

A comparative study of landfill leachate treatment using advanced oxidation photochemical processes, ozonation process and hydrogen peroxide systems

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ABSTRACT

Ozonation processes and hydrogen peroxide systems can be used as advanced oxidation photochemical processes (AOPs) to treat leachate from municipal landfills. They are efficient and effective processes in the extermination of microorganisms and elimination of viruses and pathogens, including members of the virus family coronaviridae (such as MERS-CoV, SARS-CoV-1 and SARS-CoV-2), the new coronavirus COVID-19, that all countries of the world suddenly faced recently, and the resulting emergence of many cases and injuries that exceed the absorption of hospitals, quarantine, and home self-isolation, and the abundant use of personal protective equipment, like face masks, gloves, contaminated cotton, dressings, and plaster, etc., whether in the case of infection or for prevention, and mixing of this type of waste with household waste, which increase the negative environmental impact because of the highly contaminated waste generation. This study aims to determine the most effective and safest methods of treating the leachate at optimum conditions of each process in laboratory-scale experiments at pH values (7.5–8.5). It was found that both O_3 and O_3/H_2O_2 effectively reduced the concentration of organic compounds under optimal experimental conditions. O_3/H_2O_2 was very effective in reducing the concentration of organic compounds in optimal experimental conditions. The most effective process, with the best results were obtained at 20 mg/L of H_2O_2 after increasing the treatment time to 60 min, where the efficiency of chemical oxygen demand (COD) removal was 84%–92%, the efficiency of total organic carbon removal was in the range of 29.21–58.42 mg/L. The biodegradation indicated by the biochemical oxygen demand (BOD_5)/COD ratio increased from 0.17 to 0.74, and the turbidity removal efficiency was 75.60%–82.80%.

Keywords: Ozonation; Hydrogen peroxide systems; Advanced oxidation photochemical processes (AOPs); Coronavirus; COVID-19; Treatment; Landfills; Leachate

1. Introduction

The high populations in countries with hot climates, especially those in the Arabian Gulf, Africa and the Middle East, may be characterised by high food consumption, alongside an increasing quantity of solid waste generated by rapid urban growth [1]. This growth leads to an increase in the amount of material disposed in landfills [2], and recently, a mixing of household waste with other residues potentially

contaminated by viruses [3] due to the abundant use of masks and gloves, whether in the case of infection or for the isolation. The new COVID-19 virus disease (COVID-19 or SARS-CoV-2), first appeared in Wuhan, China, in December 2019 [4]. The International Committee on Taxonomy of Viruses on 11 February 2020 officially declared the emerging coronavirus disease [5], COVID-19, a severe acute respiratory syndrome (SARS-CoV-2). This caused a pandemic according to the classification of the World Health Organization

(WHO) [6], on 11 March 2020, in many countries of the world representing 93% of the world's population (about 7.2 billion people), with 110 countries worldwide reporting 118,319 confirmed cases, [7]. This led to new challenges and the spread of a global crisis unprecedented in modern history in all parts of the world for both developed and developing countries alike, which represented a critical test of the world's public and private health care systems. In April 2020, the WHO issued directives calling on world governments to impose precautionary measures to mitigate the transmission of infectious diseases and limit the spread of the COVID-19 virus. All countries undertook maximum efforts to contain the epidemic and reduce infections in an attempt to restore normal life such methods included encouraging people to stay at home and social distancing campaigns. There were also movement restrictions on citizens, closure of schools, some workplaces and areas of human gatherings [8]. International and domestic navigation and aviation were much reduced without adopting some of the basic services countries provide to citizens and residents [9]. Countries performed their efforts in many indispensable ways, such as waste collection and water treatment [10], which contribute to the transmission and spread of infectious diseases [11]. These latter activities severely affected many aspects of human life [12], which resulted in many cases and injuries exceeding hospitals' capacity [13]. The spread of domestic infections, and the mixing of this type of hazardous and infectious waste containing significant amounts of infectious viruses with household waste [14], led to the subject of these wastes being subjected to several simultaneous physical, chemical and biological treatment changes [15] that produce a harmful liquid called spraying [16]. This liquid consists of a very concentrated mixture of organic, inorganic and viral pollutants [17], with low biodegradability [18], ammonia-nitrogen, inorganic salts, and heavy metals [19]. The leachate quantity and characteristics depend on the type of wastes disposed of and the age of the used landfill [20].

Advanced oxidation photochemical processes (AOPs) involve the production of free hydroxyl radical ($\cdot\text{OH}$) [21], which is produced from single oxidants such as ozone (O_3) [22], or a combination of strong oxidants such as (O_3) and hydrogen peroxide (H_2O_2), under optimum pH [23]. The initiation of ozone decomposition can be accelerated by increasing the pH [24] or adding hydrogen peroxide [25]. The organic pollutants are oxidised to produce biodegradable intermediates, carbon dioxide and inorganic salts [26].

The addition of hydrogen peroxide with ozone ($\text{O}_3/\text{H}_2\text{O}_2$) is one of the most effective treatment processes [27] due to the increased effectiveness and multiplication of oxidative power through the production of a hydroxyl (OH), a highly interactive process [28]. OH^- and H_2O_2 initiate a series of decomposition reactions that promote ozone degradation and result in $\cdot\text{OH}$, in the O_3/OH^- system [29]. The hydroxide ion reacts with ozone to produce a superoxide anion (O_2^-), which participates in a series of chain reactions that result in $\cdot\text{OH}$ [30]. Thus, ozone-based advanced oxidation processes are attractive to oxidise the complex leachate mixtures [31], which react rapidly to initiate a radical chain mechanism that leads to hydroxyl radicals [32].

Ozonation and the addition of hydrogen peroxide ($\text{O}_3/\text{H}_2\text{O}_2$) processes are effective for waste treatment owing to the high oxidative power and greater production of reactive hydroxyl radical species ($\cdot\text{OH}$) [33]. The OH^- and H_2O_2 initiate a series of radical reactions that enhance ozone decomposition to yield $\cdot\text{OH}$. In the O_3/OH^- system [34], the hydroxide ion reacts with ozone to yield superoxide anion radicals (O_2^-) [35], which in their turn are involved in a series of reactions that yield $\cdot\text{OH}$ [36]. Overall, 1 mol of O_3 yields 1 mol of $\cdot\text{OH}$. Hydroxyl radicals are reactive, non-selective oxidants and are the most important species in an advanced oxidation process [37]. In the $\text{O}_3/\text{H}_2\text{O}_2$ system, when H_2O_2 is dissolved in water [38], it partially dissociates into a hydro-peroxide ion (HO_2^- , the conjugate base of hydrogen peroxide) [39].

