Supporting Information

Combined column test for characterization of leaching and transport of trace elements in contaminated soils

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This supporting information consists of two sections: S.1) Supplementary method description and S.2) Supplementary data.

Summary of the numbers in Supporting Information

- Number of pages: 7
- Number of tables: 10
- Number of figures: 1

S.1. Supplementary method description

S.1.1. Bulk density

Bulk density (ρ_b) in the combined columns was calculated as

$$\rho_b = \frac{dry\ mass\ sample}{sample\ volume} \quad \text{(S1)}$$

S.1.2. Calculations pore number and porosity

The pore number and porosity in the combined columns were theoretically calculated based on the following argumentation:

The pore number (e) is the ratio between the pore volume (V_p) and the volume of solid material (V_s), equivalent to the gas filled pore volume (V_a) and the water-occupied pore volume (V_w):

$$e = \frac{V_p}{V_s} = \frac{V_a + V_w}{V_s} \quad \text{(S2)}$$

The pore volume can also be derived from the specific gravity (G_s), the volumetric water content (w) and the degree of saturation (S_r)

$$e = \frac{wG_s}{S_r} \quad \text{(S3)}$$

Under 100 % saturation (S_r = 1), where V_w is assumed to equal V_p, the pore number can be expressed as

$$e = wG_s \quad \text{(S4)}$$

Hence, the pore numbers were calculated as following:

$$e = \left(\frac{G_s \gamma_w}{\gamma_d}\right) - 1 \quad \text{(S5)}$$

where γ_w is the unit weight of water (kg/m^3), γ_d is the dry unit weight (kg/m^3) and G_s the specific gravity.

The porosity was (n) was calculated as:

$$n = \frac{e}{1+e} \quad \text{(S6)}$$

S.1.3. Calculations porewater velocity

The porewater velocity (v) is the ratio between Darcy flux (q) and porosity (n):

$$v = \frac{q}{n} \quad \text{(S7)}$$

The Darcy flux was calculated as:

$$q = \frac{pumping\ rate}{A} \quad \text{(S8)}$$

where pumping rate is the pumping rate of the peristaltic pump (mL/d), and A is the cross-sectional area of the combined columns (A).

S.2. Supplementary data

Table S1 Limits of Quantification (LOQ) for TIC and TOC analysis.

	TIC	TOC
LOQ (% dry matter)	0.010	0.01[a]

[a]Limit of reporting (LOR)

Table S2 Particle size distribution (< 4 mm).

	< 0.002 (mm)	**< 0.006 (mm)**	**< 0.02 (mm)**	**< 0.06 (mm)**	**< 0.2 (mm)**	**< 0.6 (mm)**	**< 2 (mm)**	**< 4 (mm)**
Shooting range soil (%)	7	12	23	41	64	83	95	100
Urban soil (%)	10	17	28	39	53	71	89	100

Table S3 Limits of Quantification (LOQ) for ICP-MS analysis of solid soil samples (total concentrations).

	Fe	Cu	Zn	Sb	Pb
LOQ (mg/kg)	1.5	0.42	5.30	0.002	0.26

Table S4 Packing specifications and key numbers from the combined columns.

Soil	Density	Column volume (cm^3)	Dry weight column (g)	Average pore volume (L) in columns	Pore volume corresponding to L/S 0.1	Pore volume corresponding to L/S 2	Pore volume corresponding to L/S 6	Pore volume corresponding to L/S 10
Shooting range soil	High	580.9	889.8	0.2	0.3	6.9	13.7	29.1
Shooting range soil	Low	567.2	804.7	0.3	0.3	5.9	10.6	-
Urban soil	Low	585.5	567.2	0.4	0.2	2.7	6.0	-

Table S5 Limits of Quantification (LOQ) for ICP-MS analysis of aqueous samples.

