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Evaluation of the dewaterability, heavy metal toxicity and phytotoxicity of sewage sludge in different advanced oxidation processes

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ABSTRACT

This study thoroughly evaluated six types of sludge dewatering processes in terms of sludge dewaterability, environmental risk of heavy metal release and phytotoxicity. The assessed sludge dewatering processes included two conventional coagulants (polyacrylamide (PAM); $FeCl_3$/CaO) and four advanced oxidation processes (AOPs) (Fenton's reagent/CaO; Fe^{2+}/$Na_2S_2O_8$/CaO; $Ca(ClO)_2$/$FeCl_3$/walnut shell (WS); thermal and acid-washed zero-valent iron (TA-ZVI)/H_2O_2/CaO). Compared to the conventional coagulants, the use of AOPs as conditioners presents greater potential for sludge dewatering, as AOPs have dual roles in alleviating the environmental risk of heavy metals and the phytotoxicity of dewatered sludge. Among the four tested AOPs, Fenton's reagent/CaO treatment achieved the highest sludge dewaterability improvement, with a higher water removal rate, lower specific resistance of filtration (SRF) and lower water content of dewatered sludge (Wc). This treatment was also the most cost-effective among the four AOPs. However, this method could not substantially mitigate the environmental risk due to the leaching toxicity of heavy metals and the phytotoxicity of dewatered sludge. Conversely, when considering the reduction of environmental risk, $Ca(ClO)_2$/$FeCl_3$/WS could be a better option, although its operational cost was slightly higher than that Fenton's reagent/CaO. Collectively, this study provides novel insights for the selection of sludge conditioning treatments.

1. Introduction

Sludge treatment and disposal is a major challenge affecting the operation of wastewater treatment plants (WWTPs) (Świerczek et al., 2018; Zhang et al., 2018). Sludge dewatering is an efficient strategy to reduce sludge volume and lessen the burden of subsequent sludge disposal (Brix, 2017; Li et al., 2019). Chemical conditioning is a typical and effective method of sludge treatment (Zahedi et al., 2017). Among the commonly used chemical conditioning methods, the application of conventional coagulations (Wei et al., 2018) and advanced oxidation processes (AOPs) (He et al., 2015; Maqbool et al., 2019) have received increasing attention. Polyacrylamide (PAM) and $FeCl_3$/CaO were the two commonly used conventional coagulants, however, they could not achieve satisfactory sludge dewaterability (Yu et al., 2016). This was mainly due to the fact that PAM could not apparently change the viscous structure of sludge (Ho et al., 2010), while $FeCl_3$/CaO could not efficiently destroy sludge structure (Ren et al., 2015). In recent years, AOPs including Fenton oxidation (Yu et al., 2016), Fenton-like

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oxidation (He et al., 2015), persulfate oxidation (Shi et al., 2015; Maqbool et al., 2019) and chlorine oxidation (Liang et al., 2019; Yu et al., 2019) have been developed and demonstrated to effectively improve sludge dewaterability by disrupting floc structures and removing extracellular polymeric substances (EPS).

Many studies have been performed to compare different AOP pretreatments on sludge dewaterability, but it still remains unclear which AOPs effectively improve sludge dewaterability due to the contradictory reported outcomes and conclusions. For example, some studies have demonstrated that persulfate oxidation produced superior sludge dewatering effects than Fenton oxidation and Fenton-like oxidation (Li et al., 2016; Song et al., 2016b). However, other studies have illustrated that Fenton oxidation and Fenton-like oxidation achieved better sludge dewaterability than persulfate oxidation (Song et al., 2016a; Erkan, 2019). Additionally, Zhu et al. (2018) observed that chlorine oxidation treatment exhibited better sludge dewaterability than Fenton oxidation and Fenton-like oxidation, while a study by Yu et al. (2019) showed contradictory results. Accordingly, the findings about which AOP could achieve greater sludge dewatering are still not conclusive. This may be attributed to differences in sludge characteristics or sludge sources, such as different WWTPs in different regions (Skinner et al., 2015) and different mechanical dewatering devices (e.g. centrifuges and filter press) (Pan et al., 2003). The inconsistent conclusion about sludge dewaterability may occur even though an AOP was employed to treat the sludge with different characteristics. The similar phenomenon could be observed when using different dewatering devices to treat the identical sludge (Pan et al., 2003; Ward et al., 2019). Thus, to obtain the representative and convincing result, it is indeed necessary to perform a direct comparison of AOP effectiveness in a single experiment under the same types of sludge and mechanical dewatering device.

Additionally, as a solid waste, dewatered sludge still requires appropriate disposal (Soobhany et al., 2015). The agricultural application of dewatered sludge has been predominantly adopted for sludge disposal, as the sludge contains a high content of valuable nutrients, such as organic matters, N, P and K (Kominko et al., 2019). However, dewatered sludge also contains a large amount of heavy metals (such as As, Cd, Cr, Cu, Pb and Zn), posing a potential risk to surrounding environments and human health (Vogel and Adam, 2011). Currently, many countries have drawn up a strict regulation on the limit of heavy metal contents in the dewatered sludge before its reuse (Chanaka Udayanga et al., 2018). Therefore, except for sludge dewatering, the environmental risk of heavy metals in dewatered sludge should be assessed before its disposal. The environmental risk of heavy metals in dewatered sludge was usually evaluated by the leaching toxicity and the chemical species distribution of heavy metals (Xiong et al., 2018). Reduction of the leaching toxicity and unstable fractions of heavy metals might be beneficial for mitigating the environmental risk of heavy metals (Dai et al., 2019). Moreover, it also remains unclear whether the use of AOPs effectively reduces the environmental risk posed by heavy metals in dewatered sludge. For example, some studies reported that persulfate oxidation could effectively mitigate environmental risk of heavy metals in the sludge through the release of organics matters and unstable heavy metals (Xiong et al., 2018; Liu, 2019), while it was reported the Fenton treatment did not cause obvious reduction in the environmental risk from heavy metals in the sludge (He et al., 2017). It is likely that the different characteristics of sludges assessed in different studies could result in such contradictory conclusions. Therefore, it is deemed necessary to investigate different AOPs for the reduction of environmental risk from heavy metals in a single direct comparison experiment. Phytotoxicity is also a crucial factor forecasting the feasibility of agricultural use (Kebibeche et al., 2019; Zou et al., 2019). Zou et al. (2019) found that untreated dewatered sludge induced a negative effect on plant growth. As such, the phytotoxicity of dewatered sludge should be evaluated before subsequent disposal.

In this study, two conventional coagulants (PAM and $FeCl_3$/CaO) and four representative AOPs (Fenton's reagent/CaO; Fe^{2+}/$Na_2S_2O_8$/CaO; $Ca(ClO)_2$/$FeCl_3$/walnut shell (WS); and thermal and acid-washed zero-valent iron (TA-ZVI)/H_2O_2/CaO) were assessed. This study systematically evaluated the sludge dewaterability, the sludge physicochemical characteristics, the heavy metals environmental risk, and the phytotoxicity of the conventional coagulants and the AOPs in a single experiment using the identical types of sludge and mechanical dewatering device. The aim of this study was to (a) compare the sludge dewatering efficiency by conventional coagulants and AOPs; (b) explore the changes in organic components (represented by EPS) in the raw and treated sludge; (c) evaluate the heavy metals toxicity and phytotoxicity of dewatered sludge; (d) assess the operational costs.

