Supplementary Materials: Environmental Impacts and Immobilization Mechanisms of Cadmium, Lead and Zinc in Geotechnical Composites Made from Contaminated Soil and Paper-Ash

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Table S1. ICP-MS operating parameters for determination of element concentrations.

Parameter	Type/Value	Helium mode	No gas mode
Sample introduction			
Nebuliser	Miramist		
Spray chamber	Scott		
Skimmer and sampler	Ni		
Plasma conditions			
Forward power	1550 W		
Plasma gas flow	15.0 L/min		
Carrier gas flow		1.05 L/min	0.75 L/min
Dilution gas flow		0.10 L/min	0.45 L/min
He gas flow		4.5 mL/min	
QP bias		-15 V	-3.6 V
Oct bias		-18 V	-8.0 V
Cell entrance		-40 V	-40 V
Cell exit		-60 V	-50 V
Deflect		-2.2 V	13.4 V
Plate bias		-60 V	-40 V
Sample uptake rate	0.3 mL/min		
Data acquisition parameters			
Isotopes monitored		^{66}Zn	^{111}Cd. ^{208}Pb
Isotopes of internal standards		^{72}Ge, ^{89}Y, ^{103}Rh, ^{115}In	^{72}Ge, ^{89}Y, ^{103}Rh, ^{115}In

Table S2. Concentrations of elements in standard reference material SPS-SW1 (Reference material for measurements of elements in surface waters) determined by ICP-MS. The results represent the mean concentration from three parallel samples. The uncertainty of ICP-MS determination was ± 2%.

Parameter	Certified concentration (µg/L)	Determined concentration (µg/L)
Cd	0.50±0.01	0.48±0.01
Pb	5.0±0.1	4.94±0.15
Zn	20[a]	19.7±0.6

[a]informative value

Table S3. Concentrations of elements in certified reference material CRM 320R (Trace Elements in River Sediment) determined by ICP-MS after microwave assisted digestion. The results represent the mean concentration from three parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Element	Certified (mg/kg)	Determined (mg/kg)
Cd	2.64±0.18	2.52±0.07
Zn	319±20	308±9
Pb	85±5	87±2

Table S4. Concentrations of Cd in individual fractions of extraction procedure in uncontaminated, contaminated, and remediated soils during the course of the experiment. Concentrations of Cd were determined by ICP-MS. The results represent the mean concentration from two parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Days after the start of the experiment	Extraction step	Uncontaminated soil Cd (mg/kg)	Contaminated soil Cd (mg/kg)	Remediated soil Cd (mg/kg)
7	I	0.0007	0.07	0.0007
	II	0.0007	1.30	0.0007
	III	0.015	5.90	0.07
	IV	0.012	0.32	6.80
	V	0.0007	0.01	0.03
	VI	1.01	1.20	0.80
	Total concentration	1.04	8.73	7.70
28	I	0.0007	0.57	0.0005
	II	0.0007	0.47	0.0007
	III	0.015	4.80	0.93
	IV	0.012	0.60	0.51
	V	0.0007	0.01	0.006
	VI	1.01	2.30	6.10
	Total concentration	1.04	8.73	7.70
56	I	0.0007	0.39	0.0005
	II	0.0007	0.41	0.0005
	III	0.015	2.60	1.50
	IV	0.012	0.45	3.30
	V	0.0007	0.01	0.002
	VI	1.01	4.90	2.90
	Total concentration	1.04	8.73	7.70
90	I	0.0007	0.62	0.0005
	II	0.0007	0.22	0.0005
	III	0.015	2.90	1.50
	IV	0.012	0.51	5.80
	V	0.0007	0.01	0.003
	VI	1.01	4.40	0.40
	Total concentration	1.04	8.73	7.70
120	I	0.0007	0.80	0.0008
	II	0.0007	1.40	0.006
	III	0.015	5.60	4.30
	IV	0.012	0.83	3.30
	V	0.0007	0.003	0.006
	VI	1.01	0.10	0.10
	Total concentration	1.04	8.73	7.70
365	I	0.0007	0.75	0.0005
	II	0.0007	1.50	0.007
	III	0.015	5.90	3.60
	IV	0.012	0.51	3.20
	V	0.0007	0.01	0.03
	VI	1.01	0.04	0.80
	Total concentration	1.04	8.73	7.70

Table S5. Concentrations of Pb in individual fractions of extraction procedure in uncontaminated, contaminated, and remediated soils during the course of the experiment. Concentrations of Pb were determined by ICP-MS. The results represent the mean concentration from two parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Days after the start of the experiment	Extraction step	Uncontaminated soil Pb (mg/kg)	Contaminated soil Pb (mg/kg)	Remediated soil Pb (mg/kg)
7	I	0.0003	0.04	2.1
	II	0.0003	1.60	1.2
	III	0.0021	68.4	0.31
	IV	0.18	244	634
	V	0.0043	0.45	0.0003
	VI	9.3	631	127
	Total concentration	9.5	946	767
28	I	0.0003	0.41	0.13
	II	0.0003	2.03	0.32
	III	0.0021	617	72.6
	IV	0.18	273	240.8
	V	0.0043	0.17	0.001
	VI	9.3	53.3	453
	Total concentration	9.5	946	767
56	I	0.0003	0.19	0.001
	II	0.0003	1.4	0.005
	III	0.0021	313	169
	IV	0.18	143.2	478
	V	0.0043	0.49	0.0007
	VI	9.3	488	120
	Total concentration	9.5	946	767
90	I	0.0003	0.27	0.0002
	II	0.0003	1.24	0.014
	III	0.0021	350	163
	IV	0.18	127	414
	V	0.0043	0.29	0.0002
	VI	9.3	467	190
	Total concentration	9.5	946	767
120	I	0.0003	0.24	0.002
	II	0.0003	3.7	0.030
	III	0.0021	373	177
	IV	0.18	158	341
	V	0.0043	0.0001	0.0002
	VI	9.3	411	250
	Total concentration	9.5	946	767
365	I	0.0003	0.21	0.002
	II	0.0003	7.97	0.12
	III	0.0021	670	228
	IV	0.18	180	508
	V	0.0043	0.37	0.020
	VI	9.3	57.5	31.5
	Total concentration	9.5	946	767

