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An anaerobic incubation study of metal lability in drinking water treatment residue with implications for practical reuse

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HIGHLIGHTS

- The lability of 18 metals in WTR was studied using an anaerobic incubation test.
- Most metals in WTR were stable.
- Certain metals (e.g., Mn) lability in WTR increased substantially.
- Metal lability in WTRs was recommended to be determined before application.

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ABSTRACT

Drinking water treatment residue (WTR) is an inevitable by-product generated during the treatment of drinking water with coagulating agents. The beneficial reuse of WTR as an amendment for environmental remediation has attracted growing interest. In this work, we investigated the lability of Al, As, Ba, Be, Ca, Cd, Co, Cr, Cu, Fe, Mg, Mn, Mo, Ni, Pb, Sr, V and Zn in Fe/Al hydroxide-comprised WTR based on a 180-day anaerobic incubation test using fractionation, in vitro digestion and a toxicity characteristic leaching procedure. The results indicated that most metals in the WTR were stable during anaerobic incubation and that the WTR before and after incubation could be considered non-hazardous in terms of leachable metal contents according to US EPA Method 1311. However, the lability of certain metals in the WTR after incubation increased substantially, especially Mn, which may be due to the reduction effect. Therefore, although there is no evidence presented to restrict the use of WTR in the field, the lability of metals (especially Mn) in WTR requires further assessment prior to field application. In addition, fractionation (e.g., BCR) is recommended for use to determine the potential lability of metals under various conditions.

1. Introduction

Recently, the use of a chemical immobilization technique for environmental remediation has attracted increasing attention from researchers [1]. The technique uses chemical amendments to immobilize contaminants to reduce the adverse effects of pollutants on the environment. Low-cost amendments are beneficial to the application of this technique. Therefore, many industrial wastes and by-products have been tested for reuse [2,3]. Among them, drinking water treatment residue (WTR), which is an inevitable by-product generated during potable water production, has been a focus of many studies [4]. Typically, WTR is primarily composed of Fe/Al hydroxides due to the use of coagulants to purify the raw water and impurities from the raw water (e.g., suspended solids) and other operation processes (e.g., active carbon) [5]. Currently, because WTR is considered non-hazardous material, it is typically disposed of as common waste for landfills despite the high cost and lack of available land [6]. Therefore, the successful reuse of WTR will lead to a win–win situation for environmental remediation.

Drinking water treatment residue is an effective adsorbent for many contaminants [5,6], and many studies have focused on reusing WTR as a P adsorbent. Studies have shown that WTR has a high P sorption capacity [7–9] and that the adsorbed P on the WTR

Abbreviations: Al_{ox}, oxalate-extractable Al; BCR, European Community Bureau of Reference; Fe_{ox}, oxalate-extractable Fe; ICP-AES, inductively coupled plasma-atomic emission spectrometry; SBET, simple bioaccessibility extraction test; TCLP, toxicity characteristic leaching procedure; TOM, total organic matter; WTR, drinking water treatment residue.

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is stable under various conditions [10,11]. Researchers have also attempted to reuse WTR to control P pollution in the environment. Drinking water treatment residue can be used as an amendment to reduce the loss of P from P-rich soils [12] and as the main substrate for constructing wetlands to remove excessive P from wastewater [13]. More recently, WTR has been used as an immobilizer to reduce the internal P loading of a lake for eutrophication control [14,15]. These results demonstrated that WTR has a high potential in environmental applications for P pollution control.

However, the beneficial reuse of WTR may be hampered by the potential release of some toxicants (e.g., metals) during practical applications. It has been demonstrated that the pH and redox conditions were the two main factors affecting the lability of inorganic contaminants in the environment [16]. The authors' research group has found that the lowest potential for metal release from WTR was at pH 6–9, which was also recommended as the optimal pH for WTR reuse [17]. To date, there is little information on the effects of redox status on the lability of metals in WTR. In the envisioned practical application (e.g., soil, water and sediment), the most common conditions for the application of WTR involve an anaerobic or anoxic environment. Under these conditions, the lability of metals in solid materials (e.g., soils [18] and municipal solid waste [19]) may change. Therefore, in this work, based on an anaerobic incubation test, the lability of metals in WTR was investigated using fractionation, in vitro digestion and the toxicity characteristic leaching procedure (TCLP). The results of this work will provide theoretical support for the recycling of WTR as an environmental amendment.

2. Materials and methods

2.1. Sample collection

A single sample of dewatered WTR was collected from the dewatering workshop of the Beijing City No. 9 Waterworks in China in April 2012. In this facility, both surface water and groundwater are used as drinking water sources. Coagulation, precipitation and filtration are used for conventional water treatment, and activated carbon adsorption is used for advanced water treatment. In addition, both poly aluminum chloride and $FeCl_3$ are used as coagulants. The fresh WTR was air-dried, ground and sieved to a diameter of less than 1 mm for the incubation study.

