# NICKEL AND URANIUM LOADING ESTIMATES FROM A 20-YEAR OLD WASTE ROCK PILE: A COMPARISON OF FIELD ESTIMATES AND LABORATORY LEACHING TESTS<sup>1</sup>

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Abstract. Nickel and uranium loadings from oxidized sulphide-bearing waste rock have been estimated at three successive scales: laboratory columns, a 320  $m^{3}$ -constructed pile (CPE) and the Claude waste rock pile. The Claude waste rock pile is approximately 20 years old and is composed of waste rock from the Claude open pit, which was mined for uranium on the Cluff Lake site, Northern Saskatchewan, Canada. The Claude waste rock pile is about 29 m high at its highest point and covers an area of 29.5 ha. It contains roughly 7.9 million tonnes of waste rock. Operational groundwater monitoring has been in place in the Claude pile area since 1991 with monthly hydraulic head and groundwater chemistry measurements. An assessment of the existing contaminant plume suggests that between 12% and 30% of the nickel and uranium inventory has been released to the groundwater system since initial construction of the pile. Based on sequential leach tests and unsaturated column leach tests results, these leached masses range from 25% and 45% of the total leachable mass for uranium and from 31% to 58% of the total leachable mass for nickel. This metal load was estimated be released during the first pore volume replacement of the waste rock pile. Field loading estimates for the CPE also suggested that a significant portion of the uranium and nickel mass was released during the first pore volume replacement of the waste rock mass, with calculated leached fractions on the order of 80% and 24% for uranium and nickel, respectively. For both the Claude waste rock and the waste rock used to construct the CPE, it is suspected that these high leached fractions are the result of a large quantity of readily soluble products that the waste rock has accumulated from a rapid weathering and oxidation of the sulphides. Subsequently, this suggests that the cover of the Claude pile should target an efficient reduction of infiltration rather than a reduction of oxygen ingress within the pile

Additional Key Words: acid rock drainage, geochemistry, flow, contaminant transport.

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#### **Introduction**

Predictions of acid rock drainage and subsequent metal leaching from waste rock are commonly based on laboratory tests and modeling studies. A number of factors create uncertainties in predicting metal leaching including the differences between materials tested in the laboratory and those representatives of actual site conditions. These uncertainties are inherent to the heterogeneous character of the waste rock materials, and to scale effects, both spatial and temporal. The objective of this paper is to compare at three successive scales (i.e., laboratory columns, a 320 m<sup>3</sup> constructed waste rock pile and a 4.7 Mm<sup>3</sup> actual waste rock pile) waste rock metal leaching estimates under oxidizing conditions. The comparison criterion is the leaching efficiency, defined as mass of metal released divided by original mass, as a function of the number of pore volumes of water that have infiltrated through the waste rock.

The Cluff Lake uranium mine facility has operated since 1980. Uranium ore mining and milling is now complete and the site has moved into the decommissioning phase. The site decommissioning project has been subjected to a comprehensive study (Cogema Resources Inc., 2001) and includes an obligatory extensive follow-up program according to the Canadian Environmental Assessment Act.

## Site Description

### Overview

Cluff Lake is located in the Athabasca Basin of northern Saskatchewan, approximately 75 km south of Lake Athabasca and 30 km east of the provincial border with Alberta. Air temperatures ranged from  $-40^{\circ}$ C to  $+35^{\circ}$ C, with a mean annual temperature slightly above  $0^{\circ}$ C. The average annual precipitation is 455 mm, of which 305 mm occurs as rainfall. Between late October and the end of March, the ground surface is frozen. The mining areas on the Cluff Lake project included three open pit mines, two underground mines and three surface waste rock piles. The results presented in this paper correspond to the Claude mining area, which included, from approximately 1985 to 2004, the Claude open pit, the Claude waste rock pile and the DJN waste rock pile. An overall plan of the area is shown in Fig. 1. Decommissioning activities in the Claude area started in 2004 and will be completed in 2006. Theses activities include the relocation of the DJN waste rock, pile to the Claude open pit, the complete backfilling of the Claude pit with additional waste rock, low grade ore and site demolition materials and the placement of a till cover above the backfilled Claude pit. The decommissioning of the Claude waste rock pile involves re-contouring of the pile, placement of a till cover on the re-graded and compacted waste rock, the construction of storm water management channels and revegetation.