Table S6. Concentrations of Zn in individual fractions of extraction procedure in in uncontaminated, contaminated, and remediated soils during the course of the experiment. Concentrations of Zn were determined by ICP-MS. The results represent the mean concentration from two parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Days after the start of the experiment	Extraction step	Uncontaminated soil Zn (mg/kg)	Contaminated soil Zn (mg/kg)	Remediated soil Zn (mg/kg)
7	I	0.0025	69.0	7.6
	II	0.0025	15.1	6.4
	III	0.063	7338	5.6
	IV	0.22	993	7075
	V	0.14	15.2	1.9
	VI	40.9	384	395
	Total concentration	41.3	8814	7491
28	I	0.0025	50.7	0.6
	II	0.0025	15.4	1.4
	III	0.063	7022	1613
	IV	0.22	913	877
	V	0.14	5.4	1.7
	VI	40.9	808	4998
	Total concentration	41.3	8814	7491
56	I	0.0025	34.7	0.0012
	II	0.0025	10.4	0.015
	III	0.063	5763	2932
	IV	0.22	596	3861
	V	0.14	5.1	0.074
	VI	40.9	2405	698
	Total concentration	41.3	8814	7491
90	I	0.0025	58.3	0.1362
	II	0.0025	9.3	0.161
	III	0.063	6660	3568
	IV	0.22	525.3	3298
	V	0.14	3.4	0.191
	VI	40.9	1557	624
	Total concentration	41.3	8814	7491
120	I	0.0025	63.6	0.44
	II	0.0025	41.2	0.52
	III	0.063	7563	3198
	IV	0.22	753	3793
	V	0.14	0.10	0.55
	VI	40.9	393	498
	Total concentration	41.3	8814	7491
365	I	0.0025	55.3	0.41
	II	0.0025	36.8	0.65
	III	0.063	7606	2033
	IV	0.22	656	5024
	V	0.14	1.70	12.0
	VI	40.9	458	421
	Total concentration	41.3	8814	7491

Table S7. Concentrations of Cd in individual fractions of extraction procedure in uncontaminated soil treated with CaSO₄, contaminated and remediated soils during the course of the experiment. Concentrations of Cd were determined by ICP-MS. The results represent the mean concentration from two parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Days after the start of the experiment	Extraction step	Uncontaminated soil Cd (mg/kg)	Contaminated soil Cd (mg/kg)	Remediated soil Cd (mg/kg)
7	I	0.0007	0.12	0.0007
	II	0.0007	2.00	0.0007
	III	0.015	4.70	0.072
	IV	0.009	0.23	6.40
	V	0.0007	0.01	0.01
	VI	0.97	1.90	1.30
	Total concentration	0.99	8.90	7.70
28	I	0.0007	1.5	0.0005
	II	0.0007	0.83	0.0005
	III	0.015	3.80	1.40
	IV	0.009	0.54	0.57
	V	0.0007	0.01	0.008
	VI	0.97	2.30	5.70
	Total concentration	0.99	8.90	7.70
56	I	0.0007	0.96	0.0004
	II	0.0007	0.93	0.0004
	III	0.015	3.40	1.10
	IV	0.009	0.46	2.80
	V	0.0007	0.009	0.0009
	VI	0.97	3.20	3.90
	Total concentration	0.99	8.90	7.70
90	I	0.0007	0.83	0.0005
	II	0.0007	0.22	0.0005
	III	0.015	3.40	1.30
	IV	0.009	0.51	5.30
	V	0.0007	0.007	0.002
	VI	0.97	4.00	1.20
	Total concentration	0.99	8.90	7.70
120	I	0.0007	1.00	0.002
	II	0.0007	1.60	0.006
	III	0.015	5.60	4.20
	IV	0.009	0.63	3.30
	V	0.0007	0.00	0.006
	VI	0.97	0.10	0.20
	Total concentration	0.99	8.90	7.70
365	I	0.0007	1.30	0.002
	II	0.0007	2.00	0.005
	III	0.015	5.10	3.80
	IV	0.009	0.50	3.30
	V	0.0007	0.01	0.023
	VI	0.97	0.10	0.90
	Total concentration	0.99	8.90	7.70

