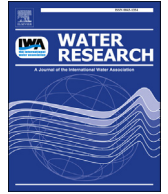




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## Watershed soil Cd loss after long-term agricultural practice and biochar amendment under four rainfall levels

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### ABSTRACT

Some heavy metals in farmland soil can be transported into the waterbody, affecting the water quality and sediment at the watershed outlet, which can be used to determine the historical loss pattern. Cd is a typical heavy metal leached from farmland that is related to phosphate fertilizers and carries serious environmental risk. The spatial-vertical pattern of Cd in soil and the vertical trend of Cd in the river sediment core were analyzed, which showed the migration and accumulation of Cd in the watershed. To prevent watershed Cd loss, biochar was employed, and leaching experiments were conducted to investigate the Cd loss from soil depending on the initial concentration. Four rainfall intensities, 1.25 mm/h, 2.50 mm/h, 5.00 mm/h, and 10.00 mm/h, were used to simulate typical rainfall scenarios for the study area. Biochar was prepared from corn straw after pretreatment with ammonium dihydrogen phosphate (ADP) and pyrolysis at 400 °C under anoxic conditions. To identify the effects of biochar amendment on Cd migration, the biochar was mixed with soil for 90 days at concentrations of 0%, 0.5%, 1.0%, 3.0%, and 5.0% soil by weight. The results showed that the Cd leaching load increased as the initial load and rainfall intensity increased and that eluviation caused surface Cd to diffuse to the deep soils. The biochar application caused more of the heavy metals to be immobilized in the amended soil rather than transported into the waterbody. The sorption efficiency of the biochar for Cd increased as the addition level increased to 3%, which showed better performance than the 5% addition level under some initial concentration and rainfall conditions. The research indicated that biochar is a potential material to prevent diffuse heavy metal pollution and that a lower addition makes the application more feasible.

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### 1. Introduction

Intensive agricultural practices enhance grain yields but also cause environmental concerns. With rapid agricultural development, phosphate fertilizer usage has increased in intensive farming areas to ensure stable crop yields (Shen et al., 2013). As phosphate fertilizer contains Cd, the continuous application of such fertilizer has been regarded as an important anthropogenic source for the accumulation of heavy metals in agricultural soils (Jones and Johnston, 1989). Leached Cd has a high probability of accumulating in the sediment at the watershed outlet, which provides a location to analyze the Cd leaching history by studying its vertical flux. To decrease soil Cd leaching, several methods have been

designed to prevent Cd loss from the soil, including a pilot study of biochar addition (Buss et al., 2016). For watershed diffuse pollution control, biochar is a potential material for pollution prevention based on the advantages of biochar treatment and its higher efficiency (Trakal et al., 2016).

Phosphate fertilizers are well known to be a major external source of soil heavy metals, resulting in serious agricultural diffuse pollution (Micó et al., 2006). Crops only absorb up to 80% of applied phosphate fertilizers, making farmland soil an important sink for residual fertilizer (Tang et al., 2010). The accumulated residual phosphate fertilizer in soils has produced detrimental environmental effects over a decade of agricultural practice (Liu et al., 2014), because this practice increases the concentration of heavy metals in the soil. Based on traditional field investigations of soil, the vertical-spatial pattern of heavy metal contamination in soil can be assessed, which is also a factor for crop production

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assessment (Xu and Chen, 2015). Because of dispersion, concealment, randomness, high uncertainty, and spatial heterogeneity, controlling agricultural heavy metal pollution is difficult due to complications in its formation and transport (Guo et al., 2014). Rapidly accumulated pollutants may be transported with water under the impacts of precipitation and tillage practice to the watershed (Razo et al., 2004). Therefore, the vertical pattern of heavy metals in sediment from a watershed outlet is hypothesized to be able to express the long-term pattern of heavy metal loss from nearby farmland soil.

Cd is a common heavy metal pollutant in agricultural watersheds, which accumulates in soil during the long-term application of phosphate fertilizers as an associated element (Shan et al., 2013). As Cd is potentially toxic and has high persistence and low solubility in water, it increases the occurrence of pollution events (Wang and Chen, 2014). Cd in farmland soils can be a pollution source to aquatic environments along surface and subsurface pathways via precipitation or irrigation. After Cd is introduced into the environment, it persists for a long time without microbial or chemical degradation compared to organic contaminants (Bolan et al., 2014). Consequently, an improved understanding of the chemical behavior and loss dynamics of Cd within a soil system is essential to create effective Cd-contaminated soil amendments (Hartley et al., 2004). The cleanup of Cd-contaminated soils and the delay of transport to waterbodies are emergency tasks for environmental protection (Wu et al., 2004), which have garnered more attention over the last few decades.