	Fe (mg/L)	Cu (µg/L)	Zn (µg/L)	Sb (µg/L)	Pb (µg/L)	Ca (mg/L)	K (mg/L)	Mg (mg/L)	Na (mg/L)
LOQ	0.060[a, b]	0.040[a] 0.527[b]	0.035[a] 0.095[b]	0.140[a] 0.0019[b]	0.385[a] 0.010[b]	0.10	0.19	0.006	0.067

[a]High density shooting range soil and batch test, [b]Low density shooting range soil and urban soil

Table S6 pH, EC, DOC, anion, and cation concentrations from the combined columns as a function of L/S ratio. Standard deviations are based on three replicates.

Soil	Density	L/S ratio	pH	EC (µS/cm)	DOC (mg/L)	Cl^- (mg/L)	NO_3^--N (mg/L)	SO_4^{2-} (mg/L)	Na (mg/L)	Mg (mg/L)	K (mg/L)	Ca (mg/L)	Fe (ug/L)	Cu (ug/L)	Zn (ug/L)	Pb (ug/L)	Sb (ug/L)
Shooting	Low	0.1	6.6 ±0.1	156 ±20	37 ±5	30 ±3	4.8 ±1.1	5.2 ±0.4	11 ±1	2.7 ±0.3	5.3 ±0.3	10 ±1	42± 28	326 ±34	989± 101	2616 ±343	338 ±8

range soil		2	6.7 ±0.2	31± 2	8.6 ±0.2	0.5 ±0.0	0.0	6.9 ±0.4	1.2 ±0.1	0.5 ±0.0	1.6 ±0.0	1.8 ±0.1	69± 24	138 ±8	189± 8	825 ±43	545 ±18
		6	6.2 ±0.0	13 ±1	2.4 ±0.2	0.0	0.0	1.9 ±0.3	0.1 ±0.1	0.2 ±0.0	1.0 ±0.1	0.6 ±0.3	53± 21	86 ±5	81±10	592 ±23	687 ±31
	High	0.1	5.9 ±0.1	178 ±7	36 ±3	33 ±3	7.1 ±0.9	4.6 ±0.6	13 ±1	3.2 ±0.3	5.3 ±0.3	21 ±13	20± 32	443 ±52	4435 ±4133	3100 ±297	327 ±27
		2	6.6 ±0.2	33± 2	8.6 ±0.4	0.5 ±0.1	0.0	7.0 ±0.3	1.6 ±0.4	0.8 ±0.4	1.7 ±0.6	16 ±19	35± 42	171 ±21	1911 ±2304	813 ±111	597 ±27
		6	6.7 ±0.0	16± 0	6.1 ±1.8	0.0	0.0	1.9 ±0.1	0.2 ±0.1	0.2 ±0.0	0.9 ±0.1	1.3 ±0.1	18± 14	106 ±5	150± 26	578 ±41	785 ±14
		10	6.7 ±0.2	12± 0	4.5 ±0.0	0.0	0.0	1.3 ±0.1	0.2 ±0.1	0.2 ±0.0	0.8 ±0.4	0.9 ±0.1	6±4	81 ±6	66±17	555 ±33	907 ±45
Urban soil	Low	0.1	8.4 ±0.0	780 ±19	31 ±2	10 ±2	3.1 ±2.1	10 ±1	18 ±1	10 ±0	43 ±2	137 ±5	42± 13	39 ±5	17±5	28± 8	6.1 ±0.6
		2	8.5 ±0.0	397 ±44	19 ±2	1.7 ±0.3	0.0	2.6 ±0.4	7± 1	5± 1	27 ±4	63 ±5	66± 16	32 ±3	0.0± 0.1	6.4± 2.2	2.6 ±0.7
		6	8.4 ±0.2	296 ±36	13 ±2	0.2 ±0.0	0.0	0.2 ±0.1	1± 1	3± 0	15 ±2	46 ±6	239 ±133	37 ±6	0.8± 1.2	4.5± 2.4	0.9 ±0.1

Table S7 pH, EC, DOC, anion, and cation concentrations in the one-stage batch test (L/S 10). Standard deviations are based on three replicates.

