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A review of the effects of ozonation process on biological sludge reduction

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ABSTRACT

The conventional activated sludge systems are commonly used in various types of wastewater treatment processes. Due to stringent environmental regulations on sludge disposal, and the high cost of conventional methods for the disposal of sludge, researches around the world have been looking for new methods to minimize sludge production in wastewater treatment plants. In this regard, the use of new processes that produce less sludge than the current processes have received much attention. This article focused on hopeful technologies, which aimed at investigating the on-site reduction of generated sludge in wastewater treatment plants. Recently, the combination of ozonation with the biological aerobic process such as activated sludge has been introduced as a strategy for reducing sludge production. However, the sludge management systems can reduce up to 60% of the total operating costs, and ozonation process reduces up to 90% of the sludge production. Finally, this review indicated that ozonation process is a promising and efficient process for zero sludge production in the wastewater treatment processes.

Keywords: Ozone; Sludge reduction; Excess sludge; Activated sludge; Zero discharge

1. Introduction

Biological treatment is one of the commonly used methods for the removal of pollutants from urban and industrial wastewaters that is based on microbial metabolism. In biological processes, principally bacteria convert dissolved and suspended solids into soluble organic, inorganic matters, carbon dioxide, water, and new cells (sludge), and a small portion of them is also decomposed to sustainable mineral compounds [1]. In recent years, excess sludge generated from biological wastewater treatment processes such as the activated sludge system has become a serious problem in the world. The sludge generation is one of the main drawbacks of the biological treatment of wastewater [2]. The excess sludge generated must be removed and treated before the final disposal or reuse. This sludge contains large amounts of volatile solids (VS) and water (95%). Because of its high solid content, the sludge disposal processes are expensive. Therefore, it is necessary to predict the volume of excess sludge generation in the design and the modeling of a wastewater treatment plant [3,4].

The activated sludge process is used to treat different types of wastewaters, and most wastewater treatment plants apply it as one of the essential processes in the wastewater treatment [4,5]. The activated sludge process consists of at least one aeration tank and a sedimentation tank. This process is considered as the initial purification step in 90% of all municipal wastewater treatment plants [6]. Clean water production is one of the advantages of this system, and its main disadvantage is associated with the excess amount of sludge

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production [7]. By overcoming the problem of excess sludge generation and its disposal, many operational problems related to sludge treatment and disposal will be solved [8]. Therefore, the excess sludge production is considered as an undesirable product in biological systems. In recent years, several mechanisms have been examined for the reduction of excess sludge produced due to the high cost of sludge treatment [9].

The supposed zero excess sludge discharge is the main target of sludge reduction. It may be difficult to achieve zero discharge in the municipal wastewater for two reasons. First, it can be attributed to the presence of inorganic and non-biodegradable solids. Second, an excessive disruption of sludge and microorganisms can reduce the amount of active biomass in the activated sludge [10]. Accordingly, the additional cost of sludge reduction needs improved techniques for wastewater treatment. In general, there are several methods to reduce excess sludge generation (Table 1):

- *Sludge reduction in the wastewater line*: in this method, the sludge reduction rate is higher than that of the final treatment, which is conducted in two ways. First, reducing yield coefficient (ozone and chemical process) and second, those are utilized in low yield (aerobic–anaerobic method).
- *Sludge reduction in the sludge line*: the purpose of this method is to reduce the amount of sludge disposal. Most studied methods increase the amount of anaerobic digestion. Some of them are the first pretreatment techniques in anaerobic reactors (e.g., mechanical decomposition, heat pretreatment, biological hydrolysis by enzymes, etc.) [11,12].
- *Sludge reduction in the final disposal sludge*: in this method, the produced sludge is converted into a stable state and the pathogens that are harmless. This method is not intended to generate sludge, but is the last treatment method for the sludge disposal. Its principle is based on energy recovery [11,12].

The use of chemical hydrolysis to reduce sludge production was first introduced by Gaudy et al. [14]. Moreover, Van Leeuwen [15] used a similar approach along with applying ozone for the sludge reduction at pilot scale. Yasui et al. [16] also developed an active sludge process via zero sludge production. A similar study was also carried out on the activated sludge process via non-real wastewater for the

Table 1
Outline of sludge reduction technologies Ramakrishna et al. [13]

