

THE ROLE OF NATURAL ATTENUATION FOR ARSENIC IN HEAP LEACH DRAINAGE

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Introduction

Natural Attenuation refers to natural processes that result in removal and/or isolation of chemical phases from mining or industrial drainage emanating. Frequently such processes are complementary to active remediation mechanisms such as source removal or control.

Within Nevada, arsenic is a ubiquitous trace element of hydrothermal mineralization (Theodore et al., 2003) and is typically the main element of concern in the management of heap leach facilities on closure (Bowell and Parshley, 2001). As there is excess draindown water on the heaps it is a common practice to use this water to recharge groundwater resources. Once cyanide is treated out of the system arsenic is the only issue prior to discharge to the environment. Fortunately hydrous ferric oxides (HFO) are common in the Great Basin alluvium and have a high affinity for arsenic and other oxyanions under the prevailing mildly oxidizing and neutral to mildly alkaline pH conditions. Other minerals present in the alluvium, such as aluminum and manganese hydroxides and clay phases can act as sorbent, although in general they show a lower efficiency for arsenic adsorption than HFO phases (Asta et al., 2009; Rait et al., 2010). In circum-neutral to mildly alkaline, oxic environments, arsenic typically forms species such as $H_nAsO_4^{-(3-n)}$. Such ions tend to form soluble products and can be adsorbed onto HFO in the pH range 4-8. Above this value, arsenic phase's solubility increases and their dissolved concentrations can rise from continued release and desorption (Bowell and Craw, 2014). Consequently pH plays a critical role in the control of natural arsenic attenuation.

In order to assess the potential for attenuation of arsenic and other constituents the infiltrated process fluids in an evapotranspiration field has been assessed at the Daisy Mine, Nevada. The numerical predictions provide a basis for evaluation of options for closure of the Daisy Heap Leach including direct discharge of the Daisy Heap draindown solution to the sub-surface. The objective of this study

is to predict geochemistry of solutes in groundwater upon interaction with draindown from the Daisy Heap to determine the potential of chemical constituents in heap solutions particularly arsenic and salts to attenuate in the unsaturated alluvium and not degrade groundwater beyond an NDEP reference value.

Methodology

The attenuation of leachate from heap draindown through 200 m of alluvium to groundwater has been modelled using advection in the geochemical code PHREEQC (Parkhurst and Appelo, 1999). Data that were used as inputs to the model were derived from the following sources; Monitoring data available from operational heap draindown chemistry; groundwater chemistry specific to the infiltration area; estimates of draindown volumes (Hydro-Engineering, 2012); and aquifer properties (i.e., depth, transmissivity, quality, etc.).

The information collected above, along with other published data (i.e. thermodynamic database, precipitation reactions) was input into the USGS-developed software, PH-REdox-EQuilibrium-Chemistry (PHREEQC, Parkhurst and Appelo, 1999). This software uses thermodynamic equilibrium chemistry and solubility calculations to determine the residual solution chemistry following interaction of a number of solutions. The software allows for mineral precipitation and sorption reactions with mineral surfaces. Dissolution and oxidation can also be factored into the model to account for reaction with solid mineral phases which can be declared in the model in finite quantities. The resulting model output predicts not only the concentration of modelled constituents but also the speciation of the aqueous solutes and the potential solubility of minerals of constituent components as well. This allows prediction of the resulting chemistry of the reactions. These results are then compared to NDEP reference values to determine if a potential impact will result from the reactions.

