Supporting info for

A critical evaluation of short columns for estimating the attachment efficiency of engineered nanomaterials in natural soils

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Formulae

Parameters

Table S1. Symbols, explanation and formulae for parameters used to calculate α

Symbol	Name	Formulae
t	Time	
z	Column depth	
θ	Porosity	
θ_{total}	Total porosity	$\frac{V_{added\ water}}{V_{column}}$
θ_{eff}	Effective porosity	
ρ	Bulk density	
D	Dispersivity	
$V_{\text{added water}}$	Water added to column during packing	
$V_{\text{column water}}$	Column volume	$\frac{\pi L d_{column}^{2}}{4}$
L	Column length	
C	Suspended ENM concentration	
C_0	Added ENM concentration	
S	ENM concentration attached to soil	
Q	Column flow rate	
U	Approach velocity	$\frac{4Q}{\pi d_{column}^{2}\theta}$
d_p	ENM diameter	
d_c	Collector diameter (grain size)	
d_{10}	Diameter than which 10 % of soil volume has lower grain size	
d_{50}	Diameter than which 50 % of soil volume has lower grain size	
A	Hamaker constant	
k	Boltzmann constant	
T	Temperature	
μ	Viscosity	
ρ_p	Density of the particle	
ρ_f	Density of the fluid	
g	Gravitational acceleration	
D_{diff}	Diffusion coefficient of the particle	$\frac{kT}{3\pi\mu d_p}$
γ		$\sqrt[3]{1-\theta}$
A_s		$\frac{2(1-\gamma^5)}{2-3\gamma+3\gamma^5-2\gamma^6}$
N_R	Aspect ratio	$\frac{d_p}{d_c}$
N_{Pe}	Peclet number	$\frac{Ud_c}{D_{diff}}$
N_{vdw}	Van der Waals number	$\frac{A}{kT}$
N_A	Attraction number	$\frac{A}{3\pi\mu d_p^2 U}$
N_G	Gravity number	$\frac{d_p^2(\rho_p-\rho_f)g}{18\mu U}$
N_{LO}	London number	$\frac{4}{3}N_A$

N_{Gi}		$\frac{1}{N_G+1}$

Models

1. Tufenkji and Elimelech (2004)[1] (Abbreviation: "TE")

$$\eta_0 = 2.4A_s^{1/3}N_R^{-0.081}N_{Pe}^{-0.715}N_{vdw}^{0.052} + 0.55A_sN_R^{1.676}N_A^{0.125} + 0.22N_R^{-0.24}N_G^{1.11}N_{vdw}^{0.053}$$

$$k = \frac{3(1-\theta)U}{2d_c\theta}\eta_0\alpha$$

2. Ma et al. (2009)[2] as corrected in [3] (Abbreviation: "MA2010")

$$\eta_0 = \gamma^2\left[2.3A_s^{1/3}N_R^{-0.028}N_{Pe}^{-0.66}N_A^{0.052} + 0.55A_sN_R^{1.8}N_A^{0.15} + 0.2N_R^{-0.047}N_G^{1.1}N_{Pe}^{0.053}N_A^{0.053}\right]$$

$$k = \frac{3(1-\theta)U}{2d_c\theta}\eta_0\alpha\left[\frac{3-\theta}{3-3\theta} - \frac{2(3-\theta)}{\pi(3-3\theta)}\cos^{-1}\left(\frac{3-3\theta}{3-\theta}\right)^{1/2} + \frac{2}{\pi}\sqrt{2\left(\frac{3-\theta}{3-3\theta}\right)^{1/2}-1}\right] = \frac{3(1-\theta)U}{2d_c\theta}\eta_0\alpha f(\theta)$$

where $f(\theta)$ a function of the porosity

3. Long and Hilpert [4] (Abbreviation: "LH")

$$\eta_0 = 15.56\frac{(1-\theta)^3}{\theta^2}N_R^{0.19}N_{Pe}^{-0.65} + 0.55A_sN_R^{1.675}N_A^{0.125} + 0.22N_R^{-0.24}N_G^{1.11}N_{vdw}^{0.053}$$

$$k = \frac{3(1-\theta)U}{2d_c\theta}\eta_0\alpha$$

4. Nelson and Ginn[5]

$$\eta_0 = \gamma^2\left[2.4A_s^{1/3}\left(\frac{N_{Pe}}{N_{Pe}+16}\right)N_{Pe}^{-0.68}N_{Lo}^{0.015}N_{Gi}^{0.8} + A_sN_R^{15/8}N_{Lo}^{1/8} + 0.7N_R^{-0.05}N_G\frac{N_{Gi}}{N_{Gi}+0.9}\right]$$

$$k = \frac{3(1-\theta)^{1/3}U}{2d_c\theta}\eta_0\alpha$$

Ma et al. (2013)[6] (Abbreviation: "MA2013")

$$\eta_0 = \gamma^2\left[\frac{8+4(1-\gamma^{1/3})A_s^{1/3}N_{Pe}^{1/3}}{8+(1-\gamma)N_{Pe}^{0.97}}N_{Lo}^{0.015}N_{Gi}^{0.8}N_R^{0.028} + A_sN_R^{15/8}N_{Lo}^{1/8} + 0.7N_R^{-0.05}N_G\frac{N_{Gi}}{N_{Gi}+0.9}\right]$$

$$k = \frac{3(1-\theta)U}{2d_c\theta}\eta_0\alpha f(\theta)$$

Derivation of model 1 for Hemisphere-in-cell geometry (used in Ma et al. (2010)[3] and Ma et al. (2013)[6])

By definition, the single-collector contact efficiency η is[7]

$$\eta = \frac{I}{AUC} \qquad \text{(eq. S1)}$$

With I the particle removal rate per collector, A the projected area of the collector including the fluid shell and C the local dispersed concentration of particles. In the case of a sphere-in-cell geometry, $A = \pi d_c^2/4$ but for the hemisphere-in-cell geometry[3]

$$A = \frac{\pi d_c^2}{4} f(\theta) \quad \text{(eq. S2)}$$

where $f(\theta)$ was defined earlier. The macroscopic particle removal is obtained by multiplying the removal rate by one collector with the number of collectors per control volume N_c[3]:

$$N_c = \frac{6(1-\theta)}{\pi d_c^3} A_c dz \quad \text{(eq. S3)}$$

where A_c is the surface perpendicular to the flow direction and z is the flow direction. The overall particle removal in the control volume is given by combining eqs. S1 and S2 to obtain the removal by one collector and multiplying this with N_c (eq. S3):

$$I = \frac{3(1-\theta)\eta f(\theta) UC}{2d_c} A_c dz \quad \text{(eq. S4)}$$

Considering I=-QdC with Q the volumetric flow, equal to UA_c, we can reorganise and integrate eq. S4 as

$$\int_{C_0}^{C_f} \frac{dC}{C} = - \frac{3(1-\theta)\eta f(\theta)}{2d_c} \int_0^L dz \quad \text{(eq. S5)}$$

where C_0 is the concentration at the column inlet, C_f is the particle concentration at the column outflow and L is the column length. Considering that $\eta = \alpha\eta_0$, Eq. S5 solves to

$$\alpha_{continuous} = - \frac{2d_c}{3(1-\theta) f(\theta) L \eta_0} lnR \quad \text{(eq. S6)}$$

with R the recovery.

