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Anaerobic co-digestion of landfill leachate and sewage sludge: Determination of the optimal ratio and improvement of digestion by pre-ozonation

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Abstract

Background: Anaerobic co-digestion (AcoD) of various wastes is a suitable method for the removal of contaminants and biogas production. The first aim of this study was to determine the optimal ratio of landfill leachate (LL) and sewage sludge (SS) for AcoD, and the second one was to evaluate the effect of pre-ozonation of the mixture on AcoD.

Methods: The LL and SS samples were taken from landfill sites and municipal wastewater treatment plants (MWTPs), respectively. In the first step, five reactors were used and named R1 (100% SS), R2(100% LL), R3 (15% LL/85% SS), R4 (25% LL/75% SS), and R5 (45% LL/55% SS). Mesophilic anaerobic digestion (AD) was performed on the reactors and the optimal ratio was determined. In the second stage, the optimal mixtures were subjected to an ozonation process before AcoD.

Results: The results of the first stage showed that the highest efficiency removal of the total solids (TS), volatile solids (VS), and chemical oxygen demand (COD), and the highest biogas production belonged to R3 digester, containing 15% LL and 85% SS. In the second stage, the results showed that the removal efficiency of COD and VS in the ozonated sample at the dosage of 7.6 gO_3/h were 29.8% and 36.6% higher than the non-ozonated sample, respectively. Furthermore, in the ozonated sample, the biogas yield and the content of methane in the gas mixture were 27% and 9% higher respectively, compared to the non-ozonated sample.

Conclusion: According to the results, the appropriate ratio of LL to SS and pre-ozonation of LL/SS mixture have a great impact on the performance of AcoD.

Keywords: Sewage, Solid waste, Ozone, Anaerobic, Methane

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Introduction

The exponential growth of population and urbanization, and the development of social economy, coupled with the improvement of living standards, have increased the amount of municipal solid waste (MSW) generation worldwide (1).

According to the World Bank report, the world generates 2.01 billion tons of MSW annually, when looking forward, global waste is expected to grow to 3.40 billion tons by 2050, more than double population growth over the same period (2). Therefore, one of the big challenges that all the countries of the world are struggling with, is providing effective and sustainable management of MSW, along with a good sanitation.

In many developed and developing countries, landfilling is widely used as a simple, low-cost, and affordable method for the final disposal of MSW (3,4).

Although approximately, up to 95% of MSW accumulated worldwide is disposed of in landfills, this method has very potential to cause environmental pollution (5).

One of the most important problems in landfill management is the generation of landfill leachate (LL). LL is produced from liquid leached from the moisture of the waste itself as primary leachate, and by water that infiltrates the landfill and permeates through the waste as secondary leachate (6).

LL can be considered as high-concentration wastewater, with various chemical and organic impurities, as well as a wide range of microorganisms (7,8).

Therefore, the leachate from the landfill area and their release into the surrounding environment cause serious environmental concerns, especially the pollution of water sources (9). The properties and conditions of LL were affected by a variety of factors, including age, rainfall, climate, and the type of waste and its constituent compounds. By its age, LL is divided into three general classes, namely, young, intermediate, and old. Leachate from young landfills contains large amounts of biodegradable organic matter and a higher BOD/COD ratio, which turns into volatile fatty acids (VFAs) in anaerobic conditions and leads to a decrease in leachate pH. As the age of the leachate increases, the leachate enters the methanogenic stage. This phase is characterized by old leachate with low COD, high concentration of ammonium nitrogen, and low BOD5/COD ratio (7,10,11).

The collection and treatment of LL is one of the basic approaches in the management of MSW landfilling sites. Until now, various physical, chemical, and biological methods have been used individually or in combination for leachate treatment (11-16). The possibility of using biological methods as a suitable, cost-effective, and environmentally friendly method is an attractive option for LL treatment. However, the biological treatment of LL has challenges due to the high concentration of organic compounds, the presence of refractory organic compounds, low biodegradability, high ammonia concentration, heavy metals and sometimes containing toxic substances (17).

Among the solutions to overcome these challenges is the co-treatment of LL with other substrates such as municipal and industrial wastewater, animal waste, agricultural waste, and sewage sludge (SS) (18-21). On the other hand, in the past few decades, the research approach in the field of waste treatment, including LL, has been focused on the simultaneous recovery of resources and removal of pollutants (22). Therefore, in this regard, the anaerobic co-digestion (AcoD) of LL with other substrates to overcome the challenges of LL treatability as well as energy obtained through biogas production has been one of the suitable solutions for LL purification.

In general, simultaneous digestion compared to single digestion has advantages such as improving the stability of the process, increasing the loading rate of organic materials, increasing the production of biogas and methane, diluting the concentration of toxic substances, balancing nutrients, and improving the synergistic effect of microorganisms (23-25).

