Predicting Metal Leaching from Selected Uranium Mine Mineralized Waste Rock Piles Samples

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Abstract
Predicting metal releases from mineralized waste rock piles presents an important challenge for the protection of the environment. It is important to improve predictions to support government decision on mine approvals. For this purpose, we collected and reviewed leach test procedures done on selected mineralized waste rock piles from Canadian Uranium mines and built a metadatabase. Simple linear regression analyses on the data indicates that total Sulfur, Molybdenum, Arsenic, Nickel and Uranium in rock samples explained 56%, 30%, 28%, 17% and 3% of the variability of the concentrations of these elements in leached water. Hence, other variables are also, if not more, important indicators for contaminant levels in leached water. However, pH and alkalinity in leaching water were not these indicators. Future efforts may be devoted to find reliable indicators by completing this metadatabase with data of all Uranium Mine waste rock piles to increase the amount of data for parameters that could affect metal levels in leached water. In our model development, we would consider: water-to-rock ratios, particle size, mineralogy; duration, leaching test type, chemistry of the water use to leach the rock, and temperature, in addition to the total metal content in rock.

Key Words: radionuclides, neutral drainage, short-term leach test, humidity cell test.

Introduction
Mining activities result in localized disturbance of landscapes and in the mobilization of elements that have been encrusted in the earth for millions of years. The most common sources of contaminant releases to the environment at mine sites are tailings and mineralized waste rock, low-grade ore stockpiles, heap leach piles, dump leach piles, and walls of open pits. Among these source terms, tailings are certainly the most prominent and hazardous source of contaminants to the environment. In the late 1990's, the Mining Association of Canada (1998) proposed guidance to minimize the likelihood of adverse environmental impacts from tailings management facilities. Since then, there is increasing preference to dispose tailings in mined out pits (Weatherhead 1988, Dixon-Hardy and Engels 2007) because such disposal practices apparently decrease the potential impacts of tailings seepage to the surface environment (Parker 2000, Tongway 2000). Despite this apparent trend in tailings disposal, sustainable development principles have been put forward for the continuous improvement of tailings management (Franks *et al.* 2011; Kempton *et al.* 2010).

Mineralized waste rock piles, on the other hand, may not be as hazardous to the environment as tailings are, but because they are more heterogeneous, they require more samples to predict releases to the environment (Broughton *et al.* 1992, Runnells *et al.* 1997, Lapakko 2002, Price 2009). Several rock samples are required for predictions because contamination of drainage water depends on the intensity and frequency of precipitation, the chemistry (pH, ionic strength) of the percolating water, the amount of rock exposed, the rock grain size, degree of aeration of the pile, the rock surface area to mass, the rock type and mineralogy, and the temperature (Lefebvre *et al.* 1993, Lefebvre *et al.* 2001a, Lefebvre *et al.* 2001b, Meast *et al.* 2005, Price 2009, Nordstrom 2011).

Waste rock management plans are based on contaminant releases to the environment, which is required to determine if the storage of waste rock on surface may be potentially problematic to exposed groundwater and surface water (Meast *et al.* 2005). These plans are approved early in the mine development phase where there is limited information to support the predictions made on drainage chemistry. Change to these initial waste work management plans are often costly/impractical to modify later because of a number of factors.

However, from the development, to operation and to decommissioning phases, there are opportunities to collect the data needed to verify the predictions made at the approval stage and implement reasonable contingency measures to the mine waste management approach, if any. Still, this comparison of actual drainage chemistry and contaminant levels in ground and surface water to predictions made at the environmental assessment stage is of importance to improve model predictions and waste rock management plans in future environmental assessment of new mine projects.

Surprisingly, these types of comparisons are scant considering the years of mining experience in North America. For instance, of the 183 mines that operated from 1975 to 2005 in the United States, Kuipers *et al.* (2005) only identified 25 mines where the comparisons of actual drainage chemistry and contaminant levels in ground and surface water to predictions could actually be made. Of these selected 25 mine sites, Kuipers *et al.* (2005) found that 60% of the case study mines had mining-related exceedences in surface water. Similarly, 77% of the mines with exceedences in groundwater predicted low groundwater quality impacts in their environmental impact statements. Overall, 21 of the 25 case study mines (84%) had exceedences of water quality standards in either surface water or groundwater or both. Hence, predictions have not been good so environmental monitoring is important but often too late to ensure the environment is protected. However, it should be noted that the 25 mines selected in their study may not necessarily reflect current mining practices because Kuipers et al. (2005) had to choose mines that had environmental assessment predictions and water quality monitoring conducted over a sufficient period of time to verify these environmental assessment predictions.

