Wet Barriers on Pyrite Uranium Tailings Laboratory Lysimeter Studies of Oxidation, Leaching and Limestone Neutralization Characteristics of Uranium Tailings and Waste Rock MEND Project 2.13.1a

- Part I: Oxidation and Leaching Characteristics of Total Mill Tailings, Coarse Tailings and Waste Rock, and Limestone Neutralization Characteristics of Tailings mixed with Limestone, under Unsaturated and Batch Leaching Conditions
- Part II: Oxidation and Leaching Characteristics of Coarse Tailings under Submerged Conditions

This work was done on behalf of MEND and sponsored by the Canada Centre for Mineral and Energy Technology (CANMET) and Rio Algom Limited Elliot Lake, Ontario

**January 1998** 

# Wet Barriers on Pyritic Uranium Tailings Part I and II Laboratory Lysimeter Studies of Oxidation, Leaching and Limestone Neutralization Characteristics of Uranium Tailings and Waste Rock

Nand K. Davé Report MMSL 98 - 001 (CR) January 1998

- Part I: Oxidation and Leaching Characteristics of Total Mill Tailings, Coarse Tailings and Waste Rock, and Limestone Neutralization Characteristics of Tailings mixed with Limestone, under Unsaturated and Batch Leaching Conditions
- Part II: Oxidation and Leaching Characteristics of Coarse Tailings under Submerged Conditions

#### Work Performed for

Rio Algom Limited, Elliot Lake, Ontario under Contract No. 50948

## WET BARRIERS ON PYRITIC URANIUM TAILINGS PART I AND II

## LABORATORY LYSIMETER STUDIES OF OXIDATION, LEACHING AND LIMESTONE NEUTRALIZATION CHARACTERISTICS OF URANIUM TAILINGS AND WASTE ROCK

Nand K. Davé\*

#### **EXECUTIVE SUMMARY**

This report describes the results of laboratory column leaching studies that were conducted as a part of a broad research program titled "Development of Wet Barriers on Pyritic Uranium Tailings for Controlling Acid Generation". The research was undertaken jointly by CANMET, Elliot Lake Laboratory and Rio Algom Limited, under the Canadian Mine Environmental Neutral Drainage (MEND) Program.

#### **Purpose:**

Column leaching studies were conducted for evaluating the oxidation and leaching characteristics of pyritic uranium tailings with and without limestone amendments and crushed waste rock, under unsaturated and submerged conditions. These experiments were designed to determine the acid generation characteristics and subsequent metal and radionuclide releases of test samples subjected to submersion underwater and exposed conditions.

#### Methods:

The leaching experiments were conducted using PVC cylindrical column lysimeters filled with total mill tailings, coarse tailings, tailings amended with 7.5% (w/w) limestone of varying screen

<sup>&</sup>lt;sup>\*</sup>Senior Environmental Research Scientist, Elliot Lake Laboratory, CANMET, Natural Resources, Canada.

sizes, and crushed waste rock samples obtained from Quirke Mine. The experiments provided unsaturated conditions for leaching of test samples as well as a submerged condition for coarse tailings where a shallow water cover, 0.45 m in depth, was established using distilled water. The submerged coarse tailings were also rinsed rapidly during the initial leaching period to remove gypsum and other soluble minerals for studying oxidation and leaching characteristics of such tailings in the absence of gypsum controlled sulphate solubility.

All tests were conducted at laboratory room temperature (22 °C) and light conditions, which aided in enhancing the chemical reactivity of test materials in laboratory simulations compared to field conditions.

#### **Duration of the Study:**

The experiments, conducted at the CANMET, Elliot Lake Laboratory, were started in April 1989 and concluded in June 1993.

#### **Results:**

#### **Coarse Tailings:**

Unsaturated Condition:

- The unsaturated coarse tailings without limestone amendment oxidized readily and produced highly acidic drainage (pH ~ 1.0 2.0 and acidity ~ 12,000 30,000 mg CaCO<sub>3</sub>/l);
- The coarse tailings amended with coarse limestone (screen size -6.3 mm) also produced highly acidic drainage (pH ~ 2.5 3.0 and acidity ~ 4,000 12,000 mg CaCO<sub>3</sub>/l), but its onset was delayed by approximately one year in comparison to coarse tailings without limestone. The acid drainage occurred with 90% of the available alkalinity still remaining in the sample;

 The coarse tailings amended with pulverized wet ground limestone did not produce any acidic drainage during the entire four year study period. Oxidation and acid generation processes were active in these tailings as well, but the fine grained limestone provided complete acid neutralization and acidic drainage was prevented during the study period.

#### Submerged Condition:

- The submerged coarse tailings did not produce any porewater acidic drainage for the first three years, but in the fourth year a weak acidic drainage (pH ~ 5.5 7.0 and acidity ~ 10 20 mg CaCO<sub>3</sub>/l) was observed ;
- In comparison to unsaturated coarse tailings, submersion of tailings underwater decreased cumulative porewater acidity, sulphate and iron loadings by factors of 295, 200 and 950 respectively;
- The submersion of coarse tailings also resulted in increased mobility and release of Ra-226 in the porewater when tailings developed acidic conditions at the surface after they were depleted of available alkalinity and gypsum. There was a 10 fold increase in the cumulative loading of Ra-226 in the porewater over that of unsaturated coarse tailings;
- The increased drainage of Ra-226 in the porewater was also accompanied by that of iron, both occurring near the end of the fourth year, where as gypsum was completely removed from the tailings during the first three months of leaching;
- The surface water above submerged tailings contained low concentrations of dissolved metals, when the water cover above the tailings was continuously maintained in a well oxygenated condition by fresh water inflow and where both surface and pore water flows were present in equal proportions. The dissolved Ra-226 concentration in the surface water was also low, ranging between 30 and 140 mBq/l.

#### **Total Mill Tailings:**

**Unsaturated Condition:** 

- The total mill tailings, in all test conditions, drained poorly and retained appreciable amounts of moisture which controlled their oxidation and acid generation rates;
- The unsaturated total mill tailings without limestone amendment also produced acid drainage, but it was moderate and less acidic (pH  $\sim 2.0 3.0$  and acidity  $\sim 500 1500$  mg CaCO<sub>3</sub>/l) than for coarse tailings under similar conditions;
- The total mill tailings amended with various screen-sized limestone did not produce acid drainages during the entire study period. The tailings were oxidizing slowly but the added limestone was actively consumed in providing complete acid neutralization in all amended samples.

#### Waste Rock:

**Unsaturated Conditions:** 

- The unsaturated crushed waste rock having a high acid generation potential (NNP = -17.8 kg CaCO<sub>3</sub>/tonne) also oxidized and started to produce highly acidic drainage ( pH ~ 2.0 - 2.5 and acidity ~ 7,000 - 20,000 mg CaCO<sub>3</sub>/l) in the fourth year of the study period;
- The unsaturated crushed waste rock having medium (NNP = -10.7 kg CaCO<sub>3</sub>/tonne) and low (NNP = +13.8 kg CaCO<sub>3</sub>/tonne) acid generation potentials did not produce acidic drainage during the entire study period.

#### **DETAILED SUMMARY OF FINDINGS**

In 1989, under the Mine Environmental Neutral Drainage (MEND) program, a joint research project titled "Development of Wet Barriers on Pyritic Uranium Tailings for Controlling Acid Generation" was established between CANMET, Elliot Lake Laboratory and Rio Algom Limited, Elliot Lake, Ontario, Canada. This research program encompassed a variety of laboratory and field studies designed to evaluate the performance of a shallow water cover or other wet barriers on pyritic uranium tailings as a close-out option for controlling acid generation and release of metals and radionuclides.

As a part of this program, extensive laboratory column leaching studies were conducted to evaluate oxidation and leaching characteristics of tailings and waste rock samples under unsaturated conditions, tailings amended with 7.5 % (w/w) limestone of varying screen sizes, and submerged coarse tailings.

For these tests, two types of tailings and three types of waste rock were chosen. The two types of tailings were: total mill tailings (unclassified), approximately 50 % less than 74  $\mu$ m; and coarse tailings, approximately 84 % greater than 74  $\mu$ m. The three types of waste rock were categorized as having, low (NNP = +13.8), medium (NNP = -10.7) and high (NNP = -17.8 kg CaCO<sub>3</sub>/tonne) acid generation potentials. The experiments were conducted in PVC columns, under unsaturated and submerged conditions. T he latter tests were conducted only for coarse tailings, where a shallow water cover, approximately 0.45 m in depth, was provided using distilled water.

For both unsaturated and submerged cases, the tailings or waste rock were allowed to weather at the exposed surface or tailing / water interface and leached with natural lake or distilled water (for submerged tailings only). The unsaturated tailings and waste rock were batch leached by adding 1 1 (litre) natural lake water every two weeks where as the submerged tailings were trickle leached with a continuous inflow of distilled water. The leachates were allowed to percolate through the test media and collected as drainage effluents from the bottom of the columns in all cases.

The experiments, conducted at the CANMET, Elliot Lake Laboratory, were started in April 1989 and concluded in June 1993.

Table S1 provides a brief summary of various test conditions and porewater drainage (seepage) quality results for this laboratory study. The detailed leaching results, comprised of both the drainage water quality and metal/radionuclide loadings are summarized below. In interpreting these results, the reader is reminded of their origin under laboratory simulated and controlled conditions, and they should be judged accordingly as well as in that context only. The laboratory conditions were designed to enhance chemical reactions and may not represent actual field conditions.

- The unsaturated coarse tailings without additional limestone (Control -2), were most amenable to oxidation and acidic drainage. These tailings drained quickly, had a high hydraulic conductivity of 4 x  $10^{-3}$  cm/s and produced a highly acidic drainage within two months of batch leaching.
- During the acidic drainage period, the effluent from these tailings had an average pH of 1.2, an oxidation-reduction potential (Eh) of 700 mV and an electrical conductance (Ec) of 40,000 µS/cm. The effluent average loading rates were: 70,000 µg CaCO<sub>3</sub> equivalent acidity, 80,000 µg SO<sub>4</sub><sup>-2</sup>, 25,000 µg Fe, 690 µg Ca and 6.2 mBq Ra-226/kg tailings/day. For the four year study period, the cumulative productions/removals (loadings) for major species were, respectively, 58 % of total acid generation potential, 67 % of total sulphur, 68.7 % of total iron, 19 % of total calcium and 0.17 % of total Ra-226 contained in the tailings.

- The acidic drainage also caused significant mobilization and release of other metals and radionuclides including: Al, Ce, Cu, Mg, Mn, Ni, Pb, Th, U, Zn and various isotopes of radium, Ra-223, Ra-224 and Ra-228.
- The acidic drainage in these tailings started prior to full consumption of the total available alkalinity, although it was present only in a small amount. At the time of acidic drainage, approximately 60 -65 % of available alkalinity was still present in the tailings.
- It was estimated that under laboratory controlled conditions, a 0.45 m high column of coarse tailings would be completely oxidized and depleted of pyrite in approximately 6.5 years.
   For Ra-226, it would take approximately 5000 years for complete removal excluding its natural decay or ingrowth in this sample.
- Prior to the acidic drainage, the Ra-226 leachability from the coarse tailings was constant and controlled by the gypsum solubility. With the onset of the acidic drainage, it increased with the leaching of thorium isotopes. In the absence of gypsum, Ra-226 leachability decreased with increasing acidity and sulphate ion concentration. The mobilization and release of thorium isotopes were accompanied by an increased release of radium isotopes including Ra-228, Ra-226, Ra-224 and Ra-223. At the peak acidic drainage period, the activities of other radium isotopes were considerably higher than that of Ra-226.
- The unsaturated coarse tailings containing 7.5 % (w/w) of coarse limestone of screen size 6.3 mm also oxidized and produced a highly acidic drainage. The onset of the acidic drainage in these tailings, however, was delayed by approximately one year in comparison to coarse tailings without the additional limestone (Control -2) which occurred in two months.
- During the acidic drainage period, the effluent from the coarse tailings coarse limestone sub-group had an average pH of 2.5, Eh of 700 mV and Ec of 6000  $\mu$ S/cm. The effluent average loading rates were: 25,000  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 28,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 8000

μg Fe, 3800 μg Ca and 7 mBq Ra-226/kg tailings/day. The cumulative productions/removals (loadings) for major species during the study period were, respectively, 17 % of total acid generation potential, 25 % of total sulphur, 17 % of total iron, 12 % of total calcium and 0.29 % total Ra-226 contained in the tailings-limestone mixture. The acidic drainage also caused significant mobilization and release of other constituents including Al, Ce, Cu, Mg, Mn, Ni, Pb, Th, U and the various isotopes of radium.

- The acid neutralization efficiency achieved for coarse limestone (-6.3 mm), prior to the occurrence of the acidic drainage, was less than 10 %. The acidic drainage in coarse tailings
   coarse limestone sub-group started with 90 % of the total available limestone alkalinity still remaining in the mixture.
- The unsaturated coarse tailings mixed with 7.5 % (w/w) wet ground and pulverized limestone did not produce any acidic drainage during the four year study period. Oxidation and acid generation were also taking place in these tailings, but the finely ground limestone provided complete acid neutralization, thus preventing acidic drainage.
- During the study period, the effluent from coarse tailings wet ground limestone sub-group was nearly neutral to slightly alkaline with a pH of ? 8, Eh ? 480 mV and Ec ? 2200  $\mu$ S/cm. The effluent was saturated with respect to gypsum and contained appreciable amounts of dissolved sulphate, calcium, and magnesium, resulting from acid neutralization, and Ra-226. In the absence of the acidic drainage, the release of other oxidation related reaction products, e.g. acidity, Al, Ce, Cu, Fe, Ni, Pb, Th, U and Zn was absent or present in trace amounts only. Mn was present in small quantities in the neutral effluent. The cumulative productions/removals (loadings) for major constituents of the effluent were: 7 % of total sulphur, 8.8 % of total calcium and 0.16 % of total Ra-226 contained in the tailings-limestone mixture.

- Approximately 13 % of the total available limestone alkalinity in the coarse tailings wet ground limestone mixture was consumed in complete acid neutralization during the entire study period. For these samples, the remaining alkalinity would provide additional acid neutralization for another 29 years assuming a 100 % limestone neutralization efficiency, and for a shorter period at reduced efficiency. The data did not permit determination of the neutralization efficiency factor and prediction of acid drainage, if any.
- The unsaturated total mill tailings without additional limestone (Control -1) also produced acid drainage, but it was moderate and less severe than for coarse tailings(Control -2). These tailings had a low hydraulic conductivity of  $8.7 \times 10^{-6}$  cm/s and retained appreciable amounts of moisture which caused slow oxidation resulting in reduced acid generation and release of oxidation reaction products.
- During the acidic drainage period, the effluent from Control 1 tailings had an average pH of ~3.0, Eh ? 780 mV and Ec ? 2500  $\mu$ S/cm. The effluent contained appreciable amounts of gypsum, and the average loading rates were: 2100  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 6000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 100  $\mu$ g Fe, 1500  $\mu$ g Ca and 4 mBq Ra-226/kg tailings/day. The cumulative productions/removals (loadings) of major species during the entire study period were: 3 % of total acid generation potential, 8 % of total sulphur, 0.33 % of total iron, 13.6 % of total calcium and 0.08 % of total Ra-226 contained in the tailings.
- The acid generation rate for total mill tailings (Control 1) was approximately 30 to 35 times less than that for coarse tailings(Control 2). The acidic drainage from these tailings also caused mobilization and release of other metals and radionuclides: Al, Ce, Cu, Mg, Mn, Ni, Pb, Th, U, Zn and other isotopes of radium, but in reduced amounts.
- At the observed low acid generation rate, the present 0.35 m high column of unsaturated total mill tailings without additional limestone would produce acid drainage for approximately 90 years.

- Disposal of tailings as a homogeneous mixture without segregation of various particle size fractions, would offer many advantages including: low hydraulic conductivity and increased surface drainage; increased water retention, high phreatic surface and shallow unsaturated zone amenable to oxidation; low acid generation and effluent loading rates, and low treatment costs; and a low risk of cover failure.
- The unsaturated total mill tailings mixed with 7.5 % (w/w) limestone of various screen sizes, from coarse (-6.3 mm) to wet ground and pulverized, did not produce any acidic drainage during the entire study period. Both, the oxidation and acid generation processes were also active in these tailings-limestone sub-groups, but a low rate of acid generation resulted in a complete acid neutralization for all limestone sizes and acidic drainage was prevented. The reactivity of the tailings decreased with the decrease in limestone particle size from coarse to pulverized and wet ground. The low rate of acid generation in combination with increased effluent retention time, contributed to the complete acid neutralization for total mill tailings mixed with coarse limestone as well.
- For all test columns containing total mill tailings amended with various screen sized limestone, the effluents were saturated with respect to gypsum and the effluent average pH, Eh and Ec, were, respectively, 7.5, 550 mV and 2200 µS/cm. After two years, the effluents were alkaline with alkalinity production (loading) rate of approximately 200 µg CaCO<sub>3</sub>/kg tailings/day. During the study period, the effluent average loading rates were: 5000 µg SO<sub>4</sub><sup>-2</sup>, 1800 µg Ca and 3 mBq Ra-226/kg tailings/day. The corresponding cumulative productions/removals (loadings) were: 8.6 % of total sulphur, 5.7 % of total calcium, 22 % of the initial gypsum and 0.06 % of total Ra-226 contained in the tailings. The release of other oxidation reaction products, acidity, Al, Ce, Cu, Fe, Ni, Pb, Th. U and Zn were absent or present only in trace amounts. Mn was present in small quantities in the neutral to alkaline effluents.
- It was estimated that the added limestone alkalinity to total mill tailings would effectively prevent acid drainage from test samples for approximately 100 years assuming a 100%

limestone neutralization efficiency, and for a reduced period of time if the efficiency was less. The data indicated that at 100% limestone neutralization efficiency, these tailings-limestone samples should not produce any acidic drainages, but the determinations of their actual neutralization efficiencies and predictions of acid drainages were not possible.

- Disposal of low sulphur tailings amended with finely ground pulverized limestone, added in stoichiometric proportion to their maximum acid generation potential, in the form of a thickened paste may provide an alternate insitu method of controlling acid generation above the water table.
- For pyritic uranium tailings mixed with pulverized limestone, there was the added advantage of immobilization and containment of the associated radioactivity in a gypsum controlled system, further reducing the risk of environmental migration of radionuclides.
- For the submerged coarse tailings, there was no acidic drainage for the first three years of the study. With the initial high porewater flow, the tailings were depleted of gypsum and other soluble minerals during the first three months of leaching. After gypsum removal, the drainage effluents were nearly neutral with a pH ~ 7.0, Eh ~ 425 mV and Ec ~ 80 µS/cm. During that period, no oxidation reaction products, most notably Fe, Al, Mn and Th, were produced or released, and the tailings produced slightly alkaline effluents with alkalinity in the range of 10 40 mg CaCO<sub>3</sub>/l.
- The submerged tailings started to produce slightly acidic porewater drainages in the fourth year of the study, with effluents having pH's of  $\sim 5.5$ , total acidities of ? 10 20 mg/l, dissolved total Fe concentrations of ? 1 20 mg/l and Ra-226 concentrations of ? 5000 25,000 mBq/l. The results indicated onset of a slow oxidation process at the surface of the tailings contributing to 10 to 30 fold increases in Ra-226 mobilization and release to the porewater associated with iron oxidation and its increased drainage.

- After the removal of gypsum and other soluble minerals, the effluent average loading rates for submerged coarse tailings were:  $180 \ \mu g \ CaCO_3$  equivalent acidity,  $300 \ \mu g \ SO_4^{-2}$ ,  $20 \ \mu g \ Fe$ ,  $200 \ \mu g \ Ca$  and  $150 \ mBq \ Ra-226/kg$  tailings/day. The corresponding cumulative productions/removals (loadings) were:  $0.2 \ \%$  of total acid generation potential,  $0.03 \ \%$  of total sulphur,  $0.07 \ \%$  of total iron,  $6 \ \%$  of total calcium and  $5.5 \ \%$  of total Ra-226 contained in the tailings.
- In comparison to unsaturated coarse tailings (Control 2) and total mill tailings (Control 1), the shallow water cover on coarse tailings reduced total acidity production by factors of 295 and 10, total sulphate production by 200 and 20, and total iron production by 950 and 3, respectively.
- For total mill tailings mixed with 7.5 % limestone of various screen sizes, the corresponding decrease in total acidity production was by factors of 1900, 50 and 7 when compared with unsaturated coarse tailings, unsaturated total mill tailings and submerged coarse tailings, respectively. For the duration of the present study, the limestone amendment to total mill tailings was more effective in preventing acid drainage than water cover on coarse tailings. However, limestone was consumed in the acid neutralization process and its addition would provide benefits for a finite period.
- For submerged tailings, the Ra-226 mobility was initially controlled by gypsum dissolution, which decreased with the depletion of gypsum. In the absence of gypsum, and without oxidation, there was no corresponding increase in Ra-226 drainage although the effluent sulphate concentrations were very low. With the onset of a slow oxidation, the mobility of Ra-226 increased significantly, up to 30 times the pre-oxidation value, and correlated with the corresponding slow increase in iron and perhaps, Th-230 release. The effluent sulphate loading also increased at the same time, but it was statistically not significant.

- The cumulative Ra-226 removal from submerged tailings during the study period was approximately 30 to 50 times greater than those for unsaturated coarse and total mill tailings without or with limestone amendments. This increased Ra-226 drainage was 3 to 6 times more than the corresponding increase in cumulative effluent volume for the submerged tailings over those of the unsaturated tailings. Perhaps, in the absence of gypsum and/or the lack of a moderate to severe acidification in the submerged tailings, the sulphate concentration was too low to suppress or adversely effect radium solubility.
- In the water column above the tailings surface, when both surface and pore water flows were present, and in the absence of gypsum, there was very little upward diffusion of dissolved metals and oxidation reaction products except Ra-226, whose concentration varied between 30 140 mBq/l in the surface water. In comparison to porewater effluents, which became slightly acidic, Ra-226 concentrations in the surface water were low and below the regulatory discharge guidelines of 1000 mBq/l.
- Because of high porewater flows in the present study, the surface water quality above the submerged tailings resembled that of the inflowing distilled water. The surface water was characterized by a pH in the range of ~ 5 5.7, Eh ~ 500 600 mV, Ec ~10 15  $\mu$ S/cm, both acidity and alkalinity in the range of ~ 3 6 mg CaCO<sub>3</sub>/l, SO<sub>4</sub><sup>-2</sup> ~ 3 8 mg/l, Fe ~ 0.01 0.05 mg/l, Ca ~ 0.1 1.0 mg/l and Mn ~ 0.08 0.1 mg/l.
- The leaching experiments for submerged tailings were conducted with distilled water which lacked in alkalinity and organic matter contents of natural waters. The observed increase in radium mobility with the oxidation of tailings underwater, after gypsum had been depleted, however suggested that additional oxidation control measures might be required in preventing tailings from going acidic.
- In shallow waters, and, in the absence of oxygen consuming covers or barriers such as organic-rich sediments, the tailings surface at the tailings-water interface was subjected to

well oxygenated water and was amenable to oxidation. In the presence of gypsum, the radium mobility and release was limited and controlled by gypsum solubility with or without oxidation of pyrite. However, after all the gypsum was dissolved and removed, further oxidation of tailings contributed to additional release of radium isotopes via both surface and ground water regimes. This scenario may occur many years after decommissioning and monitoring phases, and should be given due consideration in environmental impact assessment and modeling, as well as additional oxidation controlling measures.

- The unsaturated crushed waste rock with high acid generation potential (NNP = -17.8 kg CaCO<sub>3</sub>/tonne) also oxidized and produced a highly acid drainage in the fourth year of the study period. The leaching characteristics resembled those of coarse tailings with 7.5 % coarse limestone where delayed acid drainage occurred.
- In the acidic effluent, the average pH, Eh and Ec were, respectively, 2.2, 780 mV and 7000  $\mu$ S/cm. The effluent average loading rates were: 38,000  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 42,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 12,500  $\mu$ g Fe, 2000  $\mu$ g Ca and 3.0 mBq Ra-226/kg waste rock/day. The cumulative productions/removals (loadings) during the four year study period for major species were: 53 % of total acidity, 60 % of total sulphur, 31 % of total iron, 59 % of total calcium and 0.08 % of total Ra-226 contained in the waste rock, respectively.
- The reactivity of the high acid generating crushed waste rock was higher than that for coarse tailings, and most of its leaching occurred during the last year of the study period. The acidic drainage also caused significant mobilization and release of other constituents including Al, Ce, Cu, Mg, Mn Ni, Pb, Th, U, Zn and various isotopes of radium.
- The acid drainage in the high acid generating waste rock also occurred with 75 % of the available limestone alkalinity still remaining, resulting in an average neutralization efficiency of approximately 25% for this type of crushed waste rock. As the acid drainage related concentration peaks of various species occurred during the last year of the study period, the

data did not fully permit extrapolation and computations of reaction rate constants for acid production and metal mobilities, etc.

- The unsaturated crushed waste rock with medium acid generation potential (NNP =  $-10.7 \text{ kg CaCO}_3/\text{tonne}$ ) did not produce any acidic drainage during the study period. The results indicated that oxidation, acid generation and neutralization processes were active within the waste rock, but the acid was completely neutralized preventing acidic drainage and release of oxidation reaction products.
- During the study period, the effluent from medium acid generating waste rock was nearly neutral to slightly alkaline with a pH of ? 7.5, Eh ? 400 mV and Ec ? 1000 µS/cm. It contained mostly dissolved gypsum, resulting from acid neutralization, and Ra-226. The effluent cumulative productions/removals (loadings) for major species were, respectively: 11.9 % of total sulphur, 30 % of total calcium and 0.04 % of total Ra-226 contained in the waste rock. Some limestone dissolution contributed to alkalinity and additional calcium in the effluents.
- The acid neutralization rate for the medium acid generating waste rock was comparable to that of high acid generating waste rock, and by the end of the study period approximately 20 % of the total limestone alkalinity had also been consumed in preventing acidic drainage. From acid generation/neutralization rates, it was calculated that acidic drainage would occur from this waste rock after approximately 12 years of leaching, at 100 % limestone neutralization efficiency, and sooner if the efficiency was reduced.
- The unsaturated crushed waste rock with low acid generation potential (NNP = +13.8 kg CaCO<sub>3</sub>/tonne), also did not produce any acidic drainage during the study period. Similar to the medium acid generating waste rock, oxidation, acid generation and neutralization processes were all actively taking place in this waste rock also, but the neutralization was

complete and acidic drainage and concomitant mobilization of oxidation reaction products were absent.

- The effluents from low acid generating waste rock were slightly alkaline during the entire study period with a pH of ~ 8.0, Eh ~ 450 mV and Ec ~ 1000  $\mu$ S/cm. The drainages were mostly gypsum derived and contained appreciable amounts of calcium, and sulphate, resulting from complete acid neutralization, and Ra-226. The effluent cumulative productions/removals (loadings) for major species were, respectively: 19 % of total sulphur, 11.6 % of total calcium and 0.06 % of total Ra-226 contained in the waste rock.
- During the study period, approximately 2.5 g CaCO<sub>3</sub>/kg waste rock or 9 % of total limestone alkalinity was consumed in the complete acid neutralization process in the low acid generating waste rock. Stoichiometrically, at 100 % limestone neutralization efficiency and at the experimentally determined acid generation/neutralization rates, this waste rock should not produce any acidic drainage. However, at a decreased limestone neutralization efficiency, acidic drainage may occur in the next twenty years or so, when all the sulphur contained in the waste rock is expected to oxidize.
- The calculated acid generation and limestone neutralization rates, prior to acidic drainage, for the three waste rock types were comparable, but only the high acid generating waste rock produced acidic drainage The low total neutralization potential of 8.5 kg CaCO<sub>3</sub>/tonne for the high acid generating waste rock and perhaps, the size and distribution of alkali minerals with respect to sulphides were contributing factors in this waste rock. For the other two, low and medium acid generating waste rock, the total neutralization potentials were, respectively, 26.82 and 15.06 kg CaCO<sub>3</sub>/tonne.

In conclusion, the usefulness of column leaching tests has been well demonstrated in understanding the oxidation and leaching kinetics of reactive tailings and waste rock, and for evaluating various decommissioning options. A shallow water cover on pyritic uranium tailings is very effective in controlling/limiting their oxidation and acid generation to very low rates, but lacked in complete prevention. The tests have also shown that radium mobility increases, in the absence of gypsum, when acidic conditions develop at the surface of underwater deposited tailings. Limestone amendments to tailings , in some cases, provided short-term controls on acidic drainage.

xviii

Table S1: Summary of test conditions and porewater effluent (seepage) quality results for column leaching studies.

Experimental Arrangement	Sample Type	Material Classification	Amendments	Leaching Conditions	Leaching Solution	Porewater Effluent Characteristics		Characteristics
						Acid Mine Drainage	Effluent pH	Major Metals and Radionuclides
Group #1								
Columns #1 - #3	Tailings (Control-1)	Unclassified total mill tailings, un-oxidized	None	Unsaturated	Gravel pit lake water	Yes Moderate acidity: 500 - 1500 mg CaCO <sub>3</sub> /l	Initial 7.0 - 8.0 final 2.0 -3.0	Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Zn, Th, U and Ra-226 (500 - 3500 mBq/l)
Columns #4 - #6	Tailings	Unclassified total mill tailings, un-oxidized	7.5% (w/w)* coarse limestone, size -6.3 mm	Unsaturated	Gravel pit lake water	None	7.0 - 8.0	Ca, Mg, Mn and Ra-226 (300 - 2000 mBq/l)
Columns #7 - #9	Tailings	Unclassified total mill tailings, un-oxidized	7.5% (w/w)* medium size limestone, size -2.4 mm	Unsaturated	Gravel pit lake water	None	7.0 - 8.0	Ca, Mg, Mn and Ra-226 (300 - 2000 mBq/l)
Columns #10 - #12	Tailings	Unclassified total mill tailings, un-oxidized	7.5% (w/w)* fine limestone, size -0.84 mm	Unsaturated	Gravel pit lake water	None	7.0 - 8.0	Ca, Mg, Mn and Ra-226 (300 - 2000 mBq/l)
Columns #13 - #15	Tailings	Unclassified total mill tailings, un-oxidized	7.5% (w/w)* pulverized wet ground limestone	Unsaturated	Gravel pit lake water	None	7.0 - 8.0	Ca, Mg, Mn and Ra-226 (300 - 2000 mBq/l)

\* w/w = weight percentage

Table S1(continued): Summary of test conditions and porewater effluent (seepage) quality results for column leaching studies.

Experimental Arrangement	Sample Type	Material Classification	Amendments	Leaching Conditions	Leaching Solution	Porewater Effluent Characteristics		
						Acid Mine Drainage	Effluent pH	Major Metals and Radionuclides
Group #2								
Columns #16 and #17	Tailings (Control-2)	Coarse tailings, un-oxidized	None	Unsaturated	Gravel pit lake water	Yes, High acidity: 12,000-30,000 mg CaCO <sub>3</sub> /l	Initial 7.5 - 8.0 final 1.0 - 2.0	Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Zn, Th, U and Ra-226 (100 - 5000 mBq/l)
Columns #18 and #19	Tailings	Coarse tailings, un-oxidized	7.5% (w/w)* coarse limestone, size -6.3 mm	Unsaturated	Gravel pit lake water	Yes, 1 year delay, High acidity: 4000-12,000 mg CaCO <sub>3</sub> /l	Initial 8.0 - 8.5 final 2.5 - 3.0	Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Zn, Th, U and Ra-226 (600 - 4500 mBq/l)
Columns #20 and #21	Tailings	Coarse tailings, un-oxidized	7.5% (w/w)* pulverized wet ground limestone	Unsaturated	Gravel pit lake water	None	8.0 - 8.5	Ca, Mg, Mn and Ra-226 (500 - 2000 mBq/l)
Group #3								
Columns #22 - #24	Tailings	Coarse tailings, un-oxidized	None	Submerged	Distilled water	None for the first three years, very weak in the fourth year: acidity 10-20	Initial 7.5 - 8.0 final 5.5 - 7.0	Ca, Fe, Mg, Mn, and Ra-226 (1000 - 27,000 mBq/l)

\* w/w = weight percentage

Table S1(continued): Summary of test conditions and porewater effluent (seepage) quality results for column leaching studies.

Experimental Arrangement	Sample Type	Material Classification	Amendments	Leaching Conditions	Leaching Solution	Porewater Effluent Characteristics		
						Acid Mine Drainage	Effluent pH	Major Metals and Radionuclides
Group #4								
Columns #25 and #26	Waste rock	Crushed, size -2.0 mm, un-oxidized, medium acid generating, NNP = $-10.7$ Kg CaCO <sub>3</sub> /tonne	None	Unsaturated	Gravel pit lake water	None	7.2 - 8.0	Ca, Mg, U and Ra-226 (50 - 300 mBq/l)
Columns #27 and #28	Waste rock	Crushed, size -2.0 mm, un-oxidized, high acid generating, NNP = $-17.8$ Kg CaCO <sub>3</sub> /tonne	None	Unsaturated	Gravel pit lake water	Yes: 3 year delay, High acidity: 7,000-20,000 mg CaCO <sub>3</sub> /l	Initial 7.5 - 8.0 final 2.0 - 2.5	Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Zn, Th, U and Ra-226 (100 - 2000 mBq/l)
Columns #29 and #30	Waste rock	Crushed, size -2.0 mm, un-oxidized, low acid generating, NNP = $+13.8$ Kg CaCO <sub>3</sub> /tonne	None	Unsaturated	Gravel pit lake water	None	7.5 - 8.0	Ca, Mg, U and Ra-226 (100 - 450 mBq/l)

## **Table of Contents**

			Page
EXI	ECI	UTIVE SUMMARY	i
SUN	ЛN	IARY OF FINDINGS	v
	1.	INTRODUCTION	1-1
		1.1 Acid Generation Control - Shallow Water Cover / Wet Barriers	1-1
		1.2 Background to the Study	1-3
		1.3 Wet Barriers Research Program	1-6
	2.	LABORATORY LYSIMETER STUDIES-EXPERIMENTAL METHODOLOGY	1-8
		2.1 Objectives and Study Plans	1-8
		2.2 Column Lysimeter Leaching Studies	1-9
		2.3 Experimented Details	1-11
		2.3.1 Column Lysimeter - Experimental Arrangement	1-11
		2.3.2 Sampling Procedure: Tailings and Waste Rock	1-14
		2.3.2.1 Unclassified Total Mill Tailings	1-14
		2.3.2.2 Coarse Tailings	1-15
		2.3.2.3 Waste Rock	1-15
		2.3.3 Sample Preparation and Column Loading	1-16
		2.3.4 Column Permeability Measurements	1-17
		2.3.5 Leaching Studies: Schedule and Sampling Procedures	1-18
		2.4 Analytical Procedures and Quality Assurance / Quality Control	1-21
		2.4.1 Water Samples	1-21
		2.4.1.1 Primary Parameters	1-21
		2.4.1.2 Elemental Analysis	1-23
		2.4.1.3 Sulphate Analysis	1-23
		2.4.1.4 Ra-226 Analysis	1-24
		2.4.1.5 Minimum Detection Limits	1-25
		2.4.2 Solid Samples	1-25
		2.4.2.1 Sample Digestion	1-25
		2.4.2.2 Silica Analysis	1-26
		2.4.2.3 Sulphur Speciation	1-26

	2.4.3 Quality Assurance/Quality Control (QA/QC)	1-27
3.	RESULTS	1-28
	3.1 Column Permeability Results	1-28
	3.2 Solid Phase Composition	1-30
	3.3 Column Leaching Results	1-32
	3.3.1 Unsaturated Tailings	1-33
	3.3.1.1 Group 1: Total Mill Tailings	1-33
	3.3.1.1.1 Group 1: Total Mill Tailings Without Additional Limestone (Columns #1 - 3)	1-33
	<ul><li>3.3.1.1.2 Group 1: Total Mill Tailings Mixed With</li><li>7.5% Limestone of Various Grain Sizes</li><li>(Columns #4 - 15)</li></ul>	1-42
	3.3.1.2 Group 2: Coarse Tailings	1-49
	3.3.1.2.1 Group 2: Control - 2, Un-oxidized Coarse Tailings Without Limestone Amendment (Columns #16 and 17)	1-49
	3.3.1.2.2 Group 2: Un-oxidized Coarse Tailings Mixed With 7.5% (w/w) Coarse (-6.3 mm size) Limestone (Columns #18 and 19)	1-58
	3.3.1.2.3 Group 2: Un-oxidized Coarse Tailings Mixed With 7.5% (w/w) Wet ground and Pulverized Limestone (Columns #20 and 21)	1-69
	3.3.2 Submerged Tailings	1-76
	3.3.2.1 Group 3: Submerged Coarse Tailings	1-76
	3.3.2.1.1 Group 3: Un-oxidized Coarse Tailings Submerged Under 0.45 m Water Cover (Columns #22 - 24)	1-76
	3.3.3 Crushed Waste Rock	
	3.3.3.1 Group - 4: Un-oxidized Crushed Waste Rock (Unsaturated Conditions)	
	3.3.3.1.1 Group - 4: Un-oxidized Crushed Waste Rock With Medium Acid Generation Potential (NNP = -10.7 Kg CaCO <sub>3</sub> /tonne, Columns #25 and 26)	1-91
	3.3.3.1.2 Group - 4: Un-oxidized Crushed Waste Rock With High Acid Generation	

	Potential (NNP = $-17.8$ Kg CaCO <sub>3</sub> /tonne,	
	Columns #27 and 28) 1-96	5
	3.3.3.1.3 Group - 4: Un-oxidized Crushed	
	Waste Rock With Low Acid Generation	
	Potential (NNP = $+13.8$ Kg CaCO <sub>3</sub> /tonne,	
	Columns #29 and 30)	ł
4.	DISCUSSION	)
5.	SUMMARY AND CONCLUSIONS	2
6.	ACKNOWLEDGMENT	7
7.	REFERENCES	3

xxiv

## List of Figures

Figu	re	Page	
1.	Generalized location of uranium mine tailings sites in Elliot Lake, Ontario, Canada	2-	·1
2.	General plan of submersion of tailings at the Quirke Mine waste management area	2-	-2
3.	Schematic profile of submersion of tailings at the Quirke Mine waste management area	2-	.3
4.	General plan of submersion of tailings at the Panel Mine waste management area	2-	-4
5.	Experimental arrangement of the column leaching lysimeter for leaching under unsaturated conditions	2-	.5
6.	Experimental arrangement of the column leaching lysimeter for leaching under submerged conditions	2-	·6

## Leaching Profiles Of Total Mill Tailings Without Additional Limestone And Under Unsaturated Conditions (Control - 1, Columns #1-3)

7.	Variation of pH with time	2-7
8.	Variation of Eh with time	2-8
9.	Variation of Ec with time	2-9
10.	Variation of total acidity loading with time	2-10
11.	Cumulative acidity production with time	2-11
12.	Variation of total alkalinity loading with time	2-12
13.	Variation of dissolved $SO_4^{-2}$ loading with time	2-13
14.	Cumulative $SO_4^{-2}$ removal with time	2-14
15.	Variation of dissolved total Fe loading with time	2-15
16.	Cumulative total Fe removal with time	2-16
17.	Variation of dissolved Ca loading with time	2-17
18.	Cumulative Ca removal with time	2-18
19.	Variation of dissolved Mg loading with time	2-19
20.	Cumulative Mg removal with time	2-20
21.	Variation of dissolved Al loading with time	2-21
22.	Cumulative Al removal with time	2-22

Variation of dissolved Mn loading with time	2-23
Cumulative Mn removal with time	2-24
Variation of dissolved U loading with time	2-25
Variation of dissolved Th loading with time	2-26
Cumulative Th removal with time	2-27
Variation of dissolved Pb loading with time	2-28
Variation of dissolved Ra-226 loading with time	2-29
Cumulative Ra-226 removal with time	2-30
Average cumulative drainage volume collected with time	2-31
	Cumulative Mn removal with time Variation of dissolved U loading with time Variation of dissolved Th loading with time Cumulative Th removal with time Variation of dissolved Pb loading with time Variation of dissolved Ra-226 loading with time Cumulative Ra-226 removal with time

## Leaching Profiles Of Total Mill Tailings Mixed With 7.5% (W/W) Coarse Limestone (Screen Size -6.3 mm, -4 Mesh) And Under Unsaturated Conditions (Columns #4-6)

32.	Variation of pH with time	2-32
33.	Variation of Eh with time	2-33
34.	Variation of Ec with time	2-34
35.	Variation of total acidity loading with time	2-35
36.	Variation of total alkalinity loading with time	2-36
37.	Variation of dissolved $SO_4^{-2}$ loading with time	2-37
38.	Cumulative $SO_4^{-2}$ removal with time	2-38
39.	Variation of dissolved total Fe loading with time	2-39
40.	Variation of dissolved Ca loading with time	2-40
41.	Cumulative Ca removal with time	2-41
42.	Variation of dissolved Mg loading with time	2-42
43.	Cumulative Mg removal with time	2-43
44	Variation of dissolved Al loading with time	2-44
45.	Variation of dissolved Mn loading with time	2-45
46.	Variation of dissolved U loading with time	2-46
47.	Variation of dissolved Th loading with time	2-47
48.	Variation of dissolved Pb loading with time	2-48
49.	Variation of dissolved Ra-226 loading with time	2-49

xxvii

50.	Cumulative Ra-226 removal with time	2-50
51.	Average cumulative drainage volume collected with time	2-51
Med	hing Profiles Of Total Mill Tailings Mixed With 7.5% (W/W) ium Size Limestone (Screen Size -2.4 mm, -8 Mesh) And Under aturated Conditions (Columns #7-9)	
52.	Variation of pH with time	2-52
53	Variation of Eh with time	2-53
54	Variation of Ec with time	2-54
55	Variation of total acidity loading with time	2-55
56	Variation of total alkalinity loading with time	2-56
57	Variation of dissolved $SO_4^{-2}$ loading with time	2-57
58	Cumulative $SO_4^{-2}$ removal with time	2-58
59	Variation of dissolved total Fe loading with time	2-59
60	Variation of dissolved Ca loading with time	2-60
61	Cumulative Ca removal with time	2-61
62	Variation of dissolved Mg loading with time	2-62
63	Cumulative Mg removal with time	2-63
64	Variation of dissolved Al loading with time	2-64
65	Variation of dissolved Mn loading with time	2-65
66	Variation of dissolved U loading with time	2-66
67	Variation of dissolved Th loading with time	2-67
68	Variation of dissolved Pb loading with time	2-68
69	Variation of dissolved Ra-226 loading with time	2-69
70	Cumulative Ra-226 removal with time	2-70
71	Average Cumulative drainage volume collected with time	2-71

## Leaching Profiles Of Total Mill Tailings Mixed With 7.5% (W/W) Fine Size Limestone (Screen Size -0.84 mm, -20 Mesh) And Under Unsaturated Conditions (Columns #10-12)

72	Variation of pH with time	2-72
73	Variation of Eh with time	2-73

## xxviii

74	Variation of Ec with time	2-74
75	Variation of total acidity loading with time	2-75
76	Variation of total alkalinity loading with time	2-76
77	Variation of dissolved $SO_4^{-2}$ loading with time	2-77
78	Cumulative $SO_4^{-2}$ removal with time	2-78
79	Variation of dissolved total Fe loading with time	2-79
80	Variation of dissolved Ca loading with time	2-80
81	Cumulative Ca removal with time	2-81
82	Variation of dissolved Mg loading with time	2-82
83	Cumulative Mg removal with time	2-83
84	Variation of dissolved Al loading with time	2-84
85	Variation of dissolved Mn loading with time	2-85
86	Variation of dissolved U loading with time	2-86
87	Variation of dissolved Th loading with time	2-87
88	Variation of dissolved Pb loading with time	2-88
89	Variation of dissolved Ra-226 loading with time	2-89
90	Cumulative Ra-226 removal with time	2-90
91	Average Cumulative drainage volume collected with time	2-91

## Leaching Profiles Of Total Mill Tailings Mixed With 7.5% (W/W) Wet Ground (Pulverized) Limestone Under Unsaturated Conditions (Columns #13-15)

92	Variation of pH with time	2-92
93	Variation of Eh with time	2-93
94	Variation of Ec with time	2-94
95	Variation of total acidity loading with time	2-95
96	Variation of total alkalinity loading with time	2-96
97	Variation of dissolved $SO_4^{-2}$ loading with time	2-97
98	Cumulative $SO_4^{-2}$ removal with time	2-98
99	Variation of dissolved total Fe loading with time	2-99
100	Variation of dissolved Ca loading with time	2-100

Cumulative Ca removal with time	2-101
Variation of dissolved Mg loading with time	2-102
Cumulative Mg removal with time	2-103
Variation of dissolved Al loading with time	2-104
Variation of dissolved Mn loading with time	2-105
Variation of dissolved U loading with time	2-106
Variation of dissolved Th loading with time	2-107
Variation of dissolved Pb loading with time	2-108
Variation of dissolved Ra-226 loading with time	2-109
Cumulative Ra-226 removal with time	2-110
Average cumulative drainage volume collected with time	2-111
	Variation of dissolved Mg loading with time Cumulative Mg removal with time Variation of dissolved Al loading with time Variation of dissolved Mn loading with time Variation of dissolved U loading with time Variation of dissolved Th loading with time Variation of dissolved Pb loading with time Variation of dissolved Ra-226 loading with time Cumulative Ra-226 removal with time

# Leaching Profiles Of Coarse Tailings Without Additional Limestone Under Unsaturated Conditions (Control - 2, Columns #16 and 17)

112	Variation of pH with time	.2-112
113	Variation of Eh with time	.2-113
114	Variation of Ec with time	.2-114
115	Variation of total acidity loading with time	.2-115
116	Cumulative acidity production with time	.2-116
117	Variation of total alkalinity loading with time	.2-117
118	Variation of dissolved $SO_4^{-2}$ loading with time	.2-118
119	Cumulative $SO_4^{-2}$ removal with time	.2-119
120	Variation of dissolved total Fe loading with time	.2-120
121	Cumulative total Fe removal with time	.2-121
122	Variation of dissolved Ca loading with time	.2-122
123	Cumulative Ca removal with time	.2-123
124	Variation of dissolved Mg loading with time	.2-124
125	Cumulative Mg removal with time	.2-125
126	Variation of dissolved Al loading with time	.2-126
127	Cumulative Al removal with time	.2-127
128	Variation of dissolved Mn loading with time	.2-128

129	Cumulative Mn removal with time	2-129
130	Variation of dissolved U loading with time	2-130
131	Variation of dissolved Th loading with time	2-131
132	Cumulative Th removal with time	2-132
133	Variation of dissolved Pb loading with time	2-133
134	Cumulative Pb removal with time	2-134
135	Variation of dissolved Ra-226 loading with time	2-135
136	Cumulative Ra-226 removal with time	2-136
137	Average cumulative drainage volume collected with time	2-137
138	Alpha energy spectrum obtained for column #17 during peak acidic drainage period (Oct. 3, 1990)	2-138

## Leaching Profiles Of Segregated Coarse Tailings Mixed With 7.5% (W/W) Coarse Limestone (Screen Size -6.3 mm, -4 Mesh) And Under Unsaturated Conditions (Columns #18 and 19)

139	Variation of pH with time	2-139
140	Variation of Eh with time	2-140
141	Variation of Ec with time	2-141
142	Variation of total acidity loading with time	2-142
143	Cumulative acidity production with time	2-143
144	Variation of total alkalinity loading with time	2-144
145	Variation of dissolved $SO_4^{-2}$ loading with time	2-145
146	Cumulative $SO_4^{-2}$ removal with time	2-146
147	Variation of dissolved total Fe loading with time	2-147
148	Cumulative total Fe removal with time	2-148
149	Variation of dissolved Ca loading with time	2-149
150	Cumulative Ca removal with time	2-150
151	Variation of dissolved Mg loading with time	2-151
152	Cumulative Mg removal with time	2-152
153	Variation of dissolved Al loading with time	2-153
154	Cumulative Al removal with time	2-154
155	Variation of dissolved Mn loading with time	2-155

156	Cumulative Mn removal with time	2-156
157	Variation of dissolved U loading with time	2-157
158	Variation of dissolved Th loading with time	2-158
159	Cumulative Th removal with time	2-159
160	Variation of dissolved Pb loading with time	2-160
161	Cumulative Pb removal with time	2-161
162	Variation of dissolved Ra-226 loading with time	2-162
163	Cumulative Ra-226 removal with time	2-163
164	Average cumulative drainage volume collected with time	2-164

## Leaching Profiles Of Segregated Coarse Tailings Mixed With 7.5% (W/W) Wet Ground (Pulverized) Limestone Under Unsaturated Conditions (Columns #20 and 21)

165	Variation of pH with time	2-165
166	Variation of Eh with time	2-166
167	Variation of Ec with time	2-167
168	Variation of total acidity loading with time	2-168
169	Variation of total alkalinity loading with time	2-169
170	Variation of dissolved $SO_4^{-2}$ loading with time	2-170
171	Cumulative $SO_4^{-2}$ removal with time	2-171
172	Variation of dissolved total Fe loading with time	2-172
173	Variation of dissolved Ca loading with time	2-173
174	Cumulative Ca removal with time	2-174
175	Variation of dissolved Mg loading with time	2-175
176	Cumulative Mg removal with time	
177	Variation of dissolved Al loading with time	2-177
178	Variation of dissolved Mn loading with time	2-178
179	Cumulative Mn removal with time	2-179
180	Variation of dissolved U loading with time	2-180
181	Variation of dissolved Th loading with time	2-181
182	Variation of dissolved Pb loading with time	2-182

xxxii

183	Variation of dissolved Ra-226 loading with time	2-183
184	Cumulative Ra-226 removal with time	2-184
185	Average Cumulative drainage volume collected with time	2-185

## Leaching Profiles Of Segregated Coarse Tailings Without Additional Limestone Under Submerged Conditions (Columns #22-24)

186	Variation of pH with time2-186
187	Variation of Eh with time
188	Variation of Ec with time
189	Variation of total acidity loading with time2-189
190	Cumulative acidity production with time
191	Variation of total alkalinity loading with time2-191
192	Variation of dissolved $SO_4^{-2}$ loading with time
193	Cumulative $SO_4^{-2}$ removal with time
194	Variation of dissolved total Fe loading with time
195	Cumulative total Fe removal with time
196	Variation of dissolved Ca loading with time
197	Cumulative Ca removal with time
198	Variation of dissolved Mg loading with time2-198
199	Cumulative Mg removal with time
200	Variation of dissolved Al loading with time2-200
201	Cumulative Al removal with time
202	Variation of dissolved Mn loading with time2-202
203	Cumulative Mn removal with time
204	Variation of dissolved U loading with time
205	Variation of dissolved Th loading with time2-205
206	Variation of dissolved Pb loading with time2-206
207	Variation of dissolved Ra-226 loading with time
208	Cumulative Ra-226 removal with time
209	Average cumulative drainage volume collected with time2-209

#### xxxiii

## Leaching Profiles Of Crushed Waste Rock Having Medium Acid Generation Potential (NNP = -10.7 Kg CaCO<sub>3</sub> / Tonne) And Under Unsaturated Conditions (Columns #25 and 26)

210	Variation of pH with time2-210
211	Variation of Eh with time
212	Variation of Ec with time
213	Variation of total acidity loading with time2-213
214	Variation of total alkalinity loading with time2-214
215	Variation of dissolved $SO_4^{-2}$ loading with time
216	Cumulative $SO_4^{-2}$ removal with time
217	Variation of dissolved total Fe loading with time2-217
218	Variation of dissolved Ca loading with time2-218
219	Cumulative Ca removal with time
220	Variation of dissolved Mg loading with time2-220
221	Cumulative Mg removal with time
222	Variation of dissolved Al loading with time2-222
223	Variation of dissolved Mn loading with time2-223
224	Variation of dissolved U loading with time
225	Cumulative U removal with time2-225
226	Variation of dissolved Th loading with time2-226
227	Variation of dissolved Pb loading with time2-227
228	Variation of dissolved Ra-226 loading with time2-228
229	Cumulative Ra-226 removal with time
230	Average cumulative drainage volume collected with time2-230

## Leaching Profiles Of Crushed Waste Rock Having High Acid Generation Potential (NNP = -17.8 Kg CaCO<sub>3</sub> / Tonne) And Under Unsaturated Conditions (Columns #27 and 28)

231	Variation of pH with time	
232	Variation of Eh with time	2-232
233	Variation of Ec with time	2-233

## xxxiv

234	Variation of total acidity loading with time	2-234
235	Cumulative acidity production with time	2-235
236	Variation of total alkalinity loading with time	2-236
237	Variation of dissolved $SO_4^{-2}$ loading with time	2-237
238	Cumulative $SO_4^{-2}$ removal with time	2-238
239	Variation of dissolved total Fe loading with time	2-239
240	Cumulative total Fe removal with time	2-240
241	Variation of dissolved Ca loading with time	2-241
242	Cumulative Ca removal with time	2-242
243	Variation of dissolved Mg loading with time	2-243
244	Cumulative Mg removal with time	2-244
245	Variation of dissolved Al loading with time	2-245
246	Cumulative Al removal with time	2-246
247	Variation of dissolved Mn loading with time	2-247
248	Cumulative Mn removal with time	2-248
249	Variation of dissolved U loading with time	2249-
250	Cumulative U removal with time	2-250
251	Variation of dissolved Th loading with time	2-251
252	Cumulative Th removal with time	2-252
253	Variation of dissolved Pb loading with time	2-253
254	Cumulative Pb removal with time	2-254
255	Variation of dissolved Ra-226 loading with time	2-255
256	Cumulative Ra-226 removal with time	2-256
257	Average cumulative drainage volume collected with time	2-257

## Leaching Profiles Of Crushed Waste Rock Having Low Acid Generation Potential (NNP = +13.8 Kg CaCO<sub>3</sub> / Tonne) And Under Unsaturated Conditions (Columns #29 and 30)

258	Variation of pH with time	
259	Variation of Eh with time	2-259
260	Variation of Ec with time	

#### XXXV

261	Variation of total acidity loading with time	2-261
262	Variation of total alkalinity loading with time	2-262
263	Variation of dissolved $SO_4^{-2}$ loading with time	2-263
264	Cumulative $SO_4^{-2}$ removal with time	2-264
265	Variation of dissolved total Fe loading with time	2-265
266	Variation of dissolved Ca loading with time	2-266
267	Cumulative Ca removal with time	2-267
268	Variation of dissolved Mg loading with time	2-268
269	Cumulative Mg removal with time	2-269
270	Variation of dissolved Al loading with time	2-270
271	Variation of dissolved Mn loading with time	2-271
272	Variation of dissolved U loading with time	2-272
273	Cumulative U removal with time	2-273
274	Variation of dissolved Th loading with time	2-274
275	Variation of dissolved Pb loading with time	2-275
276	Variation of dissolved Ra-226 loading with time	2-276
277	Cumulative Ra-226 removal with time	2-277
278	Average cumulative drainage volume collected with time	2-278

# xxxvi

# List of Tables

Г	Table	Page
S1.	Summary of test conditions and porewater effluent (seepage) quality results for column leaching studies	xvii
1.	Sample arrangement of batch sampling of total mill tailings from Quirke Mill filtration circuit	1-14
2.	Minimum detection limits for various elements and Ra-226 using ICAP-AES and radiochemical methods respectively	3-1
3.	Comparison of analytical results of a Dissolved River Sediment Certified Reference Material (CRM-RS-A) for QA/QC program:	3-2
4.	Comparison of analytical results of a Trace Metal Drinking Water Certified Reference Material (CRM-TMDW) for QA/QC program	3-3
5.	Results of an inter-laboratory comparison study for the QA/QC program	3-4
6.	Column permeability (saturated hydraulic conductivity) results for group 1 - total mill tailings, columns 1-15	3-5
7.	Column permeability (saturated hydraulic conductivity) results for group 2 - coarse tailings, Columns 16-21	3-6
8.	Column permeability (saturated hydraulic conductivity) results for group 3 - submerged coarse tailings, Columns 22-24	3-7
9.	Column permeability (saturated hydraulic conductivity) results for group 4 - Crushed Waste Rock, Columns 25-30	3-8
10.	Analysis of variance of column permeability results between control tailings and limed tailings	3-9
11.	Analysis of variance of column permeability results amongst columns within the same group	3-10
12.	Solid phase composition of total mill tailings without additional limestone (Control-1), columns 1-3	3-11
13.	Solid phase composition of total mill tailings mixed with 7.5% limestone (w/w), columns 4-15	3-12
14.	Solid phase composition of segregated coarse tailings without limestone, Columns 16 and 17, and 22-24	3-13
15.	Solid phase composition of coarse tailings mixed with 7.5% (w/w) limestone, Columns 18-21	3-14
16.	Solid phase composition of Quirke Mine waste rock (QWR-A), medium acid generation potential, Columns 25 and 26	3-15
17.	Solid phase composition of Quirke Mine waste rock (QWR-B), high acid generation potential, Columns 27 and 28	3-16

18.	Solid phase composition of Quirke Mine waste rock (QWR-C),	
	low acid generation potential, Columns 29 and 30	3-17

xxxviii

# List of Attachments

1.		Leaching results for total mill tailings without additional limestone under unsaturated conditions (Control - 1, Columns #1-3).
2.	Appendix B:	Leaching results for total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (Columns #4-6).
3.	Appendix C:	Leaching results for total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (Columns #7-9).
4.	Appendix D:	Leaching results for total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (Columns #10-12).
5.	Appendix E:	Leaching results for total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone and under unsaturated conditions (Columns #13-15).
6.	Appendix F:	Leaching results for coarse tailings without additional limestone and under unsaturated conditions (Control - 2, Columns #16 and 17).
7.	Appendix G:	Leaching results for coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (Columns #18 and 19).
8.	Appendix H:	Leaching results for coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone and under unsaturated conditions (Columns #20 and 21).
9.	Appendix I:	Leaching results for coarse tailings without additional limestone and under submerged conditions (Columns #22-24).
10.	Appendix J:	Leaching results for crushed waste rock having medium acid generation potential (NNP = $-10.7$ kg CaCO <sub>3</sub> /tonne) and under unsaturated conditions (Columns #25 and 26).
11.	Appendix K:	Leaching results for crushed waste rock having high acid generation potential (NNP = $-17.8$ kg CaCO <sub>3</sub> /tonne) and under unsaturated conditions (Columns #27 and 28).
12.	Appendix L:	Leaching results for crushed waste rock having low acid generation potential (NNP = $+13.8$ kg CaCO <sub>3</sub> /tonne) and under unsaturated conditions (Columns #29 and 30).

#### **1. INTRODUCTION**

CANMET, Elliot Lake laboratory and Rio Algom Limited, Elliot Lake Division, under the umbrella of the Mine Environment Neutral Drainage (MEND) Program, signed a memorandum of understanding (MOU) in 1989, to undertake a three year joint research program dealing with the development of a shallow water cover or wet barriers on pyritic uranium tailings. The objective was to investigate the feasibility of establishing such covers on existing tailings, as a decommissioning option, for long term management and control of acid generation, and to study the associated mobilization of heavy metals and radionuclides.

During this program, CANMET, Elliot Lake Laboratory conducted extensive laboratory and field investigations for evaluating the oxidation, leaching and limestone neutralization characteristics of unclassified total mill tailings, coarse mill tailings and waste rock, and the role of a shallow water cover and wetlands in controlling acid generation and release of tailings derived contaminants into the environment. Rio Algom Limited undertook detailed geotechnical studies dealing with the establishment of a large scale field demonstration and water cover test site, its construction as a pilot project and monitoring of its performance for water cover retention and impact on surface and ground water quality. At the completion of these studies, Rio Algom Limited awarded a contract to CANMET, Elliot Lake Laboratory in late 1993 for compilation and reporting of the laboratory experimental results. This work was performed by N. Davé and the scientific and technical staff of the Elliot Lake laboratory.

#### 1.1 Acid Generation Control - Shallow Water Cover / Wet Barriers

Uranium mining waste in Elliot Lake is both acid generating and slightly radioactive. Acid generation is the single most important problem causing mobilization of iron, other metals and radionuclides resulting in a low level radioactive acid mine drainage. With the acidic conditions, most of the uranium and thorium decay series radionuclides are also mobilized, notably radioisotopes of U, Th and Ra, etc. Ra is also leached at neutral and alkaline pH conditions but

its solubility is inversely dependent upon the sulphate ion concentration. With unsaturated conditions and upon drying, the waste material is further subjected to wind erosion, where dispersal of radioactive dust particulates, and radon gas and its progeny also becomes very important factor that needs to be addressed when considering the long term management aspects of these wastes.

Acid generation from reactive sulphide wastes can be controlled by eliminating or reducing one or more of the essential components: Reactive sulphide minerals, water or moisture and oxygen; or by controlling the environment at source in order to retard the rate of acid generation to an insignificant level. The exclusion of oxygen from the reactive waste is probably the most effective long-term acid generation control technique. Appropriate cover materials for the purpose of limiting oxygen entry include water, low permeability soils and clays, synthetic liners and their combinations.

Water, with its low oxygen diffusion coefficient  $(2x10^{-9} \text{ m}^2 \text{ s}^{-1})$  and low oxygen solubility (~8.6 g m<sup>-3</sup> at 25 °C), is probably the most economical and best oxygen limiting cover that is readily available for acid generation control measures. In the absence of convective transport, the rate of oxygen diffusion through water is too slow to be of any significance for acid generation (Davé 1992).

Similar to oxygen, the diffusion coefficient of radon gas in water  $(1.13 \times 10^{-9} \text{ m}^2 \text{ s}^{-1})$  is approximately four orders of magnitude lower than in air  $(1.1 \times 10^{-5} \text{ m}^2 \text{ s}^{-1})$  (Tanner, 1964 and Nielson et. al 1984). Further, in comparison to dry tailings, the radon exhalation rates from wet and saturated uranium tailings decreased by factors of 4 to 20, depending upon the degree of moisture saturation and particle size distribution of the tailings (Silker, et al 1979, Davé and Lim 1987, Silker and Heasler 1979, Strong K. P. and Levins, D. M, 1982, Macbeth et. al, 1977). A water cover on uranium tailings, thus offers the added advantage of reducing or eliminating radon exhalations and its decay products, as well as controlling the airborne dispersal of radioactive dust particulates. Water cover or wet barriers on acid generating uranium tailings and waste rock provides the following four important control measures:

- 1. Limiting available oxygen for the oxidation process, and hence controlling acid generation,
- Eliminating surface erosion by wind and water action when placed in a depositional basin, thus controlling dusting problems,
- 3. Creating a reducing environment suitable for supporting nitrate and sulphate reducing microorganisms in sediment/substrate where, respectively, ammonia alkalinity and hydrogen sulphide gas are produced. The latter reacts with dissolved metals and precipitates insoluble metal sulphides, and
- Attenuating or eliminating radon gas exhalations and radioactive dust particulates from the uranium waste.

#### **1.2** Background to the Study

In the uranium mining district of Elliot Lake, Ontario, all the operating uranium mines, except one, are closed because of poor uranium prices and low ore grades. In 1990, Rio Algom Limited, Elliot Lake Division, closed the Quirke and Panel Mines, with the remaining Stanleigh Mine, scheduled to cease operation in 1996. In 1992, another mining company, Denison Mines, shut down its only operating mine. There are approximately 170 million tonnes of uranium tailings deposited at various surface impoundment facilities that will be decommissioned.

The uranium mill tailings in the Elliot Lake region are acid generating, low level radioactive wastes, containing approximately 5 to 7% pyrite and low concentrations (10 to 15 Bq  $g^1$ ) of uranium and thorium decay series radionuclides, e.g. U-238, U-234, Th-232, Th-230, Ra-228, Ra-226, and Pb-210 etc. Although the tailings were discharged at neutral to alkaline pH's, the resident alkalinity of the host rock was consumed in the milling process, where an acid leach process was used for uranium extraction. Consequently, these tailings have low buffering capacity. Acid generation and the concomitant release of metals and/or radionuclides are

environmental problems that need to be addressed when evaluating decommissioning options for such wastes.

In the past, many such inactive tailing sites have been successfully reclaimed by re-vegetation which has been very effective in providing physical stability and improving aesthetics. The vegetative cover, however, has not been effective in providing any control over acid generation and migration of contaminants (Veldhuizen et al 1987, Davé and Lim 1988, and Boucher et Al 1989).

With the wet barriers/water cover as an emerging technology for controlling acid generation as well as radon exhalation, both Elliot Lake mining companies have proposed decommissioning of their recently inactive tailings sites by a large scale submersion of tailings underwater and establishment of an insitu water cover. All of the sites are ideally situated in valleys with cross-valley engineered dams and favourable topography for providing a permanent water cover.

Rio Algom's plan for covering Quirke and Panel waste management areas (Fig. 1) with water consists of raising the crests of low elevation impoundment dams to submerge all the exposed waste underwater either as a single basin (Panel site) or as a multi cell terraced basin (Quirke site). In both cases, a minimum of 0.6 m deep water cover will be provided at high elevation tailings discharge locations. At far ends and low elevation locations, the depth of the water cover may vary from 1 m to several meters.

The Quirke waste management area contains approximately 48 million tonnes of uranium mine tailings and development waste rock (approximately 4 million tonnes), and covers an area of 190 ha with a drainage basin of 275 ha (Balins et 1991 and Kay et Al 1992). There is a 15 m elevation difference between the west end of the Quirke tailings basin and the east end effluent discharge area. To enable covering the tailings with water, the basin will be developed in a terraced manner by constructing four internal dykes and dividing the basin into a series of internal cells, which will effectively deal with the decrease in the tailings' surface elevation from

west to east (Figures 2 and 3). The drop in elevation from cell to cell will be about 4 m, representing an overall average gradient of approximately 0.5%. These internal cells will be retained by dykes constructed of mine waste rock, tailings and glacial till. The Quirke Waste Management Area will be allowed to submerge underwater by normal precipitation over the entire basin by collecting the area run-off in a nearby Gravel Pit Lake and diverting its overflow to the tailings basin..

The Panel Waste Management area contains (Figure 4) approximately 16 million tonnes of uranium mine tailings and waste rock (approximately 0.5 million tonnes), and covers an area of 84 ha in the main basin and 39 ha in the south basin. The south basin contains relatively small quantity of tailings that are about 30 years old and already underwater. The main basin will be covered with water as a single cell by natural precipitation (Rio Algom 1993).

In 1989, while preparing decommissioning plans for the facility, Rio Algom limited requested CANMET's participation in developing and undertaking a joint study program for evaluating oxidation and leaching characteristics of exposed and submerged tailings and waste rock, limestone neutralization of exposed tailings, and radionuclide leaching characteristics of water covered tailings. The concept of establishing wetland on submerged tailings, characterization of existing wetlands on/or near uranium tailings and their propagation and transplantation studies were also included in the study.

This research program offered a unique opportunity of studying large scale insitu establishment of water cover on reactive acid generating tailings, which was of importance to the mining industry and MEND. Consequently, the present joint research program was initiated in 1989 to evaluate the feasibility of establishing a shallow water cover on pyritic uranium tailings and determine its performance, both under laboratory controlled conditions and in the field. Funding for the program was approved by MEND and shared jointly by CANMET and Rio Algom Limited.

# **1.3** Wet Barriers Research Program

The research program consisted of the following study projects:

- Literature review on the state of the art use of wet barriers and wetlands, both natural and constructed, for controlling acid mine drainage,
- Field evaluations of established wetlands on existing tailings sites and their immediate vicinity in the Elliot Lake area,
- Development and implementation of small scale field test plots to evaluate the suitability of local wetland vegetation species for transplantation and establishment on existing tailings, and determination of a suitable planting methodology,
- Column leaching and limestone neutralization tests to determine the oxidation rate of pyrite and leaching rates of iron, calcium sulphate (gypsum) and radionuclides, both under unsaturated and saturated (submerged) conditions. Evaluation of the effectiveness of adding limestone, or other natural alkaline materials, in varying concentrations and varying screen sizes to neutralize porewater acidity in tailings, produced as a result of pyrite oxidation,
- Column leaching studies of crushed waste rock of different acid generation potentials under unsaturated conditions,
- Diffusion and oxidation studies of underwater disposed un-oxidized (fresh), and oxidized uranium mill tailings,
- Development of a large scale field demonstration and water cover test plot on existing tailings for assessing the role of water cover/wetland in controlling acid generation,
- Hydrological assessment of the field demonstration site for water retention and storage capacity of the tailings material, water balance of the area, storm flow control, and water cover management during operational and draught conditions,
- Review and assessment of various internal dykes and cells designed for water retention capabilities, mechanical stability and acid generation controls,
- Installation of field instrumentation and monitoring of the surface and ground water quality for evaluating the hydro-geochemistry of the field demonstration site and other pertinent field sites before and after the establishment of water covers/wet barriers,

 Compilation of all monitoring data needed to calibrate, confirm and verify various models developed for predicting oxidation of pyrite and leaching of metals and radionuclides from uranium tailings.

CANMET, Elliot Lake laboratory conducted the laboratory experimental at its facility in Elliot Lake, and the field survey and monitoring work was done in collaboration with Rio Algom Limited. A large 65 ha field demonstration and water cover test site for this project was developed by Rio Algom Limited at the Quirke Waste Management Area. The site has been continuously maintained underwater since October 1992 and monitored for its hydrological, meteorological and water quality parameters.

In addition to the above studies, a separate environmental monitoring and survey project consisting of evaluating a natural wetland basin containing partially submerged pyritic uranium tailings (Panel Wetland) was also undertaken by CANMET, Elliot Lake laboratory in collaboration with Rio Algom Limited. Shared funding for this program was provided by the Ontario Ministry of Northern Development and Mines (MNDM).

To-date, the following reports have been prepared and are available for distribution through MEND secretariat and Rio Algom Limited, Elliot Lake:

- Wetlands and Their Role in Treating Acid Mine Drainage A Literature Review (Davè and Lim 1989),
- Panel Wetlands A Case History of Partially Submerged Pyritic Uranium Tailings Under Water (Davé 1993),
- Investigations of the Hydrological Aspects of the Quirke Mine Waste Management Area Decommissioning Plan (Cummings and Cockburn 1991),
- Geotechnical Aspects of Decommissioning the Quirke Mine Waste Management Area (Golder Associates, 1991).

Preliminary results of this study program were reported in 1990 by Davé et al (1990) and more detailed results by Davé and Vivyurka (1993 and 1994). This report provides a comprehensive description of the laboratory lysimeter studies and in-depth analysis of the results obtained.

#### 2. LABORATORY LYSIMETER STUDIES - EXPERIMENTAL METHODOLOGY

The laboratory lysimeter studies were conducted at the CANMET, Elliot Lake Laboratory and consisted of the following separate experimental arrangements:

- Column lysimeter leaching experiments, and
- Diffusion lysimeter transfer flux experiments.

#### 2.1 Objectives and Study Plans

The column lysimeter leaching experiments were undertaken for determining the oxidation and leaching characteristics of unclassified total mill tailings, coarse tailings, and crushed waste rock, all under unsaturated conditions, and submerged coarse tailings with 0.4 m water cover. Additional studies were conducted for the evaluation of neutralization characteristics of commercial agricultural grade limestone of various grain sizes, mixed separately with unclassified total mill tailings and coarse tailings, allowed to weather and leached under unsaturated conditions.

The diffusion lysimeter studies were conducted for determining the diffusion related transfer fluxes of soluble minerals, oxidation reaction products and radionuclides, across the solid liquid interface, from un-oxidized and oxidized tailings deposited under 0.3 m water cover. These studies determined the diffusion controlled surface water quality of underwater deposited tailings in the absence of vertical advection or porewater flow. The un-oxidized tailings had been under a continuous water cover for the past twelve years and the oxidized tailings were freshly

deposited underwater in 1990., both in the laboratory diffusion lysimeters for related previous studies.

The details of the column leaching experiments are discussed in this report and those of the diffusion flux experiments are given separately in an accompanying report (Part III).

# 2.2 Column Lysimeter Leaching Studies

As mentioned previously, column lysimeter leaching tests were conducted to determine oxidation and leaching characteristics of pyritic uranium tailings under unsaturated (control) and submerged conditions, and mine development waste rock. Tests were also conducted for available net neutralization potentials for coarse and fine grained limestone mixed with tailings. The results of this study would be useful in evaluating the effectiveness of various covers, disposal methods and surface amendments in controlling acid generation, and determining the economics of their applications.

For these tests, two types of tailings and three types of waste rock were chosen. The tailings were:

- Un-oxidized total mill tailings (fresh and unclassified), approximately 50% less than 74 mm or-200 mesh, obtained from the tailings filtration circuit of Rio Algom's Quirke mill.
- Un-oxidized (fresh) coarse tailings, 94% greater than 74 μm or +200 mesh, obtained from the Quirke West Arm tailings area.

The waste rock was selected from the Quirke West Arm internal dykes and roadways and classified into the following three acid generation categories:

 High acid generation potential waste rock a having net neutralization potential (NNP) of -17.8 kg CaCO<sub>3</sub>/tonne,

- Medium acid generation potential waste rock having a net neutralization potential (NNP) of -10.7 kg CaCO<sub>3</sub>/tonne, and
- Low acid generation potential waste rock having a net neutralization potential (NNP) of +13.8 kg CaCO<sub>3</sub>/tonne.

The limestone used was of commercial agricultural grade screened to sizes of -6.3 mm (-4 mesh), -2.4 mm (-8 mesh), -0.84 mm (-20 mesh), and wet ground and pulverized.

For tailings, the leaching tests were conducted for both unsaturated and submerged conditions, and the limestone neutralization tests were conducted for the two tailings, each mixed with 7.5% (by weight) limestone of various screen sizes, and under unsaturated conditions as follows:

### **Un-saturated Conditions:**

- Un-oxidized total mill tailings (fresh and unclassified) without additional limestone (Control 1),
- Un-oxidized total mill tailings (fresh and unclassified) mixed with 7.5% (by weight) limestone of screen sizes: -6.3 mm, -2.4 mm, -0.84 m, and wet ground,
- Un-oxidized (fresh) coarse tailings without additional limestone (Control 2),
- Un-oxidized (fresh) coarse tailings mixed with 7.5% (by weight) limestone of screen size 6.3 mm, and wet ground,
- Un-oxidized crushed waste rock (-6.3 mm, 0.25 inch) having the three different acid generation potentials

The limestone, 7.5% by weight, was added in stoichiomatric proportion to the total acid generation potential of tailings containing 5% pyrite.

### Submerged Conditions with 0.45 m Water Cover:

• Un-oxidized (fresh) coarse tailings without additional limestone.

For leaching under submerged conditions, only coarse tailings were chosen for their high oxidation potential when unsaturated and low residual limestone and gypsum contents which allowed study of leaching and dissolution kinetics of Ra-226 and other radionuclides, both before and after gypsum removal.

The laboratory experimental arrangement of submerged tailings was designed to simulate conditions of the field demonstration water cover test site at the Quirke Mine Waste Management area. This test site, designated as Quirke Cell #14, is located at the high elevation western part of the Quirke Tailings basin (Fig. 2) and contained mostly coarse tailings near the surface and along the perimeter dams. The course tailings drained rapidly and are susceptible to oxidation when exposed. Upon submersion, a certain oxidation probability still existed for exposed beaches and uplands during dry summer periods or draught conditions. Also, at this site the water flow is predominantly through the groundwater regime transporting the oxidation reaction products (if any) downstream including dissolved gypsum, metals and radionuclides etc.

### 2.3 Experimental Details

#### 2.3.1 Column Lysimeter - Experimental Arrangement

The leaching experiments were conducted using cylindrical column lysimeters made from schedule 80 grey PVC pipe, of thickness 1.25 cm and nominal diameter 15.2 cm (I.D. 14.5 cm). Each column was approximately 122 cm in height and closed at the bottom end with a PVC end cap that contained, in the middle, a 1.25 cm diameter PVC fitting for drainage via a U - tube. A 60 cm long polypropylene drainage tube was connected to the fitting, and bent appropriately to form a U trap at the bottom and another inverted U at the top, whose height defined the water table elevation in the test sample. Another PVC fitting, 1.9 cm in diameter, was connected approximately 25 cm below the top of the lysimeter for overflow. This arrangement provided a water trap in the effluent drainage line for preventing air entry and waste

oxidation from the bottom. At the bottom, the lysimeter column contained two 6.5 cm thick layers, one each of clean sand and gravel, for filter and bed support purposes. Each column contained approximately 14.3 kg of the sample material which filled it to a height of 35 cm above the filter bed for unclassified total mill tailings, and 45 cm for coarse tailings and waste rock.

Figs. 5 and 6, show, the experimental arrangements of column lysimeters for leaching under unsaturated and submerged conditions, respectively.

In total 30 such columns were filled with tailings or tailings and limestone mixture or waste rock, and grouped into the following four experimental arrangements:

#### Group 1: Un-oxidized Total Mill Tailings (Unclassified), Unsaturated Conditions

Contained five sets, each with three replicates, and consisted of:

- Columns #1-3, containing un-oxidized total mill tailings without additional limestone, Control

   1,
- Columns #4-6, containing un-oxidized total mill tailings mixed with 7.5% (by weight) coarse limestone of screen size -6.3 mm (-4 mesh),
- Columns #7-9, containing un-oxidized total mill tailings mixed with 7.5% (by weight) medium size limestone of screen size -2.4 mm (-8 mesh),
- Columns #10-12, containing un-oxidized total mill tailings mixed with 7.5% (by weight) fine limestone of screen size -0.84 mm (-20 mesh), and
- Columns #13-15, containing un-oxidized total mill tailings mixed with 7.5% (by weight) wet ground limestone.

#### Group 2: Un-oxidized Coarse Tailings, Unsaturated Conditions

Contained three sets, each with two replicates, and consisted of:

- Columns #16 and 17, containing un-oxidized coarse tailings without additional limestone, Control - 2,
- Columns #18 and 19, containing un-oxidized coarse tailings mixed with 7.5% (by weight) coarse limestone of screen size -6.3 mm (-4 mesh), and
- Columns #20 and 21, containing un-oxidized coarse tailings mixed with 7.5% (by weight) wet ground limestone.

# Group 3: Un-oxidized Coarse Tailings, Submerged Conditions

Contained a single set with three replicates and consisted of:

• Columns #22-24, containing un-oxidized coarse tailings without additional limestone and covered with a 0.4 m of water cover.

# Group 4: Un-oxidized Waste Rock, Unsaturated Conditions

Contained three sets, each with two replicates, and consisted of:

- Columns #25-26, containing crushed waste rock (size -2.0 mm) of medium acid generation potential, having a net neutralization potential (NNP) of -10.7 kg CaCO<sub>3</sub>/tonne.
- Columns 27-28, containing crushed waste rock (size -2.0 mm) of high acid generation potential, having a net neutralization potential (NNP) of -17.8 kg CaCO<sub>3</sub>/tonne), and
- Columns 29-30, containing crushed waste rock (size 2.0 mm) of low acid generation potential, having a net neutralization potential (NNP) of + 13.8 kg CaCO<sub>3</sub>/tonne.

The initial set up of columns was followed, in order, by saturated hydraulic conductivity measurements, inoculation of columns with *Thiobacillus ferro-oxidans* culture solution and column leaching experiments.

#### 2.3.2 Sampling Procedure: Tailings and Waste Rock

The required tailings and waste rock samples were collected by personnel from the Research and Development department (during its operational period) of Rio Algom Limited. The following sections detail the sample collection methods for total mill tailings, coarse tailings and waste rock.

