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SUPPLEMENTARY INFORMATION

The formation of Fe colloids and layered double hydroxides as sequestration agents in the natural remediation of mine drainage

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Supplementary material, figures and tables

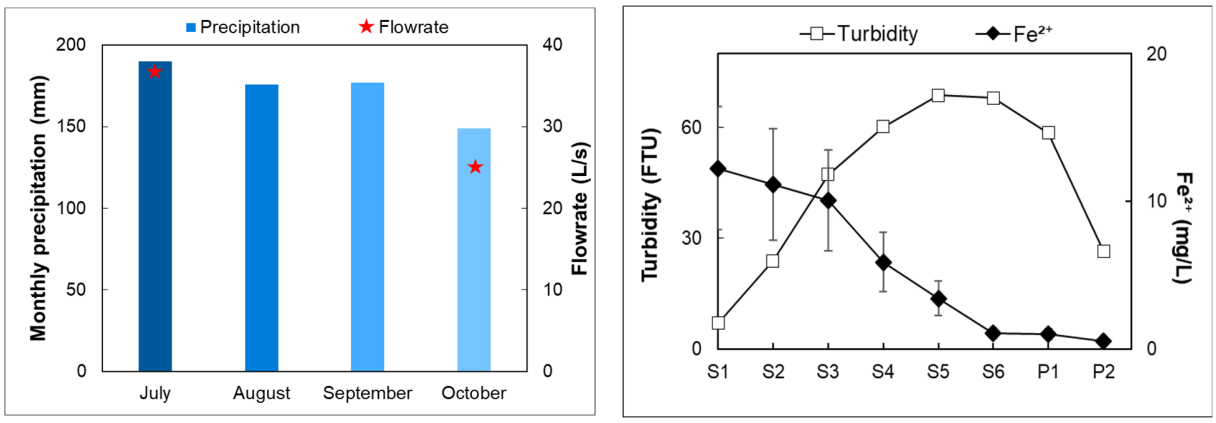

Figure S1. Average trends of the (a) Precipitation and flow rates at Ainai mine; (b) Fe^{2+} concentration (pack test) and turbidity in July 2016 and October 2018

Figure S1a summarizes the variation in rainfall and flowrate at Ainai mine drainage. Various sampling programs were carried out in July and October, from which the variations in parameters warranted further understanding on the drainage in the different months.

Figure S1b is a trend average of the turbidity and ferrous Fe concentration from pack test. An inverse relationship is observed, which represents the formation of colloids by oxidative precipitation of the ferrous Fe. The turbidity subsequently increases due to increase in particle number and size. Even when the ferrous Fe is depleted, turbidity continues to increase due to aggregation of the particles, that later settle and are removed from the drainage.

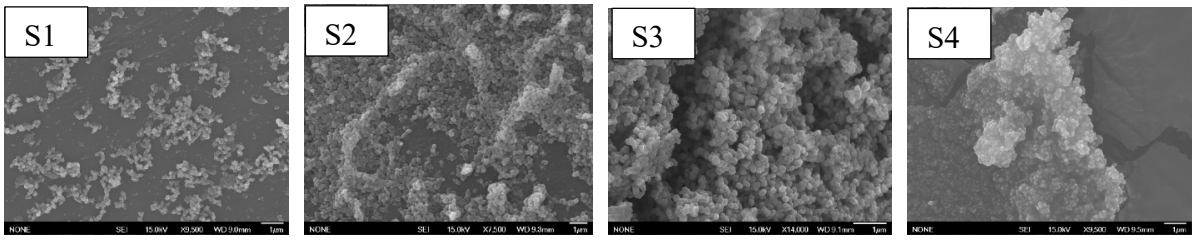

Figure S2. Size and aggregation variations in the Ainai mine drainage.

Figure S2 shows an increase in abundance and size of the colloids, initially signifying colloid formation and later colloid aggregation, therefore correlating with the turbidity trend in Figure S1b.

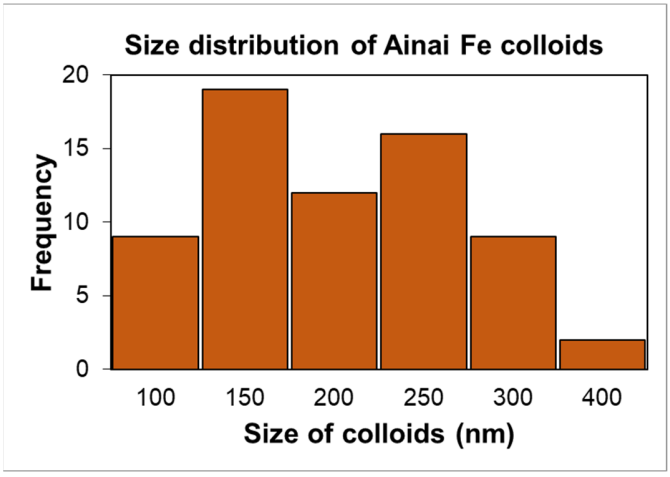

Figure S3. Size distribution of the Fe colloids in the drainage

Figure S3 shows that most of the colloids in the drainage range from 100 to 300 nm and very few exist beyond that size, therefore a size for over 300 nm may be interpreted as the ideal settling size for the colloids from the drainage. Particularly, the settling of the colloids was towards the downstream of the drainage, as seen from the increasing aggregated behavior displayed in Figure S2.

