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NaOH-Enhanced Wet Air Oxidation of Municipal Sludge for High-Quality Carbon Source Production

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Abstract: Many volatile fatty acids (VFAs) are produced after wet air oxidation, which could be a potential carbon source. In this study, we investigated the impact of NaOH on the removal of hazardous organics and the changes in the produced carbon source. The total and soluble chemical oxygen demand (SCOD) removal rates decreased to 47.9% and 55.3% with 51.6% NaOH addition. The removal rates of total suspended solids (82–85%) and volatile suspended solids (97–99%) remained stable under all conditions. Additionally, the concentrations of acetic acid and isovaleric acid increased with a high pH value. Fluorescent substances closely related to aromatic protein and fulvic acid-like substances were identified and degraded significantly with the addition of NaOH. Moreover, 41.3% NaOH addition (initial pH 13.0) could yield a VFAs/SCOD ratio of 30.5%, demonstrating the good biocompatibility of the carbon source. The effect of the nitrogen element was also considered, with the ratio of the 5-day biological oxygen demand to the total nitrogen being 7.2, indicating that the oxidation solution could provide an abundant carbon source. Thus, the sludge-derived carbon source is suitable to supply biological treatment units for municipal wastewater.

Keywords: wet air oxidation; municipal sludge; NaOH; carbon source; volatile fatty acids

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Academic Editors: Jorge Padrão and Sigrid Kusch-Brandt

Received: 21 November 2024
Revised: 17 January 2025
Accepted: 24 January 2025
Published: 26 January 2025

Citation: Fang, K.; Tong, Y.; Yao, G.; Zhu, N.; Jin, L.; Zhou, Y.; Zhao, J. NaOH-Enhanced Wet Air Oxidation of Municipal Sludge for High-Quality Carbon Source Production. *Processes* **2025**, *13*, 341. https://doi.org/10.3390/pr13020341

1. Introduction

Municipal sludge (MS) is a type of waste produced in the process of municipal sewage treatment and is mainly composed of microbial bacteria, organic residues, and in-organic particles [1]. The main features of MS include high organic content [2,3], high moisture content [1,2,4], and a cementitious structure [2]. Additionally, MS may contain harmful substances such as parasites, pathogenic microorganisms, and heavy metals [5–7], which could pose a potential threat to the environment and human health without appropriate management. With the rapid advancement of the economy and society, the demand for sustainable waste management is escalating, and MS treatment is becoming an increasingly important issue worldwide [3].

Conventional treatment approaches for MS include landfill, composting, and incineration. Landfill has become the primary treatment method for MS due to its large capacity and adaptability in the last few decades [8]. However, this method not only occupies a significant amount of land resources but also poses risks of leachate pollution and heavy metal leaching, potentially leading to secondary environmental issues such as soil and water contamination [9]. Although composting can be an effective method, the presence of heavy metals, antibiotics, harmful pollutants, and microbial pathogens in MS poses a potential threat to health over a long period, particularly if used as a fertilizer or soil amendment in agricultural production [10,11]. Incineration effectively neutralizes MS, but it requires high initial investments and drying pretreatment, which increases the operating costs and results in exhaust emissions [3,12]. For these reasons, conventional treatment techniques face challenges in adapting to the increasingly strict environmental requirements [13]. Hence, advanced oxidation processes (AOPs) have garnered more attention for the better handling of MS. Compared to conventional methods, AOPs produce hydroxyl radicals with a strong oxidizing effect, which enables them to more efficiently oxidize and degrade persistent organic compounds. Additionally, AOPs can handle various substrates without causing secondary pollution [14,15]. Specifically, AOPs include various technologies, such as ozone oxidation [16], Fenton oxidation [17], electrochemical oxidation [18], and wet air oxidation (WAO). Among these methods, WAO is particularly effective in treating waste with a high concentration and toxicity [19,20].

WAO is a highly efficient hydrothermal treatment technology that uses oxygen or air as an oxidant under high-temperature and -pressure conditions to oxidize and decompose pollutants into small organic or inorganic substances [21]. This process does not produce toxic and harmful substances such as nitrous oxide, sulfur dioxide, hydrochloric acid, dioxins, furans, or fly ash, thereby effectively avoiding secondary pollution [20,22]. WAO can also be used to deal with organic pollutants that are poorly biodegradable, such as pyridine and benzene compounds. Additionally, the WAO process holds great potential for resource recovery [19–21,23–27]. Numerous studies have demonstrated that WAO treatment can convert organic substances into volatile fatty acids (VFAs) under specific conditions [19,20,24,26,28]. These VFAs are not only soluble but also possess high energy conversion efficiency and good biocompatibility. Thus, they can be employed as an additional carbon source for biological treatment [23,29,30], particularly in promoting biological nitrogen removal [31].