SS is one of the most important and widespread substrates that have been used in AcoD with other substrates, including LL. SS is the main by-product of municipal wastewater treatment plants (MWTPs), which contains highly volatile solids (VS) and many diverse microorganisms. In this regard, some research has been conducted with a focus on co-digestion of LL and SS with various purposes such as the effect of the type and age of leachate (young, intermediate, and old), the volume or weight ratio of the mixture of leachate and sludge, digestion temperature and the effect of pre-treatment on AcoD. Regardless of the differences between these studies, the overall result of most studies implies that the AcoD of LL has been performed with higher efficiency compared to separate digestion (26-28). Various pre-treatment methods such as pre-heating, coagulation, advanced oxidation, bio-augmentation, and enzymatic pretreatment of substrates including leachate and sludge have been used to improve the anaerobic digestion (AD) (29-33). Ozone, as a strong oxidizing agent, has been used for removing chemical and microbial contaminants from water and wastewater for decades (34). Ozone may be used as a pre-treatment, complete treatment, or as a post-treatment agent. So far, many applications of ozone as a pretreatment agent to improve the removal efficiency of pollutants from various matrices including water, wastewater, SS, and LL have been reported (35-39). The results of these studies have shown that the pre-ozonation of SS and LL has reduced the toxicity and increased the biodegradability of LL or SS (40-43).

Although, several studies have been conducted regarding the use of ozone for pre- or post-treatment of LL and SS separately, but studies on the pretreatment of LL and SS mixtures for AcoD are limited to only a few studies (43-45). This study was conducted to determine the optimal ratio of LL from the MSW landfill site and SS of the MWTP of Tehran city for AcoD. Another aim of the present study was to investigate the possibility of the performance enhancement of AcoD by the mixture pre-treatment by the ozonation process.

Materials and Methods

Landfill leachate and sewage sludge sampling

The LL samples were taken from active Aradkooh municipal solid waste facilities located in *the south of Tehran.* The site is more than 40 years old and now serves more than 10 million population.

The SS samples were taken from Tehran's large sewage treatment plant located in the south of Tehran. The origin of the sludge samples was primary sludge from the first sedimentation tank and secondary sludge (after the activated sludge process) from a belt filter thickener. Each time, 20 liters of leachate and sludge were taken and placed in polyethylene containers. The samples were stored near ice while transporting them to the laboratory and kept at 4 °C before further analysis.

During the research period, LL and SS sampling was performed twice. The first sampling was used for the first stage of this study, i.e., determining the optimal leachate/sludge ratio in AcoD. The second sampling was used for the second stage of the study, i.e., the evaluation of the pre-ozonation effects on AcoD of LL/SS.

Study stages

AcoD was performed on a laboratory scale on the real samples of LL and SS in two stages. The first stage of the study was conducted to determine the optimal ratio of LL to SS for AD. Therefore, at this stage, AD was performed on volume ratios of 0%, 15%, 25%, and 45% of LL to SS. In the second step, the ozonation process was applied to the optimal ratio of LL to SS before AD, and the effect of pre-ozonation on AcoD of LL/SS was evaluated.

Preparation of mixed LL and SS for the first stage of study

Primary and secondary sludge samples with equal volume ratio (50:50) were prepared by passing through a sieve with 45 mesh (0.35 mm). Volume ratios of LL to SS were considered as 0%, 15%, 25%, 40%, and 100%. So, five anaerobic digesters were used in this research. Characteristics of substrate fed to digesters with different LL to SS ratios are shown in Table 1.

Anaerobic digestion setup and experiment

Five borosilicate glass bottles with a total volume of 250 mL and a working volume of 200 mL were considered as batch-mode anaerobic digesters. These bottles were labeled R1 to R5. So, R1 and R2 were used for the digestion of SS and LL alone, and R3 to R5 were used for the AcoD of LL and SS with a volume ratio of 15%, 25%, and 45%, respectively. Figure 1 shows the image of one of the digesters with its components. As shown in Figure 1a, each digester was tightly closed with a heat-resistant cap. The cap contains two sealed holes. Through one hole, a glass tube was submersed into the digester for sampling. Through the other hole, a pipe equipped with a valve was passed to transfer the produced gases to the SKC Tedlar bag. To measure the volume of stored gases, after closing the gas valve, the bag was separated from the digester, and the gas volume was measured by connecting it to the measuring equipment. The measurement of gas was based on the downward displacement of water. As shown in Figure 1b, all digesters were immersed in a thermostatic water bath with controlled temperature in the mesophilic digestion range (35 ± 2 °C). To ensure uniform conditions in the content of the digesters, the digesters are shaken manually at least three times a day. The hydraulic retention time (HRT) of AD was considered 21 days.

Ozonation aparatous and expriment

As mentioned earlier in the second stage of the study, to evaluate the effect of ozonation on the AcoD of LL with SS, ozonation was performed on the optimal ratio of LL to SS (resulting from the first stage of the study) before AcoD. So, three samples, each with a volume of 500 mLl of LL and SS with the optimal ratio obtained from the first stage were subjected to ozonation with the amount of 2, 3.8, and 7.6 g/h for 60 minutes. Figure 1c shows the ozonation facilities. Ozone gas was produced from pure oxygen by a laboratory ozone generator (Model SS4, Shamim Sharif Company, Iran). To control the amount of injected ozone, the amount of oxygen flow entering the ozone generator was adjusted with a rotameter (C6-DSP-CA223003 model). Ozonation was performed in a container with a volume of 1 L on samples with a volume of 500 mL. The excess ozone gas was trapped through a KI trap. The gas trap was a 250 mLErlenmeyer flask containing a 2% KI solution. After finishing the ozonation period, the contents of each trap were poured into a beaker, and then, titrated with 0.05 normal thiosulfate and starch indicator solution. After the ozonation process, residual ozone was purged out from the liquid phase and reactor headspace by bubbling nitrogen for about 5 min. Then, samples were taken from the contents of each container, and parameters including solids, VS, COD, BOD, nitrogen, phosphorus, and pH were measured. Next, AD was performed on the 200 mL contents of each of the ozonized samples. AcoD conditions were similar to those in the first phase of the study.