Figure 1. General site plan



Figure 2. Simulated steady-state distribution of hydraulic heads

# <u>Geology – Hydrogeology</u>

The surficial geology of the Claude waste rock area consists of a continuous cover of permeable, sandy till that is 2 to 15 m thick. Gravel with scattered boulders, silt and clay horizons are also represented in the overburden layer. The surficial deposits are up to 20 m thick in the Peter River valley, east of the Claude waste rock pile. A number of response tests were carried out in the surficial deposits in the Claude waste rock area. Estimated hydraulic conductivities were highly variable with values ranging from 3.0 x  $10^{-7}$  m/s to more than  $10^{-4}$  m/s. The average hydraulic conductivity of the overburden in the Claude waste rock pile area was estimated at 2.5 x  $10^{-5}$  m/s to 5 x  $10^{-5}$  m/s. Underlying the overburden are the basement aluminous gneisses. The upper 10 m of all bedrock materials is thought to be weathered and thus have a somewhat higher permeability than the deeper, unweathered bedrock. The higher permeabilities are partly attributed to paleo-weathering, weathering and fracturing. The average hydraulic conductivity of the waste rock area. The hydraulic conductivity of the weathered bedrock was estimated at approximately 4 x  $10^{-6}$  m/s. A number of fault zones are known in the waste rock area. The hydraulic conductivity of the weathered bedrock area.

Hydraulic heads and quality of groundwater in the vicinity of the Claude pile have been monitored since 1991. The current groundwater monitoring network in the Claude area includes more than 40 monitoring wells that are sampled on a monthly or quarterly basis and analyzed for major constituents, as well as dissolved trace metals (Fig. 1). The monitoring well screens were completed either in shallow sands and gravel deposits at the overburden/bedrock contact or in the upper weathered/fractured horizon of the basement gneiss. A number of trenches were also excavated and sampled for both water and soils. In addition, four observation wells were also installed within the Claude waste rock pile with screens at the base of the pile. Water levels indicate that the pile is mainly unsaturated and that the water table lies below the base of the pile. Saturated conditions appear to develop temporarily near the base of the pile as a result of

both high infiltration events and the relatively low hydraulic conductivity of the underlying materials.

Both the Claude waste rock pile and the Claude open pit influence the natural groundwater flow in the Claude mining area. The Claude pit remained dewatered throughout most of the mine life. Following mining operations, the Claude open pit was partially backfilled and was allowed to flood to the elevation 332 masl, approximately 7m below the pre-mining hydraulic head in this area. Overall, the dewatering of the Claude pit had limited effects on groundwater flow and the flow conditions were not significantly modified south and east of the Claude pile. In particular, the flow remained radial divergent from the Claude pile. Hydrogeological data collected since approximately 1990 were used in conjunction with a geological model to calibrate a three-dimensional groundwater flow and contaminant transport model for the Claude waste rock pile area. The calibrated steady-state distribution of hydraulic heads for the period with the Claude pit water level at 332 masl is shown in Fig. 2. Although the presentation of the model is not the objective of this paper, it should be noted that the model was used to verify the flow rates and travel times that contribute to the loading estimates for the Claude pile.

### The Claude Waste Rock Pile

#### **Construction**

The Claude waste rock pile was constructed between 1982 and 1989 and contains waste rock from the Claude open pit. The pile was constructed in a natural, slightly elevated area relative to the surrounding topography. From 1982 to 1984, the pile was constructed from overburden extracted from the Claude pit. This clean material was placed in the north and southwest parts of the pile. The first layers of waste rock were placed in the central part of the pile during 1985. The remaining waste rock was extracted and placed on the pile from 1986 to the end of 1989. The pile was developed by end dumping and contains well-developed traffic surfaces between the lifts of dumped material. The pile is about 29 m high at its highest point and covers an area of approximately 29.5 ha. The average thickness of waste rock in the pile is approximately 17 m. The pile contains roughly 7.9 million tonnes of waste rock for an estimated volume of approximately 4.7 Mm<sup>3</sup> and a dry density of 1.7 t/m<sup>3</sup>.

