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Effect of water management on cadmium and arsenic accumulation by rice (*Oryza sativa* L.) with different metal accumulation capacities

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Abstract

Purpose Water management affects the bioavailability of cadmium (Cd) and arsenic (As) in the soil and hence their accumulation in rice grains and grain yields. However, Cd and As show opposite responses to soil water content, but information, particularly on irrigation, is missing on a field scale. The purpose of the present study was therefore to find a water management regime that can lower accumulation of both Cd and As in grain without yield loss.

Materials and methods Two rice (*Oryza sativa* L.) cultivars, A16 and A159, with different grain Cd accumulation capacities were employed in field plot experiments with four water management regimes comprising aerobic, intermittent, conventional practice and flooded. The dynamics of Cd and As bioavailability in the soil and Cd and As concentrations in roots, straw and grains were determined at the early tillering, full tillering, panicle initiation, filling and maturity stages of crop growth.

Results and discussion The lower water content regimes (aerobic and intermittent) mostly led to higher soil HCl-extractable Cd than the higher soil water content regimes (conventional and flooded). HCl-extractable As in contrast was favoured by the higher soil water content treatments. Conventional and flooded irrigation accordingly gave higher plant As concentrations but lower Cd compared to aerobic and intermittent irrigation. Cd concentrations in roots and straw of both varieties increased with growth stage, especially in aerobic and intermittent regimes, while As concentrations in plants showed little change or a slight decrease. As the water irrigation volume increased from aerobic to flooded, brown rice Cd decreased from 1.15 to 0.02 $mg\,kg^{-1}$ in cultivar A16 and from 1.60 to 0.05 $mg\,kg^{-1}$ in cultivar A159, whereas brown rice As increased. Aerobic and flooded treatments produced approximately 10–20 % lower grain yields than intermittent and conventional treatments. Cultivars with low Cd accumulation capacity show higher brown rice grain As than those with high Cd uptake capacity.

Conclusions Of the four water management regimes, the conventional irrigation method (flooding maintained until full tillering followed by intermittent irrigation) ensured high yield with low Cd and As in the brown rice and so remains the recommended irrigation regime.

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1 Introduction

Cadmium (Cd) is a highly toxic heavy metal, and it can readily accumulate in crops and thus lead to chronic toxicity disease in livestock and humans (Satarug et al. 2003). Arsenic (As) is also hazardous to health because it is a human carcinogen and is easily taken in through the food chain (Rahman et al. 2008). Agricultural soils contaminated by Cd and As have become an important issue as a result of industrial activities in the proximity of agricultural areas, excessive application of contaminated fertilizers and manures, and irrigation with metal(loid)s-contaminated water (Roberts et al. 2007; Chen et al. 2008; Williams et al. 2009; Arao et al. 2010). Rice, the most important staple food in Asia, has been considered to be a major source of Cd and As intake by humans in some parts of Japan, India and China (Tsukahara et al. 2003; Mondal and Polya, 2008; Liang et al. 2010). Minimizing the intake of Cd and As from rice in the diet is therefore an important health issue, and it is desirable to limit the concentrations of heavy metals in rice grains to control the potential health risks (Jackson et al. 2005; Granta et al. 2008).

Uptake of metal(loid)s from the soil by plants can be influenced by a number of plant characteristics (including nutritional status) and also by edaphic factors including the bioavailability of metals which can regulate their assimilation by plants. Studies have confirmed that different varieties of rice have different capacities to accumulate Cd and As from the soil (Yu et al. 2006; Zhang and Duan 2008; Yan et al. 2010). Selection and breeding of rice varieties with low Cd and As accumulation is important for lowering Cd and As risks (Zhang and Duan 2008). Efforts have been made to lower metal concentrations in rice and metal toxicity by changing metal availability in the soil to produce safer food (Li et al. 2009; Fan et al. 2010; Gu et al. 2011). Water management is a direct method that is effective in controlling Cd and As bioavailability in soils and uptake by rice grains (Xu et al. 2008; Arao et al. 2009). Under flooded conditions, the soil Eh is very low and soil sulphur (S) will potentially be reduced to S^{2-}, and as a result, Cd will form CdS which has very low aqueous solubility (Bingham et al. 1976). In a pot experiment, it was demonstrated that flooding the soil during plant growth decreased the Cd concentrations in rice grains, whereas fast CdS oxidation when soils were drained during the grain filling stage gave high Cd in rice (Arao et al. 2009). In contrast, under low Eh (0 to −200 mV) conditions, soil As(V) is reduced to more soluble As(III) (Masscheleyn et al. 1991). Moreover, FeOOH, the main sorbent for As(V) in soils, is readily dissolved under anaerobic conditions, and as a result, As(V) is released and reduced to As(III) (Fitz and Wenzel 2002; Takahashi et al. 2004; Norra et al. 2005). Studies show that aerobic conditions decrease the availability of As in soils with a resulting decline in As concentrations in rice (Xu et al. 2008; Liu et al. 2010). However, both Cd and As can occur together as contaminants in paddy fields, and they can accumulate simultaneously in rice plants (Williams et al. 2009). Most of the studies that have been carried on the regulation of rice Cd or As uptake by water management have used pot experiments under greenhouse conditions. There is insufficient information about the effects of water management on metal(loid)s accumulation in rice at the field scale, especially investigating both Cd and As together.

