Supplementary Information

Co-Transport of Polybromodiphenyl Ethers and Soil Nanoparticles in Saturated Porous Media: Implications for the Risks of Polybromodiphenyl Ether Spreading in Groundwaters

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Table S1. Protocols of column experiments.

Column no. [a]	Background solution	Type of soil nanoparticles	Type of porous media	Ionic strength (mM)	Length (cm)	Bulk density (g/cm^3)	Porosity [b] (-)	Pore-water Velocity (m/d)	Soil nanoparticles conc.(mg/L)
1	GW	BLK	Quartz sand	3.78	18.0	1.49	0.44	10	106.8
1#	GW	BLK	Quartz sand	3.78	18.0	1.49	0.44	10	107.0
2	GW	PUR	Quartz sand	3.78	18.4	1.46	0.45	10	101.7
2#	GW	PUR	Quartz sand	3.78	18.4	1.46	0.45	10	102.3
3	GW	LOS	Quartz sand	3.78	18.0	1.49	0.44	10	104.0
3#	GW	LOS	Quartz sand	3.78	18.0	1.49	0.44	10	104.3
4	GW	BLK	HA-coated sand	3.78	17.8	1.51	0.43	10	106.1
4#	GW	BLK	HA-coated sand	3.78	17.8	1.51	0.43	10	102.7
5	GW	PUR	HA-coated sand	3.78	18.3	1.47	0.45	10	103.9
5#	GW	PUR	HA-coated sand	3.78	18.3	1.47	0.45	10	104.5
6	GW	LOS	HA-coated sand	3.78	18.0	1.49	0.44	10	104.9
6#	GW	LOS	HA-coated sand	3.78	18.0	1.49	0.44	10	104.3
7	GW	BLK	Goethite-coated sand	3.78	18.4	1.46	0.45	10	100.7
7#	GW	BLK	Goethite-coated sand	3.78	18.1	1.48	0.44	10	100.4
8	GW	PUR	Goethite-coated sand	3.78	18.3	1.47	0.45	10	101.3
8#	GW	PUR	Goethite-coated sand	3.78	18.5	1.45	0.45	10	100.6
9	GW	LOS	Goethite-coated sand	3.78	18.9	1.42	0.46	10	101.8
9#	GW	LOS	Goethite-coated sand	3.78	19.1	1.40	0.47	10	102.0

[a] The symbol # indicates that the column is the respective replicate of the column indicated by the numerical number.

[b] Porosity = (column volume - mass of sand/density of sand)/column volume.

Table S2. Recipe of artificial groundwater (GW).

Ions	Concentration (mM)
HCO_3^-	0.43
SO_4^{2-}	0.39
NO_3^-	0.62
Cl^-	0.64
Ca^{2+}	0.68
Mg^{2+}	0.24
Na^+	0.43
K^+	0.20

Table S3. Selected properties of soil nanoparticles in the influents.

Column no. [a]	Type of soil nanoparticles	Type of porous media	Background solution	pH	D_h [b] (nm)	PDI [b]	ζ potential (mV)	d_p/d_c [c]
1	BLK	Quartz sand	GW	7.5	426.2 ± 14.2	0.18	-13.4 ± 0.4	0.0016
1#	BLK	Quartz sand	GW	7.6	434.3 ± 18.8	0.15	-13.3 ± 0.3	0.0017
2	PUR	Quartz sand	GW	7.6	566.4 ± 10.2	0.15	-13.8 ± 0.4	0.0022
2#	PUR	Quartz sand	GW	7.6	578.1 ± 20.3	0.19	-13.7 ±0.4	0.0022
3	LOS	Quartz sand	GW	7.5	381.0 ± 15.6	0.17	-13.7 ± 0.3	0.0015
3#	LOS	Quartz sand	GW	7.6	364.0 ± 12.8	0.20	-13.8 ± 0.3	0.0014
4	BLK	HA-coated sand	GW	7.3	425.6 ± 19.8	0.18	-12.7 ± 0.4	0.0016
4#	BLK	HA-coated sand	GW	7.6	420.1 ± 19.7	0.22	-12.8 ± 0.4	0.0016
5	PUR	HA-coated sand	GW	7.5	608.8 ± 18.4	0.17	-13.5 ± 0.5	0.0023
5#	PUR	HA-coated sand	GW	7.6	595.4 ± 10.2	0.15	-13.5 ± 0.4	0.0023
6	LOS	HA-coated sand	GW	7.4	408.1 ± 12.8	0.15	-13.8 ± 0.3	0.0016
6#	LOS	HA-coated sand	GW	7.6	404.1 ± 18.1	0.16	-13.6 ± 0.3	0.0016
7	BLK	Goethite-coated sand	GW	7.6	519.8 ± 14.1	0.20	-12.8 ± 0.4	0.0020
7#	BLK	Goethite-coated sand	GW	7.6	544.1 ± 8.2	0.22	-12.7 ± 0.3	0.0021
8	PUR	Goethite-coated sand	GW	7.3	659.9 ± 13.3	0.18	-13.9 ± 0.3	0.0025
8#	PUR	Goethite-coated sand	GW	7.6	628.3 ± 25.3	0.18	-13.4 ± 0.4	0.0024
9	LOS	Goethite-coated sand	GW	7.5	412.2 ± 18.0	0.22	-14.1 ± 0.4	0.0016
9#	LOS	Goethite-coated sand	GW	7.6	418.2 ± 6.5	0.16	-13.8 ± 0.3	0.0016

