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Radioactivity in Municipal Wastewater and Its Behavior in Biological Treatment

U. İpek,[1] E. I. Arslan,[1] S. Aslan,[1] M. Doğru,[2] O. Baykara[2]

[1] University of Fırat, Faculty of Engineering, Department of Environmental Engineering, 23119, Elazığ, Turkey
[2] University of Fırat, Faculty of Arts and Science, Department of Physics, 23119, Elazığ, Turkey

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Since living organisms, especially those of human beings, need water to keep alive, determination and control of the radioactivity as well as the other pollutants in water are very important so as to ensure good health. The radioactivity in water generally arises from the soil composition and the additive materials that contain various radioactive minerals. The radioactivity in soil mainly rises by increasing of the amount of natural radionuclides such as ^{238}U, ^{232}Th and ^{40}K. The other way to increase the radioactivity in soil is to add artificial inseminate and manure to increase the harvest (İpek et al. 2002). The additive materials contain radioactivity from alpha and beta radioactive radionuclides thus increasing the radioactivity concentration in the water content. Drinking or tap water concentrated with high radioactive mineral may cause the different types of cancer in the human body. In view of this, many organisations over the world have put guidelines for gross alpha and gross beta radioactivity level, and also for individual radionuclides in drinking water (Dogru and Canbazoglu 2002). For practical purposes, the recommended guideline activity concentrations are 0.1 Bq/L for gross-α and 1 Bq/L for gross-β activity for drinking water (WHO 1993). In Turkey, the guidelines for similar situations were also established by the Institute of Turkish Standards (Karahan et al. 2000). According to these guidelines, the recommended gross alpha and gross beta radioactivity concentrations in drinking water are 0.037 Bq/L and 0.37 Bq/L, respectively (ITS 1997). Regrettably, these guidelines for the radioactivity concentration in municipal wastewater have not yet been met.

Municipal wastewater includes domestic water, vegetable and fruit washing water, hospital effluent, other varieties of wastewater and rainwater. Radioactivity in wastewater can arise from the natural constitution of the consumed water and the addition of various other matters to the wastewater. The wastewater from Elazığ city has been treated with the activated sludge process and the effluents with 33643, 31063, 31789 and 31594 m^3/day in April, May, June and July were discharged into Keban Dam Lake. Keban Dam Lake water is presently used for agricultural and other purposes like fishing and recreation and will probably be utilized as drinking water in near future.

Activated sludge processes are widely used for the biological treatment of sewage and are considered the appropriate means for treating radioactive organic liquid

Correspondence to: U. İpek

waste (Koyama and Nishimaki 1997). The behaviour of radioiodine and other radioactive materials released into municipal sewerage systems, such as those from large medical facilities, is not yet well understood (Martin and Fenner 1997). Most of the wastewater is re-used after it is treated by different treatment processes. Therefore, because of the above-mentioned effects of the radioactivity in water, the determination and control of the gross radioactivity containing short-long lived alpha and beta active radioisotopes in wastewater, before and after treatment, is quite important because of its effect on the receiving environment. In this study, the concentration of the gross α, gross β radioactivity and the rates of some active radioisotopes in municipal wastewater were determined, and behaviour in the conventional activated sludge treatment facility was also investigated.