It is important to note that most of the organic nitrogen in MS is converted to ammonia nitrogen (AN) instead of nitrogen gas after WAO. Baroutian et al. [19] discovered that the increase in AN content might be attributed to the hydrolysis of particulate nitrogen in sludge. Experiments conducted by Xu et al. [32] indicated that AN has strong anti-degradation properties and cannot be effectively removed, even when treated at a high temperature of 400 °C for 30 min. Zhao et al. [33] further explored the nitrogen conversion pathway during MS pyrolysis. Their results revealed that, as the reaction progressed, the protein nitrogen in MS was gradually decomposed into three types of intermediates, amine nitrogen, nitrile nitrogen, and heterocyclic nitrogen, which were all eventually decomposed into AN. When the oxidation solution is used as a supplemental carbon source to aid bioreactors, the presence of AN introduces an additional nitrogen treatment load, thereby restricting the application potential of the WAO liquid as a carbon source.

Therefore, the reaction conditions must be regulated to enhance the amount and quality of the produced carbon source. Chung et al. [28] discovered that the reaction time and temperature were crucial factors influencing the liquefaction of volatile solids. As the reaction time increased and the reaction temperature rose, the degradation efficiency of the sludge and the yield of organic acids, predominantly acetic acid, were enhanced. Baroutian et al. [34] further investigated the relative effects of operational variables in the treatment of fermented MS, finding that the temperature and oxygen ratio had the most significant impacts. Aggrey et al. [35] treated urban biosolids through a combination of thermal hydrolysis and WAO, significantly boosting the production and purity of acetic acid. Additionally, adding catalysts is an effective means to improve the treatment efficiency, particularly in AN removal [36]. However, the high cost of catalysts and the need for recycling restrict their wide application [37]. Information on the application of catalytic oxidation for complex substrates such as MS is also limited, and this needs further exploration [38]. NaOH is recognized as an effective promoter in the conversion of organic matter to VFAs in hydrothermal reactions; thus, the quality of the oxidation liquid can be increased [39]. Moreover, sodium acetate possesses better thermal stability and thus more of the carbon source can be obtained. Furthermore, the current studies on NaOH are not comprehensive enough and focus only on the removal rates, chemical oxygen demand (COD), and volatile suspended solids (VSS).

The aim of this study was to enhance the WAO process of MS by adding NaOH to obtain a high-quality carbon source. The specific goals were as follows: (1) investigating the influence of the NaOH dosage on the treatment efficiency, including pH value variations and the removal rates of total COD (TCOD), soluble COD (SCOD), total suspended solids (TSS), and VSS; (2) measuring the concentrations of various VFAs in the oxidation solution to assess the quality of the sludge-derived carbon source; (3) studying the effect of NaOH on other types of dissolved organic matter, represented by fluorescent substances; (4) analyzing the changes in total nitrogen (TN), AN, and the five-day biological oxygen demand (BOD_5) to ensure the availability of the carbon source.

2. Materials and Methods

2.1. Materials

The MS used in this study was secondary sludge from Ordos, China. After sampling, the sludge was stored at 4 °C throughout the whole research process. The basic parameters were as follows: TCOD, 19.1 ± 0.3 g/L; TSS, 16.7 ± 0.6 g/L; VSS/TSS, 72.0 ± 1.5%. NaOH and other reagents used in the experimental process were purchased from Sinopharm Chemical Reagent Co., Ltd, Shanghai, China and were of analytical purity.

2.2. WAO Process

The WAO process for MS was carried out in a 250 mL stainless steel autoclave reactor (YZPR-250, Yanzheng, Shanghai, China). The experimental procedure involved adding 40 g of sludge and 10 mL of pure water to the reactor. The amount of water replaced ranged from 2 mL to 10 mL, with 1 mol/L NaOH solution to adjust the initial pH value. The amount of NaOH added was expressed as a percentage of the mass of NaOH relative to the COD value of the substrate liquid, ranging from 0% to 51.6%. The reactor was then vented three times with O_2 at 0.5 MPa (absolute pressure) before being pressurized to 0.9 MPa. The reaction was conducted at a temperature of 240 °C for 60 min, with magnetic stirring maintained at 300 rpm throughout. The reaction conditions were determined according to our previous research and experience [40]. Under these conditions, the oxidation liquid was more likely to be a high-quality carbon source. The reaction started once the temperature in the reactor had reached the preset value. Upon completion, the oxidation solution was sampled and the rest was filtered through a 0.45 μm membrane to obtain a clear filtrate for further analysis.