Table 1. Characteristics of substrate fed to digesters with various LL to SS ratios

Parameters	Unit	R1 (100% SS)	R2 (100% LL)	R3 (15% LL, 85% SS)	R4 (25% LL, 75% SS)	R5 (45% LL, 55% SS)
TS	(g/L)	37.8	24	34	32.7	32
VS	(g/L)	31.9	19.4	30	28.5	26.3
VS/TS	-	0.84	0.80	0.84	0.83	0.82
BOD_5	(g/L)	5.6	8.15	6	6.23	6.75
COD	(g/L)	19.5	30.2	21.1	22.2	24.3
BOD_5/COD	-	0.29	0.27	0.284	0.28	0.277
Alkalinity	(g/L $CaCo_3$)	2	8.5	2.97	3.5	4.9
Ammonia	NH3-N mg/L	1.25	1.62	1.26	1.3	1.39
TP	(mg/L)	185	164	181.8	179.7	175.5
pH	-	6.8	7.57	7.1	7.1	7.2

SS, sewage sludge; LL, landfill leachate; TS; total solids; VS, volatile solids; COD, chemical oxygen demand; BOD5, biochemical oxygen demand.

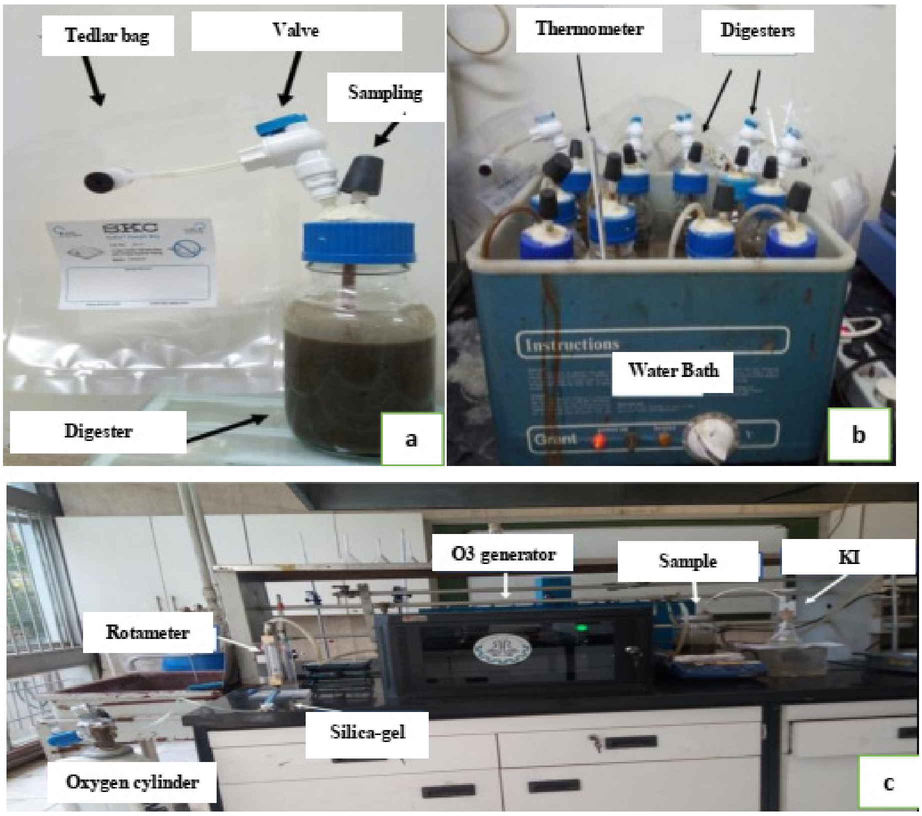

Figure 1. Images of anaerobic reactors and ozonation apparatus (a) One of the digesters and its accessories, (b) Digesters in water bath, (c) Ozonation apparatus

Analysis

Total solids (TS), VS, pH, total alkalinity (TAC), biochemical oxygen demand (BOD5), chemical oxygen demand (COD), total phosphorous (TP), and ammonia nitrogen (NH4-N) were determined according to the standard procedures (46). TS and VS were determined according to gravimetric method (2540G), pH was measured using an electrode pH meter (Corning M-220), TAC was measured by titrimetric method (2320 B), COD was analyzed by closed reflux method (5220 C) with COD instrument (DRB 200, HACH, USA), BOD5 value was examined through determining oxygen consumption by titrimetric method (5210 B), TP was determined by the persulfate + vanadomolybdophosphoric method (4500-P.C), and NH3-N was quantified using the kjeldahl digestion method (Kjeltech auto analyzer, VAP300, Gerhardt). Biogas analysis was done by gas chromatograph (GC-2552 TG, Iran).

