

Influence of thermal hydrolysis treatment on chemical speciation and bioleaching behavior of heavy metals in the sewage sludge

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ABSTRACT

In this study, the transformation of chemical speciation of Cr, Mn, As and Cd in the sewage sludge before and after thermal hydrolysis treatment was investigated using modified BCR method. The effect of thermal hydrolysis treatment and chemical speciation change on the subsequent bioleaching behavior was also researched. The results showed that the concentrations of Cr, Mn, As and Cd in oxidizable fraction decreased in the sludge treated by thermal hydrolysis. Meanwhile, the proportions of Cr, Mn and As in the mobile fractions (acid-soluble/exchangeable and reducible fraction) all decreased, while Cd was concentrated in the sludge treated by thermal hydrolysis. The final pH value of bioleached sludge treated by thermal hydrolysis was lower than that in the bioleached raw sewage sludge. And faster increase of oxidation-reduction potential (ORP) was also found in the bioleaching process of the sludge treated by thermal hydrolysis. The removal percentage of Mn and Cd increased in the bioleached sludge treated by thermal hydrolysis. Thermal hydrolysis treatment can promote the bioleaching to some extent. Furthermore, the environmental risk of Cr, Mn, As and Cd in the bioleached sludge treated by thermal hydrolysis was all alleviated according to risk assessment analysis compared with the bioleached raw sewage sludge.

Key words | bioleaching, chemical speciation, heavy metals, sewage sludge, thermal hydrolysis treatment

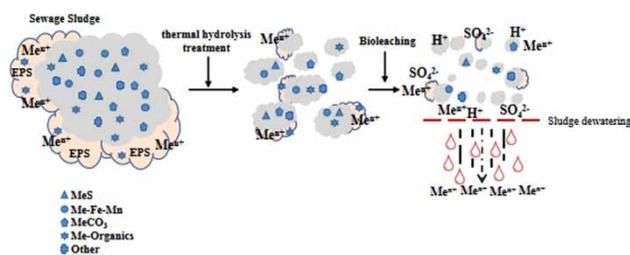
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HIGHLIGHTS

- Effect of thermal hydrolysis on heavy metals speciation in sludge was reported.
- Proportions of heavy metals in oxidizable fraction decreased after treatment.
- Metabolic activity of bioleaching bacteria was improved in treated sludge.
- Environmental risk of bioleached sludge after thermal treatment was alleviated.

GRAPHICAL ABSTRACT



INTRODUCTION

A large amount of residual sludge has been generated from the treatment process of municipal wastewater (Dewil *et al.* 2006; Zhou *et al.* 2017). Since it contains various toxic substances such as heavy metals, pathogens, organic contaminants and so on, proper treatment and disposal of sewage sludge is a major part of waste treatment (Malhotra & Garg 2019). Some of these heavy metals are persistent bio-accumulative, non-biodegradable and have high mobility in nature, which could easily cause secondary pollution to the surrounding environment, especially when adopting landfill, ocean dumping or land application for sludge disposal (Fontmorin & Sillanpaa 2015; Zhang *et al.* 2016). Therefore, the efficient removal or immobilization of heavy metals in the sewage sludge is necessary for safe disposal of sludge.

Anaerobic digestion is a cost-effective and promising technology for sewage sludge treatment due to the stabilization of sludge and the high energy recovery in the form of methane. It is a well-known fact that hydrolysis is the rate-limiting step of anaerobic digestion. Thus, various sludge pretreatment methods have been developed to accelerate the hydrolysis rate and enhance methane productivity. Thermal hydrolysis pretreatment has been reported to be the most reliable and profitable method for sludge pretreatment, which is commercially implemented (Pilli *et al.* 2015; Wu *et al.* 2015; Choi *et al.* 2018). As is well-known, there are four kinds of heavy metal chemical speciation including acid-soluble/exchangeable, reducible, oxidizable, and residual fraction (Rauret *et al.* 1999). The toxicity, bio-availability and mobility of heavy metals depend on both the quantity and chemical speciation of metals (Dong *et al.* 2013). Meanwhile, previous studies indicated that various physical and chemical treatments could influence the heavy metals' chemical speciation (Zhang *et al.* 2016, 2017; Udayanga *et al.* 2019). Obrador *et al.* (2001) reported the change of concentrations and potential lability of heavy metals after thermal treatment (180 °C–400 °C). However, the behavior of heavy metals in sludge after thermal hydrolysis pretreatment is still poorly understood (Zhang *et al.* 2016).

