

Supplementary Material for

Goethite affects phytolith dissolution through clay particle aggregation and pH regulation

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Submitted to the journal of *Geochimica et Cosmochimica Acta*

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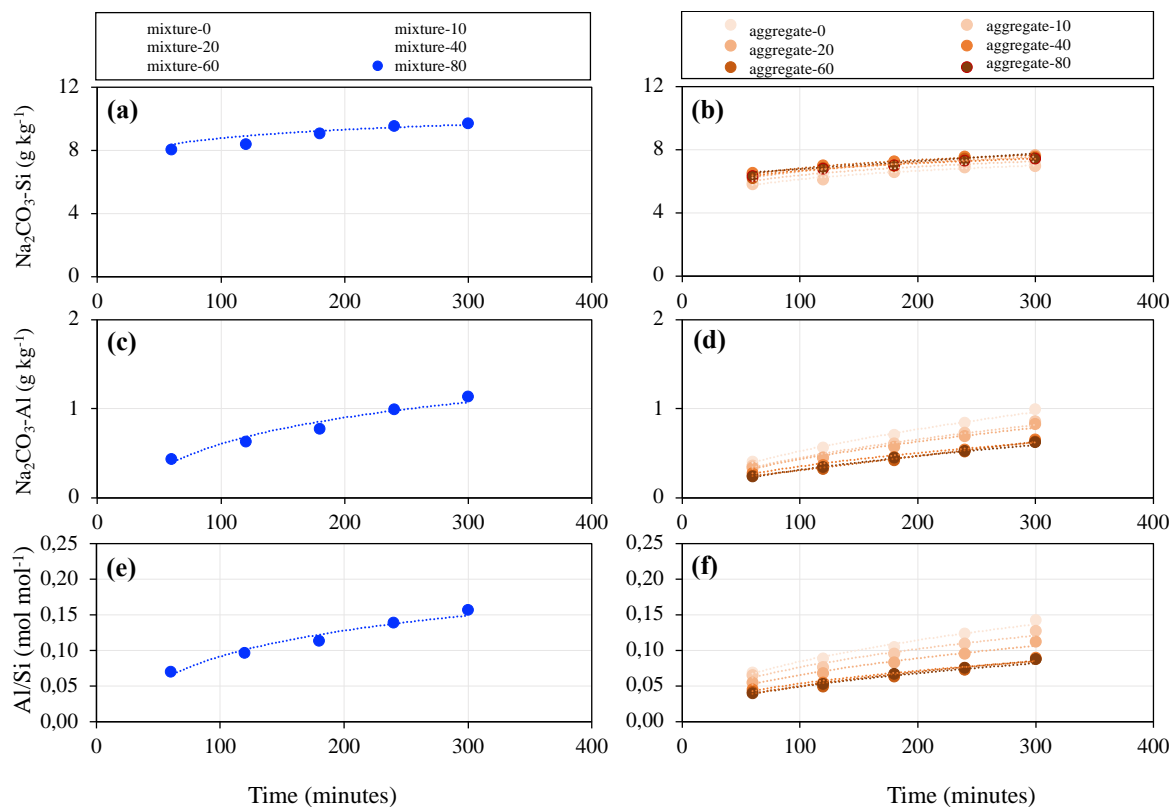


Figure S1. Time evolution of Si and Al concentrations in 0.1 M Na_2CO_3 extracts ($\text{Na}_2\text{CO}_3\text{-Si}$, $\text{Na}_2\text{CO}_3\text{-Al}$) and Al/Si molar ratio from unaggregated mixtures (left: a, c, e) and aggregates (right: b, d, f) with various goethite concentrations: $\text{Na}_2\text{CO}_3\text{-Si}$ evolution in (a) unaggregated mixtures, (b) aggregates, $\text{Na}_2\text{CO}_3\text{-Al}$ evolution in (c) unaggregated mixtures, (d) aggregates; Al/Si molar ratio in (e) unaggregated mixtures, (f) aggregates.