Results

AcoD of various ratios of LL and SS

Removal efficiency

Figures 2a-c show the inlet and outlet concentrations of TS, VS, and COD parameters and their removal efficiency in digesters R1 to R5, respectively. As shown in these figures, the highest removal efficiency for all pollutants was obtained in reactor R3 (with 15% LL/85% SS). In reactors with a ratio of LL to SS higher than 15%, the pollutants removal efficiency had a decreasing trend. Also, the lowest removal of pollutants was related to the reactor containing only LL.

According to Figure 3a, in this study, the concentration of ammonia nitrogen has increased in the effluent of all digesters. As shown in Figure 3a, the highest increase of ammonia nitrogen occurred in R2(100% LL) digester. Also, Figure 3b shows that phosphorus concentration increased during AD in all reactors. This increase was higher in reactor 2 with 100% LL and lower in the reactor with 100% SS.

pH changes

pH is known to be an important factor that affects anaerobic digester stability. The measured initial and final pH of all digesters are presented in Figure 4. The inlet pH for all reactors was in the range of 6.86-7.5. The lowest and highest increase in pH was observed in R2 (100% LL) and R3 (15% LL, 85% SS) reactors, respectively.

Biogas yield

As seen in Figure 5, the maximum biogas efficiency of 194 ml/gVS$_{removed}$ was obtained in the R3 (15% LL,85% SS) reactor. After reactor R3, the amount of biogas production in reactor R1 (SS 100%), which was fed only with sludge, was higher and equal to 138 mL/gVS$_{removed}$. Therefore, a

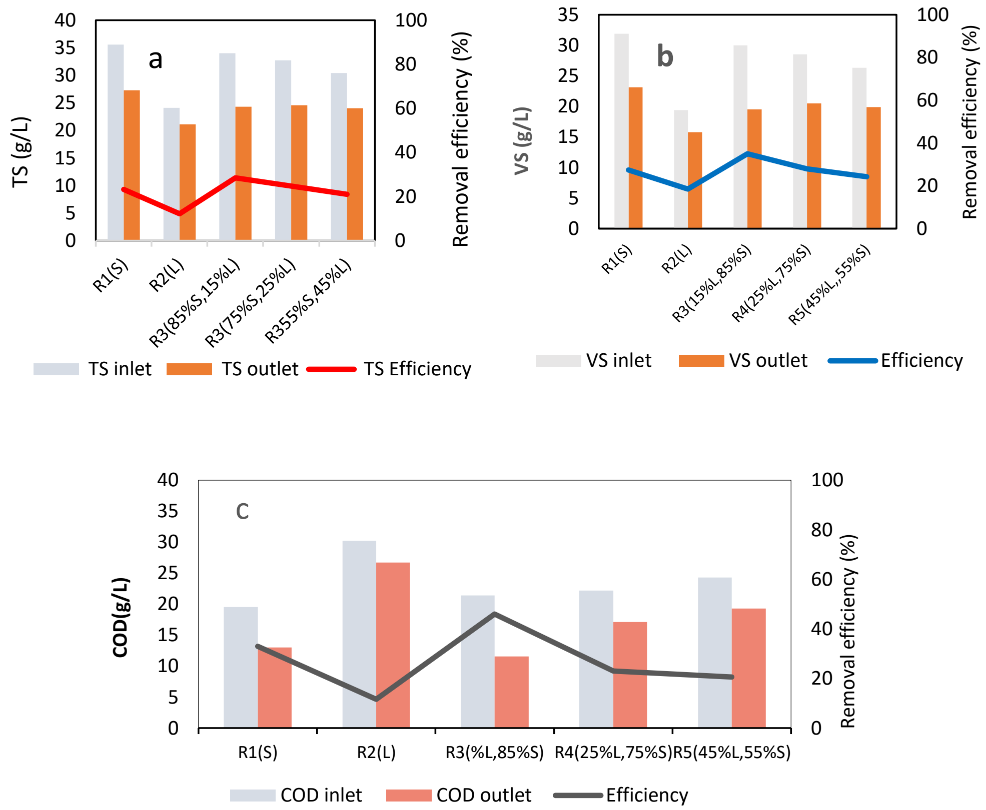

Figure 2. Inlet and outlet concentration and removal efficiency of (a) TS, (b) VS, and (c) COD in digesters containing various ratios of leachate to sludge