Type of strategy	Technique	General method type	Sludge reduction	
Sludge reduction in the waterline	Yield reduction coefficient process	Lysis cryptic growth	Chemical oxidation	Ozonation Chlorination
			Chemical–thermal treatment	
			High-purity oxygen process	
			Enzymatic reactions	
		Maintenance metabolism	Bio membrane reactors	
		Uncoupling metabolism	Chemical unmapped	
			Oxic-settling-anaerobic process	
		Predation on bacteria	Two-stage system	
			Using worms	
	Utilization in low yield	Aerobic/anaerobic systems		
Sludge reduction in the sludge line	Pre-treatment processes before anaerobic digestion	Physical pretreatment	Cavitation	
			Thermal method	
			Mechanical method	
			Radiation	
		Chemical pretreatment	Acid and alkaline hydrolysis	
			Pre-treatment using ozone	
		Biological pretreatment	Combination of heat and non-condensing	
			Combination of chemical with thermal hydrolysis	
		Combined pretreatment		
	Moderate of anaerobic digestion process	Two-stage anaerobic digestion		
		Temperature-phased anaerobic digestion (TPAD)		
		Induced air/gas flotation separator (IGF)		
Sludge reduction in the final disposal sludge	Incineration			
	Pyrolysis and gasification			
	Wet weather oxidation (WAO)			
	Supercritical water oxidation (SCWO)			

treatment of municipal and industrial wastewaters at pilot and full scales [17].

Furthermore, the use of combined ozonation–activated sludge has been reported by several studies [18]. Sludge ozonation is known as one of the most effective technologies with the highest degradation potential [19]. Ozonation process as a novel method has been used to reduce excess sludge production in the active sludge system. This method involves a sludge ozonation step and a biological degradation step [3,20]. In the mechanism of sludge reduction, ozone causes an increase in the release of oxygen, and subsequently leads to an increase in the aerobic activity of flocs [21]. As a result, the hydrolyzed biomass in floc has aerobic degradation ability and reduces sludge production [22,23]. Ozone is a strong oxidizing agent with the oxidation–reduction potential of 2.07 V. Due to its significant oxidizing power and the presence of oxygen, as a byproduct of ozonation process, it has become a good choice for oxidation and disinfection. Ozone stops the metabolism of bacterial cells and prevents their enzymatic activity. Ozone with a sufficient concentration could break down the bacterial cell wall, and then causes bacterial death [22,24]. Given that the disintegration of sludge mass, a small amount of fine particulate and organic compounds are dissolved, and then they can be easily decomposed by microorganisms. Hence the sludge reduction reaches 100% under ideal conditions [25]. In this process, a part of the recovered sludge is transferred into the ozonation unit, and the treated sludge is degraded and delivered to the biological treatment stage [16,26].

Alternative ozonation not only decreases sludge generation by 50% to 30% but also enhances the sludge settling properties and TOC amount in the effluent [26,27]. A comparative study of ozonation and the oxic settling-anaerobic processes has reported that both methods provided considerable excess sludge reduction, while showed a low decrease in biomass respiratory activity [28].

Similarly, the application of ozonation process to reduce mass and volume of sludge in activated sludge process indicated that ozone dosage of <2 g O_3 g^{-1} TS^{-1} could reduce the amounts of solids in active sludge. The mineralization rate of 50% of VS (volatile solid) occurred in the ozone dosage of 1.5 mg g^{-1} O_3 g^{-1} TS^{-1}, which was equal to the ozone dosage required for removing the insects [29].

In this paper, we reviewed the sludge ozonation processes with a main focus on preventing or reducing sludge production. The effects of some parameters, such as ozonation, activated sludge mineralization, yield coefficient, sludge settling characteristics, economic evaluation, and SOUR were also evaluated.

2. Effects of ozonation on sludge reduction

In the combined ozonation–activated sludge process, the sludge from the secondary sedimentation tank is transferred to the ozonation unit, and after complete contact with ozone is returned to the aeration tank. Under the effect of ozone, most microorganisms are destroyed and oxidized into organic material. To attain sludge reduction and soluble COD oxidation, a high amount of ozone is required. The contact time has a lower effect on sludge reduction compared with high ozone dosage. Thus, ozonation process can be carried out at short contact times, resulting in the reduction of operational costs [30]. Ozone significantly reduces the number of dry solids and sludge volume. There are three possible pathways for the reaction of ozone with sludge: (1) oxidation of extracellular polymeric materials and the formation of small particles of sludge, (2) degradation of microbial cell membranes and increasing the release of intracellular material into the liquid phase, such as protein, lipid, and nucleic acid. Generally, there is an increase in the dissolved solids, and (3) reaction of cellular interference with activated sludge [31]. When a cell disintegrates with ozone, its contents and stored nutrients (including degradable and non-degradable materials in sludge supernatant) are released into the wastewater and cause an increase in the organic load. The organic material released from cell degradation is re-entered into the metabolism cycle of other microorganisms in the aeration tank.