#### 2.3.2.1 Unclassified Total Mill Tailings

Representative, fresh (un-oxidized) and unclassified "Total Mill Tailings" samples for these tests were collected daily, in batches over a period of five days, from the solid-liquid filtration circuit of the Quirke Mill. Each day, a total of 21 sample batches, each weighing approximately 5 kg, were collected and arranged as shown in table 1. This sampling procedure was followed to ensure that representative and well mixed tailings samples were obtained, over a reasonable milling period, prior to any segregation of particle size following discharge of tailings.

 Table 1.
 Sample arrangement: batch sampling of total mill tailings from Quirke Mill filtration circuit.

DAY	SAMPLE NO.		
NO.			
1	A1, B1, C1, etc	U1	
2	A2, B2, C2, D2, etc	U2	
3	A3, B3, C3, D3, E3, etc	U3	
4	A4, B4, C4, D4, E4, F4, etc	U4	
5	A5, B5, C5, D5, E5, F5, G5, etc	U5	

A true representative mixture of unclassified total mill tailings containing a broad range of grain size from coarse to fine tailings was achieved by creating a composite sample, such as composite sample #1, which was made by mixing samples A1, A2, A3, A4 and A5, obtained during the five day sampling period.

Similarly, for composite sample #2, batch samples B1, B2, B3, B4 and B5 were mixed. A total of 21 similar composite samples were thus created as unclassified total mill tailings. These samples were first de-watered to remove the remaining liquid, followed by drying in a nitrogen environment to avoid oxidation, packed in plastic bags and transported to the CANMET Laboratory for loading the experimental columns. For the purpose of this report, to simplify the terminology, these tailings are referred throughout as total mill tailings.

# 2.3.2.2 Coarse Tailings

The un-oxidized coarse tailings samples were obtained directly, as field samples, from the West Arm tailings basins of Quirke Waste Management area, near the tailings discharge point. Random grab samples were collected of freshly discharged tailings in a bucket from the surface of the settled tailings in the basin and blended in the field. Multiple sample buckets were collected from the entire sampling area to create 20 random field samples, each containing approximately 10 kg of tailings and bagged individually for transportation to the CANMET Laboratory. Size distribution analysis on a random composite field sample showed that the tailings contained less than 4% (by weight) of size fraction less than 75  $\mu$ m (- 200 Mesh). In this report, these tailings are referred throughout as coarse tailings.

# 2.3.2.3 Waste rock

The waste rock samples were also collected in the field from the West Arm tailings basin of the Quirke Waste Management Area, where the waste rock was used in the construction of internal dykes and access roads. A total of 12 random samples approximately 15-20 kg each, were collected from different locations over a period of two months. The waste rock, originating from the Quirke Mine, was relatively un-weathered, and was sampled during the construction of the internal dyke at Cell #14, the water cover experimental test site.

Individual waste rock samples were crushed, using jaw and roller crushers, to 2 mm diameter particle size (-10 mesh) and a representative sample from each batch was collected. These representative samples were oven dried at 70 °C in dry nitrogen environment (oxygen free),

cooled and pulverized first in a disk grinder followed by a ball mill. The pulverized waste rock samples were analyzed for total sulphur content (acid generation potential) and total available alkalinity, measured as kg CaCO<sub>3</sub>/tonne waste rock, and their net neutralization potentials (NNP) were determined. Based on these results, the waste rock samples were classified into the following three categories:

- Quirke waste rock, sample A (QWR-A), medium acid generation potential, having an average net neutralization potential (NNP) of -10.7 kg CaCO<sub>3</sub>/tonne.
- Quirke waste rock, sample B (QWR-B), high acid generation potential, having an average net neutralization potential (NNP) of -17.8 kg CaCO<sub>3</sub>/tonne, and
- Quirke waste rock, sample C QWR-C), low acid generation (net acid consumption) potential, having an average net neutralization potential (NNP) of +13.8 kg CaCO<sub>3</sub>/tonne.

These samples, labeled as QWR-A, B, and C, were individually bagged and transported to the CANMET laboratory for column leaching experiments.

### 2.3.3 Sample Preparation and Column Loading

The samples were prepared for individual column sets in each experimental group by randomly selecting individual composite samples from each group and mixing them in a cement mixer for one hour. For example, for columns #1-3, (Control - 1) samples, several bags of total mill tailings were selected and mixed to produce a total of 53.5 kg of the sample. The mixed sample was divided into two or three equal portions, depending upon sample replicates, and a 14.3 kg portion of the mixed sample was placed in each column which filled it to a height of 35 to 45 cm above the sand and gravel filter depending upon the particle size of the sample material. Coarse tailings and waste rock samples filled the columns to greater heights than those of total mill tailings which, because of their particle size distribution, compacted more. The excess mixed sample was retained for physical and chemical analyses and as an additional backup sample.

For tailings samples mixed with limestone, 53.5 kg of a given tailings sample were mixed with 4 kg of limestone of certain screen size to produce a homogeneous mixture containing 7.5% of limestone by weight proportions.

All loaded columns were set upright in a wooden support frame, for column permeability and leaching experiments.

#### 2.3.4 Column Permeability Measurements

The saturated hydraulic conductivity or column permeability of each column was measured using a constant head method using the experimental arrangement for submerged tailings as shown in Figure 6.

Initially after loading, all columns were filled with distilled water from the bottom to displace trapped air. Occasionally a rubber mallet was used to tap the columns to enhance the settlement of tailings/waste rock and further release trapped air. The columns were then filled with distilled water up to the overflow level by adding water continuously from the top, allowing the test materials to settle and consolidate further, and the flow was reversed allowing a free drainage from the bottom.

For saturating hydraulic conductivity measurements, distilled water was added at an appropriate constant flow rate at the top of the column for maintaining a constant water head above the test material, and its flow volume (V, ml) in a given time (t, sec) through the column was measured. With a given hydraulic head (h, cm) above the test material of thickness (l, cm) the saturated hydraulic conductivity was calculated using the Darcy's expression for flow through saturated porous media:

$$V = \frac{K * A * (h+l) * t}{l} \qquad eq.(1)$$

Where A is the cross sectional area of the column (cm<sup>2</sup>) and K is the saturated hydraulic conductivity (cm/s), obtained by:

$$\mathbf{K} = \frac{V^*l}{A^*(h+l)^*t} \qquad \text{eq.(2)}$$

Experiments were carried out over several hours (for coarse tailings and waste rock) to several days (for total mill tailings) for collecting a sizable drainage volume from individual columns, and repeated three to four times for measuring variations within the column.

These saturated hydraulic conductivity measurements were for initial or original conditions of tailings and waste rock prior to any alterations in their phase compositions, particle size distributions and porosities as a result of oxidation/reduction and neutralization reactions taking place during the leaching studies. The production of iron hydroxide and gypsum sludge in the limestone neutralization process may decrease the available open pore channels and hence the initial hydraulic conductivity, which may increase again with the removal of gypsum and other soluble minerals from the system.

#### 2.3.5 Leaching Studies: Schedule and Sampling Procedures

At the completion of the column permeability studies, all the columns except columns #22-24, were drained to establish unsaturated hydraulic conditions within test samples by adjusting the phreatic surface (water table) just below the tailings/waste rock and sand/gravel filter interface. For the submerged tailings columns #22-24, the water level was maintained at the overflow mark. All the columns were then inoculated with 100 ml of an acidic leachate, a *Thiobacillus ferro-oxidans* culture solution, obtained from an underground leaching stope of the Quirke Mine, and the columns were allowed to acclimatize for a few days (a week) prior to draining the leaching solution.

Initially for the first year, all columns from #1-21 and #25-30 were batch leached under unsaturated conditions by daily addition of 100 ml of well aerated natural lake water, collected

from Gravel Pit Lake near the West Arm tailings basin of the Quirke Waste Management Area (Figure 2). The water was added, by pouring onto the surface of the test material, for five consecutive days with a two day rest period every week, and collected on a weekly basis. The columns drained freely through the U-tube water trap and the effluents were collected in 11 (litre) polypropylene sample bottles.

The U-tube water trap prevented air entry through the bottom of the column. This eliminated drain and column clogging problems arising from the oxidation of the sample material at the bottom, and precipitation and crystallization of dissolved salt during low or no flow conditions.

As mentioned previously, the water table or phreatic surface in the columns was adjusted near the bottom of the tailings/waste rock at the sand/gravel filter bed interface, which represented a field scenario of severe drought condition, where the water table in the basin would drop 30 to 40 cm below the surface of the waste exposing it to oxidation and leaching.

For each column, the effluent sample was collected every week and its volume and primary water quality parameters: pH, redox potential, Eh, electrical conductance, Ec, told acidity and alkalinity were measured. The effluent sample was then filtered using a 0.45  $\mu$ m membrane filter, and acidified with HCl (20 to 30 ml per litre of sample). For each column, the sample were then individually composited, every two weeks, to constitute a bi-weekly composite sample. The composite samples were then analyzed for total dissolved concentrations of Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Th, U Zn, SO<sub>4</sub><sup>-2</sup> and Ra-226.

During the first year of such a daily leaching scheme, it was observed that most of the tailings and some waste rock columns were retaining appreciable amounts of moisture or were excessively wet near the surface. This condition delayed air entry and establishment of unsaturated conditions within the tailings and waste rock substrates, thereby reducing their chances of air exposure and oxidation. The leaching scheme was then changed, at the end of the first year, to a batch leaching schedule, where 1 l of well aerated natural lake water was added in a single batch every two weeks, allowing the columns to oxidize between rinse cycles. The effluent samples were then collected on a bi-weekly basis and composited every four weeks for analyses as above.

The water covered tailings, in columns #22 to 24, were trickled leached with distilled water under submerged conditions. Distilled water was continuously added at the top of each column at a sufficient flow rate to maintain the water cover, allowing pore water drainage and collection of approximately 1 l of effluent per day. This practice was continued for a period of two years, and afterwards the pore water flow rate was decreased to allow collection of 1 l of effluent per week. The procedure represented accelerated leaching of coarse tailings under submerged conditions, where over a period of four years, approximately 250 porewater volumes were exchanged. This scenario of high porewater flow simulation was followed to rapidly leach and remove soluble minerals like gypsum to enable further studies on oxidation and leaching characteristics of residual minerals and radionuclides, especially Ra-226, post gypsum depletion and removal of

 $SO_4^{-2}$  ion solubility control.

For submerged tailings, the use of distilled water as cover as well as for leaching purposes was preferred over that of the Gravel Pit lake water to provide a continuous source of low background water supply for measuring incremental changes in parameters such as acidity, Fe,  $SO_4^{-2}$  and Ra-226, within the water column above the tailings and in the porewater

Periodically, water samples were also collected from the upper lying water column at near surface, mid depth and near bottom locations. All surface and drainage water samples were analyzed for water quality parameters as described previously.

#### 2.4 Analytical Procedures and Quality Assurance / Quality Control

#### 2.4.1 Water Samples

The water samples were analyzed for primary parameter: pH, Eh, Ec, total acidity and alkalinity, total dissolved concentration of SO<sub>4</sub><sup>-2</sup> and metals Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Th, U, Zn and Ra-226. For most analyses, the Standard methods for the examination of water and waste water (American Public Health Association, 1992) were followed. Dissolved metals/elements concentrations were measured using an Inductively Coupled Argon Plasma Atomic Emission Spectrophotometer (ICAP-AES), and SO<sub>4</sub><sup>-2</sup>, Ra-226 concentrations were determined using radiochemical methods as described below.

#### 2.4.1.1 Primary Parameters

The primary parameters were measured in water samples as collected and prior to their filtration for maintaining sample integrity, without loss of dissolved gases or oxidation of metal ions specially ferrous Fe<sup>+2</sup>. The pH was measured using a standard glass body combination pH probe with a saturated Ag/AgCl reference electrode. The redox-potential, Eh, was measured using platinum and a calomel (Hg/Hg<sub>2</sub>Cl<sub>2</sub>) reference electrodes. The measured Eh was normalized to standard hydrogen electrode reference (NHE) by adding calomel and hydrogen electrodes potential difference of +244 mV. The electrical conductivity, Ec, was measured using a small volume (5 ml) conductivity cell.

All these parameters were measured at the ambient sample temperature (as received ) and normalized to the standard room temperature of 25 °C with an automatic temperature compensator (ATC) or a temperature compensation procedure for Ec measurements.

Total acidity and alkalinity were measured by titrametric methods. The acidity in sample solutions containing hydrolizable metal ions, e.g.  $Fe^{+3}$ ,  $Al^{+3}$ , etc. and polyvalent cations, was measured as a maximum potential acidity. The pH of the sample solution was lowered below 4 by addition of a standard acid solution (H<sub>2</sub>SO<sub>4</sub>) and the reduced ionic species were oxidized

with a few drops of 30% H<sub>2</sub>O<sub>2</sub>. The sample solution was then titrated with a standard alkali (NaOH) solution to pH 8.3 using a potentiometric titration method. The sample acidity was calculated as:

Acidity, as mg CaCO<sub>3</sub>/I = 
$$\frac{[(A * B) - (C * D)] * 50000}{ml_of \_Sample\_Aliquote}$$
 eq.(3)

Where:

- A = Volume of NaOH titrant used to pH 8.3, ml
- B = Normality of NaOH
- $C = Volume of H_2SO_4 to pH 4, ml$
- $D = Normality of H_2SO_4$

The alkalinity was also measured using a potentiometric titration method. An aliquot of sample solution was titrated with a standard acid ( $H_2SO_4$ ) solution to pH 4.5. For solutions containing low alkalinity, additional titrant was carefully added to lower the pH further by exactly 0.3 pH units and the total titrant volume was recorded. The alkalinities were calculated as:

Alkalinity, as mg CaCO<sub>3</sub>/l = 
$$\frac{A * N * 50000}{ml_of_Sample_Aliquote}$$
 eq.(4)

Where A = Volume of standard acid used, ml, and

N = Normally of the standard acid,

and low alkalinity as,

Total alkalinity, as mg CaCO<sub>3</sub>/l = 
$$\frac{(2B - C) * N * 50000}{ml_of\_Sample\_Aliquote}$$
 eq.(5)

Where B = Volume of titrant (standard acid) used to lower pH to 4.5, ml C = Total volume of titrant used to reach pH 0.3 unit lower, ml, and N = Normality of the standard acid.

#### 2.4.1.2 Elemental Analysis

Quantitative analyses of dissolved metals and sulphur in solution samples were performed using a Thermo Jarrel Ash Inductively Coupled Argon Plasma - Atomic Emission Spectrophotometer (ICAP-AES), model Polyscan 61E. Elemental concentrations were measured following appropriate wave length calibration, standardization, matrix matching, inter-element and background corrections, and quality control protocol.

The samples were analyzed in triplicate and the analytical variance among replicates was typically less than 3%. A quality control (QC) sample, having a similar composition and matrix as those of the experimental samples, was also analyzed every ten samples and the system stability was checked by the observed variances in concentrations of various elements in the QC sample. A QC fail command was set at the analytical variance exceeding 5% for a given element, and upon the detection of the fail command the instrument was automatically recalibrated and the analyses repeated. Typical detection limits for most elements, using ICAP-AES, were below 0.05 mg/l.

#### 2.4.1.3 Sulphate Analysis

For solution samples, the sulphate concentrations were measured with a radiochemical tracer technique using a gamma emitter Ba-133 radioisotope.

The method consisted of precipitating sulphate as Ba (Ba-133) SO<sub>4</sub> by the addition of a precipitating solution containing a fixed ratio of BaCl<sub>2</sub>/(Ba-133)Cl<sub>2</sub> concentrations. The sulphate concentration was determined by measuring the recovery of Ba-133 radioisotope using a gamma spectroscopy system. The instrument measured the activity of Ba-133 isotope, using its 356 keV gamma energy peak, with a 10 cm x 10 cm diameter NaI (Tl) detector. By fixing the

concentration ratio of BaCl<sub>2</sub>/(Ba-133)Cl<sub>2</sub>, three precipitating solutions, A, B and C were used to measure total sulphate concentrations in the ranges of 10 - 100 mg, 1 to 10 mg and 0 to 1 mg  $SO_4^{-2}$ , in the sample aliquot, respectively.

This method was preferred over other conventional methods e.g. gravimetric, turbidity etc., for its applicability over a wide concentration range without sample dilution and the ease of sample preparation and counting. The method allowed sample preparation in large batches and counting them at a later date and time without affecting results. For very low sulphate concentrations (<10 mg/l) the above ICAP-AES method was generally used.

#### 2.4.1.4 Ra-226 Analysis

For Ra-226 determination, the alpha spectrometric technique of Lim and Davé (1981) was followed. Radium was chemically separated in a two-step process, first as a radium-barium sulphate using concentrated sulphuric acid and lead sulphate as a carrier, followed by the dissolution of the sulphate precipitate in an ammoniacal solution of EDTA (Ethylene Diamine Tetra Acetic Acid), and selective precipitation of radium from this solution, as a radium-barium sulphate, at pH 4.8 using glacial acetic acid and saturated ammonium sulphate solution.

A known amount of Ba-133 tracer (gamma emitter) was also added to the starting sample for chemical recovery factor determination and correction. The final precipitate was filtered on a 0.45 µm Millipore membrane filter and the concentration of Ra-226 (activity) on the filter was measured by alpha spectrometry using a solid-state silicon surface barrier detector. Ra-226 activity was calculated by measuring total disintegrations/counts for the 4.78 MeV alpha energy decay peak with an energy selection window of 4.0 to 4.9 MeV. The chemical recovery of Ra-226 in the precipitate was calculated by measuring the recovery of Ba-133 tracer using the gamma spectroscopy system as described above.

#### 2.4.1.5 Minimum Detection Limits

For solution samples, the measured instrumental detection limits, defined as  $3\sigma$  where  $\sigma$  is the standard deviation of the instrumental noise at background concentrations, are given in Table 2 for metals,  $SO_4^{-2}$  and Ra-226 for the analytical methods followed.

# 2.4.2 Solid Samples

The tailings and waste rock solid samples were pulverized to less 40  $\mu$ m size and analyzed for solid phase elemental and Ra-226 compositions following acid digestions and complete dissolutions of the samples. The sample solutions were individually brought to a standard volume and were analyzed using the above described procedures.

#### 2.4.2.1 Sample Digestion

For metals analyses, a pulverized tailings or waste rock sample (0.5 - 1.0 g) was completely dissolved, first by digesting it with 20 ml of HNO<sub>3</sub> in 100 ml Teflon beaker and boiling it to dryness, followed by addition of 30 ml of HF and digestion at 100 °C for silica removal. The sample was cooled and digested with 15 ml of a cocktail containing perchloric (HCLO<sub>4</sub>), nitric (HNO<sub>3</sub>) and hydrofluoric (HF) acids in a ratio of 2:1:7, slowly at 110 °C till dryness, followed by baking at 180 °C till all the white fumes of perchloric acid dissociation were expelled. The sample was cooled again and dissolved completely in 50 ml of 50% HCl, by boiling, if necessary. All sample solutions were stabilized by adding HCl to a final concentration of 10% acid. The final volume of the sample solution was brought to 11 (litre) for most metals, and 100 ml for trace metal analysis by adding de-ionized water.

For volatile elements, e.g. Pb, the samples were dissolved with nitric acid (following silica destruction) in high pressure Teflon vessels in a microwave digestion oven. The digested samples were then dissolved in 50% HCl and prepared as above.

For total Ra-226 analysis, the solid sample (0.5-1g) was first digested with hydrofluoric acid in a platinum crucible at a moderate temperature and evaporated to dryness, followed by a dilute hydrochloric acid (HCl) dissolution. The sample/solution mixture was brought to a boil, cooled and filtered. The filtrate was transferred to a pre-acid washed sample bottle. The residue (if any) was heated in a muffle furnace at 550 °C for approximately 12 hours and cooled slowly. The ashed residue was mixed with a sodium carbonate/boric acid flux in the ratio of 1 to 5, heated in the platinum crucible in a high temperature flame until a clear melt was obtained, and then cooled slowly. The mixture was dissolved in 50% HCl as above and the solution was added to the filtrate in the sample bottle. The final sample volume was brought to 11 by adding de-ionized water, if necessary. Prior to digestion, a known amount of Ba-133 radioisotope tracer solution was also added to the solid sample for chemical recovery determination, as mentioned previously.

#### 2.4.2.2 Silica Analysis

For tailings and waste rock solid samples, silica was analyzed gravimetrically by fusion and decomposition of the silicate matrix, and re-crystallization of pure SiO<sub>2</sub>. A portion, (0.5 to 1.0 g), of the finely ground solid sample was fused in a low-form zirconium crucible with sodium peroxide (Na<sub>2</sub>O<sub>2</sub>) and sodium hydroxide (NaOH) crystals (pellets) at a high temperature (650  $^{\circ}$ C) in a muffle furnace. The fused mixture was cooled and digested with 50% HCl, at a low temperature on a hot plate, and evaporated to dryness until all the fumes of HCl ceased. The solids were washed with 50% HCl and filtered on an ashless filter paper. The filter paper, together with the solid residue, was ignited in a muffle furnace at a low temperature to burn the filter paper, then heated to 1000  $^{\circ}$ C for 30 minutes. The crystallized silica residue was cooled in a desiccator for one hour and weighed for silica determination.

# 2.4.2.3 Sulphur Speciation

For solid samples, the sulphur species were determined by measuring concentrations of total sulphur, total sulphate sulphur and obtaining the total sulphide sulphur concentration by difference.

The total sulphur concentration was measured using a Leco furnace sulphur analyzer. A measured amount of dry pulverized sample was ignited at a high temperature in an electric spark furnace and the total sulphur concentration was determined by measuring the total quantity of evolved sulphur dioxide gas using gas chromatography. Occasionally, total sulphur concentration was also determined by complete destruction of the silica matrix and oxidation of sulphides to sulphates either by  $Na_2O_2$  fusion or perchloric acid digestion as described before, followed by measurement of total sulphate concentration in the sample solution by gamma spectroscopy or ICAP-AES.

The total sulphate sulphur concentration of the sample was determined by leaching it with a very dilute solution of hydrochloric acid (1%) to dissolve all water and acid soluble sulphates, and measuring the dissolved sulphate concentration by gamma spectroscopy or ICAP-AES.

#### 2.4.3 Quality Assurance/Quality Control (QA/QC)

The OA/QC program consisted of analyzing 15-20% sample replicates, sample and reagent blanks, and certified reference standards for solid and liquid samples together with an interlaboratory comparison program of unknown and reference samples.

For quality control, the experimental samples were analyzed simultaneously with matrix matched control samples, consisting of Canadian Certified Reference Material Standards (CCRMP) for Uranium, Base Metal Tailings and Ore, and appropriate sample and reagent blanks. All solid samples were digested in duplicates and processed concurrently through the analytical procedures. For dissolved metals, as discussed above, the instrumental analytical standard deviations were set at the 5% variation level of a quality control (QC) sample for an automatic QC pass /fail check and re-standardization, if necessary. The overall precision and accuracy of all measured analytical parameters were between  $\pm 5$  to 10% levels depending on the concentration range. For Ra-226 analysis, certified National Bureau of Standard (NBS) reference materials were used for the QA/QC program, and the typical standard deviations

were less than 10% at low concentrations (~300 mBq/l) and less than 3% at high concentrations (~1000 mBq/l).

Tables 3-5, give the analytical results of certified reference materials consisting of a digested river sediment solution sample, a trace metal drinking water and uranium ore standard samples, respectively, analyzed for the QA/QC program established at the Elliot Lake Laboratory. Tables 5 also gives results of an inter-laboratory comparison program.

#### 3. RESULTS

#### 3.1 Column Permeability Results

Tables 6-9, show the column permeability results for Group 1 - unclassified total mill tailings, Group 2 - coarse tailings, Group 3 - coarse tailings under submerged conditions and Group 4 - crushed waste rock, respectively. The analysis of variance and statistical significance of measured permeabilities within each group are given in Tables 10 and 11. The saturated hydraulic conductivity's were lowest for Group 1 -total mill tailings (mean average 7.03 x  $10^{-6}$  cm/sec), in the range of silty sand to till, and were highest for coarse tailings and crushed waste rock where the measured values were approximately three orders of magnitude higher (in the range of ~ 2 - 4 x  $10^{-3}$  cm/sec). For both coarse tailings and crushed waste rock, the measured hydraulic conductivity values were in the coarse sand fraction range, representing good drainage and high air entry values. Within each group, except Group 3 - submerged coarse tailings, the results were significance. For the submerged coarse tailings group, where the three columns contained same tailings, no significant variation in the measured hydraulic conductivity was observed.

For Groups 1 and 2, the variations were significant (marginally at 10%) amongst tailings without and with limestone. In general, the conductivity decreased with the addition of limestone as well as with its decreasing particle size, the finer the limestone the smaller was the observed conductivity. Although, the observed differences were marginal between limestone groups, the results showed the general trend that addition of finely ground limestone to tailings decreased their saturated hydraulic conductivity.

Similarly for crushed waste rock, the observed hydraulic conductivities were lower (8 x  $10^{-4}$  cm/s), by factors of 3 to 4, for low or non-acid producing waste rock (Quirke waste rock 'C', NNP = +13.8 kg CaCO<sub>3</sub>/tonne, columns #29 and 30), than those for acid producing waste rock (columns #25-28), where the measured values were in the range of 2.9 -3.3 x  $10^{-3}$  cm/s. Quirke waste rock 'C' contained approximately 0.42% total S and 26.82 kg CaCO<sub>3</sub> /tonne alkalinity, mostly on CaCO<sub>3</sub> or limestone (see table 18). Upon crushing, perhaps the limestone fraction of that waste rock produced fine grind leading to observed differences in the saturated hydraulic conductivity.

The saturated hydraulic conductivity, moisture retention and drainage characteristics, and air entry parameters are intrinsically dependent on the particle size distribution of the porous media. These parameters impact upon the oxidation, acid generation and neutralization, and contaminants migration characteristics of the waste material. Low hydraulic conductivity and poor drainage contribute to increased water retention, resulting in low oxidation and acid generation rates, and increased contact time for limestone neutralization and hence enhanced neutralization efficiency. Thus for similar acid generation potentials and weathering conditions, coarse tailings and waste rock are more amenable to oxidation and acidic drainage than total mill tailings.

Based on the present column permeability results, it could be predicted with a reasonable certainty that coarse tailings from the Quirke Mine Waste Management area, if allowed to oxidize, would contribute approximately 1000 times more to the acidic effluent than un-

segregated total mill tailings of the same mass and under similar conditions. This prediction will further be examined when discussing column leaching results later in this report.

#### **3.2** Solid Phase Composition

The solid phase compositions of total mill tailings (Control - 1, column #1-3), total mill tailings mixed with 7.5% limestone by weight (columns #4-15), coarse tailings (Control - 2, columns #16 and 17, and columns #22-24), coarse tailings mixed with 7.5% limestone by weight (columns #18-21), medium acid generation potential waste rock (QWR-A, columns #25 and 26), high acid generation potential waste rock(QWR-B, columns #27 and 28), and low acid generation potential waste rock (OWR-C, columns #29 and 30) are given in Tables 12-18, respectively.

The data showed that both the tailings without additional limestone, total mill tailings and coarse tailings were acid generating and the acid generation potential for coarse tailings (NNP =  $-129.2 \text{ kg CaCO}_3$ /tonne) was almost twice that for total mill tailings (NNP =  $-61.41 \text{ kg CaCO}_3$ /tonne). Coarse tailings mixed with 7.5% limestone as well as waste rock QWR-A and QWR-B was also acid generating with NNP of  $-42.8 \text{ kg CaCO}_3$ /tonne,  $-10.7 \text{ and } -17.8 \text{ kg CaCO}_3$ /tonne, respectively. Total mill tailings mixed with 7.5% limestone and Quirke Mine Waste Rock QWR-C were non acid generating having NNP of +20.44 and  $+13.81 \text{ kg CaCO}_3$ /tonne, respectively.

The coarse tailings were distinctly characterized by their high sulphide sulphur (4.22%) and iron (3.99%) contents, and low calcium (0.4%), Mg (0.06%), Ba (0.078%), Pb (0.021%), Th (0.02%) and total Ra-226 (3900 mBq/g) concentrations. The corresponding parameters for total mill tailings were, total sulphide sulphur (2.17%), iron (2.49%), Ca (1.71%), Mg (0.085%), Ba (0.1%), Pb (0.035%), Th (0.031%) and Ra-226 (7500 mBq/g) Most of Ca (as gypsum, CaSO<sub>4</sub>), Mg etc. and Ra-226 were associated with the fine fraction of the tailings. Because of their almost double sulphide (and hence pyrite) contents, the coarse tailings were

potentially two times more acid generating than total mill tailings as the residual limestone alkalinities in both the tailings were small,  $\sim 2.7$  to 6.4 kg CaCO<sub>3</sub>/tonne.

The addition of 7.5% limestone by weight to both the tailings was based on the stoichiometric limestone neutralization of total potential acidity produced by tailings containing on the average 5% pyrite (~2.5% sulphur as S, total acid generation potential 78.125 kg CaCO<sub>3</sub>/tonne or ~7.8%). These experiments were originally planned using total mill tailings, where the added limestone was sufficient for the neutralization requirements. The coarse tailings experiments were added later to establish drainage conditions similar to that of the experimental water cover Cell #14 at the Quirke Waste Management Area, which contained a significant proportion of coarse tailings in the upper most horizon. In the actual cell, the limestone was to be incorporated in the near surface layer of the tailings, up to a depth of 15 cm and at a rate of 200 tonne/ha. Thus it was considered appropriate to add the same amount of limestone to the experimental tailings as well. The field limestone application rate of 200 tonnes/ha roughly corresponded to 7.5% limestone by weight for coarse tailings having a dry bulk density of 1.8 tonne/m<sup>3</sup>. The coarse tailings limestone mixtures were thus still potentially acid generating, but were representative of field conditions for both limestone neutralization and water cover studies as stated previously in the objectives of the program.

Compared to tailings, the three types of waste rock were high in Al (4-5%), Ba (0.16-0.2%), Mg (0.16 - 0.5%), Mn (90 - 490 ppm), and U (0.005-0.008%); variable concentrations of Fe (1.5-3.8%), Ca (0.2-1.0%), Th (0.01-0.025%) and Ti (0.16-0.42%); and low concentrations of total sulphur (0.42 - 0.86%), Pb (0.006-0.01%) and Ra-226 (1750-2770 mBq/g). Waste rock (QWR-C) contained higher concentrations of Fe, Ca, Mn and Zn, etc. than those of QWR-A and QWR-B. The mineralogy of the waste rock was not examined in this study.

The waste rock was primary used, below the projected water level, in the construction of internal dykes and dams for water retention purposes. Periodically, the rock may be subjected to exposure and hence oxidation, if the water levels were to fluctuate, contributing to acid

generation and related impact on the water quality. The leaching characteristics of the waste rock were thus only studied under unsaturated conditions.

#### 3.3 Column Leaching Results

The column leaching results for total mill tailings (Control - 1, columns #1-3) are given in Appendix A. Tables A-1 to A-3, give respectively, the leachate water quality results for primary parameters and ionic concentrations (e.g. mg/l for metals), normalized loading and removal rates ( $\mu$ g/kg per day) for metals, SO<sub>4</sub><sup>-2</sup> and Ra-226, and total cumulative (as %) productions/removals (loadings) of various species in the effluents. Figures A-1 to A-16, show variations in the effluent water quality as a function of time for pH, Eh, Ec, total acidity, total alkalinity, sulphate, total iron, Ca, Mg, Al, Mn, U, Th, Pb, Ra-226 and total cumulative drainage flow volume. The corresponding tables and figures for total mill tailings mixed with 7.5% (w/w) limestone of screen sizes coarse (-6.3 mm size, columns #4-6), medium (-2.4 mm size, columns #7-9), fine (-0.84 mm size, columns #10-12) and pulverized wet ground limestone (columns #13-15) are given in appendices B-E, respectively; for coarse tailings without limestone (Control #2, columns #16 and 17), coarse tailings mixed with 7.5% (w/w) limestone of screen sizes, coarse (-6.3 mm size, columns #18 and 19) and pulverized wet ground limestone (columns #20 and 21) are given in Appendices F-H respectively; for coarse tailings under submerged conditions (columns #22- 24) in appendix I; and for the three types of waste rock having medium (QWR-A, columns #25 and 26), high (QWR-B, columns #27 and 28) and low (QWR-C, columns #29 and 30) acid generation potentials are given, respectively, in appendices J-L.

These results are further detailed and discussed according to leaching conditions for the various tailings and waste rock categories based on the group classification system described earlier in section 2.3.1.

#### **3.3.1 Unsaturated Tailings**

Initially for the first year, when the leaching scheme consisted of daily additions of 100 ml of natural lake water to the columns, no acidic drainage was observed from any of the columns including those containing waste rock. Although columns #16-21 contained coarse tailings and columns #25-30, crushed waste rock (screen size -2.0 mm), they appeared to retain appreciable amounts of moisture, in the laboratory experimental arrangement, to maintain near saturation conditions due to a shallow water table, approximately 35-40 cm below the surface, and frequent water additions. Because the objective of the study was to determine the oxidation, leaching and limestone neutralization characteristics of tailings and waste rock under different conditions, the leaching scheme for all unsaturated columns was modified after the first year, in June 1990, to batch additions of 1 l of natural lake water every two weeks for flushing and rinsing purposes. In the intermediate period, the test samples weathered and oxidized at room temperature under unsaturated conditions.

With the implementation of this batch leaching scheme, both control tailings without limestone, Control - 1, total mill tailings (columns #1-3) and Control #2, coarse tailings (columns #16 and 17), as well as coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, columns #18 and 19), all started to oxidize produced acidic drainages. The onset of the acidic drainage and its quality depended upon the nature of the tailings material and limestone amendments. No acidic drainage was observed from other tailings and limestone mixtures. The detailed results are as follows:

#### **3.3.1.1** Group 1: Total Mill Tailings

# 3.3.1.1.1 Group 1: Control - 1, Total Mill Tailings Without Additional Limestone, (Columns #1 - 3)

The total mill tailings (Control - 1) columns started to produce moderately acidic drainage six months to one year after the introduction of the batch leaching scheme, with column #2

producing acidic effluent approximately six months before Columns 1 and 3. Figures 7-31 show the oxidation and leaching profile for Control - 1 tailings, respectively, for pH, Eh, Ec, total acidity and alkalinity, effluent loading rates for SO<sub>4</sub><sup>-2</sup>, Fe, Ca, Mg, Al, Mn, U, Th, Pb and Ra-226, and cumulative productions/removals (loadings) for total acidity, SO<sub>4</sub><sup>-2</sup>, Fe, Ca, Mg, Al, Mn, Th and Ra-226, and total accumulated effluent drainage volume from April 1989 to June 1993. Except for effluent pH, Eh, and Ec, all other parameters were normalized to the effluent volume collected for a given sampling period and converted to ionic mass loading rate per unit mass of tailings per day (e.g. for total Fe as µg total Fe/kg tailings per day) and cumulated mass (%) production/removal (loading) as:

Ionic loading rate, (µg ion/kg tailings/per day) = 
$$\frac{1000 * Ci * V}{M * T}$$
 eq. (6)

Where, Ci = Ionic concentration in the effluent, mg/l,

V = Effluent volume collected during sampling period, l

M = Total mass of tailings, kg, and

T =Sampling period, d.

and

Cumulative mass (%) production/removal = 
$$\frac{\sum_{i=0}^{n} (Ci * V)}{Mi * 10}$$
 eq.(7)

where, n = Number of sampling periods,

Ci = Ionic concentration in the effluent, mg/l

V = Effluent volume collected during sampling period, l

Mi = Total ionic mass content of the tailings sample, g, and

T =Sampling period, d.

For data interpretation and understanding of various geochemical interactions within the tailings/waste rock mass, the term oxidation is used for the oxidation process in sulphide minerals producing sulphuric acid and dissolved metal sulphates irrespective of a partial or complete acid neutralization process and precipitation of gypsum and metal hydroxides. The

term acidic drainage is used when release of acid and oxidation reaction products, so produced, occur in the effluent. Thus the tailings or waste rock may be oxidizing at the exposed upper surface in the columns, but the occurrence of acid drainage may be absent or delayed depending upon the neutralizing conditions. Also, acidic drainage is further classified, for comparison purposes, as high when pH<2.5, acidity >1500 mg/l, SO<sub>4</sub><sup>-2</sup> > 3000 mg/l and Fe > 1000 mg/l, moderate for pH's between 2.5 -4, acidity 100-1500 mg/l, SO<sub>4</sub><sup>-2</sup> 1500-3000 mg/l and Fe between 100-1000 mg/l, and low when pH is between 4-6, acidity < 100 mg/l SO<sub>4</sub><sup>-2</sup> < 1500 mg/l, and Fe < 100 mg/l. Because the effluent is usually saturated with gypsum, no distinction is made based on Ca, Mg or Ra-226 concentrations. However, all data showed consistent trends of elevated Mg concentrations at the onset of oxidation and acid neutralization, and elevated concentrations of Fe, SO<sub>4</sub><sup>-2</sup> in accordance with the effluent Eh-pH phase stability relationships (see for example Figures 15-24). This classification terminology is frequently used in this report when interpreting column leaching results.

As mentioned previously, the total mill tailings (Control - 1 columns #1-3) produced moderately acidic drainage with the following characteristic leaching profiles:

#### pH, Eh and Ec:

During the first year, the effluent pH's were near neutral to slightly alkaline in the range of ~ 7-8, dropping to between 2-3 subsequently (Figure 7), with column #2 leading in all parametric changes. Acidic drainage started with column #2, approximately six months after the implementation of batch leaching, followed by columns #3 and #1 within a six month period (January-June, 1991). Near the completion of the study period, the column average pH was approximately 3. During the entire leaching period, a cumulative total of approximately 80 l of drainage effluent was collected.

The oxidation - reduction potential (ORP) or Eh (normalized to hydrogen electrode reference, NHE) increased during the same period from approximately 300 mV to 700 mV at the

occurrence of the acidic drainage (Fig. 8). It should be mentioned that Eh was measured in effluent samples, collected over a two week period in open sample bottles, that were well oxygenated. Nevertheless, Eh was a good indicator of the oxidation stages of iron ( $Fe^{+2}$  and  $Fe^{+3}$ ) whenever oxidation and acidic drainage occurred.  $Fe^{+2}$  in these samples was usually not measured because of its oxidation during and after sample collection.

The electrical conductance, Ec, for Control - 1 tailings increased slightly from approximately 2000  $\mu$ s to 3000  $\mu$ s during a six month period of peak acidic drainage and decreased afterwards to approximately 2300  $\mu$ s where it was controlled by the dissolution of gypsum (CaSO<sub>4</sub>) (see for example Figures 9 and 17). The peak in Ec was associated with increased acid generation and iron mobility but at moderate rates to impact Ec significantly.

#### Acidity and Alkalinity:

Figures A-4 and A-5 show, respectively, the total acidity and alkalinity concentration profiles for Control - 1 tailings. The acidity loading rate, total cumulative acid production and alkalinity loading rate profiles are shown in figures 10-12, respectively. The results show very illustratively the commencement of acidic drainage and termination of residual alkalinity in the effluent at the beginning of 1991, again with column #2 leading in acidic drainage, followed by columns #3 and #1, in that order.

The column average acidity initially increased with time and peaked with a maximum acidity of approximately 1500 mg CaCO<sub>3</sub>/l for a period of six months, afterwards decreasing slowly to a level of 500-600 mg CaCO<sub>3</sub>/l acidity. During the remaining 2.5 year study period when acidic drainage was occurring, the mean average column acidity generation rate was calculated as  $2100 \ \mu g$  CaCO<sub>3</sub> acidity/kg of tailings/day. During the same period, a cumulative total of ~ 3% of the maximum potential acidity was produced/removed from the Control - 1 tailings. For comparison, the average acid generation rate and cumulative acid production for coarse tailings (Control #2, columns #16 and 17), reported later in this report, were: 70,000  $\mu g$  CaCO<sub>3</sub>

acidity/kg tailings/day and 58% of the maximum acidity generation potential of the tailings, respectively. The total mill tailings, because of their homogeneous particle size distribution, were approximately 30 times less acid generating than coarse tailings.

Prior to the onset of the acidic drainage, the column average alkalinity of the effluent was approximately 25 mg/l corresponding to a daily average alkalinity loading rate of approximately  $100 \mu g \text{ CaCO}_3$  alkalinity/kg of tailings/day.

# Sulphate and Iron:

The dissolved  $SO_4^{-2}$  and total iron concentration profiles for Control - 1 total mill tailings are shown in Figures A-6 and A-7, respectively. Figures 13-16, show respectively,  $SO_4^{-2}$  loading rate, cumulative  $SO_4^{-2}$  production, total iron loading rate and cumulative total iron production characteristics.

Similar to Ec, the effluent SO<sub>4</sub><sup>-2</sup> concentrations were controlled by gypsum dissolution and there was a slight increase in dissolved  $SO_4^{-2}$  concentration near the peak acidic drainage period (Figure A-6). The change was again most pronounced and occurred first in column #2. The column average  $SO_4^{-2}$  loading or production rate increased from approximately 500 µg  $SO_4^{-2}/kg$ tailings/day to 6000  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day with the acidic drainage (Figure 13), having a column mean average SO<sub>4</sub> loading rate of 5500  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day for the entire four year leaching period. During that period, approximately 8.4 g SO<sub>4</sub><sup>-2</sup>/kg tailings of total sulphur as SO<sub>4</sub><sup>-2</sup> or 8% of the total sulphur contained in the tailings mass was leached and removed, most of it resulting from the dissolution of gypsum associated with the tailings. This total sulphate production represented approximately 21% of the soluble  $SO_4^{-2}$  content of the tailings.

Prior to the onset of acidic drainage, the dissolved iron concentrations were very low (below detection) which increased rapidly to 100-150 mg/l column average range during the maximum acid generation period, and decreased to 20-30 mg/l range thereafter. The column average iron

loading rate during the same period was approximately 100  $\mu$ g Fe/kg tailings/day with a maximum production rate of 400  $\mu$ g Fe/kg tailings/day. During the acidic drainage period, approximately 0.082 g of Fe/kg tailings or 0.33% of the total iron contained in the tailings was removed. Near the end of the study period, the iron production/removal rates from all the three columns were low, ~ 15  $\mu$ g Fe/kg tailings/day for the three columns.

## **Calcium and Magnesium:**

Figs. A-8 and A-9 show, respectively, dissolved calcium and magnesium concentration profiles, and figures 17-20, show calcium loading, cumulative calcium removal, magnesium loading and cumulative magnesium removal characteristics, respectively, for the total mine tailings. Similar to Ec and dissolved SO<sub>4</sub><sup>-2</sup>, the dissolved calcium concentration of the drainage effluent was controlled by gypsum dissolution. The effluent was saturated with respected to gypsum (~ 550 mg/l Ca) for the initial two year period, decreasing slowly to column average concentration range of 300-400 mg Ca/l. During the peak acidic drainage period, no noticeable increase in calcium concentration was observed because of gypsum saturation. The column average calcium loading or removal rate was approximately 1500 μg Ca/kg tailings/day. During the entire study period, approximately 2.33 g Ca/kg tailings or 13.6% of the total calcium contained in the tailings was removed.

The effluent magnesium concentration increased slowly from an average of 3 mg/l to approximately 30-40 mg/l with oxidation and acid generation. As mentioned previously, dissolved Mg concentration in the effluent was a good indicator of the initiation of oxidation/acid generation and neutralization processes as the solubilities of MgSO<sub>4</sub> and MgSO<sub>4</sub>.7H<sub>2</sub>O were high, approximately 260 g/l and 710 g/l, respectively. In Column #2, for example, the Mg concentration in the drainage effluent started to increase slowly at least four to six months prior to the occurrence of the acidic drainage (Figures A-9 and 19). The column average magnesium loading was in the range of 100-150  $\mu$ g/kg tailings per day in the acidic effluent. The observed Mg concentrations and loading were fairly uniform over a period of approximately two years, which suggested ongoing neutralization process. During the entire oxidation/acid generation

period, approximately 0.12 g of Mg per kg of tailings or 14% of the total amount of Mg contained in the tailings was removed. The total amounts of Ca and Mg removed, as a percentages of the tailings contents, were comparable.

Initially for the first 2 to 3 weeks trial period, columns #1-15 were washed daily with 200 ml of natural lake water (1 l per week) instead of 100 ml used afterwards, which resulted in increased loading and removal of Ca, Mg and  $SO_4^{-2}$ , as seen from Figures 13, 17 and 19.

### Aluminum and Manganese:

Figures A-10 and A-11 show, respectively, the aluminum and manganese concentration profiles, and Figures 21-24, show respectively, loading and cumulative removal characteristics for aluminum and manganese. These profiles were similar to those for column acidity and magnesium, where both concentrations and loading increased with acid generation and drainage.

In the acidic effluent, the column average Al concentration and loading were in the range of 60 or 80 mg/l, and 200-250  $\mu$ g Al /kg tailings/day, respectively. During the total acidic drainage period, approximately 0.17g Al /kg tailings or 0.6% of the total amount of aluminum contained in the tailings was only removed. The cumulative percentile removal of Al and Fe were comparable.

The column average Mn concentration and loading rate were in ranges of 2-3 mg/l, and 5-10  $\mu$ g/kg tailings/day, respectively. During the acidic drainage period monitored, approximately 6.4 mg of Mn/kg of tailings or 27% of total Mn contained in the tailings was removed. The cumulative removal of manganese was thus significantly greater than those of Al and Fe.

## Uranium, Thorium and Lead:

The leaching characteristics of uranium, thorium and lead are shown in Figures A-12 to A-13, respectively, for effluent concentrations, in Figures 25, 26 and 28, respectively, the loading characteristics of U, Th and Pb, and in Figure 27, the cumulative production/ removal characteristics of Th.

The uranium concentrations in the effluents were near detection limit of ~0.2 mg/l and no significant changes were observed with the acidic drainage. Because of such low uranium concentrations, the calculated loading rates were also very low, ~  $0.75 \mu g$  U/kg tailings/day, and the data were not processed further in calculating cumulative uranium production/removal.

The column average effluent concentrations of Th showed similar characteristics to that of total iron, increasing from very low (below detection) levels to peak concentrations of approximately 6-8 mg/l with the onset of acidic drainage and decreasing slowly afterwards to 2-4 mg/l range. The columns average Th loading rate was approximately 15 µg Th/per kg tailings/day during the acidic drainage period. Within the same interval, approximately 13.7 mg of Th per kg of tailings, or 4.4% of total thorium contained in the tailings was removed.

The effluent lead concentration and loading profiles were similar to those of Al and Mn, increasing from very low (below detection) to approximately 2 mg/l and 5  $\mu$ g Th/kg tailings/day with the occurrence of acidic drainage. During the acidic drainage period, Pb production/removal was calculated as approximately 4.6 mg Pb/kg tailings or 1.5% of total Pb contained in the tailings.

#### Other Metals, Ce, Cu, Ni and Zn:

The effluent concentrations and leaching profiles were measured for Ce, Cu, Ni and Zn but because of their low concentrations in the tailings mass, the data were not processed in greater details. Tables A-1 and A-2, together with other parameters, give the concentration and loading results for these metals. The general leaching characteristics of these metals were also very similar to those of Fe and Al, where the concentrations and effluent metal loadings

increased with the onset of acidic drainage, peaked, and decreased slowly afterwards to low but detectable levels.

# <u>Ra-226:</u>

The Ra-226 leaching characteristics of Control - 1 tailings are shown in Figure A-15 for concentration (activity), and in Figures 29 and 30, respectively, for effluent loading rate and cumulative Ra production/removal.

The data, both for effluent Ra-226 concentration and loading, showed very interesting characteristics of increased Ra-226 leaching at the initiation of the batch leaching schedule, where the parameters increased by 4 to 5 folds, peaked individually for each column and then decreased slowly (see Figure A-15 and 29). The initial column average Ra-226 concentration was in the range of 200-500 mBq/l, with oxidation and acid generation it increased to a maximum of 3000-3500 mBq/l (average ~ 1800 mBq/l) and decreased slowly to 500-1000 mBq/l range. Unlike the acid generation order of the columns, increased Ra-226 drainage started with column #1, followed by columns #2 and #3, in that order (Figure 29).

Similar to the effluent concentration profiles, the column average Ra-226 loading increased from a low of approximately 1.5 mBq/kg tailings per day to a maximum of 10 mBq/kg tailings/day (average ~ 6 mBq/kg tailings/day), then decreased slowly to 2-4 mBq/kg tailings/day range. Similar to Ca, Mg and  $SO_4^{-2}$ , Ra-226 loading were also high for the first few weeks when the columns were flushed with double the volume of natural lake water (200 ml).

During the entire study period, approximately 6200 mBq Ra/kg tailings or only 0.08% of total Ra-226 contained in the total mill tailings was removed, most of it resulting from oxidation and acidic drainage.

The results also showed that for total mill tailings, the dissolved Ra-226 concentrations in the effluent did not correlate with those of Ca and  $SO_4^{-2}$ , but with the oxidation/acid generation

reaction products. These observations will be further examined for other tailings and waste rock, and for different leaching conditions.

# 3.3.1.1.2 Group 1: Total Mill Tailings mixed with 7.5% Limestone (w/w) of Various Grain Sizes (Columns #4 - #15)

The leaching characteristics of columns #4-15, containing total mill tailings mixed with 7.5% (w/w) limestone of various screen sizes, from coarse (-6.3 mm) to wet ground and pulverized, were generally similar in trends for individual parameters in each limestone size category. During the entire study period, of approximately four years, none of these columns produced any acidic drainage.

During the monitoring period, each column received an average of 114 l of natural lake water for leaching/flushing purposes, producing a cumulative column average drainage volume of approximately 73 l, and the balance resulting in evaporation. This drainage volume represented an infiltration depth equivalent of approximately 1084 mm of annual precipitation.

Figures B-1 to B-16 (Appendix B) show respectively, the drainage effluent pH, Eh, Ec, total acidity, total alkalinity, and concentrations of dissolved SO<sub>4</sub><sup>-2</sup>, Fe, Ca, Mg, Al, Mn, U, Th, Pb, Ra-226 and cumulative effluent volume, for total mill tailings mixed with 7.5% coarse limestone (screen size -6.3 mm, columns #4-6). The corresponding parameters, expressed as effluent loading rates and cumulative mass productions/removals, for columns #4-6 are shown in Figures 32-51. Because of the absence of acidic drainage, the effluent contained only appreciable amounts of SO<sub>4</sub><sup>-2</sup>, Ca, Mg and Ra-226. Thus the cumulative mass removal characteristics are only shown for these parameters in Figures 38, 41, 43, and 50, following the corresponding loading rate figures, respectively.

Similarly, Figures C-1 to C-16 (Appendix C) and 52-71, show the corresponding parameters for total mill tailings mixed with 7.5% medium size limestone (-2.4 mm size, columns #7-9), Figures D-1 to D-16 (Appendix D) and 72-91, for fine size limestone

(-0.84 mm size, columns #10 -12), and Figures E-1 to E-16 (Appendix E) and 92-111, for wet ground and pulverized limestone (columns #13-15), respectively.

The leaching characteristics of all the above sub-groups, each group belonging to a different limestone screen size category (e.g. columns #4-6 for coarse limestone), were similar for individual parameters measured as seen from the respective figures for each category. No acidic drainage was observed from any of the above sub-groups, as the effluent pH was neither acidic nor it contained dissolved iron or other metals, mobilized as a result of oxidation and acid generation (Figures B-7, C-7, D-7, E-7). The results were thus grouped together and are presented ensemble.

#### pH, Eh and Ec:

For all columns, the initial effluent pH was near neutral to slightly alkaline, in the range of ~ 7-8, which decreased slowly during the first year to pH ~6, and increased gradually afterward to pH ~8 (Figures 32, 52, 72 and 92). In few instances, some columns and particularly columns #8 and #11, produced low pH, ~ 4-5, effluents for a period of 3 to 4 months (Figure 52 and 72), followed by a gradual increase in pH to near neutral. This occurrence was limited to the initial one and a half year period when the columns were mostly rinsed/flushed daily with 100 ml of natural lake water. During that period, it was observed that these two columns (#8 and #11) were draining very slowly, occasionally forming a permanent water pool on top of the tailings. Usually a vacuum suction was applied at the drainage tube to induce column drainage for sampling and to remove any blockage in the flow path. This procedure alleviated the problem but introduced additional air within the tailings, probably creating some channeling and limited oxidation along the flow paths. After the introduction of the batch leaching schedule, no such problems were encountered.

Not withstanding, the occasional drop in pH in few columns, generally the pH increased with time in all columns, and after the second year in the study, the drainages were slightly alkaline having pH's on the order of ~ 8, irrespective of the particle size of the added limestone.

The effluent redox potential, Eh, increased with time steadily from an average of 300 mV at the beginning to a range of ~ 500-600 mV with the implementation of the batch leaching schedule. These Eh's were, however, lower than those for columns #1-3, which were in the range of ~ 700-800 mV and where active oxidation and acid drainage were taking place.

The electrical conductance, Ec, of the effluent decreased initially with time during the first year, from approximately 3000-3500  $\mu$ S/cm to 2000-2200  $\mu$ S/cm, and remained constant afterwards it at the latter value . These values were similar to those for columns #1-3, and no effects of limestone addition or its size were observed on the electrical conductance. Ec was primarily controlled by gypsum dissolution and its nearly saturated concentration.

#### Acidity and Alkalinity:

Initially for the first two years, the effluents from all columns containing limestone had an average total acidity in the range of 20-30 mg/l, which peaked for some columns to 50-80 mg/l during the first year, and decreased gradually to zero afterwards, (Figures B - 4, to C, D, and E - 4). After approximately two years, the columns were producing slightly alkaline effluents having total alkalinities in the range of ~50-60 mg/l. For columns #4-6 containing coarse limestone, the alkalinity values peaked to approximately 100-120 mg/l range, where as in the other column sub-groups, the total alkalinities gradually increased with time (Figures B - 5 to C, D, E -5).

The increased alkalinity was also associated with the corresponding increase in Ca and Mg concentrations, probably as a result of oxidation, acid generation and neutralizing processes within the tailings-limestone mixtures, thereby introducing additional carbonate/bi-carbonate alkalinity to the effluents. This phenomenon was more pronounced for columns #4-6, where alkalinity generation rates were observed higher at ~ 300-400  $\mu$ g CaCO<sub>3</sub>/kg tailings/day than those for other columns at ~ 100-200  $\mu$ g CaCO<sub>3</sub>/kg tailings/day, as seen from Figures 36,56,76, and 96. In total mill tailings without limestone amendment (Control -1 columns #1-3),

such an initial alkalinity was rapidly consumed with the production of acidic conditions in the tailings.

The column average acid generation or loading rates for all columns were low, approximately  $50-60 \ \mu g \ CaCO_3$  acidity/kg of tailings/day, approximately corresponding to  $36 \ mg \ CaCO_3$ /per kg of total acidity production/removal during the entire leaching study. This value is to be compared with the total acid generation potential of  $61.9 \ g/kg$  tailings (kg  $CaCO_3$ /tonne tailings).

### **Sulphate and Iron:**

The sulphate concentration profiles for columns #4-15 are shown in Figures B-6, C-6, D-6, and E-6, and effluent loading rate and cumulative removal in Figures 37, 38, 57, 58, 77, 78, 97, and 98, respectively, for the four sub-groups containing various mixtures of total mill tailings and limestone. For all columns, the effluent sulphate concentrations were nearly constant in the range of 1200-1600 mg/l, and were controlled by the solubility of gypsum contained/produced in the tailings. The column average sulphate removal/loading rates were approximately 5000  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day throughout the experimental period. During that time, approximately 7.8 g of SO<sub>4</sub><sup>-2</sup> per kg of tailings or a column average of ~ 8.6% of total sulphur (as SO<sub>4</sub><sup>-2)</sup> contained in the tailings limestone mixture was removed. This cumulative sulphate loading was comparable to that for the Control-1 tailings.

The iron concentration and effluent loading profiles are shown in figures B-7, C-7, D-7 and E-7 and 39, 59, 79 and 99, respectively, for the four different groups of columns. As mentioned previously, the dissolved iron concentrations in the effluents were very low (below detection) to 0.2 mg/l range, except for occasional high values of ~ 0.5 - 0.6 mg/l for Column #11 for reasons mentioned above. The column average iron loading rates were practically zero (less than 0.05  $\mu$ g Fe/kg tailings/day). The effluents from these columns were thus classified as non acidic drainage derived.

#### **Calcium and Magnesium:**

Figures B-8 and 9 to C, D and E-8 and 9, show respectively, the effluent concentration profiles for calcium and magnesium for the four sub-groups. The corresponding loading rate and cumulative mass production/removal profiles for Ca and Mg are shown in Figures 40-43, 60-63, 80-63 and 100-103, for the four groups of columns, respectively.

The column average calcium concentrations, similar to sulphate, were nearly constant for the first three years, in the range of ~ 500 - 600 mg/l, where the were saturated with respect to gypsum. In the fourth year, Ca concentrations decreased to approximately 400-500 mg/l range. The corresponding calcium loading rates were in the range of ~ 1500-2000  $\mu$ g Ca/kg tailings/day, having a column average loading rate of approximately 1800  $\mu$ g Ca/kg tailings/day for all column sub-groups. During the four year study period, an average of approximately 2.8 g Ca/kg tailings or 5.7% of total calcium contained in the tailings-limestone mixture was removed. This corresponded to removal of approximately 22% of the total gypsum contained in the tailings.

The effluent magnesium concentrations were initially low, in the range of  $\sim 2-4$  mg/l for the first two years, which gradually increased in the third year to peak concentrations of approximately 10-20 mg/l and leveled to a column average range of  $\sim 8-10$  mg/l. The onset of increased Mg in the drainage effluent appeared earlier for columns #4-6 and #7-9 containing coarse and intermediate size limestone, respectively, than for Columns 10-15 which contained fine or pulverized limestone. Also the average Mg concentrations were higher in effluents from column #4-6 than from other column sub-groups. This indicated an early oxidation in the sub-group containing coarse limestone, but the acid neutralization was complete and acidic drainage was prevented.

For all columns, the columns average effluent loading or removal rates for Mg were in the range of  $\sim$ 40-60 µg Mg/kg tailings/day, with columns #4-6 having the highest rates. During the period of increased Mg drainage (last 2.5 years of the study), an average of approximately 36.5 mg

Mg/kg of tailings/day or 3.3% of total magnesium contained in the tailings and limestone mixture was removed.

## **Aluminum and Manganese:**

The aluminum and manganese leaching characteristics are shown, respectively, in Figures B-10 and 11 to C, D, and E-10 and 1, for concentration profiles, and in Figures 44, 45, 64,65 84, 85, 104, and 105, for effluent loadings for the four sub-groups, respectively. Similar to iron, the effluent aluminum concentrations were very low, in the range of  $\sim 0.1 - 0.2$  mg/l, with corresponding low,  $< 1 \mu g$  Al/kg tailings/day, loading rates.

The effluent Mn concentrations varied with the column sub-groups, from a low range of 0.05 - 0.1 mg/l for columns #10-12 and #13-15, to a range of ~ 0.1 - 0.4 mg/l for columns #4-6 and #7-9. Appreciable amounts of Mn in the drainage effluents were only detected for columns containing tailings mixed with coarse limestone, where Mn concentration peaks were also observed in the third year of the study. For other columns, Mn concentrations gradually increased with time during that period. The column average Mn loading rates were only significant for columns #4-6, at approximately 1 µg Mn/kg tailings per day. These results were consistent with other observations, where an early stage of oxidation was indicated in limestone amended tailings, which was more pronounced for the coarse limestone sub-group.

#### Uranium, Thorium, Lead and Other Metals:

The effluent concentrations of U, Th, Pb and other metals: Cu, Ce, Ni and Zn were below detection to very low for all columns and there were no significant concentration trends. Consequently, the data were not further analyzed. For leaching characteristics, the reader is referred to Figures B-12 to 14, to C, D and E-12 to 14, for concentration profiles and to Figures 46-48, 66-68, 86-88 and 106-108, for loading profiles, respectively, for U, Th and Pb for the four tailings-limestone sub-groups.

#### <u>Ra-226:</u>

The Ra-226 leaching profiles for total mill tailings mixed with different sized limestone are shown in Figures B-15, C, D to E-15, for concentrations and figures 49, 50, 69, 70, 89, 90, 109, and 110 for effluent loading rates and cumulative productions/removals of Ra-226, for all tailings-limestone sub-groups.

Initially for the first year, the effluent Ra-226 concentrations were nearly constant for the all subgroups, averaging in the range of 300-500 mBq/l. At the introduction of the batch leaching schedule, Ra-226 concentrations increased in effluents from all columns, for about six months, with characteristic peak concentrations in the range of ~ 1800-2000 mBq/l. The concentrations decreased afterwards to nearly constant values, between 600-800 mBq/l. These trends were similar to those for the Control -1 tailings without additional limestone, and were indicative of a very early oxidation in limestone amended tailings.

Similar leaching trends were observed for effluent Ra-226 loading profiles for all columns, having peak and column average loading rates of approximately 6 mBq Ra-226/kg tailings/day and 3 mBq Ra-226/kg tailings/day, respectively. During the complete study period, the column average cumulative loading or total production/removal of Ra-226 was calculated as ~ 4660 mBq/kg tailings or 0.06% of total Ra-226 contained in the tailings-limestone mixtures. The cumulative removal of Ra-226 was very small and comparable to that for Control -1 tailings without limestone amendment, where it was calculated as 0.08%.

Also similar to Control -1 tailings, no correlations were observed among the effluent Ra-226 and dissolved Ca, Mg or  $SO_4^{-2}$  concentrations. The effluent concentrations of these ions were still controlled by gypsum dissolution, but its contribution to total Ra-226. leachability could not be established from the observed leaching profiles.

# 3.3.1.2 Group 2: Un-oxidized Coarse Tailings

In Group 2 column leaching experiments, the coarse tailings without additional limestone (Control - 2, columns #16 and 17) oxidized rapidly producing highly acidic drainage (as defined previously) soon after and within two months of the introduction of the batch leaching schedule. The detailed leaching characteristics are shown in Figures F-1 to F-16 for concentration profiles, and in Figures 112 to 137 for effluent loadings and cumulative mass production/removal profiles, respectively. Similar to the Group 1 - total mill tailings, the results are discussed for the following parameter categories.

# pH, Eh and Ec:

Figures 112 to 114 show, respectively, the variations of pH, Eh and Ec with time for Control - 2 tailings. Initially, during the first year, the column effluent pH's were slightly alkaline at  $\sim 8$ , which decreased to <3 within one to two months after the implementation of the batch leaching schedule, and within six months decreased further to column average effluent pH's of approximately 1-2. The effluents continued to drain at very low pH's which increased very slowly to  $\sim 1.8$  near the end of the fourth year (Figure 112). These pH's were 1 to 2 orders of magnitude lower than those of the Control - 1, total mill tailings. During the four year study period, a column average cumulative total of 81 1 of drainage effluent was collected for each column. During the same period, each column had received, on an average, of approximately 104 1 of natural lake water for leaching/rinsing purposes.

During the first year, the Control -2 effluent Eh's were practically constant, within 350-450 mV range when the columns were draining at near neutral to slightly alkaline pH's. With the introduction of the batch leaching schedule and at the onset of acidic drainage, the Eh's increased rapidly to a range of ~ 680-700 mV within few months and remained constant at the high range where low pH and highly oxidized conditions prevailed through to the rest of the experimental period (Figure 113).

The effluent electrical conductance, Ec, also increased rapidly with the occurrence of the acidic drainage in the second year, from column average low Ec in the range of ~ 500-1000  $\mu$ s/cm to approximately 50,000-55,000  $\mu$ s/cm range peak conductance. Afterwards, it gradually decreased to ~ 20,000  $\mu$ s/cm level (Figure 114).

Although the Group 2 tailings experiments were conducted in duplicates, both columns #16 and 17 exhibited remarkable similarity in the observed leaching profiles for the measured parameters e.g. pH, Eh, Ec, etc., all of which indicated severe oxidation and acidic drainage for the Control - 2 tailings. The results also indicated that in coarse tailings without limestone amendment there was very little residual carbonate or silicate buffering present and the acidic drainage occurred early.

# Acidity and Alkalinity:

The acidity and alkalinity profiles for Control- 2 tailings are shown, respectively, in Figures F-4 and F-5 for concentrations, in Figures 115 and 117 for effluent loading rates, and in Figures 116 for cumulative acidity production/removal..

With the onset of the acidic drainage after the first year, the effluent acidity increased very rapidly from zero (very low) to peak concentration of approximately 25,000-30,000 mg  $CaCO_3/I$  within the first few months. It decreased slowly afterwards to a column average acidity range of ~ 12,000-15,000 mg CaCO<sub>3</sub>/l (Figure F-4). The acidity loading profiles showed similar results, increasing from 200 to peak loading rate of approximately 120,000 µg  $CaCO_3$  /kg tailings/day, and decreased gradually to ~ 50,000 µg CaCO<sub>3</sub> /kg tailings/day (Figure 115). During the three year acidic drainage period, a column average effluent production/removal (loading) of total acidity was calculated as 76.7 g CaCO<sub>3</sub> /kg tailings or approximately 58% of total acid generation potential of coarse tailings (Control - 2). As mentioned previously, the Control - 2 tailings were producing acidic drainage with total acidity loadings at a rate 30 times higher than that for Control - 1, total mill tailings.

Prior to the onset of the acidic drainage, the Control - 2 tailings were producing slightly alkaline effluents with a column average alkalinity of approximately 150 mg/l CaCO<sub>3</sub> and a alkalinity production rate of ~ 500  $\mu$ g CaCO<sub>3</sub> /kg tailings per day, which decreased to zero with the occurrence of the acidic drainage (Figures F-5 and 117). As with other tailings, the alkalinity was produced by the increased dissolution of the residual carbonate minerals e.g. CaCO<sub>3</sub> (calcite) and MgCO<sub>3</sub> (Dolomite) contained in the tailings, which provided the initial acid neutralization. The rapid acidic drainage indicated that these minerals were present only in very small quantities in Control - 2 tailings, and were either consumed fully or unavailable for further acid neutralization.

## Sulphate and Iron:

The dissolved  $SO_4^{-2}$  and total dissolved iron concentration profiles for Control -2 coarse tailings are shown, respectively, in Figures F-6 and F-7. Figures 118-121, show, respectively,  $SO_4^{-2}$  loading rate, cumulative  $SO_4^{-2}$  production/removal, total dissolved iron loading rate and cumulative total iron production/removal characteristics.

Similar to Ec, the effluent sulphate concentrations increased from a low range of ~ 500-1000 mg/l prior to acidic drainage to peak concentrations of ~ 25,000-30,000 mg/l in the acidic effluent, and then decreased slowly to ~ 12,000-15,000 mg/l range (Figure F-6). The corresponding column average effluent  $SO_4^{-2}$  loading rates were approximately 110,000 µg  $SO_4^{-2}$ /kg tailings/day during the peak acidic drainage period and ~ 60,000 - 80,000 µg  $SO_4^{-2}$ /kg tailings/day afterwards (Figure 118 and 119).

During the three year period for which acidic drainage was monitored, the column average effluent sulphate loading rate was calculated as ~ 80,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day, which corresponded to a cumulative effluent SO<sub>4</sub><sup>-2</sup> production/removal of ~ 87.9 g SO<sub>4</sub><sup>-2</sup>/kg of tailings or ~ 67% of total sulphur contained in the coarse tailings. More than 95% of this total sulphur removed resulted from the oxidation of pyrite contained in these tailings.

Prior to the acidic drainage, the dissolved total iron concentrations in the effluents were very low , < 0.3 mg/l, which increased to peak concentrations in the range of ~10,000-12,000 mg/l at the onset of acidic drainage and then decreased slowly to 5000-7000 mg/l range near the end of the study period (Figure F-7). During the same period, the column average effluent loading rate of dissolved iron was approximately 40,000  $\mu$ g Fe/kg tailings/day near the peak acidity period, and decreased gradually to a range of ~ 15,000 - 20,000  $\mu$ g Fe/kg tailings/day near the end of the study period, corresponding to an average iron removal rate of approximately 25,000  $\mu$ g Fe/kg tailings/day for the entire acidic drainage period ((Figure 118). During that time, an average of approximately 27.4 g of iron/kg tailings was removed from each of the Control -2 columns, which corresponded to a production/removal of ~ 67.7% of total iron contained in the coarse tailings.

The cumulative percentile mass productions/removals for total iron and sulphur for Control -2 tailings were very comparable, indicating good correlation between Fe and S oxidations as well as that of ionic mass balance in solid and effluent samples.

Compared to Control -1 total mill tailings, the total iron mobilization (%) in Control -2 coarse tailings was approximately 210 times greater. Because the sulphate removal rate in the former tailings was controlled by gypsum dissolution, the cumulative total sulphur mobilization, as a result of oxidation in the two tailings, was not comparable.

#### **Calcium and Magnesium:**

The calcium and magnesium leaching profiles for Control -2 tailings are shown, respectively, in Figures F-8 and F-9, for concentrations, and in Figures 122-125, respectively, for calcium loading rate, cumulative calcium removal, magnesium loading rate and cumulative removal characteristics of magnesium.

The effluent calcium concentrations increased gradually from approximately 100 mg/l at the start of the leaching experiment, to peak concentrations of  $\sim 600$  mg/l upon the implementation of the

batch leaching schedule and just before the onset of the acidic drainage. In the acidic drainage period, the calcium concentrations rapidly decreased, and within a year were in the range of ~ 10-20 mg/l, and less than 10 mg/l afterwards (Figure F-8). During the same period, the column average effluent loading/removal rate for calcium increased from approximately 200-300  $\mu$ g Ca/kg tailings/day to maximum of 2500-3000  $\mu$ g Ca/kg tailings/day, and then decreased rapidly to less than 40  $\mu$ g Ca/kg tailings/day (Figure 122). This corresponded to an estimated column average calcium removal of approximately 0.76 g Ca/kg tailings, or 19% of the total amount of calcium contained in Control -2 tailings during the entire study. period.

These results indicated that in the coarse tailings without additional limestone, the oxidation, acid generation and neutralization processes started very early in the experiment and acidic drainage occurred upon consumption of the available alkalinity. Because the total acid neutralization potential and residual gypsum content of these tailings were small, ~ 2.7 kg CaCO<sub>3</sub>/tonne (0.1% Ca) equivalent carbonate alkalinity and 0.6% (0.17% Ca) gypsum as CaSO<sub>4</sub> respectively, the gypsum dissolution and removal rates were controlled by acid generation and neutralization kinetics in the present experiments. Taking the initial (prior to acidic drainage) calcium production/removal rate of ~ 250  $\mu$ g Ca/kg tailing/day as solubility contribution of original gypsum residue in the tailings, the additional limestone neutralization and calcium removal was calculated as 0.60 g Ca/kg of tailings or approximately 60% of total limestone contained in the tailings. Thus in the coarse tailings, the acidic drainage occurred before the full consumption of the available limestone alkalinity, although, the latter was only present in a small amount.

Similar to calcium, the effluent magnesium concentrations also increased gradually, from an initial range of  $\sim$ 3-5 mg/l at the beginning of the leaching experiment, to peak concentrations of approximately 30 mg/l near the onset of the acidic drainage. In the acidic effluents, magnesium concentrations decreased slowly to approximately 5 mg/l in the fourth year of the study (Figure F-9). During the same period, the column average effluent loading/removal rates for magnesium increased from approximately 10-20 µg Mg/kg tailings/day to a maximum of 120 µg Mg/kg

tailing/day, and decreased gradually to 10-20 µg Mg/kg tailings/day range near the end of the study period (Figure 124). The calculated column average cumulative magnesium production/removal was approximately 0.077 g Mg/kg tailings or 13% of total magnesium contained in the coarse tailings. Although magnesium was easily mobilized in the oxidation and acid generation processes, its removal rate was small even under severe acidic conditions, probably because of the type of mineralization and its availability/distribution within the host rock.

## **Aluminum and Manganese:**

Figures F-10 and F-11, show respectively, the effluent aluminum and manganese concentration profiles and leaching characteristics of Control -2 tailings. The corresponding aluminum and manganese loading rate profiles are shown in Figures 126 and 128 and cumulative mass removal characteristics in Figures 127 and 129, respectively.

For Control -2 tailings, the aluminum leaching profiles were similar to those for iron, in both cases the ionic mobility was directly related to the onset of acidic drainage. Prior to acidic drainage, the effluent aluminum concentrations were very low, in the range of ~ 0.02-0.3 mg/l, which increased with the acidic drainage by many orders of magnitude to peak concentrations of approximately 500-600 mg/l. In the acidic effluent, the aluminum concentrations decreased very rapidly over a short period of time (within six months) to approximately 100 mg/l, and thereafter decreased very slowly (Figure F-10). The effluent Al loading rate profiles were also similar, with column average peak Al loading rate of ~ 2000 µg Al/kg tailings/day, which decreased afterwards to ~ 200-500 µg Al/kg tailings/day range (Figure 126). During the entire acidic drainage period, the column average cumulative production/removal of aluminum was approximately 0.46 g Al/kg tailings or 1.76% of total aluminum contained in the coarse tailings. Unlike iron and because of the alumino- silicate matrix, aluminum was not easily mobilized. Consequently, pH buffering with the dissolution of alumino-silicates was not significant in these tailings.

The leaching characteristics of manganese resembled those of magnesium, where the ionic mobility started with the oxidation and acid generation (Figures F-9 and 11). With the acidic drainage, the effluent manganese concentrations peaked in the range of 3-5 mg/l and then decreased to 0.5-1.0 mg/l range over a one year period. The corresponding column average effluent loading rates were approximately 18  $\mu$ g Mn/kg tailings/day near the peak acidic drainage period and between 2 - 5  $\mu$ g Mn/kg tailings/day elsewhere (Figure 128). During the entire study period, a column average total of approximately 6 mg Mn/kg tailings or 21% of total Mn contained in the Control -2 coarse tailings was removed. The manganese removals from both Control -1 and -2 tailings, during the study period, were significant, and in both cases were significantly greater than that of Al. In Control -2 tailings, the total removal rate iron was, however, greater than that of Mn.

#### **Uranium, Thorium and Lead:**

The leaching profiles for uranium, thorium and lead are shown, respectively, in Figures F-12 to F-14 for effluent concentrations, in Figures 130, 131 and 133 for effluent loading rates, and in Figures 132 and 134 for cumulative mass removal of thorium and lead, respectively. For uranium the cumulative mass removal profile was not computed as there some uncertainty in the measurement of solid phase uranium composition of the tailings, at low concentration range using an Inductively Coupled Plasma Argon Emission Spectrophotometer (ICAP-AES), where the minimum detection limit was high, ~ 40  $\mu$ g/g (0.2 mg/l in solution).

The leaching profiles for both uranium and thorium were quite similar in their effluent concentrations and loadings characteristics, and their mobilities resulted by the acidic drainage. The effluent peak concentrations, though very significant at ~ 16 mg/l for U and 135 mg/l for Th, decreased sharply to very low levels within four to six months of the occurrence of the acidic drainage (Figures F-12 and 13, and 132 and 133). In both columns #16 and 17, increased Th concentrations in the effluents were detected four months before U and where Th concentrations and loading rates were high for a longer period of time. In the acidic effluent, the column average peak effluent loading rates for uranium and thorium were determined as ~ 60  $\mu$ g

U and 550  $\mu$ g Th/kg tailings/day respectively. During the entire acidic drainage period, it was estimated that approximately 7 mg of total uranium/kg tailings, and 110 mg of Th/kg tailings or 54% of total thorium contained in the tailings were removed. Thus for Control -2 tailings the thorium mobility was high and comparable to that of iron.

The leaching profiles for lead were very similar to those for iron, where effluent lead concentrations and loading rates increased rapidly with the acidic drainage to peak values of approximately 3 mg/l and 12  $\mu$ g Pb/kg tailings/day, and decreased slowly in the acidic effluent (Figure F-14 and 133). The calculated column average Pb removal, during the acidic drainage period, was approximately 11 mg/kg tailings or 5% of the total lead contained in the tailings.

## Other Metals, Ce, Cu, Ni and Zn, etc.:

Similar to Control -1 tailings, the leaching profiles were also measured for Ce, Cu, Ni and Zn for Control -2 tailings, but because of their low concentrations in the tailings, the date was not processed in greater detail. Their concentrations and loading results are provided in Tables F-1 and F-2 together with other parameters measured.

For coarse tailings (Control -2), the general leaching characteristics of these metals (and presumably other rare earths that were not measured e.g. yttrium, etc.) were similar to those of iron and aluminum, where the effluent concentrations and metal loading rates increased with the onset of acidic drainage, peaked and decreased slowly afterwards to low but detectable levels. The average peak concentrations of these elements in the acidic effluents were significant at: 33 mg/l Ce, 21 mg/l Cu, 10 mg/l Ni and 9 mg/l. The corresponding column average effluent peak loading rates were, approximately 150 µg Ce, 88 µg Cu, 45 µg Ni and 40 µg Zn/kg tailings/day.

# **Ra-226:**

For Control -2 coarse tailings, the Ra-226 leaching characteristics are shown in Figure F-15 for concentration, and in Figures 135 and 136 for effluent loading rate and cumulative Ra-226 removal profiles, respectively.

Initially, in the absence of the acidic drainage, Ra-226 was leaching at a high rate with effluent concentrations in the range of 1400-1800 mBq/l, which increased with the acid generation, within few months, to peak concentrations of ~ 5000 mBq/l. Afterwards in the acidic effluent, Ra-226 concentrations decreased very rapidly, also within a short period of time, to very low values in the range of 100-200 mBq/l (Figure F-15).

These results are to be compared with Control -1, total mill tailings, which contained approximately twice the amount of Ra-226 in the solid phase (7500 mBq/g compared to 3900 mBq/g for coarse tailings), and where effluent Ra-226 concentrations were in the range of 200-500 mBq/l initially, increasing to 3000-3500 mBq/l with the acidic drainage and then decreasing slowly to a 500-1000 mBq/l range (Figure A-15). Thus in the absence of acidic drainage and before its occurrence, Ra-226 leachability was higher for coarse tailings than for total mill tailings. In the acidic effluent, the case was reversed after Ra-226 concentrations peaked with the acidic drainage. The data indicated decreased Ra-226 leachability with the development of severe acidic conditions in Control -2 tailings and in the absence of gypsum, in contrast to Control -1, total mill tailings, where the acidic conditions were moderate and gypsum was present in significant amount.

The effluent Ra-226 loading rates were similar in characteristics to its concentration profiles, where the column average effluent loading rate increased from an initial range of ~ 5.7 mBq Raof 226/kg tailings/day peak loading approximately 22 mBq to rate Ra-226/kg tailings/day, and decreased rapidly to ~ 1-2 mBq/kg tailings/day range (Figure 135). During the entire study period, a column average of approximately 6800 mBq/kg tailings of total Ra-226 or only 0.17% of total Ra-226 contained in the coarse tailings was removed. For Control -2 tailings, the cumulative (%) Ra-226 removal was approximately twice that of Control -1, total mill tailings. In the former case, most of the Ra-226 was mobilized prior to and at the onset of the acidic drainage, whereas in the latter tailings, it was mobilized after the onset of the acidic drainage.

While measuring Ra-226 concentrations (activity) using an alpha spectroscopy technique, it was observed that in the acidic effluent, Ra-226 and its progeny alpha decay peaks were accompanied by large concentration peaks of Ra-223, Ra-224 and their progeny, whose activities were one to two orders of magnitude higher than that of Ra-226. Figure 138 shows a typical alpha energy spectrum of the acidic effluent collected from column #17 near the peak acidic drainage period (July 1990), where the alpha energy peaks from various radium isotopes and their progeny, as well their relative abundances are clearly seen. It is believed that the increased drainage of radium isotopes, Ra-226, Ra-224 and Ra-223 and their progeny, resulted from leaching of their corresponding parent thorium isotopes, e.g. Th-230, Th-232 and Th-229, upon acid generation. With the leaching of Th and Ra isotopes, a new secular equilibrium was re-established in the drainage effluent samples, which was disturbed by the differential leaching of various radioisotopes from the solid phase. Such occurrences of increased Ra-223 and Ra-224 isotopes and their progeny in the acidic porewaters of tailings were common in the field, as observed by Lim (1988).

