Degradation of organics from landfill leachate by combined process of semi-aerobic aged refuse biofilter and ozone oxidation

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ABSTRACT

The effect and mechanism of the combined process of semi-aerobic aged refuse biofilter (SAARB) and ozone oxidation on landfill leachate degradation were investigated. While SAARB pretreatment alone could remove most of the organics, especially 99.97% ammonia (NH_4^+–N), high concentrations of biorefractory organics such as humic acids and fulvic acids were noted in the SAARB-pretreated leachate. Therefore, a subsequent ozone oxidation treatment was performed, which achieved 34.3% chemical oxygen demand (COD), 44.1% UV_{254}, and 67.6% color number (CN) removal efficiencies, respectively. Furthermore, the UV-vis spectroscopy and three-dimensional fluorescence spectrum analyses demonstrated that refractory organics (i.e., aromatic compounds) in SAARB-pretreated leachate were substantially degraded and removed after ozone oxidation process, leading to a significant decline of humification in the leachate. Overall, the combined process of SAARB and ozone oxidation showed a complementary effect on the removal of organics and NH_4^+–N from landfill leachate, and could achieve 95.0% COD, 99.5% CN, 98.2% UV_{254}, and nearly 100.0% NH_4^+–N removal efficiencies.

Keywords: Landfill leachate; Semi-aerobic aged refuse biofilter; Ozone oxidation; UV-Vis spectrum;
Three-dimensional excitation and emission matrix

1. Introduction

In China, the annual output of municipal waste exceeds 200 million tons, with sanitary landfill being the main technology for its management Statistics [1]. During the landfill process, the amount of landfill leachate mainly from precipitation, wastewater, and decomposed water can reach more than 100 million tons/y [1]. Landfill leachate, with its complex composition and high chemical oxygen demand (COD), ammonia (NH_4^+–N) concentration, metal ion content, color, and odor [2–5], is one of the key problems in the prevention and control of sanitary landfill pollution. Therefore, it is very important to find an effective strategy to treat large quantities of highly hazardous leachate.

The current leachate treatment methods include physicochemical methods (e.g., activated carbon adsorption, chemical precipitation, chemical oxidation, ion exchange, reverse osmosis, electrodialysis, etc.) [6,7], biological methods (e.g., bioflocculation, biological fixation, biofilm, biosorption, etc.) [3,8,9], and a combination of these processes, each of which has its own advantages and disadvantages [10,11]. When compared with biological treatments, physicochemical treatments are more stable, especially for leachate with low BOD_5/COD ratio (0.07–0.2) [6,7], however, they are expensive and consume more energy. Biological treatments employ aerobic, anaerobic, or facultative bacteria to fix or decompose organic pollutants into harmless final products, and present advantages such as being more

efficient and economical, ability to treat small molecular organics, and capability to remove nitrogen and phosphorus [3,12,13]. However, biological treatments exhibit poor adaptability and large sludge production, and are therefore often combined with physical and chemical treatments.

Semi-aerobic aged refuse biofilter (SAARB) has been frequently applied for the treatment of landfill leachate and has been proven to be a considerably efficient method to enhance the quality of wastewater, presenting high organics and nitrogen removal efficiencies and low cost [13–17]. It uses aged refuse that has been dumped in landfill for years and has high maturity and stability as a biofilter to degrade wastewater. SAARB includes anaerobic, anoxic, and aerobic areas, and can degrade most of the organics in wastewater through different kinds of microorganisms [15]. Owing to its unique anaerobic–anoxic–aerobic condition, the nitrogen in wastewater can be well-removed by SAARB through ammonification, nitrification, and denitrification. However, some biorefractory organics such as humic acids (HAs) and fulvic acids (FAs) in landfill leachate cannot be degraded by SAARB [14]. Hence, SAARB can only be used as a pretreatment method for landfill leachate, and there is a need to apply other strategies to treat SAARB-pretreated wastewater.

Chemical oxidation, with its strong oxidizing property, is considered to be one of the most effective methods to degrade biorefractory organics in wastewater [4,18–24]. Therefore, this technique can be applied to treat SAARB-pretreated wastewater. In a previous study, Han et al. [15] employed a combination of SAARB (as a pretreatment technique) for treating aged leachate, and achieved satisfactory treatment results, especially for NH_4^+–N removal.

In recent years, ozone oxidation has become one of the key oxidation technologies to remove refractory organic pollutants [25]. Studies have shown that ozone could degrade organics that are a comparatively stable constituent in some wastewaters. Furthermore, ozone oxidation does not produce secondary pollution and the treated wastewater contains only little sludge. Thus, ozone oxidation is widely applied in drinking water and wastewater treatment plants [26] and textile wastewater treatment [27], etc. However, only limited studies have investigated the application of combined process of SAARB and ozone oxidation for landfill leachate treatment.

