

## Supporting Information

### **MnFe<sub>2</sub>O<sub>4</sub>-loaded bamboo pulp carbon-based aerogel composite: synthesis, characterization and adsorption behavior study for heavy metals removal**

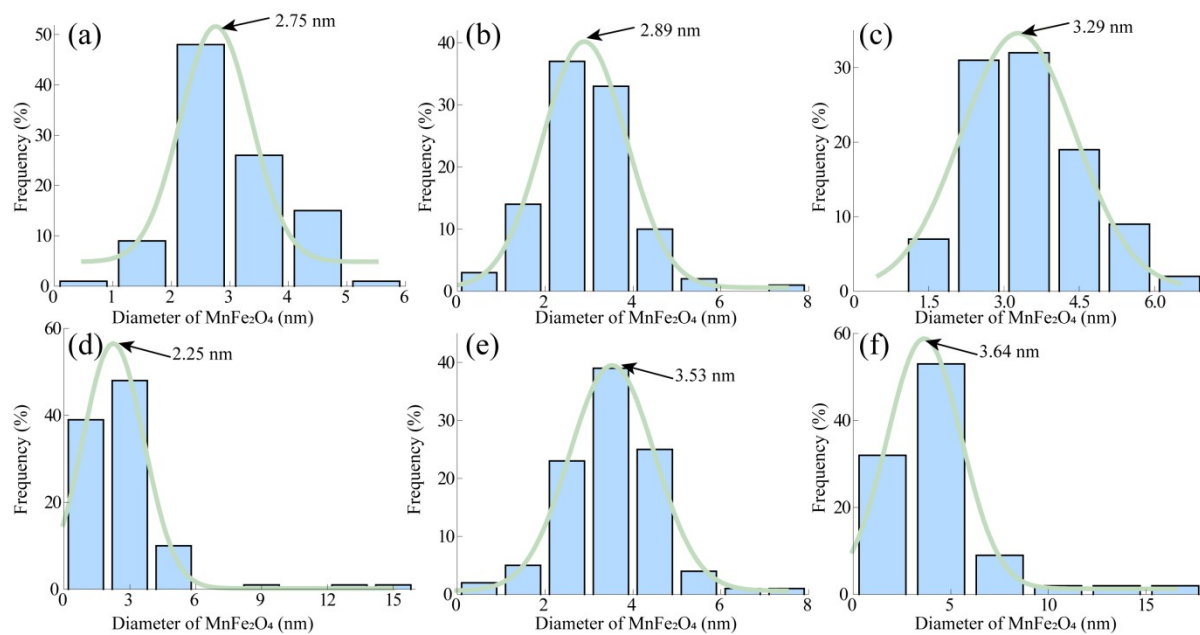
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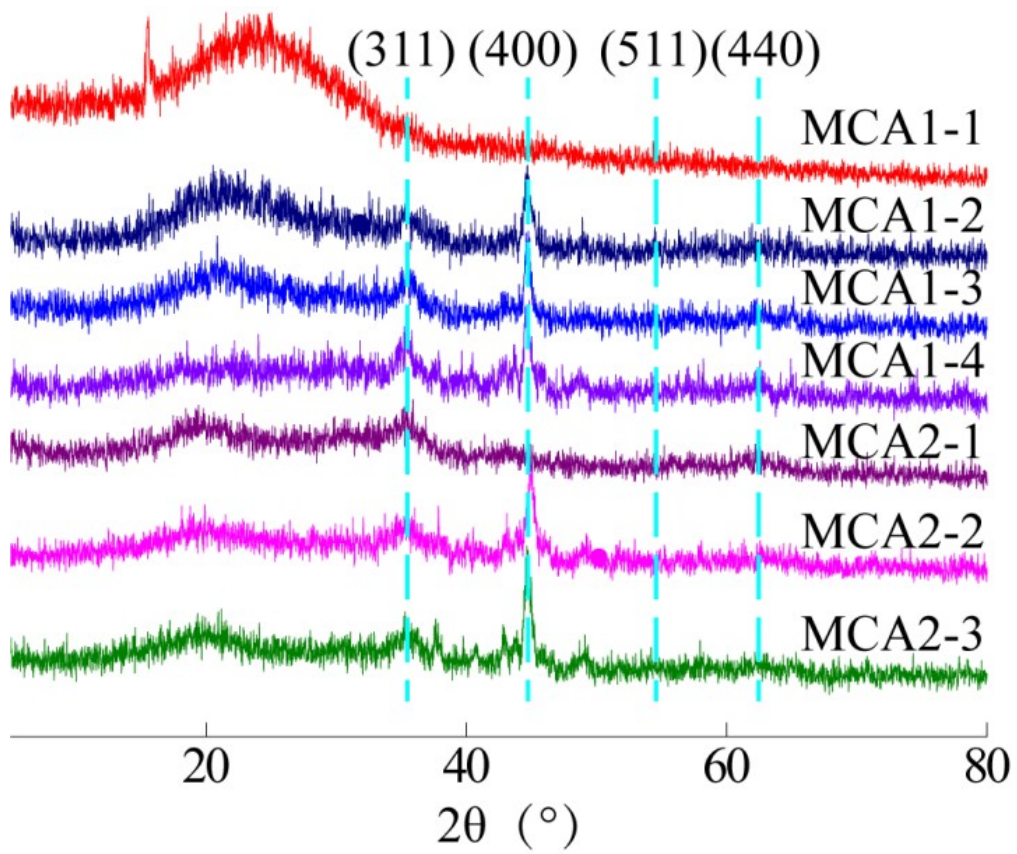
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**Fig. S1** The particle size distribution of MnFe<sub>2</sub>O<sub>4</sub> on the surface of MCA obtained by directional freezing: (a) MCA1-2; (b) MCA1-3; (c) MCA1-4; (d) MCA2-1; (e) MCA2-2; (f) MCA2-3