2. Materials and methods

2.1. Sludge sample preparation

Sewage sludge samples with water content of 95.91 wt% were collected from a domestic WWTP in Guangzhou, China. The sludge samples were treated using two conventional coagulants (PAM and $FeCl_3$/CaO) and four representative AOPs (Fenton's reagent/CaO; Fe^{2+}/$Na_2S_2O_8$/CaO; $Ca(ClO)_2$/$FeCl_3$/WS; and TA-ZVI/H_2O_2/CaO). The detailed conditioning and dosing processes for these samples are described in Table 1. Based on the pre-tests and the researches of Liu et al. (2016) and Wang et al. (2019), the PAM and $FeCl_3$/CaO treatments were set at a fixed amount of 20 mg/g dry solids (DS) PAM and at the fixed dosages of 80 mg/g DS $FeCl_3$ and 140 mg/g DS CaO, respectively. Fenton's reagent/CaO, Fe^{2+}/$Na_2S_2O_8$/CaO, $Ca(ClO)_2$/$FeCl_3$/WS, and TA-ZVI/H_2O_2/CaO were chosen as the representative AOPs to condition sludge due to its respective advantages and characteristics. For Fenton's reagent/CaO, it was an efficient strategy to achieve sludge deep dewatering (Yu et al., 2016). Respect to Fe^{2+}/$Na_2S_2O_8$/CaO, it was considered as a feasible method for enhancing the sludge dewaterability at inherent pH (Shi et al., 2015). $Ca(ClO)_2$/$FeCl_3$/WS was a novel conditioning process involving oxidation, re-flocculation, and skeleton construction to simultaneously remove EPS and bound water (Liang et al., 2019). TA-ZVI/H_2O_2/CaO was a recyclable and inexpensive method for the sludge dewaterability enhancement (Liang et al., 2020). The dosages of Fenton's reagent/CaO and Fe^{2+}/$Na_2S_2O_8$/CaO were applied according to the pre-tests and the studies of Liang et al. (2015) and Shi et al. (2015). The dosages of $Ca(ClO)_2$/$FeCl_3$/WS and TA-ZVI/H_2O_2/CaO were applied based on the pre-tests and the studies of Liang et al. (2019) and Liang et al. (2020). It should be noted that the optimization of the operational conditions for each treatment would not be further discussed in this study because this study focused on the comprehensive comparison of the sludge dewaterability, the sludge physicochemical characteristics, and the toxicity using the conventional coagulants and the AOPs in a single experiment. After treatment, the conditioned sludge was collected to analyze the specific resistance of filtration (SRF), EPS, zeta potential, and particle size.

The conditioned sludge was dewatered using an ultrahigh filter press (6 MPa) developed by Liang et al. (2019). After dewatering, the dewatered sludge and the filtrate were collected. The dewatered sludge samples were freeze-dried for 24 h at −60 °C, then ground and passed through a 150 μm sieve. The freeze-dried samples were used to analyze the transformation of heavy metals and phytotoxicity. The main characteristics of freeze-dried samples are outlined in Table 2. The filtrate was used to measure the

Table 1
Conditioning and dosing procedures of different sludge samples.

Batch test No.	Pretreatment methods	Conditioning and dosing procedures
1	PAM	20 mg/g DS PAM (10 min, 300 rpm)
2	$FeCl_3$/CaO	80 mg/g DS $FeCl_3$ (3 min, 300 rpm) → 140 mg/g DS CaO (10 min, 300 rpm)
3	Fenton's reagent/ CaO	20 mg/g DS H_2SO_4 (1 min, 300 rpm) → 50 mg/g DS Fe^{2+} (3 min, 300 rpm) → 30 mg/g DS H_2O_2 (15 min, 300 rpm) → 50 mg/g DS CaO (3 min, 300 rpm)
4	$Fe^{2+}/Na_2S_2O_8$/CaO	23.5 mg/g DS Fe^{2+} (3 min, 300 rpm) → 100 mg/g DS $Na_2S_2O_8$ (15 min, 300 rpm) → 50 mg/g DS CaO (3 min, 300 rpm)
5	$Ca(ClO)_2/FeCl_3$/WS	20 mg/g DS H_2SO_4 (1 min, 300 rpm) → 10 mg/g DS $Ca(ClO)_2$ (10 min, 300 rpm) → 60 mg/g DS $FeCl_3$ (10 min, 300 rpm) → 600 mg/g DS WS (3 min, 300 rpm)
6	TA-ZVI/H_2O_2/CaO [a]	20 mg/g DS H_2SO_4 (1 min, 300 rpm) → 84 mg/g DS TA-ZVI and 20.4 mg/g DS H_2O_2 (10 min, 300 rpm) → 50 mg/g DS CaO (3 min, 300 rpm)

Note.
[a] TA pretreatment conditions were TA temperature of 40 °C, TA reaction time of 20 min; ZVI was reused for 5 times.

Table 2
Main characteristics of freeze-dried sludge samples.

Sludge samples	C (%)	N (%)	P (%)	K (%)
Raw sludge	20.82 ± 0.11	3.64 ± 0.05	2.59 ± 0.09	0.34 ± 0.01
PAM	20.79 ± 0.16	3.61 ± 0.08	2.30 ± 0.07	0.32 ± 0.05
$FeCl_3$/CaO	19.61 ± 0.27	3.27 ± 0.02	2.46 ± 0.05	0.26 ± 0.08
Fenton's reagent/CaO	20.10 ± 0.19	3.50 ± 0.16	2.59 ± 0.03	0.17 ± 0.05
$Fe^{2+}/Na_2S_2O_8$/CaO	20.04 ± 0.26	3.53 ± 0.09	2.38 ± 0.16	0.21 ± 0.03
$Ca(ClO)_2/FeCl_3$/WS	29.02 ± 0.10	2.62 ± 0.12	1.96 ± 0.06	0.30 ± 0.11
TA-ZVI/H_2O_2/CaO	18.37 ± 0.15	3.22 ± 0.17	2.27 ± 0.01	0.15 ± 0.02

concentrations of heavy metals and total organic carbon (TOC).

2.2. Phytotoxicity tests

Phytotoxicity tests were conducted in triplicate with the plant species *Brassica rapa chinensis* (Chinese cabbage) and *Lactuca sativa* (Lettuce) (Tiquia and Tam, 1998) following the Chinese standard GB/T 23486 method and the method reported by Selim et al. (2012). The freeze-dried sludge was mixed with deionized water at a solid/liquid ratio of 1:10 and then shaken at 110 rpm for 8 h. The extracts were then filtered through a 0.45 μm membrane prior to tests. For each treatment, 30 seeds of *Brassica rapa chinensis* and *Lactuca sativa* were placed in two respective 90 mm plastic petri dishes with the filter paper at the bottom, followed by the addition of 5 mL sludge extraction solutions. Finally, these plastic petri dishes were covered with the filter paper to minimize loss from evaporation. All these plastic petri dishes were incubated at 25 °C for 72 h in the dark, with deionized water used as the control. After incubation, the percentage of seeds germination and root length of the plant species were determined. The germination index (GI) was calculated based on **Equation (1)** as follows:

$$GI\ (\%) = \frac{Seeds\ germination\ in\ samples}{Seeds\ germination\ in\ control} \times \frac{Mean\ root\ length\ in\ samples}{Mean\ root\ length\ in\ control} \times 100 \qquad (1)$$

2.3. Analytical methods

2.3.1. Sludge characteristics

The sludge dewatering efficiency was evaluated based on the water removal rate, water content of dewatered sludge (Wc) and SRF. The water removal rate was calculated according to Liang et al. (2019). The Wc was calculated following the standard methods of APHA (1998). The SRF was determined as per Qi et al. (2011). Three EPS fractions (Soluble EPS (S-EPS), loosely bound EPS (LB-EPS) and tightly bound EPS (TB-EPS)) were extracted from sludge using a modified heat method as described by Yu et al. (2008). The protein and polysaccharide concentrations in liquid S-EPS, LB-EPS and TB-EPS samples were determined after filtration through a Millipore 0.45 μm filter membrane. Briefly, the proteins were analyzed using the Coomassie Brilliant Blue G-250 method (Bradford, 1976), while polysaccharides were analyzed using the modified anthrone method described by Frolund et al. (1996). The particle size of sludge was measured using a Mastersizer 3000 instrument (Malvern, UK). The zeta potential of sludge was analyzed using a 90Plus PALS instrument (Brookhaven, USA).