Advantages of AOPs include: (i) rapid disposal of organic and inorganic compounds and reduction of its proportion with the greatest efficiency [40]; (ii) mineralisation of organic pollutants into carbon dioxide, water and mineral salts [41]; (iii) high reaction and non-selectivity of hydroxyl radicals towards contaminants [42]; (iv) production of non-toxic by-reaction products without the generation of secondary pollutants [43]; (v) these systems can also be used as treatment or pretreatment of wastewater [44], pharmaceutical wastewaters, surfactant wastewater [45], and mine wastewaters. In addition, AOPs can also be used to remove heavy metals [46], and have been successful in treating wastewater from textile processing [47], municipal wastewaters, oil refineries, pulp and paper production [48], dairy, and landfill leachate [49].

In Egypt, the quantity of solid waste is increasing due to the rapid demographic and urban growth that the country is experiencing [50]. This has been brought about in part by the migration of residents from adjacent Arab countries and the construction of many new towns [51]. The first case of COVID-19 between Egyptian nationals was reported in March 2020. After that, the number of confirmed cases per day increased rapidly from 7 March to 23 April. As a result, the Ministry of Health announced a state of severe emergency and curfew and advised citizens to take all precautionary measures in accordance with WHO instructions [52]. This resulted in the spread of cases of domestic insulation, contact with patients and the spread of domestic infections,

Table 1
Characteristics of raw and settled (4 h) leachate

| Parameter | Raw | Settled |
|-------------------------------|-------------|-------------|
| pH | 7.6–7.85 | 8.15–8.48 |
| Colour (Pt-Co) | 2,173–2,572 | 1,998–2,194 |
| Turbidity (NTU) | 312–319 | 218–276 |
| BOD ₅ (mg/L) | 175–198 | 170–194 |
| COD (mg/L) | 1,197–2,370 | 1,210–2,440 |
| Total solids (mg/L) | 6,397–6,616 | 6,337–6,583 |
| Total suspended solids (mg/L) | 172–196 | 123–172 |
| Total dissolved solids (mg/L) | 6,225–6,420 | 6,214–6,411 |
| Total phosphorus (mg/L) | 147–168 | 143–164 |
| Ammonia-nitrogen (mg/L) | 724–972 | 652–893 |

and the mixing of this type of hazardous and infectious waste containing vast amounts of infectious viruses with household wastes [53]. The adverse effects of COVID-19 extended to the solid waste sector and resulted in an increase in the amount of medical solid waste generated each day from 70 to 300 ton/d, as well as the amount of municipal solid waste generated daily [54]. The disposal of municipal solid waste (MSW) directly without treatment from its dangerous contents of hazardous contaminants components and mixed random which generate a significant massive, dangerous leachate with harmful effects on public health [55], the surrounding environment, the soil properties such as physicochemical, biological, and groundwater pollution [56].

The World Bank report for 2018 stated that the amount of solid waste produced worldwide was more than two billion tons annually. However, about 40% of this total is disposed of incorrectly through open random landfills or other unsustainable and environmentally incompatible ways, which can play a negative impact on the environment and public health; the COVID-19 pandemic has significantly impacted these poor waste management ecosystems, so many countries need to understand how COVID-19 has impacted their solid waste management efforts and systems, global solid waste management to 2050 as shown in Fig. 1a.

El Waffa Wa El Amal landfill is the largest landfill site in Cairo serving the Greater Cairo area, which is used mainly for disposal of domestic solid wastes Generated from an estimated Cairo Governorate population of 9,840,591 people according to the estimates of the Central Agency for Public Mobilization and Statistics 2019, site area nearly 350 fed, and contains accumulations of solid waste of which 79% are organic materials over a period of more than 20 y by about 13 million tons and a height of up to 25 m, regulations would not allow direct discharge of the leachates into the sewer system; therefore different ozone doses based AOPs were used. The efficiency of the selected methods depends on many factors, including the presence of inorganic and organic substances in the leachate, pH, reaction time and oxidants concentration. Therefore, studies were carried out to determine the optimum conditions of each process and its effects on biodegradability. These data might also lead to a better understanding of the chemical oxidation by different ozone doses based on AOPs and different hydrogen peroxide concentrations and contact time during treatment processes to ensure compliance and safe discharge.

2. Materials and methods

The stable filtrate leachate samples were taken and collected from the landfill site in 20-L tanks and transferred to the laboratory, where they were cooled and maintained at a temperature of 4°C. Tests were performed on 20 L of these samples for 4 h and were used in the study to reduce biological and chemical reactions. The samples consisted of (pre-coagulation) which separated sediments and other precipitated materials after a decrease in the pH as an important initial step in these tests, which carried out in a 1.5-L cylindrical reactor with ozone gas supplied continuously. Ozone was generated from atmospheric air oxygen at an oxygen flow rate of 4 L/min and converted to ozone using an ozone generator and then to an ozone conductor (1,500 mL),

which was filled with 1,000 mL of the filtrate to achieve a homogeneous state of the mixture for an increasing test period to 60 min with sampling ± 30 mL every 10 min with measurement of pH, total organic carbon (TOC), turbidity, biochemical oxygen demand (BOD₅), and chemical oxygen demand (COD), measurements were performed using standard filtrate characterisation methods, Fig. 3. Schematic diagram of equipment and experiments procedures.

The leachate samples concentrations were adjusted and diluted using Eq. (1):

$$C_i V_i = C_t V_t \quad (1)$$

where C_i is the initial concentration of CO, BOD₅ before it was diluted, C_t is the final COD concentration after dilution, V_i is the volume to be diluted, and V_t represents the sample volume after dilution, where COD, BOD₅, colour, and pH were measured at the beginning of the experiment's procedure and at times $t = 10, 20, 30, 40, 50, 60$ min according to the Standard Methods for the Examination of Water and Wastewater.

The removal efficiency of COD, colour, and pH was obtained using the following equation:

$$\text{Removal}(\%) = \frac{(X_i - X_t)}{X_i} \times 100 \quad (2)$$

where X_i = the initial COD, colour and pH concentrations, respectively and X_t = the COD, colour and PH concentrations at times $t = 10, 20, 30, 40, 50, 60$ min, respectively, according to DIN 38409a Behr COD workstation used to measure to leachate.

