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RESEARCH ARTICLE

The contents and release behavior of heavy metals in construction and demolition waste used in freeway construction

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Abstract

A large volume (more than 4.0 million m^3) of treated construction and demolition waste (CDW) is planned to be used in the construction of the Xi'an-Xianyang north loop line freeway in West China. These CDW were preliminarily separated into broken concretes, bricks, and porcelains in the treatment plants. In this study, a total of 190 CDW samples including 80 concretes, 80 bricks, 20 porcelains, and 10 mixed samples were collected from five treatment plants. Five farmland soil samples near treatment plants were collected as controls. The contents of 10 elements including cadmium (Cd), arsenic (As), copper (Cu), nickel (Ni), zinc (Zn), chromium (Cr), lead (Pb), manganese (Mn), silver (Ag), and mercury (Hg) in these samples were measured. The contents of 8 elements (Cu, Ni, Zn, Cr, Pb, Mn, Ag, and Hg) in all CDW samples were qualified for the third-level criterion of the Standard of Soil Environment (GB15618-2008). However, Cd contents in 37 concretes, 34 bricks, 6 porcelain samples, and 4 mixed CDW samples exceeded the national third-level standards (1 mg/kg) in GB15618-2008. And As contents in 28 concretes, 21 bricks, 5 porcelain samples, and 3 mixed CDW samples were higher than the national third-level standards (40 mg/kg). The total exceeding standard rates (ESRs) of Cd and As were 42.6% and 30%, respectively. The leaching tests for Cd and As were also done due to their higher ESRs. The results showed that the release amounts (μg/kg) of Cd and As from CDW were increased with increasing liquid to solid ratio (0.4–10 l/kg) but decreased with increasing pH (4–7). The leached concentrations of Cd and As from four types of CDW samples were both in a descending order: brick, mixed materials, concrete, and porcelain. The measured concentrations (μg/L) of Cd and As in leachate were all lower than second-grade criteria of Standard for Groundwater Quality (GB3838-2002). By comparing the leached concentrations of Cd and As with the value in European criteria (EU Council Decision 2003/33/EC) for hazardous wastes, all the CDW samples should be classified as inert or non-hazardous wastes. Thus, it could be concluded that heavy metals in these CDW would not pollute surrounding soil, surface water, and groundwater environment when applied in freeway construction.

Keywords Release behavior · Heavy metals · Construction and demolition wastes · Freeway construction

Introduction

Heavy metals are persistent and accumulative, which can pose potential risks to the ecosystem, food safety, and human health (Liu et al. 2015; Liang et al. 2017; Mwesigye et al. 2016; Huang et al. 2019). Heavy metals' concentration, behavior, source, and potential risk to ecological systems in surface water (Yin et al. 2015; Ezemonye et al. 2019), groundwater (Ravindra and Mor 2019; Wen et al. 2019), soil (Huang et al. 2019; Ashraf et al. 2019), sediment (Liang et al. 2019; Sun et al. 2019), and organisms (Ezemonye et al. 2019; Milenkovic et al. 2019) have always been a research hotspot all over the world.

As the rapid development of building industry, China has been the country with the largest production of CDW (construction and demolition waste) in the world (Xiao et al. 2012; Ding and Xiao 2014). It is estimated that, 150 million tons of CDW has been generated annually in China, most of which

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was delivered to suburban or rural areas for simple disposal of landfill. Increasing accumulation of CDW has been an important problem for China (Yuan 2013).

For environmental and cost reasons, CDW reusing has received great attention, especially in road construction (Lidelöw et al. 2017). Some studies have shown that treated CDW is suitable for road construction owing to their stabilized physicochemical properties and mechanical behaviors (Poon and Chan 2006; Roussat et al. 2008; Vegas et al. 2008; Lidelöw et al. 2017). Most treated CDW can be classed as inert or non-hazardous materials. However, some other CDW contains hazardous substances, such as heavy metals (Yu et al. 2018). When these CDW materials are applied to road construction, if the road surface or road bed is damaged, water percolating and runoff will lead to the leaching of heavy metals in CDW into the surface water, soil, and groundwater, which would induce toxic effects on the ecosystem and human health.