The continuing process of the dissolved suspended solids and the cryptic growth leads to a reduction in the volume of sludge [12]. In this case, the ozonated sludge has a high concentration of SCOD, which is returned to the aeration tank. The bacteria present in the aeration tank use COD as a secondary source of nutrient for the growth and maintenance of cell viability. This process converts the ozonized sludge into some products such as CO_2 and N_2, and ultimately reduces net sludge production [12,32]. It seems that the ozonation process can be affected by the volatile and non-volatile properties of the sludge. This trend is the same when using specific ozone dosage (SOD) with regard to MLSS or MLVSS. Given that the cell lysis, the MLSS decreases with increasing the volume of ozonated sludge. At ozone dosage of 10 mg g^{-1} MLSS, the volume of the excess sludge reduces by 50%, and if this amount exceeds 20 mg g^{-1} of MLSS, no sludge is produced [33]. It has been also reported that under winter conditions, adding 50 mg of ozone to activated sludge systems can reduce excess sludge production [29], and by adding 125–2,000 mg g^{-1} TS ozone, the amount of VS, TSS, and TS decreased to 45.9%, 47.9%, and 80.7%, respectively [29].

Similarly, Chiellini et al. [1] showed that the concentration of TSS and VSS in the effluent of digestion tank was constant at the outlet of both digesters, while a significant improvement was observed in the summer, and may cause more organic loading in the wastewater effluent. In the mentioned study, the concentrations of TSS and VSS in the influent and effluent of digestion tank indicated the efficiency of the contact time and ozone flow rate in ozonation process [1]. The process efficiency in the reduction of TSS and VSS was initially very low; however, a significant increase was observed by increasing the ozone dosage. The ratio of the VSS/TSS in the outlet of digestion tank was reduced from 0.79 to 0.74. Ozonation can also reduce the ratio of VSS/TSS^{-1}. At ozone dosage of 0.16 g O_3 g^{-1} TS and 0.05 g O_3 g^{-1} TS, the sludge reduction was 78% and 71%, respectively, in raw sludge and end of utilization. When the maximum ozone dosage was more than 0.05 g O_3 g^{-1} TS, no sludge was observed. Moreover, 66% of MLSS was decreased via ozone dosage of 400 mL of the return sludge [34,35]. The mean ratio of MLVSS/$MLSS^{-1}$ was equal to 0.85, which represents that the ozonation process has decreased Yobs in QR. Thus, a 39% reduction in Yobs has been attained in QR within the operation time [19].

3. Effect of ozonation on mineralization sludge

The application of ozone in sludge reduction and sludge mineralization has been widely studied (Table 2). It was revealed that various amounts of dissolved organic matters and unsettlable micro-solids (UMS) were produced at low ozone dosages, while complete mineralization was observed at higher dosages. The effect of sludge ozonation is defined in the mineralization, solubilization, and variations in remaining solid features. The level of mineralization is determined by measuring the total COD (TCOD), and the amount of solubilization is determined by measuring soluble COD (SCOD). In the study of Lee et al. [19], the effects of different doses of ozone on the consumption of chemical soluble oxygen (SCOD) and total solid (TS) were studied. The solubilization depends linearly on ozone consumption as long as mineralization happens. The release of organic materials at higher ozone dosages has not been proven yet and results in low level of solubilization. The mineralization and solubilization of organic compounds increases with increasing ozone dosage, while the amount of SCOD decreases at very high ozone dosages [19]. It has been also reported that ozone kills many heterotrophic microorganisms in the reactor and oxidizes a portion of the mass, which increases the soluble COD. Since, the concentration of primary soluble COD in the untreated sludge is high, the ozone first degrades the soluble materials [3], and then causes an increase in SCOD and a reduction in TS. A further increase in ozone concentration is attributed to the sludge lysis, which may be associated with the oxidation of the released compounds [19]. In this case, ozone oxidizes soluble materials and decreases the TCOD of sludge. Therefore, organic pollutants become more easily achievable and readily available to microorganisms and have higher biodegradability [32]. In the relationship between ozone dose, DOC and TOC, as ozone concentration reaches 32 mg L^{-1}, DOC concentration increases, while TOC concentration decreases [33]. Besides, more than 20% of TOC reduces at ozone dosage of 60 mg O_3 g^{-1} MLSS. It was also reported that almost 15% of TOC before ozonation is converted into DOC at the similar ozone dosage. These findings showed that sludge ozonation can influence both stabilization and minimization [33]. In this case, the COD removal was less than 47.5% at 35 mg of ozone concentration g^{-1} MLSS in 1 L of returned biosolids in the reactor [36].

