Supplementary Information

Transport of Graphene Oxide Nanoparticles in Saturated Sandy Soil

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Procedures used to prepare samples for AFM analysis

For each measurement, 5 μl of a GO nanoparticles suspension was deposited onto a freshly cleaved mica surface and left to adsorb for 3 min. Additionally, 30 μl of the background electrolyte solution, with which the GO nanoparticles suspension was prepared, was added to the liquid cell. The sample was scanned under tapping mode using a J scanner of a Vecco Multi-mode Nanoscope VIII AFM (Santa Barbara, CA) with a silicon nitride cantilever with sharpened pyramidal tip (OMCL-TR400PSA, Olympus).

Table S1. Fitted Parameters of Two-Site Transport Model from Breakthrough Results of Column Experiments

Column No.	Porous medium	Pore-water velocity (m/d)	Background solution	Parameters of two-site transport model			
				K_{att} (h^{-1})	S_{max} (mg/kg)	K_{str} (h^{-1})	r^2
1	Lula soil	10	DI water	3.54	0.105	0.0117	0.996
		10	10 mM NaCl	6.67	1.43	3.37	0.995
		10	25 mM NaCl	13.7	2.27	11.1	0.969
		10	35 mM NaCl	21.1	2.33	15.5	0.970
		10	50 mM NaCl	23.8	3.12	28.7	0.968
2	Sigma sand	10	DI water	3.05	0.132	1.58E-05	0.999
		10	10 mM NaCl	9.74	0.384	0.0817	0.994
		10	25 mM NaCl	11.8	0.533	3.75	0.998
		10	35 mM NaCl	16.2	0.947	5.52	0.995
		10	50 mM NaCl	22.6	1.11	15.8	0.985
3	Lula soil	10	1.5 mM NaCl	8.97	0.122	0.547	0.997
		10	0.5 mM $CaCl_2$ [a]	14.3	0.338	5.49	0.972
4	Lula soil	10	0.1 mM $CaCl_2$	9.44	0.225	11.3	0.999
		10	0.3 mM $CaCl_2$	11.8	0.366	18.6	0.993
		10	0.5 mM $CaCl_2$ [b]	12.5	0.279	11.3	0.991
5	Sigma sand	10	1.5 mM NaCl	9.91	0.242	1.25E-04	0.994
		10	0.5 mM $CaCl_2$	20.4	0.664	3.39	0.99
6	Lula soil	10	10 mM NaCl (pH 4.0)	9.90	0.843	5.15	0.965
7	Lula soil	10	10 mM NaCl (pH 7.0)	4.10	0.209	0.148	0.995
8	Lula soil	10	10 mM NaCl (pH 9.0)	7.06	0.886	0.964	0.994
9	Sigma sand	10	10 mM NaCl (pH 4.0)	24.8	1.17	0.892	0.955
10	Sigma sand	10	10 mM NaCl (pH 7.0)	15.2	0.854	0.687	0.996
11	Sigma sand	10	10 mM NaCl (pH 9.0)	20.3	0.415	1.35	0.990
12	Lula soil	10	10 mM NaCl	13.8	0.37	0.755	0.994
		10	10 mM NaCl + 10 mg/L SRHA	3.57	0.759	6.63	0.975
13	Lula soil	10	35 mM NaCl	9.09	1.61	2.16	0.968
		10	35 mM NaCl + 10 mg/L SRHA	10.5	0.376	0.821	0.991
14	Sigma sand	10	10 mM NaCl	11.8	0.363	0.342	0.953
		10	10 mM NaCl + 10 mg/L SRHA	2.97	0.109	0.179	0.997
15	Sigma sand	10	35 mM NaCl	17.3	0.853	11.0	0.925
		10	35 mM NaCl + 10 mg/L SRHA	11.0	0.411	0.914	0.929
16	Lula soil	10	10 mM NaCl	14.5	0.585	2.24	0.994
		5	10 mM NaCl	4.99	0.567	1.94	0.974
		1	10 mM NaCl	0.402	0.501	1.18	0.992
17	Sigma	10	10 mM NaCl	9.66	0.267	0.205	0.947

sand	5	10 mM NaCl	4.10	0.209	0.148	0.995
	1	10 mM NaCl	1.03	0.251	0.0629	0.996

[a] Fitted s'_{max} value is 1.05. [b] Fitted s'_{max} value is 0.794.