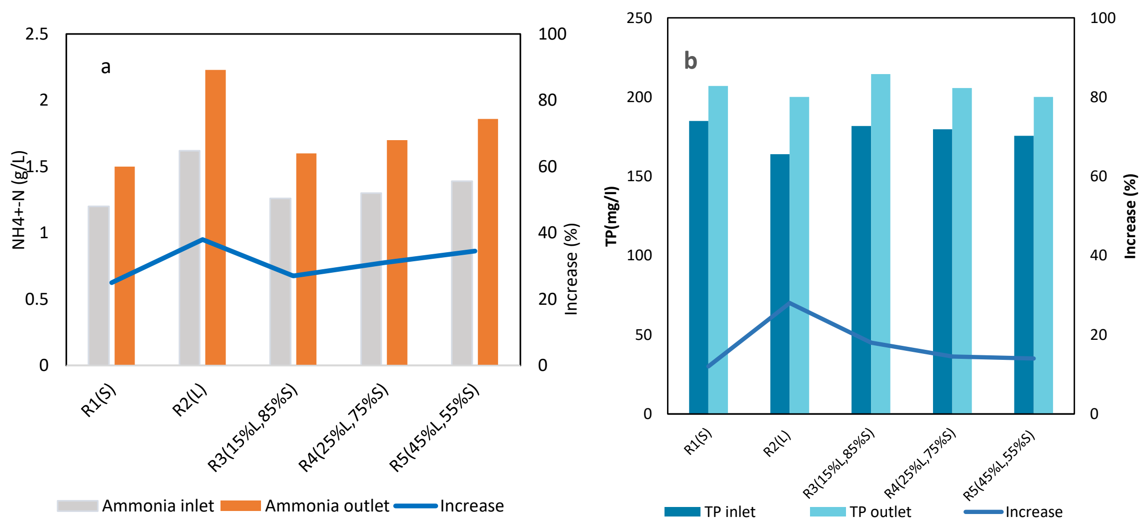

Figure 3. Inlet and outlet concentration and removal efficiency of (a) Ammonia, and (b) Phosphorous in digesters containing various ratios of leachate to sludge

decrease in biogas production was observed in reactors with higher than 15% leachate ratios. Also, according to Figure 5, the minimum biogas yield (70 mL/gVS$_{removed}$) was related to the R2 reactor fed with LL alone.

Pre-ozonation of LL and SS sludge mixture

To determine the effect of pre-ozonation on the performance of AD, three samples of SS and SS mixture with the optimal ratio obtained from the previous step (i.e., 15% LL, 85% SS) were prepared and subjected to ozonation at the dosage of 2.1, 3.8, 7.6, gO_3/h for 60 min reaction time before AD. Table 2 shows the results of ozonation on the mixture of LL and SS for 60 minutes with three doses of ozone. As can be seen in Table 2, ozonation with a low dosage (2.1 gO_3/h) had little effects on reducing the parameters. However, by increasing the ozone dosage to 3.8 gO_3/h and 7.6 gO_3/h, a relatively greater reduction of pollutants was achieved, especially

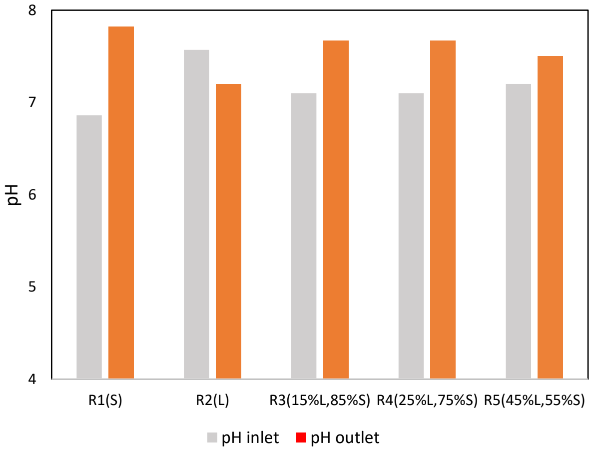

Figure 4. Inlet and outlet of pH in digesters containing various ratios of leachate to sludge

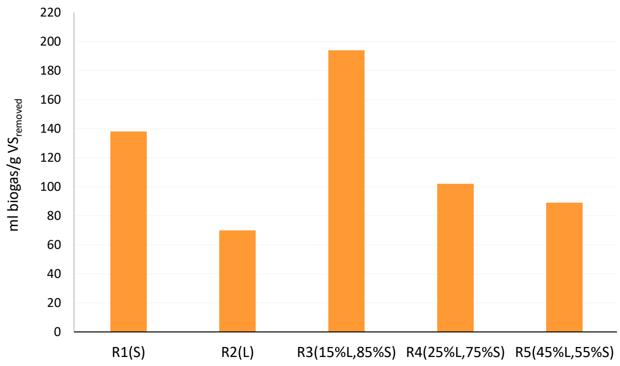

Figure 5. The value of biogas production from digesters containing various ratios of leachate to sludge

in the concentration of COD. Also, as shown in Table 2, ozonation pretreatment at a dose of 7.6 gO_3/h led to an increase in the BOD_5/COD ratio from 0.29 to 0.4, which improved the biodegradability of the substrate. However, the results showed that increasing injected ozone dosage enhances the removal efficiency of contaminants.

AcoD of pre-ozonated LL/SS mixture

AcoD was performed on the samples before and after ozonation with the characteristics shown in Table 2. The inlet and outlet concentrations as well as the removal percentage of TS, VS, and COD parameters under AD are shown in Figures 6a-c. As shown in the figures, the removal efficiency of pollutants under the AD process in the pre-ozonated samples is higher than in the non-ozonated samples. In addition, in the samples pretreated with a higher concentration of ozone, the removal of their pollutants was done more efficiently during the AD process. So that as can be concluded from Figures 6a-c, the removal efficiency of TS, VS, and COD in the reactor containing the pre-ozonized sample with the concentration of 7.6 gO_3/h was 32.8, 36.6, and 29.8% higher than the reactor containing the non-pre-ozonated sample. These results confirm the positive effect of pre-ozonation as a pre-treatment for the AcoD of SS and LL.

