



## Short Communication

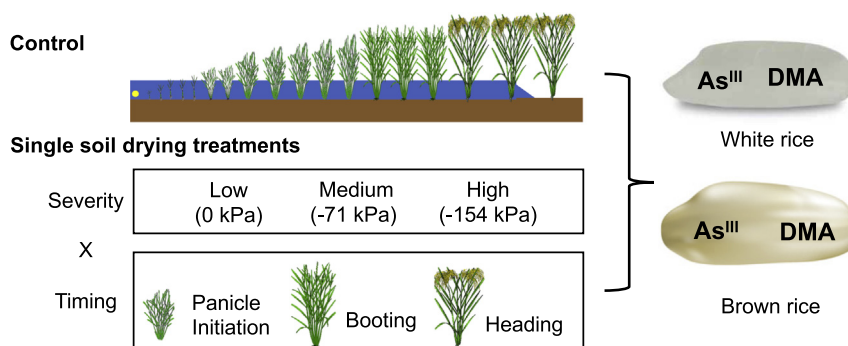
## Irrigation management for arsenic mitigation in rice grain: Timing and severity of a single soil drying

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## HIGHLIGHTS

- Continuously flooded irrigation favors the accumulation of arsenic in rice grain.
- Treatments with one soil drying period differing in timing and severity were tested.
- Across all timings, severe soil drying ( $\leq -71$  kPa) decreased total As concentration.
- However, inorganic As (the most toxic to humans) not always decreased.
- Irrigation management affects both total As and As speciation within rice grain.

## GRAPHICAL ABSTRACT



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## ABSTRACT

The accumulation of arsenic (As) in rice grain is a public health concern since As is toxic to humans; in particular, inorganic As can cause many chronic diseases including cancer. Rice crops are prone to accumulating As, in part, due to the anaerobic soil conditions triggered by the traditional continuously flooded irrigation practice. The objective of this study was to determine how the severity and the timing (i.e. crop stage) of a single soil drying period impact total As concentration and As speciation within the rice (both white and brown) grain, compared to a continuously flooded (CF) control. Drying the soil until the perched water table reached 15 cm below the soil surface (same severity as in the “Safe Alternate Wetting and Drying”), which in this study corresponded to a soil (0–15 cm) water potential of  $\sim 0$ , did not decrease grain As concentrations, regardless of timing. Drying the soil to Medium Severity [MS: soil (0–15 cm) water potential of  $-71$  kPa] or High Severity [HS: soil (0–15 cm) water potential of  $-154$  kPa] decreased total As by 41–61%. However, inorganic As did not always decrease because the severity and the timing of soil drying affected As speciation within the grain. Overall, the soil had to be dried to HS and/or late in the growing season (i.e., at booting or heading instead of at panicle initiation) to decrease inorganic As concentration in the rice grain. This study indicates that the imposition of a single soil drying period within the growing season can mitigate As accumulation in rice grain, but it depends on the severity and timing of the drying period. Further, irrigation management affects As speciation within the rice grain and this must be considered if regulations on inorganic As are based on a percentage of total As measured.

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## 1. Introduction

Rice is the primary staple food for more people on Earth than any other crop and provides one quarter of the global calorie intake

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(GRISP, 2013). However, rice can be a significant route of human exposure to arsenic (As), especially in populations with high rice consumption (Meharg, 2004; Bhowmick et al., 2018). Different forms of As are found in rice grains, the most common being the inorganic species arsenite ( $\text{As}^{\text{III}}$ ) and arsenate ( $\text{As}^{\text{V}}$ ) and the organic species monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA). While the toxicities of the organic As species are considered to be low (Hirano et al., 2004), exposure to inorganic As is associated with many types of cancer, in addition to other non-carcinogenic diseases such as diabetes and hypertension (Bjørklund et al., 2017). Inorganic As accumulates in the bran, causing paddy and brown rice to have higher inorganic As concentration than white rice (Sun et al., 2008). Maximum levels for inorganic As of 0.35 and 0.2  $\text{mg kg}^{-1}$  in paddy and white rice, respectively, have been adopted by CODEX, a joint commission of the World Health Organization and the Food and Agriculture Organization (CODEX, 2018).

Arsenic in rice grain originates from the soil, where it can be naturally present (e.g., parent rock) or carried over via irrigation (i.e., water with high As levels) or other sources (e.g., arsenical pesticides, urban residue) (Kumarathilaka et al., 2018). Arsenic tends to accumulate in rice for two reasons. First, the uptake of  $\text{As}^{\text{III}}$  and, in part, MMA are mediated by the silicon uptake pathway; rice being a silicon accumulator is therefore naturally effective in taking up As from the soil (Suriyagoda et al., 2018). Second, rice is commonly grown under flooded conditions for most of the growing season. Anaerobic soil conditions increase As bioavailability in the soil because it triggers the reduction of  $\text{As}^{\text{V}}$  to  $\text{As}^{\text{III}}$ , which is more mobile in the soil, and of  $\text{Fe}^{\text{III}}$  to  $\text{Fe}^{\text{II}}$ , dissolving iron hydro(oxides) that bind to As and, subsequently, releasing As to the soil solution (Meharg and Zhao, 2012). In addition, the high irrigation input to rice contributes to As building up in the soil if the irrigation water has high As levels (Kumarathilaka et al., 2018).