In the present study, SAARB-ozone oxidation combined technique was employed to treat landfill leachate. The objectives of this study were to (i) explore the organics and nitrogen removal efficiencies of SAARB process; (ii) determine the pollutants removal efficiency and mechanism of the ozone oxidation process; and (iii) establish the pollutants degradation effect of SAARB-ozone oxidation combined technique on landfill leachate treatment.

2. Materials and methods

2.1. Leachate sampling

The leachate used in this study was collected from a large-scale traditional anaerobic landfill in Western China. The landfill has been operated since 1993 and had a leachate daily production of 5,000 ton/d. Once collected, the wastewater was sealed and stored at room temperature and the experiment began immediately. The characteristics of the collected wastewater were as follows: COD, 5,950 mg/L; UV_{254}, 11.55 cm^{-1}; CN, 3.621; NH_4^+–N, 570 mg/L; and total nitrogen (TN), 758 mg/L. These parameters indicated that the wastewater was strongly resistant to degradation owing its high content of pollutants.

2.2. Reagents used

Concentrated sulfuric acid, ferrous ammonium sulfate, o-phenanthroline, potassium dichromate, sodium hydroxide, hydrochloric acid, mercuric sulfate, silver sulfate, etc., used in this study were analytical pure. The pure oxygen concentration employed was 99.999%.

2.3. Experimental setup

2.3.1. SAARB procedure

The laboratory-scale SAARB used in this study had an inner diameter of 300 mm and height of 1,100 mm, and was made of a PVC pipe. The middle of the reactor comprised a 15 mm diameter hole-opening air pipe. The bottom of the reactor was filled with gravel ($\Phi < 50$ mm) under aged refuse so that the water can flow out. The reactor also had functions such as measurement of temperature, settlement height, etc. The aged refuse was collected from an 8 y landfill cell, and a portion of it (≤40 mm diameter) was screened before being filled into the reactor. The acclimation period of the SAARB was 30 d, and the influent hydraulic load was about 20 L/m^3/d. After the acclimation period, the effluent was collected, its characteristics were determined (ICS-1100 ion chromatography, IDEAL, USA), and then stored as the pretreated leachate for subsequent ozone oxidation process.

2.3.2. Ozone oxidation procedure

The initial pH of the pretreated leachate was adjusted to 9.0, and 2,000 mL of the leachate were poured into the ozone reactor (KT-5G Ozone Generator; Shanghai Kangte Environmental Protection Technology Co., Ltd., China), which is a 1 m poly (methyl methacrylate) cylinder with a height of 0.5 m and total volume of 3.0 L. By adjusting the flow of pure oxygen (0.1–0.5 L/min), the amount of ozone (in ρ) was controlled to 78.00 mg/L (UV-200T ozone concentration online analyzer, IDEAL, USA and supporting ozone destruction device). Samples (10 mL) were collected from the reactor outlet at 0.5, 1.0, 2.0, 3.0, 6.0, 9.0, 12.0, 15.0, 20.0, 25.0, and 30.0 min, respectively, and their pH was adjusted to 8.0. Then, the samples were heated in a water bath at 50°C for 30 min with constant stirring to eliminate the effect of residual ozone on subsequent analyses. The samples were cooled to room temperature, and their contaminant indices were analyzed. An illustration of the experimental setup is shown in Fig. 1.

2.4. Analytical methods

The pH was measured by glass electrode method (pHS-3C + acidity meter, Chengdu Ark, China). The oxidation–reduction potential (ORP) was measured using ORP instrument. P (COD_{Cr}) was measured by potassium
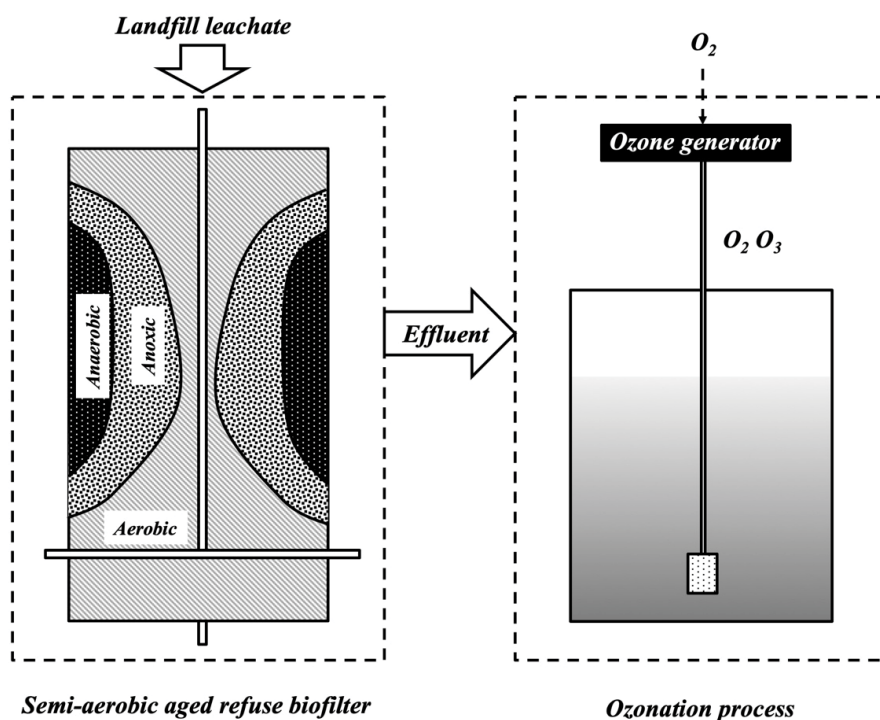
Fig. 1. Experimental setup.

