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Greenhouse gas emissions and grain arsenic and cadmium concentrations as affected by a weed control drainage in organic rice systems

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Abstract

Rice (*Oryza sativa* L.) is the primary staple food for more people than any other crop, but it is also a significant source of methane (CH_4) emissions and arsenic (As) in the human diet. For organic rice, these factors pose a greater challenge due to the use of organic fertilizers and that organic rice consumers often prefer brown rice. A major management challenge for organic rice producers is weed control. In California, organic producers have developed a practice to control weeds that involves a severe dry-down (DD) about 30 days after planting. Our objective was to determine the impact of a DD on greenhouse gas emissions and grain As and cadmium (Cd) concentrations compared to a continuously flooded (CF) control. The DD treatment reduced seasonal CH_4 emissions by 50% (from 359 to 181 kg CH_4 ha^{-1}). Only during the DD period were nitrous oxide (N_2O) emissions detected, but cumulative emissions were low (0.35 kg N_2O ha^{-1}). The global warming potential (GWP) was reduced by 49% in the DD treatment relative to the CF. Grain As and Cd concentrations were not at levels deemed a health concern in any treatment. However, grain As concentrations in the DD treatment were reduced by 30% and 37% in brown and white rice, respectively. Under the DD treatment, Cd concentrations increased but remained low. This study confirms that the weed management DD practice has a positive effect on GWP and grain As concentrations. In Cd contaminated soils, this practice may not be advisable.

1 | INTRODUCTION

Rice (*Oryza sativa* L.) is the primary staple food for more people on earth than any other crop, providing roughly one quarter of the global calorie intake (GRiSP, 2013). While rice cultivation is critically important for global food security, it is a significant source of greenhouse gas (GHG) emissions, particularly methane (CH_4) (Linquist et al., 2012). Rice systems account for 9%–11% of GHG emissions from agriculture (Smith et al., 2014). Rice can also be a significant means of human exposure to arsenic (As), especially in populations with high rice consumption (Bhowmick et al., 2018; Meharg, 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).

Abbreviations: CF, continuously flooded; DD, dry-down; GWP, global warming potential; GHG, greenhouse gas.

Both of these issues are related to rice being grown under flooded, anoxic conditions. Methane is the end product of the decomposition of organic matter under anaerobic conditions (Conrad, 2002). Anaerobic soil conditions also increase As bioavailability in the soil because it triggers the reduction of arsenate (AsV) to arsenite (AsIII), which is more mobile in the soil, and ferric oxide (FeIII) to ferrous oxide (FeII), dissolving iron hydro(oxides) that bind to As and, subsequently, releasing As to the soil solution (Meharg & Zhao, 2012).

Organic rice systems potentially have greater concerns in both of these areas. First, organic rice systems often rely on organic sources of fertilizer (cover crops, farm yard manure, etc.) to provide crop nutrition. However, these sources of fertility increase CH_4 emissions. Linquist et al. (2012) reported that CH_4 emissions increased by 26% using farm-yard manure and by 192% using green manures. Second, related to As, consumers of organic rice tend to be more health conscious and often prefer brown rice to white. Arsenic accumulates in the bran, causing brown rice to have higher As concentration than white rice (Sun et al., 2008). Interestingly, Tang et al. (2021) reported that the application of manure did not increase total As in rice grain but did increase the As content in the roots, stems, and leaves of the rice.

California is the biggest producer of organic rice in the United States in terms of area under production and amount produced (USDA, 2022). In California, both organic rice and conventional rice are typically grown using a water-seeded system. In this system, fields are tilled and fertilized to prepare a seedbed, the fields are then flooded and presoaked seed is broadcast into the flood water which is about 10-cm deep. The fields typically remain flooded until they are drained in preparation for harvest. Weeds present the biggest challenge for organic rice producers. Organic rice farmers have developed a system to help control weeds in this system, which is used by a number of organic producers. After planting and the emergence of the leaf through the surface of the water, the flood water height is slowly increased so that some rice leaf always remains above the water. This deep water helps to control grass weeds in the system (Driver et al., 2020; Galvin et al., 2022). After about a month, when the flood water can be up to 30-cm deep, the field is drained and remains so for about 1 month. The soils dry, and while the rice plant undergoes drought stress, the aquatic weeds are killed. Fields are then reflooded and remain so for duration of the season. Fertility management in these systems often involves the application of manures before planting and just before reflooding, following the weed control DD (Murray et al., 2020; Wild et al., 2011).

