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Behavior of solid matters and heavy metals during conductive drying process of sewage sludge

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Abstract: Behavior of solid matters and heavy metals during conductive drying process of sewage sludge was evaluated in a sewage sludge disposal center in Beijing, China. The results showed most of solid matters could be retained in the dried sludge after drying. Just about 3.1% of solid matters were evaporated with steam mainly by the form of volatile fatty acids. Zn was the dominant heavy metal in the sludge, followed by Cu, Cr, Pb, Ni, Hg, and Cd. The heavy metals in the condensate were all below the detection limit except Hg. Hg in the condensate accounted for less than 0.1% of the total Hg. It can be concluded that most of the heavy metals are also retained in the dried sludge during the drying process, but their bioavailability could be changed significantly. The results are useful for sewage sludge utilization and its condensate treatment.

Subjects: Environment & the City; Environmental Sciences; Technology

Keywords: sludge drying; heavy metals; solid matters; condensate; sewage sludge; disposal; utilization; mass balance; total organic carbon; total phosphorus

1. Introduction

With development of cities and swell in population, sewage sludge as a by-product in wastewater treatment is increasing sharply. The amount of sewage sludge was 30.4 million tons (moisture content of 80%) in 2010 in China, and it increased to about 40.8 million tons (moisture content of 80%) in 2013. This was expected to 47 million tons in 2015 (Chyxx net, 2014). Disposal of sewage sludge has become an environmental challenge.

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Anfeng Li, male, professor, 1976, deputy director for environmental engineering design division, Beijing Municipal Research Institute of Environmental Protection. His main research direction is the research and development and application of the technology of advanced treatment of wastewater, wastewater reuse, and waterbody remediation. With the rapidly increasing amount of sewage in China, he began to focus on the reuse of the sludge.

PUBLIC INTEREST STATEMENT

With the sharply increasing amount of sewage sludge, its disposal has become a worldwide environmental challenge. Thermal drying is one of the promising approaches in sludge disposal. Due to the abundant organic matter, N, P, and other plant nutrients, the application of dried sludge in agriculture is an attractive disposal option. However, it is considered to be a secondary pollution source of the soil and groundwater. In this study, behavior of solid matters and heavy metals during indirect conductive drying process of sewage sludge was evaluated to provide useful information on safe application of the sludge in agriculture. It is found that solid matter, total phosphorus, heavy metals, and total organic carbon could be almost retained in the dried sludge after drying. Furthermore, the drying process played an important role in heavy metal bioavailability due to pH increase, dehydration, pyrolyzation, and volatization of organic matter.

The application of sewage sludge in agriculture is an attractive option of disposal due to the abundant organic matter, N, P, and other plant nutrients. It has been an important disposal pathway in Australia, Europe, and China (Kakati, Ponmurugan, Rajasekaran, & Gnanamangai, 2013; Kelessidis & Stasinakis, 2012; Pritchard, Penney, McLaughlin, Rigby, & Schwarz, 2010). However, sewage sludge also contains many pathogens, heavy metals, persistent organic contaminants, and these sludge-derived contaminants in soil have the potential to be accumulated by plants and animals (Harrison, McBride, & Bouldin, 1999; Singh & Agrawal, 2010; Vrkoslavová et al., 2010). Sludge drying has become a necessity for agricultural application to reduce the volume of sludge, kill pathogens, and control toxic contaminants' bioavailability (Tunçal, Jangam, & Güneş, 2011). Now, sludge drying can be operated mainly by three modes: convective drying, conductive drying, and solar drying (Bennamoun, Arlabosse, & Léonard, 2013). In these processes, a large amount of steam would be evaporated. This could result in significant change of sludge properties, and consequently influence the speciation, transformation, and migration of contaminants in the sludge (McLaren & Clucas, 2001; Weng et al., 2014). Meantime, volatilization of contaminants especially for volatile ones can be expected to occur due to higher temperature maintained in convective drying or conductive drying (Chou, Lin, Hsien, Wey, & Chang, 2012; Cousins, Hartlieb, Teichmann, & Jones, 1997; Tunçal et al., 2011; Weng, Ji, Chu, Cheng, & Zhang, 2012). However, behavior of contaminants during drying process is still not very clear, and there has been little published research on behavior of metals during sludge drying. Moreover, attention is also needed to treat the condensate as a by-product of convective drying or conductive drying due to its little-known water quality characteristics now.

In this study, behavior and distribution of heavy metals and other solid matters during conductive drying of sewage sludge were investigated to provide useful information on sludge utilization and treatment of the condensate.