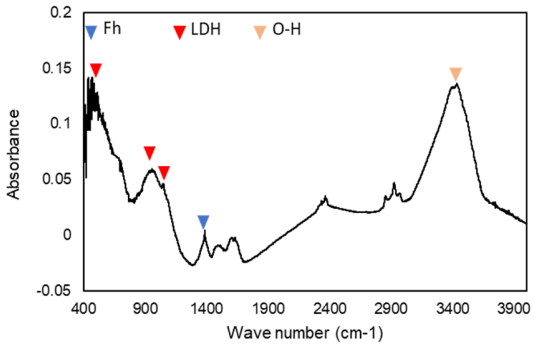

Figure S4. FTIR pattern of natural Zn–Fe LDH

The FTIR pattern of the natural Zn-Fe LDH in shown in Figure S4 and peaks corresponding to Zn-Fe LDH were observed and act as supporting data to the finding and characterization of Zn-Fe LDH at Ainai mine drainage.

Table S1. Physiochemical characteristics of Ainai mine wastewater in July 2016 and October 2018.

Site	pH		DO (mg L^{-1})		EC (ms m^{-1})		Eh (V)		Turbidity (FTU)		Fe^{2+} (ppm)		Alkalinity (mg L^{-1})		Flow rate (L s^{-1})	
	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct
S1	6.20	6.24	6.54	7.97	145.75	150.0	0.2	0.2	7.16	5.11	8.00	14.00	246.2	242.7	26.1	21.1
S2	6.43	6.55	7.79	11.28	135.50	137.0	0.1	0.1	28.77	20.90	7.50	12.00	221.9	147.5	-	-
S3	6.95	7.01	8.37	12.55	134.80	136.0	0.3	0.3	57.00	34.90	7.30	10.00	215.7	147.2	43.4	31.7
S4	7.38	7.42	9.23	11.94	131.93	137.0	0.3	0.3	68.00	50.67	1.50	5.00	221.3	156.9	-	-
S5	7.39	7.53	8.71	11.56	124.30	137.0	0.3	0.3	79.67	52.00	0.15	1.00	209.7	142.5	28.9	16.1
S6	7.34	7.52	9.23	11.46	131.96	136.0	0.4	0.4	89.66	38.50	0.05	0.10	199.1	156.8	-	-
P1	7.57	7.59	8.93	11.35	133.33	135.0	0.4	0.4	72.33	48.35	0.5	0.10	169.7	128.1	-	-
P2	7.82	7.91	9.37	11.63	273.0	148.64	0.4	0.4	19.11	2.32	0.5	0.20	169.2	127.1	-	-

- indicates locations where parameter was not measured.

Table S1 summarizes the on-site data of the Ainai mine drainage, for which some representative plots are shown in the main text figures.

Table S2. Fe, As, and Zn concentrations in dissolved and colloidal fractions in July and October.

Site	Fe concentration (mg L^{-1})				As concentration (μg L^{-1})				Zn concentration (μg L^{-1})			
	Dissolved		Colloidal		Dissolved		Colloidal		Dissolved		Colloidal	
	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct	Jul	Oct
S1	12.1	10.16	1.39	0.52	13.0	30.76	3.08	1.27	452	247	0.15	b.d.
S2	2.51	2.14	8.79	6.12	4.43	11.07	7.28	12.57	316	281	3.07	23.1
S3	0.067	b.d.	8.65	5.13	3.68	9.27	6.23	12.57	208	214	10.6	34.2
S4	0.017	b.d.	2.86	1.95	3.84	9.26	1.67	10.09	115	187	60.9	54.4
S5	0.019	b.d.	0.066	1.04	3.80	7.99	0.069	2.56	111	123	0.02	50.1
S6	0.016	b.d.	0.175	0.41	3.76	8.44	0.028	1.10	147	138	0.01	76.7
P1	0.015	b.d.	0.009	0.10	3.53	8.06	0.029	1.00	104	116	0.01	b.d.
P2	0.015	b.d.	0.004	0.05	2.43	7.73	0.000	b.d.	60.1	105	0.01	20.1

*b.d. = below detection limit

The concentrations of Fe, As and Zn in the drainage are shown in Table S2 according to their colloidal and dissolved fractions.

Table S3. pH, Fe, and Zn concentrations for synthetic experiments.

Sample	**pH**		**Fe (mg L^{-1})**		**Zn (μg L^{-1})**	
	Initial	Final (3 d)	Before	After (3 d)	Before	After (3 d)
Zn:Fe (2:1)	6.19	7.82	13.40	3.21	249.5	29.30

Table S3 summarizes the findings from the experimental aspect of Zn-Fe LDH synthesis, that utilized wastewater samples to synthesize the Zn-Fe LDH by adding a Zn reagent.