#### Waste Rock Characteristics

To obtain samples to characterize waste rock, six trenches were excavated on the Claude waste rock pile in 1999 (Fig. 1). Approximately 55 samples, each 1 m<sup>3</sup>, were taken from the trenches. Individual samples and trench composites were subjected to geochemical analysis. The tests included whole rock major oxide analysis, trace element analysis, acid base accounting (Sobek et al., 1978), modified BC SWEP tests (Province of BC, 1992), sequential extraction tests (Tessier et al., 1979), humidity-cell tests and unsaturated leach column tests. In addition saturated column tests and flushing experiments were subsequently conducted to determine initial porewater concentrations for nickel (Ni) and uranium (U) to consider the effects of sub-aqueous disposal of the waste rock (Cogema Resources Inc., 2004, Guerin et al., 2005). Solid content analysis results showed that the total sulfur content of the Claude waste rock range from less than 0.1% to approximately 0.5%. The average U and Ni contents within the Claude pile were estimated at 100 mg/kg and 53 mg/kg, respectively. The acid base accounting analysis indicated that approximately 60% of the samples tested are potentially acid generating, with NP/AP ratios less than one, and a further 25% represented uncertain acid generating potential.

The Claude waste rock reflects the characteristics of the host rock of the Claude ore body. The Claude ore body occurred in a zone of tectonized and altered aluminous gneisses. The most characteristic rock of this unit was a garnet-cordietite-sillimanite gneiss. Mineralogical and geochemical data collected during exploration of the Claude deposit (Laine et al., 1985) indicate that the gneiss was strongly altered around the mineralization with abundant argilization of the silicates. Chloritization was ubiquitous. The ore consisted essentially of pitchblende, uraninite, U-tittanates and coffinite in a matrix of phyllites and graphite. Ni was contained in the pyrite structure possibly associated with cobalt. Hematite and goethite were identified. The primary minerals positively identified in the waste rock from x-ray diffraction methods are quartz, illite, chlorite, a potassium-feldspar, kaolinite and a smectite.

### The Constructed Pile Experiment (CPE)

### Construction

An experimental waste rock pile (CPE), designed to mimic the behavior of the upper region of a much larger stockpile, was built at the Cluff Lake mine in the summer of 1998 (Nichol et al., 2000; Nichol et al, 2003). The instrumented core of the pile was 5 m high, with a footprint of 8 m by 8 m. Outflow from the base of the pile was collected in a contiguous grid of 16 zerotension lysimeters, each 2m by 2 m in area. The instrumented footprint was isolated using impermeable walls that extended the full height of the pile. The top surface was open to natural precipitation, augmented by number of artificial rainfall events. After an initial one-year period required for the water content in the pile to reach field capacity (drying occurred during construction), the experiment was continued for 3 years before a lower-permeability cover was placed on the test cell. Outflow from each lysimeter was separately piped to an instrumentation hut where outflow was monitored using tipping bucket rain gauges. Water draining from the lysimeters was then routed through a set of mixing cells. Both instantaneous grab samples and longer term composite samples were collected and analyzed.

### Waste Rock Characteristics

The pile was constructed using waste rock from the DJX pit of the Cluff site that had been mined in the fall of 1996 and exposed to natural weather conditions prior to placement in the CPE. Waste rock with a maximum diameter up to 1.5 m was randomly placed inside the test cell. Solid content analysis indicated that the total sulfur content of the DJX waste rock range from 0.44 to 0.76%. The average U and Ni contents of the DJX waste rock, based on whole-rock analysis, were estimated at 13 mg/kg and 43 mg/kg, respectively. More than 80% of the DJX samples gave NP/AP ratios substantially less than one, indicating a potential for acid generation.

### Loading Estimates from Laboratory Tests

### Sequential Extraction Tests

Leaching studies were conducted on waste rock samples from the Claude pile to determine the total inventory of soluble metals in the water within the pile. The samples were subjected to the modified Special Waste Extraction Procedure (SWEP) with a water to solid ratio of 20:1. This high ratio provides sufficient dilution that solubility limits that may be reached in pore water are not reached in the leach tests. Therefore, the soluble inventory of a particular solid can dissolve during these short-term tests. Several samples were also assessed using a water to solids ratio of 3:1 to evaluate the potential for solubility controls that may not have been evident in the 20:1 leach test results. The tests included, in addition to leach steps with de-ionized water, a leach step with a solution of hydrochloric acid adjusted to maintain the pH below 4.5. The total mass (sum of water plus HCL leach steps) extracted from the waste rock samples with this approach ranged from 30% to 58% of the average initial inventory for U and from 6% to 17% for Ni.