2.3. Analytical Methods

The TCOD and SCOD were measured using dichromate digestion–spectrophotometry. AN was measured using sodium hypochlorite–salicylic acid spectrophotometry, and TN was measured using alkaline potassium persulfate digestion–spectrophotometry. The spectrophotometer used was a DR3000, purchased from HACH, Loveland, USA. The TSS and VSS were measured by mass analysis using a muffle furnace (SX2, Lichen, Shanghai, China). The details of these measurements and the test for BOD_5 followed the standard methods [41]. Gas chromatography (G5, Persee, Beijing, China) was employed to measure VFAs using a flame ionization detector, and this was equipped with a bonded polyethylene glycol capillary column (DB-FFAP, 30 m× 0.25 mm × 0.25 µm, Agilent, Santa Clara, USA). The samples were adjusted to a pH of less than 2.0 before measurement. Helium was used as a carrier gas, with the temperature of the oven set at 160 °C. Pure organic acids were tested as standard samples, and the signal intensity was found to be directly proportional to the concentration. The R^2 value in the linear regression model was consistently higher than 0.99. Three-dimensional excitation emission matrices (3D-EEMs) were detected using a fluorescence spectrometer (F-7100, Hitachi, Tokyo, Japan) with wavelength scanning from 200 to 600 nm for both excitation (Ex) and emission (Em). The samples were diluted 500 times before measurement to avoid inner-filter correction [42]. Ultrapure water was used as a blank sample, and all data were Raman-normalized based on the peak area at 350 nm of water [40]. Following the removal of Raman and Rayleigh scattering, the data were further deciphered using fluorescence regional integration (FRI) [43].

3. Results and Discussion

The degradation of pollutants in the NaOH-enhanced WAO process was initially tested in terms of TSS and VSS removal to ensure the basic function of this sludge treatment process. Meanwhile, the pH value before and after the reaction was measured to indicate the depletion of NaOH. Subsequently, VFAs, as the main components of COD, were investigated to examine the amount and quality of the sludge-derived carbon source. Then, the effect of the alkaline environment on the dissolved organic matter was evaluated, represented by fluorescent organics shown in the form of 3D-EEMs. Finally, we examined the impact of the nitrogen element on the oxidation liquid to ensure that it was a practical and high-quality carbon source.

3.1. Removal of Organic Pollutants and pH Changes

Figure 1a shows the change in the pH of the solution before and after the NaOH-enhanced WAO. The figure indicates that the pH of the substrate increased significantly from 7.1, reaching 12.0 with 10.3% NaOH addition and peaking at 13.1 with 51.6% addition. In contrast, the pH value of the clear liquid after the reaction showed a modest increase, rising from 7.2 to 8.8. This synchronous increase in pH with the addition of NaOH was followed by a significant decrease in the pH and alkalinity after the reaction, indicating substantial hydroxide consumption during the WAO treatment. Specifically, during the hydrolysis phase of protein organic matter, hydroxide ions act as nucleophiles on peptide bonds, facilitating protein decomposition into free amino acids and resulting in a decrease in the hydroxide concentration [44]. Urrea et al. [45] found that the decomposition of organic matter forms intermediates with acidic characteristics (such as humic acids, fatty acids, and uronic acids), causing a significant initial pH drop. As the reaction progresses, these intermediates further decompose into organic acids, carbon dioxide, and water, leading to a pH rise. In this experiment, under strongly alkaline conditions, acidic intermediates reacted with the hydroxide before being completely mineralized, further lowering the pH of the oxidation solution. Furthermore, the organic matter was eventually mineralized to carbon dioxide, and the excess sodium hydroxide absorbed the carbon dioxide to form carbonates, contributing to the pH drop.