dichromate method (APLMD-6 microwave fast COD analyzer, SPL, China), ammonia nitrogen was determined by a standard method (HJ 535-2009, China) and P (ozone) was monitored by UV spectrophotometry (Lambda 950 UV-vis spectrometer, Perkin-Elmer, USA). The data were collected every 0.5 min and ozone utilization (H) was calculated as follows:

$$n = \frac{78-C}{78} \times 100\% \tag{1}$$

where C represents off-gas ρ (ozone) in mg/L.

The ozone-depleted sample was filtered through a 0.45 μm filter and diluted five times with secondary ultra-pure water for three-dimensional fluorescence spectroscopy (HORIBA Aqualog-UV-800C Three-dimensional fluorescence spectrometer, Japan) and UV-vis spectroscopy. The scanning rate was 500 nm/min, λEx (excitation wavelength) was 239–550 nm, and λEm (emission wavelength) was 250–650 nm. Map drawing, contour settings, etc., were achieved using Origin 2018 software.

The scanning range of UV-vis spectrum was 200–800 nm and the step size was 1.0 nm. Chromaticity (CN) was calculated as follows:

$$CN = \frac{A_{436}^2 + A_{525}^2 + A_{620}^2}{A_{436} + A_{436} + A_{436}} \tag{2}$$

where A_{436}, A_{525}, and A_{620} represent the absorbance at 436, 525, and 620 nm, respectively. The UV_{254} can be an indicator of the relative amount of macromolecular aromatic organics such as humus.

3. Results and discussion

3.1. SAARB pretreatment

As shown in Fig. 2, most of the pollutants in the landfill leachate were significantly removed by SAARB. For example, the COD was reduced from about 5,950 to 452 mg/L, achieving a removal efficiency of 92.4%. The high COD removal efficiency was achieved with significant removal of most of the organic pollutants by the SAARB process through adsorption and degradation, with the remaining pollutants mostly being biorefractory organic acids.
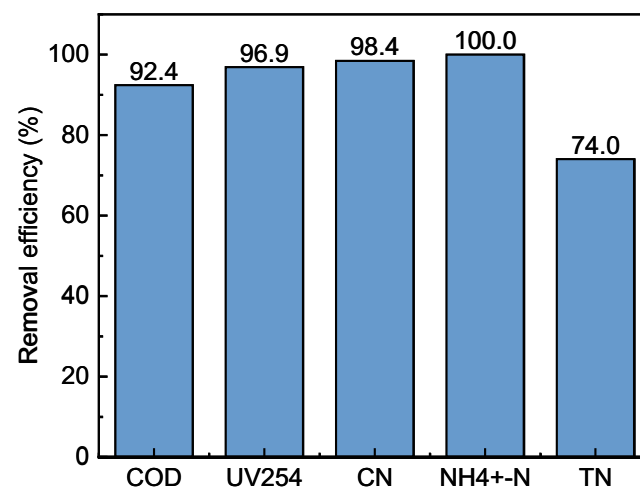

Fig. 2. Contaminant indices of the leachate after SAARB treatment.

Besides, significant TN and NH_4^+–N removal efficiencies (74.0% and 100%, respectively) were achieved by the SAARB process, with most of the NH_4^+–N being removed by nitrification in the aerobic area (near the naturally vented hole-opening air pipe in the SAARB), and TN being eliminated by simultaneous nitrification and denitrification in the anoxic and anaerobic areas of the SAARB [16].

3.2. Ozone oxidation treatment

3.2.1. Analysis of pollutants removal

As shown in Fig. 3, the increase in reaction time was favorable for the degradation of organic pollutants in the pretreated leachate. When the reaction time was 30 min, the COD declined from 452.34 to 297.31 mg/L, CN reduced from 0.056 to 0.018, and the COD and CN removal efficiencies reached 34.3% and 67.6%, respectively. During the reaction, the COD and CN removal efficiencies almost linearly increased; however, the CN removal efficiency was significantly higher than the COD removal efficiency. This was because the CN of organic wastewater was mainly affected by dissolved organic matter (DOM), which consisted of compounds with double bond and aromatic ring that were easily decomposed by ozone. In a previous study, Yu et al. [28] showed that wastewater underwent significant characteristic changes, especially decomposition of macromolecular organic matter to smaller organic molecules, during ozone oxidation treatment.

