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# Intermittent aeration incubation of drinking water treatment residuals for recycling in aquatic environment remediation

Changhui Wang [a, *], Yu Wu [a, b], Leilei Bai [a, c], Chunliu Wang [a, c], Helong Jiang [a, **], Zixun Wei [a], Xingyu Wei [a], Zechen Yuan [a], Xin Liu [b]

[a] State Key Laboratory of Lake Science and Environment, Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, Nanjing, 210008, China
[b] College of Biology and Environment, Nanjing Forestry University, Nanjing, 210037, China
[c] Graduate University of Chinese Academy of Sciences, China

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ABSTRACT

Recycling of drinking water treatment residuals (DWTRs) in aquatic environment remediation can provide numerous benefits. The hazardous effect of DWTR metals on the environment is shown to be limited; however, DWTR may release excessive nitrogen and organic matter, hindering recycling in the aquatic environment. To eliminate any potential unfavorable effect, self-purification via intermittent aeration incubation for DWTRs was developed. Results indicated that after approximately 5–6 months of incubation, the easily released nitrogen and organic matter in the DWTRs were mostly removed. The removed nitrogen was found to mainly consist of ammonium nitrogen from inorganic and organic nitrogen groups in the DWTRs. The removed organic matter consisted mainly of humic-like substances associated with the carboxyl group, distributed on the surface of the DWTRs. Biological mechanisms for effective nitrogen and organic matter removal were further explored based on the analysis of bacterial community structure, which indicated that the dominant bacteria genera (e.g., *Geothrix*, *Methylotenera*, and *Geobacter*) in the DWTRs after incubation were closely related to nitrogen and carbon cycles. In addition, the phosphorus adsorption of the DWTRs after incubation was maintained, and the specific surface area, total pore volume, microporosity, and mesoporosity of the DWTRs after incubation tended to increase because of organic matter removal. Accordingly, intermittent aeration not only addressed the concerns caused by potential excessive release of nutrients from DWTRs but also enhanced the favorable effect of DWTRs on biogeochemical cycles. Self-purification can potentially promote the beneficial recycling of DWTRs in the aquatic environment.



## 1. Introduction

The beneficial recycling of drinking water treatment residuals (DWTRs) has drawn interest worldwide (Ahmad et al., 2016; Nimwinya et al., 2016). As inevitable byproducts generated during potable water production, DWTRs often contain high concentrations of Al and Fe because of the use of Al/Fe salts as coagulants for water purification (Ippolito et al., 2011; Nair and Ahammed, 2015). Al and Fe in DWTRs mainly exist in amorphous phases, inducing the porosity of DWTRs with high adsorption (Ippolito et al., 2011). Owing to these properties, DWTRs were typically developed as adsorbents to remove various contaminants, such as P (Makris et al., 2005; Liu et al., 2016), perchloric acid (Makris et al., 2006), As (Xu et al., 2015), B (Irawan et al., 2011), heavy metals (Lin et al., 2017; Ippolito et al., 2009), and toxic organic pollutants (Punamiya et al., 2013) from aqueous solutions. DWTRs were also recycled in soil remediation (Agyin-Birikorang et al., 2009; Nagar et al., 2015), in constructed wetland to treat wastewater (Hu et al., 2014; Doherty et al., 2015; Ippolito, 2015), and in controlling the release of contaminants from sediment (Wang and Jiang, 2016) for the restoration of the aquatic ecosystem. DWTRs have long been dumped in landfills, which entails high costs, and is increasingly generated. Successful recycling of DWTRs can be beneficial both environmentally and economically.

* Corresponding author.
** Corresponding author.
*E-mail addresses:* chwang@niglas.ac.cn (C. Wang), hljiang@niglas.ac.cn (H. Jiang).

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The beneficial recycling of DWTRs should be based on the comprehensive understanding of potential pollution risks. DWTRs have often been considered non-hazardous; this conventional view has been confirmed by various studies (Ippolito et al., 2011). Owing to the high quality of raw water used in drinking water plants, DWTRs primarily consist of inorganic components (Ippolito et al., 2011). Focus has been directed toward the potential risks of metals/metalloids in DWTRs. Previous studies have indicated that DWTRs contain various quantities of Al, Fe, As, Ba, Be, Cd, Co, Cr, Cu, Mn, Mo, Ni, Pb, and Zn, whereas most metals/metalloids (except for Al and Fe) tend to exhibit low concentrations and occur in relatively stable forms (Elliott et al., 1999; Wang et al., 2014b, 2014c). The risks presented by metals/metalloids in DWTRs have been evaluated in the remediation of soils and sediments (Novak et al., 2007; Yuan et al., 2016c) and in constructed wetlands (Babatunde and Zhao, 2010). Application of DWTRs was typically found to enhance leaching of Mn from soils and sediments under certain conditions (e.g., anaerobic condition) (Novak et al., 2007; Yuan et al., 2016c) and to increase the concentrations of Al, Fe, and Mn in some plants (e.g., *Hydrilla verticillata* and *Lolium perenne* L.) (Ippolito et al., 2011; Yuan et al., 2016c); however, no reports have clearly determined the toxic effect of DWTRs on the environment. On the contrary, the addition of DWTRs to sediment has been found to induce conditions that are beneficial to nitrification and anaerobic ammonia oxidation (Wang et al., 2014a, 2013). Bioassay tests indicated that DWTRs exhibit no toxicity to *Chlorella pyrenoidosa* and *Aliivibrio fischeri* (Yuan et al., 2016a, b). These reports suggested the limited hazardous environmental effects of metals/metalloids in DWTRs during recycling.