The aim of the present study was to develop an optimum water management regime to lower both Cd and As accumulation in rice grains without yield loss. Experiments were therefore established to investigate the effects of different water management regimes under field conditions on the plant availability of soil Cd and As and their effects on Cd and As accumulation in different plant parts (roots, straw and grains) at different growth stages of two varieties with different Cd concentrations in the brown rice.

2 Materials and methods

2.1 Site description and soil properties

The experimental site was located in a conventional agricultural field near Hangzhou city, Zhejiang province, east China. The climate of the study area is moist monsoon with an average annual precipitation of approximately 1,376 mm with the maximum during June and September, and a mean temperature of 16 °C. The predominant wind direction is from the northwest in winter and southeast in summer. Agriculture is the principal land use with a typical rotation system of rice from summer to autumn and wheat or oilseed rape from winter to spring of the following year. The soil is a Typical Fe-leach-Stagnic Anthrosols (Gong 2007), selected properties of which were analysed by standard methods (Sparks et al. 1996). Soil pH (soil/water ratio of 1: 2.5) of the top 15 cm arable layer was 5.30, organic carbon content was 16.9 $g kg^{-1}$ and cation exchange capacity was 13.5 cmol $(+) kg^{-1}$. Soil total N, P and K were 1.46, 0.68 and 10.2 $g kg^{-1}$, respectively. Soil available N, P and K were 95.2, 26.6 and 66.0 $mg kg^{-1}$, respectively. Soil *aqua-regia* Cd, Zn, Cu and As were 0.48, 147, 26.0 and 6.49 $mg kg^{-1}$, respectively. The experimental soil was slightly contaminated with Cd according to the Chinese Soil Environmental Standard GB 1995–15618 of ≤0.2 $mg kg^{-1}$ (State Environmental Protection Administration of China, SEPA 1995).

2.2 Field experiment

Two rice cultivars were compared, Zhongxiang No. 1 (A16) with low Cd accumulation in the brown rice and Indonesia (A159) with high Cd accumulation (our unpublished previous data). The differences in arsenic accumulation by these two cultivars were not known. The seeds of the two cultivars were sown on May 28, 2010 and transplanted on June 21, 2010 with a distance between seedlings of 25 cm. All plots were flooded for 7 days after transplanting and then the different irrigation treatments began. There were four water management treatments: (1) aerobic (Aero.), the water was discharged to leave a water layer ~5 cm deep in the narrow ditch around the plot (about 10 cm lower than the rice field) to maintain aerobic conditions in the plot; (2) intermittent (Inter.), the plot was flooded with about 3 cm water then the water layer was allowed to decrease gradually through evaporation and leaching; when the field soil was dry and similar to the 'aerobic' treatment, it was flooded again; The duration of each cycle was about 7 days at the earlier growth stages and about 5 days from full tillering stage to harvest; (3) control (CK), the plot was irrigated following local conventional irrigation methods to maintain flooded conditions until full tillering stage (August 16) followed by intermittent irrigation; and (4) flooded (Flood.), the plot was flooded during the entire crop growth season. There were three replicates of each treatment fully randomized in the field. Each plot was 4×6 m, giving an area of 24 m^2, with a 1-m-wide protective buffer zone set up between adjacent plots, and each plot was irrigated independently. The total volumes of irrigation water used were ~300, 3,702, 6,203, and 8,704 $m^3 ha^{-1}$ in the aerobic, intermittent, control and flooded treatments, respectively (Table 1). Rainfall was recorded (Fig. 1) using an automatic rainfall recording instrument (WatchDog 2900ET Weather Station, Spectrum Technologies, Plainfield, IL), and the total rainfall over the whole crop growth period was 444.2 and 474.3 mm (equal to 4,442 and 4,743 $m^3 ha^{-1}$) for A16 and A159 (harvested on October 2 and October 16), respectively (Table 1).

