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The effect of rice residue management on rice paddy Si, Fe, As, and methane biogeochemistry

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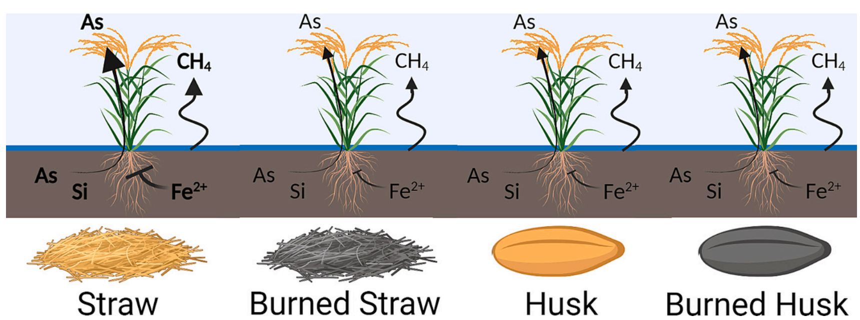
HIGHLIGHTS

- Rice straw and husk are residues that can affect biogeochemistry.
- A fully factorial design incorporated burned and unburned residues into paddy soil.
- Paddy porewater, methane emissions, and plant chemistry were measured in 2 varieties.
- Unburned straw increased plant arsenic and methane emissions.
- Husk incorporation minimally affected paddy biogeochemistry of Si, Fe, and As.

GRAPHICAL ABSTRACT



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ABSTRACT

Rice production results in residues of straw and husk, and the management of these residues has implications for the sustainability of the rice agroecosystem. Rice straw is typically incorporated into soil either as fresh residue or is burned prior to incorporation. Rice husk is not typically returned to rice fields. However, rice husk contains high levels of silicon, which has been shown to decrease rice accumulation of arsenic. In this work, we studied the resulting biogeochemical changes in rice paddy soils when paddies were amended with either straw or burned straw and either no husk, husk, or burned husk over two years. Using a full-factorial design, we observed that the higher lability of rice straw carbon controlled redox-sensitive processes despite the application of husk and straw at similar carbon rates. Amending paddies with straw, rather than burned straw, increased porewater Fe and As, plant As, and methane emissions regardless of husk amendment. Husk addition provided insignificant Si to the plant despite its high concentration of Si, suggesting limited short-term mobility of Si and that long-term additions of husk or higher rates may need to be studied.

1. Introduction

Rice is a staple crop consumed by half of the world's population; thus, its sustainable production is critical to the health of billions of people. Unique among crop plants, rice accumulates silicon (Si) in the straw and husk at concentrations exceeding those of other

Abbreviations: AWD, alternate wetting and drying; DCB, dithionite-citrate-bicarbonate; DMA, dimethylarsinic acid; DOC, dissolved organic carbon; EXAFS, extended x-ray absorption fine structure; MMA, monomethylarsonic acid; XANES, x-ray absorption near edge spectroscopy.

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macronutrients (Epstein, 2009; Savant et al., 1996). Silicon confers resistance to several biotic and abiotic stressors, resulting in a more robust rice crop (Ma, 2004), and while not considered essential for all crops, Si has been suggested to be a yield-limiting nutrient for rice (Savant et al., 1997). Increasing silicon can also decrease grain arsenic (As) through a shared root uptake pathway and suppression of Si/As transporters (Li et al., 2009; Limmer et al., 2018b; Ma et al., 2008; Seyfferth et al., 2018). Due to the flooded conditions under which rice is grown, grain typically contains ~0.1 mg/kg inorganic As, a non-threshold carcinogen that poses health risks to rice consumers (Zavala and Duxbury, 2008; Zhao et al., 2013). The environmental sustainability of rice production is also constrained by the emission of methane (CH_4) from flooded rice paddies. Globally, rice paddies comprise ~11 % of anthropogenic emissions of CH_4 (IPCC, 2013), a potent greenhouse gas. Allowing paddy soil to drain can decrease As availability and CH_4 emissions, but can mobilize cadmium (Cd), a toxic element readily accumulated in the grain (Arao et al., 2009; Honma et al., 2016). In hydroponic studies, Si has been shown to decrease rice uptake of Cd through retention of Cd in root cell walls because Si can provide a net negative charge in the cell walls (Liu et al., 2013; Ma et al., 2015). This suggests that Si may be able to decrease both Cd and As in rice grain. To improve the sustainability of rice production, globally applicable management practices are needed that provide Si, decrease grain As and Cd, and limit CH_4 emissions.

Rice residues are potential sustainable sources of Si, but how the residues are managed can affect a variety of biogeochemical cycles (Runkle et al., 2021). Rice straw is generally considered to be poor animal feed due to its high Si content (Mandal et al., 2004). Thus, straw is typically either burned, directly incorporated through tillage, or left on the field surface. Under flooded conditions during the following rice crop, the labile carbon from straw results in more strongly reducing conditions, leading to increased CH_4 emissions and As mobilization (Gutekunst et al., 2017; Yan et al., 2005). Burning straw lessens the impact of straw labile carbon on subsurface biogeochemistry (Mandal et al., 2004; Naresh, 2013), but generates approximately 13 Mg CO_2/ha, noticeable smoke plumes, CO, NO_x, SO_2, and particulate matter (Gadde et al., 2009), which can lead to adverse respiratory effects in nearby populations (Golshan et al., 2002). Burning straw also results in the loss of several nutrients from the straw, including N (up to 80 %), P (25 %), K (21 %), and S (4–60 %) (Lefroy et al., 1994; Ponnamperuma, 1984; Raison, 1979). Unburned straw contains appreciable quantities of N (6 g/kg), P (1 g/kg), K (15 g/kg), S (1 g/kg), Si (25–50 g/kg), and As (1–10 mg/kg) (Penido et al., 2016; Ponnamperuma, 1984; Savant et al., 1996), making straw a potentially valuable residue to return to the field but also a source of As.

Rice husk, the inedible outer covering of the rice grain and the other primary rice residue, is typically produced at the rice mill and not returned to the field. However, rice husk contains more Si (~50–70 g/kg), less As (~0.4 mg/kg), and less labile C than rice straw (Gutekunst et al., 2017; Penido et al., 2016; Runkle et al., 2021; Savant et al., 1996), making it an attractive soil amendment. Like rice straw, rice husk can be burned or pyrolyzed prior to field application, although burning at high temperatures can lessen the availability of the Si due to crystallization or decreased surface area (Linam et al., 2021; Teasley et al., 2017; Xiao et al., 2014). Rice husk, when applied to rice paddy soil either unburned or burned at low temperature, both increased Si concentrations in rice straw and husk 2 years after application, although Si was applied at a high loading rate (5 Mg Si/ha) (Limmer et al., 2018a; Limmer and Seyfferth, 2021). How simultaneous application of straw and husk at levels on par with crop production affect rice paddy biogeochemistry remains understudied.

The primary objective of this research was to determine how management of rice residues would affect biogeochemical cycling of C and rice accumulation of Si, As, and Cd. We hypothesized that burning both straw and husk would decrease the amendment's effect on redox-sensitive cycles, such as Fe(II), As, Cd, and CH_4 emissions. We also hypothesized that the addition of husk would have a lesser effect on the biogeochemistry than whether the straw was burned or unburned. We further hypothesized that returning both straw and husk would increase plant levels of Si, but that the lability of straw C would increase the mobility of As. Thus, we expected burned forms of amendments, which provide high levels of available Si, to decrease rice grain inorganic As.

