

## **Supplementary Material**

# **Sedimentation and Transport of Different Soil Colloids: Effects of Goethite and Humic Acid**

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## S1. Colloid transport models

The convection diffusion equation (CDE) with two kinetic retention sites was employed to describe the colloid transport and retention in the column experiments as Equation (1) [1, 2].

$$\frac{\partial C}{\partial t} = -\frac{v}{\theta} \frac{\partial C}{\partial z} + D \frac{\partial^2 C}{\partial z^2} - \frac{\rho}{\theta} \frac{\partial s_1}{\partial t} - \frac{\rho}{\theta} \frac{\partial s_2}{\partial t} \quad (1)$$

$\theta$  ( $\text{cm}^3 \cdot \text{cm}^{-3}$ ) is the volumetric water content;  $D$  is the dispersion coefficient ( $\text{m}^2 \cdot \text{s}^{-1}$ );  $\rho$  ( $\text{g} \cdot \text{m}^{-3}$ ) is the column dry bulk density;  $z$  (cm) is the spatial coordinate;  $v$  ( $\text{cm} \cdot \text{min}^{-1}$ ) is the Darcy's velocity; and  $s_1$  ( $\text{g} \cdot \text{g}^{-1}$ ) and  $s_2$  ( $\text{g} \cdot \text{g}^{-1}$ ) are colloid concentrations deposited in Site-1 and Site-2, respectively.

The Site-1, first kinetic site, on which the retention of colloid is assumed to be reversible, whereas Site-2, the second kinetic site, on which the retention is assumed to be irreversible, time-dependent retention, as described by the Langmuirian blocking or depth-dependent retention.  $s_1$  on Site-1 and  $s_2$  on Site-2 are given in Equations (2) and (3), respectively.

$$\frac{\rho}{\theta} \frac{\partial s_1}{\partial t} = k_{1a} C - \frac{\rho}{\theta} k_{1d} s_1 \quad (2)$$

$$\frac{\rho}{\theta} \frac{\partial s_2}{\partial t} = \psi_t k_{2a} C \quad (3)$$

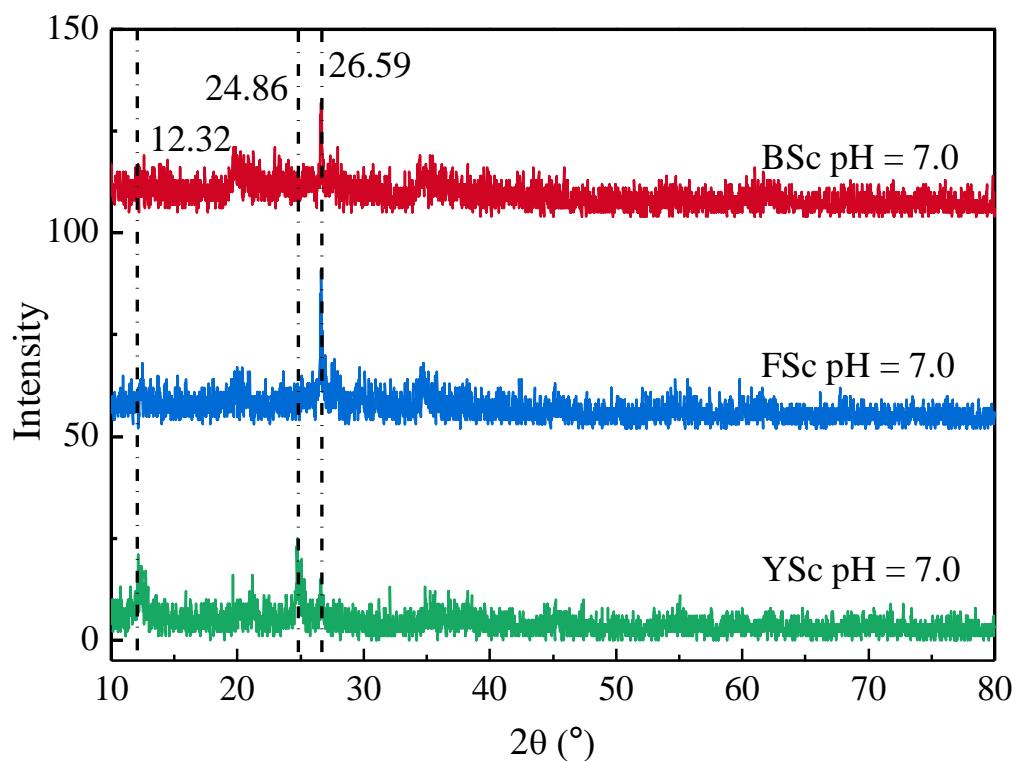
$k_{1a}$  ( $\text{min}^{-1}$ ) and  $k_{2a}$  ( $\text{min}^{-1}$ ) are first-order retention coefficients on Site-1 and Site-2, respectively;  $k_{1d}$  ( $\text{min}^{-1}$ ) is the first-order detachment coefficient;  $\psi_t$  (dimensionless) is parameters to account for time-dependent retention. The Langmuirian blocking function and the depth-dependent retention function define  $\psi_t$  are expressed by Equations (4) and (5), respectively.