These results were confirmed using sequential extraction tests (Tessier et al., 1979). The results of sequential extraction tests usually provide a basis for interpreting the geochemical association of metals with different "operationally defined" phases in the solids (i.e. "water leachable", "ion exchangeable", "acid-extractable", "reducible", "oxidizable" and "residual"). The fractions to some degree reflect the experimental conditions employed, rather than true geochemical partitioning, and multiple-component extraction schemes have been the subject of much criticism (Nirel and Morel, 1990). Nonetheless, the sequential extraction results can provide an indication of major differences in geochemical association for elements of interest. For the Claude waste rock samples, the combined "water soluble plus ion exchangeable plus acid extractable plus oxidizable" fractions were found to represent approximately 68% to 73% of the total inventory for U and 29% to 47% of the inventory of Ni (see Cogema Resources Inc, 2004, for more detailed results and interpretation). A large portion of U was associated with the acid extractable fractions. This fraction represented approximately between 25% and 50% of the total inventory for U, while it represented only 7% or less of the total inventory for Ni. The association of U with the acid extractable fraction may reflect some U bound to carbonates. However, for the Claude waste rock samples, it is suspected that this fraction reflect mainly specific adsorption of U onto clay minerals and possibly Fe oxy-hydroxides for which significant sorption can occur at pH values above 4.5.

# Unsaturated Leach Column Tests

Aerobic (oxygen-rich) column tests were performed on 35 to 40 kg composite samples of Claude waste rock and DJX waste rock. The columns were constructed out of PVC pipe approximately 150 mm in diameter and 1.5 m in length. Approximately 3 litres of water was added to the cells each week. Columns were leached over 7 days cycles. Each cycle consists of 3 days dry aeration followed by 4 days wet aeration. Each 4 day wet aeration period uses de-ionized water that was aerated to oxygen saturation before introduction into the columns. The water was added by trickle leach over a 24 hour period. Following the 4 day wet aeration, water samples were drained through a 0.45  $\mu$ m millipore filter and sent for geochemical analysis. The pH of the water draining from the columns ranged from 2.9 to 4.2 and the electrical conductivity values showed a decreasing trend during the course of testing, from about 1,000 mS/m to 100 mS/m.

The fraction of the initial metal inventory released from the waste rock is plotted for Ni and U in Fig. 3 and 4 as a function of number of replacements of the waste rock pore volume. These curves suggest a maximum leachable mass on the order of 60-70% of the initial inventory for U, for both the Claude and DJX waste rock. For Ni, the leachable mass appeared to be on the order of 30% for the DJX waste rock while values ranged from 15% to 40% for the Claude waste rock. Overall, the total leachable fractions estimated from the unsaturated column tests appeared to be similar to the leachable fractions estimated from the sequential extraction tests.



Figure 3. Estimated uranium leaching efficiency from the unsaturated leach column tests



Figure 4. Estimated nickel leaching efficiency from the unsaturated leach column tests

### Loading Estimates for the CPE

Sulfate and metal release rates can also be estimated for the CPE. The pH of the water draining from the CPE ranged from 3.2 to 4.0, across the set of 16 lysimeters. Outflow concentrations for U and Ni vary in both space and time; with average concentrations across the full data set of 242 mg/l U and 245 mg/l Ni. Average release rates of 0.052 mg/kg/week and 0.05 mg/kg/week were calculated for U and Ni, respectively, over the 2000-2003 period (see Wagner et al., this volume, Table 4). The release rates estimated for 1999 were on the order of 0.08 mg/kg/week for both U and Ni. Based on 640,000 kg of waste rock in the CPE, these results translate to approximately 6.9 kg of U and 6.6 kg of Ni leached from the CPE. The initial U and Ni inventory in the CPE was estimated to be 8.3 kg and 27.5 kg, respectively, based on the average values for whole-rock U and Ni content of 13 and 43 mg/kg, respectively. Therefore, the loading estimates for the CPE suggest that approximately 83% of the estimated initial U and 24% of the Ni was leached over the time period (1999-2003). The details of the release rates (see Wagner, this volume) show that the rates slowed during the course of the experiment. Note also that the whole-rock estimates of U and Ni concentration on which these percentage estimates are based may not be entirely representative of the U and Ni concentrations in the leachable fraction of the waste rock within the CPE. It is likely that the concentrations of these elements will be higher in the finer grain size fractions (Hollings et al, 2001).