Figure 7 shows the pH values of samples before and after AD. As shown in the figure, the pH of all samples increased after digestion and was in the desired range for AD. The volume of biogas produced from AD of ozonated and non-ozonated samples is presented in Figure 8. As presented in Figure 8, in all pre-treated samples by ozone, the volume of biogas produced is higher than in a non-pre-treated sample. According to Figure 8, the volume of biogas produced by the pre-treated sample with an ozone dose of 7.6 gO_3/h was about 27% higher than the non-ozonated sample.

In addition, in this study, pre-ozonation was conducive to an increase in the content of methane gas in the produced biogas mixture, so that in the sample ozonated with the amount of 6.7 gO_3/h, the percentage of methane gas in the biogas content was measured as 65%, while it was 58% in the non-ozonated sample.

Discussion

Leachate and sewage sludge characteristics

Table 1 represents the measured characteristics of LL, SS, and various ratios of their mixture, which were used in this research. According to the SS and LL characteristics shown in Table 1, the concentration of TS, VS, and phosphorus in the SS was higher than that of the LL, and on the contrary, the values of BOD, COD, ammonia, and TAC in the LL were higher than SS. As shown in Table 1, both the SS (the content of R1) and LL (the content of R2) had a high concentration of solids with a high proportion of VS, which indicates the presence of high organic compounds in them. The ratio of BOD_5/COD of LL was 0.27, confirming that the leachate was in the intermediate phase of decomposition (26). Although the SS ratio of BOD_5/COD was 0.29 higher than the LL but was also low and relatively close to the suitable range for biological decomposition.

As shown in Table 1, the total alkalinity (TAC) of both substrates is relatively high. TAC is an important parameter that affects AD (47). According to the results presented in Table 1, both LL and SS have a relatively high concentration of ammonia. Although ammonia is an essential nutrient for bacterial growth, at high concentrations, it is considered a potential inhibitor during AD (48).

The optimum ratio of LL and SS for AcoD

The results of this study showed that in reactor R3 containing 15% LL/85%SS, the removal efficiency of impurities was higher than in the reactor containing only sludge. But in reactors with higher than 15% leachate, lower efficiency was observed. Similarly, the results of several studies have suggested that adding LL to SS or other substrates in appropriate ratios has improved AD performance. In the studies conducted by different

Table 2. The effect of various concentrations of ozone on the removal of contaminants and the biodegradability of the mixed substrate (15% LL/85% SS)

Parameter	Unit	Before Ozonation	After Ozonation			Removal (%)		
			2.1 gO_3/h	3.8 gO_3/h	7.6 gO_3/h	2.1 gO_3/h	3.8 gO_3/h	7.6 gO_3/h
COD	g/l	22	20.4	18.08	14.52	7.27	17.8	34
BOD_5	g/l	5.8	5.73	5.47	5.04	1.2	5.6	13
VS	g/l	27	26.4	26	25.6	2	3.5	4.9
TS	g/l	39.8	39.5	37	35	2	7.56	12
pH	-	7.3	7.1	6.8	6.5	2.74	7	11
Alkalinity	g/l $CaCO_3$	3.12	2.88	2.84	2.73	7	8.9	14.2
Ammonia	g/L	1.42	1.4	1.38	1.21	1.4	2.8	14.7
BOD_5/COD	-	0.26	0.28 (7.7% increase)	0.3 (13.3% increase)	0.35 (25.7% increase)			

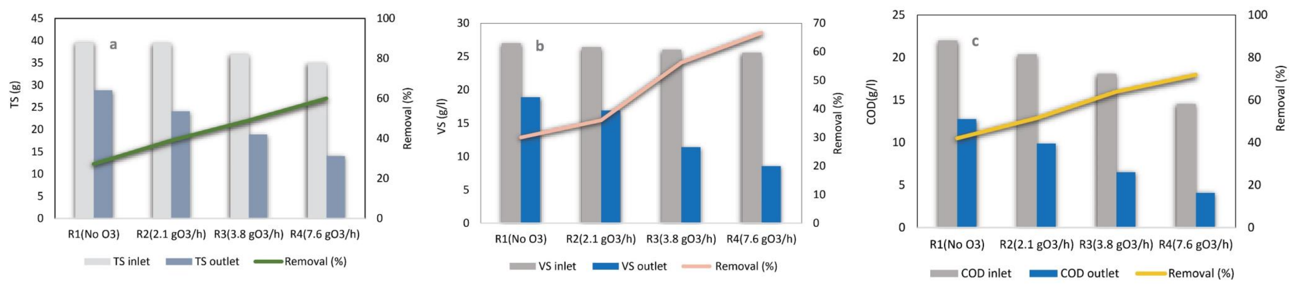

Figure 6. Inlet and outlet concentration and removal efficiency of a) TS - b) VS - c) COD in digesters containing ozonated and non-ozonated samples (15% LL/85%SS)