According to the recommendations of the US Food and Drug Administration (FDA), the adoption of a multiple analysis test called polymerase chain reaction (PCR) to diagnose the presence of the COVID-19 virus. This test, also called a molecular test, detects the genetic material of the COVID-19 virus using a laboratory technique called a polymerase chain reaction, where a specialist collects a liquid sample and sends it to the laboratory.

The results may be available within one or several days if it is sent to an external laboratory or if there are delays in some laboratories.

3. Results and discussion

Oxidation with only ozone may not reach the required effectiveness due to the complex components of the release, and sometimes there may be a need to increase the ozone dose to a high percentage value, and the resulting reactions may take a long time.

H₂O₂ can initiate a series of radical reactions that enhance ozone decomposition to yield $\cdot\text{OH}$, through the addition of H₂O₂. While the addition of H₂O₂ promotes and stimulates the initiation of a series of radical reactions that promote and accelerate ozone degradation to produce $\cdot\text{OH}$ radicals– in addition to its degradation and production of $\cdot\text{OH}$ radicals– it depends heavily on the pH, which plays a critical role in ozone degradation. The direct and rapid action of ozone in eliminating organic pollutants, viruses or

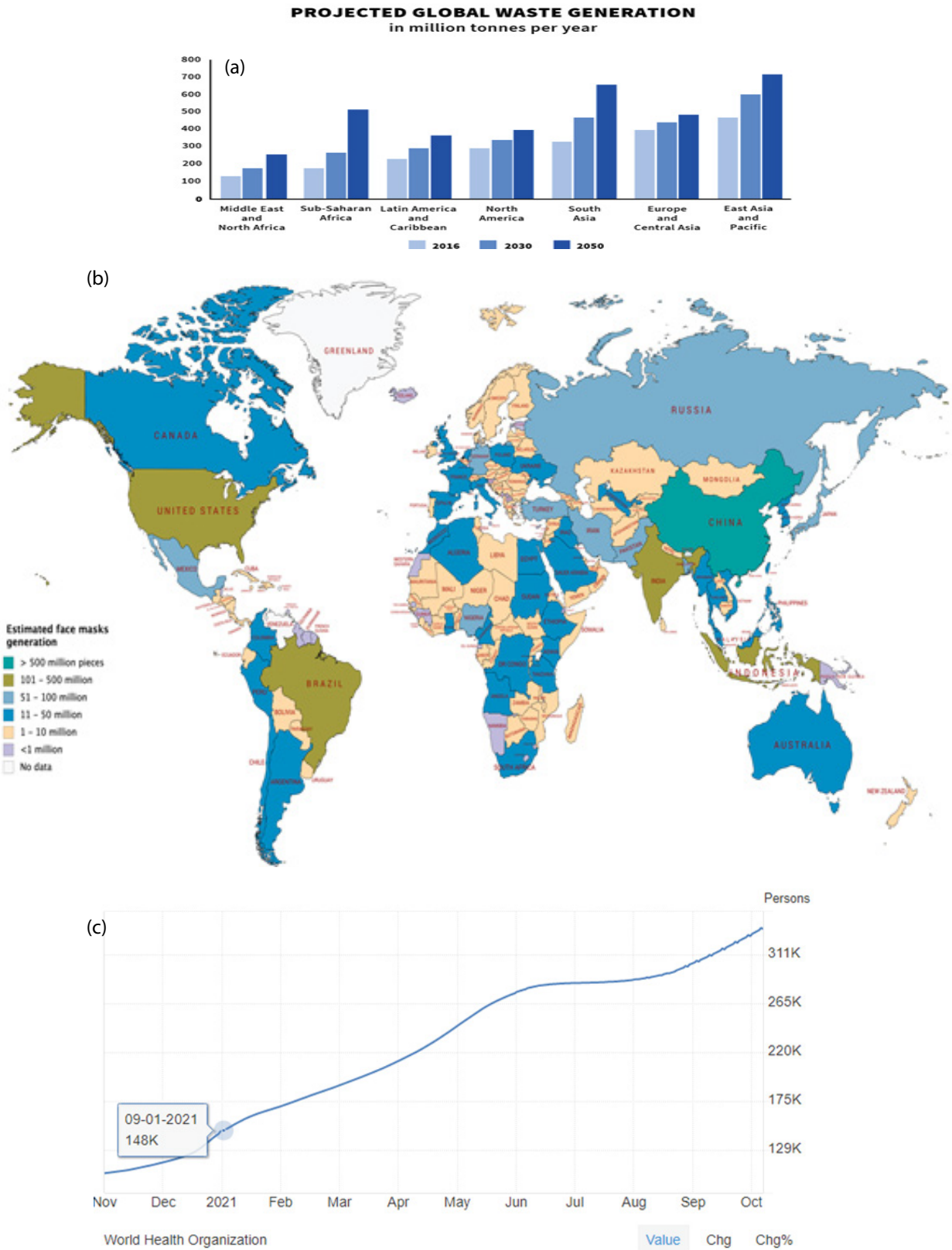


Fig. 1. (a) Snapshot of global solid waste management to 2050. Data: Source The World Bank. (b) Estimated face mask generations, Source The World Bank. (c) Trading Economics, 2020. Egypt coronavirus cases. Available at: <https://trading economics.com/egypt/coronavirus-cases> (Accessed 7 November 2021).

