



Research article

Aging of aluminum/iron-based drinking water treatment residuals in lake water and their association with phosphorus immobilization capability

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ABSTRACT

Aluminum and Fe-based drinking water treatment residuals (DWTRs) have shown a high potential for use by geoenvironmental engineers in internal P loading control in lakes. In this study, aging of Al/Fe-based DWTRs in lake water under different pH and redox conditions associated with their P immobilization capability was investigated based on a 180-day incubation test. The results showed that the DWTRs before and after incubation under different conditions have similar structures, but their specific surface area and pore volume, especially mesopores with radius at 2.1–5.0 nm drastically decreased. The oxalate extractable Al contents changed little although a small amount of Al transformed from oxidizable to residual forms. The oxalate extractable Fe contents also decreased by a small amount, but the transformation from oxidizable to residual forms were remarkable, approximately by 14.6%. However, the DWTRs before and after incubation had similar P immobilization capabilities in solutions and lake sediments. Even the maximum P adsorption capacity estimated by the Langmuir model increased after incubation. Therefore, it was not necessary to give special attention to the impact of Al and Fe aging on the effectiveness of DWTRs for geoenvironmental engineering in lakes.

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1. Introduction

Recycling of drinking water treatment residuals (DWTRs) has received increasing international attention (Ippolito et al., 2011). This could be because DWTRs are cost effective and the byproducts generated during potable water production are of low risk to the environment. The main components of DWTRs vary with water treatment processes; nevertheless, the aluminum (Al) and iron (Fe)-based DWTRs produced by applying Al and Fe salts as flocculants to remove impurities from raw water draw the most concern (Babatunde and Zhao, 2007). The DWTRs generally contained various heavy metals, e.g. arsenic, barium, beryllium, cadmium, chromium, etc., but the contents tended to be low and the metals could be stable under pH 6–9 (Wang et al., 2014a). In fact, the DWTRs have been demonstrated non-hazardous according to the

toxicity characteristic leaching procedure assessment method used by the US Environmental Protection Agency (Wang et al., 2014a). The DWTRs are amorphousness, have a high specific surface area, and also have a strong adsorption capability for many contaminants, e.g., phosphorus (P) (Makris et al., 2005b; Oliver et al., 2011), hydrogen sulfide (Wang and Pei, 2012), perchloric acid (Makris et al., 2006a), arsenic (Makris et al., 2006b; Gibbons and Gagnon, 2011), chromium (Zhou and Haynes, 2011), lead (Zhou and Haynes, 2011; Putra and Tanaka, 2011), mercury (Hovsepian and Bonzongo, 2009), and selenium (Ippolito et al., 2009), as well as organic pollutants (Zhao et al., 2013; Punamiya et al., 2013). Based on these attributes, DWTRs have also been used for environment remediation, typically for P pollution control. DWTRs can be used to immobilize excessive P in soils for off-site P pollution control (Agyin-Birikorang et al., 2009), and as main substrates for constructed wetland to remove excessive P from wastewater (Zhao et al., 2009). More recently, DWTRs have shown a high potential for use by geoenvironmental engineers in internal P loading control in lakes (Wang et al., 2012a; 2013b).

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Internal P pollution in lakes mainly refers to the release of P from lake sediments, leading to or aggravating eutrophication. A prior method for internal P loading control is classified as geo-engineering, which manipulates biogeochemical processes by using materials to achieve a desired chemical and/or ecological response (Mackay et al., 2014). The success of this method depends on selecting the appropriated P inactivating agents (Zamparas and Zacharias, 2014). The agents that attract wide interests include Al salts, Fe salts, Ca compounds (calcite), Phoslock® (a lanthanum modified bentonite). The effective control from these compounds was accomplished both in laboratory and field studies (Reitzel et al., 2003; Galvez-Cloutier et al., 2012; Kleeberg et al., 2013; Spears et al., 2013); however, all the agents more or less have exceptions (Playle, 1987; Reedyk et al., 2001; Kleeberg et al., 2012; Lürling and van Oosterhout, 2013). Their limited effectiveness, especially in field studies, could be related to inappropriate applications (Spears et al., 2014). In order to obtain satisfactory results, it is recommended to comprehensively determine the stability of P inactivating agents on pollution control before practical application (Spears et al., 2014).

Reasonably, successful application of DWTRs can lead to a “win–win” technique for lake restoration. To date, some studies have already focused on the stability of lake internal P loading control by DWTRs. Drinking water treatment residuals can substantially decrease the potential of P release from lake sediments under the effect of pH, dissolved oxygen levels, sediment organic matter, silicate, ion strength (Wang et al., 2013a), hydrogen sulfide (Wang et al., 2013d), microbial activity, light, and sediment resuspension (Wang and Pei, 2013a), and can also adsorb P from lake water during sedimentation, while the P immobilization capabilities of DWTRs after sedimentation have minor changes (Wang and Pei, 2013b). However, as an Al/Fe based agent, the aging process may be one of the most important factors affecting its P adsorption capability. It has been reported that Al salts in lake water tend to be crystallized (Berkowitz et al., 2005), and with this process, the P adsorption capability of Al decreases (Berkowitz et al., 2006). Even the maximum adsorption capacity was found to decrease by 75% of total after 90-day aging (de Vicente et al., 2008). Therefore, determination of the aging effect on the stability of internal P pollution control by DWTRs is essential.

