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Activated carbon-mediated advanced oxidation process for effective leachate treatment

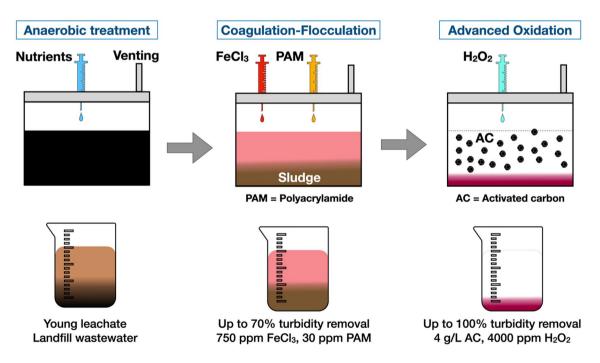
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Abstract

Leachate, a wastewater produced when rainwater percolates through landfill materials, is a global environmental concern due to its potential to contaminate groundwater and surface water. This study evaluated the use of aerobic-anaerobic, coagulation-flocculation, and advanced oxidation technologies to treat young landfill leachate collected in Chimbo, Ecuador. The treatment process effectively removed high turbidity, dark brown colour, and dissolved organic compounds. Anaerobic biodegradation reduced biochemical oxygen demand (BOD) and chemical oxygen demand (COD) by 60% and 67%, respectively, after 42 days. Coagulation with polyaluminium chloride and ferric chloride effectively removed suspended particles without adjusting the pH. The advanced oxidation process (AOP) using 4 g/L activated carbon and 4000 ppm H_2O_2 achieved 95% COD removal, 96% BOD removal, and 89% colour removal. This study suggests that AOP using activated carbon as a catalyst and H_2O_2 as an oxidant is a promising approach for high colour removal at a moderate cost.

Graphical abstract



Keywords Landfill leachate \cdot Anaerobic-aerobic biodegradation \cdot Coagulation-flocculation \cdot Activated carbon \cdot Hydroxyl \cdot Fenton-like process

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Introduction

Globally, around 2.01 billion tonnes of municipal solid waste are produced each year, of which at least 33% is not managed in a safe way and the remainder is disposed of in sanitary landfills (Kaza et al. 2021; Gautam and Agrawal 2021). By 2025, it is predicted that 4.3 billion urban residents will produce an average of 0.11-4.54 kg of municipal solid waste per day (Show et al. 2019; Nimita Jebaranjitham et al. 2022). In the European Union (EU), 505 kg of municipal garbage per capita were produced in 2020; however, the EU is moving toward recycling and other types of waste recovery to ensure a gradual reduction of waste in landfills (EU Directive (EU) 2022). Landfill mismanagement around the world has exacerbated the negative impacts of landfills, due to the emission of methane and carbon dioxide (Adelodun et al. 2021; Barlaz 2020). Landfill operations are linked to anthropogenic greenhouse gas emissions, groundwater contamination, bioaerosol formation, and wastewater leachate (Abiriga et al. 2020; Bian et al. 2021). Leachate is a liquid byproduct of decomposition processes that occurs in the solid waste management process (Staubitz et al. 2020; Ghanbari et al. 2020). It has a high concentration of organic substances such as humic acids, hydrocarbons, pharmaceuticals, hormones, pesticides, and microplastics, in addition to inorganic substances like heavy metals, the majority of which are toxic and refractory (Liu et al. 2015; Smaoui et al. 2018; Miao et al. 2019; Cirik and Gocer 2020; Cheng et al. 2021). For instance, some leachates have dissolved organic matter concentrations of up to 20,000 mg/L (Show et al. 2019; Wang et al. 2018). Besides, SARS-CoV-2 virus particles have already been found in wastewater and primary sewage sludge in several facilities, according to recent studies. It is yet uncertain if viruses may survive in waste leachate (Kitajima et al. 2020; Kweinor et al. 2020). There is limited information on the efficacy of SARS-CoV-2 concentration, extraction, and detection protocols in landfill effluent (Kitajima et al. 2020; Anand et al. 2022). Overall, releasing landfill wastewater leachate into the environment has a negative impact on aquatic life, including eutrophication, soil infertility, and human mutagenic effects (Wang et al. 2018; Asaithambi et al. 2020).

