



## Effects of wood vinegar on properties and mechanism of heavy metal competitive adsorption on secondary fermentation based composts

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### ARTICLE INFO

#### Keywords:

Wood vinegar  
Secondary compost  
Competitive adsorption  
Adsorption Isotherms  
Heavy metal speciation

### ABSTRACT

In this study, secondary municipal solid waste composts (SC) and wood vinegar treated secondary compost (WV-SC) was prepared to investigate the capability for single-heavy metals and multi-metal systems adsorption. The adsorption sequence of WV-SC for the maximum single metals sorption capacities was Cd ( $42.7 \text{ mg g}^{-1}$ ) > Cu ( $38.6 \text{ mg g}^{-1}$ ) > Zn ( $34.9 \text{ mg g}^{-1}$ ) > Ni ( $28.7 \text{ mg g}^{-1}$ ) and showed higher than that of SC adsorption isotherm. In binary/quaternary-metal systems, Ni adsorption showed a stronger inhibitory effect compared with Zn, Cd and Cu on both SC and WV-SC. According to Freundlich and Langmuir adsorption isotherm models, as well as desorption behaviors and speciation analysis of heavy metals, competitive adsorption behaviors were differed from single-metal adsorption. Especially, the three-dimensional simulation of competitive adsorption indicated that the Ni was easily exchanged and desorbed. The amount of exchangeable heavy metal fraction were in the lowest level for the metal-loaded adsorbents, composting treated by wood vinegar improved the adsorbed metals converted to the residue fraction. This was an essential start in estimating the multiple heavy metal adsorption behaviors of secondary composts, the results proved that wood vinegar was an effective additive to improve the composts quality and decrease the metal toxicity.

### 1. Introduction

Various heavy metals are toxic elements discharged into the environment and often coexist accumulate in wastewater and soil due to many industry practices such as mining, metallurgy, and printing processes, etc. (Bulgariu and Bulgariu, 2012). Thus, soils contaminated by heavy metals need ecological restoration, and potential solution for remediating mine tailings or polluted soil to improve plant growth attracted great attention recently (Park et al., 2011; Jiang et al., 2016). However, degraded soils such as abandoned mines have been destroyed the physical and nutritional structure, and rich in heavy metal (Beesley et al., 2014). The remediation of heavy metal contaminated soil by electrochemical or bioleaching methods have the disadvantages for the high restoration investment, destruction of soil structure and hazardous substance residues (Lawal et al., 2010; Anastopoulos et al., 2013). The contaminated site restoration is now more dependent on the assisted natural attenuation measures.

There have been several bioadsorbents for soil heavy metal remediation, many of which are based on lignocellulosic materials. The removal ability of Cu and Zn ions in pyrolytic biochar was evaluated,

and as a result, the pH and sulphate of a typical ranges may result in weak effect on the adsorption capacity (Jiang et al., 2016). The adsorption behavior of monometal and multimetal in sesame straw biochar was estimated and the adsorption capacities of Pb were the highest in both systems (Park et al., 2016). The potential of diverse biosorbents such as water hyacinth roots for the heavy metal adsorption in aqueous environment and adsorption behaviors were investigated (Lim et al., 2008; Zheng et al., 2016).

Composting processes related to closely with diverse microbial communities, leading to humified and stabilized organic production which are effective substance for improving soil properties. However, the organic amendments to soil such as composts, are transformed into accessible nutrients added to contaminated soil for in-situ improvement. In general, addition of composts can improve physico-chemical property of soil, and effectively decrease the mobility of soil contaminants (Gadepalle et al., 2008; Silveti et al., 2017). Compost organic matter can form insoluble metal organic compounds, reducing the amount of bioavailable metal in the soil. The functional groups of organic groups such as carboxyl, phenolic, and hydroxyl are involved in the immobilization process and can be easily interacted with metal

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<https://doi.org/10.1016/j.ecoenv.2017.12.037>

Received 12 September 2017; Received in revised form 12 December 2017; Accepted 14 December 2017

Available online 04 January 2018

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cations at neutral pH (Paradelo and Barral, 2012). It has been established that the application of organic matter to amend soils and reduce the bioavailability of heavy metals (Gadepalle et al., 2008; Soobhany et al., 2015).