Notably, DWTRs potentially release N and organic matter to the environment (Liu et al., 2016; Yuan et al., 2016b). Ammonium N ($NH_4^+$-N) was released during recycling of DWTRs in the control of internal pollution from sediments (Ichihara and Nishio, 2013) and in constructed wetlands (Bai et al., 2014). The release particularly led to the increase in $NH_4^+$-N concentrations in the effluent of the secondary effluent treated using DWTR dominant columns during the entire 260-d tests, although total N (TN) decreased in the effluent (Bai et al., 2014). The potential effects of N and organic matter release could be closely related to DWTR dosage and the requirements for receiving water. Further comparison suggested that nutrients are released from DWTRs during the treatment of surface water and secondary effluent (Ichihara and Nishio, 2013; Bai et al., 2014) but not during the treatment of livestock and domestic wastewaters (Hu et al., 2014; Doherty et al., 2015). The reason may be that the high nutrient levels of raw livestock and domestic wastewaters obscure the release of nutrients from DWTRs. Accordingly, the release of N and organic matter from DWTRs tends to be prominent during recycling in the aquatic environment. Ammonia (particularly in non-ionic state) is toxic to organisms, and excessive N promotes eutrophication (Camargo and Alonso, 2006). Organic matter is a key component in biogeochemical cycles and exerts a considerable effect on the environmental behaviors of various pollutants (Chen and Hur, 2015). Released nutrients from DWTRs were even found to enhance algal growth (Yuan et al., 2016b). These findings suggest that DWTRs exert a potential unfavorable effect of excessive release of N and organic matter from DWTRs to the environment.

Therefore, a low-cost method to eliminate the concerns caused by potential excessive nutrients released to the aquatic environment can benefit DWTR recycling. In the present study, an approach called self-purification was developed to remove N and organic matter that were easily released from DWTRs via intermittent aeration. The study mainly focused on the easily released N and organic matter in the DWTRs because nutriments were most likely to harm the environment. The removal effectiveness, the removal chemical and biological mechanisms, and the adsorption capabilities of the DWTRs before and after incubation were comprehensively determined to evaluate the feasibility of the proposed method. The results of this study can provide theoretical support for DWTR recycling in aquatic environment remediation.

## 2. Material and methods

### 2.1. Sample collection and preparation

Dewatered DWTRs were collected from a drinking water treatment plant in June 2016 in the City of Suzhou, Jiangsu, China. In the facility, Al salts were used as coagulants, and the moisture content of fresh DWTRs was approximately 81%. The collected sample was bulk material; thus, fresh DWTRs were air-dried and broken to a diameter of less than 10 mm.

Self-purification of DWTRs via intermittent aeration incubation: A detailed schematic of DWTR incubation is presented in Fig. S1 (in Supporting information). A column with an inner diameter of 120 mm and a height of 420 mm was prepared. Gravels with a diameter of less than 10 mm were first packed at the bottom of the column with a thickness of 50 mm, and 4 aerators were buried in the gravels. The air-dried DWTRs were then packed in the 250 mm thick column; porosity was approximately 55%. Tap water was added into the column and supplied accordingly, resulting in 25 mm overlying water on the DWTRs. Intermittent aeration of air was performed based on a ratio of time at 12 h:12 h at room temperature (15–20 °C) in the dark. During incubation, water solutions in the bottom layer and the upper layer of the column were collected for analysis each week after daily aeration for 1 h. After incubation, the DWTRs in the column were completely mixed. Some of the DWTRs were sampled for biological analysis, whereas the remaining DWTRs (with moisture of approximately 60%) were freeze-dried for chemical analysis. A similar column without intermittent aeration was set as the control in this test.

### 2.2. Chemical analysis

#### 2.2.1. Characterization of DWTRs before and after incubation

The $N_2$ sorption/desorption, the main structures, and the TN and total organic C (TOC) of the DWTRs were characterized using the specific surface area (SSA) and a porosity analyzer (NOVA4200e, Quantachrome, USA), a Fourier transform infrared spectroscope (FTIR, Nicolet iS5, Thermo Fisher, USA), a scanning electron microscope combined with an energy dispersive X-ray detector (SEM-EDX, ×650, Hitachi, Japan), and an elemental analyzer (EA3000, Euro Vector, Italy). The chemical states of Al2p, Fe2p, C1s, and N1s in the DWTRs were determined by X-ray photoelectron spectroscopy (XPS, ESCALAB 250, Thermo Fisher Scientific, USA). The obtained spectra were analyzed using the XPS PEAK41 software. The amorphous Al and Fe in the DWTRs, represented by the oxalate extractable Al and Fe ($Al_{ox}$ and $Fe_{ox}$), were extracted using an ammonium oxalate–oxalic acid solution (pH 3) in the dark (Shang and Zelazny, 2008). The bioavailability of Al, Fe, and Mn was determined based on 0.11 M $CH_3COOH$, which was the first step of the European Community Bureau of Reference (BCR) method (Rosado et al., 2016; Yuan et al., 2004). The extractable metals in the first step of the BCR method showed a strong correlation with sediment toxicity to *Vibrio fischeri* (Rosado et al., 2016). In addition, the pH and the total contents of Al, Fe, Ca, and Mg were measured accordingly (Wang et al., 2016).

Phosphorus adsorption tests were also performed in this study. DWTRs and solutions were mixed at a solid and solution ratio of 1:100 (g $mL^{-1}$) with shaking for 2 and 30 d, at 25 °C. In this test, solutions under pH 7 that contain 1, 2, 4, 6, 8, and 10 mg P $L^{-1}$