[a] The symbol # indicates that the column is the replicate of the column indicated by the numerical number.

[b] Hydrodynamic diameter (D_h) was determined based on dynamic light scattering analysis and PDI is the polydispersity index of hydrodynamic diameter.

[c] d_p/d_c represents the ratio of D_h to average diameter of sand grains.

Table S4. Mass balance data of soil nanoparticles in column experiments.

Column no. [a]	Type of soil nanoparticles	Type of porous media	Effluent Mass [b] (%)	Recovered mass [c] (%)	Mass balance [d] (%)
1	BLK	Quartz sand	67.6	25.3	92.9
1#	BLK	Quartz sand	63.6	25.6	89.2
2	PUR	Quartz sand	52.0	38.0	90.0
2#	PUR	Quartz sand	46.1	42.7	88.9
3	LOS	Quartz sand	17.5	75.0	92.5
3#	LOS	Quartz sand	14.9	68.7	83.5
4	BLK	HA-coated sand	84.9	5.4	90.3
4#	BLK	HA-coated sand	88.8	5.4	94.2
5	PUR	HA-coated sand	77.6	18.3	95.9
5#	PUR	HA-coated sand	76.6	19.2	95.8
6	LOS	HA-coated sand	54.7	37.3	92.1
6#	LOS	HA-coated sand	53.1	39.8	92.8
7	BLK	Goethite-coated sand	11.0	76.6	87.6
7#	BLK	Goethite-coated sand	13.5	70.6	84.0
8	PUR	Goethite-coated sand	10.8	99.2	110.0
8#	PUR	Goethite-coated sand	12.0	101.8	113.8
9	LOS	Goethite-coated sand	2.5	86.0	88.6
9#	LOS	Goethite-coated sand	2.2	91.4	93.5

[a] The symbol # indicates that the column is the replicate of the column indicated by the numerical number.

[b] Effluent mass was the percentage of soil nanoparticles passed through columns.

[c] Recovered mass was the percentage of soil nanoparticles recovered from columns.

[d] Mass balance was calculated as: effluent mass + recovered mass.

Table S5. Mass balance data of BDE-209 in column experiments.

Column no. [a]	Type of soil nanoparticles	Type of porous media	Effluent Mass [b] (%)	Recovered mass [c] (%)	Mass balance [d] (%)
1	BLK	Quartz sand	67.6	19.7	87.4
1#	BLK	Quartz sand	63.6	20.2	83.7
2	PUR	Quartz sand	49.0	33.8	82.8
2#	PUR	Quartz sand	43.7	38.2	81.9
3	LOS	Quartz sand	16.9	64.1	81.0
3#	LOS	Quartz sand	16.1	55.0	71.1
4	BLK	HA-coated sand	82.8	3.3	86.0
4#	BLK	HA-coated sand	85.1	3.3	88.4
5	PUR	HA-coated sand	76.0	9.8	85.7
5#	PUR	HA-coated sand	72.5	9.5	82.0
6	LOS	HA-coated sand	51.3	27.0	78.3
6#	LOS	HA-coated sand	47.6	29.9	77.6
7	BLK	Goethite-coated sand	8.8	66.5	75.3
7#	BLK	Goethite-coated sand	11.5	63.7	75.2
8	PUR	Goethite-coated sand	8.7	77.2	80.9
8#	PUR	Goethite-coated sand	6.9	86.2	93.1
9	LOS	Goethite-coated sand	0.6	75.8	76.4
9#	LOS	Goethite-coated sand	1.0	78.4	79.4