The environmental impact caused by the materials in question is not determined by their total content of pollutants, but by the amount of pollutants that water can dissolve and leach into the soil, thereby reaching the surface and/or underground water (Galvín et al. 2012; Lidelöw et al. 2017). Therefore, the total leachability of pollutant in the waste and the ecotoxicity of the leachate must be controlled to prevent risks to the quality of surface water or groundwater (Roussat et al. 2008; Wahlstrom et al. 2000; Galvín et al. 2012, 2013). Batch leaching tests are considered a suitable tool for quality control reasons, while percolation tests are frequently used for estimating release from solid materials under field site. Recently, some studies have focused on concentrations, release behaviors, and environmental risk of heavy metals in CDW (Townsend et al. 2004; Delay et al. 2007; Galvín et al. 2012, 2013, 2014; Butera et al. 2014, 2015; Robey et al. 2018). Townsend et al. (2004) measured 11 heavy metals in CDW debris from Florida and found that arsenic presented the greatest limitation to reuse. And the results of batch leaching tests obtained using simulated rainwater (mg/L) were compared directly with risk-based groundwater levels for Florida and were found not to pose a risk. Butera et al. (2014, 2015) analyzed the inorganic elements and organic compounds in CDW by standard up-flow column and down-flow lysimeter leaching tests. Among the different leaching parameters on the release of metals from recycled aggregates, pH is the most relevant factor due to its strong control on the pollutant release. And the recycled aggregates can be classified as inert wastes or non-hazardous material (Galvín et al. 2012, 2013, 2014). Above all, although some papers reported the concentrations and risk of heavy metals in debris, wood, screened soil, and recycled aggregates from CDW, little research on heavy metals in broken bricks and porcelain was reported. And comparative studies on contents and release behaviors of heavy metals in different types of CDW in the same area were very few.

Xi'an City, located in the Guanzhong Plain, western China, which is regarded as one of the world's four greatest ancient cities, has more than 10.0 million population and 3.0 million motor vehicles at the end of 2018. This city is bearing a great burden on the urban traffic. Xi'an-Xianyang north loop line freeway is the second Xi'an Ring Expressway, which would greatly alleviate traffic pressure. More than 4.0 million m^3 CDW is planned to apply in construction of this freeway, which is the most large-scale application of CDW in freeway construction in the world. On the whole, these CDW used in Xi'an-Xianyang north loop line freeway consists of broken concrete, brick, and porcelain. The components and sources of CDW from different areas are various. Climatic and geological variability will lead to different physicochemical properties of these materials in different areas, which will affect their leaching mechanisms and environmental effect.

Therefore, the main goals of the present work were (1) to provide experiment data of contents of 10 elements and identify the main heavy metals pollutants in different types of CDW in Guanzhong Plain, China; (2) to identify the factors (liquid to solid ratio, pH) that affected their pollutant emission in different components in CDW; and (3) to estimate the environmental effects of Cd and As in CDW applied in the construction of the Xi'an-Xianyang north loop line freeway.

Materials and methods

Sample collection

As Fig. 1 has shown, a total of 190 samples including 80 concretes, 80 bricks, 20 porcelain, and 10 mixed material samples were collected from 5 treatment plants along with Xi'an-Xianyang north loop line freeway. Five farmland soil samples near the 5 treatment plants were also collected. The particle size of all samples is less than 3 cm. The mixed materials consisting of crushed concrete, crushed bricks, and porcelain accounts for 55%, 40%, and 5% of total mass, respectively. At each sampling point, two additional subsamples were collected to increase representativity. After being transported to laboratory, all samples were crushed and sieved through a 0.3-mm fraction, then homogenized and reduced in the laboratory using a quartering method. Samples were dried at 40 °C for 2 days, then stored in desiccators during the whole experimental period.