Leachates are classified as young (less than 5 years), intermediate (5–10 years), and old (more than 10 years) based on the age of the landfill (Tałałaj et al. 2021; Lindamulla et al. 2022). The BOD/COD ratio for young landfill leachates is generally greater than 0.5, whereas for old leachates, it is generally less than 0.1 (Show et al. 2019). Due to the young leachates' high biodegradability, traditional biological treatment (aerobic, anoxic, or anaerobic) is used (Teng et al. 2021). However, intermediate



and old leachates require chemical treatment due to their high recalcitrant content (Cirik and Gocer 2020; Xu et al. 2018). Considerably, the BOD/COD ratio decreases over time, making it more challenging to biologically treat older leachates. In this context, it is challenging to appropriately design a leachate treatment process because of the variance in content and strength. Physical-chemical, biological, and other treatments are typically used to treat landfill leachate. Coagulation-flocculation, absorption, oxidation, and membrane separation are some of the physical-chemical treatments (Bandala et al. 2021). Organics and nitrogen are removed by biological treatments since they are efficient in this regard (Miao et al. 2019). However, leachate heavy metals like lead, nickel, chromium, silver, cadmium, barium, and mercury may likewise inhibit biological processes (Wijekoon et al. 2022; Karimian et al. 2021; Baun and Christensen 2004). Adsorption, chemical precipitation, electrocoagulation, ultrasonication, and electrochemical advanced oxidation processes such electrochemical oxidation, electro-Fenton, and sono-electro-Fenton process have all been investigated at the lab scale with favourable results (Asaithambi et al. 2020; Aziz et al. 2007; Kurniawan and Lo 2009; Kundariya et al. 2021; Dereli et al. 2021).

Leachate wastewater can be modelled as a closed system in which solid contaminants are dissolved and diffused throughout the water body, increasing the entropy of the final solution. Because leachate wastewater is characterized by high solids content and a variety of persistent contaminants, a large amount of energy is required to purify the water and reduce its entropy by removing the pollutants (Tai and Goda 1985). In this sense, biological processes are constrained by the age and biodegradability of the leachate pollutants (Luo et al. 2020; Mojiri et al. 2020). As a result, physical-chemical methods are complimentary in reducing toxic and refractory compounds (Rohers et al. 2021; Tan et al. 2020). However, due to limited resources for wastewater treatment, most landfills in developing countries favour biological treatments over physical-chemical ones, which have higher operational costs (Caicedo-Concha et al. 2019; Nanda and Berruti 2021; Haslina et al. 2021).

This study proposes a novel approach for the treatment of landfill leachate using activated carbon as a catalyst in a Fenton-like reaction at neutral pH. Previous studies have shown that activated carbon can be used as a catalyst in Fenton-like reactions for the degradation of organic matter in wastewater (Fan et al. 2007). However, most previous studies have conducted this reaction under acidic conditions. Herein, the hypothesis is that the effectiveness of activated carbon as a catalyst in a Fenton-like reaction at neutral pH. The use of neutral pH conditions is a significant advantage, as it eliminates the need for acidification and neutralization steps, which can be costly



Fig. 1 Leachate sample from a municipal solid waste facility in Chimbo, Ecuador

and time-consuming. In addition, the synergistic effect between activated carbon and hydrogen peroxide in a Fenton-like reaction significantly improves the removal of pollutants from landfill leachate. The main contributions of this study are: (i) the biological removal of organic matter in young leachate wastewater, (ii) the effectiveness of two coagulant products to remove suspended solids at various dosages, and (iii) the findings of an advanced oxidation process (AOP) using hydrogen peroxide as the oxidant and activated carbon as the catalyst. Overall, this study approach for the treatment of landfill leachate in a cost-effective, efficient, and environmentally friendly way.

Materials and methods

Sample collection

A landfill in Chimbo (1°42'11.4"S 79°01'30.1"W), Ecuador's highland city (2448 m.a.s.l.) with a population of 15,000 people, was the source of young leachate wastewater (Fig. 1). Samples with a leachate age of approximately one year were collected from the primary retention pond. The laboratory received samples that were transported at 4 °C. The Standard Methods for the Examinations of Water and Wastewater were used to conduct all the tests for chemical oxygen demand (COD), biochemical oxygen demand (BOD), oxidation-reduction potential (ORP), turbidity, conductivity, total and volatile solids, colour, and pH (American Public Health Association 2017). Additionally, for AOP experiments, the removal of colour was measured using a smartphone RGB detector app, and colour units were expressed in HEX colour codes (Hasnul Hadi et al. 2021).

Leachates' level of pollution is often determined by the amount of rainfall and the types of waste present. The main characteristics of the leachate from Chimbo's landfill that was used in the studies are summarized in Table 1.

Table 1 Leachate wastewater quality

Parameter	Leachate
pH	8.0
COD (mg/L)	2720
BOD5 (mg/L)	2122
Turbidity (NTU)	>4000
Colour Pt-Co	>500
Oxidation-Reduction Potential (mV)	127
Conductivity (µS/cm)	600

Table 2 Parameter conditions for biological treatments

Item	Treatment	Condition
T1	Anaerobic	100-ppm Nutrient solution, no air
T2	Aerobic	No nutrient solution, air (2 ppm dissolved air)
Т3	Anaerobic	No nutrient solution, no air
T4	Aerobic	100-ppm Nutrient solution, air (2 ppm dissolved air)

Experiments

The study was carried out in three stages: (i) biological treatment, (ii) coagulation-flocculation, and (iii) advanced oxidation. In an enclosed, rectangular, plexy glass reactor holding 10 litters of leachate, batch tests were conducted. The ambient temperature was 25 °C.