2. Methods

2.1. Rice paddies

Rice (*Oryza sativa* L.) was grown in 18 rice paddy mesocosms (Limmer et al., 2018a; Limmer and Seyfferth, 2021). These paddies were 2 × 2 m (planted area 1.5 × 1.5 m) and lined with an impermeable liner 0.5 m below ground surface. Each paddy was equipped with irrigation and a float switch and pump to manage water levels. Prior to this experiment, twelve of the paddies had been used to grow rice under continuously flooded conditions, while six of the paddies had been used to grow rice under non flooded conditions. Thus, to prepare the mesocosms for the current experiment, the top 10–15 cm of soil from each paddy was removed by hand, collectively mixed with other paddy topsoil, and randomly returned to each paddy. The soil in the rice paddies was an Ultisol with 1.5 % organic matter and a pH of 6.3. The soil contained 5.4 mg/kg As and 0.072 mg/kg Cd measured via US EPA method 3051A.

2.2. Soil amendments

The 2-year study was designed as a 2-factor randomized block design, with the paddies blocked according to previous water management. The two factors were the form of straw amendment and the form of husk. In the United States, rice straw is rarely removed from field after harvest, so rice straw from previous growing seasons was added to the paddies at two levels: either as unburned straw ('Straw') or as burned straw ('Burned Straw'). The Burned Straw was burned under ambient conditions outdoors to mimic field burning conditions and reached temperatures of 250–300 °C. The yield of the Burned Straw was 13 % of the Straw and the material contained a mixture of ash and char. The Straw contained 37 g/kg of Si and 2.0 mg/kg of As and was amended at a rate of 680 g/paddy/yr (2900 kg/ha/yr), representing returning nearly 1 year of straw into the soil (Table 1). Burned Straw was amended at 13 % of the Straw rate so that both forms of straw were applied at a Si rate of 0.11 Mg/ha/yr. The second experimental factor, rice husk, had three levels: an unamended control ('Control'), unburned husk ('Husk'), and burned husk ('Burned Husk'). Because rice husk is not commonly returned to the field, this factor included an unamended control. Rice husk was obtained from Riceland Mills (Stuttgart, AR, USA) and contained 71 g/kg Si and 0.28 mg/kg As. Husk was amended at rates approximately equal to one harvest of rice from prior studies in the mesocosms (7500 kg/ha rough rice). We assumed husk constituted 20 % of rough rice by mass, resulting in a husk application rate of 500 g/paddy/yr (2100 kg/ha/yr). Burned Husk was processed similar to straw, with open burning at 250–300 °C. The yield of Burned Husk, and thus the application rate, was 32 % of Husk. The Si loading rate was 0.15 Mg/ha/yr for both the Husk and Burned Husk, representing approximately 1 year of rice husk returned to the soil. All amendments were added in both years of the study 18 days prior to transplanting and were tilled to a depth of ~15 cm.

2.3. Rice growth and monitoring

Rice seedlings were transplanted into the paddies at the 3–4 leaf stage. In the first year the pure line variety 'Jefferson' was grown while in the second year the hybrid variety 'XL745' was grown. Seedlings were hand transplanted at a rate of 49 plants/paddy into flooded paddies. Rice paddy water was managed using safe AWD (Bouman et al., 2007;

Table 1
Elemental composition and application rate of soil amendments.

Elemental	Straw	Burned Straw	Husk	Burned Husk	Straw	Burned Straw	Husk	Burned Husk
	Concentration				Application Rate			
C (g/kg) or (Mg/ha/y)	397	106	400	381	1.15	0.04	0.84	0.26
N (g/kg) or (kg/ha/y)	8.8	3.0	3.4	5.9	25.6	1.1	7.1	4.0
P (g/kg) or (kg/ha/y)	1.7	8.8	0.71	1.9	4.9	3.3	1.5	1.2
K (g/kg) or (kg/ha/y)	26	146	2.5	5.5	75	55	5.3	3.7
Si (g/kg) or (Mg/ha/y)	39	190	68	183	0.11	0.07	0.14	0.12
As (mg/kg) or (g/ha/yr)	1.9	6.3	0.35	0.66	5.5	2.4	0.73	0.44
Cd (mg/kg) or (g/ha/yr)	0.2	0.5	<0.05	<0.05	0.7	0.2	<0.1	<0.03

Carrijo et al., 2017), where water was occasionally allowed to drain to 15 cm below the ground surface prior to reflooding. For the pure line variety two dry-downs occurred, while for the hybrid variety three dry-downs occurred. The timing of the dry-downs was weather-dependent (Table A1).

Rice paddy porewater chemistry and CH_4 emissions were monitored weekly throughout the growing season. Porewater was monitored using ceramic rhizon samplers (1910, Soilmoisture Equipment Corp. Goleta, CA, USA). Porewater was collected using a locking syringe and immediately analyzed for Fe(II) by the ferrozine method (Stookey, 1970), Si by the molybdate blue method (Derry et al., 2005), and pH and redox potential using calibrated electrodes. Additional porewater was collected into an anoxically-sealed serum vial evacuated in the field. This porewater was acidified to 2 % HNO_3 to prevent oxidation of Fe(II) prior to ICP-MS analysis (Agilent 7500cx), dissolved organic carbon (DOC) analysis (vario TOC cube), and As speciation by IC-ICP-MS (Thermo iCAP). Arsenic species were separated using a PRP-X100 column and the ammonium carbonate gradient elution described by Jackson (2015). For ICP and DOC measurements, acidified blank samples were run every 20–40 samples. Soil cores (5 cm diameter, 15 cm deep) were taken at several time points throughout the experiment (Table A1). Soil was air dried, sieved to 2 mm while removing any obvious plant roots or husk, and organic matter was measured by loss on ignition. Methane emissions were measured using the closed chamber technique with a chamber able to enclose the entire paddy (1.5 × 1.5 × 1.5 m) for five minutes between the hours of 7:30 and 12:00. Methane concentrations in the chamber were monitored using a portable FTIR gas analyzer (Gasmet DX4040, Fig. A1). Two 12 V fans were used to ensure the chamber air remained well-mixed. Fluxes of methane were calculated after manually selecting the linear portion of the methane concentration curve over time using MATLAB and the ideal gas law.

$$J = \frac{dC}{dt}\frac{V}{A}\frac{P}{RT}$$

where J is gas flux [μmol/m^2/s], dC/dt is the linear slope of the methane concentration curve [ppmv/s], V is the chamber volume [m^3], A is the soil surface area [m^2], P is atmospheric pressure (101.3 kPa), R is the ideal gas constant, and T is the paddy temperature (°C). A positive flux was defined as an emission from the paddy to the atmosphere. Methane fluxes were considered to be undetectable when $R^2 < 0.8$ and are reported as a flux of zero. Any measurements where ebullition events were observed were discarded. Cumulative emissions during the growing season were calculated using linear interpolation.

