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Key factors related to drinking water treatment residue selection for adsorptive properties tuning via oxygen-limited heat treatment

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HIGHLIGHTS

- Properties of different DWTR after oxygen-limited heat treatment changed variously.
- The treatment led to the varied lability of metals and arsenic in different DWTR.
- Oxygen-limited heat treatment may not be applicable for all DWTR improvement.
- Pre-screening processes were proposed for DWTR selection.

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ABSTRACT

The beneficial recycling of drinking water treatment residue (DWTR) for environmental remediation has attracted growing attention, and oxygen-limited heat treatment has a high potential to improve DWTR adsorptive properties for better recycling. In this study, physicochemical property variation induced by oxygen-limited heat treatment was evaluated for six DWTR obtained from different drinking water treatment plants in Australia, China, and Ireland. The results showed that the changes in many properties, typically amorphous Al and Fe contents, mesopores, specific surface area (SSA), and total pore volume (TPV) of DWTR after treatment, showed inconsistent trends. The treatment also led to the varied lability of metals and As in DWTR although the human bioaccessibility and plant bioavailability of most metals decreased. Accordingly, oxygen-limited heat treatment may not be applicable for all DWTR improvement. Based on these findings, pre-screening processes to evaluate adsorptive properties and metal and metalloid lability were determined. Oxygen-limited heat treatment was more applicable to DWTR with neutral and weak alkaline properties (pH 7.0–7.8), low humic acid (HA) content (<2.09 mg g^{-1}), and high amorphous Fe and HA ratio (>5.93), as well as with relatively high amorphous Al and HA ratio and high humin content.

1. Introduction

The beneficial recycling of drinking water treatment residue (DWTR) is of growing interest [1]. DWTR is a by-product generated during potable water production, and commonly, can be classified as coagulant, groundwater or softening, natural, or manganese residue [2]. The coagulant residue constitutes the majority of DWTR and has been studied the most. In conventional coagulation and filtration treatment process, aluminum (Al) and iron (Fe) coagulants are used to remove suspended solids from raw water, and the resulting DWTR is highly porous and contains high concentrations of amorphous Al and Fe [1]. Heavy metal and organic pollutant concentrations also tend to be low in DWTR because the applied raw water is typically high quality [3–5]. For these reasons, DWTR has been considered as a kind of recyclable resource.

The beneficial recycling of DWTR allows coagulant recovery [6,7], soil amendment [8], construction material utilization [9], and environmental remediation [10,11]. Due to the increased global problem of environmental pollution, environmental remediation using DWTR is of interest to many scientists [1]. DWTR has been found to be an effective adsorbent of phosphorus (P) [12], hydrogen sulfide [13], perchloric acid [14], arsenic (As) [15,16], boron (B) [17], chromium (Cr) [18], copper (Cu) [19], mercury (Hg) [20], lead (Pb) [18], selenium (Se) [21], zinc (Zn) [22], antibiotics [23], chlorpyrifos [24], and glyphosate [25]. Efforts have been

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made to use recycled DWTR to treat soil [26], water [27], and sediment [22,28]. To date, most DWTR is disposed in a landfill, which is costly and consumes space. Therefore, a successful recycle of DWTR will lead to environmentally friendly practices.

Efforts also have been made to tune the physicochemical properties of DWTR to facilitate recycling. Acid washing has been used to eliminate the unfavorable effect of Al to allow the use of DWTR in cultivated soil because the presence of Al in DWTR shows high adsorption capacity for P and other fertilizer components. Once washed with acid, the resulting DWTR could be used without inhibition of plant growth [8]. Ultrasonic treatment via a bath/probe sonicator changed the physical, chemical, and biological characteristics of DWTR, suggesting ultrasound could be used in pretreatment enhanced coagulation of recycling DWTR, and used for the conditioning of water and wastewater mixed sludge by ultrasound combined with polymers [29]. Pre-treatment of sludge with acid followed by the removal of organic and particulate contaminants using a 2kD ultrafiltration membrane resulted in a reusable coagulant similar in performance to that of fresh ferric sulfate for P removal from primary wastewater [30]. Similarly, we recently showed that oxygen-limited heat treatment significantly enhanced the nitrogen (N_2) sorption capacity, specific surface area (SSA), total pore volume (TPV), and porosities of DWTR, increased the amorphous Al/Fe contents in DWTR at 200–400 °C, induced organic matter to contain more aromatic carbon (C) and less aliphatic C, and showed a high potential to increase the adsorption capability of DWTR for different pollutants [31]. Overall, these studies provided guidelines for the improved recycling of DWTR.

However, the widespread use of the methods for DWTR improvement may be limited by the fact that the properties of DWTR from different plants are variable dependent on the different water sources and water treatment technologies used [5]. To address this variability, we analyzed the physicochemical properties of DWTR before and after oxygen-limited heat treatment for samples from six different drinking water treatment plants in Australia, China, and Ireland. The main objectives of this study were to determine the applicability of oxygen-limited heat treatment for DWTR improvement and provide theoretic support to increase the effective recycling of DWTR.