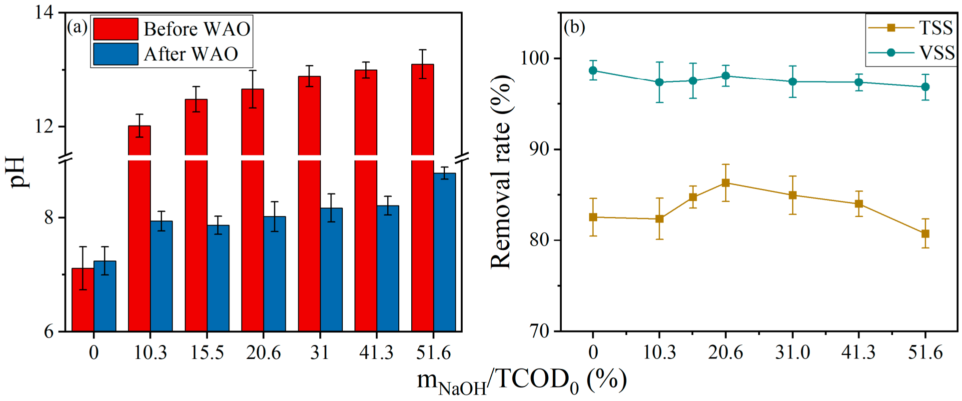

Figure 1. The influence of NaOH addition on the pH before and after the reaction (**a**) and the removal rates of TSS and VSS (**b**).

Figure 1b illustrates the trends of the TSS and VSS removal rates with the addition of NaOH after WAO treatment. The data indicate that, with increasing NaOH, the TSS removal rate initially rose slightly from 82.5% to 86.3% and then gradually decreased to 80.7%, but remained relatively stable overall. Meanwhile, the VSS removal rate remained high and was almost unaffected by the addition of NaOH, staying stable at approximately 97%. Baroutian et al. [19] achieved an 85% TSS removal rate and a 92% VSS removal rate when using WAO to treat MS at 240 °C for 60 min. Our results were consistent with theirs, indicating that the waste reduction function of WAO was robust and was not significantly affected by the addition of NaOH.

3.2. Amount and Quality of Produced Carbon Source

The composition of MS is complex, and the reaction products are also intricate mixtures due to the free radical reaction mechanism. By monitoring the TCOD and SCOD, the effectiveness of the treatment can be evaluated to some extent [46]. As shown in Figure 2a, the addition of NaOH to the substrate markedly affected the removal rates of both TCOD and SCOD, which referred to the COD difference between the substrate and total or soluble residuals. Initially, the TCOD removal rate significantly decreased from its highest value of 66.2% to the lowest of 43.6% when 31.0% NaOH was added, before slightly increasing back to 47.9%. A similar trend was observed for the SCOD removal rate, which initially decreased to a minimum of 53.9% with 41.3% NaOH addition and then slightly recovered.

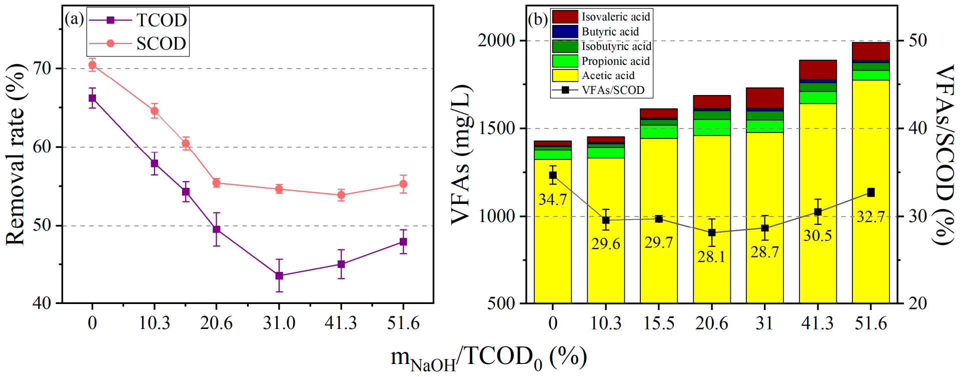

Figure 2. The influence of NaOH addition on the removal rates of TCOD and SCOD (**a**), as well as the production of VFAs and the VFAs/SCOD ratio (**b**).

The considerable variation in the removal rates of TCOD and SCOD was attributed to the enhanced biomass destruction promoted by NaOH, which further disrupted the flocculation structure and accelerated the denaturation of proteins on the cell walls [29].

As the cell walls rupture, large quantities of intracellular biopolymers, such as proteins, polysaccharides, lipids, and other intracellular polymers, dissolve into the liquid phase. At the same time, a significant amount of insoluble organic matter underwent hydrolysis and depolymerization, leading to increased concentrations of TCOD and SCOD [47]. The reduction in the TCOD and SCOD removal rates was not only due to the increased total amount but also related to the higher concentration of stable intermediates. As the pH rose, more organic matter was hydrolyzed into small organic acids, as shown by the continuous increase in VFAs production with the increasing pH, illustrated in Figure 2b. These organic acids, especially acetic acid, were highly resistant to degradation [48] and could not be completely mineralized, even at higher temperatures, thus reducing the removal rates of TCOD and SCOD.

