents present during irradiation, but also by biochemical processes

occurring over a much longer period of time. Proper pretreatment of wastewater prior to irradiation offers an advantage in maximizing the lethal effect [3]

The treatment of wastewater by irradiation can be optimized to enhance the synergistic inactivation of microorganisms and parasites. The main factors, other than the genotype of the microorganisms that influence, radio sensitivity are oxygen, chemicals present in wastewater and temperature.

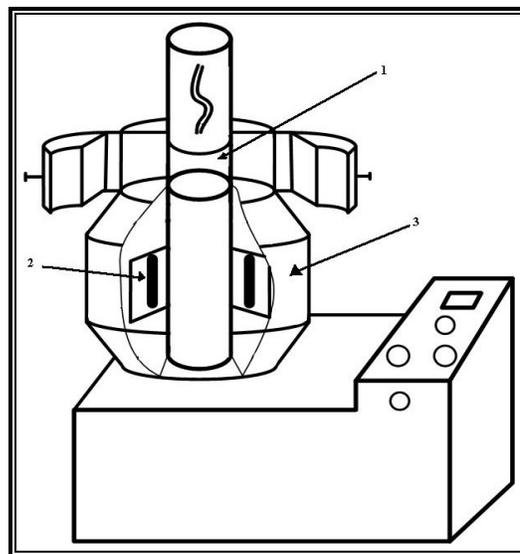
## Materials and Methods

### Sample Collection

Samples of wastewater were collected from AL-Rustamia sewage treatment plant for three months: January, February and March (2002) where  $\text{BOD}_5$  were estimated as 243mg/L, 309 mg/L and 321mg/L respectively. The samples were collected by using different types of clean containers. Samples were taken from the effluent of the primary settling tank.

### Irradiation

The facility used in irradiation is Gamma-cell 220 (Canadian made) supplied with  $\text{Co}^{60}$  which has a calculated dose rate of two Mrad/hr and radioactivity of 50 kCi in January 1985. Samples were arranged in the cylindrical irradiation chamber (16 cm in diameter and 20 cm in height). This room moved vertically down to the radiation sources of  $\text{Co}^{60}$  as rods rotate around the room to supply a homogenous dose for all samples (Fig 1) [4].



1: Irradiation room. 2: Rod of  $^{60}\text{Co}$ .  
3: Surrounding shields of lead.

**Figure 1:** Gamma cell irradiation facility type 220 Canadian made [4].

### Absorbing Dose

All tests in this study were taken as an average of three samples subjected to the same absorbing dose. The doses used were 0, 10, 25, 50, 100, 200, 300, 400, and 500 krad.

The dose rate in recent time (i.e. 2002) was approximately 0.2 Mrad/hr (Calculated on the basis that the half-life of  $\text{Co}^{60}$  is 5.26 years). The irradiation times for these dosages were 0, 3, 7.5, 15, 30, 60, 90, 120, and 150 min. respectively where:

$$D = DR \times T$$

D = Absorbing dose, (rad): DR = Dose Rate, (rad/min.): and T = Irradiation time, (min).

### Physical and Chemical Measurements

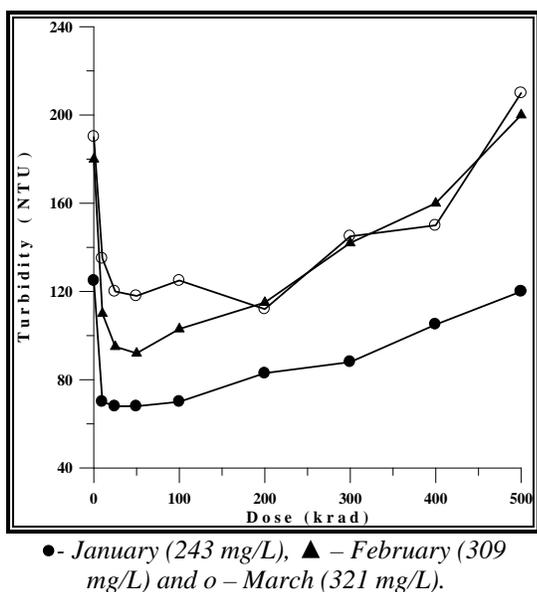
The physical and chemical measurement parameters taken for this study were: turbidity, electrical conductivity (EC) and hardness, pH, dissolved oxygen (DO), biochemical oxygen demand ( $\text{BOD}_5$ ) and chemical oxygen demand (COD), total suspended solid (TSS), total dissolved solids (TDS), fat, oil and grease (FOG), and phenolic compounds.