**Fig. S2** XRD characterization of MCA

**Table S1** Specific surface area, pore volume and porosity of MCA with different amounts of modifiers

No.	$S_{\text{BET}}$ /m <sup>2</sup> ·g <sup>-1</sup>	$D_{\text{pore}}$ /nm	$V_{\text{total pore}}$ /cm <sup>3</sup> ·g <sup>-1</sup>	$V_{<50\text{nm}}$ /cm <sup>3</sup> ·g <sup>-1</sup>	$V_{<50\text{ nm}}/V_{\text{total pore}}$	$\rho/\%$
MCA1-1	182.67	4.077	0.1772	0.1447	0.8166	95.33
MCA1-2	128.79	5.890	0.1515	0.1025	0.6766	90.51
MCA1-3	112.09	5.122	0.1358	0.1186	0.8733	80.88
MCA1-4	109.72	4.897	0.1085	0.0838	0.7724	66.18

$\rho_0$  is the density of graphite, 2.3 g/cm<sup>3</sup>

Pseudo-first-order kinetic equation (Equation S1) and pseudo-second-order kinetic equation (Equation S2) are as follows:

$$q_t = q_e(1 - e^{-k_1 t}) \quad (\text{Equation S1})$$

$$q_t = k_2 q_e^2 t / (1 + k_2 q_e t) \quad (\text{Equation S2})$$

Where  $q_e$  and  $q_t$  are the amounts of  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ , or  $\text{Cd}^{2+}$  adsorbed at equilibrium and  $t$  min, respectively, mg/g;  $K_1$  is the rate constant of the pseudo-first-order kinetic model,  $\text{min}^{-1}$ ;  $K_2$  is the rate constant of the pseudo-second-order kinetic model,  $\text{g}/(\text{mg} \cdot \text{min})$ ;  $t$  is the adsorption time, min.