The geochemical assessment was based on a conceptual model, an important precursor to the numeric calculations, is presented in Figure 1 and was developed from a review of site-specific information and a number of assumptions; these are that; ; that, the drainage water from the Daisy Heap is currently conveyed to the ET cell but will be delivered to an infiltration line system in the future; The 2011 drainage rate used in recent modelling reports for the Daisy Heap was 0.246 m³/day (Hydro-Engineering, 2012). The discharge rate is continuing to diminish and there are extended periods with no flow; Draindown chemistry is represented by average results from monitoring that occurred between 2004 and 2011 and will remain similar over time; The infiltration system will distribute the drainage water in a drain approximately 17 m long and will be oriented to place the base of the drain as close as possible to a level grade; The drain will be constructed below the evaporative zone depth; the heap drainage will migrate through the partially saturated soil profile and will gradually increase the moisture content of the soil profile; the regional groundwater aquifer is greater than 200 m below ground

surface (bgs) and represented by production well PW-1, also referred to as MW-4; the geochemical model assumes that all draindown from the Daisy Heap will migrate to groundwater, and that the draindown solution will interact with groundwater. Based on previous calculations at the current rate of draindown it will take greater than 100 years for heap draindown to reach groundwater (Hydro-Engineering, 2012); the flow path to groundwater consists entirely of alluvial material that will provide attenuation capacity. The simulation period was set at greater than 100 years to evaluate long-term depth of infiltration into the soil profile. Attenuation was represented by determining the proportion of ferric hydroxide (ferrihydrite) in the alluvial along with adsorption coefficients derived from published data (Dzombak and Morel, 1990); and that the groundwater flow rate of 25 m³/day is reasonable to define the ratio of heap draindown to groundwater for modelling (Hydro-Engineering, 2011).

The attenuation of leachate from draindown of the heap leach pad through 200 m of alluvium has been modelled using advection in the geochemical code PHREEQC (Parkhurst and Appelo, 1999). Processes that are accounted for in the models are chemical precipitation of solutes and sorption to iron hydroxides. Although sorption does not represent an indefinite sink for contaminants, the models examine whether the unsaturated alluvium has sufficient sorption capacity to attenuate contaminants until the facility is fully drained