$$\psi_t = 1 - \frac{s_2}{s_{\max 2}} \quad (4)$$

$$\psi_t = \left( \frac{d_c - x}{d_c} \right)^{-\beta} \quad (5)$$

$s_{\max 2}$  ( $\text{g} \cdot \text{g}^{-1}$ ) is the maximum colloid concentration on Site-2;  $d_c$  is the median diameter of the sand grains (cm); and  $\beta$  (dimensionless) is an empirical variable that controls the shape of the retention profile, using an optimal value of 0.432 for different sized spherical colloid and sand grains in which significant depth-dependency (hyperexponential retention profiles) occurred [1].

**S2. XRD analysis of soil colloids**



**Figure S1.** XRD analysis of soil colloids at pH 7.0.

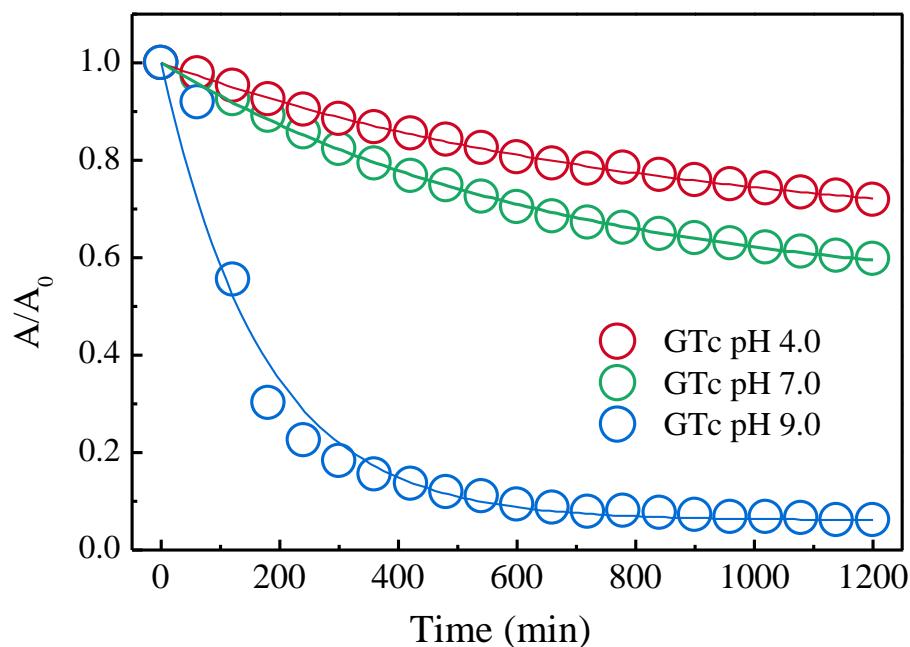
### S3. Exponential-model-fitted parameters of settling experiments

**Table S1.** Exponential-model-fitted parameters of soil colloids and mixed soil colloid-HA/GT at pH 4.0, 7.0, and 9.0.

pH	Colloid	$V_s \times 10^3$ <sup>a</sup> (m h <sup>-1</sup> )	$C'_{res}$ <sup>b</sup> (C <sub>res</sub> /C <sub>0</sub> )	R <sup>2c</sup>	Colloid	$V_s \times 10^3$ <sup>a</sup> (m h <sup>-1</sup> )	$C'_{res}$ <sup>b</sup> (C <sub>res</sub> /C <sub>0</sub> )	R <sup>2c</sup>	Colloid	$V_s \times 10^3$ <sup>a</sup> (m h <sup>-1</sup> )	$C'_{res}$ <sup>b</sup> (C <sub>res</sub> /C <sub>0</sub> )	R <sup>2c</sup>
4	BSc	1.20	0.59	0.999	FSc	4.02	0.059	0.954	YSc	1.01	0.517	0.999
7		1.05	0.375	0.999		1.61	0.384	0.997		1.24	0.432	0.999
9		0.23	-	0.930		0.772	-	0.949		0.364	-	0.995
4	BSc-GTc	1.79	0.485	0.999	FSc-GTc	4.0	0.087	0.989	YSc-GT	1.75	0.074	0.997
7		1.22	0.499	0.999		1.29	0.240	0.998		1.02	0.467	0.998
9		1.17	0.458	0.999		0.762	0.281	0.999		0.788	0.442	0.998
4	BSc-HA	0.854	0.471	0.999	FSc-HA	1.75	0.532	0.998	YSc-HA	0.923	0.453	0.999
7		0.895	0.491	0.999		2.22	0.569	0.996		1.07	0.532	0.999
9		1.11	0.559	0.999		2.08	0.578	0.994		1.12	0.561	0.999

<sup>a</sup> The sedimentation rates; <sup>b</sup> The relative nonsettling concentration; <sup>c</sup> The correlation coefficient for sedimentation model.