Table S8. Concentrations of Pb in individual fractions of extraction procedure in uncontaminated soil treated with $CaSO_4$, contaminated and remediated soils during the course of the experiment. Concentrations of Pb were determined by ICP-MS. The results represent the mean concentration from two parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Days after the start of the experiment	Extraction step	Uncontaminated soil Pb (mg/kg)	Contaminated soil Pb (mg/kg)	Remediated soil Pb (mg/kg)
7	I	0.0003	0.0461	2.43
	II	0.0003	5.50	1.28
	III	0.0028	68.00	4.48
	IV	0.20	238	621
	V	0.0067	0.15	0.0003
	VI	8.50	659	44.5
	Total concentration	8.70	970	734
28	I	0.0003	0.77	0.022
	II	0.0003	5.50	0.064
	III	0.0028	713	103
	IV	0.20	228	259
	V	0.0067	0.13	0.002
	VI	8.50	22.3	311
	Total concentration	8.70	967	734
56	I	0.0003	0.35	0.0004
	II	0.0003	6.30	0.005
	III	0.0028	491	95.3
	IV	0.20	156	470
	V	0.0067	0.19	0.0001
	VI	8.50	316	168
	Total concentration	8.70	970	734
90	I	0.0003	0.32	0.0003
	II	0.0003	2.58	0.0078
	III	0.0028	515	122
	IV	0.20	139	325
	V	0.0067	0.28	0.0002
	VI	8.50	313	287
	Total concentration	8.70	970	734
120	I	0.0003	0.37	0.0014
	II	0.0003	2.5	0.029
	III	0.0028	374	194
	IV	0.20	157	328
	V	0.0067	0.001	0.0002
	VI	8.50	436	211
	Total concentration	8.70	970	734
365	I	0.0003	0.41	0.0037
	II	0.0003	17.5	0.07
	III	0.0028	753.3	205
	IV	0.20	171.2	501
	V	0.0067	1.23	0.0032
	VI	8.50	25.9	27.4
	Total concentration	8.70	970	734

Table S9. Concentrations of Zn in individual fractions of extraction procedure in uncontaminated soil treated with CaSO₄, contaminated and remediated soils during the course of the experiment. Concentrations of Zn were determined by ICP-MS. The results represent the mean concentration from two parallel samples. The expanded uncertainty of analytical procedure was better than ± 3% (k = 2).

Days after the start of the experiment	Extraction step	Uncontaminated soil Zn (mg/kg)	Contaminated soil Zn (mg/kg)	Remediated soil Zn (mg/kg)
7	I	0.0026	101.2	10.2
	II	0.0026	50.7	8.0
	III	0.047	7596	401
	IV	0.19	847	6511
	V	0.16	10.5	5.8
	VI	38.8	77.4	88.7
	Total concentration	39.2	8782	7024
28	I	0.0026	117	0.4
	II	0.0026	32.5	0.6
	III	0.047	7235	3022
	IV	0.19	376.1	421
	V	0.16	5.6	2.6
	VI	38.8	915	3578
	Total concentration	39.2	8782	7024
56	I	0.0026	82.8	0.0012
	II	0.0026	40.8	0.0073
	III	0.047	6278	3267
	IV	0.19	566	3097
	V	0.16	4.0	0.028
	VI	38.8	1711	660
	Total concentration	39.2	8782	7024
90	I	0.0026	76.8	0.13
	II	0.0026	19.7	0.023
	III	0.047	6350	3232
	IV	0.19	558	2451
	V	0.16	4.2	0.607
	VI	38.8	1672	1340
	Total concentration	39.2	8782	7024
120	I	0.0026	97.3	0.74
	II	0.0026	56.0	0.48
	III	0.047	7715	3247
	IV	0.19	749	3512
	V	0.16	0.1	1.02
	VI	38.8	64.9	264
	Total concentration	39.2	8782	7024
365	I	0.0026	93.9	1.31
	II	0.0026	68.5	0.44
	III	0.047	7894	2264
	IV	0.19	684	4668
	V	0.16	5.5	12.6
	VI	38.8	36.1	77.2
	Total concentration	39.2	8782	7024

Table S10. Data on the statistical processing of the results.

Data on the statistical processing of the results for Cd

t-Test: Two-Sample Assuming Unequal Variances	Step I Variable 1	Variable 2	Step II Variable 1	Variable 2	Step III Variable 1	Variable 2	Step IV Variable 1	Variable 2	Step V Variable 1	Variable 2	Step VI Variable 1	Variable 2
Mean	0.533333	0.000583	0.883333	0.002567	4.616667	1.983333	3.818333	0.536667	0.012833	0.008833	2.156667	1.85
Variance	0.072427	1.77E-08	0.331147	9.39E-06	2.261667	2.643627	4.936417	0.029187	0.000179	8.17E-06	4.440867	5.307
Observations	6	6	6	6	6	6	6	6	6	6	6	6
Hypothesized Mean Difference	0		0		0		0		0		0	
df	5		5		10		5		5		10	
t Stat	4.848973		3.749041		2.912386		3.607315		0.715478		0.240595	
P(T<=t) one-tail	0.002339		0.006653		0.007751		0.007712		0.253162		0.407365	
t Critical one-tail	2.015048		2.015048		1.812461		2.015048		2.015048		1.812461	
P(T<=t) two-tail	0.004678		0.013307		0.015501		0.015424		0.506324		0.814729	
t Critical two-tail	2.570582		2.570582		2.228139		2.570582		2.570582		2.228139	