Total contents of elements

The four types of CDW samples and farmland soil samples were dissolved in three-acid mixture in four Teflon beakers,

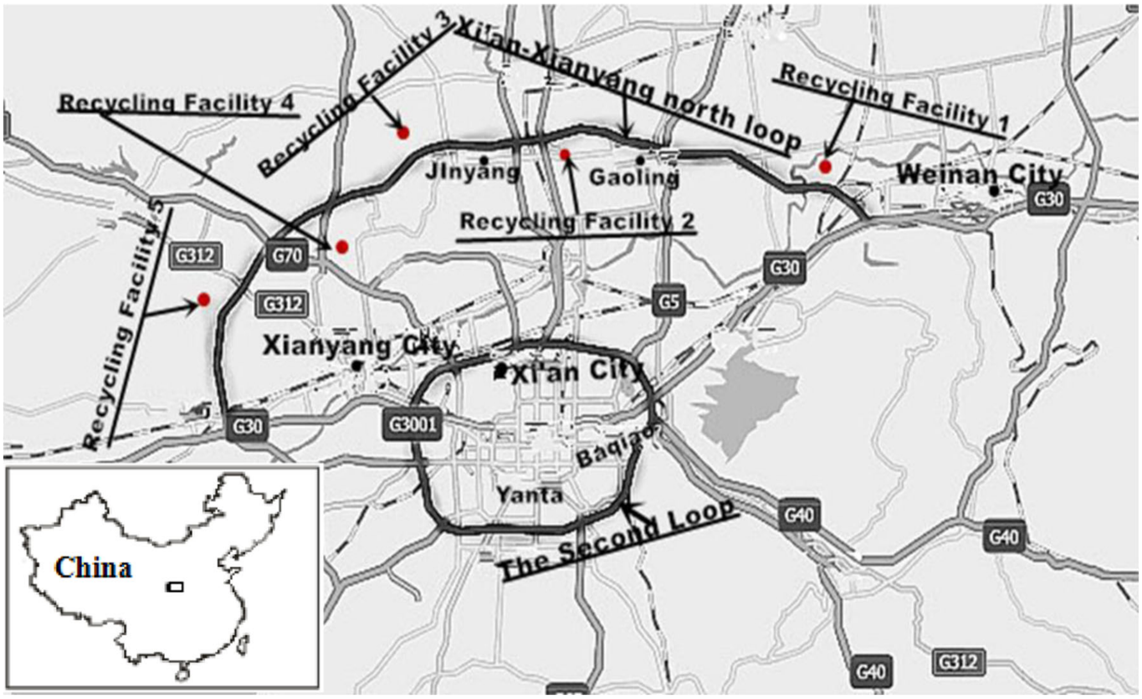

Fig. 1 Position of the Xi'an-Xianyang north loop line freeway and treatment plant

respectively. After the addition of concentrated hydrochloric acid (HCl), nitric acid (HNO_3), and hydrofluoric acid (HF), the mixture was heated until almost dry. The residue was dissolved by HNO_3 (0.2 mol/L), then transferred into volumetric flasks. Total elemental contents of these samples were determined by inductively coupled plasma mass spectrometry (ICP-MS) (Agilent-7500 CS). Parallel samples and replicated analysis were also conducted to ensure accuracy. All glassware was acid rinsed with HNO_3 (0.2 mol/L) before usage; all reagents used for analysis were of guaranteed reagent.

Leaching tests

In this study, four samples with very similar Cd contents (2.07–2.13 mg/kg) from each type of CDW were chosen to conduct leaching tests of Cd. And four samples with very similar As contents (67.58–68.24 mg/kg) from each type of CDW were chosen to conduct leaching tests of As element.

