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Microbe mediated arsenic release from iron minerals and arsenic methylation in rhizosphere controls arsenic fate in soil-rice system after straw incorporation[☆]

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ABSTRACT

Arsenic (As) contamination is a global problem. Straw incorporation is widely performed in As contaminated paddy fields. To understand how straw and straw biochar incorporation affect As transformation and translocation in the soil-microbe-rice system, a pot experiment was carried out with different dosages of rice straw and straw biochar application. Results showed that both straw biochar and straw application significantly increased As mobility. Straw biochar mobilized As mainly through increasing soil pH and DOM content. Straw incorporation mainly through enhancing As release from iron (Fe) minerals and arsenate (As(V)) reduction to arsenite (As(III)). Straw biochar didn't significantly affect As methylation, while straw incorporation significantly enhanced As methylation, elevated dimethylarsenate (DMA) concentration in soil porewater and increased As volatilization. Straw biochar didn't significantly change total As accumulation in rice grains, but decreased As(III) accumulation by silicon (Si) inhibition. Straw incorporation significantly increased DMA, but decreased As(III) concentration in rice grains. After biochar application, dissolved As was significantly positively correlated with the abundance of *Bacillus*, indicating that *Bacillus* might be involved in As release, and As(III) concentration in polished grains was negatively correlated with Si concentration. The significant positive correlation between dissolved As with Fe and the abundance of iron-reducing bacteria suggested the coupling of As and Fe reduction mediated by iron-reducing bacteria. The significant positive correlation between DMA in rice grains and the abundance of methanogenic bacteria indicated that methanogenic bacteria could be involved in As methylation after straw application. The results of this study would advance the understanding how rice straw incorporation affects As fate in soil-microbe-rice system, and provide some guidance to straw incorporation in As contaminated paddy soil. This study also revealed a wealth of microorganisms in the soil environment that dominate As mobility and transformation after straw incorporation.

1. Introduction

Arsenic (As) is among the common toxic metalloids which ubiquitously exist in the environment (Nordstrom, 2002). Inorganic As is classified as a class 1 and non-threshold carcinogen (Smith et al., 2002). Paddy fields are widely contaminated with As due to

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the use of groundwater with elevated As for irrigation, the application of As pesticides, and external pollution from industrial and mining activities (Zhao et al., 2009). Rice (*Oryza sativa* L.) is the staple food for more than half of the world's population and two thirds of the Chinese population (Kennedy, 2002), however, rice has high As accumulation potential (Williams et al., 2007) and thus poses a major health concern due to increasing the dietary As intake for populations consuming rice as the staple food (Li et al., 2011; Duan et al., 2013). Therefore, there is urgent need to reduce As accumulation in rice grains.

Arsenic accumulation in rice grains is controlled by As mobility and speciation in paddy soil, which are affected by many factors. Physicochemical properties of soil are key factors regulating As mobility in soil, such as pH, Eh, DOC (Dissolved Organic Carbon) contents, DOM (Dissolved Organic Matter) components, mineral components (Fe, Mn, Al, etc) and some nutrient elements (P, Si, S, etc) (Zhao et al., 2010; Duan et al., 2013; Rafiq et al., 2017; Xu et al., 2017; Abbas et al., 2018). While microbe mediated As biotransformation plays a key role in regulating As speciation in soil, such as arsenate (As(V)) reduction, arsenite (As(III)) oxidation and methylation, as well as organic As demethylation (Yoshinaga and Rosen, 2014; Zhang et al., 2017). The relative abundance and diversity of microbes involved in As transformation can influence the fate of As and its bioavailability to rice (Zhao et al., 2010). All these chemical and microbial factors are important for As mobility and transformation in paddy soil, any practice which could modify these factors would affect As accumulation and speciation in rice grains.

Rice straw is the main by-product of rice production, and straw incorporation into paddy fields can serve as an avenue to enhance soil fertility. Recently, with the popularization of agricultural mechanization, straw incorporation is being advocated for rice production, even in As-contaminated paddy fields. However, straw incorporation may significantly modify soil physicochemical properties, which may affect As mobility in soil. It is known that rice straw is rich in organic matter, and thus, the processes of rice straw decomposition could significantly reduce soil redox potential (Tanji et al., 2003), which could in turn result in As(V) reduction and As desorption from soil minerals (Arao et al., 2009; Sahrawat, 2012; Chen et al., 2016; Wang et al., 2017). On the other hand, rice straw is rich in silicon (Si) and phosphorus (P) (Ponnamperuma, 1984). Given that rice roots take up As(III) and As(V) by Si (Ma et al., 2008) and P transporters (Kamiya et al., 2013), respectively, thus straw incorporation might reduce As uptake by rice roots through Si or P competition. Additionally, the organic matter degraded from rice straw could stimulate the growth of indigenous microbes including those responsible for As methylation and volatilization (Huang et al., 2012; Zhao et al., 2013; Zhu et al., 2017). It has been reported that rice straw addition into paddy soil elevated the abundance of *arsM* gene in the rhizosphere soil, and increased the concentration of methylated As species in the soil solution and rice grains (Jia et al., 2013; Ma et al., 2014). Therefore, the effects of straw incorporation on rice As accumulation cannot be ignored.