Data on the statistical processing of the results for Pb

t-Test: Two-Sample Assuming Unequal Variances	Step I Variable 1	Variable 2	Step II Variable 1	Variable 2	Step III Variable 1	Variable 2	Step IV Variable 1	Variable 2	Step V Variable 1	Variable 2	Step VI Variable 1	Variable 2
Mean	0.372533	0.226667	2.99	0.2815	398.5667	134.985	435.9667	187.5333	0.295017	0.003733	351.3	195.25
Variance	0.718845	0.014427	6.74976	0.216494	48146.65	6877.631	18752.81	3410.347	0.034058	6.36E-05	57798.4	21318.98
Observations	6	6	6	6	6	6	6	6	6	6	6	6
Hypothesized Mean Difference	0		0		0		0		0		0	
df	5		5		6		7		5		8	
t Stat	0.417253		2.51365		2.752413		4.087612		3.86256		1.35895	
P(T<=t) one-tail	0.346902		0.026794		0.016593		0.002322		0.005925		0.105617	
t Critical one-tail	2.015048		2.015048		1.94318		1.894579		2.015048		1.859548	
P(T<=t) two-tail	0.693804		0.053589		0.033186		0.004645		0.011849		0.211233	
t Critical two-tail	2.570582		2.570582		2.446912		2.364624		2.570582		2.306004	

Data on the statistical processing of the results for Zn

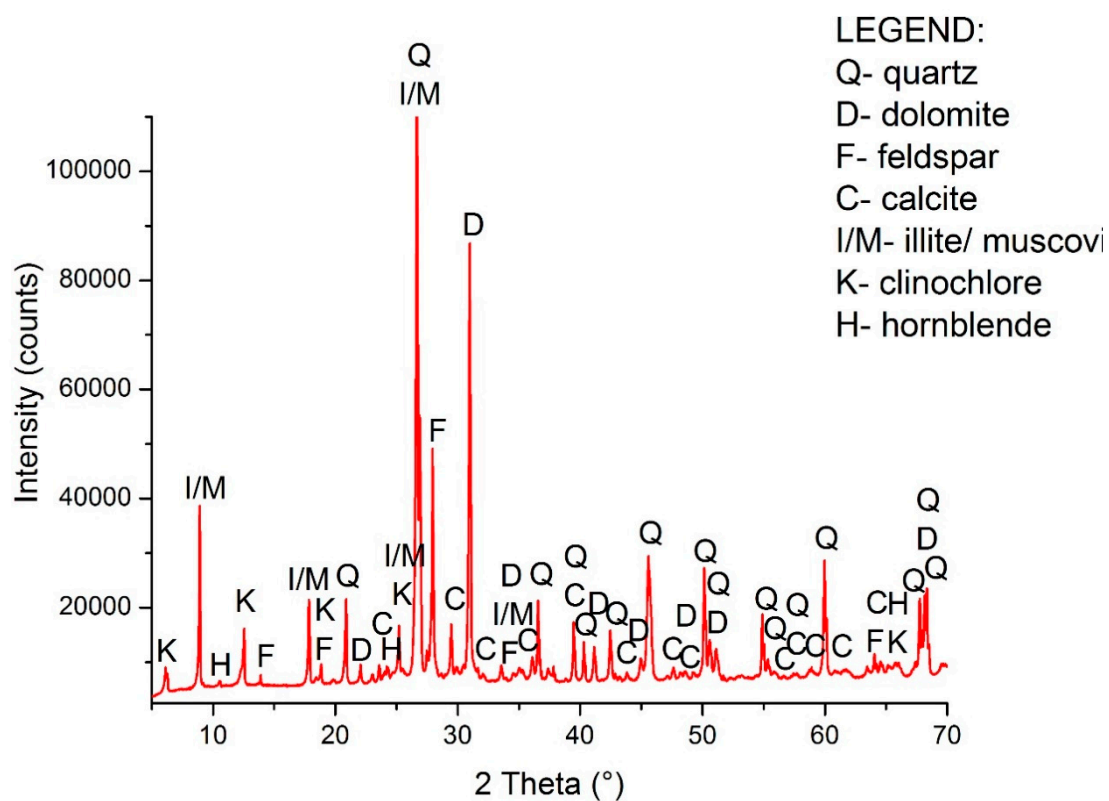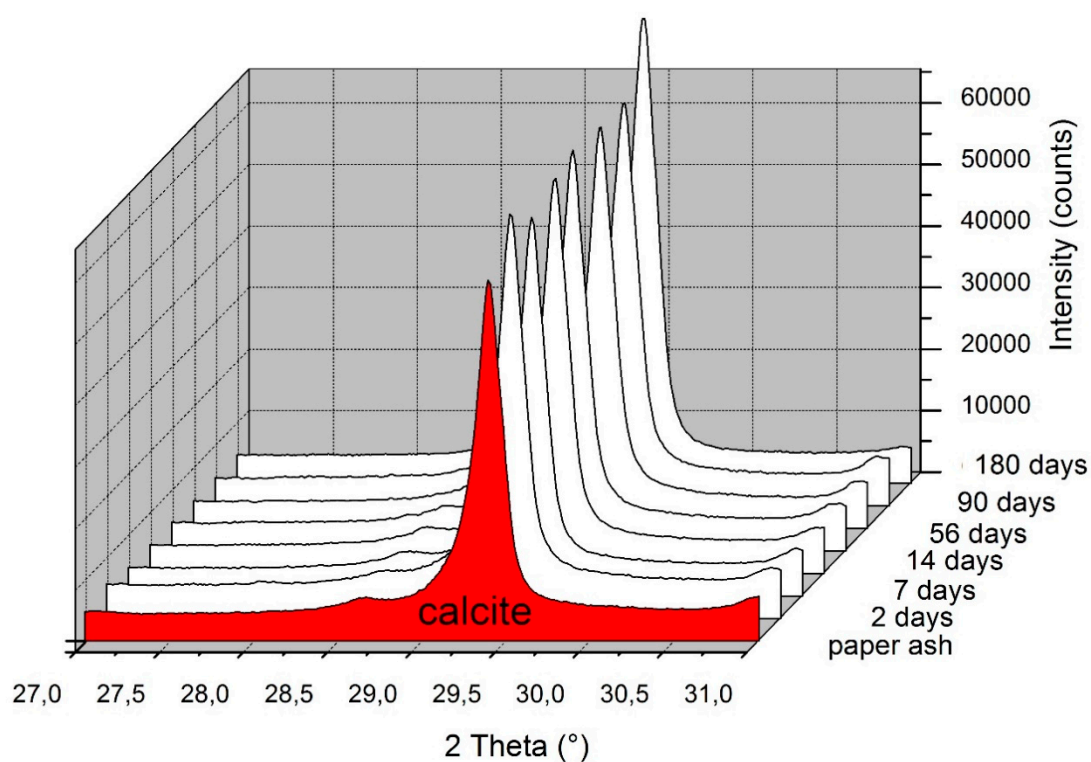| t-Test: Two-Sample Assuming Unequal Variances	Step I Variable 1	Variable 2	Step II Variable 1	Variable 2	Step III Variable 1	Variable 2	Step IV Variable 1	Variable 2	Step V Variable 1	Variable 2	Step VI Variable 1	Variable 2
Mean	55.26667	1.531233	21.36667	1.524333	6992	2224.933	3988	739.3833	5.15	2.735833	1272.333	1000.833
Variance	142.2187	8.88644	194.4587	5.939499	488863.6	1717474	4162288	33598.56	28.307	21.18715	3344972	672560.6
Observations	6	6	6	6	6	6	6	6	6	6	6	6
Hypothesized Mean Difference	0		0		0		0		0		0	
df	6		5		8		5		10		7	
t Stat	10.70772		3.433377		7.861236		3.884742		0.840555		0.331792	
P(T<=t) one-tail	1.96E-05		0.009284		2.48E-05		0.005793		0.210121		0.37488	
t Critical one-tail	1.94318		2.015048		1.859548		2.015048		1.812461		1.894579	
P(T<=t) two-tail	3.92E-05		0.018568		4.95E-05		0.011586		0.420241		0.749759	
t Critical two-tail	2.446912		2.570582		2.306004		2.570582		2.228139		2.364624	