Soil properties

Table S2: Properties of the soils used in the experiments as well as the methods used to obtain these properties.

Soil property		Lufa 2.2	Woburn	Dorsett	Chiltern	North Wales	Method	Ref
pH		6.49	6.41	4.08	7.63	4.77	Water	[8]
Texture (weight %)	Sand	67.25	75.56	91.8	29.5	57.7	Stokes settling	[8]
	Silt	17.51	11.81	4.7	46.8	29.7		
	Clay	15.23	12.62	3.5	23.7	12.6		
Total organic carbon (weight %)		1.53	0.61	2.18	3.26	9.63	Leco Trumac CN analyser	[8]
Cation exchange capacity (cmol kg^{-1})		13.3	13.3	7.83	22.94	33.32	Ammonium acetate extraction	[9]
ζ -potential (mV)		-25.3	-27.5	-43.7	-24	-25.9	Electrophoretic mobility ($\zeta_{n.d.}$)	See main text
		-14.4	-15.2	-23.9	-2.7	-6.3	Streaming potential ($\zeta_{dispersed}$)	
d_{10} (μm)		69.2	104.4	47.1	35.9	NA*	Dry-sieving	
		45.0	26.3	47.2	8.7	43.7	Mastersizer	
d_{50} (μm)		184.2	200.4	201.9	482.6	249.8	Dry-sieving	
		207.3	211.0	221.0	141.3	279.7	Mastersizer	
Amorphous Fe, Al and Mn minerals (mg kg^{-1})	Fe	1097.4	2706.9	208.2	1123.5	1588.9	Oxalate extraction	[10]
	Al	415.8	379.6	145.5	1471.9	787.0		
	Mn	120.1	130.0	0.8	259.6	27.4		
Quartz (weight %)		78.8	88.2	91.8	10.8	72.3	XRPD analysis	[11]
K-feldspar (weight %)		9.1	0.4	0	0.3	1		
Plagioclase (weight %)		2.1	0.9	0.3	0.6	0.7		
Calcite (weight %)		0.5	0.1	0.1	75.7	0		
Aragonite (weight %)		0	0	0	1.6	0		
Goethite (weight %)		0.9	3.4	0.5	0.5	0.3		
Illite (weight %)		1.2	0.6	0.3	0.0	2.1		
Kaolinite (weight %)		0.5	0.4	0.2	0.0	0.5		
Chlorite (weight %)		0.1	0.0	0.0	0.0	0.0		
2:1 clays (weight %)		2.8	3.6	0.4	6.2	2.2		
Amorphous (weight %)		4	2.4	6.4	4.3	20.8		

* Not available because a negative d_{10} was calculated from the available dry sieving data

Some additional results on soil properties are shown below such as the boxplots of total and effective variance for each of the individual soils (Figure S1), a comparison of some soil properties (Figure S2), and a comparison of the grain size distribution obtained using laser scattering and dry sieving (Figure S3).

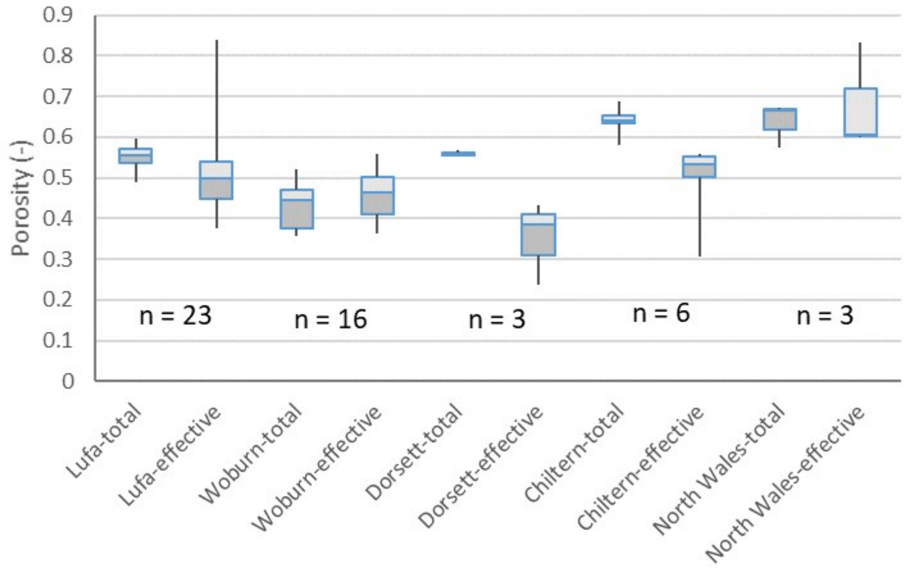

Figure S1. Boxplots of total and effective porosity for different soils showing also the number of data points for which these boxplots were calculated

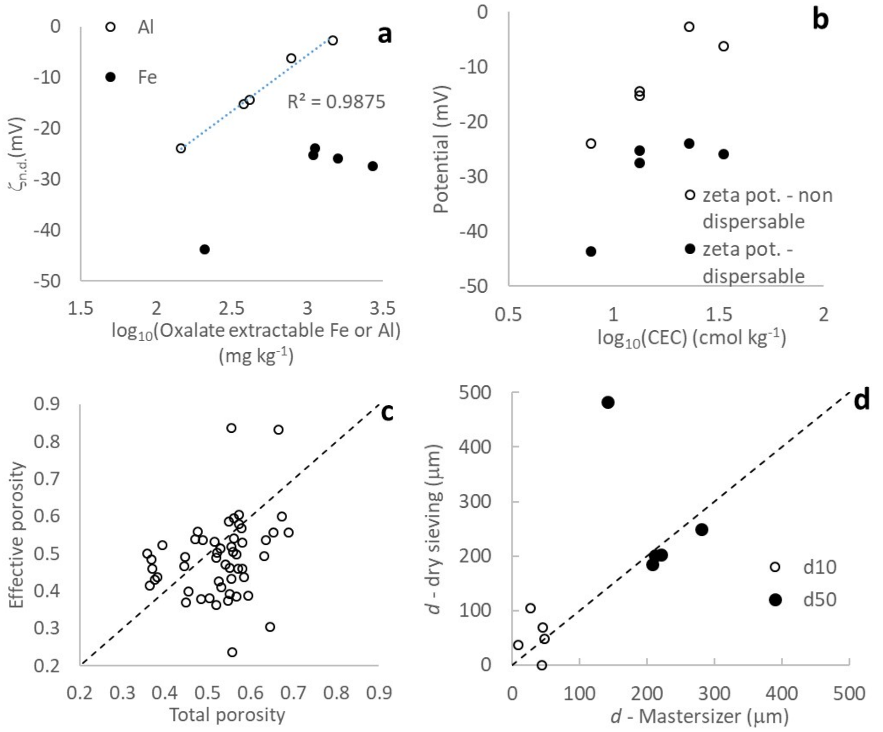

Figure S2. Interrelation of soil properties. a) ζ-potential of the non-dispersable soil fraction versus oxalate extractable Al or Fe; b) CEC versus ζ-potentials c) Effective versus total porosity showing also the 1:1 relation and d) d_{10} *or* d_{50} *determined using light scattering versus using dry sieving also showing the 1:1 relation.*

