Bio-weathering Using *Shewanella oneidensis* MR-1 Enhances Selective Recovery of Rare Earth Elements from Alaskan Coal Mines

Ankur Sachan[1,2,3], Subhabrata Dev[3,4], Brandon R. Briggs[5], Tathagata Ghosh[2,3*],
Srijan Aggarwal[3,4,5], Fahimeh Dehghani[2,3,6]

[1]Freeport McMoRan Technology, Central Mine Planning, Tucson, AZ, 85706, United States
[2]Department of Mining and Mineral Engineering, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[3]Mineral Industry Research Laboratory, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[4]Water and Environmental Research Center, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[5]Department of Biological Sciences, University of Alaska Anchorage, Anchorage, AK 99508, United States
[6]Department of Civil, Geological, and Environmental Engineering, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[7]Freeport-McMoRan, Chino Mine, Silver City, NM 88023, United States

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**Corresponding author.*
Tathagata Ghosh, 1760 Tanana loop, University of Alaska Fairbanks, AK 99775, United States, +1-907-474-6917, tghosh@alaska.edu

Abstract

Coal mines in Alaska with high rare earth elements (REEs) levels (286-524 mg/kg) serve as an alternative domestic source for REEs. Existing leaching/separation technologies fail to selectively recover REEs from the feedstock and require downstream multiple purification stages that increase the overall operational cost. This study aims at bio-weathering coal from two Alaskan coal mines (Wishbone Hill and Healy) at three specific gravity fractions (1.3 float, 1.3 and 1.5 sink) using *Shewanella oneidensis* MR-1 for achieving higher selective REEs recovery in a one-step process. Optimizing the bio-weathering process by varying solids percentages (5.7 to14.3% w/v), particle size (-14 to -200 M), incubation temperatures (30 to 34 °C), and inoculum dosing (0.2 to 1% v/v) resulted in highest recovery of Neodymium (75.3%) and total REEs (98.4%) from 1.3 float Wishbone Hill and 1.3 sink Healy coal, respectively. When compared to the chemical leaching process, the bio-weathering enhanced the selective recovery of REEs including Scandium, Yttrium, Ytterbium, Terbium, Erbium, and Lutetium from Healy coal at lower specific gravity, and Yttrium from Wishbone Hill coal at higher specific gravity. The results indicate the future scope for developing cost-effective selective REEs recovery processes that may address the global critical minerals supply chain risk.

Keywords. *Rare Earth Elements; Metal reduction; Biorecovery; Shewanella oneidensis MR-1.*

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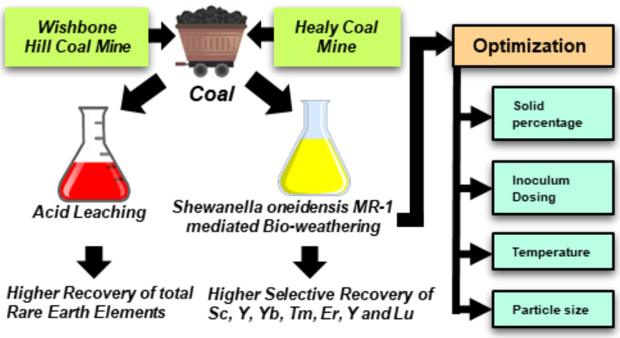

1. Introduction

The modern world economy is almost exclusively dependent on technology, from electronics, defense, medical equipment, and other industries like ceramics, electrical, chemical, nuclear, optical, catalytic, and metallurgical applications.[1] The raw materials for most of these industries require affluent supply of a group of critical elements known as the 'rare earth elements' (REEs) comprising of scandium (Sc) and yttrium (Y), and fifteen lanthanide series elements, namely, lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu).[2] The major producers of REEs are China (85%), followed by Australia (10%), Russia (2%), India (1%), Brazil (1%), and Malaysia.[2] The global REEs import to the United States in the year 2015 was reported majorly from China (77%), followed by Estonia (7%), France (4%), and Japan (4%).[3] Strict export norms on REEs by China in 2011 resulted in severe supply chain risk and economic setbacks in the countries highly dependent on the global imports of REEs.[4] Extraction of REEs from alternative sources including coal could be a suitable strategy to ease such supply scarcity.[5-7]

Accumulation of REEs into coal follows four genetic types including terrigenous, tuffaceous, infiltrational, and hydrothermal.[7] The REEs may be present in coal as polygenetic and multi-stage form. The presence of tonstein layer in the fire coal seam is correlated to higher levels of REEs in the coal samples.[8] The genetic types and chemical nature of REEs cause the fractionation of REEs in the coal seam.[9, 10] The heavy REEs (HREEs) including Eu, Tb, Gd, Y, Lu, Dy, Tm, Ho, and Er are reported to be present at high concentration in the coals with low ash content, whereas their concentrations are low in the coals with high ash content.[11] Desorption and leaching of HREEs from tonsteins, clays, and other rocks are higher than light REEs (LREEs).[12] Water percolating through these natural components gets enriched with HREEs, and may circulate within coal basins.[13, 14] Inclusion of humic substances during the coalification process has provided the stronger complexing ability of coal with HREEs than LREEs.[15]

Different physico-chemical strategies including gravity separation,[16] flotation,[17] magnetic separation,[18] solvent extraction,[19] roasting,[20] and leaching,[21] have been adopted for recovering REEs from coal and its byproducts. All these processes show less specificity for REEs with poor recovery capacity and require multi-stage downstream purification processes that increase overall operation costs.[2] Additionally, generation of heavy metals-rich sludge makes these processes environmentally unfavorable.[2, 22] Recovery of REEs from coal using environmentally sustainable bio-weathering processes has gained significant attention in the recent times due to its cost-effectiveness, and higher specificity allowing selective recovery of REEs.[23-25] In the recent times, reductive dissolution of metals by *Shewanella oneidensis* through extracellular electron transport (EET) system has been utilized for enhanced biorecovery of different metals including Cr, Mn, Fe, Cu, Pd, and Zn.[26-29] The reduction of metals can be achieved by accepting electrons from several electron donors including lactate, pyruvate, acetate, and formate.[30-32]

Total amount of coal mined in a year throughout the world can provide a long-lasting and alternative source of REEs. The US has large reserves of coal containing an average of 66 mg/kg REEs.[33] Moreover, Alaska contains 40% more coal than the whole of the contiguous United States and previous research has shown that coal from some of the Alaskan coal mines contains up to 525 mg/kg REEs which makes it a perfect candidate for extraction using novel bio-weathering methods that are cost-effective and environment-friendly.[34] Because of enhanced bio-weathering capacity and ability to grow at acidic conditions[35], *S. oneidensis* MR-1 strain has been selected for the first time as a suitable microorganism for recovering REEs from coal. This study aims at developing a bio-weathering process using *S. oneidensis* MR-1 for enhanced recovery of REEs in one-step process from coal collected from two different coal mines namely Wishbone Hill and Healy - both are located in the State of Alaska. In this work we considered optimizing the REEs biorecovery from coal by varying particle size, solid percentage, incubation temperature, and inoculum dosing at three different specific gravity fractions. These parameters are considered as important factors contributing to the microbial growth[2, 36] and metals recovery efficiency[37] during bio-weathering process. Successful development of this process could open doors for deploying REEs bioleaching and recovery at larger pilot and commercial scales in a cost-effective and environment-friendly manner from unconventional sources including coal, and address the global REEs supply demand.