MATERIALS AND METHODS

The wastewater of Elazığ city is collected and then treated in the biological treatment plant. The biological treatment plant includes the units of conventional activated sludge. The effluent from the city treatment facility is discharged into Keban Dam Lake. Sewage sludge is used as fertilizer and/or ultimate disposal on the land. The influents of the biological treatment plant and effluents of the primary and final settling tank were sampled as composites with 2 hours at the different seasons. The samples were collected in 500 ml sterilised glass bottles and the residues from each sample were prepared in accordance with the standard evaporation method. Three 50 ml aliquots of each water sample were poured into 250 ml beakers. 0.5 ml 3N nitric acid was added to prevent the precipitation and absorption of the samples by the container walls and they were evaporated slowly at 60 ^{o}C to near dryness (approximately 2-3 ml). Then, each was transferred quantitatively to a 1.9 cm diameter stainless steel counting planchette and exposed by ultraviolet rays to dryness. The residues obtained were counted for gross-α, gross-β, ^{137}Cs, ^{90}Sr and ^{129}I radioactivity in low background alpha and beta counters. Measurements of the radioactivity level of all samples were performed in accordance with the Krieger method (Krieger 1975) using the gross-α and gross-β counting system. The beta radioactivity concentration in the prepared samples was determined by using a β-radiation-sensitive plastic scintillator (2059 plastic scintillator from NE Tech. Inc.) supported by a suitable photo multiplier tube and SR-8 Low Level Radiation Counter from the same company. β-radiation sensitive plastic scintillators are preferable for use for this purpose than the other solid-state detectors (Dogru 1997). Window counting based on their individual energies confirmed the individual radioisotopes activity. The instrumentation used to count global-α was a ZnS(Ag) alpha scintillator was supported by a photomultiplier tube from NE Technology. The counting time was 300 min for gross-α and gross-β for each counting period. Six counting periods were selected to determine total counts for the radioactivity of the gross-α, gross-β and other radioisotopes. The background was subtracted from the gross count to obtain the net counts used for calculation. The accuracy of the system used was about 5% of the standard deviation for both alpha and beta radioactivity determinations. The

gross-α and gross-β activities were calculated using the subtracted (net) counts in the following equations (Canbazoglu et al. 2000; Dogru et al. 2002).

$$A_\alpha = (N \times ECF) / 2.22 \qquad A_\beta = (0.391 \times R \times N_m) / N_0 \qquad (1)$$

Where, A_α and A_β are the activities of alpha and beta in pCi, respectively (Karahan 1997), N is the sample net count per minute for alphas. ECF is the efficiency correction factor (Knoll 1979), R is the sample net count for betas per minute, N_m is the specific mass of the sample in mg/cm^2, N_0 is the count corresponding to the specific activity which is determined from the standard calibration curve obtained by using KCl source by (Alkan 1989; Karahan 1997; Canbazoglu 1998). The correlation co-efficient (r) of the curve was calculated to be 0.998. The calculation of the efficiency correction factor (ECF) for the determination of alpha activity of the residue in the aluminium planchette is given in equation 2.

$$ECF = 1 / (E \times T) \qquad (2)$$

Where, the quantity of T is determined from the U_3O_8 self-absorption lines, that obtained by (Karahan 1997) in mg/cm^2 and E is the absolute efficiency.

RESULTS AND DISCUSSION

The radioactivity concentrations in the influent and effluent of the treatment plant are given in Table 1 as gross alpha and gross beta radioactivity. As shown in Table 1, gross α and gross β radioactivity of the raw wastewater ranges from 0.0772 to 0.2221 Bq/L, and from 0.1041 to 0.3161 Bq/L, respectively.

Table 1. The radioactivity levels of the wastewater and treatment effluents.

Time	Parameter	Influent	The Effluent of the Primary Settling Tank	The Final Effluent
April	α, Bq/L	0.2221 ± 0.0311	0.1573 ± 0.0303	0.0568 ± 0.0254
May	α, Bq/L	0.1605 ± 0.0341	0.0624 ± 0.0279	0.0487 ± 0.0231
June	α, Bq/L	0.0772 ± 0.0269	0.0447 ± 0.0224	0.0374 ± 0.0197
July	α, Bq/L	0.0991 ± 0.0134	0.0547 ± 0.0187	0.0428 ± 0.0147
December	α, Bq/L	0.1301 ± 0.0184	0.0746 ± 0.0329	0.0220 ± 0.0102
January	α, Bq/L	0.1792 ± 0.0295	0.0481 ± 0.0308	0.0168 ± 0.0158
April	β, Bq/L	0.2270 ± 0.0180	0.1008 ± 0.0212	0.0865 ± 0.0164
May	β, Bq/L	0.1485 ± 0.0136	0.1392 ± 0.0137	0.1311 ± 0.0116
June	β, Bq/L	0.3161 ± 0.0148	0.1636 ± 0.0181	0.1218 ± 0.0165
July	β, Bq/L	0.3045 ± 0.0133	0.2602 ± 0.0083	0.0782 ± 0.0073
December	β, Bq/L	0.1041 ± 0.0027	0.0257 ± 0.0031	0.0051 ± 0.0035
January	β, Bq/L	0.1353 ± 0.0031	0.1074 ± 0.0037	0.0044 ± 0.0025