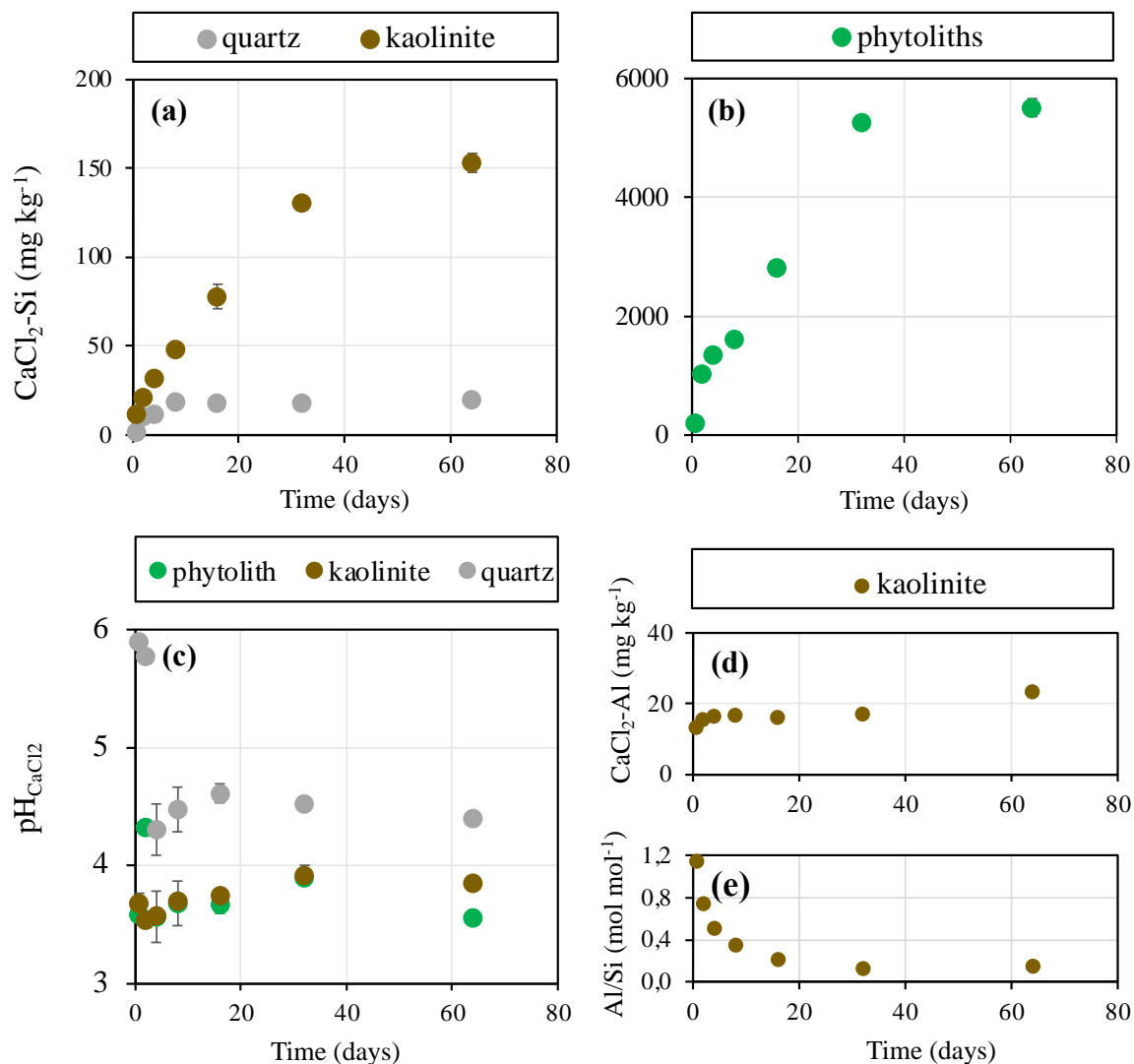


Figure S2. Time evolution, in the 0.01 M CaCl₂ extract, of (a-b) Si concentration (CaCl₂-Si), (c) pH (pH_{CaCl₂}) for individual constituents (quartz, *Charentes* kaolinite, and rice phytolith). Time evolution of (d) Al concentration (CaCl₂-Al), and (e) Al/Si ratio for *Charentes* kaolinite.