As shown in Fig. 3c, ozone utilization decreased with reaction time from 0 to 930 s, reaching a lowest point at 930 s when the ozone utilization rate was 35.8%. These results indicated that high organics concentrations, especially macromolecular refractory organics (e.g., HAs and FAs), were beneficial for ozone oxidation in the early stage of the reaction. A previous study demonstrated that ozone could degrade macromolecular organics into smaller ones (e.g., small molecular acids) owing to its strong oxidizing property [29]. However, ozone has a relatively poor ability to further oxidize small molecular organics, leading to their accumulation and decrease in ozone utilization [25]. However, as shown in Fig. 3d, the ozone oxidation reaction was conducted under alkaline condition in the present study, which has been shown to be favorable for the generation of hydroxyl radical (•OH) that can continuously oxidize the ozonation products [25], thus maintaining high organics removal efficiency even when ozone utilization decreased.
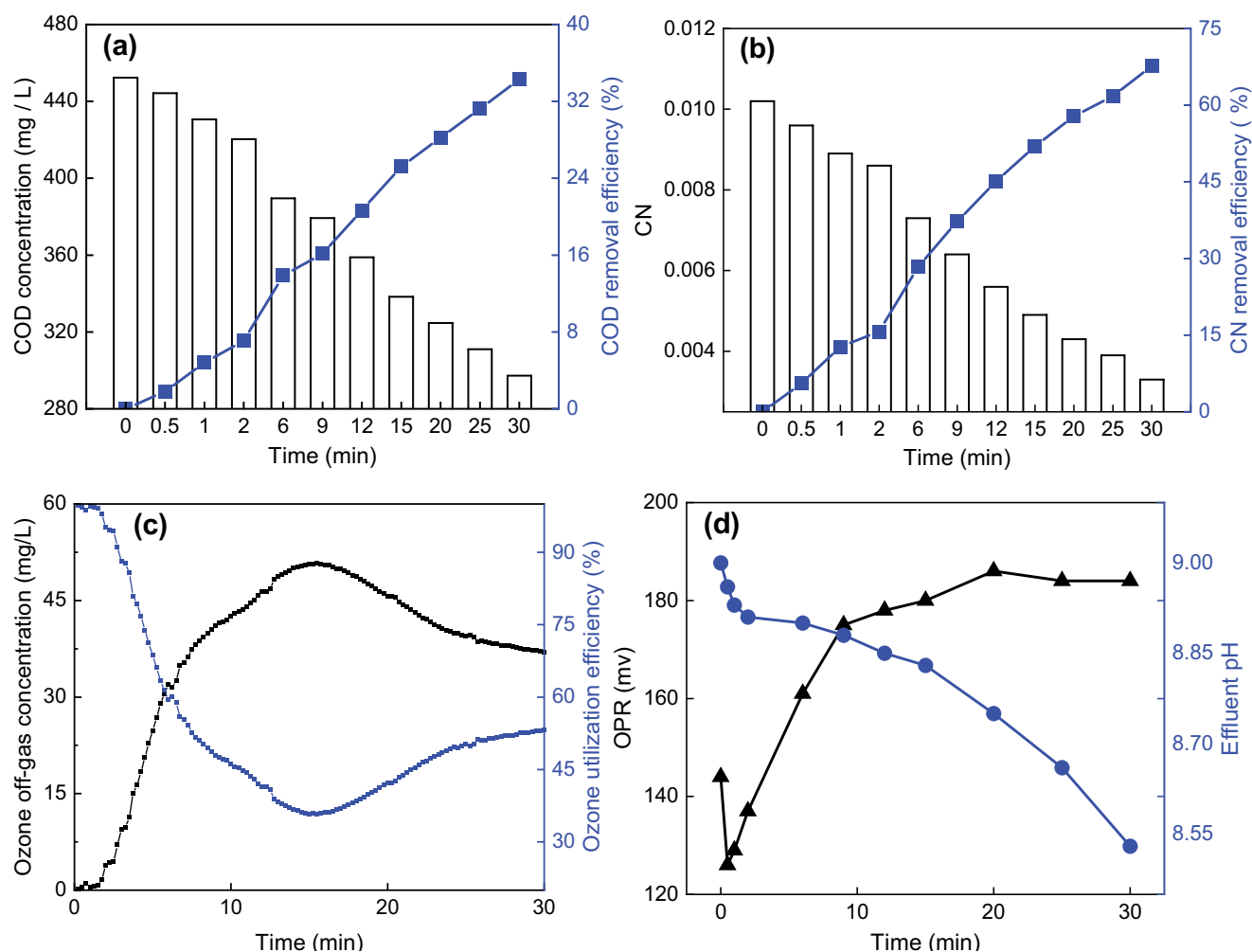

Fig. 3. Variations in (a) COD removal efficiency, (b) CN removal efficiency, (c) ozone utilization, and (d) ORP and effluent pH.

