



## Research article

# Optimization of typical diffuse herbicide pollution control by soil amendment configurations under four levels of rainfall intensities



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## ABSTRACT

Herbicides are a main source of agricultural diffuse pollution due to their wide application in tillage practices. The aim of this study is to optimize the control efficiency of the herbicide atrazine with the aid of modified soil amendments. The soil amendments were composed of a combination of biochar and gravel. The biochar was created from corn straw with a catalytic pyrolysis of ammonium dihydrogen phosphate. The leaching experiments under four rainfall conditions were measured for the following designs: raw soil, soil amended with gravel, biochar individually and together with gravel. The control efficiency of each design was also identified. With the designed equipment, the atrazine content in the contaminant load layer, gravel substrate layer, biochar amendment layer and soil layer was measured under four types of rainfall intensities (1.25 mm/h, 2.50 mm/h, 5.00 mm/h and 10.00 mm/h). Furthermore, the vertical distribution of atrazine in the soil sections was also monitored. The results showed that the herbicide leaching load increased under the highest rainfall intensity in all designs. The soil with the combination of gravel and biochar provided the highest control efficiency of 87.85% on atrazine when the additional proportion of biochar was 3.0%. The performance assessment under the four kinds of rainfall intensity conditions provided the guideline for the soil amendment configuration. The combination of gravel and biochar is recommended as an efficient method for controlling diffuse herbicide pollution.

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## 1. Introduction

The intensive agricultural practices in recent years improve grain yields, but they also involve an excessive use of agrochemicals, which leads to herbicide accumulation in soils and, as a result, is a source of agricultural diffuse pollution (Tang et al., 2010; Panagopoulos et al., 2012). As a low-affinity element for soil, the herbicide leaching from farmland has become an important factor in agricultural diffuse pollution. The leaching process of herbicides is directly affected by rainfall intensity, irrigation patterns, soil properties, and related management practices (Mudhoo and Garg, 2011). Leached herbicides eventually result in aquatic environmental pollution and exerts eco-toxicity effects even at low concentrations (Sandoval-Carrasco et al., 2013). A study of diffuse herbicide pollution control with soil amendments is necessary

because the addition is regarded as an effective method to prevent the discharge of herbicides into water bodies.

Atrazine (2-chloro-4-(ethylamino)-6-isopropylamino-s-triazine) is a widely used herbicide for controlling broadleaf weeds and annual grasses (Lasserre et al., 2009), which can improve crop yields and tillage efficiencies. Atrazine is usually applied as a 38% suspending agent at a concentration of 1.5–2.0 L/ha. As a persistent and highly mobile herbicide, atrazine has been found in both groundwater and drinking water in the environment (Jablonowski et al., 2008). It has also been detected in freshwater and estuarine fauna due to its acute and chronic toxicity (Kadian et al., 2008). It is reported to act as an endocrine disruptor and, thereby, has the potential to trigger the cascading effects on human health throughout the food chain (Eykelbosh et al., 2015). Hence, it is necessary to control the transport of atrazine, which has adverse impacts on environmental security and human health.

By improving the interaction between chemicals and the soil matrix, soil amendments play a positive role in herbicide management (Ghosh and Singh, 2012). Biochar is considered to be an effective and versatile environmentally friendly soil amendment

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with potential soil conditioning properties and beneficial physical-chemical characteristics (Beesley et al., 2010). Being the most abundant agricultural waste material in China with an annual output of more than 0.2 billion tons (Chen et al., 2011), corn straw is usually chosen as the raw material for biochar. When the corn straw is preprocessed with a catalyst of ammonium dihydrogen phosphate (ADP), it has wide availability at a very low cost (Gao et al., 2011). The pretreated corn straw with ADP has been confirmed to prompt the development of the pore structure of biochar (Li et al., 2010). Due to the advantages of reuse of agricultural wastes and water pollution mitigation (Cao et al., 2011), the application of biochar is considered to be an effective measure to control herbicide diffusion.

Previous studies have explored the potential sorption for controlling pollutants by biochar production (Meng et al., 2013; Mohan et al., 2014). However, the simple addition of biochar to soil cannot be advocated in farmland, and the combined application of biochar and gravel is more suitable for field engineering. The natural gravel has the effective diameter of particles ranging from 1 to 100 mm. Gravel existence in soil can impact the soil infiltration process by influencing the soil bulk density, soil moisture content and other soil properties (Abrahams and Parsons, 1994). With its good adsorption performance, extensive availability and good resistance to impact load, it has the potential to reduce surface runoff and pollution load and increase the removal of suspended particles (Abou-Elela and Hellal, 2012; Poesen and Lavee, 1994). However, few studies have been conducted of gravel applied to farmland soil; more study is needed into this issue.