Broadly, introducing a drying period during the rice growing season has been shown to reduce both seasonal CH_4 emissions (Jiang et al., 2019) and grain As concentrations (Carrijo et al., 2022). The weed control dry-down described above is very similar to a mid-season drain, which has also been shown to reduce CH_4 emissions and grain As in conventional water-seeded systems in California (Carrijo et al., 2019; Perry et al., 2022). However, introducing drying periods can have the negative effect of increasing cadmium (Cd) concentrations in rice grain (da Silva et al., 2020). The effect of mid-season drainage, such as the weed control dry-down (DD), has not been investigated under organic rice production systems such as those used in California. Therefore, the objective of this research was to quantify the effect of a DD for weed control on GHG emissions and grain As and Cd concentrations in an organic production system.

Core Ideas

- A weed control dry-down was compared to continuously flooded rice in an organic rice system.
- The dry-down treatment reduced the GWP by 49% compared to continuous flood.
- Grain As levels in the dry-down treatment were reduced by 30% and 37% in brown and white rice, respectively.
- Grain Cd levels increased with the dry-down, but they were all below critical limits.

2 | METHODS

2.1 | Site characteristics

A field study was conducted in a registered organic rice field near Richvale, CA, during the 2018 growing season. Following the 2017 rice harvest, the straw was incorporated and winter flooded during the winter fallow period, as is common in California rice systems. This is a common practice to help facilitate the decomposition of rice straw (Linquist et al., 2006) and it also provides habitat for overwintering waterfowl. The soil was classified as an Esquon-Neerdobe which are clay soils (47% clay), typically having 2.1% organic matter and a pH of 4.7 (1:1 water; Web Soil Survey). The climate is Mediterranean, with cool wet winters and dry hot summers.

2.2 | Field management

The field was tilled in early spring to prepare the seedbed. During this period, about 5.5 Mt ha^{-1} of dry chicken manure (4.5% N) was applied and tilled in. After tillage, the field was leveled and rolled. The field was then flooded (May 14) and seed (high-yielding short grain variety, S-102) was aerially broadcast onto the field on May 15. During the first month after planting, the flood water was raised slowly to a final depth of about 30 cm, allowing the rice plant to have some leaves out of the water. On June 18, the field was drained for a 1-month period. At the end of this period, the field was reflooded and the field remained so until about 3 weeks before harvest when the field was drained to prepare for harvest.

2.3 | Experimental setup

The experiment was set up as a randomized complete block design with two treatments and four replications. The two treatments were continuously flooded (CF) and a weed control DD. The experiment was set up in a field where the weed control DD was being implemented; therefore, the CF treatment was established using 60-cm diameter PVC rings. These same PVC rings were used in the DD treatment to account for any possible "ring" effect. Rings were forced into the ground to a depth of 20 cm to create a water seal. The rings extended about 10 cm above the soil surface. Each PVC ring had holes at soil level, which could be plugged or opened using rubber stoppers. These holes allowed the DD rings to be drained when the main field was drained. These holes were left open at all other times of the season so that the water level in the rings was the same as in the field. When the main field was dried down, the DD rings were allowed to drain, while water was maintained in the CF ring. Water in the CF rings was maintained by adding irrigation water at least every other day to maintain a water height between 5- and 10-cm depth using a drip irrigation system and float valves inside each ring.

Yields between treatments could not be determined rigorously due to rat damage late in the season. Grain yields were taken within the 60-cm diameter PVC ring but variability was high. Yields across both treatments averaged about 8.9 Mt ha^{-1}.

2.4 | GHG sampling and analysis

A static closed vented flux chamber method was used for GHG measurements. The chamber system consisted of an open PVC base, extensions of variable lengths to accommodate growing plants and a sealed lid equipped with a vent tube, fan and thermocouple. The PVC base (29.5-cm in diameter) was permanently inserted to a depth of 15 cm into the soil before planting and was positioned in the middle of the larger 60-cm diameter rings which were to maintain the water treatments (described above). Two holes were drilled on the aboveground side of the base to allow for water movement between the inside and outside of the base. These holes were plugged with rubber stoppers during sampling when the water level was below the holes to ensure that the chambers were airtight. Four 11-cm diameter holes were drilled in the belowground portion of the chamber base to prevent restriction of water and root movement. Boardwalks were used to reach the sampling locations to ensure minimal soil disturbance which can artificially inflated gas fluxes.