Bioleaching has been considered as one of the most economical and promising methods for bioremediation of heavy metals from sewage sludge (Rastegar *et al.* 2014; Gu *et al.* 2018). It uses chemolithotrophic iron- or sulfur-oxidizing bacteria to solubilize metallic compounds such as metal oxides and sulfides (Chang *et al.* 2019). During bioleaching process, metal sulfide (oxidizable fraction) is oxidized directly or indirectly to soluble metal sulfate.

Meanwhile, the elemental sulfur and metal sulfides can be oxidized to sulfuric acid, leading to the partial dissolution of metal oxide and metals bound to carbonates (reducible fraction) and organic matter (oxidizable fraction), due to the low pH coupled with highly oxidizing conditions (Gu *et al.* 2018; Potysz *et al.* 2018). Therefore, the chemical speciation of metals can affect the bioleaching efficiency to a certain extent. However, there is little literature available on the influence of thermal hydrolysis pretreatment on the chemical speciation distributions and bioleaching behavior of heavy metals in the sewage sludge.

The aim of this paper was to investigate the change of heavy metals chemical speciation in the sewage sludge after thermal hydrolysis treatment and the effect of pretreatment on the bioleaching behavior. The chemical speciation distribution of Cr, Mn, As and Cd in the raw sewage sludge (RSS) and sludge treated by thermal hydrolysis (THS) was compared using modified BCR method (Rauret *et al.* 1999). Then, the effects of chemical speciation on removal efficiency of heavy metals in the RSS and THS were compared through bioleaching experiments.

MATERIALS AND METHODS

Sewage samples and thermal hydrolysis pretreatment step

The RSS used in this study was collected from the thickening tank gathering secondary sludge in a local municipal wastewater treatment plant (Tianjin, China) and stored at 4 °C for further use. The total solids (TS) and volatile solid (VS) of the RSS were 42.62 g·L⁻¹ and 22.76 g·L⁻¹, respectively.

The RSS was treated by thermal hydrolysis at a temperature of 90 °C and reaction time of 150 min in a thermostatic water bath.

Inoculum preparation and bioleaching procedure

The mixed bioleaching culture (mainly acidophilic iron-oxidizing bacteria) was isolated and enriched on the basis of previous study (Pathak *et al.* 2009). The seed sludge (200 mL) collected from the secondary sedimentation tank was added into 500 mL Erlenmeyer flasks with FeSO₄·7H₂O (20 g·L⁻¹) as substrate. The flasks were placed in a constant

temperature shaker set at 200 rpm and 28 °C. When the pH value of the mixed culture was reduced from an initial value of 6.5 to below 3.0, 20 mL mixed culture was transferred into other flasks filled with 200 mL fresh feed sludge. The bioleaching mixed culture was enriched twice again in the constant temperature shaker under the same conditions. The acidified mixed culture obtained, which contained abundant acidophilic iron-oxidizing bacteria, was used as the inoculum for subsequent bioleaching experiments.

The bioleaching experiments were conducted using 300 mL of sludge (RSS or THS) mixed with 10% (v/v) of the inoculum. FeSO₄·7H₂O at a concentration of 20 g·L⁻¹ was added as energy source. The bioleaching samples were cultured in a constant temperature shaker at 200 rpm and 28 °C. During the experiments, the change of oxidation-reduction potential (ORP) and pH was monitored with time. Every other day, 10–20 mL of sludge sample was taken out for heavy metal analysis.