Biological treatment

Anaerobic and aerobic treatments were tested in accordance with Table 2. Aerobic treatments (T2 and T4) received intense aeration (2 ppm dissolved air) for 42 days. Nutrient solution (100 ppm, 10% N, 10% P, 30% K) was added to anaerobic treatment T1 and aerobic treatment T4 for 2 days each. No activated sludge was used in either anaerobic or aerobic treatments. Supernatant from each treatment was



collected to measure total solids (TS), volatile suspended solids (VSS), and turbidity. Standard procedures were applied to measure total solids (TS) and volatile suspended solids (VSS) at 105 °C and 550 °C, respectively (American Public Health Association 2017).

Coagulation-flocculation

Two coagulants were tested independently in the coagulation process: polyaluminium chloride (PAC, $Al_2O_3 > 30\%$) and ferric chloride (Fengbai, China, 99% purity). Anionic high molecular weight polyacrylamide (Henan, China) was used as a flocculant at a constant concentration of 30 ppm. Separate additions of the coagulants were performed using a syringe at various doses ranging from 250 to 6000 ppm. No pH adjustment was performed. Instead of a conventional jar test, rapid mixing was done in a 10-L reactor using a recirculation pump for 10 min. After mixing, the entire volume was allowed to settle without stirring for 30 min. The supernatant was collected to measure changes in physical–chemical and biological properties. Turbidity values were used to calculate removal efficiency using Eq. (1).

$$Removal(\%) = \left[\left(T_o - T_f \right) / T_o \right] * 100 \tag{1}$$

where:

 T_o —initial turbidity of leachate wastewater.

 T_f —final turbidity of treated wastewater.

Advanced Oxidation Process (AOP)

A 6-L supernatant from the coagulation stage was used for the AOP. Hydrogen peroxide 50% (Merck, USA) was dosed at 4000, 8000, and 12,000 ppm as the oxidant, and 5 mm granular activated carbon (Merck, USA) was added at 4, 8, and 20 g/L as the catalyst. The oxidation treatment was run for a maximum of 4 h. To reduce operational costs, neither pH changes nor UV radiation effects were examined.

Data analysis

The observational data were analysed by descriptive and inferential statistics using R-project and R-studio with ggplot2 package (R Core Team 2022; Wickham 2016). The effects of aerobic or anaerobic process either with nutrients or no- nutrients (Factor A) and time (Factor B) on the final quality of water for SS, TS and turbidity (response variables), and their interaction, were studied using ANOVA. In the AOP, a Tukey's HSD test was performed to assess the effect of different dosages of GAC catalyst and H_2O_2 oxidant on pollutant removal.



Results and discussion

This study investigated the effects of aerobic and anaerobic biological treatments, with or without nutrients, followed by coagulation-flocculation for the removal of turbidity. Subsequently, activated carbon was evaluated as a catalyst and hydrogen peroxide was evaluated as an oxidant for the removal of residual color from previously treated samples. Chemicals were avoided throughout the treatment process to buffer the reaction medium or make additional pH adjustments, to reduce operational costs.

Effect of biological treatment

In both aerobic and anaerobic treatments, with or without nutrients, the removal of TS and VSS increased with time (Fig. 2A). The average reduction in TS and VSS across all treatments was 33% and 92%, respectively. Anaerobic treatment with N-P-K nutrient addition (T1) removed more TS than anaerobic treatment without nutrient addition (T3) after 42 days (39% vs. 33%). Besides, VSS removal was higher in T1 (94%) than in T3 (90%). Aerobic treatment without nutrients (T2) removed 32% of total solids (TS), while aerobic treatment with N-P-K nutrients (T4) removed 29% of TS. VSS removal was 89% in T2 and 94% in T4. Figure 2B shows the reduction of solids over a 42-day period, measured by the ratio of volatile suspended solids (VSS) to total solids (TS). The VSS/TS ratio was initially 0.76 for both aerobic and anaerobic treatments but decreased to 0.06 in the anaerobic treatment with N-P-K nutrients (T1) and 0.12 in the anaerobic treatment without nutrients (T3) after 42 days. In the aerobic treatments, the VSS/TS ratio was 0.12 in T2 without nutrients and 0.07 in T4 with N-P-K nutrients. Overall, this indicates that, on average, 94% of the organic matter (VSS) was stabilized. Figure 2C shows the reduction in turbidity for all biological treatments, as an indication of microbial activity for the degradation of organic compounds. Turbidity was reduced by 72% in anaerobic treatment T1 and 60% in anaerobic treatment T3. In aerobic treatments T2 and T4, turbidity was reduced by 79% and 60%, respectively.

Based on the results, anaerobic treatment with N-P-K nutrient addition (T1) was the most effective option, achieving the highest reductions in both TS and VSS, and the lowest VSS/TS ratio after 42 days.