In the use of combined ultrasonic (US)-ozonation, the amount of solubilization depends on the ozone dosage. The results of a study on US process showed that a low frequency of the US is needed for the degradation of excess sludge and distribution of DOC solubilized. At the ozone dosage range 0.02–0.09 g O_3 g^{-1} TS^{-1}, the amount of organic matter is largely dissolved due to bacteria cell lysis [19]. In the US process, SCOD enhanced by increasing TS from 1.5% to 3%. Ozone penetrates into the cell wall of microorganisms and damages their cell walls. Hence, increasing DOC is due to the solubilization of intra-cellular organic materials [19].

In a study on the use of combined ozonation and hydrogen peroxide for the sludge reduction, it was found that by adding 6 mL of H_2O_2, the TSS and VSS decreased to 64% and 65%, respectively. Moreover, by adding 4 mL of H_2O_2, COD and sludge settling volume decreased to 58% and 75%, respectively [37]. In the mentioned study, the highest decrease in COD was obtained at an ozone dosage of 0.39 mg O_3 mg^{-1} TSS, and then by increasing ozone dosage and contact time, SCOD increases. According to the changes in the COD, the amount of SCOD increases due to cell lysis and the release of the intracellular organic substances. The amount of breakdown in cells is associated with contact time and ozone dosage. In another study, it was found that ozonation does not enhance SCOD in the effluent, which is consistent with the above study [37].

4. Effect of different ozone dosages on yield coefficient

In biological treatment processes, simultaneous oxidation of organic and non-organic materials, and cell growth occurs. An appropriate solution for reducing excess sludge production reduces the yield coefficient [36]. In the use of biological processes for wastewater treatment, a part of organic pollutants is consumed for respiratory activity in catabolism. The other part is absorbed and thereby generates new biological mass, which is shown with yield coefficient (Y) [38]. Y indicates the change of organic materials into biomass and is described as the biomass formed per unit of the substrate (g biomass/g substrate) or $Y = Y_x/Y_p$ [38,39]. The production of the new biomass is considered as a major drawback of activated sludge process, because the excess sludge must be safely disposed to reduce environmental impacts. The sludge

Table 2
COD removal for reducing excess sludge production

References	Wastewater type	Ozonation dosage	Sludge Reduction (%)	COD removal (%)
Kamiya and Hirotsuji [33]	Synthetic	60 mg O_3 g^{-1} MLSS	100%	15%
Yasui et al. [16]	Industry	0.05 g O_3 g^{-1} MLSS	100%	78%
Takdastan et al. [3]	Industry	20 mg g^{-1} MLSS	52%	61.4%
Torregrossa et al. [28]	Municipal	0.015 g O_3 g^{-1} TSS RAS	42%	80%
Demir and Filibeli [34]	Synthetic	0.05 g O_3 g^{-1}	40%	99.42%
Isazadeh et al. [23]	Municipal	2.3 g O_3 g^{-1} VSS	19%	50%
Lin et al. [21]	Industry	500 mg O_3 g^{-1} SS	95%	68.2%
Chen et al. [32]	Synthetic	from 0.02 to 0.3 g O_3 g^{-1} TSS	95%	54.58%
Ehsanifar et al. [37]	Municipal	0.39 mg O_3 mg^{-1} TSS	64%	75%

yield for aerobic biomass process ranges from 0.4 to 0.8 g BOD or COD (biomass)/g COD (removed) and for anaerobic process ranges from 0.02 to 0.08 COD (biomass)/g COD (removed) [33].

When an external oxidizing agent (such as ozone) is used in a part of the sludge, it kills the microbial cells and releases their substances into the bulk liquid. The released substances are then absorbed by microorganisms. Solubilization of cell products are greater than growth on the lysis products (released substrates), which leads to a reduction in the generation of cell mass. When treated sludge is returned to the aeration tank, degradation of the secondary nutrient occurs, resulting in reduced Y coefficient production. However, their mechanisms are still not well understood [40]. Decreasing yield coefficient with cell lysis is obtained through thermal, mechanical, enzymatic, or ozone treatments. However, the low yield coefficient system depends on restricting/limiting sludge growth and enhancing non-growth biomass metabolic activity [40].

