A Periodic Table of Passive Treatment for Mining-Influenced Water[1]

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ABSTRACT

The technical community of regulators and engineers that specializes in passive water treatment should be familiar with the passive treatment "decision tree" that was published by the former US Bureau of Mines about 14 years ago. The decision tree was originally intended to address mining influenced water (MIW) from coal mines. But since then, the breadth of passive treatment has expanded to embrace precious and base metal mines, uranium mines, and even gravel pits. Each MIW has its unique signature, either imposed by the natural geochemical conditions of the ore body and surrounding mine waste, or by resource recovery processes that may include heap leaching or traditional hydrometallurgical technologies. As such, the decision tree is no longer applicable to the range of constituents being treated.

With apologies to Dmitri Ivanovich Mendeleev, a "Periodic Table of Passive Treatment" could become a useful design tool. This revised table would focus on identifying passive treatment methods that have been observed to work on specific elements or species of elements based on the author's experience or other practitioners of the technology, supported by geochemical analysis and modeling. This paper provides an update on a continuing effort to compile a simple, science-based reference that can be used in preliminary design of passive treatment systems for mining influenced water.

Additional Key Words: constructed wetlands, acid rock drainage, bioreactors

INTRODUCTION

The field of passive treatment is constantly growing and finding both new applications and new technologies applicable to mining. Passive treatment may encompass a range of processes or a combination of processes because no single treatment cell type works in every situation or with every solution geochemistry. Attempts have been made in the past to simplify the realm of passive treatment options through methods such as flow charts. A recent version of a decision flow chart may be found in Gusek (2008); some decision flow charts have provided limited information about the range of metals a passive treatment designer may wish to address.

This paper provides an update on a continuing effort to compile a simple, science-based reference that can be used in preliminary design of passive treatment systems for mining influenced water. Modeling the reference after the Periodic Table of the Elements (PTE) is a natural choice. The PTE was first introduced by the Russian chemist Dmitri Ivanovich

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Mendeleev in 1871. He brilliantly organized the elements into similar groups which we now know are governed by how their atomic structures are arranged.

The elements have been organized many ways since Mendeleev's time. Some of these groupings are particularly relevant to mining, such as classification of metals cations as hard or soft type cations (Stumm and Morgan (1996) provides an excellent explanation), where hard cations preferentially complex with fluoride or ligands that have oxygen as the donor atom and soft cations complex preferentially with sulfur, nitrogen, or iodide. Another example is "An Earth Scientist's Periodic Table of the Elements" (Railsback 2004), where elements may appear multiple times based on their redox state or hard and soft properties. However, these alternate groups still take their roots from Mendeleev's original table, which is elegant and simple to use.

MINING INFLUENCED WATER

The concept of "mining influenced water" was first introduced by Schmiermund and Drozd (1997). It covers the breadth of solutions ranging from acid rock drainage (ARD) to neutral mine drainage to the mining process solutions, such as alkaline sodium cyanide solutions used in recovery of gold or silver. The definition of MIW may be driven by regulations, by the processes employed, or by the geology of the site being mined. Sources such as geo-environmental models (e.g., du Bray 1995; Plumlee and Nash 1995; Plumlee 1999; Plumlee et al. 1999; and Seal and Foley 2002) will provide an indication of typical constituents that may be observed at a site based on its geology. Regardless, MIW may include a large range of constituents beyond which passive treatment systems initially targeted.

PASSIVE TREATMENT AND REMOVAL MECHANISMS

To paraphrase Gusek (2002), passive treatment is defined as:

> "*a process of sequentially removing contaminants and/or acidity in a natural-looking, man-made bio-system that capitalizes on ecological, and/or geochemical reactions coupled with physical sequestration. The process does not require power or chemicals after construction, and lasts for decades with minimal human help.*

Passive treatment is a sequential process because no single treatment cell type works in every situation or with every MIW geochemistry. It is an ecological/geochemical process because most of the reactions (with the exception of limestone dissolution) that occur in passive treatment systems are biologically assisted. Lastly, it is a removal process because the system must involve the filtration or immobilization of the metal precipitates that are formed. Otherwise, they would be flushed out of the system, and the degree of water quality improvement would be compromised.

From a passive treatment system designer's perspective, there are several basic components available "off-the-shelf", as shown in Table 1. How the passive treatment systems in Table 1 immobilize constituents is pertinent for application to the periodic table. Elements of a single group tend to behave similarly in chemical reactions; this should apply to biogeochemical behavior in passive treatment systems as well. If a removal mechanism can be established for an element, it may apply to other elements within a PTE group. In general, passive treatment systems operate by pH adjustment and alkalinity generation under either aerobic or anaerobic conditions[2]. Table 1 provides classification of treatment systems by aerobic or anaerobic conditions.