2. Materials and methods

2.1. Sample collection and pre-treatment

Dewatered sludge, dried sludge, and condensate samples were obtained from a sewage sludge disposal center in Beijing, China. The center receives dewatered sludge from different municipal wastewater treatment plants in Beijing, such as Qinghe, Jiuxianqiao, Beixiaohe. Dewatered sludge with the moisture of about 77.7% was treated by indirect conductive drying at 250°C with heated oil, and the dried sludge's moisture was about 29.5%. The dewatered sludge was viscous and odorless with a few visible impurities, while the dried sludge was granular with bad smell.

Sludge and condensate samples were continuously collected one time per day with seven replicates one time for one week for analysis of heavy metals, and general parameters such as pH, total organic carbon (TOC), and total phosphorus (TP) were also detected. The collected sludge samples including dewatered sludge and dried sludge were immediately air-dried, ground, and passed through a 100-mesh nylon sieve, then stored in dry glass bottles at room temperature. The condensate samples were stored in the sealed polyethylene bottle at 4°C in the dark and analyzed within one week.

2.2. Analytical methods

Sludge pH was measured in 1:10 (w/v) sludge and water suspension using a pH meter (Orion 3-Star, Thermo, USA). TP of the sludge was determined by molybdenum antimony colorimetric method, while TP of the condensate was detected by ammonium molybdate spectrophotometric method (721, Sunny Hengping scientific instrument Co., China). The moisture of sludge was determined by drying at 105°C to constant weight. TOC of the sludge was determined by oxidation with potassium dichromate (TOC-Torch, Teledyne-Tekmar, USA), while TOC of the condensate was analyzed by combustion oxidation non-dispersive infrared absorption method. Concentration of volatile fatty acids (VFAs) in the condensate was measured by titrimetric analysis with sodium hydroxide preceded by alkali distillation for ammonia removal and high purity nitrogen for hydrogen sulfide removal. Atomic absorption spectrometry (AAS) (GBC932 & GBC908AA, GBC, Australia) was used to determine the total metal concentrations of Cu, Zn, Pb, Cr, Ni, and Cd in the sludge (GB/T17138-1997, 1998). 0.5 g of the air-dried sludge was heated with 10-mL HCl (1.19 g/mL) at room temperature (25 ± 1°C). And when about 3-mL liquid was left, then 5-mL HNO_3 (1.42 g/mL), 5-mL HF (1.49 g/mL), and 3-mL $HClO_4$ (1.68 g/mL) were added and the mixture was heated at a middle temperature for 1 h (170 ± 2°C). Hg in the sludge was determined by atomic fluorescence spectrometry (AFS-2202E, Kechuang haiguang instrument Co., China) (GB/T22105-2008, 2008). 0.5 g of the air-dried sludge was digested with 10-mL (1 + 1) aqua regia in the boiling water bath for 2 h. Cu, Zn, Pb, Cr, Ni, and Cd in the condensate were determined by AAS with 100-mL water samples being digested with 5-mL HNO_3 (1.42 g/mL), then 5-mL HNO_3 (1.49 g/mL), and 2-mL $HClO_4$ (1.68 g/mL) were used for Cd, Cu, Zn, and Pb (GB7475-87, 1987), 5-mL HNO_3 (1.49 g/mL) and 2-mL H_2O_2 (30%) for Cr (HJ 757-2015, 2015), and 5-mL HNO_3 (1.49 g/mL) for Ni (GB11912-89, 1989). Hg in the condensate was detected by AFS with 50-mL water samples being digested with 5-mL (1 + 1) HNO_3 (1.42 g/mL)-$HClO_4$ (1.68 g/mL), and 10-mL $KMnO_4$ (4%) (Ministry of Environmental Protection of the People's Republic of China, 2002).

The mass balance chart was calculated and developed with the content and weight of elements before and after drying (Steger & Meisner, 1996).

3. Results and discussion

3.1. Mass balance of the drying process

The mass balance during the conductive drying process of sewage sludge in this study is shown in Figure 1. 500.0 tons per day (t/d) of dewatered sludge with a solid content of 22.3% was fed into the dryer. 153.2 t/d of dried sludge with a solid content of 70.5% and 346.8 t/d of evaporated fraction were produced. Therefore, about 97% of solid matters were retained in the dried sludge and only 3.5 t were pyrolyzed and evaporated with steam during the drying process. About 98.1% of the evaporated fraction (340.3 t/d) could be condensed and the residual 1.9% (6.5 t/d) was considered as non-condensate. More than 95% of the non-condensate was water vapor. Thus, most of the evaporated solid matters were transferred into the condensate.