2 Materials and Methods

2.1 Collection and preparation of coal samples

We obtained the coal and ash samples from Wishbone Hill and Healy Coal Mines (also known as Usibelli Coal Mine), both located in the State of Alaska. The ash and moisture contents of coal from both these mines vary between 45.8-2.7% and 23.7-9.4%, respectively.[34, 38] Samples from both the coal mines also have low sulfur content ranging from 0.2- 0.4%.[34] The collected coal samples were crushed and screened to 200 mesh size based on the previous report on effective bio-weathering at size 200 mesh.[1] Coal samples from both the coal mines were segregated into three specific gravity ranges, namely 1.3 float, 1.3 sink, and 1.5 sink. The coal was pasteurized by alternating between 80 and 4 °C, each with one hour holding time. This was repeated three times to prevent the contamination of *S. oneidensis* MR-1 pure culture during bio-weathering process.

2.2 Bio-weathering of coal

Bio-weathering of coal and ash samples was performed using *S. oneidensis* MR-1 inoculum grown in “Luria-Bertani” (LB) broth at 30 °C for 24 h. The inoculum was then added to a 50 ml serum bottle with a minimal media containing 20 mM of sodium L-lactate ($C_3H_5NaO_3$, 98%), 10 mM of sodium bicarbonate ($NaHCO_3$), 29.76 mM of 1, 4 piperazinediethanesulfonic (PIPES, ≥99% titration), 1.34 mM of potassium chloride (KCl, BioXtra, ≥99%), 0.2 mM of anthraquinone-2, 6-disulfonate (AQDS), and coal (5.7% w/v). After inoculation, the serum bottles were capped with a butyl rubber stopper and sealed with a crimp cap. Resazurin was used to identify oxidation states of the media. We performed float-sink tests[39] to segregate the coal and ash samples into following three specific gravity fractions; 1.3 float, 1.3 sink, and 1.5 sink. We compared REEs biorecovery from coal and ash samples at all the three specific gravity fractions by conducting bio-weathering experiment in a batch condition for twenty days. We also performed additional batch experiments to optimize REEs biorecovery from coal samples by varying solids percentages, inoculum dosing, incubation temperature, and particle size (Table 1). Three different experimental conditions (A) -200M, 5.7% w/v, 30ºC, 1% v/v, (B) -200M, 10% w/v, 32ºC, 1% v/v, and (C) -48M, 14.3% w/v, 30ºC, 0.2% v/v, were designed to compare the yield of REEs bio-recovery from Wishbone and Healy coal samples. Samples from each specific gravity were divided into five sets, from which three were inoculated with bacteria, and the other two served as controls. As the experiment progressed, the serum bottles were sparged with air passed through 0.22 µm syringe filter at five-days intervals.

2.4 Comparison of acid leaching and bio-weathering

A comparative batch experiment was performed to determine the efficiency of REEs recovery using both acid leaching and *S. oneidensis* MR-1 mediated bio-weathering process. In the acid leaching process, coal samples with -14M and +48 M particle size were added with 1.2 M sulfuric acid for each specific gravity fractions and incubated at 75 °C for 48 hours. The percentage recovery of the REEs by acid leaching process was compared with that of the bio-weathering process.

2.4 REEs analysis

Samples from both bio-weathering and acid leaching were filtered (0.22 μm) using vacuum filtration units and analyzed for REEs. Before bio-weathering and leaching experiments, the coal and ash samples were also characterized for REEs content. The samples were sent to ALS Geochemistry (Fairbanks, USA) where REEs analysis were performed using Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The detection limit of the method used for REEs analysis was 0.005 μg/L.

2.5 Statistical analysis

Data obtained from control and *S. oneidensis* mediated bio-weathering experiment for REEs recovery from both Wishbone Hill and Healy coal were tested for normality using Kolmogorov Smirnov test (Table S1) and considered normally distributed if the null hypothesis of normality was not rejected (P>0.05). Subsequently, Mann Whitney Wilcox test (α =0.05) was used to test significance differences (for non-normal samples) in REEs recovery.

3. Results and Discussion

3.1 Residual REEs in coal and ash

The residual concentration of REEs in the coal and ash samples, on a whole coal basis, collected from Wishbone Hill and Healy coal mines are presented in table 2. The REEs, including Ce, Y, Nd, La, and Sc constituted the majority of total REEs concentrations in each of the samples analyzed. The total REEs concentrations in the coal samples from Wishbone Hill mine were slightly higher than that of Healy coal mine, except for 1.3 float specific gravity. On ash basis Healy coal has been found to have significantly higher concentrations of REEs.[34]

Coal byproducts, fly ash, cinders and bottom ash, from the University of Alaska Fairbanks' power plant were previously analyzed for REEs content.[34] The cinders samples were found to contain high amounts of fixed carbon and volatile matter with 67% ash content. Some amount of carbon and volatile matter were retained in the fly ash samples, along with a higher concentration of sulfur and 84% ash value. The bottom ash samples were devoid of carbon and contained trivial concentrations of volatile matter and sulfur, with an ash value of 99%. The REEs content of the power plant products ranged from 217-231 mg/kg, which was almost similar to that we observed in ash, samples from Wishbone Hill and Healy coal mines. The ash samples, upon investigation were found to be fused in a glassy matrix, around 10 μm in size, requiring significant grinding energy to liberate the particles, making the process cost prohibitive.

3.2 Biorecovery of REEs using S. oneidensis

S. oneidensis MR-1 resulted in higher biorecovery of REEs from coal and ash samples than controls that did not contain any bacteria (Fig. 1). While some REEs were released into the solution in the control samples, this amount (0.6-230 mg/L total REEs) was significantly lower ($p=1\times10^{-2}$-9.4×10^{-6}) than the experimental samples from *S. oneidensis* mediated bio-weathering (Table 3). The release of REEs in experimental samples could potentially be influenced by AQDS which may act as an electron shuttle for *S. oneidensis* and has been shown to increase iron reduction rates.[26, 40] AQDS is also known to chelate iron and some REEs, and could thus potentially cause the release of REEs from the coal matrix in control samples.[41-45] However, the efficiency of AQDS is very low and does not account for the higher REEs extractions observed in the experimental samples.

The percentage of REEs recovered was, in general, higher for coal samples from Healy than Wishbone Hill. Statistical p-values for the difference in the REEs recovery between the Wishbone Hill and Healy samples are presented in Table 3. This higher yield from Healy coal samples was despite Wishbone Hill coal having higher REEs content, on a whole coal basis, for most size fractions (Table 4). Additionally, Wishbone Hill coal contained a higher concentration of heavy metals including As, Cd, and Pb (Table S2) that may have inhibited *S. oneidensis* MR-1.[46, 47]

3.3 Effect of solids percentage

Effect of solids percentage on the biorecovery of REEs from Wishbone Hill and Healy coal is presented in Fig. 2. In Wishbone Hill coal, the highest total REEs recovery was attained at 10% (w/v) solids percentage, followed by 5.7 and 14.3% (w/v) (Fig. S1). At fixed solids percentage, the total REEs biorecovery decreased with increasing specific gravity from 1.3 float to 1.5 sink for Wishbone Hill coal samples. One of the critical REEs, Nd showed best recovery (75.3%), followed by Sc (67.2%) from Wishbone Hill coal with specific gravity of 1.3 float. The optimum biorecovery of total REEs from Wishbone Hill coal samples was obtained at 10% (w/v) solids percentage for all specific gravity values.

The biorecovery of total REEs were significantly higher from Healy than Wishbone coal samples at all solids percentages considered in this experiment. The optimum biorecovery of total REEs was attained at 10% (w/v) solid percentage from Healy coal samples for both composite and 1.5 float, whereas for 1.3-1.5 sink samples, it was attained at 5.7% (w/v) solid percentage. The highest biorecovery was obtained for La (96%) at 5.7 % (w/v) solids percentage from Healy coal with a specific gravity of 1.3 float and 1.3 sink, followed by Lu (93%), and Y (90%) at 5.7 % (w/v) solids percentage from samples with a specific gravity of 1.3 sink.