Understanding the exact removal mechanism for a constituent, however, is a daunting task. Several mechanisms may be occurring within a single treatment cell and biologically mediated reactions are not necessarily well understood or adequately quantified for inclusion into standard modeling techniques. Nonetheless, geochemical modeling was performed on effluents from passive treatment cells to provide an indication of removal mechanisms. Modeling was performed using PHREEQC or The Geochemist's Workbench (Parkhurst and Appello 1999 and Bethke 2008, respectively). Effluent water quality was speciated and plotted on Eh-pH diagrams to examine saturation indices (SI) and stability fields for various minerals. The SI value (unit-less) provides an indication as to mineral saturation by comparison of the ion activity product to the equilibrium constant for a mineral. SI values at or near zero indicate that a mineral is in equilibrium with a solution. Negative SI values indicate a solution is under saturated with respect to the mineral (and the mineral will dissolve) and positive SI values indicate super-saturation with respect to a solution. If the effluent water quality is in equilibrium with a mineral (based on the SI value) or plots within the stability field, it is possible this mineral is present within the passive treatment system. Hence, while we are unable to model the exact reactions, this modeling provides confirmation as to what has occurred within the passive treatment cells. Specific removal mechanisms are described further below.

Table 1 – Oxidation Reduction Conditions Prevalent in Conventional Passive Treatment System Components

Passive System Component	**Aerobic (ORP > zero mV) Oxidizing Conditions**	**Anaerobic (ORP < zero mV) Reducing Conditions**
Biochemical bioreactors	X (upper 2-3 cm)	X (most of the cell mass)
Aerobic wetlands	X	
Oxidation & Settling ponds	X	
Anoxic limestone drains		X?
Successive alkalinity producing systems	X (upper 2-3 cm)	X (most of the cell mass)
Open limestone channels and limestone beds	X	

Notes: Biochemical bioreactor are also referred to as sulfate reducing bioreactors. Table 1 above does not include emerging technologies that hold promise but may still be under development. Also, while an anoxic limestone drain may exhibit mildly reducing conditions, it is sole design goal is to add alkalinity, not remove metals. Thus it should not be inferred that ALD's are appropriate for the precipitation of certain metals beyond their ability to remove aluminum and iron. ORP values assume standard hydrogen electrode.

pH Adjustment

For some constituents, simply altering the pH results in mineral precipitation and immobilization. Aluminum is one typical acidic MIW constituent; it is fully expected to precipitate as the pH rises, regardless of redox conditions. However, the exact mineral precipitating may be difficult

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[2] It is worth noting that other removal mechanisms are also likely occurring in passive treatment systems, such as organic complexation, plant uptake, and adsorption. However, these likely play a minor role relative to the carbonate dissolution and microbial facilitated redox reactions.

to pinpoint. Figures 1A & 1B, developed using the effluent chemistry from a passive treatment system, indicate that aluminum hydroxysulfates found in standard geochemical thermodynamic databases may be precipitating.