### Results and Discussion

Ionizing radiation has a drastic effect on the organic materials in the wastewater, because of the strong activity of gamma radiation energy that changes the characteristics of pollutants in the wastewater [3]. The results were found in the undetectable levels for most of the tests, but there were some tests which did not respond to the radiation energy because of inorganic compounds. The result of this research consisted of 320 tests. All tests involved 1008 irradiation processes.

#### Turbidity

Figure 2 and Table 1 indicated a decrease in turbidity as the radiation dose increased to a certain absorbing dose, thereafter, turbidity started to increase with the increase of radiation doses.

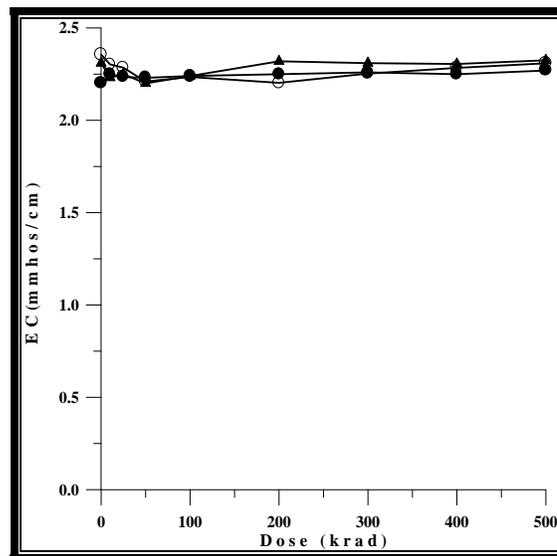


**Figure 2:** Turbidity values of irradiated sample of wastewater for  $\text{BOD}_5$

Turbidity decreased to 68 NTU at 25 krad for January and 92 NTU at 50 krad for February and 118 NTU at 50 krad for March, respectively. Then the values increased to 120, 200 and 210 NTU for January, February and March, respectively at an irradiation dose of 500 krad. Sometimes turbidity surpassed the starting value. This increase in turbidity was attributed to the dissolved organic materials in the sludge [3].

#### Electrical Conductivity (EC) and Hardness:

Results revealed that EC values and hardness remained stable with the increase in radiation dose (Fig. 3). This is because EC and hardness are functions of dissolved salts in water. It is known that the organic salts are not conductive materials, but the inorganic salts are very conductive materials when dissolved in water since they are electrolytes. Moreover, the radiation doses do not degrade the inorganic materials consequently the salinity value does not change with the increase in radiation dose. Therefore, EC (2.25mmhos/cm) and hardness (800mg/L as  $\text{CaCO}_3$ ) values remained stable (Table 1) as previously shown in an earlier I.P.S. work, [7].



● - January (243 mg/L), ▲ - February (309 mg/L) and ○ - March (321 mg/L).

**Figure 3:** EC values of irradiated sample of wastewater for  $\text{BOD}_5$

#### pH

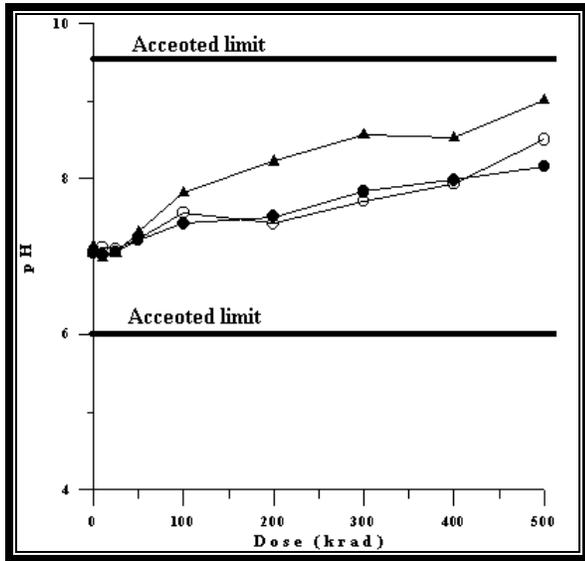
pH increased as the radiation dose increased. Figure (4) shows that pH values tended to increase as a result of the radiolysis of water. These values reached 8.1, 9.0 and 8.5 for January, February and March, respectively. This pH increase resulted from the increase of hydroxyl radicals at very large quantities due to the hydrolysis of water. The G-value of OH (G-OH) is high when compared with the other radicals, which caused a large concentration of free hydroxyl radicals in water consequently increasing the pH value. All pH measurements remained within accepted values, as shown in (Tables 1 and 2).

