SUPPORTING INFORMATION

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Laboratory investigations of enhanced sulfate reduction as a groundwater arsenic remediation strategy.

Five pages, including this cover sheet.

Eight tables.

Table S1.

| | Observed Concentrations (mM) | | | Concentrations in |
Parameter	Mean ± Std. Dev.	Median	(N)	artificial GW (mM)[a]
pH	7.2±1.1	7.1	(25)	7.53 (initial)
Sodium	3.57 ± 4.14	1.25	(17)	3.89
Potassium	0.32 ± 0.20	0.28	(20)	0.26
Calcium	1.88 ± 0.61	1.59	(16)	1.47
Magnesium	0.71 ± 0.13	0.70	(55)	0.62
Chloride	0.67 ± 0.29	0.73	(17)	0.86
TIC[b]	0.33 ± 0.00	0.33	(6)	0.32
Sulfate	2.97 ± 3.92	0.97	(17)	3.56
Phosphate[c]	--	--		0.05

Geochemical parameters from groundwater monitoring (1999-2000) and artificial groundwater composition.

[a] No iron or arsenic were added to the artificial groundwater; concentrations were assumed to be negligible.
[b] Total inorganic carbon was measured; carbonate was added to artificial groundwater as the only source of inorganic carbon.
[c] Phosphate was not measured for field monitoring, so a typical groundwater concentration was added to artificial groundwater.

Table S2.

#	Reaction	Log K
1	$As(OH)_3^0 + HS^- + H^+ \leftrightarrow As(OH)_2SH + H_2O$	8.69
2	$As(OH)_3^0 + HS^- \leftrightarrow As(OH)_2S^- + H_2O$	3.54
3	$As(OH)_2S^- + HS^- \leftrightarrow As(OH)S_2^{2-} + H^+$	5.06
4	$As(OH)S_2^{2-} + HS^- + H^+ \leftrightarrow AsS_3H^{2-} + H_2O$	11.78
5	$As(OH)S_2^{2-} + HS^- \leftrightarrow AsS_3^{3-} + H_2O$	3.89

Reactions reported in Wilkin et al (24) for formation of thioarsenites. Reactions 1 and 4 are less favorable under the incubation conditions reported here due to the relatively high pH (low [H+]).

Table S3.

Incubation period	Geochemical Conditions	_[As]	_[Fe]	_[Mn]
0 to 8 days	Rising As, Fe, Mn	1.66	110.34	4.65
8 to 13 days	Falling As, Fe, Mn	(1.58)	(102.85)	(3.37)
13 to 28 days	Rising As, stable Fe, Mn	1.13	(7.37)	(2.05)
28 to 39 days	Falling As, stable Fe, Mn	(1.17)	0.14	0.02
39 to 79 days	Stable As, Fe, Mn	(0.08)	(0.11)	0.00
0 to 79 days		(0.04)	0.14	(0.74)
Maximum concentration to 79 days		(1.70)	(110.19)	(5.58)

Arsenic, iron, and manganese mass balances for the microcosm incubations during key geochemical time periods. The change in aqueous concentrations is shown, with decreasing concentrations shown in parentheses; net changes are assumed to be due to transfer of mass to the solid phase. Concentrations are in μmol L^{-1}.

Table S4

Incubation period	_[SO$_4$] (by IC)	_[HS] (by colorimetry)	_ Σ[S] ([SO$_4$] + [HS])	_ΣS (by ICPMS)
0 to 8 days	0[a]	0	0.00	2.23
8 to 13 days	(0.79)	0.02	(0.77)	(1.09)
13 to 28 days	(2.88)	1.25	(1.63)	(7.26)
28 to 39 days	0.08	(0.52)	(0.44)	(0.42)
39 to 79 days	0.11[b]	(0.41)	(0.30)	(0.04)
0 to 79 days	(3.70)[b]	0.34	(3.36)	(8.81)

Sulfur mass balance for the microcosm incubations. The change in aqueous concentrations is shown, with decreasing concentrations shown in parentheses; net changes are assumed to be due to transfer of mass to the solid phase and some transfer of sulfide to the gas phase. Concentrations are in mmol L^{-1}.

[a]As noted in the text, the IC sample at 0 days was lost, so the initial SO$_4$ concentration was estimated as the first measured concentration.
[b]No IC sample was available for 79 days, so the final SO$_4$ concentration was taken to be that measured at 44 days.

Table S5.

Incubation Period	_[Acetate]	_[Carbonate] from alkalinity	Σ_C
0 to 8 days	(5.57)[a]	5.40	(5.74)
8 to 13 days	(7.35)	(2.89)	(17.58)
13 to 28 days	(7.08)	2.64	(11.53)
28 to 39 days	0.00[a]	(0.15)	(0.15)
39 to 79 days	0.00	1.72	1.72
0 to 79 days	(20.00)	6.72	(33.28)

Carbon mass balance for the microcosm incubations. The change in aqueous concentrations is shown, with decreasing concentrations shown in parentheses; net changes are assumed to be due to transfer of mass to the solid phase as carbonate minerals as well as increasing CO_2 and CH_4 concentrations in the headspace. Concentrations are in mmol L^{-1}.

[a]As noted in the text, the IC sample at 0 days was lost, so the initial acetate concentration was estimated as the first measured concentration.
[b]Acetate was below detection from 28 days until the end of the experiment.

Table S6.

Incubation period	Geochemical Conditions	_[As]	_[Fe]	_[Mn]
0 to 12 days	Rising As, Fe, Mn	0.82	28.78	1.96
12 to 28 days	Falling As, Fe, Mn	(0.96)	(28.85)	(2.63)
28 to 40 days	~Steady As, Fe, Mn	0.39	0.14	0.11
0 to 40 days		0.25	0.07	(0.56)
Maximum concentration to 40 days		(0.57)	(30.11)	(2.53)

Arsenic, iron, and manganese mass balances for the reactor incubation. The change in aqueous concentrations is shown, with decreasing concentrations shown in parentheses; net changes are assumed to be due to transfer of mass to the solid phase. Concentrations are in μmol L^{-1}

Table S7.

Incubation period	_[SO₄] (by IC)	_[HS] (by colorimetry)	_Σ[S] ([SO₄] + [HS])
0 to 12 days	(1.65)	0.00	(1.65)
12 to 28 days	(0.65)	0.44	(0.21)
28 to 40 days	(2.29)	0.18	(2.11)
0 to 40 days	(4.60)	0.63	(3.97)

Sulfur mass balance for the reactor incubation. The change in aqueous concentration is shown, with decreasing concentrations shown in parentheses; net changes are assumed to be due to transfer of mass to the solid phase and transfer of sulfide to the gas phase. Concentrations are in mmol L^{-1}.

Table S8.

Incubation Period	_[Acetate]	_[Carbonate] from alkalinity	Σ_C
0 to 12 days	(11.12)	(0.42)	(22.65)
12 to 28 days	(11.48)	0.64	(22.33)
28 to 40 days	(10.16)	0.62	(19.71)
0 to 40 days	(32.76)	0.84	(64.69)

Carbon mass balance for the reactor incubation. The change in aqueous concentration is shown, with decreasing concentrations shown in parentheses; Net changes are assumed to be due to transfer of mass to the solid phase as carbonate minerals as well as CO_2 and CH_4 which may have escaped from the reactor. Concentrations are in mmol L^{-1}.