2.4. Plant and Fe plaque analysis

Rice was harvested at maturity, separated into various parts, and analyzed for elemental composition. Harvesting the pure line variety occurred 107 days after transplanting while harvesting the hybrid variety occurred 114 days after transplanting. Rice plants were separated into rough rice, flag leaves, straw, the upper-most node (node I), and the next lower node (node II). Biomass and yield measurements were taken using all the plants in the paddy. Dried rough rice was dehusked and polished to give bran and polished rice. Nodes and flag leaves were separated from the straw because they control loading of many elements to the filling grain (Chen et al., 2015; Yamaji and Ma, 2014, 2009). Plant parts were oven dried, finely ground, and digested in trace metal grade HNO_3 and microwave digested (Seyfferth et al., 2016). Acid-soluble elements were analyzed by ICP-MS (Agilent 7500 and Thermo iCAP), while Si was analyzed colorimetrically using the molybdate blue method after dissolving in 2 M NaOH (Kraska and Breitenbeck, 2010). Blanks and certified reference materials (NIST1568b rice flour, NIST 1570a spinach leaves, and WEPAL IPE 188 oil palm) were included in each digestion run (Table A2). Rice grain and bran were also analyzed for As species after extraction using 2 % HNO_3 (Maher et al., 2013), filtration, and dilution to 1 % HNO_3. Chromatographic conditions were the same as described for porewater analysis. Blanks and NIST1568b were used in each digestion for quality assurance (Table A2).

Rice root Fe plaque was further characterized to analyze the elemental composition, Fe mineral composition, and As speciation. At harvest, entire root systems from three plants collected diagonally across the paddies, avoiding the edges, were removed from the soil and roots were gently separated from the soil and washed twice to remove soil. Roots were composited for each paddy and allowed to air dry. Iron plaque from half of the roots was removed using a dithionite-citrate-bicarbonate extraction (Taylor and Crowder, 1983), which was analyzed using ICP-MS after 100× dilution for Fe, Mn, Si, As, K, Mg, Al, Cr, P, Zn, and Cd. Both Zn and Cd were below the limit of detection. The other half of the root system was sonicated to remove intact Fe plaque that was captured on a nitrocellulose filter in preparation for x-ray absorption spectroscopy (XAS) (Amaral et al., 2017). Samples from the pure line variety were analyzed at the Stanford Synchrotron Radiation Lightsource (SSRL) on beamline 11–2. Samples from the hybrid variety were analyzed at the National Synchrotron Light Source (NSLS-II) on beamline 6-BM (BMM). At the beamlines, As x-ray absorption near-edge spectroscopy (XANES) spectra were obtained in fluorescence while Fe extended x-ray absorption fine structure (EXAFS) spectra were obtained in transmission. For measurement of As fluorescence, beamline 11–2 was equipped with a 100-element Ge detector while beamline 6-BM was equipped with a 4-element vortex detector. EXAFS and XANES spectra from both beamlines were background subtracted and normalized using Athena v 0.9.26. Athena was also used for linear combination fitting (LCF) of normalized sample spectra to normalized standards. For As XANES spectra, data were fit from −20 to 30 eV from the As edge and standards included sodium arsenite and sodium arsenate. For Fe EXAFS, LCF fitting of the k^3 spectra occurred from k = 2–12 and standard spectra included 2-line ferrihydrite, lepidocrocite, goethite, and siderite, which have been shown to comprise Fe plaque (Amaral et al., 2017; Seyfferth et al., 2019).

2.5. Statistical analysis

Data were analyzed using PROC Mixed in SAS 9.4. Average porewater concentrations were calculated during the reproductive period (>40 days after transplant) until the paddies were drained for harvest. The model included the following fixed effects: block, Husk, Straw, and

Husk*Straw interaction. The interaction term was never significant and was removed from the final models. Data from each variety were analyzed separately because differences between varieties were confounded by differences between years. Soil organic matter was modeled similarly but with the addition of a covariate: the soil organic matter present prior to amending to control for differences in background levels of soil organic matter. When significant amendment effects were present for Husk, comparisons between amendment levels were made with Tukey's adjustment.

3. Results

3.1. Yield and biomass

Rice yield was consistent across amendments for each year (Table A3). For the pure line variety, rough rice yield averaged 8400 ± 800 kg/ha (12 % moisture content, wet basis) while straw biomass averaged 5300 ± 1000 kg/ha (dry weight). Neither was significantly affected by the form of straw amendment ($p > 0.61$) or the form of husk amendment ($p > 0.30$). For the hybrid variety, rough rice yield averaged 12,000 ± 1100 kg/ha (12 % moisture content, wet basis) while straw biomass averaged 9900 ± 900 kg/ha (dry weight). Neither was significantly affected by the form of straw amendment ($p > 0.77$) or the form of husk amendment ($p > 0.51$).

3.2. Porewater chemistry

In both varieties, porewater chemistry was more strongly affected by the form of straw amendment than the form of husk amendment (Figs. 1 and A2 and Table A4). Porewater Si was not significantly affected by either amendment in the pure line variety (Straw $p = 0.06$, Husk = 0.43) and averaged 128 ± 49 μM, with porewater Si decreasing during the growing season. In the hybrid variety average porewater Si was lower (76 ± 31 μM) and was not significantly affected by either amendment ($p \geq 0.15$). In both varieties, burning the straw decreased average porewater Si.

The form of straw addition strongly affected the redox chemistry. Burning the straw decreased porewater Fe(II), As, and Mn, and also increased porewater S and redox potential. Porewater Fe(II) was significantly affected by the form of straw amendment in both varieties ($p \leq 0.012$, Fig. 1), with Straw averaging 33 % more Fe(II) than Burned Straw in the pure line variety and 45 % more Fe(II) in the hybrid variety.