Increasing the solids percentage from 5.7 to 10% (w/v) increased the REEs biorecovery, probably because of the increase in REEs content. Further increasing the solids percentage to 14.3% (w/v) decreased the REEs biorecovery, possibly because of the inhibition of microbial growth associated with increasing shear force, limited oxygen flux, and higher toxic metal load.[40] Additionally, the high solids percentage interfered with the medium's pH, which might have affected the bio-weathering process.[41] The decrease in REEs biorecovery upon increasing specific gravity was probably because of an increase in ash content (Table S3) that may contain higher concentrations of As, Cd, and Pb (Table S2).

3.4 Effect of inoculum dosing

The biorecovery of total REEs increased significantly with increasing inoculum dosing from 0.2-1% v/v for both Wishbone Hill and Healy coal samples at all the specific gravities (Fig. 3). In Healy coal, the highest biorecovery of total REEs was obtained from composite coal samples (67.1%), followed by 1.3 float (53.5%), 1.5 sink (16.4%), and 1.3 sink (14.3%). Among all the REEs present in Healy coal, the highest recovery was obtained for Sc (81.4%) from composite samples, followed by La (77.1%) from 1.3 float samples, and Lu (73.8%), Pr (71.7%) and Yb (71.3%) from composite samples. In Healy coal, the highest biorecovery of total REEs was obtained from 1.3 float samples (43.2%), followed by 1.3 sink (21.2%), composite (3.6%), and 1.5 sink (1.8%). Among the REEs present in Wishbone Hill coal, the highest recovery was obtained for Nd (75.3%), followed by Sc (67.2%), and Ce (49.8%) from 1.3 sink samples. Increasing inoculum dosing enhanced the activity of the microorganism and organic acids production that aided in the solubilization of REEs, and enhanced REEs biorecovery.[2, 42]

3.5 Effect of incubation temperature

Incubation temperature was varied to 30, 32, and 34 °C. Increasing the temperature from 30 to 32 °C increased the total REEs biorecovery, which decreased subsequently upon increasing the temperature to 34 °C (Fig. 4). The highest total REEs biorecovery from Wishbone Hill coals was obtained at a specific gravity of 1.3 float (43.2%), followed by 1.3 sink (21.2%), composite (3.6%), and 1.5 sink (1.8%) (Fig. S2). Among the REEs present in Wishbone Hill coal, the highest biorecovery was obtained for Sc (67.2%), followed by Ce (49.8%), Sm (38.8%), Lu (38.5%), and Gd (33.6%) at a specific gravity of 1.3 float.

Highest total REEs biorecovery from Healy coals was obtained from composite samples (67.1%), followed by 1.3 float (53.5%), 1.5 sink (16.4%), and 1.3 sink (14.3%) (Fig. S2). The highest REEs recovery was obtained for Sc (81.4%), followed by La (77.1%), and Pr (71.7%) from composite samples, and Ce (67.8%) from 1.3 float.

3.6 Effect of particle size

The highest total REEs biorecovery was observed at a particle size of -14M, followed by -48M, and -200M from both Wishbone Hill and Healy coal samples (Fig. 5). In Wishbone Hill coal, the highest total REEs biorecovery was obtained for 1.3 float (43.2%), followed by 1.3 sink (21.2%), composite (3.6%), and 1.5 sink (1.8%) (Fig. S3). Among all the REEs from Wishbone Hill coal, the highest biorecovery was obtained for Nd (75.3%), followed by Sc (67.2%), Ce (49.8%), and Sm (38.8%) from 1.3 float samples.

3.7 Comparison of yield between Wishbone Hill and Healy coal

The yield was calculated for three sets of experiments with the following conditions, (i) -200M, 2 g, 30ºC, 1% v/v, (ii) -200M, 3.5 g, 32ºC, 1% v\v, and (iii) -48M, 5 g, 30ºC, 0.2% v/v. For tabular and graphical purposes, the three conditions are named A, B, and C, respectively. The yield of REEs biorecovery was <2% for all the conditions studied (Table 4). The highest yield was obtained from Healy coal samples for 1.3 float (1.73%), followed by 1.3 sink (1.70%), and 1.5 sink (1.69%) at condition C. For conditions A and B, higher yields were obtained from Healy coal samples at all the specific gravity fractions. The composite samples from Wishbone Hill and Healy coal showed comparable yields at condition C. There is no definite pattern between yield percentages in relation to the different conditions, although the yields for Wishbone Hill are less than 1% for all the conditions.

3.8 Comparison of bio-weathering and acid leaching process

In Wishbone Hill coal, the total REEs recovery is relatively higher for acid leaching than the bio-weathering process for all the samples (Fig. 6). The highest total REEs recovery (58.3%) was obtained from Wishbone Hill for 1.3 float samples by acid leaching. In case of the recovery of individual REEs from Wishbone Hill coal, the recovery of Nd, Sc, Er, and Lu was higher using bio-weathering than acid leaching process from 1.3 float samples (Fig. 6A). Similarly, the recovery of Lu, Y, and Sc from 1.3 sink samples was higher using the bio-weathering process than the chemical/acid leaching. Recovery of REEs using both the processes from Wishbone Hill coal was much lower in composite and 1.5 sink samples as compared to 1.3 float and 1.3 sink samples.

As compared to Wishbone Hill coal samples, the Healy coals responded better to the bio-weathering process for composite samples. The total REEs recovery was higher using bio-weathering than the acid leaching process for the composite sample, and comparable in case of 1.5 sink sample, whereas total REEs recovery was lower compared to acid leaching for 1.3 float and 1.3 sink samples (Fig. 6B). All the individual REEs showed higher recovery using the bio-weathering process from composite Healy coal samples. Similarly, higher recovery using bio-weathering was obtained for Y from 1.3 sink samples, and Ce, La and Y from 1.5 sink samples. The results indicate that Sc, Yb, Er, Tm, Y, and Lu were selectively leached out in the bio-weathering process more efficiently than any other REEs from Healy coal samples.

Conclusions

Coal from two mines in Alaska, Wishbone Hill and Healy, was used as feedstock for a novel bio-weathering approach performed at circumneutral pH. Cycling between oxic and anoxic conditions allowed *S. oneidensis* MR-1 to extract REEs from the coal matrix significantly greater than abiotic controls. The REEs biorecovery increased upon increasing solid percentage upto 10% w/v for Wishbone Hill coal independent of specific gravity, whereas increasing solid percentage showed a decrease in REEs biorecovery from Healy coal at higher specific gravity. The optimum inoculum dosing, temperature, and particle size were 1% v/v, 32°C, and -14 M, respectively, for coal samples from both the mines. Comparison of the chemical and bio-weathering process showed that higher selective recovery of specific REEs including Sc, Y, Yb, Tm, Er, and Lu occurred upon bio-weathering than the chemical leaching process. This study establishes the efficient REEs bio-weathering process using *S. oneidensis* MR-1, and creates the future potential for developing cost-effective and environmental-friendly technology for selectively recovering REEs from low-grade coals and coal rejects.

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Tables and Figures

Table 1. Variation of parameters for optimizing the bio-weathering of REEs from coal samples.

Parameters	**Variables**			
Particle size (M)	-200	-48	-14	+200
Temperature (°C)	30	32	34	
Inoculum dosing (% v/v)	0.2	0.4	1	
Solid percentage (% w/v)	5.7	10	14.3	

Table 2. The concentration of REEs in coal samples, on a whole coal basis, from Wishbone Hill and Healy coal mines.