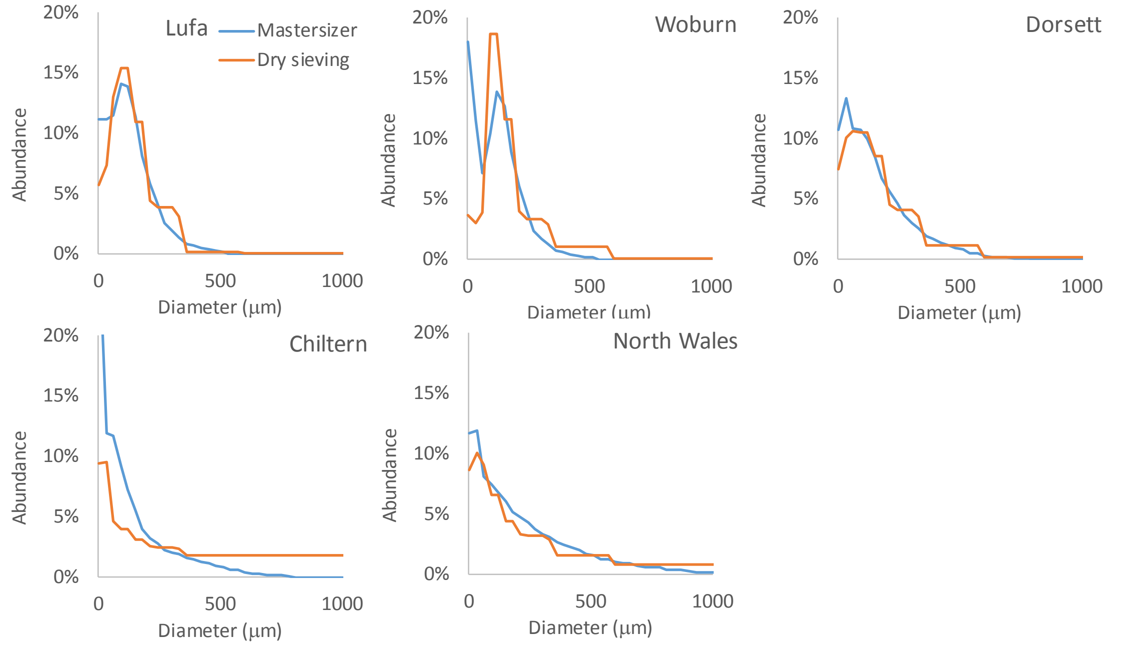

Figure S3. Grain size distributions determined using either mastersizer or dry sieving. Note that the distribution were rebinned to the same bin size of 50 µm.

spTOF-ICP-MS analysis

Table S3 shows the results of the spTOF-ICP-MS analysis that were done using 20 and 80 nm Au ENMs and saturated solutions of five soils. Shown are how much the saturated soil solution was diluted prior to mixing with diluted 20 nm or 80 nm Au suspensions and how many of the detected particle events of Al, Si, Ti, Mn and Fe, elements commonly found in mobile colloids, overlap with particle events of Au.

Table S3. Fraction of detected Au particle events that co-occur with particle events of common elements found in mobile colloids.

Au ENM diameter	Dilution Soil solution	Soil	% of particle events overlapping with Au					
			Al	Si	Ti	Mn	Fe	Sum
20	100	Chiltern	0	0	0	0	0	0
20	100	Chiltern	0	0	0	0	0	0
20	100	Dorset	0	0	0	0	0	0
20	10	Lufa	0	0	0	20	40	60
20	100	Lufa	10	0	0	0	10	20
20	100	North Wales	4	17	0	0	8	29
20	100	Woburn	0	0	0	0	13	13
20	100	Woburn	0	0	0	0	0	0
20	100	Woburn	0	0	0	0	0	0
20	1000	Woburn	0	0	0	0	29	29
80	100	Chiltern	0	0	0	0	0	0
80	100	Chiltern	2	8	0	0	0	10
80	200	Chiltern	3	7	1	0	2	13
80	10	North Wales	0	1	0	0	2	3
80	100	North Wales	0	1	0	0	1	2
80	100	North Wales	0	4	1	0	1	6
80	10	Woburn	1	4	0	0	3	7
80	10	Woburn	0	4	0	0	8	13
80	10	Woburn	1	1	0	0	4	5
80	10	Woburn	1	1	0	0	5	7
80	100	Woburn	1	3	0	0	5	9
80	100	Woburn	1	1	0	0	4	6

Figure S4 shows how the observed % of coincidences of Au with other elements (last column in Table S3) as a function of predicted fraction of coincidences in case of random arrival that was calculated as the joint (empirical) probability of a particle event occurring together with a Au particle event, summed over all elements other than Au. Most predicted probabilities are higher than the observed fractions, suggesting that very few detected coincidences are in fact heteroaggregates.

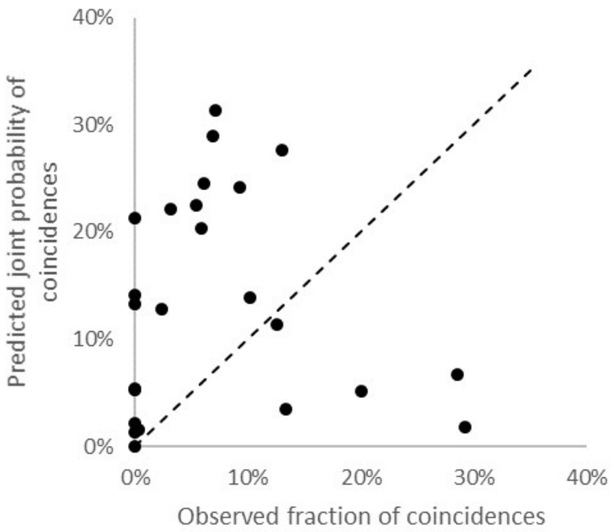

Figure S4. Total predicted probability of random coincidences of Au particle events as a function of observed fraction of total coincidences. The 1:1 relation is indicated as well.

Examples of BTC and associated fits

All figure below show experimental breakthrough curves (BTCs) and fitted models of inert tracer conductivity data on the left and ENM BTCs and fitted models on the right. All conductivity and concentration data is normalised (divided by input conductivity or ENM concentration). Elution time was recalculated into number of eluted pore volumes that were calculated using the effective porosity as deducted from inter tracer model fitting. The fitted model in the case of the inert tracer is a zero interaction model, whereas the three models that were fitted to ENM BTCs using Hydrus 1D are shown, i.e. a model assuming irreversible attachment only ("Attach"), a model assuming only straining ("Straining") and a model assuming that types of attachment sites exist where either irreversible or reversible sorption occurs ("2-sites").

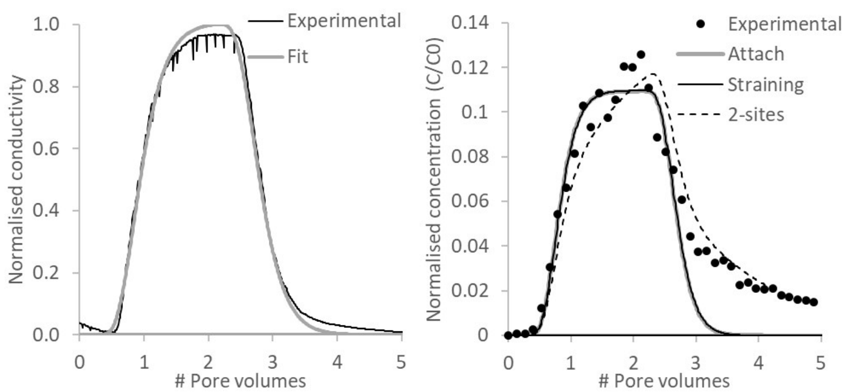

Figure S5. 23.22 mg kg^{-1} 20 nm Au ENMs eluting from a Lufa 2.2 soil column.