Table S2. ζ Potential Values of Working Suspensions of GONPs

Solution chemistry	ζ potential [a] (mV)
DI water	-49.0 ± 2.1
10 mM NaCl	-23.8 ± 0.4
25 mM NaCl	-23.2 ± 0.7
35 mM NaCl	-13.9 ± 0.8
50 mM NaCl	-14.6 ± 0.4
1.5 mM NaCl	-23.8 ± 1.2
0.1 mM $CaCl_2$	-23.2 ± 1.2
0.3 mM $CaCl_2$	-19.6 ± 0.4
0.5 mM $CaCl_2$	-14.0 ± 0.9
10 mM NaCl (pH 4.0)	-21.9 ± 1.8
10 mM NaCl (pH 7.0)	-27.4 ± 0.9
10 mM NaCl (pH 9.0)	-27.4 ± 0.5
10 mM NaCl + 10 mg/L SRHA	-21.4 ± 1.3
35 mM NaCl + 10 mg/L SRHA	-17.8 ± 0.8

[a] Values after ± sign represent standard deviation of five replicates.

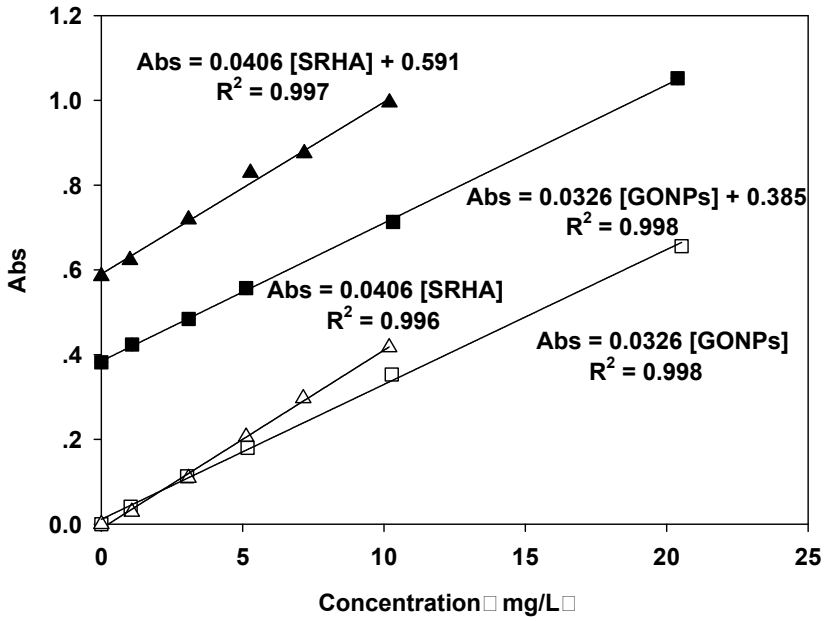

Fig. S1 Calibration curves of GONPs and SRHA used to calculate the concentrations of GONPs in the presence of SRHA and vice versa.[S1] Hollow square (□): UV absorbance of GONPs (at 230 nm) as a function of GONPs concentration in the absence of SRHA. Filled square (■), UV absorbance of GONPs (at 230 nm) as a function of GONPs concentration in the presence of 10 mg/L SRHA. Hollow triangle (Δ): UV absorbance of SRHA (at 208 nm) as a function of SRHA concentration in the absence of GONPs. Filled triangle (▲): UV absorbance of SRHA (at 208 nm) as a function of SRHA concentration in the presence of 20 mg/L GONPs. According to Chen et al.,[S1] because the standard curves of GONPs in the absence and presence of SRHA have nearly identical slopes, the presence of SRHA does not interfere the determination of GONPs concentration; likewise, the identical slopes of SRHA in the absence and presence of GONPs indicate that the presence of GONPs does not affect the determination of SRHA concentration. The SRHA stock solution was prepared by dissolving 50 mg SRHA powder in 500 mL DI water and stirring overnight. The solution was filtered through 0.45-μm filters and then kept in the dark at 4 °C.