Note that the experimental data were similar to those reported by Li et al. (2022), though they have been acquired in a new experimental design, as notified in the section *Materials and methods*.

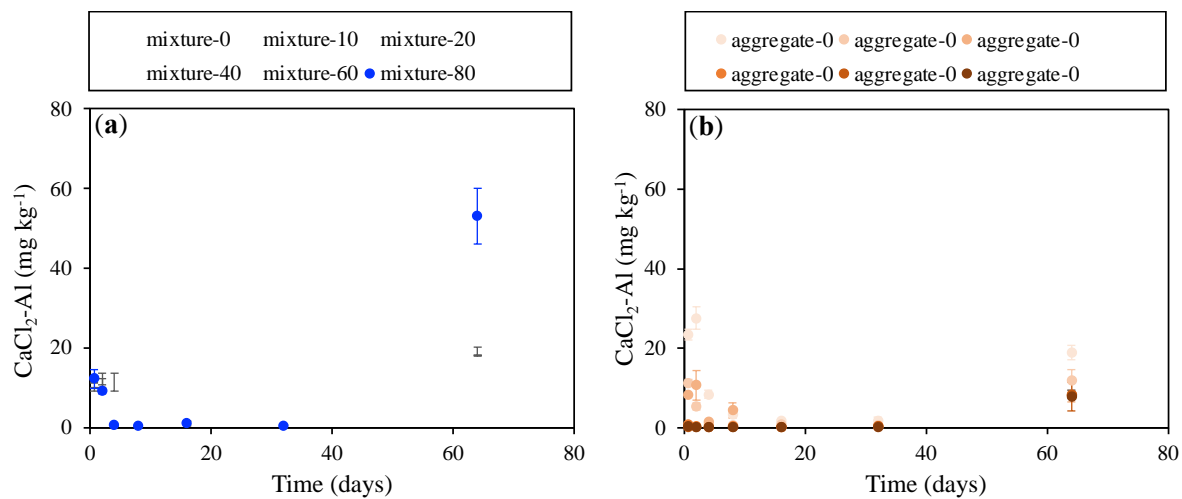


Figure S3. Time evolution of Al concentrations in 0.01 M CaCl₂ extracts (CaCl₂-Al) from unaggregated mixtures (a) and aggregates (b) with various goethite proportion.

Table S1. Soil organic C budget in unaggregated mixtures and aggregates at each goethite concentration. A different lowercase letter (a, b) resulting from the independent samples *t*-separation test, indicates a significant difference between unaggregated mixtures and aggregates at a given goethite content ($p < 0.05$); a different uppercase letter (A, B, C) resulting from *LSD*'s multiple comparison test, indicates a significant difference in either unaggregated mixtures or aggregates with various goethite contents ($p < 0.05$).

Treatment	Total initial C (TC) g C kg ⁻¹	DOC g C kg ⁻¹	DOC contribution to TC %	C release rate* mg C kg ⁻¹ TC hour ⁻¹
mixture-0	21.90	3.92 Aa	17.88 Aa	11.18Aa
mixture-10	21.90	3.74 Aa	17.07 Aa	10.67Aa
mixture-20	21.90	3.71 Aa	16.95 Aa	10.60Aa
mixture-40	21.90	3.30 Ba	15.05 Ba	9.40B7a
mixture-60	21.90	3.40 Ba	15.53 Ba	9.70Ba
mixture-80	21.90	3.44 Ba	15.70 Ba	9.81Ba
aggregate-0	21.90	2.40 Ab	10.96 Ab	6.85Ab
aggregate-10	21.90	1.58 Bb	7.23 Bb	4.52Bb
aggregate-20	21.90	1.54 Bb	7.03 Bb	4.39Bb
aggregate-40	21.90	1.32 Bb	6.04 Bb	3.78Bb
aggregate-60	21.90	1.03 Cb	4.71 Cb	2.94Cb
aggregate-80	21.90	1.04 Cb	4.77 Cb	2.98Cb