2. Materials and methods

2.1. Sample collection and preparation

The six dewatered DWTR samples were collected from drinking water treatment plants in Perth, Australia; Beijing, Guangzhou, Hangzhou, and Lanzhou in China; and Dublin, Ireland, and the samples were denoted as Australia-R, China-BR, China-GR, China-HR, China-LR, and Ireland-R, respectively. The drinking water treatment plant in Perth uses groundwater as the water source and uses Al- or Fe-based flocculant; the plant in Beijing uses surface and ground water as its water source and uses Al-or Fe-based flocculant for water purification; and the plants in Guangzhou, Hangzhou, Lanzhou, and Dublin use surface water as the water source and use Al-based flocculant. The fresh dewatered DWTR was air-dried, and then ground and sieved to a diameter less than 0.25 mm.

Oxygen-limited heat treatment [31] was performed by placing 20 g of the DWTR in a quartz flacon covered with a quartz lid. The samples were heated under a nitrogen atmosphere with a heating rate of 15 °C $\min^{-1}$ to reach 350 °C for 4 h. The produced solid was washed by deionized water until the supernatant was almost colorless and then was oven-dried at 50 °C. We refer to the DWTR before treatment as the raw DWTR and the DWTR after treatment as the treated DWTR.

2.2. Properties characterization

The N_2 sorption/desorption, the main structures of organic matter, the total organic C (TOC) content, and the surface morphologies and elemental distribution of DWTR were characterized by using the SSA and porosity analyzer (NOVA4200e, Quantachrome, USA), Fourier transform infrared spectroscopy (FTIR, Nicolet iS5, Thermo Fisher, USA), elemental analyzer (EA3000, Euro Vector, Italy), and scanning electron microscope combined with energy dispersive X-ray detector (SEM-EDX, X650, Hitachi, Japan), respectively. The organic matter in DWTR was obtained by demineralizing the DWTR with 1 M hydrochloric acid and 10% (v/v) hydrofluoric acid at 1:5 solid/liquid ratio seven times [32]. In addition, fulvic acid (FA), humic acid (HA), humin (HM), pH, and surface charge properties of DWTR before and after treatment were also analyzed [31,33].

2.3. Metal and metalloid extractability

The amorphous Al and Fe in DWTR, represented by the oxalate extractable Al and Fe (Al_{ox} and Fe_{ox}), were extracted by ammonium oxalate-oxalic acid solution (pH 3) in the dark [34]. The lability of As and metals, including barium (Ba), cadmium (Cd), cobalt (Co), Cr, Cu, manganese (Mn), molybdenum (Mo), nickel (Ni), Pb, and Zn, was determined by a simple bioaccessibility extraction test (SBET) [35] and a diethylenetriaminepentaacetic acid (DTPA) extraction method [36] that indicate human bioaccessibility and plant bioavailability, respectively. In addition, the total concentrations of As and other metals in DWTR were determined after digestion with nitric acid-hydrofluoric acid-perchloric acid. The As and other metals contents in extracts were measured using inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700x, USA).

2.4. Statistical analysis

In order to understand the basis for the variation of DWTR properties after oxygen-limited heat treatment, linear correlations analysis was applied to the data using IBM SPSS Statistics 19. In addition, the standard errors for As and metals extractability determination were within 10% (n = 3) and for other properties, including pH, TOC, HA, and FA, they were within 5%.

3. Results

3.1. The basic properties of DWTR

The basic properties of DWTR before and after oxygen-limited heat treatment are presented in Table 1. The pH of Australia-R, China-HR, and Ireland-R after treatment decreased from 5.93–7.26 to 5.38–7.06, and that of China-BR, China-GR, and China-LR increased from 7.44–8.10 to 7.67–8.65. The TOC of DWTR after treatment generally decreased from 9.17–61.5 to 6.12–45.2 mg g^{-1}, but the TOC of Ireland-R slightly increased from 111 to 117 mg g^{-1}. The FA and HM for all DWTR after treatment decreased from 1.66–19.8 to 0.44–7.09 mg g^{-1} and from 6.34–76.3 to 3.90–62.8 mg g^{-1}, respectively. In contrast, HA generally increased from 1.17–14.9 to 1.78–47.2 mg g^{-1}, but HA in China-GR slightly decreased from 1.27 to 1.18 mg g^{-1}.

The total contents of Al and Fe for all DWTR after treatment increased from 93.3–172 to 96.5–222 mg g^{-1} and from 6.44–93.7 to 9.05–125 mg g^{-1}, respectively. The Al_{ox} contents were found to increase from 13.3–104 to 13.6–115 mg g^{-1} for China-BR, China-GR, and China-GR, and decreased from 16.7–147 to 15.8–127 mg g^{-1} for Australia-R, China-LR, and Ireland-R. The Fe_{ox} contents increased from 4.69–87.7 to 5.52–96.6 mg g^{-1} for China-BR,