Samples	**REEs (mg/Kg)**																
	Sc	**Y**	**La**	**Ce**	**Pr**	**Nd**	**Sm**	**Eu**	**Gd**	**Tb**	**Dy**	**Ho**	**Er**	**Tm**	**Yb**	**Lu**	**Total**
Composite																	
Wishbone coal	13.1	21.1	16.4	35.3	4.2	17.5	4.1	0.9	4	0.6	3.8	0.8	2.2	0.4	2.1	0.3	126.8
1.3 Float																	
Wishbone coal	7.1	13.3	4.8	10.6	1.3	5.7	1.4	0.3	1.7	0.3	2.1	0.5	1.4	0.2	1.5	0.2	52.4
Healy coal	3.4	9.7	8.6	17.3	2	8.6	2.1	0.4	2	0.3	1.6	0.3	0.9	0.1	0.8	0.1	58.2
1.3 Sink																	
Wishbone coal	11.9	19.4	15.3	31.9	3.8	15.8	3.7	0.8	3.6	0.6	3.4	0.7	2.1	0.4	2.2	0.3	115.9
Healy coal	4.6	16.6	16.8	33.8	4	16.2	3	0.8	3.4	0.5	3	0.6	1.6	0.3	0.6	0.2	106
1.5 Sink																	
Wishbone coal	17	26.4	23.7	51.1	6.1	25.1	5.8	1.2	5.4	0.9	4.9	1	2.7	0.4	2.5	0.4	174.6
Healy coal	9	19.9	28.1	52.1	6.2	23.4	4.8	0.9	4	0.6	3.7	0.8	2.1	0.3	2	0.3	158.2

Table 3. Mann Whitney Wilcox test to quantify the significant difference in the percentages of REEs recovery between the control and *S. oneidensis* mediated bio-weathering as well as between Wishbone and Healy coal.

Sample			**p**
Control vs *S. oneidensis*			
Wishbone Hill	Coal	1.3 float	2×10^{-3}
		1.3 sink	5.8×10^{-4}
		1.5 sink	**0.3**
	Ash	1.3 float	1.7×10^{-3}
		1.3 sink	2.7×10^{-5}
		1.5 sink	5.4×10^{-4}
Healy	Coal	1.3 float	1.8×10^{-4}
		1.3 sink	2.4×10^{-5}
		1.5 sink	9.4×10^{-6}
	Ash	1.3 float	5.3×10^{-5}
		1.3 sink	1×10^{-2}
		1.5 sink	8×10^{-3}
Wishbone Hill vs Healy			
Coal	1.3 float	Control	7.9×10^{-4}
		S. oneidensis	1.9×10^{-4}
	1.3 sink	Control	1.6×10^{-5}
		S. oneidensis	2.6×10^{-6}
	1.5 sink	Control	1.6×10^{-6}

		S. oneidensis	6.5×10^{-7}
Ash	1.3 float	Control	3.9×10^{-3}
		S. oneidensis	8.2×10^{-3}
	1.3 sink	Control	7.2×10^{-4}
		S. oneidensis	**0.7**
	1.5 sink	Control	1.7×10^{-5}
		S. oneidensis	3.6×10^{-6}

Table 4. Yield of REEs from Wishbone and Healy coal at different conditions; (A) -200M, 2 g, 30ºC, 1 % v/v, (B) -200M, 3.5 g, 32ºC, 1% v/v, and (C) -48M, 5 g, 30ºC, 0.2 % v/v.

Samples	**Specific gravity**	**Yield (%)**		
		A	**B**	**C**
Wishbone Hill	Composite	0.29	0.13	0.03
	1.3 float	0.23	0.45	0.36
	1.3 sink	0.25	0.16	0.35
	1.5 sink	0.77	0.15	0.84
Healy	Composite	0.33	0.46	0.02
	1.3 float	0.46	1.10	1.73
	1.3 sink	0.53	0.21	1.70
	1.5 sink	0.61	0.22	1.69

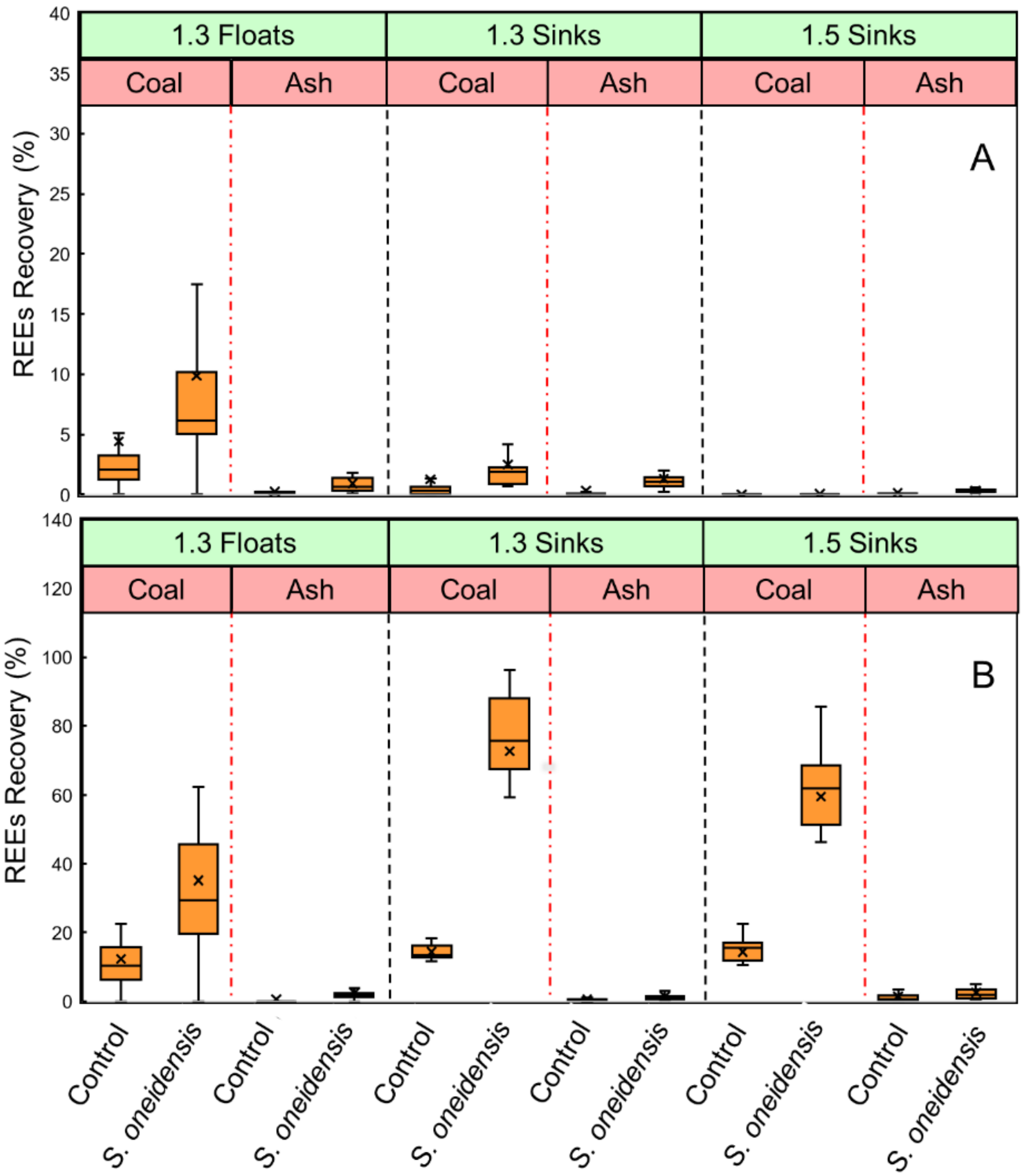

Figure 1. Percentage recovery of total REEs from Wishbone Hill coal (A), and Healy coal (B) samples using *S. oneidensis*. The serum bottles containing minimal media and coal but without *S. oneidensis* was used as a control.

