Electronic Supplementary Material (ESI) for Environmental Science: Processes & Impacts.

Supporting Information

Effects of Aluminum Incorporation on Schwertmannite Structure and Surface Properties

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Table S1. Weight percent of stoichiometric water and weight loss (%) from TGA data.

Sample	% w w (OH^{-} + H_2O)	% w lost TGA
Al-Sch$_{0.00}$	24.41	25.02
Al-Sch$_{0.03}$	21.94	24.51
Al-Sch$_{0.08}$	20.80	24.67
Al-Sch$_{0.32}$	27.22	24.79

Table S2. Calculated As(V) species in the different arsenic solutions used in adsorption experiment using the PHREEQC code (Mintq.v4 database). The model was run at 25 °C, pH 3.5 and an ion strength of 100 mmol L^{-1}

	As 1	As 2	As 3	As 4	As 5	As 6	As 7	As 8	As 9	As 10
Total As(V) (mmol L^{-1})	0.06	0.1	0.5	0.8	1	1.5	3	5	7	10
$H_2AsO_4^-$ (%)	96.33	96.32	96.31	96.32	96.35	96.29	96.22	96.14	96.06	95.92
H_3AsO_4 (%)	3.57	3.57	3.58	3.59	3.60	3.60	3.68	3.76	3.86	4.01
$HAsO_4^{-2}$ (%)	0.10	0.10	0.10	0.10	0.10	0.10	0.09	0.09	0.08	0.07
AsO_4^{-3} (%)	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Table S3. Calculated saturation index (SI) for supersaturated Fe(III)-As(V)-Al- mineral phases of reacted solution using the PHREEQC code (Mintq.v4 database). The model was run at 25 °C, pH 3.5 and with ion strength of 100 mmol L^{-1}. Bold values represent the supersaturated phases for each adsorption experiment.

Phases	Formula	SI	Phases	Formula	SI
Al(OH)3(am)	$Al(OH)_3$(am)	-5.10	Gibbsite	$Al(OH)_3$	-2.59
Al2O3	Al_2O_3	-8.25	Goethite	FeOOH	**3.37**
Basaluminite	$Al_4(OH)_{10}SO_4$	-11.64	H-Jarosite	$(H_3O)Fe_3(SO_4)_2(OH)_6$	**2.78**
AlAsO4:2H2O	$AlAsO_4{:}2H_2O$	-2.73	Halite	NaCl	-4.28
AlOHSO4	$AlOHSO_4$	-1.52	Hematite	Fe_2O_3	**9.14**
As2O5	As_2O_5	-13.97	Lepidocrocite	FeOOH	**2.49**
Boehmite	AlOOH	-2.88	Maghemite	Fe_2O_3	**1.34**
Diaspore	AlOOH	-1.17	Mirabilite	$Na_2SO_4{:}10H_2O$	-5.03
Fe(OH)2.7Cl.3	$Fe(OH)_{2.7}Cl_{0.3}$	**5.45**	Na-Jarosite	$NaFe_3(SO_4)_2(OH)_6$	**4.04**
Fe2(SO4)3	$Fe_2(SO_4)_3$	-19.90	Schwertmannite	$Fe_8O_8(OH)_{4.32}(SO_4)_{1.84}$	**3.66**
FeAsO4:2H2O	$FeAsO_4{:}2H_2O$	-0.17	Thenardite	Na_2SO_4	-6.45
Ferrihydrite	$Fe(OH)_3$	**0.67**			

Table S4. Fitted parameters of As(V) adsorption in schwertmannite (Sch) by Langmuir models and exchange isotherm between sulfate (SO_4^{-2}) and arsenate ($H_2AsO_4^-$).