When a monthly comparison of the radioactivity concentration of the raw wastewater is made, it is shown that contrary to the gross α radioactivity; the gross β radioactivity is at its highest level during the summer months. While the β

radioactivity increases greatly in June and July, the α radioactivity reaches its maximum level in the April and May months during the studying periods. The variations result from the diversities of the consumed foods, rain conditions and the characteristics of the other wastes received into the sewerage. The increases in the concentrations of the gross α radioactivity in the spring months can be explained by the foods consumed in this season: vegetables such as carrots and cabbages containing the α radioisotopes at high levels (Kuo et al. 1997) are consumed in much greater amounts than in other months. Similarly, the contents of the β active radioisotopes (such as ^{43}K and ^{137}Cs) of the foods consumed during the winter months are smaller in comparison to those of the summer months (Badran et al. 2003).

It was shown that α and β radioactivity in the effluent of the biological treatment sludge varied between 0.0168 and 0.0568 Bq/L, and 0.0044 and 0.1311 Bq/L, respectively. While the α radioactivity decreased by 52-90%, (except for May) β radioactivity decreased by 62-96%. Decreases in the radioactivity level occurred by the settling and adsorption of the radionuclides and the biological degradation of the minerals (Ipek et al. 2002) such as ^{14}C, and ^{40}K. To detect the reduction in the radioactivity level by settling, the radioactivity in the effluent of the primary settling tank is also determined. The α radioactivity, particularly, reduced much more by settling in January than the other months. This can be explained by minerals or particular matters causing α radioactivity prevalent in this month. The α radioactivity decreased by 29%, 61%, 42%, 44% and 42% in April, May, June, July and December, respectively. The β radioactivity reduced the minimum proportion by 6% in May and by 56%, 48%, 15%, 75% and 21% in April, June, July, December and January.

If it is accepted that the radioactivity levels in the analysed samples are the same as the radioactivity concentration of the discharged effluent, the amount of the α radioactivity given to Keban Dam Lake is 7.47, 4.99, 2.45, 3.13 MBq/day and 1.91, 1.51, 1.19, 1.35 MBq/day before and after treatment in April, May, June, July, respectively. The β radioactivity is also reduced from 7.64 to 2.91 MBq/day, from 4.61 to 4.07 MBq/day, from 10.05 to 3.87 MBq/day and from 9.62 to 2.47 MBq/day in April, May, June, and July, respectively. The loadings of radioactivity given to Keban Dam Lake were not determined in December and January because the flowmeter of the treatment plant was out of order.

The variation of the gross beta radioactivity concentration with ^{129}I, ^{137}Cs and ^{90}Sr radioisotopes and the reduction ranges of grossβ, ^{129}I, ^{137}Cs and ^{90}Sr activities are given in Table 2 and Table 3, respectively.

Table 2. Activity concentrations of gross β and some radioisotopes in wastewater, collected in various months.