2.3.2. Metals analysis

Based on the control standard of pollutants in sludge for agricultural use (GB 4284-2018, China) and the study reported by Dai et al. (2019), six heavy metals (As, Cd, Cr, Cu, Pb, and Zn) were analyzed. The total contents of heavy metals in the dewatered sludge were extracted using the microwave-assisted leaching method as described by Link et al. (1998). The toxicity of heavy metals in dewatered sludge were determined using a toxicity characteristic leaching procedure (TCLP) (Li et al., 2019). Detailed protocols about leaching experiments are described by Tsang et al. (2013). The chemical speciation of heavy metals in dewatered sludge were analyzed according to the modified Community Bureau of Reference (BCR) method (Ye et al., 2017). The detailed BCR extraction procedures are shown in Table S1. After extraction, the heavy metal species were divided into four fractions, the exchangeable and weak acid soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3) and residual fraction (F4). The environment risk of heavy metals in dewatered sludge was evaluated via the risk assessment code (RAC), based on the proportion of F1 concentration (Xiong et al., 2018). The environment risk was classified as no risk (risk index (%) < 1), low risk (1 < risk index (%) < 10), medium risk (11 < risk index (%) < 30), high risk (31 < risk index (%) < 50) and very high risk (risk index (%) > 50) (Liu, 2019). The heavy metal contents of the dewatered sludge, the liquid phase, the leachate, and the extracting solution samples were analyzed using a Prodigy 7 Inductively Coupled Plasma Optic Emission Spectrometer (ICP-OES) (Leeman Labs, USA).

2.3.3. Other analysis

The C and N contents in the dewatered sludge samples were analyzed using an organic elemental analyzer (Vario Macro, Germany). The TOC of the filtrate was analyzed using a TOC-VCPH analyzer (Shimadzu, Japan). The K concentrations were determined using a 7500 ICP-OES (Leeman Labs, USA) and the P concentrations were analyzed using the molybdate blue colorimetric method described by Kara et al. (1997). The pH and oxidation-reduction potential (ORP) were determined using a STARTER 2100 pH analyzer (Ohaus, USA) and an ORP electrode with an Ag/AgCl reference (STORP1, Ohaus, USA), respectively.

3. Results and discussion

3.1. Sludge dewaterability

The water removal rate of treated sludge was increased from 53.12% in raw sludge to 80.45% in PAM treated sludge, 87.21% in $FeCl_3$/CaO treated sludge, 94.15% in Fenton's reagent/CaO treated sludge, 92.46% in $Fe^{2+}/Na_2S_2O_8$/CaO treated sludge, 88.88% in $Ca(ClO)_2/FeCl_3$/WS treated sludge and 91.46% in TA-ZVI/H_2O_2/CaO treated sludge (Fig. 1a). These results illustrated that the AOPs achieved higher water removal rate in sludge than conventional coagulants, which could be due to the disintegration of sludge flocs and the breakdown of EPS after treatment with AOPs (Xiao et al., 2017). The water removal rate of sludge treated by AOPs followed the order of Fenton's reagent/CaO > $Fe^{2+}/Na_2S_2O_8$/CaO > TA-ZVI/H_2O_2/CaO > $Ca(ClO)_2/FeCl_3$/WS (Fig. 1a), indicating that the Fenton oxidation exhibited the highest water removal rate in sludge compared to the other three AOPs. It may be attributed to the strong flocculation and oxidation capacity of Fenton oxidation process (Song et al., 2016a; Erkan, 2019), which was supported by the higher ⊿ORP and filtrate TOC results in the present study (Fig. S1).

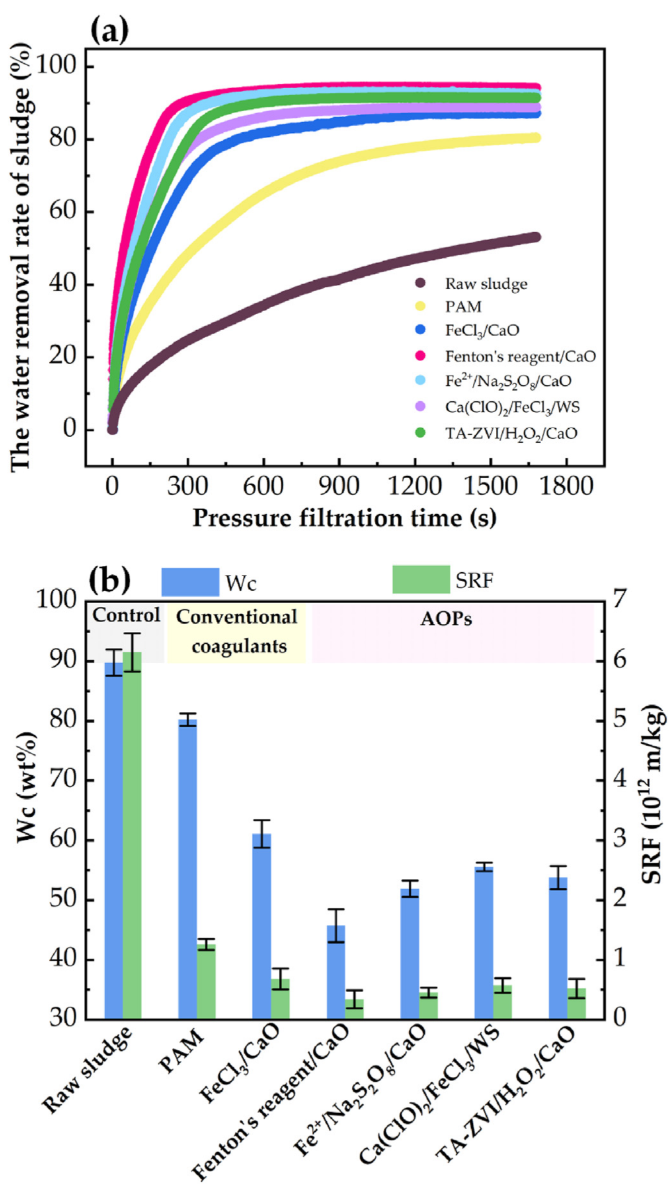

Fig. 1. Sludge dewaterability of the conventional coagulants and the AOPs treatments: (a) The water removal rate of sludge, and (b) Wc and SRF. Abbreviation: TA = thermal-acid-washed pretreatment; ZVI = zero-valent iron.

Similarly to the trend in the water removal rate of sludge, the Wc values were 80.22 wt% for PAM, 61.08 wt% for $FeCl_3$/CaO, 45.73 wt% for Fenton's reagent/CaO, 51.90 wt% for $Fe^{2+}/Na_2S_2O_8$/CaO, 55.56 wt% for $Ca(ClO)_2/FeCl_3$/WS and 53.78 wt% for TA-ZVI/H_2O_2/CaO (Fig. 1b), indicating that the AOPs achieved a lower Wc than the conventional coagulants. These results showed that among the AOPs, the Wc value was lowest with the Fenton's reagent/CaO treatment, followed by an increasing order of $Fe^{2+}/Na_2S_2O_8$/CaO, TA-ZVI/H_2O_2/CaO and $Ca(ClO)_2/FeCl_3$/WS. Therefore, Fenton's reagent/CaO treatment exhibited a greater sludge dewatering efficiency (Wc of 45.73 wt%) than other AOPs, allowing effective sludge deep dewatering. This Wc value was far below Chinese standard limit for sludge landfilling (Wc < 60.00 wt%, GB/T 23485-2009).