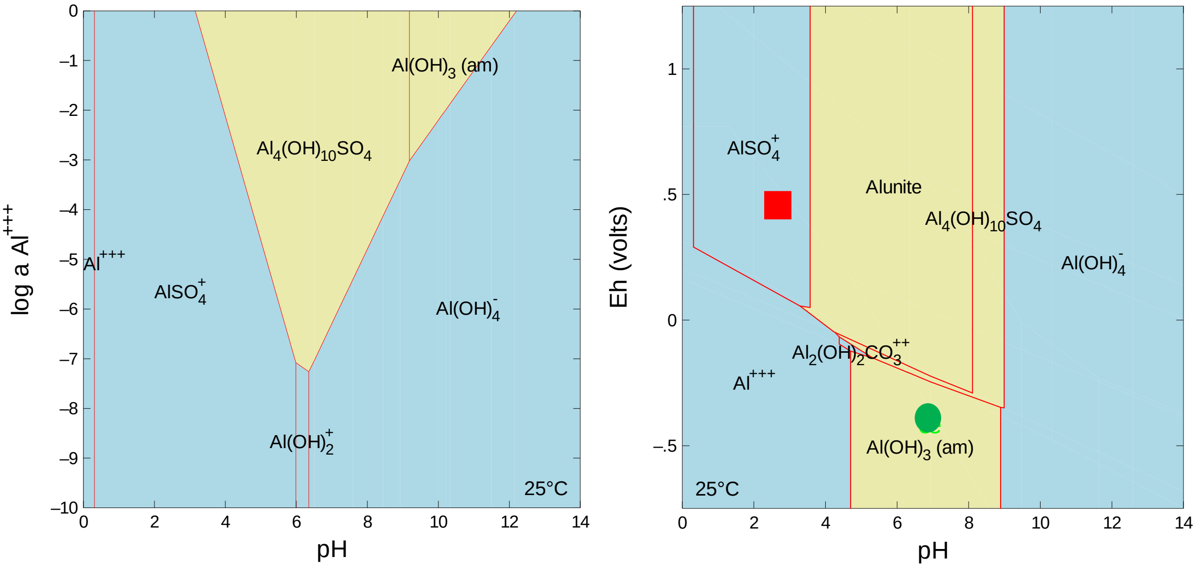


Figure 1. A) Example of a potential stability diagram for aluminum generated using specific water chemistry from a passive treatment system. B) Example of an Eh-pH diagram for aluminum species using specific water chemistry from a passive treatment system. The red square represents influent water to the system; the green circle represents effluent water. Both diagrams were created in GWB using the Minteq database with suppression of non-credible geochemical minerals.

Aerobic Systems

In aerobic systems, dissolution of carbonates plays a significant role, adding alkalinity and raising the pH. However, a number of metals are sensitive to both pH and redox conditions. Oxidation of metals, in particular iron, results in precipitation of metal hyrdroxides. Iron hydroxide precipitation can be significant, and co-precipitation or sorption associated with the iron hydroxides may remove additional metals from solution.

Anaerobic Systems

In anaerobic systems, metals and sulfate reduction to form metal sulfides is significant. For example, geochemical modeling of a sulfate reducing bioreactor (SRBR) effluent indicates that the effluent solution is near equilibrium with zinc sulfide (sphalerite), as shown in Table 2, but undersaturated with respect to with smithsonite (zinc carbonate) or zinc hydroxides or zinc oxides. Values shown in Table 2 provide a guide or indication as to the actual conditions. The geochemical modeling relies on thermodynamic databases, which do not have complete data for every mineral possible. The zinc sulfide formed in the SRBR may not be the crystalline or ore mineral of sphalerite, but lacking data for other zinc sulfides (e.g., an amorphous zinc sulfide precipitate), the sphalerite thermodynamic data provides an indication as to what is occurring.

This removal mechanism also applies to other constituents in the same PTE group as zinc, such as cadmium and mercury. Modeling data for cadmium also indicated near equilibrium with greenockite (cadmium sulfide) and undersaturation with respect to otavite (cadmium carbonate), as shown in Table 2. Data were not available for mercury for this effluent, but formation of a solid such as metacinnabar (mercury sulfide) would be a possibility.

Table 2. Examples of SI Index values generated by geochemical modeling of SRBR effluent.

Mineral	**Formula**	**SI**	**Mineral**	**Formula**	**SI**
Sphalerite	ZnS	1.31	Malachite	Cu2(OH)2CO3	-42
Smithsonite	ZnCO3	-9.73	Galena	PbS	1.35
Zinc hydroxide	Zn(OH)2	-12.70	Cerrusite	PbCO3	-9.1
Greenockite	CdS	0.84	Nickel sulfide	NiS (alpha)	1.98
Otavite	CdCO3	-11.12	Nickel carbonate	NiCO3	-6.33
FeS(ppt)	FeS(ppt)	-0.62	Rhodocrhosite	MnCO3	-0.22
Ferrihydrite	Fe(OH)3	-10.32	Manganese sulfide	MnS (grn)	-1.49
Siderite	FeCO3	-2.6	Cr(OH)3 *(amorphous)*	Cr(OH)3	0.54
Cobalt sulfide	CoS (alpha)	3.60	Amorphous gibbsite	Al(OH)3	-0.85
Cobalt carbonate	CoCO3	-3.4	Calcite	CaCO3	0.07
Covellite	CuS	-1.54	--	--	--

Notes:
-SI values = Saturation Index, calculated using PHREEQC and the Minteq database
-As described in the text, modeling not intended to show that the exact minerals above (such as ore minerals) are necessarily present.
-SI values of 0 indicates equilibrium; however, values are approximate given the limitations of the thermodyanic database and modeling and other sources of error (e.g., analysis error)