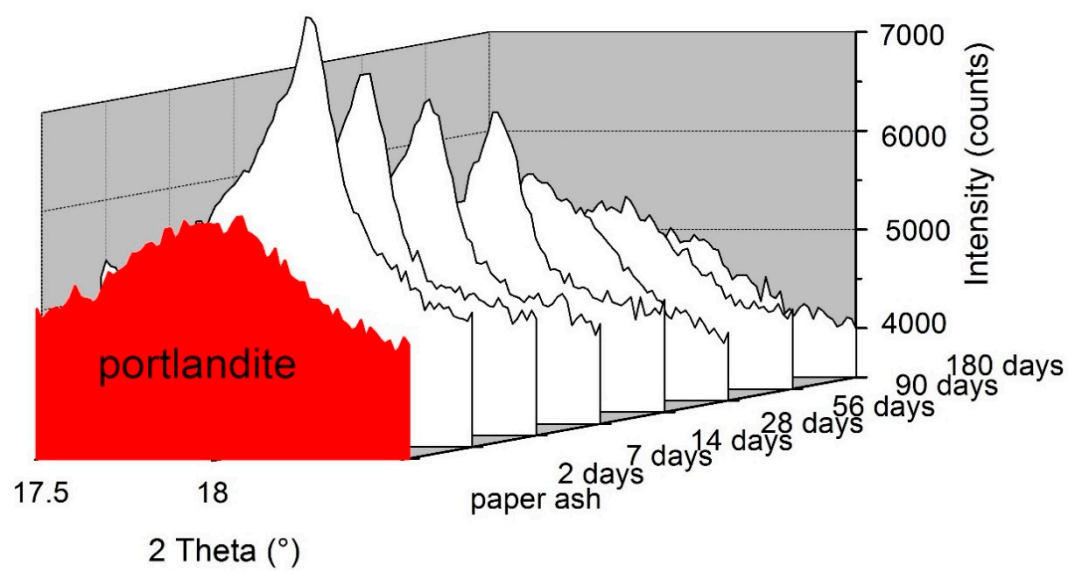**Figure S1.** XRD pattern of uncontaminated soil.