Time	Influent				The Effluent of the Primary Settling Tank				Final Effluent			
	Grossβ (Bq/L)	^{129}I (Bq/L)	^{137}Cs (Bq/L)	^{90}Sr (Bq/L)	Grossβ (Bq/L)	^{129}I (Bq/L)	^{137}Cs (Bq/L)	^{90}Sr (Bq/L)	Grossβ (Bq/L)	^{129}I (Bq/L)	^{137}Cs (Bq/L)	^{90}Sr (Bq/L)
April	0.2270 ± 0.0180	0.0182 ± 0.0014	0.0272 ± 0.0022	0.0840 ± 0.0067	0.1008 ± 0.0212	0.0051 ± 0.0011	0.0141 ± 0.0029	0.0312 ± 0.0066	0.0865 ± 0.0116	0.0060 ± 0.0008	0.0156 ± 0.0021	0.0242 ± 0.0032
May	0.1485 ± 0.0136	0.0178 ± 0.0012	0.0134 ± 0.0016	0.0564 ± 0.0052	0.1392 ± 0.0137	0.0097 ± 0.0016	0.0167 ± 0.0096	0.0585 ± 0.0057	0.1311 ± 0.0116	0.0079 ± 0.0007	0.0120 ± 0.0025	0.0393 ± 0.0035
June	0.3161 ± 0.0148	0.0126 ± 0.0006	0.0253 ± 0.0012	0.0790 ± 0.0037	0.1636 ± 0.0181	0.0147 ± 0.0016	0.0245 ± 0.0027	0.0524 ± 0.0058	0.1218 ± 0.0165	0.0034 ± 0.0018	0.0207 ± 0.0028	0.0438 ± 0.0059
July	0.3045 ± 0.0133	0.0243 ± 0.0011	0.0609 ± 0.0026	0.0914 ± 0.0046	0.2602 ± 0.0083	0.0130 ± 0.0041	0.0390 ± 0.0012	0.1041 ± 0.0033	0.0782 ± 0.0073	0.0047 ± 0.0004	0.0094 ± 0.0009	0.0235 ± 0.0022
December	0.1041 ± 0.0027	Not detectable (N.D)	N.D	N.D	0.0257 ± 0.0031	N.D	N.D	N.D	0.0050 ± 0.0035	N.D	N.D	N.D
January	0.1353 ± 0.0031	N.D	N.D	N.D	0.1074 ± 0.0037	N.D	N.D	N.D	0.0044 ± 0.0025	N.D	N.D	N.D

Table 3. The reduction ranges of gross β and radioisotopes in the wastewater.

Time	Grossβ (%)	^{129}I (%)	^{137}Cs (%)	^{90}Sr (%)
April	61	67	42	41
May	11	55	10	30
June	61	73	18	44
July	74	80	84	74
December	95	N.D	N.D	N.D
January	96	N.D	N.D	N.D

The mean radioactivity concentration is reduced by 69%, 39% and 48% in ^{129}I, ^{137}Cs and ^{90}Sr radioisotopes during the biological treatment of wastewater. The variations according to obtained data for different months are not so significant. The variation between the monthly data is spread widely. It was found that 57, 58, 37, 58 % and 50, 60, 55, 60 % and 52, 48, 55, 48 % of gross β activity, according to influent, effluent and final effluent respectively, in April, May, June and July arose from the radioisotopes such as ^{129}I, ^{137}Cs and ^{90}Sr. The mean ratio of (^{129}I + ^{137}Cs + ^{90}Sr)/gross β in wastewater is about 50%. This means that the half of the gross β radioactivity is coming from these radioisotopes.

Although radioactivity reduces by the classic biological treatment method, the radioactivity given to the receiving environment by effluent is still at important levels. The decreases of radioactivity in wastewater result in increases of radioactivity in treatment sludge. Therefore, radioactivity must also be taken into consideration when treatment sludge is used for agricultural purpose. It can be said that the radioactivity removal at the wastewater treatment plant could occur by adsorption of radioactive matters to the activated sludge granules, their biological degradation, and final settling. The final effluent for all months without relation to influent radioactive values had a quite similar value for α and β radioactivity levels.

As a result, this study has shown that the radioactivity in municipal wastewater is at important levels and is reduced by biological treatment. The radioactivity in the municipal wastewater must be considered particularly at high flow rates, because, the small amount of doses in the wastewater may lead to significant doses in the receiving environment. The radioecological effects caused by the raw wastewater and effluent in the receiving environment should also be investigated.

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