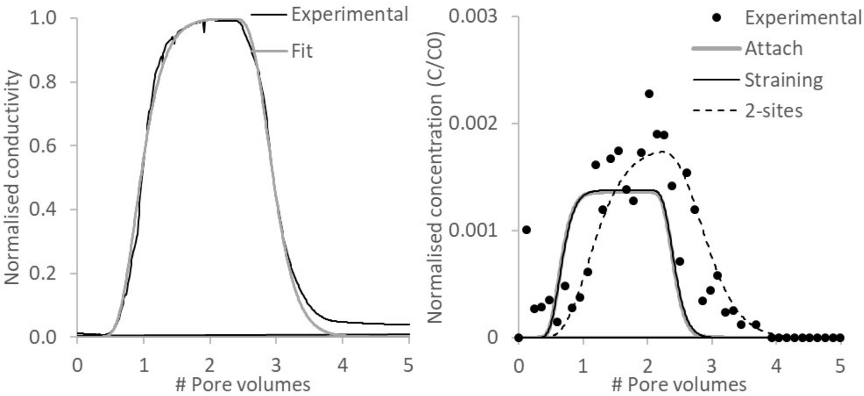

Figure S6. 33.95 mg kg^{-1} 20 nm Au ENMs eluting from a Woburn soil column.

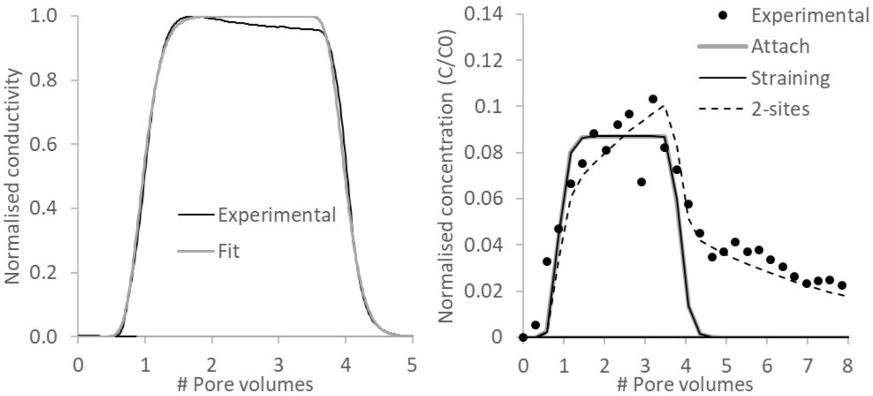

Figure S7. 2.65 mg kg^{-1} 80 nm Au ENMs eluting from a Dorsett soil column.

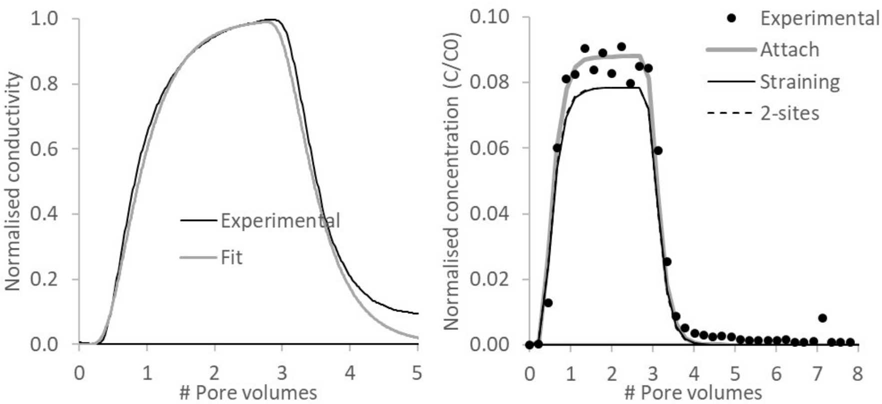

Figure S8. 1.66 mg kg^{-1} Ag_2S ENMs eluting from Chiltern soil columns. The 2 sites model overlaps with the straining model.

PLS1 Analysis

PLS is a more appropriate regression analysis than multiple linear regression in cases such as in the current study where many, often intercorrelating, predictor variables exist versus a relatively low (here $n = 52$) number of observations. The PLS approach is termed PLS1 because, in this case, there is only a single response variable y. In PLS1, the X (nxp) matrix of predictor values and a response vector y (nx1) are expressed in terms of a number (l) of latent variables, sometimes also called principal components in analogy with PCA, according to the following equations:

$X = TP^t + E$ Equation S1

$y = Tb + f$ Equation S2

Note that matrices are denoted using capital letters, whereas vectors are denoted using lower-case letters. In the equation above, the pxl matrix $\boldsymbol{P}$ describes the latent X-variables, called **loadings**, the **scores** are contained in the nxl matrix $\boldsymbol{T}$ and E (nxp) represents random errors. The y vector is related to the loadings via the regression coefficient vector b (1 x l) with some residual f. It is the purpose of PLS, given a number of latent variables l, to find the weights W (pxl) for which the scores $T = X$xW so that for each latent variable t_a, i.e. each column a of T and row a of P, the covariance between y and t_a is maximal. An estimated $X_{est} = t_a p_a$' is then subtracted from E producing a new variable X_{a+1} for which the procedure is repeated producing a new latent variable (and thus a new column, reps. Row for T and P). This procedure is repeated until the set value of l is reached or E equals the zero matrix.

Prior to PCA and PLS, all predictor and response variables were logarithmically (base 10) transformed if skewness was larger than 1 (Table S4). If skewness was larger than 0.5, transformation was only done if this operation reduced skewness. The untransformed variable was also used when logarithmic or any other (arcsin, square root) transformation did not result in any improvement in skewness. Transformed variables were subsequently centered and rescaled producing z-scores.

Table S4. Independent variables and how these were transformed prior to PLS1 analysis.

Independent Variables	Transformation	Skewness of transformed variable
Column length	None	0.35
Approach velocity	Log_{10}	-0.07
Effective porosity	Log_{10}	-0.30
Total porosity	None	-0.47
Dispersivity	Log_{10}	0.78
ENP size	None	0.38
ENP concentration	Log_{10}	0.20
pH*	None	1.38
Sand content*	None	-1.14
Clay content	None	0.10
d_{10} (Mastersizer)	None	-0.23
d_{50} (Mastersizer)	None	-0.24
Total carbon	Log_{10}	0.76
$\zeta_{dispersed}$*	None	-3.40
$\zeta_{n.d.}$	None	0.78
Oxalate extractable Fe	None	0.47
Oxalate extractable Al	Log_{10}	0.78

* No transformation resulted in reduced skewness

The optimal number of latent variables l was found in this study by removing one observation from the data set and then predicting the response for this variable based on a PLS analysis with l latent variables of the remaining experiments. Predicted variables for all experiments can thus be obtained and a mean squared error of prediction (MSEP) with observed variables can thus be obtained as a function of l. The optimal l was thus the number where the highest MSEP was found.

p (the number of predictor variables) was also optimized because a high number tends to spoil PLS analyses reducing their consistency and introducing unnecessary variance. In a first step, intercorrelating variables were divided in groups based on their PCA scores and if a physical explanation could be found for the observed covariance. One predictor variable representative for each group was retained. A PLS analysis was subsequently run with the remaining predictors and the variable importance in projection (VIP) of each variable was then calculated according to equation S3.

$$VIP_j = \sqrt{\frac{p\sum_{k=1}^{l}\left[SS_k\left(\frac{w_{jk}}{\|w_k\|}\right)^2\right]}{\sum_{k=1}^{l}SS_k}}$$ Equation S3

Here, p is the number of predictor variables, l is the number of latent variables, SS is the sum of squares relative to the k^{th} latent variable calculated as $b_k^2 t_k^2$ where b_k and t_k are the predictor scores and response loadings, respectively. w_{jk} is the weight of the j'th predictor variable, $\|w_k\|$ is the Euclidian norm of the weight vector of the k^{th} latent variable. Variables having VIP < 1 were removed and PLS1 was repeated, including optimizing the number of latent variables, with the z-scores of selected variables.