Is has been reported that the Y coefficient was reduced from 0.58 to 0.28 (mg biomass mg^{-1} COD) at ozone dosage of 20 mg ozone g^{-1} MLSS in a batch reactor. In fact, excess sludge production was decreased by 52%, and at ozonation dosage of 25 mg, no sludge was observed. In the ozone concentrations over 25 mg L^{-1} min^{-1}, the Y coefficient of organic substance was decreased as a result of the effect of ozone on microorganisms [41,42]. In this case, the yield coefficient was lower than the optimal range (0.4–0.8 mg mg^{-1}), which is due to the type of substrate consumption. When the substrate is BOD, there are two reasons for reducing of yield coefficient: the lower biodegradable substrate than COD and the F/M ratio. This issue is agreed with various studies because substrate and bacterial populations affect the cellular efficiency, increase the F/M ratio, decrease MLSS concentration to 1,700 mg L^{-1}, and decrease the value of Y. Also, low Y coefficients occur under low MLSS conditions and with increasing the concentration of MLSS to 3,000 mg L^{-1}, the yield coefficients are calculated in the optimum range [42].

The amount of Y_H has been reported to be in the range 0.38–0.75 mg COD$_{cell}$ mg^{-1} COD$_{ox}$. After ozonation, the Y_H was 0.63 mg COD$_{cell}$ mg^{-1} COD$_{ox}$, although sludge treatment and ozone had the effect to reduce the heterotrophic growth yield to an average value equal to 0.54 mg COD$_{cell}$ mg^{-1} COD$_{ox.}$ The lower values for Y_H suggested that the exposure to ozone could likely favor the selection of a heterotrophic population with a lower growth efficiency because more energy is required to produce enzymes for supporting cells from high oxidation activities as well as to repair damaged cells [28]. The value of Y_H obtained using respirometry differs from the Y_{obs} values, because the observed yield coefficient is obtained based on the mass balances and can lead to less net growth. Moreover, the different behavior of biomass with real wastewater and the synthetic substrate is a possible explanation. The sludge age increases and the yield coefficient decreases over time [28]. According to the previous studies, the percentage of excess sludge reduction is not high at low ozone dosages, and this reduction increases with the change in ozone dosage per mg of MLSS. This phenomenon can be attributed to the presence of heterotrophic bacteria that have a high resistance to the ozone power, and this resistance disappears at higher concentrations [43].

In a study, the synthetic efficiency of Y in various cell retention times was also investigated. In the mentioned study, the effects of 4 COD concentrations including 300, 400, 600, and 800 mg L^{-1} were tested during 10 d of cell time in a stable state with high yield. The temperature was maintained constant in the range 20°C–22°C, and the amount of dissolved oxygen was maintained in the range 1.5–2 mg h^{-1}. To determine the biosynthetic yields, especially biomass generation (Y), the changes of COD consumed was measured during 10 d with Y = 0.51 mg biomass mg^{-1} COD without ozone. The amounts of Y with ozone concentrations of 5, and 20 mg ozone g^{-1} MLSS were 0.53 and 0.32 mg biomass mg^{-1} COD, respectively [44]. The removal efficiency of COD was 81.7% without ozone via COD = 600 mg L^{-1}, Y was 0.51 mg biomass mg^{-1} COD. However, by adding ozone, the yield coefficient reduced. In fact, by adding 15 mg ozone g^{-1} MLSS, the yield coefficient was reduced to 0.42 mg biomass mg^{-1} COD. The possible reason for the low coefficient is attributed to the consumption of ozone as a disinfectant and oxidant, and therefore many microorganisms are killed in the reactor [44].

5. Effect of ozonation on sludge settling characteristics

In the secondary wastewater treatment, organic pollutants are removed as a result of the metabolic activity of microorganisms (biomass) in the activated sludge tank. In the activated sludge process, the characteristics of the biological flocs (excess sludge) affect their settling quality. Sludge volume index (SVI) is the most common parameter that is used to determine the settling characteristics of activated sludge solids with suspended growth and function of sludge handling equipment in specific thickeners. Settling properties are defined as the mass flow of MLSS m^{-2} in the clarifier tank. This parameter is used to determine the sludge recycle rate. Ozonation can improve sludge settling by decreasing the presence of filamentous bacteria in the microbial communities. A considerable enhance in the concentration of activated sludge in the secondary settling tank was the main cause of poor sludge settling. Ozone can be used to solve the difficulty of poor settling in the return activated sludge line [45]. SVI is a characteristic that helps operators recognize the sludge settling ability in the treatment plant and is used for bulking and sporadic growth phenomena. This index ranging 80–120 indicates the ability of good sludge settling, and the values below 100 represent ideal conditions of sludge settling. The values below 80 indicate the beginning of scattered growth and the absence of flocs, which is one of the problems of activated sludge systems. The values greater than 120 indicate poor settling conditions and the occurrence of the bulking phenomenon [46].