The aim of this study was to optimize an impactful configuration for removing atrazine diffusion under different rainfall intensities. The leaching experiments were designed to compare the retention capabilities of soil, gravel and biochar individually and in combination with gravel. The results can provide guidance for the use of soil amendments and enhance water security assurance. Furthermore, understanding the vertical distributions of diffuse herbicides with the application of soil amendments in intensive tillage zones is an innovative way to reduce the risk of agrochemical pollution.

## 2. Materials and methods

### 2.1. Soil collection and analysis

The soil samples were collected in June 2014 from dryland on the Bawujiu Farm in NE China. The sampling site was located in a typical intensive agricultural zone, which has suffered from a decreasing pattern of the organic matter content (Ouyang et al., 2015). Fifty grams of the sampled soil were immediately stored at a temperature of 4 °C in a dark refrigerator for background value analysis of atrazine. Other soil was sieved to 2 mm and stored at room temperature for soil column filling. To obtain the basic characteristic properties of the sampled soil, the moisture content (MC), carbon (C), soil organic matter (SOM) and particle size were measured (Jiao et al., 2014). The MC was measured with the aid of a gravimeter after drying in an oven at 105 °C. Elemental analysis was conducted using a Vario El Elemental Analyzer. SOM was determined by measuring weight loss on ignition at 400 °C. The soil particle size distributions were determined with the aid of a laser particle size analyzer.

### 2.2. Biochar preparation

The biochar (ACS) was produced from corn straws, which were washed with tap water, air-dried, smashed into powder and dipped in 5% (w/w) ADP solution at a solid to solution ratio (g/mL) of 1:10 for 24 h. After drying in an oven at 105 °C for 24 h, the samples were

put into ceramic pots with lids. In order to provide an oxygen-free condition required for producing the biochar, the ceramic pots were covered with silver paper and corn straw powders were pyrolysed at 400 °C in an oxygen-limited muffle furnace for 4 h. The following steps of ACS biochar preparation are delineated in an earlier paper (Hao et al., 2013). The biochar properties of elementals (C, N, H and O), pore diameter (PD), specific surface area (SSA), total pore volume (TPV) and micropore volume (MV) were analyzed (Zhao et al., 2013).

### 2.3. Soil column installation

Plexiglas enclosed columns (25 cm tall and 5 cm in diameter) were used to study the transport and leaching process of atrazine (Fig. 1). The soil column was filled with six layers from the bottom to the top: inverted filter layer, soil layer, biochar amendment layer, gravel substrate layer, contaminant load layer and primary filter layer. Under different soil improvement conditions, the biochar amendment layer and the gravel substrate layer were adjusted. In order to improve infiltration capacity and to prevent soil migration (Eykelbosh et al., 2015), a porous glass plate was designed and placed at the bottom of the installation. In further detail, the inverted filter layer, from bottom to top, consisted of two pieces of filter paper, a layer of 60 mm nylon membrane and 10 g of quartz sand. Then the 20 cm tall soil layer was formed with approximately 400 g of soil which was mixed several times to create consistency. The column was treated with 200 mL of distilled water to guarantee the natural soil bulk density condition and to avoid variations in the soil water content between different treatments.

After the ACS biochar mixed with soil was loaded into the column, the 2 cm biochar amendment layer was capped with a 2 cm substrate layer, which was composed of 1–3 mm, 2–4 mm and 4–8 mm grain diameter gravels. Whether the biochar amendment layer and gravel substrate layer were added depended on the specific control designs. Then 10 g of contaminated soil was loaded into the column. It was pretreated with 2 mg of atrazine, which was dissolved in 20 mL of methanol, and left overnight at 4 °C. The additional amount of atrazine was selected according to the local field conditions, 0.76 kg/ha. The top of the column was covered with a 0.5 cm thick primary filter layer using a piece of filter paper, 5 g of quartz sand and a 2 mm of porous water outlet. The covered layers allowed the even infiltration and minimized the soil surface disturbance. On each end of the column, there was a dismountable and telescoping thread plug, which could be used to control the height of the soil column. Each end of the column was connected to a 0.50 cm diameter pipe to allow the leachates to pass through. The CaCl<sub>2</sub> solution (0.01 M) was introduced at the top of the soil columns.

### 2.4. Leaching experiments with different soil amendment configurations

Batch simulated rainfall leaching experiments were designed to investigate the impact of different amendment configurations on the migration of atrazine in soil. The leaching experiment was conducted in a completely randomised design with four treatments: (1) A0, control, 24 cm of raw soil, (2) A1, 2 cm of gravel substrate layer +22 cm of soil, (3) A2, 2 cm of biochar amendment layer +22 cm of soil, (4) A3, 2 cm of gravel substrate layer +2 cm biochar amendment layer +20 cm of soil. Each treatment was subjected to four types of rainfall intensities (1.25 mm/h, 2.50 mm/h, 5.00 mm/h and 10.00 mm/h), which were set according to the local precipitation pattern. Each group had three parallel replicates. There were 16 groups and 48 samples in total.

Then the batch experiments were set up to optimize the

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