**Table 1:** Summary for the conventional result of experimental work

<i>Parameter</i>	<i>Month</i>	<i>Initial value ( mg/l)<sup>a</sup></i>	<i>Affect dose (krad)</i>	<i>Final Value ( mg/l)<sup>a</sup></i>	<i>Percentage to initial value</i>
Turbidity	January	125 (NTU)	25	68 (NTU)	54.4
	February	180 (NTU)	50	92 (NTU)	51.11
	March	190 (NTU)	50	118 (NTU)	62.1
EC	January	2.2 (mmhos/cm)	Does not affect	--	--
	February	2.31 (mmhos/cm)	Does not affect	--	--
	March	2.356 (mmhos/cm)	Does not affect	--	--
PH	January		No more 500	8.15 * (unit less)	--
	February	7.13 (unit less)	No more 500	9.01 * (unit less)	--
	March	7.06 (unit less)	No more 500	8.5 * (unit less)	--
DO	January	9.5	50	4.5	--
	February	9.3	50	4.4	--
	March	10	50	4.7	--
BOD <sub>5</sub>	January	243	500	30 *	12.3
	February	309	500	44	14.23
	March	321	500	59	18.38
COD	January	1400	500	75 *	5.36
	February	1705	500	112	6.56
	March	1809	500	130	7.18
FOG	January	1750	500 and more	409	23.4
	February	1725	500 and more	482	27
	March	2017	500 and more	29	1.4
TSS	January	308	50	56 *	18.2
	February	422	100	50 *	11.85
	March	450	200	51 *	11.33
TDS	January	10580	10	1360	11.4
	February	12435	25	1100	8.85
	March	12210	25	1121	9.18
Hardness	January	800 (mg/l as CaCO <sub>3</sub> )	Does not affect	--	--
	February	820 (mg/l as CaCO <sub>3</sub> )	Does not affect	--	--
	March	850 (mg/l as CaCO <sub>3</sub> )	Does not affect	--	--
Phenol	January	0.1449	500	0.0121 *	8.4
	February	0.3115	500	0.0192 *	6.2
	March	0.3091	500	0.041 *	13.16

<sup>a</sup> All unit in mg/l except those shown in brackets.

\* Accepted values within the Iraqi limits.



● - January (243 mg/L), ▲ - February (309 mg/L) and ○ - March (321 mg/L).

**Figure 4:** pH values of irradiated sample of wastewater for BOD<sub>5</sub>

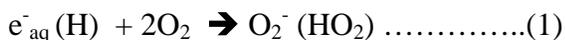
**Table 2:** Iraqi allowable values of conventional concentrations that can be disposed to the surface water [19].

Parameter	Limit
BOD <sub>5</sub>	40 mg/l
COD	100 mg/l
TSS	60 mg/l
Phenol	0.01 - 0.05 mg/l
pH	6 - 9.5

*Dissolved Oxygen (DO)*

The reduction in DO concentration values with increasing radiation doses are shown in Fig. 5 and Table 1. The AL-Rustamia treatment plant used a primary aeration tank, hence the value of DO increased in the raw samples. On the other hand, the irradiation process decreased the values of DO. The decrease in DO concentration resulted from the following mechanism:

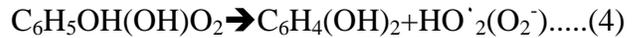
The irradiation in aqueous solutions forms solvated electrons. It may react by neutralization reaction or may add to molecules. These electrons react with oxygen in the presence of hydrogen, as in the following reaction (1) [5]:



Pulses radiolysis data confirm the assumption that C<sub>6</sub>H<sub>5</sub>OH(OH) radicals formed in reaction (2), and reacted with dissolved oxygen, [6].