Table S5 summarizes the results of the PLS1 analysis, showing the % variance explained by each latent variable that was retained after cross validation and selection of predictor variables.

Table S5 summary of the PLS1 results including for each model, the number of retained data points, the optimal number of components (latent variables),the total % of the variance explained and the weights different variables have on the first latent variable.

Porosity	Diameter	Correlation	Model	n	Total variance explained	# com-po-nents	Weights on first comp by NM and soil props									Weights on first comp by operational parameters					
							NM size	pH	Sand	Clay	TOC	Zeta Poten-tial	Streaming Potential	Oxalate Fe	Oxalate Al	Column length	C0	dc	θ	Dis-persivity	Approach velocity
Effective	Single d10	TE	Continuous	34	26%	1	0.059	0	0	0	0	0	0	0	0	0	0.080	0.081	0	0	0.057
			Pulse	34	27%	1	0.059	0	0	0	0	0	0	0	0	0	0.081	0.079	0	0	0.058
			Attachment	47	42%	2	0	0.029	0	0	-0.036	0.030	0	0	0	0.045	0.045	0.033	0	0	0.034
			Straining	47	36%	1	0	0.033	0	0	-0.043	0.028	0	0	0	0.052	0.043	0	0	0	0.034
			2-sites	44	8%	1	0	0	0	0	0.054	0	0	-0.044	0	0	0.048	0.058	0.052	0	0
		LH	Continuous	34	28%	3	0	0	0	0	0	0	0	0	0	0	0.078	0.080	0.050	0	0.060
			Pulse	34	39%	4	0	0	0	0	0	0	0	0	0	0	0.079	0.078	0.050	0	0.061
			Attachment	47	49%	2	0	0	0	0	0	0.040	0	0	0	0.039	0.045	0.041	0.051	0	0.037
			Straining	47	33%	2	0	0.030	0	0	-0.032	0.035	0	0	0	0.045	0.041	0	0.039	0	0.035
			2-sites	44	11%	1	0	0	0	0	0.054	0	0	-0.041	0	0	0.039	0.055	0.067	0	0
		MA2010	Continuous	34	26%	1	0.058	0	0	0	0	0	0	0	0	0	0.080	0.081	0	0	0.058
			Pulse	34	27%	1	0.058	0	0	0	0	0	0	0	0	0	0.081	0.079	0	0	0.059
			Attachment	47	42%	2	0	0.029	0	0	-0.035	0.031	0	0	0	0.045	0.045	0.034	0	0	0.035
			Straining	47	37%	2	0	0.033	0	0	-0.042	0.029	0	0	0	0.052	0.043	0	0	0	0.034
			2-sites	44	8%	1	0	0	0	0	0.054	0	0	-0.043	0	0	0.048	0.058	0.053	0	0
		NG	Continuous	34	24%	1	0.054	0	0	0	0	0	0	0	0	0	0.082	0.081	0	0	0.059
			Pulse	34	25%	1	0.054	0	0	0	0	0	0	0	0	0	0.083	0.079	0	0	0.060
			Attachment	47	43%	2	0	0.031	0	0	-0.036	0.031	0	0	0	0.045	0.045	0.031	0	0	0.034
			Straining	47	38%	2	0	0.031	0	0	-0.038	0.026	0	0.025	0	0.047	0.038	0	0	0	0.030
			2-sites	44	8%	1	0	0	0	0	0.056	0	0	-0.043	0	0	0.049	0.056	0.054	0	0
		MA2013	Continuous	34	20%	1	0.052	0	0	0	0	0	0	0	0	0	0.083	0.081	0	0	0.060
			Pulse	34	22%	1	0.052	0	0	0	0	0	0	0	0	0	0.084	0.078	0	0	0.061
			Attachment	47	43%	2	0	0.033	0	0	-0.036	0.032	0	0	0	0.046	0.045	0.029	0	0	0.034
			Straining	47	38%	2	0	0.032	0	0	-0.038	0.026	0	0.026	0	0.047	0.038	0	0	0	0.029
			2-sites	44	8%	1	0	0	0	0	0.057	0	0	-0.043	0	0	0.050	0.054	0.055	0	0
	Single d50	TE	Continuous	34	26%	1	0.050	0	0	0	0	0	0	0	0	0.068	0.069	0	0	0.042	0.062
			Pulse	34	27%	1	0.047	0	0	0	0	0	0	0	0	0.068	0.066	0	0	0.050	0.061
			Attachment	47	42%	2	0	0.030	0	0	-0.035	0	0	0.049	0	0.050	0	0	0	0.046	0
			Straining	47	36%	1	0	0.030	0	0	-0.038	0	0	0.049	0	0.050	0	0	0	0.042	0
			2-sites	44	8%	1	0	0	-0.030	0	0.041	0	0.043	0	0.038	0	0	0	0.035	0.030	0
		LH	Continuous	34	34%	3	0	0	0	0	0	0	0	0.036	0	0.047	0.058	0	0.068	0	0.060
			Pulse	34	39%	4	0	0	0	0	0	0	0	0.038	0	0.048	0.057	0	0.066	0	0.059
			Attachment	47	49%	2	-0.039	0	0	0	0	0	0	0.058	0	0.055	0	0	0.044	0.053	0
			Straining	47	34%	2	-0.035	0	0	0	-0.033	0	0	0.052	0	0.050	0	0	0	0.043	0
			2-sites	44	12%	1	0	0	-0.029	0	0.045	0	0.042	0	0.037	0	0	0	0.055	0	0
		MA2010	Continuous	34	26%	1	0.049	0	0	0	0	0	0	0	0	0.067	0.069	0	0	0.041	0.063
			Pulse	34	27%	1	0.047	0	0	0	0	0	0	0	0	0.068	0.067	0	0	0.049	0.061
			Attachment	47	42%	2	0	0.030	0	0	-0.035	0	0	0.049	0	0.050	0	0	0	0.046	0
			Straining	47	37%	2	0	0.030	0	0	-0.038	0	0	0.050	0	0.050	0	0	0	0.042	0
			2-sites	44	8%	1	0	0	-0.030	0	0.041	0	0.042	0	0.038	0	0	0	0.036	0.030	0
		NG	Continuous	34	25%	1	0.041	0	0	0	0	0	0	0.035	0	0.063	0.063	0	0	0.040	0.056
			Pulse	34	26%	1	0.038	0	0	0	0	0	0	0.036	0	0.063	0.060	0	0	0.047	0.054
			Attachment	47	43%	2	0	0.029	0	0	-0.035	0	0	0.049	0	0.050	0	0	0	0.046	0
			Straining	47	38%	2	0	0.029	0	0	-0.038	0	0	0.050	0	0.050	0	0	0	0.042	0
			2-sites	44	8%	1	0	0	-0.030	0	0.041	0	0.042	0	0.038	0	0	0	0.036	0.030	0
		MA2013	Continuous	34	24%	1	0.039	0	0	0	0	0	0	0.036	0	0.062	0.063	0	0	0.039	0.057
			Pulse	34	25%	1	0.036	0	0	0	0	0	0	0.037	0	0.062	0.061	0	0	0.046	0.055
			Attachment	47	43%	2	0	0.029	0	0	-0.035	0	0	0.050	0	0.050	0	0	0	0.046	0
			Straining	47	37%	2	0	0.030	0	0	-0.038	0	0	0.050	0	0.050	0	0	0	0.042	0
			2-sites	44	8%	1	0	0	-0.030	0	0.041	0	0.042	0	0.038	0	0	0	0.037	0.030	0