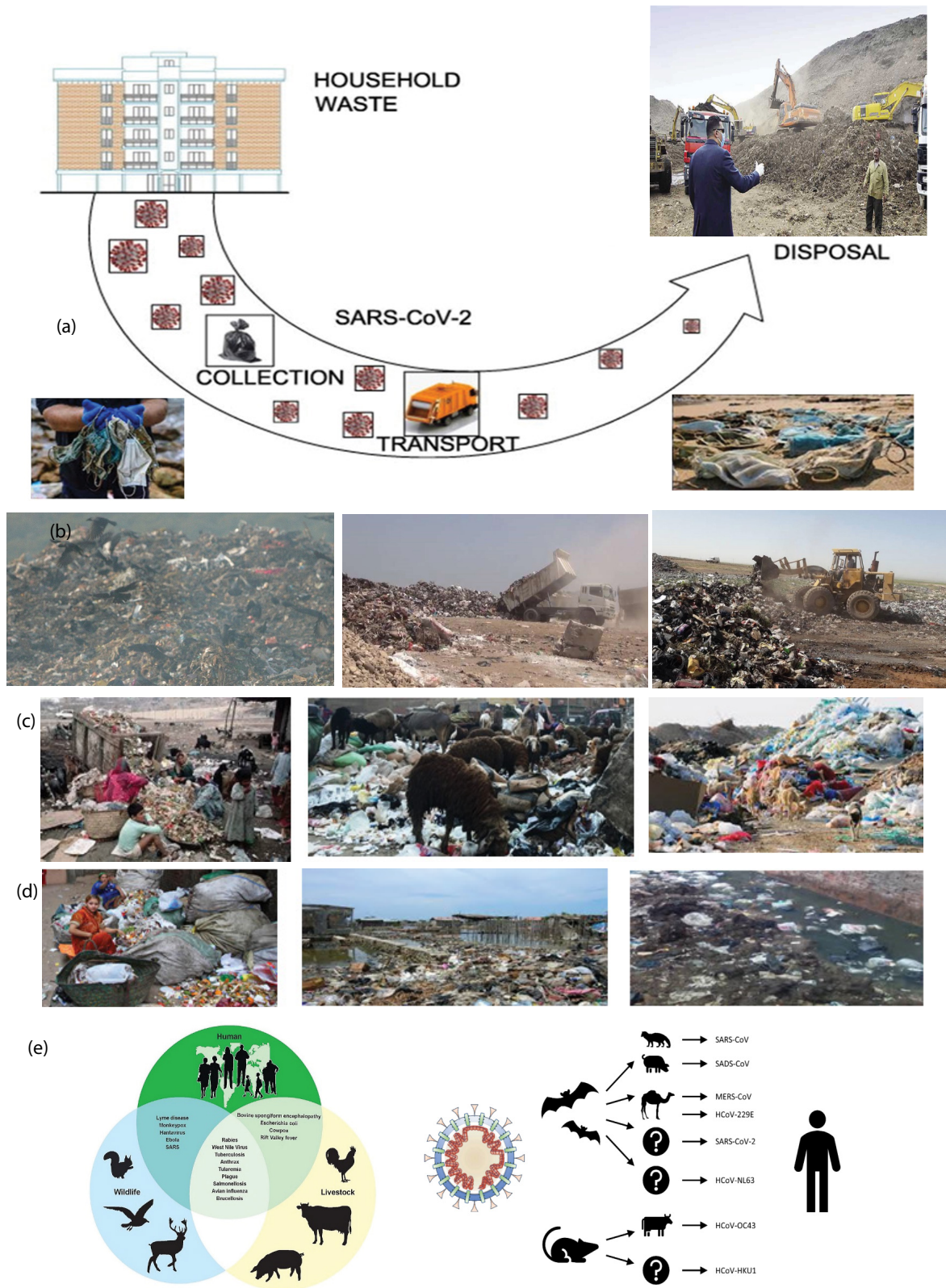


Fig. 2. (a–d) Examples of zoonotic diseases and their affected population, pollution due to leachate. (e) Examples of zoonotic diseases and their affected living creatures, animals, birds and population. *Source:* GAO analysis of USGS data.

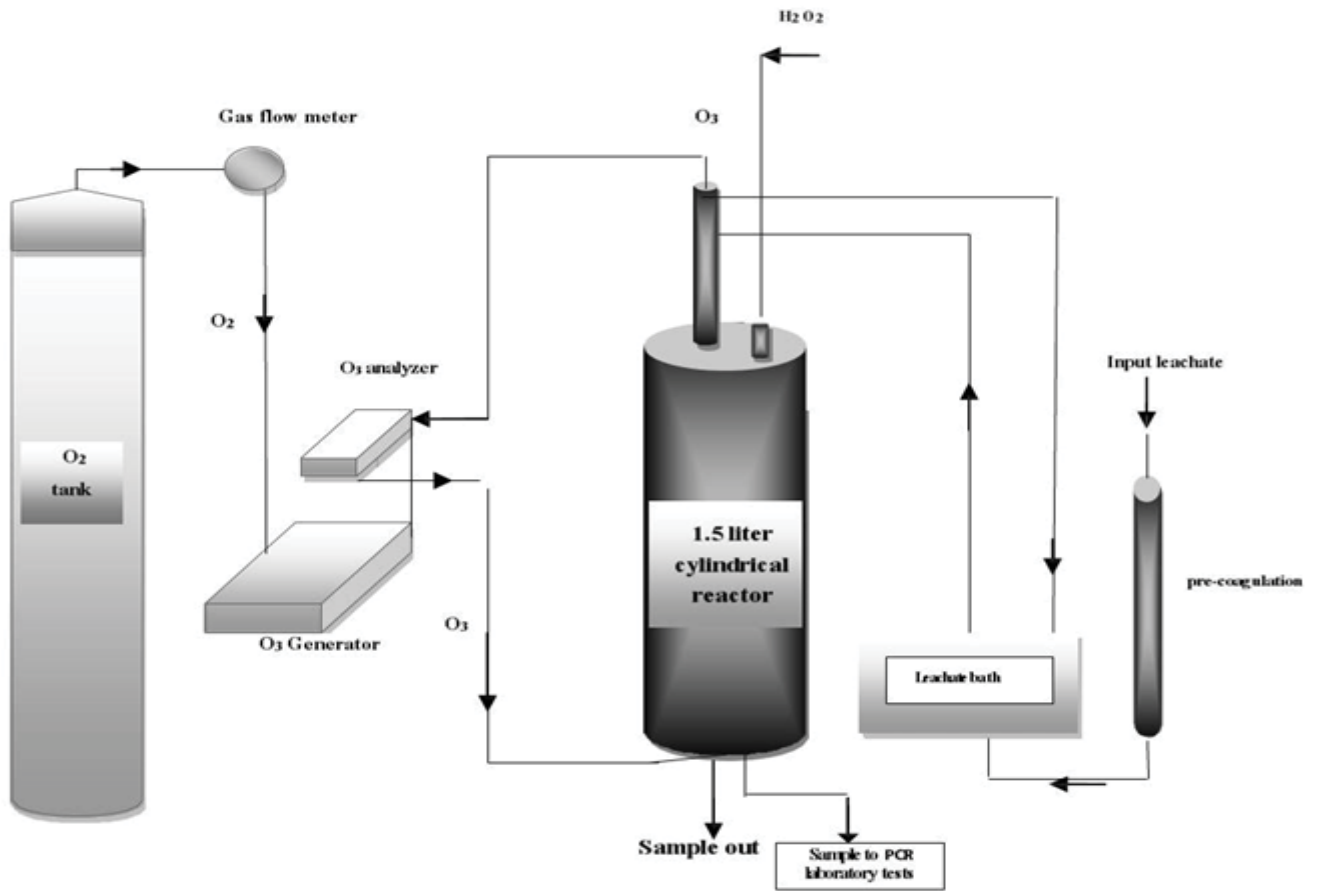


Fig. 3. Schematic diagram of equipment and experimental procedure.