**Table S2** MCA adsorption kinetics fitting parameters for  $\text{Pb}^{2+}$  in single, binary, and ternary systems

adsorbates	concentration (mg/L)	Pseudo-first-order model			Pseudo-second-order model		
		$K_1$ ( $\text{min}^{-1}$ )	$q_e$ (mg/g)	$R^2$	$K_2$ (g/mg·min)	$q_e$ (mg/g)	$R^2$
$\text{Pb}^{2+}$	20	0.0253	16.74	0.985	0.0014	19.91	0.974
	40	0.0346	29.24	0.997	0.0013	33.34	0.994
	60	0.0319	38.88	0.957	0.0010	43.83	0.971
$\text{Pb}^{2+}/\text{Cu}^{2+}$	20	0.0261	9.55	0.981	0.0027	11.24	0.966
	40	0.0301	16.87	0.988	0.0020	19.41	0.992
	60	0.0383	18.93	0.994	0.0022	21.55	0.974
$\text{Pb}^{2+}/\text{Cd}^{2+}$	20	0.0381	12.10	0.950	0.0035	13.73	0.916
	40	0.0474	19.88	0.981	0.0029	22.20	0.997
	60	0.0249	25.59	0.987	0.0010	30.15	0.983
$\text{Pb}^{2+}/\text{Cu}^{2+}/\text{Cd}^{2+}$	20	0.0266	7.12	0.972	0.0039	8.35	0.986
	40	0.0188	13.74	0.979	0.0011	17.27	0.966
	60	0.0268	16.53	0.993	0.0015	19.73	0.979

**Table S3** MCA adsorption kinetics fitting parameters for Cu<sup>2+</sup> in single, binary, and ternary systems

adsorbates	concentration (mg/L)	Pseudo-first-order model			Pseudo-second-order model		
		K <sub>1</sub> (min <sup>-1</sup> )	q <sub>e</sub> (mg/g)	R <sup>2</sup>	K <sub>2</sub> (g/mg·min)	q <sub>e</sub> (mg/g)	R <sup>2</sup>
Cu <sup>2+</sup>	20	0.0196	22.29	0.981	0.0007	28.09	0.963
	40	0.0304	32.97	0.969	0.0009	38.89	0.937
	60	0.0438	37.46	0.985	0.0016	41.40	0.973
Cu <sup>2+</sup> /Pb <sup>2+</sup>	20	0.0363	11.70	0.935	0.0035	13.32	0.903
	40	0.0574	16.53	0.995	0.0048	18.05	0.988
	60	0.0402	20.44	0.950	0.0023	23.05	0.983
Cu <sup>2+</sup> /Cd <sup>2+</sup>	20	0.0421	16.13	0.962	0.0032	18.03	0.927
	40	0.0170	24.38	0.975	0.0005	31.16	0.961
	60	0.0250	30.47	0.971	0.0007	36.67	0.945
Cu <sup>2+</sup> /Pb <sup>2+</sup> /Cd <sup>2+</sup> +	20	0.0398	8.31	0.963	0.0059	9.31	0.986
	40	0.0247	14.70	0.989	0.0017	17.41	0.986
	60	0.0201	20.02	0.997	0.0009	24.60	0.994

**Table S4** MCA adsorption kinetics fitting parameters for Cd<sup>2+</sup> in single, binary, and ternary systems

adsorbates	concentration (mg/L)	Pseudo-first-order model			Pseudo-second-order model		
		K <sub>1</sub> (min <sup>-1</sup> )	q <sub>e</sub> (mg/g)	R <sup>2</sup>	K <sub>2</sub> (g/mg·min)	q <sub>e</sub> (mg/g)	R <sup>2</sup>
Cd <sup>2+</sup>	20	0.0167	15.28	0.920	0.0007	20.11	0.892
	40	0.0127	29.46	0.972	0.0003	40.68	0.961
	60	0.0199	36.52	0.983	0.0004	45.61	0.969
Cd <sup>2+</sup> /Pb <sup>2+</sup>	20	0.0214	11.95	0.966	0.0015	14.74	0.943
	40	0.0234	18.03	0.998	0.0012	21.77	0.987
	60	0.0384	18.73	0.964	0.0021	21.58	0.938
Cd <sup>2+</sup> /Cu <sup>2+</sup>	20	0.0284	7.63	0.913	0.0045	8.69	0.955
	40	0.0228	12.69	0.990	0.0016	15.30	0.977
	60	0.0188	17.03	0.974	0.0010	20.82	0.983
Cd <sup>2+</sup> /Pb <sup>2+</sup> /Cu <sup>2+</sup>	20	0.0137	6.09	0.930	0.0017	7.82	0.941
	40	0.0195	9.63	0.988	0.0016	12.06	0.983
	60	0.0295	12.83	0.989	0.0024	14.93	0.992