The optical absorption shows the first order decay dependent on oxygen with a spectrum of indicated long-live O<sub>2</sub><sup>-</sup> identification. These observations could best be the sequence of reactions such as reactions (3), (4) and (5), [6]:



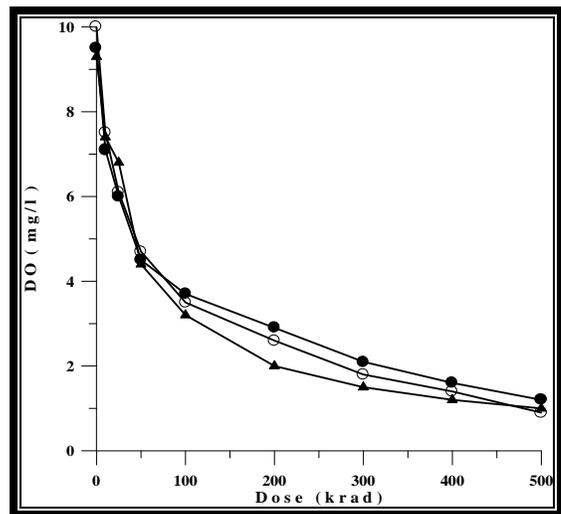
Also, the energy of radiation applied to hydrogen radicals in water leads to the reaction indicated in (6), that will react with DO to form HO<sub>2</sub>, as in the following reaction, [3]:



In addition, the free electrons react with oxygen to form O<sub>2</sub><sup>-</sup>, as in the following reaction: -



It is well known that HO<sub>2</sub> and O<sub>2</sub><sup>-</sup> are the most reactive species, and they react with wastewater pollutants, although they are less powerful reactants compared with H<sup>·</sup> and e<sub>aq</sub><sup>-</sup> [3]. For all the above reactions and others, it is clear that a large number of receivers for oxygen cause a decrease in dissolved oxygen concentration.

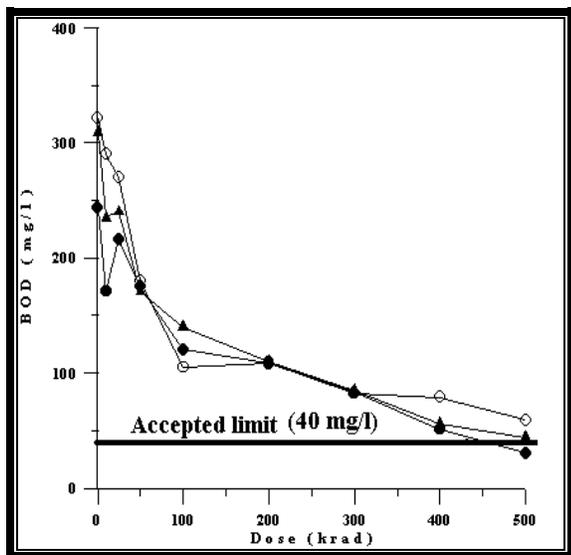


● - January (243 mg/L), ▲ - February (309 mg/L) and ○ - March (321 mg/L).

**Figure 5:** DO values of irradiated samples of wastewater for BOD<sub>5</sub>

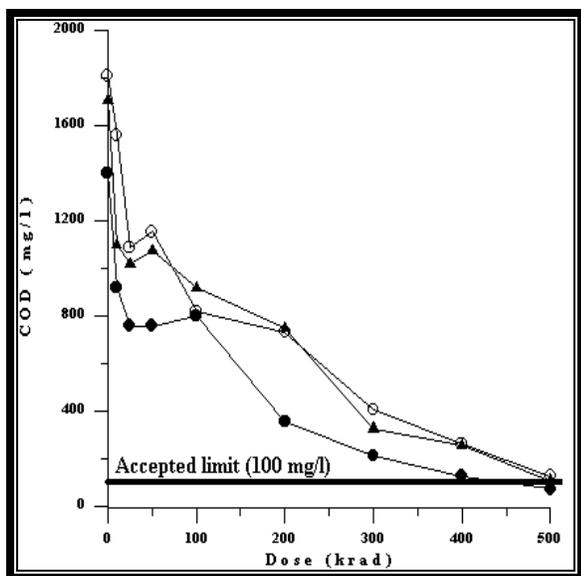
Biochemical Oxygen Demand (BOD<sub>5</sub>) and Chemical Oxygen Demand (COD)

(BOD<sub>5</sub>): - Figures 6 and 7 show the decrease in BOD<sub>5</sub> and COD concentration values respectively with respect to the increase of radiation dose. This decrease resulted from the destruction of microorganisms responsible for oxygen consumption. It is known that microorganisms are very sensitive to radiation pulses. In addition, the radiation energy can degrade organic compounds found in biological systems.