Model	**Laqngmuir Adsorption Isotherm**					**SO_4^{-2} - $H_2AsO_4^-$ Exchange Paramenter**			
	Slope	**Intercept**	**R^2**	**Γ_{max} (mol_{As} mol_{Fe}^{-2})**	**Log K_L**	**Slope**	**Intercept**	**R^2**	**% Exchange**
Al-$Sch_{0.00}$	5.1272	0.000010	0.999	0.195	5.71	0.5333	42.239	0.986	59
Al-$Sch_{0.03}$	4.2978	0.000100	0.995	0.233	4.63	0.5695	51.500	0.992	72
Al-$Sch_{0.08}$	4.5233	0.000002	0.999	0.221	6.35	0.5445	46.906	0.988	68
Al-$Sch_{0.32}$	3.8502	0.000020	0.999	0.260	5.28	0.4899	42.200	0.979	75

Figure S1. Thermogravimetric analysis results from Al-Sch samples. The weight lost during the measurement is normalized to the initial sample weight. The dashed horizontal line indicates the average weight lost for all the solid samples.

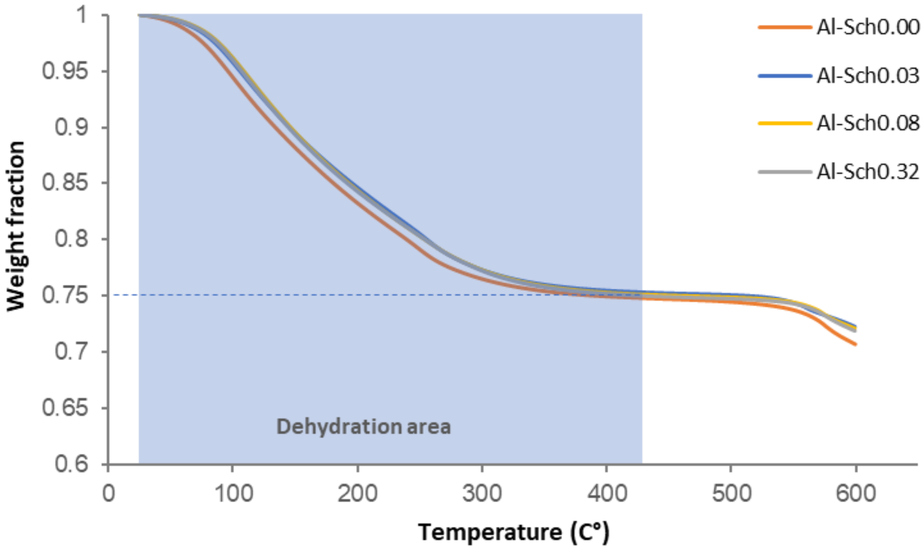

Figure S2: Correlation between Al^{+3} contained in Al-Sch (mmol g^{-1}_{sch}) with (a) sulfate and (b) OH^- (mmol g^{-1}_{sch}). The solids selected for As(V) adsorption experiments are indicated with black arrows.

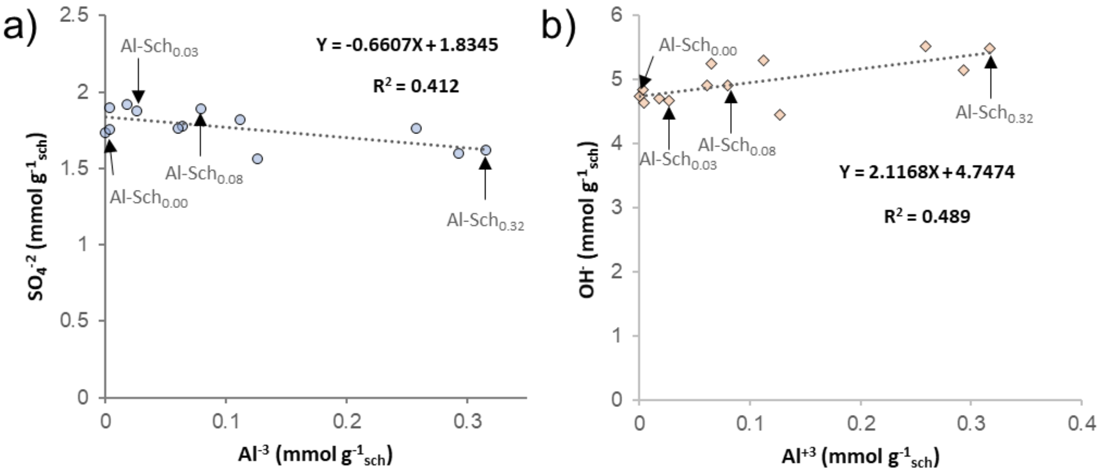

Figure S3: Ion exchange isotherm: exchange between As(V) with sulfate onto (a) Al-$Sch_{0.00}$ (b) Al-$Sch_{0.03}$ (c) Al-$Sch_{0.08}$ (d) Al-$Sch_{0.32}$. The experiment was conducted with an ionic strength of 100 mmol L^{-1}, $6{\cdot}10^{-2}$ to 10 mmol L^{-1} oxyanion concentration and 2.5 g L^{-1} of solid phase.