The adsorption behavior of heavy metals depends not only on the nature of the compost, but also on the properties of the adsorbed metal and the competitive behavior on the adsorbent adsorption sites. Therefore, methods of soil remediation including adding polyvalent metal cations to the soil were conducted to reduce the heavy metal mobility in organic matter (Udovic and McBride, 2012). It has also been suggested that the addition of compost to the contaminated soil with arsenic and lead can contribute to reduce the metal bioavailability (Sharma et al., 2010). However, the multi-metal adsorbed structure and mechanisms by the organic matters, and their stability in composts are still obscure.

Wood vinegar is produced by pyrolysis of charcoal and contains more than 200 types of chemical components (Wei et al., 2010). Wood vinegar can be used as soil amendment, composting fermentation, plant growth conditioner. Recently, wood vinegar has been studied as an excellent additives to remove pollutants. The addition of biochar and bamboo vinegar reduced mobility of copper and zinc in pig manure composts (Chen et al., 2010). However, little attention has been paid so far on the effect of wood vinegar on the changes of organic matter during composting, and research on improving the nature of adsorbent needs to be further explored.

Taking into account that metal pollution can take place affecting to various pollutants simultaneously, it is interesting to perform studies focusing on multi-metal systems. Therefore, the aim of this work was to i) determine the characteristics of secondary municipal solid waste composts and wood vinegar treated secondary compost (SC and WV-SC), compare the sorption capacity of single and competitive metal systems; ii) to analyze the competitive effect involved in the multimetal adsorption processes, compare the adsorption isotherm models; iii) to examine the role of wood vinegar commonly present in composting and metal adsorption process; Finally, using three-dimensional simulation to characterize the competitive adsorption behavior of heavy metals, identify the changes in metal mobility and fractionation after adsorption.

## 2. Materials and methods

### 2.1. Materials

The municipal waste primary composts material used in this study was obtained from Asuwei Waste Sanitary Landfill Site which is located in the Beijing Changping District. It was composted with municipal solid waste with 2% (v/w) microorganism agents for 35 days. Part of primary compost was air dried, then ground and sieved through 2-mm diameter sieve, for determination of chemical analyses (Table 1). Wood vinegar was provided by Shijiazhuang Hongsen Activated Carbon Co. Ltd. It was refined from fruit shells and the specific gravity of wood vinegar was 1.035 g mL<sup>-1</sup>, pH was 2.40–3.00, formic acid, phenol and acetaldehyde concentration were lower than 0.01%, and the concentration of acetic acid was 9.53%.

### 2.2. Incubation experiment

The secondary compost procedure was performed as followed. Primary composts were collected, chopped, and air-dried, and then, grounded and sieved to a particle size < 2 mm. And the municipal waste primary compost materials were added with wood vinegar (v/w) of 0% (i.e. secondary compost, SC) and 2% (i.e. wood vinegar treated secondary compost, WV-SC), and mixed thoroughly. The composting experiments were carried out with 300 g primary compost, treated with mixing the wood vinegar or distilled water and then placed in the 1 L compost bin. Triplicate each secondary composting treatment and the

**Table 1**

Properties of secondary compost (SC) and the wood vinegar treated secondary compost (WV-SC).