Figure 2b shows the changes in the content of different VFAs and the VFAs/SCOD ratio with the increase in NaOH addition. The composition of VFAs remained relatively stable, with acetic acid consistently being the most significant component, accounting for more than 85%, followed by propionic acid. The formation and behavior of acetic acid align with previous studies, confirming it as the main product of WAO [48,49]. Due to its relatively stable properties and resistance to oxidation, acetic acid is regarded as a stable intermediate that remains in the system as a by-product [46]. As the NaOH dosage increased, the VFA concentrations exhibited a rapid growth trend. The most significant effects of NaOH were observed in the concentrations of acetic acid and isovaleric acid, with acetic acid increasing from 1323 mg/L to 1774 mg/L and isovaleric acid rising from 24 mg/L to a peak of 113 mg/L. This might be influenced by the carbonate generated when NaOH captures carbon dioxide, which attenuated the activity of the hydroxyl radical [50]. Consequently, organic matter was less likely to be fully mineralized to carbon dioxide and remained in the form of small-molecule intermediates, thereby increasing the concentration of VFAs. With the addition of NaOH, the proportion of VFAs/SCOD decreased slightly from 34.7% to 29.6% before rebounding to 32.7%. This slight decrease could be attributed to the significant rise in the SCOD concentration. Since VFAs are small molecules that can be easily utilized by microorganisms, higher VFA concentrations result in a better carbon source [23]. Therefore, the VFAs/SCOD ratio can reflect the quality of the oxidation liquid as a carbon source. In conclusion, the VFAs/SCOD ratio remained stable after NaOH addition, indicating that the quality of this carbon source converted from MS was assured.

3.3. Changes in Fluorescent Organics

Fluorescent substances are important components of the oxidation liquid as they can be generated during the reaction as intermediates. Meanwhile, many types of dissolved organic matter, other than VFAs, remain unknown and require further investigation. Therefore, 3D-EEM coupled with FRI was employed to study the impact of NaOH addition on the trends of other types of dissolved organic matter, represented by fluorescent substances.

Figure 3 illustrates the visualized 3D-EEMs of all samples, while Figure 4 shows the detailed data of these samples after FRI analysis. The plot space is divided into five regions: I, aromatic protein I; II, aromatic protein II; III, fulvic acid-like; IV, soluble microbial by-product-like; V, humic acid-like [43]. Four peaks could be clearly observed as the main components. The primary peak, centered at Ex/Em of 230/400 nm, was a fulvic acid-like substance. The secondary peak, centered at Ex/Em of 300/395 nm, was a humic acid-like substance. The other two peaks, weaker in intensity, were at Ex/Em of 215/295 nm and 275/310 nm, corresponding to aromatic protein I and tyrosine and protein-like substances, respectively. It was also observed that, with the addition of NaOH, the intensity of these peaks initially rose and then decreased sharply.