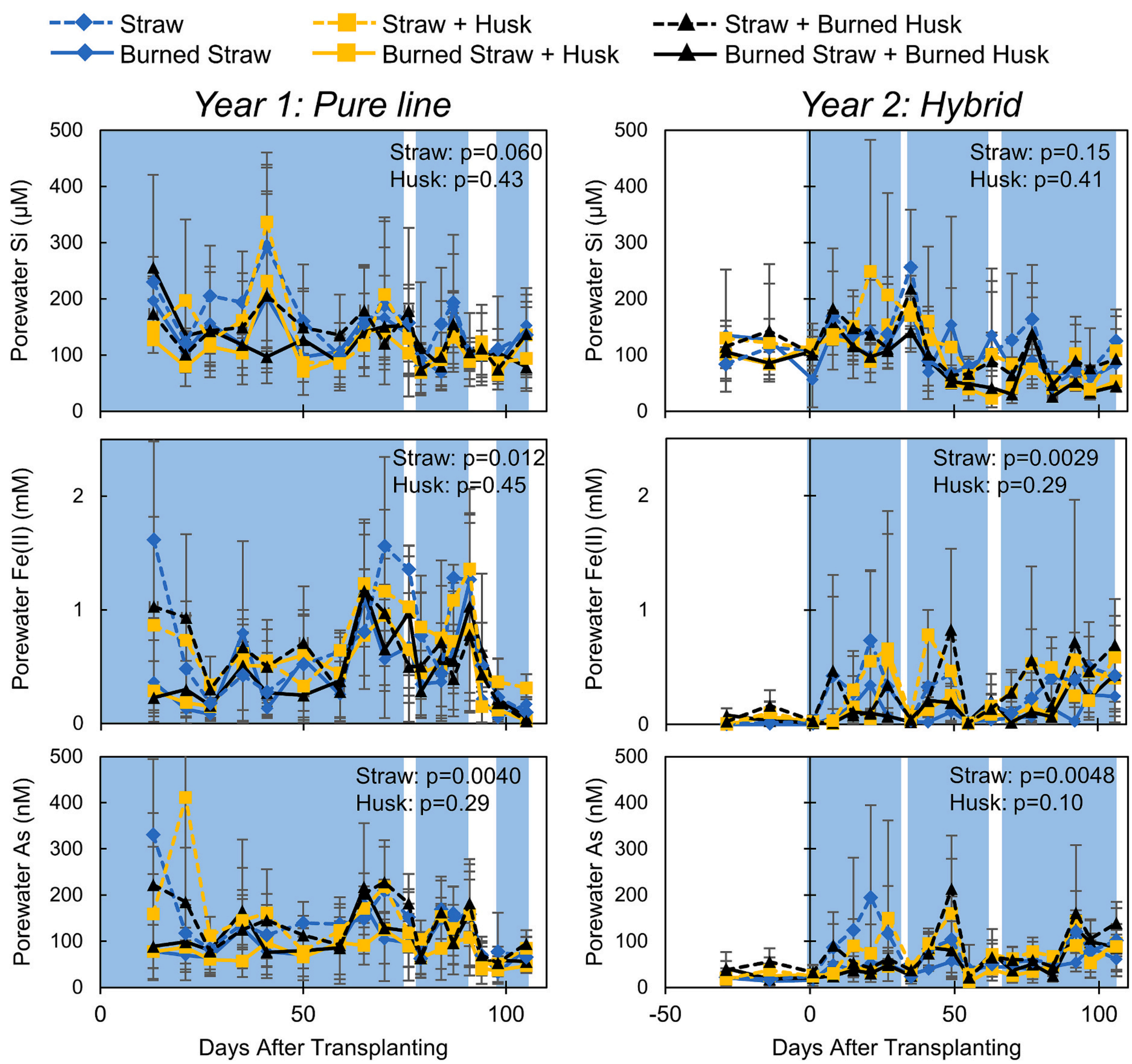

Fig. 1. Porewater Si, Fe(II), and As during the two year experiment. Straw addition tended to increase average porewater Si, Fe(II), and As in both varieties compared to Burned Straw. For each panel, p values are calculated for each main effect from the average value during reproduction. White shaded areas indicate when paddies were not flooded. Error bars are ±1 standard deviation ($n = 3$). Bar graphs of average porewater per treatment are provided in Fig. A2.

Porewater redox potential reduced (Fig. A3) with Straw amendment in the pure line variety (24 % decrease from Burned Straw, $p = 0.16$) and in the hybrid variety (17 % decrease from Burned Straw, $p = 0.019$). Porewater S similarly decreased with Straw amendment in the pure line and hybrid varieties (21 % and 39 % decrease from Burned Straw, respectively, Fig. A3). Porewater Mn increased with Straw relative to Burned Straw amendment for the pure line and hybrid varieties (17 % and 45 % increase, respectively, Fig. A3). Porewater As significantly increased in both years with Straw addition ($p \leq 0.0048$, Fig. 1). In the pure line variety, Straw increased average porewater As by 32 %, while in the hybrid variety Straw increased porewater As by 36 %. In both varieties, the porewater was predominately inorganic As, although the organic species monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), and trimethylarsine oxide (TMAO) were also present at lower levels (Fig. A4). Compared to Burned Straw, Straw increased porewater organic As (pure line: 52 %, $p = 0.08$; hybrid: 99 %, $p = 0.02$) and inorganic As (pure line: 38 %, p = 0.01; hybrid: 48 %, p = 0.08). The increased organic As was the result of increases in porewater MMA, DMA, and TMAO (Table A4).

The porewater molar ratio of As:Si was used to investigate the relative effect of amendment form on As release compared to Si release. In both varieties, Straw increased As:Si relative to Burned Straw (Fig. A5). In the pure line variety, Straw increased average porewater As:Si 20 % relative to Burned Straw ($p = 0.06$). In the hybrid variety, Straw increased average porewater As:Si by only 13 % relative to Burned Straw ($p = 0.91$). Both Husk and Burned Husk amendment tended to increase average porewater As:Si. In the pure line variety, Husk and Burned Husk increased average porewater As:Si by 2 and 14 %, respectively, relative to Control ($p = 0.58$). In the hybrid variety, Husk and Burned Husk increased average porewater As:Si by 25 and 92 %, respectively, relative to Control ($p = 0.09$).

The form of husk addition did not significantly affect other measured elements in the porewater, except for Ca. In the pure line variety, porewater Ca was significantly affected by the form of husk ($p = 0.017$, Fig. A3), with Husk decreasing porewater Ca by 22 % relative to the unamended Control. Burned Husk negligibly decreased porewater Ca by 1 % relative to the Control. In the hybrid variety, Husk decreased porewater Ca by 24 %, but the effect was not significant ($p = 0.19$).

3.3. Plant Si

Plant Si was more strongly affected by the form of straw amendment than the form of husk amendment (Fig. 2 and Table A5). In all plant parts measured, Si was not significantly affected by the form of husk amendment in either year ($p \geq 0.13$). In both varieties, burning the straw significantly decreased flag leaf Si ($p \leq 0.0076$). In the hybrid variety, burning the straw also significantly decreased root Si and husk Si ($p = 0.024$ and $p = 0.021$, respectively).

3.4. Plant As

Plant As was more significantly affected by the form of straw amendment than by the form of husk amendment (Fig. 3 and Table A6). In the pure line variety, only root As was significantly affected by the form of straw amendment, where burning the straw significantly decreased root As ($p = 0.0084$). In the pure line variety, burning the straw also decreased flag leaf As, but this effect was not significant ($p = 0.070$). In the hybrid variety, burning the straw significantly decreased As in all plant parts ($p \leq 0.031$). These decreases were larger in the straw, roots, and node I (29–34 % decrease) and smaller in the grain and bran (14–17 % decrease).

Arsenic speciation in the polished grain and bran was not significantly affected by either amendment in pure line variety but was affected by the form of straw amendment in the hybrid variety (Fig. 4). In the hybrid variety, burning the straw significantly decreased grain and bran total As. Burning the straw decreased grain inorganic As by 5 % ($p = 0.25$) and organic As by 21 % ($p = 0.0042$), but due to higher concentrations of grain inorganic As, these different percent increases resulted in similar absolute decreases in concentration. Similarly, bran As significantly decreased when straw was burned, with inorganic As decreasing 14 % ($p = 0.0035$) and organic As decreasing 30 % ($p = 0.0004$).