Soil	pH	EC (µS/cm)	DOC (mg/L)	Cl^- (mg/L)	NO_3^--N (mg/L)	SO_4^{2-} (mg/L)	Na (mg/L)	Mg (mg/L)	K (mg/L)	Ca (mg/L)	Fe (mg/L)	Cu (ug/L)	Zn (ug/L)	Pb (ug/L)	Sb (ug/L)
Shooting range soil	6.2 ±0.1	14± 2	8.1 ±0.8	0.5 ±0.0	2.0 ±0.1	0.8 ±0.1	0.5 ±0.1	0.2 ±0.0	0.7 ±0.0	1.1 ±0.2	0.1 ±0.1	89 ±6	408 ±96	346 ±35	480 ±20
Urban soil	7.9 ±0.1	178 ±7	9.6 ±2.3	0.4 ±0.1	0.1 ±0.0	1.4 ±0.1	2.1 ±0.2	3.4 ±2.2	12± 1	28± 1	6±7	48 ±24	233 ±105	232 ±283	5±2

Table S8 Hydraulic conductivity derived from the combined columns (K_{exp}) as a function of L/S ratio.

Shooting range soil (high density)						**Shooting range soil (low density)**						**Urban soil (low density)**					
Replicate 1		Replicate 2		Replicate 3		Replicate 1		Replicate 2		Replicate 3		Replicate 1		Replicate 2		Replicate 3	
L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)	L/S	K_{exp} (m/s)

0.1	5.8E-07	0.0	5.6E-07	0.0	4.7E-07	0.0	1.5E-06	0.1	1.6E-06	0.1	1.2E-06	0.1	1.5E-04	0.1	2.4E-04	0.1	2.0E-04
0.3	5.8E-07	0.3	5.2E-07	0.2	4.0E-07	0.6	1.0E-06	0.7	9.8E-07	0.7	4.6E-07	2.0	6.2E-05	2.0	1.2E-04	2.0	9.0E-05
0.5	5.4E-07	0.5	4.8E-07	0.4	3.4E-07	1.0	8.7E-07	1.0	8.3E-07	1.1	4.8E-07	5.0	9.8E-05	5.0	2.1E-04	5.0	1.5E-04
0.8	5.0E-07	0.8	4.4E-07	0.5	4.2E-07	1.9	9.3E-07	2.1	5.7E-07	1.7	1.7E-06						
1.9	3.5E-07	1.9	3.7E-07	1.5	4.1E-07	2.3	7.4E-07	2.5	6.4E-07	4.2	3.0E-07						
2.1	3.2E-07	2.3	3.3E-07	1.8	2.7E-07	3.4	5.5E-07	3.5	4.3E-07	4.4	8.9E-07						
2.3	3.0E-07	3.7	2.4E-07	2.8	2.2E-07	4.8	5.2E-07	4.8	3.7E-07	4.6	4.2E-06						
3.5	3.1E-07	3.8	2.8E-07	3.2	2.2E-07	5.7	5.7E-07	5.5	3.1E-07								
4.8	3.0E-07	5.4	2.5E-07	4.7	1.9E-07			6.0	2.7E-07								
5.4	2.0E-07	5.8	2.4E-07	5.2	1.8E-07												
5.8	1.9E-07	6.7	2.4E-07	6.2	1.6E-07												
6.8	1.8E-07	7.7	3.6E-07	7.3	1.5E-07												
7.8	1.6E-07	8.8	4.5E-07	8.0	1.5E-07												
8.3	2.1E-07	8.9	5.1E-07	8.8	1.4E-07												
8.6	2.1E-07	9.4	9.4E-07	9.6	1.4E-07												
9.1	2.1E-07	9.7	1.1E-06	9.8	1.3E-07												
10.0	2.3E-07			9.9	1.3E-07												

Table S9 Cumulative releases of DOC, Cu, Zn, Pb, and Sb from the combined columns as a function of L/S ratio. Standard deviations are based on three replicates.