Other agronomic techniques were the same for all treatments. Basal nutrients consisted of 375 $kg ha^{-1}$ mixed fertilizer ($N/P_2O_5/K_2O$=15:15:15) applied 1 day before transplanting. The first topdressing was applied on July 1 (10 days after transplanting) at a rate of 120 $kg ha^{-1}$ of urea (containing 46 % N). The second topdressing was applied on July 7 and comprised 150 $kg ha^{-1}$ urea, 150 $kg ha^{-1}$ mixed fertilizer and 120 $kg ha^{-1}$ KCl (equivalent to 62 % K_2O).

2.3 Sample collection and pre-treatment

Plant samples and the corresponding 0–15-cm arable layer fresh soil samples were collected during plant growth at the early tillering (July 22), full tillering (August 16), panicle initiation (September 3), filling (September 26 for A16, October 12 for A159) and maturity stages (October 2 for A16, October 16 for A159). All plant and soil samples were refrigerated and taken to the laboratory within 4 h after collection. Plant samples were rinsed with deionized water, wiped with absorbent paper and dried below 50 °C. About 14 g of the fresh soil samples was weighed and mixed with 46 ml 0.1 M HCl to extract the soil available Cd and As (Sparks et al. 1996). The water content of the fresh soil was determined simultaneously.

2.4 Chemical analysis

Soil total and available nitrogen were determined by Kjeldahl digestion and distillation. Soil available phosphorus (P) was extracted with 0.1 M HCl. Total P was determined by $H_2SO_4/HClO_4$ digestion and analysed by the molybdenum

Table 1 Volumes of irrigation water and rainfall over the whole crop growth period

Water management treatment	Irrigation water volume ($m^3 ha^{-1}$)	Rainfall[a] (mm)		Rainfall ($m^3 ha^{-1}$)		Total volume: irrigation water plus rainfall ($m^3 ha^{-1}$)	
		A16	A159	A16	A159	A16	A159
Aerobic	300	444.2	474.3	4,442	4,743	4,742	5,043
Intermittent	3,702	444.2	474.3	4,442	4,743	8,144	8,445
Control	6,203	444.2	474.3	4,442	4,743	10,645	10,946
Flooded	8,704	444.2	474.3	4,442	4,743	13,146	13,447

[a] A16 and A159 were harvested on October 2 and October 16, 2010, respectively

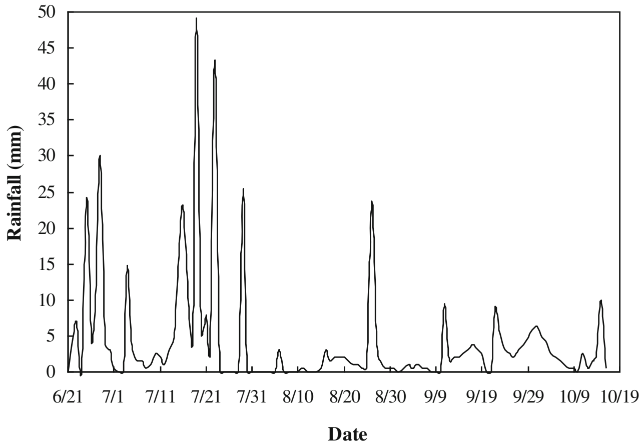


Fig. 1 Rainfall record during crop growth

blue method. Soil available K was determined by flame photometry (Model 6400-A, Shanghai Analytical Instrument Factory, Shanghai) after extraction with 1 M NH_4OAc. Total K was determined using flame photometry after *aqua-regia* digestion. Soil total heavy metal concentrations were determined by atomic absorption spectrophotometry (Varian SpectrAA 220FS, 220Z; Varian, Palo Alto, CA) after digestion of 0.25 g sub-samples with 12 ml of HCl-HNO_3 (4:1, *v*/*v*). Soil HCl-extractable cadmium was determined by flame atomic absorption spectrophotometry using a graphite furnace (Varian SpectrAA 220Z). Soil total As was digested with HNO_3 (10 ml) and H_2SO_4 (2 ml), and available As was extracted with 0.1 M HCl, and both were determined by atomic fluorescence spectrometry (AFS-930, Beijing Jitian Instruments). Replicate samples, blanks and a certified reference material (GBW07401, provided by the Institute of Geophysical and Geochemical Exploration, Langfang, Hebei province, China) were included in all analyses for quality control.

Plant (separate husk, brown rice, straw and root) sub-samples (0.5 g) were digested using a mixture of 6 ml HNO_3 and 4 ml $HClO_4$, and a certified reference material (GBW07603, Institute of Geophysical and Geochemical Exploration, Langfang, Hebei province, China) was included for quality control. All chemicals used were of analytical reagent grade, and the reference material results obtained by the methods described above were within the certified ranges of Cd. Straw, root, husk and brown rice samples (0.2–0.3 g) were digested with 6 ml high-purity HNO_3 and 2 ml H_2O_2 in a microwave digester, and the As concentration in the digest solution was determined by atomic fluorescence spectrometry. Concentrations of Cd were determined by flame atomic absorption spectrophotometry.