Similar profiles for radium isotopes in the acidic drainage were also observed for other tailings and waste rock groups.

# 3.3.1.2.2 Group 2: Un-oxidized and Coarse Tailings Mixed With 7.5% (w/w) Coarse (-6.3 mm Size) Limestone (Columns #18 and 19)

The coarse tailings mixed with 7.5% by weight coarse limestone of screen size -6.3 mm (column #18 and 19), also oxidized and produced highly acidic drainage but its onset was delayed by approximately one year in comparison to Control -2 coarse tailings without limestone, where it occurred soon after the introduction of the batch leaching schedule. The detailed leaching characteristics for columns #18 and 19 are shown in Figures G-1 to G-16 for concentration, and in Figures 139-164 for effluent loading rates and cumulative mass removal profiles, respectively. Similar to Control -1 tailings, a column average cumulative total of 75 l of effluent was collected from each column during the four year study period. During the same period

each column had received an average of approximately 104 l of natural lake water for leaching/rinsing purposes. The results are discussed below in detail:

### pH, Eh and Ec:

Figures 139-141 show, respectively, the variations of pH, Eh and Ec with time for coarse tailings mixed with coarse limestone. As mentioned previously, these columns were producing slightly alkaline effluents with pH's ~ 8.0 during the first two years of the study. Acidic drainage started near the end of the second year (April 1991), where the pH dropped from ~8 to ~ 2.5 within a two month period and remained low thereafter. The two columns differed slightly in their effluent characteristics, with column #19 leading by few months in the occurrence of acidic drainages as well as accompanied chemical species. (Fig. 139).

Although, the tailings were mixed with an additional 7.5% coarse limestone (NNP-42.8 kg CaCO<sub>3</sub>/Tonne), its quantity was, however, not sufficient enough to provide a acid complete neutralization for a prolonged period. In comparison to coarse tailings without limestone (Control - 1), the addition of coarse limestone to coarse tailings prevented acidic drainage for an extra year. In these tailings-limestone mixtures, some acid neutralization was still taking place, as seen from effluent pH's and dissolved calcium concentrations, but because of their high hydraulic conductivities and rapid drainage characteristics, the effluent residence times in the tailings were not long enough for complete acid neutralization and utilization of the available alkalinity in preventing acidic drainage.

Similar to Control -2 tailings, the effluent Eh's were within 350-450 mV prior to the occurrence of the acidic drainage. With the acidic drainage, the Eh's increased rapidly to a range of ~ 650-700 mV, corresponding well oxidized conditions, and remained constant at these values in the acidic effluent throughout the experiment (Figure 140). The transition in effluent characteristics for column #19 was once again early.

In contrast to the above two parameters, the effluent Ec increased slowly from 400 to 3000  $\mu$ s/cm before the occurrence of the acidic drainage, and then at a higher rate in the acidic effluent, increasing from approximately 3000  $\mu$ s/cm to a peak range of ~ 8000-10,000  $\mu$ s/cm. Towards the end of the study, the effluent Ec decreased to a column average of ~ 6000  $\mu$ s/cm (Figure 141). For Control -2 tailings, the corresponding Ec values were 3 to 6 times higher, indicating that for coarse tailings-coarse limestone mixture, there was an ongoing acid neutralization and the effluent was saturated with respect to the gypsum.

## Acidity and Alkalinity:

The acidity and alkalinity profiles for coarse tailings mixed with coarse limestone are shown in Figures G-4 and G-5 for concentrations, in Figures 142 and 144 for effluent loading rates, and in Figure 143 for cumulative acid production/removal characteristics, respectively.

Prior to the onset of acidic drainage during the first two years, the effluent acidities were zero. Acidities increased very rapidly with the acidic drainage, and within few months, were in the range of ~ 5000-6000 mg CaCO<sub>3</sub>/l, and continued to increase for the next two years to a column average peak acidity of ~ 12,000 mg CaCO<sub>3</sub>/l, and decreased afterward to ~ 4000 mg CaCO<sub>3</sub>/l near the end of the study period (Figure G-4). These acidities were lower by factors of 3-4 than those for Control -2 tailings.

The acidity loading profiles showed similar results, increasing from zero to a peak value of approximately 40,000 - 60,000  $\mu$ g CaCO<sub>3</sub> acidity/kg tailings/day, then decreased to ~ 20,000  $\mu$ g CaCO<sub>3</sub> acidity/kg tailings/day near the end of the study period (Figure 142). During the acidic drainage period of approximately two and a quarter year, lasting till the end of the study, a column average acidity load of the effluent was calculated as 20.5 g CaCO<sub>3</sub> equivalent acidity/kg tailings or approximately 17% of the total acid generation potential of the coarse tailings and limestone mixture. Because of a shorter acidic drainage period and available limestone buffering, the total acidities produced by such tailings - limestone mixtures were

approximately 3 to 4 times less than those for Control - 2 tailings during the study period, as observed earlier for effluent acidities.

As mentioned previously, the added 7.5% limestone was not stoichiometrically equivalent to the maximum acid generation potential of coarse tailings (118.75 kg CaCO<sub>3</sub>/tonne). The additional alkalinity, which provided a total neutralization potential of 75.9 kg CaCO<sub>3</sub>/tonne, was, however, comparable to the total column average acidity production of 76.7 kg CaCO<sub>3</sub>/tonne tailings by Control -2 tailings during the entire experimental leaching period of approximately years (see Section 3.2.1.2.1). The limestone addition to coarse tailings had thus not only delayed the onset of acidic drainage by one additional year but suppressed the production of additional acidity by ~ 56.2 kg CaCO<sub>3</sub>/tonne tailings, or 47% of maximum potential acidity. These results indicated some initial delay and suppression of acid generation in tailings mixed with limestone because of prevailing alkaline conditions, as well as acid neutralization prior to the occurrence of acidic drainage. As shown below, the total limestone neutralization achieved in these tailings was much less.

As seen from effluent calcium concentrations, the drainage effluents from all Group -2 coarse tailings and coarse tailings-limestone mixtures, columns (#16-21), were initially unsaturated with respect to gypsum and before occurrences of substantial oxidation and acid generation conditions (see for example Figures F-8, G-8 and H-8). With acid generation and its neutralization, both effluent calcium concentrations and loading rates increased with time until complete saturation with respect to gypsum and remained constant thereafter. For both Control -2 tailings (columns #16 and 17) and coarse tailings - coarse limestone (columns #18 and 19) groups, the effluent gypsum saturation was observed near the occurrence of the acidic drainage. Based on the measured column average effluent calcium loading rate of  $\sim 2500 \ \mu g \ Ca/kg$  tailings/day for columns #18 and 19, the total acid generation and hence limestone neutralization of this sub-group of tailings-limestone, prior to acidic drainage, was calculated as 4.6 g CaCO<sub>3</sub>/kg tailings mixed with coarse limestone, only less than 10% of the available alkalinity was

utilized before the occurrence of the acidic drainage. Thus the neutralization efficiency of coarse limestone was very low.

Similar to Control -2 tailings, the coarse tailings - coarse limestone subgroup was also producing slightly alkaline effluents at the beginning and prior to acidic drainage with column average alkalinity in the range of 150-180 mg CaCO<sub>3</sub>/l and an alkalinity production rate of approximately 600  $\mu$ g CaCO<sub>3</sub>/kg tailings/day. The effluent alkalinity decreased rapidly to zero with the occurrence of the acidic drainage (Figures G-5 and 144), further indicating insufficient neutralization.

## Sulphate and Iron:

The leaching characteristics for coarse tailings mixed with coarse limestone for dissolved sulphate and total dissolved iron are shown, respectively, in Figures G-6 and G-7 for concentrations, in Figures 145 and 147 for effluent loading rates, and in Figures 146 and 148 for cumulative mass removal profiles.

The dissolved sulphate concentration profiles were very similar to those for electrical conductance, Ec's, where during the non acidic drainage period, the sulphate concentration increased slowly with time, from an initial range of ~ 100-200 mg/l to approximately 2000 mg/l. With the onset of acidic drainage, sulphate concentrations increased gradually but at a higher rate to peak concentration in the range of 1200-1600 mg/l over a two year period and then decreased to 6000-8000 mg/l range near the end of the study period (Figure G-6). The sulphate loading rate profiles were similar, increasing from a low range of ~ 2000-5000  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day to peak levels of 40,000-50,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day. During the entire study period, a column average of approximately 28 g of total sulphate/kg of tailings or 25% of total sulphur contained in the tailings-coarse limestone mixture was removed. More than 85% of the sulphur removed resulted from the oxidation of these tailings.

The iron concentration and loading rate profiles were similar to those of total acidity, increasing from near zero to very low values prior to acidic drainage, then to peak concentrations and loading rates of approximately 5000 mg/l and 15,000  $\mu$ g Fe/kg tailings/day, respectively, with the acidic drainage, and after a two year period, decreased to ~ 2000 mg/l and 10,000  $\mu$ g Fe/kg tailings/day, respectively (Figures G-7 and 147). During the entire acidic drainage period monitored, a column average effluent loading and cumulative removal of iron was calculated as 6.6 g of iron/kg tailings or approximately 17% of total iron contained in the tailings limestone mixture. Because of continued limestone reactivity and acid neutralization to a certain extent, the total amount of iron removed was somewhat less than that of sulphur.

Compared to Control - 2 coarse tailings, the cumulative (%) sulphur and iron removals for coarse tailings - coarse limestone mixtures were reduced by factors of 3 to 4, resulting from delayed acid generation and continued limestone neutralization. Further monitoring of these columns is required for evaluating the impact of coarse limestone in controlling the release of acidity, iron, sulphate and other oxidation derived reaction products.

# **Calcium and Magnesium:**

The calcium and magnesium leaching profile for coarse tailings - coarse limestone subgroup are shown in Figures G-8 and G-9, for concentrations, and in Figures 149-152, respectively, for calcium loading rates and cumulative calcium removal, magnesium loading rates and cumulative removal characteristics of magnesium.

As mentioned previously in the acidity section, the effluent calcium concentrations increased gradually from ~100-150 mg/l range at the beginning of the leaching experiments to peak concentrations of ~ 580 mg/l near the onset of the acidic drainage. Since then, the calcium concentrations remained constant and were controlled by gypsum dissolution (Figure G-8). During the same period, the effluent calcium loading rates increased gradually from ~ 300  $\mu$ g Ca/kg tailings/day to maximum of 3500-4500  $\mu$ g Ca/kg tailings/day (Figure 149). This corresponded to an estimated column average calcium

removal of 4.3 g Ca/kg tailings, or 12% of total calcium contained in the tailings coarse limestone mixture during the entire study period. In comparison, the cumulative (%) sulphur removal was higher because of acidic drainage and lack of adequate neutralization.

Similar to calcium, the effluent magnesium concentrations also increased, at first slowly from an initial 3-5 mg/l range at the beginning of the leaching experiments, then rapidly to peak concentrations of ~ 80 mg/l near the occurrence of the acidic drainage and in the acidic effluent. Near the end of the fourth year, magnesium concentration gradually decreased to 20-30 mg/l range (Figure G-9). During the same period, the column average effluent loading rates for magnesium increased from ~ 25  $\mu$ g Mg/kg tailings/day to peak values of 300-300  $\mu$ g Mg/kg tailings/day, decreasing gradually to 75-100 µg Mg/kg tailings/day range near the end of the experiment (Figure 151). During the entire study period, the column average magnesium calculated removal 166 mg/kg tailings was as or  $\sim 26\%$  of total magnesium contained in tailings-coarse limestone mixture. Compared to Control -2 tailings, the increased removal of magnesium in coarse tailings-coarse limestone mixture resulted from acid neutralization by the additional magnesium alkalinity contained in the limestone, and increased solubility of MgSO<sub>4</sub> which also resulted in higher magnesium removal than that for calcium.

# Aluminum and Manganese:

Figures G-10 and G-11 show, respectively, the effluent aluminum and manganese concentration profiles and leaching characteristics of coarse tailings-coarse limestone subgroup. The corresponding effluent loading rate profiles are shown in Figures 153 and 155, and the cumulative mass removal characteristics in Figures 154 and 156, respectively.

Similar to Control -2 tailings, the aluminum leaching profiles for coarse tailings-coarse limestone mixtures were similar to those for iron, increasing rapidly from below detection and very low concentrations in the absence of acidic drainage to 150-200 mg/l range in the acidic effluent. Near the end of the study, the aluminum concentrations decreased to 50-80 mg/l range (Figure

G-11). The corresponding column average effluent aluminum loading rates increased from zero to peak loadings of ~ 600  $\mu$ g Al/kg tailings/day, decreasing afterward to 150-200  $\mu$ g Al/kg tailings/day (Figure 153). During the acidic drainage period, a column average total of 206 mg of Al/kg tailings or approximately 0.8% of total aluminum contained in the tailings-limestone mixture was removed. Thus with the acidic drainage, the mobilization of aluminum was limited to less than 1% of Al content of coarse tailing-coarse limestone test samples, which was approximately half that for Control -2 tailings due to the delayed onset of acidic drainage. In both cases, the cumulative removal of aluminum removal was much less than that of iron.

The leaching characteristics of manganese, similar to coarse tailings (Control -2), resembled those of magnesium, where both were mobilized with the oxidation and acid generation (Figures G-9 and G-11). In the neutral to alkaline drainage the effluent manganese concentrations were very low, ~ 0.2-0.5 mg/l, which increased with the increased acidity to peak concentrations in the range of 3-4 mg/l, decreasing thereafter to approximately 1.0 mg/l near the end of the fourth year of the study. The corresponding effluent manganese loading rates increased from 1 µg Mn/kg tailings/day to peak maximums in the range of ~ 12-14 µg Mn/kg tailings/day, and then decreased to approximately 4 µg Mn/kg tailings/day. For coarse tailings-coarse limestone samples, a column average effluent manganese removal during the study period was calculated as approximately 7.0 mg/kg tailings or 75% of total manganese contained in the mixture. Although, the total mass of manganese removed for Control -2 and coarse tailings-limestone sub groups was comparable, the cumulative percentile value was higher in the later case because of low to near detection manganese concentration in the solid phase. In both cases, the cumulative removal of manganese was much greater than that of aluminum.

#### Uranium, Thorium and Lead:

The leaching profiles for uranium, thorium and lead for coarse tailings-coarse limestone mixture are shown, respectively, in Figures G-12 to G-14 for effluent concentrations, in Figures 157, 158, and 160 for effluent loading rates, and in Figures 159 and 161, respectively, for cumulative mass removal characteristics for thorium and lead. Similar to Control -2 tailings, because of low

solid phase uranium concentrations, the cumulative mass removal profiles for uranium were not computed.

As seen from Figures G-12 and 157, the effluent uranium concentrations and loading rate profiles were somewhat poorly defined with occasional peaks at different times for the two columns (#18 and 19). During the acidic drainage period, some mobility of uranium was indicated where effluent concentrations and loading rates increased from below detection to approximately 2.5 mg/l and 12  $\mu$ g U/kg tailings/day, respectively.

The effluent thorium concentrations and loading rates increased from below detection and very low values to approximately 30-40 mg/l range and 100-150  $\mu$ g Th/kg tailings day range peaks, respectively, in the acidic effluent (Figures G-13 and 158), then decreased to ~ 3 mg/l and 25  $\mu$ g Th/kg tailings/day, respectively, near the end of the study period. Unlike uranium, the leaching characteristics of thorium were better defined. During the acidic drainage period monitored, a column average effluent thorium removal was calculated as 41 mg Th/kg tailings or approximately 22% of total thorium contained in the tailings-coarse limestone mixture. Similar to Control -2 tailings, the cumulative (%) thorium and iron removals were comparable.

Similar to thorium and iron, the effluent lead concentration and loadings increased from below detection levels to peak values of approximately 0.8 mg/l and 6  $\mu$ g Pb/kg tailings/day, then decreased to 0.1 mg/l and 1.0  $\mu$ g Pb/kg tailings/day, respectively. The occurrence of lead drainage in the effluent was delayed, and where column #18 was most active in comparison to acidity, iron and aluminum drainages (Figures G-14 and 160). A column average effluent lead removal with the acidic drainage was calculated as ~ 1.3 mg Pb/kg tailings or only 0.5% of total lead contained in the tailings-limestone mixture.

Compared to Control -2 coarse tailings, the effluent loadings and mass removals for most parameters for the coarse tailings-coarse limestone sub group were low because of the delayed

acidic drainage and to a certain extent continued limestone neutralization, although at a reduced rate.

# Other Metals, Ce, Cu, Ni and Zn, etc.:

The concentrations and loading results for other metals are provided in Tables G-1 and G-2, and as in the previous cases, the data were not processed in greater details because of low solid phase compositions of these metals. Similar to Control -2 tailings, the leaching profiles for these metals for coarse tailings-coarse limestone sub-group were also similar to those for Fe and Al, etc. where increased acidic drainage resulted in increased mobilities of various metals and then decreased to low but detectable levels. The column average peak concentrations of these elements in acidic effluent also significant the were at: 6 mg/l Ce, 6 mg/l Cu, 7 mg/l Ni and 3 mg/l Zn. The corresponding column average effluent peak loading rates for various metals were: ~ 20  $\mu$ g Ce, 20  $\mu$ g Cu, 25  $\mu$ g Ni and 10  $\mu$ g Zn/kg tailings/day. These values were lower than those for Control -2 tailings. Also similar to observations made for other parameters, the leaching profiles obtained for column #19 were characterized by higher effluent concentrations, loading rates and early occurrences in the drainage than those for column #18.

#### **Ra-226:**

For coarse tailings mixed with coarse limestone, the Ra-226 leaching characteristics are shown in Figure G-15 for concentrations, and in Figure 162 and 163 for effluent loading rates and cumulative Ra-226 removal profiles, respectively.

Initially during the first year when the effluent was near neutral to alkaline, the effluent Ra-226 concentrations were in the range of 1000-1500 mBq/l, which similar to other tailings above, increased rapidly with the introduction of the batch leaching schedule to peak concentrations in the range of 3000-4000 mBq/l, and then decreased equally rapidly to 1200-1500 mBq/l range before the occurrence of the acidic drainage. In the acidic effluent, Ra-226 concentration increased again to a second peak with concentrations in the range of 3500-4500 mBq/l and decreased somewhat differently for the two columns in the fourth year. For column #18, Ra-

226 continued to leach at a higher rate for about a year than for column #19 where Ra-226 concentrations decreased rapidly in the acidic effluent (Figure G-15). Near the end of the study period, the column average effluent Ra-226 concentrations were  $\sim$  600-800 mBq/l.

In the acidic effluent, similar to Control-2 tailings, the Ra-226 concentrations were controlled by the effluent acidity, where Ra-226 leaching and drainage decreased with severe acidic conditions.

The effluent Ra-226 loading profiles were similar to those for concentrations, where the effluent loading rates increased from approximately 4 mBq/kg tailing/day in the first year to approximately 12 mBq/kg tailings/day at the introduction of the batch leaching schedule, and then to second peak loadings in the range of 12 - 18 mBq/kg tailings/day. In the fourth year, the column average effluent loading rates of Ra-226 decreased to a range of 3 - 6 mBq/kg tailings/day (Figure 162). The corresponding column average removal of Ra-226 during the entire study period was calculated as approximately 10,800 mBq/kg tailings/day or approximately 0.29 % of total Ra-226 contained in the tailings -coarse limestone mixture. Thus for the coarse tailings - coarse limestone sub-group, the cumulative Ra-226 removed was approximately double that for Control - 2 tailings because of decreased cumulative total acidity production in the former case as explained above. However, similar to Control - 2 tailings, the acidic effluent was characterized by an increased mobility of other radium isotopes, e.g. Ra-223, Ra-224 and Ra-228 and their progeny, resulting from increased leaching of corresponding thorium parent radionuclides. The increased mobility of radium isotopes with the acidic drainage also correlated with the oxidation of pyrite and iron mobility in the tailings.

# 3.3.1.2.3 Group - 2: Un-oxidized and Coarse Tailings Mixed With 7.5 % (w/w) Wet Ground and Pulverized Limestone (Columns #20 and 21)

The coarse tailings mixed with wet ground and pulverized limestone did not produce any acidic drainage during the study period. Similar to total mill tailings mixed with various screen sized limestone, the effluents from columns #20 and 21 were near neutral to slightly alkaline and

contained appreciable amounts of dissolved SO<sub>4</sub><sup>-2</sup>, Ca, Mg and Ra-226. The results further indicated that although oxidation was also taking place in the coarse tailings - pulverized limestone sub-group, the acid so produced was completely neutralized by the finely ground limestone in combination with the increased retention time resulting from slightly decreased hydraulic conductivities (Tables 7 and 10). The mobilization of other oxidation related reaction products, e.g. acidity, SO<sub>4</sub><sup>-2</sup>, Fe, Al, U, Th, Pb, Ce, Cu, Ni and Zn was absent or present in trace amount except Mn which was present in small quantity. During the study period, the leaching characteristics of coarse tailings mixed with wet ground limestone were very similar to those of the coarse tailings mixed with coarse limestone prior to the occurrence of the acidic drainage, where sufficient acid neutralization was taking place in the latter sub-group.

The detailed leaching characteristics of coarse tailings-wet ground limestone subgroup (columns #20 and 21) are shown in Figures H-1 to H-16 for concentration profiles, and in Figures 165 to 185 for effluent loading rates and cumulative mass removed profiles, respectively. Similar to other Group - 2 tailings, a column average cumulative total of 75 l of effluent was collected from each column during the four year study period. During the same period, each column had received an average of approximately 104 l of natural lake water for leaching and rinsing purposes. The detailed results are as follows:

#### pH, Eh and Fe:

Figures 165 to 167 show, respectively, the variations of pH, Eh and Ec with time for coarse tailings mixed with wet ground limestone (columns #20 and 21). As seen from Figure 165, these columns were producing slightly alkaline effluents with pH's in the range of 8 - 8.5 throughout the study period.

The effluent Eh values increased very slowly from an initial range of  $\sim 350 - 400$  mV to approximately 500 - 500 mV near the end of the study, which indicated a small degree of

oxidation in these tailings limestone mixtures (Figure 168). At these observed pH - Eh values, the effluents were characterized by precipitation of solid phase  $Fe(OH)_3$  and near the precipitation boundary of Mn with a slight solubility of  $Mn^{+2}$ .

Initially, the effluent electrical conductivities, Ec's, increased from approximately 500  $\mu$ S/cm to 1500  $\mu$ S/cm during the first six months, and then dropped to approximately 1000  $\mu$ S/cm range and remained constant at that range for a further period of 6 to 8 months. After the introduction of the batch leaching schedule, the effluent Ec's increased steadily to approximately 3000  $\mu$ S/cm over a one year period, decreasing afterward to 2200 - 2500  $\mu$ S/cm range for the rest of the study period (Figure 167). Similar to other tailings-limestone mixtures, the effluent Ec's in columns #20 and 21, prior to acidic drainage, were indicative of gypsum formation and its solubility near saturation levels. After the introduction of the batch leaching schedule, the effluent quality results indicated oxidation, acid generation and complete limestone neutralization in this sub-group of coarse tailings-wet ground limestone, although this mixture had a rather high net acid generation potential (NNP = - 43 kg CaCO<sub>3</sub> /tonne). This observation was further substantiated by acidity, Ca, Mg, Fe and SO4<sup>-2</sup> drainage results. The long-term monitoring of these columns would be interesting for evaluating the degree of limestone neutralization provided prior to the occurrence of the acidic drainage, if any.

#### Acidity and Alkalinity:

The acidity and alkalinity profiles for coarse tailings mixed with wet ground limestone are shown in Figures H-4 and H-5 for concentrations, and in Figures 168 - 169 for effluent loading rates, respectively.

As seen from Figures H-4 and 168, the effluent acidities were absent throughout the experimental period and there was no acidic drainage. The columns were producing alkaline effluents with alkalinities in the range of 180 - 200 mg CaCO<sub>3</sub> /1 for the first year, which

decreased gradually to a range of 80 - 100 mg CaCO<sub>3</sub> /l after the implementation of the batch leaching schedule and were constant at these levels for the rest of the study period. The alkalinity production during this period increased from an initial 400 - 600  $\mu$ g CaCO<sub>3</sub> /kg tailings/day range to peak alkalinities of 800 - 900  $\mu$ g CaCO<sub>3</sub> /kg tailings/day, decreasing afterward to a range of 300 - 400  $\mu$ g CaCO<sub>3</sub> /kg tailings/day (Figures H-5 and 169). The results indicated existence of steady state conditions amongst oxidation, acid generation an complete acid neutralization. Thus it was not possible to speculate on the consumption of the available buffering capacity and start of the acidic drainage from these test samples.

## Sulphate and Iron:

The leaching characteristics of dissolved sulphate and total iron for coarse tailings mixed with wet ground limestone are shown, respectively, in Figures H-6 and H-7 for concentrations and in Figures 170 - 172 for effluent sulphate loading rates, cumulative sulphate removals and iron loading rates.

The effluent sulphate profiles were similar to those for Ec, where sulphate concentrations and loading rates increased uniformly from initial 200 - 400 mg/l and 1000 - 1500  $\mu$ g SO<sub>4</sub><sup>-2</sup> /kg tailings/day ranges to steady states and uniform values in the ranges of 1600 - 1800 mg/l and 6000 - 6500  $\mu$ g SO<sub>4</sub><sup>-2</sup> /kg tailings/day, respectively (Figures H-6 and 170). The observed sulphate concentrations and loading profiles were comparable to those for coarse tailings (Control - 2)as well as coarse tailings mixed with coarse limestone prior to the occurrence of the acidic drainage from them. During the entire study period, a column average cumulative sulphate loading or total sulphate production/removal in the effluent was calculated as approximately 8.2 g SO<sub>4</sub><sup>-2</sup> /kg tailings or 7 % of total sulphur contained in the tailings-wet ground limestone mixture. Unlike the previous two sub-groups of coarse tailings and coarse tailings - coarse limestone, where acidic drainage occurred, the total sulphur removed in the present case was comparable to those for total mill tailings mixed with various screen sized limestone (columns #4-15), and where effluent sulphate drainage was controlled by gypsum dissolution.

The effluent dissolved iron (total) concentrations were very low, less than 0.3 mg/l during the first year and then below detection levels for the rest of the study period. The effluent iron loading rates were similarly low and of little significance for iron drainage and mass removal computations (see Figures H-7 and 172).

## **Calcium and Magnesium:**

The effluent calcium and magnesium leaching profiles are shown, respectively in Figures H-8 and H-9 for concentrations, in Figures 173 and 175 for loading rates, and in Figures 174 and 176 for cumulative mass removal characteristics.

Similar to electrical conductance and sulphate, the effluent calcium concentrations were characterized by initial increase from ~ 100 mg/l levels to approximately 300 mg/l peaks, followed by a slight decrease to 200 mg/l levels during the first year. After the implementation of the batch leaching schedule, calcium concentrations increased steadily to maximum gypsum solubility levels during the subsequent year and remained constant at 600 - 700 mg/l range for the rest of the study period. Similarly, the effluent calcium loading rates increased from an initial range of ~ 500 - 1000  $\mu$ g Ca/kg tailings/day to saturation values in the range of 2500 - 3000  $\mu$ g Ca/kg tailings/day (Figures H-8 and 173). During the entire study period, the column average cumulative calcium loading or total mass production/removal was calculated as approximately 3.1 g Ca/kg tailings or

8.8 % of total calcium contained in the tailings-wet ground limestone mixture.

Stoichiometrically, the cumulative mass removal of total calcium was higher than that of total sulphate sulphur (7 %) as noted above, perhaps resulting from the increased solubility of limestone in the natural lake water contributing to moderate effluent alkalinities as observed. In comparison to the coarse tailings - coarse limestone sub-group (columns #18 and 19), the total calcium removal in the present case was less, ~ 75 % of that for the former case, resulting from delayed establishment of gypsum saturation conditions, by approximately 8 - 10 months, in tailings mixed with wet ground limestone (Figures 149 and 173).

Although not very accurate, the effluent calcium loading results were extrapolated further to estimate the degree of limestone neutralization achieved for the present sub-group. Assuming that the oxidation and hence the acid generation rates were increasing linearly with time, as observed during the first two years of leaching and before gypsum saturation conditions, the average acid generation or its neutralization rate during the study period was estimated as approximately 6250 µg CaCO<sub>3</sub>/kg tailings/day from calcium equivalent rate of 2500 µg Ca kg tailings/day (Figure 173). The corresponding limestone neutralization, for the duration of the study, was thus estimated as 9.7 g CaCO<sub>3</sub> /kg tailing or 13 % of total limestone contained in the tailings-wet ground limestone mixture. After approximately four years of leaching under unsaturated conditions, approximately 87 % of residual limestone alkalinity was still available in this tailings-limestone subgroup, which would provide further acid neutralization for an additional period of 29 years. Further monitoring of these columns, for an additional period, would verify predictions of continued acid neutralization and the occurrence of acidic drainage, it any.

The effluent magnesium profiles for concentrations and loading rates were also similar to those of calcium, except during the first year where small peaks in magnesium parameters were observed. Both concentrations and loading rates increased very rapidly from initial ranges of 2 mg/l and 10 µg Mg/kg tailings/day to peak values of 12 mg/l and 40 µg Mg/kg tailings/day, respectively, and decreased back to the initial values. After the implementation of the batch leaching schedule, the parameters increased again, over a one year period, to peak values and then decreased slightly to ranges of 6 - 7 mg/l and 20 - 30 µg Mg/kg tailings/day, respectively (Figures H-9 and 175). During the complete study period, the column average magnesium drainage estimated approximately was as 43 mg/kg tailings or 7 % of total magnesium contained in the tailings. The effluent cumulative magnesium removal was comparable to that of calcium, but was significantly less than that for tailings mixed with coarse limestone, where it was high at ~23 % resulting from increased acidic drainage in the latter case. Total magnesium loadings for coarse tailings - pulverized wet ground limestone and total mill tailings mixed with various screen sized limestone were comparable.

#### **Aluminum and Manganese:**

The effluent aluminum and manganese leaching profiles are shown in Figures H-10 and H-11, respectively, for concentrations and in Figures 177 - 179 for loading rates and cumulative manganese removals.

Similar to iron, the effluent aluminum concentrations and loading rates were very low, less than 0.2 - 0.3 mg/l and  $1 - 2 \mu g$  Al/kg tailings/day, respectively, as the acidic drainage was absent.

The effluent manganese concentrations and loading rates were also low but at detectable levels, in the ranges of 0.1 - 0.5 mg/l and  $1 - 2 \mu g$  Mn/kg tailings/day, respectively, and were indicative of oxidation activities within these tailings-limestone mixtures. As stated previously, the effluent Eh and pH values were near the manganese precipitation boundary which affected Mn solubility in the effluent. During the study period, the column average cumulative manganese removal was calculated as 0.3 mg Mn/kg tailings or approximately 3 % of total manganese contained in the tailings-wet ground limestone mixture.

### U, Th and Pb:

For the coarse tailings - wet ground limestone sub-group, the effluent U, Th and Pb leaching profiles are shown in Figures H-12 to H-14 for concentrations and in Figures 180 - 182 for loading rates, respectively. The results for uranium indicated some initial mobilization during the first year, where concentrations and loading rates increased from below detection to ranges of  $\sim$  1 - 2 mg/l and 4 -10 µg U/kg tailings/day, and decreased gradually to below detection levels in the third year (Figure H-12 and 180). Similar to iron and aluminum, no mobilization was observed for thorium and lead in the absence of the acidic drainage.

## Other Metals Ce, Cu, Ni and Zn:

The leaching characteristics of other metals: Ce, Cu, Ni and Zn are given in Tables H-1 and H-2, where in the absence of the acidic drainage, these metals were not mobilized from the coarse tailings mixed with wet ground limestone.

# **Ra-226:**

The effluent Ra-226 leaching characteristics are shown in Figure H-15 for concentrations (activities) and in Figures 183 and 184, respectively, for effluent loading rates and cumulative production/removal profiles, for coarse tailings mixed with wet ground and pulverized limestone.

Generally, the effluent Ra-226 concentrations (activities) increased gradually with time from an initial range of ~ 500 - 1000 mBq/l to 1200 - 1500 mBq/l during the study period. During the first year and at the start of the batch leaching schedule, two concentration peaks at ~ 1200 mBq/l and 2000 mBq/l, respectively, were also observed (Figure H-15). Similarly, the effluent loading rates increased from approximately 2 mBq/kg tailings/day to 4 mBq/kg tailings/day with intermediate peak loading rates at ~ 4 and 8 mBq/kg tailings/day (Figure 183). The occurrence of these peaks was coincident with those for calcium and magnesium. During the entire study period, the column average cumulative Ra-226 production/removal (loading) was calculated as approximately 5900 mBq/kg tailings or 0.16 % of total Ra-226 contained in the tailings - limestone mixture. The cumulative Ra-226 removal was comparable to coarse tailings without limestone amendment (Control - 2), and less than that for the coarse tailings - coarse limestone sub-group. For all group - 2 coarse tailings-limestone mixtures, the radium removals were more than double of those for group - 1 total mill tailings, although the Ra-226 contents of latter tailings sub-group were higher.

For coarse tailings without or mixed with limestone, there appeared to be no correlation between total Ra-226 removal and effluent acidity/alkalinity.

## 3.3.2 Submerged Tailings

## **3.3.2.1 Group 3: Submerged Coarse Tailings**

# 3.3.2.1.1 Group 3: Un-oxidized Coarse Tailings Submerged Under 0.45 m Water Cover (Columns #22-24)

The leaching characteristics of submerged coarse tailings are shown in Figures I-1 to I-16 for concentrations, and in Figures 186 - 209 for primary parameters, loading rates and cumulative mass removal profiles for various ions and radionuclides. As mentioned in the experimental methods section, these columns were leached under submerged conditions by continuous addition of distilled water at the top of the columns, letting the water run through the tailings and collecting as porewater drainage. The planned leaching scheme consisted of accelerated leaching at the rate of 1 I/day porewater flow for the first two years for removal of water soluble minerals (i.e. gypsum), followed by decreased porewater flow at the rate of 1 I/week. In the actual practice, because of manual flow control valves, the effluent volume varied between ~ 0.4 1 to 1 I/day with an yearly cumulative effluent flow volume of approximately 190 1 for the first two years, followed by 100 I/y for the balance of the study period. At the end of the four year study period, a column average cumulative flow volume of 580 1 of effluent was collected for each column, which represented approximately 270 pore volume exchanges. This leachate volume was approximately 7 times more than that used for leaching/rinsing purposes for the other unsaturated tailings and waste rock groups.

It should also be mentioned that because of the manual flow control valves, there had been occasional difficulty in maintaining a constant height of the water cover above the tailings. In one or two instances, around the middle of study the period, the water cover fell close to the tailings surface. The problem was quickly corrected by increasing the distilled water inflow appropriately and frequent monitoring of water levels in the columns. The inflow of the distilled water was, however, not metered in this study.

With the accelerated porewater flow, the soluble minerals (i.e. gypsum) were mobilized very rapidly and were mostly depleted, within a short time interval of initial three months, with a

cumulative effluent volume of approximately 50 l. During the first three years, the submerged coarse tailing produced no acidic drainages. The effluents were nearly neutral with pH's in the range of ~ 6.5 - 7.5 and zero to very low acidities. There was no mobilization of oxidation reaction products most notably Fe, Al, Mn and Th during this period. In the fourth year, however, the columns started to produce slightly acidic drainages with dissolved iron and manganese concentrations in the ranges of 2 - 20 mg/l and 0.1 - 0.3 mg/l, respectively. Also, there was about a 10 - 20 fold increase in Ra-226 mobilization from the submerged tailings. Further, the effluents had low dissolved oxygen, where concentrations decreased from near saturation, ~8 - 10 mg/l, in the well oxygenated feed water to approximately 2 - 4 mg/l in the drainage effluents. The water columns above the tailings surfaces were well oxygenated and there were no oxygen gradients. As well, there was no evidence of sulphate reduction in the submerged tailings, as noted by the absence of characteristic H<sub>2</sub>S odour in the effluents.

The submerged tailings were leached directly with distilled water which lacked the natural alkalinity and organic matter contents of natural run-off and lake waters. If the leaching water had contained alkalinity levels typical of the local run-off, some moderation in acidic drainage from submerged tailings was expected. Also, in the absence of any oxygen consuming organic layer covering the tailings, there was no other oxygen sink except tailings which were directly in contact with well oxygenated surface water, and to a certain extent porewater because of its accelerated flow rate. In the field, natural alkalinity and organic matter would be present in water covers of submerged tailings. The water column above the tailings was also monitored periodically at various depths and the results are given in Table I-4, together with drainage effluent water quality in Table I-1 to I-3 (Appendix I). The detailed results are as follows:

#### pH, Eh and Ec:

Figures 180 -188, show the drainage effluent pH, Eh and Ec profiles for submerged coarse tailings (columns #22-24). Initially during the first two years, the effluents were near neutral to slightly alkaline with pH's in the range of 7.5 and 8. Afterward, the pH's started to decrease gradually, and near the end of the study period, the effluent pH's were in the range of  $\sim 5.5$  -

7.0. As seen from Figure 186, it was observed that column #24 was producing slightly more acidic effluents, which indicated an early start of oxidation, than the other two columns. This column also had early occurrences of acidity, total iron, manganese and Ra-226, as discussed below. Because many of these changes were occurring near the end of the study period, it was difficult to predict if the oxidation process would continue and would also occur in the other two columns.

In the water columns above the tailings surface, the pH's were fairly uniform and decreased slightly with time from a column average of 5.7 to 5.0. These pH's were in the range of aerated distilled water containing dissolved carbon dioxide (Table I-4).

The drainage (porewater) effluent Eh's were fairly uniform around 400 mV with a slight decreasing trend from an initial range of 450 - 500 mV in the first year to 350 - 400 mV in the final year of the study. Similarly, in the water column above the tailings, the Eh's were in the well oxygenated range of 500 - 600 mV.

The effluent electrical conductances, Ec's, corresponded to rapid dissolution and leaching of soluble minerals, most notably gypsum, during the initial three month period. It increased rapidly from initial values of ~ 1000  $\mu$ S/cm to peak conductances in the range of ~ 2100 - 2400  $\mu$ S/cm during the first month, and then decreased during two to three month period that followed to constant conductances in the range of approximately 70 - 100  $\mu$ S/cm (Figure 188). Generally, the drainage Ec's were fairly uniform, following the initial gypsum dissolution peaks, during the rest of the study period with minor fluctuations occasionally.

In the water columns above the tailings, the electrical conductance peaked at a column average of approximately 115  $\mu$ S/cm around the middle of the study period, decreasing afterwards to 10 - 15  $\mu$ S/cm range, which was slightly above the feed water range of 3 - 6  $\mu$ S/cm. Because of the high power water flow and flushing of the soluble minerals, the water columns were not monitored for the first two years. In the following years, there appeared to be insignificant

upward diffusion of oxidation reaction products from submerged tailings to the water column above, although a limited oxidation was taking place at the surface of the tailings at the interface.

## **Acidity and Alkalinity:**

The acidity and alkalinity profiles for the effluent drainage are shown, respectively, in Figures I-4 and I-5 for concentrations, and in Figures 189 - 191 for acidity loading rates, cumulative acidity production and alkalinity loading rates respectively.

At the start of the experiments, columns #22-24, produced slightly acidic effluents which were flushed within the first few weeks. Since then, and for the next three years, the drainage effluent acidities were very low to zero. At the beginning of the fourth year of the underwater leaching study, the columns started to produce slightly acidic effluents, most notably column #24, in the range of ~ 10 - 20 mg/l CaCO<sub>3</sub> acidity. During this period, the drainage effluent acidity loading rates were in the range ~ 500 - 1500  $\mu$ g CaCO<sub>3</sub>/kg tailings/day (Figures I-4 and 189). During the entire study period, the column average cumulative acidity production/removal (loading) was calculated as 0.26 g CaCO<sub>3</sub>/kg tailings or 0.2 % of total acid generation potential of the coarse tailings.

The total acidity produced by the submerged coarse tailings is to be compared with those for Control - 1, total mill tailings, and Control - 2, coarse tailings, without limestone amendments where the acidities were, respectively,  $1.9 \text{ g CaCO}_3$ /kg tailings and 76.7 g CaCO<sub>3</sub>/kg tailings. During the experimental leaching period, a shallow distilled water cover on coarse tailings in the laboratory studies reduced acidity production by factors of approximately 10 and 300 in comparison to those for total mill tailings and coarse tailings leached under unsaturated conditions, respectively. However, for total mill tailings mixed with 7.5 % limestone of various sizes, and also leached under unsaturated conditions, the total acidity production was only 0.04 g CaCO<sub>3</sub> /kg tailings. The latter combination was thus more effective in preventing acidic drainage, for a shorter time period, than water cover on coarse tailings. The benefits of

limestone amendments in the surface layer of the tailings, however, would be only temporary unless appropriate quantity of limestone was incorporated in the tailings above the minimum elevation of the phreatic surface for complete acid neutralization purposes.

Initially during the first year, the submerged tailings were producing alkaline effluents with column average alkalinity in the range of ~ 40 - 100 mg CaCO<sub>3</sub> /l, which gradually decreased in subsequent years to a range of approximately 8 - 10 mg CaCO<sub>3</sub> /l at the end of the study. During the same period, the corresponding alkalinity production rate decreased from 8000 to 500  $\mu$ g CaCO<sub>3</sub>/kg tailings/day. As was observed for other leaching columns under unsaturated conditions, most of the alkalinity production resulted from the dissolution of residual limestone in the coarse tailings, estimated to be approximately 2 g CaCO<sub>3</sub> /kg tailings (2 kg CaCO<sub>3</sub> /tonne tailings or 0.2 %) from drainage effluent alkalinities (Figures I-5 and 191). The total available neutralization potential for coarse tailings was measured as 2.7 kg CaCO<sub>3</sub> /tonne tailings.

In the water cover above the tailings, the column average acidity and alkalinity were in the range of 3 - 6 mg CaCO<sub>3</sub> /l, mostly resulting from aerated distilled water and absorption of atmospheric gases, e.g.  $CO_2$  etc.

#### **Sulphate and Iron:**

The drainage effluent sulphate and iron leaching profiles for submerged tailings are shown, respectively, in Figures I-6 and I-7 for concentrations, in Figures 192 and 194 for effluent loading rates, and in Figures 193 and 195 for cumulative mass removal profiles for total sulphate and iron, respectively.

Similar to Ec's, the effluent sulphate concentrations were gypsum solubility derived and were quickly depleted from peak concentration of ~1500 mg/l to a range of ~ 10 - 20 mg/l during the first three months. For the remainder of the study period, sulphate concentrations were fairly constant at the lower range of ~ 10 - 20 mg/l. The corresponding column average effluent sulphate loading rates were approximately 70,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day at the peak and

between 200 - 400  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg tailings/day following gypsum removal (Figures I-6 and 192). For the duration of the study period, the columns average cumulative sulphate production/removal (loading) was estimated as approximately 2.6 g SO<sub>4</sub><sup>-2</sup>/kg tailings or 2.0 % of total sulphur contained in the coarse tailings. The corresponding total sulphate loading for coarse tailings under unsaturated leaching conditions was 87.9 g SO<sub>4</sub><sup>-2</sup>/kg tailings or ~ 67% of total sulphur content.

The total amount of sulphate removed in the drainage effluent, 2.6 g  $SO_4^{-2}$ /kg tailings, for submerged tailings corresponded to approximately 0.87 g soluble sulphur (as S) per kg tailings or 0.09 % total soluble sulphur, which was equivalent to 0.38 % CaSO<sub>4</sub> (gypsum) or 0.11 % Ca as soluble gypsum. These values were closely comparable to those measured in bulk coarse tailings samples as shown in Table 14.

The drainage effluent total iron concentrations were zero to very low during the first three years of the study, and increased gradually in the fourth year in effluents from all column, to a range of  $\sim 1 - 20$  mg/l. As mentioned previously, the largest iron drainage was observed from column #24, ~ 5 - 20 mg/l, followed by columns #23 and #22 in that order, where the effluent iron concentrations were in the range of  $\sim 0.1 - 1.0$  mg/l. Near the end of the study period, the iron concentrations in effluents #24 had from column peaked at ~ 20 mg/l and then decreased to a range of ~ 6 - 8 mg/l (Figures I-7). The corresponding drainage effluent iron loading rates were in the column average range of ~ 50 - 200  $\mu$ g Fe/kg tailings/day in the fourth year (Figure 194). The column average cumulative iron production/removal (loading) for the duration of the study period was calculated as approximately 29 mg Fe/kg tailings/day or 0.07 % of total iron contained in coarse tailings.

The results clearly indicated that for coarse tailings under a shallow water cover, oxidation and iron mobilization processes have established, although, to a very limited extent. However, in comparison to Control - 2, coarse tailings, where total iron removal was very high at 68.9 %, the total iron mobilization and removal from the submerged tailings was insignificant at 0.07 %.

For Control - 1, total mill tailings, total iron removal was also low at approximately 0.33 %. No iron mobilization was observed for total mill tailings mixed with 7.5 % limestone of various screen sizes and coarse tailings mixed with wet ground limestone.

Similar to total effluent acidity results, the performance of a shallow distilled water cover on coarse tailings was second to total mill tailings mixed with limestone, for a limited time period, when drainage effluent water quality results were compared.

In the water column above the tailings, dissolved total iron and sulphate concentrations were at background levels, in ranges of 0.01 - 0.5 mg/l and 3 - 8 mg/l, respectively.

#### **Calcium and Magnesium:**

For submerged columns, the leaching profiles for calcium and magnesium are shown, respectively, in Figures I-8 and I-9 for concentrations, in Figures 196 and 198 for drainage effluent loading rates, and in Figures 197 and 199 for cumulative mass removal characteristics.

Similar to Ec and sulphate leaching characteristics, the effluent calcium drainage was controlled by gypsum dissolution, reaching a peak calcium concentration of approximately ~ 600 mg/l during the first month, and decreasing very rapidly afterwards to a range of approximately 10 -15 mg/l during two to three months period that followed. For the remainder of the study period, calcium concentrations were constant at the above low range (Figure I-8). Correspondingly, the drainage effluent calcium loading rates were approximately 30,000 µg/kg tailings/day at the peak and between 100 - 300 µg/kg tailings/day range elsewhere (Figure 196). For the duration of the study period, the column average cumulative calcium production/removal (loading) was calculated as approximately 1.3 g Ca/kg tailings or 32 % of total calcium contained in the coarse tailings. Most of the calcium removed resulted from the dissolution of gypsum from the tailings which contained ~ 1.1 g Ca/kg tailings or 0.11 % Ca as gypsum. The cumulative calcium removed from submerged tailings was higher than that from Control - 2, unsaturated coarse tailings, where it was approximately 19 %. The drainage effluent magnesium concentration and loading rate profiles were also similar to those for calcium and SO<sub>4</sub><sup>-2</sup>, but for Mg the drainage peaks lasted for a shorter duration and were at ~ 16 mg/l and 700  $\mu$ g Mg/kg tailings/day, respectively. After the first three months, magnesium concentrations and loading rates decreased to ranges of ~ 0.1 - 0.3 mg/l and 2 - 10  $\mu$ g Mg/kg tailings/day, respectively. The magnesium data indicated slightly increase drainage in the third year, perhaps a indicating an onset of very limited oxidation (Figure 198). The drainage effluent cumulative magnesium production/removal (loading) during the study period was calculated as approximately 20 mg/kg tailings/day or 3.4 % of total magnesium contained in coarse tailings. Approximately half of the total magnesium removed resulted from the initial dissolution of readily soluble magnesium salts contained in the tailings. For comparison, the column average magnesium removed from Control - 2 unsaturated coarse tailings, which produced acidic drainage, was approximately 13 %.

In the water column above the tailings surface, the dissolved calcium and magnesium concentrations were very low in ranges of 0.1 - 1.0 mg/l and 0.08 - 0.1 mg/l, respectively.

## **Aluminum and Manganese:**

Figures I-10 and I-11, show respectively, the drainage effluent aluminum and manganese concentration profiles, Figures 200 and 202, the corresponding effluent loading rates, and Figures 201 - 203 the cumulative mass removal profiles, respectively.

The drainage effluent aluminum concentrations were very low to below detection levels, < 0.03 mg/l, during most of the study period, except for a few weeks at the beginning of the study and occasional small peaks where dissolved aluminum concentrations were less than 0.5 mg/l (Figures I-10). Similarly, the effluent aluminum loading rates were less than 3 µg/kg tailings/day range except a few peak loading rates in the range of approximately 10 - 30 µg/kg tailings/day (Figure 200). The corresponding column average cumulative aluminum production/removal (loading) was calculated as 6 mg Al/kg tailings or less than 0.02 % of total

aluminum contained in coarse tailings. Thus, for submerged coarse tailings aluminum removal was insignificantly small.

The drainage effluent manganese profiles were combinations of those for calcium and iron. Initially, during the first three months and similar to gypsum, soluble manganese salts were removed with peak manganese concentrations and loading rates in ranges of 0.6 - 0.8 mg/l and  $25 - 30 \ \mu g$  Mn/kg tailings/day, respectively. In the period that followed, the effluent manganese levels were near background or below detection for approximately six months, and increased slightly in drainage effluents from all three columns, at different times, to concentrations and loading rates in ranges of ~ 0.1 - 0.4 mg/l and 5 - 20  $\ \mu g$  Mn/kg tailings/day, respectively (Figures I-11 and 202). Similar to iron, the effluent manganese drainage was most significant from column #24 where it increased to peak concentrations and loading rates of approximately 0.4 mg/l and 20  $\ \mu g$  Mn/kg tailings/day at the end of the third year, and decreased gradually to 0.2 mg/l and 5  $\ \mu g$  Mn/kg tailings/day, respectively, in the fourth year. During the entire study period, the column average cumulative manganese production/removal (loading) was calculated as 4 mg Mn/kg tailings or 14 % of total manganese contained in coarse tailings. It was estimated that approximately 50 % of the observed Mn drainage resulted from surface oxidation of tailings underwater.

Similar to iron, the slight drainage of manganese in the effluent indicated active oxidation and Mn mobilization at very low rates in the submerged tailings. During this period, the dissolved concentrations of Fe and Mn were controlled by effluent Eh - pH phase stability relationship which, for example, indicated  $Mn^{+2}$  solubility and Fe near Fe(OH)<sub>3</sub> precipitation boundary for column #24, whose drainage effluent had a pH of ~ 5.5 and Eh ~ 350 mV. During the last year of the study the effluent sample bottles for column #24 were stained with characteristic Fe(OH)<sub>3</sub> orange colouration.

In the upper-lying water column above the tailings, aluminum was detected in trace amounts, 0.03 - 0.6 mg/l and manganese concentrations were very low, to below detection levels.

### **Uranium, Thorium and Lead:**

The drainage effluents leaching profiles for submerged coarse tailings for uranium, thorium and lead are shown in Figures I-12 to I-14 for concentrations, and in Figures 204 to 206 for effluent loading rates, respectively.

The effluent uranium concentrations were slightly elevated and above detection limits, approximately 0.3 - 0.4 mg/l, for the first few weeks at the very beginning of the experiment, where residual uranium salts from the tailings were mobilized. Since then, the uranium concentrations were fairly uniform and below detection levels, <0.2 mg/l, during the rest of the study period. The drainage effluent loadings were similarly near background levels, < 10  $\mu$ g U/kg tailings/day, except during the initial leaching period of few weeks, where the loading rates were in the range of ~ 20 - 25  $\mu$ g U/kg tailings/day. Because of flow variations and occasional detection of uranium in the effluent, some fluctuations in both parameters were observed (Figures I-12 and 204).

The drainage effluent thorium and lead concentrations were generally below detection levels and no loadings above background levels were noted. Because of low concentrations and loadings of these trace elements, no cumulative mass removal profiles were plotted.

## Other Metals Ce, Cu, Ni and Zn:

The leaching results for other metals, Ce, Cu, Ni and Zn, are given in Tables I-1 to I-3 (Appendix I) for submerged coarse tailings. Similar to thorium and lead, no mobilization of these metals in the drainage effluent was observed.

In the water column above the tailings surface, the concentrations of U, Th, Pb, Ce, Cu, Ni and Zn were very low to below detection levels.

## <u>Ra-226:</u>

The leaching characteristics of Ra-226 for the submerged coarse tailings are shown in Figure I-15, for concentrations (activities), and in Figures 207 and 208, respectively, for effluent loading rates and cumulative removal profiles.

For submerged coarse tailings, Ra-226 leaching characteristics resembled those of manganese, where ionic mobilites were observed both at the beginning of the experiment and near the end of the study period. Initially, at the start of the leaching and accelerated porewater flow, the column average Ra-226 concentrations in the drainage effluent decreased very rapidly, from approximately 5000 mBq/l to 1000 mBq/l during the first three months. During this period and unlike gypsum dissolution characteristics, no Ra-226 concentration peaks were observed. For the next two years or so, Ra-226 concentrations in the drainage effluents were fairly constant in the range of ~ 800 - 1000 mBq/l, which increased subsequently to peak concentrations in the range of ~ 5000 - 27,000 mBq/l near the end of the study period. The increase was initially gradual and then very rapid, and was most significant for column #24, where Ra-226 concentrations in the drainage effluents increased by approximately 25 times, followed by columns #23 and #22 (Figure I-15). The increase in Ra-226 drainage was coincident with acidic conditions occurring in the leachates. The corresponding Ra-226 loading profiles were similar, with column average loadings rates during initial, intermediate and end of the study periods were  $\sim 200, 40$  and 800 mBq Ra-226/kg tailings/day, respectively (Figure 207). For column #24, the peak loading rate was approximately 1700 mBq/kg tailings/day.

The cumulative column average production/removal (loading) for Ra-226 removal during the study period was calculated as ~ 215,000 mBq/kg tailings or 5.5 % of total Ra-226 contained in coarse tailings. Of the cumulative total Ra-226 removed; approximately 0.23 % was associated with the initial removal of gypsum, 1.5 % with intermediate leaching at neutral pH conditions, and 3.7 % with the on-set of slow oxidation conditions underwater and concurrent with the increased drainage iron in the effluent. The last process, however, was most significant for #24. Thus, comparison column in to Control - 1, total mill tailings and Control - 2, coarse tailings without limestone amendments, where cumulative Ra-226 removals with acidic drainages under unsaturated conditions were, respectively, 0.08 % and 0.17 %, Ra-226 drainage from coarse tailings under submerged conditions increased by factors of ~ 69 and 32, respectively.

For submerged coarse tailings, there were many interesting and important features associated with the leaching of Ra-226. Firstly, in the initial gypsum dissolution phase during accelerated leaching, the effluent Ra-226, gypsum and hence sulphate concentrations decreased simultaneously. During this period, the residual gypsum in the tailings was mostly depleted within the first three months, where the corresponding effluent sulphate concentrations decreased from approximately 1500 mg/l to 10 - 20 mg/l, and Ra-226 concentrations decreased from 5000 mBq/l to 1000 mBq/l. There was no increase in the solubility and mobilization of Ra-226 with decreasing sulphate concentrations in the gypsum removal process, where both Ra-226 and sulphate concentrations were controlled by gypsum dissolution. Secondly, during the intermediate leaching period, and in the absence of gypsum, where effluent pH's were nearly neutral, both sulphate and Ra-226 concentrations were fairly constant. Finally, near the end of the study period, where a slow oxidation process had started, there was a significant increase in Ra-226 concentration in the drainage effluent which was associated with the development of slow oxidation and slight acidic conditions at the tailings surface, and coincident with the mobilization of iron in low concentration in drainage effluents. During this period, sulphate concentrations had increased slightly but the change was not statistically significant.

These observations were in non-conformity with the most commonly held view that in a simple solid-liquid phase partition, the liquid phase Ra-226 solubility was controlled and inversely related to the sulphate ion concentration. However, with the development of slow oxidation and acidic conditions in submerged coarse tailings under a shallow water cover, the large increase in Ra-226 release was most probably associated with the mobility of parent Th-230 isotope but in low chemical (molecular) concentration similar that of iron. Perhaps, during this period, radium solubility was high because of low sulphate concentrations. As described previously, the

association of radium and thorium mobilities with oxidation and acidic conditions was also observed in other unsaturated tailings sub-groups. However, the above mentioned Ra/Sulphate inverse solubility relationship was observed to be valid only for severely acidic effluents from unsaturated coarse tailings (Control - 2, columns #16 and 17).

The increased effluent loading rate and cumulative removal of Ra-226 from submerged tailings, each by a factor of 32, in comparison to Control - 2 tailings did not correspond to the increased porewater in the former case. The cumulative effluent flow volumes for the two cases were, respectively, 580 l and 75 l, contributing a volume increase factor of ~ 8 for the submerged tailings. As noted above, the slow oxidation and acidification of submerged tailings at the surface increased Ra-226 mobility significantly.

In the water column above the submerged tailings, some upward diffusion of Ra-226 was observed, where dissolved Ra-226 concentrations varied between 30 - 140 mBq/l. Compared to Ra-226 concentrations in porewater drainage effluents, the surface water concentrations were low and below the regulatory guidelines of 1000 mBq/l for effluent discharge.

Although the leaching experiments for submerged tailings were conducted with distilled water, which lacked in natural alkalinity and organic matter contents, the radium mobility results post gypsum depletion implied a futuristic scenario of increased radium leachability and release from underwater deposited tailings upon development of acidic conditions. In shallow water and in the absence of an oxygen consuming cover layer or barrier such as organic rich sediments, the surface at the tailings - water interface is subjected to well oxygenated water and is thus amenable to oxidation and acidification. In the presence of gypsum, the radium mobility and release are limited and controlled by gypsum dissolution irrespective of tailings oxidation. However, post gypsum dissolution and depletion, further oxidation and acidification of tailings may lead to increased release of radium isotopes to both surface and ground water regimes.

This scenario may occur many years past decommissioning and monitoring phases, and should be given due consideration in environmental impact assessment and modeling.

It is believed that in the field, the natural alkalinity of run-off water and gradual development of an organic cover layer at the surface of the submerged tailings may play a significant role in controlling the surface oxidation and acidification of the tailings, and hence radium drainage. Additional leaching tests using natural lake water and an organic cover layer on gypsum depleted submerged tailings will further improve the understanding of radium dissolution and its partitioning dynamics amongst various solid-liquid phases.

## 3.3.3 Crushed Waste Rock

# 3.3.3.1 Group - 4: Un-oxidized Crushed Waste Rock (Unsaturated Conditions)

The leaching results for the three types of crushed waste rock (particle size - 2.0 mm) are given in Appendices J, K, and L, Tables J(1-3) to L(1-3) and in Figures J(1-16) to L(1-16), respectively, for medium (NNP = -10.7 kg CaCO<sub>3</sub>/tonne), high (NNP = -17.8 kg  $CaCO_3/tonne)$  and low (NNP = +13.8 kg CaCO<sub>3</sub>/tonne) acid generating waste rock. As mentioned previously in the experimental method section, the waste rock was only leached under unsaturated conditions, similar to those for the two types of tailings, and the drainage effluent monitored similarly. The study period lasted for four and a quarter years, during that time each column received an average of 104 l of natural lake water for leaching/rinsing purposes and produced a cumulative effluent volume of approximately 76 l.

The leaching characteristics of the waste rock varied according to its neutralization potential and were different for the three waste rock sub-groups. For both medium (columns #25 and 26) and low (columns #29 and 30) acid generating waste rock, no acidic drainage was observed during the entire study period, although their net neutralization potentials (NNP's) were on

different sides of the scale, as mentioned above. The leaching characteristics were quite similar to those of coarse tailings mixed with 7.5 % pulverized limestone (columns #20 and 21), as well the results indicated that oxidation, acid generation and neutralization processes were all active within these waste rock sub-groups. For the waste rock with a high acid generation potential (columns #27 and 28), no acidic drainage was observed during the first three years of the study. The occurrence of acidic drainage started at the end of the third year and severe acidic conditions prevailed thereafter. The leaching characteristics of the high acid generating waste rock resembled those of coarse tailings mixed with 7.5 % coarse limestone where severe acidic drainage conditions developed after two years of leaching. The detailed results for the waste rock sub-groups are given below. Because of similarities between coarse tailings and waste rock sub-groups, greater details are discussed only where significant differences existed among them.

# 3.3.3.1.1 Group - 4: Un-oxidized Crushed Waste Rock with Medium Acid Generation Potential (NNP = -10.7 kg CaCO<sub>3</sub>/tonne, Columns #25 and 26)

The leaching characteristics of the medium acid generating waste rock are shown in Figures J-1 to J-16 (Appendix J) for concentration profiles, and in Figures 210 - 230, for effluent loading rates and cumulative mass or Ra-226 activity removal profiles respectively. As shown in Table 16, the acid-base accounting components of the medium acid generating waste rock were: total sulphur (as S) 0.82 %, equivalent total acid generation potential 25.77 kg CaCO<sub>3</sub>/tonne, total neutralization potential equivalent 15.06 kg CaCO<sub>3</sub>/tonne and net neutralization potential of - 10.7 kg CaCO<sub>3</sub>/tonne. As mentioned previously, the waste rock did not produce any acidic drainage during the four year study period.

# pH, Eh and Ec:

The drainage effluent pH, Eh and Ec profiles for the medium acid generating waste rock are given, respectively, in Figures 210 - 212. Throughout the study period, the effluents were near neutral to slightly alkaline with pH's in the range of  $\sim 7.2$  to 8.0 (Figure 210). The column

average effluent Eh increased from 300 mV to 400 mV during the initial six month period, remained fairly uniform at approximately 400 mV for the next two years, and afterwards increased gradually to approximately 500 mV (Figure 211). The effluent Ec's increased slightly during the first few months of the study, from 200 to 600  $\mu$ S/cm peak, and decreased slowly to approximately 400  $\mu$ S/cm in another few months. After the introduction of the batch leaching scheme in the second year, Ec's increased rapidly to a peak range of 1400 - 1700  $\mu$ S/cm and decreased slightly afterwards to a constant column average conductance of approximately 1250  $\mu$ S/cm. Similar to coarse tailings mixed with wet ground limestone (columns #20 and 21), the effluent Ec was derived and controlled by gypsum formation and its dissolution (Figure 212).

### Acidity and Alkalinity:

The drainage effluent acidity and alkalinity profiles for the medium acid generating waste rock are shown in Figures J-4 and J-5, respectively, and in Figures 213-214 for effluent acidity and alkalinity loading rates. The effluents had no acidities and were slightly alkaline. The column average effluent acidity and acidity loading rates were zero throughout the leaching period, except for one composite sample where the measured values were, respectively, 18 mg CaCO<sub>3</sub>/l and 170 µg CaCO<sub>3</sub>/kg tailings/day (Figures J-4 and 213).

The column average effluent alkalinity and alkalinity loading rates increased slightly from initial values of 100 mg CaCO<sub>3</sub>/l and 400  $\mu$ g CaCO<sub>3</sub>/kg tailings/day to peak values of approximately 160 mg CaCO<sub>3</sub>/l and 700  $\mu$ g CaCO<sub>3</sub>/kg tailings/day in the second year after the introduction of the batch leaching schedule, and decreased afterwards to uniform ranges of 60 - 70 mg CaCO<sub>3</sub>/l and 200 - 300  $\mu$ g CaCO<sub>3</sub>/kg tailings/day, respectively (Figures J-5 and 214). Similar to columns #20 and 21, coarse tailings with wet ground limestone, the effluent alkalinity was produced by the dissolution of carbonate minerals present in the waste rock.

#### Sulphate and Iron:

The sulphate and iron leaching profiles for the medium acid generating waste rock are shown, respectively, in figures J-6 and J-7 for concentrations, and in Figures 215 - 217, respectively, for effluent sulphate loading rates, sulphate mass removal characteristics and iron loading rates.

The drainage effluent sulphate concentrations increased, after the introduction of the batch leaching scheme, from an initial range of ~ 100 - 120 mg/l to 600 - 800 mg/l, during the first year that followed and remained fairly uniform thereafter (Figure J-6). The corresponding sulphate loading rates increased from column average loading rate of approximately 500 to 2500  $\mu$ g SO<sub>4</sub><sup>-2</sup> /kg tailings/day (Figure 215). During the four year study period, the column average cumulative sulphate production/removal (loading) was calculated as 2.9 g SO<sub>4</sub><sup>-2</sup> /kg tailings or approximately 11.9 % of total sulphur (as SO<sub>4</sub><sup>-2</sup>) contained in the medium acid generating waste rock. Most of the sulphate drainage resulted from complete acid neutralization by the available limestone alkalinity in the waste rock during the study period.

The results indicated that although some oxidation was taking place within the waste rock samples(as seen above from sulphate and Ec results), the available alkalinity provided complete acid neutralization preventing acidic drainage and release of iron and other metals. The effluent iron concentrations were below detection levels, < 0.01, to very low <0.05 mg/l, with negligible loading rates (Figures J-7 and 217).

## **Calcium and Magnesium:**

The drainage effluent calcium and magnesium profiles are shown, respectively, in Figures J-8 and J-9 for concentrations, in Figures 218 and 220 for effluent loading rates, and in Figures 219 and 221 for cumulative mass removal characteristics.

Similar to coarse tailings mixed with wet ground and pulverized limestone (columns #20 and 21), the effluent Ca,  $SO_4^{-2}$  and Ec leaching profiles were similar, gypsum derived and controlled by its dissolution. The effluent calcium concentrations increased gradually after the

introduction of the batch leaching schedule, from an initial range of ~ 30 - 80 mg/l to nearly constant drainage range of 300 - 400 mg/l, and were below the gypsum saturation limit of approximately 600 mg/l. The corresponding effluent loading rates increased from approximately 250  $\mu$ g Ca/kg tailings/day at the beginning of the experiment to 1200  $\mu$ g Ca/kg tailings/day near the end of the study period. The column average cumulative calcium production/removal (loading) during the study period was calculated as approximately 1.4 g Ca/kg tailings or 30 % of total calcium contained in the waste rock.

Stoichiometrically, approximately 1.2 g Ca/kg tailings or 25 % g total calcium contained in the waste rock was mobilized as a direct result of limestone neutralization and gypsum drainage. Because the effluents were not saturated with respect to gypsum, the average acid production or limestone neutralization rate was calculated as approximately 2600  $\mu$ g CaCO<sub>3</sub>/kg tailings/day equivalent from gypsum formation and its release. Theoretically at 100 % limestone neutralization efficiency, the available alkalinity in the medium acid generating waste rock would be sufficient for approximately 12 to 15 years of acid neutralization, corresponding to oxidation of 60 % of total sulphur contained in the waste rock. Thus, acidic drainage from such a waste rock would occur after approximately 12 years of exposure in a vertical pile configuration, similar to the column arrangement, where the drainage occurs only from the bottom. The waste rock, however, may produce acidic drainage early, depending upon the geometry of the waste rock pile and efficiency of the limestone neutralization.

The effluent magnesium concentration and loading rate profiles were also similar to those for the coarse tailings - wet ground limestone mixture. The column average magnesium concentrations peaked at ~ 4 mg/l during the first three months of leaching, then increased slowly near the end of the second year, and decreased afterwards to approximately 2 mg/l (Figure J-9). The corresponding column average magnesium loading rates were between 20 - 25  $\mu$ g Mg/kg tailings/day in the peak regions and approximately 5 - 6  $\mu$ g Mg/kg tailings/day elsewhere. During the experimental period, the cumulative total magnesium drainage was calculated as approximately 13 mg Mg/kg tailings or 0.8 % of the total magnesium contained in the waste

rock. The cumulative removal of magnesium was significantly less than that of calcium during the same leaching period.

## **Aluminum and Manganese:**

The drainage effluent aluminum and manganese profiles are shown, respectively in Figures J-10 and J-11 for concentrations and in Figures 222-223 for loading rates. Similar to iron, the effluent aluminum concentrations were low and less than 0.2 mg/l throughout the study period. The corresponding effluent aluminum loading rates were also low and less than 1  $\mu$ g Al/kg tailings/day, except near the end of the study period where they increased slightly to 2  $\mu$ g Al/kg tailings/day (Figures J-10 and 223).

The effluent manganese concentrations were slightly elevated at the beginning of the study period, in the range of  $\sim 1 - 2$  mg/l, which increased to peak manganese drainage in the range of 2 - 4 mg/l with the implementation of the batch leaching schedule. Manganese drainage slowly decreased to below detection after the second year, except one isolated instance where the concentration in one of the columns (column #26) was 16 mg/l (Figure J-11). The corresponding effluent manganese loading rates were also low, and during the first year varied in the range of 2 - 15 µg Mn/kg tailings/day (Figure 223). Because of low drainage rates for aluminum and manganese, no cumulative effluent loadings and metal removals were calculated.

#### Uranium, Thorium and Lead:

The leaching characteristics of uranium, thorium and lead for the medium acid generating waste rock are shown, respectively, in Figures J-12 to J-14 for concentrations, in Figures 224, 226 and 227 for effluent loading rates, and in Figure 225 for cumulative uranium removal profile.

The effluent uranium concentrations increased gradually in the first year, from below detection levels, < 0.02 mg/l, to peak levels in the column average range of 3 - 5 mg/l in the second year of the study, and decreased afterward to 1 - 2 mg/l. The effluent loading rates increased correspondingly to column average range of 15 - 20 µg U/kg tailings/day and then decreased to approximately 7 - 8 µg U/kg tailings/day. The column average cumulative uranium drainage

was calculated as approximately 15 mg U/kg tailings or 20 % of total uranium contained in the waste rock. Although the waste rock did not produce any acidic drainage, there was significant mobilization of uranium after the introduction of the batch leaching schedule (Figures J-12 and 224).

In the absence of acidic drainage, the effluent thorium and lead concentrations and loadings were below detection levels(Figures J-13, J-14, 226 and 227).

#### Other Metals Ce, Cu, Ni and Zn:

The leaching characteristics of other metals, Ce, Cu, Ni and Zn, are given in Table J-1. Similar to iron, thorium and lead, mobilization and drainage of Ce, Cu, Ni and Zn were not observed for the medium acid generating waste rock.

## <u>Ra-226:</u>

The leaching characteristics of radium-226 for the medium acid generating waste rock are shown, respectively, in Figure J-15 for concentrations and in Figures 228 and 229 for effluent loading rates and cumulative removal profiles.

Initially, during the first year, the effluent Ra-226 concentrations were practically constant in the range of 80 - 120 mBq/l, which increased after the implementation of the batch leaching schedule to approximately 200 mBq/l peaks. Near the end of the second year, Ra-226 concentrations again increased slowly from 50 mBq/l to a range of approximately 200 - 300 mBq/l during the rest of the study period (Figure J-11). The corresponding effluent Ra-226 loading rates increased from approximately 0.5 to 1.5 mBq Ra-226/kg tailings/day. During the entire study period, the column average cumulative Ra-226 drainage was calculated as approximately 1020 mBq/kg tailings or less than 0.04 % of total Ra-226 contained in the waste rock. Thus in the absence of acidic drainage from the medium acid generating waste rock, the mobility of Ra-226 was very low and controlled by dissolved gypsum. Similar results were

obtained for the two unsaturated tailings groups, coarse and total mill tailings mixed with limestone, where acidic drainages were also absent.

# 3.3.3.1.2 Group - 4: Un-oxidized Crushed Waste Rock with High Acid Generation Potential (NNP = -17.8 kg CaCO<sub>3</sub>/tonne, Columns #27 and 28)

The leaching characteristics of the high acid generating waste rock are shown in Figure K-1 to K-16 (Appendix K) for concentrations, and in Figures 231 - 257, for effluent loading rates and cumulative mass or activity (Ra-226) removal profiles, respectively. As shown in Table 17, the acid base accounting components of the high acid generating waste rock were: total sulphur (as S) 0.86 %, equivalent total acid generation potential (AP) 26.34 kg CaCO<sub>3</sub>/tonne, total neutralization potential (NP) equivalent 8.5 kg CaCO<sub>3</sub>/tonne, and net neutralization potential (NNP) of -17.8 kg CaCO<sub>3</sub>/tonne. As mentioned previously, this waste rock produced highly acidic drainage near the end of the third year of the study. The detailed results are as follows:

## pH, Eh and Ec:

Figures 231 - 233, show, respectively the pH, Eh and Ec profiles for the high acid generating waste rock sub-group, where the leaching characteristics were similar to those for coarse tailings mixed with coarse limestone (columns #18 and 19). During the first three years, the effluent pH's were near neutral to slightly alkaline ~ 7.5 - 8, which decreased very rapidly to a range of ~ 2.0 - 2.5 at the beginning of the fourth year with the start of a severe acidic drainage (Figure 231). The acidic drainage appeared early from column #28 and three months later from column #27. Similarly, at the on-set of acidic drainage, the effluent Eh's increased from approximately 400 mV to a range of ~ 750 - 800 mV and were reflective of severe oxidation and mobilization of iron (Figure 232). During the same period, the electrical conductances, Ec's, increased from column average range of 300 - 800  $\mu$ S/cm in the neutral effluents to 6000 - 8000  $\mu$ S/cm with the acidic drainage having peak conductances of approximately 9500  $\mu$  S/cm, which were observed after transition to acidic conditions had been complete (December 1992), as seen from pH and Eh results.

## Acidity and Alkalinity:

The effluent acidity and alkalinity profiles are shown, respectively, in Figures K-4 and K-5 for concentrations, in Figures 234 and 236 for effluent loading rates and in Figure 235 for cumulative acidity production. The effluent acidities were zero or absent (neutral to alkaline effluents) for the first three years and increased very rapidly, with the on-set of acidic drainage, to peak acidities in the range of ~ 18,000 - 20,000 mg CaCO<sub>3</sub>/l, decreasing afterwards to column average acidity of approximately 7000 mg CaCO<sub>3</sub>/l. The corresponding acidity loading rates were 60,000 - 65,000 and 30,000  $\mu$ g CaCO<sub>3</sub>/kg tailings/day, respectively (Figures K-4 and 234). With the occurrence of the acidic drainage at the beginning of the fourth year and during the balance of the study period, a column average cumulative acidity production/loading was calculated as approximately 13.9 g CaCO<sub>3</sub>/kg waste rock or 53 % of total acidic generation potential of the waste rock. Although, the waste rock contained lower concentrations of sulphide (pyrite) and available limestone alkalinity than coarse tailings, its reactivity was higher than that of coarse tailings mixed with coarse limestone fraction (columns #18 and 19).

The effluent alkalinities increased initially from a range of ~ 30 - 50 mg/l to peak concentrations of approximately 150 mg/l during the first year, and decreased slowly to zero at the on-set of acidic drainage (Figure K-5). The corresponding columns average alkalinity production rates were in the range of 200 to 600 µg CaCO<sub>3</sub>/kg waste rock/day equivalents, respectively. Similar to coarse tailings, the alkalinity was being produced by the dissolution of carbonate minerals. During the alkaline drainage period of initial three years, the corresponding cumulative alkalinity production was calculated as 0.44 g CaCO<sub>3</sub>/kg waste rock or approximately 3 % of total neutralization potential of the waste rock.

## Sulphate and Iron:

The leaching and mobilization characteristics of sulphate and iron for the high acid generating waste rock are shown, respectively, in Figures K-6 and K-7 for effluent concentrations, in Figures 237 and 239 for effluent loading rates and in Figures 238 and 240 for cumulative mass removal profiles, respectively.

The column average effluent sulphate concentration increased from an initial range of 80 - 100 mg/l, first gradually and then very rapidly with the occurrence of the acidic drainage, to a peak value of approximately 20,000 mg/l (average 16,000 mg/l). Near the end of the study period, the sulphate concentration decreased to approximately 8,000 mg/l. The corresponding effluent sulphate loading rates were 70,000 (60,000 average) at the peak and 38,000  $\mu$ g SO<sub>4</sub><sup>-2</sup> /kg waste rock/day near the end of the study period. For the duration of the study period, the column average cumulative sulphate production/removal (loading) was calculated as approximately 15.4 g SO<sub>4</sub><sup>-2</sup> /kg waste rock or 60 % of the total sulphur contained in the waste rock. Similar to total acidity, the corresponding sulphate production rate for the high acid generating waste rock sub-group was very significant, most of it resulting from oxidation and acid generation.

The effluent iron concentrations were zero to below detection levels, <0.01 mg/l, prior to the occurrence of the acidic drainage, and increased very rapidly in the acidic effluent to peak values in the range of ~ 7000 - 9000 mg/l (column average 6000 mg/l) during the fourth year of the study. The corresponding column average effluent iron loading rates were in the range of 20,000 - 25,000  $\mu$ g Fe/kg waste rock/day. The calculated total iron loading or cumulative iron production/removal during the acidic drainage period monitored was ~ 4.6 g Fe/kg waste rock or 31 % of total iron contained in the waste rock. The iron mobilization and release were also high for the waste rock.

## **Calcium and Magnesium:**

The leaching characteristics of calcium and magnesium are shown, respectively, in Figures K-8 and K-9 for concentrations, in Figures 241 and 243 for effluent loading rates, and in Figures 242 and 244 for cumulative metal removal profiles, respectively.

The effluent calcium concentrations for the high generating waste rock increased gradually during the initial two and a half years period, from approximately 50 mg/l to 200 mg/l, then very rapidly to peak calcium concentrations in the range of 800 - 900 mg/l (column average approximately 750 mg/l) near the on-set of the acidic drainage. At the end of the study period, the column average calcium concentration had decreased to 400 mg/l. The effluent calcium concentration peaks were concurrent with transition to acidic pH's, and their occurrences preceded those of iron and sulphate peaks by approximately six months. The corresponding column average effluent calcium loading rate increased from an initial range of 250 - 750  $\mu$ g Ca /kg waste rock/day to a peak value of 3200  $\mu$ g Ca /kg waste rock/day. During the entire study period, the column average cumulative calcium production/removal (loading) was calculated as approximately 1.59 g Ca/kg waste rock or 59 % of total calcium contained in the waste rock.

Prior to the on-set of acidic drainage (approximately June 1992), the effluent calcium drainage resulted from complete neutralization of the acid produced and formation of gypsum. At the peak concentration, the effluent was saturated with respect to gypsum and calcium concentration was controlled by gypsum dissolution. Using the observed calcium loading rates, the total limestone neutralization, prior to the occurrence of the acidic drainage from the waste rock, was calculated as approximately 2 g CaCO<sub>3</sub> (0.8 g Ca)/kg waste rock or 24 % of total limestone alkalinity present in the waste rock. Thus, similar to coarse tailings without and with added limestone (coarse), the acidic drainage from the occurred before complete utilization of the available alkalinity.

The effluent magnesium profiles were similar to those for sulphate and calcium, where Mg concentrations increased slowly at the beginning from a range of  $\sim 1 - 5$  mg/l, and then rapidly

1-100

with the on-set of acidic drainage to peak values in the range of 70 - 80 mg/l (column average 70 mg/l). Near the end of the study period, the column average magnesium concentration was approximately 50 mg/l (Figure K-9). It should be noted that magnesium concentration peaks were concurrent with those of sulphate and iron, and were preceded calcium by approximately six months. The corresponding effluent magnesium loading rates were in the range of ~ 10 - 30  $\mu$ g Mg/kg waste rock/day before acidic drainage, ~ 250 - 300 at the peak concentrations , and 160 - 200  $\mu$ g Mg/kg waste rock/day near the end of the study. The column average cumulative magnesium production/removal (loading) during the study period was calculated as approximately 0.08 g Mg/kg waste rock or 6.5 % of total magnesium contained in the waste rock. The reactivity of available magnesium alkalinity was less than that of calcium in the high acid generating waste rock. This result differed from that of coarse tailings mixed with coarse limestone, where magnesium removal was higher.