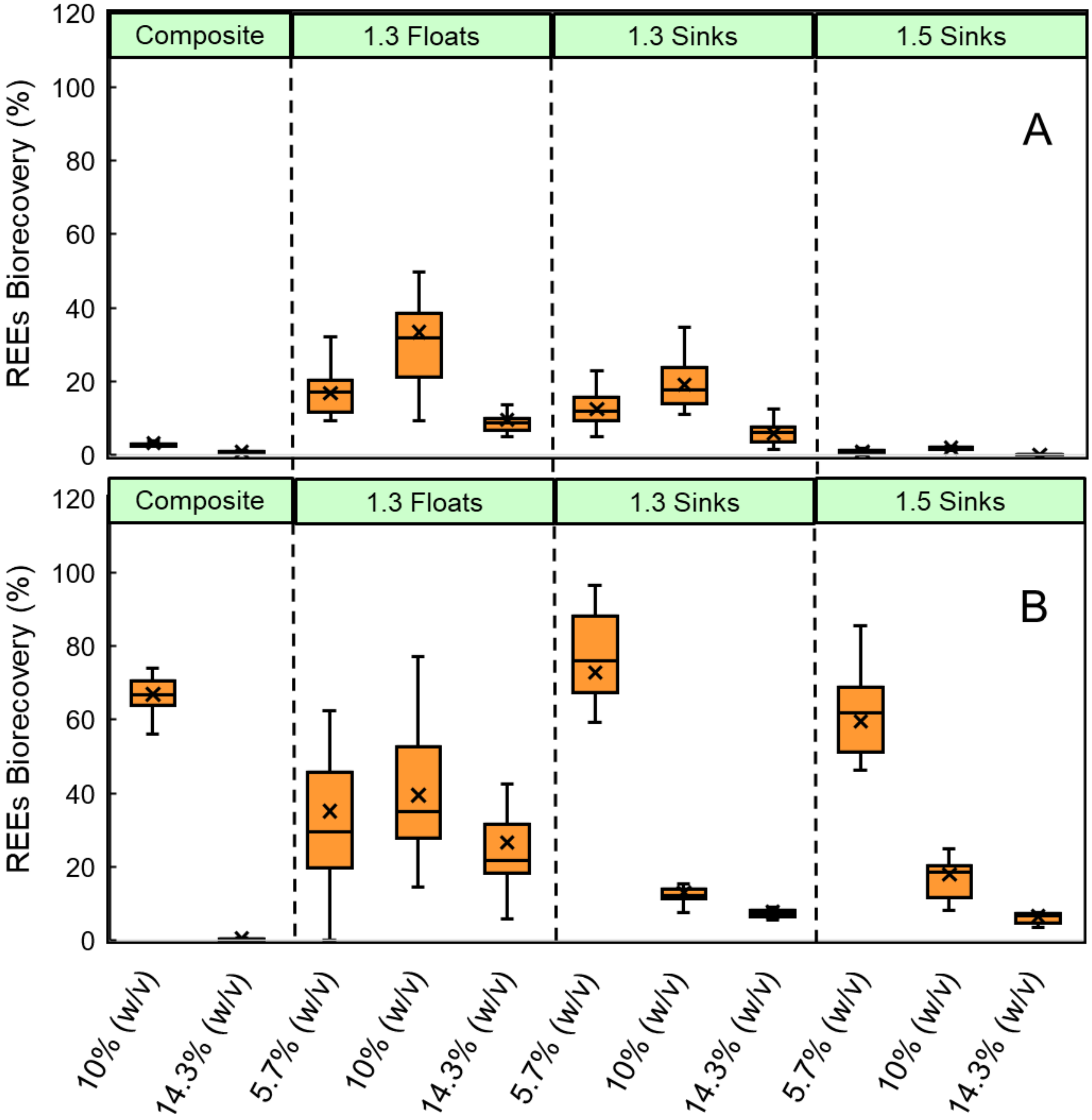

Figure 2. Recovery (%) of total REEs from Wishbone (A), and Healy (B) coal samples at varying solid percentages (% v/w).

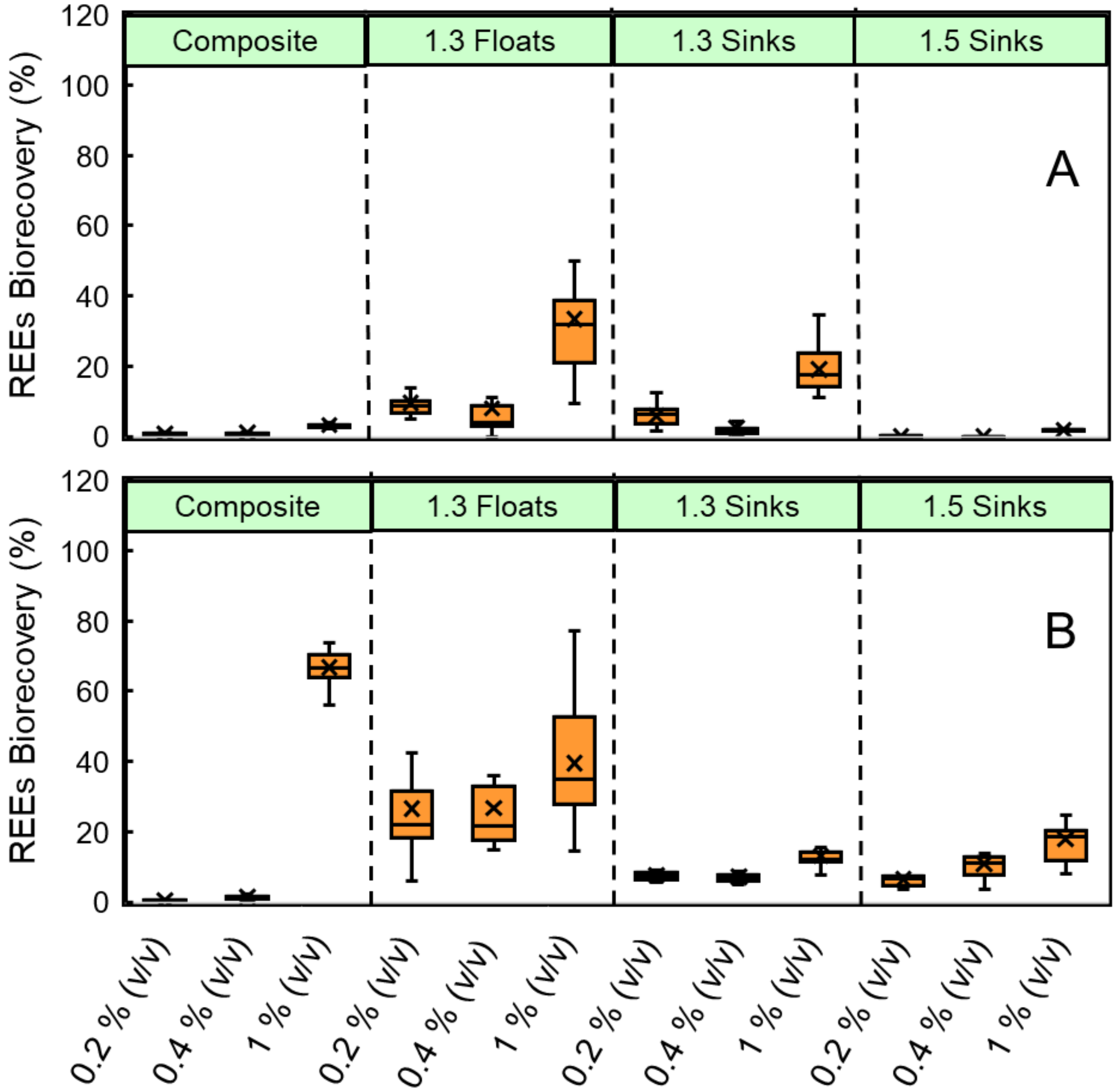

Figure 3. Recovery (%) of total REEs from Wishbone (A), and Healy (B) coal samples at varying inoculum dosings.