Porosity	Diameter	Correlation	Model	n	Total variance explained	# components	Weights on first comp by NM and soil props									Weights on first comp by operational parameters					
							NM size	pH	Sand	Clay	TOC	Zeta Potential	Streaming Potential	Oxalate Fe	Oxalate Al	Column length	C0	dc	θ	Dispersivity	Approach velocity
Effective	Distributed	TE	Continuous	34	26%	1	0.048	0	0	0	0	0	0	0	0	0.065	0.069	-0.047	0	0.064	0.070
			Pulse	34	27%	1	0.045	0	0	0	0	0	0	0	0	0.064	0.066	-0.050	0	0.071	0.067
			Attachment	47	43%	2	0	0	0	0	0	0	0	0.053	0	0.048	0	-0.052	0	0.054	0
			Straining	47	36%	1	0	0	0	0	0	0	0	0.056	0	0.050	0	-0.050	0	0.048	0
			2-sites	44	8%	1	0	0	-0.034	0	0	0	0.034	0	0.035	0.048	0.037	0	0	0.060	0.042
		LH	Continuous	34	34%	2	0	0	0	0	0	0.008	0	0	0	0.058	0.066	-0.042	0	0.062	0.081
			Pulse	34	39%	4	0	0	0	0	0	0.009	0	0	0	0.057	0.063	-0.044	0	0.067	0.077
			Attachment	47	48%	2	0	0	0	0	0	0	0	0.052	0	0.048	0	-0.047	0.043	0.057	0
			Straining	47	37%	2	-0.034	0	0	0	0	0	0	0.051	0	0.046	0	-0.043	0	0.046	0
			2-sites	44	11%	1	0	0	-0.035	0	0	0	0.034	0	0.035	0.046	0.037	0	0	0.059	0.045
		MA2010	Continuous	34	26%	1	0.048	0	0	0	0	0	0	0	0	0.064	0.069	-0.046	0	0.064	0.071
			Pulse	34	27%	1	0.045	0	0	0	0	0	0	0	0	0.064	0.066	-0.049	0	0.070	0.068
			Attachment	47	42%	2	0	0	0	0	0	0	0	0.053	0	0.048	0	-0.051	0	0.054	0
			Straining	47	37%	2	0	0	0	0	0	0	0	0.056	0	0.050	0	-0.050	0	0.048	0
			2-sites	44	8%	1	0	0	-0.035	0	0	0	0.034	0	0.035	0.048	0.037	0	0	0.060	0.042
		NG	Continuous	34	25%	1	0.043	0	0	0	0	0	0	0	0	0.065	0.068	-0.049	0	0.066	0.069
			Pulse	34	27%	1	0.040	0	0	0	0	0	0	0	0	0.064	0.065	-0.052	0	0.073	0.066
			Attachment	47	43%	2	0	0	0	0	0	0	0	0.053	0	0.048	0	-0.052	0	0.054	0
			Straining	47	38%	2	0	0	0	0	0	0	0	0.056	0	0.050	0	-0.051	0	0.048	0
			2-sites	44	8%	1	0	0	-0.034	0	0	0	0.034	0	0.035	0.048	0.037	0	0	0.060	0.042
		MA2013	Continuous	34	24%	1	0.041	0	0	0	0	0	0	0	0	0.064	0.069	-0.048	0	0.066	0.070
			Pulse	34	25%	1	0.039	0	0	0	0	0	0	0	0	0.064	0.065	-0.051	0	0.072	0.067
			Attachment	47	43%	2	0	0	0	0	0	0	0	0.053	0	0.048	0	-0.052	0	0.054	0
			Straining	47	37%	2	0	0	0	0	0	0	0	0.056	0	0.050	0	-0.050	0	0.048	0
			2-sites	44	8%	1	0	0	-0.034	0	0	0	0.034	0	0.035	0.048	0.037	0	0	0.060	0.042
Total	Single d10	TE	Continuous	34	33%	2	0.055	0	0	0	0	0	0	-0.041	0	0	0.070	0.076	0	0	0.050
			Pulse	34	35%	2	0.055	0	0	0	0	0	0	-0.040	0	0	0.071	0.074	0	0	0.052
			Attachment	47	44%	2	0	0.036	0	0	-0.039	0	0	0	0	0.047	0.051	0.041	0	0	0.036
			Straining	47	36%	1	0	0.034	0	0	-0.040	0	0	0	0	0.047	0.040	0	-0.032	0	0.030
			2-sites	44	8%	1	0.041	0	0	0	0.056	0	0	-0.052	0	0	0.042	0.058	0	0	0
		LH	Continuous	34	45%	4	0.041	0	0	0	0	0	0	-0.064	0	0	0.057	0.080	0	0	0.045
			Pulse	34	44%	3	0.041	0	0	0	0	0	0	-0.064	0	0	0.058	0.078	0	0	0.046
			Attachment	47	39%	2	0	0.038	0	0	0	0.035	0	0	0	0.031	0.050	0.054	0	0	0.038
			Straining	47	34%	2	0	0.037	0	0	-0.030	0.029	0	0	0	0.040	0.042	0.027	0	0.029	0.034
			2-sites	44	13%	1	0	0	0	0	0.056	0	0	-0.053	0	0	0	0.052	0.034	0	0
		MA2010	Continuous	34	33%	2	0.055	0	0	0	0	0	0	-0.042	0	0	0.069	0.076	0	0	0.050
			Pulse	34	35%	2	0.055	0	0	0	0	0	0	-0.041	0	0	0.070	0.074	0	0	0.051
			Attachment	47	44%	2	0	0.037	0	0	-0.038	0	0	0	0	0.046	0.052	0.042	0	0	0.037
			Straining	47	36%	1	0	0.034	0	0	-0.040	0	0	0	0	0.047	0.041	0	-0.031	0	0.031
			2-sites	44	9%	1	0.041	0	0	0	0.056	0	0	-0.052	0	0	0.041	0.057	0	0	0
		NG	Continuous	34	31%	2	0.051	0	0	0	0	0	0	-0.044	0	0	0.070	0.076	0	0	0.051
			Pulse	34	33%	2	0.051	0	0	0	0	0	0	-0.043	0	0	0.071	0.074	0	0	0.053
			Attachment	47	44%	2	0	0.040	0	0	-0.038	0	0	0	0	0.047	0.051	0.038	0	0	0.036
			Straining	47	36%	1	0	0.034	0	0	-0.038	0	0	0	0	0.045	0.038	0	-0.030	0.027	0.029
			2-sites	44	8%	1	0.038	0	0	0	0.059	0	0	-0.053	0	0	0.042	0.056	0	0	0
		MA2013	Continuous	34	30%	2	0.049	0	0	0	0	0	0	-0.044	0	0	0.071	0.076	0	0	0.051
			Pulse	34	32%	2	0.049	0	0	0	0	0	0	-0.043	0	0	0.073	0.074	0	0	0.053
			Attachment	47	45%	2	0	0.041	0	0	-0.039	0	0	0	0	0.047	0.051	0.036	0	0	0.035
			Straining	47	36%	1	0	0.034	0	0	-0.038	0	0	0	0	0.045	0.038	0	-0.030	0.027	0.028
			2-sites	44	8%	1	0.037	0	0	0	0.060	0	0	-0.053	0	0	0.044	0.055	0	0	0