To decrease diffuse pollution leaching from farmland soil, several amendments have been suggested to immobilize heavy metals (Cao et al., 2009; Liu et al., 2017). Biochar produced from waste biomass has economic and environmental benefits and is superior for the remediation of contaminated soils and the mitigation of water pollution (Cao et al., 2011). Meanwhile, recycling and reusing agricultural solid waste has the benefit of soil organic matter conservation, which helps mitigate the decreased organic matter content caused by long-term agricultural development and agricultural practices (Ouyang et al., 2014). Biochar addition is an effective measure to improve soil quality and sequester soil Cd pollution (Lucchini et al., 2014; Park et al., 2016). Potential differences in the biochar-amended sorption rates on typical soil pollution have been observed, and the sorption mechanism has been determined (Huggins et al., 2016; Ouyang et al., 2016). In this study, we further investigate the typical heavy metal pollution control performance of soils amended with agricultural waste-derived biochars under diverse precipitation conditions.

For agricultural areas with high contents of soil organic matter, the vertical-spatial pattern of soil Cd after long-term tillage practice is an interesting issue, as its loss pattern can be demonstrated by the Cd flux in sediment from the watershed outlet (Doriz et al., 1998). Therefore, it is necessary to identify the amount of Cd lost from agricultural soil using sediment analysis and to assess the performance of biochar amendment under diverse rainfall conditions. The detailed purposes of this study are to: (1) explore the spatial-vertical pattern of the Cd concentration in farmland soils after long-term agricultural practices; (2) identify the Cd loss by using the vertical pattern of the Cd concentration in sediment from the watershed outlet; and (3) evaluate the control efficiency of biochar from corn straw after pretreatment by a catalyst under different amendment levels and rainfall intensities.

## 2. Materials and methods

### 2.1. Study area description

The agricultural farm under study is intensely developed and

situated in northeast China (47°18' N to 47°50' N, 133°50' E to 134°33' E) (Fig. 1). The main soil type in this area is albic soil, which has a high content of organic matter (Xing and Dudas, 1992). This farm has experienced long-term agricultural expansion over nearly 40 years, starting in the late 1970s, and is the largest commodity grain base in China. To ensure high crop yields, an average amount of 87 kg P/ha of phosphate fertilizer is applied in this area (Jiao et al., 2014). The averaged Cd concentration in fertilizer is approximately 7 mg/kg, which has created environmental concerns. Consequently, the farmland soil is affected by surface runoff erosion and leaching during the summer storm season (Ouyang et al., 2013). According to monitoring data, the local daily rainfall intensity ranges from 0 to 300 mm, with an annual average precipitation of 595.32 mm. Precipitation, which mostly occurs in summer, may reduce the utilization rate of fertilizers and increase their diffusivity risk, especially during extreme rainfall events.

### 2.2. Soil sampling and analysis

To highlight the spatial differences in the Cd concentration in the farmland soils after decades of tillage practices, 47 sampling sites were selected using a 1.5 km grid system over the whole watershed (Fig. 1). The on-site farmland raw soil for column experiments was also collected in the autumn of 2013. At each site, three replicate samples were taken from both the surface soils (0–20 cm depth) and subsoils (20–40 cm depth) (Fig. 1). All the soil samples were air dried, sieved to 0.2 mm, and stored at room temperature (approximately 25 °C) until treatment with a HNO<sub>3</sub>–HF–HClO<sub>4</sub> mixture. After digestion, the samples were analyzed using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). A reagent blank, parallel sample, and Cd standard (National Standard Material GBW-07401, National Standard Material GBW-07402) were run as well for quality control.

### 2.3. Watershed main-stream sediment sampling

To highlight the long-term soil Cd loss to the entire watershed, the watershed main-stream outlet was chosen as the sediment sampling site (Fig. 1). A sediment core (0–30 cm) was taken with a columnar sampler (PVC tube, 7.5 cm in diameter). To avoid turbulence at the sediment-water interface, the sampler was pressed slowly into the water during sediment acquisition. The sediment core sample was divided immediately into 1 cm thick sections using a plastic knife and then sealed in plastic bags (Ahmed et al., 2015). All samples were transported immediately after collection to the laboratory for further analysis. To analyze the Cd concentration in the sediment, the samples were digested with an acid mixture of HF–HNO<sub>3</sub>–HClO<sub>4</sub> and measured using inductively coupled plasma-atomic emission spectroscopy (ICP-AES) (IRIS Intrepid II XSP, Thermo Electron, USA).

### 2.4. Preparation and characteristics of biochar

The sorption capability of corn straw biochar has been widely analyzed. Corn straw is abundant in the studied agricultural area, and thus, the fresh local biomass was used to produce biochar. After pretreatment with ammonium dihydrogen phosphate (ADP), the biochar was pyrolyzed at 450 °C under anoxic conditions. ADP can greatly improve the biochar sorption capacity (Zhao et al., 2013). The original structure was destroyed, generating a rougher surface morphology, which can be seen in the scanning electron microscopy images of the corn straw and biochar at 1000× magnification (Fig. 2). The surface morphology of the biochar was rougher and had cracks and multiple holes. The elemental composition and physicochemical properties were analyzed. The specific surface

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