Langmuir isotherm model (Equation S3) and Freundlich isotherm model (Equation S4)

are as follows:

$$q_e = q_m K_L C_e / (1 + b C_e) \quad (\text{Equation S3})$$

$$q_e = K_F C_e^{1/n} \quad (\text{Equation S4})$$

Where  $q_e$  is the amount of  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ , or  $\text{Cd}^{2+}$  adsorbed by MCA at equilibrium and  $q_m$  is saturation capacity of MCA for  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$ , or  $\text{Cd}^{2+}$ , mg/g;  $C_e$  is the concentration of  $\text{Pb}^{2+}$ ,  $\text{Cu}^{2+}$  and  $\text{Cd}^{2+}$  in the solution at equilibrium,  $\mu\text{g/mL}$ ;  $K_L$  is the adsorption equilibrium constant of Langmuir isothermal model, L/mg;  $K_F$  is the adsorption equilibrium constant of Freundlich isothermal model,  $\text{mg} \cdot \text{g}^{-1} \cdot (\text{L} \cdot \text{mg}^{-1})^{1/n}$ ;  $1/n$  is the adsorption intensity.

**Table S5** MCA adsorption isotherm curve fitting parameters for  $\text{Pb}^{2+}$  in single, binary, and ternary systems

adsorbates	temperature (°C)	Langmuir isotherm model			Freundlich isotherm model		
		$q_m$ (mg/g)	$K_L$	$R^2$	n	$K_F$	$R^2$
$\text{Pb}^{2+}$	25	53.18	0.0568	0.997	2.037	6.02	0.983
	35	61.70	0.0771	0.999	2.034	8.07	0.970
	45	74.38	0.0982	0.993	1.971	10.55	0.947
$\text{Pb}^{2+}/\text{Cu}^{2+}$	25	30.47	0.0444	0.946	2.064	3.20	0.990
	35	34.31	0.0055	0.975	2.167	4.26	0.991
	45	37.01	0.0751	0.963	2.384	5.98	0.942
$\text{Pb}^{2+}/\text{Cd}^{2+}$	25	38.91	0.0352	0.984	1.805	2.90	0.951
	35	44.37	0.0434	0.982	1.916	4.07	0.947
	45	55.26	0.0466	0.995	1.887	5.31	0.968
$\text{Pb}^{2+}/\text{Cu}^{2+}/\text{Cd}^{2+}$	25	24.68	0.0353	0.886	1.930	2.08	0.943
	35	32.30	0.0340	0.922	1.837	2.45	0.973
	45	37.49	0.0372	0.995	1.788	2.83	0.966

**Table S6** MCA adsorption isotherm curve fitting parameters for  $\text{Cu}^{2+}$  in single, binary, and ternary systems

adsorbates	temperature (°C)	Langmuir isotherm model			Freundlich isotherm model		
		$q_m$ (mg/g)	$K_L$	$R^2$	n	$K_F$	$R^2$
$\text{Cu}^{2+}$	25	44.54	0.1402	0.978	2.668	9.95	0.990
	35	63.86	0.1103	0.957	2.291	11.21	0.999
	45	84.21	0.1014	0.933	2.160	13.47	0.996
$\text{Cu}^{2+}/\text{Pb}^{2+}$	25	34.54	0.0249	0.910	1.651	1.80	0.853
	35	53.18	0.0206	0.957	1.484	1.98	0.926
	45	67.07	0.0226	0.963	1.489	2.67	0.940
$\text{Cu}^{2+}/\text{Cd}^{2+}$	25	44.57	0.0550	0.985	1.950	4.73	0.942
	35	49.26	0.0834	0.992	2.071	6.95	0.948
	45	52.63	0.1317	0.971	2.239	9.97	0.898
$\text{Cu}^{2+}/\text{Pb}^{2+}/\text{Cd}^{2+}$	25	28.56	0.0390	0.988	2.005	2.67	0.956
	35	32.50	0.0447	0.978	2.008	3.26	0.928
	45	36.14	0.0574	0.949	2.128	4.43	0.875