Analysis

pH and ORP of the bioleaching sludge were measured using a multi-parameter tester with pH/ORP electrode (Multi 3510 IDS, WTW, Germany). VS content was quantified by standard method (APHA 1998).

The concentration of heavy metals in the sludge was determined using inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7700). Prior to analysis, the sludge samples were centrifuged at 5,000 r/min for 15 min. The centrifuged deposit was freeze-dried for 24 h. Then, 0.5 g of dried deposit sample was digested with 2.0 mL of H₂O₂ (30%) and 6.0 mL of HNO₃ using a microwave digestion instrument (Milestone ETHOS A, Italy). The sample after microwave digestion was filtered with 0.45 μm cellulose syringe filters. The filtrate was analyzed using ICP-MS to determine heavy metal concentrations. The concentrations of heavy metals in different chemical forms were analyzed according to the improved BCR procedures (Rauret et al. 1999). In this technique, chemical speciation of metals is divided into acid-soluble/exchangeable, reducible, oxidizable (organic-bound and sulfide), and residual forms.

Risk assessment of heavy metals in sludge

Risk assessment code (RAC) was used to evaluate the environmental risk of heavy metals in the sewage sludge (Huang et al. 2011). RAC is calculated as follows:

$$\text{RAC}(\%) = \frac{\text{FA}}{\text{HM}} \times 100 \quad (1)$$

where FA is the concentration of heavy metals present in acid-soluble/exchangeable form, HM is the total concentration of heavy metals.

Modified potential ecological risk index (MRI) is a heavy metal pollution assessment method that takes into account the toxicity, total concentration, and bioavailability of heavy metals (Ji et al. 2019). The calculation equation of MRI was presented as follows:

$$\text{MRI} = \sum_{i=1}^m E_r^i = \sum_{i=1}^m T_r^i \cdot \frac{C_D^i \cdot \Omega}{C_r^i} \quad (2)$$

$$\Omega = A\delta + B \quad (3)$$

where C_D^i : the concentration of heavy metal in sludge; C_r^i : the background or pristine value of individual heavy metal; T_r^i : the heavy metal toxic response factor. T_r^i values of the heavy metals in the sludge were as follows: Cr (2), Mn (1), As (10), Cd (30) (Hakanson 1980; Huang et al. 2011; Ji et al. 2019). E_r^i : the monomial potential ecological risk factor; Ω is the correction coefficient of heavy metal contents, A is the percentage of acid-soluble/exchangeable form, $B = 1 - A$. $\delta = 1.0$ ($1 < \text{RAC} \leq 10$), $\delta = 1.2$ ($11 \leq \text{RAC} \leq 30$), $\delta = 1.4$ ($31 \leq \text{RAC} \leq 50$), $\delta = 1.6$ ($50 < \text{RAC}$). The standard of MRI is shown in Table 1 (Jin et al. 2016; Ji et al. 2019).

RESULTS AND DISCUSSION

Chemical speciation change of heavy metals in RSS and THS

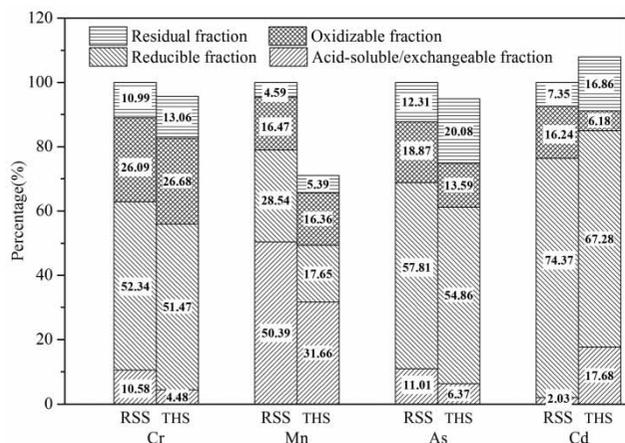
The total concentrations of Cr, Mn, As and Cd in the RSS and THS are given in Table 2. The chemical speciation of heavy metals in the binding fractions given as % of this total of the RSS and THS is presented in Figure 1, and