ORP reflects the macroscopic oxidation–reduction of all substances in aqueous solution. As illustrated in Fig. 3d, with the increase in reaction time, the ORP value rapidly increased at 0–9 min, then slowly increased at 9–20 min, and finally stabilized after 20 min. This trend was owing to the initial oxidation of easily oxidizable organics (low-ORP organic matter) in wastewater by ozone, resulting in rapid increase in ORP; subsequently, the recalcitrant organics (high-ORP organic matter) were oxidized by ozone, leading to slow increase in the ORP value until it reached stability. This finding is in accordance with that reported in a previous study on improving biodegradability of landfill leachate by using ozonation [30].

3.2.2. Degradation and transformation mechanism of refractory organics

3.2.2.1. UV-vis spectroscopic analysis

Although SAARB exhibited significant organics removal through adsorption and microbial degradation, large amount of dissolved biorefractory organics with complex structure (e.g., HAs and FAs) still existed in the SAARB-pretreated leachate. Therefore, UV-Vis light absorbance was measured at wavelengths from 200 to 400 nm to analyze the degradation mechanism of these refractory organics during the later ozone oxidation process.

As shown in Fig. 4, the pretreated leachate influent exhibited strong absorbance at 220–250 nm, indicating the presence of conjugated two unsaturated bonds in the leachate. This absorption band corresponded to the *K* absorption band generated by $\pi–\pi^*$ transition [14], which contained conjugated diene structure or α, β unsaturated aldehydes, and ketones structure. Furthermore, moderate intensity absorption was noted at 250–290 nm [31], suggesting the presence of a fine structure (possibly an aromatic ring), and this absorption band could be attributed to a characteristic *B* band of monocyclic rings or a typical band of heterocyclic
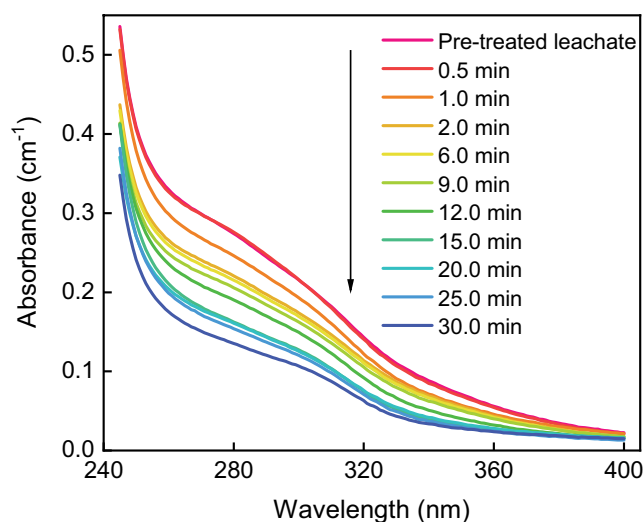
rings. At the wavelength of 290–350 nm [32], a certain intensity of absorption was noted, showing the presence of carbonyl or conjugated groups. Overall, the ozone oxidation effluent showed a substantial decline in absorbance at different wavelengths with the reaction time, indicating that ozone can significantly reduce aromaticity and humification degree.

Some characteristic absorbances of the pretreated leachate before and after ozone oxidation process were determined to thoroughly investigate the degradation effect of aromatic organics. Typically, the absorbance at 254 and 280 nm (expressed as UV_{254} and UV_{280}) indicated the degree of aromaticity of organics, with higher absorbance values denoting higher aromaticity of the organics. In addition, as the absorbance at 226–400 nm could be attributed to multiple conjugated systems of benzene rings, the integral from 226 to 400 nm (expressed as $A_{226–400}$) was used to determine the relative content of aromatic compounds in wastewater. As shown in Fig. 5, obvious downward trends were observed for UV_{254}, UV_{280}, and $A_{226–400}$. For example, after 30 min of ozone oxidation process, UV_{254} and UV_{280} decreased to 0.204 and 0.1353 cm^{-1} from the initial 0.3652 and 0.2471 cm^{-1}, while the $A_{226–400}$ presented a decrease of 17.375, indicating that the aromaticity of the leachate can significantly decrease after ozone oxidation.

3.2.2.2. Three-dimensional excitation and emission matrix analysis

The three-dimensional excitation and emission matrix (3D-EEM) analysis reflects the spectral intensity with changes in excitation and emission wavelengths, providing more complete spectral information, with advantages such as high sensitivity, high selectivity, rapid detection, no use of chemical reagents, etc. Most of the organic pollutants in wastewater contain fluorescent groups, which emit light with characteristic wavelengths under specific wavelengths of excitation light. Therefore, 3D-EEM has potential wide applications in qualitative and quantitative analyses of pollutants in sewage. As shown in Fig. 6, the effluent presented two main light emission regions, among which E_x/E_m = 235–255/410–450 nm was the UV FAs fluorescence region and E_x/E_m = 310–360/370–450 nm was the visible FAs fluorescence region. The fluorescence of FAs in UV region is mainly caused by organic matter with low molecular weight and high fluorescence efficiency (FAs-like), while that in visible region is produced by relatively stable and high molecular weight aromatic substances (HAs-like).