**Table S7** MCA adsorption isotherm curve fitting parameters for Cd<sup>2+</sup> in single, binary, and ternary systems

adsorbates	temperature (°C)	Langmuir isotherm model			Freundlich isotherm model		
		q <sub>m</sub> (mg/g)	K <sub>L</sub>	R <sup>2</sup>	n	K <sub>F</sub>	R <sup>2</sup>
Cd <sup>2+</sup>	25	61.05	0.0338	0.953	1.692	4.00	0.977
	35	64.01	0.0450	0.959	1.788	5.39	0.971
	45	73.63	0.0509	0.941	1.753	6.47	0.956
Cd <sup>2+</sup> /Pb <sup>2+</sup>	25	20.20	0.3478	0.878	4.421	8.21	0.977
	35	24.12	0.4406	0.923	4.328	10.22	0.988
	45	26.47	0.6602	0.926	4.707	12.77	0.982
Cd <sup>2+</sup> /Cu <sup>2+</sup>	25	31.02	0.0187	0.993	1.496	1.10	0.981
	35	37.46	0.0212	0.991	1.458	1.37	0.980
	45	55.35	0.0158	0.989	1.307	1.31	0.980
Cd <sup>2+</sup> /Pb <sup>2+</sup> /Cu <sup>2+</sup>	25	30.71	0.0252	0.984	1.417	0.84	0.977
	35	31.00	0.1049	0.991	1.509	1.37	0.965
	45	36.29	0.0331	0.974	1.501	1.92	0.947

The thermodynamic parameters of the adsorption process are calculated by Equations (S5)-(S8).

$$\ln K = \frac{\Delta S^\theta}{R} - \frac{\Delta H^\theta}{RT} \quad \text{(Equation S5)}$$

$$\Delta G^\theta = -RT \ln K \quad \text{(Equation S6)}$$

$$\Delta G^\theta = \Delta H^\theta - T \Delta S^\theta \quad \text{(Equation S7)}$$

$$K_d = \frac{q_e}{C_e} \quad \text{(Equation S8)}$$

Where R is the ideal gas constant, 8.314 J/(mol • K); T is the absolute temperature, K;  $\Delta G^\theta$  is the change of Gibbs free energy, J/K;  $\Delta H^\theta$  is the change of enthalpy, J/K;  $\Delta S^\theta$  is change of entropy, J/(mol • K); K is the equilibrium constant, (L/mol);  $q_e$  is the amount of  $Pb^{2+}$ ,  $Cu^{2+}$  or  $Cd^{2+}$  in the MCA at equilibrium;  $C_e$  is the concentration of  $Pb^{2+}$ ,  $Cu^{2+}$  or  $Cd^{2+}$  in the solution at equilibrium, mg/L.

**Table S8** Thermodynamic parameters of Pb<sup>2+</sup>adsorption by MCA in single, binary, and ternary systems

adsorbates	$\Delta G^\theta$ (kJ/mol)			$\Delta H^\theta$ (kJ/mol)	$\Delta S^\theta$ (kJ/mol·K)
	25°C	35 °C	45 °C		
Pb <sup>2+</sup>	-1.152	-2.324	-3.589	35.162	0.122
Pb <sup>2+</sup> /Cu <sup>2+</sup>	-0.311	-0.354	-1.001	20.551	0.066
Pb <sup>2+</sup> /Cd <sup>2+</sup>	-0.072	-0.656	-0.879	23.517	0.077
Pb <sup>2+</sup> /Cu <sup>2+</sup> /Cd <sup>2+</sup>	-0.631	-1.093	-1.770	18.782	0.057

**Table S9** Thermodynamic parameters of Cu<sup>2+</sup>adsorption by MCA in single, binary, and ternary systems

adsorbates	$\Delta G^\theta$ (kJ/mol)			$\Delta H^\theta$ (kJ/mol)	$\Delta S^\theta$ (kJ/mol·K)
	25°C	35 °C	45 °C		
Cu <sup>2+</sup>	-1.958	-2.906	-3.829	25.933	0.094
Cu <sup>2+</sup> /Pb <sup>2+</sup>	-0.040	-0.819	-1.656	26.941	0.085
Cu <sup>2+</sup> /Cd <sup>2+</sup>	-0.189	-1.573	-3.004	41.776	0.141
Cu <sup>2+</sup> /Pb <sup>2+</sup> /Cd <sup>2+</sup>	-0.236	-0.987	-1.704	23.581	0.073