Leaching test as a function of liquid to solid ratio

The test was performed with distilled water and without any pH adjustment during the test. When the L/S ratio was less than 0.4, the lixivium was very difficult to obtain according to preliminary experiment. So, L/S ratios chosen were 0.4, 0.55, 0.7, 0.85, 1, 2, and 10 l/kg, respectively. The solid-water system, for each of three replicate series, was shaken for 18 h, centrifuged at 3000 rpm for 10 min, and then the liquid supernatant was filtered (0.45 μm). The obtained leaching solutions were analyzed by the ICP-MS. The test was performed with distilled water and without any pH adjustment during the experiment.

Leaching test as a function of pH

The pH-dependent test was defined to examine pollutant solubility release as a function of pH. In this experiment, pH-dependent set-points were pre-defined (pH 4–7). Ten grams of brick, concrete, porcelain, and mixed material samples were put into 4 Erlenmeyer flasks together with 100 mL of solutions (pH 4–7) and distilled water (pH 7), respectively, shaken for 18 h, and centrifuged for 10 min (3000 rpm). Then, the liquid supernatant was filtered through a 0.45-μm membrane filter. The obtained leaching solutions were analyzed by the ICP-MS. The pH of water-solid system was kept constant by feedback control and the addition of HNO_3.

Results and discussion

Total element analysis

The total contents of elements of Mn, Ag, Hg, Cu, Ni, Zn, Cr, Pb, Cd, and As in CDW samples are shown in Table 1. Among these elements, the concentrations of Mn, Ag, and Hg were less than their detection limits (Mn, 0.05 mg/kg; Hg, 1.0 mg/kg; Ag,

Table 1 Total contents of heavy metals in four types of materials

Metals (mg/kg)	Brick (*n* = 80)		Concrete (*n* = 80)		Porcelain (*n* = 20)		Mixed materials (*n* = 10)		Farmland soil (*n* = 5)		Limit value[a]	ESR[b] (%)
	Range	Average	Range	Average	Range	Average	Range	Average	Range	Average		
Mn	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	< 0.05	—[c]	0.00
Ag	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	–	0.00
Hg	< 1.0	< 1.0	< 1.0	< 1.0	< 1.0	< 1.0	< 1.0	< 1.0	< 1.0	< 1.0	1.5	0.00
Cr	71.6–131	107	20.8–95.2	71.2	39.5–248	132	32.5–96.9	81.6	95.3–115	110	300	0.00
Cu	25.3–38.7	27.0	9.7–60.5	19.2	5.81–55.9	44.5	19.2–27.7	22.2	28.5–40.2	36.8	400	0.00
Zn	68.3–183	96.8	26.5–208	66.7	30.0–486	194	64.1–103	82.6	89.4–111	101	500	0.00
Ni	40.0–47.1	42.9	10.9–23.9	19.2	12.6–109	20.6	18.1–27.6	22.5	25.3–61.2	42.2	200	0.00
Pb	14.5–244	47.6	7.9–238	58.7	1.34–150	50.8	32.9–66.0	48.3	34.2–40.2	37.3	500	0.00
As	12.8–229	38.6	4.58–626	77.8	1.42–113	21.2	7.07–78.3	45.7	34.2–44.4	39.5	40	30.0
Cd	0.29–3.7	1.27	0.16–10.4	1.68	0.25–3.18	1.05	0.19–3.81	1.38	0.89–1.82	1.26	1	42.6

[a] Limitation of third-level standard of the China Environmental Quality Standard for Soils

[b] Exceeding standard rates

[c] There is no limit value for Mn and Ag in standard of the China Environmental Quality Standard for Soils