The gas samples (20 mL) were taken at weekly intervals during the growing season. However, at the start and end of the DD treatment period, samples were taken every other day. Gas samples were taken at 20, 40, and 60 min between 9:00 and 13:00 h and were injected into pre-evacuated glass vials (12.5 mL). During each sampling event, four ambient samples were also taken at 0 min. In order to prevent leakage between sampling and analysis, each glass vial was sealed with the rubber septa and silicon.

Gas sampling followed the methodology presented by Adviento-Borbe et al. (2013). All samples were analyzed for nitrous oxide (N_2O) and CH_4 on a GC-2014 gas chromatograph equipped with an electron capture detector and a flame ionization detector (Shimadzu Scientific, Inst.). The detection limits of the GC instrument were 2.2 pg s^{-1} for CH_4 and 0.3 pg s^{-1} for N_2O. Standards were inserted after every 10 samples for quality assurance. Results from the GC were considered acceptable when the standard gas calibrations produced $r^2 > 0.99$. The peak area for each sample was converted to concentration using the calibration curve. Fluxes were estimated from the linear increase of gas concentration over time and converted to an elemental mass per unit area using the ideal gas law. For this calculation, we used the chamber volume measured at each sampling event, the chamber air temperature measured for each gas sample taken, and an atmospheric pressure of 0.101 MPa. Gas fluxes with a linear correlation below the predetermined threshold of $r^2 = 0.9$ were treated as missing data, and those that were below the GC detection limits were set to zero flux for data analysis. Individual flux values were integrated across all time points using linear interpolation to calculate cumulative growing season emissions.

2.5 | Grain arsenic and cadmium analysis

When plants were mature, all the grain from inside each 60-cm diameter treatment ring was sampled. Brown rice was obtained by removing the husk from paddy grain using a laboratory dehusker (Model FC2K-Y, Yamamoto Co. Ltd.). White rice (i.e., polished rice) was obtained by removing the husk and the bran from paddy grain using a laboratory mill (Paz-1/DTA; Zaccaria). Grains were ball milled to pass a 250-μm sieve and stored in the dark at 4°C prior to analysis, which was done separately for brown and white rice.

Total As and Cd in the rice grain was determined following the method of Sun et al. (2008) and modifications described by Carrijo et al. (2019). In brief, samples of 0.5 g (two

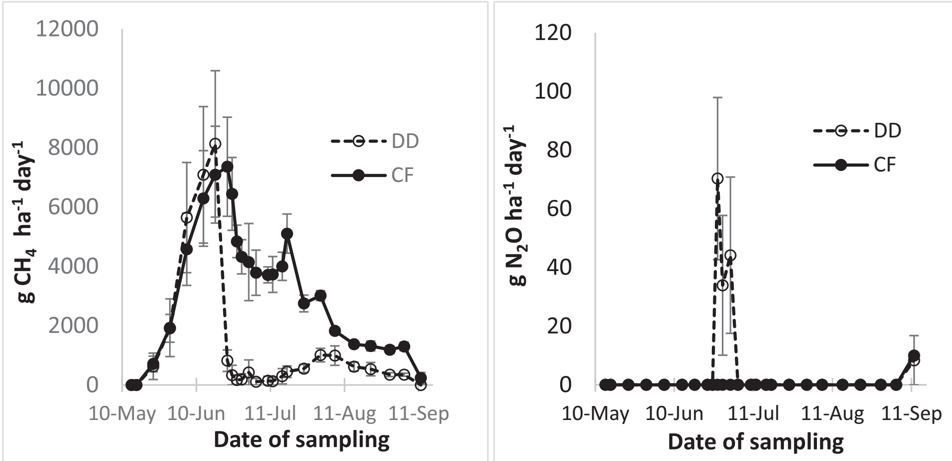

FIGURE 1 Daily methane (CH_4) and nitrous oxide (N_2O) fluxes from the continuously flooded (CF) and dry-down (DD) treatments. Error bars are the standard error.