Table 1 | Standard of modified potential ecological risk index

E_r^i	Potential ecological risk	MRI	Sludge contamination
$E_r^i < 40$	Low	MRI < 150	Low
$40 < E_r^i < 80$	Moderate	$150 < \text{MRI} < 300$	Moderate
$80 < E_r^i < 160$	Considerate	$300 < \text{MRI} < 600$	Considerable
$160 < E_r^i < 320$	High	MRI > 600	High
$E_r^i > 320$	Very high		

Table 2 | Total concentrations of heavy metals in the RSS and THS

Parameter	RSS	THS
Cr (mg/kg-TS)	414.85 ± 6.85	396.94 ± 8.38
Mn (mg/kg-TS)	604.49 ± 16.13	429.78 ± 14.92
As (mg/kg-TS)	24.70 ± 0.81	23.44 ± 0.48
Cd (mg/kg-TS)	1.78 ± 0.25	1.92 ± 0.07

**Figure 1** | Chemical speciation (%) of heavy metals in the RSS and THS; total concentration of heavy metals in the RSS was set as 100%.

the total concentration of heavy metals in the RSS is set as 100%.

It can be seen that there was a slight decrease in the total concentrations of Cr and As, and significant reduction of Mn after thermal hydrolysis treatment. It was reported that part of the heavy metals in the sewage sludge were from the absorption action of extracellular polymeric substance (EPS), while some of the heavy metals were absorbed and accumulated in the protoplasm of the microbial cells (Li & Yu 2014). The thermal hydrolysis treatment could disintegrate the EPS and even the microbial cells; thereby releasing part of the adsorbed and weak-bonded heavy metals to the liquid phase from the zoogloae (Choi *et al.* 2018). However, the concentrations of Cd in most sludge samples after thermal hydrolysis treatment were increased in the parallel tests, probably due to the loss of organic matter; similar phenomena have also been found when investigating the effects of thermal hydrolysis and anaerobic treatment on the heavy metal distributions in sewage sludge (Dong *et al.* 2013; Zhang *et al.* 2016). The VS of the THS was 20.03 g·L⁻¹, which was 11.99% less than the RSS. Thus, Cd was concentrated due to the loss of organic matter in the sewage sludge, which resulted from the decomposition

and degradation of organic matter caused by thermal hydrolysis treatment.

The acid-soluble/exchangeable fraction of Cr decreased from 10.58% to 4.48% after thermal hydrolysis treatment (Figure 1). Cr in acid-soluble/exchangeable fraction was unstable in mild acid conditions (Karwowska *et al.* 2015) and could be dissolved out by volatile fatty acids produced during the thermal hydrolysis treatment process (Zou *et al.* 2019). The pH value of the sludge decreased from 7.75 to 6.60 after treatment in the study; this may be one of the reasons for the reduction of Cr in the acid-soluble/exchangeable fraction. In addition, some of the lipids, carbohydrates and proteins were hydrolyzed into small molecules and lost binding sites of heavy metals (Wei *et al.* 2019), which might be another reason for the decrease of acid-soluble/exchangeable fraction of Cr. Most of the Cr existed in the reducible fraction in the RSS, and its content barely changed after treatment. Similarly, despite the loss of organic matter, the content of Cr present in the oxidizable fraction also remained constant in the THS.

The concentration of Mn existing in the acid-soluble/exchangeable fraction decreased from 304.60 mg/kg to 191.38 mg/kg after thermal hydrolysis treatment, which could account for the decrease of the total concentration of Mn in the THS. The proportion of Mn present in oxidizable form (organic-bound and sulfide) decreased slightly from 16.47% (RSS) to 16.36% (THS). One possible reason might be that most of the Mn was present in sulfide rather than bound to organic matter, and metal sulfide is generally considered to be relatively stable (Zhang & Gu 2007; Nguyen *et al.* 2018).