[a] The symbol # indicates that the column is the replicate of the column indicated by the numerical number.

[b] Effluent mass was the percentage of BDE-209 passed through columns.

[c] Recovered mass was the percentage of BDE-209 recovered from columns.

[d] Mass balance was calculated as: effluent mass + recovered mass.

Table S6. Mineral compositions and contents (wt%) of the soil nanoparticles based on the semi-quantitative analysis of XRD patterns.

	Illite	IS[a]	Kaolinite	Quartz	Calcite	Hematite	Clinochlore	Albite
BLK	26.5	61.4	4.7	4.4	0.0	1.1	1.9	0.0
PUR	18.9	65.2	0.9	8.9	0.0	5.3	0.9	0.0
LOS	24.5	44.2	6.3	12.2	5.5	3.2	4.0	0.1

[a] Interstratified illite-smectite.

Table S7. Selected properties of coated sand.

Type of porous media	Background solution	ζ potential (mV)	Specific surface area (m^2/g)	Amount of coating	
				mg-C/g-sand	mg-Fe/g-sand
Quartz sand	GW, pH 7.5	-19.8 ± 0.6	0.28 ± 0.02	ND	ND
HA-coated sand	GW, pH 7.5	-23.1 ± 0.4	0.36 ± 0.04	1.40±0.24	ND
Goethite-coated sand	GW, pH 7.5	-16.6 ± 0.5	0.79 ± 0.11	ND	1.58±0.12

ND represents “non-detection”.

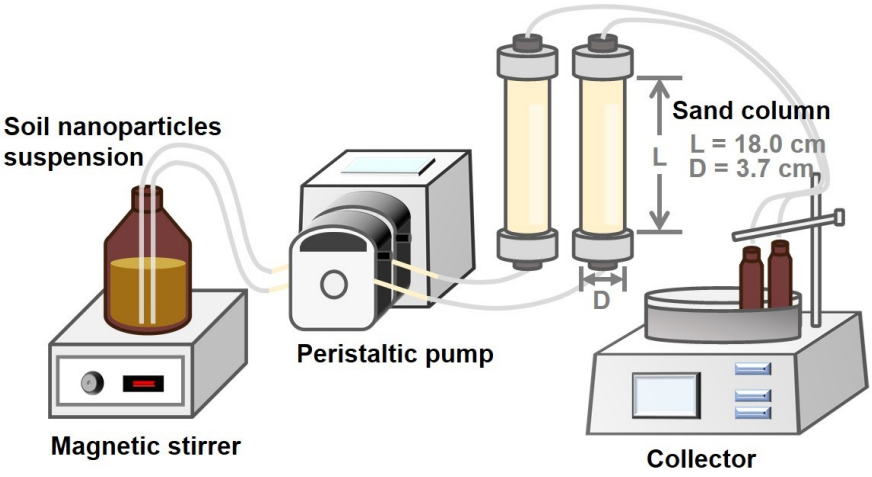

Fig. S1. Schematic diagram of column transport experiments.