## **Aluminum and Manganese:**

The effluent aluminum and manganese profiles are shown in Figures K-10 and K-11 for concentrations, in Figures 245 and 247 for effluent loading rates, and in Figures 246 and 248 for cumulative mass removal characteristics, respectively.

The effluent aluminum profiles were similar to those for iron, where aluminum concentrations increased from below detection levels, <0.03 mg/l, to a peak range of ~ 350 - 400 mg/l with the acidic drainage, and then decreased to approximately 200 mg/l near the end of the study period. The corresponding column average effluent loading rates were less than 0.5  $\mu$ g Al/kg waste rock/day at the beginning, 1600-1800 near the peak drainage and 600-800  $\mu$ g Al/kg waste rock/day near the end of the study. During the acidic drainage period monitored, the column average cumulative aluminum production/removal (loading) was calculated as approximately 0.33 g Al/kg waste rock or 0.8 % of total aluminum contained in the waste rock. Similar to coarse tailings, the mobilization of aluminum with the acidic drainage was also limited in the waste rock.

The effluent manganese profiles resembled those of calcium, where Mn concentrations increased slowly at the beginning in the range of 0.1 - 0.5 mg/l, then rapidly to peak values of approximately 40 - 50 mg/l at the on-set of acidic drainage, and decreased to 5 - 8 mg/l range near the end of the study period. The effluent loading rates, similarly, increased to column average peak loading rate of approximately 150 µg Mn/kg waste rock/day and decreased rapidly to 20 µg Mn/kg waste rock/day afterwards. The corresponding column average cumulative manganese production/removal (loading) during the study period was calculated as approximately 64 mg Mn/kg waste rock or 69 % of total manganese removal with the acidic drainage was also high for the waste rock.

# **Uranium, Thorium and Lead:**

The leaching characteristics of uranium, thorium and lead for the high acid generating waste rock are shown, respectively, in Figures K-12 to K-14, for concentrations, in Figures 249, 251 and 253 for effluent loading rates, and in Figures 250, 252 and 254 for cumulative mass removal profiles, respectively.

The effluent uranium and thorium leaching characteristics were very similar to those of iron and aluminum, where all of them were significantly mobilized with the acidic drainage. Uranium concentrations in the effluents increased from below detection levels, <0.2 mg/l, at the beginning of the experiment to 1 - 5 mg/l in the neutral effluents, to peak concentrations in the range of 150 - 200 mg/l (column average 150 mg/l) with the acidic drainage, and then decreased to 10 -20 mg/l of the end the experiment range near (Figure K-12). The corresponding column average effluent loading rates were, ~ 550  $\mu$ g U/kg waste rock/day near the peak drainage and approximately 50  $\mu$ g U/kg waste rock/day near the end of the study period. During the entire study period, a column average cumulative production/removal (loading) of calculated uranium was as  $\sim$  0.10 g U/kg waste rock or approximately 88 % of total uranium contained in the waste rock. As mentioned in the analytical section, because of high uranium detection limit using the

ICAP-AES, the mass balance for uranium was not very accurate. Nonetheless, the mobility of uranium was very high in the high acid generating waste rock with the occurrence of the acidic drainage.

The thorium results were slightly different than those for uranium, where thorium mobility and release were entirely associated with the acidic drainage. Thorium concentrations in the effluent increased with the acidity, from below detection levels, <0.05 mg/l, to peak levels in the range of 200 - 300 mg/l and decreased to 5 - 10 mg/l near the end of the study period (Figure K-13). The corresponding column average effluent loading rates were, respectively, 1000 and 200  $\mu$ g Th/kg waste rock/day at the peak drainage and afterwards. During the acidic drainage period monitored, the cumulative thorium production/removal (loading) was calculated as ~ 0.18 g Th/kg waste rock or 83 % of total thorium contained in the waste rock. Similar to uranium, the thorium mobility and release were also high with the acidic drainage in the waste rock.

Similarly, the effluent lead concentrations increased with the acidic drainage from below detection levels, <0.06 mg/l, to peak concentrations in the range of ~ 2 - 3 mg/l and decreased to 0.5 mg/l level near the end of the study period. The effluent lead loading rates were, correspondingly, 12 - 15  $\mu$ g Pb/kg waste rock/day at the peak drainage and ~ 1 - 2  $\mu$ g Pb/kg waste rock/day near the end of the study period. With the acidic drainage, a column average cumulative lead production/removal (loading) was calculated as 1.8 mg Pb/kg waste rock or 1.8 % of total lead contained in the waste rock. Similar to coarse tailings, the lead mobility with the acidic drainage was low for the acid generating waste rock as well.

### Other metals Ce, Cu, Ni and Zn:

The leaching characteristics of Ce, Cu, Ni and Zn for the high acidic generating waste rock are given in Table K-1 and were similar to those of iron, aluminum and thorium where the metal releases were entirely caused by the acidic drainage. In the acidic effluents, the concentration peaks for these metals were, respectively, 50 mg/l for Ce,

12 mg/l for Cu, 10 mg/l for Ni and 10 mg/l for Zn. Similar to coarse tailings, the mobilities of these metals with the acidic drainage were significantly high.

## <u>Ra-226:</u>

Ra-226 leaching characteristics for the high acid generating waste rock are shown in Figure K-15 for concentrations (activities), and in Figures 255 and 256, respectively, for effluent loading rates and cumulative Ra-226 removal profiles.

The leaching profiles for Ra-226 for the waste rock were different than those for coarse tailings without or mixed with limestone, where for the high acid generating waste rock and in the absence of gypsum, the effluent Ra-226 concentrations also increased with the acidic drainage. In the neutral to slightly alkaline effluents, Ra-226 concentrations increased from an initial range of ~ 100 - 300 mBq/l to approximately 500 mBq/l, which increased with the acidic drainage to peak concentrations of 1000 - 2000 mBq/l (column average 1000 mBq/l) and then decreased rapidly to 200 - 300 mBq/l range near the end of the study period (Figure K-15). Similarly, the effluent Ra-226 loading rates increased from a range of ~ 0.5 - 1.0 mBq/kg waste rock/day to peak rates of ~ 4 - 8 mBq/kg waste rock/day. The column average cumulative production/removal (loading) of Ra-226 during the entire study period was calculated as approximately 2200 mBq/kg waste rock or 0.08 % of the total Ra-226 contained in the high acid generating waste rock. Similar to medium acid generating waste rock and coarse tailings, Ra-226 mobility and release in the present case were also very low.

# 3.3.3.1.3 Group - 4: Un-oxidized Crushed Waste Rock with Low Acid Generation Potential (NNP = +13.8 kg CaCO<sub>3</sub>/tonne, Columns #29 and 30)

The leaching results for the low acid generating waste rock (columns #29 and 30) are given in Tables L-1 to L-3 (Appendix L). The characteristics are shown in Figures L-1 to L-16 for concentration profiles, and in Figures 258 - 278 for effluent loading rates, and cumulative mass or activity (Ra-226) removal profiles. The acid base accounting components of the low acid generating waste rock were: total sulphur (as S) 0.42 %, equivalent total acid generation

potential (AP), 13.01 kg CaCO<sub>3</sub>/tonne, total neutralization potential (NP) equivalent, 26.82 kg CaCO<sub>3</sub>/tonne and net neutralization potential (NNP) of +13.8 kg CaCO<sub>3</sub>/tonne (Table 18). Similar to the medium acid generating waste rock, the low acid generating waste rock subgroup did not produce any acidic drainage during the four year study period, and the parametric profiles for the two waste rock sub-groups were quite similar. The description of results and discussions are thus limited to key features of this group.

## pH, Eh and Ec:

The drainage effluents for the low acid generating waste rock were slightly alkaline with pH's in the range 7.5 - 8 for the entire study period (Figure 258). The Eh's increased from approximately 300 mV at the beginning to 500 mV near the end of the study period (Figure 259). During the same time, the electrical conductances, Ec's, increased first gradually and then rapidly from approximately 250 to 500  $\mu$ S/cm, then to peak conductances of 1500 - 2200  $\mu$ S/cm in the second year, decreasing afterwards to 1000  $\mu$ S/cm level (Figure 260) near the end of the study period. The effluent was mostly controlled by gypsum formation in the limestone neutralization process and its subsequent dissolution.

## Acidity and Alkalinity:

The effluent acidities were zero throughout the experimental period of four years (Figure 261). The alkalinities increased initially during the first year from a range of ~ 50 - 60 mg CaCO<sub>3</sub>/l to 120 - 150 mg CaCO<sub>3</sub>/l, then decreased to uniform alkalinity levels of 70 - 80 mg CaCO<sub>3</sub>/l. Similar to medium acid generating waste rock (columns #25 and 26) and coarse tailings mixed with limestone, the effluent alkalinities resulted from dissolution of carbonate minerals contained in the waste rock (Figure 262).

# Sulphate and Iron:

Similar to Ec, the effluent sulphate concentrations were controlled by gypsum formation and its dissolution, and increased from a range of 100 - 200 mg/l during the first year to peak sulphate drainage in the range of 700 - 1300 mg/l, decreasing afterwards to 400 - 600 mg/l level (Figure

L-6). The corresponding column average effluent sulphate loading rates were approximately 500  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg waste rock/day at the beginning, 2500 - 4000 near the peak drainage and approximately 1500  $\mu$ g SO<sub>4</sub><sup>-2</sup>/kg waste rock/day at the end of the study period (Figure 263 and 264). The column average cumulative sulphate production/removal (loading) during the study period was calculated as 2.4 g SO<sub>4</sub><sup>-2</sup>/kg waste rock or 19 % of total sulphur contained in the waste rock. The cumulative sulphate loading of 2.4 g SO<sub>4</sub><sup>-2</sup>/kg waste rock for the low acid generating waste rock was comparable to that for medium acid generating waste rock, 2.9 g SO<sub>4</sub><sup>-2</sup>/kg waste rock, and indicated active oxidation, acid generation neutralization processes within the two sub-groups.

The effluent iron concentrations were generally below detection levels, < 0.01 mg/l, except in few instances where they were between 0.02 - 0.14 mg/l for one column (#29) at the beginning of the leaching period (Figure L-7). The corresponding effluent loading rates were insignificantly small (Figure 265).

## **Calcium and Magnesium:**

Similar to Ec and sulphate, the effluent calcium concentrations (Figure L-8) increased slowly during the first year from 20 to 100 mg/l, then rapidly to peak calcium drainage range of 350 - 500 mg/l and decreased afterwards, in the third year, to 200 - 300 mg/l range. The corresponding column average calcium loading rates were, respectively, between  $100 - 400 \mu g$  Ca/kg waste rock/day at the beginning, 1200 - 1700 in the peak drainage region and  $700 - 1000 \mu g$  Ca/kg waste rock/day near the end of the study period (Figure 266 and 267). The column average cumulative calcium production/removal (loading) was calculated as ~ 1.2 g Ca/kg waste rock or approximately 11.6 % of total calcium contained in the waste rock. Because of acid neutralization, gypsum formation and its dissolution, the effluent cumulative calcium loadings for low and medium acid generating waste rock comparable.

The effluent calcium profiles clearly indicated ongoing oxidation, acid generation and complete neutralization processes within the waste rock. The effluents were also not saturated with

respect to gypsum and most of calcium and sulphate resulted from limestone neutralization. As seen from alkalinity profiles, small amounts of calcium drainage, estimated to be approximately  $0.6 \text{ g CaCO}_3$  or 0.2 g Ca/kg waste rock, were produced by limestone dissolution. The balance of calcium and total sulphate loadings were then in stoichiometric equivalent compositions corresponding to gypsum dissolution.

Similar to the medium acid generating waste rock, the observed effluent calcium loading rates were used to calculate the average acid generation and hence limestone neutralization rates, which were obtained as  $2250 \ \mu g$  CaCO<sub>3</sub> or 900  $\ \mu g$  Ca/kg waste rock/day. Thus for low acid generating waste rock, the total available alkalinity was theoretically sufficient for 33 years of acid neutralization at 100 % limestone neutralization efficiency. At the above calculated acid generation rate, the total sulphur contained in the waste rock would be oxidized and mobilized within the first twenty years at an estimated rate of 5 % total sulphur/year. Thus the low acid generating waste rock, at 100 % neutralization efficiency, should not produce any acidic drainage. At lower neutralization efficiencies, however, acidic drainage may occur at some time in the future. The observed limestone neutralization efficiencies for high acid producing waste rock and tailings have seldom exceeded 25 % of the available alkalinity. It remains to be seen if the low acid generating waste rock would produce acidic drainage in the future, and further monitoring of these columns may be necessary.

The effluent magnesium concentrations (Figure L-7) increased in the first year to peak drainages of approximately 8 mg/l, then decreased to a range of  $\sim 2 - 3$  mg/l, and increased again to second drainage peaks, in the range of  $\sim 5 - 8$  mg/l, near the end of the second year. Afterwards, the concentrations decreased gradually to a range of  $\sim 1 - 2$  mg/l near the end of the study period. The effluent magnesium and calcium profiles were complimentary in indicating active oxidation-neutralization processes.

The corresponding effluent magnesium loading rates were, respectively, at 30 and 20  $\mu$ g Mg/kg waste rock/day for the two peak regions and approximately 8  $\mu$ g Mg/kg waste rock/day

elsewhere. The column average cumulative production/removal (loading) of magnesium was calculated as ~ 0.02 g Mg/kg waste rock or approximately 0.4 % of total magnesium contained in the waste rock. Similar to the medium acid generating waste rock, the cumulative removal of magnesium was considerably lower than that of calcium (Figures 268 and 269).

#### **Aluminum and Manganese:**

For the low acid generating waste rock, a minor amount of aluminum drainage was observed, although the effluent pH's were slightly alkaline (Figures L-10 and 270). The effluent aluminum concentrations were low and varied between 0.1 - 0.4 mg/l with corresponding loading rates in the range of  $0.5 - 1.0 \mu g$  Al/kg waste rock/day.

The effluent manganese concentrations and loadings were also low and were in the ranges 0.1 - 0.6 mg/l and  $1 - 2 \mu \text{g}$  Mn/kg waste rock/day, respectively. Similar to calcium and magnesium, the manganese drainage substantiated ongoing oxidation-neutralization processes within the low acid generating waste rock (Figures L-11 and 271).

#### Uranium, Thorium and Lead:

Similar to the medium acid generating waste rock, the effluent uranium concentrations (Figure L-11) increased from the initial low levels of <0.2 mg/l to 4 - 5 mg/l peak drainage and then decreased slowly to ~ 2 mg/l near the end of the study period. Similarly, the column average effluent loading rates increased from a very low drainage to 20 - 25  $\mu$ g U/kg waste rock/day near the peak region and between 8 - 10  $\mu$ g U/kg waste rock/day afterwards. In the alkaline effluent, and with carbonate buffering, the column average effluent uranium loading or cumulative uranium production/removal rock during the study period was calculated as ~ 16 mg U/kg waste rock or 32 % of the total uranium contained in the waste. Similar to the medium acid generating waste rock, significant amount of uranium in the present case was mobilized, even though in both cases acidic drainages were absent (Figures 272 and 273). For the low acid generating waste rock, no mobilization of thorium and lead was observed during the study period, as seen from Figures L-13, L-14, 274 and 275.

#### Other Metals Ce, Cu, Ni and Zn:

The leaching characteristics of other metals, Ce, Cu, Ni and Zn are given, together with other water quality results, in Tables L-1 and L-2 (Appendix L). Similar to iron, aluminum, thorium and lead, there was no mobilization of Ce, Cu, Ni and Zn, in the absence of acidic drainage from the low acid generating waste rock.

#### <u>Ra-226:</u>

The leaching characteristics of Ra-226 for the low acid generating waste rock are shown in Figure L-15 for concentrations (activities) and in Figures 276 and 277 for effluent loading rates and cumulative Ra-226 removal profiles, respectively.

The column average effluent Ra-226 concentrations increased slightly from an initial range of ~ 100 - 200 mBq/l to peak drainages between 300 - 450 mBq/l near the end of the first year, and then decreased to a range of ~ 100 - 150 mBq/l range. The corresponding effluent loading rates were low and varied between 0.5 - 1.0 mBq/kg waste rock/day (Figures 276 and 277). During the four year study period, the column average cumulative production/removal (loading) of Ra-226 was calculated as approximately 1100 mBq/kg waste rock or ~ 0.06 % of total Ra-226 activity contained in the waste rock. Thus, the mobilization and release of Ra-226 for the low acid generating waste rock were also low.

#### 4. DISCUSSION

The column leaching experiments were conducted under climatically controlled laboratory conditions of warm room temperature (22 °C) and cool fluorescent lighting, providing favourable conditions for both bacterial and chemical oxidations of sulphide minerals. Also, the

columns were subjected to a constant leaching schedule, designed to promote oxidation and drainage under unsaturated conditions and high porewater flow and oxygenation for submerged tailings. In the field, such conditions only exist during warm summer months where maximum reactivity and release of oxidation reaction products takes place. The laboratory test were thus simulations of enhanced chemical activity and leaching conditions.

In actual field conditions there are seasonal and other factors that need to be considered. For example, for unsaturated and exposed tailings and waste rock, additional variables are: diurnal surface temperature cycling, solar heating and drying, wind desiccation, freeze-thaw cycling and frost penetration, snow cover, and spring and fall recharge events, etc. For submerged tailings consideration should be given to wind/wave induced mixing and surface erosion, freezing of the water cover and tailings surface, ice formation and break-up, and temperature dependence of mineral solubilities etc. The laboratory results should, therefore, be weighted appropriately when comparing with those in the field under different conditions. The most useful and directly applicable leaching result for a given test sample is expressed in term of an effluent loading rate, as µg/kg tailings (waste rock)/day, for various ionic species and mBq/kg tailings (waste rock)/day for radionuclides. This result could be converted easily to kg/hectare/year for metals and sulphate and kBq/hectare/year for radionuclides by multiplying the corresponding column loading rates, obtained in this study, by 3.165 and 3,165 respectively. In converting these results it was assumed that the oxidation rate was limited by the available oxygen and hence the exposed surface area. For other tailings and waste rock, having different sulphide and mineral compositions, results normalized to unit elemental concentration should be used.

The laboratory experimental conditions of continuous weathering at warm temperature and batch leaching, in comparison to those existing in the field on degree-days basis, provided a 3 fold scaled up leaching period. Therefore, one year on the laboratory scale was approximately equivalent to three years of activity in the field. The unsaturated tailings and waste rock columns were rinsed/flushed with 1 l of natural lake water every two weeks or 26 l/year of water was applied to each column having a surface area of 0.0165 m<sup>2</sup>. This corresponded to an equivalent

annual precipitation of approximately 1600 mm. The average porewater drainage and hence infiltration per column was approximately 18.75 l or 1140 mm per year, and as there was no surface drainage, the balance, 460 mm/yea, resulted in surface evaporation. For a nominal porosity or void volume of 33 % for the tailings, the annual infiltration depth corresponded to 3.4 m or approximately 3 times that of the average field value. For both, the unsaturated tailings and waste rock sub-groups, the temperature and drainage conditions in the laboratory thus provided, on an actual time scale, accelerated weathering and leaching by factors of 3.

Similarly for submerged tailings, the accelerated weathering time factor of 3 applied equally, but for gypsum leaching and flushing requirements, the effluent drainage rates were high. In the experimental columns, containing coarse tailings to a height of approximately 0.35 m, gypsum was completely removed within the first fifty days with the application of 50 l of drainage water. In the actual field scenario, gypsum removal time may vary between 50 - 300 years, depending upon basin surface area, hydrology, tailings tonnage and their gypsum content, etc. Thus for submerged tailings, the Ra-226 mobilization results obtained in the laboratory would be applicable only after complete removal of gypsum from underwater disposed and decommissioned tailings. Hence, appropriate judgment should be exercised when applying laboratory results to field cases.

Although the detailed laboratory column leaching results have been discussed for each tailings and waste rock sub-group previously, a brief overview of salient features is provided below along with relevant discussion.

For unsaturated tailings, the coarse tailings without additional limestone (Control - 1) were most amenable to oxidation and acidic drainage. These tailings drained quickly, had a high hydraulic conductivity of 4 x  $10^{-3}$  cm/s and hence a high air entry value, and produced highly acidic drainage within two months of the introduction of a batch leaching schedule. During the acidic drainage period, the effluent average pH, Eh and Ec were, respectively, 1.5, 700 mV and 40,000 µS/cm. The effluent average loading rates for various species were: approximately 70,000 µg CaCO<sub>3</sub> equivalent acidity, 80,000 µg  $SO_4^{-2}$ , 25,000 µg Fe, 690 µg Ca and 6.2 mBq Ra-226/kg tailings/day. The cumulative production/removal during the four year study period the above species were: 58.0 % of acidity, 67.0 % of total sulphate, 68.7 % of total iron, 19.0 of total Ca and 0.17 % of total Ra-226 contained in the coarse tailings.

The acidic drainage caused mobilization and release of other metals and radionuclides including Al, Mn, Mg, U, Th, Pb, Ce, Cu, Ni, Zn and various isotopes of radium (Ra-223, Ra-224 and Ra-228, etc.). Although these tailings contained only small amounts of alkaline materials, the acidic drainage started prior to full consumption of the available limestone alkalinity. It was estimated that at the time of acidic drainage, approximately 60 - 65 % alkalinity was still present in the tailings.

Because the initial gypsum content of these tailings was small, most of the calcium drainage occurred as a result of oxidation and limestone neutralization during the first one-and-one-half year. After that period, the calcium removal rate was small even under severe acidic conditions.

Based on yearly average production/removal rates of acidity, iron and sulphate, obtained near the end of the study period as ~ 13.8 % of total acidity production/iron/sulphur contents of coarse tailings, it was estimated that with the acidic drainage the Control-2 coarse tailings (columns #16 and 17) would be completely oxidized and depleted of total iron and sulphur in an additional 2.5 years (total 6.75 years) of acid generation. For Ra-226, the corresponding complete removal time was calculated as 5,000 years. This result was based on the observed cumulative Ra-226 removal rate during the acidic drainage period, which was very small (0.02 % per year). It is believed that this removal time for Ra-226 has been severely over-estimated as the Ra-226 removal rate post acidic drainage and pyrite depletion periods was unknown. Additional monitoring of Control-2 tailings would offer further insight in the leaching of Ra-226 and other radium isotopes as pyrite depletes and acidic drainage ceases.

At the on-set of the acidic drainage and because of mobilization and release of thorium isotopes (Th-232, Th-230 and Th-229), there were corresponding mobilization and drainage of thorium progeny including Ra-228, Ra-226, Ra-224 and Ra-223. At the peak acidic drainage period, the activities of other radium isotopes because of their shorter half lives, were considerably higher than that of Ra-226. The results for coarse tailings further showed that with the continued acidic drainage, and in the absence of gypsum, Ra-226 leachability decreased with the increased acidity and sulphate concentration.

The results substantiated the well known fact that exposure of coarse tailings leads to rapid acidic generation problem. For coarse tailings covered with a shallow water cover, the loss of water cover and exposure of the tailings for a prolonged period of time may result in some surface oxidation and acidification impacting the surface water quality. Additionally, there exits the potential of ferric ions introduction to the submerged tailings and oxidation of pyrite in the absence of oxygen underwater. Thus, the exposure of coarse tailings to air for an extended period should be avoided, otherwise consideration should be given to providing an additional oxygen consuming or a barrier layer at the surface of the underwater deposited tailings.

The unsaturated coarse tailings containing additional 7.5 % (w/w) coarse limestone of screen size - 6.3 mm, also oxidized and produced highly acidic drainage. The on-set of the acidic drainage was, however, delayed by approximately one year in comparison to coarse tailings without additional limestone (Control-2), where it occurred within two months of the implementation of the batch leaching schedule.

During the acidic drainage period, the leaching characteristics of coarse tailings - coarse limestone sub-group were similar to those of Control-2 coarse tailings, where the effluent average pH, Eh and Ec were, respectively, 2.5, 700 mV and 6,000  $\mu$ S/cm. The effluent average loading rates were: 25,000  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 28,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 8,000  $\mu$ g Fe, 3,800  $\mu$ g Ca and 7 mBq Ra-226/kg tailings/day. The cumulative productions/removals during the four year study period for the major species were: 17% of total acidity, 25 % of total

sulphur, 17 % of total iron, 12 % of total calcium and 0.29 % of total Ra-226 contained in the coarse tailings- coarse limestone mixture. The acidic drainage also caused mobilization and release of other constituents including Al, Ce, Cu, Mg, Mn, Ni, Pb, Th, U, Zn and various isotopes of radium.

The results clearly indicated that for coarse tailings mixed with coarse limestone, only less than 10 % of the available alkalinity was utilized before the occurrence of the acidic drainage. Thus, the neutralization efficiency of the coarse limestone was very low. During the acidic drainage period, some acid neutralization was still occurring but at a reduced rate caused most probably by decreased surface area of limestone particles and low contact time with the effluent. The effect of gypsum and other metal hydroxides, armouring the coarse limestone particles was not investigated in this study.

The unsaturated coarse tailings mixed with wet ground and pulverized limestone did not produce any acidic drainage during the four year study period. The results indicated that oxidation was also taking place in this coarse tailings limestone mixture, but the acid so produced was completely neutralized by the finely ground limestone in combination with the increased retention time resulting from slightly decreased hydraulic conductivity for this sub-group. The effluent was nearly neutral to slightly alkaline during the entire study period with pH of approximately 8, Eh approximately 480 mV, and Ec 2200  $\mu$ S/cm. It also contained appreciable amounts of dissolved sulphate, calcium and magnesium, resulting from acid neutralization and gypsum dissolution, and Ra-226. The effluent was also saturated with respect to gypsum. The mobilization of other oxidation related reaction products, e.g. acidity, Fe, Al, Ce, Cu, Ni, Pb, Th, U and Zn was absent or present only in trace amounts, except Mn which was present in low concentration in the neutral effluent.

During the entire study period the cumulative productions/removals of major species in the drainage effluent, in relation to their solid phase compositions, were: 7 % of total sulphur, 8.8 % of total calcium and 0.16 % of total Ra-226 contained in the tailings.

From the experimentally acid generation/neutralization rates, it was also estimated that during the entire study period approximately 13 % of total limestone alkalinity present in the tailings limestone mixture was consumed. The balance 87 % available alkalinity would provide further acid neutralization for an additional 29 years, assuming a 100 % (full) limestone neutralization efficiency. The data did not permit evaluation of the efficiency factor, and hence, it was not possible to predict the occurrence of acidic drainage from coarse tailings mixed with wet ground limestone in stoichiometrically equivalent proportion to their total acid generation potential. It is, however, clear that the benefits of such limestone additions are for limited time only.

The unsaturated total mill tailings (Control-1) without additional limestone also produced acidic drainage, but it was moderate and less severe in comparison to Control-2, coarse tailings. These tailings had a low hydraulic conductivity of 8.7 x  $10^{-6}$  cm/s and retained appreciable amounts of moisture, causing reduced acid generation as well as mobilization and release of oxidation reaction products. During the acidic drainage period, the effluent average pH, Eh and Ec were, respectively, 3.0, 780 mV and 2500 µS/cm. The effluent average loading rates were: approximately 2100  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 6000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 100  $\mu$ g Fe, 1500  $\mu$ g Ca and 4 mBq Ra-226/kg tailings/day. The corresponding cumulative productions/removals during the study period were: 3 % of total acidity, 8 % of total sulphur, only 0.33 % of total iron, 13.6 % of total calcium and 0.08 % of total Ra-226 contained in the total tailings. The acid generation rate for these tailings was approximately 30 - 35 times less than that for Control - 2 coarse tailings.

Similar to Control - 2 coarse tailings, the acidic drainage also caused mobilization and release of other metals and radionuclides: Al, Ce, Cu, Mg, Mn, Ni, Pb, Th, U, Zn, Ra-226 and other isotopes of radium, but in reduced amounts.

Theoretically, Control - 1 tailings columns, containing tailings to a height of 0.35 m, would continue to produce acidic drainage for another 85 years or so but at a decreased rate.

Disposal of tailings as a homogenous mixture without segregation of their constituent particle sizes, i.e. paste like, would offer many advantages over the conventional end spill slurry discharge method. Some of these are:

- Low hydraulic conductivity resulting into decreased water infiltration and porewater flow, and hence, increased surface drainage,
- Increased water retention contributing to a high phreatic surface or water table, high capillary fringe and a shallow unsaturated zone amenable to oxidation,
- Low acid generation and metal mobilization rates contributing to decreased effluent loadings, and hence, lower treatment costs. For example, in comparison to coarse tailings, the acid neutralization cost alone would be reduced by a factor of 30,
- A possible decommissioning option of covering the reactive tailings above the phreatic surface with non reactive tailings or geological materials or de-suphurized tailings mixed with additional limestone, if required, for controlling acidic drainage,
- Increased storage capacity of the tailings basin without raising the height of impoundment dams.

The total mill tailings mixed with 7.5 % (w/w) limestone of various screen sizes, from coarse (-6.3 mm) to wet ground and pulverized, columns #4 - 15, did not produce any acidic drainage during the entire study period. The results indicated that oxidation and the acid generation processes were also active in these columns, but at low rates, and complete acid neutralization was taking place (as seen from effluent Mg and Mn drainage profiles). The reactivity of the tailings-limestone mixture decreased with the decrease in particle size of the mixed limestone from coarse to wet ground and pulverized. The low rate of acid generation in combination with increased effluent retention time contributed to complete acid neutralization for tailings containing coarse limestone. For all columns, the effluents were saturated with respect to gypsum and the column average pH, Eh and Ec were, respectively, ~ 7.5, 550 mV and 2200  $\mu$ S/cm. After two years, all columns were producing alkaline effluents with alkalinity production rates of approximately 200  $\mu$ g CaCO<sub>3</sub>/kg tailings/day. During the study period, the average effluent loadings were: 5000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 1800  $\mu$ g Ca and 3 mBq Ra-226/kg tailings/day. The corresponding cumulative productions/removals were: 8.6 % of total sulphur, 5.7 % of total calcium, 22 % of initial total gypsum and 0.06 % of r total Ra-226 contained in the tailings limestone mixture. Similar to coarse tailings mixed with wet ground limestone, the mobilization of other oxidation related reaction products, e.g. acidity, Fe, Al, Ce, Cu, Ni, Pb, Th, U and Zn was absent or present in trace amounts, except Mn, which was present in small quantities in the neutral to alkaline effluents.

Because of gypsum saturation, the drainage effluent characteristics did not permit estimation of the rates of acid generation and limestone neutralization taking place in these tailings limestone mixtures. Assuming the rates to be the same as those for total mill tailings without additional limestone, approximately 2100  $\mu$ g CaCO<sub>3</sub> equivalent acidity/kg tailings/day, the added limestone to these tailings was sufficient for approximately 100 years of acid neutralization at 100 % limestone neutralization efficiency, and for a shorter time at decreased efficiency. It is also believed that, because of neutral to alkaline pH's, the actual acid generation rates in the these tailings-limestone mixtures were lower than that for total tailing without limestone amendment.

In the laboratory testing over a four year period, and based on the effluent quality results for all tailings, tailings and limestone mixtures and those with a shallow water cover, the total mill tailings mixed with pulverized and finely ground limestone were judged, in the short term, as the best performer in preventing acid mine drainage. Disposal of limestone amended total mill tailings above the phreatic surface offers another latitude to mine waste management but the long-term limestone neutralization potential in such a scenario remains to be investigated further.

For the submerged coarse tailings, there was no acidic drainage during the first three years of the study. After the removal of gypsum and other soluble minerals, within first three months with a high porewater flow, the drainage effluents were nearly neutral having pH, Eh and Ec in ranges of 6.5 - 7.5, 400 - 450 mV and 70 - 100  $\mu$ S/cm, respectively. During that period, there was no mobilization of oxidation reaction products most notably Fe, Al, Mn and Th, and the tailings produced slightly alkaline effluent with alkalinity in the range of 10 - 40 mg CaCO<sub>3</sub>/l.

In the fourth year, however, the submerged columns started to produce slightly acidic effluents with pH's, total acidities, iron and Ra-226 concentrations in ranges of 5.5 - 7.0, 10 - 20 mg/l, 1 - 20 mg/l and 5000 - 25,000 mBq/l, respectively. The results indicated on-set of a slow oxidation and development of slight acidic conditions at the surface of the tailings, causing 10 to 30 fold increases in Ra-226 mobilization and release to the porewater, which were associated with iron oxidation and accompanied by its increased drainage. After the removal of gypsum and other soluble minerals, the effluent average loading rates were: 180  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 300  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 20  $\mu$ g Fe, 200  $\mu$ g Ca and 150 mBq Ra-226/kg tailings/day. The corresponding cumulative productions/removals were: 0.2 % of total acidity, 0.3 % of total sulphur, 0.7 % of total iron, 6 % of calcium and 5.5 % of total Ra-226 contained in the submerged coarse tailings.

In comparison to unsaturated coarse tailings (Control-2) and total mill tailings (Control-1), the shallow water on coarse tailings reduced total acidity production by factors of 295 and 10, total sulphate production by 200 and 20, and total iron production by 950 and 3, respectively. For total mill tailings mixed with 7.5 % limestone of various screen sizes, the corresponding decrease in total acidity production was by factors of 1900, 50 and 7 when compared with those for unsaturated coarse tailings, total mill tailings and submerged coarse tailings, respectively. The combination of total mill tailings and limestone was more effective in preventing acid drainage than water cover on coarse tailings during the short laboratory testing period. However, the efficiency of water cover in preventing acidic drainage from total mill tailings remains to be evaluated.

For the submerged coarse tailings, the Ra-226 mobility was initially controlled by gypsum dissolution and decreased with the depletion of gypsum. During the gypsum removal period, the Ra-226 leaching rates for submerged and unsaturated coarse tailings were similar. In the absence of gypsum and without oxidation, there was no corresponding increase in Ra-226 drainage although the effluent sulphate concentrations were very low. With the on-set of a slow oxidation, the mobility of Ra-226 increased significantly, up to 30 times its pre-oxidation value, and correlated with the corresponding slow increase in iron mobilization and perhaps Th-230 as well. The effluent sulphate loading also increased at the same time, but it was statistically not significant.

The cumulative Ra-226 removal from submerged coarse tailings during the study period was approximately 30 to 50 times greater than those for unsaturated coarse and total mill tailings without or mixed with limestone. This increased Ra-226 drainage was 3 to 6 times greater than the corresponding increase in cumulative effluent volumes for the two cases. Perhaps in the absence of gypsum and/or that of a moderate to severe acidification in the submerged case, the sulphate concentration was too low to adversely effect or suppress radium solubility. The dynamics of Ra-226 leaching under various conditions of oxidation, reduction and gypsum depletion etc., need to be further investigated.

In the water columns above the tailings surface and in the absence of gypsum, there was very little upward diffusion of dissolved metals and oxidation reaction products except Ra-226, whose the concentration varied between 30 - 140 mBq/l. Because of high porewater flow, the surface water quality resembled that of the distilled water inflow, with pH in the range of 5 - 5.7, Eh 500 - 600 mV, Ec 10 - 15  $\mu$ S/cm, acidity and alkalinity in the range of 3 - 6 mg CaCO<sub>3</sub>/l, SO<sub>4</sub><sup>-2</sup> ~ 3 - 8 mg/l, Fe ~ 0.01 - 0.05 mg/l, Ca ~ 0.1 - 1.0 mg/l and Mg ~ 0.08 - 0.1 mg/l. For other conditions of low porewater flow, natural lake water cover and other oxygen consuming barriers placed on the submerged tailings, there may be a further improvements/changes in both surface as well as porewater qualities, which need to be established.

The unsaturated and crushed waste rock with high acid generation potential (NNP = -17.8 kg CaCO<sub>3</sub>/tonne) also oxidized and produced highly acidic drainage in the fourth year of the study. The leaching characteristics resembled those of coarse tailings with 7.5 % coarse limestone, where delayed acidic drainage occurred. In the acid effluent, the average pH, Eh and Ec were, respectively, 2.2, 780 mV and 7000  $\mu$ S/cm. The effluent average loading rates were: 38,000  $\mu$ g CaCO<sub>3</sub> equivalent acidity, 42,000  $\mu$ g SO<sub>4</sub><sup>-2</sup>, 12,500  $\mu$ g Fe, 2000  $\mu$ g Ca and 3.0 mBq Ra-226/kg waste rock/day. The cumulative productions/removals during the four year study period for major species were: 53 % of total acidity, 60 % of total sulphur, 31 % of total iron, 59 % of total calcium and 0.08 % of total Ra-226 contained in the waste rock. The reactivity of the crushed waste rock was higher than that of coarse tailings, and most of the leaching occurred during the last year of the study period. Similar to coarse tailings, the acidic drainage also caused significant mobilization and release of other constituents including Al, Ce, Cu, Mg, Mn, Ni, Pb, Th, U, Zn and various isotopes of radium.

Prior to acidic drainage, the average acid generation/limestone neutralization for this waste rock was estimated as  $1800 \ \mu g \ CaCO_3/kg$  waste rock/day. Also similar to coarse without or mixed with coarse limestone, the acidic drainage from the high acid generating waste rock occurred with 75 % of the equivalent limestone alkalinity still remaining in the waster rock. The waste rock was crushed to a particle size of -2.0 mm, and the results established a acid neutralization efficiency of approximately 25 % for the size and type of the waste. Since the acidic drainage related species peaks occurred during the last year of the study period, the data did not permit extrapolations and computations of oxidation related steady state reaction rates for calculating total period of acidic drainage and metal mobilities, etc.

The unsaturated and crushed waste rock with medium acid generation potential (NNP = -10.7 kg CaCO<sub>3</sub>/tonne) did not produce any acidic drainage during the study period. The results indicated that oxidation, acid generation and neutralization were active processes within the waste rock, but the acid neutralization was complete in preventing acidic drainage and release of

oxidation reaction products. During the study period, the effluent was nearly neutral to slightly alkaline with pH ~ 7.5, Eh ~ 400 mV and Ec ~ 1000  $\mu$ S/cm. The effluent contained mostly dissolved gypsum, resulting from acid neutralization, and Ra-226. The cumulative effluent loadings/removals were, respectively: 11.9 % of total sulphur, 30 % of total calcium and 0.04 % of for total Ra-226 contained in the waste rock. Some limestone dissolution contributed to alkalinity and additional calcium in the effluents.

The average acid generation or limestone neutralization rate for this waste rock was approximately  $2100 \ \mu g \ CaCO_3/kg$  waste rock/day, which was comparable to that of high acid generating waste rock, and by the end of the study period approximately 20 % of the total limestone alkalinity had also been consumed in preventing acidic drainage. From the calculated acid generation/neutralization rates, it was estimated that acidic drainage from this waste rock would occur after approximately 12 additional years of leaching, assuming a limestone neutralization efficiency of 100 %, and sooner at lower efficiencies.

The unsaturated and crushed waste rock with low acid generation potential (NNP = +13.8 kg CaCO<sub>3</sub>/tonne) also did not produce any acidic drainage during the study period. Similar to the medium acid generating waste rock, oxidation, acid generation and neutralization processes were also actively taking place in this waste rock, but the neutralization was complete and acidic drainage and concomitant mobilization of oxidation reaction products were prevented. The effluents were slightly alkaline during the entire study period with pH ~ 8.0, Eh ~ 450 mV and Ec ~ 1000  $\mu$ S/cm. The drainages were mostly gypsum derived and contained appreciable amounts of calcium and sulphate, resulting from complete acid neutralization, and Ra-226.

The cumulative effluent loadings/removals were, respectively: 19 % of total sulphur, 11.6 % of total calcium and 0.06 % of total Ra-226 contained in the waste rock. The average acid production or limestone neutralization rate for this waste rock was 2250  $\mu$ g CaCO<sub>3</sub>/kg waste rock/day and, during the study period, approximately 2.5 g CaCO<sub>3</sub>/kg waste rock or 9 % of total limestone alkalinity present in the waste rock was consumed in complete acid

neutralization. Stoichiometrically, at 100 % limestone neutralization efficiency and the experimentally determined acid generation/neutralization rates, this waste rock should not produce any acidic drainage. However, at lower neutralization efficiencies, acidic drainage may occur in the next twenty years or so, the estimated time for complete oxidation of pyrite in test samples of the low acid generating waste rock.

Prior to acidic drainage, the calculated acid generation and limestone neutralization rates for the three waste rock types were comparable, but acidic drainage occurred only in the high acid generating waste rock. Perhaps, in this waster rock, the low total neutralization potential of 8.5 kg CaCO<sub>3</sub>/tonne and the distribution and particle size of the alkali minerals in relation to sulphides were contributing factors in the early occurrence of acidic drainage. For the other two, low and medium acid generating waste rock, the total neutralization potentials were, respectively, 26.82 and 15.06 kg CaCO<sub>3</sub>/tonne.

#### 5. SUMMARY AND CONCLUSIONS

Column leaching studies were conducted for evaluating the oxidation and leaching characteristics of pyritic uranium tailings and waste rock under unsaturated conditions, coarse tailings with a shallow water cover, and limestone neutralization characteristics of tailings mixed with 7.5 % (w/w) limestone of various screen sizes. The results are summarized as follows:

The unsaturated coarse tailings without additional limestone (Control - 2) oxidized readily and produced highly acidic drainage with a pH of ~ 1.2. The acidic drainage caused significant mobilization and release of all oxidation reaction products including: Al, Ca, Ce, Cu, Fe, Mg, Mn, Ni, Pb, Th, U, Zn, SO<sub>4</sub><sup>-2</sup> and various isotopes of radium (Ra-223, Ra-224, Ra-226 and Ra-228).

- The acidic drainage in coarse tailings started prior to full consumption of the total available alkalinity, and, at the time of its occurrence, approximately 60 65 % of the available alkalinity was still present in the tailings.
- The Ra-226 leachability was controlled by gypsum solubility, and decreased with increased acidity and sulphate concentration in the absence of gypsum. The acidic drainage also resulted in an increased mobility and release of thorium and other radium isotopes.
- Prolonged exposure of coarse tailings to air would contribute toward significant acid generation and water quality problems. For submerged coarse tailings, it is important to maintain a minimum depth of water cover to prevent acid production and minimize Ra-226 mobility. This may require additional alkalinity in the incoming water or incorporation of an oxygen consuming cover or barrier layer at the surface of the underwater deposited tailings for permanently controlling their surface oxidation and development of acidic conditions.
- The unsaturated coarse tailings containing an additional 7.5 % (w/w) coarse limestone of screen size -6.3 mm, also oxidized and produced severe acidic drainage with a pH of approximately 2.5. The onset of the acidic drainage was delayed by approximately one year from its occurrence in the Control 2 coarse tailings.
- The acidic drainage in coarse tailings-coarse limestone sub-group occurred with 90 % of the total available alkalinity still remaining. Addition of coarse limestone to coarse tailings was considered unsuitable for prevention of the acidic drainage.
- The unsaturated and coarse tailings mixed with 7.5 % (w/w) wet ground and pulverized limestone did not produce any acidic drainage. Oxidation and acid generation were also taking place in these tailings, but the finely ground limestone provided complete acid neutralization preventing acidic drainage and release of oxidation reaction products.

- During the four year study period, approximately 13 % of total available limestone alkalinity was consumed in acid neutralization in the coarse tailings wet ground limestone sub-group. It was estimated that the remaining alkalinity would provide additional acid neutralization for another 29 years.
- The unsaturated total mill tailings without additional limestone (Control 1) also produced acidic drainage, but it was moderate at a pH of ~ 3.0 and less severe than for Control 2 coarse tailings. These tailings had a low hydraulic conductivity and retained high moisture, which reduced oxidation and acid generation rates and controlled the release of oxidation reaction products.
- The acid generation rate for Control 1 total tailings was approximately 30 35 times less than that for Control - 2 coarse tailings, which would result in much lower treatment and sludge handling costs.
- The unsaturated and total mill tailings mixed with 7.5 % (w/w) limestone of various screen sizes, from coarse (-6.3 mm) to wet ground and pulverized, did not produce any acidic drainage during the entire study period. Both oxidation and acid generation processes were also active in these tailings limestone sub-groups, but the acid neutralization was complete for all limestone sizes and acidic drainage was prevented.
- It was estimated that the added limestone alkalinity to total mill tailings would be sufficient for acid neutralization for approximately 100 years. At 100 % limestone neutralization efficiency, these tailings in the laboratory test columns would not produce any acidic drainage.
- The total mill tailings mixed with pulverized and wet ground limestone were judged very effective in preventing acidic drainage during the short laboratory testing period. For pyritic uranium tailings, there was an additional advantage of the containment of associated radioactivity in a gypsum controlled system.

- The submerged coarse tailings did not produce any acidic drainage and release of oxidation reaction products during the first three years. In the fourth year, the submerged coarse acidic effluents with pH's of tailings started to produce slightly  $\sim$  5.5. There was an onset of a slow oxidation process at the surface of the tailings resulting in 10 to 30 fold increases in Ra-226 mobilization and release to the porewater associated with iron oxidation and its increased drainage. The presence of an organic substrate or additional alkalinity in the water column would be beneficial in controlling acid generation.
- The water cover on coarse tailings reduced acid generation by factors of 295 and 10, total sulphate production by 200 and 20, and total iron production by 950 and 3, in comparison to unsaturated coarse (Control 2) and total mill tailings (Control 1), respectively.
- The Ra-226 leachability in submerged tailings was very high with the onset of slow oxidation and development of acidic conditions in the absence of gypsum.
- In the water cover above the tailings surface and in the absence of gypsum, there was very little upward diffusion of dissolved metals and other oxidation reaction products except Ra-226, whose concentration was slightly elevated.
- The unsaturated and crushed waste rock with a high acid generation potential, oxidized and produced highly acid drainage (pH approximately 2.2) in the fourth year of the study period. The acidic drainage also caused significant mobilization and release of all oxidation reaction products.
- The acidic drainage in the high acid generating waste rock occurred with 75 % of the available limestone alkalinity still remaining in the waste rock.
- The unsaturated and crushed waste rock with a medium acid generation potential did not produce any acidic drainage or released oxidation reaction products during the study

period. Oxidation, acid generation and neutralization were active processes within the waste rock, but the acid neutralization was complete and acidic drainage absent.

- During the study period, approximately 20 % of the total available limestone alkalinity was consumed for acid neutralization in the medium acid generating waste rock. At 100 % limestone neutralization efficiency, it was estimated that acidic drainage from this waste rock would occur in approximately 16 years, and somewhat sooner if the efficiency was reduced.
- The unsaturated and crushed waste rock with a low acid generation potential also did not produce any acidic drainage or released oxidation reaction products during the study period. Oxidation, acid generation and complete acid neutralization processes were active in this waste rock as well.
- During the entire study period, approximately 9 % of the total available limestone alkalinity was consumed for complete acid neutralization in the low acid generating waste rock. At 100 % limestone neutralization efficiency, this waste rock should not produce any acidic drainage, but if the efficiency was reduced acidic drainage may occur in the next twenty years or so.

In conclusion, the usefulness of column leaching tests has been well demonstrated in understanding the oxidation and leaching kinetics of reactive tailings and waste rock, and for evaluating various decommissioning options.

A shallow water cover on pyritic uranium tailings is very effective in controlling/limiting their oxidation and acid generation to very low rates, but lacked in complete prevention. The tests have also shown that radium mobility increases, in the absence of gypsum, when acidic conditions develop at the surface of underwater deposited tailings. Limestone amendments to tailings , in some cases, provided short-term controls on acidic drainage.

### 1-126

#### 6. ACKNOWLEDGMENT

The author wishes to gratefully acknowledge the participation of Rio Algom Limited, Elliot Lake Division, in this project, and for their financial and continue support during the study period. Special thanks are due to Mr. A. Vivyurka for his invaluable help at all stages of the laboratory and field studies and logistic support, Messrs. R. Lipinski, L. Lance and W. Stefanich for their technical support, and Messrs. J. Nightingale, K. Kay, R. Payne and Ms. M. Wiber for their understanding and support of the environmental research at the Elliot Lake laboratory. The participation and support of Messrs. B. Palmer, B. Bihari and D. Kay (all formerly of Rio Algom Limited) during initial studies are equally well acknowledged. Sincere thanks and deep appreciation are also due to the staff of the surface environment research group at the Elliot Lake Laboratory: Ms. Y. Boucher, Mr. T. Lim, Ms. D. Horne, Ms. L. Coté and Ms. A. Eden (both formerly of the Elliot Lake Laboratory) for their excellent technical and analytical support, and for a job well done. Last but not the least, the continued help and support of the CANMET management are always well appreciated.

#### 7. REFERENCES

- American Public Health Association. 1992. Standard Methods for the Examination of Water and Waste Water. Published by American Public Health Association, American Water Works Association and Water Environment Federation. Ed. by Arnold F. Greenberg, Lenore S. Clesceri, and Andrew D. Eaton. American Public Health Association, Publication, Washington, DC, U.S.A.
- Balins, J. K., Welch, D. F., Knapp, R. A., Ashley, C.F. and Maltby, J. M. 1991. Decommissioning of Quirke Mine Uranium Tailings Basin to Eliminate Acid Production. In Proc. 2nd International. Conference on the Abatement of Acid Drainage, Montreal, Quebec, Canada, September, 1991, vol. 1, pp. 378-398.
- Boucher, Y. M., Davé, N. K., and Lim, T. P. 1989. Surface Environmental Research at the Elliot Lake Laboratory - A Compilation of Papers. Mining Research Laboratory, Division Report MRL, 89-79 (LS), CANMET, Energy Mines and Resources, Canada, 1989.
- Cummings and Cockburn Ltd. 1991. Investigations of the Hydrological Aspect of the Quirke Mine Waste Management Area Decommissioning Plan. Summary Report # 7249, prepared for Rio Algom Ltd. and Golder Associates, January 1991.
- Davé, N. K. and Lim, T. P. 1987. Effects of Various Tailings Covers and Radon Gas Emanation from Pyritic Uranium Tailings, In Proc. 1987. National Symposium on Surface Mining Hydrology, Sedimentology and Reclamation, Springfield, Illinois, 1987, pp. 99-104. Division Report MRP/MRL 87-84 (OP).

- Davé, N. K., and Lim, T. P. 1988. Environmental Research at the Mining Research Laboratory, CANMET, Elliot Lake, Ontario, Canada. Division Report, MRL, 88-84 (OPJ), CANMET, Energy Mines and Resources, Canada, 1988.
- Davé, N. K., Lim, T. P., Vivyurka, A. J., Bihari, B. and Kay, D. A. 1990. Development of Wet Barrier on Pyritic Uranium Tailings Elliot Lake, Ontario: Phase 1. Division Report MRL 90-50 (OPS), 1990.
- Davé, N. K. 1992. Water Cover on Acid Generating Mine/Mill Waste A Technical Review. Division Report MSL 92-38(CR), CANMET, Energy Mines and Resources, Canada, November 1992.
- Davé, N. K., and Vivyurka, A. J. 1993. Shallow Water Cover and Acid Generating Uranium Tailings. In Proc. International Symposium on Material Processing Technology and Environmental Protection in Metallurgical Processing. CIM Annual Conference of Metallurgists, Quebec City, August 1993. Canadian Institute of Mining, Metallurgy and Petroleum, September 1993, pp. 287-305.
- Davé, N. K., and Vivyurka, A. J. 1994. Water Cover on Acid Generating Uranium Tailings -Laboratory and Field Studies. In Proc. International Land Reclamation and Mine Acid Drainage Conference on the Abatement of Acid Drainage, Pittsburgh, U.S.A., April 1994, vol. 1, pp. 297-306.
- Golder Associates Ltd. 1991. Summary Report on Geotechnical Aspects of Decommissioning of the Quirke Mine Waste Management Area. Summary Report #901-1824. Prepared for Rio Algom Limited, February 1991.
- Kay, K. R., Payne, R. A. and Vivyurka, A. J. 1992. Elliot Lake Uranium Mine Division Closure Planning and Decommissioning of the Quirke and Panel Mines and Waste

Management Facilities: A Case History. Rio Algom Limited, Environment Health and Safety Report for Rio Tinto Zinc (RTZ), 1992.

- Lim, T. P. and Davé, N. K. 1981. A Rapid Method of Ra-226 Analysis in Water Samples using Alpha Spectroscopic Technique. CIM Bull. 74, pp. 97-105, 1981.
- Lim, T. P. 1988. Paradoxical Secular Disequilibrium of Radioisotopes in Uranium Tailings. M. Sc. Thesis. Laurentian University, Sudbury, Ontario, Canada. September 1988. Also Division Report MRL 89-34 (OP)., CANMET, Energy Mines and Resources, Canada, 1989.
- Macbeth, P. J., Rogers, V. C., Chater, A., Sandquist, G. M. and Jensen, C. M. 1977. Measurements of Surface Radon Fluxes from Uranium Mill Tailings. American Nuclear Society Transactions, vol. 26, pp. 143, 1977.
- Nielsen, K. K., Rogers, V. C., and Gee, G. W. 1984. Diffusion of Radon through Soils -A Pore Distribution Model. Soil Science Soc., Am. J., vol. 48, pp. 482-487, 1984.
- Silker, W. B., Heasler, P. G., and Santos L. E. 1979. Diffusion and Exhalation of Radon from Uranium Tailings. NUREG/CR-1138. PNL-3207. Pacific Northwest Laboratory, Richland, WA. 99352.
- Silker, W. B., Wogman, N. A., Thomas, C. W., Carr, D. B., and Heasler, P. G., 1979. Measurement of Radon Diffusion and Exhalation from Uranium Mill Tailings Piles, Environmental Science and Technology, vol. 13, No. 8, 1979, pp. 962-964.
- Strong, K. P. and Levins, D. M. 1982. Effect of Moisture Content on Radon Emanation from Uranium Ore and Tailings. Health Physics, 42, pp. 27-32, 1982.

- Tanner, A. B. 1964. Radon Migration in Ground A Review. The Natural Radiation Environment. Eds. John A. S. Adams and Wayne M. Lowder, Chicago, University of Chicago Press. Rice University Semi-centennial Publication, pp. 161-190.
- Veldhuizen, H., Blowes, D. W. and Siwik, R. 1987. The Effectiveness of Vegetation in Controlling Acid Generation in Base Metal Tailings. In Proc. Seminar/Workshop on Acid Mine Drainage. Supply and Services Canada., Nova Scotia, 1987, pp. 507-536.

# FIGURES

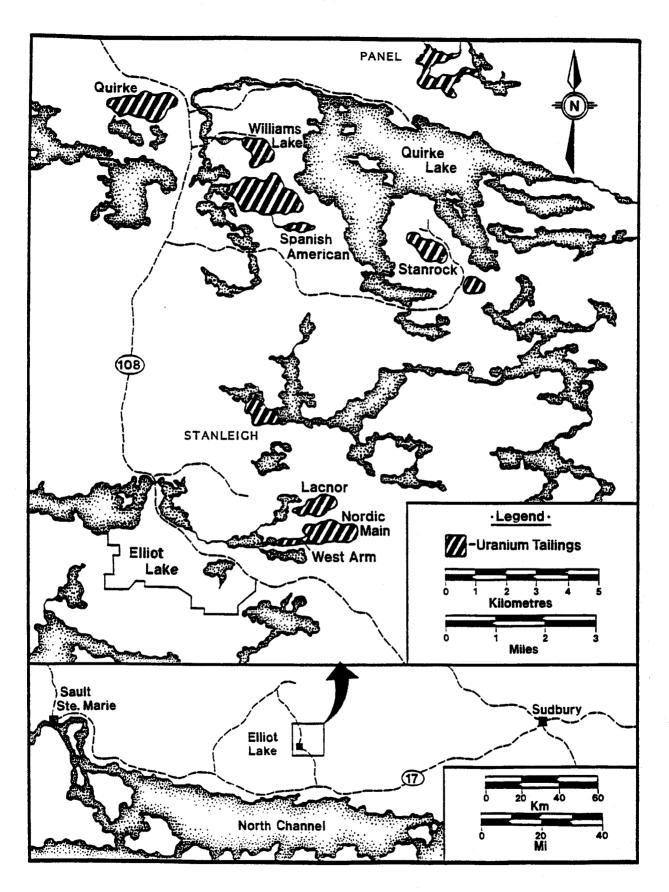
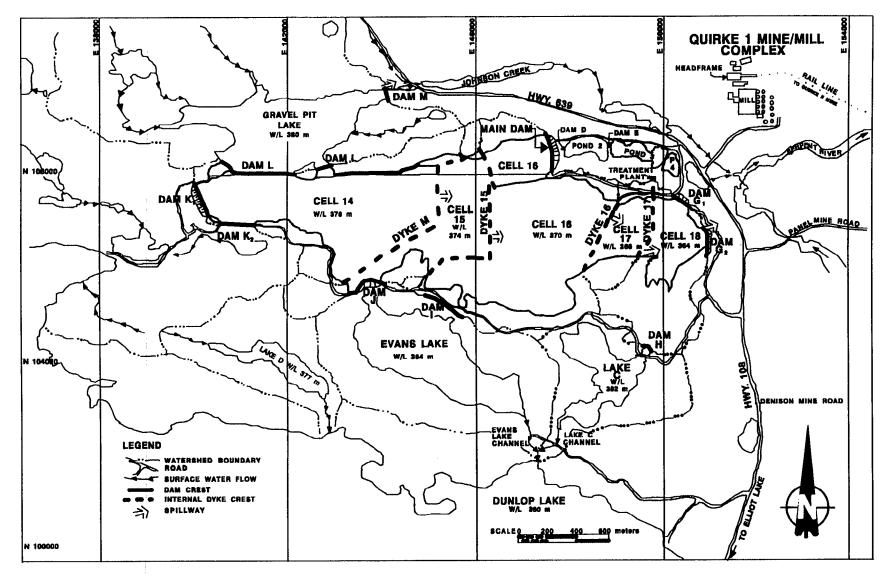


Fig. 1 Generalized location of uranium mine tailings sites in Elliot Lake, Ontario, Canada.



4

Fig. 2 General plan of submersion of tailings underwater at the Quirke Mine waste management area.



÷

.

ŧ

I

.

1

)

;

1

1



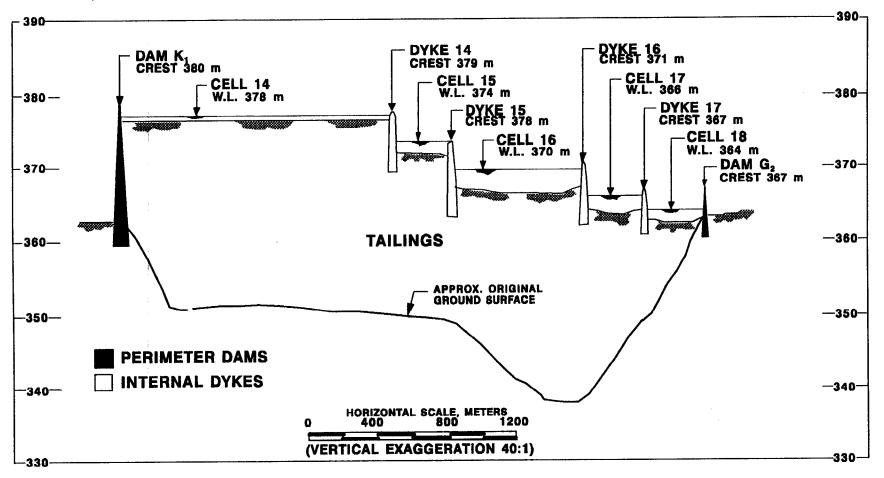
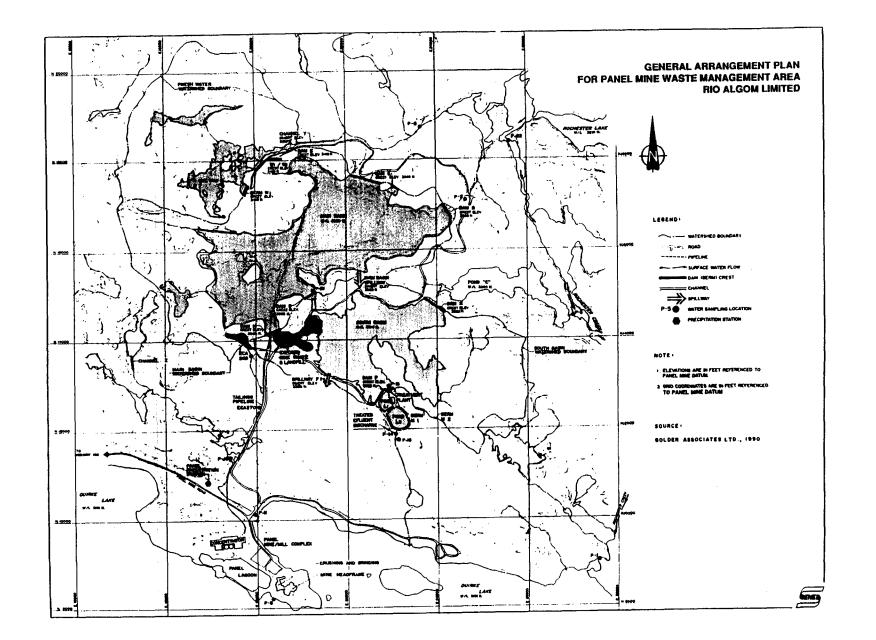


Fig. 3 Schematic profile of submersion tailings underwater at the Quirke Mine waste management area.



1

Fig. 4 General plan of submersion of tailings at the Panel Mine waste management area.

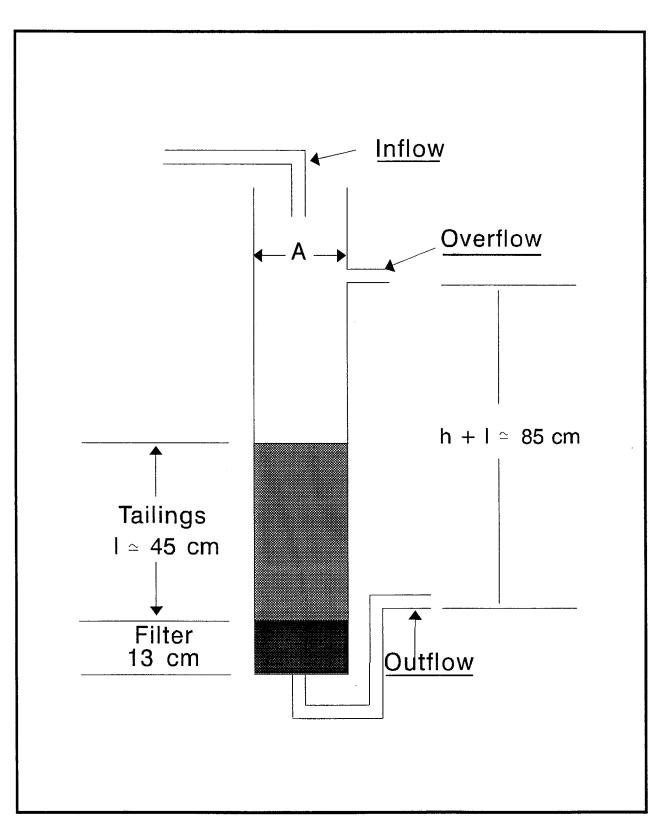


Fig. 5 Experimental arrangement of the column leaching lysimeter for leaching under unsaturated conditions.

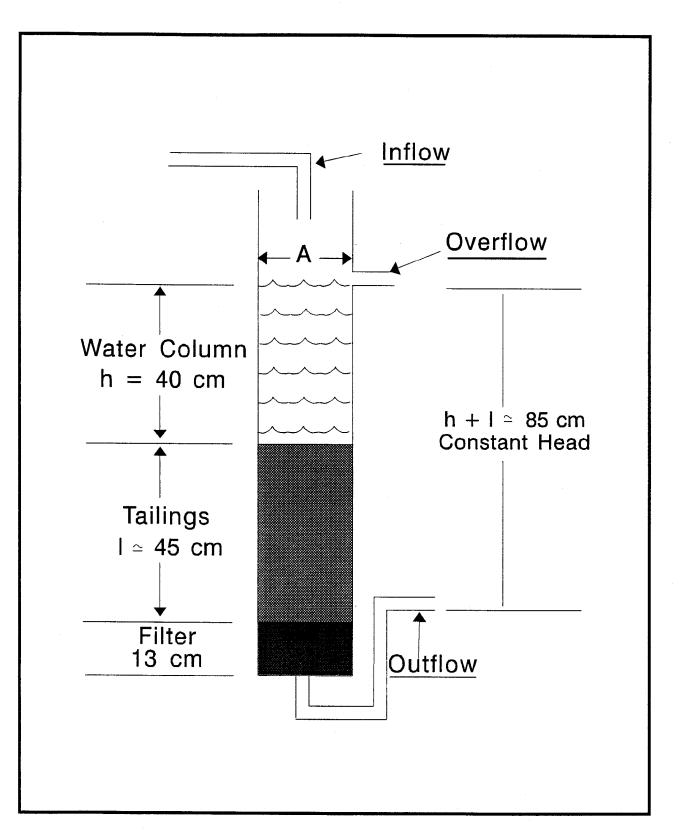
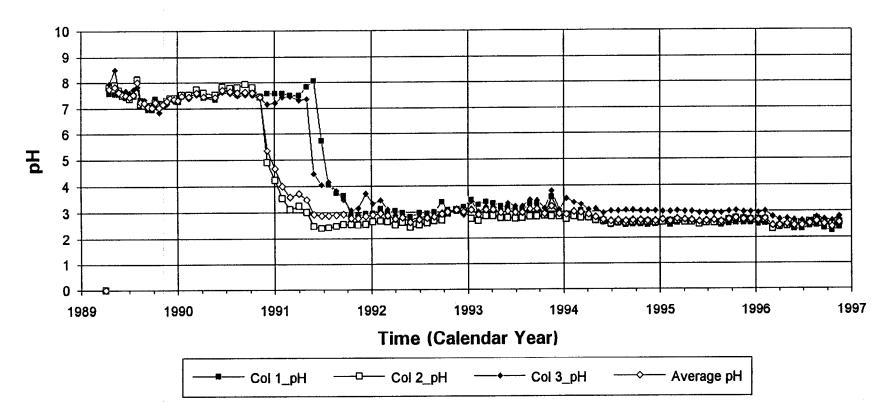


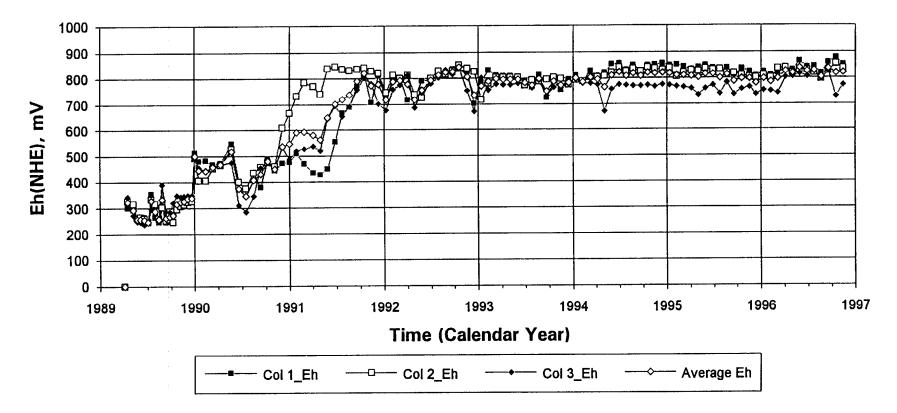
Fig. 6 Experimental arrangement of the column leaching lysimeter for leaching under submerged conditions.



1

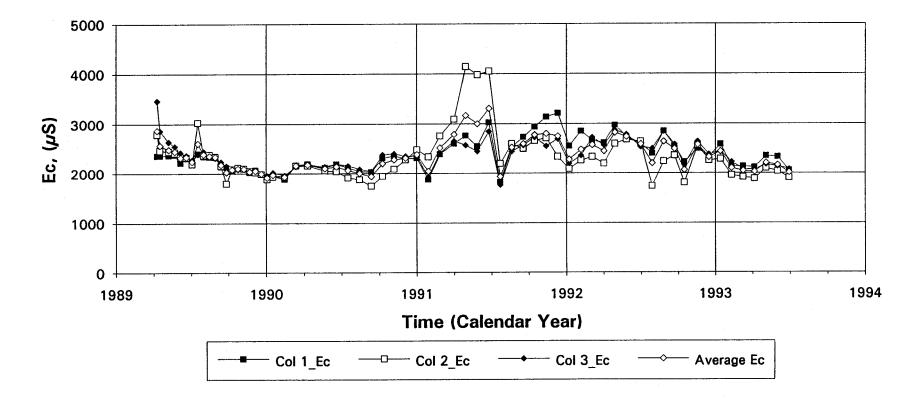
# Homogenized Total Mill Tailings (Control-1) pH vs. Time

Fig. 7 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of pH with time.



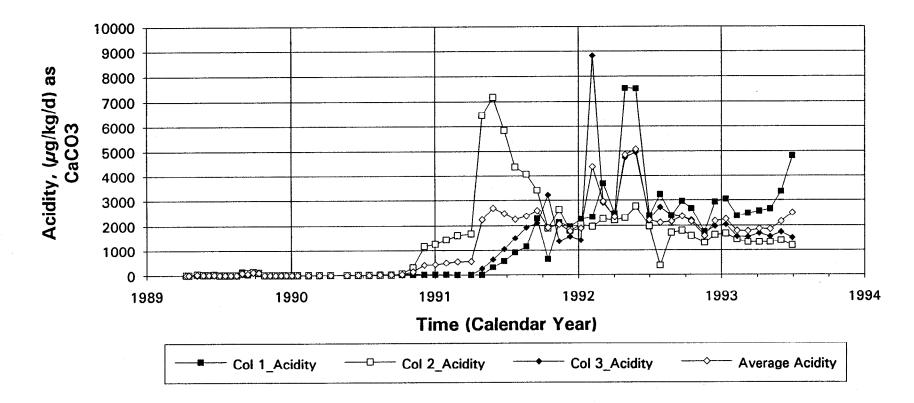
Homogenized Total Mill Tailings (Control-1) Eh(NHE) vs. Time

Fig. 8 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of Eh with time.



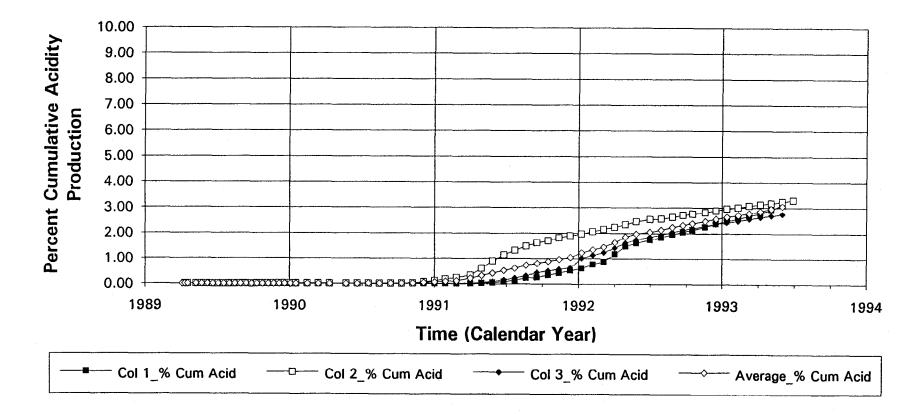
## Homogenized Total Mill Tailings (Control-1) Electrical Conductivity vs. Time

Fig. 9 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of Ec with time.



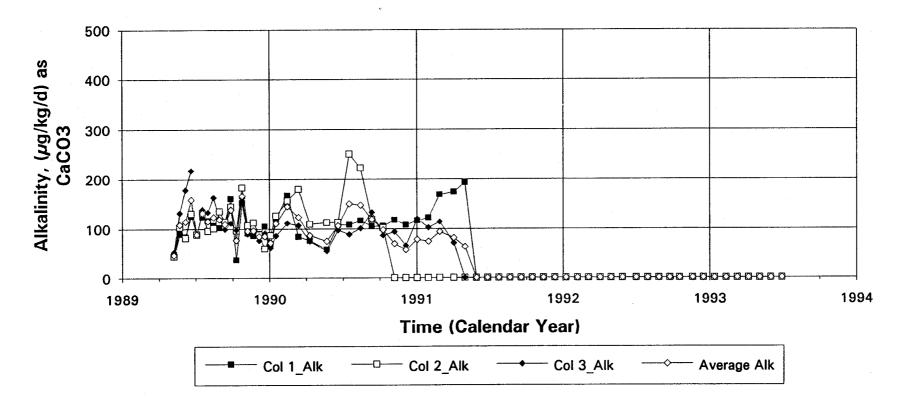
# Homogenized Total Mill Tailings (Control-1) Acidity Loading vs. Time

Fig. 10 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of total acidity loading rate with time.



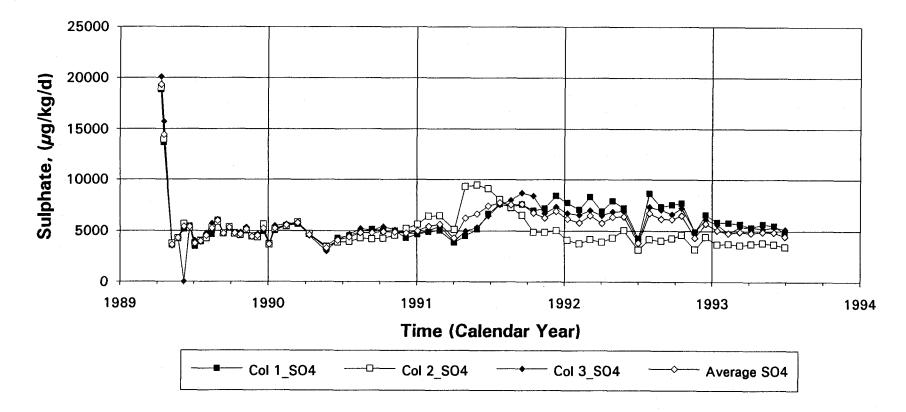
## Homogenized Total Mill Tailings (Control-1) Cumulative Acidity Production vs. Time

Fig. 11 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative acidity production with time.



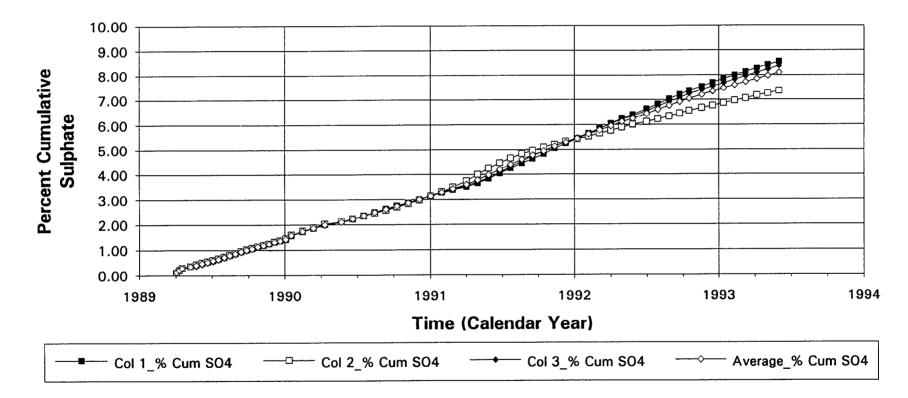
## Homogenized Total Mill Tailings (Control-1) Alkalinity Loading vs. Time

Fig. 12 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of total alkalinity loading rate with time.



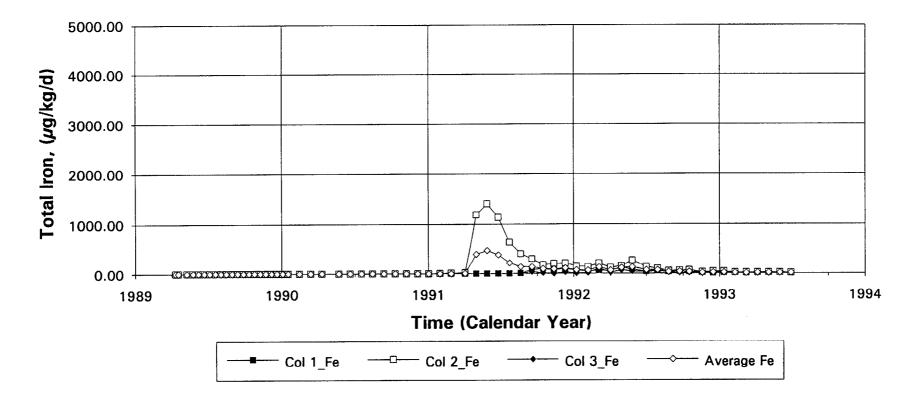
## Homogenized Total Mill Tailings (Control-1) Sulphate Loading vs. Time

Fig. 13 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved SO<sub>4</sub><sup>-2</sup> loading rate with time.



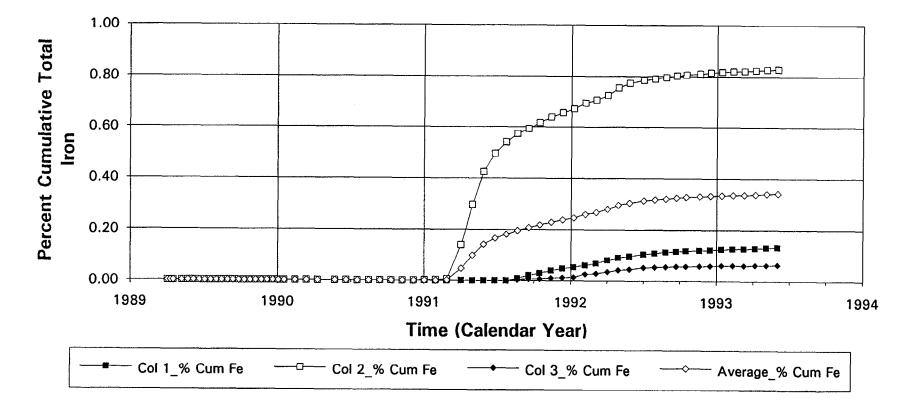
## Homogenized Total Mill Tailings (Control-1) Cumulative Sulphate Removal vs. Time

Fig. 14 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative SO<sub>4</sub><sup>-2</sup> removal (as % total S) with time.



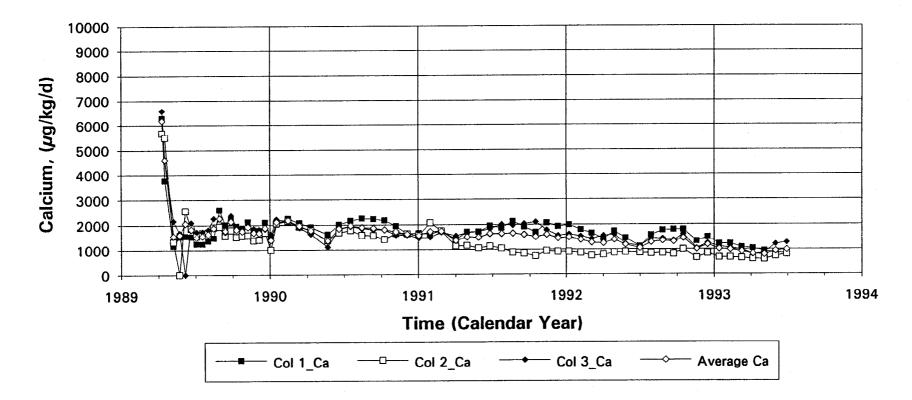
## Homogenized Total Mill Tailings (Control-1) Total Iron Loading vs. Time

Fig. 15 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved total Fe loading rate with time.



