Application of leachate recirculation on the concentration of landfill gases

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Abstract. Leachate is the product of the biodegradation process in the landfill. On-site treatment of leachate using leachate recirculation is one of the alternative methods to reduce the hazard. The operation of leachate recirculation provides benefits such as speeding up biodegradation, lowering pollutant concentrations, and increasing gas production. This study aims to evaluate the application of leachate recirculation on the concentration of CO2 and CH4 produced. Experiments were performed in a laboratory using 20 lysimeters, with 1 L in volume for 365 days. The lysimeter was divided into two groups, with 10 reactors, each group arranged in series. Leachate recirculation will be given to the second reactor until the tenth reactor, using a high leachate concentration for the first group and a low concentration of leachate for the second. The addition of leachate in the two reactor treatment groups significantly increased the organic content in the leachate. Leachate recirculation does not cause a significant escalation in CO2 and CH4 concentration compared to reactors without leachate recirculation. In the reactor group with high leachate concentrations, reactors with leachate recirculation produced a more stable gas concentration than those without leachate recirculation which produced more volatile CH4 concentration.

1. Introduction
Waste management problems are issues that have not been fully resolved until now. Various efforts have been made to manage waste, such as 3R (reducing, reusing, recycling), composting, burning waste (incinerators), and landfilling. Of all the available methods, landfills' final waste processing method is Indonesia's most acceptable and widely used waste management system.

Leachate is the primary material produced by a landfill and gas production, resulting from water percolation in the waste heap [1]. Leachate contains many organic and inorganic materials and other contaminants in the landfill that the leachate produced by a landfill must be processed first. Researches on leachate treatment are still less attentive even though leachate contains highly polluted materials [2]. The leachate management method commonly used at landfills in Indonesia is an off-site management method with high operating and maintenance costs, such as human resources, consumables, equipment, energy sources, and others.
Another alternative leachate management method is the on-site or in-situ management method: leachate treatment by returning the generated leachate to the garbage dump. This method allows the waste pile to be used as a bioreactor at the same time. Research on landfills as bioreactors has been conducted for more than 20 years, including laboratory-scale (laboratory-scale reactors), pilot-scale tests (pilot-scale lysimeters), and comprehensive demonstrations of landfills (full-scale lysimeters). The study results show that the operation of the bioreactor can control the decomposition process of waste in the landfill and minimize the long-term risks to humans and the environment [3]. In addition, based on various previous studies, the recirculation of leachate to landfills can provide various advantages, such as increasing the moisture in the waste, accelerating biodegradation, reducing the time required for the stabilization process and reducing the total amount of leachate: volume and concentration. Disposal must be done after the landfill is closed, increasing the speed at which the landfill is closed. In addition, total gas production, accelerating the growth of microbial populations, increasing the distribution of nutrients and enzymes, controlling pH, diluting materials that inhibit the decomposition rate (inhibitors), recycling and distribution of methanogenic bacteria, storage of leachate, and increasing percolation Chance of filtrate evaporation [1, 3, 4, 5, 6]. This study aims to evaluate the application of leachate recirculation on the concentration of CO$_2$ and CH$_4$ produced.

2. Methodology
The research was carried out on a laboratory scale (experimental laboratory), using 60 lysimeters (waste reactors), of which 30 were used for high-concentration leachate, and 30 were used for low-concentration leachate. The leachate used in this study is artificial leachate with high concentration (BOD 3,758.19 mg/L and COD 7,406.67 mg/L) and low concentration (BOD 641.30 mg/L and COD 1,279.33 mg/L). Recycling and fruit waste in the container. The research is carried out in three phases: the preparation phase, the implementation phase, and the data analysis has been carried out for one year, or the waste has reached a steady-state (the leachate concentration has reached <1000 mg/L). Leachate recycling is carried out under two operating or processing conditions. The first treatment is a reactor with a high leachate concentration, and the second treatment is a reactor with a low leachate concentration.
3. Results and discussion

3.1. CO₂ concentration
CO₂, H₂, and CH₄ are the leading gases produced in the anaerobic decomposition of organic matter. The presence of these gases can describe the stages that occur in the decomposition of organic material. Changes in the concentration of CO₂ in each of the reactor groups studied can be seen in Figure 2 and Figure 3 below.

Figure 1. Reactor operational conditions.
Figure 2. CO$_2$ concentration in a reactor with low C leachate addition.

Based on the graph above, it can be seen that the CO$_2$ concentration in the reactor group with the addition of low concentration leachate, in the first month, the highest CO$_2$ concentration occurred in the R8 reactor with a value of 404.441 ppm and the lowest in the R5 reactor with a concentration of 108.398 ppm. In the second month, the concentration of CO$_2$ in all reactors increased, with the highest value of 529.888 ppm in reactor R10. In the 4th month, the concentration of CO$_2$ in all reactors decreased quite sharply, with the lowest concentration of 23.235 ppm occurring in reactor R8. The new R1 reactor decreased in the 5th month with a concentration of 114.201 ppm and reached the lowest value in the 7th month with a concentration of 4.628 ppm. Furthermore, the CO$_2$ concentration was relatively stable until the 17th month.