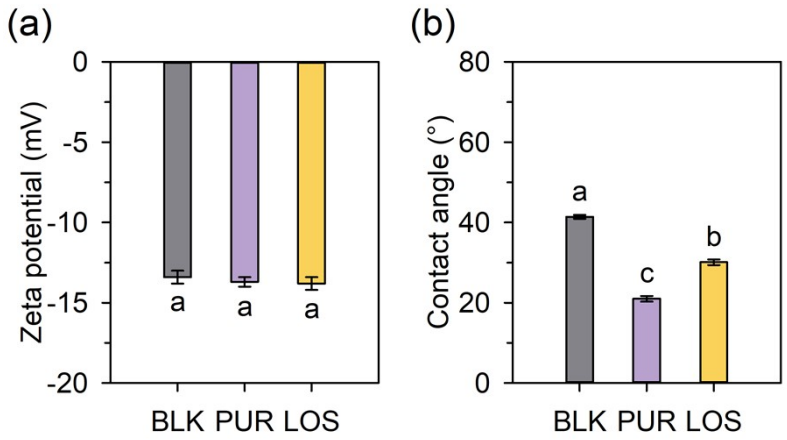

Figure S2. Selected properties of soil nanoparticles. (a) ζ (Zeta) potential in artificial groundwater. (b) Water contact angle. Error bars indicate variations of duplicates.

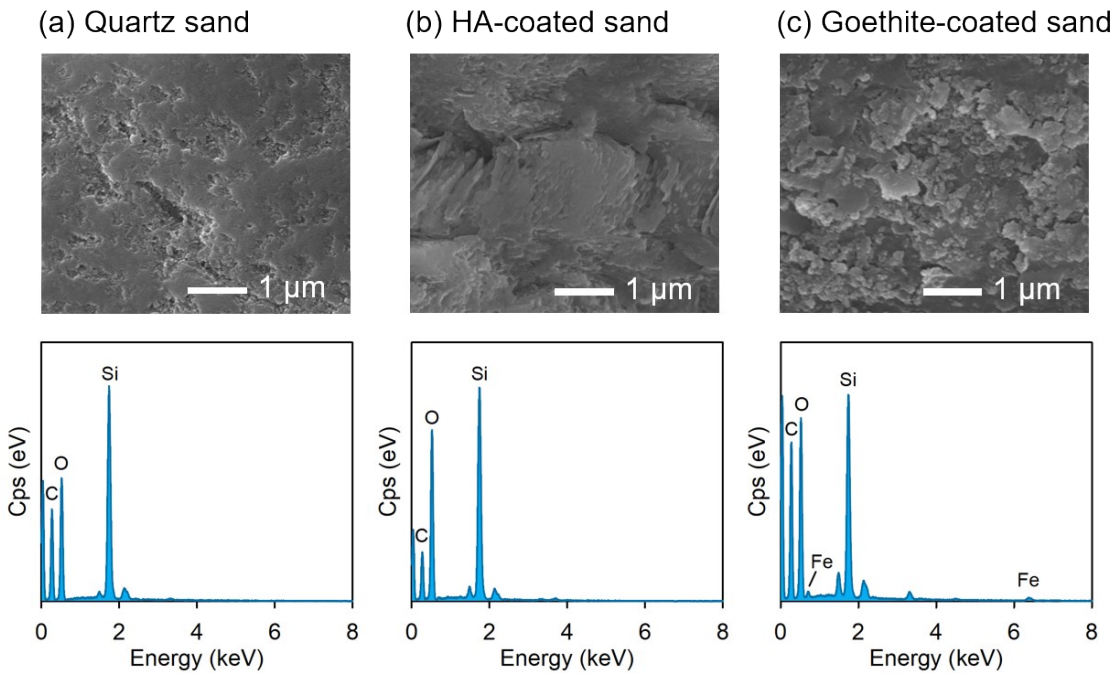

Figure S3. SEM image and energy dispersive X-ray spectrometry (EDS) spectra of different porous media. (a) Quartz sand, (b) Humic acid-coated sand, and (c) Goethite-coated sand.

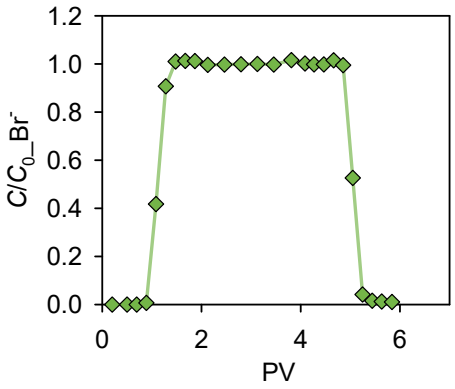

Figure S4. Representative breakthrough curve of conservative tracer (Br^-).