It is necessary to understand the specific effect of ozone on different components of mixed liquor suspended solids (MLSS). A very low dosage of ozone is needed to improve the oxidation process, but extra oxygen is necessary for the sludge stabilization. At low dosages, ozone destroys the cell wall of bacteria in the flocs and makes them disappear. At higher dosages, ozone is considered as the cause of flocs destruction, which directly affects the bacterial extracellular polymeric matrix and plays an important role in the accumulation of bacteria. In addition, ozone helps oxidize the dissolved organic material, and subsequently improves sludge settling [1].

Ozone covers the surface of filamentous bacteria, degrades them and changes their structure [33]. Under ideal conditions, a layer of biosolids and supernatant are formed on top of the sludge. The supernatant can be returned to the treatment plant, while biosolids can be discharged or used for different purposes if its solid content is adequately stabilized [29]. Previous studies demonstrated that the ozonation process has a positive impact on sludge reduction and improves the settling characteristics or inactivates bacteria [47]. Improving the sludge settling ability causes a reduction in the bonded water and filamentous bacteria. The amount of bonded water has a key effect in dehydration and volume reduction. Ozonation process decreases the banded water and the mass of microorganisms, because of releasing the bonded water of cells and flocs [29]. After ozonation, the rate of sludge settling increases and the amount of SVI reduces. The use of ozone leads to a decrease in the SVI index, which can cause of sporadic growth. Under sludge balking conditions, limited availability to organic matters in low loading conditions causes an increase in the number of filamentous bacteria, and subsequently increases the SVI index, while the reduction of sludge settling has been observed under such conditions [48].

In a study, the effect of continuous ozonation with a concentration of 21.4 mg O_3 g^{-1} MLSS d^{-1} was studied on SVI. In the control step, a fast enhancement was observed in the SVI and the atrophy of sludge settling happened. In the sludge ozonation step, the increase of SVI was extremely small, and good settling properties remained constant [33]. The microscopic examination showed that despite the low density of sludge in the ozonation step, filamentous bacteria were present in the sludge. The dependent SVI is defined as the value of SVI after ozonation. By increasing ozone dosage, relative SVI decreased. The ozonation gradually improved the settling properties of the sludge, especially in a concentration less than 1 g O_3 g^{-1}. In the ozone dosages more than 2 g O_3 g^{-1} TS, the number of settleable solids decreased to 110 g O_3 g^{-1} TS. Also, the improvement of sludge settling can be attributed to the ozone characteristics [29]. There is no adequate data regarding ozone concentration range 2 to 8 mg O_3 g^{-1} MLSS. This can result in up to 60% decrease in SVI at the ozone concentration of 9.5 mg O_3 g^{-1} MLSS, and thus sludge settling properties improved. In the ozone dosage of 0.05 g O_3 g^{-1} TS, the SVI decreased to 82 mL mg^{-1} MLSS [29].

Previous studies also reported the same results for the effect of the ozone on settling properties of sludge. The sludge reduction was very low at the low ozone dosages. The maximum sludge settling formed in the SOD of 12.5 kg MLSS m^{-2} and sludge reduction reached 64% [6]. A 40% drop in the SVI rate was observed at ozone dosage of 20–60 mg g^{-1} SS. When TOD reached 10 and 10–20 mg g^{-1} SS, the amount of SVI increased to 2%–45% and 14%–49%, respectively [49]. The ozone acted as a possible oxidant for the filamentous bacteria. The results from the several study are presented in Table 3.

6. Effect of the ozonation process on SOUR

The oxygen uptake rate (OUR) is a characteristic of sludge activity and represents the volumetric oxygen conception for biological degradation [50]. It is also used for the toxicity of different chemical factors. Ozonation can be used for the pretreatment of biodegradable materials. Specific oxygen uptake rate (SOUR) is known as the rate of oxygen absorbed per unit mass of volatile suspended solids and also defined as a measure of biomass respiration. The SOUR is defined as a specific OUR (SOUR; mg O_2 g^{-1} VSS h^{-1}), which explains specific bioactivity [51]. The average SOUR for the reference system is 50 mg O_2 g^{-1} VSS h^{-1}, which is the natural range of SOUR for an active sludge process (40–70 mg O_2 g^{-1} VSS h^{-1}). As long as there is enough oxygen, SOUR depends on the availability of degradable carbon and the existence or lack of competing compounds. High concentrations of dissolved oxygen produce more active biomass, and therefore generate lower amount of activated sludge. Because the increase in the amount of dissolved oxygen in the reactor increases the oxygen permeation rate, and thereby a larger portion of the biomass is hydrolyzed aerobically, resulting in a decrease in sludge volume. When ozone is contacted with sludge, some microbes are inactivated and extinguished (exception for a limited number of encapsulated microorganisms), and the microbial activity