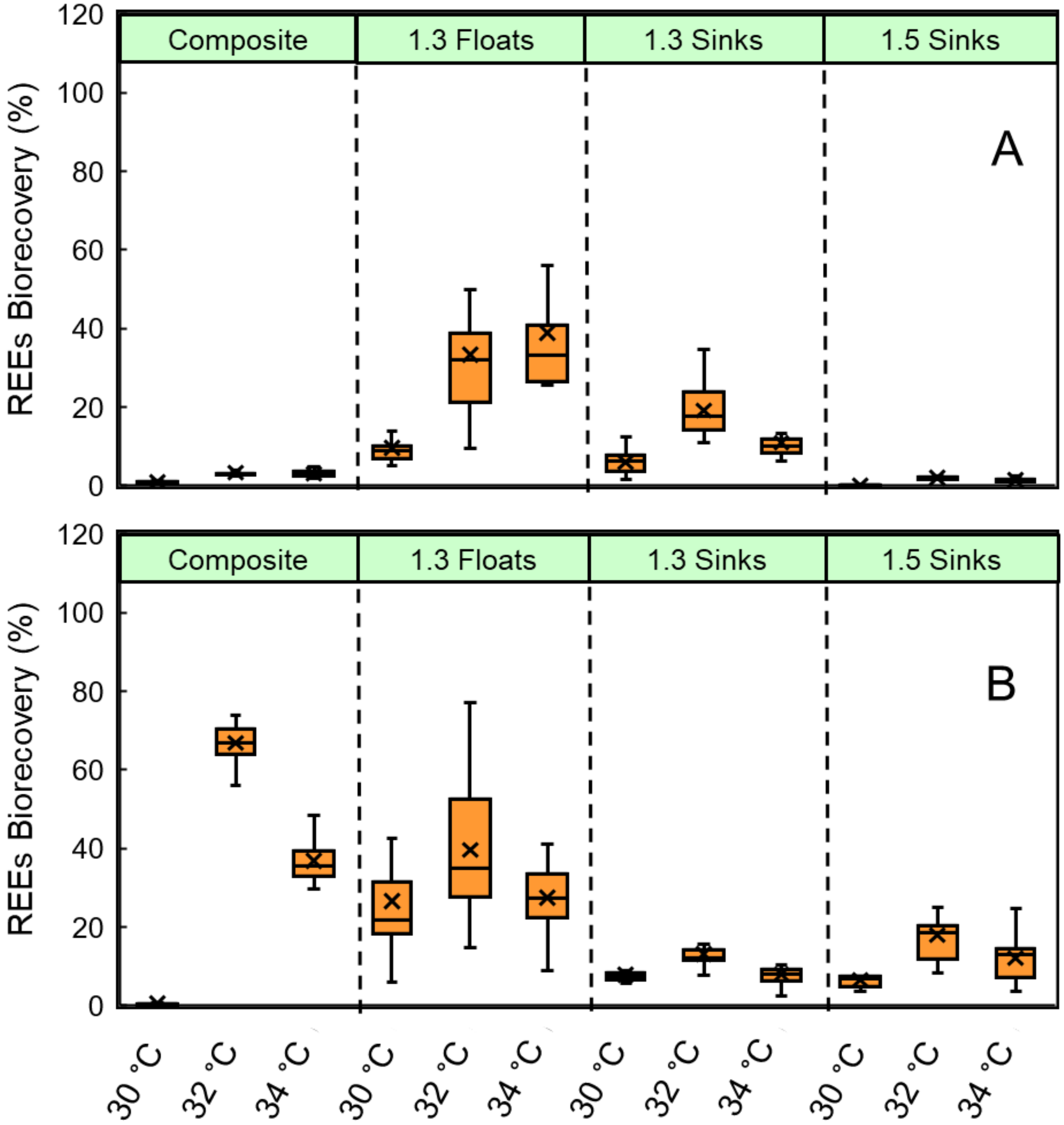

Figure 4. Recovery (%) of total REEs from Wishbone (A), and Healy (B) coal samples at varying temperatures.

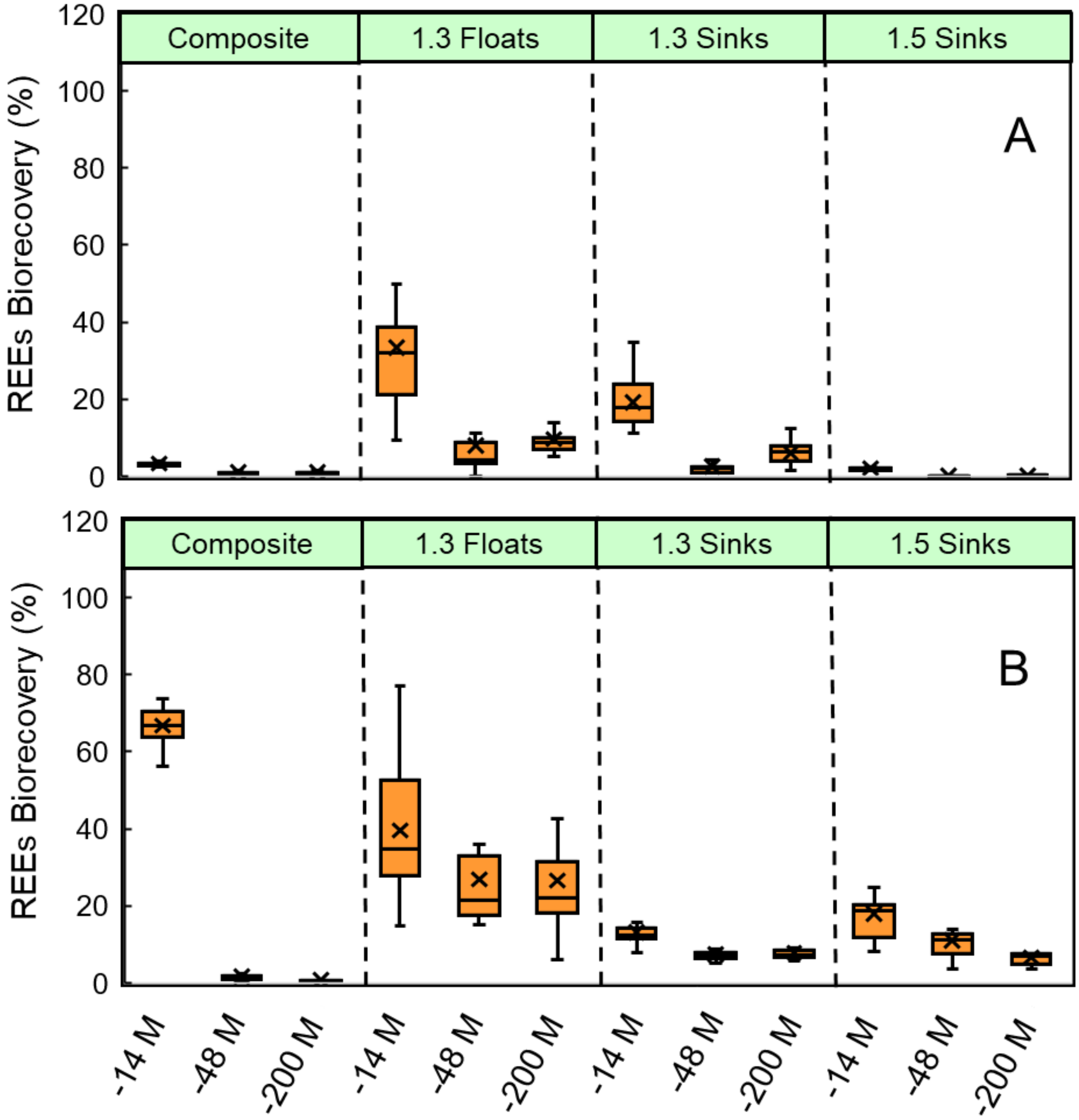

Figure 5. Recovery (%) of total REEs from Wishbone (A), and Healy (B) coal samples at varying particle sizes.