B

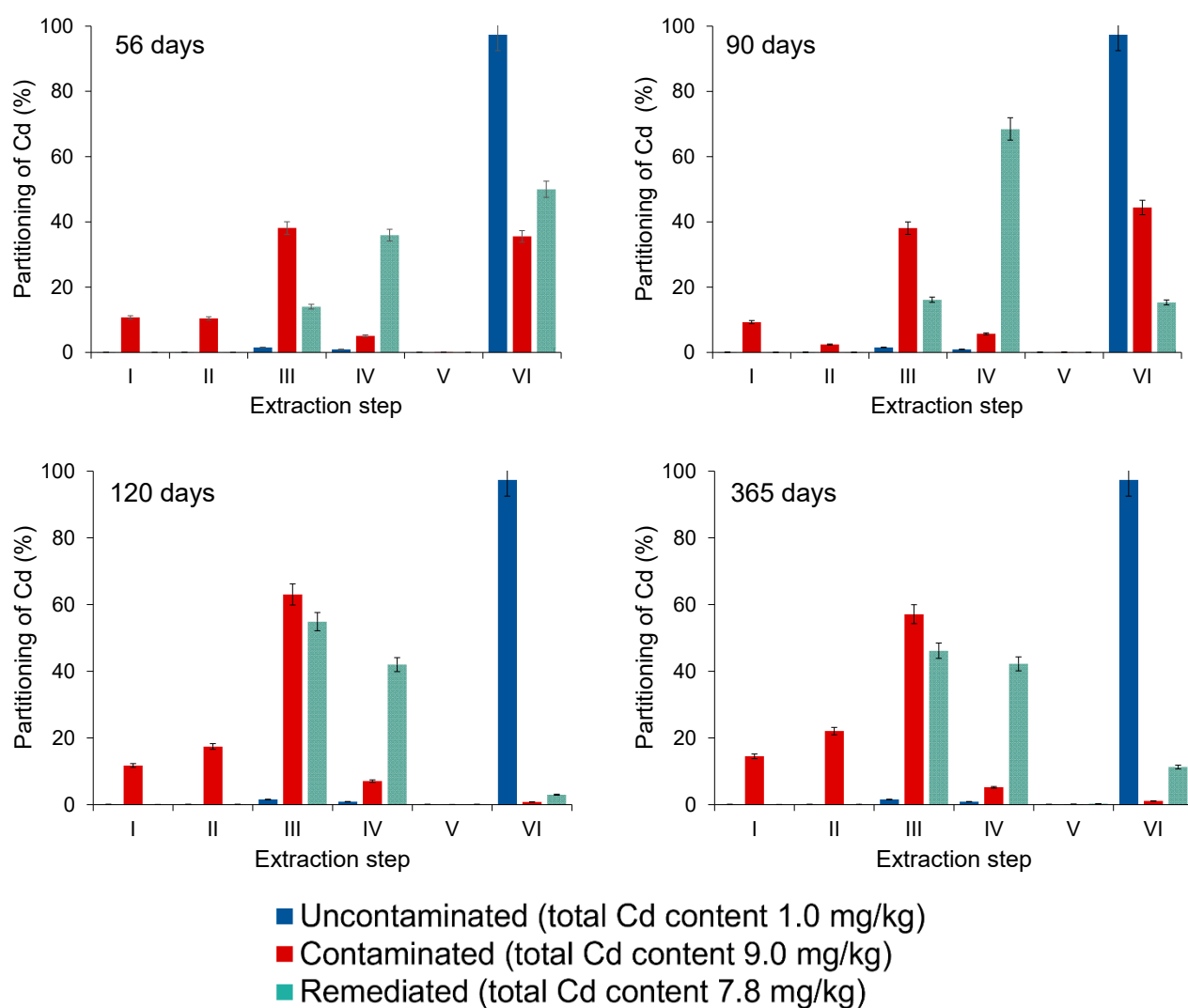**Figure S2.** Sections of the XRD patterns of the paper ash paste.

Figure S3. Partitioning of Cd in sulphate-treated uncontaminated, contaminated, and remediated soil samples with time elapsed after the remediation.

