

## Supplementary Information

### Multi-mode soil chemical passivation and crop protection of severe cadmium and arsenic polluted soils with engineered silica

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**Text S1.** Information of plant pot trial.

**Soil treatment:** To study the performance of FMS amendment high pollution risk soil, we

added CdCl<sub>2</sub> to increase BJ soil Cd content to 5 mg/kg, this approach is similar to that reported in the study by Wang et al (2019).<sup>1</sup> For rice and pakchoi pot trial, nitrogen fertilizer (urea), phosphorus fertilizer (superphosphate), and potassium fertilizer (potassium chloride) were 1, 0.4, and 0.46 g/pot, respectively.

**Plant growth:** Rice seeds were sterilized in 30% H<sub>2</sub>O<sub>2</sub> solution for 15 min, then washed with deionized water. The seeds were germinated in Petri dishes (100 mm diameter) each containing moist filter paper, these were kept in a greenhouse with shade cover for one week. Each day ~10 ml deionized water was added into the dishes to maintain sufficient moisture for growth. After that, seeds were transfer to Yoshida nutrient solution for one week for sprouting.<sup>2</sup> Finally, rice seedlings were transplanted to the mesocosms (2 seedling each mesocosm), where long-term continuous flooding water treatments were maintained: for example 3-5 cm submerged water layer. Two weeks before harvest soils were allowed to dry out.<sup>3</sup> Pakchoi seeds were directly transferred into the pot (10-15 seeds each pot), and the soil water holding capacity (WHC) was maintained around 60% until pakchoi harvest.

**Harvest:** The entire fresh rice roots were collected for iron plaque extraction. Rice tissues (grain, leaf, straw and plaque-free root) and pakchoi were washed three times with ultra-pure water (Milli-Q water, resistivity: 18.2 MΩ.cm). Then the samples were dried at 60°C for 3 days until a constant weight was obtained. The dried rice grain was mechanically de-husked (LTJM-2099 Rice Dehusker) without polishing/removal of the

bran layer, then dried rice tissues and pakchoi were ground into a fine powder (DonLim DL-MD18 mill). The soil was dried at room temperature after plant harvest, and dried soil was sieved (< 2 mm) before BCR sequential extraction and anti-aqua regia digestion.<sup>4</sup> All containers used in the experiment were soaked in 5% HNO<sub>3</sub> bath for 24 hrs before use.

***DCB extraction and soil analysis:*** Iron plaque on the surface of the fresh roots was extracted using the dithionite–citrate–bicarbonate (DCB) method. Soil pH/Eh was determined in a 1:2.5 soil/water suspension. BCR (Community Bureau of Reference) sequential extraction was used to identify the different fractions of elements in the soil,<sup>5</sup> performed using a three-stage modified procedure plus the residual aqua regia digestion, detailed progress show in Table S7. Additionally, the Anti-Aqua Regia digestion (AAR) was used to validate the BCR extraction and determine the pseudo total metal content: 100 mg of each dried and ground soil sample was weighed on the digital weighing scales (Sartorius – BSA2245. Max: 220g, d: 0.1mg) and placed into a Teflon tube, with the precise weight being recorded (0.001g). 7.5 ml conc HNO<sub>3</sub> and 2.5 ml HCl was added to each tube. The same volume of acid was also added to 3 tubes designated as blank and the 3 tubes containing certified reference material (GBW07405 soil flour). Tubes were then placed into the carousel of the microwave digester (PreeKem TOPEX+). The program heated the samples up to 190°C; detailed setup is shown in Table S8. After cooling, samples are made up to a final weight of about 20g with deionized water with these weights being recorded precisely. Then, the sample was filtered (PES membrane, pore size: 0.45 μm,

270mm\*10m) and diluted finally 1000 times.

**Plant sample digestion:** 100 mg of each dried and powdered sample was weighed on digital weighing scales (Sartorius – BSA2245. Max: 220g, d: 0.1mg) and placed into a precleaned Teflon tube, with the precise weight being recorded (0.001 g). 2mls of BDH Prolabo Aristar 69% nitric acid (HNO<sub>3</sub>) was added to each tube. The same volume of nitric acid was also added to 8 tubes designated as blank and the 8 tubes containing certified reference material (GBW10010 rice flour). Tubes were briefly shaken, and the mixtures were allowed to soak overnight. After that, 2mls BDH Prolabo Analar Normapur 30% hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) was added to each Teflon tube to enhance digestion. Then the open tubes were outgassed for 15 minutes. Tubes were then placed into the carousel of the microwave digester (PreeKem TOPEX+). The program heated the samples up to 95°C; detailed setup is shown in Table S8. After cooling, add deionized water to the Teflon tube to bring the sample weight in the Teflon tube to 20 g, the samples were then transferred to the PVC tube for further processing. Samples were filtered (PES membrane, pore size: 0.45 µm, 270mm\*10m) and diluted (final dilution: 500 times) before analysis.

**Text S2.** Information of soil incubation experiments.

**Soil incubation:** The soil was dried and sieved (< 2mm), and mixed FMS was added to

the dried soil and shaken for 60 minutes using a DVX-2500 Multi-Tube Vortexer at a speed of 500 RCF to ensure thorough mixing with the soil. The soil-FMS mixture was then wetted to 100% maximum water holding capacity (MWHC) and transferred to the incubator (New Brunswick Innova 44 Stackable Incubator Shaker), with the temperature set at 24°C. During the initial week, pots were weighed daily to monitor the evaporation rate within the incubator. In subsequent weeks, the pots were weighed and watered according to a predetermined schedule based on these initial observations.

**DGT deployment and collection:** Chelex-100 and Metsorb mixed binding layer DGT devices (standard DGT holder with 0.78 mm APA diffusive gel, polyethersulphone filter membrane) assembled by DGT Research Ltd. DGT devices were deployed on the soil surface at the end of the incubation period, after c. 24 h, DGT devices were collected, jet washed with Milli-Q water (18.2 MΩ.cm) to remove soil particles and stored at < 4°C in a refrigerator (Liebherr Mediline LKexv 3910) before disassembling. Resin gels, once removed from the DGT device, were placed in 1.5 ml PVC tubes before elution. Chelex-Metsorb gel was first eluted in 1 mL 1 M HNO<sub>3</sub> for 24 h to extract cations, then wash the binding gel surface with ca. 5 mL deionised water, followed by elution in 1 mL 1 M NaOH for 24 h to extract anions, the eluents were mixed prior analysis.<sup>6</sup> Samples were filtered (PES membrane, pore size: 0.45 µm, 270mm\*10m) and diluted 5 times with 2% HNO<sub>3</sub> before analysis.

**Porewater collection:** after the deployment of the DGT was completed, soil was

centrifuged (30 mins, 4500 RCF) (centrifuge: fisherbrand CT 2R Expert Centrifuge), and the supernatant was collected and filtered (PES membrane, pore size: 0.45  $\mu\text{m}$ , 270mm\*10m) by using 10ml syringes (Terumo Sterile Syringes – Luer Slip). Then, samples were diluted 2 times with 2%  $\text{HNO}_3$  in labelled polypropylene (PP) centrifuge tube (15ml) and stored at  $< 4^\circ\text{C}$  in a refrigerator (Liebherr Mediline LKexv 3910) until analysis.

**Calculation of DGT:** The DGT-measured concentration ( $C_{\text{DGT}}$ ,  $\mu\text{g/L}$ ) was calculated using the DGT equation (Eq. 1).<sup>7</sup>

$$C_{\text{DGT}} = M\Delta g/D_dAT \text{ (Eq. 1)}$$

M (ng) is the mass of ions that diffused into the resin layer, which can be calculated from Eq. 2,  $\Delta g$  is the thickness of the diffusive layer (0.78 mm) and filter membrane (0.13 mm),  $D_d$  is the element diffusive coefficient in the diffusive layer (At 25  $^\circ\text{C}$ ,  $A_s = 6.02 \times 10^{-6} \text{ cm}^2/\text{s}$ ,  $C_d = 5.56 \times 10^{-6} \text{ cm}^2/\text{s}$ ).<sup>6</sup> A is the diffusive area of the device (3.14  $\text{cm}^2$ ), and T is the deployment time (24h).

$$M = C_e(V_g + V_e)/f_e \text{ (Eq. 2)}$$

$V_e$  is the volume of eluted solution (2 mL),  $C_e$  is the concentration of ions in eluted solution, which was measured by ICP-MS in this study,  $f_e$  is the elution efficiencies for binding layer ( $A_s = 80.4$ ,  $C_d = 93.0$ ).<sup>6</sup>  $V_g$  is the volume of gel in the resin gel layer. In

practice,  $V_g$  is often negligible.<sup>7</sup>

**DGT induced fluxes in sediments and soils (DIFS):** The DIFS model provides insights into the kinetics of the resupply process of an element in the soil-porewater-DGT system.<sup>8</sup> In this study, 2D-DIFS was used to calculate  $R_{diff}$ ,<sup>9</sup> which helps in analysing the ability of soil to replenish metals(loid) in the soil solution and improve the understanding of the FMS-treated soil. Input parameters:  $\Delta g$ , the porosity of the diffusive gel ( $\phi^d$ ) was set at 0.95, as estimated by Zhang and Davison (1995),  $D_d$  (At 25 °C,  $A_s = 6.02 \times 10^{-6} \text{ cm}^2/\text{s}$ ,  $C_d = 5.56 \times 10^{-6} \text{ cm}^2/\text{s}$ ) and  $D_0$  (At 25 °C,  $A_s: 5.26 \times 10^{-6} \text{ cm}^2/\text{s}$ ,  $C_d = 5.52 \times 10^{-6} \text{ cm}^2/\text{s}$ ),<sup>6</sup>  $K_d$  ( $0.0 \text{ cm}^3/\text{g}$ ),  $T_c$  ( $1E10 \text{ s}$ ), particle concentration ( $P_c$ ).