2.5 Statistical analysis

Statistical analysis was performed using one-way analysis of variance, and the LSD test or independent samples *t* test was used to compare mean values using the SPSS version 16.0 for Windows software package. Data are presented as mean± standard error of the mean.

3 Results

3.1 Dynamics of soil HCl-extractable Cd and As concentrations

Within the growth period, soil HCl-extractable Cd increased with time and reached a maximum concentration at maturity except in the aerobic and intermittent regimes for A16 and the aerobic treatment for A159 (Fig. 2). These two irrigation regimes led to higher HCl-extractable Cd in the soil than the flooded treatment at all growth stages of both rice cultivars.

In contrast to Cd, soil HCl-extractable As concentrations in the control and flooded plots were significantly higher than in the aerobic treatment at all growth stages of both cultivars and in the intermittent regime at panicle initiation and grain filling (Fig. 2). The aerobic and intermittent treatments showed a decreasing trend in soil HCl-extractable As over the whole growing period. However, in the control and flooded plots the highest soil HCl-extractable As was found at the panicle initiation stage (Fig. 2).

3.2 Cd and As concentrations in different plant parts

Cd concentrations in straw of both cultivars increased in all treatments as plant growth proceeded but decreased at maturity under flooding, particularly in cultivar A159 (Fig. 3). In both cultivars, the Cd concentrations in straw of the aerobic and intermittent flooding treatments were significantly higher than those of the control and flooded plots from panicle initiation to maturity. In contrast to a high Cd accumulation in A159 grains, A159 straw showed a lower Cd concentration at maturity than A16 in the aerobic and intermittent flooding treatments. Total straw As concentrations decreased at early growth stages but increased at the grain filling stage of A16 with the exception of the aerobic treatment in which the increase was observed at panicle initiation (Fig. 3). In A159, this increase was found at panicle initiation. Overall, straw As concentrations in the four water regimes followed the sequence aerobic<intermittent<control<flooded.

Root Cd concentrations increased over the whole growth period with significantly increased concentrations in the aerobic and intermittent over the control and flooded plots (Fig. 4). The aerobic irrigation regime gave the highest root Cd concentrations followed by intermediate>control> flooded for both rice varieties. Root As concentrations showed the opposite trend to Cd. Arsenic decreased at early growth stages but increased slightly at the filling stage. Root As followed the sequence aerobic<intermittent<control< flooded under the different water management treatments (Fig. 4).

Brown rice and husk Cd concentrations showed the same trend, i.e. aerobic>intermittent>control>flooded (Fig. 5). The flooding treatment and the control led to slightly or significantly higher brown rice and husk As than the aerobic or intermittent (Fig. 5). Cultivar A16 had lower Cd concentrations but higher As in the brown rice than A159. Arsenic concentrations in both varieties followed the sequence roots>straw>husk>grain and Cd gave roots>straw> grain>husk (Figs. 3, 4 and 5).

3.3 Effects of water management on grain yields

The different water regimes influenced the grain yields of both varieties. The grain yields of both varieties in the control and intermittent plots were significantly higher than those in the aerobic and flooding treatments (Fig. 6). The two varieties did not differ significantly in grain yield except for the flooded plots in which A159 had higher grain yields than A16.

4 Discussion

The present study demonstrates that on a field scale, soil Cd and As bioavailability and their accumulation in rice were greatly affected by the water management regime. The flooded and conventional treatments had lower soil HCl-extractable Cd than the intermittent and aerobic treatments at all growth stages of both rice varieties. However, soil HCl-extractable As increased in the flooded plots (Fig. 2). These results are similar to those obtained in a pot experiment conducted by Arao et al. (2009) who found that flooded conditions increased As and decreased Cd concentrations in the soil solution. As rice growth proceeded, soil HCl-extractable Cd increased with some exceptions at panicle initiation and grain filling in the low water content (aerobic and intermittent) treatments, and the fraction of As extracted by HCl showed the highest concentration in the flooded treatment and the control at panicle initiation in