Table 3
Compare the results of the same research

References	SRT (d)	Ozonation dosage	SVI (mg L^{-1})	Sludge Reduction	SVI Reduction
Kamiya and Hirotsuji [33]	20	21.4 mg O_3 g^{-1} MLSS d^{-1}	250–300	100%	60%
Yasui et al. [16]	51	0.05 g O_3 g^{-1} MLSS	–	94.4%	30%
Wei et al. [26]	28	0.05 g O_3 g^{-1} SS	60	60%	–
Zhao et al. [54]	–	0.02–0.06 g O_3 g^{-1} MLSS	60	80%	53%
Takdastan et al. [3]	10	25 mg O_3 g^{-1} MLSS	0	100%	100%
Torregrossa et al. [28]	100	0.02 g O_3 g^{-1} TSS	25	42%	90%
Takdastan and Eslami [41]	10	–	13	56%	72%
Demir and Filibeli [34]	30	0.05 g O_3 g^{-1} TS	82	73%	17%
Muz et al. [60]	4	1.27 mg O_3 g^{-1} MLSS	82.7	84%	53%
Nilsson [17]	45	2.8–5.0 g O_3 kg^{-1} SS	170	80%	41%
Marce et al. [49]	6	0.03 g O_3 g^{-1} SS	–	24%	40%

decreases, which causes in a decrease in SOUR (an indicator of bacteria breathe) [51].

In this regard, a study on the use of ozone as a powerful cell lyses factor showed a decrease in the microbial population in the reactor. When the live microbial population decreased, the $SOUR_{end}$ decreased dramatically. This may be attributed to the oxygen concentration that is used by the cells for storing purposes [52]. Endogenous breathing explains the cell metabolism in the absence of the outer substrate. The enhancement in the endogenous phase is due to living microorganisms which consume lysed sludge as a substrate, and consequently increase $SOUR_{end}$ [52]. In this case, the primary OUR_{end} varied in the range 8–12.3 mg O_2 g $MLVSS^{-1}$ h^{-1}. As the ozone was used, the measured $SOUR_{end}$ without adding external material enhanced up to 1.8 times compared with primary OUR_{end} [52].

SOUR lower than 12 mg oxygen h^{-1} g of suspended solid represents a toxic substance in the reactor. 20 mg ozone g^{-1} MLSS in 1 L circulated sludge in the reactor can decrease SOUR to 3 mg O_2 h^{-1} g VSS [41]. In the ozone dosage of 1,476 mg L^{-1}, the amount of SOUR was enhanced from 9 to 35.1 mg O_2 g^{-1} MLSS h, respectively [47]. However, increasing ozonation times of higher than 30 min led a negligible decrease in the BOD_5 rate. In other words, SOUR did not show considerable variations after 30 min of ozonation [47]. Enhancement in SCOD increased OUR rate in the range of 0.05–0.07 g O_3 g^{-1} SS. By increasing ozone dosage to more than 20 min the microbial respiratory activity decreased due to a smaller amount of organic solvents. In this ozone rate, dosage has more impact than contact time, and OUR was quickly reduced at contact times over 20 min [30]. After about 3 months, the COD and VSS reached less than 42% and the high rate of stabilization occurred. At this time, live microbes in the endogenous phase were significantly reduced, which was related to low SOUR. The SOUR value was 21 ± 3 mg O_2 g^{-1} VSS h^{-1} [53,54].

Another study claimed that the amount of SOUR was relatively low. In this case, it may be attributed to the repressive impact of phenol on respirometric oxygen usage. The preventive function of phenol biodegradation is associated with the hydrophobic disorder of microbial membrane. Moreover, the SOUR rate indicated an exponential enhancement in the operation period until the maximum rate was achieved. In effect, the maximum SOUR rate was obtained at nearly 10 h of breathing period for 500 mg L^{-1} phenol. Therefore, it can be concluded that the time before reaching the higher rate was related to phenol dosage. There are two probably reasons to justify this phenomenon: first, a specific rate of active biomass was reduced and the SOUR was negative. Thus, since the phenol was available for biodegradation, the SOUR enhanced. The second possible cause is that as the mechanism of biomass deceased, the SOUR rate reduced. Further research is needed to confirm this assumption [55].