2.2. Anaerobic incubation experiment

A 500-g sample was weighed into a 1-L flask, and deionized water was added to saturate the WTR with water (with approximately 1 mm depth of water above the WTR). Then, the flask was covered with a gas-permeable film (12#, Shanghai Yongxing Biological Science Co., LTD., China) and placed in a culture tank. The gas (air) extraction and replacement processes were completed 3 times by a Unijar Suction System (Unitech BioScience Co., Ltd, China) in the culture tank to create anaerobic conditions. The replacement gases were N_2 (80%), CO_2 (10%) and H_2 (10%). Previous studies have shown that the above processes were able to maintain anaerobic conditions in the culture tank [20]. The culture tanks were placed in constant-temperature incubators at 25 °C. Finally, the incubated WTR was sampled at 30, 60, 120 and 180 d. During sampling, the water-saturated WTR was homogenized and collected with a plastic spoon. After each sampling, deionized water was added to keep the WTR saturated with water, and gas (air) extraction and replacement processes were applied again to maintain anaerobic conditions in the culture tank. The sampled water-saturated WTR (approximately 50 g in dry weight) was directly freeze–dried, ground and sieved to a diameter of less than 0.15 mm for further analysis (i.e., samples characterization, fractionation, in vitro digestion and TCLP analysis).

2.3. Solids analysis

2.3.1. Samples characterization

The pH of the WTR before and after incubation was determined in the supernatant of a solid and solution (deionized water) ratio of 1:2.5 ($g\,mL^{-1}$) suspensions using a pH meter (pH-10, Sartorius, Germany). The contents of total metal, oxalate-extractable Al and Fe (Al_{ox} and Fe_{ox}) and total organic matter (TOM) in the WTR were also determined. The total metals in WTR were digested according to US EPA Method 3051 [21], and then their contents were measured using inductively coupled plasma-atomic emission spectrometry (ICP-AES, ULTIMA, JY, France). Notably, the quality control for WTR digestion was achieved using standard addition method [21]. Spiked samples were prepared by WTR with addition of standard solutions. The total metals contents in the spiked samples were analyzed as those in WTR. Then, the standard curve was drawn based on the added amounts and detected values. The results showed that R^2 for standard cure of each metal was >0.99. Therefore, US EPA Method 3051 was appropriate to be used to digest WTR for total metal contents determination. In fact, the total digestion method for WTR has also been used by other studies [22,23]. In addition, the Al_{ox} and Fe_{ox} contents were analyzed according to the methods of Shang and Zelazny [24]. The TOM content was measured using the potassium dichromate oxidation method [25].

2.3.2. Fractionation of metals

The European Community Bureau of Reference (BCR) sequential extraction procedure was used to extract the metals from the WTR before and after incubation [26]. Briefly, WTR was sequentially extracted using: (1) 0.11 M CH_3COOH (pH 2.85) for 16 h at 25 °C, (2) 0.1 M $NH_2OH{\bullet}HCl$ (pH 2) for 16 h at 25 °C and (3) 30% H_2O_2 for 2 h at 85 °C, followed by 1 M CH_3COONH_4 (pH 2) for 16 h at 25 °C. The metals in the extracts were determined using ICP-AES. This sequential extraction technique separated the extracted metals into the nominal three fractions of acid-soluble metal, reducible metal and oxidizable metal, respectively. The residual metal content in the WTR (non-extractable by the BCR procedure) was calculated as the difference between the sum of each fraction and the total content quantified using US EPA Method 3051 [20].

2.3.3. In vitro digestion

A simple bioaccessibility extraction test (SBET) was an in vitro digestion method that simulated mobilization of contaminants in the acid conditions of the stomach [27]. The method has been validated to the in vivo situation with swine studies [28]. Therefore, the SBET method was used to examine the bioaccessibility of metals in the WTR before and after incubation. Briefly, the WTR was mixed with 0.4 M glycine ($C_2H_5NO_2$, pH 1.5) at a solid and solution ratio of 1:100 ($g\,mL^{-1}$). The mixtures were rotated end-over-end at 30 ± 2 rpm for 1 h at 37 °C. The WTR extracts were filtered using 0.45-μm micropore filter paper, and the metal content in the filtered extracts was determined using ICP-AES.

2.3.4. Leaching toxicity of metals

Toxicity characteristic leaching procedure was used to investigate the leaching potential of metals from the WTR before and after incubation [29]. In accordance with the TCLP method, the pH of the supernatant of a 5 g WTR: 96.5 mL deionized water slurry was first determined, and as this was less than pH 5, WTR samples before and after incubation were extracted with an acetic acid-sodium hydroxide solution (pH 4.93 ± 0.05) at a ratio of 1:20 ($g\,mL^{-1}$) at 25 °C for 18 h. The detailed methods for the pH determining and the extraction solution preparing can be referred to Supporting