The calculation of  $P_c$  was based on Menezes-Blackburn et al. (2016),<sup>10</sup> considering the ratio between the dry weight of soil and the mass of water at the soil saturation point (Eq. 3).

$$p_c = \frac{m}{m_1 - m/\rho} \text{ (Eq. 3)}$$

Where  $m$  represents the mass (g) of dry-weight soil,  $m_1$  is the mass (g) of soil after saturation with deionised water, and  $\rho$  ( $\text{g}/\text{cm}^3$ ) is the water density at the temperature of the experiment.

$R_{diff}$  refers to the ratio of the theoretical interface concentration between the DGT device and soil (or sediment) to the soil solution in the presence of only solution diffusion without particulate supplementation (single diffusion type). The effective concentration ( $C_E$ ,  $\mu\text{g/L}$ ) is the effectively available concentration from both soil solution and the solid phase labile pool, calculated according to Eq. 4.

$$C_E = C_{DGT}/R_{diff} \text{ (Eq. 4)}$$

The ratio ( $R$ ) between  $C_{DGT}$  and the pore-water concentration ( $C_{sol}$ ) (Eq. 5) was used to indicate the extent of the depletion of solution concentration at the DGT interface and describe the ability of soil particles to replenish metals when metals in the soil solution are transferred or consumed.

$$R = C_{DGT}/C_{sol} \text{ (Eq. 5)}$$

An  $R$ -value of  $> 0.95$  demonstrates that soil buffering is sufficient to maintain porewater element concentrations, whereas a value equal to or close to  $R_{diff}$  represents a diffusion-only supply to the DGT.

**Text S3.** Preparation of spiked FMS.

**Pre-lording process of K/Mn/P spiked FMS**

Weigh 1 g of FMS and add it to 40 ml of nutrition element solution (container: 50 ml glass bottle); detailed information is shown in Table ST1. The oscillator (Jipad-200TLMS) was used for loading experiments, shaking speed set up as 200 RCF, and shaking for 4 h at room temperature. After shaking, the supernatant was collected and filtered (PES membrane, pore size: 0.45  $\mu\text{m}$ , 270mm\*10m), and ICP-OES was used to determine the remaining element content in the solution. Add 20 ml of Milli-Q water (18.2 M $\Omega$ .cm) to the glass bottle to rinse the spiked FMS, repeating 3 times, then place the spiked FMS in an oven at 65  $^{\circ}\text{C}$  to dry for 3 days.

Loading capacity (LC) is calculated by Eq. 6.

$$LC = (C_0 - C_t) \times V_0 / m_{FMS} \text{ (Eq. 6.)}$$

Where  $C_0$  is the initial concentration of standard solution,  $C_t$  is the concentration of standard solution after FMS adsorbed,  $V_0$  is the volume of standard solution, and the  $m_{FMS}$  is the weight of FMS in the adsorption experiment.

#### **Salicylic acid (SA) spiked FMS experimental design**

Weigh 5 g of FMS and add it to 40 ml of SA solution (container: 50 ml glass bottle); detailed information is shown in Table ST 1. The constant temperature oscillator (Jipad-200TLMS) was used, shaking speed set up as 200 RCF, and shaking for 7 h at room temperature. After shaking, the supernatant was collected and filtered (PES membrane, pore size: 0.45

$\mu\text{m}$ , 270mm\*10m), and HPLC was used to determine the remaining SA content in the solution. Add 20 ml of Milli-Q water (18.2 M $\Omega$ .cm) to the glass bottle to rinse the spiked FMS, repeating 3 times, then place the spiked FMS in an oven at 65 0C to dry for 3 days.

Add SA-spiked FMS (5 g) to 40 ml of Milli-Q water (container: 50 ml glass bottle), the constant temperature oscillator (Jipad-200TLMS) was used, shaking speed set up as 200 RCF, and shaking for a total of 24 h at room temperature. Sampling was carried out at 3 h, 6 h, 21 h, and 24 h. The specific sampling operation was as follows: collect the supernatant, 20 ml of Milli-Q water (18.2 M $\Omega$ .cm) was used to rinse the spiked FMS in the glass bottle, repeating 3 times. Then add 40 ml of Milli-Q water (18.2 M $\Omega$ .cm) again and continue shaking. All supernatant was filtered (PES membrane, pore size: 0.45  $\mu\text{m}$ , 270mm\*10m), and HPLC was used to determine the content of SA in the supernatant.

**Table ST1.** FMS Loading experiment set up

Target	Solution concentration (mg/kg)	Solution pH	FMS code	Replication
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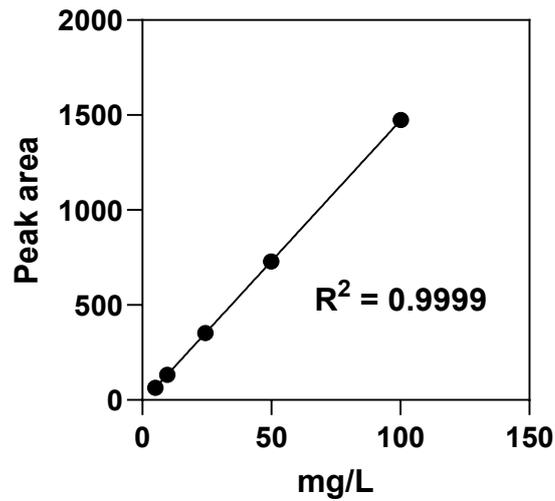
K	503	6.94	FSP-1, FSP-2, FSN-2, FSN-3, FSN-4	3
Mn	498	6.85	FSP-1, FSP-2, FSPN-1, FSPN-2	3
P	391	5.56	FSN-2, FSN-3, FSN-4	3
Salicylic acid	2863	2.77	FSA	3

Potassium chloride (KCl) was used for K solution preparation; manganese(II) sulfate monohydrate ( $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ ) was used for Mn solution preparation; and sodium dihydrogen phosphate ( $\text{NaH}_2\text{PO}_4$ ) was used for P solution preparation. Weigh 1.5 g salicylic acid (CAS 69-72-7, Aladdin) and dissolved in 70 ml ethanol, then Milli-Q water (18.2 M $\Omega$ .cm) was used to make the volume to 500 ml. Ultrasonic treatment of the dissolved liquid removes bubbles and accelerates the dissolution. Then, transfer the SA solution to the fume hood and stand it overnight.

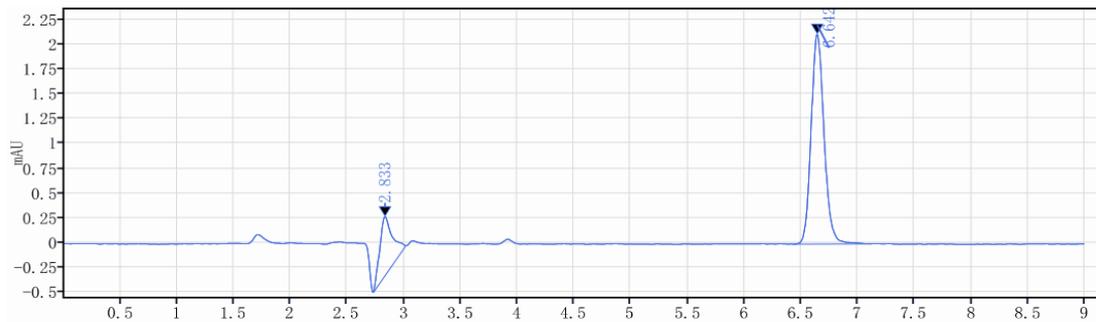
**Text S4.** HPLC and XRF measurement.

**Salicylic acid (SA) HPLC measurement**

SA solution sample was analyzed by HPLC (Agilent 1260 infinity 2 series HPLC). The column used is Agilent 5HC-C18(2), 250 x 4.6 mm. The mobile phase's volume ratio is water: methanol: acetic acid = 40: 59: 1. After cooling to room temperature, a 40 µm mixed-phase filter membrane was used to filter the mobile phase, which was then ultrasonicated for 15 min (mainly to eliminate air bubbles). After that, a 40 µm filter was used to pump the diluted sample into a 1 ml sample bottle, then put it into the sample tank, adjust the sequence, and change the flow of the injected mobile phase before the sample injection. Open the gas valve (gas flow: ~5 ml/min) and exhaust for 5 min to balance the pressure, then set up the gas flow to ~1 ml/min. After the pressure value of the instrument and the absorbance of the UV lamp are stabilized, the sample analysis can be started. 5 standards were prepared (5 mg/L, 10 mg/L, 25 mg/L, 50 mg/L, 100 mg/L) using Salicylic Acid (CAS 69-72-7, Aladdin). The standard curve is shown on Fig. ST 1. ( $R^2 > 0.999$ ). The flow rate was ~1 ml/min, the wavelength was 306 nm UV detection, and the sampling volume was 10 µg. Fig. ST 2. shows the peak of SA appearing during 6.5 – 7 min, and the peak of 2.5 – 3 min is mainly the solvent peak of the mobile phase. The process of testing a sample can be completed within 8 min.