A comparison of environmental assessment predictions of metal releases to groundwater and surface water will help improve model predictions. Another approach to improve predictions is to properly monitor mineralized waste rock piles as was done by Lefebvre *et al.* (2001a,b). Unfortunately, field monitoring of mineralized waste rock piles are very complex and hence, very few state-of-the-art monitoring studies like the one of Lefebvre *et al.* (2001b) are available to improve current model predictions. In addition, these field monitoring studies are likely very site specific and cannot be applied readily to improve predictive models. Since, Kuipers *et al.* (2001) indicated that environmental assessment predictions could be improved and that actual monitoring indicates contamination that was not predicted, then regulatory authority often rely on very conservative assumptions for any risk-based decisions. Improved modeling predictions would allow for more realistic decisions based on science.

Another approach is to create metadatabases that include results of short-term and long-term leach tests. Such a database may be useful in improving predictions of drainage chemistry if it includes information that captures the variability in the parameters that can affect drainage chemistry in full scale mineralized waste rock piles.

The objective of this study was to investigate if we could develop a model algorithm that would predict metal concentration in leachate of static or kinetic tests from mineralized waste rock characteristics, which could then be eventually used as an independent tool to predict releases of contaminants from mineralized waste rock piles.

Methodology

Data Collection

We selected seven mineralized waste rock characterization reports on seven piles at different uranium mines in Canada. For each waste rock pile, we collected information on waste rock samples such as rock type, grain size, sulfur levels, metal levels (Ni, As, Mo, Se) and radionuclide levels (U, ^{226}Ra, ^{210}Po) (Table 1). Table 2 presents detailed information on the leach tests done on each of the chosen rock samples such as duration, type of water used to leach the mineralized waste rock, the quantity of water and rock used, as well as concentration of metals and radionuclides in the leachate. All this information was used to build our database.

Statistical analysis

We performed simple regression analysis on our database to investigate variables that were related to the levels of contaminants in leached water. Linear regression analysis was done using sigma plot. To use both leach test and kinetic test data, we only used the first week leaching results for the kinetic tests in all regression analyses because the static tests generally lasted a week (or less).

Results

Contaminant levels in waste rock

Table 1 indicates that more modern mines or proposed projects (Site 4&5) have waste rock segregation programs that separate rock into clean and special waste based on levels of As, Ni and U. Older operations (Sites 1-2-3) did not necessarily have these segregation programs. Special waste rock generally dissolves higher levels of Ni, As or U in leached water than clean waste rock (Table 2), which confirms that modern mines (Sites 4-5) are better able to manage special waste, rather than piling clean and special waste together in one pile.

Variables affecting contaminant levels in leached water

Regression analysis of total constituent levels in rock versus levels in leached water is presented in Figure 1. Total Sulfur in rock predicts 56% of the variability of sulfate leachate. In addition, total metal levels in rock predicted 30 and 28% of Mo and As in leached water, respectively. Total metal concentration was a much less reliable predictor of metal leachate for U and Ni (R^2=0.17 and 0.03, respectively). Total selenium level in rocks was not related to selenium levels in leached water. Therefore, other variables appear to affect the levels of metals in leached water.

We first investigated whether the pH or alkalinity in the leached water could explain the metal levels measured in leached water and did not find any significant relationship. For other variables such as temperature, mineralogy, water to rock ratios, or rock grain size, there was insufficient information in our database to further explore the role of these variables on the levels of metal leached in percolating water.

Table 1. Metal and radionuclide content in different rock type at the selected waste rock pile samples from Uranium mines in Canada

Site	Rock type	Number of entries	Grain size (<1: 1-10: >10cm; %)	As (mg/Kg)	Ni (mg/Kg)	S (mg/Kg)	U (mg/Kg)	Ra-226 (Bq/g)	Po-210 (Bq/g)
Site 1	Basement	17	10-100:0-50:0-80	12-781	43-1180	0.01-0.31	6.5-148	0.14-1.9	0.09-1.8
Site 1	Sandstone	10	40-80:15-40:0-40	2.4-142	0.2-49	0.01-0.02	0.7-23	0.04-0.61	0.05-0.7
Site 2	Bleached lim. Sandstone (50%)[1]	2	NA	13-17	3-5	NA	25.7	0.25	NA
Site 2	Friable sandstone (10%)	2	NA	14-17	4-5	NA	47.1	0.35	NA
Site 2	Chloritic gneiss (25%)	2	NA	10-13	14	NA	4.6	0.09	NA
Site 2	Graphitic gneiss (5%)	2	NA	1-2	43-46	NA	14.9	0.25	NA
Site 2	Hematitic sandstone (5%)	2	NA	17-23	5-7	NA	36	0.30	NA
Site 2	Pegmatite (%5)	2	NA	1	30-36	NA	6.6	0.10	NA
Site 3	NA	22	NA	2-708	8-289	NA	10	NA	NA
Site 4	Clean rock	4	NA	11-74	15-92	NA	5-67	NA	NA
Site 4	Special waste	4	NA	1600-4000	380-1300	NA	120-1100	NA	NA
Site 5	Clean rock	4	NA	14-160	11-87	NA	4-19	NA	NA
Site 5	Special waste rock	4	NA	420-1700	270-1400	NA	208-535	NA	NA

[1]Indicates the percentage of the rock type in the composite samples presented in table 2 for site 2.