Porosity	Diameter	Correlation	Model	n	Total variance explained	# com-po-nents	Weights on first comp by NM and soil props: NM size	pH	Sand	Clay	TOC	Zeta Poten-tial	Streaming Potential	Oxalate Fe	Oxalate Al	Weights on first comp by operational parameters: Column length	C0	dc	θ	Dis-persivity	Approach velocity
Total	Single d50	TE	Continuous	34	33%	2	0.056	0	0	0	0	0	0	0	0	0.061	0.068	0	0.000	0.051	0.063
			Pulse	34	35%	2	0.053	0	0	0	0	0	0	0	0	0.062	0.065	0	0	0.059	0.061
			Attachment	47	44%	2	0	0.027	0	0	-0.028	0	0	0.037	0	0.039	0	-0.027	-0.025	0.045	0
			Straining	47	36%	1	0	0.027	0	0	-0.032	0	0	0.040	0	0.042	0	0	-0.030	0.041	0
			2-sites	44	8%	1	0	0	-0.035	0	0.043	0	0.044	0	0.040	0	0	0	0	0.032	0
		LH	Continuous	34	39%	3	0	0	0	0	0.078	0	0	0	0	0	0.071	0	0	0.086	0.086
			Pulse	34	40%	3	0	0	0	0	0.074	-0.007	0	0	0	0	0.068	0	0	0.096	0.083
			Attachment	47	39%	2	0	0.038	0	0	0	0	0	0.037	0	0.038	0	-0.042	0	0.063	0
			Straining	47	34%	2	0	0.038	0	0	0	0	0	0.045	0	0.045	0	-0.037	0	0.057	0
			2-sites	44	12%	1	0	0	-0.036	0	0.047	0	0.042	0	0.039	0	0	0	0	0.027	0
		MA2010	Continuous	34	33%	2	0.057	0	0	0	0	0	0	0	0	0.059	0.068	0	0	0.052	0.064
			Pulse	34	35%	2	0.054	0	0	0	0	0	0	0	0	0.060	0.065	0	0	0.060	0.062
			Attachment	47	44%	2	0	0.030	0	0	-0.031	0	0	0.041	0	0.043	0	-0.030	0	0.050	0
			Straining	47	36%	1	0	0.028	0	0	-0.032	0	0	0.040	0	0.042	0	0	-0.030	0.041	0
			2-sites	44	9%	1	0	0	-0.035	0	0.043	0	0.043	0	0.040	0	0	0	0	0.032	0
		NG	Continuous	34	32%	2	0	0	0	0	0	0	0	0	0	0.061	0.068	0	0	0.054	0.063
			Pulse	34	34%	2	0	0	0	0	0	0	0	0	0	0.062	0.065	0	0	0.061	0.061
			Attachment	47	44%	2	0	0.027	0	0	-0.028	0	0	0.037	0	0.039	0	-0.027	-0.025	0.045	0
			Straining	47	36%	1	0	0.027	0	0	-0.032	0	0	0.040	0	0.042	0	0	-0.030	0.041	0
			2-sites	44	8%	1	0	0	-0.035	0	0.043	0	0.043	0	0.040	0	0	0	0	0.033	0
		MA2013	Continuous	34	32%	2	0.051	0	0	0	0	0	0	0	0	0.060	0.069	0	0	0.055	0.063
			Pulse	34	34%	2	0.047	0	0	0	0	0	0	0	0	0.061	0.066	0	0	0.062	0.061
			Attachment	47	44%	2	0	0.030	0	0	-0.031	0	0	0.041	0	0.043	0	-0.030	0	0.050	0
			Straining	47	36%	1	0	0.028	0	0	-0.032	0	0	0.040	0	0.042	0	0	-0.030	0.041	0
			2-sites	44	8%	1	0	0	-0.035	0	0.044	0	0.043	0	0.040	0	0	0	0	0.032	0
	Distributed	TE	Continuous	34	33%	2	0.056	0	0	0	0	-0.020	0	0	0	0.056	0.070	-0.050	0	0.077	0.071
			Pulse	34	34%	2	0.052	0	0	0	0	-0.016	0	0	0	0.055	0.065	-0.053	0	0.083	0.068
			Attachment	47	44%	2	0	0	0	0	0	0	0	0.047	0	0.041	0	-0.059	0	0.060	0
			Straining	47	36%	1	0	0	0	0	0	0	0	0.052	0	0.045	0	-0.056	0	0.052	0
			2-sites	44	8%	1	0	0	-0.035	0.029	0	0	0.033	0	0.034	0.030	0	0	0	0.052	0.030
		LH	Continuous	34	39%	3	0	0	0	0	0	-0.010	0	0	0	0	0.059	-0.046	0	0.109	0.089
			Pulse	34	40%	3	0	0	0	0	0	-0.006	0	0	0	0	0.055	-0.050	0	0.113	0.083
			Attachment	47	40%	2	0	0	0	0	0	0	0.040	0	0.041	0	0	-0.054	0	0.060	0
			Straining	47	34%	2	0	0	0	0	0	0	0.032	0.039	0.033	0.032	0	-0.049	0	0.050	0
			2-sites	44	12%	1	0	0	-0.037	0.030	0	0	0.033	0	0.034	0	0	0	0	0.047	0.026
		MA2010	Continuous	34	33%	2	0.058	0	0	0	0	-0.020	0	0	0	0.053	0.070	-0.049	0	0.078	0.072
			Pulse	34	35%	2	0.054	0	0	0	0	-0.016	0	0	0	0.053	0.066	-0.053	0	0.085	0.068
			Attachment	47	44%	2	0	0	0	0	0	0	0	0.046	0	0.041	0	-0.059	0	0.060	0
			Straining	47	36%	1	0	0	0	0	0	0	0	0.052	0	0.045	0	-0.056	0	0.053	0
			2-sites	44	9%	1	0	0	-0.036	0.029	0	0	0.033	0	0.034	0.029	0	0	0	0.051	0.030
		NG	Continuous	34	32%	2	0.051	0	0	0	0	-0.020	0	0	0	0.055	0.069	-0.052	0	0.079	0.070
			Pulse	34	34%	2	0.047	0	0	0	0	-0.016	0	0	0	0.055	0.064	-0.055	0	0.086	0.066
			Attachment	47	44%	2	0	0	0	0	0	0	0	0.046	0	0.041	0	-0.059	0	0.060	0
			Straining	47	36%	1	0	0	0	0	0	0	0	0.052	0	0.045	0	-0.056	0	0.053	0
			2-sites	44	8%	1	0	0	-0.035	0.029	0	0	0.033	0	0.034	0.030	0	0	0	0.052	0.030
		MA2013	Continuous	34	32%	2	0.051	0	0	0	0	-0.019	0	0	0	0.053	0.069	-0.052	0	0.082	0.071
			Pulse	34	34%	2	0.047	0	0	0	0	-0.016	0	0	0	0.052	0.065	-0.055	0	0.088	0.067
			Attachment	47	44%	2	0	0	0	0	0	0	0	0.046	0	0.040	0	-0.059	0	0.060	0
			Straining	47	36%	1	0	0	0	0	0	0	0	0.052	0	0.045	0	-0.056	0	0.053	0
			2-sites	44	9%	1	0	0	-0.036	0.029	0	0	0.033	0	0.034	0.029	0	0	0	0.052	0.029