Nichol et al (2005) estimated a median residence time of water in the CPE on the order of 4 years for natural rainfall conditions. Thus, loading estimates and mass removal derived from chemical data collected prior to placement of the cover on the CPE reflect conditions where the outflow was approaching 1 pore volume for the waste rock inside the CPE.

### Loading Estimates for the Claude Pile

#### Pore Volume Estimate for Claude Pile

There are no hydrologic data available to quantify the movement of water through the interior of the Claude waste rock pile. Bellehumeur (2001) and Smith and Beckie (2003) described infiltration processes that occur on the surface of the Claude pile. Substantially higher infiltration rates occur on the western part of the pile, where the surface texture is much coarser. During an exceptionally high rainfall event in 2000, the emergence of several seeps at the base of the pile with relatively low TDS provided evidence that macropore pathways are present in the Claude pile. These pathways can transmit some component of newly-infiltrated water across the full extent of the pile within a period of a few days.

In the absence of more specific data than this, infiltration through the CPE was used to guide the development of a conceptual description of the flow within the Claude pile. The pile was constructed from 1982 to 1989, reaching a total height of 29 m, with an average thickness of approximately 17 m. For simplicity, we assume that during each year of construction, approximately 3 - 5 m of waste rock was added to the pile as it was built upwards. The objective here is to develop a first-order estimate of the number of pore volumes of water that may have infiltrated across the Claude pile since it was constructed. In the period from April to October each year, water would have been available to re-wet the waste rock that had been placed the previous year. For the CPE, it took approximately one year for drainage to be established from all 16 lysimeters (i.e., re-wetting the 5 m high test pile), although small amounts of drainage occurred in several of the basal lysimeters within a few months of waste rock placement. This response suggests the possibility that most of the water that infiltrated during pile construction went into storage to reach field capacity as each layer was added to the pile. Once the full height of the pile was attained in 1989, infiltrating water was no longer allocated to storage in newly-We speculate that the water content may have been sufficient to form a placed rock. hydraulically continuous pathway between the upper surface of the pile, and its base. This condition then creates the situation where in each subsequent year since 1989, wetting fronts in response to the larger rainfall events could propagate across the full extent of the Claude pile, displacing older resident water from the base of the pile that infiltrated during the construction phase. Thus, metal loadings to groundwater seem likely to have been occurring across most of the footprint of the Claude pile since the early 1990's. During the construction period, we would expect a limited amount of macropore flow through each lift that would have carried metals from the base of the pile toward the underlying groundwater system, perhaps as early as 1985, assuming that there was no time lag in the onset of ARD.

Infiltration through the surface of the completed Claude pile exhibited a high degree of spatial variability (Smith and Beckie, 2003). This factor creates difficulties in quantifying the volume of reactive waste rock within the pile that has been flushed by infiltrating water. To proceed, we apply the observed median residence time of water in the CPE, computed for each of the 16 basal lysimeters, indicating water moved downward through the matrix materials in the test cell at a rate of approximately 1.5 m/yr. Note this estimate neglects the influence of any zones that could potentially impede the downward infiltration of water, such as buried traffic surfaces. Using this value, we estimate that the water that infiltrated the surface of the Claude pile since completion has, on average, moved down to a depth of 20 - 25 m. Thus, we anticipate that in most regions of the pile, but especially on the thinner eastern part of the pile, a volume of

water somewhat greater than one pore volume of water may have passed through the Claude pile. The CPE results also demonstrated significant spatial variability in the outflow at the base of the pile (Nichol et al., 2005). Therefore, it seems likely that in some regions of the pile, where matrix flow rates are higher than average, one pore volume of water may have moved through those regions of the pile at least several years prior to 2005. It seems also likely that there will be other regions of the pile where this condition has not yet been met.