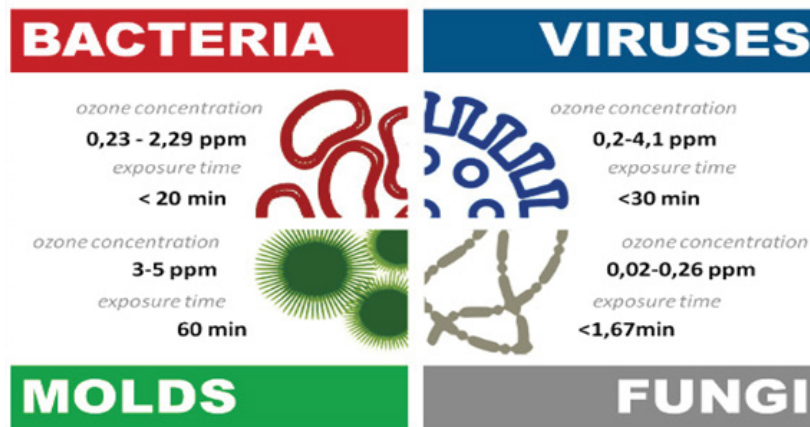


Fig. 4. Ozone effect. Source: GAO analysis of USGS data.

organic matter occurs by dismantling the fatty wall, dismantling the DNA or RNA and direct union with it through a direct electrophilic attack of the ozone molecule.

The effectiveness of ozone (O_3 / O_3/H_2O_2) in the elimination of bacteria, viruses and fungi, and in many protozoa species, including SARS and COVID-19, has been ascertained by experiments upon families of viruses,

including non-enveloped viruses called “naked” viruses (Adenoviridae, Picornaviridae, namely poliovirus, Coxsackie, Echovirus, Rhinovirus, hepatitis A and E, and Reoviridae Rotavirus). These consist of a DNA nucleus (made of DNA or RNA) and a DNA coat, or capsid, made of Picornaviridae protein. Destruction occurs by disrupting the external protein cortex by oxidation. When ozone comes

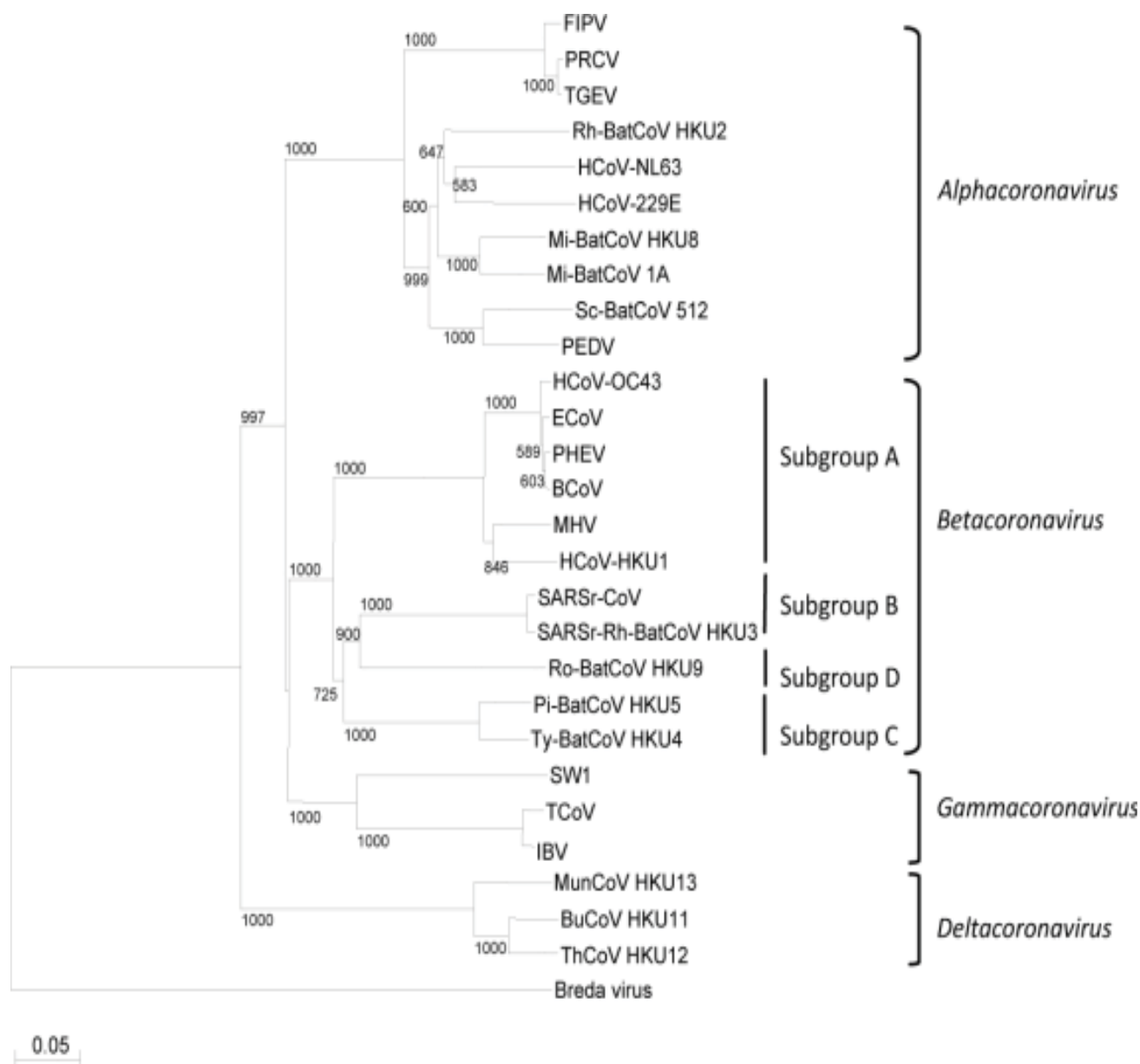


Fig. 5. Phylogenetic tree of coronavirus. Source: GAO analysis of USGS data.

in contact with capsid proteins, protein hydroxyatand interaction with protein is formed in DNA, leading to damage to viral RNA, which, once the fat envelope is fragmented from the virus, DNA or basic RNA cannot survive, since viruses, unlike bacteria, only multiply within the host cell.