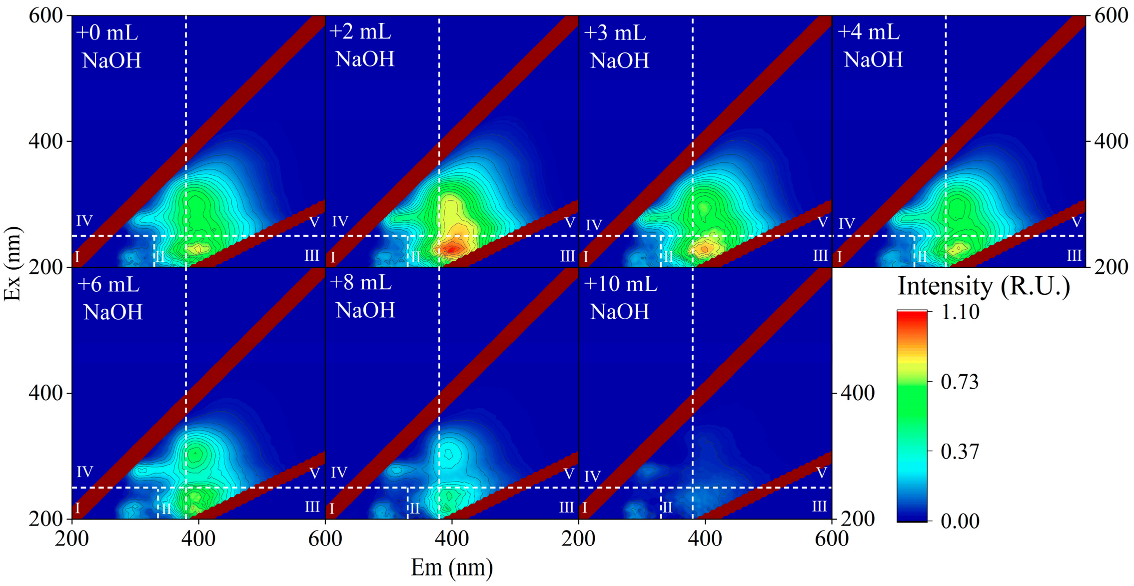

Figure 3. The 3D-EEMs of samples with different amounts of NaOH solution addition (I–V refers to the region division in FRI).

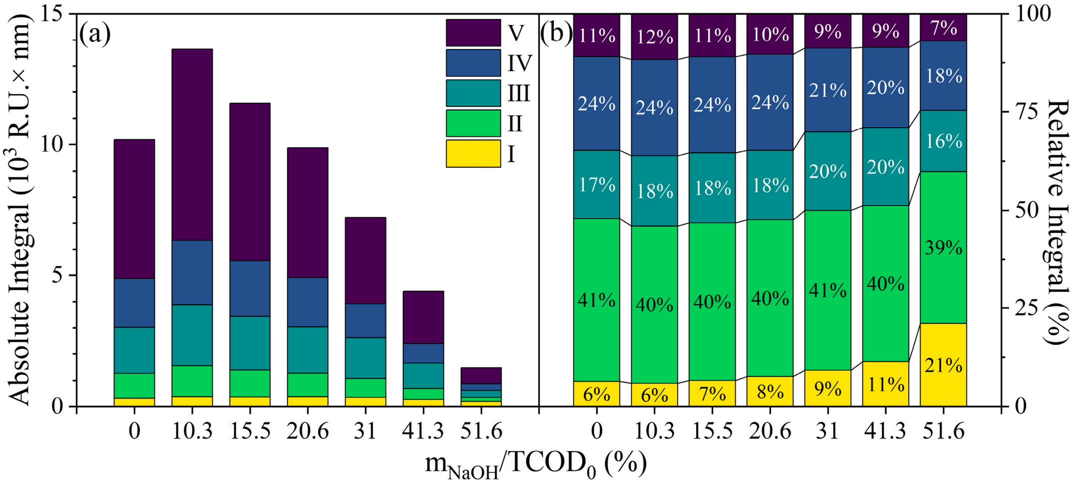

Figure 4. The absolute integral (**a**) and relative integral (**b**) of samples with varying amounts of NaOH solution addition, categorized by different FRI regions.

With the help of FRI, further investigation was conducted. Initially, the absolute integral increased by 34% after 10.3% NaOH addition, likely due to enhanced sludge cell decomposition, generating more fluorescent intermediates. The integral then decreased to 15% of the no-NaOH sample, suggesting that more NaOH primarily converted organics into smaller molecules. Generally, the distribution of the relative integral (Figure 4) remained stable, with aromatic protein II (region II) as the main component, followed by fulvic acid-like and soluble microbial by-product-like substances. Notably, region I increased from 6% to 21% after 51.6% NaOH addition, indicating that aromatic protein I was more resistant to degradation than the other fluorescent substances through WAO enhanced by NaOH.

Combining this with the SCOD trend shown in Figure 5b, the SCOD remained stable after 20.6% NaOH addition, while the fluorescent substances were significantly reduced. This suggested that NaOH initially increased the SCOD and then promoted its degradation. In other words, the quantity of the carbon source was increased at first, and then the quality was enhanced.