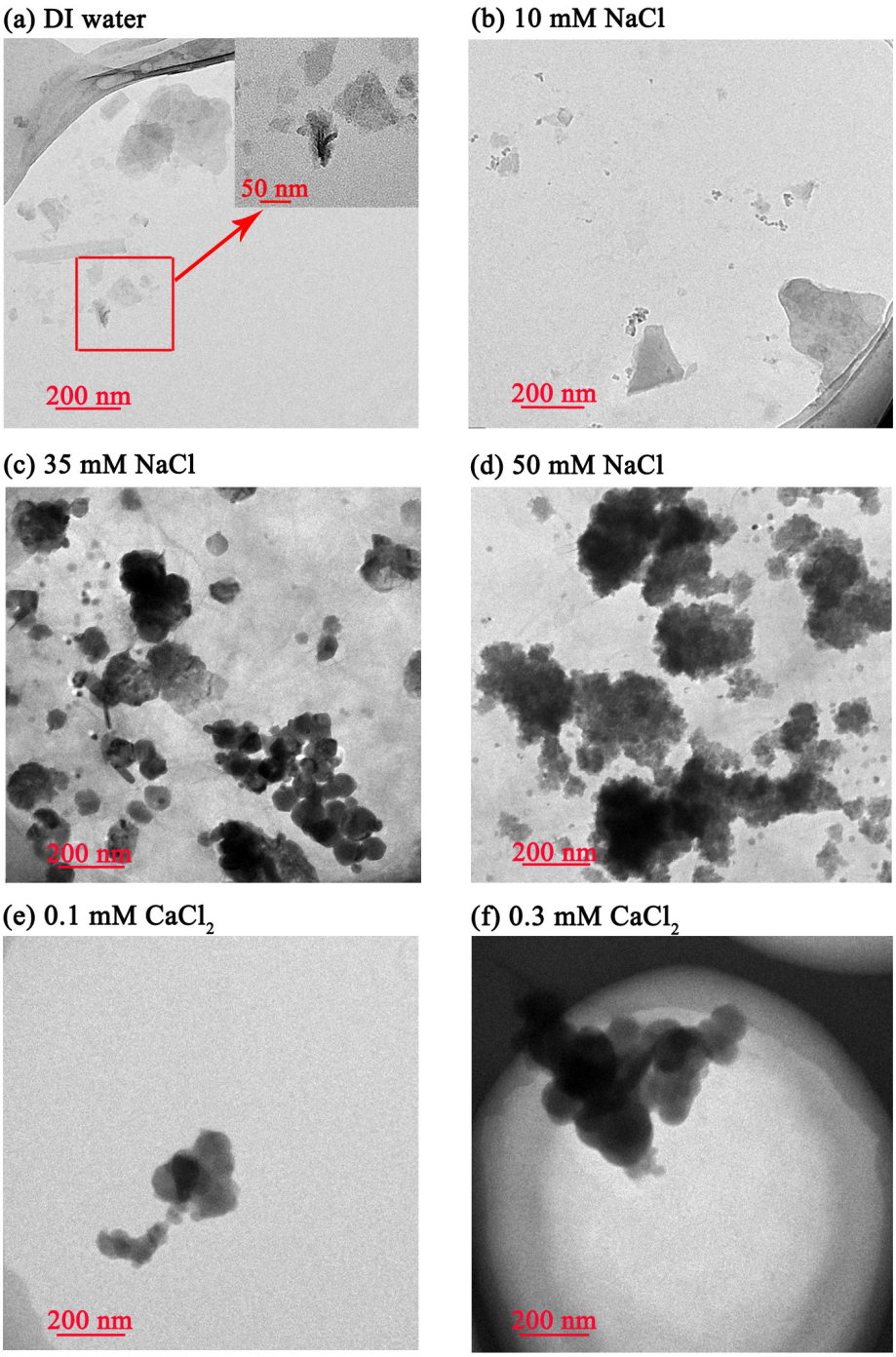

Fig. S2 (part 1) Representative TEM images of GONPs (~20 mg/L) under different solution chemistry conditions.