The US Environmental Protection Agency (EPA) defines residual sludge as biostabilized or mature if the change in the organic fraction indicated by VSS relative to TS is less than 0.6. This quality indicator is used to control the quality of biostable products. Although our method used a liquid phase, we used this limit value to assess

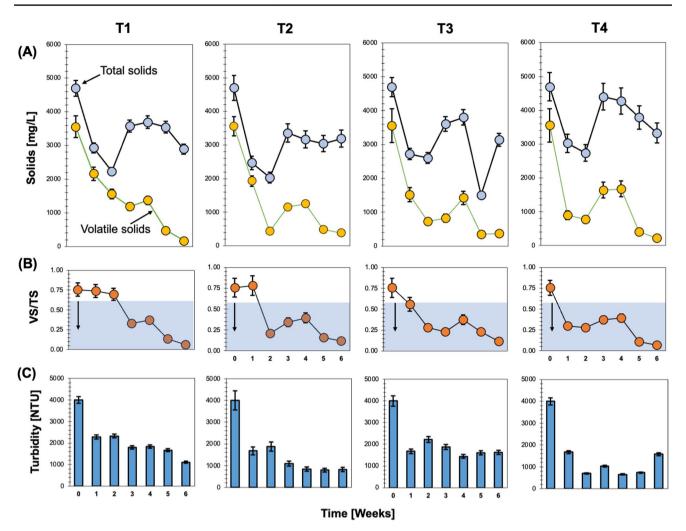


Fig. 2 Effects of aerobic and anaerobic treatments with or without N-P-K nutrients on the removal of (A) TS and VSS in mg/L; (B) VSS/TS ratio; and (C) Turbidity. Where T1 = Anaerobic treatment

the biodegradation criterion (Berenjkar et al. 2019). After 42 days, the VSS/TS ratio ranged from 0.06 to 0.12 for all treatments. Unpleasant odours and the original dark brown colour disappeared, and no abnormal changes in pH were observed. The fact that over 92% of volatile suspended solids were stabilized on average demonstrates that the reactors successfully removed organic components. This proves that microbial metabolism in both aerobic and anaerobic treatments effectively reduced the concentration of solids in young leachate.

ANOVA in Table 3 showed no significant difference (p > 0.01) in TS and VSS removal among aerobic and anaerobic treatments (Factor A). However, a significant response (p < 0.01) was observed in terms of biodegradation time, indicating that both aerobic and anaerobic treatments are effective in removing TS and VSS over time. VSS removal, which indicates the breakdown of organic matter,

with a 100-ppm nutrient solution; T2 = Aerobic treatment with no nutrient addition; T3 = Anaerobic treatment with no nutrient addition; T4 = Aerobic treatment with a 100-ppm nutrient solution

 Table 3
 Summary of the analysis of variance (ANOVA) for aerobic/ anaerobic treatment, biodegradation time according to VSS, TS, and turbidity

Source of variation	Df	Suspended solids	Total solids	Turbidity
Treatment	3	0.978 (ns)	0.511 (ns)	0.558 (ns)
Time	1	0.000 (***)	0.000 (***)	0.000 (***)
Treatment:Time	3	0.931 (ns)	0.897 (ns)	0.891 (ns)

Df Degrees of freedom. Significance codes: 0 (***) 0.001 (**) 0.01 (*) 0.05 (.) 0.1 (-) no significance (ns)

is not significantly affected by the presence or absence of nutrients, suggesting that wastewater typically contains sufficient nutrients for microbial growth. Both aerobic and anaerobic biological treatments were effective, but anaerobic treatments may be more cost-effective because they do not require aeration.



Anaerobic biological treatment is the most common for leachate, producing less sludge, treating young leachates, and generating methane, which is beneficial for the environment and the economy, but it requires a well-balanced macro- and microelement environment for cell metabolism (Smaoui et al. 2018; Liikanen et al. 2018; Nabi et al. 2022). Co-digestion with different substrates, including intermediate or mature leachate, enhances efficient anaerobic digestion (Dereli et al. 2021; Berenjkar et al. 2019; Aromolaran et al. 2022). For example, a study of sewage sludge co-digested with different amounts of mature landfill leachate showed that anaerobic co-digestion with high organic matter load and VS concentration is most efficient, as lower concentrations can significantly decrease COD, TS, and VS removal efficiency (Dereli et al. 2021; Berenjkar et al. 2019). Another drawback of biological processes is that high salt concentrations in leachates can harm wastewater microbes, reducing ammonia and nitrogen removal efficiency, and affect sludge floc structure, settling properties, oxygen solubility, and transfer (Wang et al. 2018; Deng et al. 2022). Our findings agree with other studies, showing that solids removal by biological means was not significantly affected by high leachate salinity (Tałałaj et al. 2021; Gao et al. 2022). According to our results, and those of other researchers, activated sludge for landfill leachates requires long aeration times of more than 24 h, which is expensive due to the high energy demand (Tyagi and Ojha 2023). Despite extensive research as a potential leachate treatment method, activated sludge is impractical for large-scale implementation due to extended retention times and considerable energy consumption. However, technologies such as Membrane and Sequencing Batch Bioreactors have a smaller footprint, higher treatment efficiency, and produce less sludge, while optimizing the process can reduce costs such as energy consumption by using more efficient aeration systems and optimizing process parameters.