Figure 3. CO$_2$ concentration in a reactor with high C leachate addition.
In the reactor group with the addition of high concentration leachate, the concentration of CO₂ in the first month reached the highest value in reactor R5 of 372.748 ppm, higher than the group with the addition of low concentration leachate. In comparison, the lowest CO₂ concentration occurred in reactor R1 with a value of 670 ppm. In the 2nd month, the CO₂ concentration in almost all reactors experienced the same pattern as in the reactor group with low concentration leachate, which increased and then decreased sharply in the 4th month. The highly fluctuating CO₂ concentrations pattern occurred in the R1 and R2 reactors, where the R1 reactor fluctuated throughout the study period. In contrast, the R2 reactor only experienced a sharp increase in the 7th month with a value of 324.513 ppm, which decreased in the 8th month. Followed the pattern of decreasing CO₂ concentration as in other reactors. Furthermore, the CO₂ concentration was relatively stable until the 17th month.

This phenomenon can be explained as follows: In the early stages of the waste decomposition process, the oxygen content trapped in the waste heap will be depleted gradually because it is consumed by aerobic bacteria, where there is a transition process from the aerobic phase to the anaerobic phase \[7\]. At this stage, the fermenting bacteria carry out hydrolysis and fermentation on organic material, producing dissolved sugars, amino acids, carboxylic acids, and glycerol. The gases produced at this stage are H₂ and CO₂ \[8, 9\]. It can be seen both in the reactor with the addition of leachate at low and high concentrations. In the first month to the fourth month (hydrolysis and acetogenesis phase), the largest concentration of CO₂ gas was detected. At this stage, the CO₂ produced will jump to the next stage, namely acetogenesis, which together with H₂ will be converted into CH₄ by hydrophilic methanogen bacteria. In the acetogenesis phase itself, acetogenic bacteria will also change the decomposition products of organic material produced from the previous phase into Acetic Acid, H₂, and CO₂, which are then converted into CH₄ by hydrophilic methanogen bacteria \[7\]. This happened in both reactor treatments, which can be seen in the 6th to 17th month, CO₂ gas was still detected in lower concentrations.

An anomaly occurred in reactor 1 (control reactor) in the treatment group with a high concentration of leachate addition, where CO₂ was still detected with a relatively high fluctuating concentration. This shows that the R1 reactor is still experiencing the initial phase of the organic material decomposition process, namely the hydrolysis, acidogenesis, and acetogenesis phases, where CO₂ is still produced and has not been converted into CH₄. This is also an indicator that the organic content in the R1 reactor is still relatively high, with the decomposition process running slowly. The high organic content will trigger acids in high concentrations, which will produce CO₂ and H₂ gases in high concentrations \[10\]. This acidic condition will also suppress the activity of methanogenic bacteria to slow down the conversion of Acetic Acid, CO₂, and H₂ to CH₄. Even in extreme conditions, it can stop the whole process of CH₄ formation \[6\].

3.2. CH₄ concentration

CH₄ is also the primary product gas produced in the anaerobic decomposition process besides CO₂. The formation of CH₄ occurs at the final stage of the organic material decomposition process, which can also indicate the maturity level of organic waste. In this study, apart from CO₂, the production of CH₄ in each treatment reactor group was also observed to determine how the decomposition process occurred in each reactor. The graph of changes in CH₄ in the treatment of the addition of low and high concentrations of leachate can be seen in Figure 4 and Figure 5 below.
Figure 4. CH$_4$ concentration in a reactor with low C leachate addition.

Based on the graph above, in the reactor with the addition of low concentration leachate, it can be seen that in the first month, all reactors produced CH$_4$, especially reactor one, which produced CH$_4$ with the highest concentration compared to other reactors with a value of 55.69 ppm. The lowest concentration occurred in reactor 5, with a value of 20.95 ppm. Furthermore, the concentration of CH$_4$ in all reactors decreased and tended to be stable until the end of the study, except for reactor 1, which in the 16th to 17th months experienced a sharp increase in CH$_4$ concentration up to 126.11 ppm.