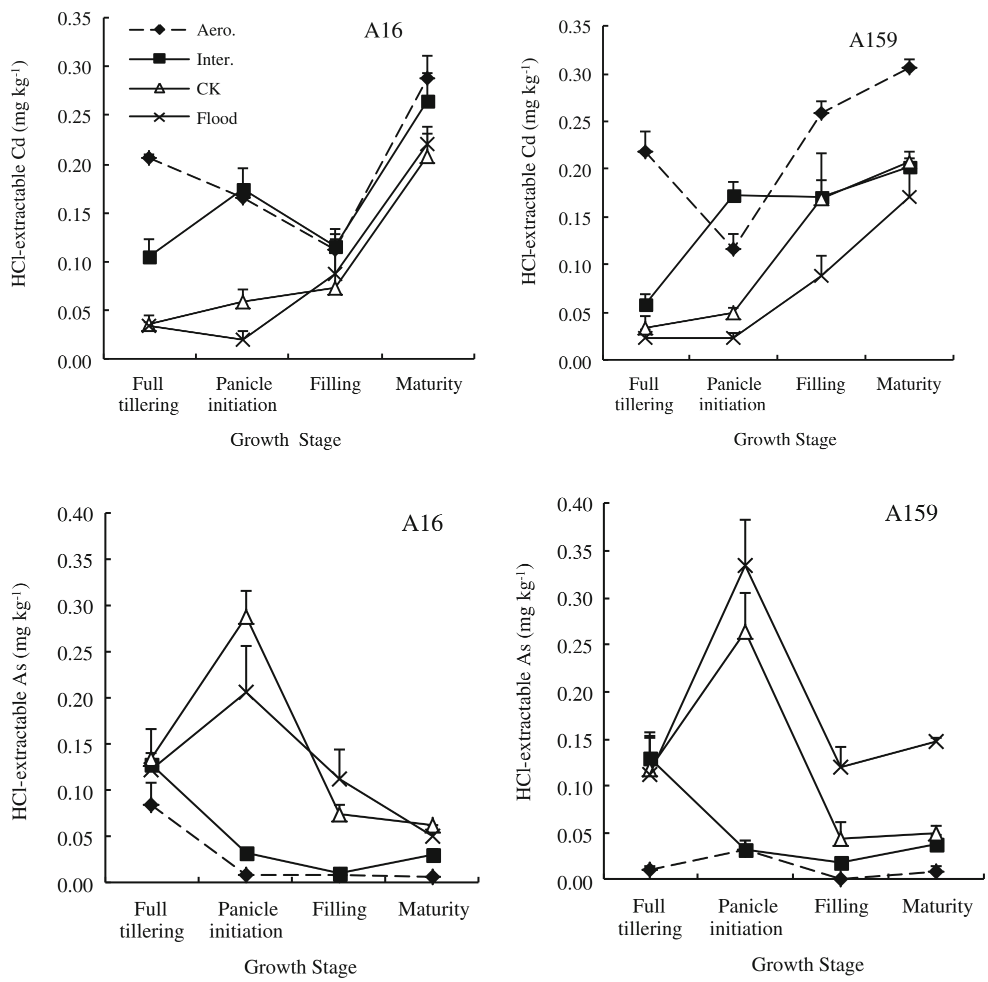
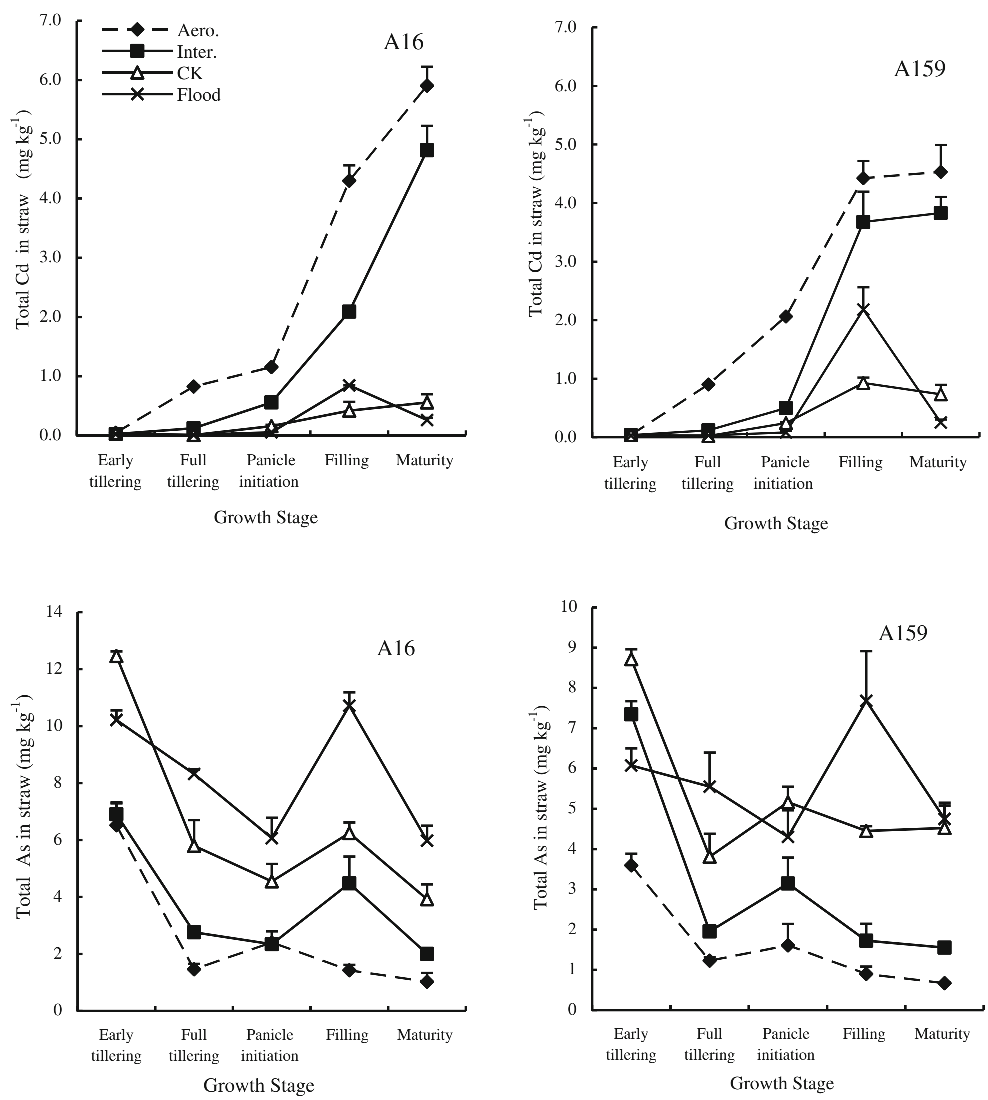