## Soluble mass

Groundwater monitoring results indicate that metals released from the Claude pile since 1985 have infiltrated the underlying formations and then migrated both vertically and horizontally according to the groundwater flow. Therefore the current extent of U and Ni concentrations in groundwater can be used to estimate the soluble mass of U and Ni that was released and transported from the waste rock pile in approximately 20 years. As an illustration, the estimated extent of the current U plume in groundwater is shown in Fig. 5. The contour map was generated from the concentrations measured in the monitoring wells and test pits shown in Fig. 1. Average concentrations over the 2004 – 2005 period were considered in order to smooth the monthly fluctuations in concentrations. The logarithm of the concentrations was then kriged using a simple linear model variogram and no nugget effects. It should be noted that the plume shown in Figure 5 corresponds only to the contribution of the Claude waste rock pile. The metal contributions from the nearby DJN pile were identified and subtracted using the groundwater flow model results and identification of specific "tracers" from the DJN pile (i.e., in particular radium-226 associated with the presence of special waste in the DJN pile). From a geochemical perspective, the extent and intensity of the U plume appears to be governed by the pH of the groundwater. The excellent correlation between pH and U concentrations in groundwater is illustrated in Fig. 5. The U concentrations are plotted versus the pH on a log-scale for three monitoring wells located approximately 50 m from the Claude pile. The pH at these wells oscillates around a value of approximately 4. This value is a good geochemical signature consistent with the dissolution of Al. The bedrock in the Claude pile area is composed of aluminous gneisses and it is reasonable to consider that Al hydroxides buffer the pH in the groundwater system.

The maximum concentrations of U and Ni in the monitoring wells surrounding the pile were recorded during the nineties with values as high as 500 mg/l for U and 80 mg/l for Ni. Since approximately 1998 the tendency is toward stabilization or a decrease of the concentrations. In the most recent data available (2005), the concentrations in groundwater underneath the pile are on the order of 15 mg/l for U and Ni for a pH of 4.5.

Based on the contour map in Fig. 5, the average U concentration over the current plume area was estimated at 9.3 mg/l. The corresponding average saturated thickness affected by the U concentrations was estimated at approximately 4 m, including both the overburden material and the weathered bedrock. The porosity of the sandy till at the Cluff Lake site was measured to be about 30%. Assuming a porosity of 25% for the combined till and weathered bedrock, the total mass of uranium currently in solution was estimated at 16 tonnes. A similar approach was used for Ni and the total mass of Ni in solution was estimated at 6.5 tonnes.



Figure 5. The distribution of uranium in groundwater for the Claude waste rock pile area in 2004-2005 and illustration of the uranium vs pH relationship in monitoring wells surrounding the pile.

These soluble inventories of U and Ni in the groundwater were verified using the time varying concentrations and water levels measured in the monitoring wells located immediately downgradient from the pile since 1991. Each monitoring well was considered to be indicative of a section of the aquifer through which the plume was transported. The flow model was used to estimate the flow associated with each section. Subsequently the U and Ni mass flux were calculated based on the measured concentrations. The calculated U and Ni cumulative release curves are illustrated in Fig. 6. Totals of about 18 tonnes of U and 6 tonnes of Ni were calculated with this approach. These values compare relatively well with the previous estimates, which were based on the current extent of the plumes.



Figure 6. Estimated soluble uranium and nickel mass released from the Claude waste rock pile since 1990.

### Sorbed mass

The sorbed component associated with U and Ni concentrations in groundwater needs also to be considered in order to calculate the total mass released from the pile. Batch tests were conducted on samples of weathered gneisses in order to evaluate the potential for sorption of Ni and U. The tests were based on the EPA procedure EPA/530/SW-87/006-F. In order to simulate conditions relatively similar to those found in groundwater adjacent to the Claude waste rock pile, the tests were carried out at a constant target pH of 4.2. The rock was in contact with the solutions for 72 and 75 hours in order to establish equilibrium concentrations. The tests showed limited sorption of both U and Ni by the gneisses. The batch tests gave estimates of K<sub>d</sub> on the order of 1.5 L/kg for U and K<sub>d</sub> values ranging from 0.6 L/kg to 4.7 L/kg for Ni.

In addition to the batch tests, the overburden till material was sampled south and east of the Claude pile within the groundwater plume area, in the vicinity of monitoring wells. Concentrations of U in the solids ranging from 40 to 50 mg/kg were associated with concentrations in groundwater ranging from about 6 to 10 mg/L and a pH around 4.5. Concentrations of Ni in the solids ranging from 15 to 40 mg/kg were associated with concentrations in groundwater of approximately 12 mg/L. Assuming equilibrium concentrations, this translates to  $K_d$  values on the order of 4 L/kg to 8 L/kg for U and 1 L/kg to 4 L/kg for Ni.