**Fig. ST 1. HPLC SA standard curve**



**Fig. ST 2. HPLC chromatogram of SA analysis**

**Plant sample XRF measurement**

A 3g subsample of dried, powdered rice tissues/pakchoi was transferred into a prepared XRF cup (32 mm double open-ended plastic rings with 4  $\mu$ m thick propylene X-ray film at the base), then a manual press (PANAPRESS) was used to crush them into a pellet with approximately 300 pascal (Pa) for 10 seconds, after that the height of the sample was recorded. The instrument we used is Cartesian geometry energy dispersive X-ray fluorescence spectrometer (ED-XRF) (Rigaku NEX CG). The analysis method we choose is FS47 CuMgFe, the measurement time of each sample is 13.30 mins. The concentration

of Fe and Si in rice tissues and pakchoi samples were obtained from ED-XRF.

**Table S1.** BJ-soil and LPS-soil information (Mean  $\pm$  s.d.).

		BJ-soil (mg/kg) (n=9)				LPS-soil (mg/kg) (n=3)		
As		30.8	$\pm$	1.38		47.9	$\pm$	5.19
Cd		1.88	$\pm$	0.25		9.54	$\pm$	0.27
Cu		52.6	$\pm$	2.19		58.7	$\pm$	6.14
Mn		671	$\pm$	27		1069	$\pm$	40
Zn		176	$\pm$	6		943	$\pm$	44

**Table S2.** CZ soil, GD soil, and GG soil elements content and pH.

		<b>As</b>	<b>Cu</b>	<b>Fe</b>	<b>Mn</b>	<b>P</b>	<b>Pb</b>	<b>Si</b>	<b>Zn</b>	<b>pH</b>
		(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	
<b>Chenzhou soil</b>										
<b>CZ</b>	Average	73.3	36.8	35933	360	931	90.7	271889	146	6.91
	Std dev.	2.2	1.2	608	21	14	1.8	2315	6	
<b>Dabaoshan soil</b>										
<b>GD (a)</b>	Average	56.1	321	44133	273	791	199	289667	381	5.72
	Std dev.	3.2	29	2376	9	34	15	6028	20	
<b>GD (b)</b>	Average	40.7	309	36600	234	1557	165	301000	370	6.18
	Std dev.	2.3	17	1836	8	76	8	9644	24	
<b>GD (c)</b>	Average	37.7	364	29100	185	862	207	315667	547	4.98
	Std dev.	2.8	24	1539	9	31	16	5686	42	
<b>Gaoming soil</b>										
<b>GG (a)</b>	Average	24.6	33.2	33233	150	2073	80.0	245333	146	5.68
	Std dev.	2.9	1.9	2371	19	191	5.9	24007	8	
<b>GG (b)</b>	Average	12.4	29.9	18767	226	965	63.1	331000	61.9	5.18
	Std dev.	0.8	3.6	153	12	34	0.4	2646	2.6	
<b>GG (c)</b>	Average	11.0	42.3	15333	117	1053	62.7	348333	61.6	5.34
	Std dev.	0.4	1.3	751	8	47	2.3	4509	1.8	
<b>GG (d)</b>	Average	7.9	29.0	14200	104	878	46.2	357667	48.6	5.05
	Std dev.	1.2	1.6	854	9	26	2.6	1528	1.6	

**Table S3.** Cd concentration in Dabaoshan soil and Gaoming soil. Cd<sup>R</sup> : Cd concentration reported by Williams et al. (2012).<sup>11</sup> Cd\*: Calculated Cd concentration in each mixed soil sample.

<b>Field</b>	<b>Subsample</b>	<b>Wt. (g)</b>	<b>Cd<sup>R</sup> (mg/kg)</b>	<b>Cd* (mg/kg)</b>
<b>Dabaoshan soil</b>				
<b>GD (a)</b>	1	144	1.09	1.01
	2	161	0.94	
<b>GD (b)</b>	1	112	0.74	0.74
	2	148	0.84	
	3	165	0.64	
<b>GD (c)</b>	1	180	1.29	1.14
	2	38	1.31	
	3	164	0.94	
<b>Gaoming soil</b>				

<b>GG (a)</b>	1	133	1.13	
	2	131	1.44	1.06
	3	144	0.65	
<b>GG (b)</b>	1	141	0.18	
	2	137	0.19	0.25
	3	153	0.36	
<b>GG (c)</b>	1	120	0.14	
	2	142	0.17	0.19
	3	98	0.28	
<b>GG (d)</b>	1	172	0.21	
	2	165	0.15	0.20
	3	239	0.22	

**Table S4.** Soil properties of Dabaoshan soil and Gaoming soil, reported by Williams et al. (2012) (Mean  $\pm$  s.e.).<sup>11</sup>

	<b>Dabaoshan soil</b>				<b>Gaoming soil</b>			
<b>Clay (%)</b>	27	$\pm$	5.3		28	$\pm$	1.8	
<b>Silt (%)</b>	46	$\pm$	9.2		39	$\pm$	0.5	
<b>Sand (%)</b>	27	$\pm$	5.5		32	$\pm$	1.6	
<b>Organic C (<math>\mu\text{g/g}</math>)</b>	20	$\pm$	4.0		23	$\pm$	0.9	
<b>Texture</b>	Clay Loam				Clay Loam			

**Table S5.** Seed information (China Rice Data Center).<sup>12</sup>

Rice code	Whole		Chalky grain rate (%)	Chalkyness (%)	Amylose content (%)	Aspect ratio	National Approval Rice Number
	white rice rate (%)	rice rate (%)					
CY67	□	54	23	2.8	17.7	3.5	20196039
YY17	□	60	22	2.4	16.6	3.1	20190055

The product name of the pakchoi seed is Feicuikuaicai, variety purity > 96%; germination rate > 85%; moisture content < 7%.

**Table S6.** FMS and MS characteristics. (\*FSN-3: higher -NH<sub>2</sub> content. \*FSN-4: higher -SO<sub>3</sub>H content).

Work package	FMS code	Functional group	Functional group loading (mmol/g)	Specific surface area (m <sup>2</sup> /g)	Particle size (μm)	Capacity of channel in the hole (ml/g)	Diameter of channel in hole (Å)
Plant pot trial	□	□	□	□	□	□	□
	MS	□	□	350~550	180-600	□	□
	□	□	□	□	≥95%	0.6-1	40-75
Soil incubation (mixed FMS = FS-1:FS-2:FS-3 = 2:1:1)	FS-1	-SH	1.2	395	180-600	□	□
	□	□	□	□	□	□	□
	FS-1	-SH, -	1.2	395	180-600	□	□
	FS-2	PO <sub>3</sub> H <sub>2</sub>	1.0-1.3	400-450	180-600	□	□
	□	□	□	□	≥95%	0.5-0.8	6.0-8.0
	FS-3	-SH, -SO <sub>3</sub> H	1.0-1.3	420-440	180-600	□	□
Pre-loading and release of FMS	□	□	□	□	□	□	□
	FSN-2	-SO <sub>3</sub> H, -NH <sub>2</sub>	0.9-2.2	240-280	180-600	□	□
	□	□	□	□	□	□	□
	*FSN-3	-SO <sub>3</sub> H, -NH <sub>2</sub>	0.9-2.2	240-280	180-600	□	□
	□	□	□	□	□	□	□
*FSN-4	-SO <sub>3</sub> H, -NH <sub>2</sub>	0.9-2.2	240-280	180-600	□	□	

		-SH, -			180-600	
FSP-1		PO <sub>3</sub> H <sub>2</sub>	1.0-1.3	410-430	≥95%	0.3-0.5 4.0-6.0
		-SO <sub>3</sub> H, -			180-600	
FSP-2		PO <sub>3</sub> H <sub>2</sub>	0.9-1.4	400-430	≥95%	0.6-0.9 6.0-9.0
		-SH, -			180-600	
FSPN		SO <sub>3</sub> H, -			180-600	
-1		PO <sub>3</sub> H <sub>2</sub>	1.0-1.6	310-330	≥95%	0.4-0.6 5.0-6.0
FSPN					180-600	
-2		-SH, -NH <sub>2</sub>	1.6-2.2	330-370	≥95%	0.5-0.8 5.0-7.0
					180-600	
FSA		-NH <sub>2</sub>	1.6-2.2	240-260	≥95%	0.2-0.4 4.0-7.0

**Table S7.** BCR sequential extraction.

Extraction step	Reagent(s)	Nominal target phase(s)
1	HOAc (0.11 mol/L)	Soil solution, carbonates, exchangeable metals
2	NH <sub>2</sub> OH.HCl (0.1 mol/L)	Oxides Fe/Mn
3	H <sub>2</sub> O <sub>2</sub> (8.8 mol/L) then NH <sub>4</sub> OAc (1.0 mol/L) at pH 2	Organic matter and sulphides
Residual	HCl + HNO <sub>3</sub>	Remaining, non-silicate bound metals

**Step 1** (Acid-extractable/exchangeable fraction): Dried soil (1 g) was added to 40 ml of 0.11 mol/L acetic acid (HOAc), shaken for 16 hrs at room temperature. Separation of the extract: separate extract from the solid residue by centrifugation at 3000 RCF for 20 min, and the resultant supernatant liquid was transferred into a polyethylene volumetric flask. The residue was washed by adding 20 mL of deionized water, shaken for 15 min on the end-over-end shaker, and centrifuged for 20 min at 3000 RCF. Subsequently, the supernatant was decanted.