Most of the As was distributed in reducible and oxidizable forms in the RSS. Thermal hydrolysis treatment reduced the oxidizable fraction of As in the RSS from 18.87% to 13.59% in the THS, as shown in Figure 1, mainly due to the release of organic matter. Correspondingly, the concentration of As in the residual fraction was increased from 3.04 mg/kg to 4.96 mg/kg. In the presence of aqueous Fe³⁺ in the sludge, the dissolved As³⁺ could be oxidized to As⁵⁺ (AsO₄³⁻) and then react with Fe³⁺ to generate insoluble FeAsO₄ (Wang *et al.* 2015), thus increasing the As content in the residual fraction in the THS.

The concentration of Cd present in the oxidizable fraction decreased from 0.29 mg/kg in the RSS to 0.11 mg/kg in the THS, which indicated that most of the oxidizable Cd was bound to organic substances in the RSS. The portions of Cd in reducible fraction decreased from 74.37% to 67.28% after treatment, while the portions of Cd distributed in other forms all increased.

Bioleaching process of the RSS and THS

Evolution of pH and ORP during bioleaching process

The changes of pH and ORP are the most important indicators to reflect the metabolic activity of acidophilic bioleaching microorganisms (Zhu *et al.* 2013). Figure 2 shows the evolution of pH and ORP in the bioleaching process of the RSS and THS. After a slight increase of the mixed culture pH, a sharp decline of the pH value of the RSS and THS was observed in the following 6 days (4th–10th day), which was the result of oxidation of Fe^{2+} and generation of H^+ due to the metabolism of iron-oxidizing bacteria. The pH of the RSS and THS leaching samples decreased to 2.25 and 2.00 on the 10th day, respectively, and remained stable till the end of the bioleaching process. The generation of sulfuric acid from metal sulfide oxidation, and the formation of ferric hydroxide and jarosite from the hydrolysis of Fe^{3+} , were the main reasons for the decrease in pH (Zhu *et al.* 2013). As shown in Figure 2, the ORP of the THS (−162.5 mV) was much lower than that of the RSS (25.0 mV) due to the generation of reducing substances (such as organic acids and amino acids) during the thermal hydrolysis process (Chen *et al.* 2019). With the oxidation of Fe^{2+} and production of Fe^{3+} , the ORP of the RSS increased from the initial 25.0 mV to 602.0 mV (maximum value) after 12 days, while the ORP of the THS reached 596.0 mV (maximum value) on the 14th day. The bioleached THS showed a faster increase of ORP as well as lower final pH value compared with the RSS, probably because the thermal pre-treatment of sewage sludge could decrease the sludge floc size and improve the sludge flowability (Pilli *et al.* 2015; Choi *et al.* 2018), thus providing more suitable

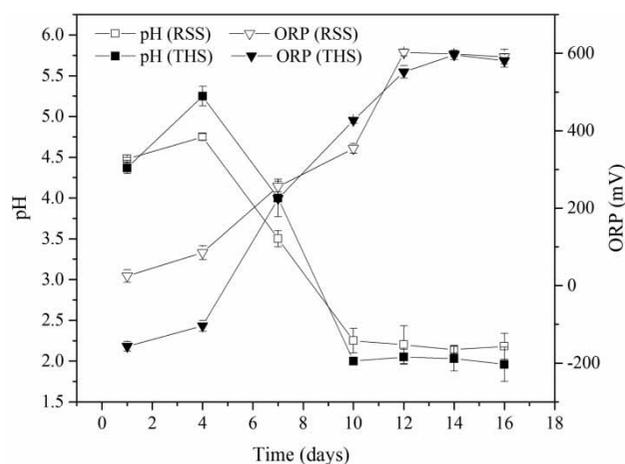


Figure 2 | Changes of pH and ORP in the RSS and THS during bioleaching process.

mass transfer conditions for the microorganisms. The decreasing pH and the increasing ORP are indicators of the proliferation of the bioleaching microorganisms (Fontmorin & Sillanpaa 2015).