**S4. Sedimentation kinetics of goethite colloid at different pH**



**Figure S2.** Sedimentation kinetics of goethite (GT) colloid at pH 4.0, 7.0, and 9.0. Symbols show observed data and lines show simulation fitting.

**Table S2.** Particle size and zeta potential of the goethite (GT) colloid and exponential-model-fitted parameters of goethite (GT) colloid at pH 4.0, 7.0, and 9.0.

pH	Colloid	Particle Size (nm)	Zeta Potential (mV)	$V_s \times 10^3$ <sup>a</sup> (m h <sup>-1</sup> )	$C'_{res}$ <sup>b</sup> ( $C_{res}/C_0$ )	$R^2$ <sup>c</sup>
4		199.5	32.0	1.54	0.519	0.998
7	GTc	382.8	22.1	1.26	0.643	0.998
9		4290.0	7.0	5.94	0.061	0.962

<sup>a</sup> The sedimentation rates; <sup>b</sup> The relative nonsettling concentration; <sup>c</sup> The correlation coefficient for sedimentation model.

## S5. Fitted and experimental parameters of transport experiments

**Table S3.** Fitted and experimental parameters of transport of soil colloids in the sand, 0.5% GT mixed sand, and 0.5% GT–0.2% HA sand columns at pH 7.0.

Column	$k_{1a}^a$ (min <sup>-1</sup> )	$k_{1d}^b$ (min <sup>-1</sup> )	$k_{2a}^c$ (min <sup>-1</sup> )	$S_{max}^d$	$k_{1d}/k_{1a}$	$R^2 e$	recovery <sup>h</sup> (%)
Q-BS	0.19	0.12	0.02	0.07	0.64	0.99	100.1
Q-GT-BS	0.15	0.09	0.13	0.16	0.60	0.983	93.0
Q-GT-HA-BS	0.18	0.14	0.02	0.12	0.82	0.999	100.5
Q-FS	0.23	0.21	0.00	0.09	0.92	0.999	102.0
Q-GT-FS	0.20	0.17	0.55	1.40	0.83	0.967	68.8
Q-GT-HA-FS	0.17	0.19	0.41	35.57	1.14	0.979	75.8
Q-YS	0.09	0.16	0.00	0.00	1.72	0.990	105.4
Q-GT-YS	0.20	0.08	0.15	0.66	0.41	0.991	87.8
Q-GT-HA-YS	0.19	0.18	0.07	0.55	0.94	0.996	96.6

<sup>a</sup> The first-order retention coefficient on Site-1; <sup>b</sup> The first-order detachment coefficient on Site-1; <sup>c</sup> The first-order retention coefficient on Site-2; <sup>d</sup> Maximum solid phase concentration of nanoparticle on Site-1; <sup>e</sup> Squared Pearson's correlation coefficient; <sup>h</sup> Recovery of soil colloids in the effluent.

## 66. The DLVO energy barriers for settling and transport systems

**Table S4.** The DLVO energy barriers for settling systems.

pH	Colloid	$\Phi_{MAX}^a$ (kt)	Colloid	$\Phi_{MAX}$ (kt)	Colloid	$\Phi_{MAX}$ (kt)
4		-		-		38.2
7	BS	107.7	FS	-	YS	215.6
9		544.2		-		396.4
4		-		-		-
7	BS-GT	202.4	FS-GT	-	YS-GT	182.3
9		279.2		-		368.6
4		11.8		-		50.2
7	BS-HA	307.6	FS-HA	3.8	YS-HA	388.9
9		366.0		1.8		487.0

<sup>a</sup> The primary energy barriers, its value less than 0 kt express as “-”.

**Table S5.** The primary energy barriers for transport systems.

Column	$\Phi_{MAX}$ (kt)
Q-BS	385.7
Q-GT-BS	-
Q-GT-HA-BS	407.2
Q-FS	276.8
Q-GT-FS	-
Q-GT-HA-FS	286.4
Q-YS	753.5
Q-GT-YS	-
Q-GT-HA-YS	829.7

## References

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