In this study, based on a 180-day incubation test, the aging effect of DWTRs under different pH and redox conditions was investigated. The study was mainly carried out from determining the structures, specific surface area and porosity, and the Al and Fe forms, as well as the P adsorption capability in solutions and the immobilization capability for mobile P in lake sediments. The results of this study will provide theoretic supports for recycling DWTRs in geoengineering of lakes for eutrophication control.

2. Materials and methods

2.1. Sample collection

Dewatered DWTRs were collected from Beijing City No. 9 Waterworks in China in Apr. 2012. In this facility, Al or Fe salts were used as coagulants and the moisture content of fresh DWTRs was 78%. The fresh DWTRs were air-dried, and then were ground and sieved to a diameter less than 1 mm to reduce experimental errors because the air-dried samples were bulk materials. The DWTRs contained 0.070 mg g⁻¹ arsenic, 0.423 mg g⁻¹ barium, 0.00047 mg g⁻¹ beryllium, 20.3 mg g⁻¹ calcium, 0.00072 mg g⁻¹ cadmium, 0.0101 mg g⁻¹ cobalt, 0.774 mg g⁻¹ chromium, 0.0649 mg g⁻¹ copper, 1.52 mg g⁻¹ magnesium, 2.54 mg g⁻¹ manganese, 0.00613 mg g⁻¹ molybdenum, 0.0319 mg g⁻¹ nickel, 0.0215 mg g⁻¹ lead, 0.134 mg g⁻¹ strontium, 0.127 mg g⁻¹

vanadium, and 0.100 mg g⁻¹ zinc, but the silver, mercury, stibonium, and selenium were not detectable (Wang et al., 2014c). Besides P, the DWTRs used in this study were also found to have high adsorption capability for hydrogen sulfide and chlorpyrifos. The adsorption capacities for hydrogen sulfide were approximately 14–15 mg g⁻¹ (Wang and Pei, 2012), and the affinity (log K_{oc}) for chlorpyrifos were 4.76–4.90 (Zhao et al., 2013).

Sediments were sampled at Zhainan in Lake Baiyangdian (38°53' N, 115°59' E) in North China in July 2012. The surface sediments, at a depth of 0–5 cm, were sampled by a column sampler (Eijkkelkamp Co., Giesbeek, Netherlands) in the centre of the lake with a boat. The collected sediments were packed into aseptic valve bags, put into an incubator containing ice bags, and then transported to laboratory. The sediments were filtered through a 1.8-mm sieve, homogenized, and stored at 4 °C. The aseptic valve bag, made by polyethylene particles, was used to prevent the contamination and to preserve the physical, chemical, and biological characteristics of sediments. The sediment pH and electrical conductivity were 7.25 and 1482 us cm⁻¹, and also contained 63.2 mg g⁻¹ organic matter, 56.3 mg g⁻¹ Al, and 34.2 mg g⁻¹ Fe, respectively. Other basic characteristics of the sediments can be found elsewhere (Wang et al., 2014b). Lake water was sampled at the same location at a depth of approximately 0.5 m. The lake water was filtered through a 0.45 μm Millipore filter paper and stored at 4 °C.

2.2. Incubation test

Air-dried DWTR samples with a weight of 10 g were added into a series of 100 mL head space bottles containing 50 mL of lake water. The bottles were incubated at 15 °C, and were sampled on the 30th and 180th days. The mixtures were filtered through 0.45 μm Millipore filter paper to separate the solids and solutions for further analysis. The incubated DWTRs were freeze-dried, ground, and completely mixed for further analysis.

The experimental conditions are listed as follows: (1) Redox conditions: Two redox conditions were investigated. One was an aerobic condition based on a natural oxygen exchange between air and lake water by covering sample bottles with a gas-permeable film. The other was anaerobic condition accomplished by covering a gas-permeable film on the bottles and cultured in an anaerobic tank. The anaerobic condition was created by a gas extraction and replacement processes using a Unijar Suction System (Unitech BioScience Co., Ltd, China) for the culture tank. The replacement gases were N₂ (80%), CO₂ (10%), and H₂ (10%), and the gas extraction and replacement processes were applied three times. The gas-permeable film could ensure the achievement of anaerobic condition in the bottles, prevent the contamination by other impurities, and also reduce water evaporation. (2) pH: The pH of overlying lake water was adjusted by NaOH and HCl every 7 days. Two pH ranges investigated in this study were 5.5–7.0 and 7.5–9.0. The pH of the overlying water incubated was determined by a pH meter (pH-10, Sartorius, Germany). The pH range (5.5–9) in this study was selected just based on the normal nature environment pH, which was within 5–9.

2.3. Sample characterization

The indices for lake water analysis included oxidation-reduction potential (ORP) (PY-C01, Sartorius, Germany), pH (pH-10, Sartorius, Germany), and dissolved oxygen (DO) levels (320D-01, Thermo Orion, USA), as well as the concentrations of soluble Al, Fe, and P. Aluminum and Fe concentrations were determined using inductively coupled plasma-atomic emission spectrometry (ICP-AES, ULTIMA, JY, France) after the water samples being acidified (approximately containing 1% HCl); the ICP-AES used inductive

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