As illustrated in Fig. 7, the intensities of peak *A* and *C* in the pretreated leachate were 4,651.79 and 1,813.85, respectively, denoting relatively high concentration of FAs-like and HAs-like substances. It can be observed from Fig. 7a that peak *A* significantly decreased after 6 min of ozone treatment, indicating that FAs-like substances could be oxidized by ozone in a short time. Furthermore, at 30 and 20 min of ozone treatment, the removal efficiency of Peak *A* and *C* reached 88.4% and 83.2%, respectively, indicating that the refractory FAs in visible region were also significantly oxidized by ozone (Fig. 7b). Moreover, the π-electron system of water changed after ozone treatment, suggesting that the FA aromatic ring reduced and gradually oxidized into small
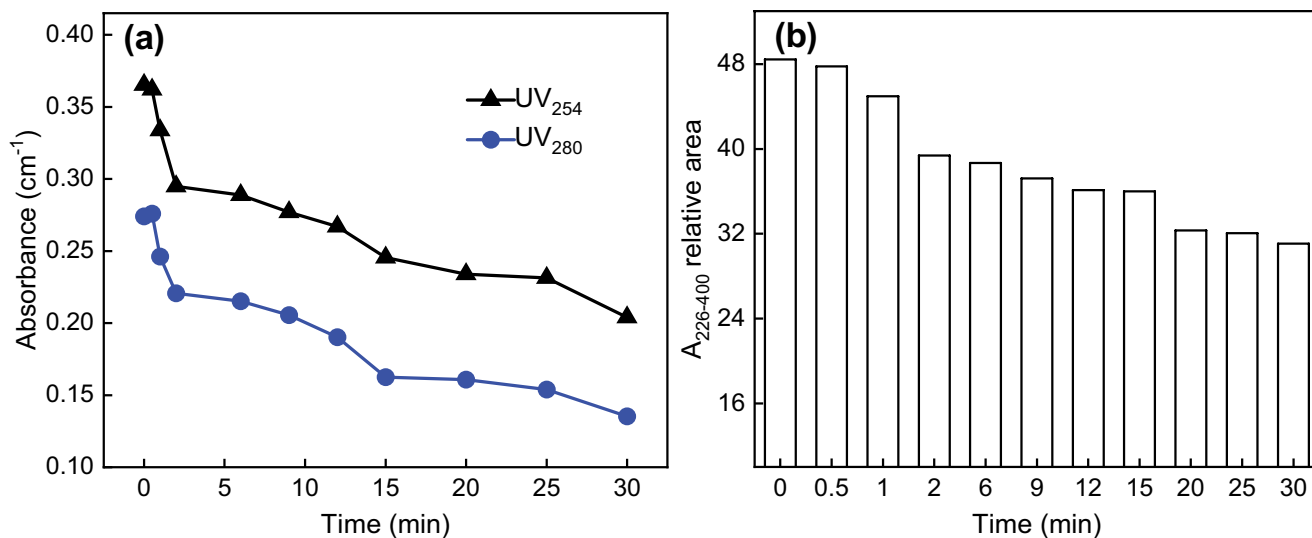
Fig. 4. UV-vis spectra of the pretreated leachate and effluent after ozone oxidation process at different times.

Fig. 5. Variations in characteristic absorbance indices of (a) UV_{254}, E_{280}, and (b) $A_{226-400}$ relative area with reaction time.
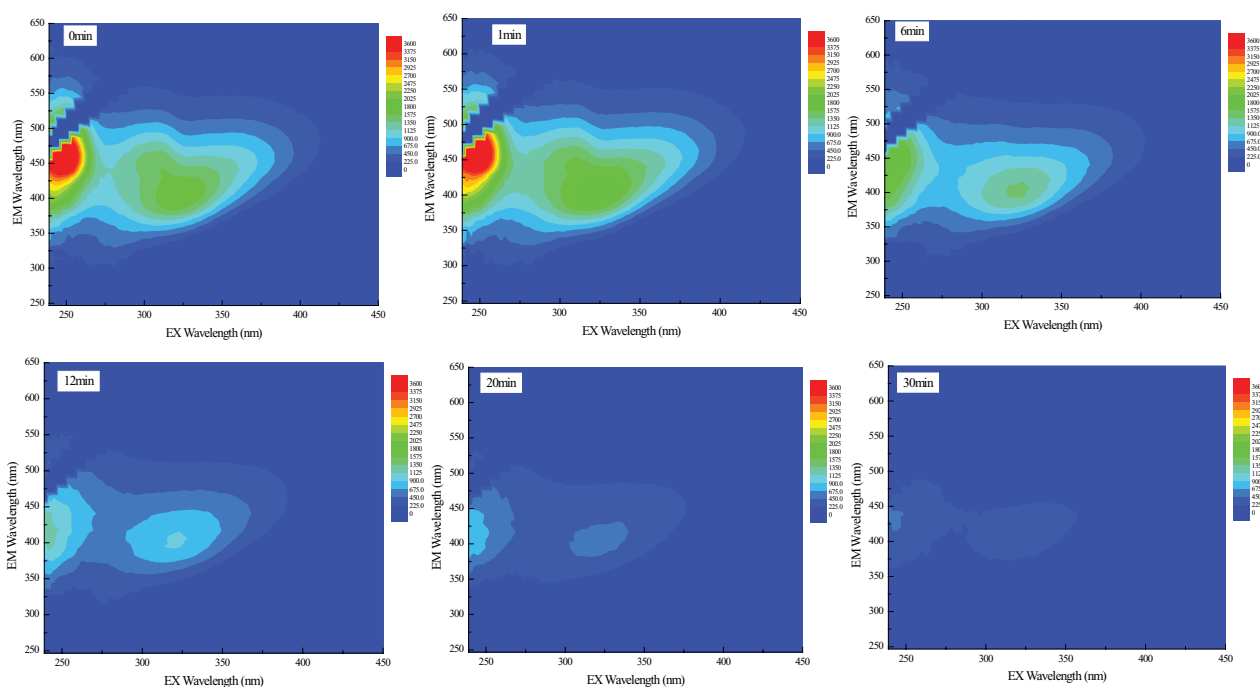