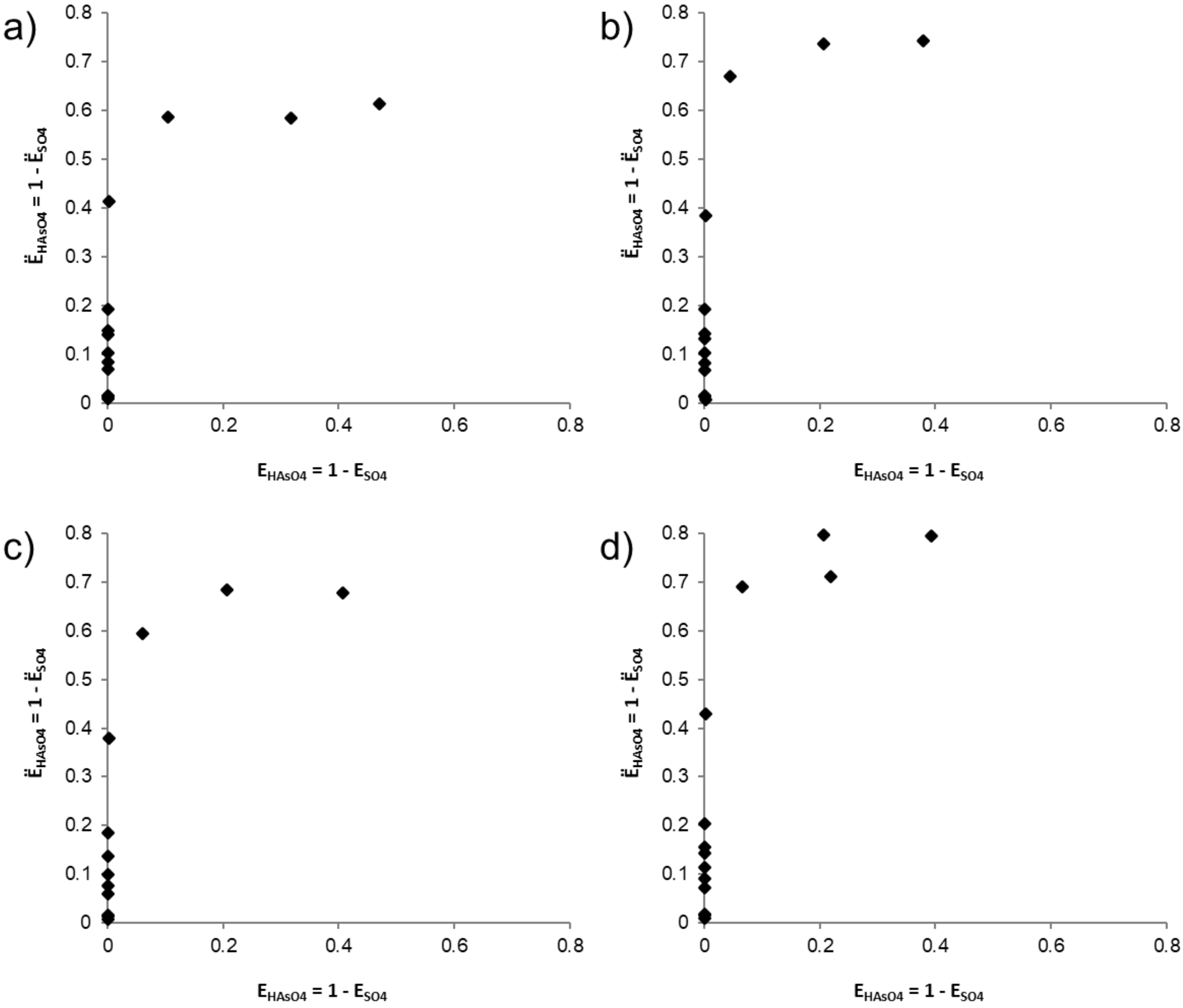

Figure S4: d-PDFs of arsenate onto (a) Al-Sch$_{0.00}$ (b) Al-Sch$_{0.03}$ (c) Al-Sch$_{0.08}$ (d) Al-Sch$_{0.32}$ with 1.5, 3.0, 5.0, 7.0 mmol L^{-1} of arsenate.