**Table S10** Thermodynamic parameters of Cd<sup>2+</sup>adsorption by MCA in single, binary, and ternary systems

adsorbates	$\Delta G^\theta$ (kJ/mol)			$\Delta H^\theta$ (kJ/mol)	$\Delta S^\theta$ (kJ/mol·K)
	25°C	35 °C	45 °C		
Cd <sup>2+</sup>	-0.804	-1.519	-2.327	21.878	0.076
Cd <sup>2+</sup> /Pb <sup>2+</sup>	-0.531	-1.719	-2.832	33.784	0.115
Cd <sup>2+</sup> /Cu <sup>2+</sup>	-1.130	-1.600	-2.387	21.167	0.063
Cd <sup>2+</sup> /Pb <sup>2+</sup> /Cu <sup>2+</sup>	-0.628	-1.752	-2.803	35.207	0.109

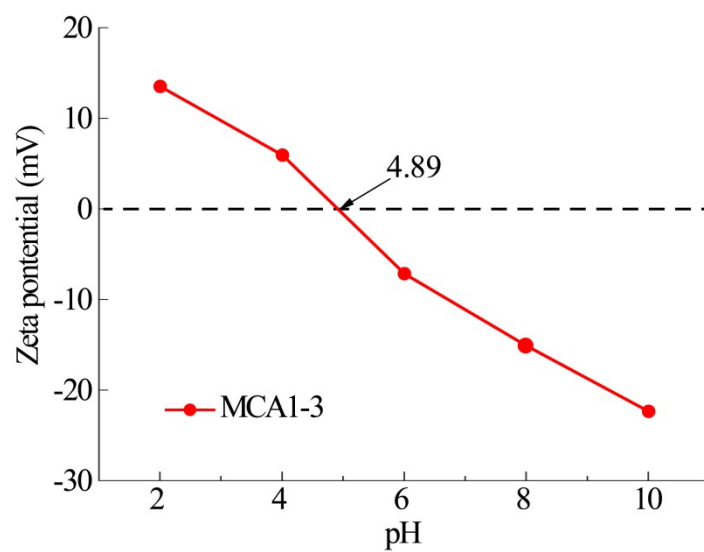


Distribution coefficient ( $K_d$ ) and adsorption selectivity factor  $\alpha_M^A$  are calculated by Equations (S9)-(S10).  $K_d$  represents the strength of the affinity of the adsorbent to the adsorbate, L/g.  $\alpha_M^A$  represents the adsorption selectivity factor of the adsorbent on the target ion (A) in the solution with interfering ions (M).

$$K_d = (C_0 - C_e) / C_e \times V / m \quad \text{(Equation S9)}$$

$$\alpha_M^A = \frac{q_A C_M}{q_M C_A} \quad \text{(Equation S10)}$$

Where  $C_0$  is the concentration of heavy metals in the solution before adsorption, mg/mL,  $C_e$  is the concentration of heavy metals in solution at adsorption equilibrium, g/ml,  $V$  is the volume of sample solution, mL;  $m$  is the weight of sample, g;  $q_A$  is the adsorption amount of A at adsorption equilibrium, mg/g;  $q_M$  is the adsorption amount of M at adsorption equilibrium, mg/g;  $C_A$  is the concentration of A in solution at adsorption equilibrium, mg/L;  $C_M$  is the concentration of M in solution at adsorption equilibrium, mg/L.



**Fig. S3** The zeta potentials of MCA1-3 at different pH in the presence of  $\text{KNO}_3$  ( $10^{-3}\text{mol}\cdot\text{L}^{-1}$ ).

**Table S11** Comparison of MCA with other biomass based materials.

Adsorbent	Initial concentration ( $\mu\text{g/mL}$ )			Adsorption capacity ( $\text{mg/g}$ )			Ref.
	$\text{Pb}^{2+}$	$\text{Cu}^{2+}$	$\text{Cd}^{2+}$	$\text{Pb}^{2+}$	$\text{Cu}^{2+}$	$\text{Cd}^{2+}$	
Magnetic carbon quantum dots from Pomegranate peel ( $\text{Fe}_3\text{O}_4$ -PPCQDs)	150	/	150	17.92	/	23.75	[1]
magnetic multi-functional biochar adsorbents (MMF-BC)	25	/	25	61.25	/	53.72	[2]
Wood ash amended biochar	150	150	150	61.5	38.9	10.2	[3]
Garden waste biochar	300	300	/	45.9	25.2	/	[4]
MCA	60	60	60	74.38	84.21	73.63	This work