**Step 2** (Reducible fraction): To Step 1 residue add 40 ml of 0.1 mol/L freshly prepared hydroxylammonium chloride ( $\text{NH}_4\text{OH}\cdot\text{HCl}$ ) and re-suspended by shaking for 16 hrs at room temperature. The separation of the extract was the same as described in Step 1.

**Step 3** (Oxidizable fraction): the residue in Step 2 was treated twice with 10 mL of 8.8 mol/L hydrogen peroxide ( $\text{H}_2\text{O}_2$ ). First, 10 mL of  $\text{H}_2\text{O}_2$  was added to the residue from Step 2 in the centrifuge tube. The digestion was allowed to proceed at room temperature for 1 h with occasional manual shaking, followed by digestion at  $85 \pm 2$  °C for another 1 h in a water bath. During the digestion, the centrifuge tube was loosely covered to prevent a substantial loss of  $\text{H}_2\text{O}_2$ . Following that, the centrifuge tube was uncovered, and heating was continued until the volume reduced to about 2–3 ml. An additional 10 mL of  $\text{H}_2\text{O}_2$  was added to the tube, covered, and digested with cover at  $85 \pm 2$  °C for another hour. Heating was continued as before until the volume was reduced to 2–3 ml. Finally, 50 mL of 1 mol/L ammonium acetate ( $\text{NH}_4\text{OAc}$ ) was added to the cold mixture and shaken for 16 hrs at room temperature. The separation of the extract was the same as described in Step 1.

**Step 4** (Residual fraction): the residue from Step 3 was digested using aqua regia (7.5 ml 6 mol/L HCl and 2.5 ml 14 mol/L  $\text{HNO}_3$ ) and microwave digestion: the temperature was increased to 200°C in 20 min and then maintained constant for 40 min.

The extractants were prepared according to the following procedure:

**Solution I** (acetic acid, 0.11 mol/L): Redistilled glacial acetic acid,  $25 \pm 0.2$  mL, was added (in a fume cupboard), to about 500 mL of deionized water in a 1000 mL polyethylene volumetric flask and made up to the mark. 250 mL of this aliquot (0.43 mol/L acetic acid) was diluted to 1.0 L to obtain an acetic acid concentration of 0.11 mol/L.

**Solution II** (hydroxylammonium chloride, 0.5 mol/L, pH 1.5): Hydroxylammonium chloride (34.75 g) was dissolved in deionized water. The solution was acidified with concentrated nitric acid to pH 1.5 and made up to 1000 mL with deionized water.

**Solution III** (hydrogen peroxide, 8.8 mol/L): Hydrogen peroxide was used as supplied by the manufacturer.

**Solution IV** (ammonium acetate, 1.0 mol/L): Ammonium acetate (77.08 g) was dissolved in 900 mL of deionized water. The solution was acidified to pH 2.0 with concentrated nitric acid and made up to 1000 ml.

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The extraction step of 1, 2, and 3 was reported by Nemati et al., (2011),<sup>5</sup> the extraction step of residual was reported by Yang et al., (2020).<sup>13</sup>

**Table S8.** Microwave digester (PreeKem TOPEX+) set up.

	Temperature (°C)	Ramping (mins)	Holding (mins)
Soil AAR digestion	120	5	15
	150	5	10
	190	5	50
Plant digestion	55	5	30
	75	5	10
	95	5	30

**Table S9.** Instrumental parameters for ICP-MS (Agilent 7700X and NexION 300X) and ICP-OES (Varian Vista MPX CCD) analysis.

	Instrument condition	
ICP-MS (Agilent 7700X)	Measure mode	KED
	RF power	1500 W
	Plasma gas flow	15 L/min
	Auxiliary gas flow	0.40 L/min
	Nebulizer gas flow	0.80 L/min
	Collision gas	Helium (He)
	Collision gas flow	5.0 ml/min
	Internal standard	<sup>103</sup> Rh
	Isotopes monitored	<sup>75</sup> As, <sup>111</sup> Cd, <sup>63</sup> Cu, <sup>55</sup> Mn, <sup>66</sup> Zn
ICP-MS (NexION 300X, PerkinElmer)	Measure mode	KED
	RF power	1550 W
	Plasma gas flow	17L/min
	Auxiliary gas flow	1.2 L/min
	Nebulizer gas flow	0.85 L/min
	Collision gas	Helium (He)
	Collision gas flow	4.0 ml/min
	Internal standard	<sup>115</sup> In
	Isotopes monitored	<sup>75</sup> As and <sup>111</sup> Cd
ICP-OES (Varian Vista MPX CCD)	Polychromator	Echelle
	Grating	94.74 lines/mm
	Power	1.30kW
	Frequency	40 MHz
	Plasma flow	15.0 L/min
	Auxiliary gas flow	1.5 L/min
	Nebulizer gas flow	0.72 L/min
	Viewing height	7 mm

Replicate read time	10 s
Instrument stabilization delay	45 s
Sample uptake delay	30 s
Pump rate	15 rpm
Rinse time	30 s
Analyte (wavelength)	Fe (238.2 nm), K (766.5 nm), Mn (257.6 nm), and P (213.6 nm)

**Table S10.** Quality control of ICP-MS, ICP-OES, and ED-XRF analysis.

		Measured Concentration (mg/kg)	Certified Value (mg/kg)	Recovery (%)	Limits of Detection ( $\mu\text{g/L}$ )
<b>ICP-MS</b>					
GBW10010 rice flour					
	As	0.09 $\pm$ 0.02	0.102	90	0.158
	Cd	0.09 $\pm$ 0.01	0.087	104	0.033
	Cu	4.74 $\pm$ 0.29	4.9	97	0.720
	Mn	15.56 $\pm$ 0.82	17	92	0.403
	Zn	20.28 $\pm$ 1.61	23	88	3.698
GBW07405 soil flour					
	As	419.63 $\pm$ 7.72	412	102	0.148
	Cd	0.50 $\pm$ 0.01	0.45	112	0.003
	Cu	166.35 $\pm$ 2.66	144	116	0.189
	Mn	1624.63 $\pm$ 48.69	1360	119	0.280
	Zn	528.17 $\pm$ 17.42	494	107	1.548
SRM 1640a water					
	As	0.008 $\pm$ 0.000	0.008	102	0.029
	Cd	0.004 $\pm$ 0.000	0.004	101	0.003
<b>ICP-OES</b>					
GBW07405 soil flour					
	Fe	96796 $\pm$ 2272	98000	99	0.09
<b>ED-XRF</b>					
NCS ZC73018 Citrus leaves					

flour		Fe	426 ± 4	480	89
□	□	Si	3749 ± 22	4100	91

**Table S11.** BCR extraction result validation (Mean ± s.d.).

	BCR extraction (mg/kg)	Anti aqua regia (AAR) digestion (mg/kg)	Recovery (%) (BCR/AAR)
<b>CY67 (n=4)</b>			
<b>CT</b>			
As	39.1 ± 0.8	33.5 ± 0.5	117
Cd	4.2 ± 0.6	4.3 ± 0.5	96
Cu	60.0 ± 1.0	60.5 ± 2.9	99
Fe	62002 ± 1409	65414 ± 1949	95
Mn	936.4 ± 64.6	994.5 ± 64.9	94
Zn	222.5 ± 4.6	187.4 ± 3.5	119
<b>FMS</b>			
As	36 ± 1.8	30.5 ± 1.6	118
Cd	2.9 ± 0.6	3.3 ± 0.6	90
Cu	52.8 ± 1.0	51.7 ± 1.6	102
Fe	55586 ± 656	56623 ± 1560	98
Mn	680.7 ± 93.9	764.9 ± 92.4	89
Zn	197.4 ± 5.0	165.3 ± 4.8	119
<b>YY17 (n=4)</b>			
<b>CT</b>			
As	37.9 ± 1.5	32.2 ± 1.0	118
Cd	4.7 ± 0.2	4.6 ± 0.1	101
Cu	60.9 ± 1.3	58.8 ± 2.1	104
Fe	60093 ± 2436	64384 ± 1098	93
Mn	906.5 ± 49.0	935.2 ± 45.0	97
Zn	222.3 ± 4.1	181.8 ± 2.4	122
<b>FMS</b>			
As	35 ± 1.1	30.9 ± 1.5	112
Cd	3.0 ± 0.7	3.2 ± 0.7	93
Cu	50 ± 2.1	52 ± 1.4	96
Fe	52064 ± 240	56470 ± 2044	92
Mn	622.3 ± 77.1	715.6 ± 89.3	87
Zn	187.1 ± 7.0	166.8 ± 6.8	112

**Pakchoi (n=5)****CT**

As	52.50	±	4.07	48.06	±	3.72	109
Cd	9.84	±	0.38	10.59	±	0.40	93
Cu	45.11	±	1.93	48.53	±	2.29	93
Fe	56950	±	2220	61778	±	2288	92
Mn	901	±	19	1079	±	40	84
Zn	806	±	58	758	±	60	106

**FMS**

As	44.80	±	4.37	40.85	±	2.19	110
Cd	8.53	±	0.32	9.20	±	0.40	93
Cu	40.73	±	2.70	43.81	±	1.07	93
Fe	49899	±	2524	55941	±	1052	89
Mn	857	±	30	979	±	25	88
Zn	731	±	65	698	±	38	105

**Table S12.** Plant yield (Mean ± s.d.). Statistical analysis was performed using a GLM. Post hoc analysis was carried out using Tukey Pairwise Comparisons. Mean ± s.d. that does not share a letter are significantly different.