Effect of coagulation

Figure 3 illustrates the effectiveness of PAC and FeCl₃ coagulant dosages while maintaining the flocculant dosage constant at 30 ppm. At 250 ppm, FeCl₂ reduced 50% of the turbidity, while PAC reduced approximately 38%. At 500 ppm, FeCl₃ was still 66% more effective at removing turbidity than PAC, which was just 52% effective. To achieve maximum particle removal, the coagulant dosage was doubled at this step. At a dosage of 750 ppm, both PAC and FeCl₃ reached essentially the same efficiency (69%). Both coagulants were efficient in removing turbidity between 84% and 97% up to 3000 ppm. With increasing doses, contaminant removal increased and eventually reached its maximum asymptote at 3000 ppm. At 3000 ppm, FeCl₃ reached its maximum efficiency for turbidity removal (97%). Beyond this level, FeCl₃ dosages caused a charge reversal and prevented colloid destabilization from proceeding. There was no requirement for pH modification because the initial pH was 7.0. In addition to turbidity removal, aluminium polychloride and ferric chloride also removed colour, assisting in the removal of both dissolved and fine suspended particles while maintaining the original pH. Both PAC and FeCl₃ successfully removed turbidity. Ferric salts, as opposed to aluminium, were shown to be more effective. The particle surface charge, which is considered to have been initially negative, was reversed, however, in response to a high dose of the coagulant. According to experimental results, coagulation had the ability to neutralize the electrostatic charge that was present in leachate colloids, which lessened the attraction between negatively charged particles by generating protons.

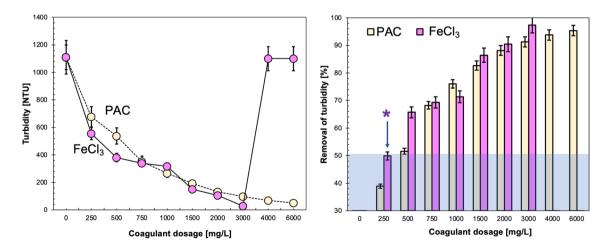


Fig. 3 Removal percentage of turbidity from municipal wastewater using PAC and FeCl_3 coagulants. Samples came from biological processes with a turbidity of 1110 NTU

According to reaction (I), an aluminium salt dissolves in water and losses three electrons from its final orbitals. Due to the aluminium atom's remaining 6 electrons in its electrical state, the hydration of the ion takes the form $Al(H_2O)_6^{+3}$. In reaction (II), the bond between the positively charged aluminium cation and an oxygen atom of one of the six water molecules in the $Al(H_2O)_6^{+3}$ results in a complex ion that behaves like a proton donor due to the increased polarity of the O–H bonds of the water molecule. As a result, hydrogen atoms are more likely to ionize. The solution becomes acidic because of the hydrolysis of the metal cation and proton generation. The same complex ionization occurs for ferric chloride as shown in (III) and (IV).

$$Al_{(s)}^{+3} + 6H_2O_{(l)} \rightarrow Al(H_2O)_{6}^{+3}{}_{(aq)} + 3e^-$$
 (2)

$$Al(H_2O)^{+3}_{6(aq)} + H_2O_{(l)} \rightleftharpoons Al(OH)(H_2O)^{+2}_{5(aq)} + H_3O^{+}_{(aq)}$$
 (3)

$$\operatorname{Fe}^{+3}_{(s)} + 5\operatorname{H}_2\operatorname{O}_{(l)} \to \operatorname{Fe}(\operatorname{H}_2\operatorname{O})_5^{+3}_{(aq)} + 3e^-$$
(4)

$$Fe(H_2O)_{5(aq)}^{+3} + H_2O_{(l)} \Leftrightarrow Fe(OH)(H_2O)_{4(aq)}^{+2} + H_3O_{(aq)}^{+}$$
(5)

