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Influence of the inherent properties of drinking water treatment residuals on their phosphorus adsorption capacities

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ABSTRACT

Batch experiments were conducted to investigate the phosphorus (P) adsorption and desorption on five drinking water treatment residuals (WTRs) collected from different regions in China. The physical and chemical characteristics of the five WTRs were determined. Combined with rotated principal component analysis, multiple regression analysis was used to analyze the relationship between the inherent properties of the WTRs and their P adsorption capacities. The results showed that the maximum P adsorption capacities of the five WTRs calculated using the Langmuir isotherm ranged from 4.17 to 8.20 mg/g at a pH of 7 and further increased with a decrease in pH. The statistical analysis revealed that a factor related to Al and 200 mmol/L oxalate-extractable Al (Al_{ox}) accounted for 36.5% of the variations in the P adsorption. A similar portion (28.5%) was attributed to an integrated factor related to the pH, Fe, 200 mmol/L oxalate-extractable Fe (Fe_{ox}), surface area and organic matter (OM) of the WTRs. However, factors related to other properties (Ca, P and 5 mmol/L oxalate-extractable Fe and Al) were rejected. In addition, the quantity of P desorption was limited and had a significant negative correlation with the ($Fe_{ox} + Al_{ox}$) of the WTRs ($p < 0.05$). Overall, WTRs with high contents of Al_{ox} , Fe_{ox} and OM as well as large surface areas were proposed to be the best choice for P adsorption in practical applications. © 2014 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences.

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Introduction

Excessive phosphorus (P) in water bodies is one of the main causes of eutrophication (Edwards and Withers, 2007). Chemical adsorption can remove P effectively. Common adsorbents include natural minerals, engineering materials and industry by-products (Pant et al., 2001; Liu et al., 2007). Drinking water treatment residuals (WTRs) are inevitable by-products of drinking water treatment plants, which have attracted substantial attention in recent years due to their high P adsorption capacity. The reuse of WTRs to remove P is a win-win technology for waste management and water environment restoration. Several applications for WTRs have been developed (Agyin-Birikorang et al., 2007; Zhao et al., 2009; Wang et al., 2012a).

Therefore, a comprehensive understanding of the P adsorption capacity of WTRs is crucial to their effective utilization.

The factors affecting the P adsorption capacity of WTRs are mainly derived from two categories. One is solution chemistry, including pH, temperature, dissolved organic carbon, low molecular weight organic acids and P species (Razali et al., 2007; Wang et al., 2012b; Gao et al., 2013). The pH has been considered the most important factor, exhibiting a negative correlation with the adsorption capacity of WTRs (Wang et al., 2011a). The other category is the inherent properties of the WTRs, such as their surface area, particle size and contents of Fe, Al, P, Ca and other elements (Dayton and Basta, 2005; Makris et al., 2005). The physicochemical characteristics of WTRs are related to the source

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water, the chemicals used for treatment and the strategy of operations. However, limited data are available regarding the influence of the inherent properties on the P adsorption of WTRs.

Five types of WTRs were collected from different regions in China. Their physicochemical properties were determined. Batch experiments were carried out to investigate the P adsorption and desorption on different WTRs. Afterward, the integrated relationship of the inherent properties (contents and fractions of elements, pH and surface area) and P adsorption characteristics of the WTRs was calculated using statistical analysis. This work could be beneficial toward understanding P adsorption on WTRs and promoting their practical application.

1. Materials and methods

1.1. Preparation and characterization of the WTRs

The BJ1-WTRs and BJ2-WTRs were sampled from the Beijing NO. 9 Water Treatment Plant in 2011 and 2012, respectively. This plant used a combination of FeCl₃ and polymeric aluminium (PAC) as a coagulant, while supplying 60% of the water requirement for Beijing. Active carbon was used as an adsorbent for advanced treatment. The HZ-WTRs and LZ-WTRs were sampled from the Hangzhou Yuxi Water Treatment Plant and Lanzhou Water Treatment Plant, respectively. Both plants used PAC as a coagulant. The SD-WTRs sampled from the Shandong Yuxing Water Treatment Plant used PAC as a coagulant and Ca(HCO₃)₂ as a softening agent. The five WTRs were air-dried and sieved (<2 mm) to create homogeneous sub-samples before analysis.