	Plant yield (g/pot)											
	CT		FMS		MS		BC					
CY67 (d. wt.)												
Grain	4.2	± 0.5	c	6.0	± 0.4	b	8.4	± 0.8	a	5.7	± 0.4	b
Husk	2.0	± 0.6	b	3.1	± 0.2	a	3.6	± 0.3	a	2.3	± 0.5	b
Leaf	8.4	± 0.9	ab	8.8	± 0.5	a	9.0	± 0.5	a	7.1	± 0.7	b
Straw	17.6	± 0.9	a	17.7	± 1.1	a	18.4	± 0.7	a	17.5	± 0.7	a
Root	9.8	± 2.0	a	9.2	± 1.7	a	12.5	± 1.6	a	9.7	± 1.3	a
Total	42.0	± 2.2	b	44.8	± 1.5	b	51.9	± 1.9	a	42.3	± 2.4	b
YY17 (d. wt.)												
Grain	5.2	± 0.4	b	5.2	± 0.3	b	8.7	± 0.4	a	5.2	± 0.3	b
Husk	2.7	± 0.2	b	2.6	± 0.1	b	4.3	± 0.2	a	2.3	± 0.4	b
Leaf	8.3	± 0.8	a	8.4	± 0.5	a	9.0	± 0.6	a	7.5	± 0.9	a
Straw	17.4	± 0.9	a	17.4	± 1.0	a	17.6	± 1.7	a	16.4	± 0.7	a
Root	11.3	± 0.9	a	10.5	± 0.7	a	9.6	± 0.6	a	9.4	± 1.8	a
Total	44.8	± 0.7	b	44.2	± 1.2	b	49.2	± 1.6	a	40.8	± 1.1	c
Pakchoi (w. wt.)	31.6	± 2.5	a	33.1	± 2.6	a	30.7	± 4.6	a	29.6	± 3.6	a

**Table S13.** Translocation factor (TF) of Cd in rice tissues (Data calculated based on Log10 of Cd concentration) (Mean  $\pm$  s.d. n=4). (ANOVA, one-way, Turkey's Multiple).

	TF <sub>(Grain/straw)</sub>				TF <sub>(Leaf/straw)</sub>				TF <sub>(Straw/root)</sub>			
<b>CY67</b>												
CT	0.88	$\pm$	0.03	a	0.92	$\pm$	0.04	a	0.79	$\pm$	0.07	a
BC	0.87	$\pm$	0.05	a	0.92	$\pm$	0.02	a	0.79	$\pm$	0.03	a
MS	0.88	$\pm$	0.06	a	0.91	$\pm$	0.06	a	0.68	$\pm$	0.02	b
FMS	0.89	$\pm$	0.14	a	0.95	$\pm$	0.03	a	0.64	$\pm$	0.05	b
<b>YY17</b>												
CT	0.91	$\pm$	0.05	a	0.87	$\pm$	0.03	a	0.82	$\pm$	0.03	a
BC	0.87	$\pm$	0.02	a	0.88	$\pm$	0.03	a	0.82	$\pm$	0.04	a
MS	0.93	$\pm$	0.03	a	0.93	$\pm$	0.04	a	0.70	$\pm$	0.02	b
FMS	0.95	$\pm$	0.12	a	0.92	$\pm$	0.04	a	0.70	$\pm$	0.03	b

TF is calculated as Cd concentration in tissue 1 divided by the corresponding value in tissue 2 (Wang et al., 2015)<sup>14</sup>:  $TF = C_{\text{tissue1}}/C_{\text{tissue2}}$

**Table S14.** As offtake in plant (Mean  $\pm$  s.d.). Statistical analysis was performed using a GLM. Post hoc analysis was carried out using Tukey Pairwise Comparisons. Mean  $\pm$  s.d. that does not share a letter are significantly different.

	CT ( $\mu\text{g}/\text{pot}$ )				FMS ( $\mu\text{g}/\text{pot}$ )				MS ( $\mu\text{g}/\text{pot}$ )				BC ( $\mu\text{g}/\text{pot}$ )			
<b>CY67</b>																
	147	$\pm$	23	bc	97	$\pm$	40	c	219	$\pm$	17	a	178	$\pm$	25	ab
<b>YY17</b>																
	147	$\pm$	11	a	106	$\pm$	16	a	112	$\pm$	22	a	114	$\pm$	29	a
<b>Pakchoi</b>																
	0.7	$\pm$	0.4	b	1.1	$\pm$	0.6	ab	0.6	$\pm$	0.3	b	1.9	$\pm$	0.7	a

**Table S15.** Elements in the rice root IP (n=4) (Mean  $\pm$  s.d.). Statistical analysis was performed using a GLM. Post hoc analysis was carried out using Tukey Pairwise Comparisons. Mean  $\pm$  s.d. that does not share a letter are significantly different.

	CT					FMS			
<b>CY67</b>									
<b>As</b> ( $\mu\text{g/kg}$ )	49.8	$\pm$	6.74	a		28.2	$\pm$	9.92	b
<b>Cd</b> ( $\mu\text{g/kg}$ )	1.52	$\pm$	0.1	a		0.32	$\pm$	0.13	b
<b>Fe</b> ( $\text{mg/kg}$ )	21.2	$\pm$	5.37	a		50.9	$\pm$	10.7	b
<b>Mn</b> ( $\text{mg/kg}$ )	2.38	$\pm$	0.2	a		3.86	$\pm$	0.61	b
<b>YY17</b>									
<b>As</b> ( $\mu\text{g/kg}$ )	47.9	$\pm$	5.2	a		41.0	$\pm$	11.4	a
<b>Cd</b> ( $\mu\text{g/kg}$ )	2.11	$\pm$	0.37	a		1.22	$\pm$	1.15	a
<b>Fe</b> ( $\text{mg/kg}$ )	18.3	$\pm$	4.42	a		49.9	$\pm$	12.8	b
<b>Mn</b> ( $\text{mg/kg}$ )	3.28	$\pm$	0.33	a		2.53	$\pm$	1.85	a

**Table S16.** Chenzhou soil DGT characteristics.  $C_{\text{sol}}$  and  $C_{\text{DGT}}$  are shown as mean  $\pm$  s.d.,  $R_{\text{diff}}$ ,  $R$ , and  $C_E$  shown as mean ( $n=4$ ). MBL-DGT method detection limits (MDL) were reported by Panther et al. (2014).  $^{\text{As}}\text{MDL}$ : 0.02  $\mu\text{g/L}$ ,  $^{\text{Cd}}\text{MDL}$ : 0.04  $\mu\text{g/L}$ .