For the purpose of estimating the sorbed mass,  $K_d$  values ranging from 1 L/kg to 2 L/kg were assumed for both U and Ni. These values were derived from the batch tests and solid content analysis of the till but also from the calibration of a contaminant transport model (not presented in this paper). Assuming a bulk dry density of 1.85 t/m<sup>3</sup> for the overburden and weathered bedrock, the sorbed masses associated with the leaching from the pile were estimated at 115 tonnes and 48 tonnes for U and Ni, respectively, for a K<sub>d</sub> of 1 L/kg, and at 230 tonnes and 96 tonnes for U and Ni, respectively, using a K<sub>d</sub> of 2 L/kg. Significant uncertainty may be

associated with the sorbed masses estimated in this fashion because of the large variability of U and Ni adsorption in the pH range from 3.5 to 6. For example, U sorption on hydrated ferric oxides or ferric hydroxides calculated from data presented by Dzombak and Morel (1990) shows that greater than 90% of U can be sorbed at pH values greater than 4.5 and only 50% of U may be sorbed at pH values near 3.5. EPA (1999) reviewed uranium K<sub>d</sub> values obtained for a number of soils, crushed rock material, and single-mineral phases. The review indicated that, although uranium K<sub>d</sub> values can exhibit a great deal of scatter at any fixed pH value, the adsorption of uranium increase rapidly with increasing pH from pH 3 to 5 and reaches a maximum in adsorption in the pH range from pH 5 to 8. Given the orders of magnitude variability reported for uranium values, it is considered that a K<sub>d</sub> of 2 L/kg for U significantly underestimates sorption at distance from the pile where the pH of groundwater is greater than 5.

#### **Discussion and Conclusions**

A summary of laboratory and field-derived uranium and nickel leached fractions is provided in Table 1 for both the Claude waste rock (Claude pile) and the DJX waste rock (CPE). Field estimates suggest that the total mass of U released from the Claude waste rock pile in 20 years ranged from approximately 130 tonnes to 245 tonnes, representing on the order of 16% to 30% of the initial U inventory in the solids. Similarly, field estimates suggest that the total mass of Ni released from the Claude pile ranged from approximately 55 tonnes to 130 tonnes, representing on the order of 12% to 25% of the initial Ni inventory. Based on leaching test results, this translates to leached masses ranging from 25% to 45% % of the total leachable mass for U, and from 31% to 58% of the total leachable mass for Ni. Although there are no hydrologic data available to quantify the movement of water through the interior of the Claude pile, it seems likely that these estimates correspond to the flushing of approximately one pore volume of the Claude pile in 20 years. Table 1 shows that the field loading estimates appear to be relatively consistent with the loading estimates from unsaturated column tests (Fig 2. and 3.).

At an intermediate scale, the loading estimates for the CPE show that approximately 80% and 24% of the estimated initial U and Ni inventory were leached, respectively, during one flush of pore volume through the constructed pile. Based on leaching test results, this represents approximately 80% of the total leachable mass for Ni. For U, the calculated leached mass (80%) appears to be greater than the maximum leachable mass (70%) estimated from the unsaturated leach column test (Fig 3.). It is suspected that the whole-rock estimates of U and Ni concentrations on which these estimates are based may not be entirely representative of the U and Ni solid concentrations in the leachable fraction of the waste rock.

Both the U and Ni loading estimates suggest that a significant portion of the leachable mass was released during the first pore volume replacement of the waste rock mass. For both the Claude waste rock and the waste rock used to construct the CPE, it is suspected that these relatively high leaching efficiencies result from the solid phases controls on U and Ni concentrations, which may be more reasonably associated with sorption than with individual mineral phases. It is also suspected that the limited scale effect between laboratory and field results is the result of a relatively large quantity of readily soluble products that the waste rock has accumulated as a result of a rapid weathering and oxidation of the sulfides. Subsequently,

this suggests that the cover of the pile should target an efficient reduction of infiltration rather than a reduction of oxygen ingress within the pile.

	Leaching (percent of estimated initial mass)	
	Uranium	Nickel
Claude waste rock (Claude Pile)		
Sequential extraction	68 - 73%	29 - 48 %
Leach columns		
1 pore volume	22 - 25%	5 - 19%
11 pore volumes	54 - 60%	17 - 42%
Field estimates (~ 1 pore volume)	16 - 30%	12 - 25 %
DJX waste rock (CPE)		
Leach columns		
1 pore volume	~42%	~15 %
11 pore volumes	~70 %	~31 %
Field estimates (~ 1 pore volume)	~ 83%	~24 %

Table 1: Summary of uranium and nickel leached fractions from laboratory and field experiments

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