Fig. 6 shows the evolution of pH as a function of reaction time from 0 to 60 min with ozonation by O_3 alone and by ozonation combined with hydrogen peroxide O_3/H_2O_2 at various H_2O_2 concentrations (5, 10, 15, and 20 mg/L). In the cases of O_3/H_2O_2 , a decrease in pH followed by a rise is noticed. With O_3 , a much greater pH decrease is seen, followed by a rise with average pH values of 7.79–8.80. This is contrary to what is expected of ozonation and its effect on pH, where it is understood to reduce lower pH and reach acid values, attributed to the formation of carboxylic

acids. However, this was not noticed in the oxidation of organic materials from construction and demolition waste, which is destined to increase in the future. The limited decrease in pH may be due to hydrogen peroxide because it is a weak acid ($H_2O_2 \rightleftharpoons HO_2^- + H^+$). As pH values refer to H_2O_2 activity, its effectiveness in ozone degradation and the production of many ions (OH^-) gives a high positive effect in the electrolyte currencies of all forms of organic matter. Furthermore, it gives a positive effect on the rate of removal of COD according to the following interactions:





When H_2O_2 is added to O_3 , in addition to the production of hydroxyl ions, the process of ozone degradation is accelerated, depending on reactions (11–13):



Fig. 7 shows the COD removal efficiency with ozonation by O_3 alone and ozonation combined with hydrogen peroxide $\text{O}_3/\text{H}_2\text{O}_2$ with various concentrations of H_2O_2 over different reaction times (0–60 min). The figure shows how to eliminate organic compounds, especially non-midwives, for biodegradation, where it is clear that the increase in decomposition of organic matter and the increase in the rate of removal of COD is high at the beginning of the experiment due to the availability of oxidised compounds and the significant increase in the level of hydrogen peroxide OH^* . This gives rise to a high rate of increase in COD removal with an increased concentration of hydrogen peroxide through free radical reactions. Ozone combined with hydrogen peroxide removed COD by 60.23%–92%, higher than ozone

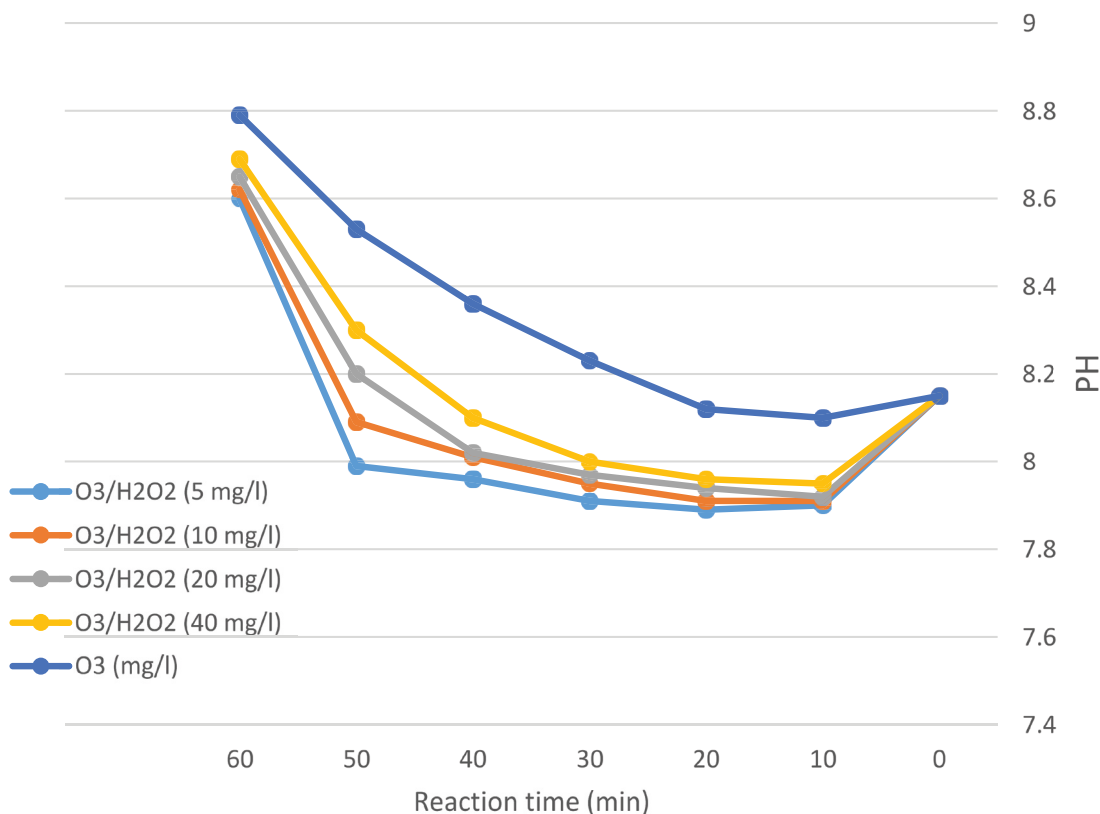


Fig. 6. pH change as a function of reaction time.

alone with a COD removal efficiency of 46.12%–84%. The maximum removal efficiency of COD, 92%, was obtained by ozone combined with hydrogen peroxide at 40 mg/L, but by ozone alone the efficiency was 84%, at an ozone dosage of 2 g O₃/L. The decrease in COD concentration is caused by non-biodegradable organic pollutants, oxidised either directly with ozone (selectively) or indirectly by OH[•] (non-selectively) and converted into an inorganic product such as CO₂.