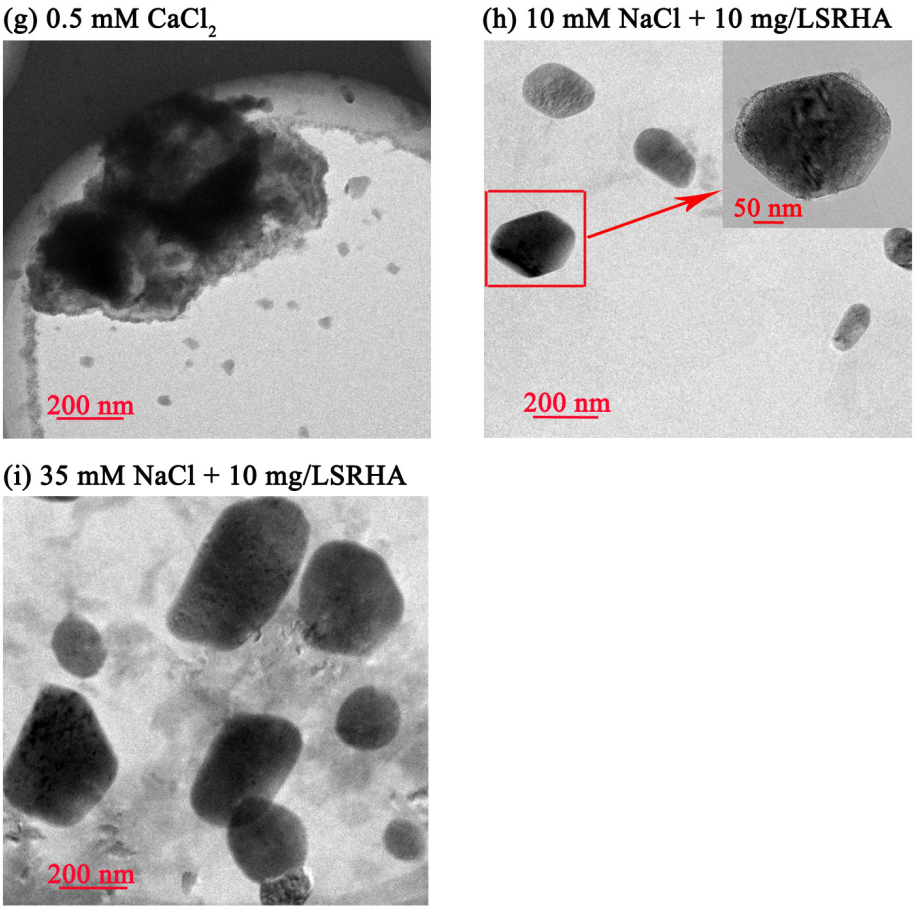

Fig. S2 (part 2) Representative TEM images of GONPs (~20 mg/L) under different solution chemistry conditions.