The dosage of coagulants for leachate treatment depends on the leachate's hydrogen bonding system and colloidal electrostatic charge, which vary from city to city. Therefore, directly applying the conclusions of other studies is inappropriate. However, other studies have found that coagulant dosages for leachate coagulation range from 100 to 5000 mg/L of Me³⁺ ions (where Me³⁺ is either Al³⁺ or Fe³⁺) or even higher dosages, with optimal pH values of 4.0-8.0 and 3.0-9.0 for Al-based and Fe-based coagulants, respectively (Turan et al. 2023; Djeffal et al. 2021; Tousizadeh et al. 2022). A study of leachate from Jeram Sanitary Landfill (Kuala Selangor, Malaysia) found that alum was effective in removing 54% of the COD at a dosage of 750 mg/L and a pH of 8.5 (Cheng et al. 2021). Additionally, a dark brown leachate from a landfill in Ranchi, Jharkhand, India, with an initial COD of 4300 mg/L and total suspended solids of 4400 mg/L was effectively treated with 2800 mg/L of alum or 470 mg/L of ferric chloride as coagulants and polyacrylamide grafted gum ghatti as flocculant (Kumar et al. 2023). In the present study, FeCl₃ at a dosage of 3000 ppm effectively removed turbidity, with an efficiency of 97%. This demonstrates that $FeCl_3$ is a viable coagulant, making it a useful tool for leachate treatment. In general, several studies have been conducted

on leachate coagulation-flocculation, but little is known about the mechanisms that optimize and coagulate landfill leachate utilizing aluminium or iron as coagulants (Cheng et al. 2021; Aziz et al. 2007).

Effect of an advanced oxidation process

The supernatants with the lowest turbidity from coagulationflocculation using PAC and FeCl₃ were used as the feedstock for the following AOP. The coagulation-flocculation step was optimized to remove as much turbidity as possible, as turbidity can interfere with the effectiveness of AOPs. Different doses of a 50% H₂O₂ oxidant were used: 4000, 8000, and 12,000 ppm. Commercial granular activated carbon (GAC) was added as a catalyst at dosages of 4, 8, and 20 g/L (without any chemical modification). Each treatment was limited to a maximum of 4 h of sun exposure. The supernatant's original colour was light brown, its turbidity was 0 NTU, and its initial pH was 6.5. The effects of pH and UV light were not tested.

This study evaluated the effects of GAC and oxidant dosage on colour removal to assess the efficiency of the AOP. Figure 4 shows that the optimal colour removal was achieved in samples treated with 4, 8, and 20 g/L GAC and $4000 \text{ mg/L H}_2\text{O}_2$, using FeCl₃ in the coagulation step. These results align with previous research, which has shown that the presence of iron oxide in GAC significantly enhances the oxidative ability of H₂O₂, leading to more effective removal of humic acids, fulvic acids, and non-humic substances from leachate (Fan et al. 2007; Khalil et al. 2001). Increasing the dosages of H₂O₂ and GAC did not significantly improve colour removal, especially for samples that were treated with PAC in the coagulation step (Fig. 4). This in contrast information in which is reported that, increasing concentrations of H₂O₂ have a positive effect on removal efficiencies, as previously reported (Fan et al. 2007). In terms of colour removal, the optimal treatment in the current study was activated carbon at 4 g/L and 4000 ppm H₂O₂.

Besides the assessment of colour removal, Fig. 5 revealed the effects of AOP on final turbidity. According to results, the AOP was ineffective to clarify water that was previously treated with PAC. It is hypothesized that an environment containing Al^{+3} residues from the coagulation stage did not increase AOP at all; on the other hand, an increase in turbidity was observed in all treatments. Turbidity increased from 0 to a maximum of 389 NTU, as shown in Fig. 5. Since there was no colour removal in this instance, the objective was not accomplished when using PAC.

The 4 g/L dosage of activated carbon in all H_2O_2 concentrations had no effect on the turbidity from the supernatant

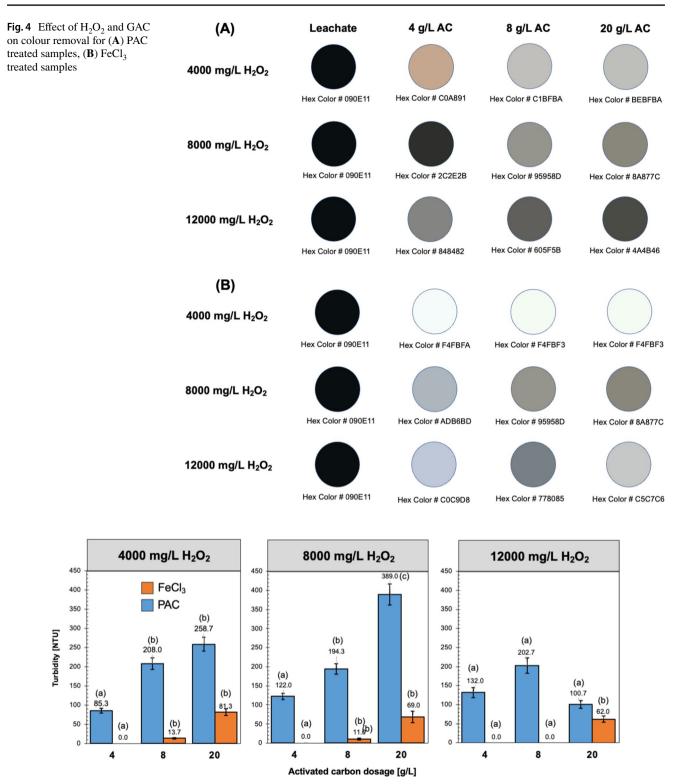


Fig. 5 Effects of AC and H_2O_2 dosages on the removal of turbidity. In the upper part of the bars a summary of turbidity means from Tukey HSD test is presented, in which identical letters per turbidity mean indicate non-significant differences at the significance level of 95%