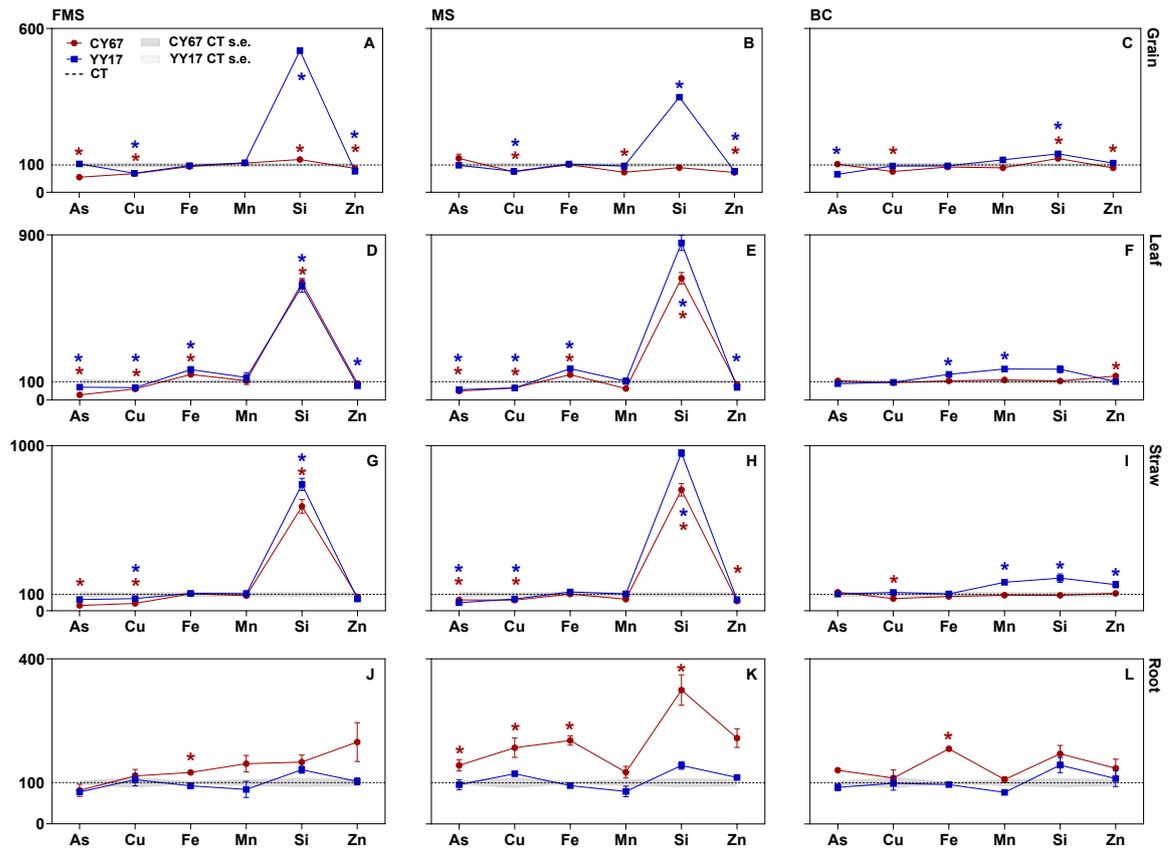
	$C_{sol}$ ( $\mu\text{g/L}$ )			$C_{DGT}$ ( $\mu\text{g/L}$ )			$R_{diff}$	$R$	$C_E$ ( $\mu\text{g/L}$ )
<b>As</b>									
<b>CZ-L</b>									
<i>Dose (wt%)</i>									
0%	231	$\pm$ 43	89	$\pm$ 9.5	0.061	0.39	1466		
0.1%	342	$\pm$ 72	87	$\pm$ 8.9	0.061	0.25	1418		
0.5%	134	$\pm$ 18	38	$\pm$ 3.8	0.061	0.28	617		
1.0%	60	$\pm$ 4	16	$\pm$ 3.0	0.061	0.27	270		
<i>Time (weeks)</i>									
2	134	$\pm$ 18	38	$\pm$ 3.8	0.061	0.28	617		
4	78	$\pm$ 20	25	$\pm$ 11.5	0.061	0.32	412		
8	67	$\pm$ 58	17	$\pm$ 12.9	0.061	0.25	279		
<b>CZ-S</b>									
<i>Dose (wt%)</i>									
0%	175	$\pm$ 52	98	$\pm$ 10.3	0.060	0.56	1640		
0.1%	210	$\pm$ 64	101	$\pm$ 21.2	0.060	0.48	1687		
0.5%	174	$\pm$ 31	39	$\pm$ 3.0	0.060	0.22	656		
1.0%	75	$\pm$ 18	18	$\pm$ 2.4	0.060	0.23	294		
<b>Cd</b>									
<b>CZ-L</b>									
<i>Dose (wt%)</i>									
0%	0.041	$\pm$ 0.009	0.017	$\pm$ 0.001	0.075	0.42	0.227		
0.1%	0.074	$\pm$ 0.021	0.017	$\pm$ 0.003	0.075	0.23	0.224		
0.5%	0.074	$\pm$ 0.045	0.017	$\pm$ 0.002	0.075	0.23	0.224		
1.0%	0.057	$\pm$ 0.011	0.017	$\pm$ 0.001	0.075	0.30	0.229		
<i>Time (weeks)</i>									
2	0.074	$\pm$ 0.045	0.017	$\pm$ 0.002	0.075	0.23	0.224		
4	0.255	$\pm$ 0.094	0.021	$\pm$ 0.003	0.075	0.08	0.282		
8	0.275	$\pm$ 0.146	0.038	$\pm$ 0.024	0.075	0.14	0.512		
<b>CZ-S</b>									
<i>Dose (wt%)</i>									
0%	0.043	$\pm$ 0.012	0.015	$\pm$ 0.002	0.073	0.34	0.200		
0.1%	0.051	$\pm$ 0.014	0.020	$\pm$ 0.011	0.073	0.40	0.277		
0.5%	0.077	$\pm$ 0.007	0.019	$\pm$ 0.002	0.073	0.25	0.258		
1.0%	0.071	$\pm$ 0.034	0.019	$\pm$ 0.001	0.074	0.27	0.257		

**Table S17.** Dabaoshan soil and Gaoming soil  $^{As}DGT$  characteristics.  $C_{sol}$  and  $C_{DGT}$  are shown as mean  $\pm$  s.d.,  $R_{diff}$ ,  $R$ , and  $C_E$  shown as mean ( $n=3$ ).  $^{As}MDL$ : 0.02  $\mu\text{g/L}$ .

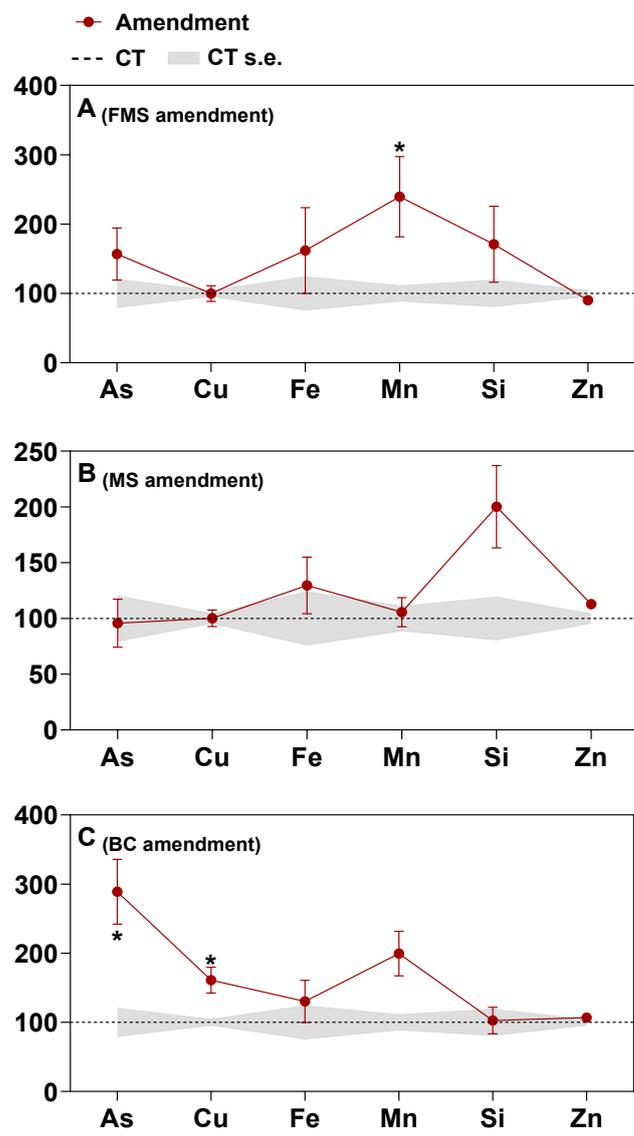
	$^{As}C_{sol}$ ( $\mu\text{g/L}$ )	$^{As}C_{DGT}$ ( $\mu\text{g/L}$ )	$^{As}R_{diff}$	$^{As}R$	$^{As}C_E$
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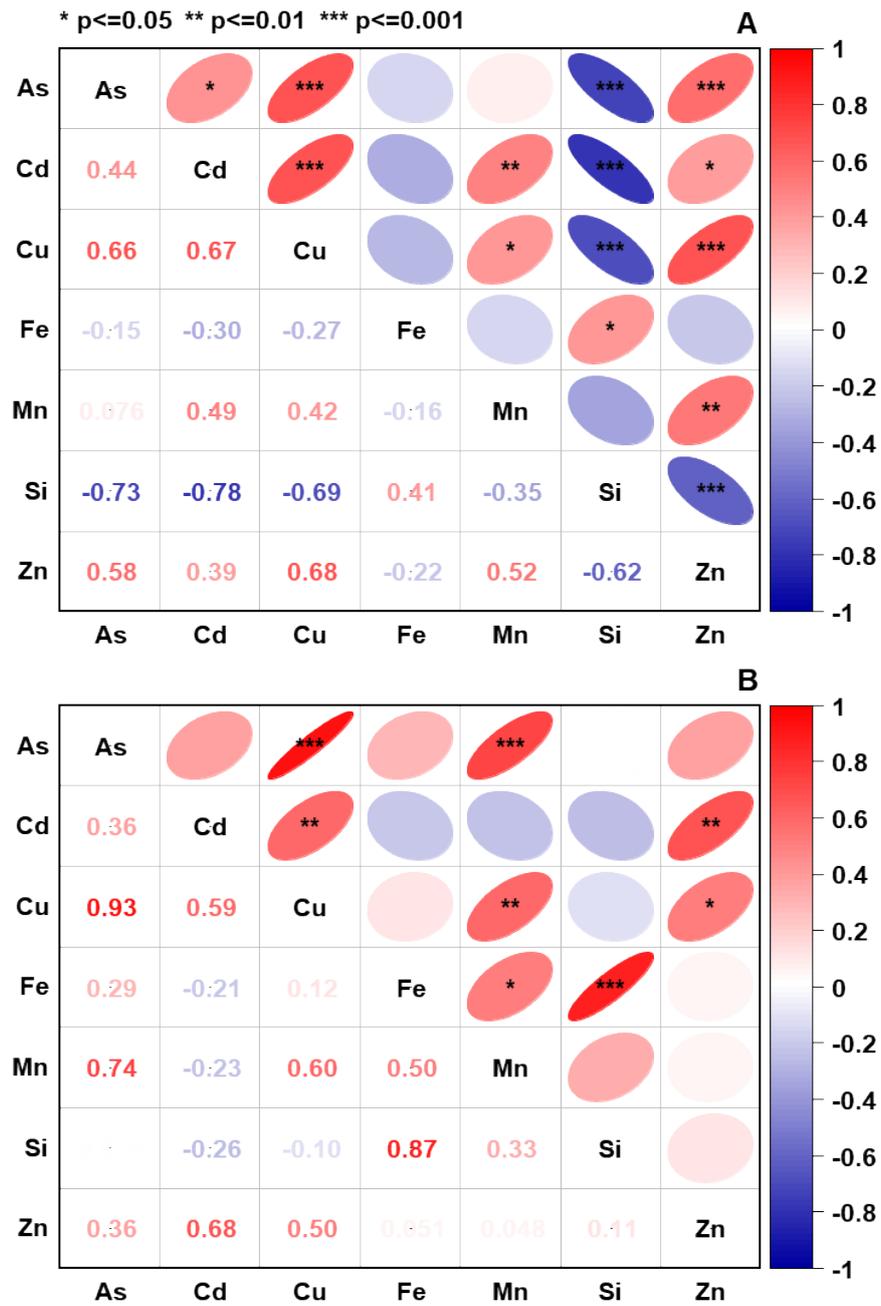
<b>GD (b)</b>													
CT	1.324	±	0.551	□	0.108	±	0.017	□	0.071	□	0.08	□	1.53
FMS	1.067	±	0.269	□	0.094	±	0.007	□	0.071	□	0.09	□	1.33
<b>GD (c)</b>													
CT	5.188	±	1.000	□	3.432	±	1.053	□	0.071	□	0.66	□	48.13
FMS	0.511	±	0.036	□	0.058	±	0.013	□	0.072	□	0.11	□	0.81
<b>Gaoming</b>													
<b>GG (a)</b>													
CT	0.169	±	0.108	□	0.014	±	0.005	□	0.074	□	0.08	□	0.19
FMS	0.398	±	0.066	□	0.020	±	0.003	□	0.073	□	0.05	□	0.27
<b>GG (b)</b>													
CT	0.770	±	0.086	□	0.016	±	0.001	□	0.067	□	0.02	□	0.24
FMS	0.477	±	0.160	□	0.013	±	0.002	□	0.067	□	0.03	□	0.20
<b>GG (c)</b>													
CT	0.574	±	0.230	□	0.020	±	0.005	□	0.069	□	0.04	□	0.29
FMS	0.401	±	0.046	□	0.018	±	0.003	□	0.068	□	0.05	□	0.27
<b>GG (d)</b>													
CT	0.365	±	0.191	□	0.012	±	0.002	□	0.068	□	0.03	□	0.18
FMS	0.499	±	0.107	□	0.014	±	0.002	□	0.067	□	0.03	□	0.21