I water-soluble

II exchangeable

III bound to carbonates

IV bound to Fe/Mn oxides and hydroxides
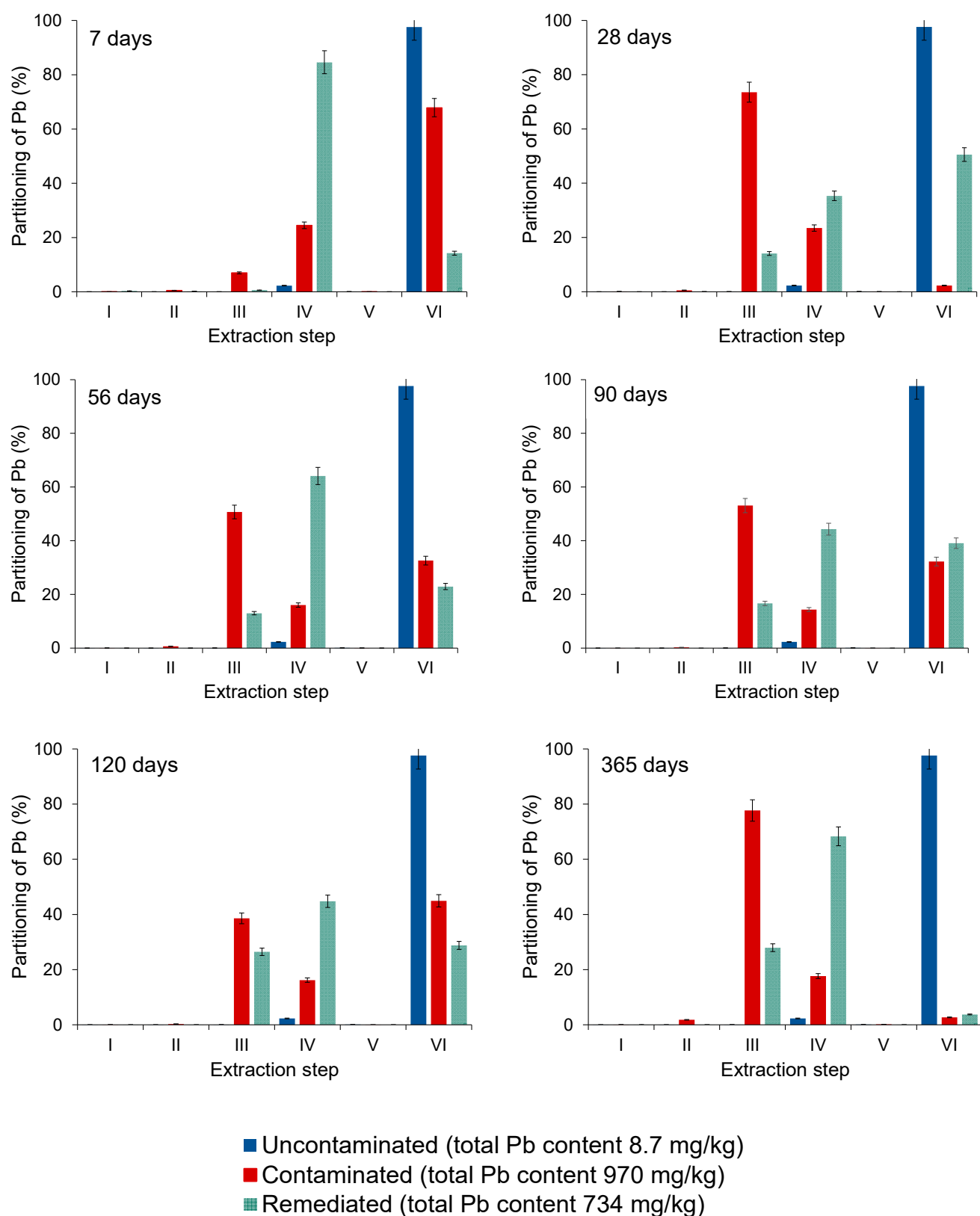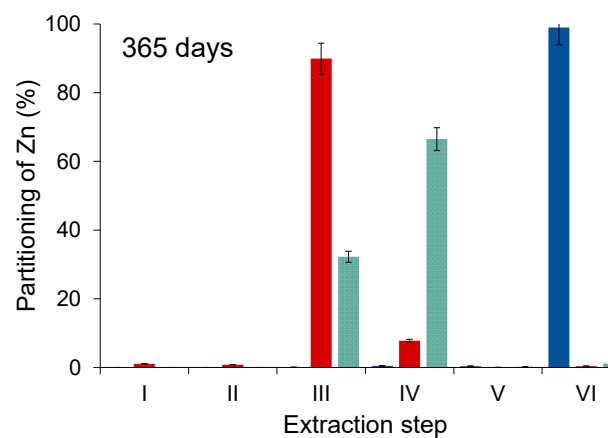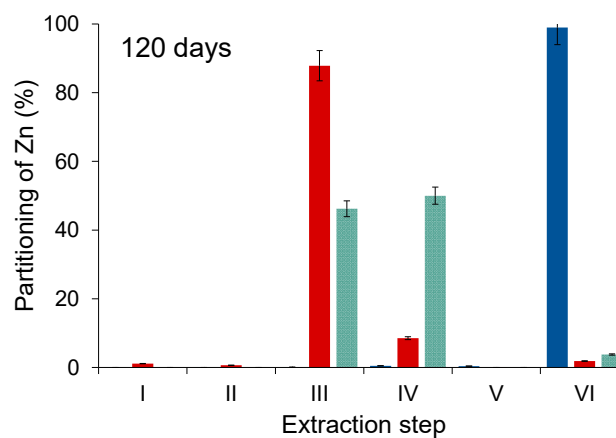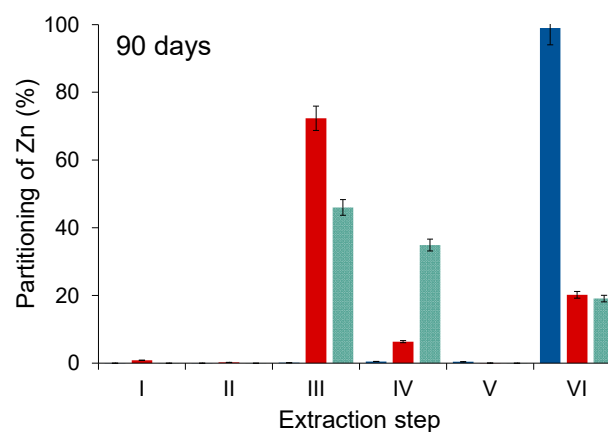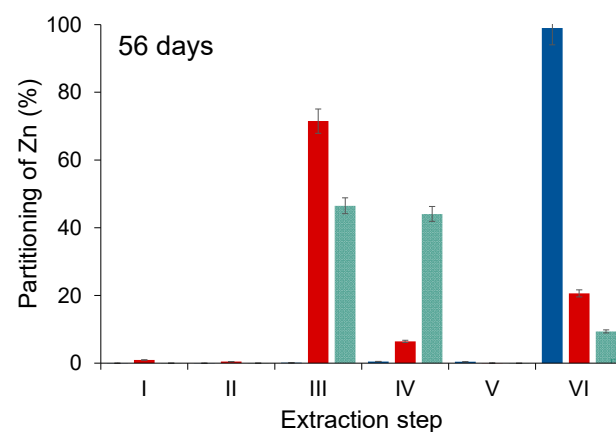
V bound to organic matter

VI bound to silicate lattice

Figure S4. Partitioning of Pb in sulphate-treated uncontaminated, contaminated, and remediated soil samples with time elapsed after the remediation.