The above conditions can be explained as follows: because in the first month, the presence of CH$_4$ has been detected, it can be concluded that there has been a rapid decomposition process of organic material in all reactors (already in the final phase). At the same time, CO$_2$ has also been detected, that it can be assumed that the decomposition process in the first month has simultaneously reached the stages of acetogenesis and methanogenesis. According to research [4], the presence of CO$_2$ and CH$_4$ occurs in the acid formation phase. Acid formation phase and maturation phase. This condition can also be caused by the decomposition process of organic material that is not simultaneously even in the same reactor, where some organic material is still decomposed in the initial phase. There is some organic material that has decomposed in the final phase. This condition can also be caused by the leachate recirculation process that cannot guarantee that the leachate can be spread evenly throughout the organic material in the reactor. Based on the higher CO$_2$ concentration compared to CH$_4$, it can be assumed that the organic material that has reached the final phase of the decomposition process is relatively less than the material still in the initial phase. In the reactors with a low concentration of leachate treatment, reactor 1 had a higher CH$_4$ concentration than the other reactors. It was appropriate when compared to the relatively low concentration of CO$_2$ in the same reactor. Under these conditions, it can be assumed that some of the CO$_2$ in reactor 1 has turned into CH$_4$. [8, 9].
Figure 5. CH₄ concentration in a reactor with high leachate addition.

The reactor group that received high leachate concentration had a different CH₄ gas production pattern compared to that of the reactor with a low leachate concentration. In the first month, all reactors produced CH₄ gas with the highest concentration of 18.48 ppm, which occurred in reactor 6, while the lowest concentration occurred in reactor 1 with a value of 11.55 ppm. This value is lower than the highest concentration achieved in the reactor group with low concentration leachate. Reactor 3 to reactor 10 experienced an average peak of CH₄ concentration in the 3rd and fourth months, which then decreased and remained stable until the end of the study. Reactor 2 experienced a sharp decrease in CH₄ concentration until the 6th month, which increased quite sharply to 39.99 ppm in the 7th week. In the 8th week, the concentration of CH₄ in reactor two again decreased sharply and then stabilized until the end of the study. Reactor R1 is the reactor that experiences the most fluctuations in CH₄ concentration changes. The highest concentrations were achieved in the 12th and 15th months with 62.63 ppm and 62.69 ppm. The concentration of CH₄ then decreased sharply in the 16th month with a value of 15.41 ppm.

The above conditions in some reactors have the same pattern when compared to the reactor group with the addition of low concentration leachate, where in the first month, the presence of CH₄ has also been detected, which means there has also been a rapid decomposition process of organic material in all reactors (already in the final phase). At the same time, CO₂ has also been detected, so it is assumed that the decomposition process in the first month has simultaneously reached the stages of acetogenesis and methanogenesis. According to research [4], the presence of CO₂ and CH₄ occurs in the acid formation phase, methane formation (acid formation phase), and maturation phase. This condition can also be caused by the decomposition process of organic material that is not simultaneously even in the same reactor, where some organic material is still decomposed in the initial phase, and some organic material has decomposed in the final phase. This condition can be caused by the leachate recirculation process that cannot guarantee that the leachate can be spread evenly throughout the organic material in the reactor. Based on the higher CO₂ concentration compared to CH₄, it can be assumed that the organic material that has reached the final phase of the decomposition process is relatively less than the material still in the initial phase. In the reactors with a low concentration of leachate treatment, reactor R1 had a higher CH₄ concentration than the other reactors. It was appropriate when compared to the relatively
low concentration of CO$_2$ in the same reactor. Under these conditions, it can be assumed that some of the CO$_2$ in reactor 1 has turned into CH$_4$ [7, 8].

Similar to the CO$_2$ concentration in the R1 reactor, the CH$_4$ concentration fluctuated quite a lot, increasing and decreasing throughout the study period. These conditions can illustrate a partial decomposition process that occurs in the organic material in the R1 reactor. At the final stage of the study, the concentration of CH$_4$ still experienced a sharp increase. This indicates that there is still organic material that has not been wholly decomposed and produces CH$_4$. The concentration then decreased quite sharply and had a relatively the same concentration as the concentration of CH$_4$ in other reactors. At this stage, all organic material and nutrients have been converted into CH$_4$ and CO$_2$. CH$_4$ and CO$_2$ gas formation will also decrease significantly because most nutrients have been lost with the leachate during the previous stages. The residual material in the reactor will decompose slowly [1, 10, 11].

4. Conclusion

1. The leachate recirculation did not increase the CO$_2$ concentration significantly compared to the reactor without leachate recirculation treatment.
2. Leachate recirculation did not cause a significant increase in CH$_4$ concentration compared to the reactor without leachate recirculation treatment. In the reactor group with high leachate concentrations, reactors R2 – R10 which received leachate recirculation treatment, produced a more stable gas concentration than the reactor without recirculation (R1) more fluctuating CH$_4$ concentrations. The leachate recirculation did not cause a significant difference in pH value compared to the reactor without leachate recirculation, both in the reactor group with the addition of high concentration leachate and low concentration leachate.

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