Fig. 6. 3D-EEM spectra of the pretreated leachate before and after ozone oxidation process.

molecules. These 3D-EEM results showed that ozone could effectively degrade refractory organics (i.e., HAs and FAs) in pretreated leachate, consistent with the findings of the UV-Vis spectra.

3.2.3. Nitrogen transformation

The transformation tendencies of different forms of nitrogen are shown in Fig. 8. It can be observed from the figure that the nitrate–nitrogen (NO_3^-–N) concentration was stable, NH_4^+–N level gradually decreased, and nitrite nitrogen (NO_2^-–N) content slightly increased with the reaction time. When the reaction time reached 30 min, a maximum of 76.0% NH_4^+–N removal was achieved, and the NH_4^+–N concentration decreased from 0.150 to 0.036 mg/L (Fig. 8). On the one hand, ozone itself could oxidize NH_4^+–N to NO_x^-–N (Eq. 3), possibly leading to a slight increase in the NO_2^-–N concentration. On the other hand, NH_4^+–N oxidation by ozone is limited, and that the alkaline condition and blow-off effect of ozone may be the main reasons for NH_4^+–N removal. It must be noted that NH_4^+ is the main form of NH_4^+–N in the leachate under neutral pH. However, under
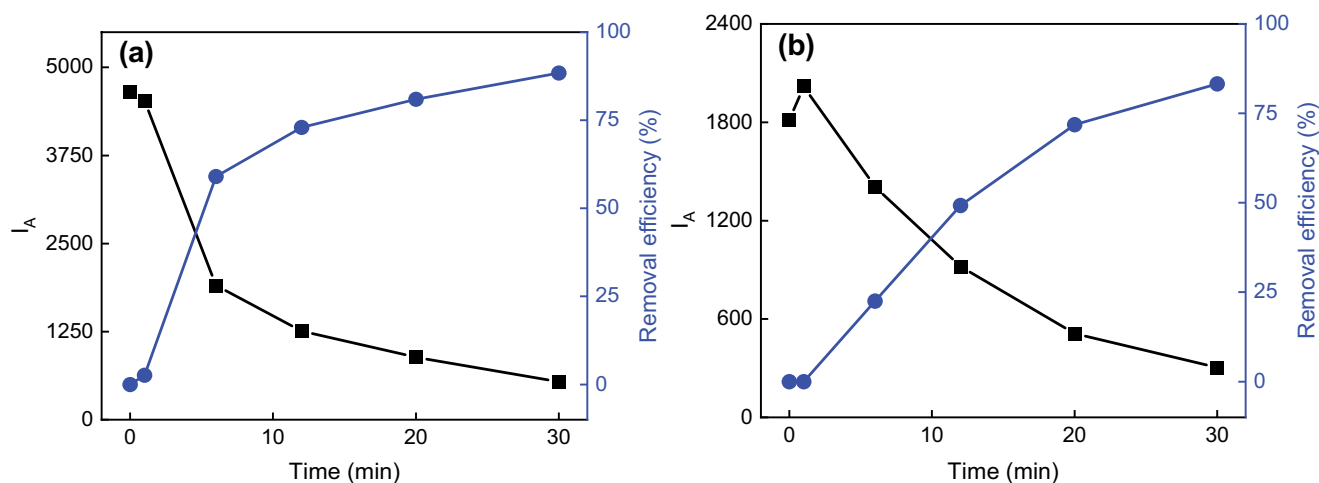
Fig. 7. Intensity variation of (a) peak *A* and (b) peak *C* of the pretreatment leachate after ozone treatment.

alkaline condition, NH_4^+ is inclined to transform to NH_3 (Eq. 4), and with ozone addition into the leachate from the bottom of the experimental column, NH_3 was transformed from liquid phase to gas phase, resulting in high NH_4^+–N removal efficiency.