NA: data not available

Table 2. Metal concentrations in 1 week duration static leach test and the leached concentration in the first week of kinetic leach tests

Site	Rock Type	Number of entries	Water to rock ratio	pH	Alkalinity (mg $CaCO_3$/L)	As (mg/L)	Ni (mg/L)	U (mg/L)	Ra226 (Bq/L)
Site 1	Basement	17	3	4.13-7.01	2-31	0.1-6790	0.01-45.40	0.6-117	NA
Site 1	Sandstone	10	3	3.8-6.1	10-17	0.8-140	0.01-6.5	1-593	NA
Site 2	composite samples	18	0.94-5.93	4.88-6.89	0.6-1.5	0.2	0.02-0.597	0.065-0.345	0.65-3.5
Site 2	Composite samples	18	0.81-9.33	4.54-7.34	0.6-5.0	0.0034-0.0057	0.02-0.157	0.0042-0.068	0.04-2.5
Site 2	Composite samples	18	0.91-1.72	2.63-2.99	0	0.0033-0.0118	1.07-14.7	1.05-29.6	0.4-1.2
Site 3	NA	22	0.27-0.45	5.67-9.54	NA	2-708	8-289	10	NA
Site 4	Clean rock	4	0.71-1	4.21-7.05	0-18	0.004-0.960	0.002-0.510	0.002-4.4	NA
Site 4	Special waste	4	0.57-1	3.78-6.45	0-18	2.9-130	0.9-200	0.003-9.1	NA
Site 5	Clean rock	4	1-1.33	4.00-7.01	0.5-21	0.1-13	0.002-1.3	0.0006-0.066	NA
Site 5	Special waste	4	0.58-2.76	3.34-7.45	0-26	0.9-70	0.03-120	0.003-1.5	NA

NA: data not available

Discussion

Linear regression for each metal in static leach tests and the first week kinetic tests indicated that total metal in rock can predict, to some extent, the amount of As and Mo leached by water (Figure 1). The relatively high partial coefficient for As is not surprising. In another study, it was found that 88% of the total leachable As concentration was explained by the total As content in special waste rock of Midwest, McClean, Cigar Lake, Sue E and Caribou projects (Cogema and CLRC 2001, Ecometrix 2008). The lower partial correlation coefficient obtained in our study is likely the result of including older waste rock piles which did not necessarily segregate waste rock into special and clean and hence, piled clean and special waste rock together in one pile. Including these samples of mixed clean and special waste rock may have lead to higher variability in As levels in leachate.