The concentration and gross amount of VFAs in the condensate were 6,840.3 mg/L and 2.3 t/d (Table 1 and Figure 1), respectively, which accounted for about 67.0% of the evaporated solid matters. It was indicated that VFAs should be one main component of the evaporated fraction and condensate besides water, which were mainly formed through pyrolyzation of the lipids and proteins in the sludge (Bougrier, Delgenès, & Carrère, 2008; Deng et al., 2009; Shanableh & Jones, 2001). Due to the large amount of VFAs, the condensate was acidic, and the pH was 5.2. Accordingly, the sludge pH increased from 6.7 to 7.0 after drying, and about 6.9% of TOC was evaporated and concentrated in the condensate. About 50% of sulfide in the dewatered could be volatilized into the condensate, and the residual was retained in the dried sludge (Table 1 and Figure 1). There was no significant content difference of TP concentration between dewatered and dried sludge (Table 1). The amount of TP in dried sludge accounted for about 99.1% of that in the dewatered sludge, and the amount of TP in condensate was negligible.

Figure 1. Mass balance during conductive drying process of sewage sludge.

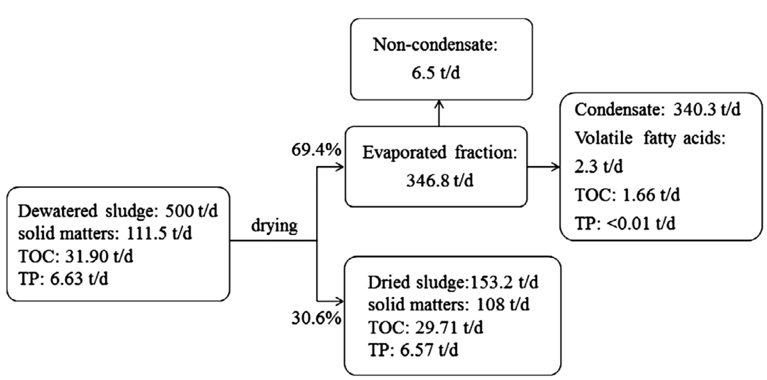

Table 1. General parameters of the dewatered sludge, dried sludge, and condensate

Samples	pH	TOC (g/kg DS)	TP (g/kg DS)	Solid content (%)	VFAs (mg/L)	Sulfide (S^{2-}) (mg/kg DS)
Dewatered sludge	6.7 ± 0.2	286.1 ± 10.72	59.5 ± 3.93	22.3 ± 0.59	–	0.031 ± 0.009
Dried sludge	7.0 ± 0.1	275.1 ± 7.70	60.8 ± 0.96	70.5 ± 1.28	–	0.019 ± 0.002
Condensate[a]	5.2 ± 0.1	4,860.6 ± 79.72[a]	8.9 ± 1.18[a]	–	6,840.3 ± 104.49	0.008 ± 0.001

[a]Units of TOC, TP, and Sulfide in condensate are mg/L.

3.2. Concentrations of heavy metals in the dewatered sludge

The highest concentrations of 469.5 and 82.7 mg/kg DS were observed for Zn and Cu in the dewatered sludge, respectively, and followed by Cr, Pb, Ni, Hg, and Cd (Table 2). Metal concentrations in the dewatered sludge were all below Chinese permitted limits of pollutants for sludge to be used for agriculture (GB18918-2002, 2003) in neutral or alkaline soil (pH ≥ 6.5). Only Hg concentration was higher than the limit for agriculture in acid soil (pH < 6.5). The concentration order of heavy metals corresponded well with the results discovered by Dai, Xu, Chen, Yang, and Ke (2007) in sludge from six wastewater treatment plants in Beijing. However, concentrations of the heavy metals became significantly lower. Two reasons may explain the lower level of the metals in the sludge. One is that galvanized pipe has been partially replaced by polyvinyl chloride polymer, polypropylene, and stainless steel tubes in wastewater pipeline in Beijing. This could reduce the release of Zn into wastewater before treatment. The other reason is that use of clean production technologies and separation of industrial wastewater from domestic sewerage networks are very universal, and sewage sludge is typically domestic in origin. Meantime, stricter discharged standard of heavy metals in China has been enforced recently and leads to lower concentration in discharge.