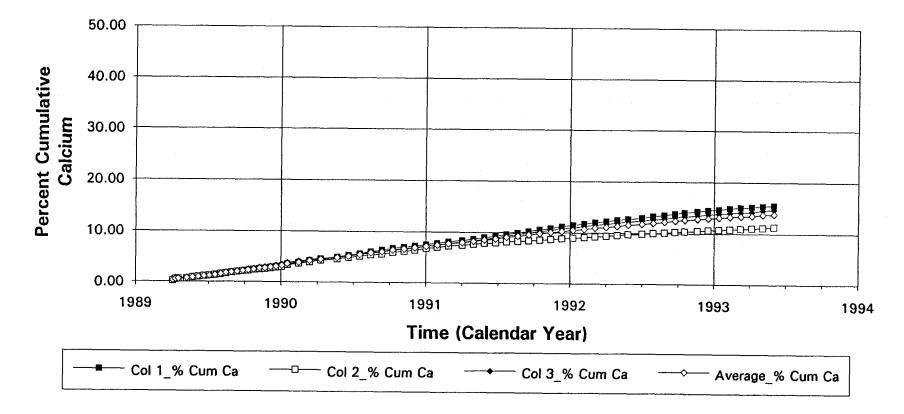
## Homogenized Total Mill Tailings (Control-1) Cumulative Total Iron Removal vs. Time

Fig. 16 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative total Fe removal with time.



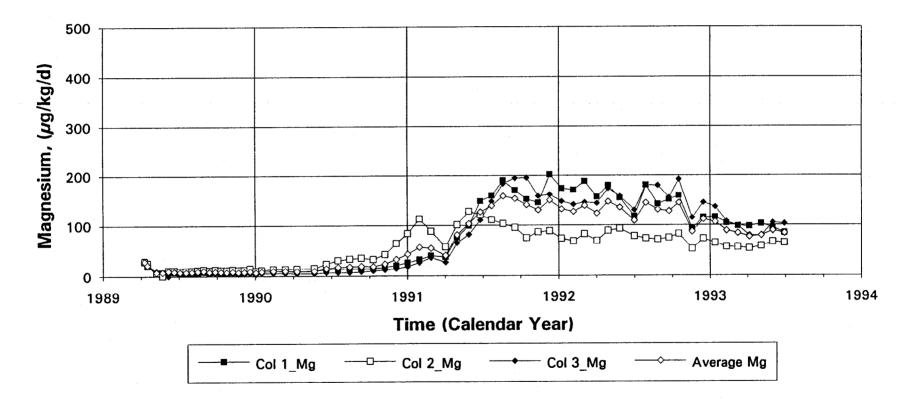
#### Homogenized Total Mill Tailings (Control-1) Calcium Loading vs. Time

Fig. 17 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Ca loading rate with time.



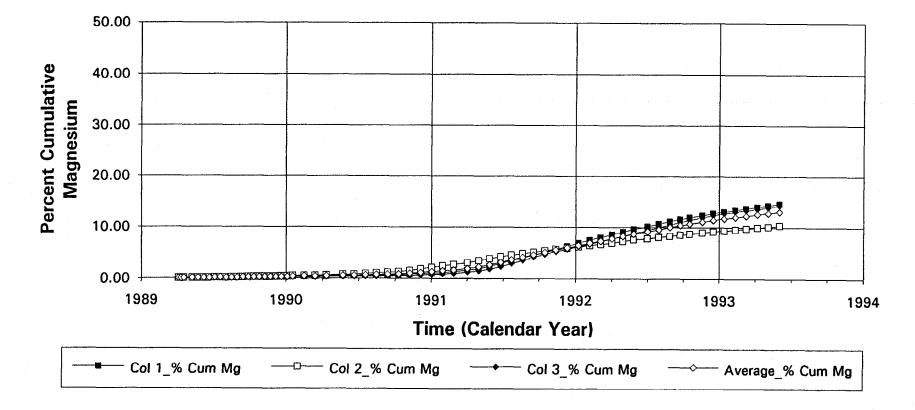
# Homogenized Total Mill Tailings (Control-1) Cumulative Calcium Removal vs. Time

Fig. 18 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative Ca removal with time.



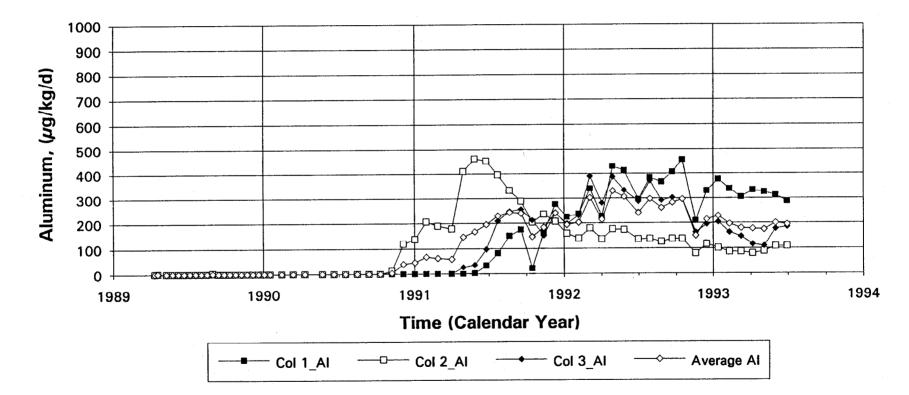
Homogenized Total Mill Tailings (Control-1) Magnesium Loading vs. Time

Fig. 19 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Mg loading rate with time.



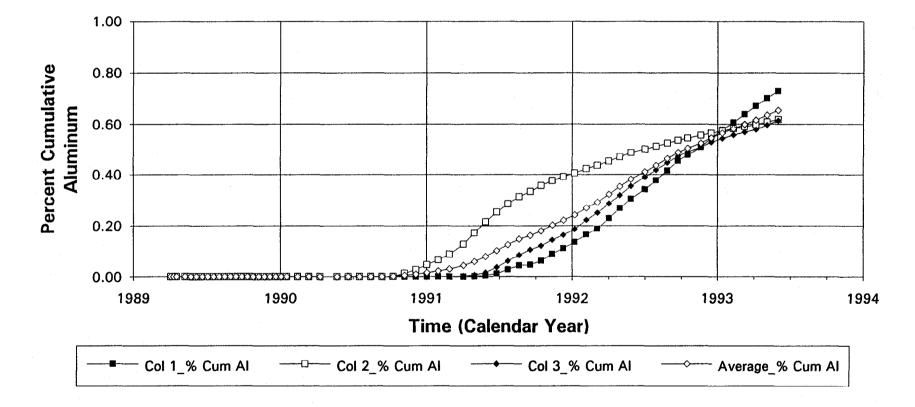
## Homogenized Total Mill Tailings (Control-1) Cumulative Magnesium Removal vs. Time

Fig. 20 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative Mg removal with time.



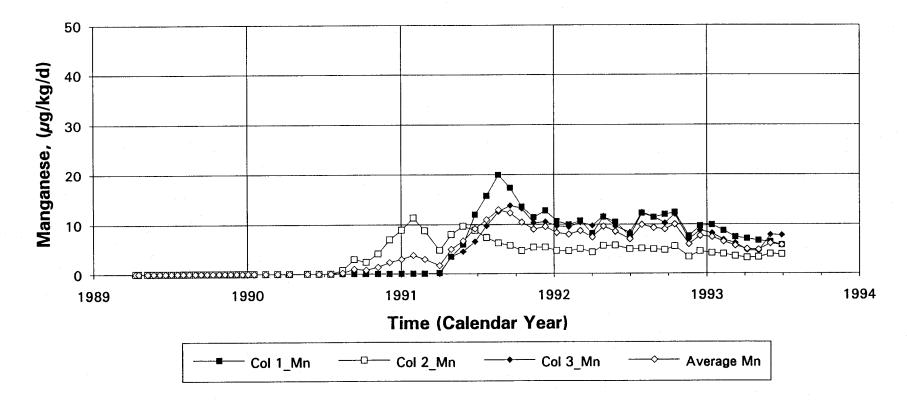
#### Homogenized Total Mill Tailings (Control-1) Aluminum Loading vs. Time

Fig. 21 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Al loading rate with time.



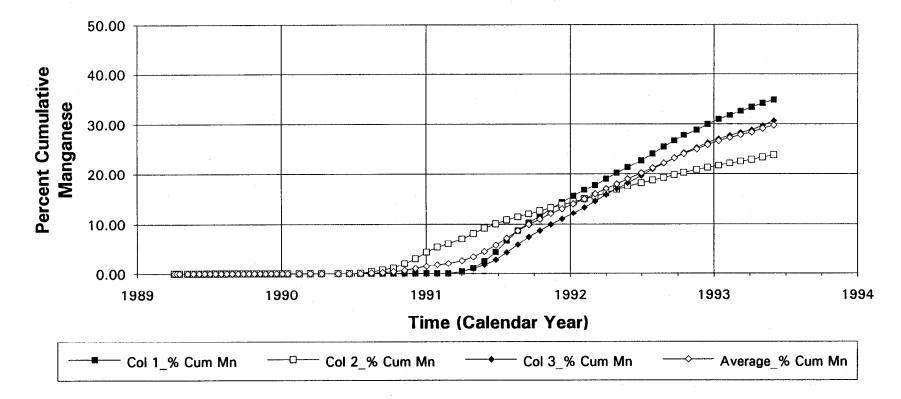
Homogenized Total Mill Tailings (Control-1) Cumulative Aluminum Removal vs. Time

Fig. 22 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative Al removal with time.



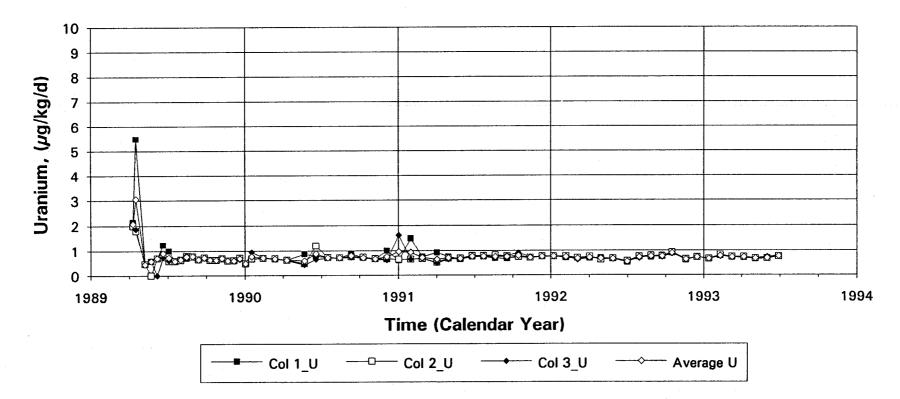
Homogenized Total Mill Tailings (Control-1) Manganese Loading vs. Time

Fig. 23 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Mn loading rate with time.



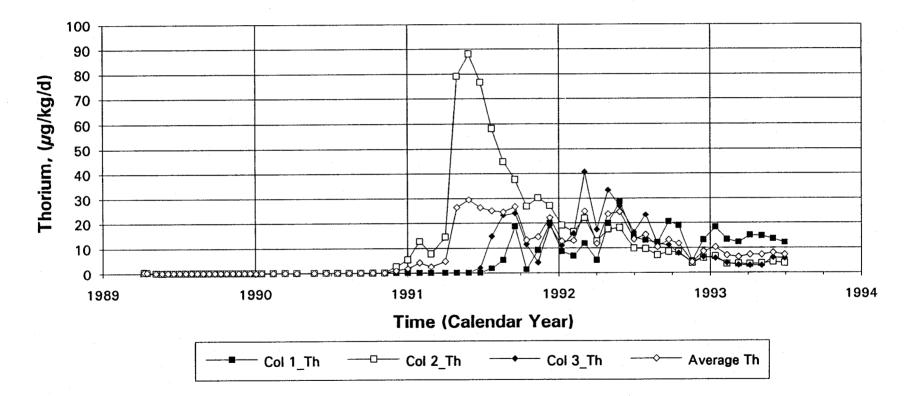
Homogenized Total Mill Tailings (Control-1) Cumulative Manganese Removal vs. Time

Fig. 24 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative Mn removal with time.



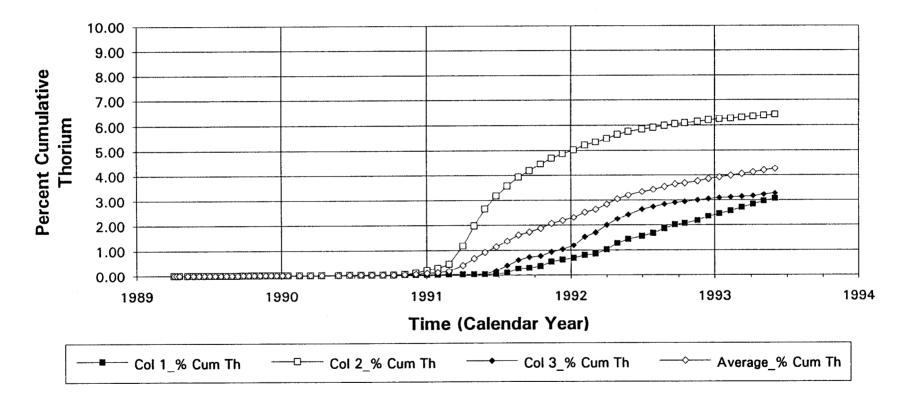
# Homogenized Total Mill Tailings (Control-1) Uranium Loading vs. Time

Fig. 25 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved U loading rate with time.



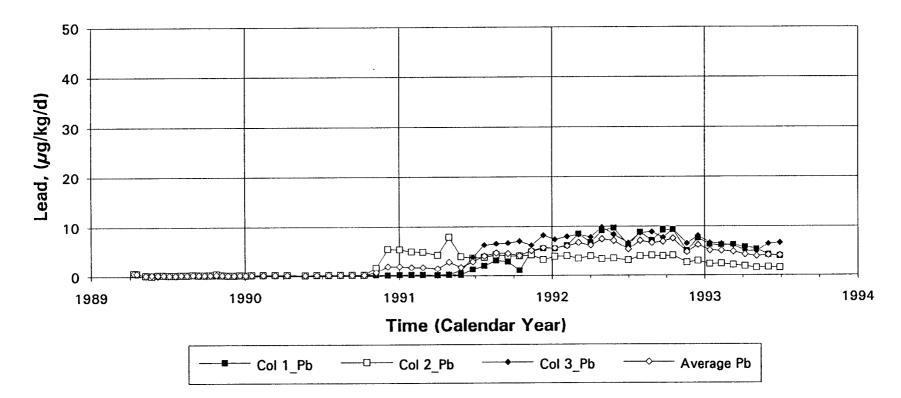
#### Homogenized Total Mill Tailings (Control-1) Thorium Loading vs. Time

Fig. 26 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Th loading rate with time.



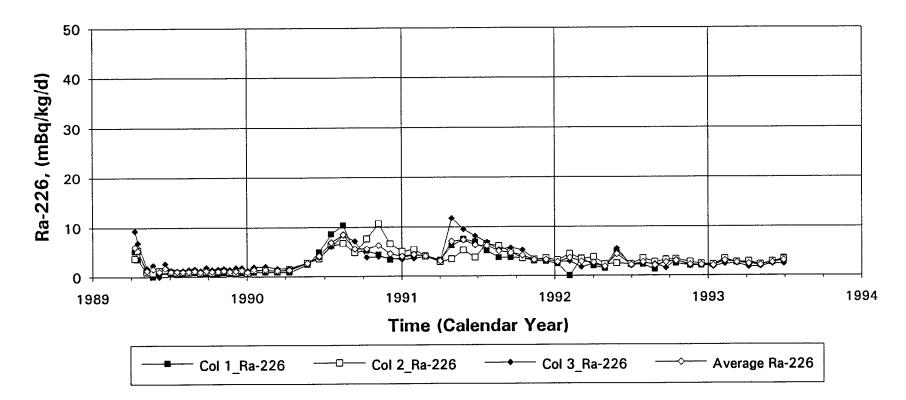
#### Homogenized Total Mill Tailings (Control-1) Cumulative Thorium Removal vs. Time

Fig. 27 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative Th removal with time.



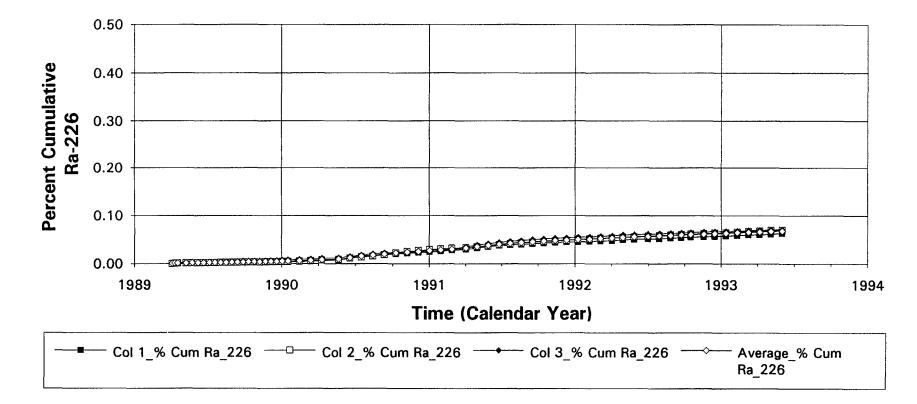
Homogenized Total Mill Tailings (Control-1) Lead Loading vs. Time

Fig. 28 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Pb loading rate with time.



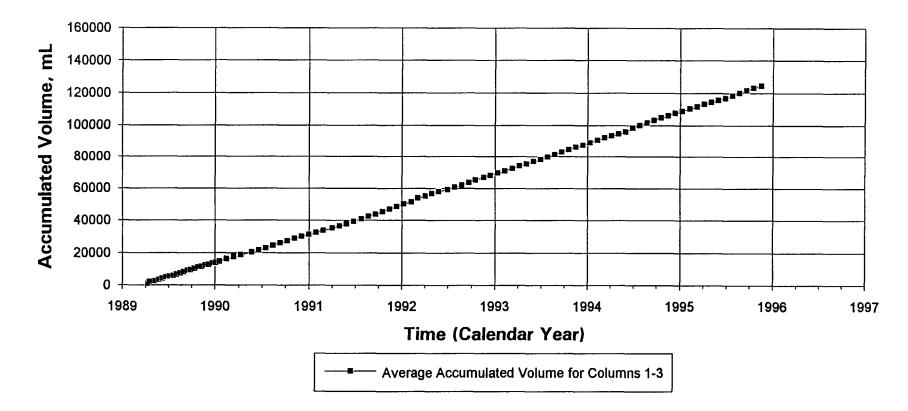
Homogenized Total Mill Tailings (Control-1) Ra-226 Loading vs. Time

Fig. 29 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), variation of dissolved Ra-226 loading rate with time.



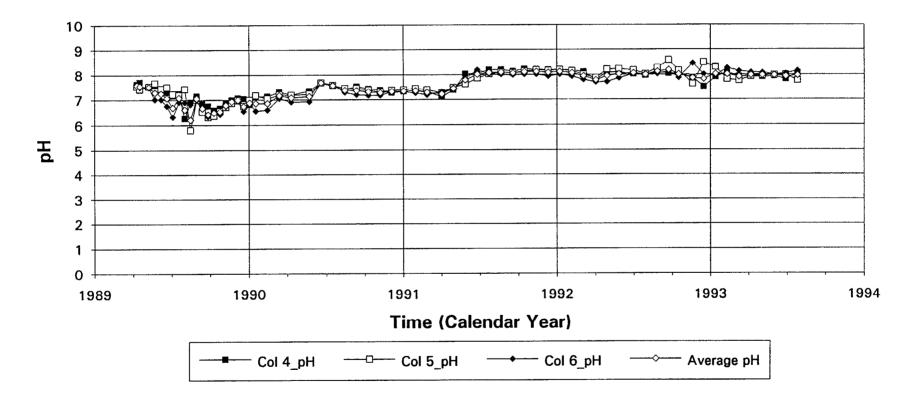
Homogenized Total Mill Tailings (Control-1) Cumulative Ra-226 Removal vs. Time

Fig. 30 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), cumulative Ra-226 removal with time.



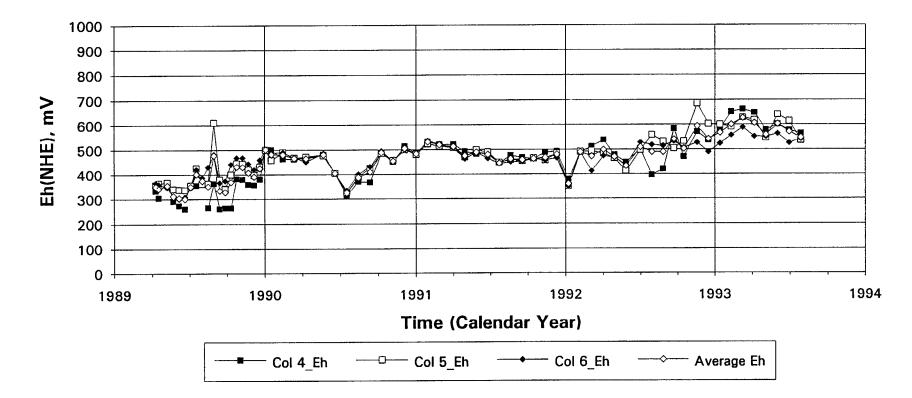
## Homogenized Total Mill Tailings (Control-1) Accumulated Volume vs. Time

Fig. 31 Leaching profiles of total mill tailings without additional limestone and under unsaturated conditions (Control - 1, columns #1-3), average cumulative drainage volume collected with time.



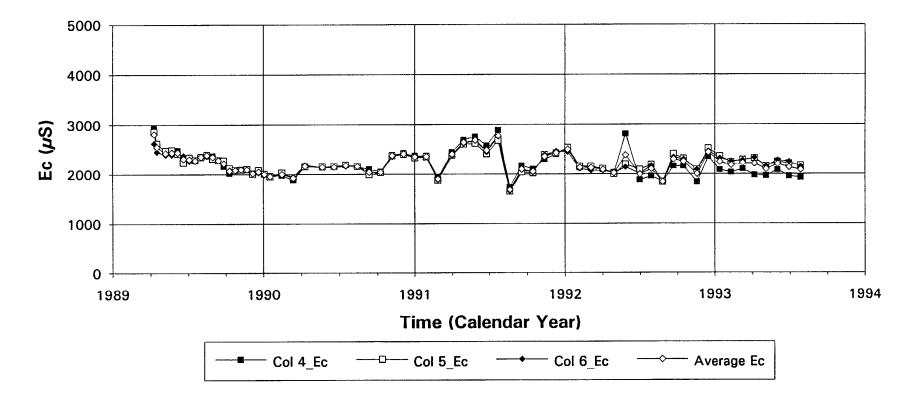
#### Homogenized Total Mill Tailings, Coarse Limestone pH vs. Time

Fig. 32 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of pH with time.



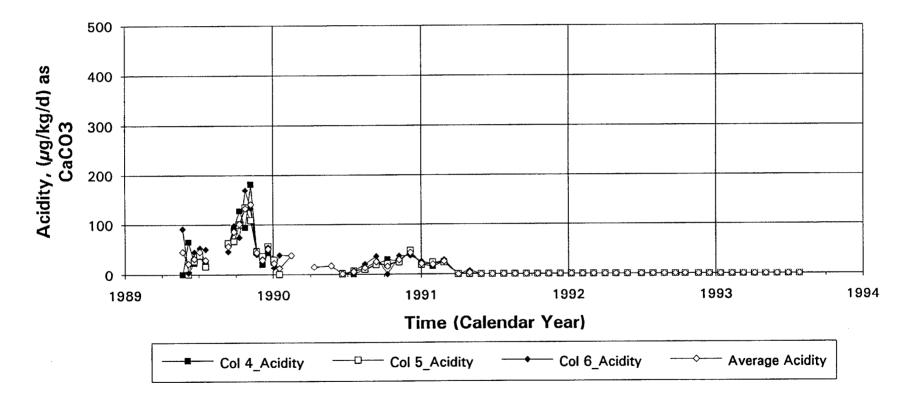
#### Homogenized Total Mill Tailings, Coarse Limestone Eh(NHE) vs. Time

Fig. 33 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of Eh with time.



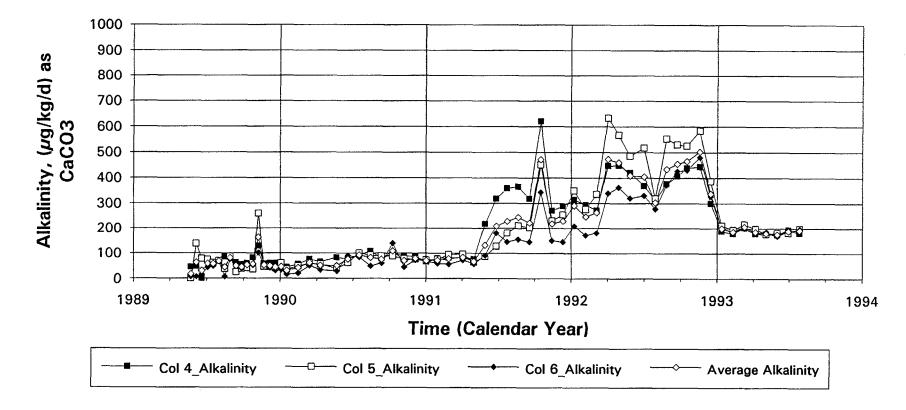
## Homogenized Total Mill Tailings, Coarse Limestone Electrical Conductivity vs. Time

Fig. 34 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of Ec with time.



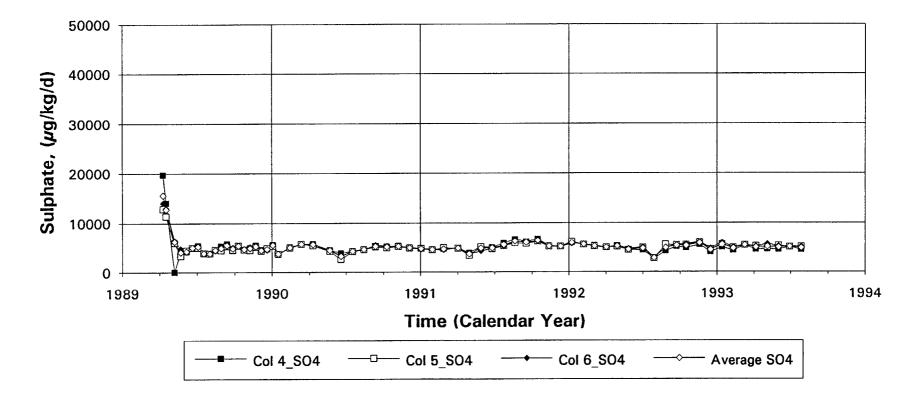
## Homogenized Total Mill Tailings, Coarse Limestone Acidity Loading vs. Time

Fig. 35 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of total acidity loading rate with time.



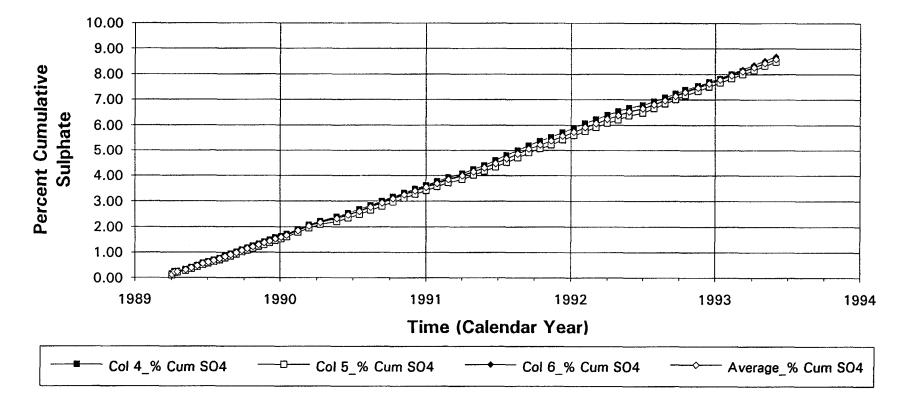
# Homogenized Total Mill Tailings, Coarse Limestone Alkalinity Loading vs. Time

Fig. 36 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of total alkalinity loading rate with time.



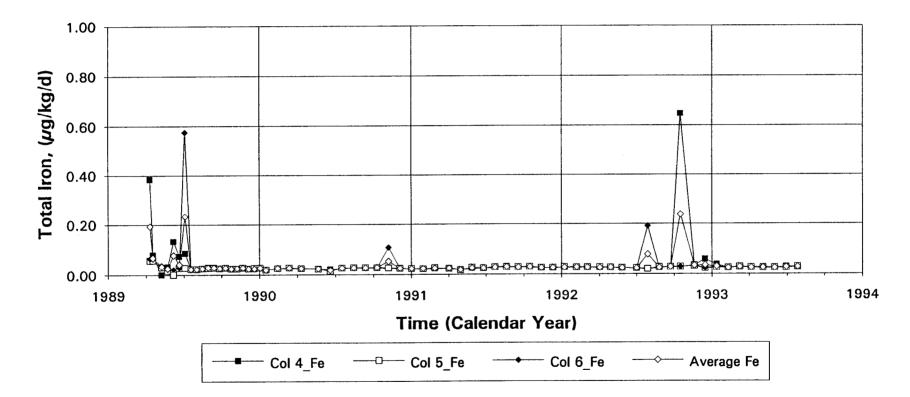
#### Homogenized Total Mill Tailings, Coarse Limestone Sulphate Loading vs. Time

Fig. 37 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved  $SO_4^{-2}$  loading rate with time.



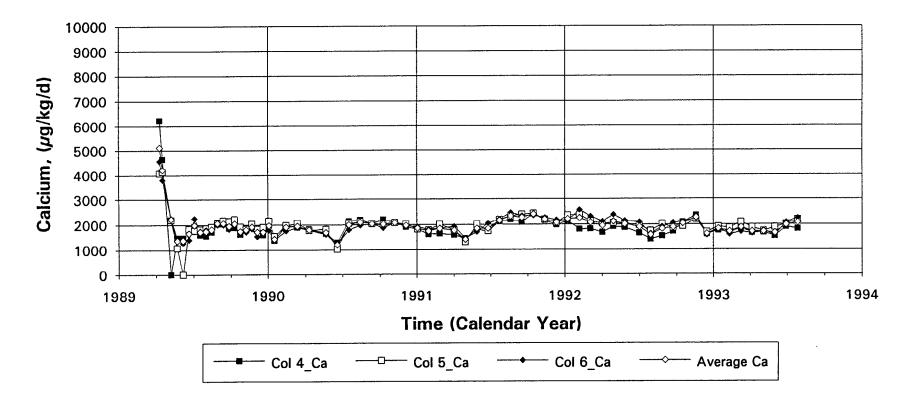
## Homogenized Total Mill Tailings, Coarse Limestone Cumulative Sulphate Removal vs. Time

Fig. 38 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



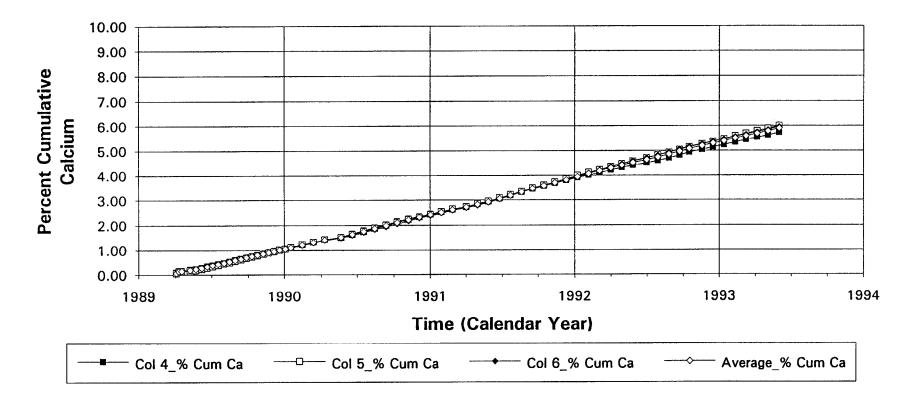
### Homogenized Total Mill Tailings, Coarse Limestone Total Iron Loading vs. Time

Fig. 39 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved total Fe loading rate with time.



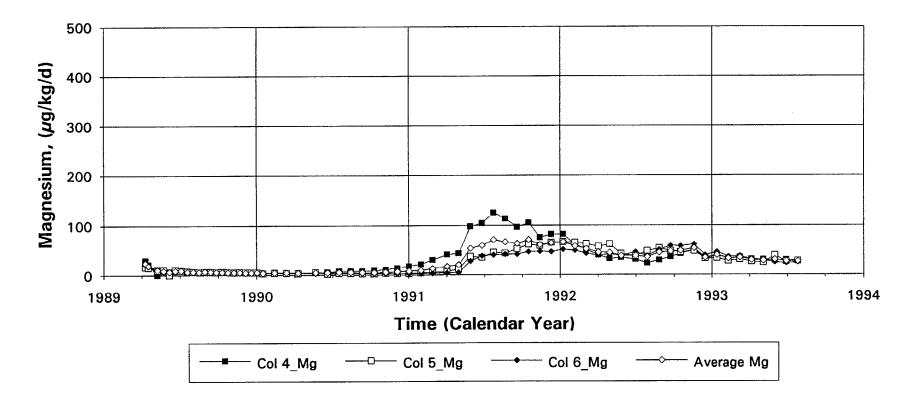
## Homogenized Total Mill Tailings, Coarse Limestone Calcium Loading vs. Time

Fig. 40 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Ca loading rate with time.



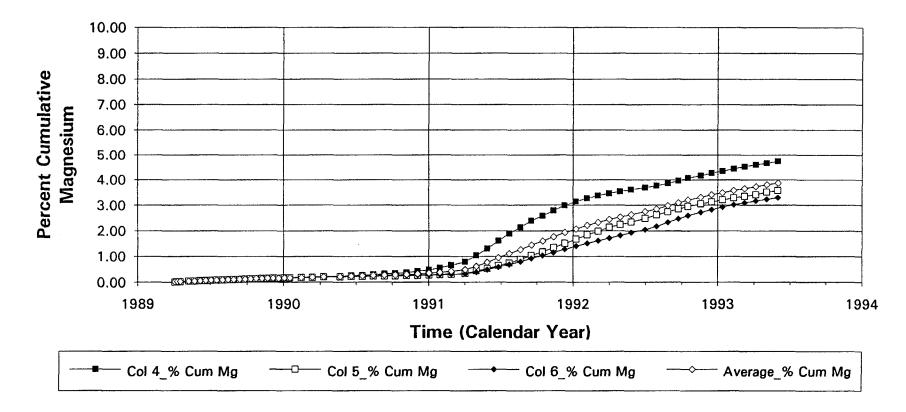
#### Homogenized Total Mill Tailings, Coarse Limestone Cumulative Calcium Removal vs. Time

Fig. 41 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), cumulative Ca removal with time.



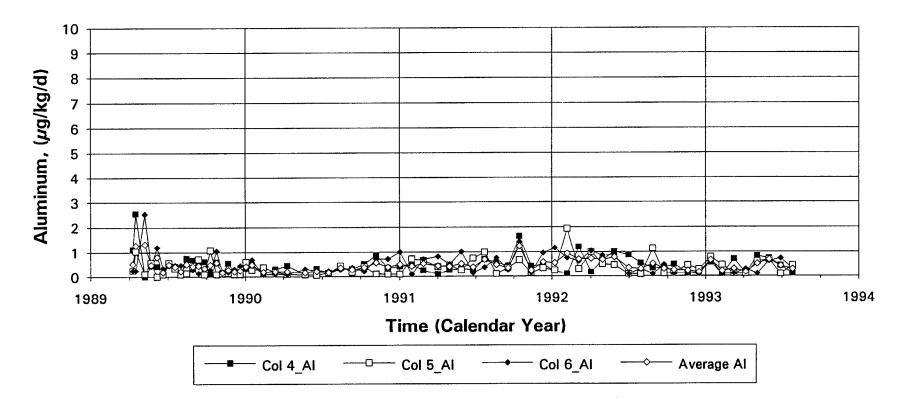
#### Homogenized Total Mill Tailings, Coarse Limestone Magnesium Loaded vs. Time

Fig. 42 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Mg loading rate with time.



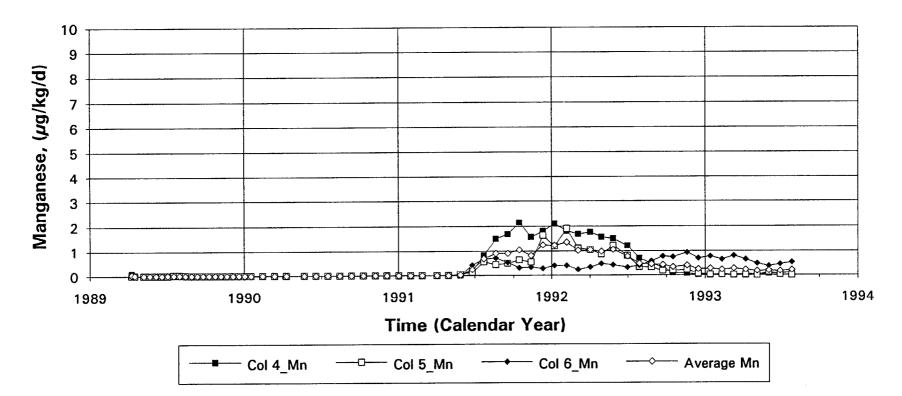
## Homogenized Total Mill Tailings, Coarse Limestone Cumulative Magnesium Removal vs. Time

Fig. 43 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), cumulative Mg removal with time.



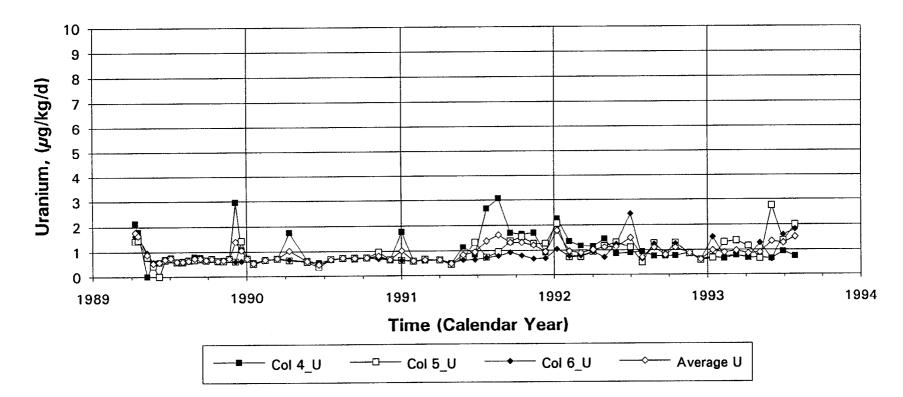
## Homogenized Total Mill Tailings, Coarse Limestone Aluminum Loading vs. Time

Fig. 44 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Al loading rate with time.



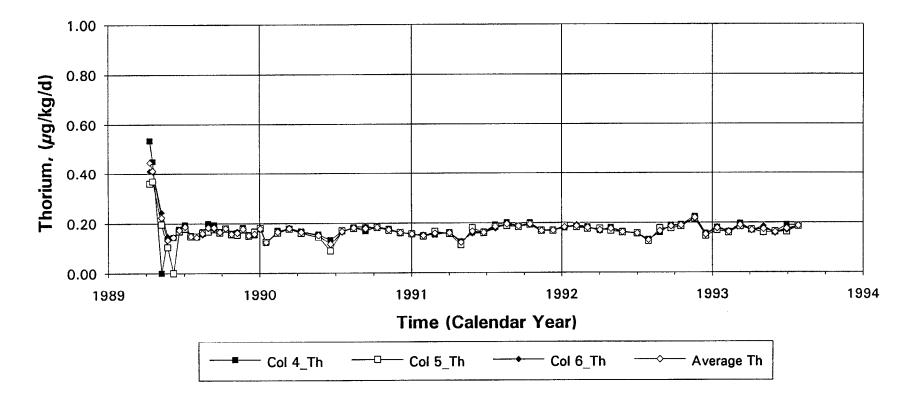
## Homogenized Total Mill Tailings, Coarse Limestone Manganese Loading vs. Time

Fig. 45 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Mn loading rate with time.



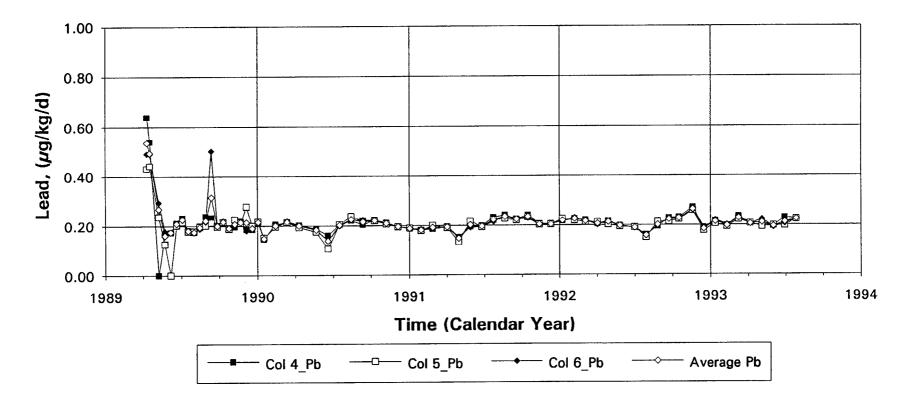
# Homogenized Total Mill Tailings, Coarse Limestone Uranium Loading vs. Time

Fig. 46 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved U loading rate with time.



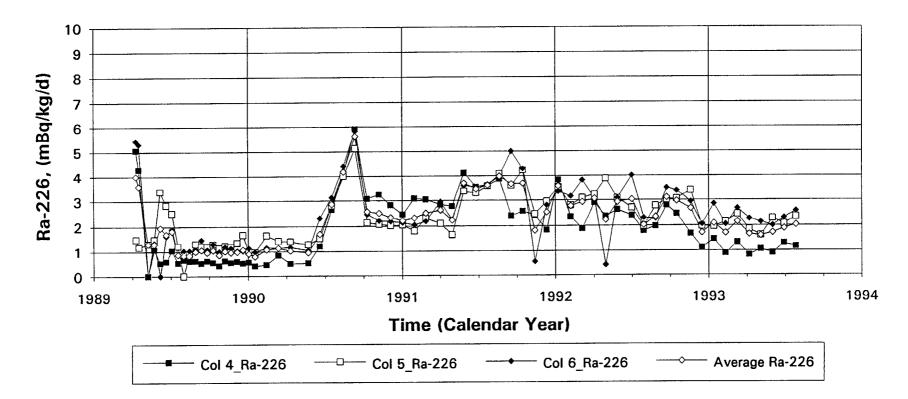
# Homogenized Total Mill Tailings, Coarse Limestone Thorium Loading vs. Time

Fig. 47 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Th loading rate with time.



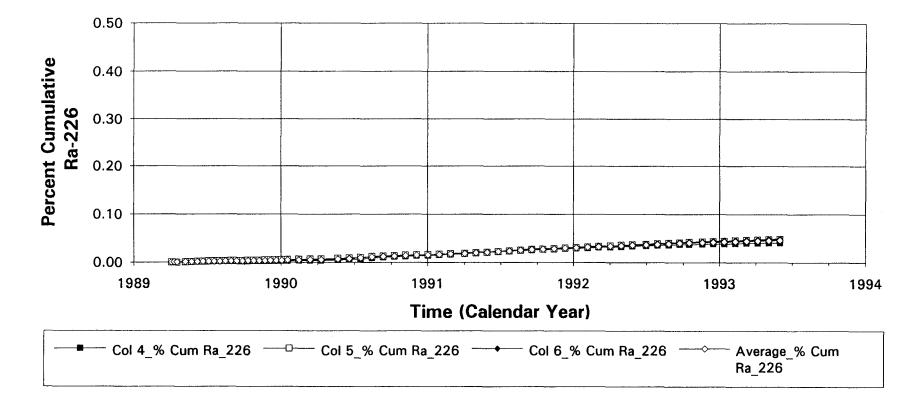
Homogenized Total Mill Tailings, Coarse Limestone Lead Loading vs. Time

Fig. 48 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Pb loading rate with time.



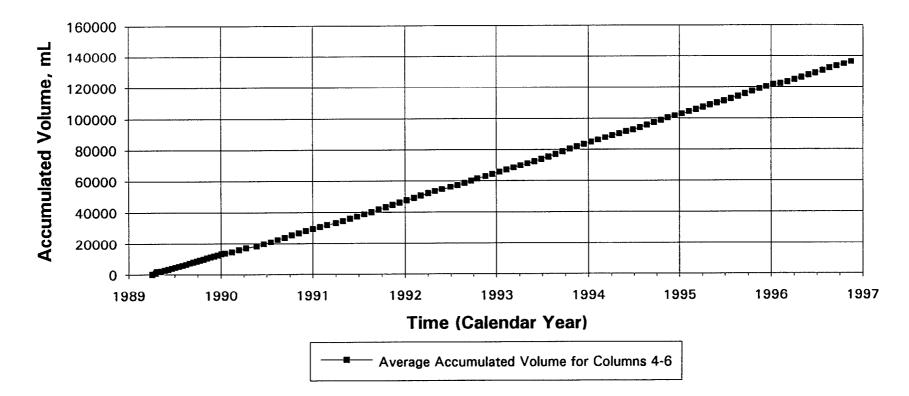
#### Homogenized Total Mill Tailings, Coarse Limestone Ra-226 Loading vs. Time

Fig. 49 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), variation of dissolved Ra-226 loading rate with time.



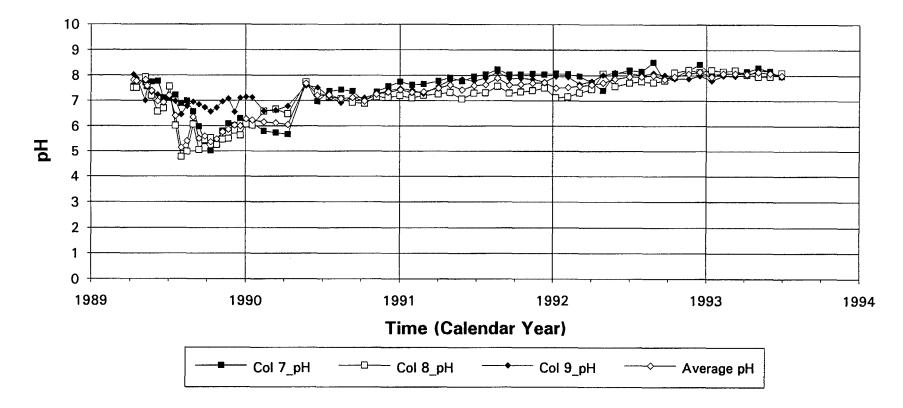
## Homogenized Total Mill Tailings, Coarse Limestone Cumulative Ra-226 Removal vs. Time

Fig. 50 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), cumulative Ra-226 removal with time.



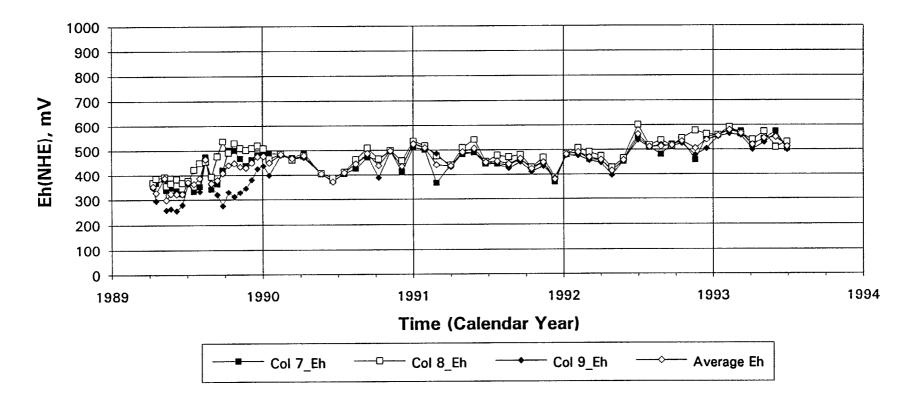
# Homogenized Total Mill Tailings, Coarse Limestone Accumulated Volume vs. Time

Fig. 51 Leaching profiles of total mill tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #4-6), average cumulative drainage volume collected with time.



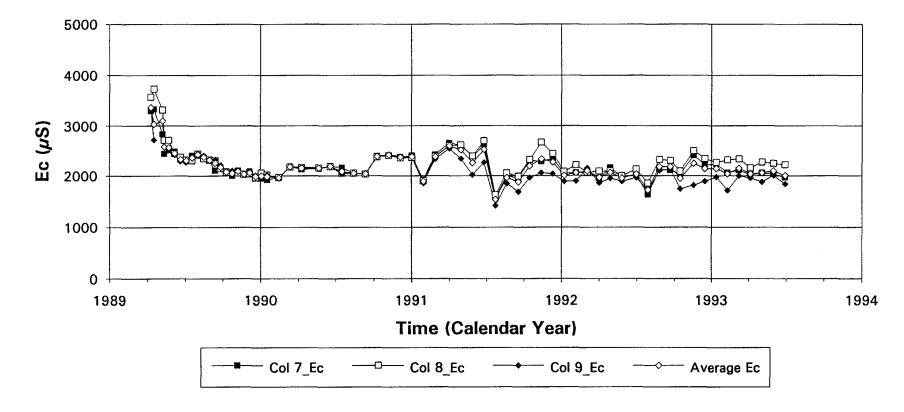
Homogenized Total Mill Tailings, Medium Size Limestone pH vs. Time

Fig. 52 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of pH with time.



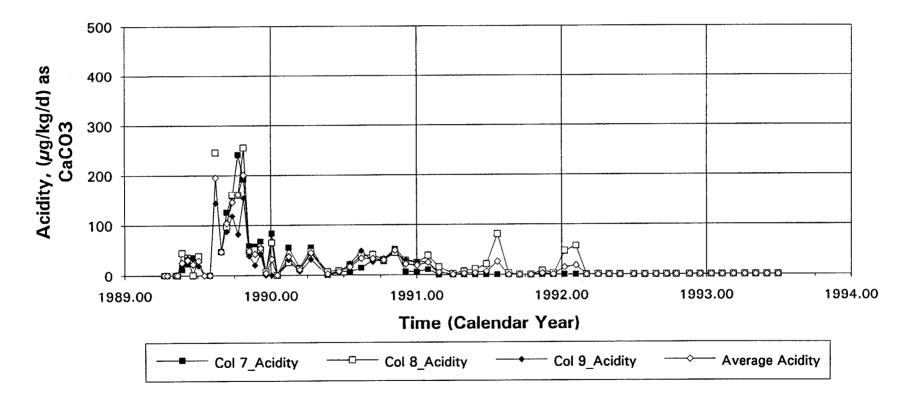
## Homogenized Total Mill Tailings, Medium Size Limestone Eh(NHE) vs. Time

Fig. 53 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of Eh with time.



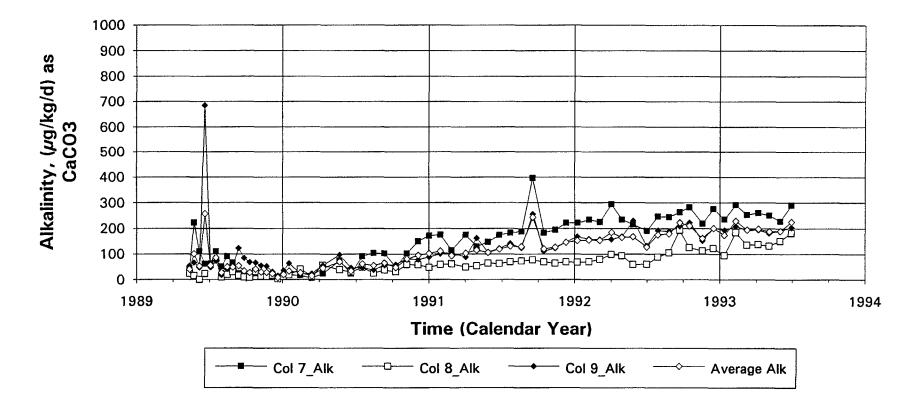
## Homogenized Total Mill Tailings, Medium Size Limestone Electrical Conductivity vs. Time

Fig. 54 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of Ec with time.



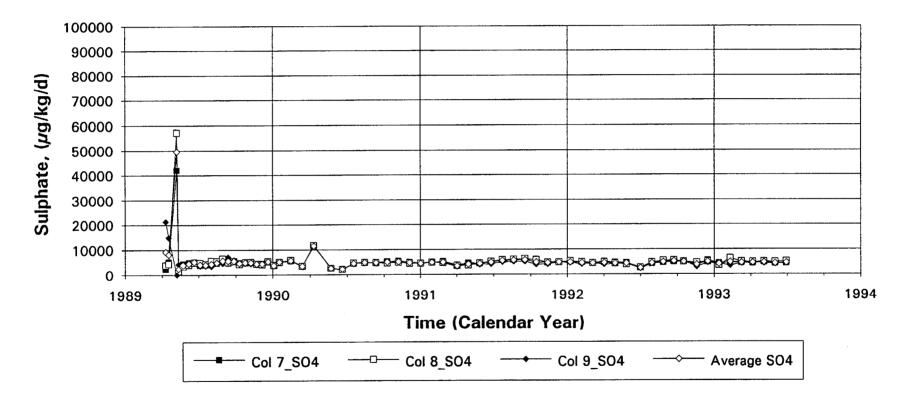
# Homogenized Total Mill Tailings, Medium Size Limestone Acidity Loading vs. Time

Fig. 55 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of total acidity loading rate with time.



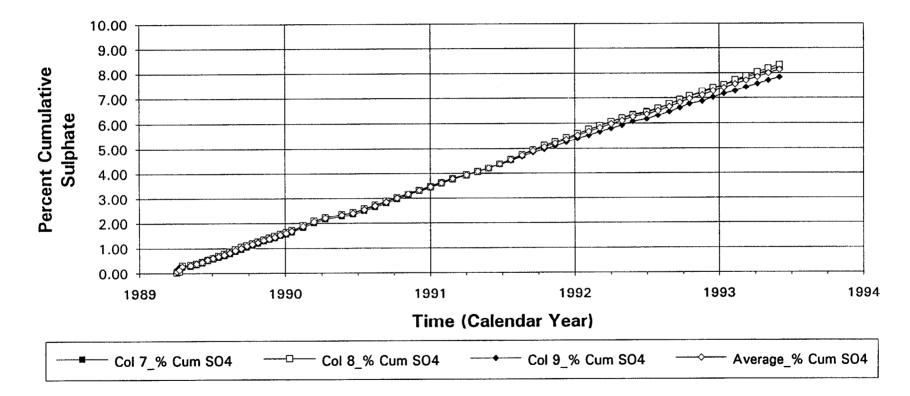
# Homogenized Total Mill Tailings, Medium Size Limestone Alkalinity Loading vs. Time

Fig. 56 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of total alkalinity loading rate with time.



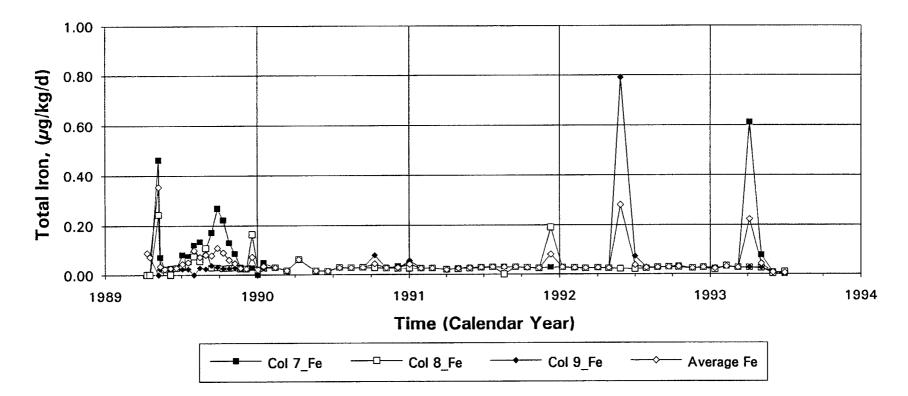
# Homogenized Total Mill Tailings, Medium Size Limestone Sulphate Loading vs. Time

Fig. 57 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved  $SO_4^{-2}$  loading rate with time.



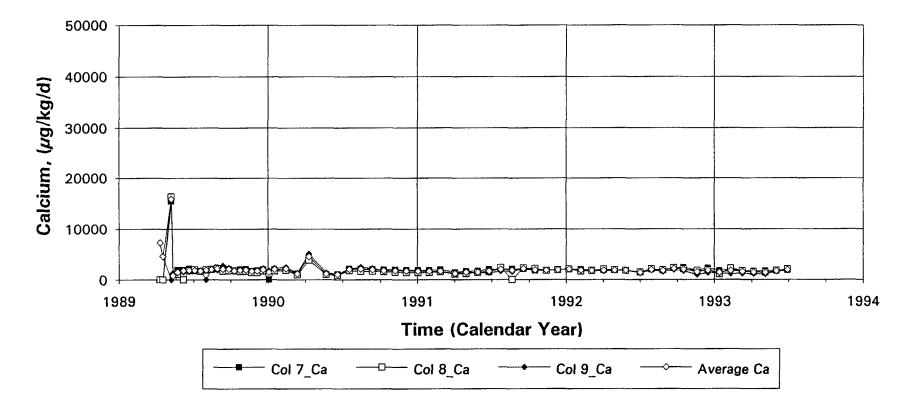
### Homogenized Total Mill Tailings, Medium Size Limestone Cumulative Sulphate Removal vs. Time

Fig. 58 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



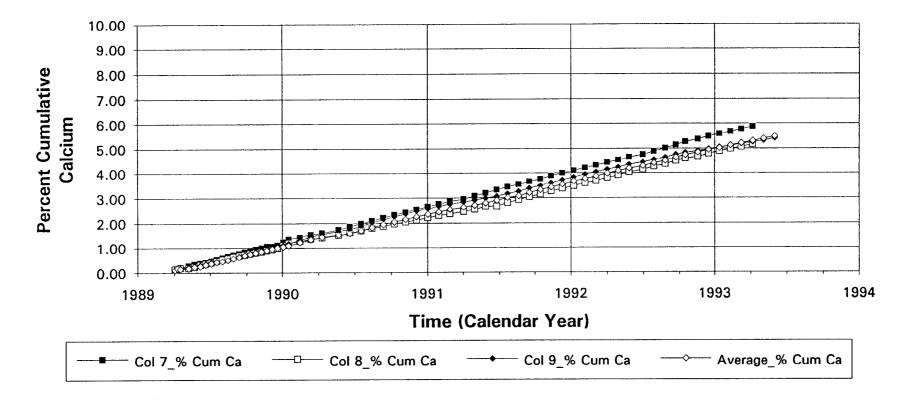
Homogenized Total Mill Tailings, Medium Size Limestone Total Iron Loading vs. Time

Fig. 59 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved total Fe loading rate with time.



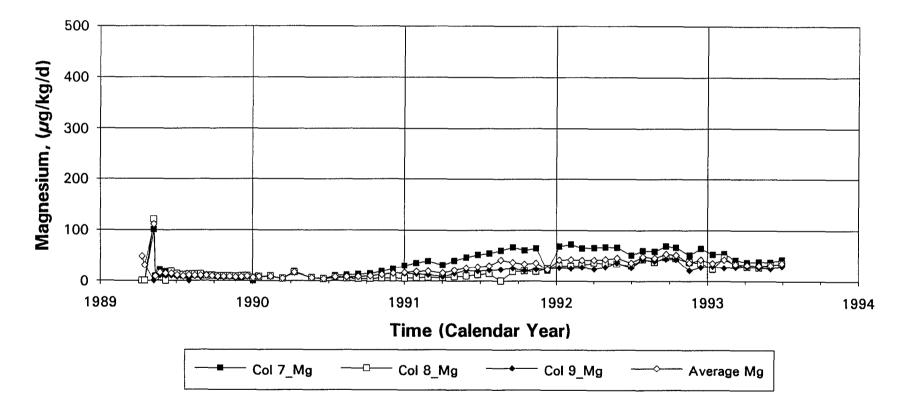
# Homogenized Total Mill Tailings, Medium Size Limestone Calcium Loading vs. Time

Fig. 60 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Ca loading rate with time.



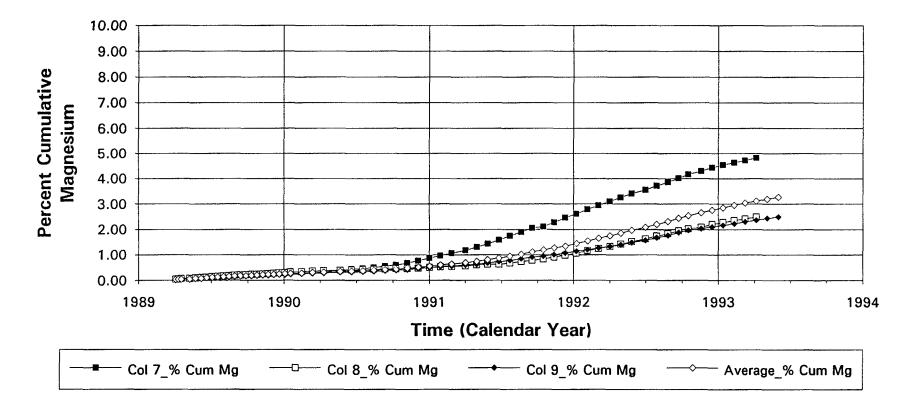
#### Homogenized Total Mill Tailings, Medium Size Limestone Cumulative Calcium Removal vs. Time

Fig. 61 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), cumulative Ca removal with time.



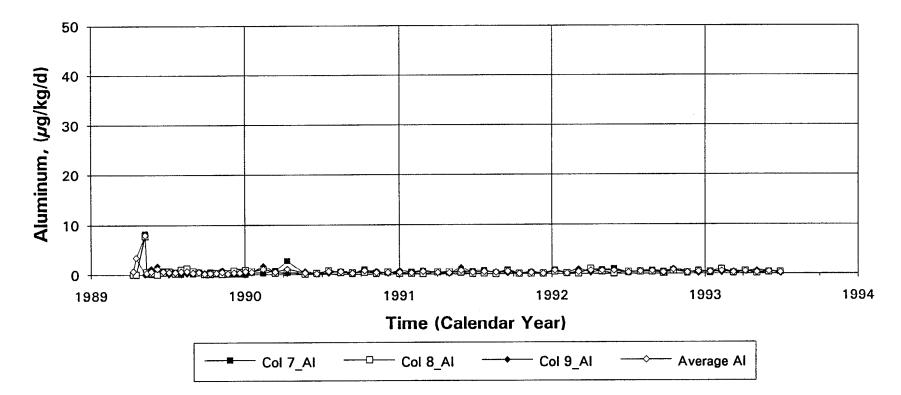
## Homogenized Total Mill Tailings, Medium Size Limestone Magnesium Loading vs. Time

Fig. 62 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Mg loading rate with time.



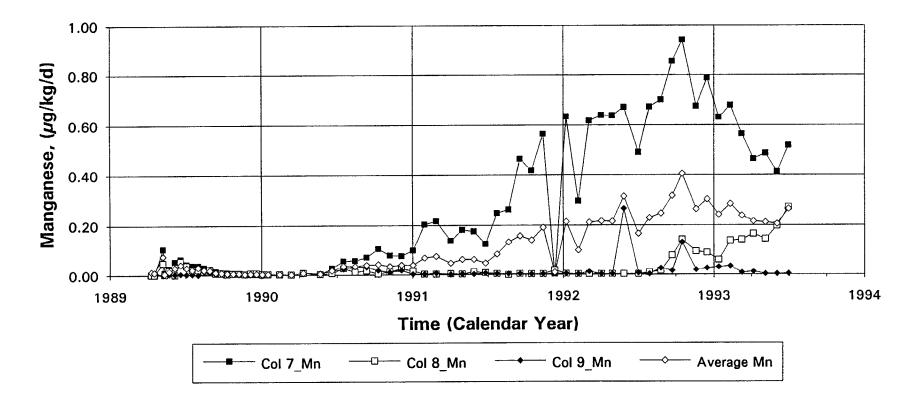
## Homogenized Total Mill Tailings, Medium Size Limestone Cumulative Magnesium Removal vs. Time

Fig. 63 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), cumulative Mg removal with time.



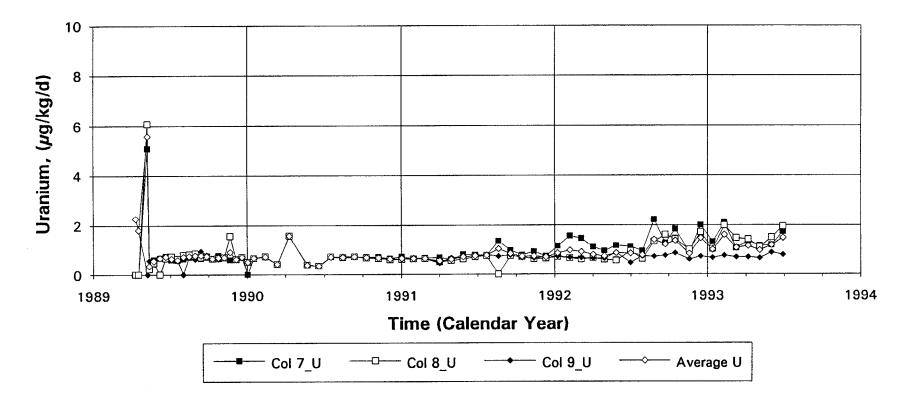
## Homogenized Total Mill Tailings, Medium Size Limestone Aluminum Loading vs. Time

Fig. 64 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Al loading rate with time.



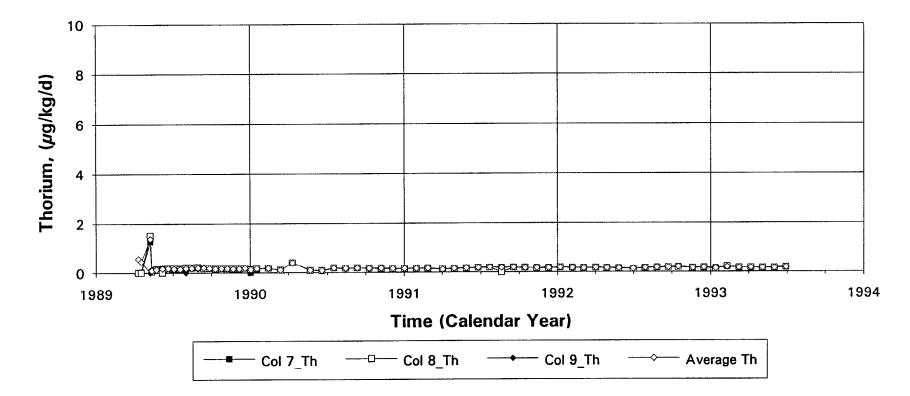
### Homogenized Total Mill Tailings, Medium Size Limestone Manganese Loading vs. Time

Fig. 65 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Mn loading rate with time.



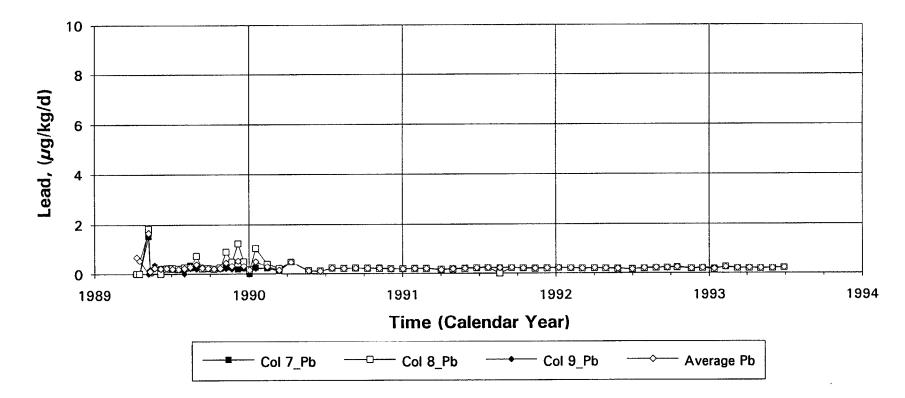
#### Homogenized Total Mill Tailings, Medium Size Limestone Uranium Loading vs. Time

Fig. 66 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved U loading rate with time.



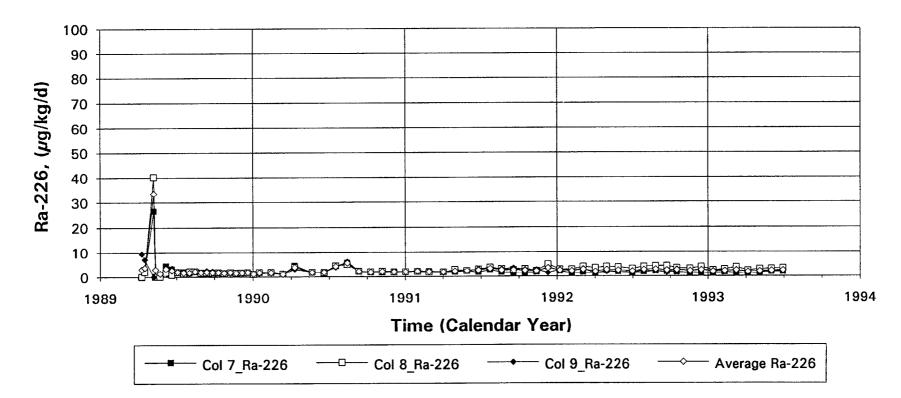
#### Homogenized Total Mill Tailings, Medium Size Limestone Thorium Loading vs. Time

Fig. 67 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Th loading rate with time.



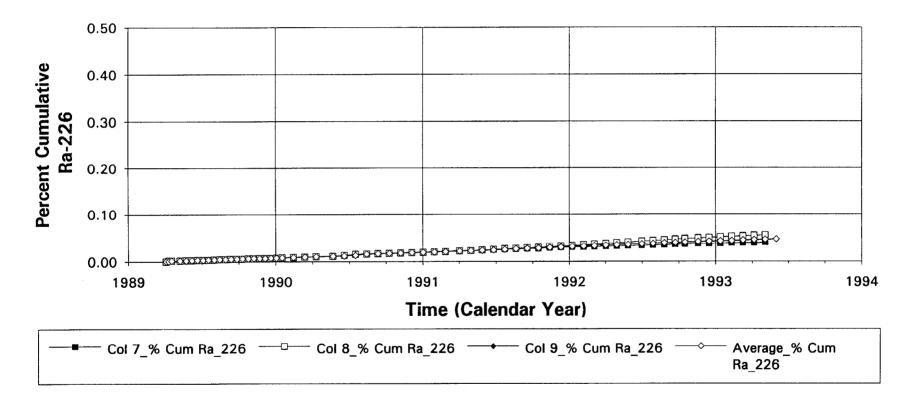
## Homogenized Total Mill Tailings, Medium Size Limestone Lead Loading vs. Time

Fig. 68 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Pb loading rate with time.



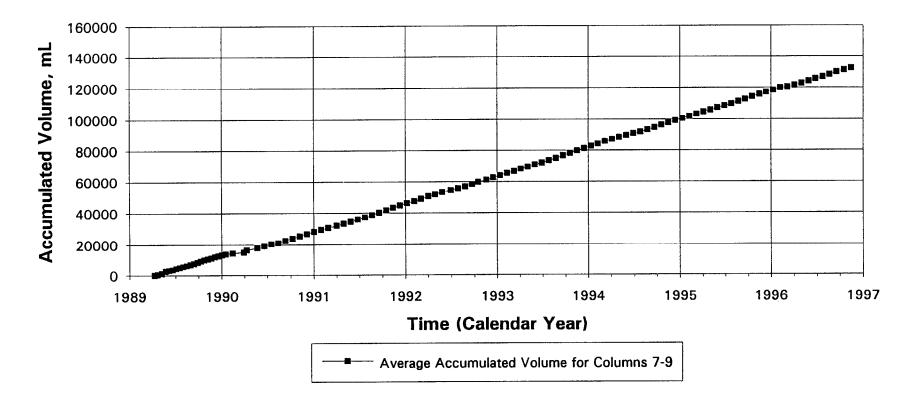
# Homogenized Total Mill Tailings, Medium Size Limestone Ra-226 Loading vs. Time

Fig. 69 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), variation of dissolved Ra-226 loading rate with time.



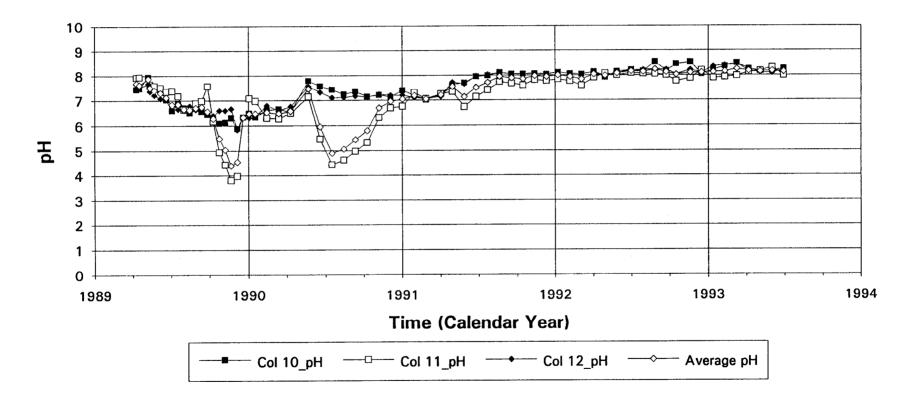
#### Homogenized Total Mill Tailings, Medium Size Limestone Cumulative Ra-226 Removal vs. Time

Fig. 70 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), cumulative Ra-226 removal with time.



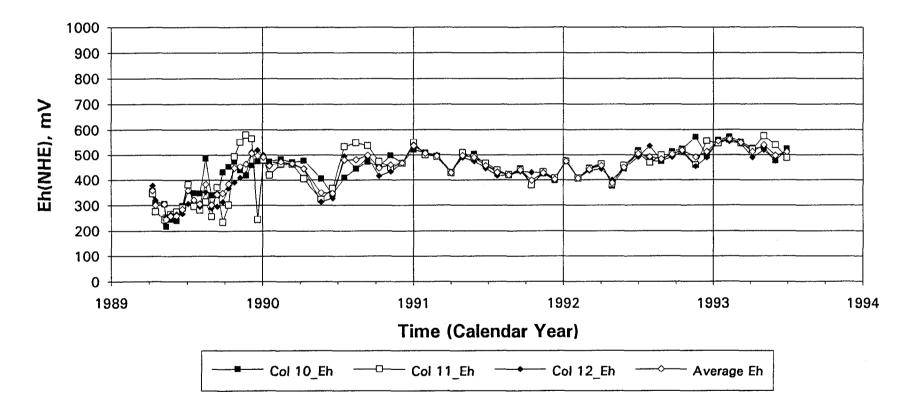
#### Homogenized Total Mill Tailings, Medium Size Limestone Accumulated Volume vs. Time

Fig. 71 Leaching profiles of total mill tailings mixed with 7.5% (w/w) medium size limestone (screen size -2.4 mm, -8 mesh) and under unsaturated conditions (columns #7-9), average cumulative drainage volume collected with time.



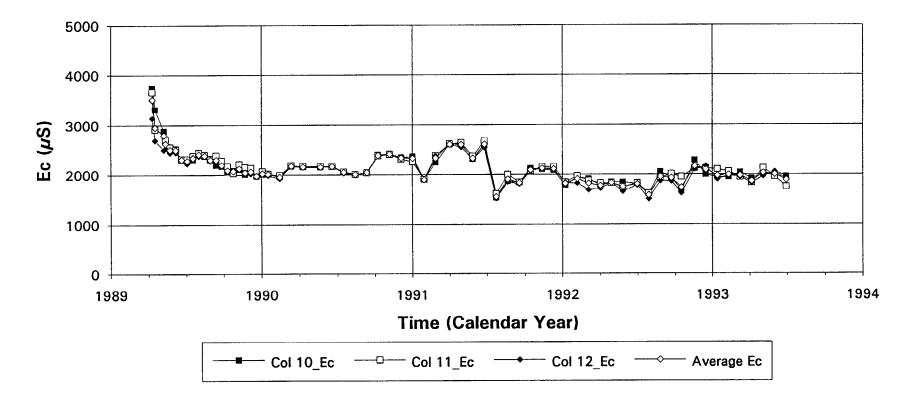
## Homogenized Total Mill Tailings, Fine Limestone pH vs. Time

Fig. 72 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of pH with time.



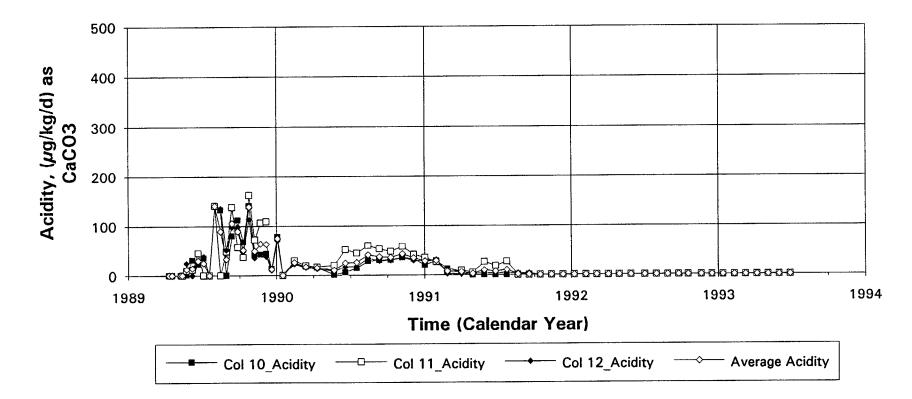
## Homogenized Total Mill Tailings, Fine Limestone Eh(NHE) vs. Time

Fig. 73 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of Eh with time.



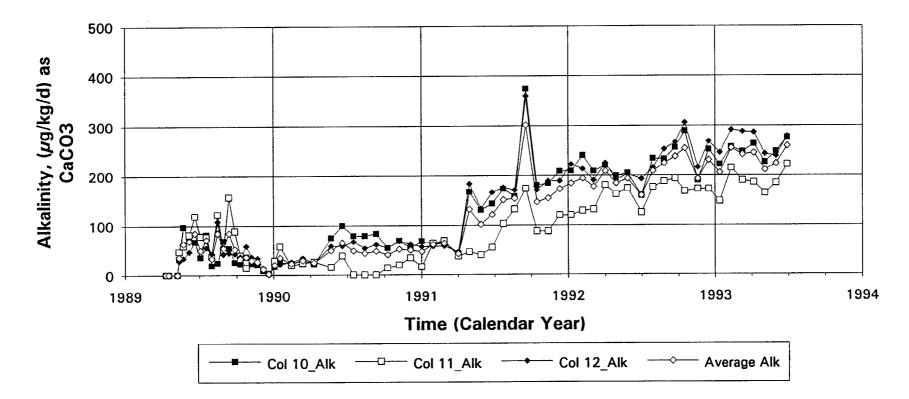
#### Homogenized Total Mill Tailings, Fine Limestone Electrical Conductivity vs. Time

Fig. 74 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of Ec with time.