Fig. 8 shows that the COD decreased gradually during the initial ozonation with O₃ alone and with ozonation combined with hydrogen peroxide O₃/H₂O₂ at various concentrations of H₂O₂. The increased concentration of H₂O₂ leads to more ozone degradation and more OH ions, which in turn processes organic matter and promotes biodegradable processes due to the availability of OH radicals. Furthermore, an increase in COD concentration was observed at 60 min, indicating the formation of organic

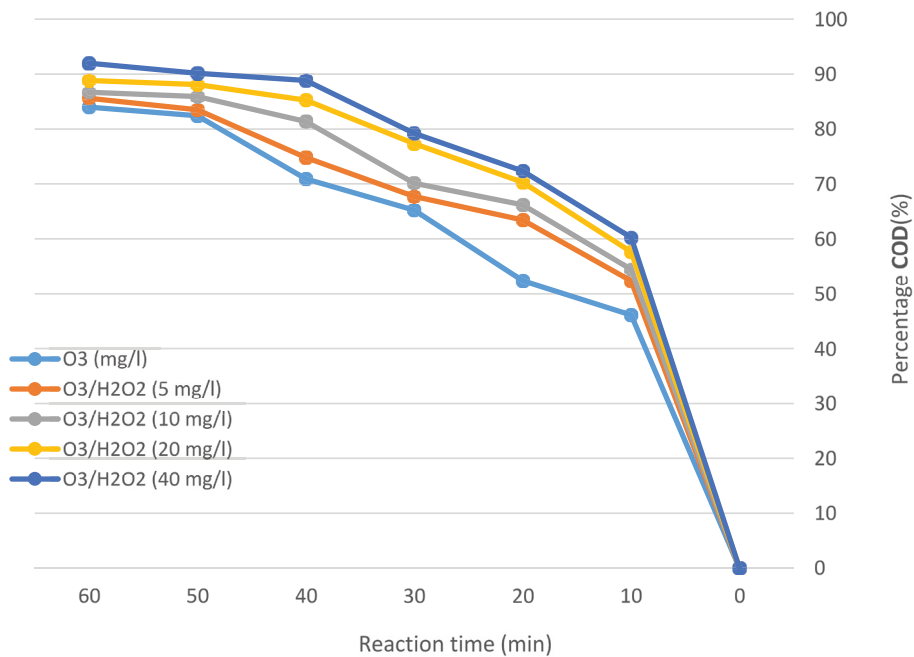


Fig. 7. COD removal efficiency by time.

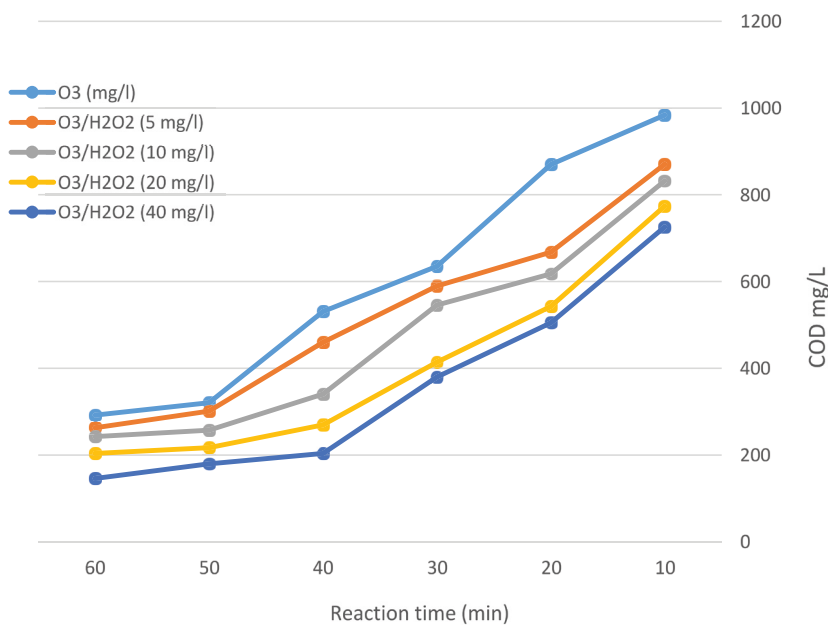


Fig. 8. COD mg/L variation with time.

acids. An improvement in COD removal from 1,825.67–1,533.56 mg/L and a COD value of 292.11 mg/L was found using O_3 treatment for 60 min. The maximum COD removal was 1,825.67–1,679.62 mg/L, and the COD value was 146.05 mg/L when using O_3/H_2O_2 at a dose of 40 mg/L. The presence of H_2O_2 resulted in increased COD removal, which in turn mineralised the organic compounds.

Fig. 9 shows results for the effect of ozonation by O_3 and ozonation combined with hydrogen peroxide O_3/H_2O_2 with various concentrations of H_2O_2 . We found that biodegradability of the leachate after adding O_3/H_2O_2 at a hydrogen peroxide dose of 40 mg/L enhanced biodegradability from 0.1 to 0.73 and reduced COD by 92%. The ozonation process alone enhanced biodegradability from 0.1 to 0.57 and reduced COD by about 84%. At an ozone dose of 2 g/L biodegradability increased due to an increase in BOD_5 concentration and a decrease in the concentration of COD down to the value of that gave a biodegradability value of 0.73. At pH 7 and the highest score rate in the degree of 8.4 pH, due to the selective nature of ozonation by O_3 . In the interaction with and oxidation of organic matter, the rate of biodegradability decreases due to the low amount of oxidised substances available due to the presence of hydroxyl ions. There is low biodegradability ($BOD_5/COD = 0.1$), which shows that spraying still contains many complex and diverse compounds that cannot be analysed and disposed.

Fig. 10 shows that turbidity removal efficiency with ozonation by O_3 alone and ozonation combined with hydrogen peroxide O_3/H_2O_2 with various concentrations of H_2O_2 (5, 10, 20, and 40 mg/L) during different reaction times (0, 10, 20, 30, 40, 50 and 60 min). The curves present two kinetic periods: the first period, (from time zero up to about 11 min). At this stage, the fast kinetics reaction is rapid, and the quick removal of the turbidity occurs due to

higher hydroxyl radical production in the O_3/H_2O_2 process. The second period, from around 11 min up to 60 min, is because ozone reacts very quickly with primary and organic compounds but slowly reacts with acids and aldehydes, which may be formed as a result of the interaction of ozone with aromatic compounds, phenols, aromatic acids and the residues of organic detergents, which may be found in the spray. This may cause the formation of aromatic acids and aldehydes, which in turn slow the reaction. The maximum removal efficiency of turbidity obtained by ozone alone was 75.60%, but ozone combined with hydrogen peroxide at 40 mg/L was 82.80%, demonstrating that H_2O_2 increased the turbidity removal rate by almost as much as O_3/H_2O_2 as compared to ozone alone.