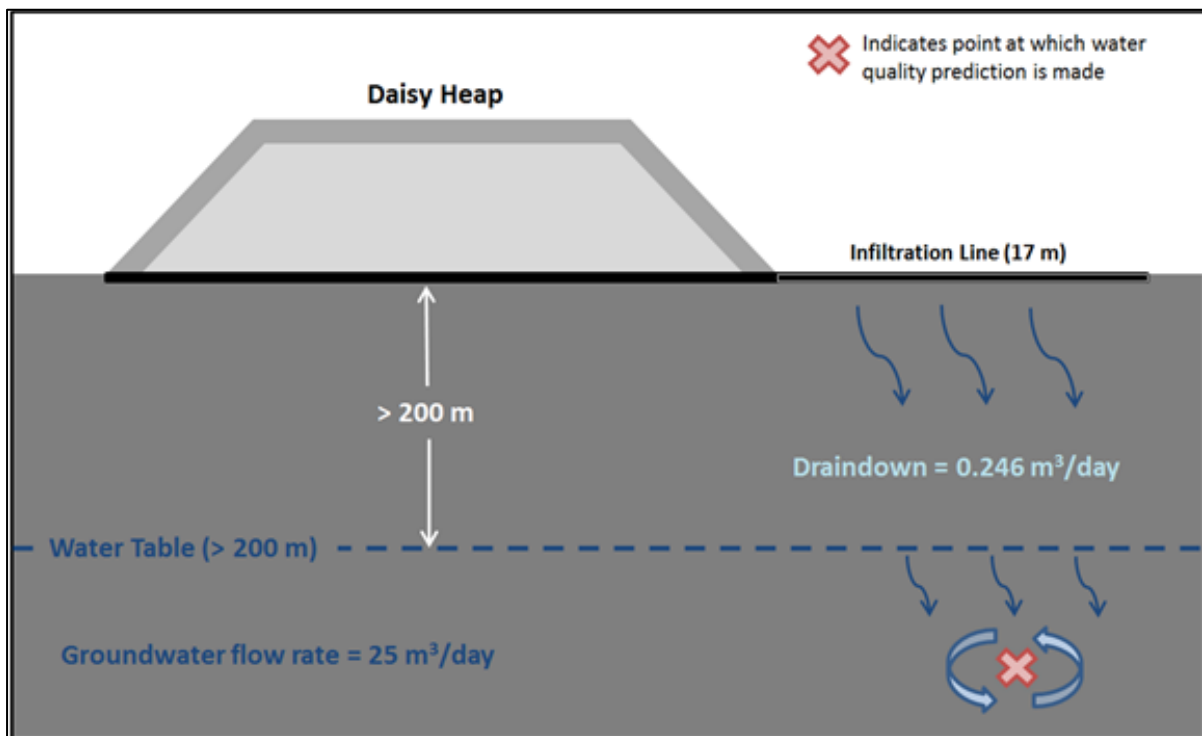


Figure 1. Conceptual model of Daisy Heap solution infiltration

The Daisy Heap covers approximately 17,000 m². The final cover was placed on the heap in 2002 and the draindown rate from the Daisy Heap has declined since then from approximately 76 m³/day to 0.246 m³/day by 2011. The discharge rate continues to diminish and there are extended periods with no flow. This draindown rate is expected to continue to decrease with time. For the sensitivity analysis additional model runs were performed with incremental increases in the draindown rates to determine the rate at which constituents exceed the NDEP reference values.

Draindown chemistry for the Daisy Heap was monitored on a quarterly basis and water quality data is available from 2004 through 1st Quarter 2011. Because there was not a significant change in chemistry over time observed during this period, the average chemistry was used to represent heap draindown chemistry in the models. The average draindown chemistry is summarized in Table 1.

Table 1. Average draindown chemistry, all parameters in mg/L except pH

Parameter	NDEP Value	Min	Max	Average	Standard Deviation
Alkalinity (Total)	--	95	160	130	17
Bicarbonate	--	120	200	150	22
Carbonate	--	<1.0	<1.0	<1.0	0
Aluminum	0.2	0.03	0.22	0.067	0.048
Antimony	0.006	0.0057	0.011	0.0072	0.0012
Arsenic	0.01	0.27	0.613	0.52	0.062
Barium	2.0	0.016	0.05	0.02	0.0067
Beryllium	0.004	<0.001	<0.002	<0.001	0.0004
Bismuth	--	<0.06	<0.5	<0.01	0.12
Boron	--	0.19	0.5	0.31	0.056
Cadmium	0.005	<0.001	<0.005	<0.001	0.0011
Calcium	--	360	450	400	26
Chloride	400	82	120	96	11
Chromium	--	0.005	0.025	0.013	0.0056
Cobalt	--	0.2	0.31	0.23	0.03
Copper	1.3	<0.01	<0.25	<0.05	0.043
Fluoride	4	0.29	8.7	1.8	1.6
Gallium	--	<0.02	<0.5	<0.1	0.12
Iron	0.6	0.067	1	0.2	0.24
Lead	0.015	<0.0025	<0.01	<0.01	0.0029
Lithium	--	0.095	0.5	0.13	0.12
Magnesium	150	93	140	110	11
Manganese	0.1	0.005	0.23	0.085	0.069
Mercury	0.002	0.0023	0.02	0.0089	0.0046
Molybdenum	--	0.01	0.05	0.033	0.013
Nickel	0.1	0.01	0.05	0.012	0.0078
Nitrate + Nitrite	10.0	48	230	160	30
pH	6.5- .5	6.89	8.01	7.7	0.3