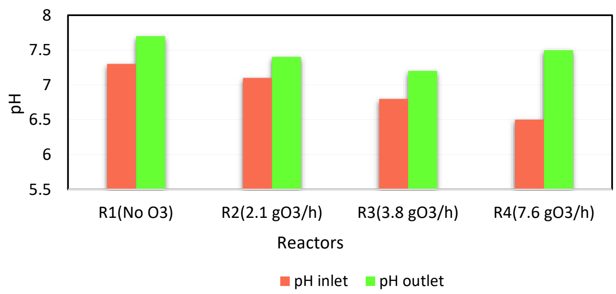

Figure 7. The inlet and outlet of pH in digesters containing ozonated and non-ozonated samples (15% LL/85% SS)

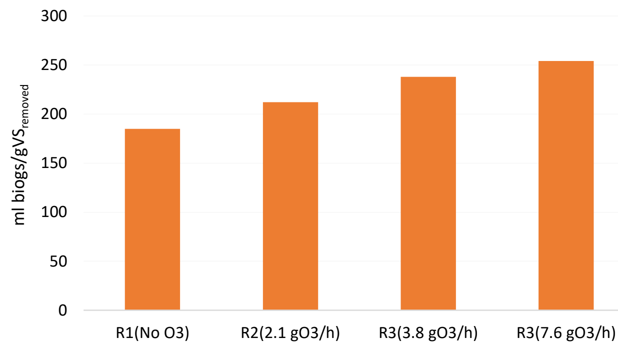

Figure 8. The value of biogas production from digesters containing ozonated and non-ozonated samples (15% LL/85% SS)

researchers, the appropriate ratio of leachate to substrate has been reported differently. According to the research by Montusiewicz and Lebiocka, the best results of AcoD of intermediate LL and SS were reported in a volumetric ratio of 1:20 (27). The results of the study by Gao et al showed that in the AcoD of LL and waste-activated sludge, adding the LL with a ratio of 18.2% resulted in the highest efficiency of the hydrolysis process and breakdown of large organic macromolecules. This issue has led to the improvement and acceleration of the acid production processes, and ultimately, to increase the activity of methanogenic bacteria and biogas production. In the present study, the lowest removal of pollutants was related to the reactor containing only LL. Based on the results of the present study and other reports, high ammonium concentration, low BOD_5/COD and VS/TS ratios, and the presence of resistant and possibly toxic organic compounds in the LL lead to an unsuitable substrate composition for AD (49-51). The use of high proportions of LL has imposed negative effects on all stage processes, especially in the stage of methane production (26).

In general, studies have shown that by adding the appropriate LL to SS ratio, microorganisms have access to more diverse nutrients that enhance growth and improve the efficiency of AD and biogas production. In addition, LL may contain adapted bacteria that help break down complex organic compounds. These bacteria are often different from the natural bacteria present in the SS and can provide a bacterial population with a higher potential to perform the AD process (18,52).

In this study, the concentration of ammonia nitrogen has increased in the effluent of all digesters. As shown in Figure 3a, the highest increase of ammonia nitrogen occurred in R2(100% LL) digester. As previously stated, the lowest efficiency of contaminant removal has occurred in this reactor. Ammonium is released during the anaerobic hydrolysis of organic nitrogen compounds and in high concentrations may lead to the inhibition of the biological process (53-55).

As mentioned in the results, phosphorus concentration increased during AD in all reactors. This increase was higher in reactor 2 with 100% LL and lower in the reactor with 100% SS. Similarly, in the study conducted by Guven et al, titled AD of organic fraction of MSW and LL, the release of phosphorus and the increase in its concentration was reported (23). In anaerobic conditions, some microorganisms can take up carbon sources such as VFAs, and store them intracellularly as poly b-hydroxyalkanoates (PHAs). The energy for these biotransformations is mainly generated by the cleavage of polyphosphate and the release of phosphate from the cell (56).

In the AD process, the stable performance of the process during the digestion period is determined by indicators such as pH, the ratio of TAC/VFA, and the amount of biogas production.

pH is known to be an important factor that affects anaerobic digester stability (57). In this study, pH reduction occurred only in digester R2 containing 100% LL. VFA and TAC play an important role in maintaining a stable pH in the AD process. VFAs are produced by microorganisms during the decomposition of organic matter and are important intermediates in biogas production. On the other hand, the appropriate amount of TAC as a provider of buffering capacity of the system versus VFA production is necessary for proper operation and by breaking down the VFA as a result of the AD process, the pH value of the samples increases (58).

Biogas production as an energy source is one of the advantages of AD of SS and LL. On the other hand, the biogas yield in the AD process is considered as an indicator of the degradability of the substrate that is fed to the system and if the process is well operated or not.

Based on the results, adding LL to SS with a suitable ratio (15% in this study) improves process performance, and also, increases biogas yield. In this study, biogas yield efficiency in the reactor with a ratio of 15% LL:85% SS was increased by 64% and 28.8%, compared to LL alone and SS alone, respectively. The results of most studies confirm the increase of biogas yield by adding a certain amount of LL to SS or other substrates. In a study by Nikiema et al, regarding the optimization of co-digestion of SS and bovine dung for biogas production, the addition of leachate improved the digestion performance and increased biogas production (28). In a study titled AcoD of intermediate LL and SS, at the optimum ratio of 20 SS:1 LL, the biogas yield was 130 $m^3/kg\ VS_{removed}$, which was 13% higher than SS alone. In addition, methane yield was exceeded 16.9% (18).