Soil	Density	L/S ratio	DOC (mg/kg)	Cu (mg/kg)	Zn (mg/kg)	Pb (mg/kg)	Sb (mg/kg)
Shooting range soil	Low	0.1	2.3±0.7	0.0	0.1±0.0	0.2±0.0	0.0
		2	20±2	0.3±0.0	0.5±0.0	1.9±0.1	1.1±0.0
		6	29±1	0.6±0.0	0.7±0.0	3.9±0.1	3.5±0.5
	High	0.1	2.8±0.2	0.0	0.4±0.4	0.2±0.0	0.0±0.0
		2	19±1	0.4±0.0	3.8±4.0	1.8±0.2	1.2±0.1

		10	55±1	1.0±0.1	4.4±0.4	6.2±0.4	8.4±0.3
Urban soil	Low	0.1	3.6±0.5	0.01±0.00	0.00	0.00	0.00
		2	37±2	0.07±0.01	0.00	0.02±0.01	0.01±0.00
		6	87±15	0.27±0.06	0.01±0.01	0.03±0.02	0.01±0.00

Table S10 Comparison leaching and transport for the combined column test (CCT) versus leaching and transport from the batch test (BT) combined with Hazen's equation.

Leaching	Transport	Pb (mg/y)	Sb (mg/y)	Cu (mg/y)
CCT	CCT	490	801	72
BT	Hazen's Equation	263	364	68

Figure S1

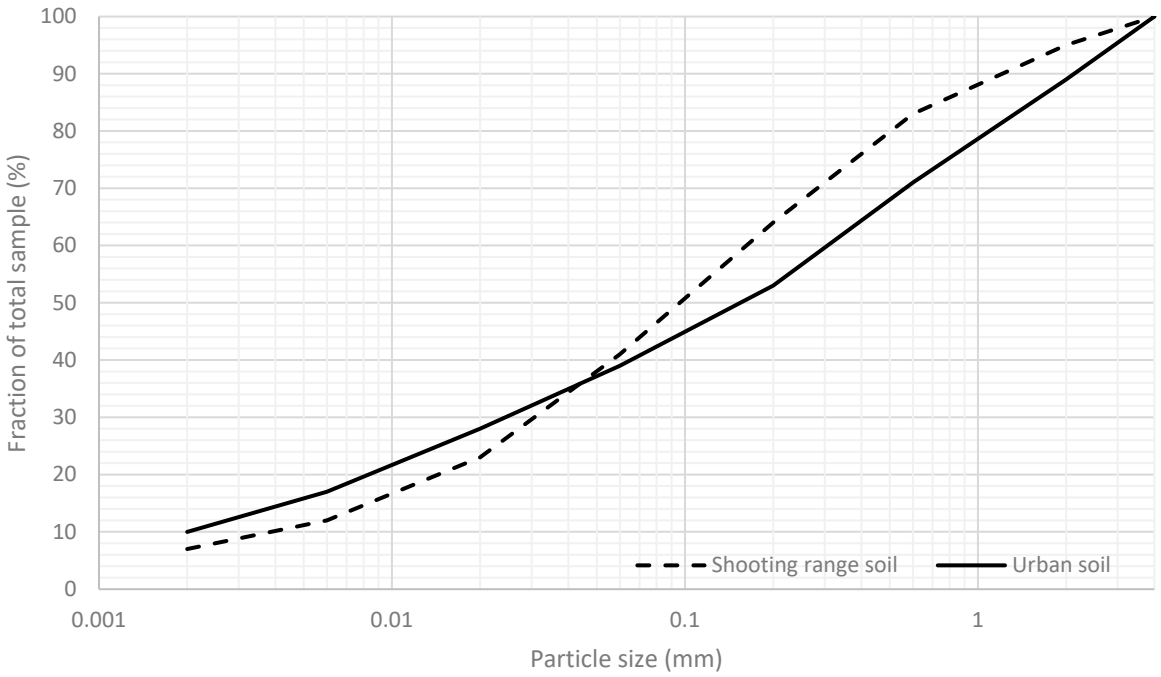

Figure S1: Particle size distribution curves.