that resulted from the FeCl₃ treatment. The removal of colour with 8 g/L activated carbon at 12,000 ppm H_2O_2 was successful without causing any turbidity to rise. However, turbidity increased, and colour was not removed when GAC was used at 8 g/L, and H_2O_2 at 4000, and 8000 ppm. Besides, all H_2O_2 dosages resulted in an increase in turbidity at 20 g/L activated carbon. Efficiency dropped as more H_2O_2 , and AC were supplied to the system.

As mentioned earlier, it is important to note that a high concentration of H_2O_2 does not always guarantee an enhancement in colour and turbidity removal, as evidenced by the Tukey HSD test in Fig. 4 and supported by existing literature (Asaithambi et al. 2020). In terms of residual turbidity, the best treatment in the current study was activated carbon at 4 g/L and 4000 ppm H_2O_2 . The degree of treatment needed to remove dissolved solids, including coloured particles, was not provided by coagulation-flocculation

operations. The results show that AOP significantly accelerated the degradation of emerging pollutants. When H_2O_2 and catalysts like Fe⁺³ and Fe⁺² ions are combined, the peroxide decomposes to produce hydroxyl radicals (•OH) (Wang et al. 2018). These radicals are highly reactive species that can aggressively degrade organic matter (Yilmaz et al. 2010; Safarzadeh-Amiri et al. 1996; Miklos et al. 2018; Ramirez Zamora et al. 2000). The use of AC as a catalyst in the current work was made possible by the material's high porosity, which would encourage the production of \bullet OH. A reaction mechanism using activated carbon, zeolite or any other mineral like granite as catalysts is not yet entirely researched (Banchón 2022; Banchón et al. 2022). However, it is herein suggested in reactions (V) to (VIII) that H_2O_2 decomposes catalytically by the presence of AC (Kurniawan and Lo 2009).

 Table 4
 Quality and operational parameters after leachate treatment technologies

Landfill site	Coagulation	AOP	References
Sanitary Landfill (Chimbo, Ecua- dor)	3000 ppm FeCl ₃ coagulation 30 ppm Anionic polyacrylamide Young leachate Turbidity removal = 97% COD removal = 80% BOD removal = 78% Colour removal = 53%	4 g/L GAC and 4000 ppm H_2O_2 Solar-UV COD removal = 95% BOD removal = 96% Colour removal = 89% Initial pH = 6.5	Present study
Sanitary Landfill (Guarapuava, Brazil)	Electrocoagulation (Al-electrodes) Colour removal = 55–83% COD removal = 40% Turbidity removal = 92% Current density = 128 A/m ² Initial pH = 7.9 Reaction time = 90 min	_	Galvão et al. 2020)
Municipal Landfill (Ahvaz, Iran)	Electrocoagulation (Fe/Al-elec- trodes) COD removal = 60% Ammonia removal = 43% Colour removal = 85% Initial pH = 6.4	Electro-oxidation (Stainless steel); peroxymonosulfate (PMS)/UV/ $CuFe_2O_4 AOP$ COD removal = 95.6% Ammonia removal = 99.8% Colour removal = 99.9%	Ghanbari et al. 2020)
Municipal Landfill (Lages, Brazil)	-	Fenton process Old leachate Colour reduction = 97.2% Initial pH = 3.1 $1.2 H_2O_2/FeSO_4$	Göde et al. 2019)
Municipal Landfill (Caldas, Colombia)	_	75 °C, 3 Bar Cavitation time = 30 min 200 ppm H_2O_2 COD removal = 30–53% Suspended solids removal = 99% Initial pH = 8.9	Gutiérrez-Mosquera et al. 2022)
Municipal Solid Waste Sanitary Landfill (Selangor, Malaysia)	-	Photoelectro-Fenton process COD removal = 97% Current density = 0.30 A/dm^2 pH = 3.0 H ₂ O ₂ = 300 mg/L Reaction time = 4 h	Asaithambi et al. 2020)



Table 5 Landfill leachate analysis before and after biological, and physical-chemical treatments

Parameter	WW	Bio	C-F	AOP
рН	8.0	7.0	6.5	6.5
COD (mg/L)	2720	1088	217	10
BOD ₅ (mg/L)	2122	707	152	6
Turbidity (NTU)	>4000	1110	0	0
Colour Pt-Co	> 500	350	283	30
ORP	127	180	130	238
Conductivity (µS/cm)	600	210	253.1	1.2