I water-soluble

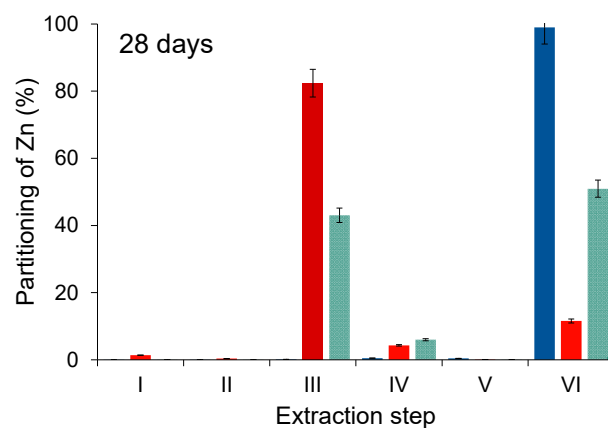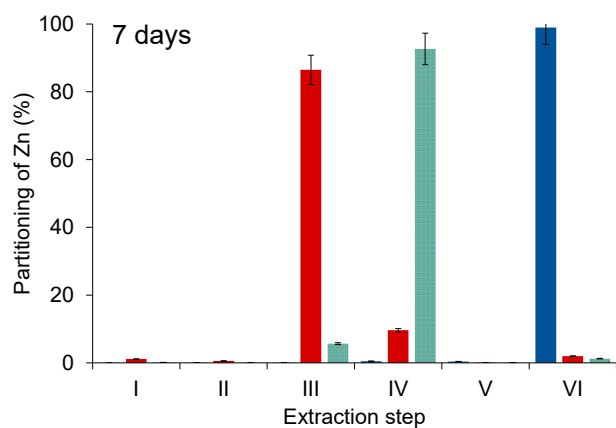II exchangeable

III bound to carbonates

IV bound to Fe/Mn oxides and hydroxides

V bound to organic matter

VI bound to silicate lattice

■ Uncontaminated soil (total Zn content 39.2 mg/kg)
■ Contaminated soil (total Zn content 8782 mg/kg)
■ Remediated soil (total Zn content 7024 mg/kg)

Figure S5. Partitioning of Zn in sulphate-treated uncontaminated, contaminated, and remediated soil samples with time elapsed after the remediation.

I water-soluble

II exchangeable

III bound to carbonates

IV bound to Fe/Mn oxides and hydroxides

V bound to organic matter

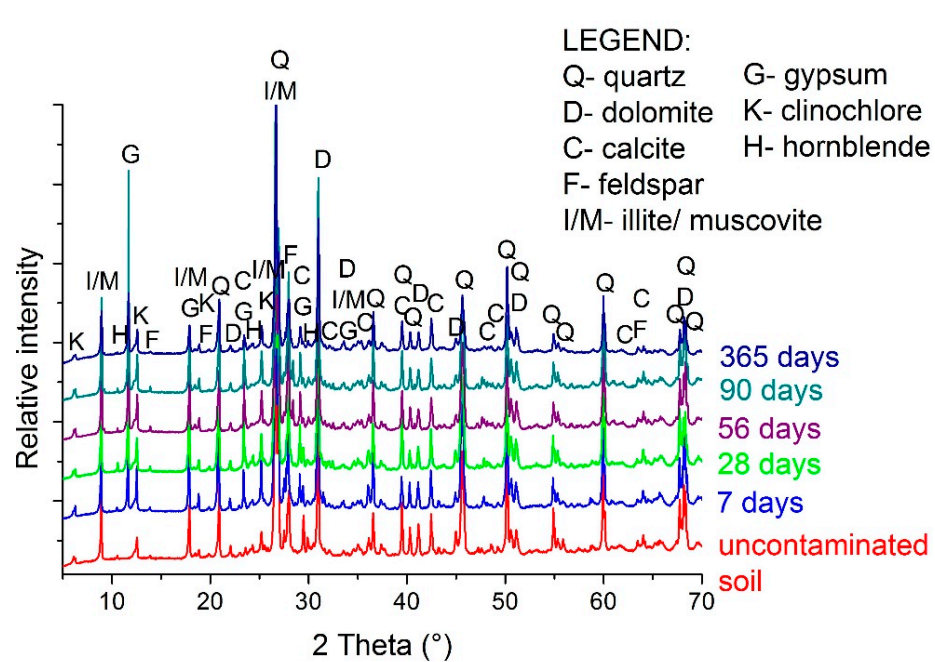VI bound to silicate lattice

Figure S6. XRD patterns of contaminated soil (**A**) and sulphate-treated contaminated soil (**B**) with time elapsed.