4. Conclusions

Treatment of landfill leachates using O_3/H_2O_2 was more effective than using ozonation by O_3 alone. Within the current or future circumstances of the spread of viral infections and the resulting multiplication, enveloped viruses are usually more sensitive to physicochemical challenges than naked viruses. The new coronavirus is an enveloped virus. Although ozone's effect upon unsaturated lipids is one of its best-documented biochemical actions, ozone interacts with proteins, carbohydrates, and nucleic acids. O_3/H_2O_2 here is reported as more efficient than ozonation alone because hydrogen peroxide is intended to accelerate the decomposition of ozone in liquid and thus produce more hydroxyl radicals. The best results were obtained from an O_3/H_2O_2 mix at an ozone dose of 20 and 40 mg/L of H_2O_2 . This produces highly reactive compounds after increasing the treatment time to 60 min. The efficiency of removing organic matter and organic compounds from leachate was

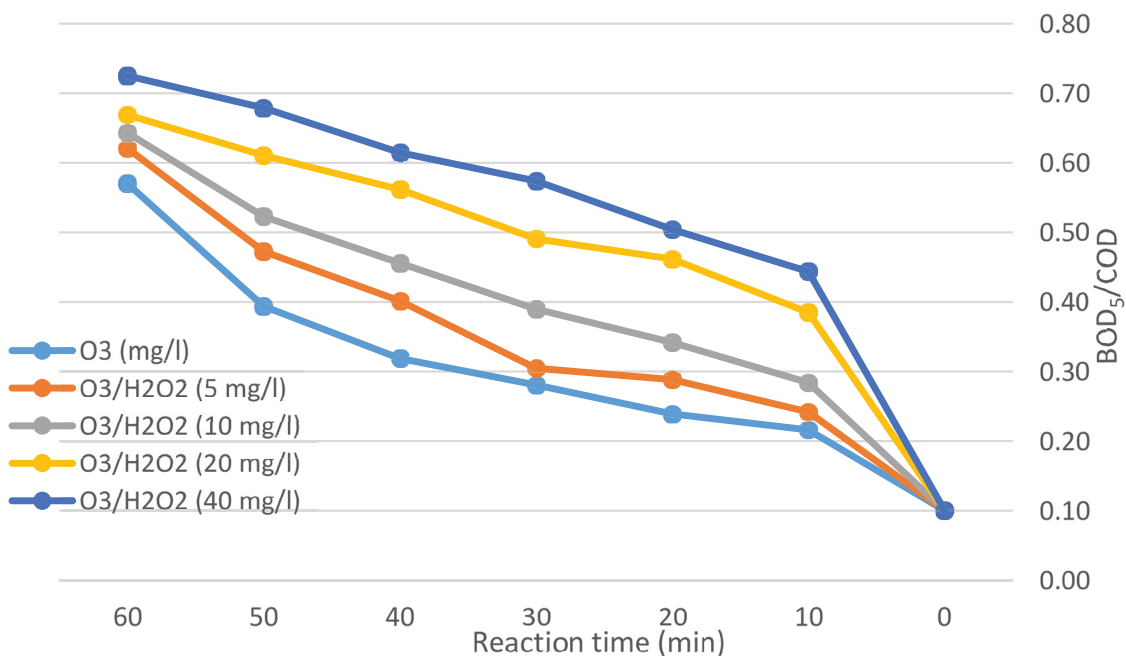


Fig. 9. Effect of O_3 and O_3/H_2O_2 systems on biodegradability BOD_5/COD .

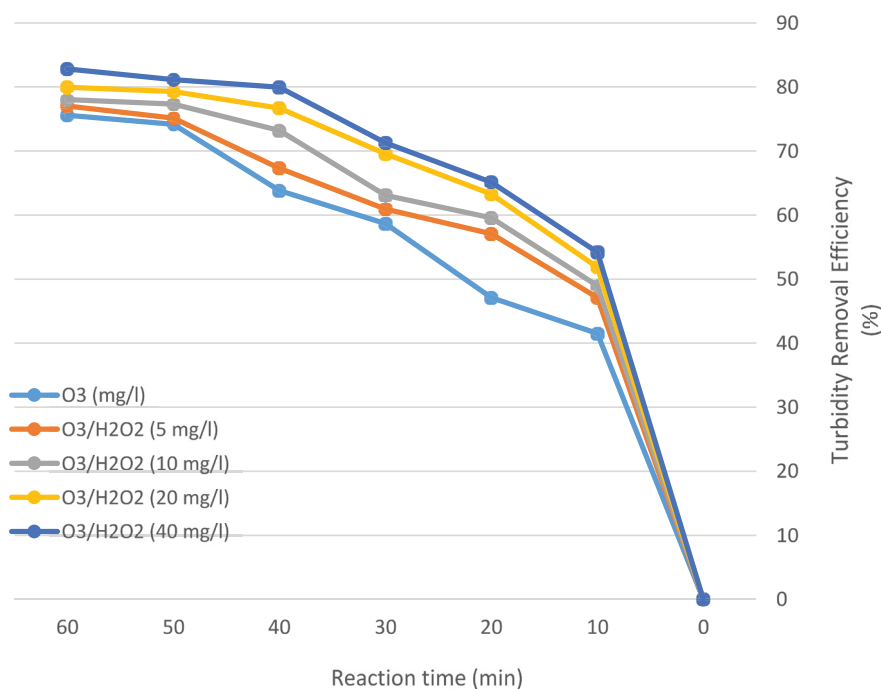


Fig. 10. Turbidity removal efficiency expressed in NTU (nephelometric turbidity units).

characterised through physicochemical parameters. The efficiency of COD removal with O_3/H_2O_2 was up to 92%, greater than O_3 alone, which reached 84%. The efficiency of TOC removal was 29.21–58.42 mg/L. The biodegradation indicated by BOD_5/COD ratio increased from 0.17 to 0.74 after the process, and the turbidity removal efficiency was 75.60%–82.80%. O_3/H_2O_2 offers several advantages, such as free radical production and high bactericide activity; the method is less time consuming and provides high removal efficiencies. Its drawbacks include low solubility of O_3 in aqueous solutions, limited mass transfer, ineffectiveness at high toxic pollutants concentrations, and a high dependence on regulations and leachate characteristics. Thus, combining AOPs with a biological process would significantly decrease the overall cost of leachate treatment research considering the toxicity reduction was involved. However, the toxicity assessment of landfill leachate is very important, which determines the effect of the subsequent biological treatment or the influence on the environment. So, the toxicity reduction of AOPs should be evaluated in future research.

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