● - January (243 mg/L), ▲ - February (309 mg/L) and ○ - March (321 mg/L).

Figure 6: BOD<sub>5</sub> concentration of irradiated sample of wastewater for BOD<sub>5</sub>.



● - January (243 mg/L), ▲ - February (309 mg/L) and ○ - March (321 mg/L).

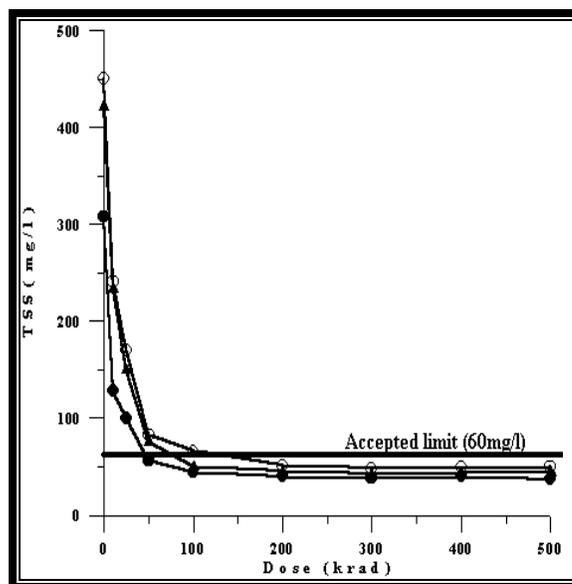
Figure 7: COD concentration of irradiated sample of wastewater for BOD<sub>5</sub>.

The BOD<sub>5</sub> values reached 12.3% of their initial value through the use of 500 krad absorbing dose, while the accepted value of BOD<sub>5</sub> is 40 mg/l, (Tables and 2). However, in March, the concentration of BOD<sub>5</sub> at the same radiation dose remained high, but they were within the accepted value. This observation can be explained by the fact that sometimes samples contain surfactant pathogens such as Bacillus spores and Micrococcus radiodurant and mineral organics, which are not degraded by radiation. To reduce these types of pathogens, the combined processes must be used (i.e., radiation with either heat or ozone) [8].

(COD) values show the attainment of the acceptable value for January but were higher than the accepted value in February and March (see Tables 1 and 2). To achieve the accepted values in February and March, higher radiation doses were needed. This decrease in BOD<sub>5</sub> and COD matched previous research reports from I.P.S. [7], Compton et. al [9]; Etzel & Condern [10], Mytelka [11].

Total Suspended Solid (TSS):

Figure 8 shows the decrease in total suspended solids (TSS) as the absorbed radiation dose increased. For the three months, January, February and March, the removal percentage reached 18.2 %, 11.85% and 11.23% of the initial values, respectively. All these values were within the accepted range (see Tables 1 and 2). The total suspended solids (TSS) were decreased, as they were converted to precipitates resulting from the degradation of organic substances and suspended matter in wastewater. These results are in agreement with previous work of I.P.S. [7], Compton [12], and Undynamic Corporation (U.C.) [13].

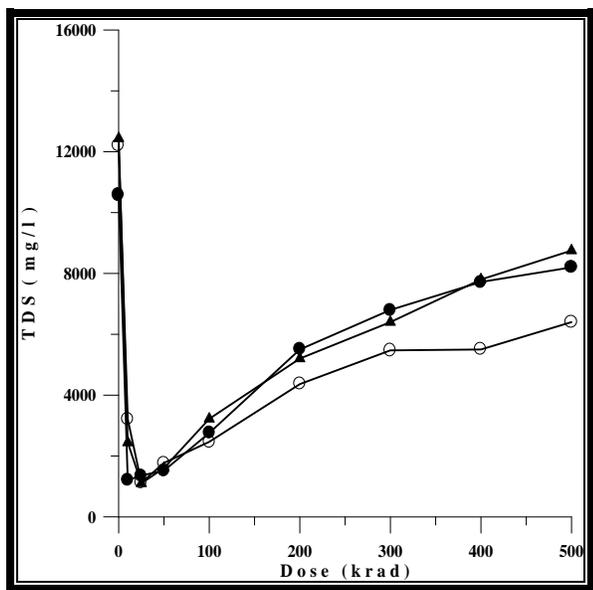


● - January (243 mg/L), ▲ - February (309 mg/L) and ○ - March (321 mg/L).