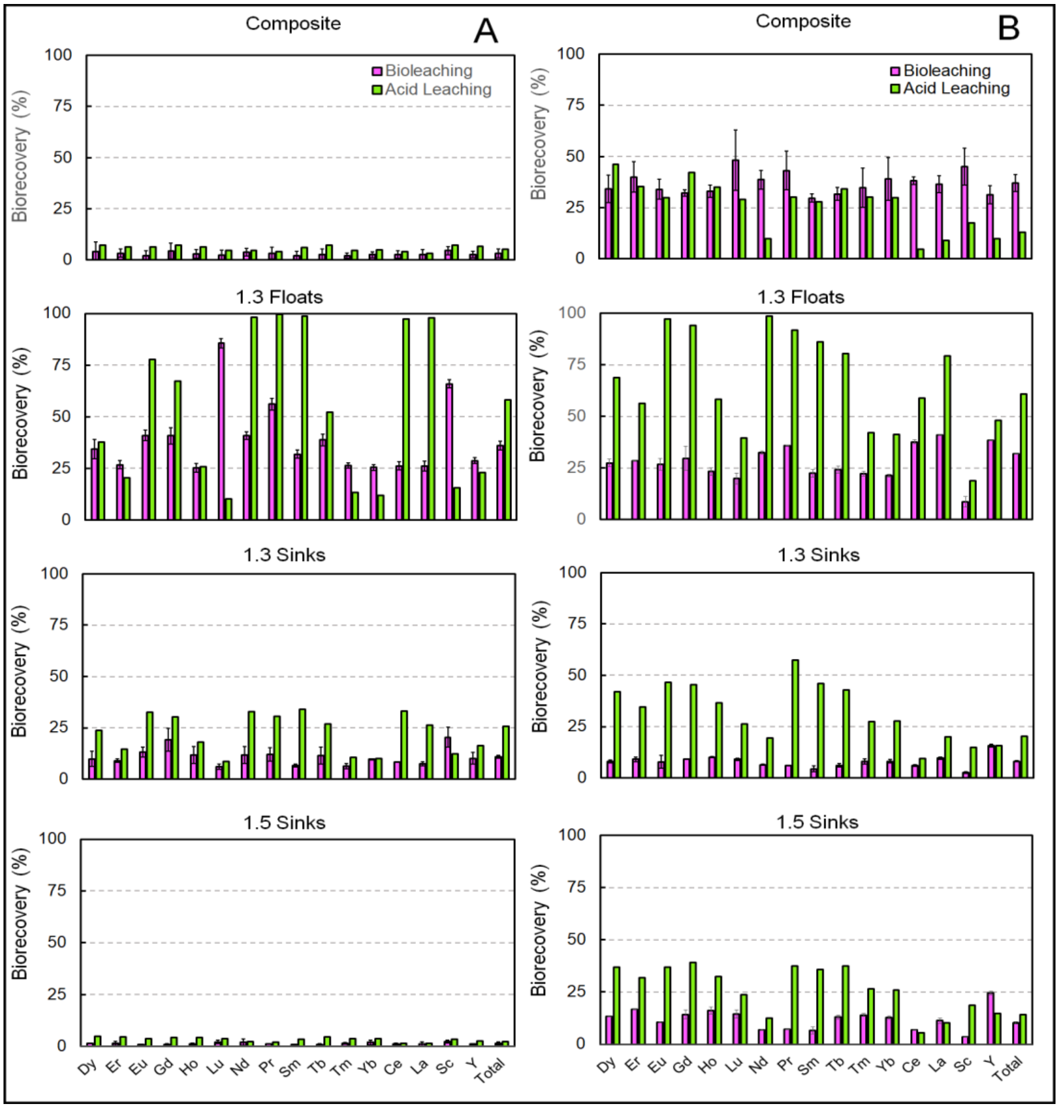

Figure 6. Comparison of chemical leaching and bio-weathering process for REEs recovery (%) from Wishbone Hill (A) and Healy coal (B) samples at different specific gravities.

Supplementary information

Bio-weathering Using *Shewanella oneidensis* MR-1 Enhances Selective Recovery of Rare Earth Elements from Alaskan Coal Mines

Ankur Sachan[1,2,3], Subhabrata Dev[3,4], Brandon R. Briggs[5], Tathagata Ghosh[2,3], Srijan Aggarwal[3,4,5], Fahimeh Dehghani[2,3,6]

[1]Freeport McMoRan Technology, Central Mine Planning, Tucson, AZ, 85706, United States
[2]Department of Mining and Mineral Engineering, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[3]Mineral Industry Research Laboratory, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[4]Water and Environmental Research Center, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[5]Department of Biological Sciences, University of Alaska Anchorage, Anchorage, AK 99508, United States
[6]Department of Civil, Geological, and Environmental Engineering, University of Alaska Fairbanks, Fairbanks, AK 99775, United States
[7]Freeport-McMoRan, Chino Mine, Silver City, NM 88023, United States

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Corresponding author. Tathagata Ghosh, 1760 Tanana loop, University of Alaska Fairbanks, AK 99775, United States, +1-907-474-6917, tghosh@alaska.edu

Table S1. Normality test (Kolmogorov Smirnov) to determine the distribution of REEs recovery (%) data.

Sample				***p***
Wishbone Hill	Coal	1.3 float	Control	9.2×10^{-8}
			S. oneidensis	1.1×10^{-13}
		1.3 sink	Control	3.1×10^{-4}
			S. oneidensis	4.2×10^{-9}
		1.5 sink	Control	3.1×10^{-4}
			S. oneidensis	3.1×10^{-4}
	Ash	1.3 float	Control	3.1×10^{-4}
			S. oneidensis	6.8×10^{-5}
		1.3 sink	Control	3.1×10^{-4}
			S. oneidensis	1.2×10^{-5}
		1.5 sink	Control	6.8×10^{-5}
			S. oneidensis	6.8×10^{-5}
Healy	Coal	1.3 float	Control	1.1×10^{-13}
			S. oneidensis	1.1×10^{-13}
		1.3 sink	Control	1.1×10^{-13}
			S. oneidensis	1.1×10^{-13}
		1.5 sink	Control	1.1×10^{-13}

		S. oneidensis	1.6×10^{-15}
Ash	1.3 float	Control	3.1×10^{-4}
		S. oneidensis	5.4×10^{-9}
	1.3 sink	Control	3.1×10^{-4}
		S. oneidensis	2.9×10^{-7}
	1.5 sink	Control	6.8×10^{-5}
		S. oneidensis	4.2×10^{-9}

Table S2. Concentration of As, Cd, and Pb in Wishbone and Healy coal samples

Coal samples	Specific gravity	As ppm	Cd ppm	Pb ppm	Total ppm
Wishbone Hill	Composite	4.62	0.364	12.05	17.03
	1.3 float	0.96	0.123	5.97	7.05
	1.3 sink	0.87	0.25	8.45	9.57
	1.5 sink	6.37	0.706	24.7	31.78
Healy	Composite	0.94	0.526	2	3.47
	1.3 float	0.28	0.217	1.38	1.88
	1.3 sink	0.67	0.481	2.16	3.31
	1.5 sink	1.45	0.434	0.78	2.26

Table S3. Ash content of Wishbone and Healy coal samples at different specific gravities.