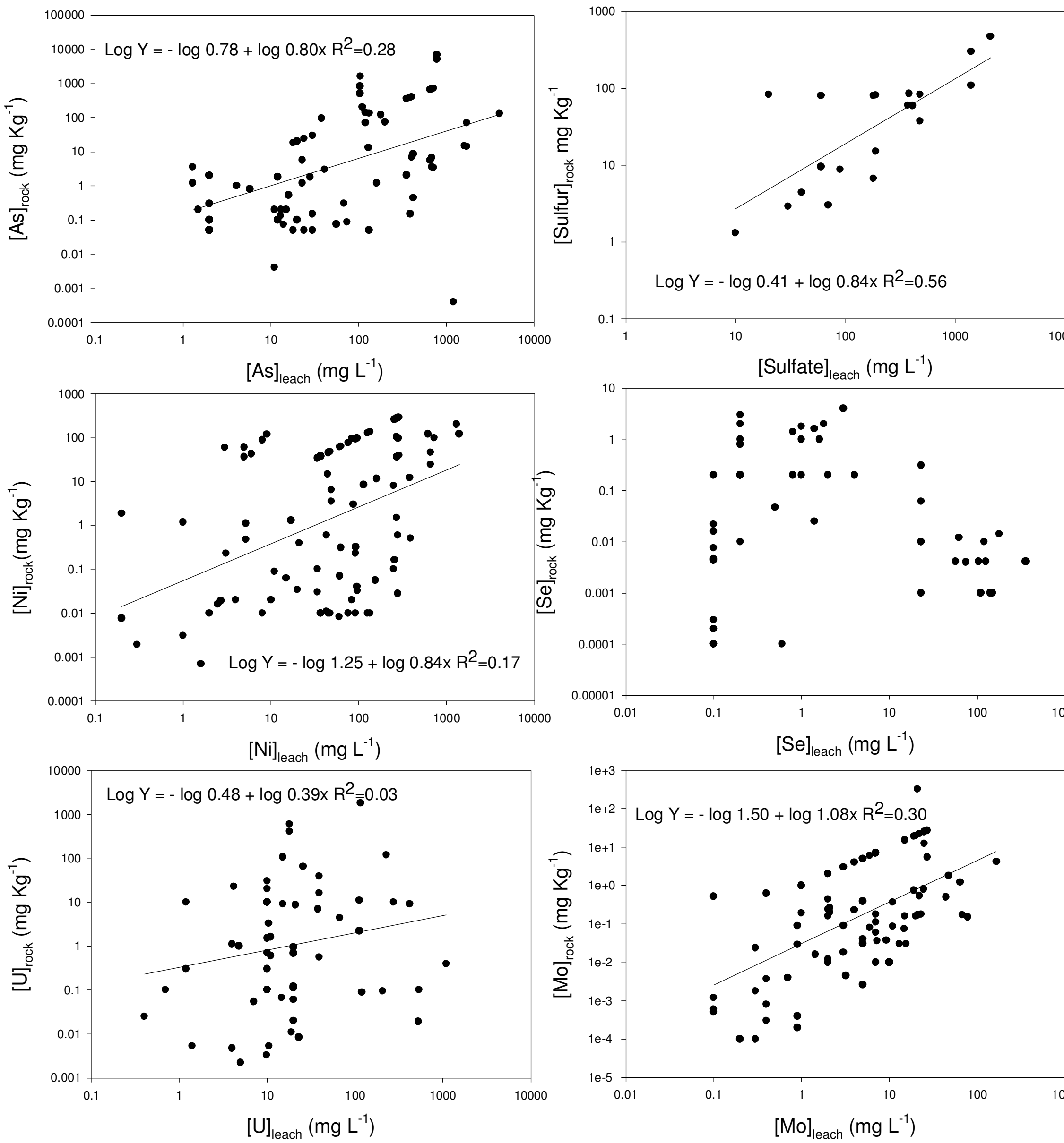

Figure 1. Relationships between total concentration of metals in rock samples with metal concentrations in leached water.

The partial coefficient was generally low for Ni, U, and Se indicating that total metal levels in rock will have modest potential to predict metal levels in leached water. Therefore, it appears that other variables need to be considered in order to improve our power of predicting metal levels in leached water. We have attempted to link metal levels in leached water and alkalinity of the leach water as Uranium, for instance, is well known to bind effectively to carbonates and form carbonate minerals (Goulet *et al.* 2011) but the regression analysis was not statistically significant. We then investigated if the effect of pH in the leach water could improve our predictions of metals in leached water and the results indicated that pH did not affect the concentrations of metals in leached water.

At this point in time, it was not possible to get enough information from the seven reports to investigate if other variables such as the water/rock ratios, the effect of particle size, mineral solubility at the surface of rock samples; duration of exposure; static vs. kinetic leaching, and the effect of temperature in the exposure water could improve the prediction of metals in leached water. The next step would then to collect all data from reports of all waste rock piles regulated by the CNSC to determine if there would be enough information to conduct a credible statistical analysis on the role of these variables in predicting metal levels in leached water.

However, it should be noted that the main uncertainty of the approach discussed in this paper is that it is entirely possible that laboratory derived leachate data may not give accurate representation of expected water quality in the field. Thus an ability to predict result of leach tests accurately may not translate to an ability to predict field seepage quality. Despite this argument, we believe that a credible statistical analysis on a metadatabase as proposed in this paper could perhaps reduce the uncertainty in extrapolating laboratory leachate data to levels of contaminants in leached water in the field. For instance, a complete database would provide a range in variability in the different parameters that affect metal leaching in the laboratory. Once a waste rock pile has been constructed, it could be possible that the database based on laboratory data would capture the measured parameters in this new created mineralized waste rock pile and predict drainage issues before seepage is actually collected. The creation of such a complete database could even decrease the necessary requirements of collecting several rock samples since the database would likely capture the type of geochemical, hydrogeological and physical parameters of a newly created waste rock pile. Finally, such a database could also be employed to improve waste rock segregation programs at mine sites.

Conclusion

This initial study has demonstrated that total metal content in rock can predict, with a relatively low level of accuracy, the levels of contaminants in leached water. However, this was achieved by grouping data from different mine sites and older and recent waste rock piles, which indicates that although some claims that release from waste rock piles is site specific, there are some possible advantages to collecting data from several sites in order to better predict metal releases from mineralized waste rock piles. As a result, we may attempt to complete this database by including reports on all mineralized waste rock pile at Uranium mines in Canada in order to investigate if other variables such as the water:rock ratio, the effect of particle size, mineral availability; duration of exposure; static vs. kinetic leaching, and the effect of temperature in the exposure water could improve the prediction of metals in leached water.

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