The SRF values of the sludge conditioned using PAM, $FeCl_3$/CaO, Fenton's reagent/CaO, $Fe^{2+}/Na_2S_2O_8$/CaO, $Ca(ClO)_2/FeCl_3$/WS and TA-ZVI/H_2O_2/CaO decreased by 79.51%, 88.94%, 94.47%, 92.68%, 90.73% and 91.54%, respectively (Fig. 1b), compared to the raw sludge (6.14×10^{12} m/kg). This indicated that the AOPs enhanced sludge dewaterability to a greater degree than conventional coagulants. Furthermore, the sludge dewaterability with Fenton's reagent/CaO treatment exceeded the dewaterability with other AOPs. These results are consistent with the results of water removal rate and Wc tests (Fig. 1a and b).

3.2. Sludge fertilizing properties and physicochemical characteristics

3.2.1. Sludge fertilizing properties

Sludge fertilizing property preservation was evaluated using the nutrient elements including the total C, N, P and K (Table 2). As illustrated in Table 2, compared to the raw sludge, the contents of total C, N, P and K in dewatered sludge slightly decreased after treatment, indicating that the great mass of the nutrients were hold in the dewatered sludge. Moreover, the result showed that the total C content in dewatered sludge were highest after treatment with $Ca(ClO)_2/FeCl_3$/WS, which might be due to the carbon source released from the biomass (WS) (Liang et al., 2019).

3.2.2. EPS

The special structure of EPS in sludge plays an important role in sludge dewaterability and the release of organic matter and heavy metals from stable layers (Dai et al., 2019). To understand the changes in EPS of sludge treated using conventional coagulants and AOPs, the protein and polysaccharide concentrations were determined.

The protein concentrations of raw sludge in the S-, LB- and TB-EPS fractions were 10.72, 13.57 and 67.59 mg/g DS, respectively (Fig. 2a). After respective treatments by the conventional coagulants and AOPs, the protein concentrations in the LB- and TB-EPS decreased distinctly, while concentrations in S-EPS increased gradually. Compared to the raw sludge, protein concentrations in the TB-EPS of sludge treated by PAM, $FeCl_3$/CaO, Fenton's reagent/CaO, $Fe^{2+}/Na_2S_2O_8$/CaO, $Ca(ClO)_2/FeCl_3$/WS and TA-ZVI/H_2O_2/CaO reduced by 13.57%, 49.90%, 87.48%, 82.14%, 73.00% and 79.91%, respectively. Similar to the changes in protein contents of the TB-EPS, the lowest protein concentration in LB-EPS was observed

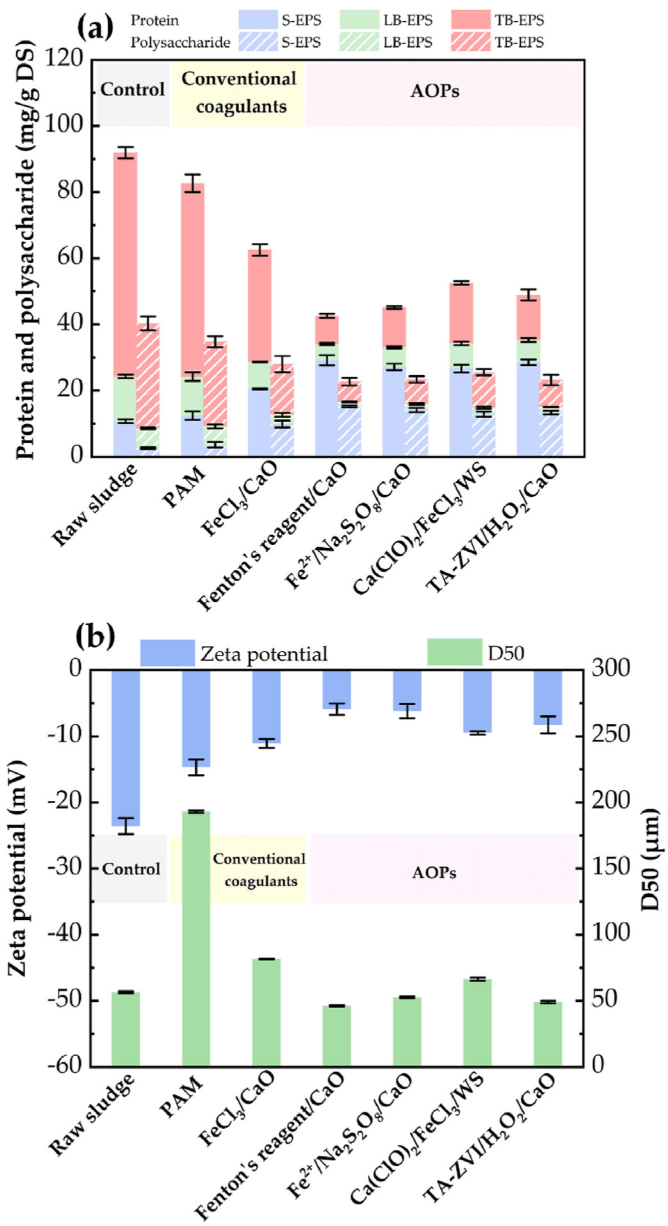

Fig. 2. Influence of the conventional coagulants and the AOPs treatments on sludge characteristics: (a) EPS concentrations, and (b) zeta potential and particle size. Abbreviation: TA = thermal-acid-washed pretreatment; ZVI = zero-valent iron.

following the Fenton's reagent/CaO treatment, followed by the $Fe^{2+}/Na_2S_2O_8/CaO$ treatment, TA-ZVI/H_2O_2/CaO treatment, $Ca(ClO)_2/FeCl_3$/WS treatment, $FeCl_3$/CaO treatment and PAM treatment. These results indicated that AOPs induced a higher removal rate of proteins in the LB- and TB-EPS than conventional coagulants, thus enhancing dewaterability (Masihi and Gholikandi, 2018). Treatment using AOPs produces numerous oxidizing radicals, which results in breakdown of EPS and changes in the chemical properties of EPS (Maqbool et al., 2019). Furthermore, decreases in the protein contents of the LB- and TB-EPS following treatment with AOPs, occurred in the ranked order of: Fenton's reagent/CaO > $Fe^{2+}/Na_2S_2O_8$/CaO > TA-ZVI/H_2O_2/CaO > $Ca(ClO)_2/FeCl_3$/WS. This result showed that Fenton's reagent/CaO treatment most effectively and simultaneously decreased protein contents in the LB- and TB-EPS, which was also consistent with the results for the improvement of sludge dewaterability (Fig. 1). This indicates that the destruction of proteins in the LB- and TB-EPS layer may be associated with the improvement in sludge dewaterability. Moreover, the protein content of the S-EPS of sludge conditioned using Fenton's reagent/CaO treatment, increased to 29.19 mg/g DS, which was higher than other AOPs. This suggested that Fenton's reagent/CaO effectively destroyed flocs and EPS, causing cell lysis and the release of proteins (Xiao et al., 2017), which is in agreement with the ⊿ORP and filtrate TOC results (Fig. S1).