Figure 8: TSS concentration values of irradiated sample of wastewater for BOD<sub>5</sub>.

Total Dissolved Solid (TDS)

Total Dissolved Solids (TDS) concentrations showed different patterns of interaction with radiation doses. The TDS concentration decreased with respect to the increase in the absorbed radiation doses to certain values, after which TDS concentration increased with increases in absorbed radiation dose. Based from Fig 9 and Table 1, the TDS concentration decreased to 11.4% of its initial value at 10 krad absorbing dose then it increased to 77.5% at 500 krad for January and February. The concentration decreased to 8.85% of its the initial value at 25 krad then it increased to 70.4%. For March, it decreased to 9.2% at 25 krad then it increased to 52.4% at 500 krad. This might be due to the conversion of the dissolved organic substances to simple molecular compounds or formation of dimmers or trimmers. Also, the high doses may have dissolved the suspended solids and sludge that were at the bottom of the container, thus increasing the concentration of TDS. It was noted however that the TDS values never reached the starting values [1]. Since the inorganic materials were not altered by radiation, our results are similar to an earlier I.P.S. finding [7].



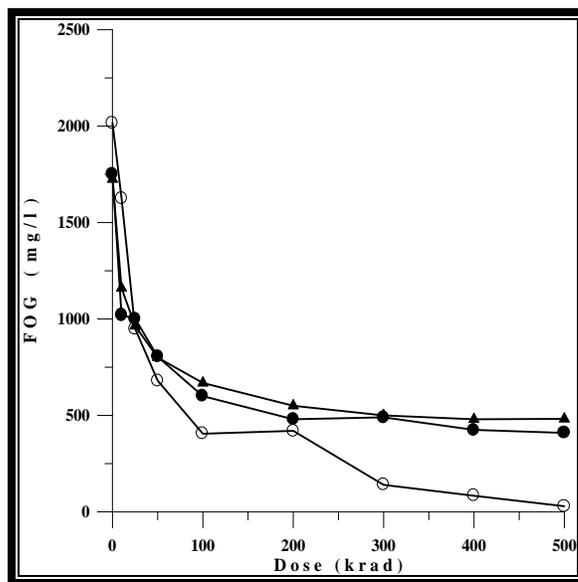
●- January (243 mg/L), ▲ – February (309 mg/L) and o – March (321 mg/L).

Figure 9: TDS values of irradiated sample of wastewater for BOD<sub>5</sub>.

Fat, Oil and Grease (FOG)

Figure 10 and Table 1 show the decrease of FOG concentrations as the absorbed radiation dose increased. Fat, oil and grease values were reduced by radiation to 23.4%, 27% and 1.4% of their initial values for January, February and March, respectively. FOG is composed of long chains of hydrocarbons. The radiation energy lysed the bonds of these hydrocarbon chains converting the FOG to shorter hydrocarbon fragments. These findings agree with previous results [7, 14]. The degradation of FOG depends on the

presence of mineral FOG. It is known that radiation does not degrade the mineral FOG [3]. These new compounds have certain characteristics and composition that differ from FOG, and have fewer impacts as wastewater hazards.

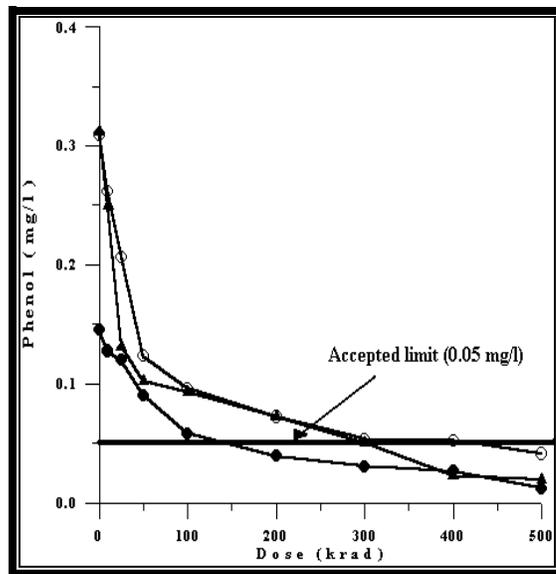


●- January (243 mg/L), ▲ – February (309 mg/L) and o – March (321 mg/L).