analytical replicates per plot) were digested in glass digestion tubes by adding 5 mL of nitric acid and allowing it to dissolve overnight at room temperature. Samples were further digested in a heating block at 105°C until the cessation of a brown fog, and then at 120°C until complete dryness. The ash was redissolved with 10 mL of 0.28 mol L^{-1} nitric acid and filtered using a syringe filter (0.45 µm), taking care to discard the first 1 mL of the filtrate. The extract was then diluted fivefold with water. Total As and Cd in samples were quantified by inductively coupled plasma mass spectrometry (ICP-MS 7900; Agilent Technologies) with a detection limit of 0.01 µg L^{-1} for both As and Cd. As was monitored at *m/z* of 75 and selenium was also monitored (*m/z* 77, 78, and 82) to check for polyatomic $^{40}Ar^{35}Cl$ interferences on *m/z* 75. All water used for analysis was 18.2 MΩ•cm (Barnstead Nanopure). Trace metal grade nitric acid (67%–70%), ammonium phosphate dibasic (≥99%), and ammonium hydroxide (28%–30%) were from Fisher Chemical. For quality assurance, certified reference material rice flour (CRM 1568b) from the National Institute of Standards and Technology was used as a check.

2.6 | Data analysis

The global warming potential (GWP) was calculated using the radiating forcing potential relative to CO_2, which was 265 for N_2O and 28 for CH_4 (IPCC, 2014). An analysis of variance on the data (response variables: cumulative CH_4 and N_2O emissions, GWP, and grain As and Cd concentration) was conducted in Statisix (Version 10, Analytical Software). Blocks (replications) and treatment × block interactions were considered as random effects. Mean separation was conducted using Tukey's mean separation test ($p \leq 0.05$).

3 | RESULTS AND DISCUSSION

3.1 | Greenhouse gas emissions

In the CF control, CH_4 emissions were detected within 1 week of flooding (about 700 g CH_4 ha^{-1} day^{-1}) and by June 18 had risen to between 7000 and 8000 g CH_4 ha^{-1} day^{-1} (Figure 1). In late June, CH_4 emissions declined to 4000 g CH_4 ha^{-1} day^{-1} during the mid-season, followed by a short spike in mid-July and then a steady decline to the end of the season. This overall pattern of CH_4 fluxes is fairly typical of conventional California water-seeded rice systems (Adviento-Borbe et al., 2016; Balaine et al., 2019; LaHue et al., 2016; Perry et al., 2022; Pittelkow et al., 2013, 2014; Simmonds et al., 2015). However, compared to what has been reported from conventional rice systems, CH_4 emissions were detected earlier in this study. In conventional systems, CH_4 emissions are usually detected 2–4 weeks after flooding instead of 1 week after flooding as seen here. Furthermore, peak seasonal fluxes in the range of 7000–8000 g CH_4 ha^{-1} day^{-1} are on the high end of what has been reported from conventionally managed California rice systems, which have ranged from about 500 (Adviento-Borbe et al., 2016) to as high as 10,700 g CH_4 ha^{-1} day^{-1} (Balaine et al., 2019). Cumulative seasonal CH_4 emissions from the CF treatment were 359 kg CH_4 ha^{-1} (Table 1), and they are also on the high end of what has been reported for California conventional rice systems. Linquist et al. (2018), based on an analysis of GHG emissions from rice systems, reported average California seasonal emissions of 218 kg CH_4 ha^{-1} (95% CI of 153 to 284 kg CH_4 ha^{-1}) and a maximum reported value of 446 kg CH_4 ha^{-1}. The high early season fluxes, peak fluxes, and cumulative CH_4 emissions observed in this study are likely due to the 5.5 Mt ha^{-1} of dry chicken manure applied (Linquist et al., 2012) in addition to the rice

TABLE 1 Seasonal methane (CH_4) and nitrous oxide (N_2O) emissions and global warming potential (GWP) for the continuously flooded (CF) and dry-down (DD) treatments. Values in brackets represent the standard error.

Treatment	CH_4 (kg ha^{-1} season^{-1})	N_2O[a] (kg ha^{-1} season^{-1})	GWP (kg CO_2-eq ha^{-1} season^{-1})
CF	359a	0b	10,052 (1361)a
DD	181b	0.352a	5161 (1638)b

Note. Different letters within the same column indicate difference ($p < 0.05$) between treatments.
[a]Does not include end of season flux.

straw being incorporated from the previous season (Fitzgerald et al., 2000; Linquist et al., 2018).