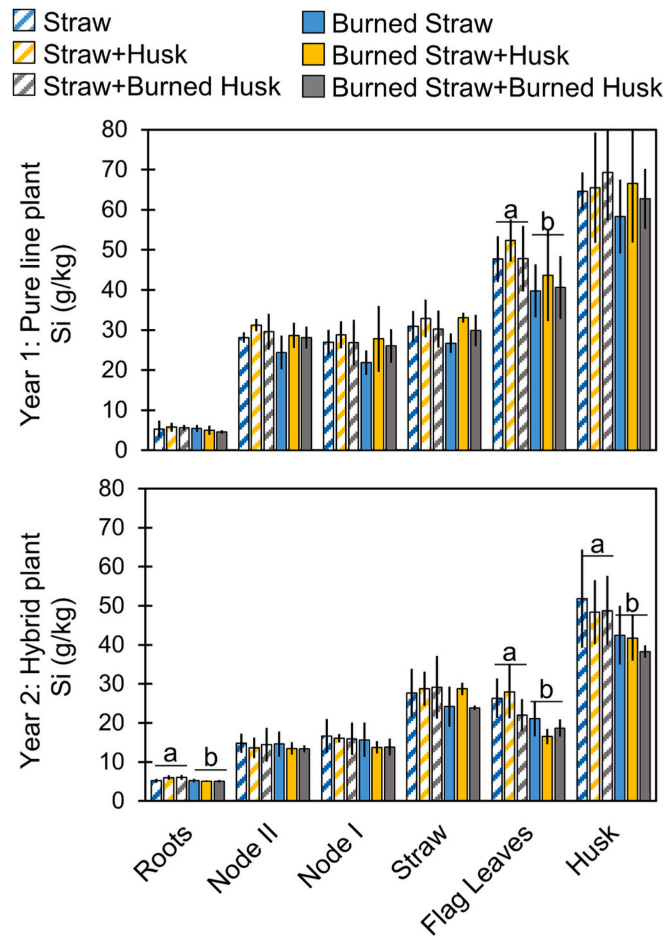

Fig. 2. Straw addition, as compared to Burned Straw addition, significantly increased plant Si in some plant parts, as shown by differing letters above groups of bars. The form of husk amendment did not significantly affect plant Si. Error bars are ±1 standard deviation ($n = 3$).

3.5. Plant cadmium

Plant Cd concentrations were not substantially affected by the forms of the amendments. In the pure line variety, Cd was only detectable in the nodes, but was significantly increased by Burned Straw ($p = 0.023$–0.10) and the form of husk ($p = 0.035$–0.036). The application of Husk increased Cd in the nodes by 60–98 % relative to Control. In the hybrid variety and using a more sensitive ICP-MS, Cd was detectable in all plant parts but was not significantly affected by the form of straw ($p > 0.27$) or the form of husk ($p > 0.59$). For the hybrid variety, average grain Cd (0.022 ± 0.009 mg/kg) was lower than average bran Cd (0.045 ± 0.014 mg/kg, Table A7). The low concentrations of plant Cd are likely due to the low soil Cd concentration and insufficiently aerobic soils. These low plant Cd concentrations preclude further discussion of the effect of amendments on Cd uptake.

3.6. Plant nutrients

The form of straw and the form of husk amendment did not

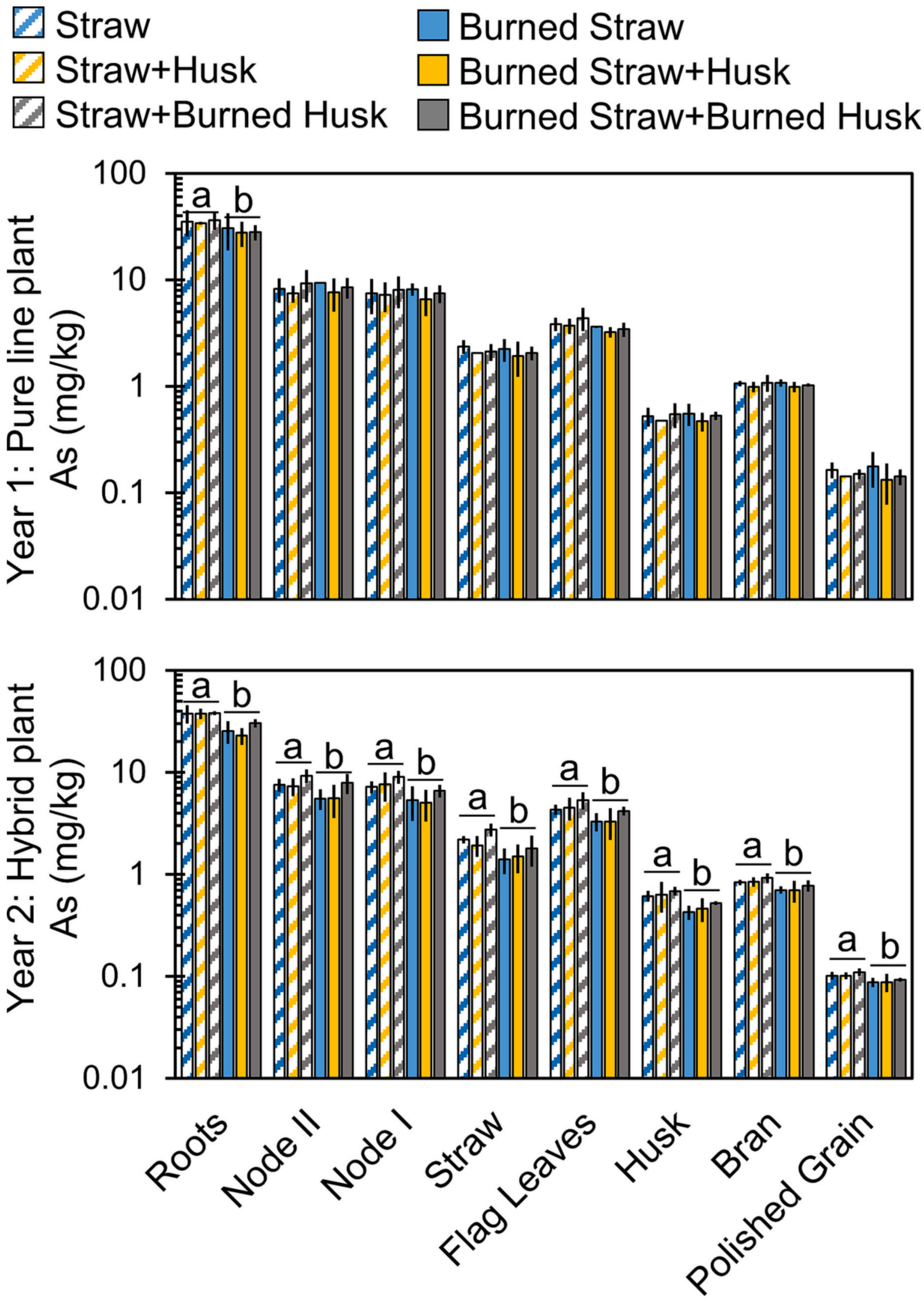

Fig. 3. Straw addition, compared to Burned Straw addition, significantly increased plant As, mostly in the hybrid variety, shown by differing letters above groups of bars. The form of husk amendment did not significantly affect plant As. Note data are plotted on a log_{10} scale. Error bars are ±1 standard deviation ($n = 3$).