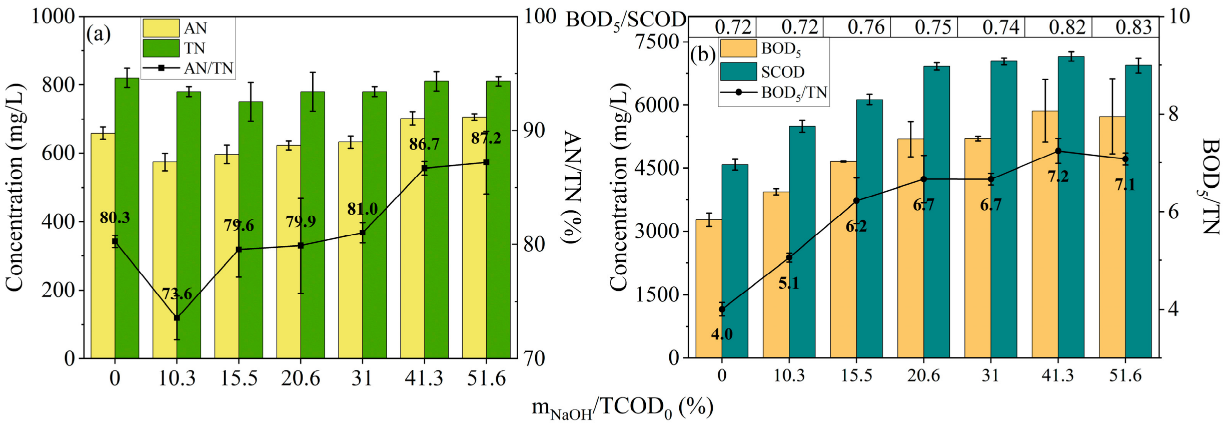

Figure 5. The influence of NaOH addition on the concentrations of AN, TN, and AN/TN (**a**), as well as the concentrations of BOD_5, SCOD, BOD_5/SCOD, and BOD_5/TN (**b**).

3.4. Effects of Nitrogen on the Carbon Source

Figure 5a shows the changes in the AN and TN concentrations, as well as the AN/TN ratio, with the addition of NaOH. The figure indicates that the effect of NaOH on TN was not significant, with its concentration remaining around 800 mg/L. However, the addition of NaOH had a noticeable effect on AN, reflected in both the concentration change and the trend of AN/TN. When 10.3% NaOH was added, the AN concentration significantly decreased from 658 mg/L to 574 mg/L, and the corresponding AN/TN ratio dropped from 80.3% to 73.6%, suggesting that a small amount of NaOH did not favor the decomposition of nitrogen-containing organic matter. However, this trend was reversed with further NaOH addition. As the amount of NaOH increased, the AN concentration rebounded, peaking at 706 mg/L. Simultaneously, the AN/TN ratio rose to 87.2%, indicating that more NaOH could promote the decomposition of nitrogenous organic matter and its conversion to AN.

Figure 5b illustrates the correlation between NaOH addition and the BOD_5 and SCOD concentrations, as well as the BOD_5/SCOD (B/C) and BOD_5/TN (B/N) ratios. With increasing NaOH addition, both the BOD_5 and SCOD concentrations rose. B/C also rose with a particularly pronounced increase in BOD_5. Specifically, BOD_5 escalated from 3280 mg/L to 5860 mg/L with 41.3% NaOH addition, while the SCOD rose from 4580 mg/L to 7150 mg/L. Consequently, B/C increased from 0.72 to 0.82, signifying an improvement in both the quality and quantity of the carbon source. Previous studies have indicated that the optimal B/N ratio to achieve the maximum denitrification efficiency for bioreactors typically ranges between 4 and 5 [51,52]. The B/N of the oxidation solution without NaOH remained at only 4.0 due to the low organic carbon content, rendering it nearly ineffective as a supplementary carbon source for denitrification bioreactors. However, following the addition of 41.3% NaOH, there was a significant increase in B/N to a peak value of 7.2, which demonstrates the powerful effects of NaOH in producing a carbon source.

3.5. Discussion

As mentioned in the Introduction, although incineration seems to be a prevalent and promising method for MS disposal, the initial investment is very high and it is considered a major source of carbon dioxide emissions [12,53]. Nowadays, there is increasing concern about the resource utilization of waste to reduce carbon dioxide emissions and promote sustainability. While anaerobic digestion is considered the most suitable pathway among the most common disposal routes due to its relatively low economic cost, it suffers from low time efficiency and a low conversion rate for organic matter, typically ranging between 35% and 60% [54]. In the field of AOPs, significant efforts have been devoted to convert-ing sludge into practical and useful carbon sources due to the high VFA concentrations remaining in the liquid phase. For example, Liao et al. [55] used a microwave–hydrogen peroxide–sulfuric acid enhanced process to treat sludge and recover volatile fatty acids, achieving a COD removal rate of 53.5% and an acetic acid/COD ratio of 21.0% with the addition of 5.8 mg/L H_2O_2 per mg/L COD at 120 °C. While this addition was substantial, the NaOH addition in our study was minimal, with a maximum of 51.6%.