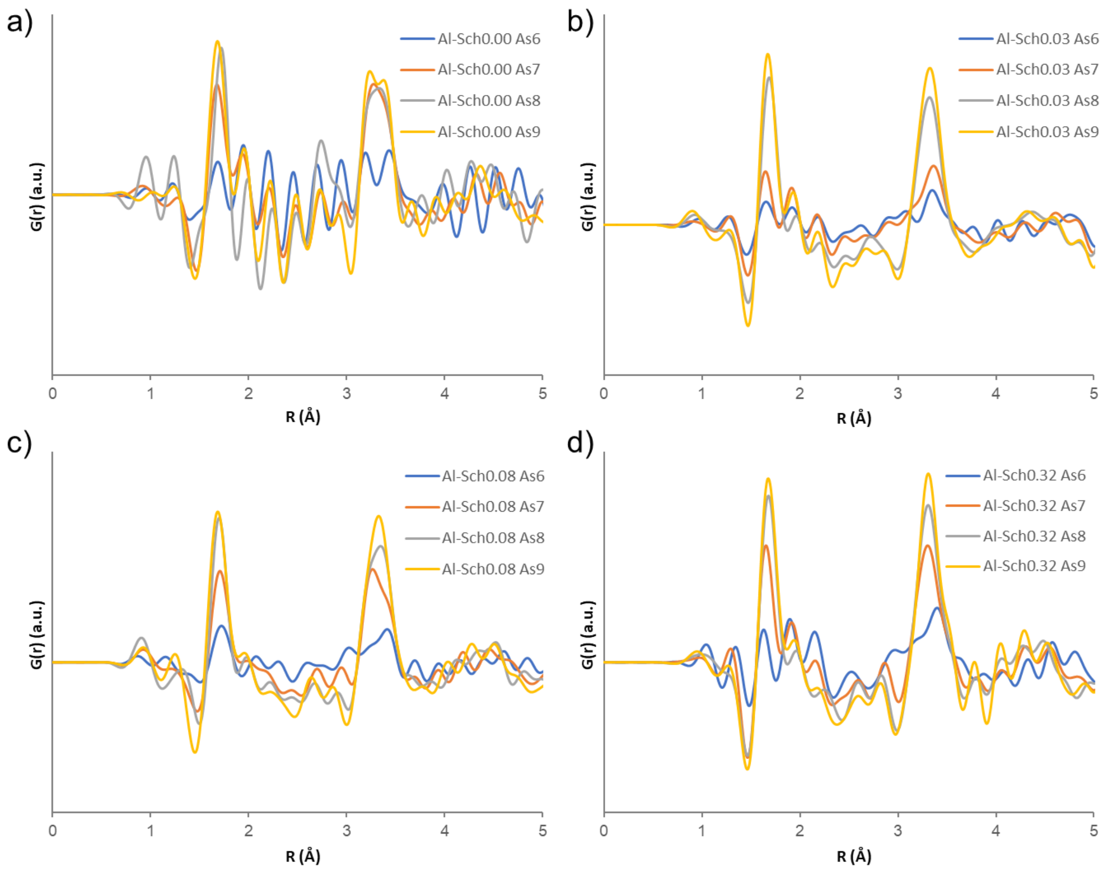

Figure S5: d-PDFs of arsenate onto Al-$Sch_{0.32}$ with 1.5, 3.0, 5.0, 7.0 mmol L^{-1} of arsenate. Zoom in the Fe-Fe and Fe-O peaks where Al incorporation affect the intensity of these atoms' pairs.