The changes in polysaccharide contents were similar to those of proteins. For LB- and TB-EPS, after treatment with AOPs, the polysaccharide concentrations were lower than that of the conventional coagulants. After treated with AOPs, the polysaccharide concentrations in the S-EPS increased as compared to the conventional coagulants. A possible explanation is that AOPs could effectively remove the organic matter in the internal layers, which was then released to the outer layer (Liang et al., 2020). Furthermore, the polysaccharide concentrations in the LB- and TB-EPS after Fenton's reagent/CaO treatment decreased to the lowest level compared with the other AOPs. This indicates that Fenton's reagent/CaO has a stronger capacity for reducing the polysaccharide contents of LB- and TB-EPS, due to the stronger level of oxidation.

3.2.3. Zeta potential and particle size

The zeta potential reflects the stability of the floc colloidal matrix, which can influence sludge dewaterability (Miryahyaei et al., 2019). The zeta potential of treated sludge increased to −14.7 mV with PAM, −11.1 mV with $FeCl_3$/CaO, −5.9 mV with Fenton's reagent/CaO, −6.2 mV with $Fe^{2+}/Na_2S_2O_8$/CaO, −9.5 mV with $Ca(ClO)_2/FeCl_3$/WS and −8.3 mV with TA-ZVI/H_2O_2/CaO, compared to the raw sludge (−23.6 mV) (Fig. 2b). This indicated that the AOPs induced better charge neutralization than the conventional coagulants. This might be because the oxidation process disrupted flocs and the positive ion would reduce the electro-static repulsion of flocs, leading to charge neutralization (Yu et al., 2016; Wang et al., 2019). The present results showed that the zeta potential was highest following treatment with Fenton's reagent/CaO, followed by $Fe^{2+}/Na_2S_2O_8$/CaO, TA-ZVI/H_2O_2/CaO and $Ca(ClO)_2/FeCl_3$/WS, indicating that the Fenton's reagent/CaO treatment induced better charge neutralization. A possible explanation for this is that a significant decrease in TB-EPS could effectively decrease the negative zeta potential of sludge (Masihi and Gholikandi, 2020).

Particle size of sludge is related to sludge dewaterability (Kim et al., 2016). The particle sizes (D50) of sludge treated by PAM and $FeCl_3$/CaO increased to 193.0 and 81.6 μm, compared to the raw sludge (D50 of 56.4 μm) (Fig. 2b). This result could be due to charge neutralization and bridging (Zhang et al., 2017). Moreover, the D50 of AOPs were obviously smaller than those of the conventional coagulants, which may be due to the fact that the strong oxidizing radicals destroyed the flocs and EPS structures (Kim et al., 2016). Furthermore, the particle size of sludge treated by Fenton's reagent/CaO (46.3 μm) was smaller than the particle size observed with the other three AOPs, showing that the flocs were severely destroyed.

3.3. Transformation and species distribution of heavy metals in sludge

A high concentration of heavy metals with strong bio-accumulation risk restricts the potential for sludge utilization (Dai et al., 2019). To further evaluate the subsequent disposal of dewatered sludge, the transformation and species distribution of heavy metals in sludge were analyzed and compared.

3.3.1. Total heavy metal contents

Compared to the raw sludge, the heavy metal contents increased in the filtrate and decreased in the dewatered sludge after the

Table 3
Heavy metals contents in liquid phase and dewatered sludge before and after treatments.

Phase	Heavy metals	Raw sludge	PAM	$FeCl_3/CaO$	Fenton's reagent/CaO	$Fe^{2+}/Na_2S_2O_8/CaO$	$Ca(ClO)_2/FeCl_3/WS$	TA-ZVI/H_2O_2/CaO	Limit value [a]
Liquid phase (mg/L)	As	0.0140 ±0.0015	0.0228 ±0.0208	0.0374 ±0.0157	0.0514 ±0.0090	0.0576 ±0.0114	0.0559 ±0.0138	0.0351 ±0.0185	0.1
	Cd	0	0	0	0.0088 ±0.0006	0.0006 ±0.0009	0.0086 ±0.0002	0.0003 ±0.0001	0.01
	Cr	0	0	0	0	0	0	0	0.1
	Cu	0	0	0	0	0	0	0	0.5
	Pb	0.0017 ±0.0001	0.0020 ±0.0043	0.0071 ±0.0006	0.0053 ±0.0003	0.0069 ±0.0011	0.0016 ±0.0001	0.0058 ±0.0009	0.1
	Zn	0	0	0	0.2350 ±0.0027	0.1887 ±0.0013	1.0401 ±0.0043	0.0913 ±0.0020	1.0
Dewatered sludge (mg/kg)	As	21.75 ±3.47	19.44 ±3.55	17.89 ±5.13	10.46 ±2.19	13.81 ±1.64	8.91 ±1.48	11.51 ±1.89	30
	Cd	1.61 ±0.05	1.52 ±0.03	0.65 ±0.09	0.73 ±0.04	0.66 ±0.01	0.46 ±0.03	0.94 ±0.04	3
	Cr	214.95 ±1.37	208.19 ±0.38	168.59 ±0.41	112.42 ±0.93	97.67 ±0.39	86.77 ±0.48	100.51 ±0.80	500
	Cu	86.21 ±1.29	78.39 ±0.61	67.65 ±0.35	59.78 ±0.61	72.76 ±0.78	56.48 ±0.97	63.23 ±0.63	500
	Pb	64.18 ±2.56	34.36 ±0.49	30.44 ±1.03	25.23 ±0.87	28.03 ±2.23	24.92 ±1.49	28.59 ±2.25	300
	Zn	417.73 ±3.20	395.96 ±1.08	208.30 ±1.19	164.98 ±1.11	202.37 ±1.83	104.04 ±0.06	195.37 ±2.40	1200

Note.
[a] Discharge standard of pollutants for municipal wastewater treatment plant, GB 18918-2002, China (Liquid phase); Agricultural A level, GB 4284-2018, China (Dewatered sludge).

conventional coagulants and AOPs treatments, respectively (Table 3), indicating that conditioning with conventional coagulants and AOPs, released heavy metals from the sludge into the filtrate. These results agreed with the trend in EPS variation, with the release of EPS from the inner layers to the outer layer (Fig. 2a). This is probably because EPS had a strong adsorption effect on heavy metals (Sheng et al., 2013).

Low concentrations of heavy metals (As and Pb) in the raw sludge and sludge treated by the conventional coagulants were extracted into solution (Table 3). Compared to the raw sludge and conventional coagulants, the concentrations of As, Cd, Pb and Zn in the liquid phase significantly improved after treatment with AOPs. The highest heavy metals concentrations were found with the Fenton's regent and CaO, in which the liquid-phase contents of As, Cd, Pb and Zn were 0.0514, 0.0088, 0.0053 and 0.2350 mg/L, respectively. A possible reason for this trend could be that the oxidation process disrupted flocs and released the heavy metals contained within EPS (Wang et al., 2019). The trend of increased heavy metals in the filtrate following treatment by AOPs are consistent with the studies reported by Xiong et al. (2018) and Liu (2019), who found that sulfate radical-based oxidation also was beneficial for solubilizing heavy metals from sludge to the liquid phase. The present result showed that all heavy metals concentrations in the liquid phase in the raw and treated sludge were below the Chinese discharge standard of pollutants for WWTPs (GB 18918-2002), indicating low environmental risk.