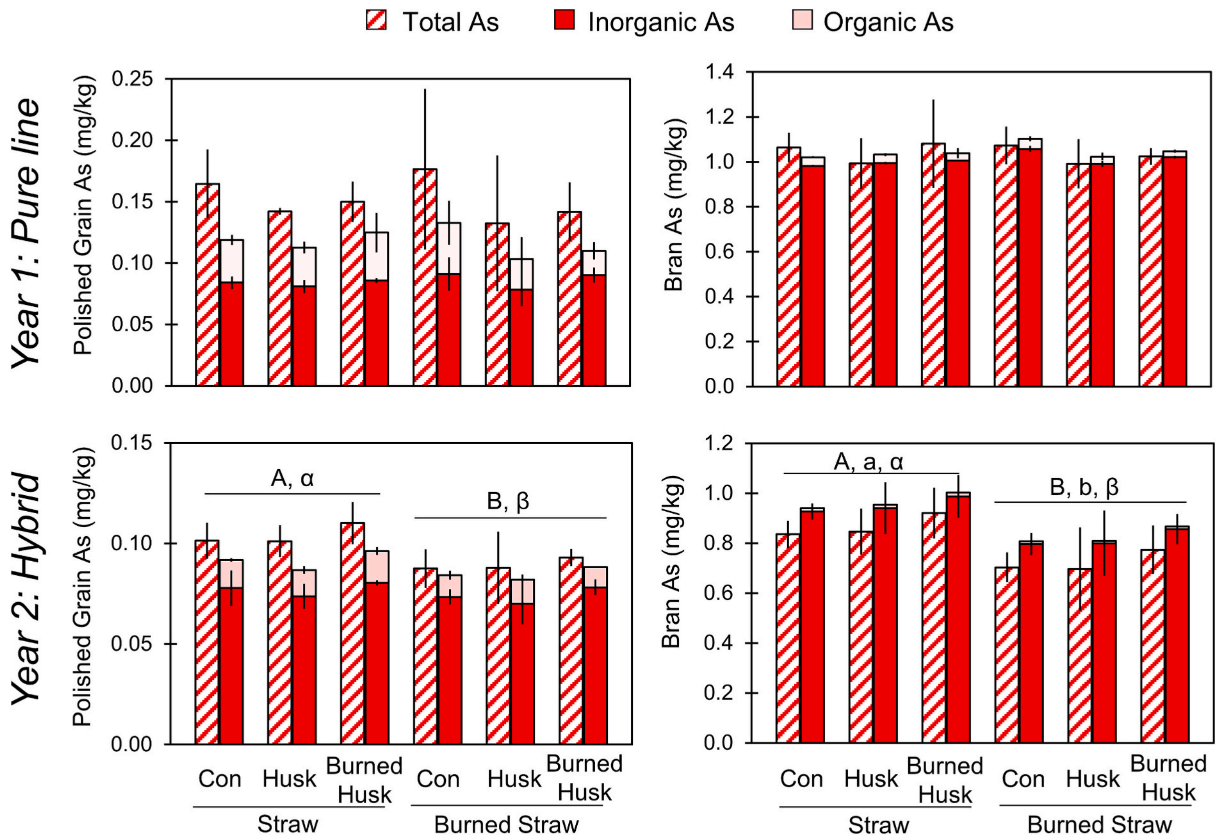

Fig. 4. Grain As concentration and speciation. Straw addition, compared to Burned Straw addition, significantly increased grain As in the hybrid variety, shown by differing letters above groups of bars. Comparisons between total As, As_i, and As_o are denoted by uppercase Latin, lowercase Latin, and Greek letters, respectively. The form of husk amendment did not significantly affect grain As. Note that y-axes are scaled differently for each panel. Error bars are ±1 standard deviation (n = 3). The overall recovery of As species was 96 ± 15 % of the total As (average ± standard deviation, n = 72).

substantially affect the concentrations of other plant nutrients measured (Figs. S4-S12, Tables S8-S16). The nutrients measured included Ca, Cu, Fe, K, Mg, Mn, P, S, and Zn in the following plant parts: polished grain, bran, husk, flag leaves, straw, node I, node II, and roots. The only nutrient that showed consistent amendment effects across most plant parts was copper (Cu) for the hybrid variety. Plant Cu significantly increased in the nodes, straw, flag leaves, husk, and polished grain when straw was burned (Fig. A7).

3.7. Iron plaque

Iron plaque accumulation on the roots was minimally affected by the form of the amendments. For the pure line root plaque Fe, neither the form of straw (p = 0.19) nor the form of husk (p = 0.54) affected the amount of Fe in the root DCB extract (overall average 51 ± 11 g Fe/kg root). In the hybrid rice, neither the form of straw (p = 0.17) nor the form of husk (p = 0.40) affected the amount of Fe in the root DCB extract (overall average 81 ± 10 g Fe/kg root). Concentrations of other elements in the DCB extract were divided by the concentration of Fe in the DCB extract and are reported as mg of element per kg of Fe (Fig. A15). In both the pure line and hybrid rice, concentrations of elements in the Fe plaque generally decreased in the order of K, Ca, P, Mn, Mg, Si, Al, and As. In the pure line variety, the form of straw and the form of husk amendment did not significantly affect the concentration of any element in the Fe plaque, while in the hybrid variety burning the straw significantly increased the concentration of Ca and Mg in the Fe plaque.

Iron plaque mineral composition was significantly affected by the form of straw amendment, but not the form of husk amendment. Using linear combination fitting of Fe EXAFS spectra, the Fe plaque was primarily ferrihydrite (FHY), lepidocrocite (LEP), and goethite (GOE), with minor amounts of siderite (SID) (Fig. 5). In the pure line variety, only the fraction of LEP in the Fe plaque was significantly affected by the form of straw amendment (p = 0.0048), with Burned Straw increasing the fraction of LEP relative to Straw. This increase in LEP primarily corresponded with a decrease in FHY (p = 0.11). In the hybrid variety, amendments had similar levels of FHY, but the form of straw amendments significantly affected LEP and GOE, with Burned Straw increasing LEP (p = 0.0004) and decreasing GOE (p = 0.0004) relative to Straw. The form of husk did not significantly affect Fe mineral composition in either year.

Arsenic speciation in the iron plaque was only affected by the form of straw amendment in the hybrid variety (Fig. A16). In the pure line variety, plaque As was relatively equal mixtures of arsenite and arsenate, but these concentrations were not affected by the form of straw amendment (p = 0.78) or the form of husk amendment (p = 0.42). In the hybrid variety, plaque As was predominately arsenate (72 ± 5.7 %), and plaque arsenite significantly decreased when straw was burned (p = 0.0024) but was not affected by the form of husk amendment (p = 0.40).