0.2 mg/kg) and were negligible. The concentrations obtained were compared with the limit values from the China Environmental Quality Standard for Soils (GB 15618- 2008). The concentration range of Cd in concrete, brick, porcelain, and mixed samples were 1.68 mg/kg, 1.27 mg/kg, 1.05 mg/kg, and 1.38 mg/kg, respectively, which were all higher than the limitation (1 mg/kg) of third level. And their exceeding standard rates (ESRs) were 46.25%, 42.25%, 30%, and 40%, respectively. Compared with Cd concentration in CDW from other areas in the word, the result of this study is at a moderate level. The mean values of Cd in concrete, brick, porcelain, and mixed samples in this study are all lower than the results of Nakamura et al. (1996) (mean 2.8 mg/kg), Chandler et al. (1997) (mean 3.8 mg/kg), and Townsend et al. (2004) (mean ± SD 2.0 ± 1.7 mg/kg). However, the result of this study is higher than those of Yu et al. (2018) (< 0.3 mg/kg) and Rotter et al. (2004) (mean 1 mg/kg). The average contents of As in concrete, brick, porcelain, and mixed samples were 77.8 mg/kg, 38.6 mg/kg, 21.2 mg/kg, and 45.7 mg/kg, and their ESRs were 35%, 26.25%, 25.0%, and 30%, respectively. The mean values of As in brick and porcelain samples were lower than its limit value (40 mg/kg), while those in concrete and mixed material samples exceeded its limit value. Compared with results of other researches, As concentration in this study is in a high level, which is higher than the results of Yu et al. (2018) (< 7 mg/kg) and Townsend et al. (2004) (mean ± SD 4.4 ± 6.1 mg/kg). The total ESRs of Cd and As were 42.6% and 30%, respectively. For the remaining elements (Cr, Cu, Zn, Ni, and Pb), their highest concentrations in all the samples were lower than the corresponding threshold values. Thus, Cd and As could be considered main heavy metal pollutants in these CDW. So, it is necessary for further study to obtain leaching behaviors and environmental effect of Cd and As elements.

The mean content (39.5 mg/kg) of As in farmland soil samples was very close to third-level standards in GB15618-2008 and the mean content (1.26 mg/kg) of Cd was even higher than its limited value. Local soil was one of the main raw materials to produce brick, porcelain, and concrete indirectly or directly, which may be one of the reasons why the total contents of Cd and As in these materials were at a higher level.

Liquid to solid ratio analysis

Leached concentrations of Cd and As from four types of samples as a function of liquid to solid ratio were given in Fig. 2. From the results, it can be found that Cd and As leached concentrations were both in a descending order: brick, mixed materials, concrete, and porcelain. The released concentrations (μg/L) of Cd and As were decreased as L/S ratio increased. This result was in line with many previous studies (Delay et al. 2007; Garrabrants et al. 2004; Roussat et al. 2008). This decrease might be due to the reduction of exchange area and depletion of the elements within the porous structure, which can cause decrease of heavy metals into water (Roussat et al. 2008).

The unit of released concentrations was converted from μg/L to μg/kg for further analysis. Figure 3 illustrates the leached contents (μg/kg) of Cd and As at seven different L/S ratios. From Fig. 3, the leached concentrations of Cd and As had the similar changing tendency, and their concentrations increased with increasing L/S ratio from 0.4 to 10 L/kg. The results were consistent with the reports of other authors (Galvín et al. 2012, 2013; Allegrini et al. 2015). Galvín et al. (2013) analyzed the leaching behaviors of heavy metals through three different leaching methods and found that