Fig. 2 Soil HCl-extractable Cd and As concentrations at different rice growth stages

the case of both rice varieties, but decreased at filling and maturity (Fig. 2). This was also similar to the findings of Arao et al. (2009). Liu et al. (2010) also found that flooding led to very high As concentrations in the soil solution at the booting stage compared to non-flooded and alternating flooded and non-flooded treatments, but low As concentrations were found at maturity, indicating that sometime after flooding a new equilibrium was established between the different As species in the soil. This equilibrium may be related to soil and rhizosphere conditions at different stages of plant growth, and this requires further research.

Metal(loid)s concentrations in rice grains are controlled by two pathways, uptake by the roots and translocation from roots to shoots and then to the rice grains (Liu et al. 2007). Thus, Cd and As in the roots and straw can have large effects on their concentrations in the grain. Li et al. (2009) concluded that aerobic conditions can lower As accumulation in rice straw during either the vegetative or the reproductive growth stage, and similarly, the present experiment shows that the straw and roots of both rice cultivars in the aerobic and intermittent irrigation regimes had higher Cd and lower As concentrations than those in the high water content treatments (flooded and control; Figs. 3, 4 and 5). In our field study, As concentrations in straw and roots decreased during the vegetative growth stages (early tillering to full tillering) because of biomass dilution effects in all treatments, but there was greater As accumulation during reproductive growth (full tillering to panicle initiation or panicle initiation to filling). In the present field study, the increase in Cd in straw and roots was more pronounced during reproductive growth from panicle initiation to filling (Figs. 3, 4 and 5). Our results confirm earlier work by Arao et al. (2009) who also concluded that during the heading period of rice water management can regulate Cd and As concentrations in the grains under pot experiment conditions. In conclusion, these results indicate that the reproductive growth period plays a more important role than earlier growth stages in controlling metal(loid) accumulation in rice straw and roots.

Fig. 3 Cd and As concentrations in the straw at different rice growth stages



Some studies have shown that aerobic conditions can depress As uptake by rice plants (Li et al. 2009; Hua et al. 2011). In our experiment, the intermittent and aerobic treatments also lowered As concentrations in the brown rice, but these two treatments also increased Cd concentrations substantially compared to the control and the flooded plots (Fig. 5). Arao et al. (2009) found that aerobic conditions increased Cd concentrations in rice grains. However, Yang et al. (2009) drew the opposite conclusion that moderate soil drying increased Cd concentrations in rice roots but decreased them in rice grains. This disagreement may result from the use of different experimental conditions. The intermittent and control plots produced higher grain yields than the aerobic and flooded plots (Fig. 6), and this is consistent with the findings of Yang et al. (2009). Considering the effects of water management on accumulation of the two heavy metal(loid)s in grain and the grain yields together, the control gave moderate Cd and As concentrations and higher grain yields, and this may be the best water regime for safe crop production in our experimental soil.