3.8. Carbon

The form of soil amendments did not significantly affect soil organic matter or porewater DOC. Soil organic matter was determined at 10 time points over the 2-year study but was rarely significantly affected by the form of amendment (Fig. A17). Only in the pure line variety at 29 days after transplanting did burning the straw significantly decrease soil organic matter (p = 0.049). In both years, soil organic matter was transiently elevated during grain filling. In the pure line variety, porewater DOC averaged 4.1 ± 1.6 mM C over the entire growing season and was not significantly affected by the form of husk (p = 0.89) or the form

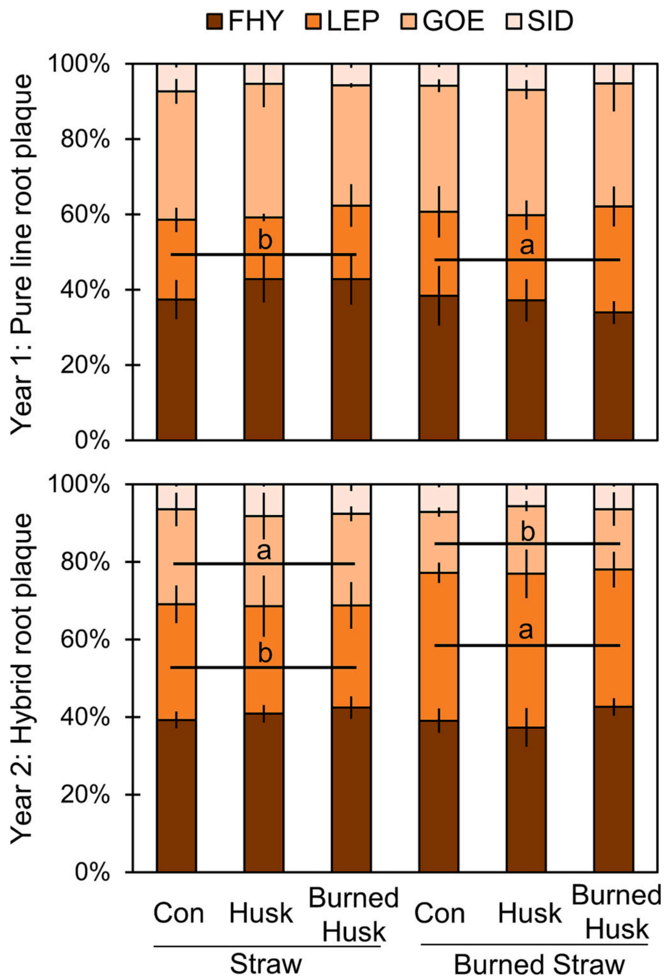

Fig. 5. Root plaque Fe mineral composition. Straw addition, as compared to Burned Straw addition, significantly decreased lepidocrocite, as shown by differing letters above groups of bars. The form of husk amendment did not significantly affect Fe mineral composition. Error bars are ±1 standard deviation ($n = 3$).

of straw ($p = 0.86$) amendments. In the hybrid variety, average porewater DOC dropped to 2.0 ± 0.63 mM C and was slightly decreased when straw was burned ($p = 0.057$) but was not affected by the form of husk amendments ($p = 0.15$).

Methane emissions were strongly influenced by the form of straw amendment, but not the form of husk amendment (Fig. 6 and Table A4). In both varieties, methane emissions were highest early in the growing season, after incorporation of amendments and flooding. In the pure line variety, cumulative CH_4 emissions over the growing season for Straw (2370 mmol/m^2) exceeded cumulative CH_4 emissions for Burned Straw (533 mmol/m^2, $p < 0.0001$). In the hybrid variety, a similar pattern was observed, with Straw amendments cumulatively emitting 1550 mmol/m^2 and Burned Straw emitting a significantly lower 356 mmol/m^2 ($p < 0.0001$).

4. Discussion

4.1. The effect of amendments on redox-sensitive parameters

Despite amending the soil with similar rates of carbon, Straw more strongly affected redox-sensitive parameters than Husk in their unburned forms. This is consistent with our hypothesis that the addition of husk would have a lesser effect on the biogeochemistry than whether the straw was burned or unburned. While the application rate of Straw (1.15 Mg C/ha/yr) was slightly higher than Husk (0.84 Mg C/ha/yr), Straw strongly affected redox-sensitive soil parameters. Compared to Burned Straw, application of Straw resulted in lower porewater redox potentials ($p = 0.049$–0.16), higher porewater Mn ($p = 0.028$–0.25), higher porewater Fe(II) ($p = 0.0029$–0.012), higher porewater As ($p = 0.0040$–0.0048), and lower porewater S ($p = 0.074$–0.0040) (Figs. 1 and A3). All these trends in porewater chemistry are consistent with the burning process decreasing the labile C concentration in the straw, resulting in noticeable differences in the mobility of redox-sensitive elements in the porewater. In contrast, the addition of Husk or Burned Husk did not significantly affect any of the redox-sensitive porewater constituents, implying that despite similar rates of C amendment (Table 1), the C in Husk is less labile than Straw C (Penido et al., 2016). The lability of Straw C was particularly evident in elevated porewater Fe (II) and As early in the growing season for Straw amendments (Fig. 1). The iron plaque data also supports the porewater data. Burning the straw increased root plaque lepidocrocite (Fig. 5). Lepidocrocite is known to transform to goethite when Fe(II) is present (Boland et al., 2014), suggesting that roots in the Straw amendment were exposed to higher levels of Fe(II) than the Burned Straw amendment. The more

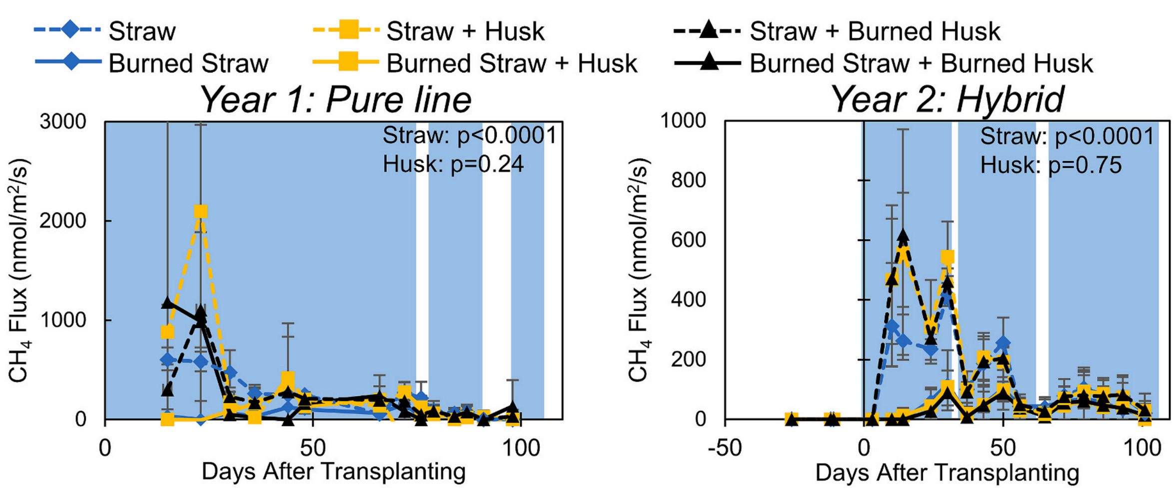

Fig. 6. Methane emissions during the growing season measured by the closed chamber technique. Straw addition, as compared to Burned Straw addition, increased cumulative CH_4 emissions. The form of husk amendment did not significantly affect cumulative CH_4 emissions. For each panel, p values are for the cumulative CH_4 emissions due to each main effect. White shaded areas indicate when paddies were not flooded. Error bars are ±1 standard deviation ($n = 3$).

reducing conditions created by Straw also increased the fraction of As (III) in the Fe plaque for the hybrid rice compared to Burned Straw (Fig. A16). Interestingly, the hybrid rice accumulated more Fe plaque on its roots (mostly in the form of LEP) compared to the pure line rice, despite being exposed to lower porewater Fe(II) and higher porewater ORP.