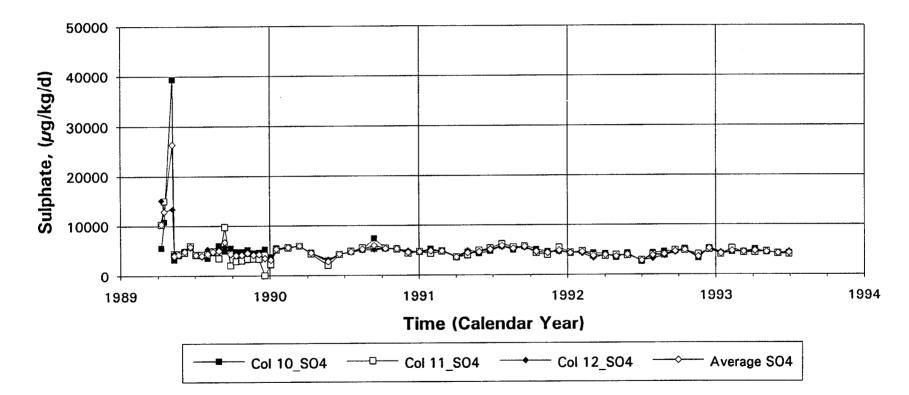
#### Homogenized Total Mill Tailings, Fine Limestone Acidity Loading vs. Time

Fig. 75 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of total acidity loading rate with time.



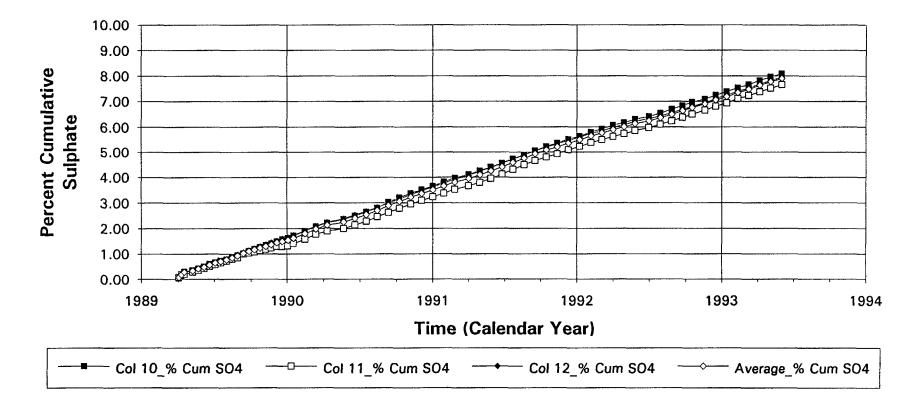
#### Homogenized Total Mill Tailings, Fine Limestone Alkalinity Loading vs. Time

Fig. 76 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of total alkalinity loading rate with time.



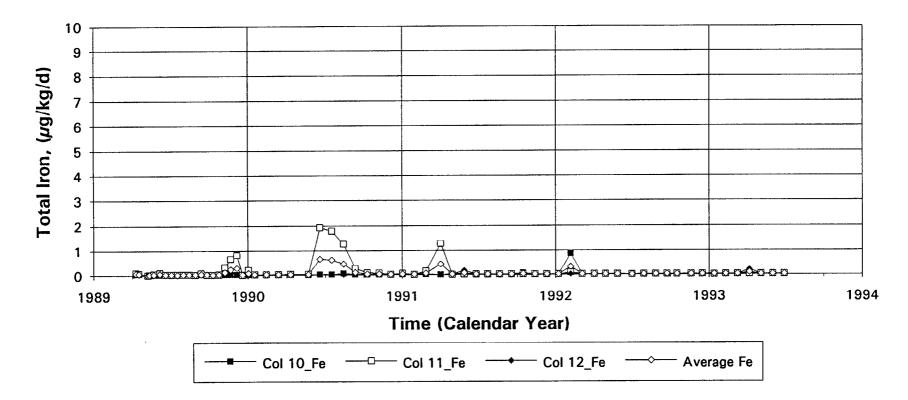
## Homogenized Total Mill Tailings, Fine Limestone Sulphate Loading vs. Time

Fig. 77 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved  $SO_4^{-2}$  loading rate with time.



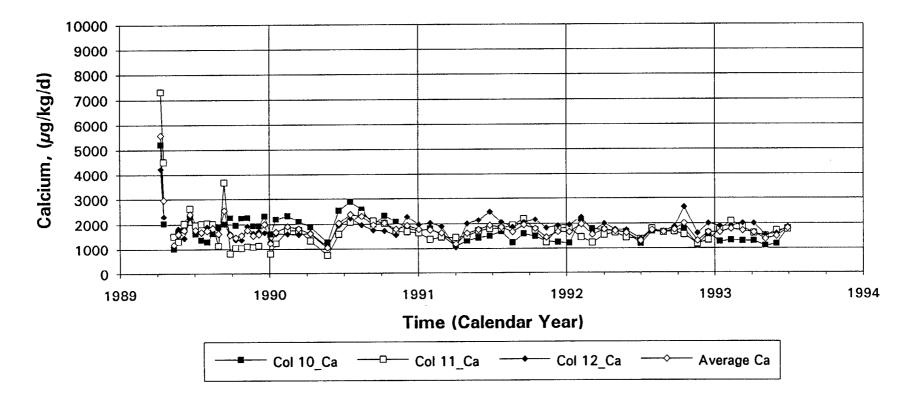
## Homogenized Total Mill Tailings, Fine Limestone Cumulative Sulphate Removal vs. Time

Fig. 78 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



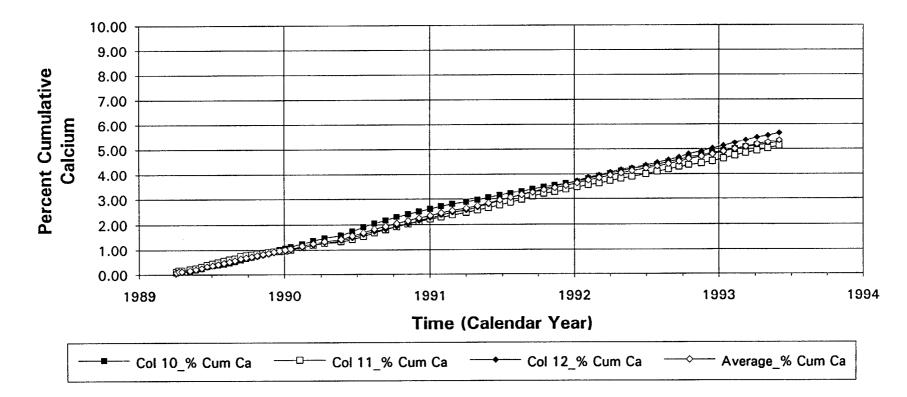
#### Homogenized Total Mill Tailings, Fine Limestone Total Iron Loading vs. Time

Fig. 79 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved total Fe loading rate with time.



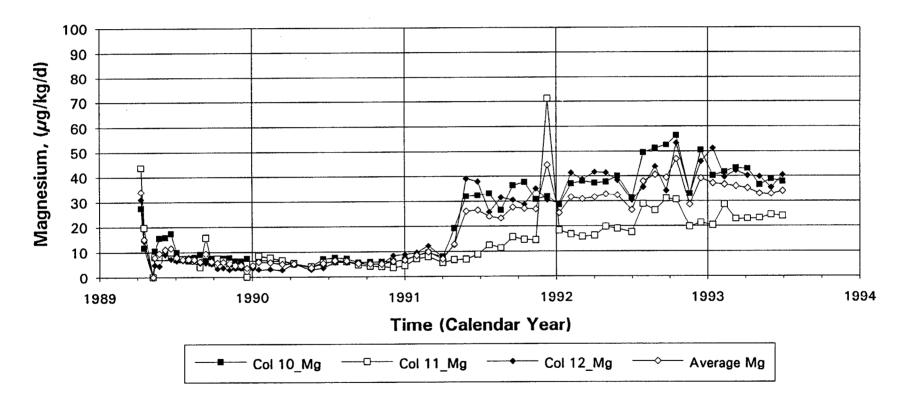
### Homogenized Total Mill Tailings, Fine Limestone Calcium Loading vs. Time

Fig. 80 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Ca loading rate with time.



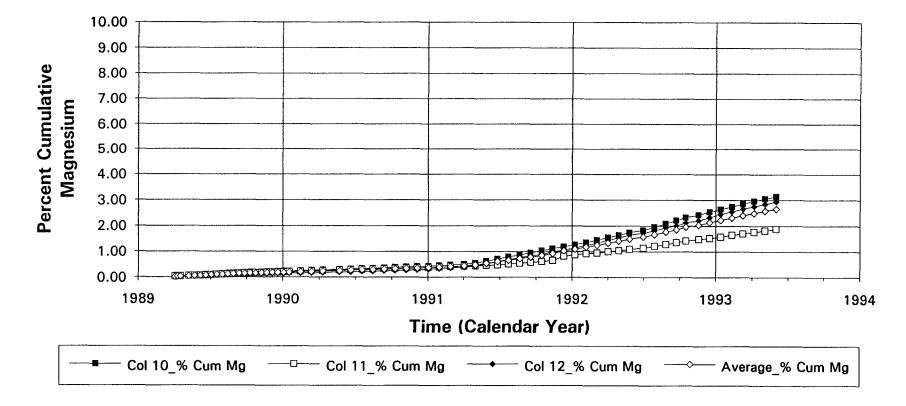
### Homogenized Total Mill Tailings, Fine Limestone Cumulative Calcium Removal vs. Time

Fig. 81 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), cumulative Ca removal with time.



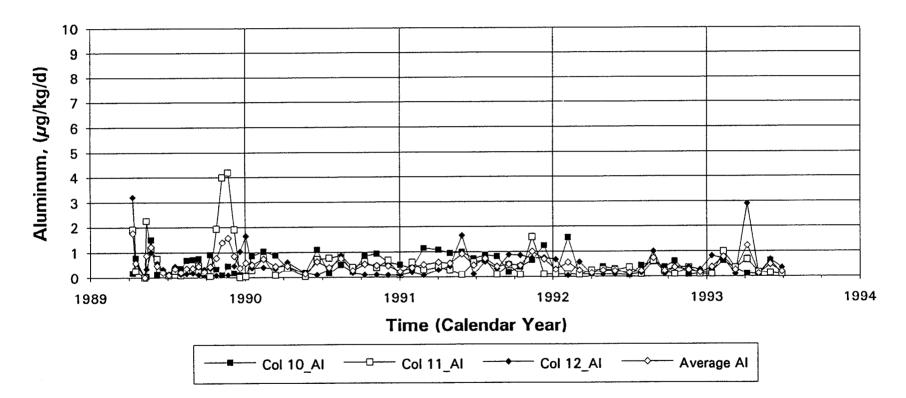
## Homogenized Total Mill Tailings, Fine Limestone Magnesium Loading vs. Time

Fig. 82 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Mg loading rate with time.



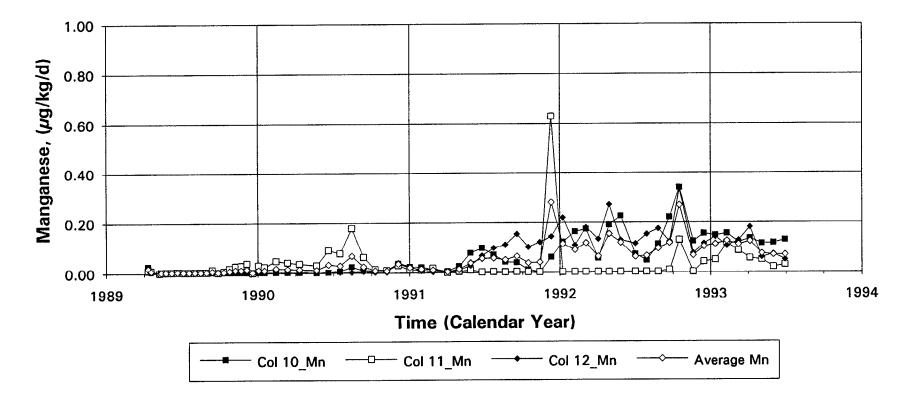
# Homogenized Total Mill Tailings, Fine Limestone Cumulative Magnesium Removal vs. Time

Fig. 83 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), cumulative Mg removal with time.



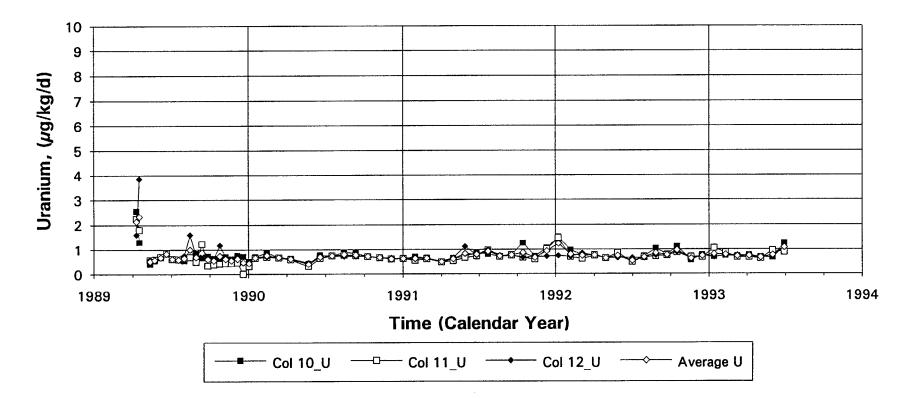
## Homogenized Total Mill Tailings, Fine Limestone Aluminum Loading vs. Time

Fig. 84 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Al loading rate with time.



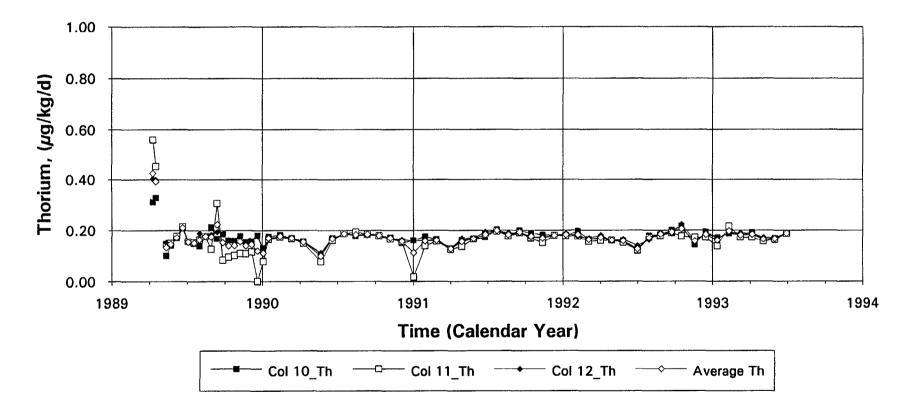
## Homogenized Total Mill Tailings, Fine Limestone Manganese Loading vs. Time

Fig. 85 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Mn loading rate with time.



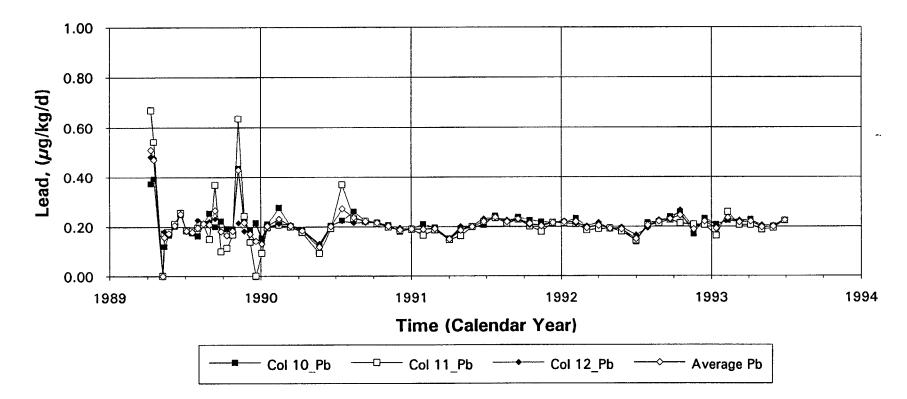
#### Homogenized Total Mill Tailings, Fine Limestone Uranium Loading vs. Time

Fig. 86 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved U loading rate with time.



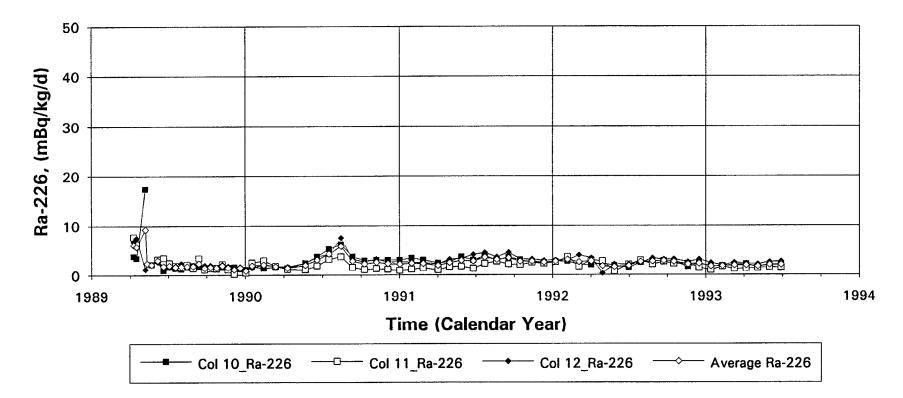
## Homogenized Total Mill Tailings, Fine Limestone Thorium Loading vs. Time

Fig. 87 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Th loading rate with time.



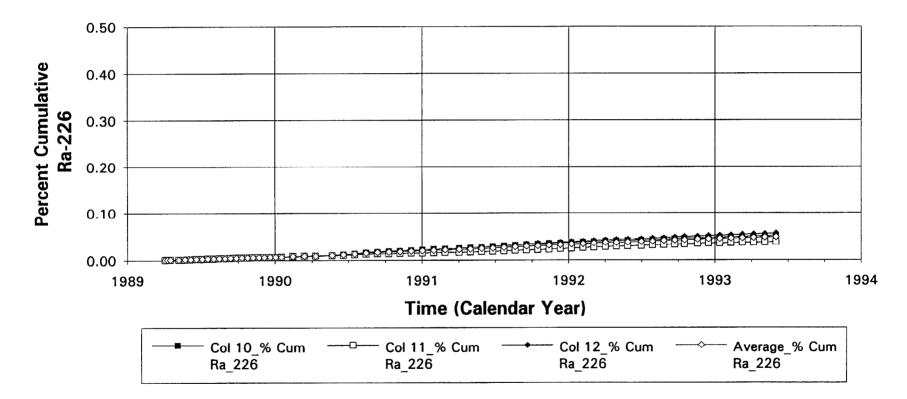
## Homogenized Total Mill Tailings, Fine Limestone Lead Loading vs. Time

Fig. 88 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Pb loading rate with time.



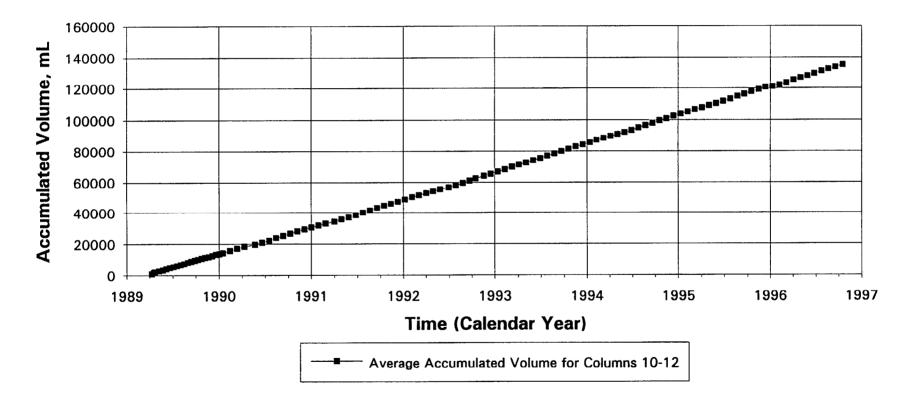
## Homogenized Total Mill Tailings, Fine Limestone Ra-226 Loading vs. Time

Fig. 89 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), variation of dissolved Ra-226 loading rate with time.



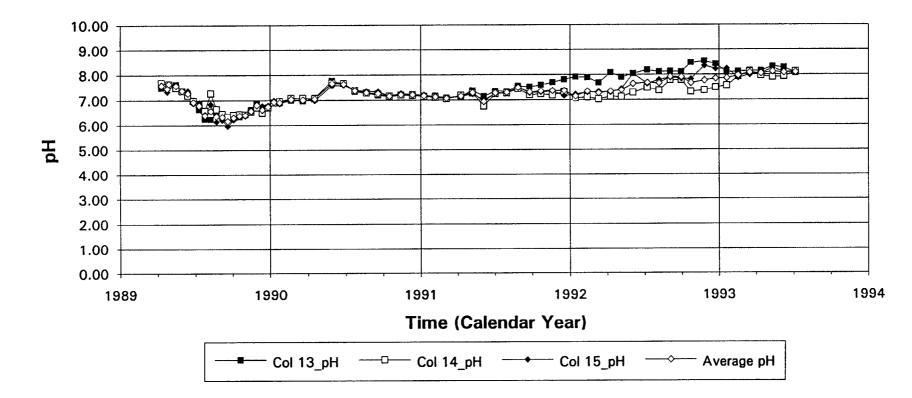
### Homogenized Total Mill Tailings, Fine Limestone Cumulative Ra-226 Removal vs. Time

Fig. 90 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), cumulative Ra-226 removal with time.



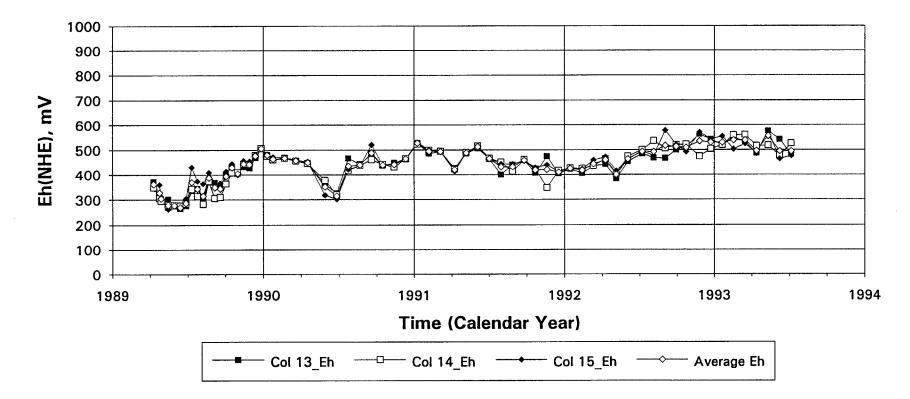
#### Homogenized Total Mill Tailings, Fine Limestone Accumulated Volume vs. Time

Fig. 91 Leaching profiles of total mill tailings mixed with 7.5% (w/w) fine size limestone (screen size -0.84 mm, -20 mesh) and under unsaturated conditions (columns #10-12), average cumulative drainage volume collected with time.



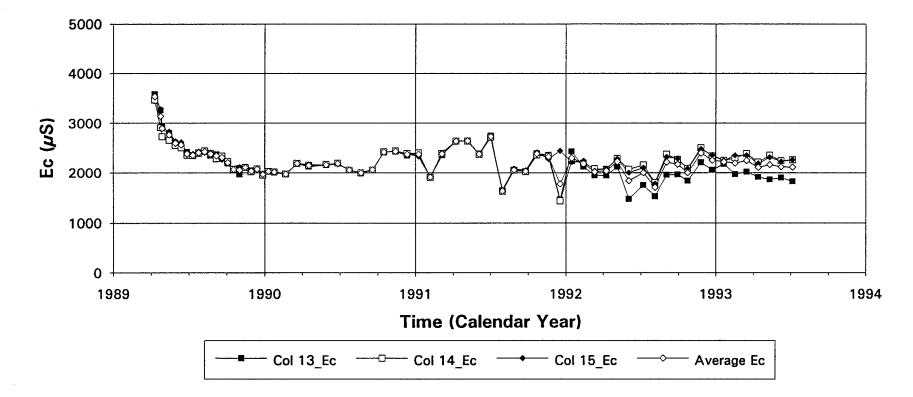
## Homogenized Total Mill Tailings, Wet Ground Limestone pH vs. Time

Fig. 92 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of pH with time.



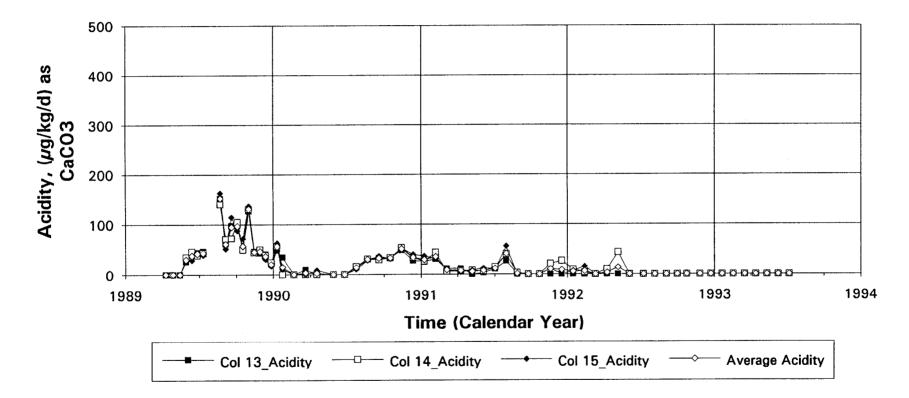
### Homogenized Total Mill Tailings, Wet Ground Limestone Eh(NHE) vs. Time

Fig. 93 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of Eh with time.



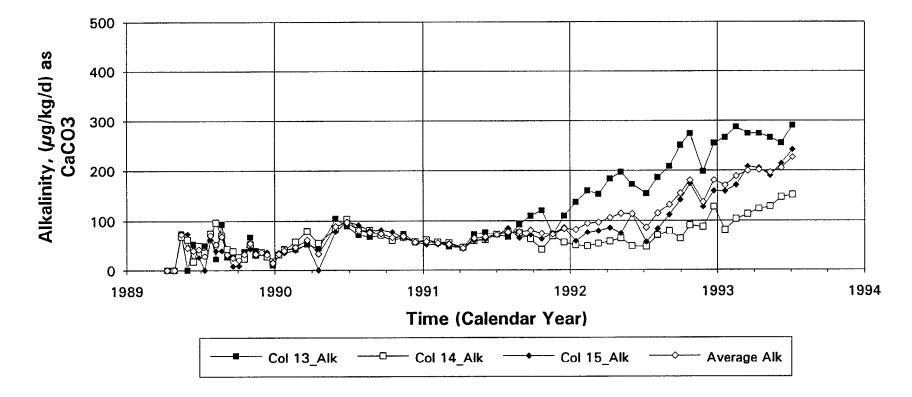
# Homogenized Total Mill Tailings, Wet Ground Limestone Electrical Conductivity vs. Time

Fig. 94 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of Ec with time.



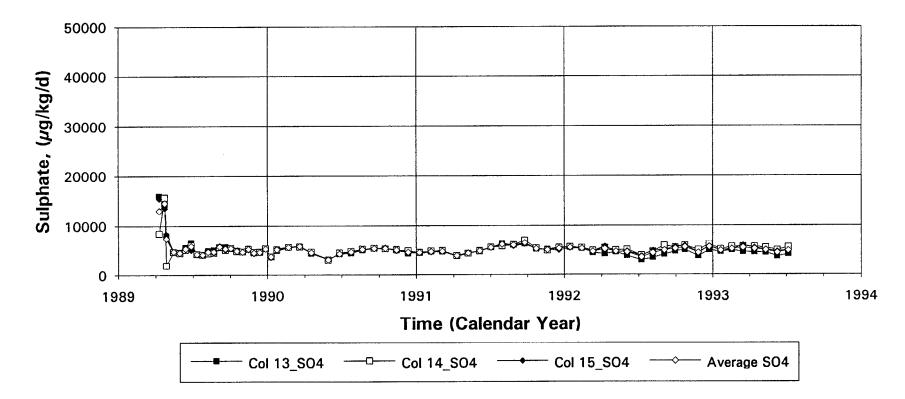
## Homogenized Total Mill Tailings, Wet Ground Limestone Acidity Loading vs. Time

Fig. 95 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of total acidity loading rate with time.



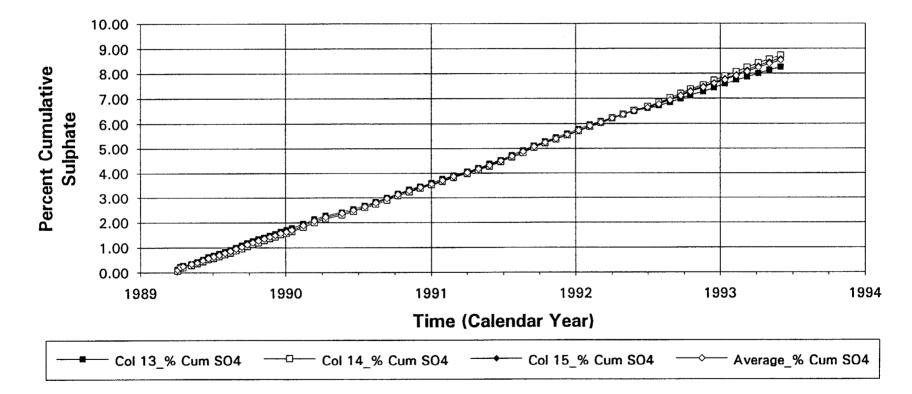
## Homogenized Total Mill Tailings, Wet Ground Limestone Alkalinity Loading vs. Time

Fig. 96 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of total alkalinity loading rate with time.



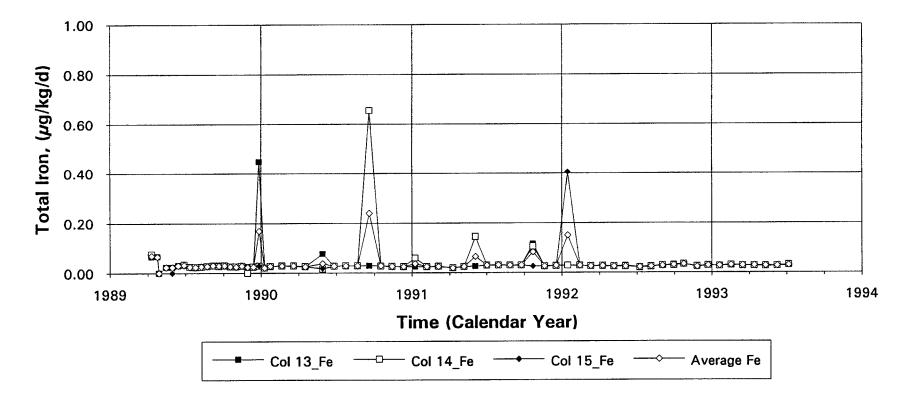
# Homogenized Total Mill Tailings, Wet Ground Limestone Sulphate Loading vs. Time

Fig. 97 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved  $SO_4^{-2}$  loading rate with time.



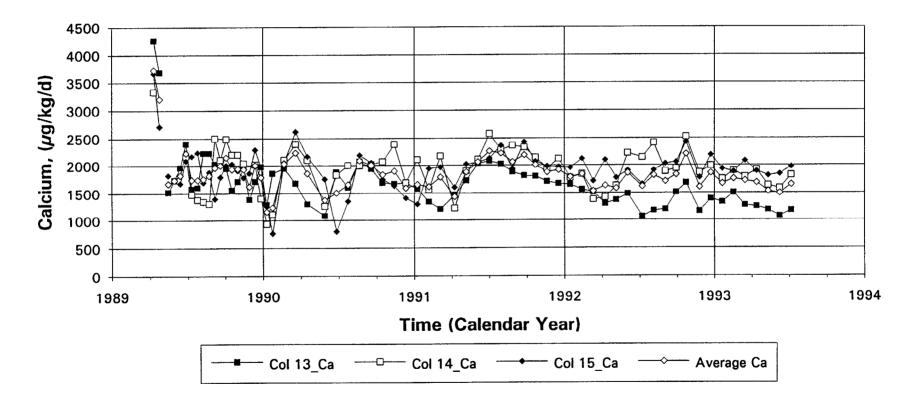
### Homogenized Total Mill Tailings, Wet Ground Limestone Cumulative Sulphate Removal vs. Time

Fig. 98 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



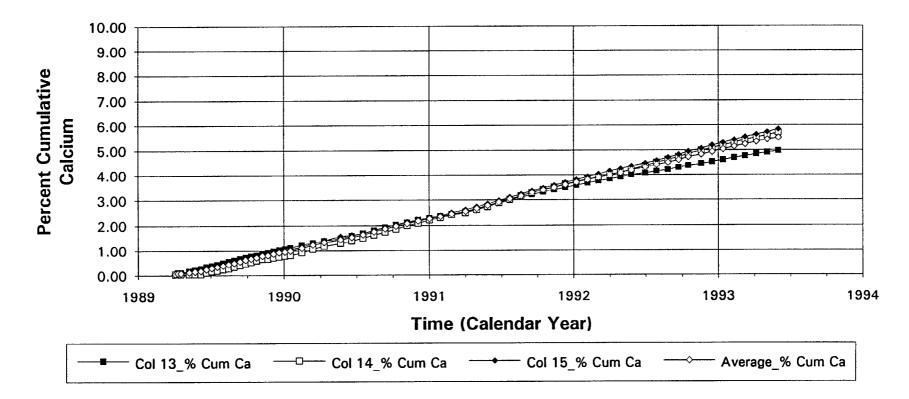
# Homogenized Total Mill Tailings, Wet Ground Limestone Total Iron Loading vs. Time

Fig. 99 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved total Fe loading rate with time.



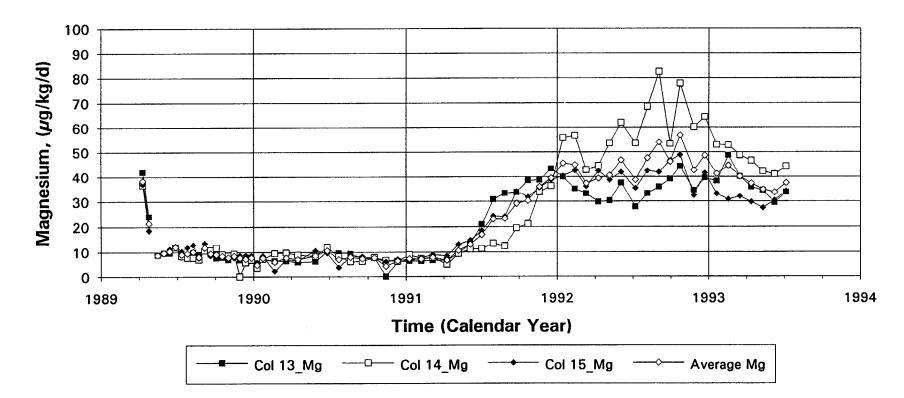
### Homogenized Total Mill Tailings, Wet Ground Limestone Calcium Loading vs. Time

Fig. 100 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Ca loading rate with time.



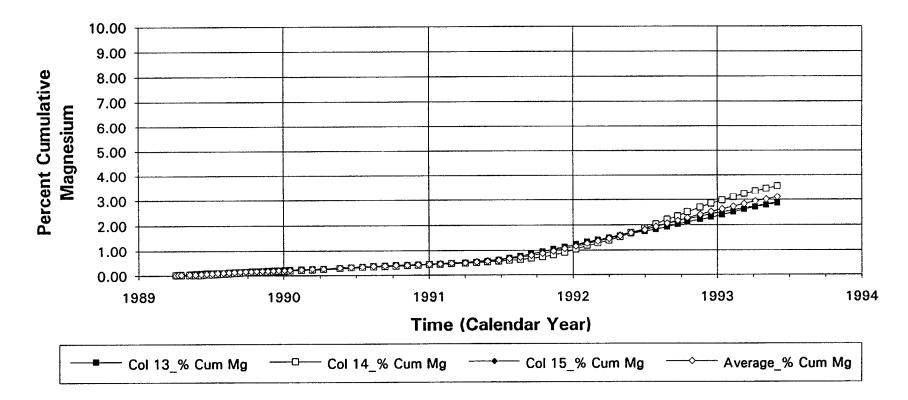
#### Homogenized Total Mill Tailings, Wet Ground Limestone Cumulative Calcium Removal vs. Time

Fig. 101 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), cumulative Ca removal with time.



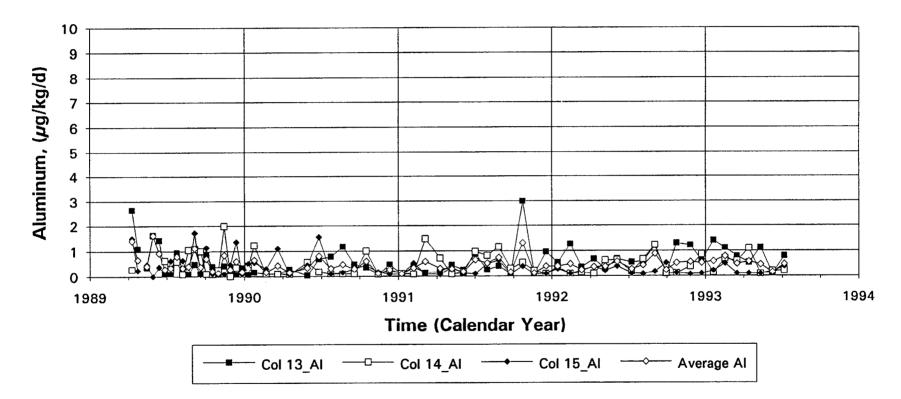
## Homogenized Total Mill Tailings, Wet Ground Limestone Magnesium Loading vs. Time

Fig. 102 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Mg loading rate with time.



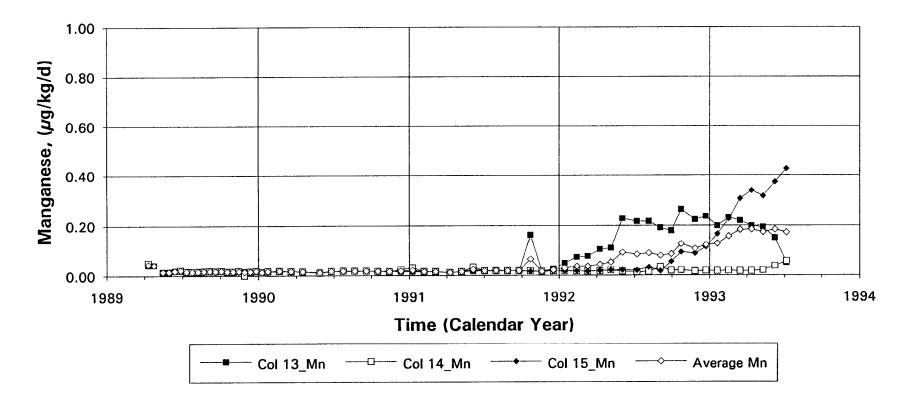
## Homogenized Total Mill Tailings, Wet Ground Limestone Cumulative Magnesium Removal vs. Time

Fig. 103 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), cumulative Mg removal with time.



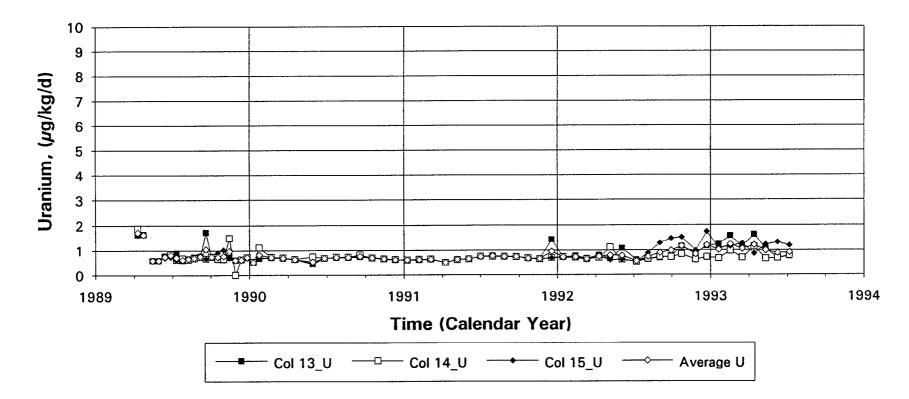
## Homogenized Total Mill Tailings, Wet Ground Limestone Aluminum Loading vs. Time

Fig. 104 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Al loading rate with time.



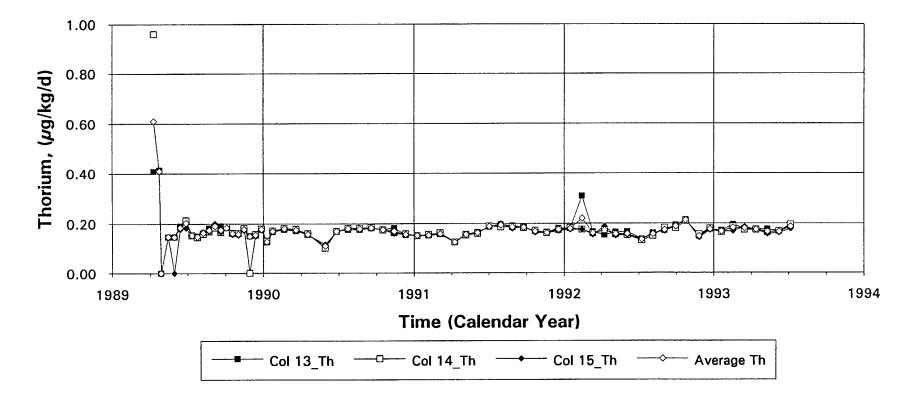
### Homogenized Total Mill Tailings, Wet Ground Limestone Manganese Loading vs. Time

Fig. 105 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Mn loading rate with time.



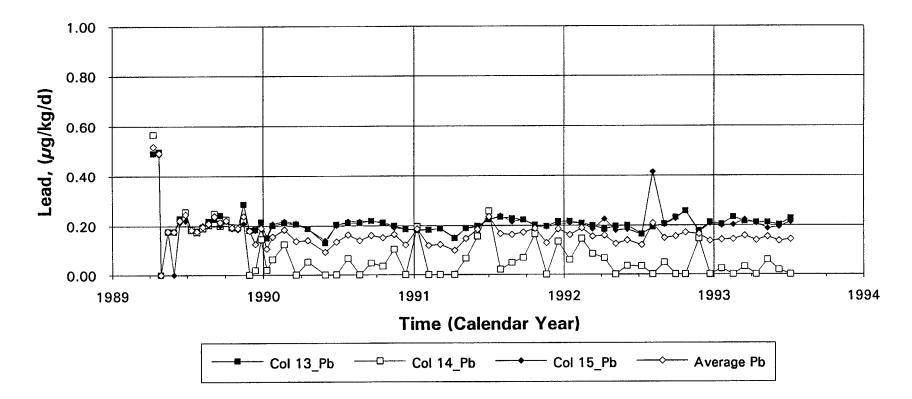
## Homogenized Total Mill Tailings, Wet Ground Limestone Uranium Loading vs. Time

Fig. 106 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved U loading rate with time.



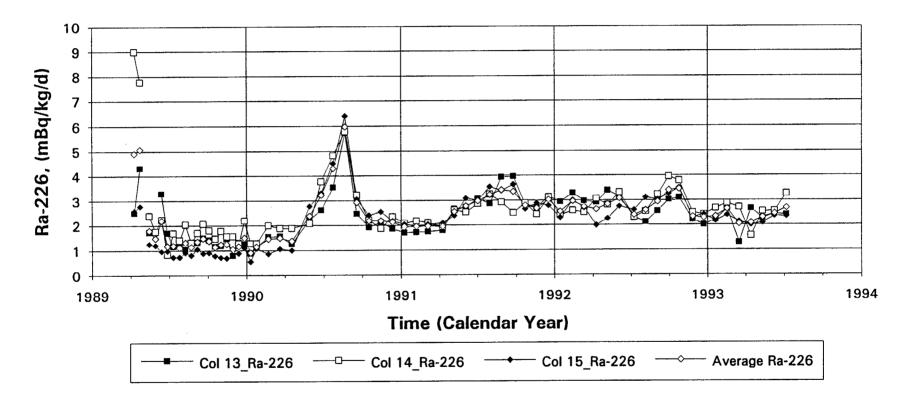
## Homogenized Total Mill Tailings, Wet Ground Limestone Thorium Loading vs. Time

Fig. 107 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Th loading rate with time.



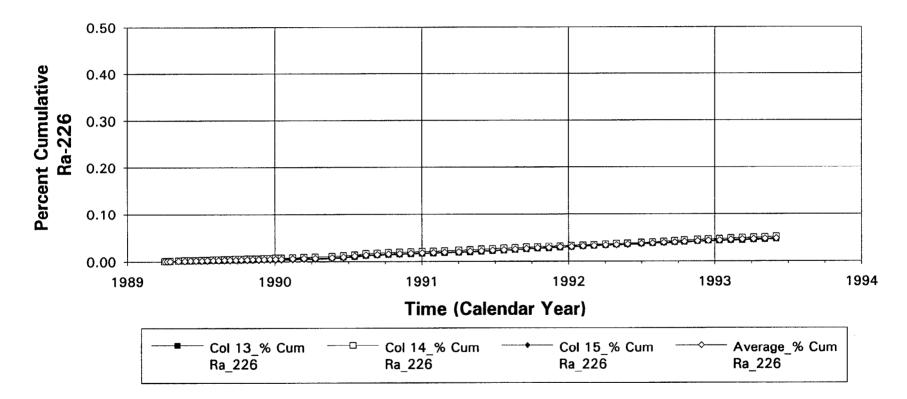
# Homogenized Total Mill Tailings, Wet Ground Limestone Lead Loading vs. Time

Fig. 108 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Pb loading rate with time.



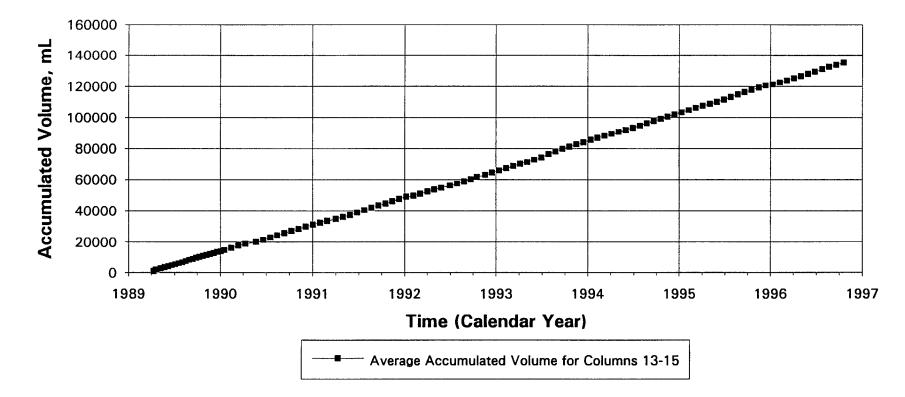
## Homogenized Total Mill Tailings, Wet Ground Limestone Ra-226 Loading vs. Time

Fig. 109 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), variation of dissolved Ra-226 loading rate with time.



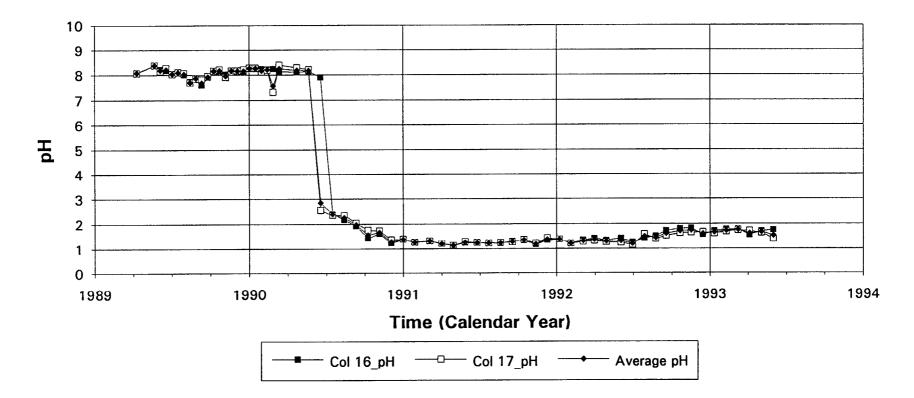
#### Homogenized Total Mill Tailings, Wet Ground Limestone Cumulative Ra-226 Removal vs. Time

Fig. 110 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), cumulative Ra-226 removal with time.



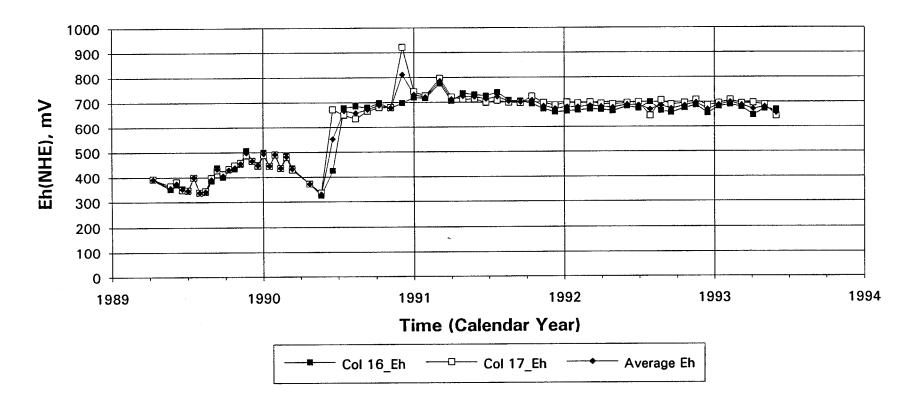
#### Homogenized Total Mill Tailings, Wet Ground Limestone Accumulated Volume vs. Time

Fig. 111 Leaching profiles of total mill tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #13-15), average cumulative drainage volume collected with time.



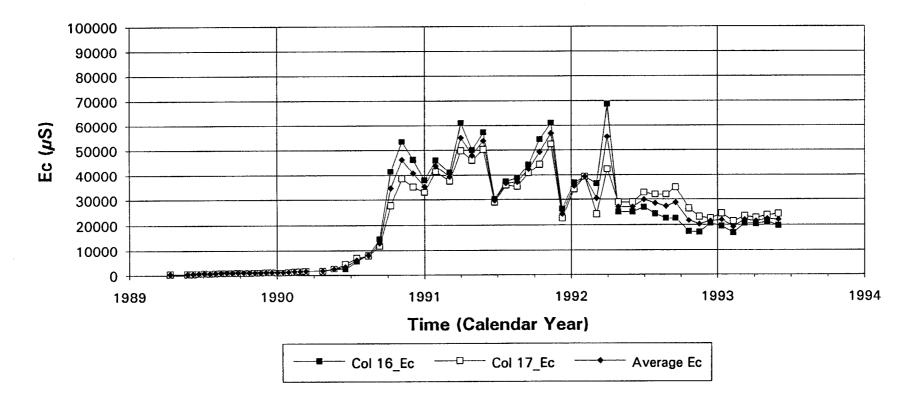
Segregated Coarse Tailings (Control-2) pH vs. Time

Fig. 112 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of pH with time.



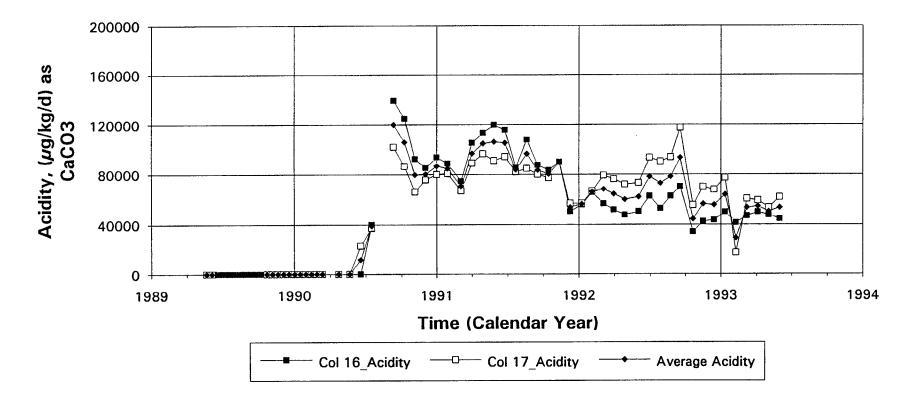
# Segregated Coarse Tailings (Control-2) Eh(NHE) vs. Time

Fig. 113 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of Eh with time.



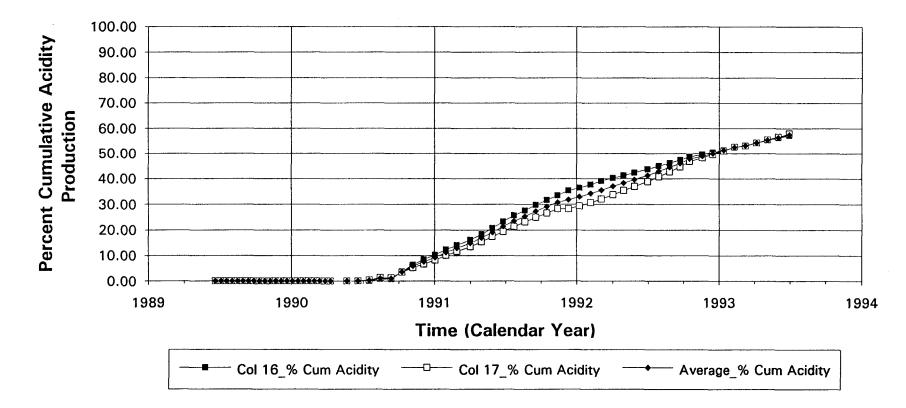
## Segregated Coarse Tailings (Control-2) Electrical Conductivity vs. Time

Fig. 114 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of Ec with time.



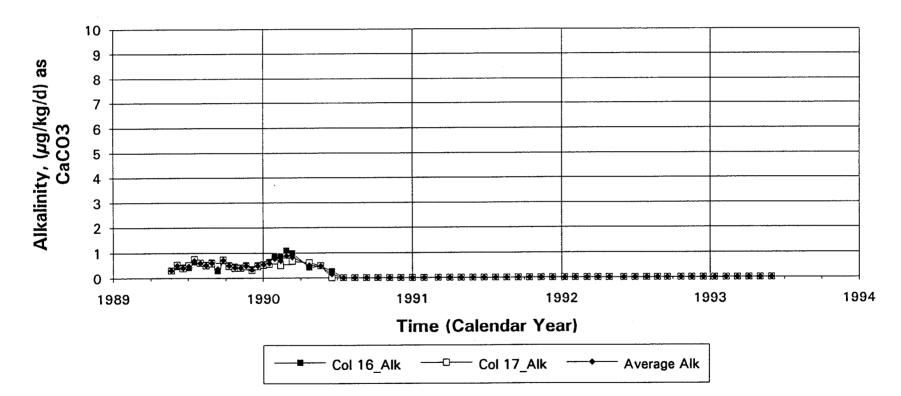
Segregated Coarse Tailings (Control-2) Acidity Loading vs. Time

Fig. 115 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of total acidity loading rate with time.



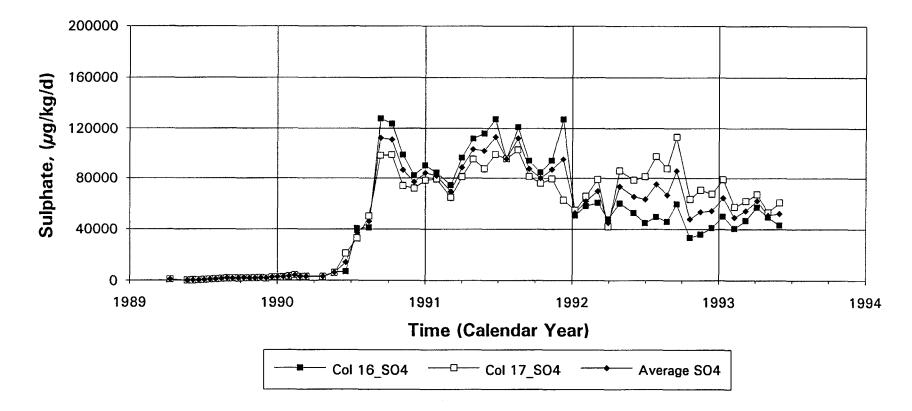
# Segregated Coarse Tailings (Control-2) Cumulative Acidity Production vs. Time

Fig. 116 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative acidity production with time.



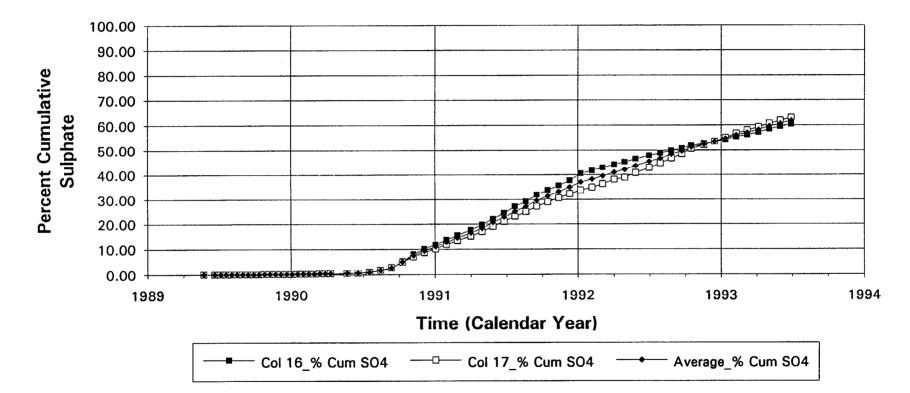
## Segregated Coarse Tailings (Control-2) Alkalinity Loading vs. Time

Fig. 117 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of total alkalinity loading rate with time.



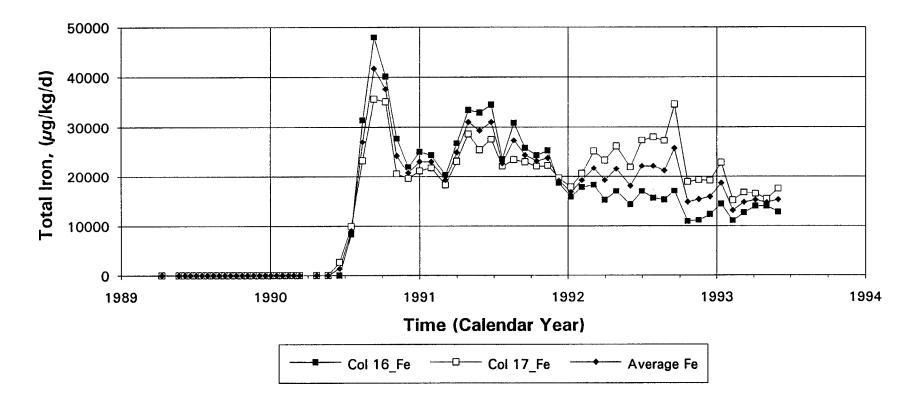
## Segregated Coarse Tailings (Control-2) Sulphate Loading vs. Time

Fig. 118 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved  $SO_4^{-2}$  loading rate with time.



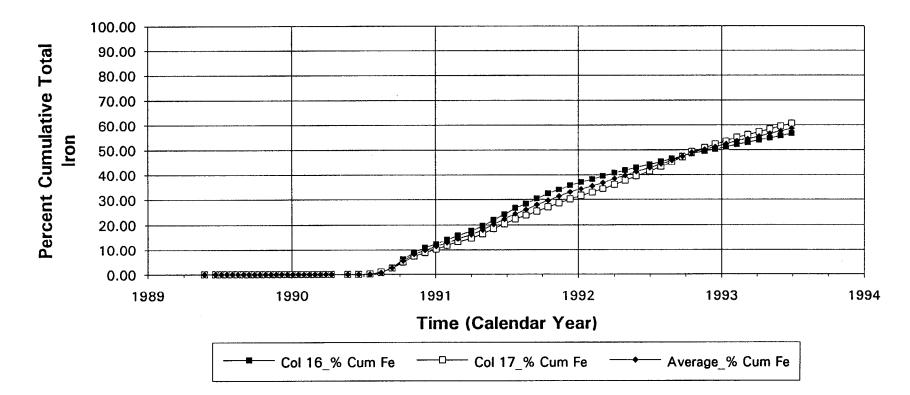
Segregated Coarse Tailings (Control-2) Cumulative Sulphate Removal vs. Time

Fig. 119 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



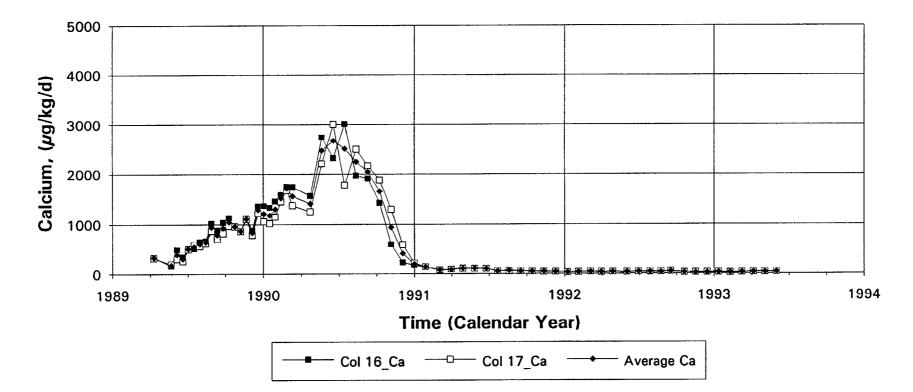
Segregated Coarse Tailings (Control-2) Total Iron Loading vs. Time

Fig. 120 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved total Fe loading rate with time.



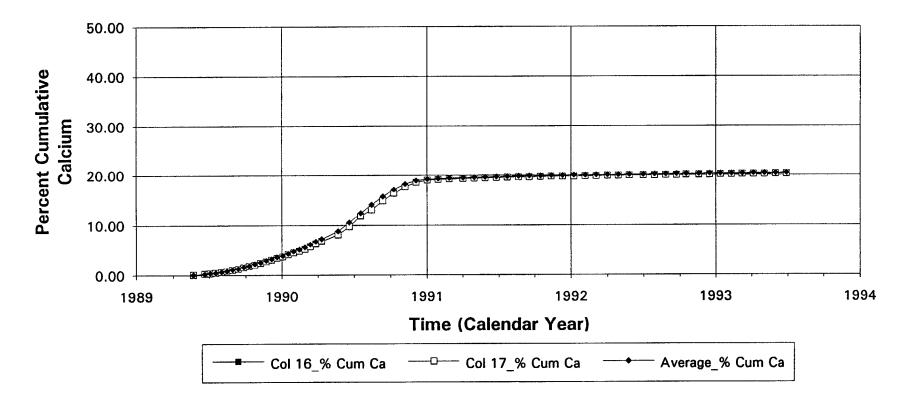
Segregated Coarse Tailings (Control-2) Cumulative Total Iron Removal vs. Time

Fig. 121 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative total Fe removal with time.



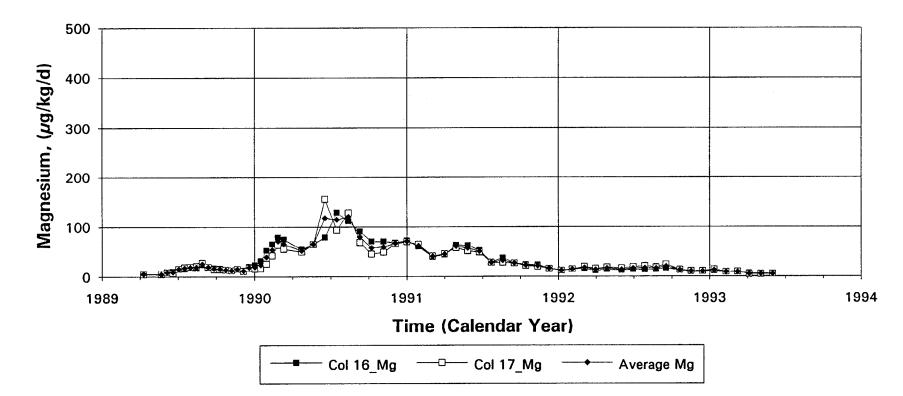
#### Segregated Coarse Tailings (Control-2) Calcium Loading vs. Time

Fig. 122 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Ca loading rate with time.



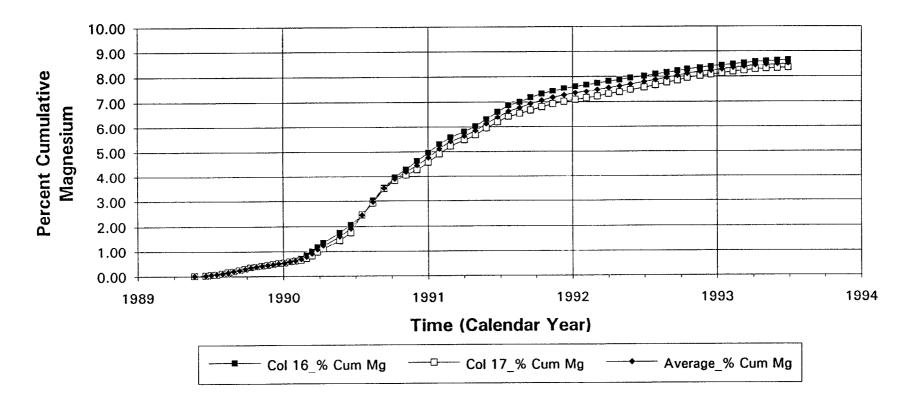
## Segregated Coarse Tailings (Control-2) Cumulative Calcium Removal vs. Time

Fig. 123 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Ca removal with time.



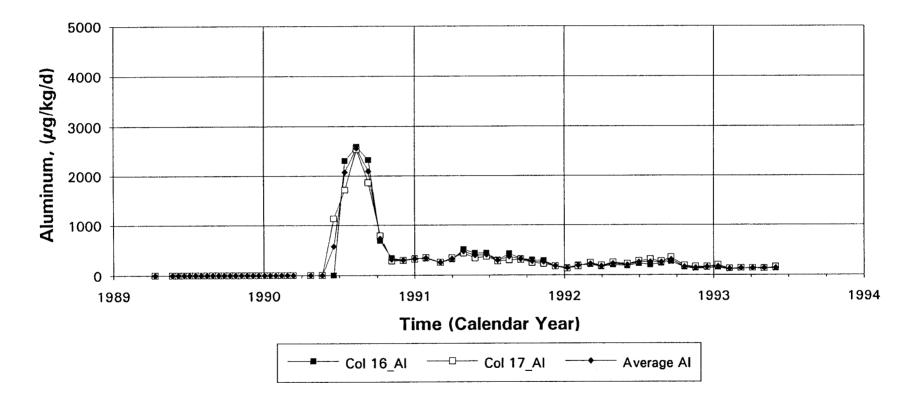
Segregated Coarse Tailings (Control-2) Magnesium Loading vs. Time

Fig. 124 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Mg loading rate with time.



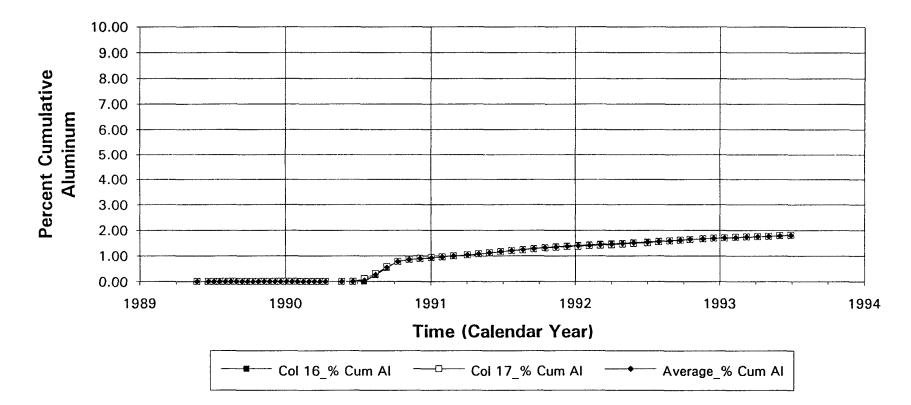
## Segregated Coarse Tailings (Control-2) Cumulative Magnesium Removal vs. Time

Fig. 125 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Mg removal with time.



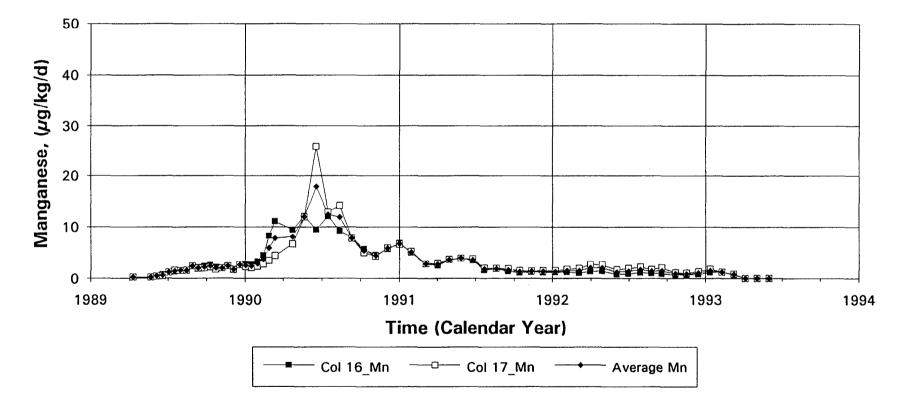
Segregated Coarse Tailings (Control-2) Aluminum Loading vs. Time

Fig. 126 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Al loading rate with time.



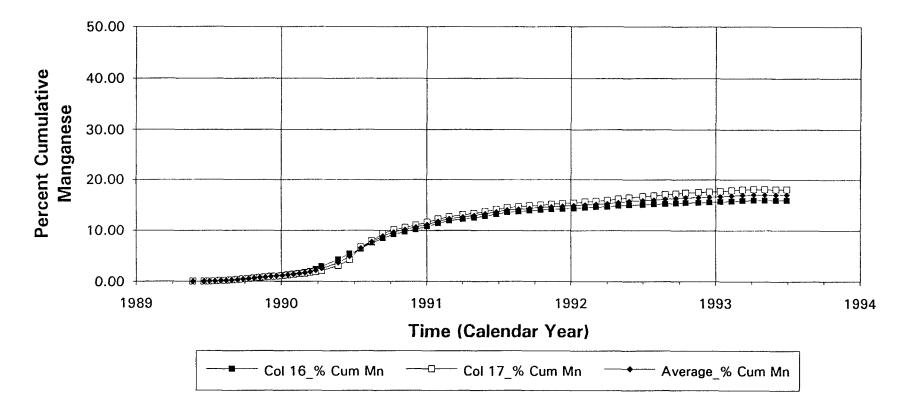
## Segregated Coarse Tailings (Control-2) Cumulative Aluminum Removal vs. Time

Fig. 127 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Al removal with time.



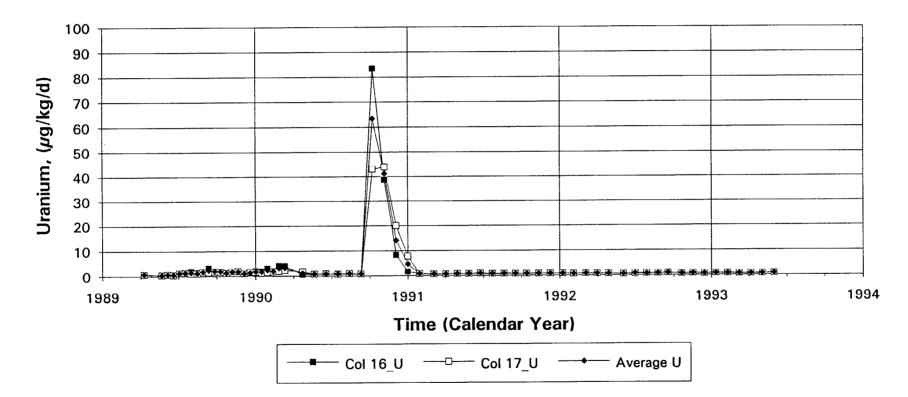
## Segregated Coarse Tailings (Control-2) Manganese Loading vs. Time

Fig. 128 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Mn loading rate with time.



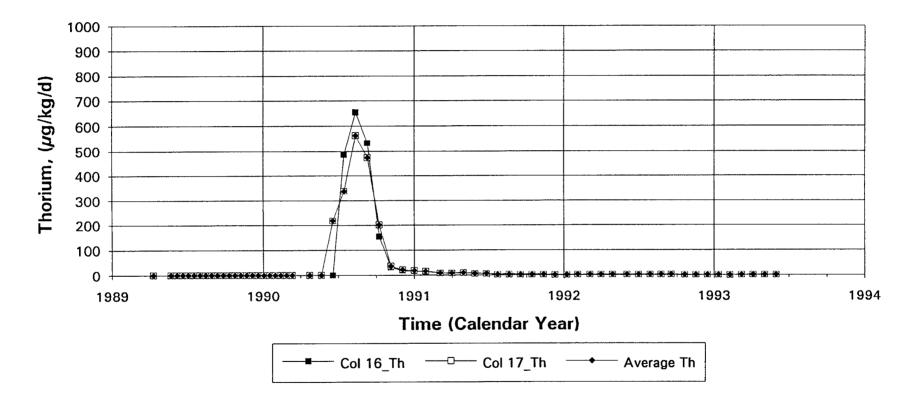
Segregated Coarse Tailings (Control-2) Cumulative Manganese Removal vs. Time

Fig. 129 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Mn removal with time.



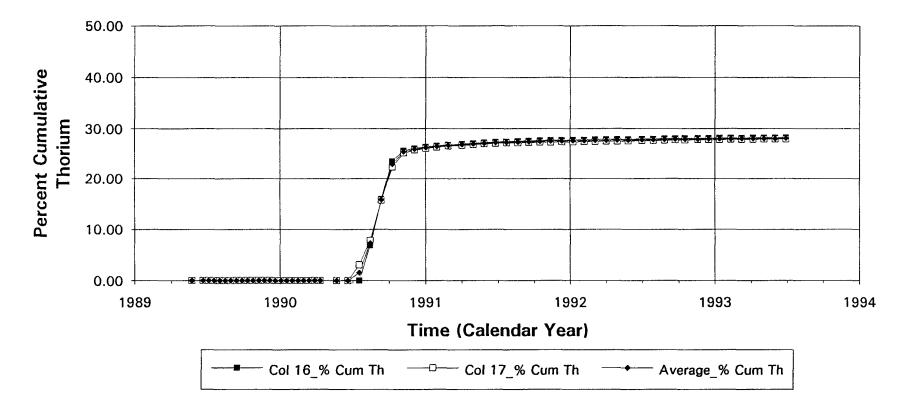
#### Segregated Coarse Tailings (Control-2) Uranium Loading vs. Time

Fig. 130 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved U loading rate with time.



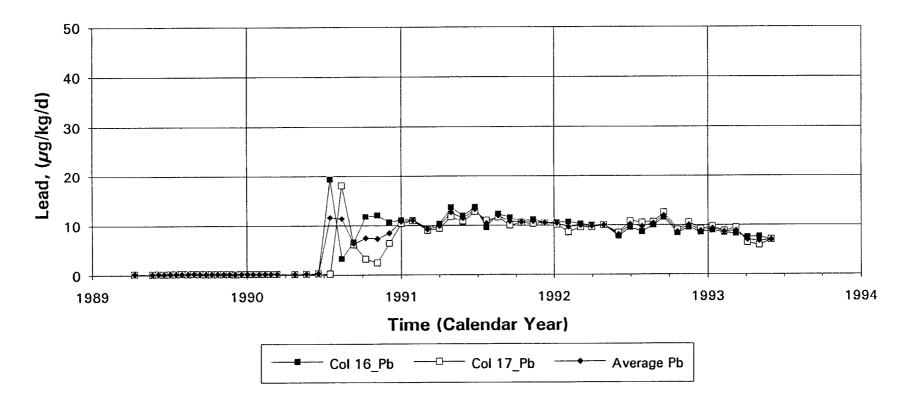
## Segregated Coarse Tailings (Control-2) Thorium Loading vs. Time

Fig. 131 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Th loading rate with time.



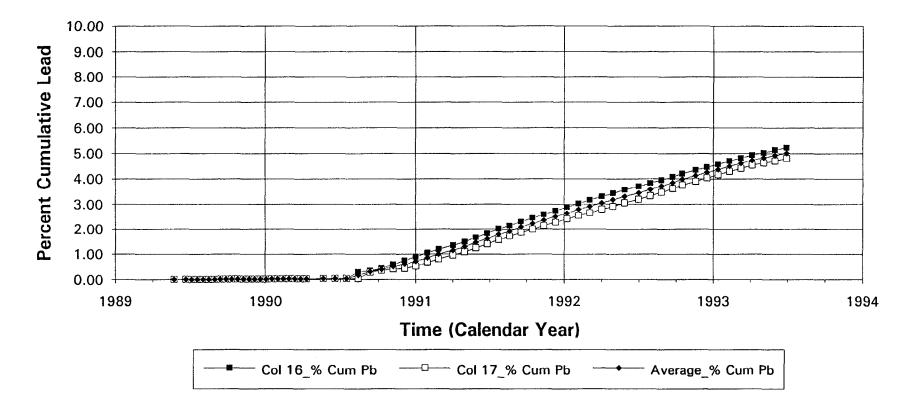
## Segregated Coarse Tailings (Control-2) Cumulative Thorium Removal vs. Time

Fig. 132 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Th removal with time.



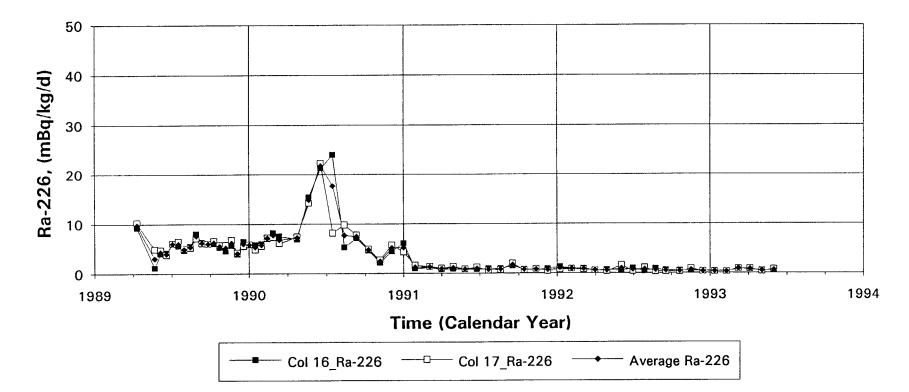
Segregated Coarse Tailings (Control-2) Lead Loading vs. Time

Fig. 133 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Pb loading rate with time.