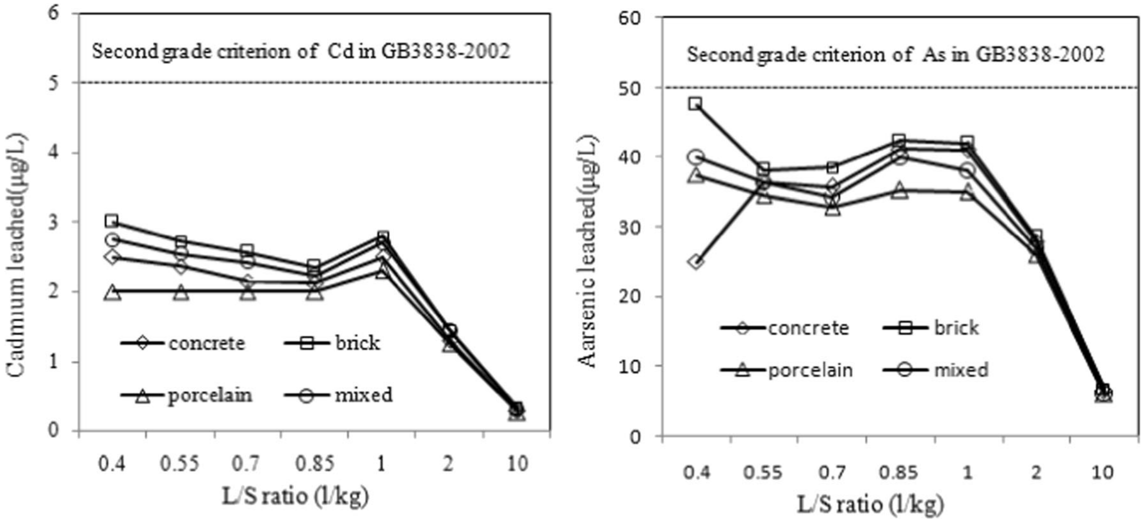

Fig. 2 Leached concentrations in accordance with the L/S ratio (without any pH adjustment)

element-leached concentrations increased with L/S ratio increasing. For most heavy metals, characterization of leaching behaviors shows similar pattern as a function of L/S ratio, despite the considerable difference of the test procedure. It suggested that the metal element would be more easily leached from the solid at high L/S ratio.

pH analysis

As recently studies reported, pH plays a crucial role in determining the solubility of heavy metals (Galvín et al. 2012), and largest amounts of contaminants were leached at low pH values (at pH < 7) (Townsend et al. 2003, 2004). In general, recycled materials from CDW were often with high initial natural pH. However, carbonation and acid rain will cause considerable loss of alkalinity and decrease the pH value when they are exposed. Furthermore, pH value of recycled materials in field site was much lower than laboratory conditions (Delay et al. 2007; Roussat et al. 2008). In this study, aggressive conditions were imposed to better identify the effect of pH on release behaviors of Cd and As from CDW. The leaching experiments of four types of CDW samples were performed under pH range from 4 to 7. This pH range is often regarded as the worst-case scenario compared with the real situation in which a recycled material is applied in road construction (NEN 7341 1994).

In Fig. 4, pH-dependent release of Cd and As were shown. In spite of the different types of CDW samples, the leaching trends showed very similar characteristics according to the pH increase. It was found that the leachate concentrations of Cd and As exhibited equilibrium values that depend strongly on leachate pH.