As illustrated in Table 3, compared to the raw sludge, the contents of As, Cd, Cr, Cu, Pb and Zn in dewatered sludge decreased after treatment. These results showed that the heavy metals contents in dewatered sludge were lowest after treatment with $Ca(ClO)_2/FeCl_3$/WS, followed by Fenton's reagent/CaO, TA-ZVI/H_2O_2/CaO, $Fe^{2+}/Na_2S_2O_8/CaO$, $FeCl_3/CaO$ and PAM. This indicated that a distinct decrease in heavy metal contents was observed in dewatered sludge after treatment with AOPs, especially $Ca(ClO)_2/FeCl_3$/WS. The contents of As, Cd, Cr, Cu, Pb and Zn in dewatered sludge treated by $Ca(ClO)_2/FeCl_3$/WS decreased by 59.03%, 71.43%, 59.63%, 34.49%, 61.17% and 75.09%, respectively, as compared to the raw sludge. This might be due to the dilution effect caused by the addition of biomass (WS), as confirmed by Liu (2019). Furthermore, according to the agricultural A level (GB 4284-2018, China), heavy metals concentrations of the dewatered sludge after treatment were below the threshold level, indicating that dewatered sludge could meet the requirements for agricultural use.

3.3.2. Leaching behavior

To examine the environmental risk and immobilization of heavy metals in dewatered sludge, a TCLP test was conducted and the results are shown in Table 4. For the raw sludge, the contents of As, Cd, Cr, Cu, Pb and Zn in the leachate were 0.3946, 0.0057, 0, 0.4033, 0.0084 and 3.4064 mg/L, respectively. Compared with the raw sludge, the leached concentrations of heavy metals in dewatered sludge dramatically reduced after treatment with the conventional coagulants and AOPs. This may arise from the metallic fixation capability of calcium and biomass (WS) (Xiong et al., 2018). Moreover, the leaching contents of heavy metals after treatment were below the limit values established by wastewater quality standards for discharge to municipal sewers at the A level (CJ 343−2010, China). This result suggested that the conventional coagulants and AOPs treatments could enhance the immobilization of heavy metals, thus reducing the environmental risk of heavy metal leaching. After treatment, the leached contents of heavy metals in dewatered sludge followed the order of $Ca(ClO)_2/FeCl_3$/WS < Fenton's reagent/CaO < TA-ZVI/H_2O_2/CaO < $Fe^{2+}/Na_2S_2O_8$/CaO < $FeCl_3$/CaO ≪ PAM. These results suggested that the reduction in heavy metal leaching toxicity after treatment with AOPs were better than with conventional coagulants. It was observed that $Ca(ClO)_2/FeCl_3$/WS was more effective at reducing the leaching toxicity of heavy metals than other AOPs. Compared to the raw sludge, the leaching toxicity reductions of As, Cd and Zn treated by $Ca(ClO)_2/FeCl_3$/WS were 85.45%, 84.21% and 91.30%, respectively, while the Cr, Cu and Pb in the leachate were not detected. The reason for this might be related to the addition of biomass (WS), which might reduce the heavy metal concentrations (Liu, 2019). The present result indicated that $Ca(ClO)_2/FeCl_3$/WS exhibited an improved capacity to mitigate the environmental risk of heavy metals and enhance heavy metals immobilization, which was favorable for subsequent disposal.

Table 4
Heavy metals in TCLP tests before and after treatments.

Heavy metals	Raw sludge	PAM	$FeCl_3$/CaO	Fenton's reagent/CaO	$Fe^{2+}/Na_2S_2O_8$/CaO	$Ca(ClO)_2/FeCl_3$/WS	TA-ZVI/H_2O_2/CaO	Limit value [a]
As	0.3946 ±0.0007	0.3719 ±0.0014	0.1696 ±0.0018	0.0679 ±0.0028	0.1207 ±0.0015	0.1174 ±0.0035	0.0127 ±0.0034	0.5
Cd	0.0057 ±0.0001	0.0051 ±0.0002	0.0026 ±0.0001	0.0008 ±0.0001	0.0016 ±0.0002	0.0008 ±0.0001	0.0007 ±0.0001	0.1
Cr	0	0	0	0	0	0	0	1.5
Cu	0.4033 ±0.0020	0.2481 ±0.0016	0.1006 ±0.0001	0.0164 ±0.0012	0.02307 ±0.0008	0	0.0198 ±0.0008	2.0
Pb	0.0084 ±0.0020	0	0	0	0	0	0	1.0
Zn	3.4064 ±0.0179	3.1674 ±0.0035	1.9883 ±0.0066	1.0803 ±0.0061	1.2599 ±0.0013	0.2963 ±0.0022	1.0344 ±0.0049	5.0

Note.
[a] Wastewater quality standard for discharge to municipal sewers A level, CJ 343−2010, China.

3.3.3. *Chemical species distribution and environment risk assessment*

The chemical species distribution of heavy metals also reflects the environmental risk (Dai et al., 2019). F1, F2, F3 and F4 content of heavy metal ranges represent strong mobility and toxicity, high potential mobility, moderate stability and strong stability, respectively (Xiong et al., 2018).

As was major in the F1 and F4, which accounted for 79.26% of total heavy metal content in raw sludge (Fig. 3a). After treatment with the conventional coagulants and AOPs, the percentage of As in F1 significantly decreased, while in F4 the As content obviously increased, suggesting that the unstable As (F1) dissolved into filtrate and the stable As (F4) remained in the dewatered sludge. It is of note, that after AOPs conditioning, the decrease in the As fraction in F1 was greater than following conventional coagulant treatment due to the degradation of flocs and EPS by strong oxidizing radicals (Liu, 2019). After conditioned by Fenton's reagent/CaO, $Fe^{2+}/Na_2S_2O_8$/CaO, $Ca(ClO)_2/FeCl_3$/WS and TA-ZVI/H_2O_2/CaO, the percentage of As in F1 fractions decreased to 9.38%, 3.55%, 9.58% and 16.19%, respectively. This showed that $Fe^{2+}/Na_2S_2O_8$/CaO more effectively reduced the direct toxicity of As in dewatered sludge than other three AOPs.

For raw sludge, the percentages of Cd and Zn in the total F1 and F2 fractions were 71.28% and 68.28%, indicating that F1 and F2 were the primary fractions of Cd and Zn in the raw sludge (Fig. 3b and f), which was consistent with the findings of Ignatowicz (2017) and Liu (2019). After treatment, the total contents of Cd and Zn in F1 and F2 significantly decreased, while the total percentages of F3 and F4