*Hourly release rate of DOC in 0.01 M CaCl₂

Table S2. pH, concentrations (mol l⁻¹) of H₄SiO₄ and Al, and Ge (μmol l⁻¹) in 0.01 M CaCl₂ extracts from 16 h to 64 d for phytolith, quartz, kaolinite, and mixtures and aggregates.

	days	16h	2d	4d	8d	16d	32d	64d
phytolith	log ₁₀ (H ₄ SiO ₄)	-3.14	-2.45	-2.33	-2.26	-2.01	-1.74	-1.72
	pH	3.6	4.3	3.6	3.7	3.7	3.9	3.6
	log ₁₀ (Ge)	-2.66	-	-2.58	-2.50	-2.51	-2.38	-2.30
quartz	log ₁₀ (H ₄ SiO ₄)	-5.20	-4.44	-4.39	-4.19	-4.20	-4.21	-4.16
	pH	5.9	5.8	4.5	4.6	4.7	4.5	4.4
	log ₁₀ (Ge)	nd	nd	nd	nd	nd	nd	nd
kaolinite	log ₁₀ (H ₄ SiO ₄)	-4.39	-4.13	-3.96	-3.78	-3.57	-3.35	-3.28
	log ₁₀ (Al ³⁺)	-6.31	-6.24	-6.22	-6.21	-6.23	-6.20	-6.06
	log ₁₀ (Ge)	-3.09	-	-3.01	-2.98	-3.04	-2.98	-3.04
	pH	3.7	3.5	3.6	3.7	3.7	3.9	3.8
mixtures-0	log ₁₀ (H ₄ SiO ₄)	-4.02	-3.63	-3.44	-3.21	-3.06	-3.05	-2.93
	log ₁₀ (Al ³⁺)	-4.25	-4.15	-4.16	-3.89	-3.99	-4.57	-4.45
	log ₁₀ (Ge)	-3.04	-	-2.96	-2.95	-3.03	-3.99	-4.09
	pH	4.9	4.8	5.0	5.0	5.3	5.2	5.5
mixtures-10	log ₁₀ (H ₄ SiO ₄)	-3.90	-3.62	-3.39	-3.20	-2.98	-2.96	-2.82
	log ₁₀ (Al ³⁺)	-4.22	-4.35	-4.30	-4.05	-3.79	-4.90	-4.03
	log ₁₀ (Ge)	-3.07	-	-2.99	-2.98	-3.06	-3.59	-3.39
	pH	4.9	5.0	5.0	5.7	5.7	5.8	6.3
mixtures-20	log ₁₀ (H ₄ SiO ₄)	-3.87	-3.39	-3.33	-3.06	-2.95	-2.86	-2.82
	log ₁₀ (Al ³⁺)	-4.27	-4.24	-5.36	-5.64	-5.29	-5.32	-4.47
	log ₁₀ (Ge)	-3.17	-	-3.09	-3.08	-3.16	-2.92	-3.27
	pH	4.9	5.4	5.7	6.6	5.8	5.9	6.6
mixtures-40	log ₁₀ (H ₄ SiO ₄)	-3.84	-3.41	-3.14	-3.05	-2.90	-2.84	-2.75
	log ₁₀ (Al ³⁺)	-4.38	-4.36	-4.37	-5.85	-5.23	-5.58	-4.15
	log ₁₀ (Ge)	-2.94	-	-2.86	-2.85	-2.93	-2.84	-2.97
	pH	5.0	5.5	5.1	6.6	5.9	5.6	6.6
mixtures-60	log ₁₀ (H ₄ SiO ₄)	-3.67	-3.26	-3.09	-2.96	-2.89	-2.80	-2.76
	log ₁₀ (Al ³⁺)	-4.45	-4.37	-5.75	-5.76	-5.69	-5.78	-4.17
	log ₁₀ (Ge)	-2.90	-	-2.82	-2.81	-2.89	-2.53	-2.69
	pH	5.1	5.5	5.7	5.7	6.0	6.8	7.3
mixtures-80	log ₁₀ (H ₄ SiO ₄)	-3.63	-3.25	-3.12	-2.95	-2.87	-2.81	-2.70
	log ₁₀ (Al ³⁺)	-4.34	-4.47	-5.68	-5.82	-5.45	-5.76	-3.71
	log ₁₀ (Ge)	-2.86	-	-2.78	-2.77	-2.85	-2.63	-2.57
	pH	5.1	5.8	5.6	6.4	6.4	6.7	7.5
aggregate-0	log ₁₀ (H ₄ SiO ₄)	-3.78	-3.40	-3.29	-3.16	-3.07	-2.88	-2.82
	log ₁₀ (Al ³⁺)	-4.06	-3.99	-4.50	-4.89	-5.16	-5.18	-4.15
	log ₁₀ (Ge)	-3.03	-	-2.96	-2.95	-3.02	-3.46	-3.66
	pH	5.2	5.3	5.9	6.6	6.2	6.5	7.0
aggregate-10	log ₁₀ (H ₄ SiO ₄)	-3.68	-3.37	-3.21	-3.12	-2.98	-2.85	-2.82
	log ₁₀ (Al ³⁺)	-4.38	-4.69	-5.43	-5.53	-5.45	-5.55	-4.36
	log ₁₀ (Ge)	-3.34	-	-3.26	-3.24	-3.31	-3.29	-3.79
	pH	5.5	6.8	6.6	6.8	7.2	7.2	7.7
aggregate-20	log ₁₀ (H ₄ SiO ₄)	-3.65	-3.28	-3.15	-3.07	-2.98	-2.84	-2.81