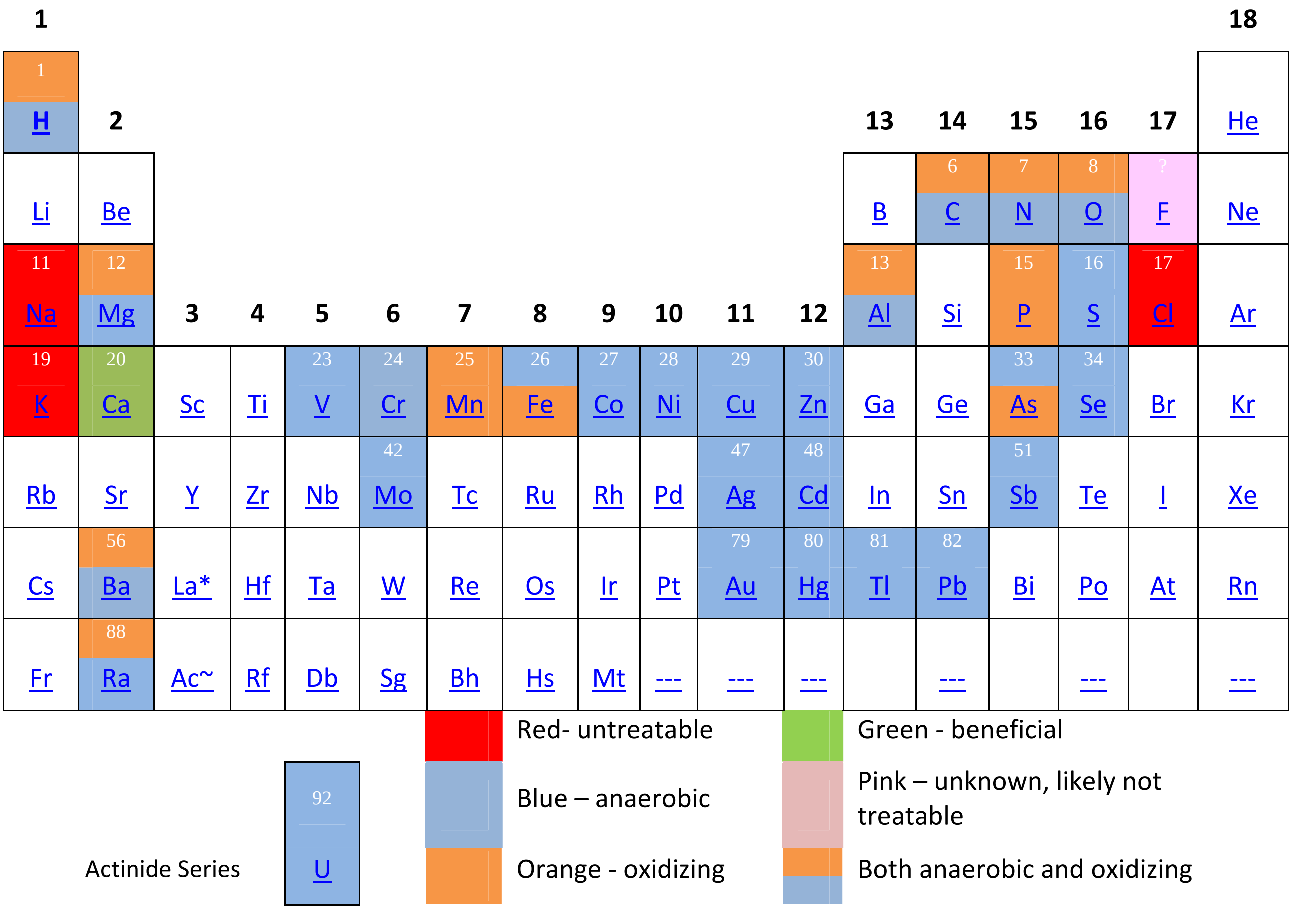
Precipitation of metal sulfides in passive treatment systems applies to many of the transition metals on the PTE, not just the zinc-cadmium-mercury group. Practical experience has show that metals such as nickel, lead, iron, copper, and cobalt are also removed by passive treatment systems. In support of this, the geochemical modeling of SRBR effluent indicated near equilibrium conditions with minerals such as nickel sulfide, galena, covellite, and cobalt sulfide.