Methane is the end product of organic matter decomposition under anaerobic soil conditions; thus, by introducing a drying period, soil oxygen availability is altered, thereby affecting various processes underlying CH_4 production (Conrad, 2007). In the DD treatment, CH_4 emissions were similar to the CF treatment until fields were drained for the weed treatment on June 18 with CH_4 fluxes peaking at 8000 g CH_4 ha^{-1} day^{-1} (Figure 1). Following the drain, CH_4 fluxes dropped to near negligible levels during the drain period and then following reflood, CH_4 fluxes slowly increased, but not to the levels of the CF treatment. This seasonal pattern of emission is similar to what is seen for an extended mid-season drain in conventional systems (Perry et al., 2022). When drain periods are long and the soil dries out to a greater degree, CH_4 fluxes tend to remain low for the rest of the season; however, when the drain period is short, CH_4 emissions tend to increase rapidly after reflooding (Balaine et al., 2019; Perry et al., 2022). Shorter drain periods are common for practices such as "Safe-AWD" where the perched water table is only allowed to recede to 15 cm below the soil surface (Lampayan et al., 2015). Total seasonal emissions in the DD treatment were 181 kg CH_4 ha^{-1} (Table 1), which represents a 50% reduction compared to the CF. This level of reduction is similar to what is seen in other water management practices such as alternate wetting and drying (AWD), intermittent flooding or mid-season drains (Jiang et al., 2019; Linquist et al., 2018; Perry et al., 2022).

In the CF treatment, during the growing season, N_2O emissions (10 g N_2O ha^{-1} day^{-1}) were only detected at the end of the season when the fields were drained in preparation for harvest (Figure 1). It was not possible to determine quantitatively the cumulative amount of N_2O emissions because sampling ceased after this event. However, in other studies, it is common to see a brief spike in N_2O emissions shortly after fields are drained (Adviento-Borbe et al., 2015). Flooded rice systems generally emit less N_2O than dryland crops because flooding results in most N being lost as N_2 rather than N_2O due to the highly reduced conditions (Firestone & Davidson, 1989). In California, N_2O emissions have averaged 0.15 kg N_2O ha^{-1} season^{-1} in CF water-seeded conventional systems (Linquist et al., 2018).

The introduction of aerobic periods during the flood period increases the likelihood of N_2O emissions because N_2O emissions can occur during both nitrification and denitrification (Klemedtsson et al., 1988). However, in this study, N_2O emissions were only observed after the field was drained as the soil went from anaerobic to aerobic conditions as the soil began to dry out (fluxes up to 70 g N_2O ha^{-1} day^{-1}) (Figure 1). They were not observed when the soil was reflooded. This observation is similar to what others have found (Adviento-Borbe et al., 2013; Towprayon et al., 2005; Yao et al., 2012). The range in soil moisture when N_2O emissions are most likely is from 60% to 88% water filled pore space (Adviento-Borbe et al., 2015; Hou et al., 2012; Linn & Doran, 1984). Thus, it would be expected for N_2O fluxes to diminish during long DD events when the soil can get very dry. Also, N_2O fluxes are generally low when soil mineral N is low during drain periods (LaHue et al., 2016; Perry et al., 2022). In this study, cumulative N_2O emissions were relatively low and averaged 0.35 kg N_2O ha^{-1} (Table 1). While we did not quantify mineral soil N, it is likely that the soil mineral N pool was low due to the low mineralization rate of poultry manure in these flooded systems (Wild et al., 2011) and continued plant uptake of mineral N during the drying period. Given that N_2O emissions made up only 6% of total GWP in the DD treatment, the change in GWP (49%) between these systems was similar to the change in the amount of CH_4 reduction (50%).

3.2 | Heavy metals

The grain total As concentrations in brown and white rice for the CF treatment were 125 and 86 µg kg^{-1}, respectively (Figure 2). Higher concentrations of As in brown rice are commonly reported, as the rice bran contains more As than the endosperm (Sun et al., 2008). The Food and Drug Administration (FDA) set a limit on inorganic As for infant rice cereal at 100 µg kg^{-1} (FDA, 2016). While inorganic As was not determined in this study, the proportion of inorganic to total As in US white rice is about 42% (Zhao et al., 2013). Thus, it is likely that both the brown and white rice samples in the CF treatment of this study were below the FDA critical limit for infant cereals. Under the DD treatment, grain As concentrations were reduced to 87 (30% reduction) and 54 µg kg^{-1} (37% reduction) for brown and white rice, respectively. This level of