Property	Units	Primary compost	SC	WV-SC
pH (1:25)	–	7.40 ± 0.37 <sup>a</sup>	7.76 ± 0.41 <sup>b</sup>	7.52 ± 0.45 <sup>ab</sup>
Organic Carbon (OC)	g kg <sup>-1</sup>	189 ± 16 <sup>b</sup>	167 ± 20 <sup>a</sup>	159 ± 23 <sup>a</sup>
Organic matter (OM)	g kg <sup>-1</sup>	325 ± 26	287 ± 36	274 ± 42
Total Nitrogen (TN)	%	1.35 ± 0.14	1.68 ± 0.25	1.70 ± 0.19
Available phosphorus (AP)	g kg <sup>-1</sup>	61.3 ± 9.6	42.8 ± 3.7	47.33 ± 14.7
C/N	–	14 ± 2.2	10 ± 0.8	9.4 ± 1.1
Humic Acid (HA)	g kg <sup>-1</sup>	9.2 ± 0.8 <sup>a</sup>	16.9 ± 1.2 <sup>b</sup>	26.9 ± 2.1 <sup>c</sup>
Fulvic acids (FA)	g kg <sup>-1</sup>	34.9 ± 2.7 <sup>b</sup>	25.2 ± 2.1 <sup>a</sup>	25.2 ± 5.4 <sup>a</sup>
HA/FA (H/F)	–	0.26 ± 0.1 <sup>a</sup>	0.67 ± 0.2 <sup>b</sup>	1.07 ± 0.3 <sup>c</sup>
CEC	cmol(+) kg <sup>-1</sup>	21.1 ± 2.4 <sup>a</sup>	29.11 ± 3.7 <sup>b</sup>	32.29 ± 2.8 <sup>b</sup>
K	mg kg <sup>-1</sup>	4173 ± 126 <sup>a</sup>	6537 ± 215 <sup>b</sup>	4976 ± 653 <sup>a</sup>
Cu	mg kg <sup>-1</sup>	163 ± 23	170 ± 31	168 ± 21
Zn	mg kg <sup>-1</sup>	389 ± 39 <sup>a</sup>	455 ± 42 <sup>b</sup>	459 ± 35 <sup>b</sup>
Cd	mg kg <sup>-1</sup>	4.76 ± 0.8 <sup>b</sup>	3.46 ± 1.0 <sup>a</sup>	4.19 ± 0.9 <sup>b</sup>
Ni	mg kg <sup>-1</sup>	30.1 ± 2.3 <sup>a</sup>	32.8 ± 3.9 <sup>a</sup>	38.4 ± 3.3 <sup>b</sup>

**Notes:** Data are reported as Mean ± SD with n = 3. Different letters following the mean values indicate a significant difference ( $p < 0.05$ ) between different composts using ANOVA.

reactors were kept and incubated in the thermostat incubator (RXZ-500A-LED, Ningbo Jiangnan Instrument, China) at 45 °C. The moisture of the mixing materials was adjusted with distilled water and maintained to the same level of 60%. In order to ensure the aerobic composting process, the composting material were turned over daily. At the end of the composting, the composts from each reactor were collected and air dried.

### 2.3. Characterization of composts

The compost samples were sieved through 1-mm sieve, and the pH and electric conductivity (EC) value were determined by a pH meter (pH/ISE meter, MP522, Shanghai) in 1:2.5 (W/V) suspension in DI water. Ammonium acetate method was used for measure the cation exchange capacity (CEC), and the relevant CEC was equal to the amount of exchangeable ammonium per 100 g sample. Organic carbon (OC) was determined by potassium dichromate oxidation in sulfuric acid solution (Nelson and Sommers, 1996). Total nitrogen (TN) was measured by Kjeldahl method. The humic substances extracted from composts were determined according the method of International Humic Substances Society, the acid (HA) and fulvic acid (FA) content was tested by photometric determination (Helmke et al., 1996). The spectral properties and functional groups of composts were observed by Fourier transform infrared (FTIR, Spectrum 100D, PERKINELMER, USA). The morphologies of the SC and WV-SC, and the SEM-EDX analyses of composts treated with heavy metals were observed with the field emission scanning electron microscopy (Hitachi S-4800). Detailed properties of the SC and WV-SC used in follow-up experiments were presented in Table 1. The measure results were carried out on triplicate for each compost samples with an average ± standard deviation.

Total content of selected heavy metal of composts were determined using ICP-OES (Prodigy XP, LEEMAN LABS INC. USA) after digestion by wet-ashing method (HNO<sub>3</sub> + HCl + HF). The heavy metal speciation was determined using sequential extraction procedure, and exchangeable fractions were extracted with 0.11 mol L<sup>-1</sup> HAc extractable, residue from above step was extracted with 0.1 mol L<sup>-1</sup> NH<sub>2</sub>OH·HCl extractable for the reducible fractions, oxidizable fraction was extracted with 8.8 mol L<sup>-1</sup> H<sub>2</sub>O<sub>2</sub> and followed by 1 mol L<sup>-1</sup> CH<sub>3</sub>COONH<sub>4</sub>

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