Parameter	NDEP Value	Min	Max	Average	Standard Deviation
Phosphorus	--	<0.05	<2.5	<0.6	0.6
Potassium	--	9.9	27	15	3
Scandium	--	<0.002	<0.5	<0.1	0.12
Selenium	0.05	0.047	0.074	0.056	0.0067
Silver	0.1	<0.005	<0.025	<0.006	0.0039
Sodium	--	720	940	800	68
Strontium	--	1.6	2.2	1.9	0.17
Sulfate	500	2160	3100	2400	190
Thallium	0.002	0.001	0.005	0.0016	0.0011
Tin	--	<0.05	<0.5	<0.1	0.12
Titanium	--	<0.005	<0.5	<0.1	0.12
Total Dissolved Solids	1000	3500	5400	4500	380
Vanadium	--	0.005	0.5	0.091	0.13
WAD Cyanide	0.2	0.023	0.28	0.086	0.063
Zinc	5.0	0.01	0.1	0.028	0.033

All values reported in mg/L except pH which is in standard units (s.u.).

< Denotes less than the specified laboratory method detection limit.

Shaded values exceed the respective comparative value from NDEP Form 0190 for Profile II constituents. The groundwater conditions in the upper portion of the project area have not been well defined due to the very limited use of groundwater in this area. Information on groundwater was taken from a US Geological Survey study on two wells, VH-1 VH-2 in Crater Flat (USGS, 1985; 1987).

Groundwater chemistry data is available for the regional groundwater aquifer (from annual monitoring conducted during 1998 to 2000). The average water chemistry for this well is provided in Table 2. The water quality for this well is similar to the water quality defined by the US Geological Survey (1987). Arsenic is the only constituent elevated above NDEP reference standards.

Table 2. Average Groundwater Chemistry, all parameters in mg/L except pH

Parameter	NDEP Value	Min	Max	Mean	Standard Deviation
Alkalinity (Total)	--	119	123	120	2.1
Bicarbonate	--	119	123	120	2.1
Carbonate	--	<1.0	<1.0	<1.0	0
Aluminum	0.2	<0.02	<0.037	<0.03	0.0089
Antimony	0.006	0.002	0.003	0.0023	0.00058
Arsenic	0.01	0.014	0.017	0.016	0.0015
Barium	2.0	0.024	0.025	0.024	0.00058
Beryllium	0.004	<0.002	<0.002	<0.002	0
Boron	--	0.19	0.253	0.23	0.032
Cadmium	0.005	<0.002	<0.0024	<0.002	0.00023
Calcium	--	10.9	11.6	11	0.35
Chloride	400	10	10.6	10	0.32
Chromium	0.1	<0.005	<0.008	<0.006	0.0015
Copper	1.3	<0.003	<0.004	<0.003	0.00058
Fluoride	4	3.1	3.3	3.2	0.1
Iron	0.6	<0.019	<0.02	<0.02	0.00058
Lead	0.015	<0.001	<0.001	<0.001	0
Magnesium	150	0.22	0.274	0.24	0.027
Manganese	0.1	0.002	0.004	0.0027	0.0012
Mercury	0.002	<0.0002	<0.0002	<0.0002	0
Nickel	0.1	<0.005	<0.023	<0.01	0.0091
Nitrate + Nitrite	10.0	0.03	0.41	0.26	0.2
pH	6.5 - 8.5	8.1	8.2	8.1	0.051
Potassium	--	2.8	3.7	3.4	0.49
Selenium	0.05	<0.001	<0.002	<0.002	0.00058
Silver	0.1	<0.005	<0.006	<0.006	0.00058
Sodium	--	66.2	78.3	73	6.2
Sulfate	500	43.8	45.8	45	1
Thallium	0.002	<0.001	<0.001	<0.001	0
Total Dissolved Solids	1000	237	307	260	39
WAD Cyanide	0.2	<0.01	<0.01	<0.01	0
Zinc	5.0	0.003	0.039	0.016	0.02

All values reported in mg/L except pH which is in standard units (s.u.)

< Denotes less than the specified laboratory method detection limit.

Results

For the purpose of the predictive calculations SRK assumed that the draindown waters would interact evenly and completely with groundwater underlying the facility. Under these circumstances the solutes in these waters will react with each other and may form chemical precipitates if the concentrations and macro-geochemical conditions (Eh, pH, $p\text{CO}_2$, $p\text{O}_2$, and ionic strength) allow saturation to occur. The models require specification of potential equilibrium phases that were allowed to precipitate if they become saturated.