The exact precipitation mechanism of these metals sulfides is not entirely certain—whether they are actively precipitated by microbes or are the result of local high sulfide, low Eh environments generated by the microbes. In either case, the role of microbes is critical in explaining the appearance of some of these sulfides. The microbiology of passive treatment has become better understood in the past five years and perhaps a “periodic table of microbial activity” might be a logical extension of this paper. Such a paper would link the microbial communities most responsible for the removal of parameters of interest in passive treatment system components.

Effective removal of metals by sulfide precipitation does have exceptions. A notable example of this is manganese. Manganese sulfides are not stable at pH values obtained by the passive treatment systems which has made manganese removal difficult (Walton-Day 2003). Manganese removal, when successful, may be due to rhodochrosite (manganese carbonate) precipitation, based on limited speciation modeling, or as oxides in aerobic systems (Clayton et al. 1999). In addition, a chromium hydroxide, rather than a chromium sulfide, may be forming based on the geochemical model SI values (Table 2).

PERIODIC TABLE OF PASSIVE TREATMENT

Figure 2 provides the current periodic table of passive treatment. The general ORP categories introduced in Table 1 were applied to the PTE based on the authors' experience, limited geochemical modeling, and the available literature and conventional wisdom. The periodic table of passive treatment will continue to grow. Geochemical modeling of all MIW is beyond the scope of this paper and incomplete data would make this task misleading as well. In addition, the technology and its application are also broadening. Specific references for each element on the periodic table of passive treatment are provided in Table 3; the references are provided to provide guidance for cursory additional research and are not intended to be all-inclusive.



1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 H 🟧🟦																	He
Li	Be											B	6 C 🟧🟦	7 N 🟧🟦	8 O 🟧🟦	? F 🩷	Ne
11 Na 🟥	12 Mg 🟧🟦											13 Al 🟧🟦	Si	15 P 🟧	16 S 🟦	17 Cl 🟥	Ar
19 K 🟥	20 Ca 🟩	Sc	Ti	23 V 🟦	24 Cr 🟦	25 Mn 🟧	26 Fe 🟦🟧	27 Co 🟦	28 Ni 🟦	29 Cu 🟦	30 Zn 🟦	Ga	Ge	33 As 🟦🟧	34 Se 🟦	Br	Kr
Rb	Sr	Y	Zr	Nb	42 Mo 🟦	Tc	Ru	Rh	Pd	47 Ag 🟦	48 Cd 🟦	In	Sn	51 Sb 🟦	Te	I	Xe
Cs	56 Ba 🟧🟦	La*	Hf	Ta	W	Re	Os	Ir	Pt	79 Au 🟦	80 Hg 🟦	81 Tl 🟦	82 Pb 🟦	Bi	Po	At	Rn
Fr	88 Ra 🟧🟦	Ac~	Rf	Db	Sg	Bh	Hs	Mt	---	---	---		---		---		---

Actinide Series: 92 U 🟦

🟥 Red- untreatable

🟦 Blue – anaerobic

🟧 Orange - oxidizing

🟩 Green - beneficial

🩷 Pink – unknown, likely not treatable

🟧🟦 Both anaerobic and oxidizing

Figure 2 – Proposed Periodic Table for Passive Treatment of MIW

On Figure 2, the red-shaded elements (Na, K, Cl), which may be associated with elevated TDS, are not affected by the off-the-shelf passive treatment processes. Calcium, which is also conserved or involved in the generation of hardness, is a beneficial ion and it is therefore color coded in green. There are specialized situations where elevated fluorine (as fluoride) has been a component of MIW. Being a halide immediately above chlorine, passive fluoride remove is not straightforward. In acidic MIW, fluoride solubility is known to be sensitive to pH, but a solid precipitate can be formed only in a very restricted pH range. This condition may be difficult to maintain in a passive treatment system; fluorine is thus color coded a shade of pink.

The proposed Periodic Table of Passive Treatment (PT2) offers another view of the sometimes complicated picture of conflicting priorities in treating MIW passively. As suggested earlier, the proposed PT2 is a starting point to a more complete understanding of the complicated bio-geochemistry behind the passive treatment design process. It should be considered a logical expansion of the former USBM passive treatment decision tree and like Mendeleev's original work over 130 years ago, should be the focus of future enhancement.