WW=Landfill leachate wastewater

Bio = Anaerobic treatment using N-P-K nutrients

C-F=Coagulation-flocculation treatment using 3000 ppm FeCl₂

AOP=Advanced oxidation process using 4 g/L AC and 4000 ppm H_2O_2

$$AC + H_2O_2 \rightarrow AC^+ + OH - + \bullet OH$$
(6)

$$AC^{+} + H_2O_2 \rightarrow AC + H^{+} + \bullet O_2H$$
(7)

Additionally, the residual Fe^{+3} ions from the coagulation step contribute to the Fenton-like reaction that produces •OH.

$$\mathrm{Fe}^{+3} + \mathrm{H}_2\mathrm{O}_2 + hv \to \mathrm{Fe}^{+2} + \mathrm{OH} + + \bullet \mathrm{O}_2\mathrm{H}$$
(8)

$$Fe^{+2} + H_2O_2 + hv \rightarrow Fe^{+3} + OH - + \bullet OH$$
(9)

Research has explored different approaches for achieving colour and COD removal from leachate, including lightbased (UV) procedures, electro-Fenton, and photo-electro-Fenton as detailed in Table 4. These approaches use UV radiation exposure and electrochemical reactions to remove pollutants (Asaithambi et al. 2020). Studies have reported that the highest removal efficiencies for colour, COD, and total dissolved organic carbon were achieved at 70 °C, pH 5, with 200 mg/L of zirconia-supported copper catalyst, and $30 \text{ mL/L of } H_2O_2 \text{ for } 150 \text{ min (Hussain et al. 2022).}$

Physico-chemical properties of the treated leachate

A high organic load and high biodegradability of the leachate are indicated by the ratio of BOD to COD, which has a mean value of 78% in Table 5. While a low BOD/COD ratio (between 20% and 40%) denotes the existence of lowbiodegradable recalcitrant compounds, a high BOD/COD

ratio (between 40% and 60%) defines a good biodegradability of wastewater (Cirik and Gocer 2020; Tchobanoglous et al. 2014). Heterotrophic microorganisms break down a significant percentage of biodegradable organic matter from young landfills into fatty acids. While BOD was reduced by 67% (2122 to 707 mg/L), COD was lowered by 60% (from 2720 to 1088 mg/L) during the anaerobic treatment employing nutrients (T1) (Table 5). According to some authors, anaerobic treatment may decrease COD by up to 74% and colour by up to 98% (Cirik and Gocer 2020). As a result, the anaerobic condition with nutrient addition (T1) was determined to be the optimal treatment in this work in terms of low energy consumption and operational expenses. A biological treatment alone is not a practical method to eliminate virus particles in leachate. However, it is still uncertain if viruses may survive in the environment, including sewage and landfill leachate (Kweinor et al. 2020; Anand et al. 2022). Coagulationflocculation is necessary to remove all turbidity, and accelerated oxidation to destroy pathogens and minimize virus particles is also important.

The coagulation treatment at 3000 ppm FeCl₃ decreased COD by 80% from 1088 to 217 mg/L and BOD by 78% from 707 to 152 mg/L. Furthermore, this treatment removed 53% of the initial colour. According to other investigations, a suspended particles reduction up to 95% was observed at 1509 ppm FeCl₃ at a pH of 7.02 (Moradi and Ghanbari 2014). The AOP treatment reduced 95% COD, 96% BOD, and 89% colour using activated carbon at 4 g/L and 4000 ppm H_2O_2 . Another study found that COD was removed by 82% applying 15 g/L of chemically modified granular AC and 4000 ppm H₂O₂ at pH 8.0 (Wijekoon et al. 2022; Kurniawan and Lo 2009).

Conclusion

This study determined the minimum physicochemical (Advanced oxidation, coagulation-flocculation) and biological (Aerobic-anaerobic) processes for degrading total and suspended volatile solids from a dark brown leachate from Chimbo, Ecuador. N-P-K nutrition addition under anaerobic conditions increased COD and BOD removal by 67%. PAC and FeCl₃ provided substantial solids removal without pH modification, while 3000 ppm FeCl₃ was the most effective coagulant for the following AOP, removing up to 97% COD, 78% BOD, and 53% color. In the



oxidation step, high H_2O_2 concentration does not necessarily improve color and turbidity removal, although 4000 ppm H_2O_2 with 4 g/L activated carbon reduced COD, BOD, and colour by 95%, 96%, and 89%. Peroxide decomposes by producing extremely reactive hydroxyl radicals, hence activated carbon performed optimally as a catalyst with H_2O_2 and residual Fe⁺³ ions. This study shows that activated carbon is capable of being used as a catalyst for large-scale applications in a wide dose range, however its reaction mechanism and reusability are still being studied. This research focused on the minimal requirements for leachate clarification using activated carbon and hydrogen peroxide in a Fenton-like reaction for high organic matter concentrations in landfill leachate to reduce operational costs for large-scale treatment plants.

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Declarations

Conflicts of interest The authors declare that they have no conflict of interest.

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