Specific Gravity	Ash content (%)	
	Wishbone Hill	**Healy**
1.3 float	6.09%	11.89%
1.3 sink	28.81%	21.09%
1.5 sink	73.23%	56.09%

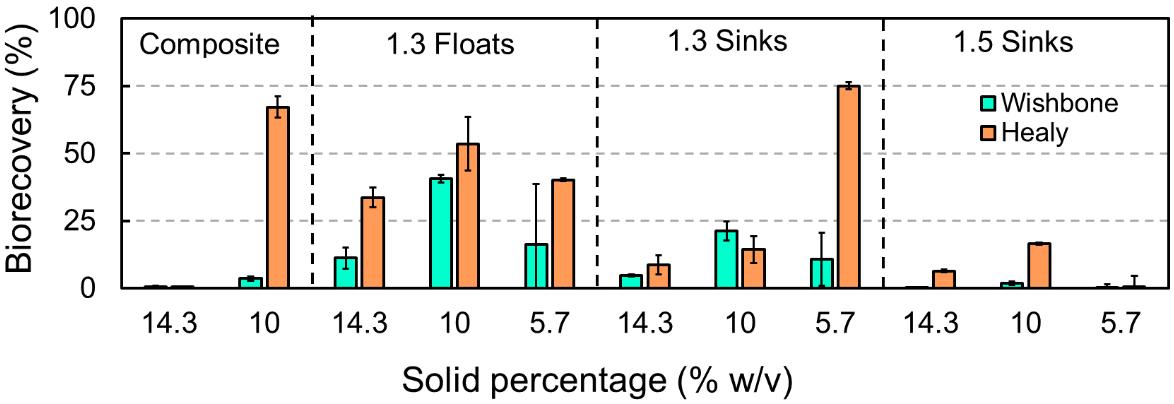

Figure S1. Total REEs recovery from Wishbone and Healy coal at varying solid percentages (% w/v).

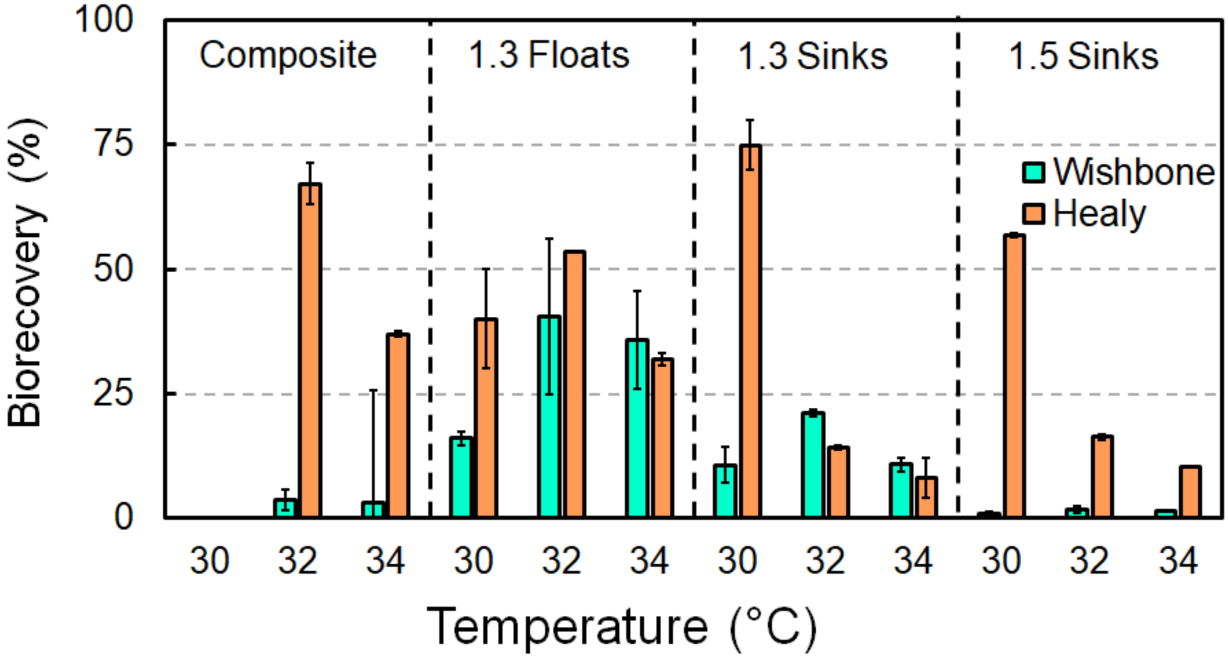

Figure S2. Total REEs recovery from Wishbone and Healy coal at varying incubation temperatures.

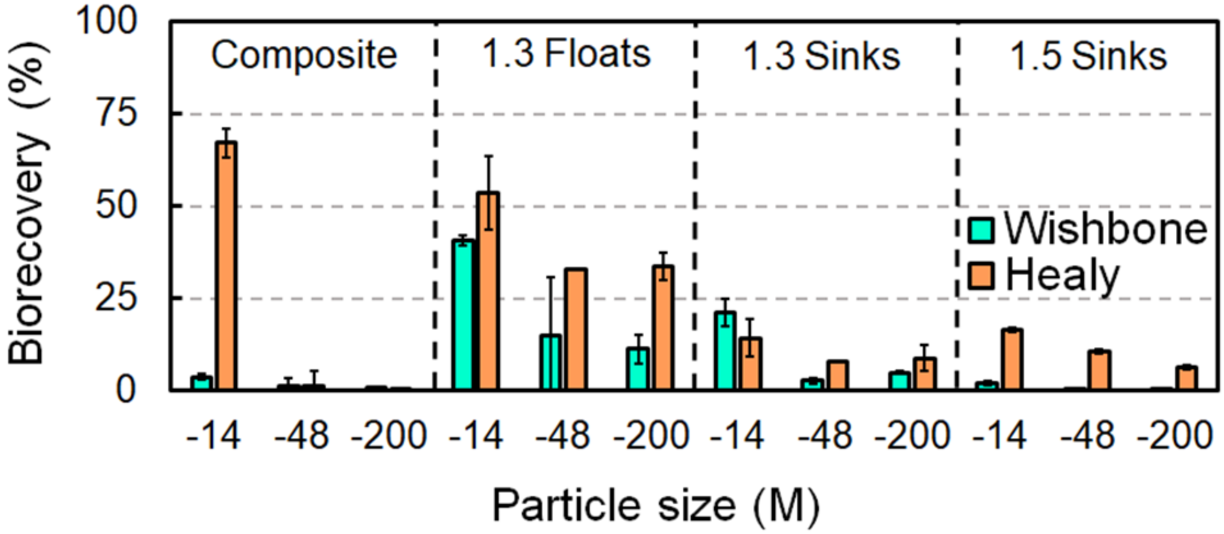

Figure S3. Total REEs recovery from Wishbone and Healy coal at varying particle sizes.