Since soil flooding is a major contributor to As accumulation in rice grains, irrigation practices that include one or more periods of soil drying within the growing season [e.g., alternate wetting and drying (AWD), intermittent flooding, mid-season drain] have been proposed as mitigation strategies (Bakhat et al., 2017). However, the impact of these irrigation practices on grain As concentration is highly variable, with decreases of 0 to 90% in total grain As being reported (Arao et al., 2009; Linquist et al., 2015; Honma et al., 2016; Yang et al., 2016; Norton et al., 2017a; Carrijo et al., 2018). This variability may be attributed, at least in part, to differences in the severity and timing (i.e. crop stage) of soil drying.

The severity of soil drying affects soil redox potential (Eh), which is intrinsically related to soil As bioavailability, and subsequently As uptake. Grain As concentration decreases sharply with an increase in soil Eh from  $-200$  to  $-100$  mV and tends to plateau at very small concentrations ( $<0.02 \text{ mg kg}^{-1}$ ) as soil Eh increases above 0 (Honma et al., 2016). Carrijo et al. (2018) found that total grain As concentration decreased with increasing soil drying severity, although drying the top soil (0–15 cm) to a water potential lower than  $-33$  kPa did not translate into a further decrease in grain As concentration. However, their study was limited to total As concentrations and, given that As speciation may be affected by soil drying (Yamaguchi et al., 2014), their conclusions may not apply to all As species present in the grain.

The effect of timing of soil drying on grain As concentration has been scarcely investigated, with the exception of a few pot studies (e.g., Arao et al., 2009; Li et al., 2009). Soil drying would be most effective in minimizing grain As concentration if imposed when As uptake is highest under flooded conditions. However, predicting temporal As uptake is a difficult task considering the many variables regulating As availability and uptake. Reports that the expression of Lsi1, a root transporter involved in the uptake of  $\text{As}^{\text{III}}$ , is enhanced around the heading stage (Yamaji and Ma, 2007; Ma et al., 2008), suggest that this could be a key stage for As uptake. In contrast, Li et al. (2015) reported that of the total As present in aboveground tissues at harvest, 64% had been taken up at the jointing (~panicle initiation) and booting stages, and

they attributed that to enhanced nutrient uptake and root size during this period. In addition, As uptake is strongly influenced by the formation of iron plaques on the surface of rice roots, and their formation and capacity of sequestering As is dependent on crop stage (Garnier et al., 2010; Awasthi et al., 2017; Yu et al., 2017). For example, Mei et al. (2012) found that plants at the bolting (~booting) stage showed higher root radial oxygen loss and higher root porosity than plants at the tillering stage, and this translated into higher As sequestration in the root plaque and lower As uptake.

In this study, we sought to quantify how the severity and the timing of soil drying impact total As and As speciation within the rice grain. We hypothesized that grain arsenic concentration (total and individual species) would decrease with increasing soil drying severity independent of crop stage.

## 2. Material and methods

### 2.1. Study site characteristics

A field experiment was conducted at the Rice Experiment Station ( $39^{\circ}27'47''\text{N}$ ,  $121^{\circ}43'35''\text{W}$ ) in Biggs, California, USA, during the summer of 2016. The soil at the site is a Vertisol, comprised of fine, smectitic, thermic, Xeric Epiaquerts and Duraquerts, with a soil texture of 29% sand, 26% silt and 45% clay, a pH of 5.3, 1.06% organic C and 0.08% total N (Pittelkow et al., 2012). Total As concentration in the soil was  $3.85 \text{ mg kg}^{-1}$  (Carrijo et al., 2018) and in the irrigation water it averaged  $1 \mu\text{g L}^{-1}$  in the growing season. Total As in irrigation water was measured by collecting water samples from the main irrigation canal on four sampling dates (July 8th and 25th, August 9th and September 16th). Samples were immediately filtered [through glass microfiber filter paper (Whatman GF/F)] and acidified with nitric acid (67–70%, trace metal grade) to a pH of 2 prior to storage at  $4^{\circ}\text{C}$ . Total As in water was quantified by inductively coupled plasma (ICP) mass spectrometry (MS), following the same methodology described for rice grains in Section 2.6.2. The climate is Mediterranean, and the total precipitation and average daily temperature over the growing season (May through October) was 10.1 mm and  $21.7^{\circ}\text{C}$ , respectively (CIMIS, 2018).

### 2.2. Treatments

There were ten irrigation treatments: a continuously flooded (CF) control, which was maintained flooded (i.e., standing water maintained at  $\sim 12$  cm above the soil surface) from sowing to three weeks before harvest (pre-harvest drain), and nine treatments in which a single soil drying period was imposed, and that represented a combination of three timings and three severities of soil drying. The three timings, which determined the onset of the soil drying period, were: at panicle initiation, during booting and at 50% heading (this extended into the early grain filling period for most treatments). We did not include soil drying timings that were earlier than panicle initiation due to the risk of potential for high fertilizer nitrogen losses (LaHue et al., 2016). The severities were: Low Severity (LS - reflooded when the perched water table reached 15 cm below the soil surface), Medium Severity (MS - reflooded when the soil volumetric water content at the 0–15 cm soil depth reached 35%), and High Severity (HS - reflooded four days after the MS treatments). The choice on severity treatments aimed at representing a wide range of severities, from the LS, which is the severity used in a common form of AWD known as “Safe-AWD” (widely adopted in some Asian countries and considered to not limit rice yields) (Bouman et al., 2007; Lampayan et al., 2015), to the HS, which is considered to limit rice yields (Carrijo et al., 2018). All soil drying treatments underwent a single drying period according to their respective timing/severity, and except for this period, followed the same water management as in the CF. Within each timing, all plots started drying together and plots of the same severity were reflooded at the same time when, on average, the targeted severity was reached across plots. No

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