The total Fe, Al and Ca contents of the WTRs were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES, Jobin Yvon, Paris, France) according to USEPA-METHOD 3051. Total P was determined using ICP after digestion by a mixture of HNO₃, HClO₄ and HCl. The 200 mmol/L oxalate-extractable Fe (Fe_{ox1}), Al (Al_{ox1}), and P (P_{ox1}), and the 5 mmol/L oxalate-extractable Fe (Fe_{ox2}), Al (Al_{ox2}), and P (P_{ox2}) were determined by ICP after extraction at a 1:60 solid:solution ratio following the procedures described in a previous study (Schoumans, 2000). The phosphorus saturation index (PSI) was determined using the P_{ox1}/(Al_{ox1} + Fe_{ox1}), where the units of P_{ox1}, Al_{ox1} and Fe_{ox1} are mol/kg (Elliott et al., 2002). The Mehlich 3 extractable Ca (Ca_M) was determined as described in the literature (Mehlich, 1984). The total carbon was determined using the Dumas method with a 1500 series dry combustion analyzer (Crilo Erba, Milan, Italy). The organic matter (OM) was determined using a potassium dichromate oxidation method (Nelson and Sommers, 1982). The pH was determined in a 1:2 WTRs to 0.01 mol/L CaCl₂ solution. The electrical conductivity (EC) was determined in a 1:2 WTRs to deionized water solution. The SEM, XRD and surface area were analyzed to determine the structure of the different WTRs. The tests were repeated twice, and the average values are reported.

1.2. Adsorption characteristics

For adsorption experiment, P working solutions with initial P concentrations (P₀) at eight common levels (5, 10, 15,

20, 30, 40, 50 and 100 mg/L) were prepared by dissolving pre-determined amounts of KH₂PO₄ in a 0.01 mol/L KCl solution.

The batch experiments were conducted by pouring 0.5 g of the WTRs and 50 mL of the P working solutions with different P₀ into 100 mL plastic bottles. The pH values of the P working solutions were adjusted to 5.0, 7.0 and 9.0 with 0.01 mol/L HCl and 0.01 mol/L NaOH. The mixed samples were shaken at 200 r/min for 48 hr to attain equilibrium (Wang et al., 2012b). Afterward, the samples were removed from the shaker and filtered using a 0.45-μm millipore membrane filter to separate the solid from the liquid. The residual P in suspensions was measured using an ammonium molybdate spectrometry method. The amount of P adsorbed (Q) was determined using Eq. (1):

$$Q = \frac{(P_0 - P_e)V}{m} \quad (1)$$

where, P₀ (mg/L) and P_e (mg/L) are the initial P and final P concentrations, respectively; V (L) is the solution volume, and m (g) is the mass of WTRs.

The desorption was facilitated by adding 50 mL of 0.01 mol/L KCl solution to 0.5 g of P-loaded WTRs from adsorption test that had been washed with ethanol three times. The initial pH was maintained at 7. Afterward, the samples were shaken at 200 r/min for 48 hr. The P concentrations of the suspensions were measured. The amounts of P desorbed (Q_d) were also determined using Eq. (1).

The Langmuir isotherm was used:

$$Q_e = \frac{Q_{\max} b P_e}{1 + b P_e} \quad (2)$$

where, Q_e (mg/g) is the mass of P adsorbed per unit mass of WTRs, b is an empirical constant related to the entropy, Q_{max} (mg/g) is the maximum adsorption capacity, and P_e (mg/L) is the equilibrium P concentration. It has been proved that the P adsorption by WTRs fits the Langmuir isotherm well (Dayton and Basta, 2005; Wang et al., 2011a). The Q_{max} was calculated by fitting Eq. (2) to the experimental data.

1.3. Statistical analyses

The statistical analyses were performed using SPSS software version 20 (SPSS Inc., Chicago, Illinois, USA). The measured inherent characteristics of the five WTRs were analyzed using rotated principal component analysis. This procedure reduced the number of independent variables from 12 individual abiotic variables to three or four principal component factors, which were independent linear combinations of the 12 original variables. The resulting factor scores and pH values of the P working solution were used as independent variables during the multiple regression analysis while using the P removal properties of the five WTRs as dependent variables. Afterward, the relationship between the inherent properties of the WTRs and their P adsorption capacities was obtained (Arias et al., 2001).

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