3.3. Behavior of heavy metals during drying process

Like TP, no significant difference of metal concentrations was found in dewatered and dried sludge (Table 2). This was different from composting, digestion, and liquefaction processes. Because during these processes, a mass of organic matters could be decomposed and mineralized, concentrations of heavy metals would increase accordingly (Dong, Liu, Dai, & Dai, 2013; Wagner, Bacon, Knocke, & Switzenbaum, 1990; Yuan et al., 2011). By rough calculation of mass flow, amounts of heavy metals in the dried sludge accounted for about 88–111% of those in the dewatered sludge (Table 3). Meanwhile, the heavy metals were hardly detected in condensate except Hg. Moreover, Hg concentration was very low (Table 2). Only less than 0.1% of Hg in the dewatered sludge could be migrated

Table 2. Total concentrations of heavy metals in dewatered sludge, dried sludge (mg/kg DS), and condensate (mg/L)

Heavy metal	Dewatered sludge	Dried sludge	Condensate	pH < 6.5[b]	pH ≥ 6.5[b]
Zn	469.5 ± 24.7	503.7 ± 31.6	<0.005[a]	2,000	3,000
Cu	82.7 ± 5.1	78.1 ± 4.3	<0.001[a]	800	1,500
Cr	35.1 ± 2.0	32.9 ± 3.6	<0.03[a]	600	1,000
Pb	27.5 ± 2.3	30.6 ± 2.6	<0.01[a]	300	1,000
Ni	12.6 ± 1.4	11.5 ± 0.8	<0.05[a]	100	200
Hg	6.5 ± 0.3	6.6 ± 0.5	0.0014	5	15
Cd	0.14 ± 0.01	0.16 ± 0.02	<0.001[a]	5	20

[a]Lower than detection limit.

[b]Chinese permitted limits for agricultural application of sludge in acid soil (pH < 6.5) and neutral or alkaline soil (pH ≥ 6.5).

Table 3. Mass balance of heavy metals during drying (g/d)

Samples	Zn	Cu	Cr	Pb	Ni	Hg	Cd
Dewatered sludge	52,349.3	9,221.1	3,913.7	3,066.3	1,404.9	724.8	15.6
Dried sludge[a]	54,399.6 (103.9%)	8,434.8 (91.5%)	3,553.2 (90.8%)	3,304.8 (107.8%)	1,242.0 (88.4%)	712.8 (98.3%)	17.3 (110.9%)
Condensate	<1.70[b]	<0.34[b]	<10.21[b]	<3.40[b]	<17.02[b]	0.48	<0.34[b]

[a]Mass percentages of the heavy metals in dried sludge to ones in dewatered sludge are shown in brackets.
[b]Lower than detection limit.

to the condensate. It was indicated heavy metals during the drying process were difficult to be volatilized from the sludge, and would be almost retained in the dried sludge.

The condensate was also found to be severely lack of trace elements such as Mn and Co, which could significantly affect the activity and settleability of activated sludge in the biological treatment system (data not shown). Therefore, it is necessary to take appropriate measures in condensate biological treatment to avoid trace element deficiency and secondary pollution by volatile metals such as Hg (Zorpas, Vlyssides, Zorpas, Karlis, & Arapoglou, 2001).

Although metals were retained in dried sludge during drying process, their bioavailability could be changed significantly. As well known, the speciation of heavy metals in sludge includes exchangeable/acid soluble fraction, reducible fraction, oxidizable fraction, and residual fraction (Tessier, Campbell, & Bisson, 1979). First, the pH value of the dried sludge increased in this study, probably due to release of a large amount of VFAs. With pH increasing, the mobility and bioavailability of heavy metals might decrease (Wang, Li, Ma, Qian, & Zhai, 2006). Second, dehydration during drying process was an important factor causing the conversion of acid soluble and reducible fractions into the residual form of the heavy metals due to precipitation of heavy metals with minerals and change of iron/manganese amorphous oxyhydroxides to stable crystalline forms (Weng et al., 2014). Third, organic matter and sulfide were found to exhibit a significant positive correlation not only with exchangeable and reducible fractions of heavy metals but also with oxidizable fraction (Wang et al., 2006). Thus, the volatilization of sulfide and pyrolyzation of organic matter, and evaporation of resulting VFAs were one important factor causing conversion of exchangeable, reducible, and oxidizable fractions into residual one. Overall, these changes of sludge and heavy metal speciation indicated that the heavy metals in the dried sludge might be more stable than in the dewatered sludge (Obrador, Rico, Alvarez, & Novillo, 2001; Zorpas et al., 2001).

4. Conclusion
Solid matter, TP, heavy metals, and TOC were almost retained in the dried sludge after indirect conductive drying process of sewage sludge. However, due to the increase in pH, the dehydration and volatilization of sulfide, pyrolyzation of organic matter, and evaporation, the indirect conductive drying process might play an important role in reducing heavy metal bioavailability. Furthermore, as numerous VFAs and little metals during the thermal drying could be transferred into the condensate, appropriate pre-treatment might to be used for its biological treatment.

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