In solution, trace element concentrations are mostly controlled by adsorption onto common mineral phases or are removed from solution through a process of co-precipitation. The models assumed that trace metals may be removed from solution via sorption onto iron oxides contained within the alluvium underlying the facility. Ferrihydrite ($5\text{Fe}_2\text{O}_3 \cdot 9\text{H}_2\text{O}$) was selected as a sorption surface because it is a common sorption substrate in oxygenated natural waters and because the trace element sorption thermodynamic properties of these reactions are well defined by numerous empirical studies.

The model assumes that between 10% and 100% of the total iron content of the alluvium (derived from whole rock analysis) is FeOH_3 (Parkhurst and Appelo, 1999). It assumes a ratio of 0.025 strong sites to 0.975 weak sites on the FeOH_3 surface (Dzombak and Morel, 1990), a total of 2.27 sites per nm^2 and a surface area of $244 \text{ m}^2/\text{g}$ (Liger et al., 1999). This amounts to a total calculated mass of FeOH_3 of $1.99 \text{ kg}/\text{m}^3$. The density of alluvium was assumed to be $1900 \text{ kg}/\text{m}^3$ (Geological Survey, 1960; 1968). A number of model iterations were run assuming that either 10%, 50% and 100% of iron within the alluvium is present as FeOH_3 . Assessment of these different scenarios provides a range for the expected attenuation capacity of the alluvium.

The results of the base case geochemical model (i.e. most probable scenario at $0.246 \text{ m}^3/\text{day}$ draindown) and additional draindown scenarios are provided in Tables 3 to 5. The results indicate that at the 2011 draindown rate (0.045 gpm), the concentration of arsenic is predicted to slightly exceed the NDEP reference value, with a predicted concentration of $0.015 \text{ mg}/\text{L}$ compared to a reference value of $0.01 \text{ mg}/\text{L}$. However, arsenic is elevated in the regional groundwater ($0.016 \text{ mg}/\text{L}$). With higher flow rates greater attenuation is observed. This may appear counter intuitive but in fact reflects the fact that complete consumption of attenuation sites is not achieved in any of the models so with higher flow more attenuation apparently occurs.

Predicted values of all other constituents remain below the NDEP reference values with draindown rate below $0.545 \text{ m}^3/\text{day}$; a flow rate that has not occurred since 2009. At this draindown rate, total nitrate plus nitrite ($\text{NO}_3 + \text{NO}_2$ as N) are predicted to exceed the NDEP reference value, with modelled concentrations above

the reference value of 10 mg/L. With more ferrihydrite in the system the level of arsenic attenuation shows no appreciable increase and this is also considered to reflect the fact that there are excess attenuation sites in the system compared to arsenic species predicted to be available for adsorption onto ferrihydrite. The results indicate there is mass solubility control on solute concentration upon interaction of heap draindown with groundwater with minimal saturation of solids other than calcite, goethite and clays (i.e., all other constituents are present as insignificant concentrations or in mineral phases that are not predicted to be saturated under resulting water chemistry).

Table 3. Selected Predictions for Draindown chemistry (10% FeOH3)