Solubilization of heavy metals in the RSS and THS

The bioleaching behavior of heavy metals in the RSS and THS is shown in Figure 3. After 4 days' lag phase, all the heavy metals began to dissolve out along with the decreasing pH of the mixed culture. Cr is reported to be one of the relatively stable metals, with a low solubilization threshold pH (Villar & Garcia 2002); it could only be solubilized efficiently when the pH of the sludge dropped below 3.0 (Karwowska *et al.* 2015). In addition, the content of Cr that is easy to be dissolved (present in the acid-soluble/exchangeable fraction) was low in the RSS and THS. Thus, the rapid solubilization of Cr started on the 8th day in the RSS and on the 12th day in the THS. The final solubilization rate of Cr in the THS (51.53%) was lower than that in the RSS (58.18%), because 57.66% of the Cr in the acid-soluble/exchangeable fraction was dissolved out during the thermal hydrolysis treatment process. Furthermore, the residual content of Cr in the bioleached RSS and THS was approximately equal.

The highest removal rate of Mn in the RSS was 43.86%, obtained on the 16th day, which was almost equal to that in the THS (44.80% on the 16th day). However, the final content of Mn in the bioleached THS was 333.80 mg/kg, which was much lower than that in the bioleached RSS (339.90 mg/kg). This was because part of the acid-soluble/exchangeable and reducible fractions of Mn in the sludge was transferred to the liquid phase after thermal hydrolysis treatment. Despite previous studies showing that Mn was easily dissolved out in the bioleaching process of mine tailings, river sediment and fly ash with low organic matter content (Nguyen *et al.* 2015; Gan *et al.* 2016; Funari *et al.* 2017), the solubilization efficiency of Mn was reported to be relatively low when treating sewage sludge and contaminated soil with high organic matter content (Lombardi & Garcia 2002; Deng *et al.* 2013).

The decrease of As content was observed on the 6th day in the RSS and THS. These results were consistent with previous studies; a certain pH value should be reached to dissolve out As from the solid phase (Nguyen *et al.* 2018). Final removal rate of As was 35.40% in the RSS, but this value was decreased to 21.79% in the THS. As previously mentioned, the generation of insoluble FeAsO_4 could lead to the decreasing solubilization efficiency of As (Wang