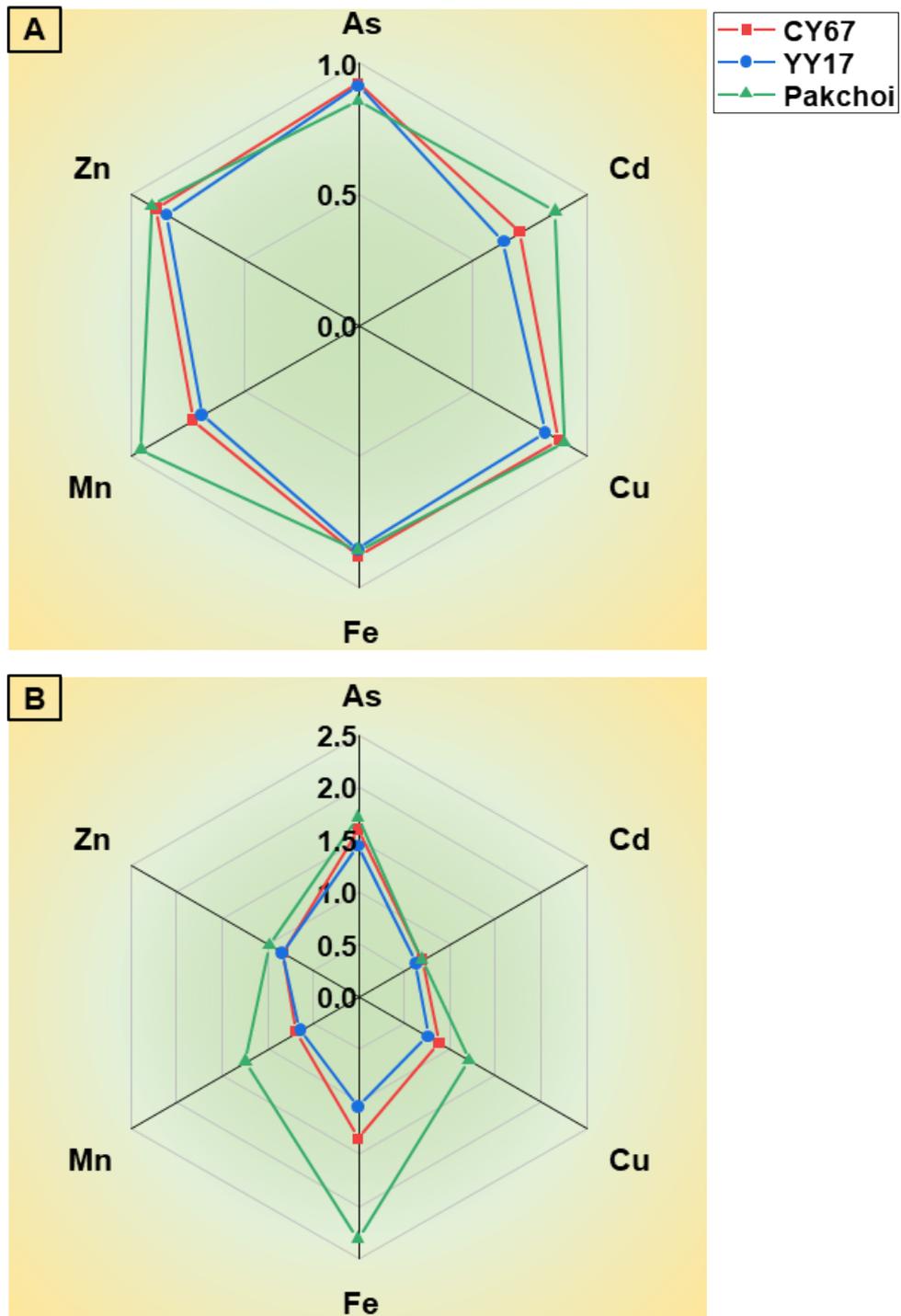
**Fig. S1.** Element concentration in different amendment group rice tissues expressed as a percentage relative to the mean of the control group for each element. (Mean  $\pm$  s.e. n=4). (GLM, Dunnett Multiple/Compared with CT). \* Indicated significant differences between treatment and control (P<0.05).



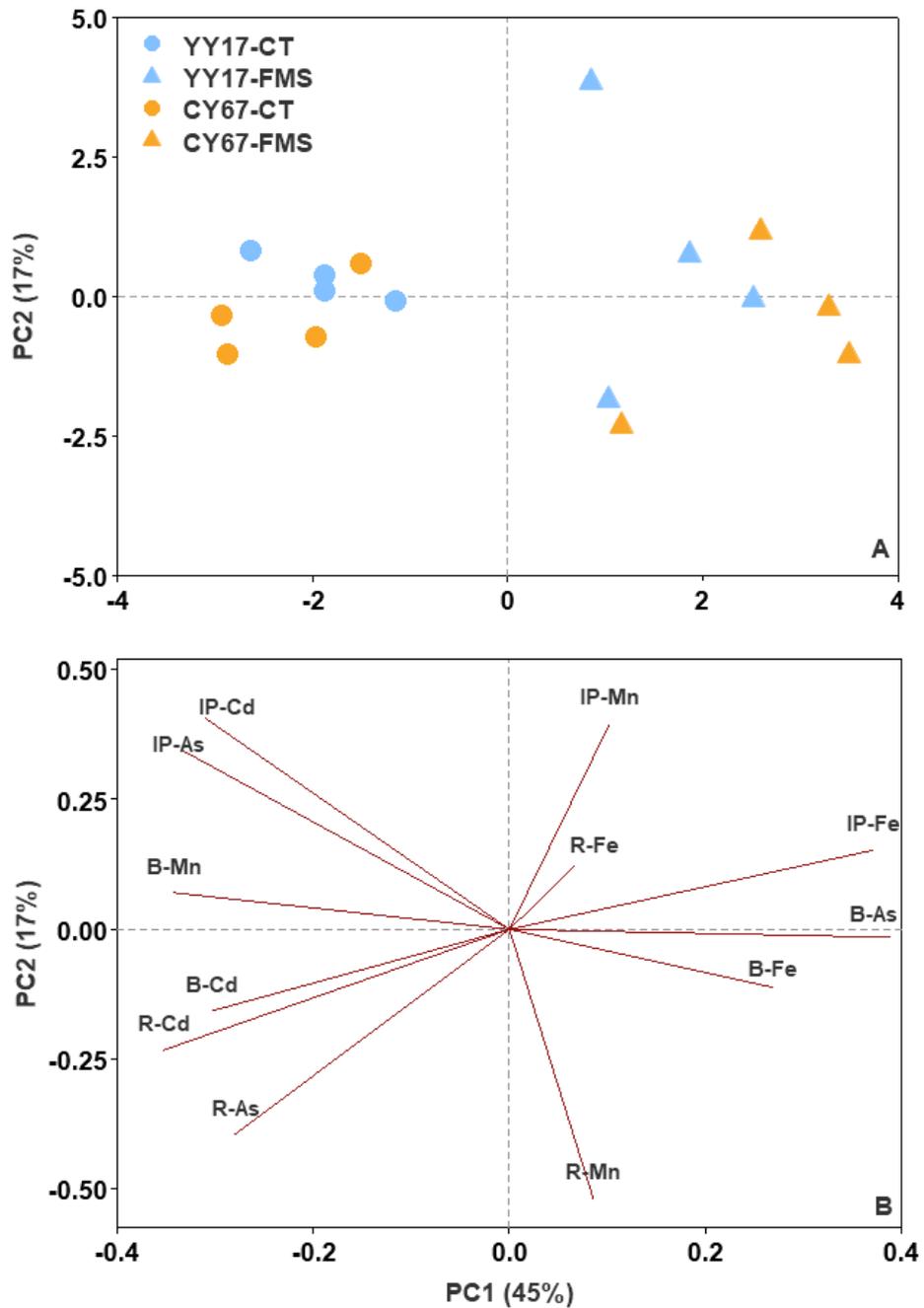
**Fig. S2.** Element concentration in different amendment group pakchoi expressed as a percentage relative to the mean of the control group for each element (Mean  $\pm$  s.e. n=5). (GLM, Dunnett Multiple/Compared with CT).