7. Economic evaluation

Economic efficiency and energy rate are significant tools that can be used for a cost–benefit analysis of minimization in a sludge generation system. To produce 1 kg of ozone gas, the energy consumption is about 15 Yuan = US $1.92. The operating expense of sludge ozonation was estimated with flow rate q = 0.0067 Q, OD = 0.05 kg O_3 kg^{-1} MLSS, and ozone yield of 95% as a sample, and then this cost is calculated as follows:

15 Yuan kg O_3 × 8.168 kg MLSS m^{-3} × 0.05 kg O_3 kg^{-1} MLSS 95% = 6.13 Yuan (US $0.79) m^{-3} sludge

The extra ozonation expense for wastewater treatment is as follows:

6.13 Yuan m^3 sludge × (0.0067 Q sludge Q wastewater) = 0.041 Yuan (US $0.0053) m^{-3} wastewater [22]

The main disadvantage of ozonation was the high costs of repair, maintenance, and operation. The operation cost for ozone included ozone concentration, treated sludge rate, etc. As expected, the capital and operating costs for activated sludge–ozonation systems is high due to high energy consumption for ozone production, while by considering the costs of dewatering and disposal sludge, it will be much more affordable than the conventional activated sludge system [56]. The activated sludge–ozonation process is considered as one of the most productive and useful technologies for reducing excess sludge production and improving sludge extraction, but it has still several unsolved problems. For example, ozone is not a selective oxidatant, so that it can react with any reductant material in the environment, and this may reduce its efficiency for the oxidation of excess sludge. On the other hand, ozone in reaction with low degradability compounds may produce toxic compounds at the outflow of wastewater. Moreover, the high-speed ozone consumption, which sometimes reached 30 mg ozone g^{-1} VSS, is another drawback of the combined ozonation–activated sludge process [33].

In a laboratory-scale research, it has been predicted that the cost of installing an ozone generator was 3.75 EUR or 4.20 USD per kg of O_3 generated (1 EUR USD as of 2016). The sum cost of sludge ozonation was comparable with the conventional CAS utilization. The sludge ozonation cost of 538 USD $tonne^{-1}$ DS for ozone generation and 56 USD $tonne^{-1}$ DS for excess aeration, which the total cost was about 594 USD $tonne^{-1}$ DS [25]. A similar rate was observed in CAS for excess sludge dewatering (202 USD $tonne^{-1}$ DS), transporting (56 USD $tonne^{-1}$ DS), and incineration (280 USD $tonne^{-1}$ DS) [25]. A full-scale ozonation unit consumes 14% of the total energy use of the plant [6]. In this case, the real cost of ozonation was 0.05 EUR or 0.06 USD per kg of O_3; 1 EUR = 1.12 USD as of 2016, which seems to be considerably lower than that reported by Huysmans et al. [57]. This difference can be due to the change in the technical features including ozone concentration, and generator lay out for annual production of 3,276 kg O_3, which was used to decrease sludge by 10%, the total cost of the ozonation process was found to be 6,605 USD $year^{-1}$ [6].

Applying the life-cycle index of sludge treatment and sludge disposal showed that the annual ozonation expense could be simply offset with economy sludge dewatering. Additional saving can be achieved by decreasing in sludge drying, digestion, transportation, and other downstream systems [58]. Besides, ozone-related costs could be reduced by preforming the ozonation process under optimum conditions. For instance, the increase of ozone concentrations in aerobic digester was 1.23 to 1.40 g O_3 kg^{-1} TSS and caused a nominal decrease in the rate of sludge disposal. However, the operating expense during the process was enhanced [59]. The real cost of ozonation increased with increasing energy consumption for wastewater quality (e.g., extra aeration in CAS to degrade the excess COD in ozonated sludge) [57,60]. Despite the additional costs, several studies have shown that ozonation is economically feasible because it can decrease expenses related to the downstream system (e.g., dewatering and emanation) as well as the excess sludge produced [25,61].

8. Conclusion

This study showed that ozonation is an effective method in excess sludge reduction because of its advantages such as being compact, clean, and powerful. In this method, the sludge reduction rate can reach 100%. In addition to the sludge reduction, it has a positive impact on solubility, dewatering, and filtration. This can be successfully applied on a pilot and full scale. Ozone decreases the sludge volume by disintegrating return sludge as the largest portion of the final sludge and increases its settling rate. After ozonation, the release of the bonded water in cells or flocs is responsible for decreasing of banded water and the increase of solid content of dewatered cake. Moreover, the minimization of sludge volume is helpful to decrease the expense of sludge handling. As a result, the combined ozonation–activated sludge processes can be effectively applied for decreasing excess sludge generation and improving sludge settleability.

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