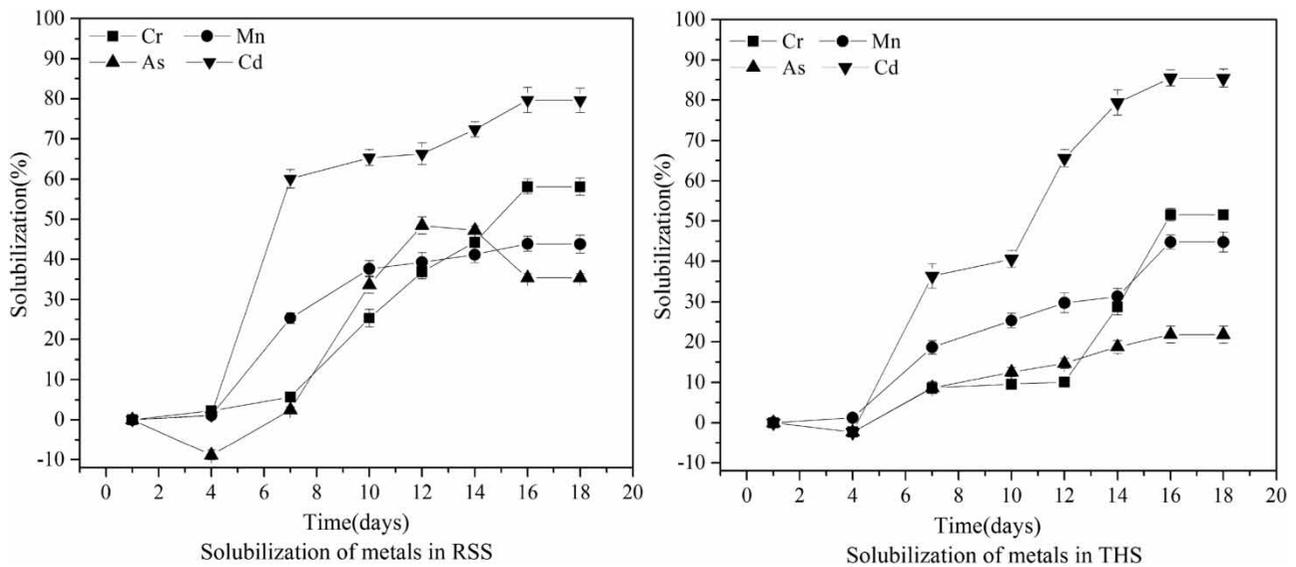


Figure 3 | Solubilization efficiency of heavy metals in the RSS and THS during bioleaching process.

et al. 2015) in the RSS after the 14th day, so the leaching period should be optimized to obtain high removal rate of As.

Previous study showed that the acid dissolution was the main solubilization mechanism of Cd during bioleaching process and it could be dissolved out easily with threshold pH of 3.0–4.9 (Wang *et al.* 2015). Figure 3 also showed that after a lag phase of 4 days, Cd was dissolved out quickly. A higher solubilization rate of 85.40% in the THS was observed compared with 79.55% in the RSS. This is mainly because part of the Cd bound to organic matters (oxidizable fraction) was dissolved after thermal hydrolysis treatment as shown in Figure 1.

Chemical speciation change of heavy metals in the RSS and THS after bioleaching

The distributions of Cr, Mn, As and Cd in the bioleached RSS and THS are shown in Figure 4. The chemical speciation distribution was one of the major influencing factors for the solubilization efficiency of heavy metals. The speciation analysis showed that bioleaching dissolved out Cr in the reducible fraction by 69.47% in the RSS and 71.97% in the THS. The lower bioleaching efficiency of Cr in the THS was mainly due to the decreasing solubilization percentage of the acid-soluble/exchangeable fraction after thermal hydrolysis treatment as shown in Figure 1.

The solubilization of the reducible (90.49%) and oxidizable fraction (69.24%) could account for the removal of Mn in the RSS, while most of the Mn present in the reducible fraction (96.53%) was dissolved out in the THS, which

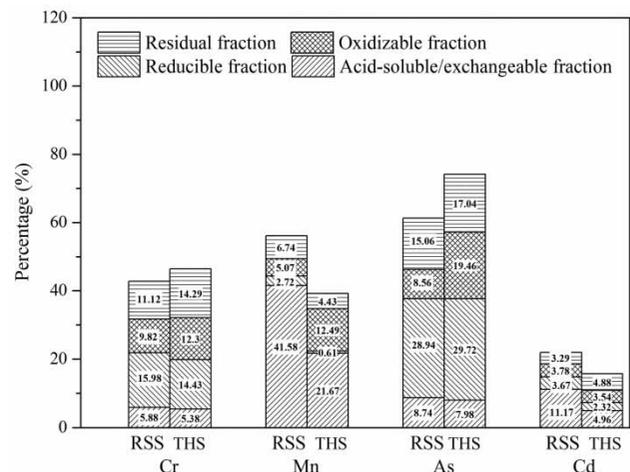


Figure 4 | Distributions of heavy metal chemical speciation (%) in the bioleached RSS and THS; total concentration of heavy metals in the RSS was set as 100%.

was usually observed under acidic conditions. However, the percentage of Mn present in the acid-soluble/exchangeable fraction in the bioleached RSS and THS was still high. The reason may be that part of the Mn in the exchangeable fraction existed in the EPS. Similar results have been found when investigating the distribution of Mn in the anaerobic sewage sludge after bioleaching; plenty of Mn present in the exchangeable fraction was detected in the bioleached sludge (Lombardi & Garcia 2002).