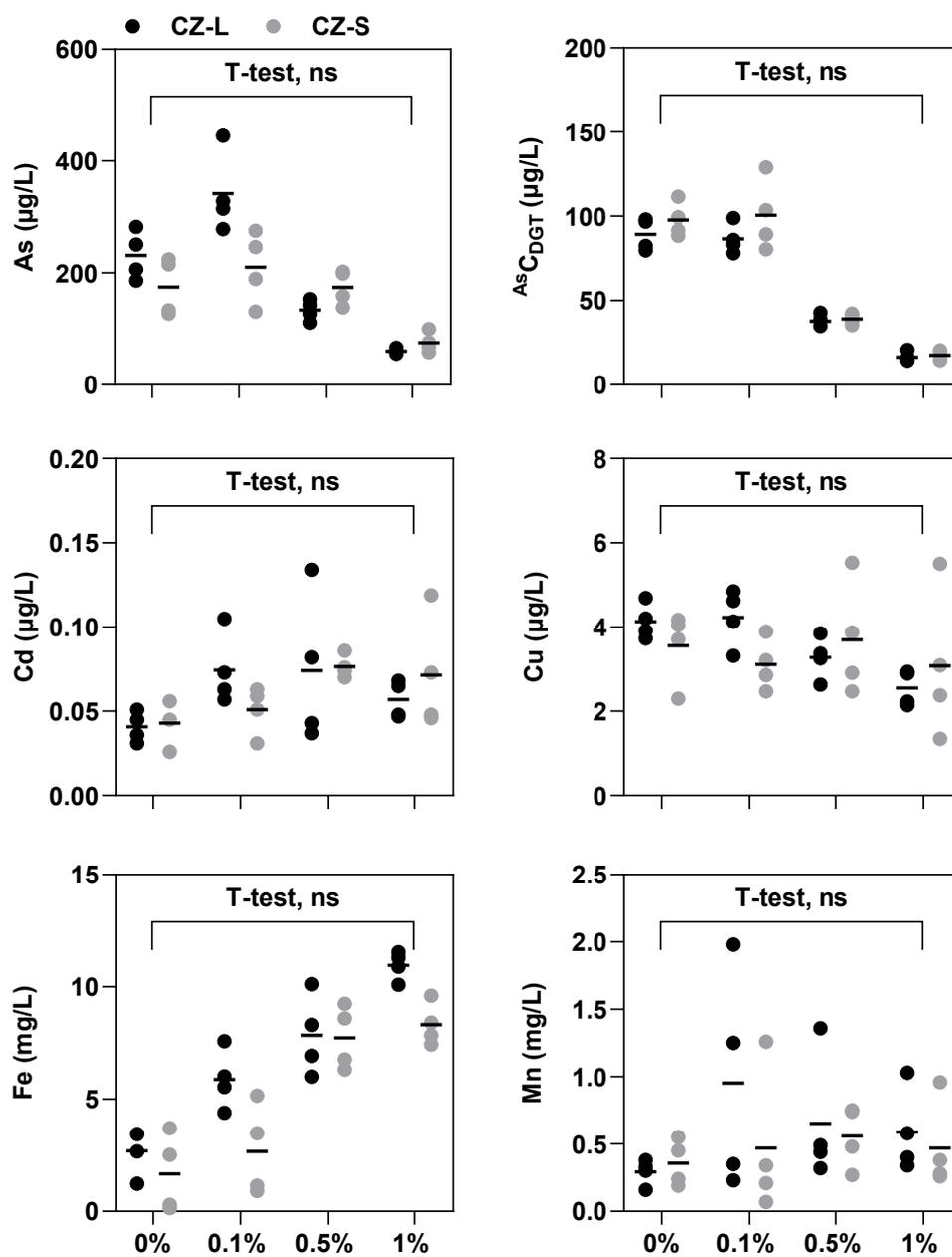
**Fig. S3.** Pearson's correlation coefficients for plant multi-element concentrations (A: rice straw. B: pakchoi.).



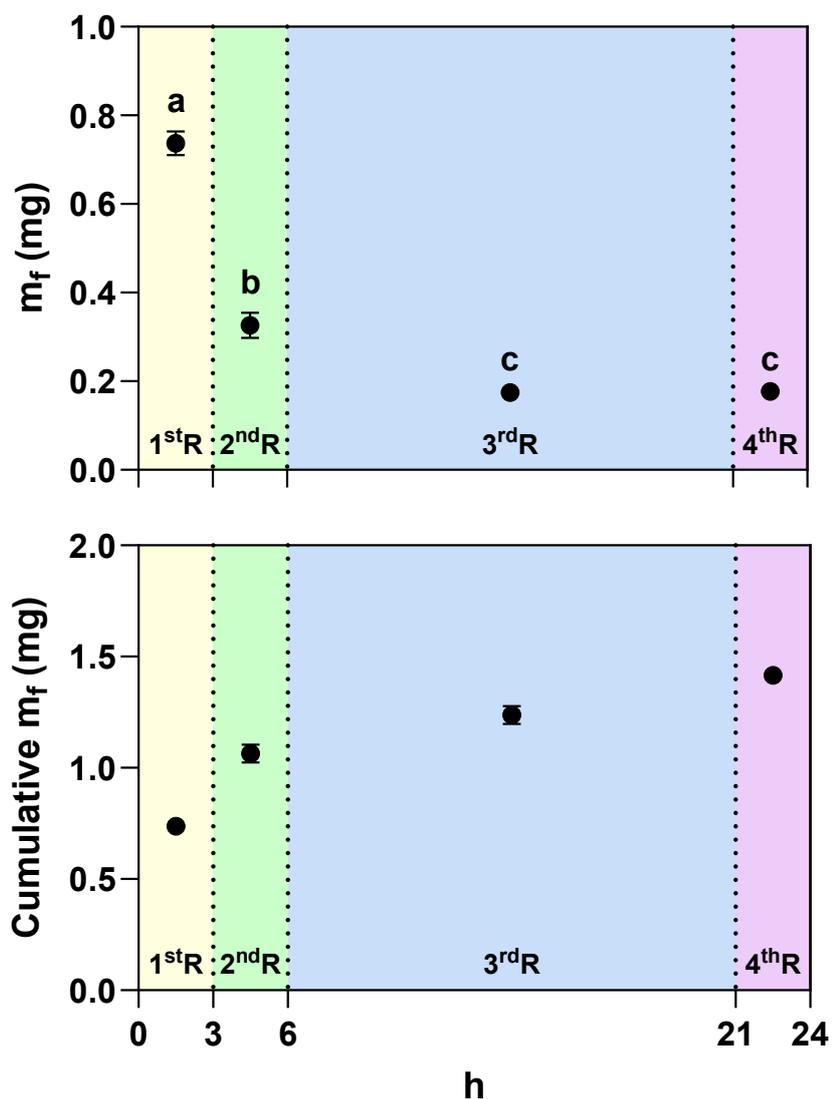
**Fig. S4.** Element concentration in FMS-amended soils is expressed as a ratio relative to each element's mean of the control group. A: BCR extracted. B: bioavailability.



**Fig. S5.** Rice rhizosphere elements PCA. A: Score plot. B: Loading plot. (R-: element concentration in rice root. IP-: element concentration in iron plaque. B-: element bioavailability in soil.)



**Fig. S6.** FMS performance in different scale soil (150g vs 50g). (T-test [paired] was conducted between CZ-L and CZ-S).



**Fig. S7.** Release behavior of SA spiked FMS (Mean  $\pm$  s.e.) (n=3). 1<sup>st</sup> R: First sampling. 2<sup>nd</sup> R: Second sampling. 3<sup>rd</sup> R: Third sampling. 4<sup>th</sup> R: Fourth sampling. Symbols that do not share a letter are significantly different.

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