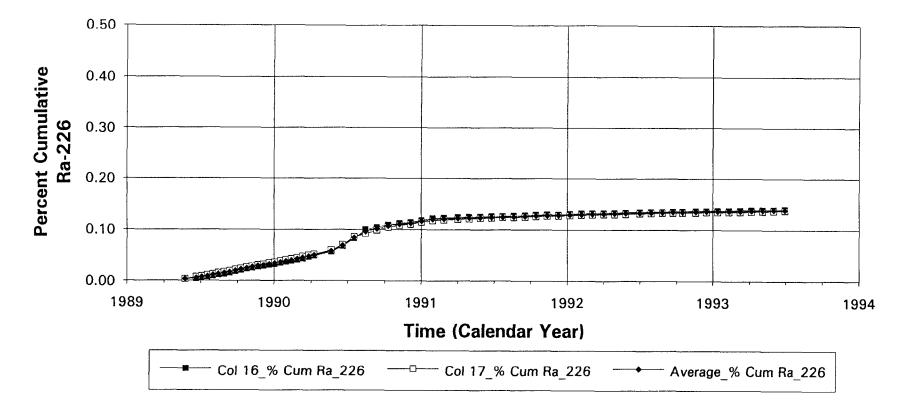
#### Segregated Coarse Tailings (Control-2) Cumulative Lead Removal vs. Time

Fig. 134 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Pb removal with time.



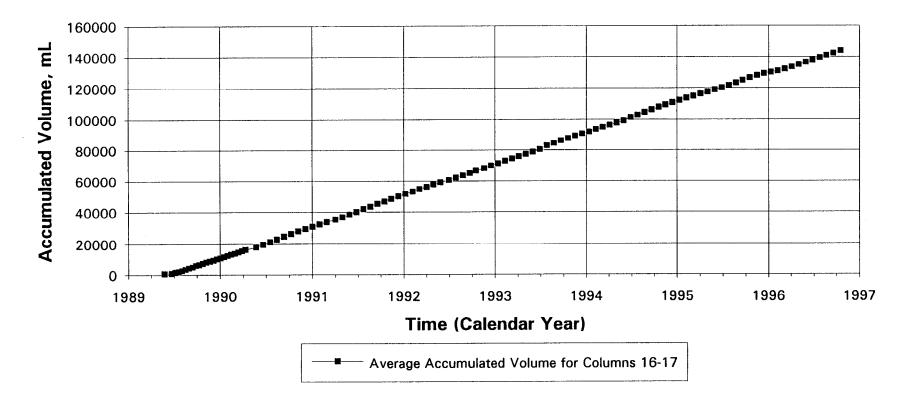
Segregated Coarse Tailings (Control-2) Ra-226 Loading vs. Time

Fig. 135 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), variation of dissolved Ra-226 loading rate with time.



Segregated Coarse Tailings (Control-2) Cumulative Ra-226 Removal vs. Time

Fig. 136 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), cumulative Ra-226 removal with time.



#### Segregated Coarse Tailings (Control-2) Accumulated Volume vs. Time

Fig. 137 Leaching profiles of coarse tailings without additional limestone under unsaturated conditions (Control - 2, columns #16 and 17), average cumulative drainage volume collected with time.

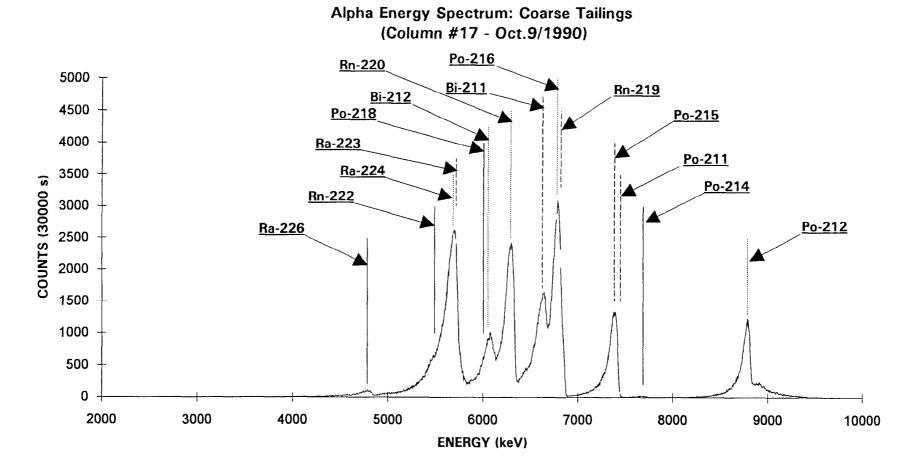
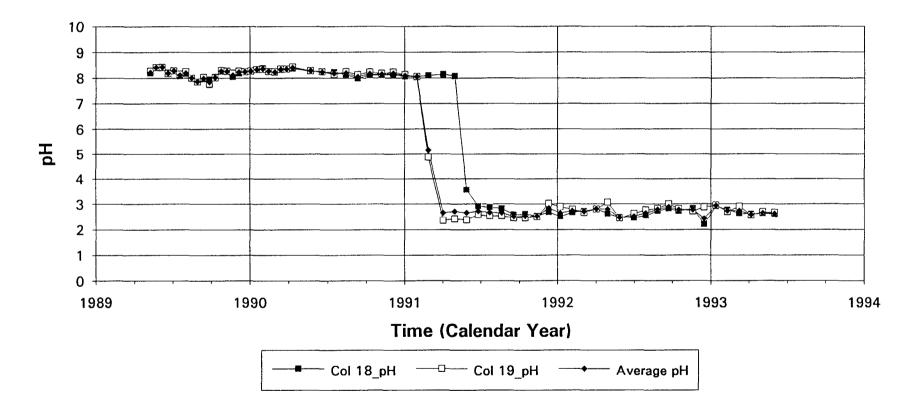
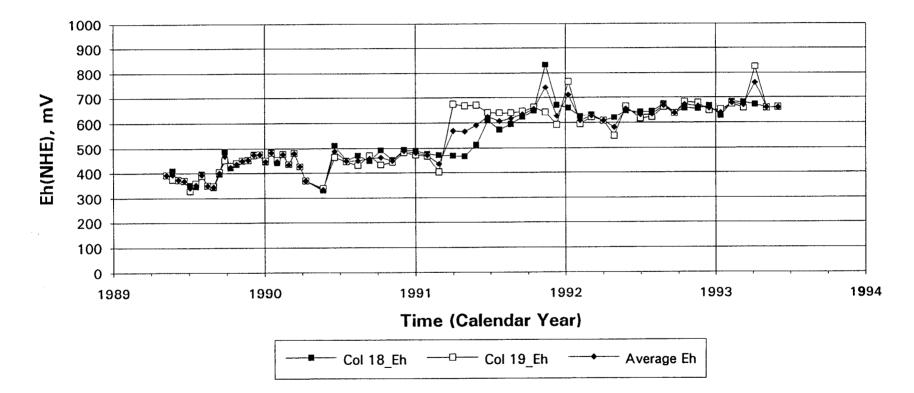


Fig. 138 Alpha energy spectrum obtained for coarse tailings (column #17) during peak acidic drainage period (Oct. 3, 1990).



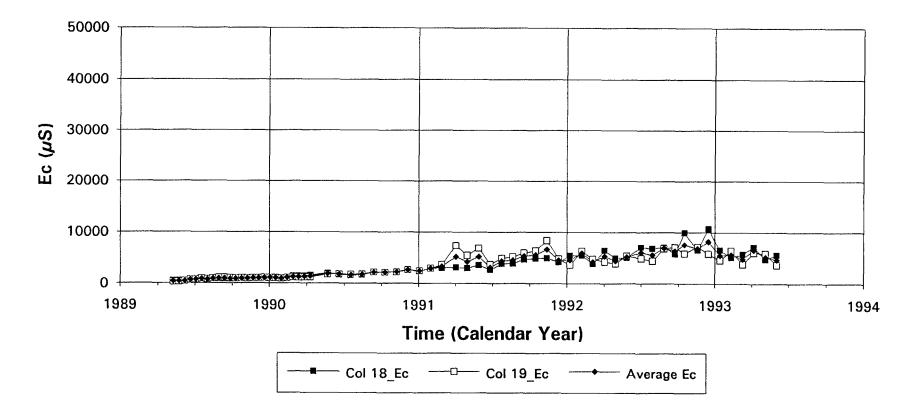
Segregated Coarse Tailings, Coarse Limestone pH vs. Time

Fig. 139 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of pH with time.



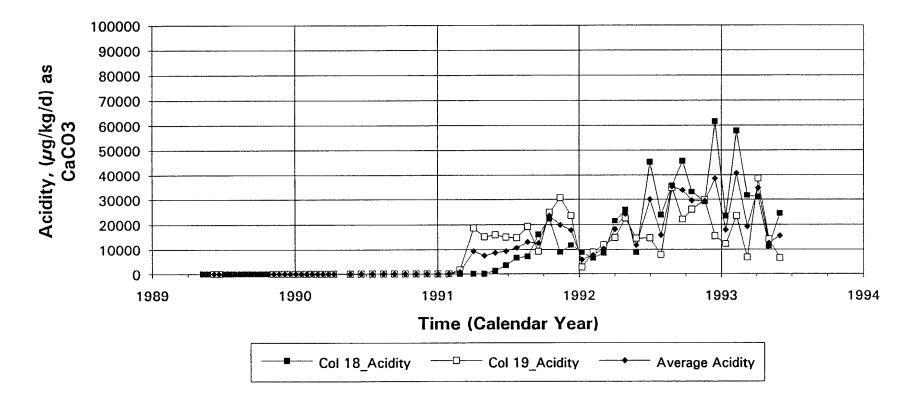
# Segregated Coarse Tailings, Coarse Limestone Eh(NHE) vs. Time

Fig. 140 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of Eh with time.



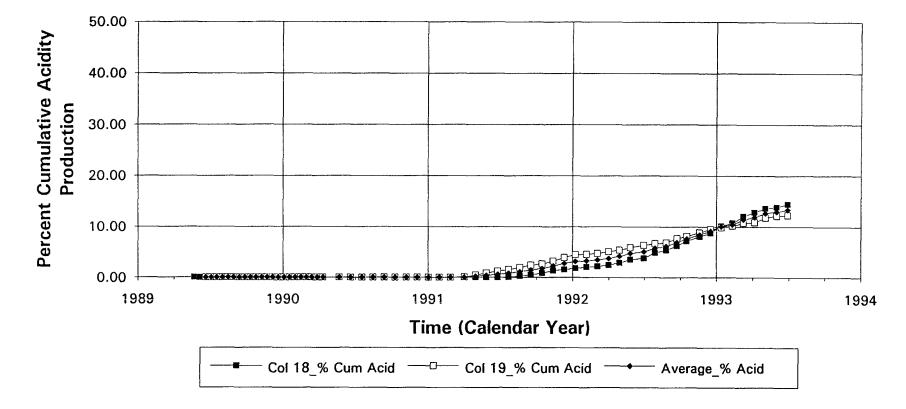
# Segregated Coarse Tailings, Coarse Limestone Electrical Conductivity vs. Time

Fig. 141 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of Ec with time.



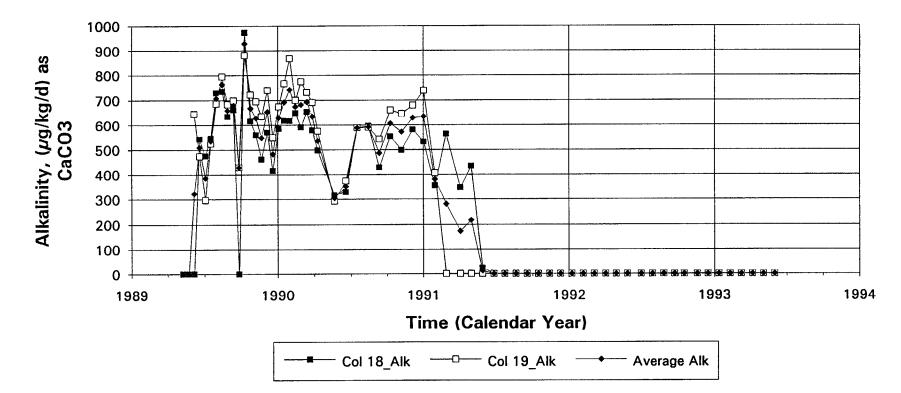
## Segregated Coarse Tailings, Coarse Limestone Acidity Loading vs. Time

Fig. 142 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of total acidity loading rate with time.



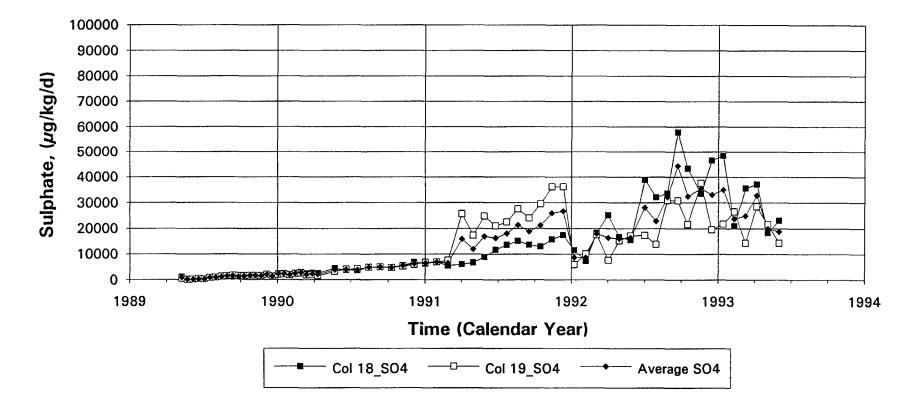
## Segregated Coarse Tailings, Coarse Limestone Cumulative Acidity Production vs. Time

Fig. 143 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative acidity production with time.



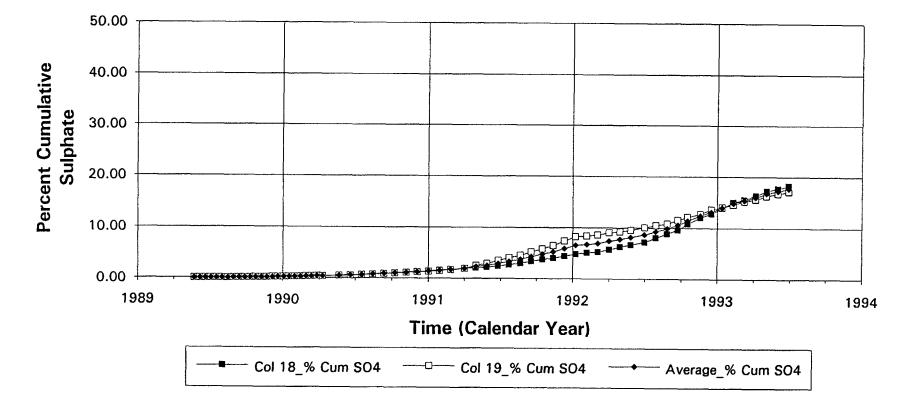
## Segregated Coarse Tailings, Coarse Limestone Alkalinity Loading vs. Time

Fig. 144 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of total alkalinity loading rate with time.



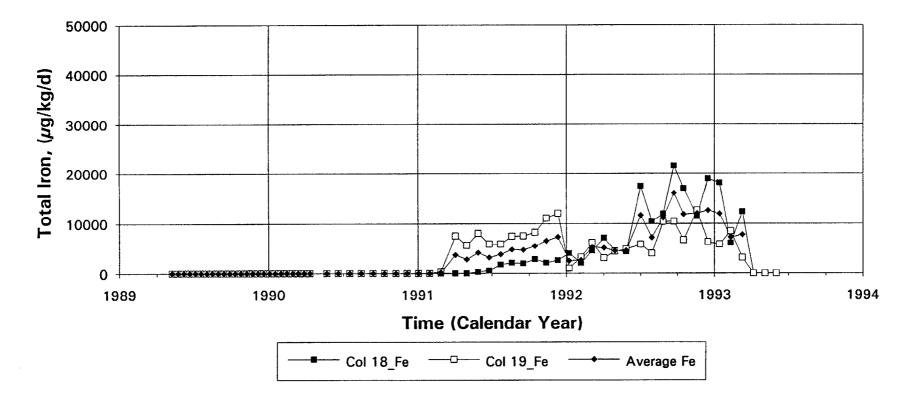
# Segregated Coarse Tailings, Coarse Limestone Sulphate Loading vs. Time

Fig. 145 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved  $SO_4^{-2}$  loading rate with time.



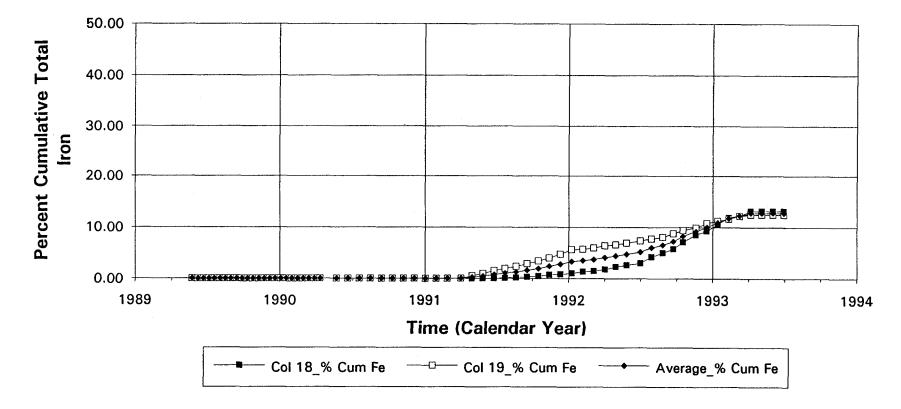
# Segregated Coarse Tailings, Coarse Limestone Cumulative Sulphate Removal vs. Time

Fig. 146 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



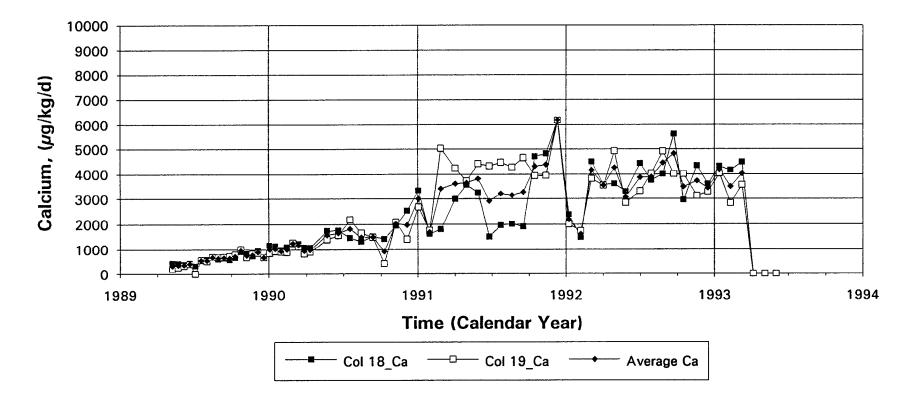
Segregated Coarse Tailings, Coarse Limestone Total Iron Loading vs. Time

Fig. 147 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved total Fe loading rate with time.



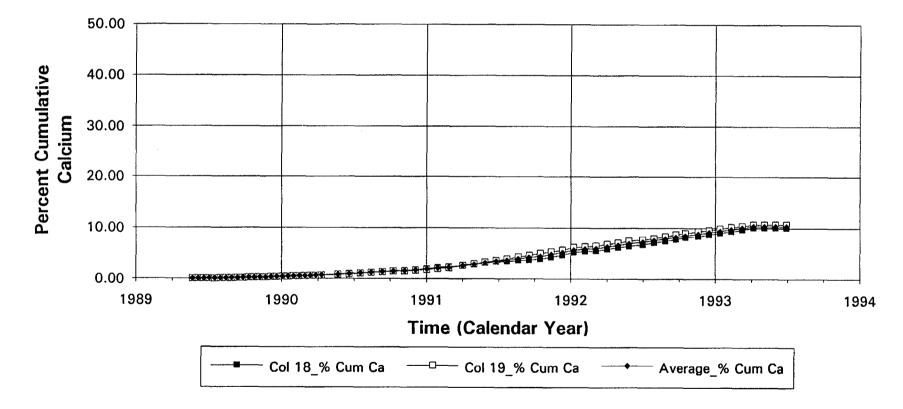
# Segregated Coarse Tailings, Coarse Limestone Cumulative Total Iron Removal vs. Time

Fig. 148 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative total Fe removal with time.



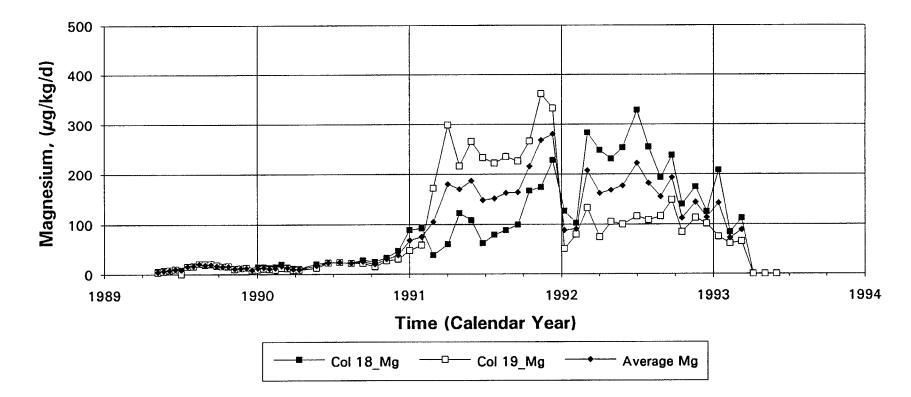
## Segregated Coarse Tailings, Coarse Limestone Calcium Loading vs. Time

Fig. 149 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Ca loading rate with time.



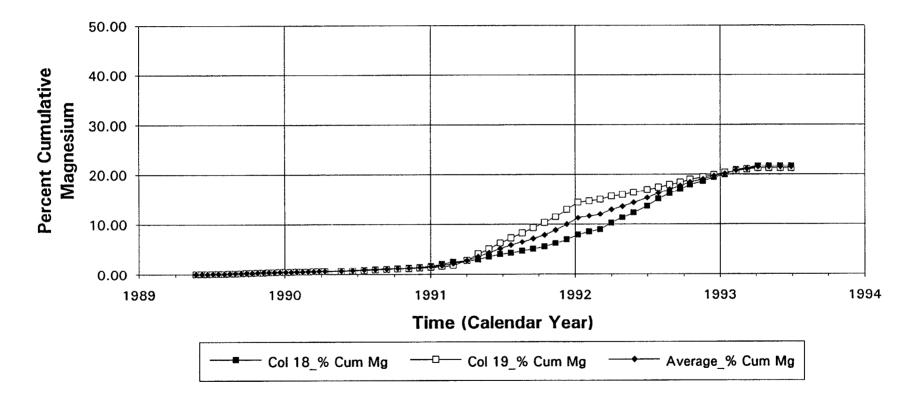
## Segregated Coarse Tailings, Coarse Limestone Cumulative Calcium Removal vs. Time

Fig. 150 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Ca removal with time.



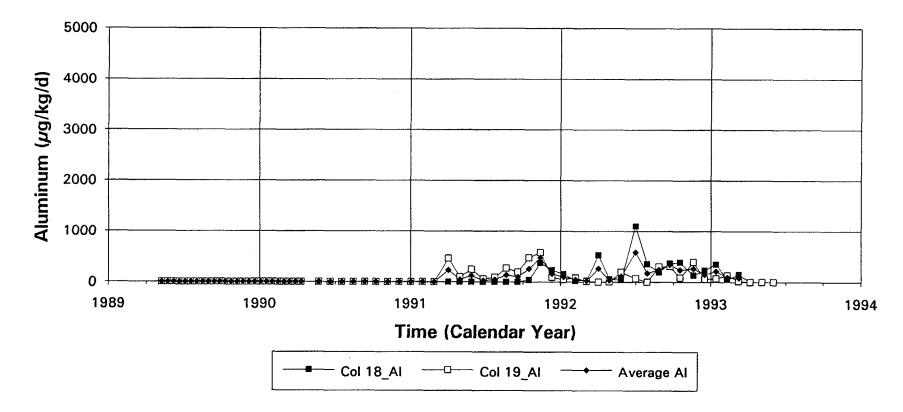
#### Segregated Coarse Tailings, Coarse Limestone Magnesium Loading vs. Time

Fig. 151 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Mg loading rate with time.



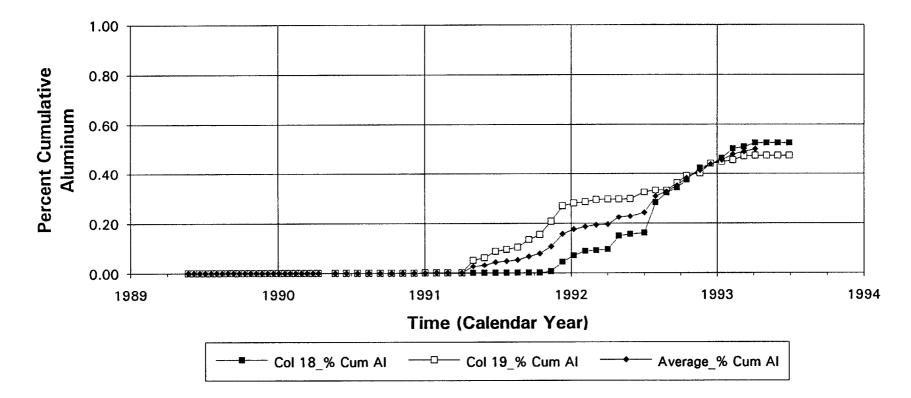
#### Segregated Coarse Tailings, Coarse Limestone Cumulative Magnesium Removal vs. Time

Fig. 152 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Mg removal with time.



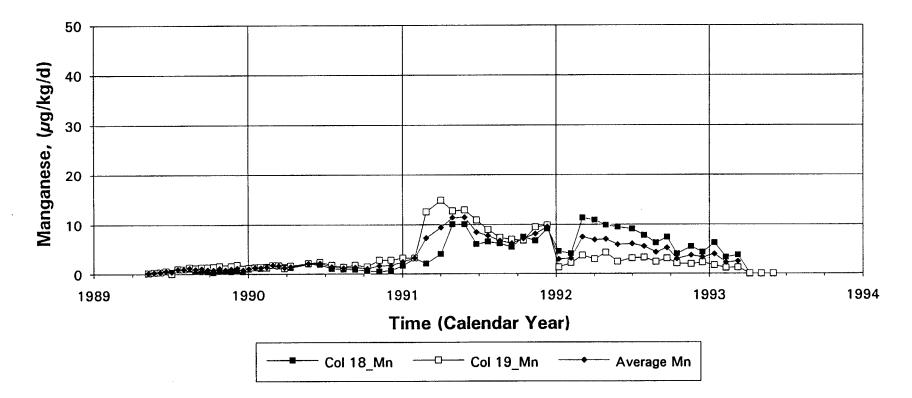
# Segregated Coarse Tailings, Coarse Limestone Aluminum Loading vs. Time

Fig. 153 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Al loading rate with time.



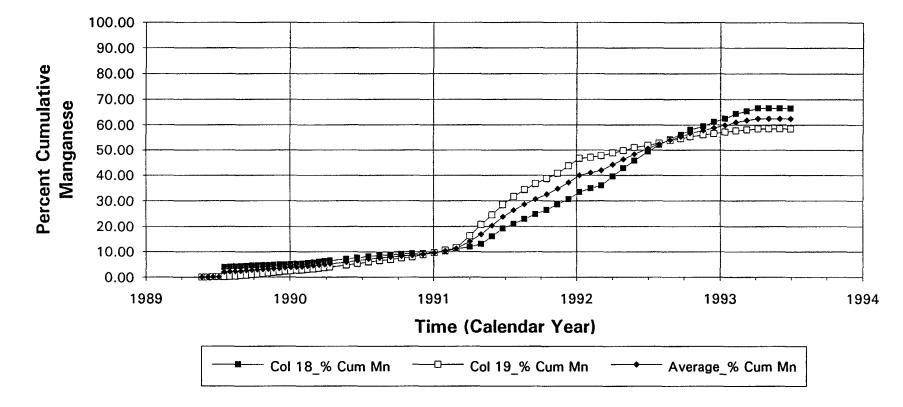
#### Segregated Coarse Tailings, Coarse Limestone Cumulative Aluminum Removal vs. Time

Fig. 154 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Al removal with time.



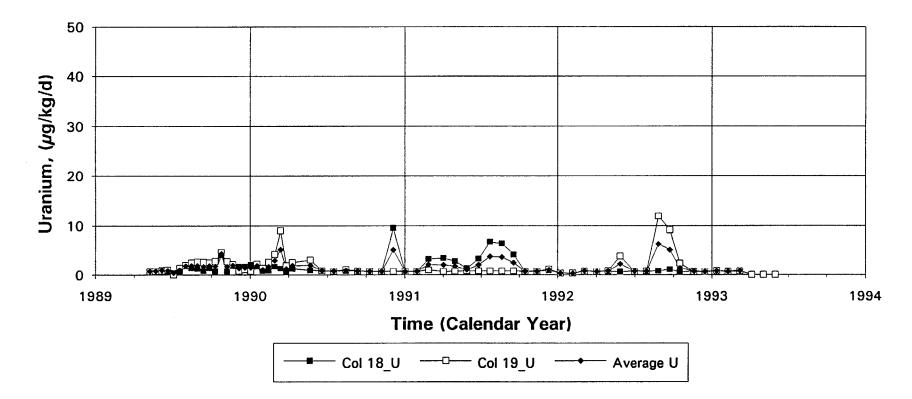
## Segregated Coarse Tailings, Coarse Limestone Manganese Loading vs. Time

Fig. 155 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Mn loading rate with time.



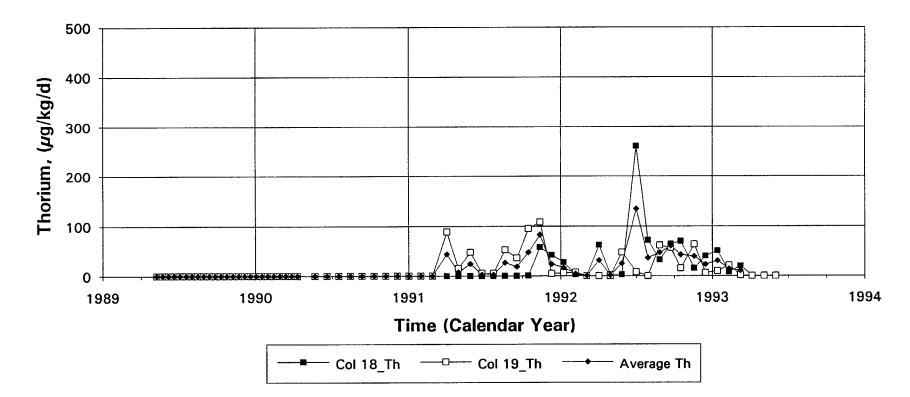
# Segregated Coarse Tailings, Coarse Limestone Cumulative Manganese Removal vs. Time

Fig. 156 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Mn removal with time.



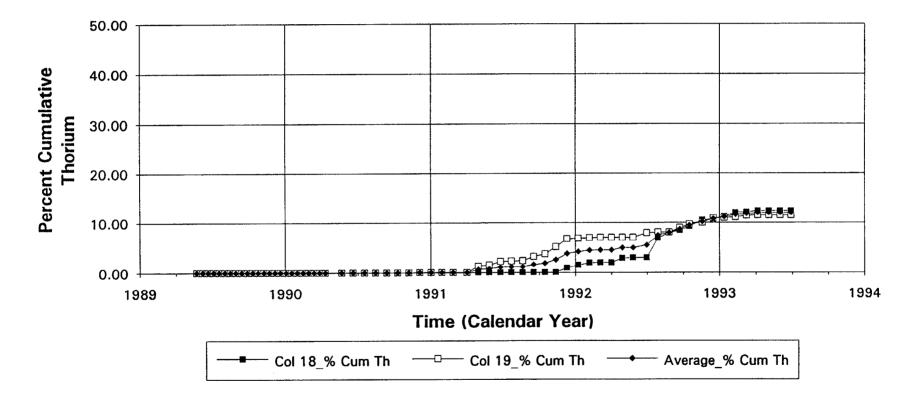
## Segregated Coarse Tailings, Coarse Limestone Uranium Loading vs. Time

Fig. 157 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved U loading rate with time.



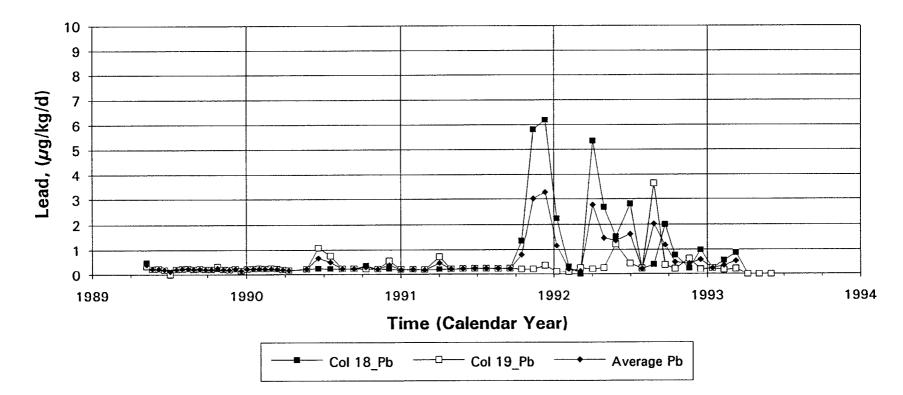
## Segregated Coarse Tailings, Coarse Limestone Thorium Loading vs. Time

Fig. 158 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Th loading rate with time.



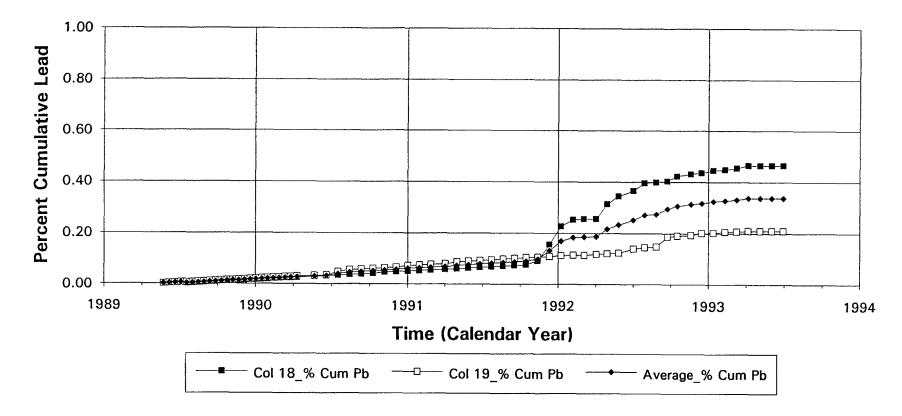
## Segregated Coarse Tailings, Coarse Limestone Cumulative Thorium Removal vs. Time

Fig. 159 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Th removal with time.



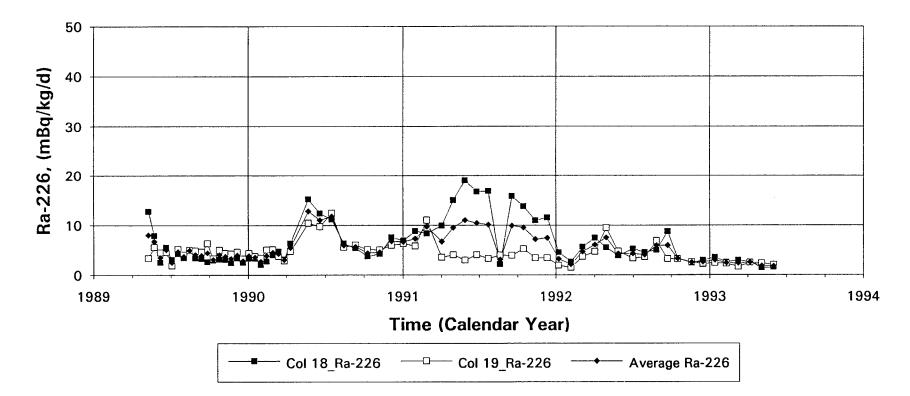
# Segregated Coarse Tailings, Coarse Limestone Lead Loading vs. Time

Fig. 160 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Pb loading rate with time.



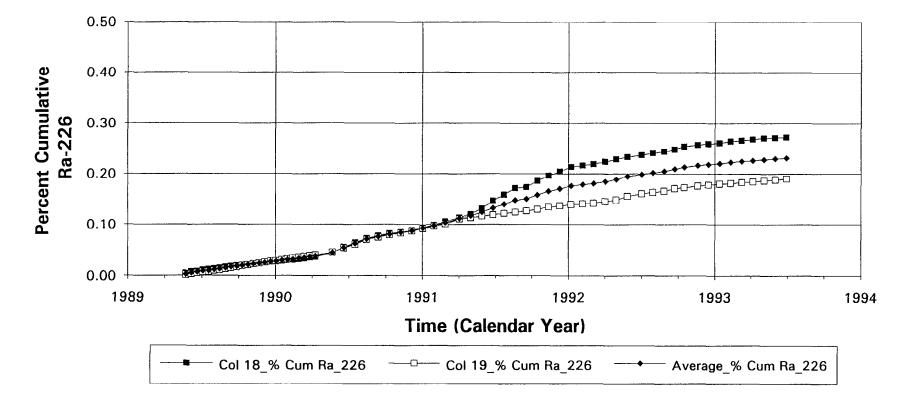
# Segregated Coarse Tailings, Coarse Limestone Cumulative Lead Removal vs. Time

Fig. 161 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Pb removal with time.



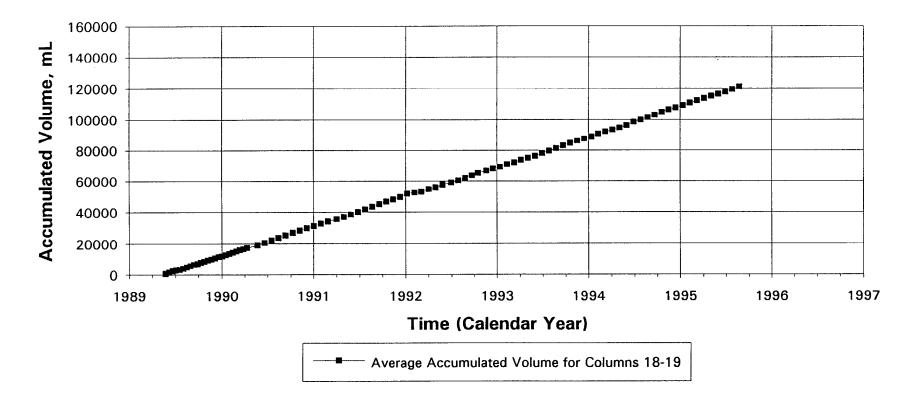
Segregated Coarse Tailings, Coarse Limestone Ra-226 Loading vs. Time

Fig. 162 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), variation of dissolved Ra-226 loading rate with time.



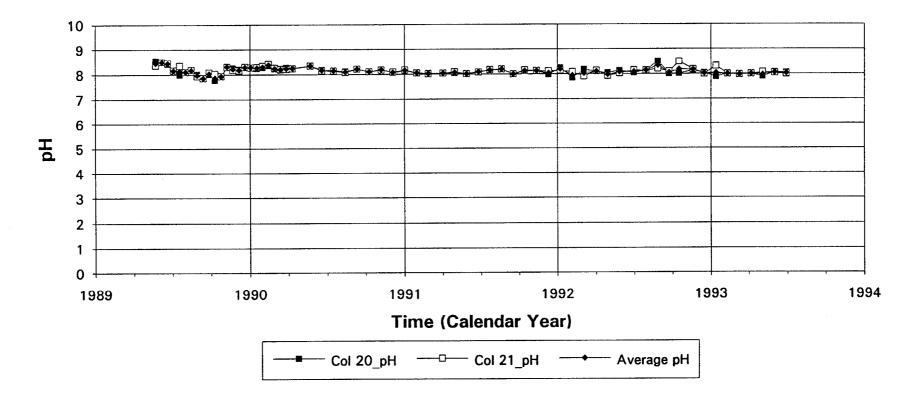
# Segregated Coarse Tailings, Coarse Limestone Cumulative Ra-226 Removal vs. Time

Fig. 163 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), cumulative Ra-226 removal with time.



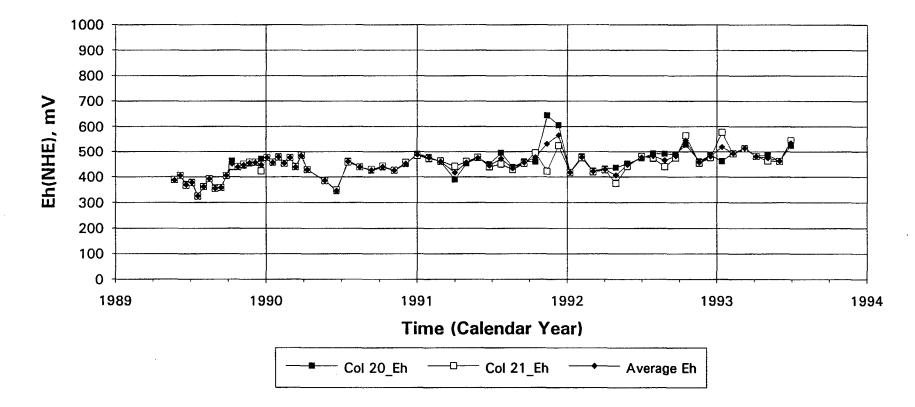
#### Segregated Coarse Tailings, Coarse Limestone Accumulated Volumn vs. Time

Fig. 164 Leaching profiles of coarse tailings mixed with 7.5% (w/w) coarse limestone (screen size -6.3 mm, -4 mesh) and under unsaturated conditions (columns #18 and 19), average cumulative drainage volume collected with time.



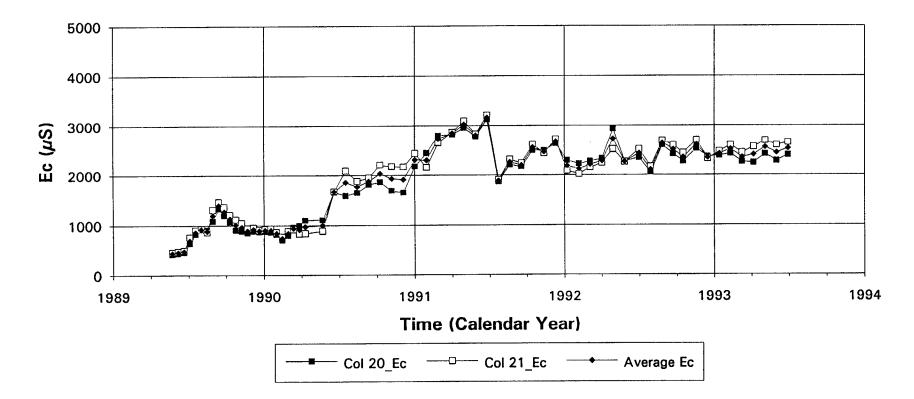
# Segregated Coarse Tailings, Wet Ground Limestone pH vs. Time

Fig. 165 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of pH with time.



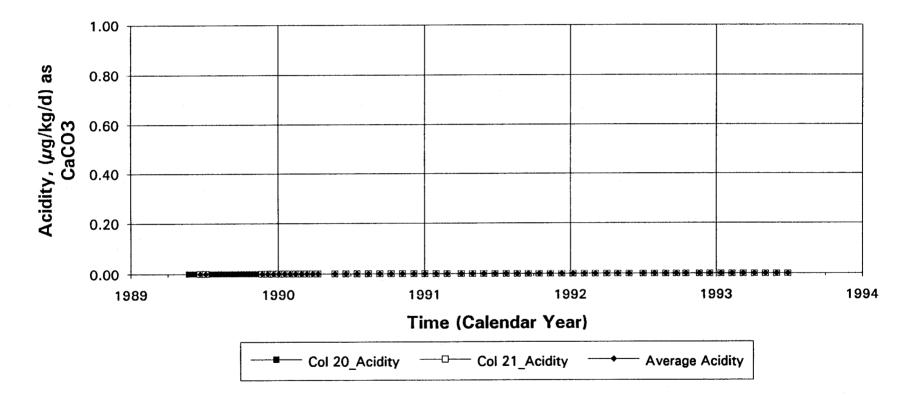
# Segregated Coarse Tailings, Wet Ground Limestone Eh(NHE) vs. Time

Fig. 166 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of Eh with time.



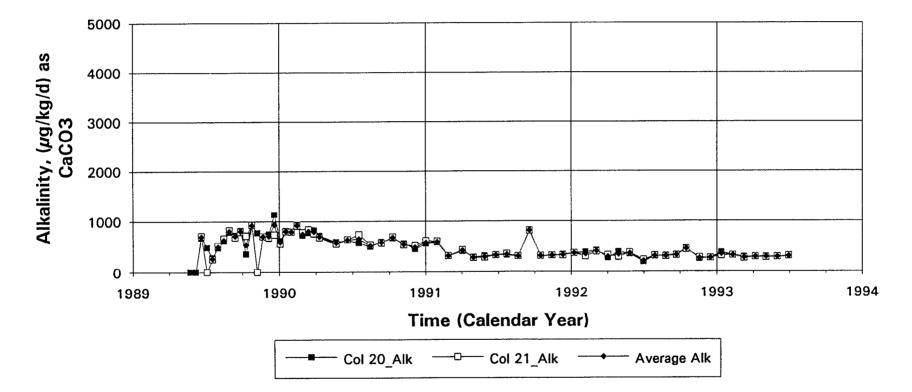
## Segregated Coarse Tailings, Wet Ground Limestone Electrical Conductivity vs. Time

Fig. 167 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of Ec with time.



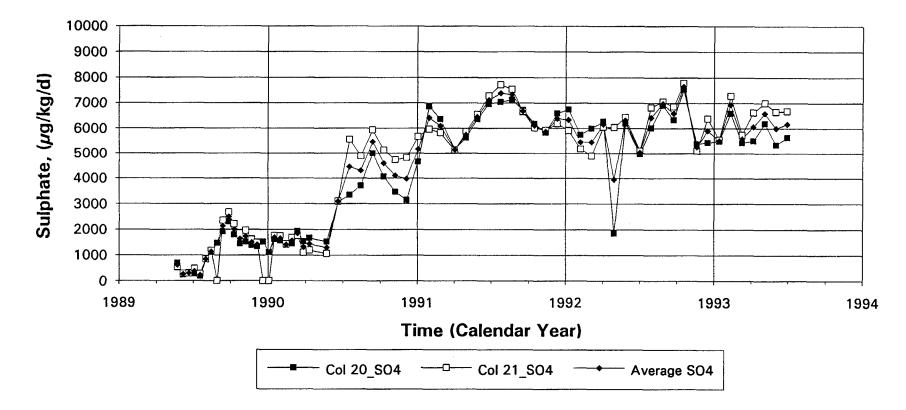
# Segregated Coarse Tailings, Wet Ground Limestone Acidity Loading vs. Time

Fig. 168 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of total acidity loading rate with time.



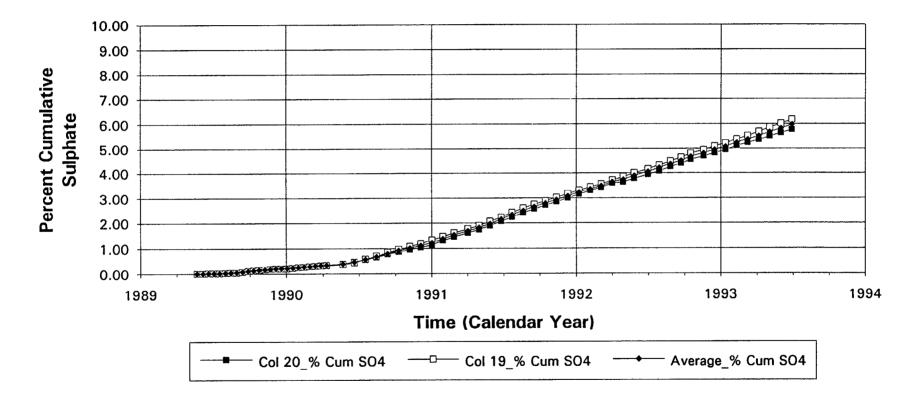
# Segregated Coarse Tailings, Wet Ground Limestone Alkalinity Loading vs. Time

Fig. 169 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of total alkalinity loading rate with time.



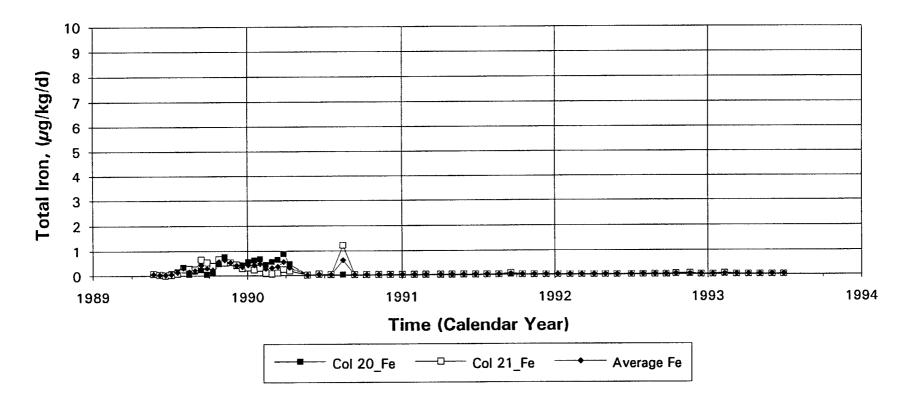
## Segregated Coarse Tailings, Wet Ground Limestone Sulphate Loading vs. Time

Fig. 170 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved  $SO_4^{-2}$  loading rate with time.



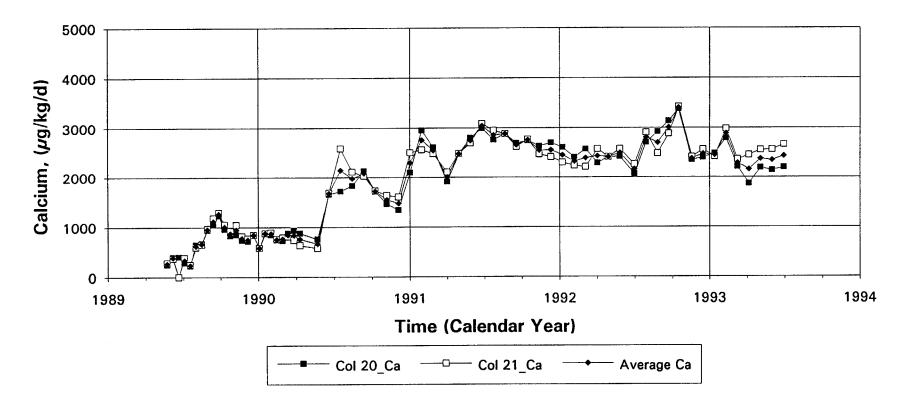
## Segregated Coarse Tailings, Wet Ground Limestone Cumulative Sulphate Removal vs. Time

Fig. 171 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



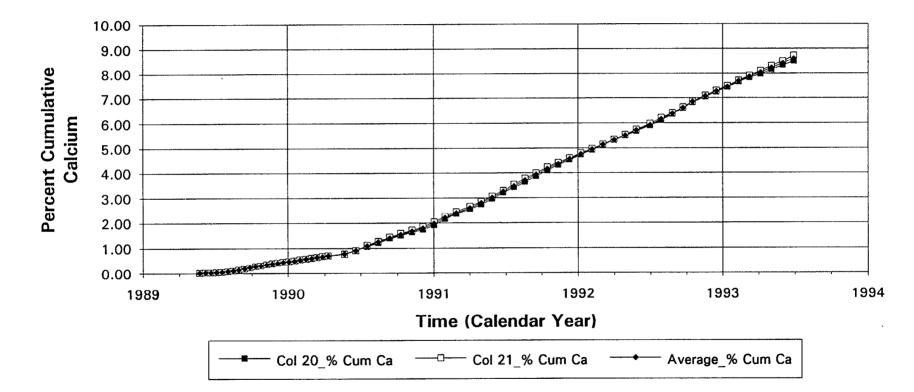
## Segregated Coarse Tailings, Wet Ground Limestone Total Iron Loading vs. Time

Fig. 172 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved total Fe loading rate with time.



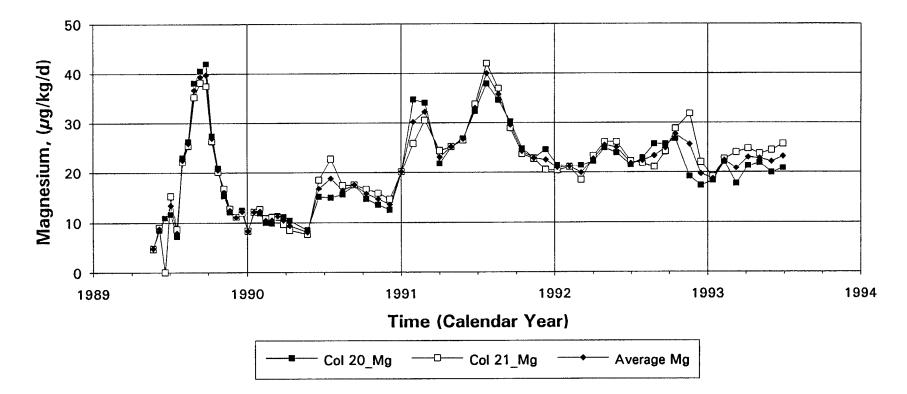
Segregated Coarse Tailings, Wet Ground Limestone Calcium Loading vs. Time

Fig. 173 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Ca loading rate with time.



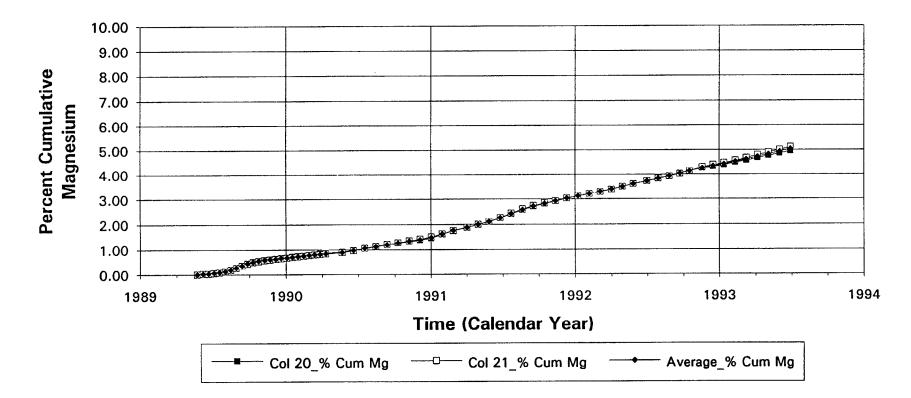
#### Segregated Coarse Tailings, Wet Ground Limestone Cumulative Calcium Removal vs. Time

Fig. 174 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), cumulative Ca removal with time.



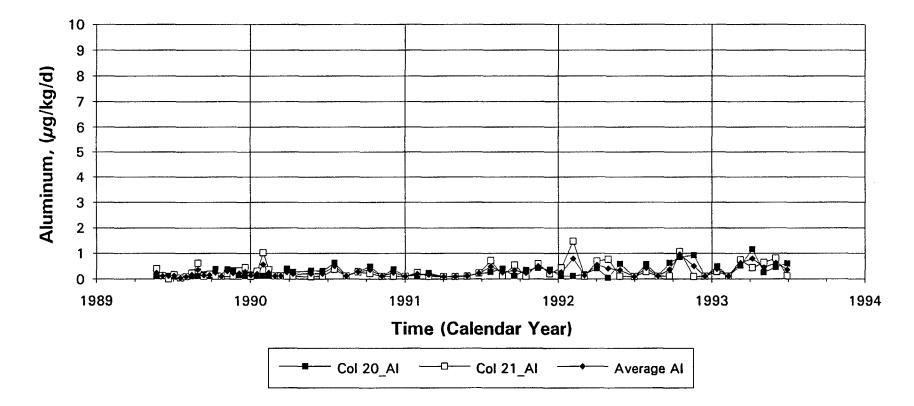
## Segregated Coarse Tailings, Wet Ground Limestone Magnesium Loading vs. Time

Fig. 175 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Mg loading rate with time.



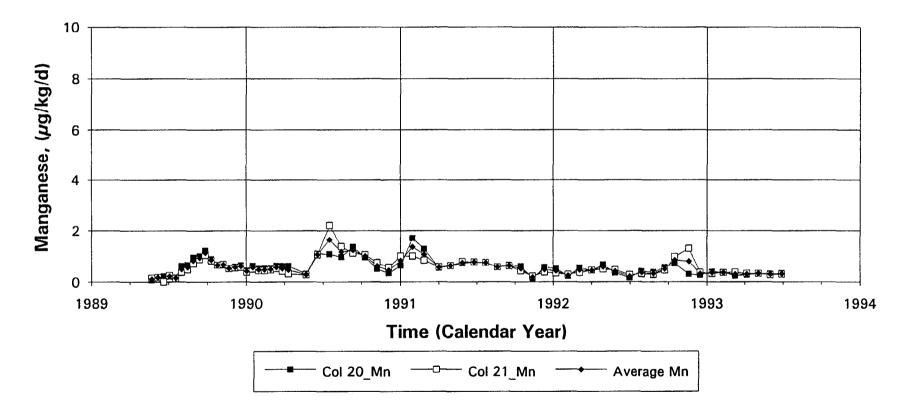
## Segregated Coarse Tailings, Wet Ground Limestone Cumulative Magnesium Removal vs. Time

Fig. 176 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), cumulative Mg removal with time.



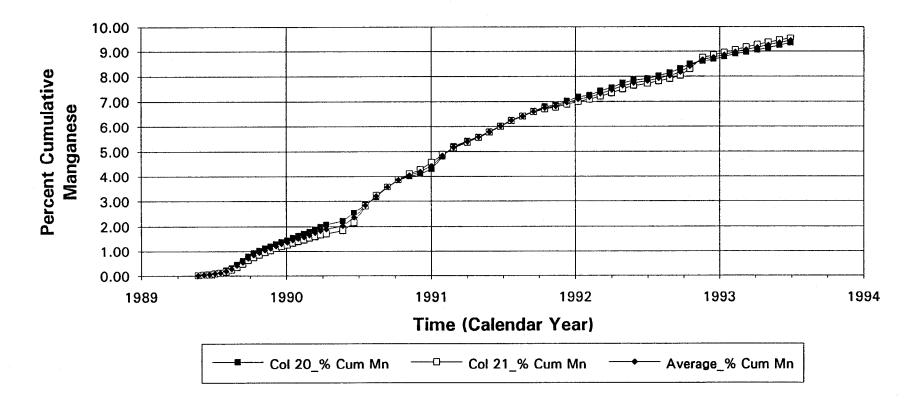
# Segregated Coarse Tailings, Wet Ground Limestone Aluminum Loading vs. Time

Fig. 177 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Al loading rate with time.



# Segregated Coarse Tailings, Wet Ground Limestone Manganese Loading vs. Time

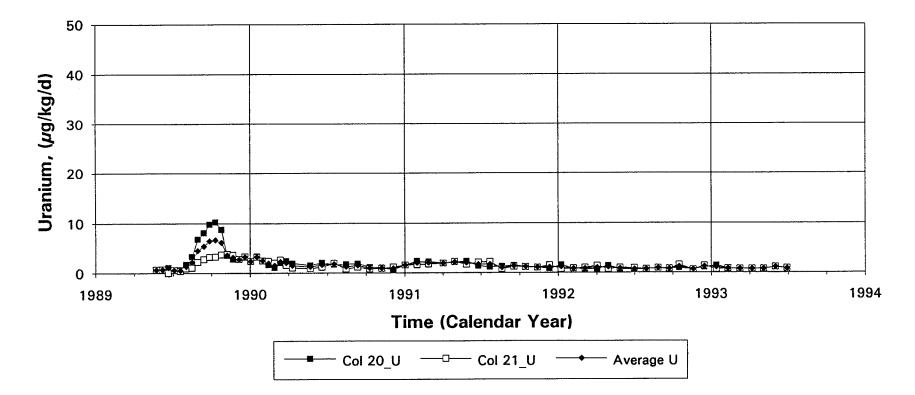
Fig. 178 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Mn loading rate with time.



#### Segregated Coarse Tailings, Wet Ground Limestone Cumulative Manganese Removal vs. Time

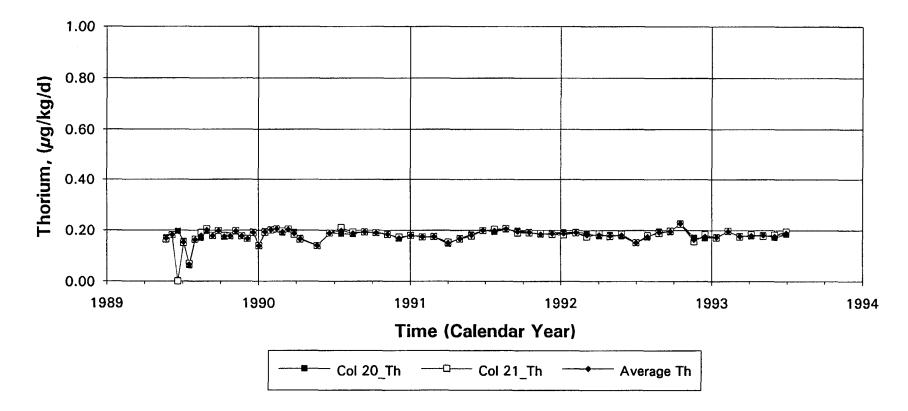
Fig. 179 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), cumulative Mn removal with time.

2-179



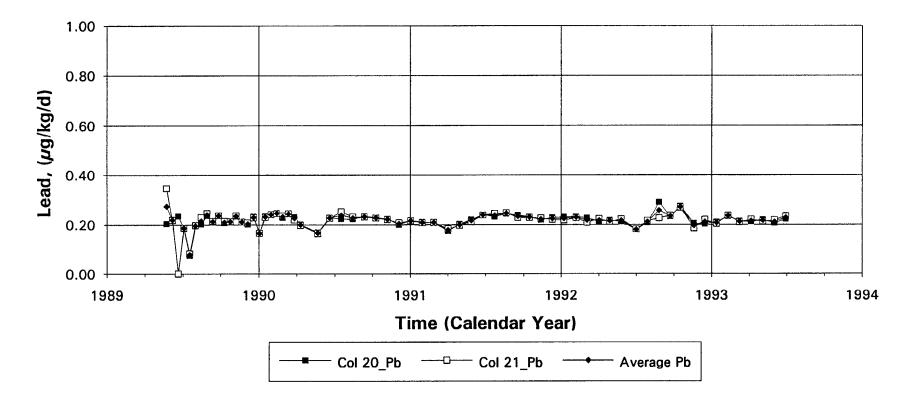
#### Segregated Coarse Tailings, Wet Ground Limestone Uranium Loading vs. Time

Fig. 180 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved U loading rate with time.



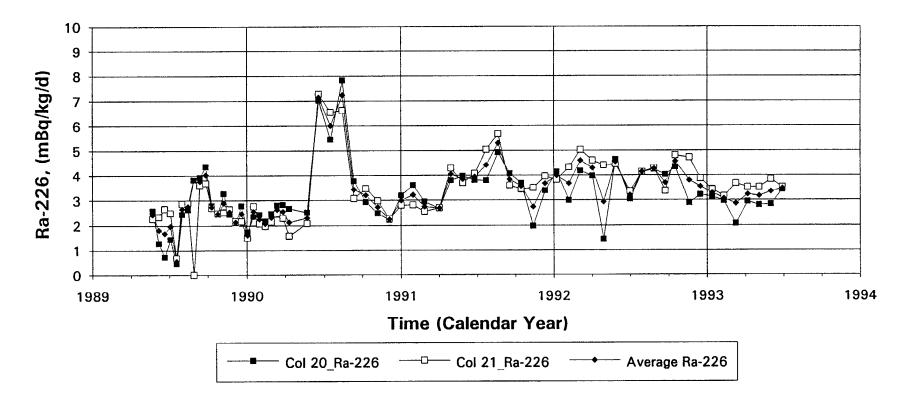
## Segregated Coarse Tailings, Wet Ground Limestone Thorium Loading vs. Time

Fig. 181 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Th loading rate with time.



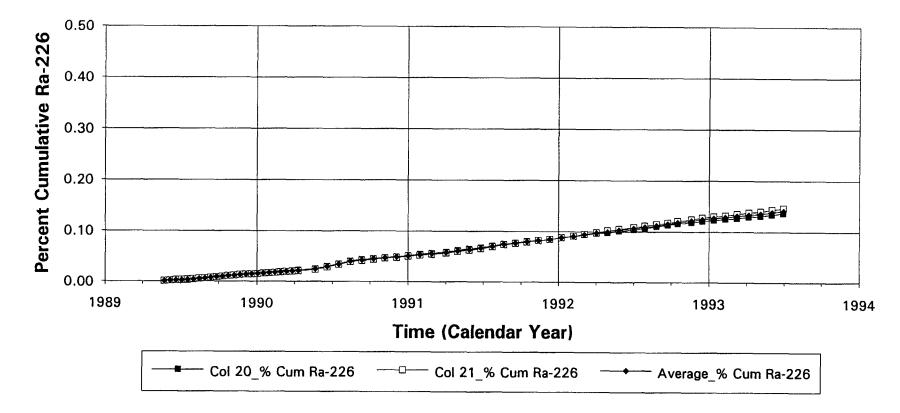
## Segregated Coarse Tailings, Wet Ground Limestone Lead Loading vs. Time

Fig. 182 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Pb loading rate with time.



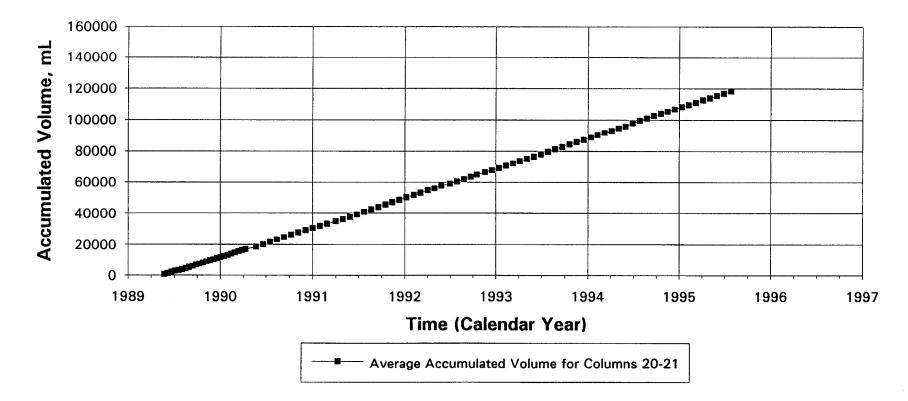
#### Segregated Coarse Tailings, Wet Ground Limestone Ra-226 Loading vs. Time

Fig. 183 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), variation of dissolved Ra-226 loading rate with time.



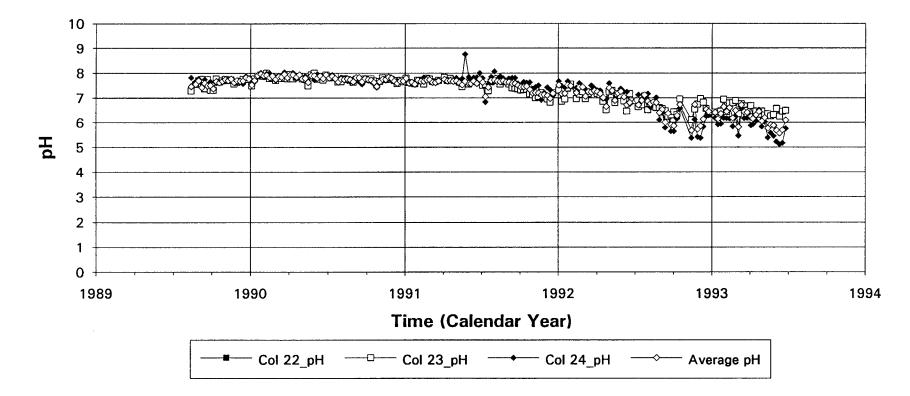
# Segregated Coarse Tailings, Wet Ground Limestone Cumulative Ra-226 Removal vs. Time

Fig. 184 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), cumulative Ra-226 removal with time.



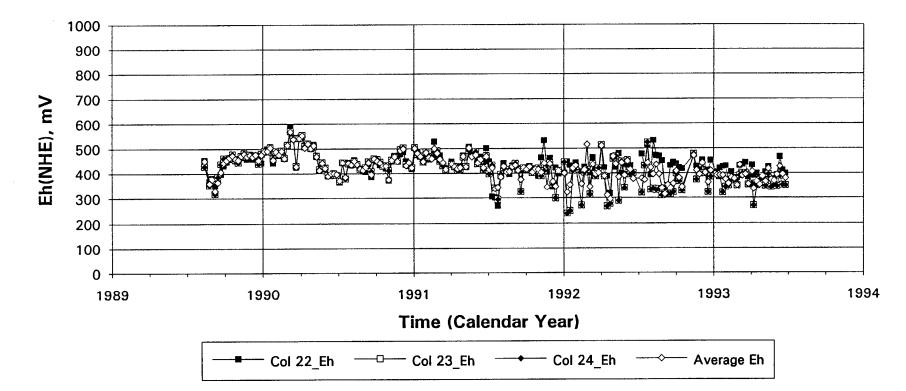
#### Segregated Coarse Tailings, Wet Ground Limestone Accumulated Volume vs. Time

Fig. 185 Leaching profiles of coarse tailings mixed with 7.5% (w/w) wet ground (pulverized) limestone under unsaturated conditions (columns #20 and 21), average cumulative drainage volume collected with time.



Submerged Coarse Tailings pH vs. Time

Fig. 186 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of pH with time.



## Submerged Coarse Tailings Eh(NHE) vs. Time

Fig. 187 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of Eh with time.

Submerged Coarse Tailings Electrical Conductivity vs. Time

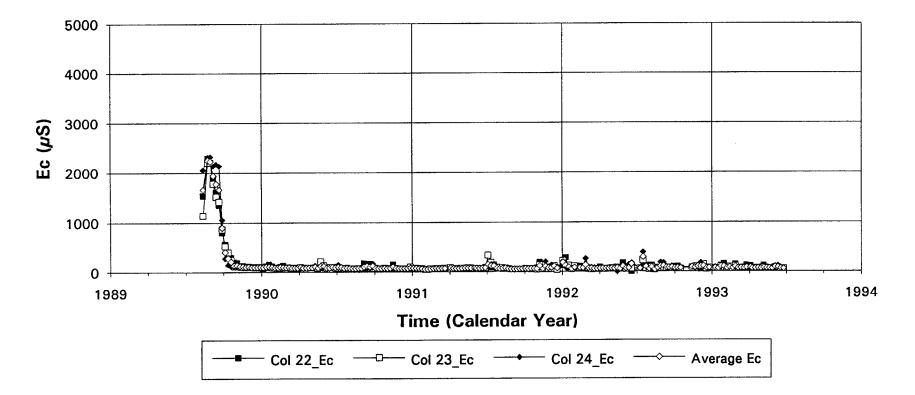


Fig. 188 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of Ec with time.

Submerged Coarse Tailings Acidity Loading vs. Time

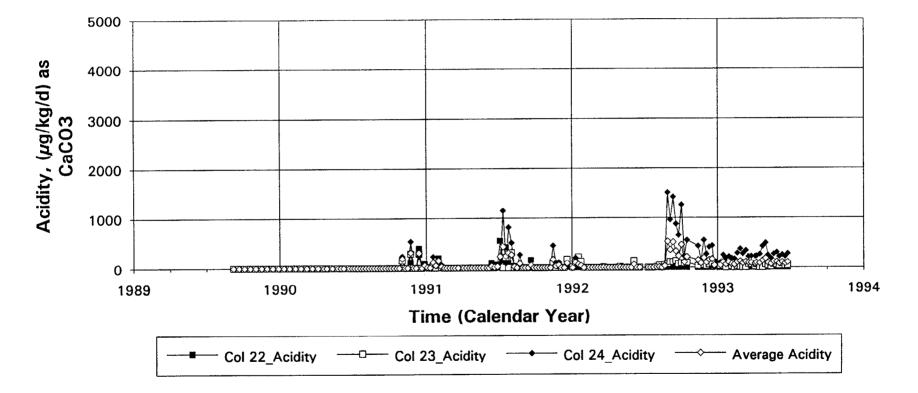
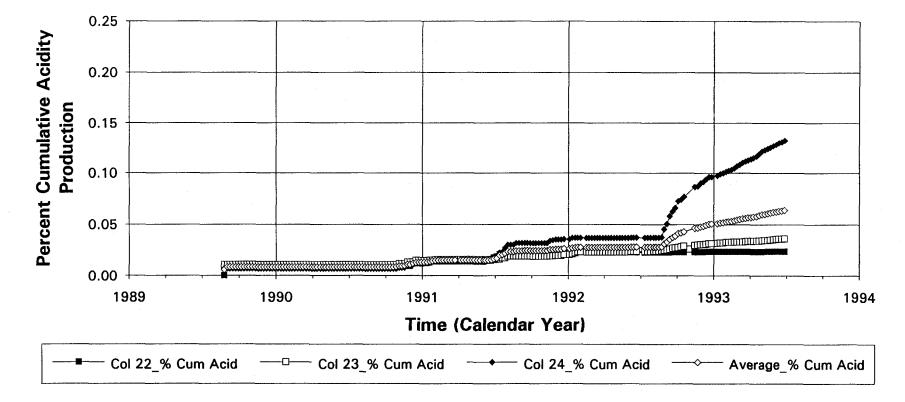


Fig. 189 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of total acidity loading rate with time.



# Submerged Coarse Tailings Cumulative Acidity Production vs. Time

Fig. 190 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative acidity production with time.



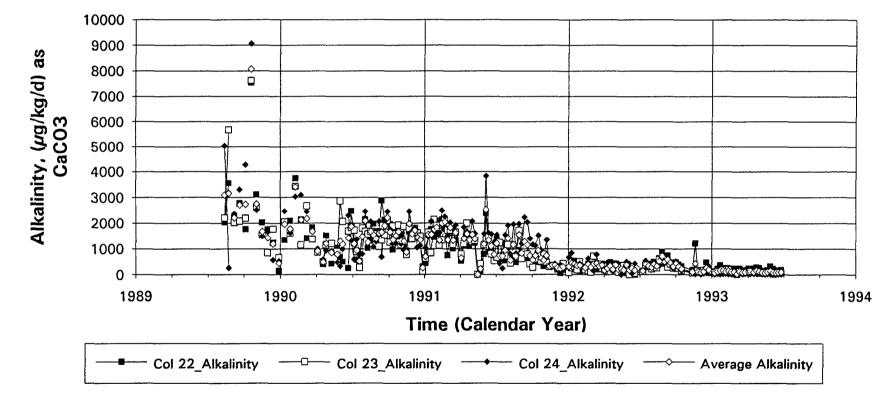


Fig. 191 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of total alkalinity loading rate with time.



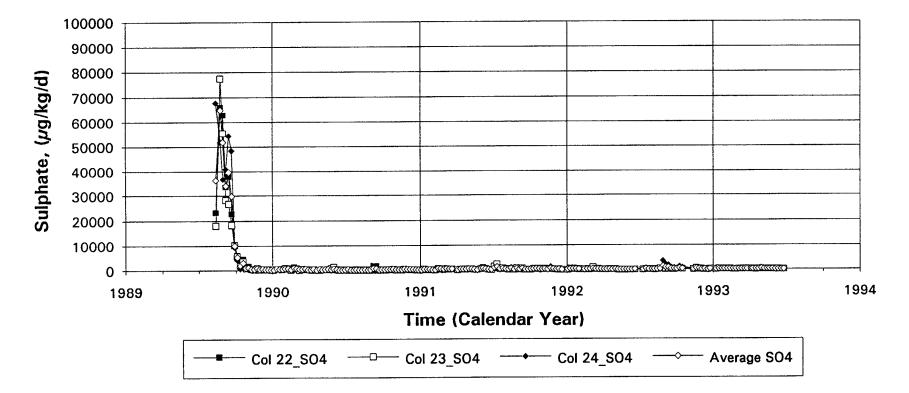


Fig. 192 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved  $SO_4^{-2}$  loading rate with time.



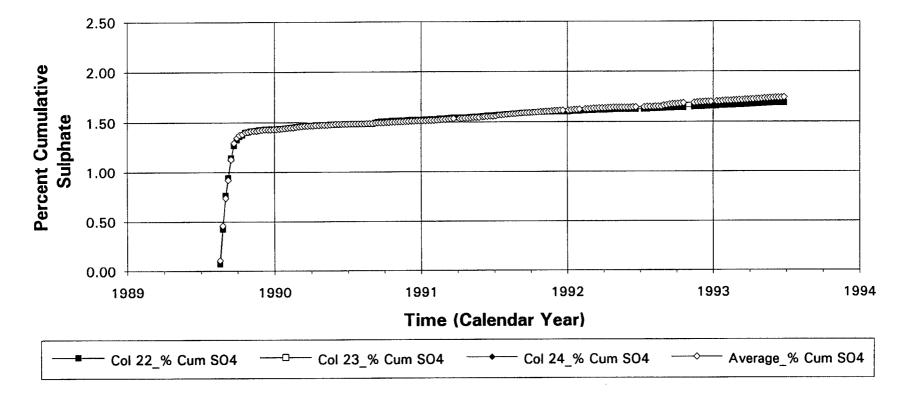


Fig. 193 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



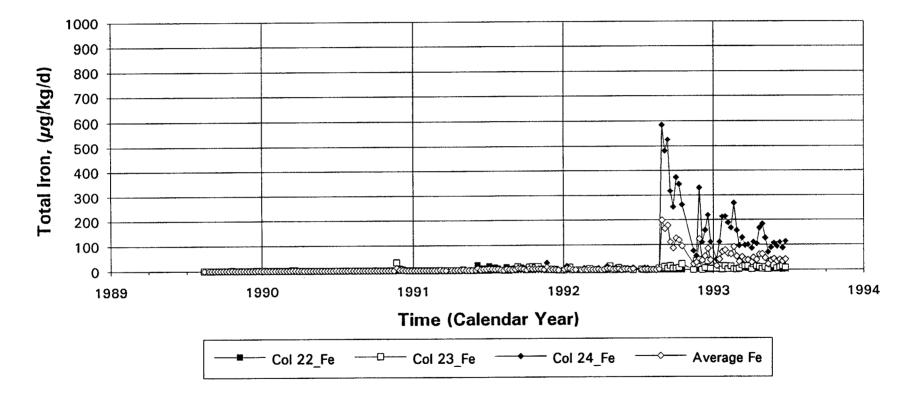
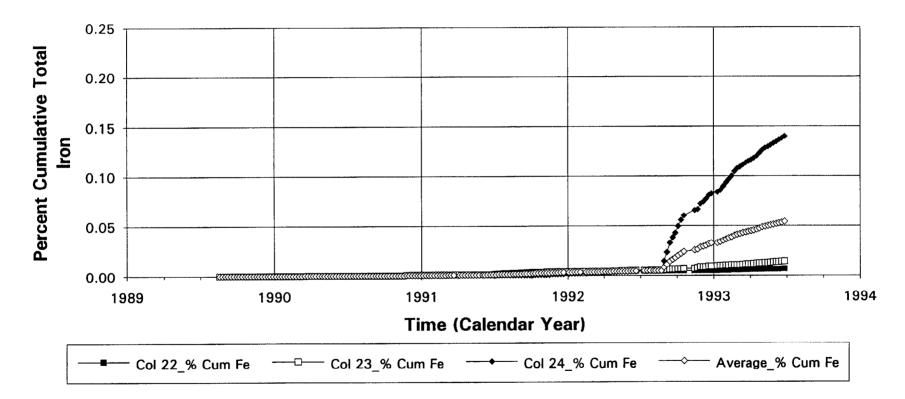


Fig. 194 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved total Fe loading rate with time.