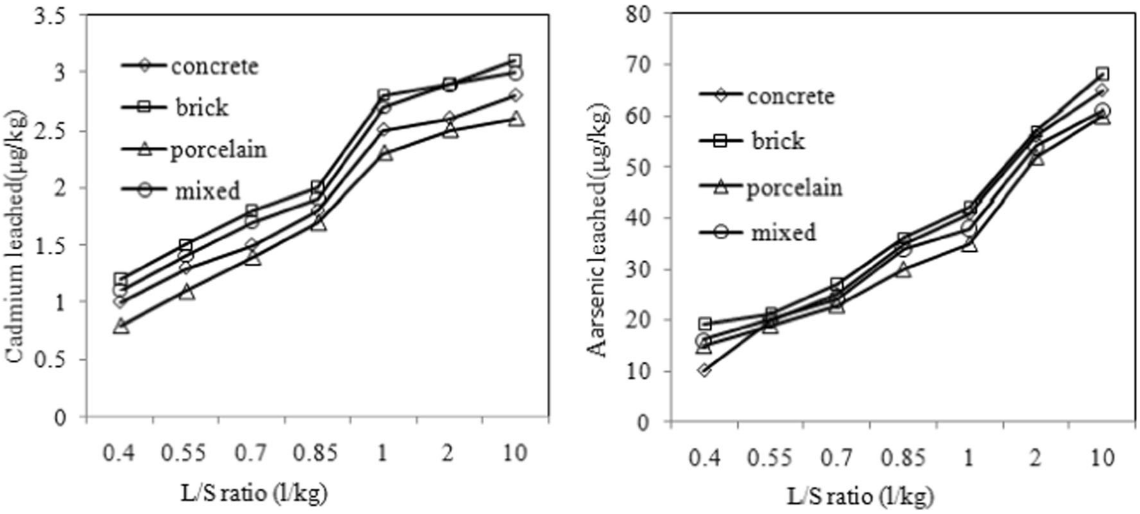

Fig. 3 Leached concentrations in accordance with the L/S ratio (without any pH adjustment)

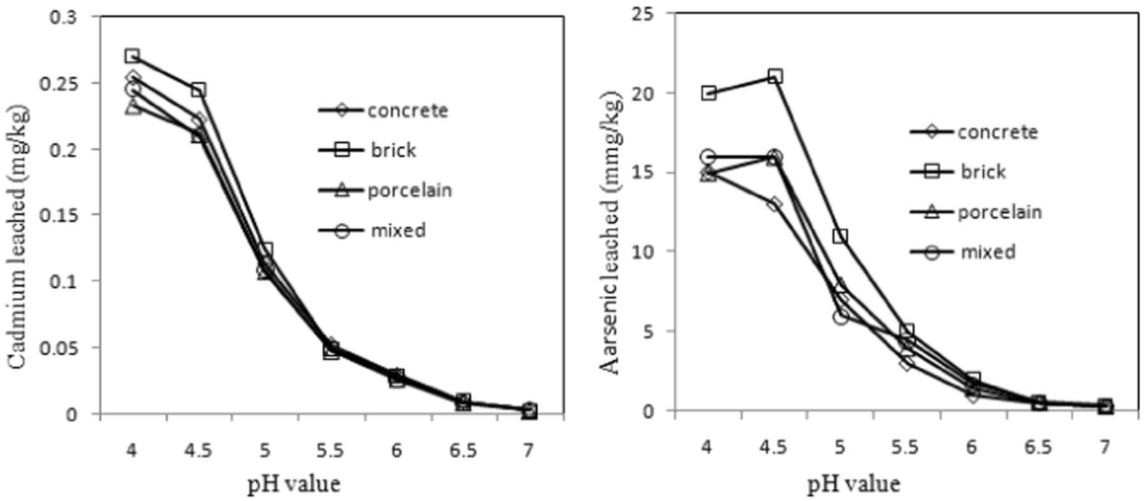

Fig. 4 Release of Cd and As for different types of materials as a function of pH(L/S = 10 l/kg)

This research study observed that leached concentrations of Cd and As were both decreased with pH increasing. This result was in line with previous findings at lower pH value (pH < 7) (Townsend et al. 2003; Garrabrants et al. 2004). Zhao and Huang (2017) also found that the concentration pattern and cumulative release of Cu, Cr, and Cd were arranged in the order of neutral rain (pH = 4.8) < weak acid rain (pH = 5.8) < strong acid rain (pH = 3.8) condition. Cd and As were changed to corresponding cations and (oxy) anions in their leachate, respectively (Mulugeta et al. 2011). Both (oxy) anion and cations were decreased with increasing pH at lower pH. While at high pH, only the (oxy) anion increased with pH increasing (Engelsen et al. 2009, 2010). Furthermore, release of metal cations and (oxy) anions was affected by interactions with different constituents within solid materials (Engelsen et al. 2009, 2010). Metal leaching behavior was mainly controlled by metal hydroxide solubility (Li et al. 2001).

It is also worth to note that the released concentrations of Cd and As showed obvious difference between the four types of samples. Although the contents of Cd and As in four types of samples were very close, their leached concentrations showed a visible difference. Cd and As were most easily released from brick samples than other type of samples in this study. CDW is a highly heterogeneous material and several types of interactions may occur simultaneously in the solidified systems (Cocke and Mollah 1993). Released properties of metals were significantly influenced by heterogeneous compositions (Van der Sloot et al. 2002). Previous literatures suggested that the presence of masonry might affect mineral solubility to some extent (Alonso-Santurde et al. 2010; Galvín et al. 2013; Butera et al. 2014, 2015). The different released concentrations of Cd and As in four types of materials may be caused by the difference of their sources, production process, and heterogeneity.