Parameter	Units	Baseline groundwater	NDEP reference value	Draindown rate (m ³ /day)					
				0.246	0.545	1.363	2.725	4.088	5.451
pH	-	8.1	6.5 - 8.5	8.49	8.45	8.36	8.27	8.20	8.16
Alkalinity	mg/L	120	-	102	94.7	78.5	63.9	56.2	51.7
Aluminium	mg/L	0.03	0.2	0.03	0.03	0.03	0.02	0.02	0.02
Antimony	mg/L	0.0023	0.006	0.002	0.002	0.003	0.003	0.003	0.003
Arsenic	mg/L	0.016	0.01	0.015	0.014	0.014	0.013	0.013	0.012
Cadmium	mg/L	0.002	0.005	0.002	0.002	0.002	0.002	0.002	0.002
Chloride	mg/L	10	400	10.9	11.9	14.6	18.8	22.6	25.9
Chromium	mg/L	0.006	0.1	0.006	0.006	0.006	0.005	0.005	0.005
Copper	mg/L	0.003	1	0.003	0.003	0.003	0.003	0.003	0.002
Fluoride	mg/L	3.2	4	3.18	3.17	3.12	3.05	2.98	2.92
Iron	mg/L	0.02	0.6	0.00004	0.00004	0.00004	0.00004	0.00004	0.00004
Lead	mg/L	0.001	0.015	0.001	0.001	0.0009	0.0009	0.0008	0.0008
Manganese	mg/L	0.0027	0.1	0.003	0.003	0.003	0.003	0.003	0.003
Nitrate (total)	mg/L	0.26	10	6.40	12.5	29.1	54.3	77.4	97.4
Mercury	mg/L	0.0002	0.002	0.0003	0.0004	0.0007	0.001	0.001	0.002
Selenium	mg/L	0.002	0.05	0.002	0.002	0.002	0.002	0.002	0.002
Sulfate	mg/L	45	500	68.3	93.8	164	270	368	453
Thallium	mg/L	0.001	0.002	0.001	0.001	0.001	0.001	0.001	0.001
Cyanide (total)	mg/L	0.01	-	0.01	0.01	0.01	0.009	0.009	0.009
Zinc	mg/L	0.016	5	0.016	0.016	0.015	0.014	0.014	0.013
Indicates value is above the NDEP reference value									

Table 4. Selected Predictions for Draindown chemistry (50% FeOH3)

Parameter	Units	Baseline groundwater (MW4)	NDEP reference value	Draindown rate (m ³ /day)					
				0.246	0.545	1.363	2.725	4.088	5.451
pH	-	8.1	6.5 - 8.5	8.50	8.48	8.42	8.34	8.29	8.25
Alkalinity	mg/L	120	-	105	100.1	88.3	75.2	67.0	61.9
Aluminium	mg/L	0.03	0.2	0.03	0.03	0.03	0.03	0.02	0.02
Antimony	mg/L	0.0023	0.006	0.002	0.002	0.003	0.003	0.003	0.003
Arsenic	mg/L	0.016	0.01	0.015	0.014	0.014	0.013	0.012	0.012
Cadmium	mg/L	0.002	0.005	0.002	0.002	0.002	0.002	0.002	0.002
Chloride	mg/L	10	400	10.8	11.6	13.9	17.4	20.6	23.3
Chromium	mg/L	0.006	0.1	0.006	0.006	0.006	0.005	0.005	0.005
Copper	mg/L	0.003	1	0.003	0.003	0.003	0.003	0.003	0.002
Fluoride	mg/L	3.2	4	3.18	3.16	3.11	3.02	2.94	2.87
Iron	mg/L	0.02	0.6	0.00004	0.00004	0.00004	0.00004	0.00004	0.00004
Lead	mg/L	0.001	0.015	0.001	0.001	0.0009	0.0009	0.0008	0.0008
Manganese	mg/L	0.0027	0.1	0.003	0.003	0.003	0.002	0.002	0.002
Nitrate (total)	mg/L	0.26	10	5.61	10.8	25.1	46.7	66.6	83.8
Mercury	mg/L	0.0002	0.002	0.0002	0.0003	0.0004	0.001	0.001	0.001
Selenium	mg/L	0.002	0.05	0.002	0.002	0.002	0.002	0.002	0.002
Sulfate	mg/L	45	500	61.4	79.3	129	203	272	331
Thallium	mg/L	0.001	0.002	0.001	0.001	0.001	0.001	0.001	0.001
Cyanide (total)	mg/L	0.01	-	0.01	0.01	0.01	0.009	0.009	0.008
Zinc	mg/L	0.016	5	0.016	0.016	0.015	0.014	0.014	0.013
Indicates value is above the NDEP reference value									