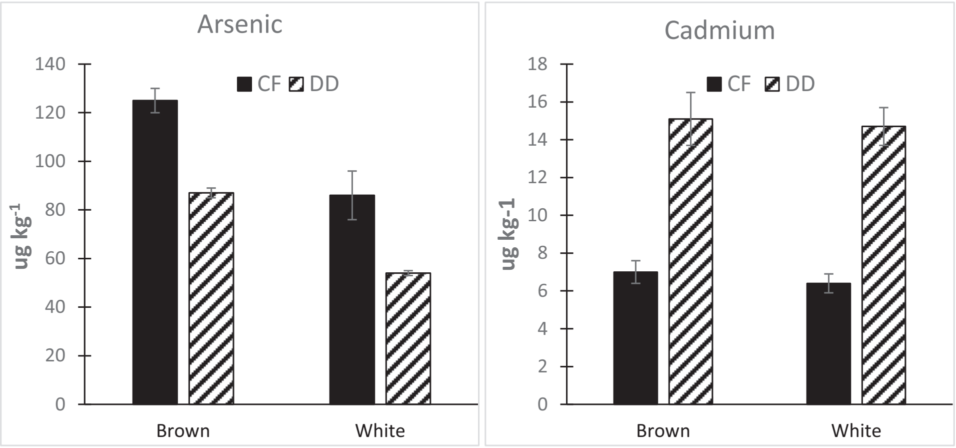

FIGURE 2 Total arsenic and cadmium in brown and white (polished) rice in continuously flooded (CF) and dry-down (DD) treatments. Error bars are the standard error. In all cases, the differences between the CF and DD treatments were statistically significant ($p \leq 0.05$).

reduction is similar to what has been reported by Carrijo et al. (2022) based on a global analysis of the effects of AWD. During the drying events, soils become aerobic and AsV is the predominate As species. Arsenate is less strongly absorbed to soil minerals, limiting its movement in the soil solution and plant uptake relative to AsIII, which prevails under anaerobic conditions (Li et al., 2019; Meharg, 2004).

One concern with introducing aerobic events to reduce CH_4 or grain As concentrations is that grain Cd can increase. This occurs for a number of reasons, including (1) the oxidation of sulfur in Cd sulfide precipitates and the consequent release of Cd^{2+} into soil solution; (2) weaker adsorption of Cd^{2+} to manganese and iron hydrous oxides at lower pH values that are typical of aerobic soils, compared to anaerobic soils; and (3) the oxidation (and subsequent removal from soil solution) of Mn^{2+}, which competes with Cd^{2+} and inhibits its uptake by rice roots (Li et al., 2021; Simmons et al., 2008). In this study, the grain Cd concentrations in brown and white rice were similar (Figure 2). For the CF treatment, Cd concentrations were 7 and 6 μg kg^{-1}, respectively. Grain Cd concentrations increased to 15 μg kg^{-1} for both brown and white rice, respectively, under the DD treatment. This was an average increase in grain Cd concentration of 122% and in agreement with other studies (Carrijo et al., 2022). Importantly, the maximum Cd levels in this study were 15 μg kg^{-1} which is well below the CODEX standard of 400 μg kg^{-1} (Codex Alimentarius Commission, 2006).

4 | CONCLUSION

Seasonal CH_4 emissions and daily fluxes in the CF control treatment were high relative to reported values for conventional rice fields in California. However, the weed control DD practiced by organic farmers resulted in a 49% reduction in GWP (mostly due to a reduction in CH_4 emissions) and over a 30% reduction in grain As concentrations. While Cd grain concentrations increased in the DD treatment, the concentration of Cd in grain was much below levels that are of concern for human health. This is the first research that has been conducted examining this practice for its effect on GHG emissions and heavy metal uptake; however, our findings with respect to introducing aerobic cycles into the system are similar to other research in conventional rice systems. Only 1 year of data are presented here. It is important to confirm these results on other soils and with other practices that may be used by organic growers such as the use of cover crops.

AUTHOR CONTRIBUTIONS

Bruce Linquist: Conceptualization; funding acquisition; methodology; project administration; supervision; visualization; writing—original draft. **Henry Perry**: Data curation; formal analysis; investigation; validation.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

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