Environment effect

When CDW was applied in road construction, environment effect was caused by the amount of pollutants that water can dissolve and leach into the soil, thereby reaching the surface and/or underground water. Although high ESRs of Cd and As were found in each types of CDW samples, their leached concentrations were lower than second-grade criteria of the Standard for Groundwater Quality (GB3838-2002) (see Fig. 2).

In order to further identify the environmental effect of each type of CDW by leaching test, the European criteria (EU Council Decision 2003/33/EC, 200) (Table 2) were used as a reference. Wastes can be classified as inert, non-hazardous, and hazardous materials based on results of leaching tests according to the European criteria. By comparing the data in Fig. 4 with European criteria (Table 2), released concentrations of Cd and As from the test materials were all lower than the limit value of inert or non-hazardous, so the CDW materials in this study can be classified as inert or non-hazardous materials.

Results above suggested that all these materials could be regarded as inert or non-hazardous materials for their leaching behaviors, regardless of liquid to solid ratio or pH values. It could be concluded that heavy metals in these CDW would not cause environment pollution when applied in freeway construction.

Table 2 Acceptance criteria (μg/L) of EU Council Decision 2003/33/EC(L/S = 10 l/kg)

	Inert	Non-hazardous	Hazardous
Cd	0.4	10	50
As	5	20	250

Conclusions

In recent years, the utilization of CDW in construction industry, specifically in road pavements, embankments, backfills around buried pipes, and incorporating them as recycled aggregates in the manufacture of non-structural concrete, has gained widespread attention (Juan-Valdés et al. 2018; Cardoso et al. 2016; Tavira et al. 2018; Barbudo et al. 2012; Rahman et al. 2013; Kianimehr et al. 2019; Zhang et al. 2019).

In this study, following an extensively sampling campaign of CDW in Xi'an City, a total of 190 samples from five treatment plants were investigated for quantification of 10 heavy metal contents. Regarding their total concentration in CDW, 8 heavy metals (Mn, Ag, Hg, Cu, Ni, Zn, Cr, and Pb) would not result in pollution to surrounding environment. However, high ESRs of Cd and As were observed in this study, which suggested that Cd and As were major heavy metal pollutants in CDW in this area. By comparing the leached concentrations of Cd and As with Chinese and European criteria, all of the four types of CDW in this study should be classified as inert or non-hazardous wastes. Thus, as it was expected, all analyzed heavy metals would not pollute surrounding environment from the obtained data when applied in freeway construction.

CDW accounts for around 40% of total urban waste in mainland China (China Strategic Alliance of Technological Innovation for Construction Waste Recycling Industry or CSATICWRI 2014). The average recycling rate of CDW in China is only about 5%, so there is a high potential for recycling of CDW in China. Many studies (e.g., Jin et al. 2017; Duan et al. 2015; Huang et al. 2018) have recognized the huge benefits of recycling of CDW, so recycling is often cited as the best way to manage CDW (Silva et al. 2014). Despite the widely recognized benefits, the sustainable management of CDW are facing many barriers (Jin et al. 2017; Duan et al. 2015; Huang et al. 2018; Domingo and Luo 2017; Jia et al. 2017). Based on these literature review and stakeholder interview, strategies for improving CDW management in China are proposed.

Although the bulk of CDW is non-hazardous, there are environmental problems during demolition and disposal phases. Each approach to dispose mixed CDW has potential environmental impact. However, it has not been fully characterized by Chinese researchers (Duan et al. 2015). Not only economic and social benefits but also environmental impacts must be considered in future CDW management in China. CDW must be first characterized to examine potential risks to human health and the environment before disposal.

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