Table 3 – Passive Treatment of Typical Elements and Species in MIW

Group	Element or Species	Suspected or Documented Mechanisms & Passive System Components and Reference
1	Hydrogen ion	H^+ can be addressed aerobically and anaerobically: limestone dissolution in ALDs, OLCs, BCRs and SAPS and microbial bicarbonate alkalinity in BCRs or SAPS; Conventional Wisdom (multiple refs)
2	Magnesium	Mg has been observed being removed by the replacement of calcium in limestone to form suspected dolomitic limestone in a pilot BCR in Slovakia; this may occur aerobically too
2	Barium	Dissolved barium can be precipitated aerobically or anaerobically as the insoluble barium sulfate by comingling with slightly elevated sulfate bearing MIW; Gusek, et al. 2000
2	Radium	Ra 226 was observed being removed in a BCR pilot in 1993, probably as RaSO4 with barite; Unpublished BCR data & Wanty et al. 1999
5	Vanadium & Uranium	V and U occur naturally in uranium roll front deposits which form in reducing conditions prevalent in BCRs; Whitmer and Saunders 2000
6	Chromium	Reduction to Cr^{+3} with hydrolysis/ precipitation of chromium hydroxide in BCRs; Ozawa et al. 1995
6	Molybdenum	Molybdenum removal in a pilot BCR was observed in 1994; Unpublished data
7	Manganese	Precipitation of MnO_2 facilitated by algae; presence of limestone is recommended but not required; $MnCO_3$ (rhodochrosite) formation suspected in over-loaded BCRs; Conventional Wisdom, Robbins & Ziemkiewicz 1999, & other refs
8	Iron	Fe precipitation as ferric oxy-hydroxide in aerobic wetlands, OLCs, oxidation ponds, and the surface zone of BCRs; iron sulfide (FeS) precipitation in BCRs and reducing zone of SAPS; Conventional Wisdom (multiple refs)
9	Cobalt	Cobalt sulfide formation in BCRs; Eger 1992
10	Nickel	Nickel sulfide formation in BCRs; Hammack and Edenborn 1991
11	Copper	Copper sulfide formation in BCRs; Wildeman et al., 1990
11	Silver	Silver sulfide formation in BCRs
11	Gold	Native gold precip. in BCRs is possible but undocumented
12	Zinc	Precipitation of sphalerite (ZnS), Wildeman, et al. 1990; Barnes et. al.
12	Cadmium	Cd removal in a pilot BCR suspected to be as greenockite (CdS) observed in 1994; Unpublished data
12	Mercury	Meta-Cinnabar (HgS) in BCRs – some uncertainty of Hg methylation in BCRs; Unpublished data
13	Aluminum	Al hydroxide (gibbsite) precipitates in well-buffered MIW in aerobic wetlands, OLCs, SAPS, ALDs; aluminum hydroxy-sulfate precipitation in BCRs; Conventional Wisdom & (Thomas 2002)
13	Thallium	Tl sulfide co-precipitation with FeS in BCRs; (Blumenstein, et al. 2008)
14	Cyanide	CN degradation anaerobically in BCRs; Cellan, et al. 1997
14	Cyanide	CN degradation aerobically by UV light in aerobic wetland; Wildeman, et al. 1994
14	Biochem. Ox. Demand (BOD)	By-product of BCRs – polished with aerobic wetlands; Conventional wisdom (multiple refs)

14	Lead	PbS (galena) precipitation in BCRs; Wildeman, et al. 1993
15	Ammonia	NH_3 is oxidized to nitrate in aerobic wetlands and is also utilized by plants; EPA 1988
15	Nitrate/ Nitrite	NO_3 and NO_2 are denitrified in BCRs to N_2; EPA 1988
15	Phosphate	Plant uptake in aerobic wetlands, sorption to ferric oxy-hydroxides; EPA 1988
15	Arsenic	Removal in aerobic conditions by sorption to or co-precipitation with iron oxy-hydroxide and possibly by scorodite formation. Removal in anaerobic (BCR) conditions as sulfide; Wildeman et al. 1994, EPA 2007
15	Antimony	Stibnite (Sb_2S_3) formation in hot springs environments may be similar to conditions in a BCR – removal data lacking
16	Oxygen	Depressed dissolved oxygen from BCRs is polished with aerobic wetlands; oxygen is required in aerobic wetlands and other situations to precipitate iron. Conventional wisdom (multiple refs)
16	Sulfate	Sulfate is removed by microbial conversion to sulfide in a BCR; Conventional wisdom (multiple refs)
16	Sulfide	Sulfide is scavenged by sacrificial metals such as zero valent iron; Conventional wisdom (multiple refs)
16	Selenium	Selenium is removed by microbial conversion to elemental selenium or iron selenide precipitates in a BCR; Conventional wisdom (multiple refs)

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