According to the results of studies including this study, LL has both synergistic and antagonistic effects on the process of AD and biogas production. In general, the addition of fresh or medium LL in a small amount due to the high content of VFA as well as nutrients and trace elements and increasing the diversity of decomposing microorganisms increase biogas production. The negative effects of LL in high proportions can be attributed to the release of ammonia, inhibitory organic substances, and heavy metal toxicity (18,26).

Pre-ozonation of LL and SS sludge mixture

Based on the results, it was observed that ozonation of the mixture of LL and SS caused the removal of a percentage of pollutants, and improved the biodegradability of the mixture. However, the results showed that increasing injected ozone dosage enhances the removal efficiency of contaminants. Similar results have been obtained in other studies. In the study by Wu et al, LL after the coagulation process was subjected to ozonation process at the rate of 0.6 g/L for 30 minutes. In the study by Wu et al, a COD removal rate of about 30% was achieved, and the ratio of BOD/COD increased from 0.08 to 0.4 (59). In another report, the ozonation of the LL at a dose of 50 g/m^3 concentration for 60 minutes resulted in 25% efficiency in removing COD. Also, the ratio of BOD/COD increased slightly and reached from 0.1 to 0.15 (60).

In an experiment study conducted by Derco et al, investigating the effect of ozone treatment on the biodegradability of recalcitrant pollutants contained in the biologically pretreated LL, ozonation was performed for up to 30 hours, but the maximum COD removal of about 65% was achieved in the initial 11.5 hours of reaction and was not significant in longer times. Also, the BOD_5/COD ratio increased from 0.03 to 0.21 (61).

Ozone as a powerful oxidant (2.07 V) can degrade organic pollutants using two mechanisms: (1) Direct electrophilic attack by molecular ozone; and (2) indirect attack by •OH radicals produced through the ozone decomposition process. It was observed that raising the ozone concentration promotes the degradation rate of some pollutants but has no obvious effect on the degradation of some other pollutants (62).

Effect of pre-ozonation on AcoD

Based on the results of the present study, it was found that pre-ozonation of the mixture of LL and SS improves the digestion process and leads to the removal of more organic matter as well as higher yields of biogas. The most important positive effects of pretreatment with ozone can be attributed to increasing the ratio of BOD/

COD, breaking macromolecules into simpler molecules, solubilization of VSS and COD, and decreasing viscosity of substrate (42,45,61). Increasing the ratio of BOD/COD as a result of pre-ozonation increases the decomposition of organic matter. Also, pre-ozonation causes the release of soluble substances in the aqueous environment and increases the availability of compounds for microorganisms. Ozonation can disintegrate the SS mass and release COD, proteins, and polysaccharides from the solid phase to the aqueous phase, thus, improving and accelerating the decomposition of organic matter, and subsequently, increasing the amount of methane production during AD (43). In the study conducted by Carballa et al, pre-ozonation of pharmaceutical waste and personal care products before AD resulted in the dissolution of about 8% of VS and 60% of COD. The use of this process led to an increase in the biogas yield and the soluble organic matter removal efficiency during AD (44). In another study, pre-ozonation of waste-activated sludge before AD led to 22% and 25% solubilization of COD and TS, respectively. Moreover, the apparent viscosity of sludge was decreased (63).

As shown in this study, the highest biogas production was achieved in the reactor with a 15% LL/85% SS ratio, which compared to AD of SS and LL alone, the amount of biogas production increased by 28.9% and 64%, respectively. Therefore, from the point of view of energy recovery from waste, this issue can be one of the important advantages of co-digestion of LL and SS with an optimal ratio. Furthermore, in the ozonated sample containing 15% LL/85% SS, the biogas yield and the content of methane in the gas mixture were 27% and 9% higher, respectively, compared to the non-ozonated sample.

Conclusion

This study was conducted to determine the optimal ratio of the LL and sewage SS for the AcoD. Also, the impact of pre-ozonation on the improvement of the AcoD was evaluated. AD was conducted on three ratios of LL to SS (15% LL/85% SS, 25% LL/75% SS, and 45% LL/55% SS). Among the digested samples, in the AcoD of 15% LL/85% SS sample, the highest removal efficiency of VS, COD as well as the highest biogas production was achieved. AD of SS alone was the next rank in the removal efficiency of pollutants and gas production. Also, in the reactor containing LL alone, the lowest efficiency was achieved. The assessment of the impact of pre-ozonation on the AcoD identified that pre-treatment of mixture (15% LL/85% SS) with ozone can improve the AD performance through the increase in the ratio of BOD/COD, breaking macromolecules into simpler molecules and solubilization of COD and TS. The results showed that the removal efficiency of COD and VSS in the pre-ozonated sample at the dosage of 7.6 gO_3/h was 29.8% and 36.6% higher than in the non-ozonated sample, respectively. According to the results of the present study, the appropriate ratio of leachate to sludge can have a great impact on the amount of biogas produced.

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Competing interests

The authors declare that they have no conflict of interests.

Ethical issues

The authors hereby certify that all data collected during the research are as expressed in the manuscript, and no data from the study has been or will be published elsewhere separately. This research was approved by the Vice-Chancellor for Research and Technology of Shahid Beheshti University of Medical Sciences, Tehran, Iran (Ethical code: IR.SBMU.PHNS.REC.1400.0176).

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