	$\log_{10}(\text{Al}^{3+})$	-4.51	-4.40	-5.24	-4.78	-5.92	-5.90	-4.52
	$\log_{10}(\text{Ge})$	-3.32	-	-3.24	-3.22	-3.30	-3.33	-3.61
	pH	5.6	6.3	6.6	6.2	6.9	7.5	7.9
aggregate-40	$\log_{10}(\text{H}_4\text{SiO}_4)$	-3.46	-3.05	-3.02	-2.98	-2.90	-2.82	-2.80
	$\log_{10}(\text{Al}^{3+})$	-5.50	-5.81	-6.07	-5.77	-6.01	-5.69	-5.04
	$\log_{10}(\text{Ge})$	-3.38	-	-3.30	-3.22	-3.23	-2.87	-3.25
	pH	6.3	6.9	6.9	6.7	6.1	6.5	7.9
aggregate-60	$\log_{10}(\text{H}_4\text{SiO}_4)$	-3.40	-3.15	-3.04	-3.03	-2.87	-2.82	-2.86
	$\log_{10}(\text{Al}^{3+})$	-5.98	-5.87	-5.87	-5.87	-5.87	-5.75	-4.51
	$\log_{10}(\text{Ge})$	-3.42	-	-3.34	-3.22	-3.20	-3.07	-3.22
	pH	6.8	7.1	6.9	6.9	6.4	7.2	7.8
aggregate-80	$\log_{10}(\text{H}_4\text{SiO}_4)$	-3.44	-3.11	-3.06	-3.03	-2.91	-2.83	-2.83
	$\log_{10}(\text{Al}^{3+})$	-5.68	-5.87	-5.90	-5.90	-5.90	-5.85	-4.53
	$\log_{10}(\text{Ge})$	-3.20	-	-3.12	-3.07	-3.10	-2.99	-3.11
	pH	7.2	6.7	6.8	7.2	7.0	7.1	7.2

S-References

Li, Z., Meunier, J.-D., Delvaux, B., 2022. Aggregation reduces the release of bioavailable silicon from allophane and phytolith. *Geochim. Cosmochim. Acta.* 325, 87-105.