Energy consumption is also a major concern for the WAO process, as the substrate needs to be heated. It has been reported that the energy released during oxidation reactions can replenish part or even all of the energy demand [56]. Specifically, it was calculated that a substrate with 10,000 mg/L COD could enable the reaction to automatically take place at 150 °C [22]. Therefore, at higher reaction temperatures, the energy required is acceptable and can be replenished as long as the concentration of the substrate is increased. Meanwhile, the energy consumed can be recycled by adding a heat exchanger between the hot effluent and cold influent in industrial applications. Furthermore, the energy demand can also be reduced by adjusting the reaction conditions and combining WAO with other advanced techniques such as sub-millimeter bubbles [57,58]. In comparison with the mono-incineration of MS, where the substrate must be dried first, consuming multiple times the energy of the whole WAO process [53], WAO technology has great potential for commercialization [59].

NaOH is recognized as an efficient additive in promoting the degradation of pollutants. However, the advantage of increasing VFA concentrations has not been sufficiently emphasized, and several related studies are not comprehensive enough [56,60]. In this study, we not only examined the effect of NaOH addition on common parameters, including the COD, TSS, VSS, and VFAs, but also investigated changes in fluorescent organics and the impact of nitrogen. Therefore, our study is more comprehensive and can help scholars to better understand the effect of NaOH. A key finding of our study was that 51.6% NaOH addition can lead to a 39% increase in the VFA concentrations and a 78% increase in B/C, indicating that MS could be converted into a practical and high-quality carbon source. For sludge with 80% moisture content, the reagent cost is about USD 15 per ton based on the parameters gained in this study. The energy required can be primarily supplied by the oxidation reaction itself, with green power providing a solid supplement. Through this process, each ton of sludge treated saves about USD 35 in disposal fees and USD 14 in purchasing carbon sources, making it highly economically viable.

4. Conclusions

In this study, we discussed the effect of NaOH addition on the WAO treatment of MS. The results showed that (1) the addition of NaOH had no negative effect on TSS and VSS removal, while the concentration of SCOD was elevated; (2) the production of VFAs increased with rising NaOH addition, with acetic acid being the main component; isovaleric acid also increased significantly; (3) the intensity of fluorescent substances initially increased and then rapidly decreased with the addition of NaOH, indicating that NaOH addition could help to degrade refractory organics; (4) the maximum B/N of 7.2 could be achieved with the addition of 41.3% NaOH (initial pH 13.1). Simultaneously, the VFAs/SCOD and B/C ratios were 30.5% and 0.82, ensuring both the quantity and quality of the carbon source.

As mentioned in Section 3.5, NaOH-enhanced WAO is economically acceptable in terms of both chemical addition and energy consumption. It also offers multiple advantages compared to other disposal methods, such as higher efficiency than anaerobic digestion and lower carbon dioxide emissions than incineration.

The utilization of NaOH-enhanced WAO can be expanded to other toxic and hazardous wastes, such as pharmaceutical sludge and wastewater, to achieve greater economic benefits. However, while continuous flow reaction systems are more accurate for practical applications, the data in this study were obtained from a batch reactor. This might be a limitation of our research.

In conclusion, NaOH-enhanced WAO could produce a potential high-quality carbon source to aid biological treatment units in denitrification. This study could help to move the concept of resource utilization for municipal sludge through WAO closer to practical application and industrialization.

Author Contributions: K.F. and Y.T. contributed equally to this study. Conceptualization, J.Z. and G.Y.; methodology, G.Y. and L.J.; formal analysis, N.Z. and K.F.; investigation, K.F. and Y.T.; resources, Y.Z.; writing—original draft preparation, K.F. and Y.T.; writing—review and editing, G.Y. and K.F.; visualization, Y.T. and N.Z.; supervision, Y.Z. and J.Z.; project administration, L.J. and Y.Z.; funding acquisition, G.Y. All authors have read and agreed to the published version of the manuscript.

Funding: This work was financially supported by the Jiaxing Public Welfare Research Project (2024AY10039 & 2023AY11050) and the Key R&D and Achievement Transformation Project of Inner Mongolia Autonomous Region (2022YFHH0051).

Data Availability Statement: Data are available on request.

Conflicts of Interest: The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Abbreviations

AN	ammonia nitrogen
AOPs	advanced oxidation processes
BOD_5	five-day biological oxygen demand
COD	chemical oxygen demand
Em	emission wavelength
Ex	excitation wavelength
FRI	fluorescence regional integration
MS	municipal sludge
SCOD	soluble chemical oxygen demand
TCOD	total chemical oxygen demand
TN	total nitrogen
TSS	total suspended solids
VFAs	volatile fatty acids
VSS	volatile suspended solids
WAO	wet air oxidation
3D-EEMs	three-dimensional excitation emission matrices

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