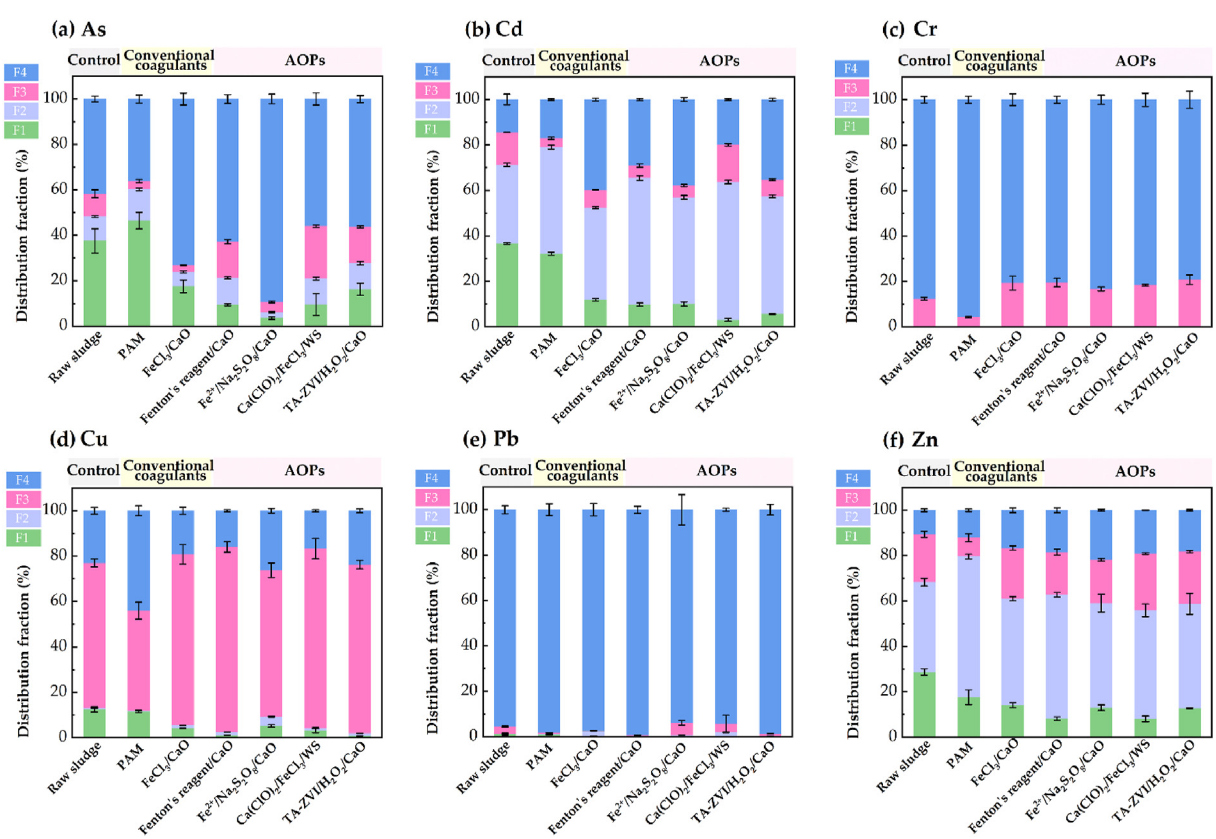

Fig. 3. Chemical species distribution of heavy metals in the dewatered sludge of the conventional coagulants and the AOPs treatments. Abbreviation: TA = thermal-acid-washed pretreatment; ZVI = zero-valent iron.

increased, suggesting the low mobility and good stabilization of Cd and Zn. Furthermore, after AOPs conditioning, the percentages of Cd and Zn in the F1 fraction were lower than that of conventional coagulants, indicating the low mobility and toxicity of Cd and Zn after treatment with AOPs. It is of note, that after conditioning by $Ca(ClO)_2/FeCl_3$/WS, the percentages of Cd and Zn in F1 decreased to 2.97% and 8.11%, respectively, which were lower than following treatment with other three AOPs. This indicated that the direct toxicity of Cd and Zn was decreased after $Ca(ClO)_2/FeCl_3$/WS treatment, which might be due to the adsorption and fixation effect by biomass (WS) (Xiong et al., 2018).

For raw and treated sludge, Cr (Fig. 3c), Cu (Fig. 3d) and Pb (Fig. 3e) were mainly distributed in F3 and F4, accounting for 85%–100% of the total heavy metal content. These results show the high stability of Cr, Cu and Pb in the environment, which is confirmed by the decrease in Cr, Cu and Pb contents in dewatered sludge (Table 3). The present results are consistent with the results reported by Dai et al. (2019). Moreover, AOPs were found to be more effective at improving the stabilization of Cu than conventional coagulants. It is of note, that the stabilization of Cu in sludge after treatment with AOPs was most effective with TA-ZVI/H_2O_2/CaO, followed by Fenton's reagent/CaO, $Ca(ClO)_2/FeCl_3$/WS and $Fe^{2+}/Na_2S_2O_8$/CaO.

Agricultural use is the mainstream approach for sludge disposal worldwide (Song et al., 2016b). To apply the sludge in agricultural scenarios, the environmental risk of heavy metals in dewatered sludge has to be evaluated using the RAC (Liu, 2019). The RAC of As, Cd, Cr, Cu, Pb and Zn in raw sludge posed a high risk, high risk, no risk, medium risk, low risk and medium risk, respectively (Table S2). After treatment, the environment risk of heavy metals in dewatered sludge was reduced. The RAC of heavy metals in dewatered sludge treated using conventional coagulants reduced to the medium risk level, while the RAC following treatment with AOPs decreased to a low risk level. The present results demonstrated that environmental risk of heavy metals was reduced obviously following treatment with AOPs. Moreover, after the Fenton's reagent/CaO and $Ca(ClO)_2/FeCl_3$/WS treatments, the RAC of As, Cd, Cr, Cu, Pb and Zn in sludge presented a low risk, low risk, no risk, low risk, no risk and low risk, respectively, indicating a reduced environmental risk from heavy metals.

Consequently, treatment with AOPs significantly enhanced the solubilization of heavy metals and reduced both the leaching toxicity and the environmental risk of heavy metals. Among the AOPs, the $Ca(ClO)_2/FeCl_3$/WS treatments were more effective at improving the immobilization of heavy metals, decreasing the heavy metals content and alleviating both the leaching toxicity and environmental risk of heavy metals, which was beneficial to agricultural use.

3.4. Phytotoxicity in the dewatered sludge

Phytotoxicity is also a crucial factor influencing the use of dewatered sludge in agriculture (Kebibeche et al., 2019). Therefore, the responses of two representative plant species (*Brassica rapa chinensis* and *Lactuca sativa*) to potential phytotoxicity in terms of the GI were measured (Fig. 4). The raw sludge samples induced the strongest inhibition in the two plant species (*Brassica rapa chinensis* (GI = 50.15%); *Lactuca sativa* (GI = 33.98%)). This difference may arise from the phytotoxic effects of ammonia and the presence of harmful substances (Kebibeche et al., 2019). For *Brassica rapa chinensis*, the GI values were 63.31%, 73.16%, 103.74%, 88.11%, 150.08% and 101.32% for PAM, $FeCl_3$/CaO, Fenton's reagent/CaO, $Fe^{2+}/Na_2S_2O_8$/CaO, $Ca(ClO)_2/FeCl_3$/WS and TA-ZVI/H_2O_2/CaO treatments, respectively. For *Lactuca sativa*, the GI values were 38.67%, 57.99%, 87.28%, 79.75%, 95.06% and 85.53% for PAM, $FeCl_3$/CaO, Fenton's

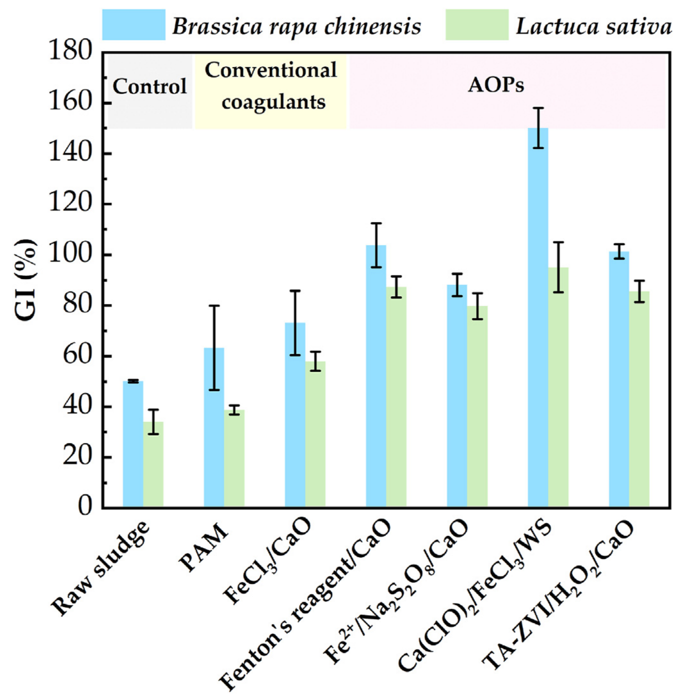

Fig. 4. Phytotoxicity in the dewatered sludge of the conventional coagulants and the AOPs treatments. Abbreviation: TA = thermal-acid-washed pretreatment; ZVI = zero-valent iron.