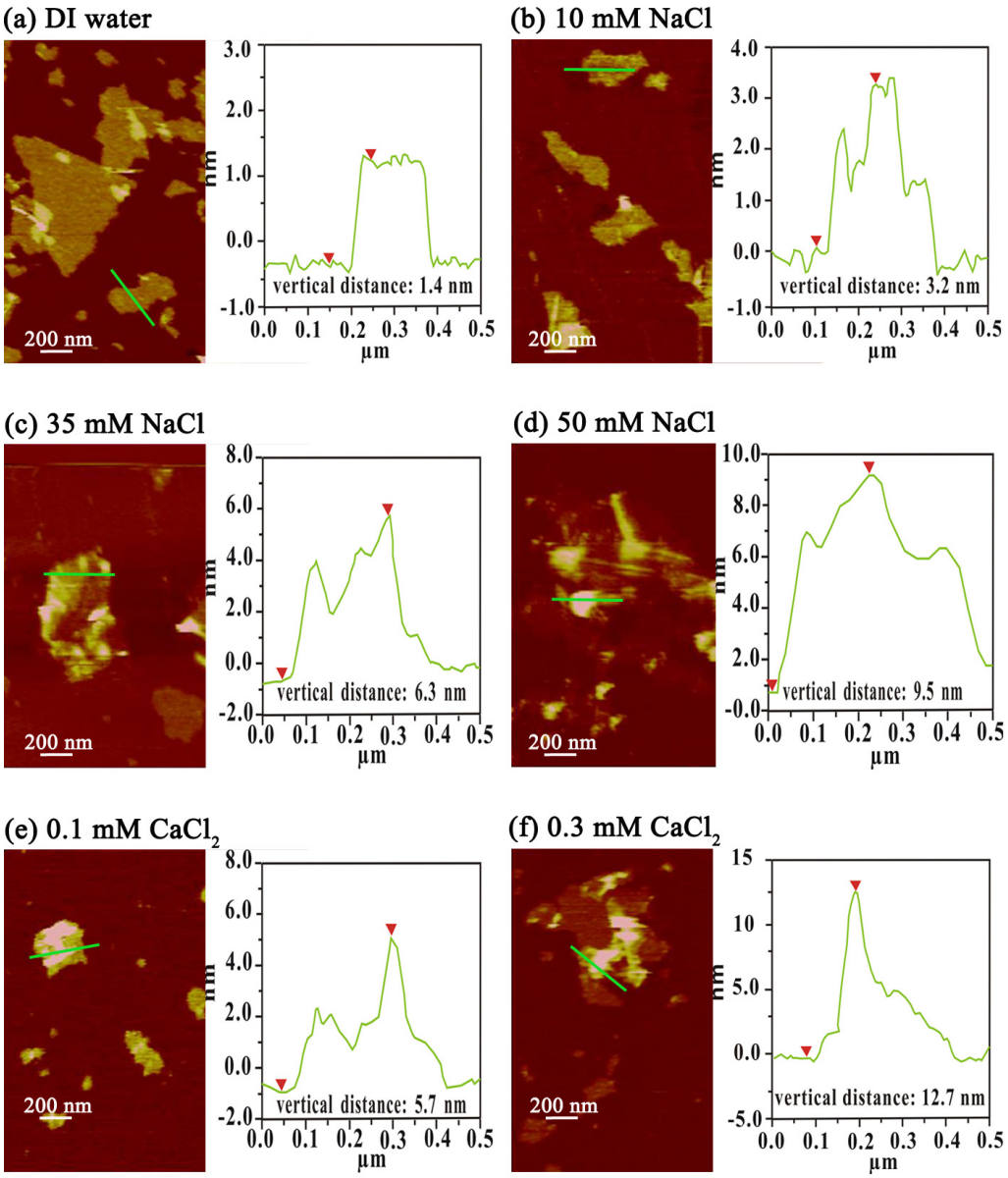

Fig. S3 (part 1) Representative AFM images of GONPs (~20 mg/L) under different solution chemistry conditions and associated cross-section analyses showing the thickness.