Table 5. Selected Predictions for Draindown chemistry (100% FeOH₃)

Parameter	Units	Baseline groundwater (MW4)	NDEP reference value	Draindown rate (m ³ /day_					
				0.246	0.545	1.363	2.725	4.088	1.0
pH	-	8.1	6.5 - 8.5	8.51	8.50	8.46	8.42	8.38	8.35
Alkalinity	mg/L	120	-	108	104.8	97.8	88.9	82.2	77.4
Aluminium	mg/L	0.03	0.2	0.03	0.03	0.03	0.03	0.03	0.03
Antimony	mg/L	0.0023	0.006	0.002	0.002	0.002	0.003	0.003	0.003
Arsenic	mg/L	0.016	0.01	0.015	0.014	0.014	0.013	0.012	0.012
Cadmium	mg/L	0.002	0.005	0.002	0.002	0.002	0.002	0.002	0.002
Chloride	mg/L	10	400	10.6	11.3	13.2	16.1	18.7	21.0
Chromium	mg/L	0.006	0.1	0.006	0.006	0.006	0.005	0.005	0.005
Copper	mg/L	0.003	1	0.003	0.003	0.003	0.003	0.003	0.002
Fluoride	mg/L	3.2	4	3.18	3.16	3.09	3.00	2.91	2.83
Iron	mg/L	0.02	0.6	0.00004	0.00004	0.00004	0.00004	0.00004	0.00004
Lead	mg/L	0.001	0.015	0.001	0.001	0.0009	0.0009	0.0008	0.0008
Manganese	mg/L	0.0027	0.1	0.003	0.003	0.003	0.002	0.002	0.002
Nitrate (total)	mg/L	0.26	10	4.90	9.3	21.4	39.8	56.7	71.4
Mercury	mg/L	0.0002	0.002	0.0002	0.0002	0.0003	0.000	0.000	0.000
Selenium	mg/L	0.002	0.05	0.002	0.002	0.002	0.002	0.002	0.002
Sulfate	mg/L	45	500	56.8	69.7	105	159	208	251
Thallium	mg/L	0.001	0.002	0.001	0.001	0.001	0.001	0.001	0.001
Cyanide (total)	mg/L	0.01	-	0.01	0.01	0.01	0.009	0.009	0.008
Zinc	mg/L	0.016	5	0.016	0.016	0.015	0.014	0.014	0.013
Indicates value is above the NDEP reference value									

Discussion & Conclusions

Geochemical predictive calculations have demonstrated that based on published estimates of attenuation that there is unlikely to be any impacts to groundwater chemistry from the infiltration of heap leach process solutions once the flow rate of the heap has declined to less than 0.545 m³/day, approximately double the current flow rate observed. At this rate most constituents, including arsenic show concentrations close to or at groundwater quality. Although arsenic is predicted to be higher than the Nevada reference value for water quality it is due to groundwater or elevated baseline reflecting the natural geochemistry in the area. Salt content of the water, such as sulfate is not elevated significantly above baseline values indicating that salination of groundwater would not be predicted. The elevated nitrate is likely to be an overestimate as the calculations only include inorganic chemical reactions and cannot account for the high utilization of nitrogen that would be expected in alluvium soils.

Natural attenuation is a viable method for the long term management of spent process solutions in arid environments like Nevada once a low flow rate has been attained on the facilities and a “steady state” chemistry and flow observed. Such passive measures of managing water will allow for recharge of groundwater without degrading water quality and could be used to augment irrigation water for many crops such as alfalfa.

At the current negligible draindown rates, this geochemical assessment indicates that the interaction of Daisy Heap draindown with groundwater will not appreciably alter baseline groundwater chemistry in the vicinity of the Daisy Heap.

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