As mentioned above, straw incorporation might increase rice As accumulation through promoting As release and As(V) reduction to As(III). On the contrary, straw incorporation could also decrease As uptake and accumulation through increasing the content of available Si and P in soil. Therefore, it is of great importance to fully understand the effects of straw incorporation on rice As accumulation. Although, so far, several studies have investigated the effects either on soil physicochemical properties or on microbial As transformation, it is still unclear which are the predominant factors after straw incorporation. In addition, previous studies referring to microbial effects mostly focused on *arsM* and As methylation, while other factors and processes were largely ignored. Moreover, it is still unknown how straw incorporation affects the microbial community in rice rhizosphere, and which microbial species are the predominant ones and how these microbes affect As biochemical dynamics in rice rhizosphere after straw incorporation.

In addition, biochar is recently becoming a popular material for soil remediation, because biochar has large surface area, and high capacity to absorb heavy metals and organic pollutants (Beesley et al., 2011, 2014; Ahmad et al., 2014; Khan et al., 2014; Rizwan et al., 2016; Niazi et al., 2018). Several studies have demonstrated that biochar could increase the mobility of As in soil (Chen et al., 2016; Wang et al., 2017; Qiao et al., 2017, 2018), but decreased As uptake by tomato plants (Beesley et al., 2013), or had little effect on As accumulation in rice plants (Ma et al., 2014), or increased As accumulation in rice shoots (Zheng et al., 2012). The results are inconsistent. In addition, it is more economic for famers to incorporate straw directly, rather than conversing straw to straw biochar. Hence, it is of practical demand to compare different methods of straw incorporation on As behaviors in soil-rice system, and clarify which incorporation method of rice straw is more beneficial for As remediation.

Therefore, the objectives of this study were to (1) clarify the effects of rice straw and straw biochar addition on both chemical and microbial factors that may influence As fate in rice rhizosphere, and find out which factors are predominant in regulating As mobility in rhizosphere and As accumulation in rice grains after straw and straw biochar application, (2) characterize the response of rhizosphere microbial community to straw and straw biochar incorporation, and identify the potential microbes controlling As biochemical behaviors in rice rhizosphere after straw and straw biochar incorporation, and (3) compare the effects of direct straw incorporation and straw biochar incorporation on As mobility and transformation in soil-rice system, and clarify which incorporation method of rice straw is more appropriate for As-contaminated paddy soil.

2. Materials and methods

2.1. Pot experiments

Pot experiments were carried out in a greenhouse setting from June to November of 2016 in Jiaxing city (Zhejiang province, China). The soil used for pot experiments was collected from an As contaminated paddy field, located at Qiyang city, Hunan province. Prior to potting, the soil, straw biochar and straw were air dried, passed through a 2 mm sieve and mixed together. The As concentration of the soil was 84.7 mg kg^{-1}. Rice straw used for incorporation was collected from paddy fields in Jiaxing (Zhejiang province, China). Rice straw biochar was bought from a company in Nanjing (Jiangsu province, China). The physicochemical properties of the soil, straw and straw biochar are shown in Table 1. Each experimental pot (20 cm diameter, 30 cm height) was filled with 5 kg soil. Approximately 1 kg soil was placed in the nylon mesh (24 μm) bag (height of 16 cm, diameter of 8 cm) which was regarded as the rhizosphere soil, and the other 4 kg soil outside the bag was regarded as the bulk soil. The Weiyou 8 rice cultivar was used for the potted plants. The cultivar is a hybrid rice that is widely grown in southern China. There were five rice seedlings in each pot. There were 7 treatments and 4 replicates for each treatment, totally 28 pots. To collect the maximum amount of gaseous As, the straw and biochar application rate were higher than local practical application. The treatments were as follows: (1) Control (No straw biochar nor straw); (2) 1% straw biochar; (3) 2% straw biochar; (4) 4% straw biochar; (5) 1% straw; (6) 2% straw; (7) 4% straw. The rice planting process and soil Eh measurement are described in detail in