All metadata

Table S6. All metadata of the column experiments

	Soil	Column length	Flow rate	Total porosity	effective porosity	NP core	NP size	C_0	mass recovery	Dis-persivity
		cm	mL min^{-1}	-	-	-	nm	mg kg^{-1}	-	cm
1	Chiltern	4.6	0.2219	0.645	0.3049	Au	80	3.96	0.0594	1.214
2	Chiltern	2.7	0.4000	0.580	0.5305	Au	20	1.56	0.7320	0.1487
3	Chiltern	3.3	0.4000	0.654	0.5575	Ag_2S	27	2.08	0.0214	0.6874
4	Chiltern	3.3	0.4000	0.689	0.5582	Au	20	1.68	0.8607	0.3264
5	Chiltern	2.7	0.4000	0.636	0.5366	Au	80	1.23	1.0703	0.4772
6	Chiltern	3.7	0.4000	0.632	0.4939	Ag_2S	27	1.66	0.0912	0.5906
7	Dorset	3.8	0.2563	0.557	0.2365	Au	80	2.65	0.1411	9.80E-02
8	Dorset	3.7	0.4000	0.566	0.3852	Au	20	1.10	1.0123	0.2138
9	Dorset	3.3	0.4000	0.556	0.4324	Ag_2S	27	2.45	0.6888	0.1143
10	Lufa 2.2	6	0.4183	0.488	0.5377	Au	20	23.22	0.1401	0.3212
11	Lufa 2.2	5.4	0.5522	0.560	0.5414	Au	80	296.47	0.3697	0.3973
12	Lufa 2.2	4.3	0.5564	0.561	0.5953	Au	80	16.46	0.6169	0.1152
13	Lufa 2.2	4.2	0.5241	0.568	0.4997	Au	20	95.92	0.6267	0.1486
14	Lufa 2.2	3.8	0.3730	0.579	0.5687	Au	80	4.22	1.4660	0.08604
15	Lufa 2.2	3.9	0.4935	0.519	0.4888	Au	80	1.65	0.6351	0.07586
16	Lufa 2.2	4.25	0.3780	0.559	0.5047	Au	20	22.79	1.0373	0.1732
17	Lufa 2.2	3.5	0.3914	0.552	0.4621	Au	80	36.98	1.6653	0.04795
18	Lufa 2.2	3.8	0.3863	0.580	0.4602	Au	80	8.46	2.7883	0.08915
19	Lufa 2.2	3.9	0.3685	0.584	0.4378	Au	80	6.02	2.3583	0.1259
20	Lufa 2.2	4.7	0.6000	0.542	0.4726	Au	20	4.61	0.9923	0.1405
21	Lufa 2.2	4.7	0.6000	0.529	0.5147	Au	20	0.96	1.2126	0.1136
22	Lufa 2.2	3.5	0.4000	0.532	0.4115	Ag_2S	27	0.83	0.9186	0.3718
23	Lufa 2.2	4.6	0.4000	0.526	0.4266	Au	80	1.39	1.2222	0.2097
24	Lufa 2.2	3.8	0.4000	0.516	0.5325	Ag_2S	27	0.99	1.1797	0.2454
25	Lufa 2.2	4.6	0.5230	0.571961347	0.5798	Au	80	12.65	0.4892	0.08169
26	Lufa 2.2	3.9	0.1924	0.550248425	0.3926	Au	80	2.49	0.9971	0.07876
27	Lufa 2.2	3.9	0.4087	0.594857515	0.3883	Au	80	14.04	0.4311	0.08378
28	Lufa 2.2	3.4	0.2149	0.571549742	0.4593	Au	20	24.01	1.2664	0.05004
29	Lufa 2.2	3.9	0.4218	0.546435236	0.3747	Au	20	17.31	0.0276	0.1172
30	Lufa 2.2	3.8	0.4000	0.555829373	0.8381	Au	20	1.17	1.1578	0.2137
31	Lufa 2.2	3.8	0.4000	0.555829373	0.5188	Au	20	1.17	1.2704	0.1565
32	Lufa 2.2	4.6	0.5564	0.54826802	0.587	Au	80	4.60	NA	0.1474
33	North Wales	4	0.4000	0.672626987	0.60006	Au	80	4.50	0.4643	0.1275
34	North Wales	3.5	0.4000	0.573	0.6047	Au	20	1.53	1.4618	0.1069
35	North Wales	2.8	0.4000	0.665	0.8319	Ag_2S	27	1.71	0.7328	0.06512
36	Woburn	6	0.5662	0.447	0.4928	Au	20	20.22	0.0050	0.3164
37	Woburn	5.7	0.5250	0.445	0.467	Au	20	8.79	0.0322	0.3123
38	Woburn	5.4	0.5753	0.469	0.54	Au	20	18.07	0.0059	0.35
39	Woburn	5.7	0.5667	0.449	0.3694	Au	20	0.68	0.1974	1.113
40	Woburn	5.7	0.5615	0.394	0.5232	Au	80	14.41	0.0018	2.38E-01
41	Woburn	5.5	0.6122	0.369	0.4603	Au	80	5.92	0.0023	0.1535

42	Woburn	5.4	0.2177	0.369	0.4849	Au	20	5.27	0.0250	0.206
43	Woburn	5.6	0.4229	0.381	0.4381	Au	20	7.10	0.0280	0.1834
44	Woburn	5.3	0.4103	0.376	0.4304	Au	80	6.67	0.2440	0.125
45	Woburn	5.9	0.1600	0.364	0.4141	Au	20	25.46	0.0281	0.1214
46	Woburn	5.8	0.3703	0.358	0.501	Au	80	8.77	0.0004	0.1318
47	Woburn	6.3	0.6172	0.477	0.5589	Au	20	33.95	0.0021	2.71E-01
48	Woburn	5.5	0.4580	0.455	0.39999	Au	20	35.81	2.8710	9.88E-02
49	Woburn	3.7	0.3386	0.519	0.3629	Au	80	0.09	1.0127	0.1354
50	Woburn	4.3	0.4000	0.521	0.5032	Au	20	0.98	0.9998	0.07877
51	Woburn	4.1	0.4000	0.503	0.3813	Ag_2S	27	1.28	0.5075	0.1439

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