Bioleaching decreased the concentration of As in reducible fraction by 49.93% in the RSS and 45.83% in the THS. Higher solubilization efficiency of As in the RSS was

Table 3 | Risk assessment of heavy metal pollution in RSS, THS, bioleached RSS and bioleached THS

Heavy metal	RAC				MRI	E_r			
	RSS	THS	Bioleached RSS	Bioleached THS		RSS	THS	Bioleached RSS	Bioleached THS
Cr	10.58	4.68	13.73	11.60	10.15	9.52	4.39	4.71	
Mn	50.39	44.56	74.12	55.27	1.09	0.76	0.52	0.47	
As	11.01	6.71	14.25	10.05	27.03	25.15	16.73	19.67	
Cd	2.03	16.37	50.98	42.75	586.81	651.96	154.29	104.31	
					625.08	687.74	175.93	129.16	

mainly because of the dissolving out of As in the oxidizable fraction (54.51%). Meanwhile, the generation of FeAsO_4 could lead to the re-sedimentation of As, and increased the portion of As distributed in the residual fraction after bioleaching (Wang *et al.* 2015).

Over 95% of Cd present in oxide and hydroxide (reducible fraction) was dissolved out from both the RSS and THS after bioleaching. The higher removal rate of Cd in the THS could be attributed to the increase of Cd existing in mobile fractions (acid-soluble/exchangeable and reducible fraction) after treatment; the content of Cd present in the mobile fraction was increased from 76.40% to 84.96% (Figure 1).

Risk assessment of heavy metals in sludge

The risk assessment results of RSS and THS before and after bioleaching are displayed in Table 3. Heavy metals in THS were assessed to have lower risk than those in RSS, except Cd according to RAC. Thermal hydrolysis treatment could decompose part of the EPS and reduce the pH of the sewage sludge, leading to the release of heavy metals in the acid-soluble/exchangeable fraction (Pilli *et al.* 2015; Choi *et al.* 2018). Furthermore, despite the dissolution percentage of Cr and As decreasing in the THS after bioleaching, the environmental risk of Cr, Mn, As and Cd in the bioleached THS was all alleviated compared with the heavy metals in the bioleached RSS due to the reduction of heavy metals in the acid-soluble/exchangeable fraction.

The E_r^i values' change of Cr, Mn, As and Cd was similar to those of RAC after thermal hydrolysis. After bioleaching treatment, the E_r^i values of the four heavy metals in raw sludge and thermal hydrolyzed sludge decreased significantly. Cd, with the highest toxicity, was reduced by 73.71% and 82.22%, respectively (compared with RSS). As shown in Table 3, the E_r^i value of Cd plays a major role in the MRI value. After bioleaching, MRI values of RSS and THS were reduced dramatically. The MRI value of

bioleached THS was lower than that of bioleached RSS, indicating that heavy metals in bioleached THS caused less environmental pollution than in bioleached RSS.

CONCLUSIONS

The concentrations of Cr, Mn and As in the sewage sludge was decreased after thermal hydrolysis treatment, because part of the heavy metals had been released to the water phase. But the concentration of Cd was increased due to the loss of organic matter in the sewage sludge. Thermal hydrolysis treatment could also change the chemical speciation of metals in the sludge. The concentrations of all the heavy metals detected (Cr, Mn, As and Cd) in the oxidizable fraction were decreased in the THS. Lower final pH of the bioleached THS and faster ORP increase during bioleaching duration of the THS were also found. The removal efficiency of the Mn and Cd was improved after thermal hydrolysis treatment, while the final concentration of Cr and As was higher in the bioleached THS than that in the bioleached RSS. However, the environmental risk of heavy metals in the bioleached THS was alleviated compared with that of the bioleached RSS. This suggests that proper treatment could reduce the environmental risk of sewage sludge.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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