In the present study, the control plots were irrigated using the local conventional irrigation method, i.e. maintenance of flooding after transplanting until the full tillering stage and then followed by intermittent irrigation. Thus, at the later growth stages the soil was mainly under intermittent irrigation conditions which would result in increased soil Eh and promote plant growth. Moreover, these conditions lower the bioavailability of both Cd and As (Fig. 2), resulting in lower Cd and As concentrations in the brown rice (Fig. 5).

There have been few studies on differences in both Cd and As concentrations in the brown rice. Similar to our previous study (data not published), the Cd concentrations in the brown rice of A16 (a low Cd uptake cultivar) were significantly lower than in A159 (a higher Cd uptake cultivar) regardless of water management regime. In contrast, As concentrations in A16 were higher than in A159, especially

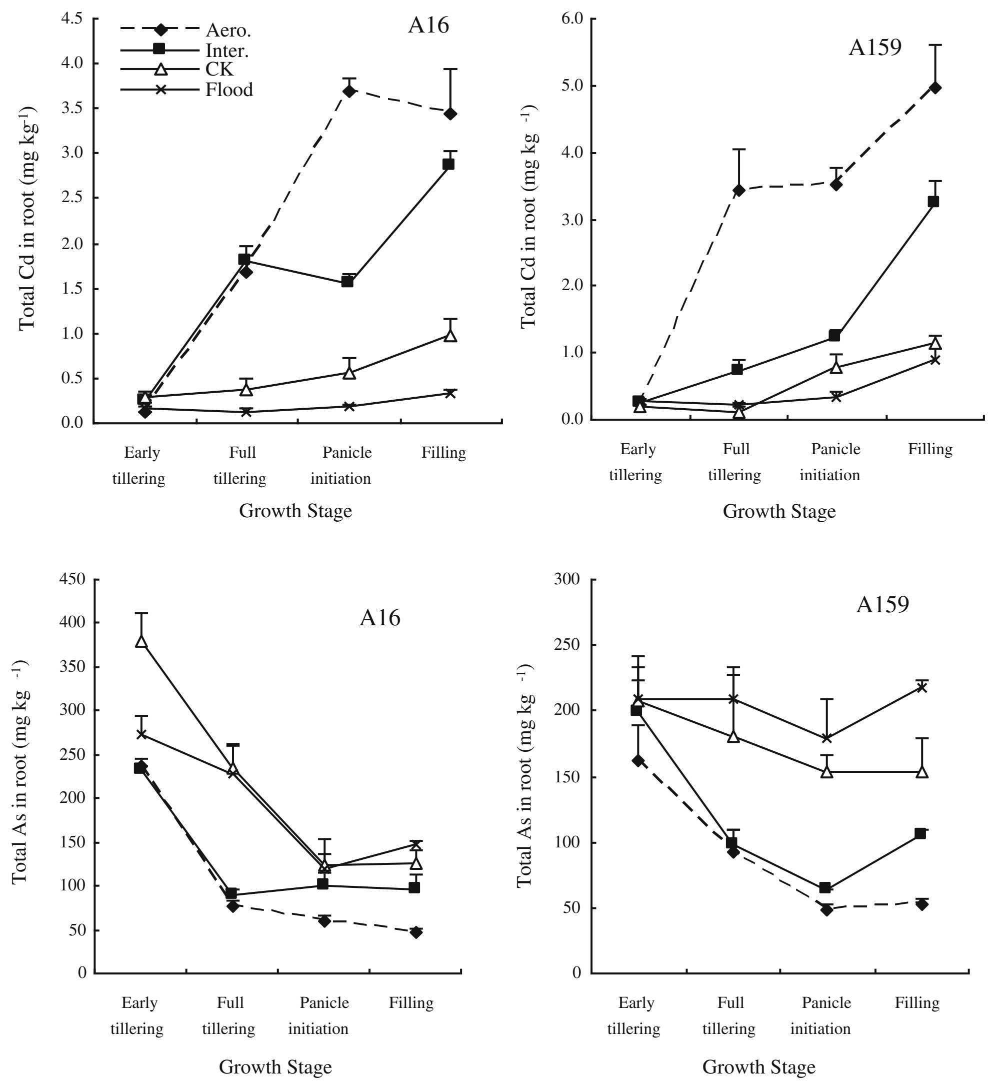


Fig. 4 Cd and As concentrations in the roots at different growth rice stages

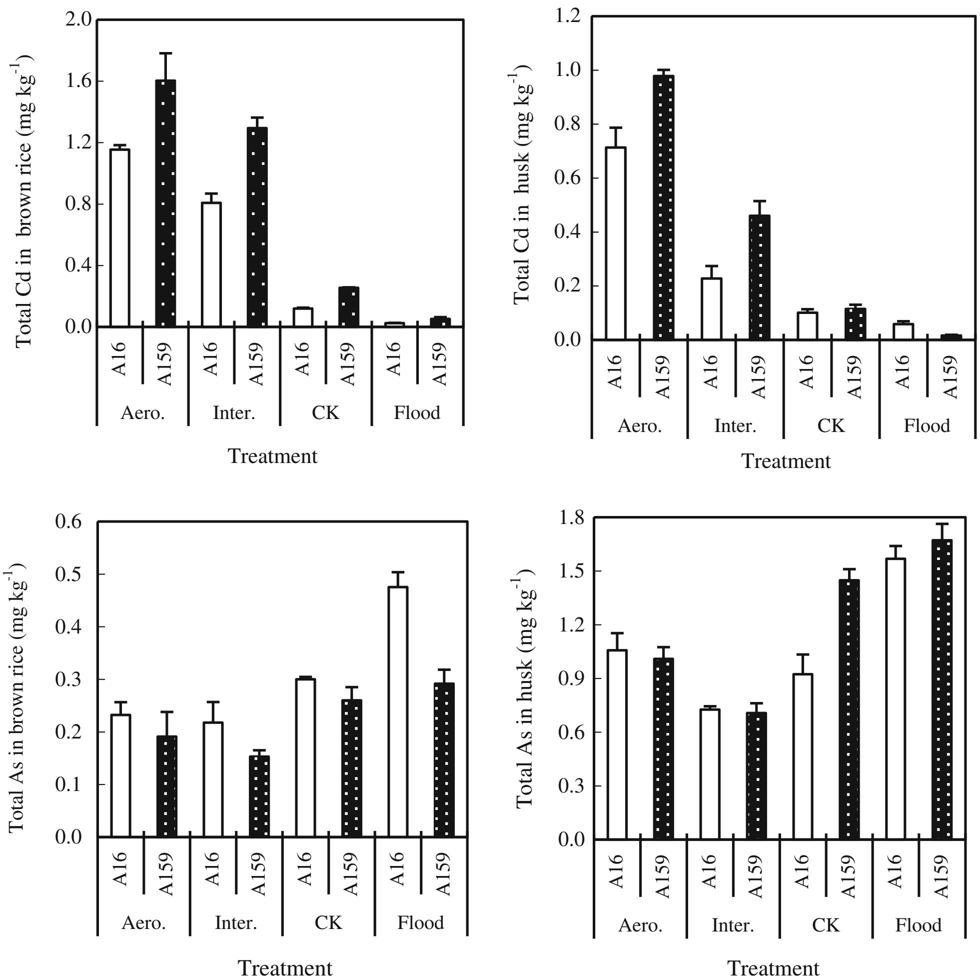


Fig. 5 Cd and As concentrations in the brown rice and husk of different rice cultivars

in the flooded treatment. The relationship between Cd and As concentrations in the brown rice was found to be significant using correlation analysis of the power function:

$$As = 0.2Cd^{-0.193}, \left(R^2 = 0.8218,\ n = 8,\ p < 0.05\right)$$

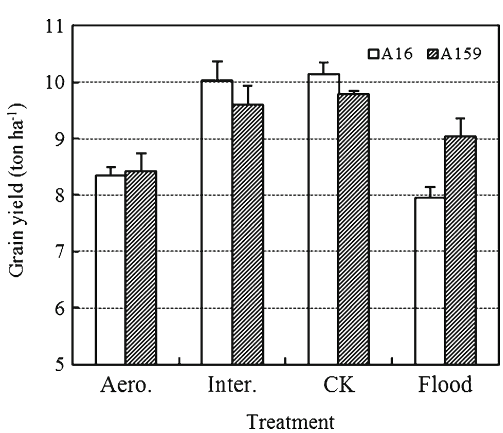


Fig. 6 Yields of the two rice varieties under the four different irrigation treatments

These results indicate that an antagonism might exist between As and Cd accumulation in rice grains (Sun et al. 2008). This requires further research, but our work does suggest that low Cd accumulating rice varieties should be avoided or used with great caution (analyse prior to use) in areas where the soils have high concentrations of bioavailable As.

5 Conclusions

The reproductive growth period plays a more important role than earlier growth stages in controlling Cd and As accumulation in rice straw and roots. Comparing the four water management regimes, conventional irrigation, i.e. flooding maintained until full tillering followed by intermittent irrigation, ensured high grain yield combined with relatively low Cd and As in the brown rice and was therefore the optimum field water management method. Cd and As uptake by rice showed opposite trends under the four water management regimes, with the low Cd cultivar showing high As concentrations in the brown rice. Further research is required to fully elucidate the mechanisms involved.

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