Figure 10: FOG concentration of irradiated sample of wastewater for BOD<sub>5</sub>.

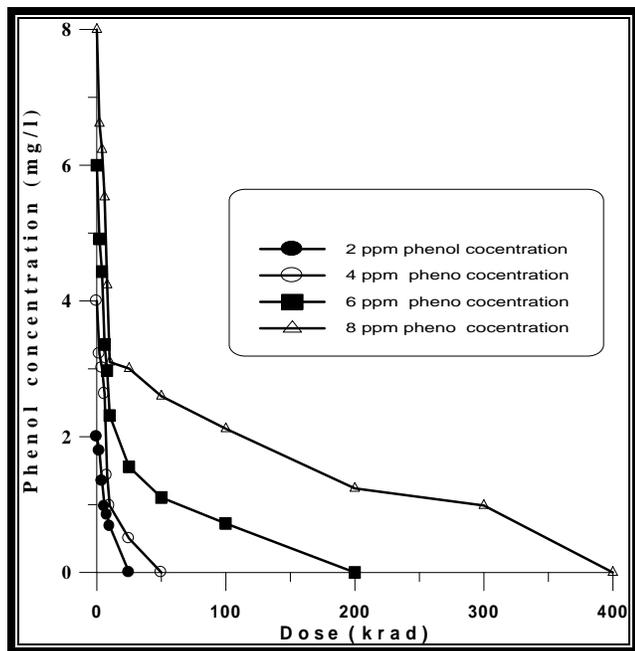
Phenolic Compounds and Standards of Phenols

Concentrations of irradiated phenolic compounds and standard phenol solutions decreased as the radiation dose increased (Figs. 11 and 12).



●- January (243 mg/L), ▲ – February (309 mg/L) and o – March (321 mg/L).

Figure 11: Phenol concentration of irradiated sample of wastewater for BOD<sub>5</sub>.



**Figure 12:** Phenol concentration values of standard solution of phenol with respect to radiation dose.

All these values at final absorbing dose of radiation are within the in accepted range (see Tables 1 and 2). They varied from 8.4% of their initial value in January, 6.2% for February and 13.16% for March, respectively. The decrease in the concentration of phenol agreed very well with the results of Takehtsa & Sakumoto [15] and Machi [16]. The degradation of phenol samples never reached zero concentration value, while standard solutions of phenol reached zero concentration. This is because the standard solutions of phenol are pure, while the samples of wastewater contain different types of phenols such as chlorophenols and nitrophenols that contain surfactants.

**Conclusions**

Irradiation by gamma radiation is an efficient physical way to destroy organic compounds, as long as the radiation parameters delivered are correctly suited to the application. Under the experimental conditions imposed in the present study, the optimal doses are mentioned in Table 3. Even at these values, the radiation efficiency for most parameters is, surprisingly, not completely satisfactory. The applicability of a laboratory experiment like this should be further tested in the field using, a pilot plant. The established pilot plant must be supplied by wastewater from the effluent of the primary sedimentation tank of the Baghdad wastewater treatment plant. All the laboratory-tested parameters, including sludge management should be considered in order to evaluate the multiple benefits of ionizing radiation technologically for treating municipal wastewater in Baghdad city.

**Table 3:** Suitable Radiation dose to minimize each parameter to accepted value

Parameter	Initial value <sup>a</sup> (mg/l)	Suitable dose (krad)	Final value <sup>a</sup> (mg/l)
BOD <sub>5</sub>	210	500	44.33
COD	1638	500	105.67
TSS	393.33	100-150	44
TDS	11741.67	25	972
Phenol	0.2551	500	0.0241
FOG	1830.67	500	306.67
Turbidity	165 (NTU)	100-200	103.33 (NTU)
EC	2.288 (mmhos/cm)	Does not response	2.30 (mmhos/cm)
Hardness	823.33 (mg/l as CaCO <sub>3</sub> )	Does not response	800 (mg/l as CaCO <sub>3</sub> )
DO	9.6	Decreased	1.03
pH	7.08 (unitless)	Increased	8.553 (unitless)

<sup>a</sup> All unit in mg/l except those shown in brackets. All values are an average of three months January, February, and March.

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