$$NH_3 + O_3 \rightarrow NO_x + H_2O \qquad (3)$$

$$NH_4^+ + OH^- \rightarrow NH_3 + H_2O \qquad (4)$$

3.3. Pollutants removal performance of the combined treatment

As shown in Fig. 9, in the combined SAARB and ozone oxidation process, the COD removal efficiency was exceptionally high, reaching 95.0%, with influent and effluent COD concentrations of 5,950 and 297 mg/L, respectively. A remarkable CN removal of 99.5% was noted, from initial 3.621 to a final value of 0.018. The UV_{254} was reduced to 0.20 from initial 11.55, with a decline efficiency of 98.2%. The NH_4^+–N removal efficiency was nearly 100.0%, from the initial value of 570 mg/L to a final value of 0.036 mg/L. In particular, the SAARB achieved a significantly higher removal of organics and NH_4^+–N, when compared with the ozone oxidation process. The SAARB presented strong ability to remove organics through adsorption and microbiological degradation, and eliminated most of the NH_4^+–N through nitrification and denitrification owing to its unique anaerobic–anoxic–aerobic environment, however, it could not eliminate dissolved biorefractory organics, which was demonstrated by UV-vis and 3D-EEM analyses. Ozone oxidation process played a crucial role in degradation of dissolved biorefractory organics. As a result, SAARB and ozone oxidation processes were complementary to a certain extent, and the combined treatment exhibited remarkable pollutants removal performance.

4. Conclusion

This study revealed the mechanism of degradation of landfill leachate by combined SAARB and ozone oxidation
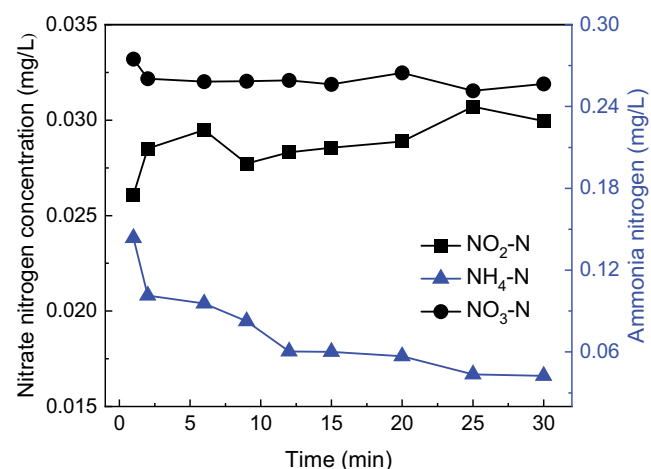

Fig. 8. Nitrogen transformation tendencies during ozone oxidation process.
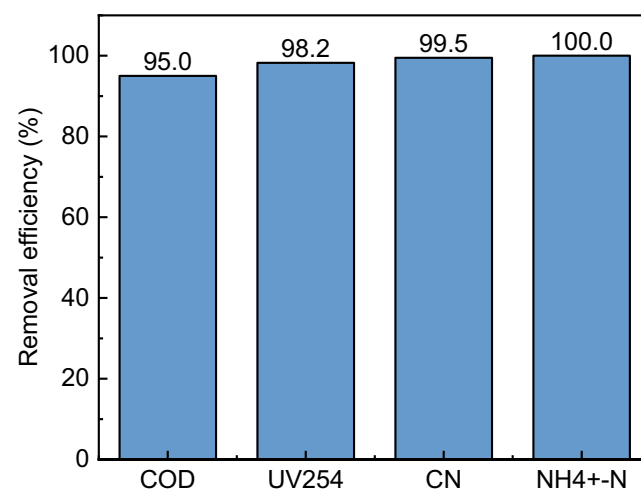

Fig. 9. Contaminant indices of the leachate after combined SAARB and ozone oxidation process.

process. The SAARB pretreatment achieved 92.4% COD, 96.9% UV_{254}, 98.4% CN, nearly 100.0% NH_4^+–N, and 74.0% TN removal efficiencies, respectively. Furthermore, under the reaction time of 30 min, pH of 9.0, and ozone dosage (ρ) of 78.00 mg/L, the CN, and COD removal efficiencies reached 67.6% and 34.3%, respectively. The total UV-Vis spectra showed that ozone could significantly reduce the aromaticity of organics in the SAARB-pretreated leachate, reducing the amount of benzene ring compounds and enhancing their biodegradability. The 3D-EEM analysis revealed that ozone could reduce FAs aromatic ring of the pretreated leachate and gradually oxidize it to small molecules. With the reaction time, the degree of humification in the leachate gradually reduced. The COD, CN, UV_{254}, and NH_4^+–N removal efficiencies of the combined process reached 95.0%, 99.5%, 98.2%, and nearly 100.0%, respectively. These results demonstrated that the SAARB and ozone oxidation process showed complementary effect on the removal of organics and NH_4^+–N, and their combined use could have promising applications in treatment of landfill leachate.

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