Similar to porewater data, the incorporation of Straw was the primary driver of CH_4 emissions. This partially supports our hypothesis that burning both straw and husk would decrease the amendment's effect on methane emissions. Incorporation of rice straw is well known to increase CH_4 emissions from flooded rice paddies (Linquist et al., 2018; Naser et al., 2007; Wang et al., 1992). In this work, Straw increased CH_4 emissions by 100–340 % compared to Burned Straw ($p < 0.0001$) and most of the CH_4 emissions occurred early in the growing season (i.e., shortly after incorporation, Fig. 6). Despite the relatively large input of Husk C, Husk CH_4 emissions were only 17–46 % higher than Control ($p = 0.24$–0.75). In previous studies at this field site, husk addition increased CH_4 emissions by 270 % compared to control when husk was applied at 30-fold the rate described in this study and rice was grown under continuously flooded conditions, (Limmer et al., 2018a), or Husk increased CH_4 emissions by 54 % when applied at 7-fold the rate and grown under AWD conditions (Linam et al., 2023). Burning rice straw prior to soil incorporation has also previously been shown to decrease CH_4 emissions. A meta-analysis of US rice methane emissions found a 57 % decrease in CH_4 emissions when rice straw was burned before incorporation (Linquist et al., 2018). This is comparable to the 50–77 % decrease in CH_4 emissions observed when rice straw was burned in this study. Few data are available comparing CH_4 emissions between straw and husk incorporation into rice soils. Penido et al. (2016) found that after 6 weeks of incubation in flooded soil, porewater in soil amended with rice straw contained ~80 μM dissolved CH_4 while soil with rice husk contained ~2 μM dissolved CH_4, a concentration similar to soil with rice straw ash. The straw amendment was also the only amendment where PCR of DNA extracted from soil identified expression of *mcrA* and *mrtA* (Penido et al., 2016), the functional marker genes for methanogenesis (Lueders et al., 2001).

4.2. The effect of amendments on Si

The type of soil amendment affected the accumulation of Si in the plant tissues, but in ways differing from our hypothesis that returning both straw and husk would increase plant levels of Si. All Si-rich amendments were applied at similar rates, with Straw and Burned Straw applied at 0.11 Mg Si/ha/yr while Husk and Burned Husk were applied at 0.15 Mg Si/ha/yr. Neither form of husk significantly affected porewater Si ($p = 0.41$–0.43) while Straw provided more porewater Si than Burned Straw ($p = 0.060$–0.15, Fig. 1). A similar trend was observed in plant accumulation of Si (Fig. 2). Straw significantly increased plant Si relative to Burned Straw in the flag leaves of both varieties and in the roots and husk of the hybrid rice. Husk tended to increase plant concentrations of Si compared to Control, but this effect was not significant ($p \geq 0.13$). The effect of Straw increasing Si in the flag leaves suggests it was able to provide more Si during periods of high Si demand, such as the reproductive growth phase (Ma and Takahashi, 2002). Note that a control without any form of straw addition was not used because removal of straw from the paddies in the United States is rarely practiced. Thus, we cannot assess the Si contribution of Burned Straw relative to a straw-less control.

Despite application of Si-rich materials at similar Si rates, Straw released more plant-available Si than Husk and both burned materials. This implies that the Si in rice husk is more recalcitrant than Si in rice straw, similar to what was observed for carbon. Thus, the dissolution of the Si from the residues appeared to be controlled by degradation of the residue carbon. Linam et al. (2021) found that 17 repetitive extractions of husk with 10 mM $CaCl_2$ for 24 h extracted <20 % of the total Si and that solution Si took 1–2 months to reach equilibrium in soil or water. Repeated 24-h extractions (28) with rice straw found Si was quickly released after 10 extractions (Xiao et al., 2014). Seyfferth et al. (2013) found that fallow season porewater Si was elevated relative to the growing season, due to the decomposition of rice straw. Marxen et al. (2016) also reported rapid degradation of rice straw, as 2–2.5 % of the rice straw phytolith (SiO_2 minerals) dissolved per day over a 33-day experiment. Few studies compare Si dissolution of both rice husk and rice straw. A 6-week flooded soil incubation study found higher porewater Si with rice husk (1 Mg Si/ha) after 4 weeks than with rice straw (1 Mg Si/ha), and rice straw resulted in strong reductions in porewater redox, unlike rice husk (Penido et al., 2016). The disagreement between this work and Penido et al. (2016) may arise from the higher Si concentration of rice husk in their study (113 g/kg) compared to this study (71 g/kg) because higher Si concentration in rice tissue has previously been found to result in more rapid degradation of the phytoliths (Marxen et al., 2016). Previous field studies with Husk in this soil has shown substantial increases in plant Si, but much higher rates of Husk were amended into the soil (5 Mg Si/ha) (Limmer et al., 2018a; Limmer and Seyfferth, 2021). In this work, neither burned material provided appreciable Si for plant uptake. Burned Straw resulted in decreased porewater Si and plant Si as compared to Straw, likely due to Si crystallization at high burning temperatures or slowed dissolution kinetics (Linam et al., 2021; Xiao et al., 2014).

4.3. The effect of amendments on plant As accumulation

Plant accumulation of As was primarily driven by changes in redox caused by Straw addition, partially supporting our hypothesis that burned forms of amendments, which provide high levels of available Si and lower levels of porewater As, would decrease rice grain inorganic As. Straw resulted in elevated porewater As in both varieties ($p = 0.0040$–0.0048, Fig. 1). For both varieties, Straw significantly increased root As; however, As only significantly increased in the aboveground tissues for hybrid rice (Fig. 3). Similarly, Straw only significantly affected grain and bran As for the hybrid rice (Fig. 4), with Straw significantly increasing total As and As_o in the polished grain and bran and also significantly increasing As_i in the bran. Silicon has frequently been demonstrated to increase grain concentrations of organic As (Seyfferth et al., 2018), likely due to increased desorption of arsenite leading to additional formation of DMA (Dykes et al., 2020). In most studies, increasing the availability of Si results in lower grain As_i (Fleck et al., 2013; Liu et al., 2014; Seyfferth et al., 2018), which did not occur in this study. However, in this work, increased porewater Si was confounded with decreased redox and increased porewater As (Fig. 1). The porewater As:Si ratio (Fig. A5) showed that Straw had 13–20 % higher As:Si, although this effect was not significant. Thus, the increase of porewater and plant Si by Straw appeared to be offset by the increase in porewater As due to increased reductive dissolution of oxidized Fe by the labile C present in Straw. Husk and burned amendments performed similarly, providing similar levels of porewater Si and As, resulting in similar plant levels of As.

5. Conclusion

Collectively, we observed that the addition of Straw more strongly affected redox conditions and accumulation of As than Burned Straw or any form of husk addition. The labile C supplied by Straw drove reductive dissolution of Fe oxides, mobilized As, and increased methane emissions. Burning straw, rather than incorporating unburned straw into the soil, can decrease grain arsenic and rice paddy methane emissions. The inability of husk to provide additional Si, despite similar application rates to straw, suggests that the lability of Si in rice husk is low and may require long-term studies with annual applications or high rates of application to observe significant effects.

CRediT authorship contribution statement

MAL and ALS designed the experiment; MAL and FAL conducted the experiment and analyzed the data; MAL wrote the paper with input from all co-authors.

Declaration of competing interest

The authors declare no competing financial conflicts of interest.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.166496.

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