reagent/CaO, $Fe^{2+}/Na_2S_2O_8$/CaO, $Ca(ClO)_2/FeCl_3$/WS and TA-ZVI/H_2O_2/CaO, respectively. The present results revealed that, compared to conventional coagulants, the sludge treated by AOPs presented lower phytotoxicity in both plants. This may be because AOPs destroyed the cellular structure and released numerous nutrients, such as carbon, proteins and polysaccharides (Fig. 2a and Table 2), which were favorable to plant growth (Zou et al., 2019). After AOPs treatments, the reduction in phytotoxicity in both exposed plants followed the order of $Ca(ClO)_2/FeCl_3$/WS > Fenton's reagent/CaO > TA-ZVI/H_2O_2/CaO > $Fe^{2+}/Na_2S_2O_8$/CaO. This indicated that the sludge treated by $Ca(ClO)_2/FeCl_3$/WS significantly mitigated the phytotoxicity in both plants (GI>87%). Moreover, the GI of the dewatered sludge treated by AOPs was greater than 80%, indicating the non-phytotoxicity of sludge (Tiquia and Tam, 1998). Accordingly, the $Ca(ClO)_2/FeCl_3$/WS significantly depressed the phytotoxicity of dewatered sludge, exhibiting a potential for application in agricultural use.

3.5. Cost analysis

To compare the economic potential of treatment using conventional coagulants and AOPs, a desktop scaled-up study was conducted in the scenario of a 400,000 m^3/d WWTP with a daily dry sludge production of 60.0 ton. The total annual cost of the conventional coagulants were USD $8,853,638.5 and USD $4,732,279.4 for PAM and $FeCl_3$/CaO treatments, respectively (Table 5). The total annual cost of AOPs were USD $3,862,391.7 for Fenton's reagent/CaO, USD $4,836,344.1 for $Fe^{2+}/Na_2S_2O_8$/CaO, USD $4,247,265.5 for $Ca(ClO)_2/FeCl_3$/WS and USD $3,933,740.4 for TA-ZVI/$H_2O_2$/CaO. These results suggested that the total annual cost of AOPs were much lower than conventional coagulants, especially for PAM treatment. It is of note, that the annual total cost of Fenton's reagent/CaO treatment was the lowest, which was lower than $Fe^{2+}/Na_2S_2O_8$/CaO by 20.14%, lower than $Ca(ClO)_2/FeCl_3$/WS by 9.06% and lower than TA-ZVI/H_2O_2/CaO by 1.81%. Therefore, the Fenton's

Table 5
Economic analyses of the conventional coagulant and the AOPs for the sludge treatment and disposal.

Sludge treatment methods [a]	Total annual sludge production (ton DS/year)	Wc (wt%)	Treatment cost [b] (USD$/year)	Disposal cost [c] (USD$/year)	Total cost (USD$/year)
PAM	21,900.0	80.22	1,933,770.0	6,919,868.5	8,853,638.5
$FeCl_3$/CaO		61.08	1,215,450.0	3,516,829.3	4,732,279.4
Fenton's reagent/CaO		45.73	1,340,280.0	2,522,111.6	3,862,391.7
Fe^{2+}/$Na_2S_2O_8$/CaO		51.90	1,990,710.0	2,845,634.1	4,836,344.1
$Ca(ClO)_2$/$FeCl_3$/WS		55.56	1,167,270.0	3,079,995.5	4,247,265.5
TA/ZVI/H_2O_2/CaO		53.78	972,360.0	2,961,380.3	3,933,740.4

Note.
[a] Sludge treatment conditions were according to Table 1.
[b] Price of chemical agents was according to http://www.alibaba.com/. Treatment cost was calculated according to Table S3.
[c] Dewatered sludge disposal fee is USD $62.5/ton, which gathered from the local market.

reagent/CaO treatment was the most cost-effective, feasible and environmental-friendly process for improving sludge dewaterability, which is highly beneficial for sludge disposal.

3.6. Environmental implications

The tested AOPs are more preferable than conventional coagulants for sludge treatment and disposal, as AOPs significantly enhance sludge dewaterability, alleviate heavy metal toxicity and phytotoxicity of dewatered sludge, while having lower operational costs. Among the AOPs, Fenton's reagent/CaO treatment achieved the greatest sludge dewaterability and had the lowest operational costs (Table 5 and Fig. 1). However, $Ca(ClO)_2$/$FeCl_3$/WS treatment had the lowest environment risk from heavy metals and the lowest phytotoxicity of dewatered sludge (Tables 3, 4, and S2 and Fig. 4). In addition, sewage sludge is the main carrier of heavy metals in WTTPs and the distribution of heavy metal contents in sludge varies in different regions (Chen et al., 2008). Therefore, in order to minimize operational costs, environmental risk and phytotoxicity as well as maximizing sludge dewaterability, different sludge conditioning measures should be employed. For instance, Fenton's reagent/CaO treatment could be preferentially utilized if the content of heavy metals in sludge was relatively low. When the content of heavy metals in sludge is high, $Ca(ClO)_2$/$FeCl_3$/WS treatment would be a preferred option as it can effectively reduce heavy metal toxicity and phytotoxicity in the dewatered sludge with only a slight increase in operational costs and a slight decrease in sludge dewaterability. It is of note, that the respective superiority of Fenton's reagent/CaO and $Ca(ClO)_2$/$FeCl_3$/WS treatments should be further testified via long-term field trials. It is also worth noting that tracing the changes in the crystalline structure of sludge flocs during different AOPs would be beneficial to further understand the sludge conditioning and dewatering mechanism (Maqbool et al., 2019).

4. Conclusions

In this study, the sludge dewatering efficiency and the potential toxicity of both conventional coagulants and AOPs were systematically compared. The main conclusions of this study are shown as follows:

- The AOPs exhibited a higher sludge dewatering efficiency than the conventional coagulants under ultrahigh pressure. The Wc values of AOPs were lower than 56.0 wt%. Compared to the conventional coagulants, the AOPs significantly reduced the environmental risk of heavy metals and the phytotoxicity of dewatered sludge. From the economic analysis results, the total annual cost of AOPs were lower than the conventional coagulants.
- Greater sludge dewaterability improvement and cost savings were obtained using the Fenton's reagent/CaO treatment. However, the $Ca(ClO)_2$/$FeCl_3$/WS treatment could more effectively reduce the environmental risk of heavy metals and the phytotoxicity of dewatered sludge.

In summary, the Fenton's reagent/CaO is a feasible, alternative and inexpensive method to realize sludge deep dewatering, which is better suited to low heavy metal concentration sludge. The $Ca(ClO)_2$/$FeCl_3$/WS treatment is an effective and feasible method to alleviate sludge heavy metal toxicity and phytotoxicity, being more applicable to high heavy metal content sludge. The findings obtained in this study could provide useful information for optimizing sludge deep dewatering and sludge disposal processes.

Declaration of competing interests

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.

CRediT authorship contribution statement

Jialin Liang: Conceptualization, Methodology, Software, Investigation, Writing - original draft. **Liang Zhang:** Validation, Writing - review & editing. **Maoyou Ye:** Writing - review & editing. **Zhijie Guan:** Investigation. **Jinjia Huang:** Investigation. **Jingyong Liu:** Writing - review & editing. **Lei Li:** Writing - review & editing. **Shaosong Huang:** Supervision, Conceptualization. **Shuiyu Sun:** Supervision, Resources, Supervision, Project administration, Funding acquisition.

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Appendix A. Supplementary data

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