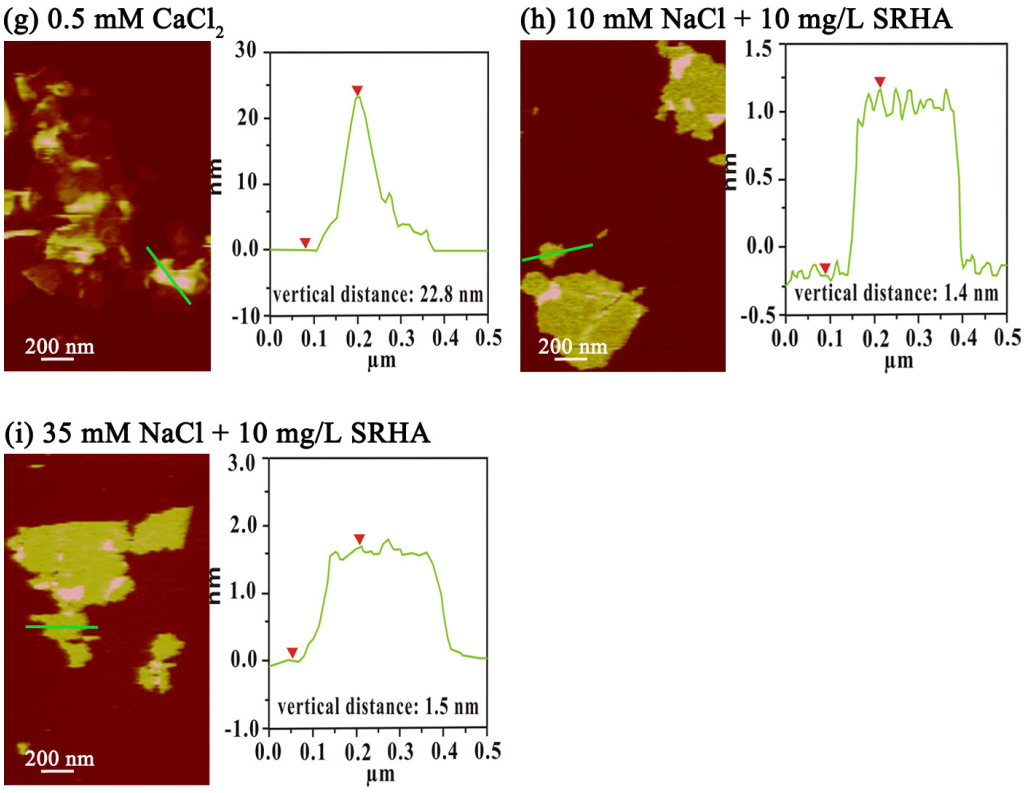

Fig. S3 (part 2) Representative AFM images of GONPs (~20 mg/L) under different solution chemistry conditions and associated cross-section analyses showing the thickness.

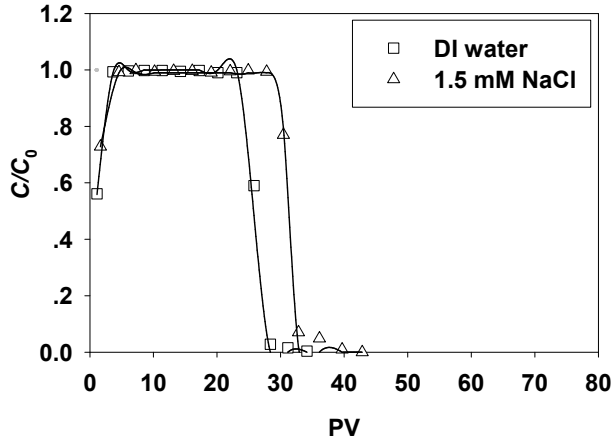

Fig. S4 Comparison between BTCs of GONPs in Lula soil: the influent of one experiment was GONPs in DI water (Column 1); the influent of the other was GONPs in 1.5 mM NaCl (Column 3). Solid lines (—) was plotted by fitting the BTCs with the two-site transport model (Equations 1–5).

References

S1 G. Chen, X. Liu and C. Su, Distinct effects of humic acid on transport and retention of TiO_2 rutile nanoparticles in saturated sand columns, *Environ. Sci. Technol.*, 2012, **46,** 7142–7150.