Submerged Coarse Tailings Cumulative Total Iron Removal vs. Time

Fig. 195 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative total Fe removal with time.

Submerged Coarse Tailings Calcium Loading vs. Time

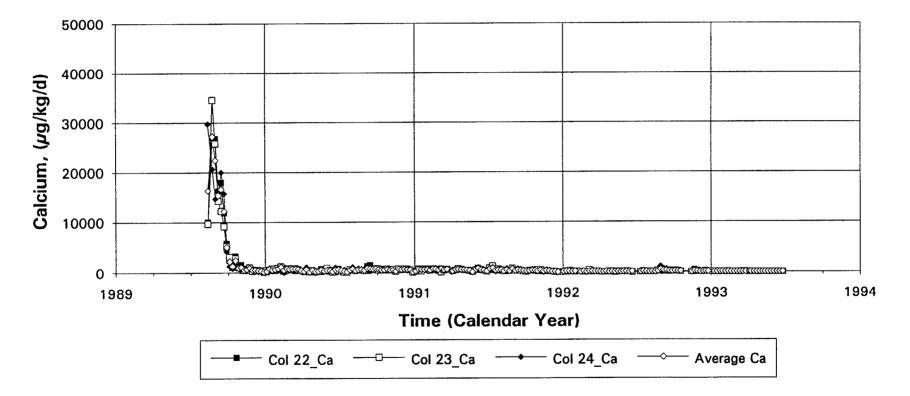
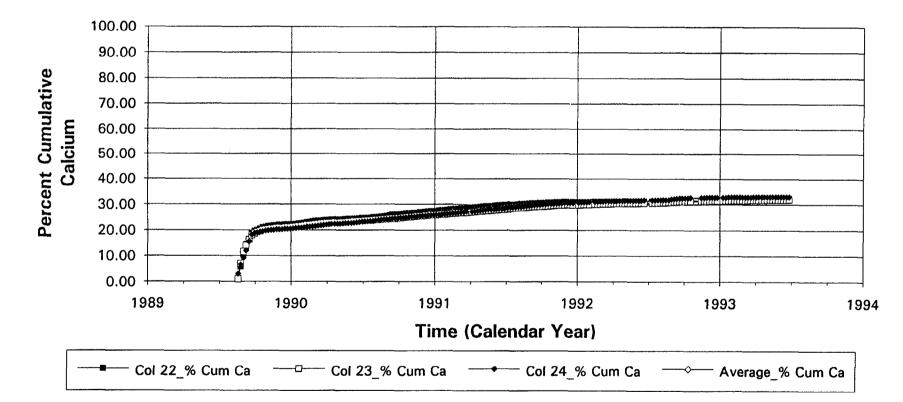


Fig. 196 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Ca loading rate with time.

.



# Submerged Coarse Tailings Cumulative Calcium Removal vs. Time

Fig. 197 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative Ca removal with time.

#### Submerged Coarse Tailings Magnesium Loading vs. Time

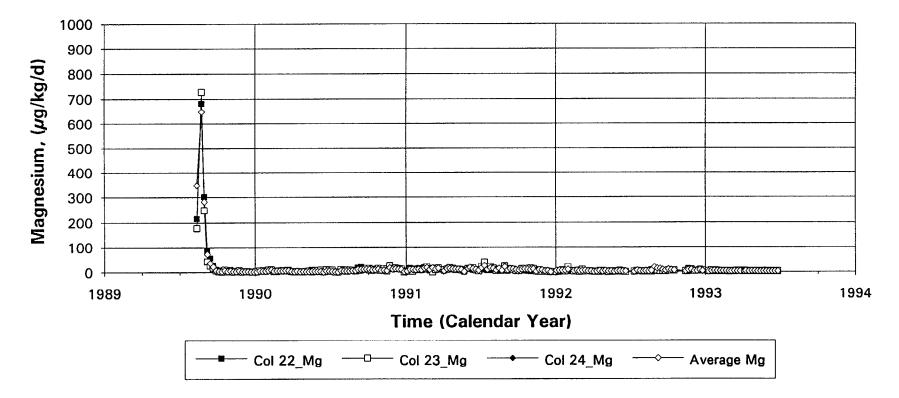
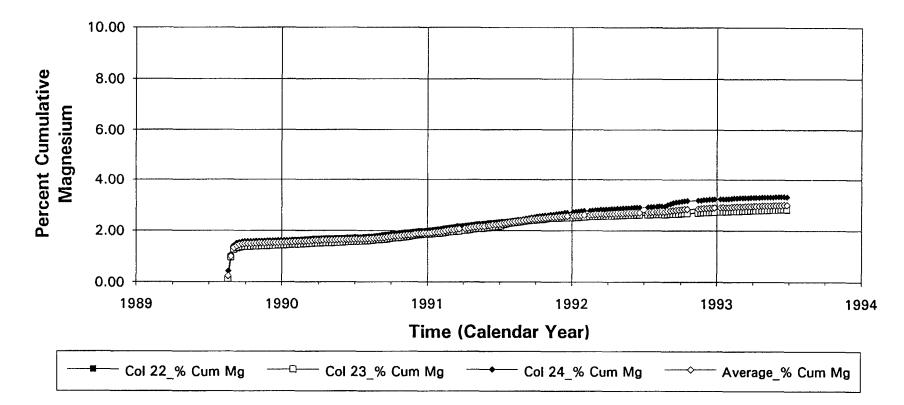


Fig. 198 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Mg loading rate with time.



# Submerged Coarse Tailings Cumulative Magnesium Removal vs. Time

Fig. 199 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative Mg removal with time.



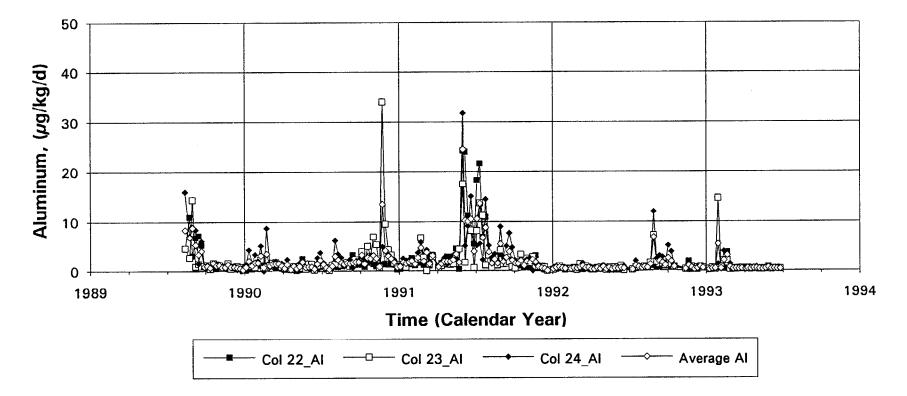
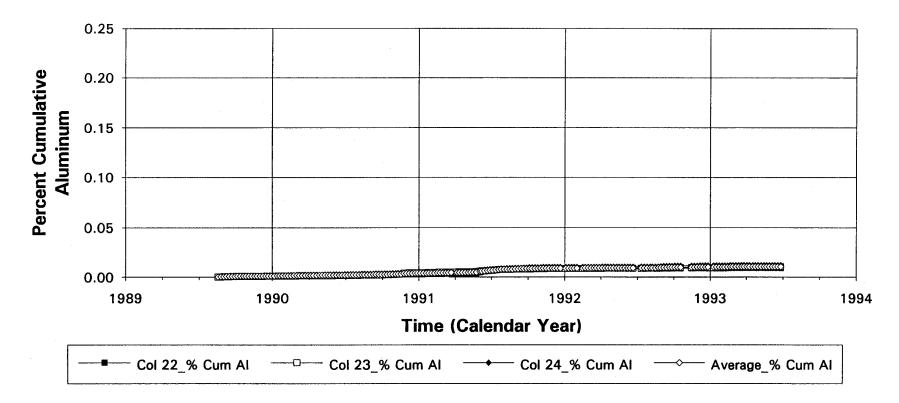


Fig. 200 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Al loading rate with time.



## Submerged Coarse Tailings Cumulative Aluminum Removal vs. Time

Fig. 201 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative Al removal with time.



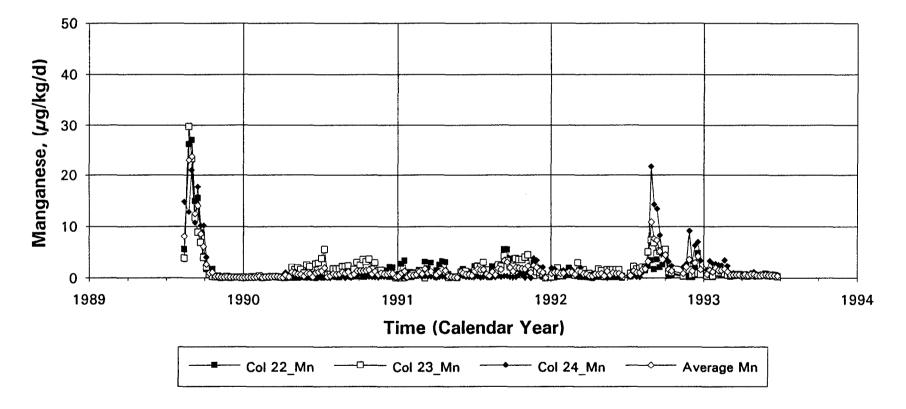
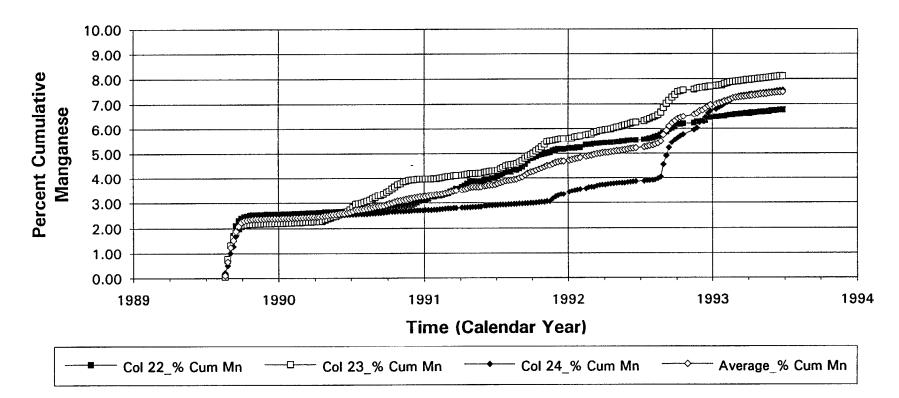


Fig. 202 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Mn loading rate with time.



Submerged Coarse Tailings Cumulative Manganese Removal vs. Time

Fig. 203 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative Mn removal with time.



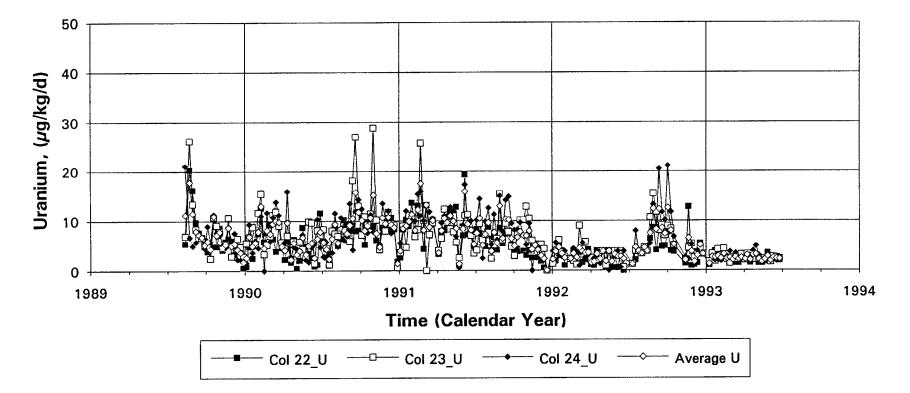


Fig. 204 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved U loading rate with time.



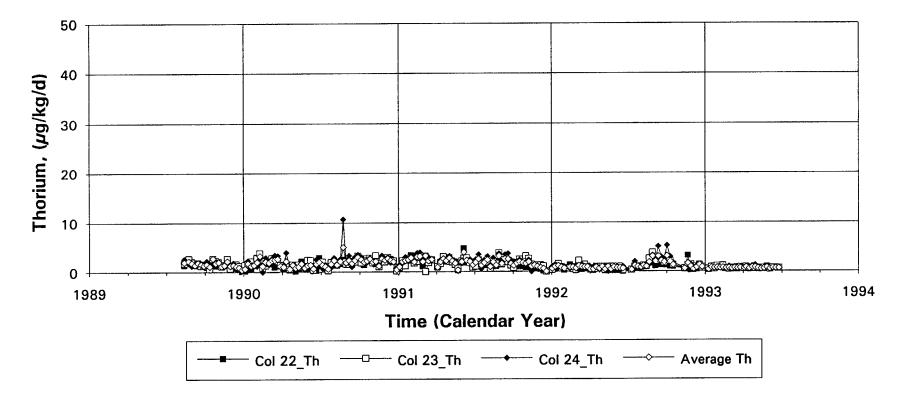


Fig. 205 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Th loading rate with time.



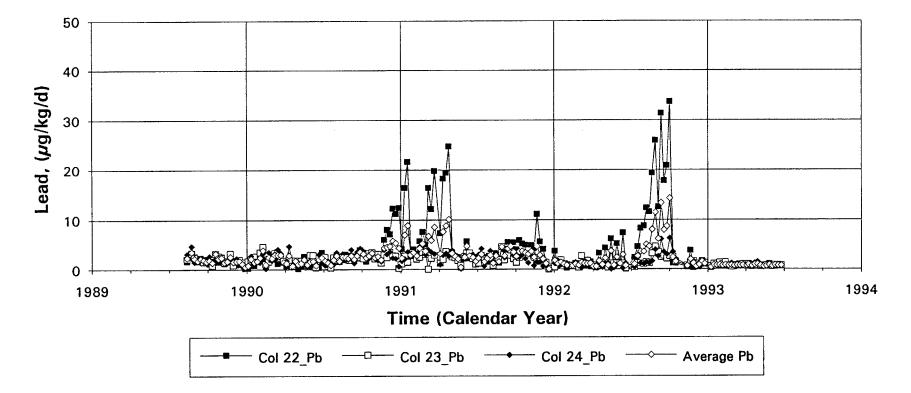


Fig. 206 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Pb loading rate with time.

Submerged Coarse Tailings Ra-226 Loading vs. Time

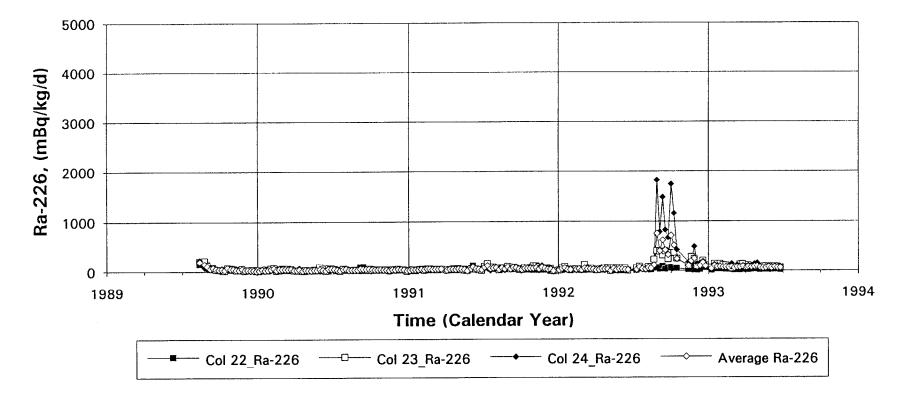
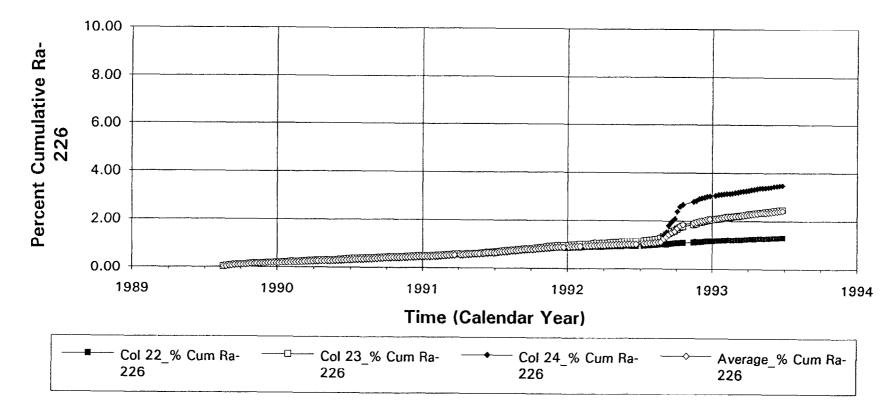


Fig. 207 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), variation of dissolved Ra-226 loading rate with time.



# Submerged Coarse Tailings Cumulative Ra-226 Removal vs. Time

Fig. 208 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), cumulative Ra-226 removal with time.



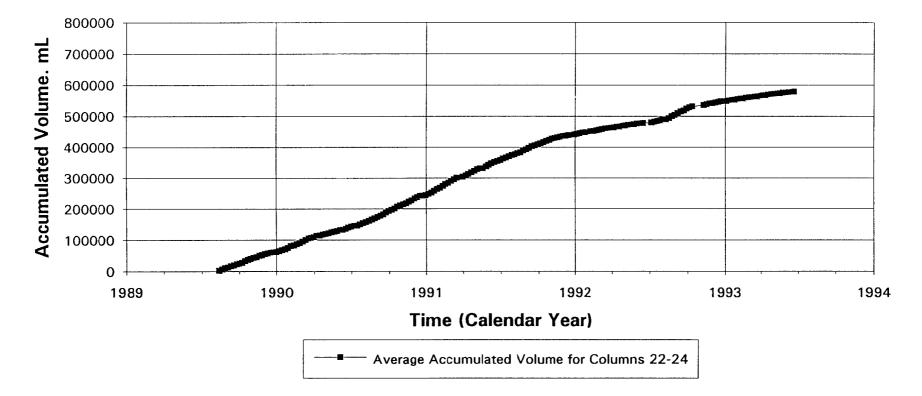
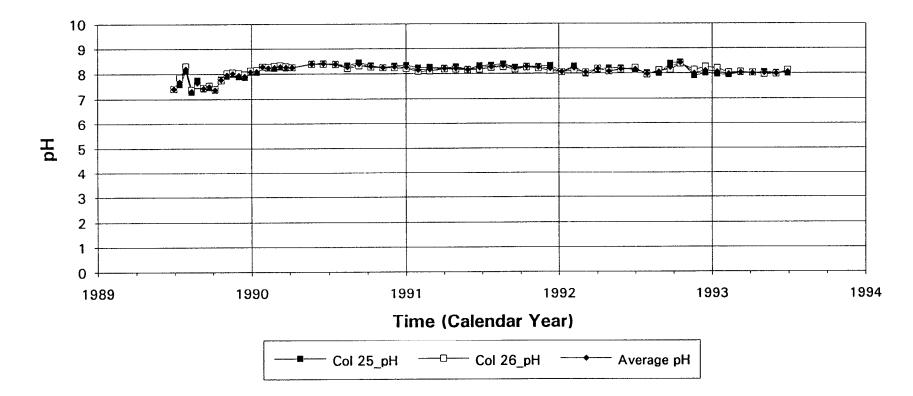
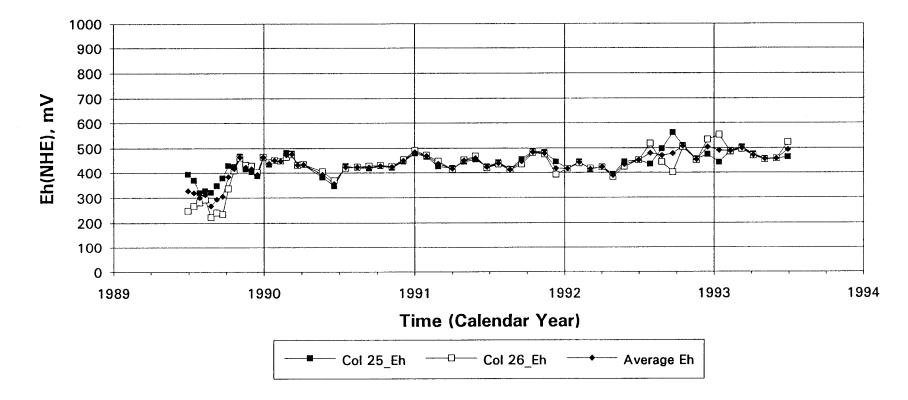


Fig. 209 Leaching profiles of coarse tailings without additional limestone under submerged conditions (columns #22-24), average cumulative drainage volume collected with time.



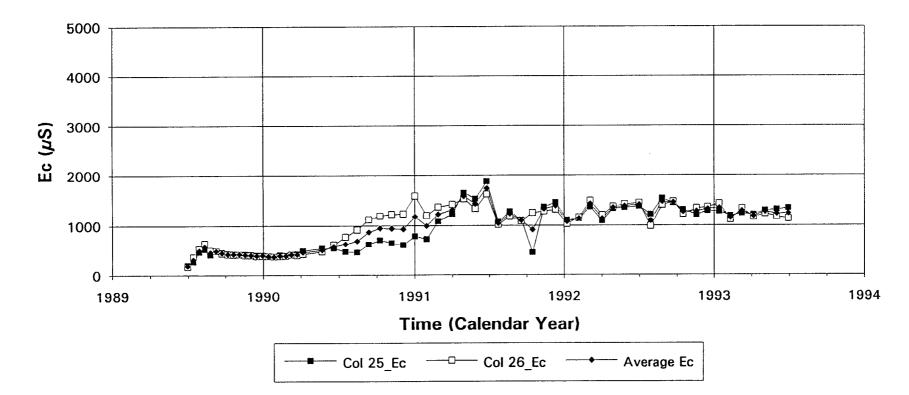
### Medium Acid Generating Crushed Waste Rock pH vs.Time

Fig. 210 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of pH with time.



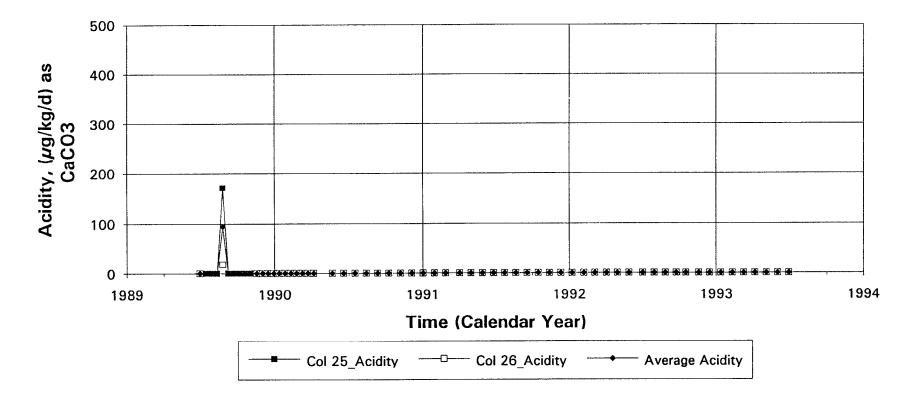
### Medium Acid Generating Crushed Waste Rock Eh(NHE) vs.Time

Fig. 211 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of Eh with time.



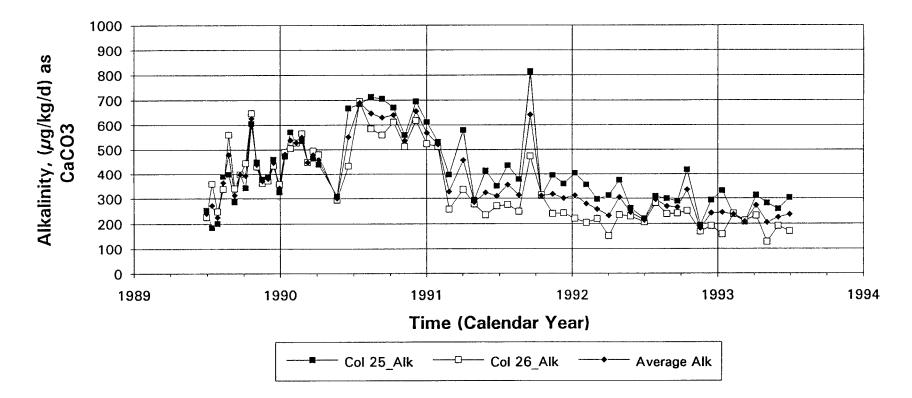
### Medium Acid Generating Crushed Waste Rock Electrical Conductivity vs.Time

Fig. 212 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of Ec with time.



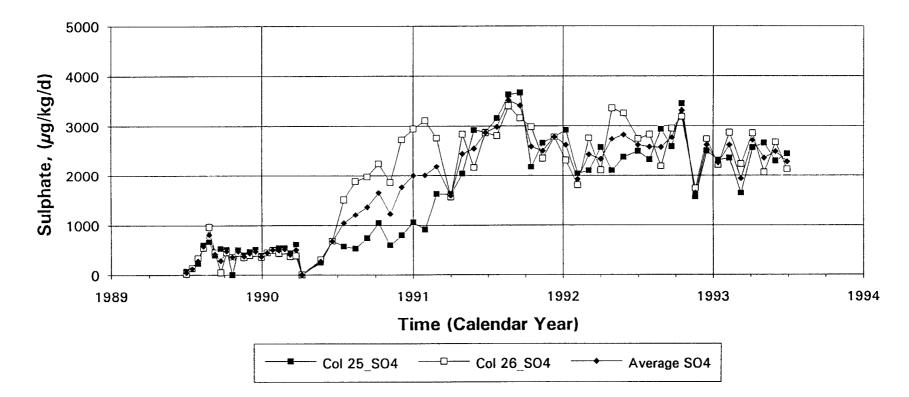
# Medium Acid Generating Crushed Waste Rock Acidity Loading vs.Time

Fig. 213 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of total acidity loading rate with time.



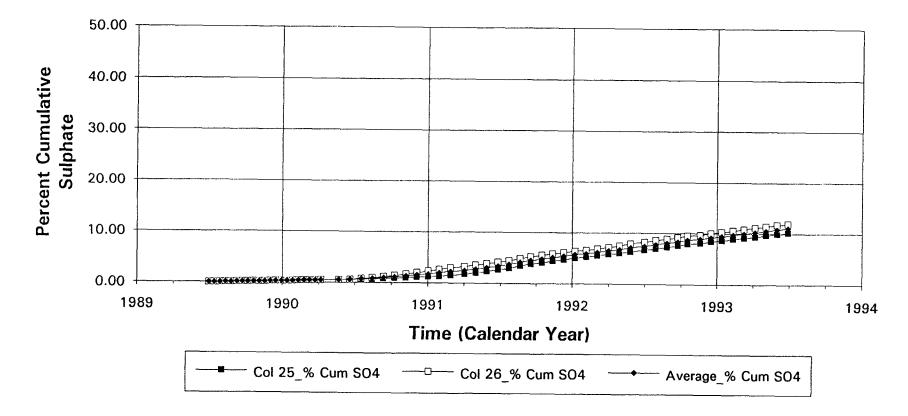
### Medium Acid Generating Crushed Waste Rock Alkalinity Loading vs.Time

Fig. 214 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of total alkalinity loading rate with time.



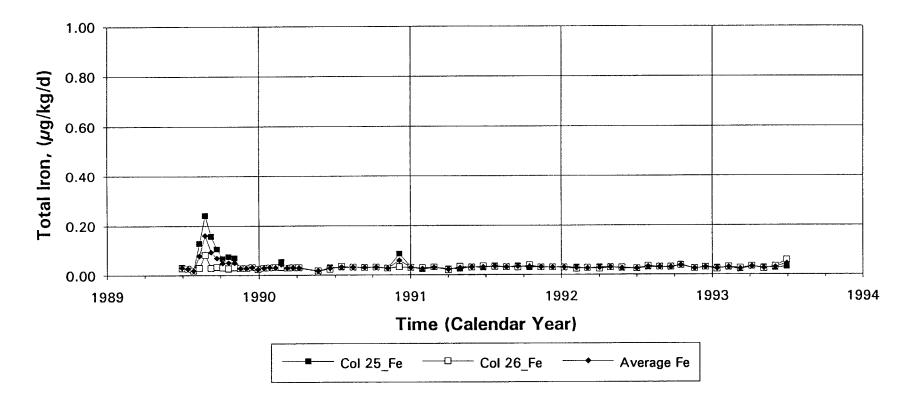
#### Medium Acid Generating Crushed Waste Rock Sulphate Loading vs.Time

Fig. 215 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved SO<sub>4</sub><sup>-2</sup> loading rate with time.



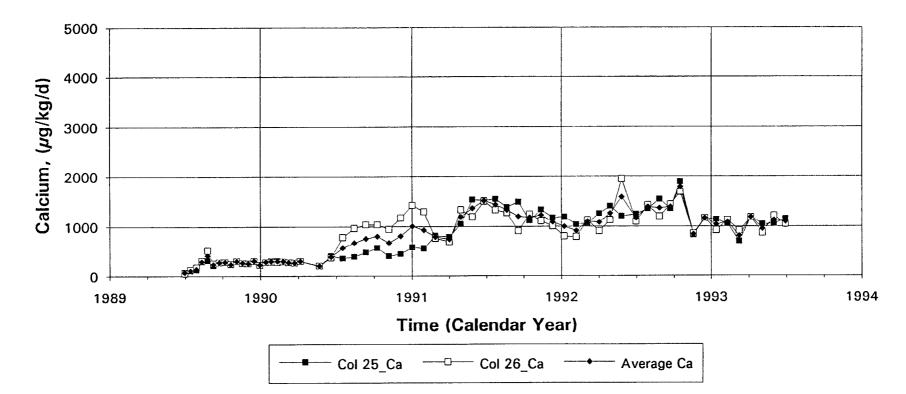
# Medium Acid Generating Crushed Waste Rock Cumulative Sulphate Removal vs. Time

Fig. 216 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), cumulative SO<sub>4</sub><sup>-2</sup> removal (as % total S) with time.



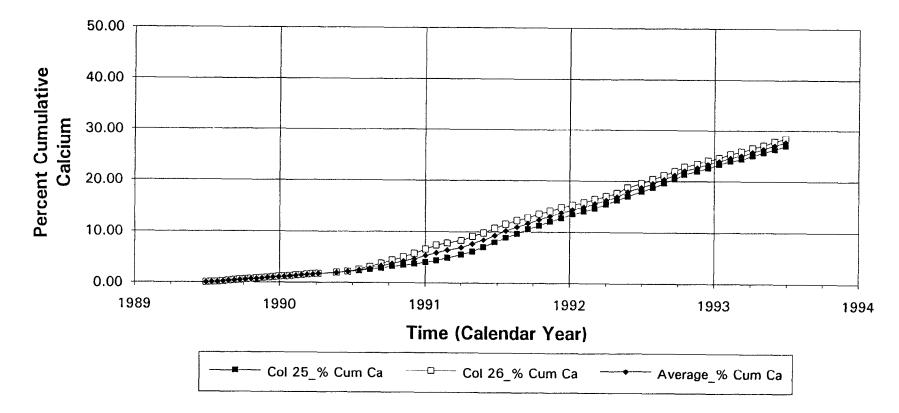
# Medium Acid Generating Crushed Waste Rock Total Iron Loading vs.Time

Fig. 217 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved total Fe loading rate with time.



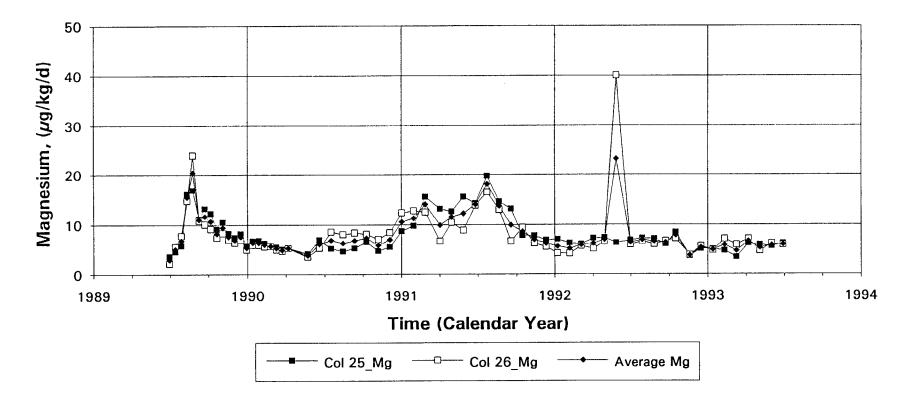
# Medium Acid Generating Crushed Waste Rock Calcium Loading vs.Time

Fig. 218 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Ca loading rate with time.



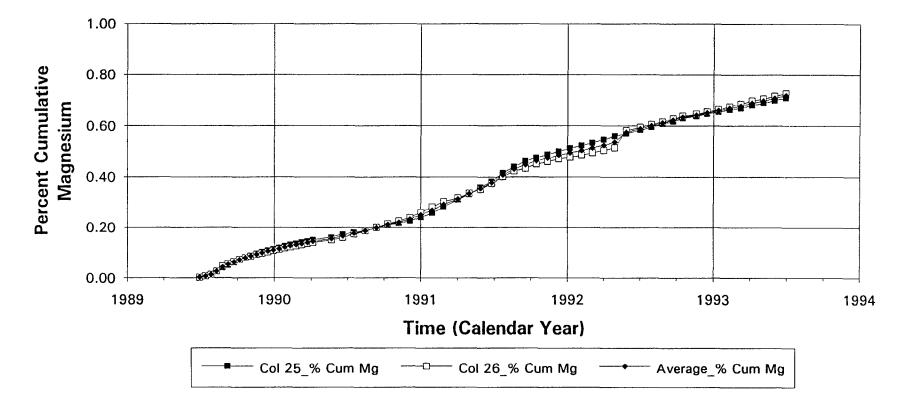
# Medium Acid Generating Crushed Waste Rock Cumulative Calcium Removal vs. Time

Fig. 219 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), cumulative Ca removal with time.



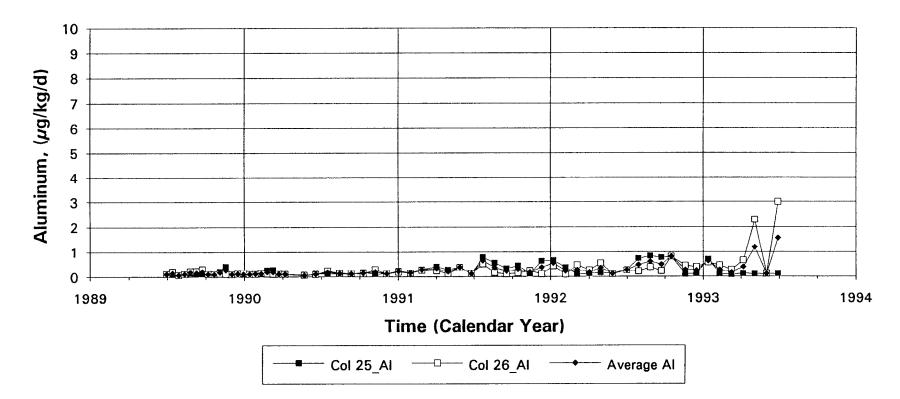
### Medium Acid Generating Crushed Waste Rock Magnesium Loading vs.Time

Fig. 220 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Mg loading rate with time.



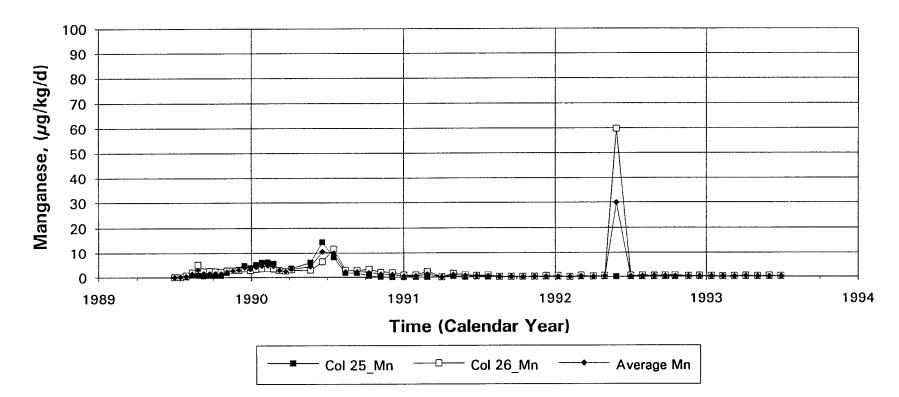
#### Medium Acid Generating Crushed Waste Rock Cumulative Magnesium Removal vs. Time

Fig. 221 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), cumulative Mg removal with time.



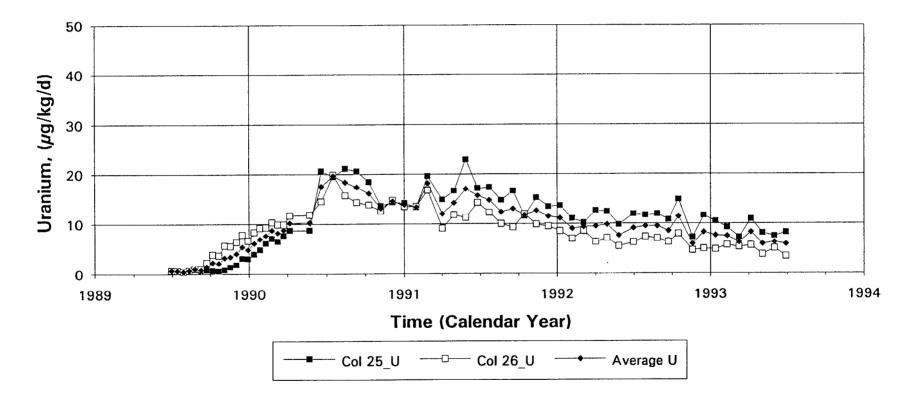
### Medium Acid Generating Crushed Waste Rock Aluminum Loading vs.Time

Fig. 222 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Al loading rate with time.



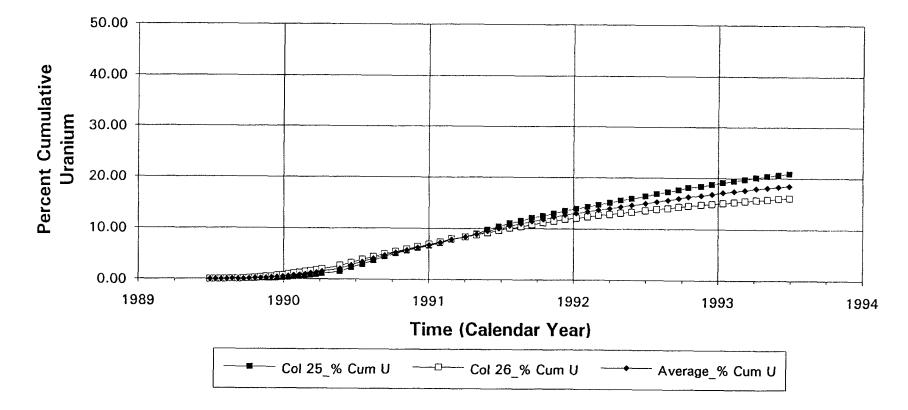
### Medium Acid Generating Crushed Waste Rock Manganese Loading vs.Time

Fig. 223 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Mn loading rate with time.



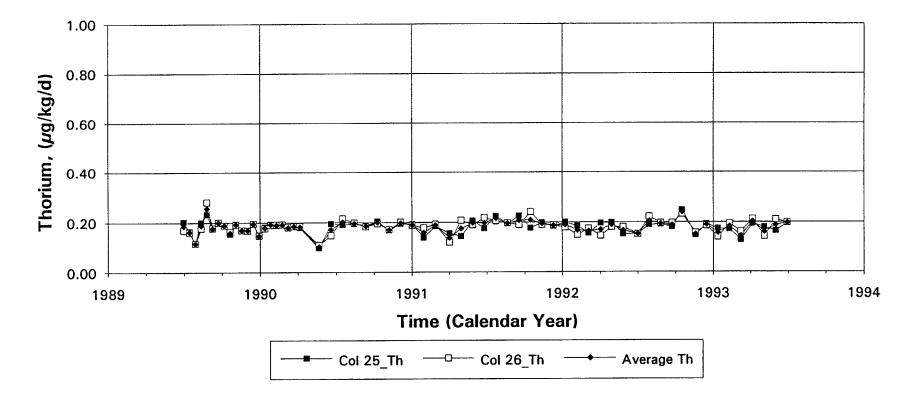
### Medium Acid Generating Crushed Waste Rock Uranium Loading vs.Time

Fig. 224 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved U loading rate with time.



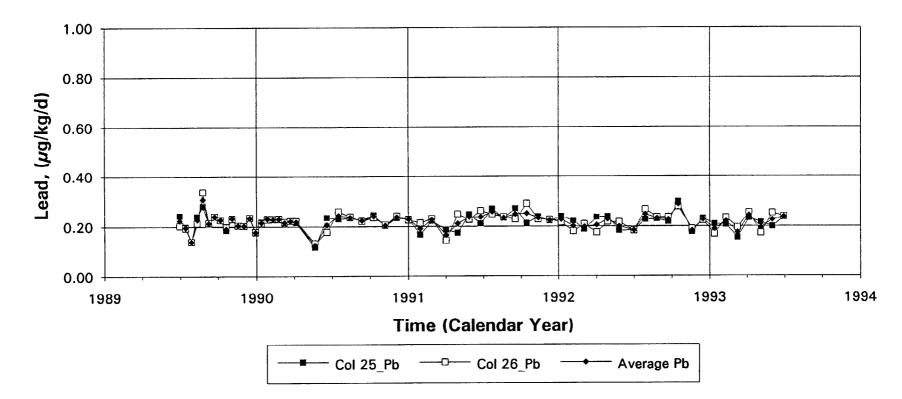
## Medium Acid Generating Crushed Waste Rock Cumulative Uranium Removal vs. Time

Fig. 225 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), cumulative U removal with time.



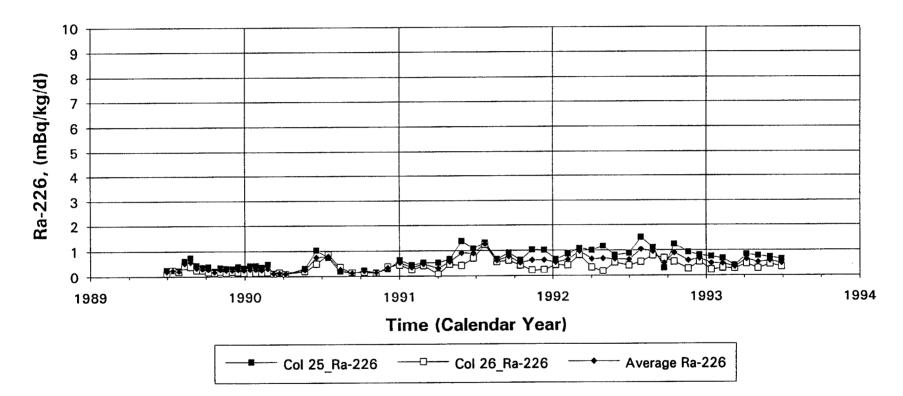
### Medium Acid Generating Crushed Waste Rock Thorium Loading vs.Time

Fig. 226 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Th loading rate with time.



# Medium Acid Generating Crushed Waste Rock Lead Loading vs.Time

Fig. 227 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Pb loading rate with time.



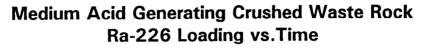
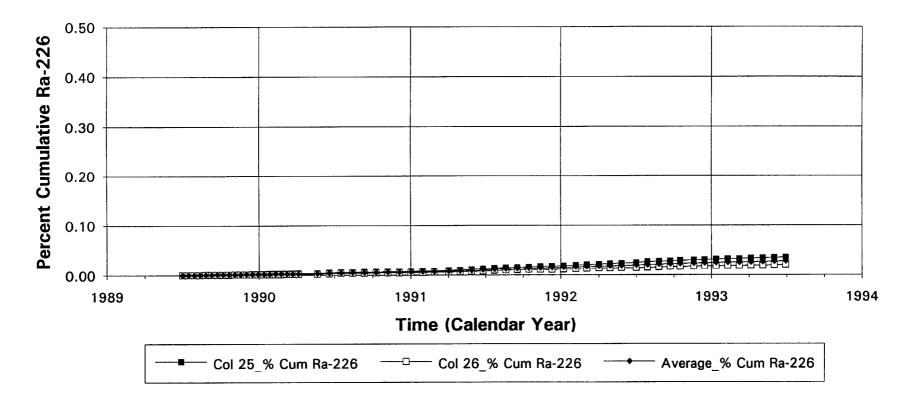
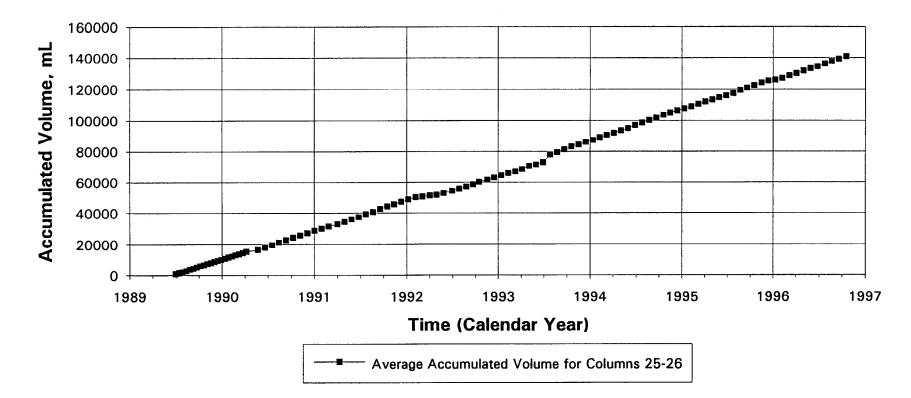


Fig. 228 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), variation of dissolved Ra-226 loading rate with time.



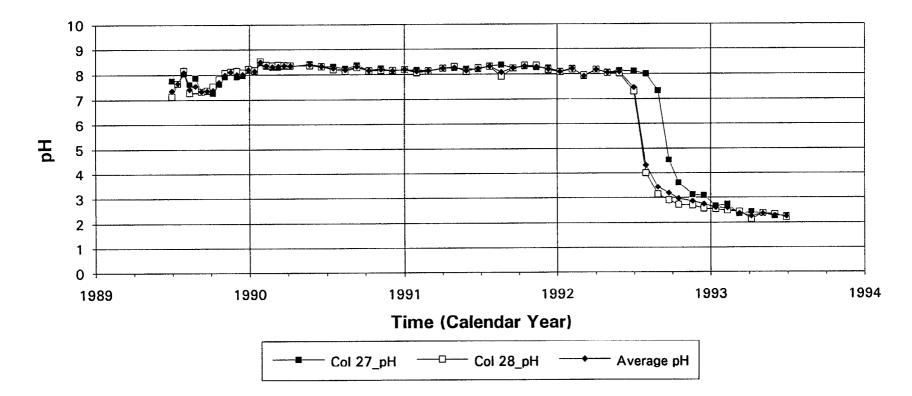
### Medium Acid Generating Crushed Waste Rock Cumulative Ra-226 Removal vs. Time

Fig. 229 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), cumulative Ra-226 removal with time.



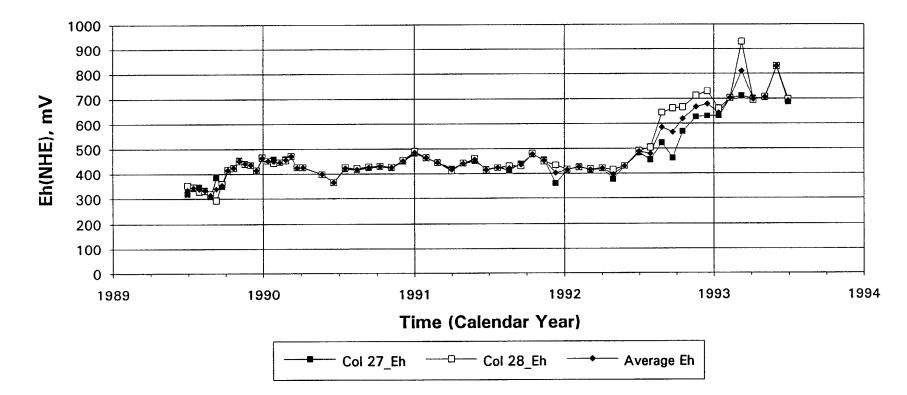
#### Medium Acid Generating Crushed Wasterock Accumulated Volumn vs. Time

Fig. 230 Leaching profiles of crushed waste rock having medium acid generation potential (NNP = -10.7 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #25 and 26), average cumulative drainage volume collected with time.



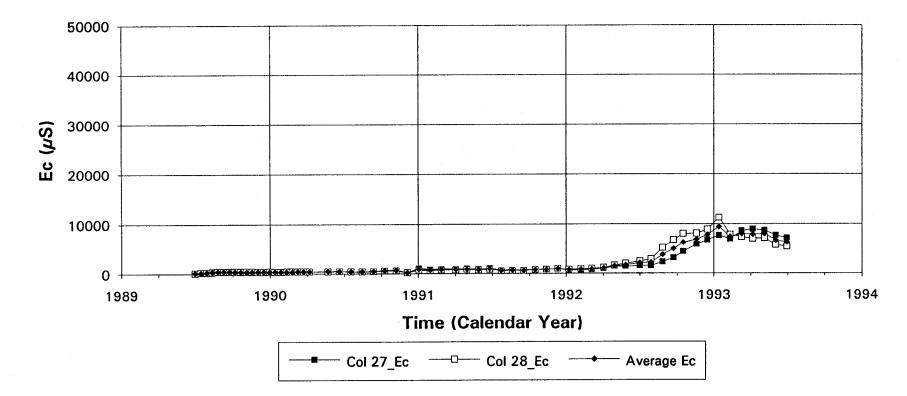
# High Acid Generating Crushed Waste Rock pH vs. Time

Fig. 231 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of pH with time.



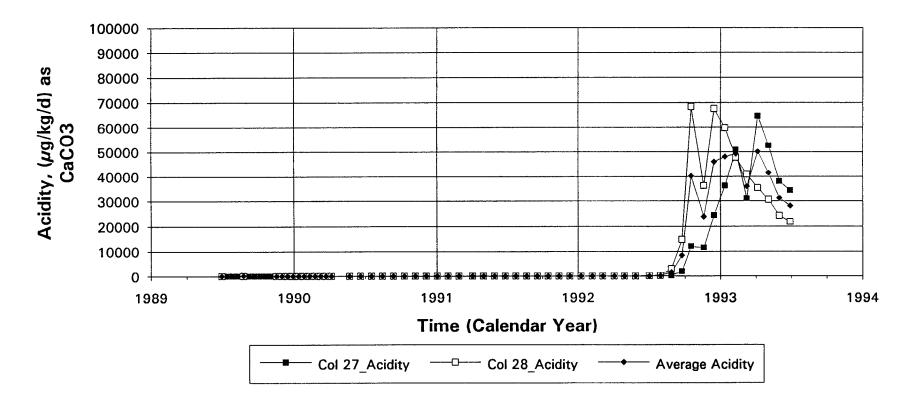
# High Acid Generating Crushed Waste Rock Eh(NHE) vs. Time

Fig. 232 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of Eh with time.



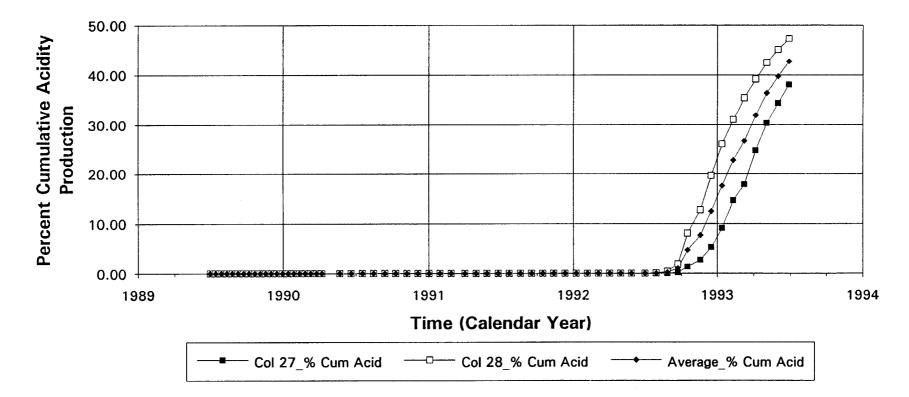
High Acid Generating Crushed Waste Rock Electrical Conductivity vs. Time

Fig. 233 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of Ec with time.



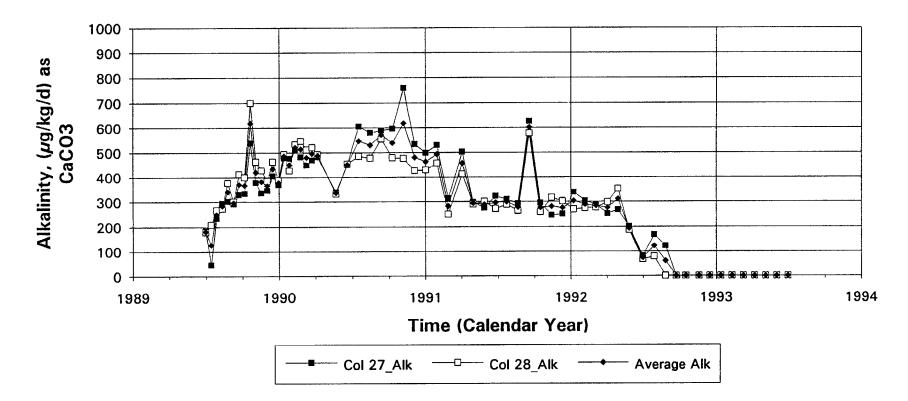
# High Acid Generating Crushed Waste Rock Acidity Loading vs. Time

Fig. 234 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of total acidity loading rate with time.



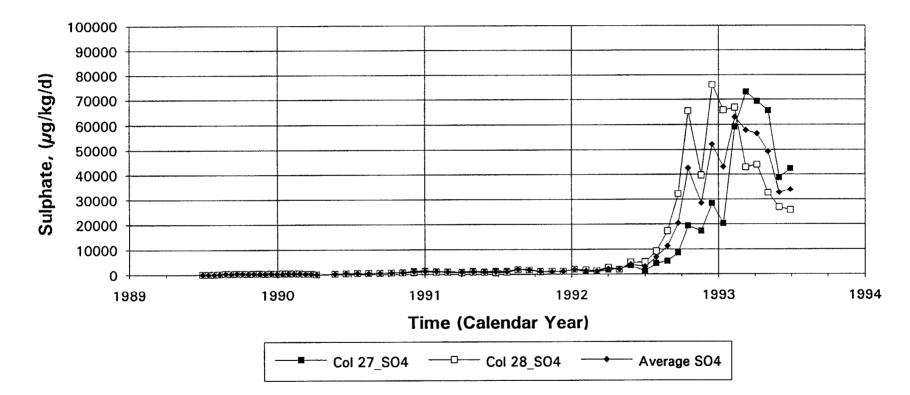
High Acid Generating Crushed Waste Rock Cumulative Acidity Production vs. Time

Fig. 235 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative acidity production with time.



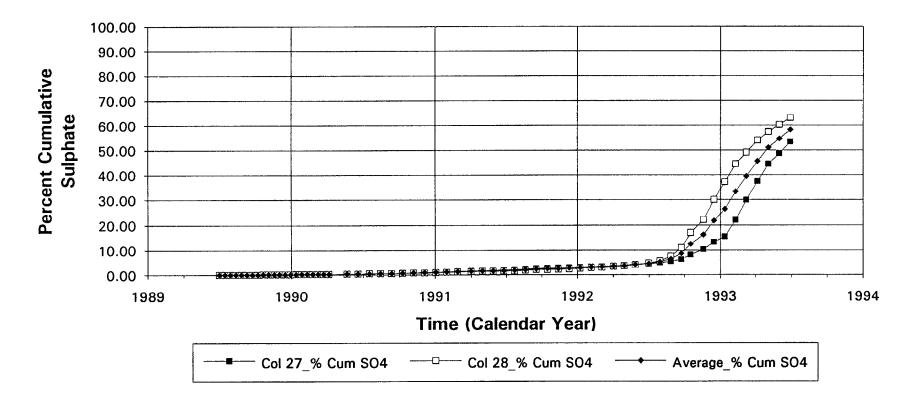
#### High Acid Generating Crushed Waste Rock Alkalinity Loading vs. Time

Fig. 236 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of total alkalinity loading rate with time.



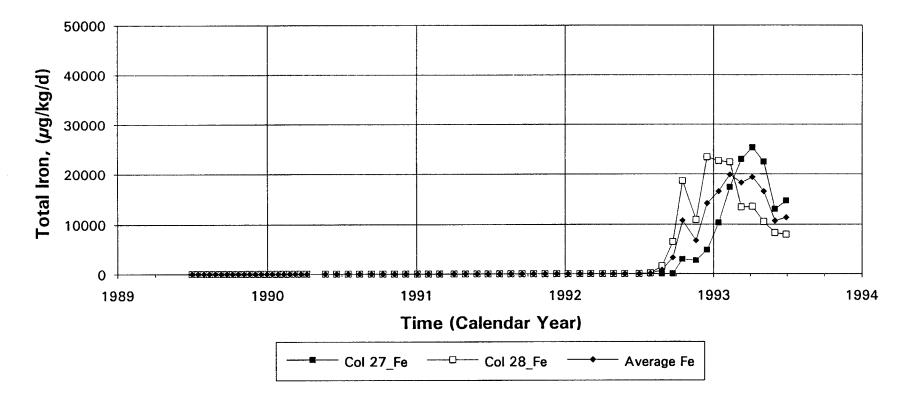
# High Acid Generating Crushed Waste Rock Sulphate Loading vs. Time

Fig. 237 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved SO<sub>4</sub><sup>-2</sup> loading rate with time.



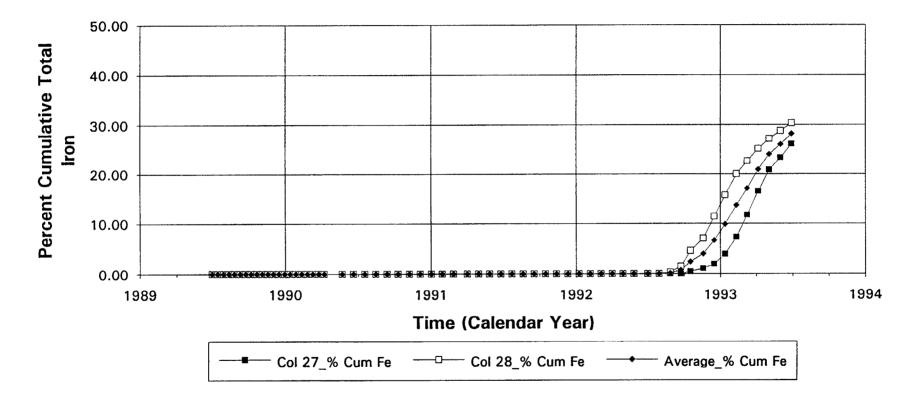
# High Acid Generating Crushed Waste Rock Cumulative Sulphate Removal vs. Time

Fig. 238 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative SO<sub>4</sub><sup>-2</sup> removal (as % total S) with time.



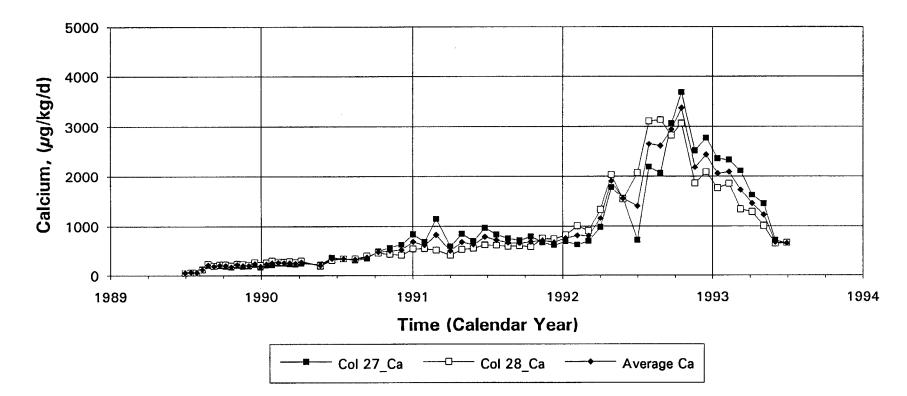
#### High Acid Generating Crushed Waste Rock Total Iron Loading vs. Time

Fig. 239 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved total Fe loading rate with time.



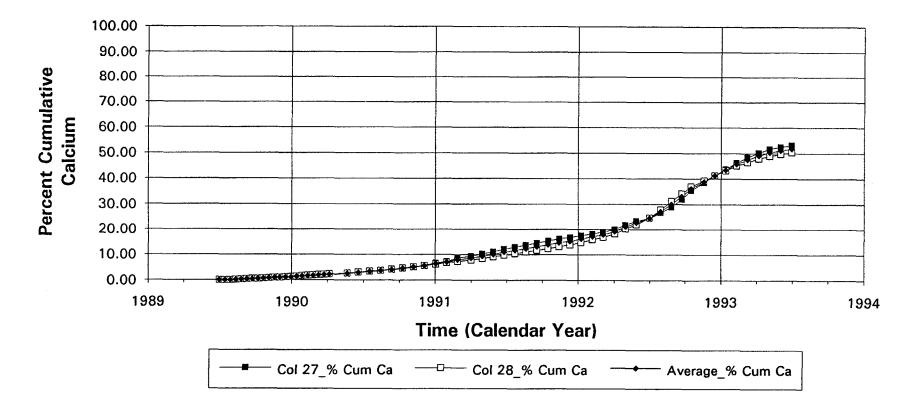
#### High Acid Generating Crushed Waste Rock Cumulative Total Iron Removal vs. Time

Fig. 240 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative total Fe removal with time.



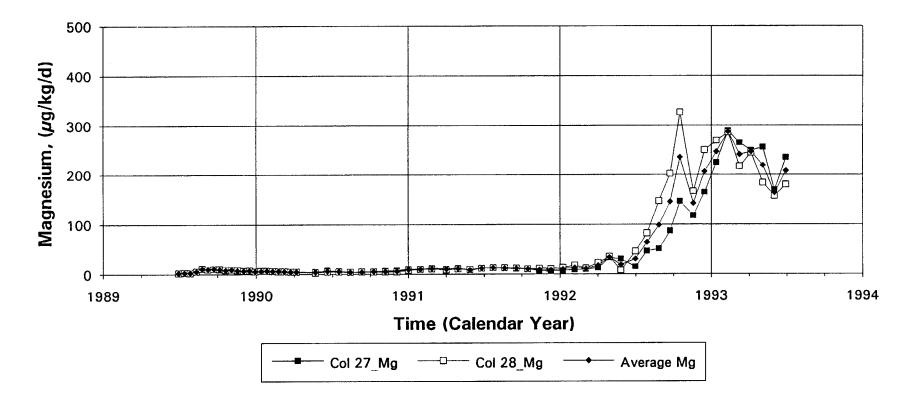
#### High Acid Generating Crushed Waste Rock Calcium Loading vs. Time

Fig. 241 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Ca loading rate with time.



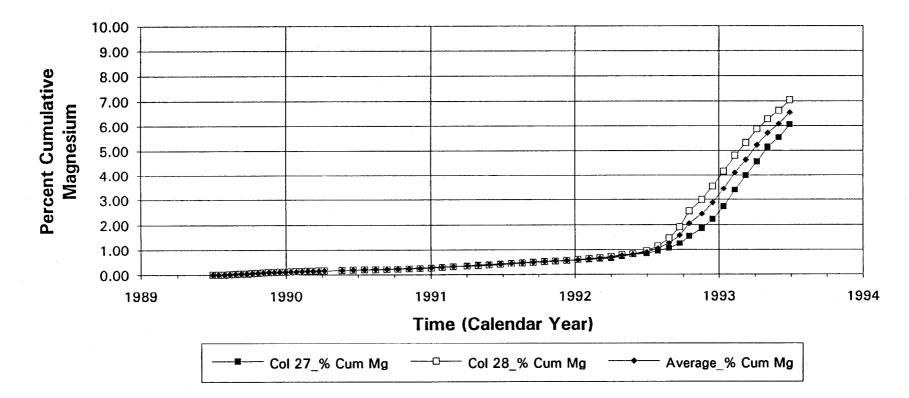
### High Acid Generating Crushed Waste Rock Cumulative Calcium Removal vs. Time

Fig. 242 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Ca removal with time.



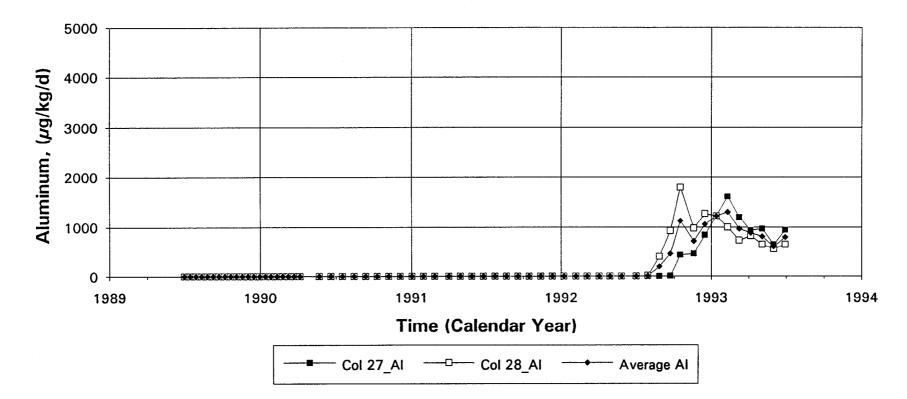
### High Acid Generating Crushed Waste Rock Magnesium Loading vs. Time

Fig. 243 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Mg loading rate with time.



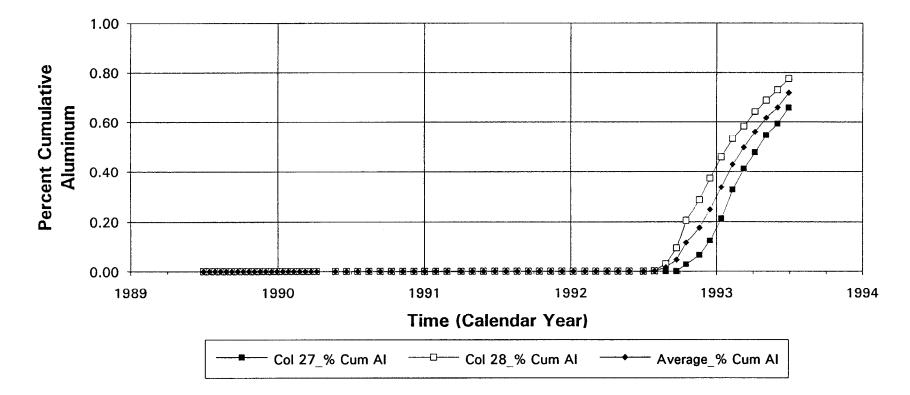
#### High Acid Generating Crushed Waste Rock Cumulative Magnesium Removal vs. Time

Fig. 244 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Mg removal with time.



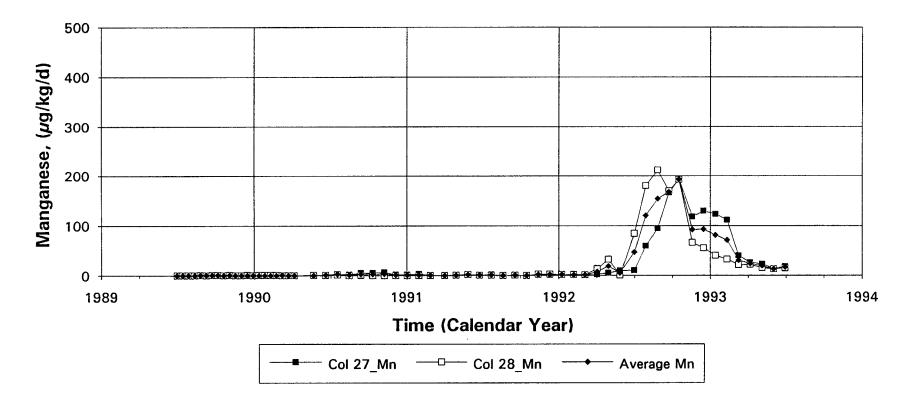
High Acid Generating Crushed Waste Rock Aluminum Loading vs. Time

Fig. 245 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Al loading rate with time.



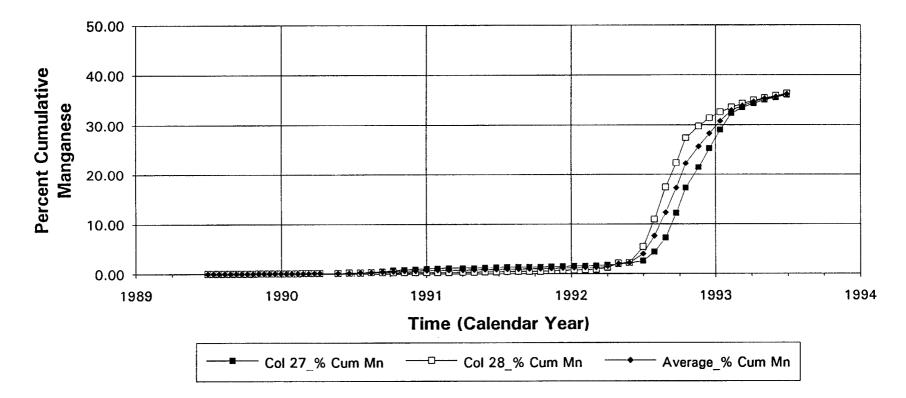
High Acid Generating Crushed Waste Rock Cumulative Aluminum Removal vs. Time

Fig. 246 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Al removal with time.



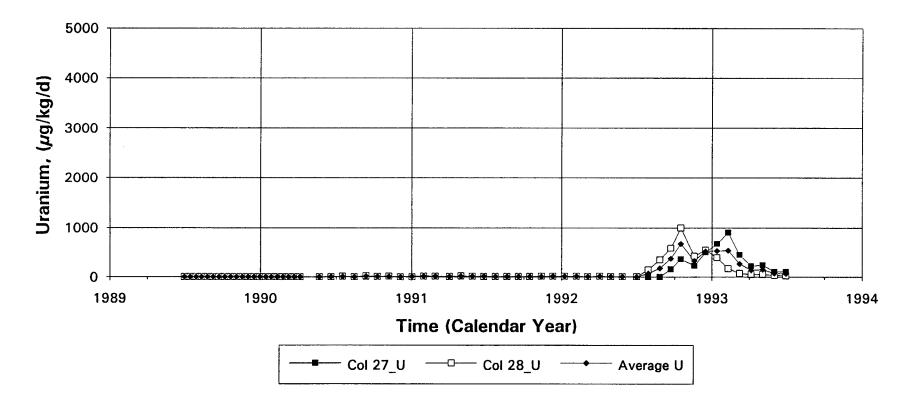
### High Acid Generating Crushed Waste Rock Manganese Loading vs. Time

Fig. 247 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Mn loading rate with time.



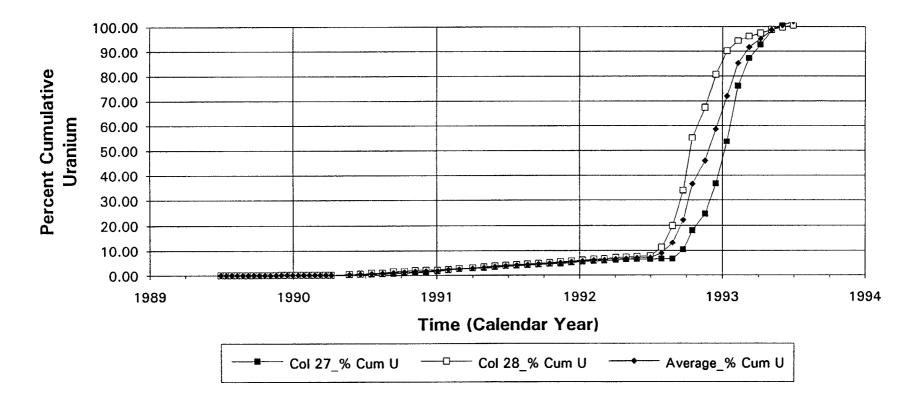
# High Acid Generating Crushed Waste Rock Cumulative Manganese Removal vs. Time

Fig. 248 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Mn removal with time.



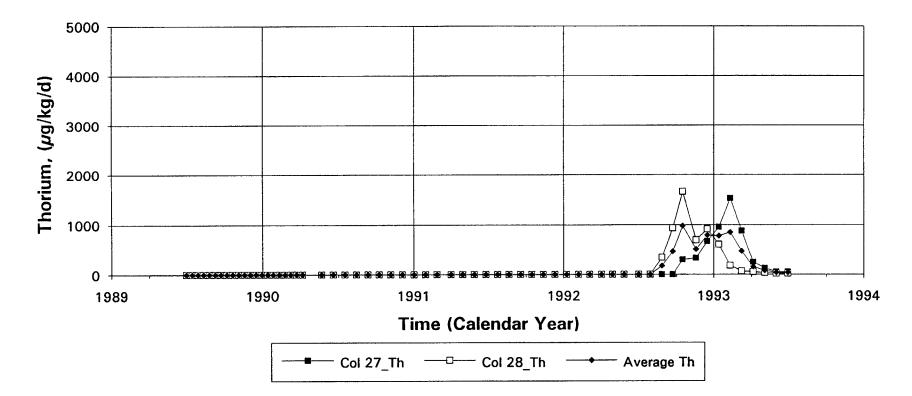
#### High Acid Generating Crushed Waste Rock Uranium Loading vs. Time

Fig. 249 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved U loading rate with time.



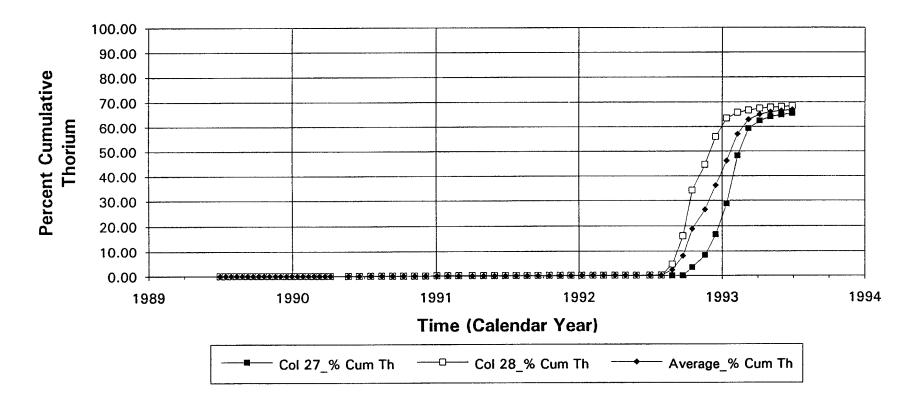
### High Acid Generating Crushed Waste Rock Cumulative Uranium Removal vs. Time

Fig. 250 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative U removal with time.



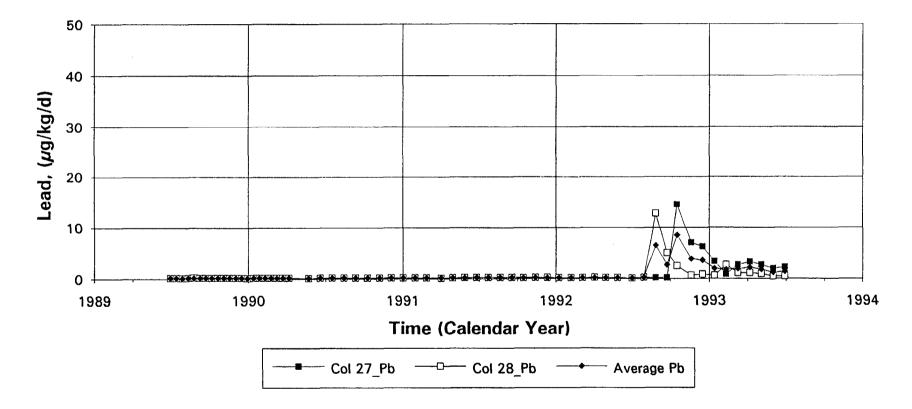
### High Acid Generating Crushed Waste Rock Thorium Loading vs. Time

Fig. 251 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Th loading rate with time.



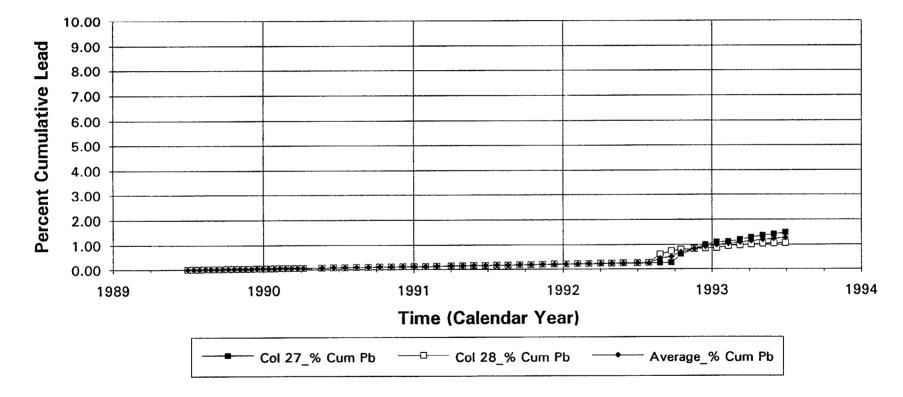
### High Acid Generating Crushed Waste Rock Cumulative Thorium Removal vs. Time

Fig. 252 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Th removal with time.



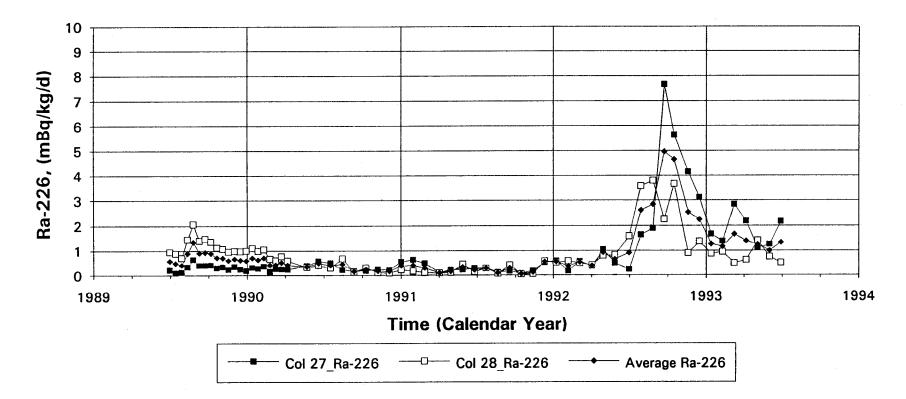
# High Acid Generating Crushed Waste Rock Lead Loading vs. Time

Fig. 253 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Pb loading rate with time.



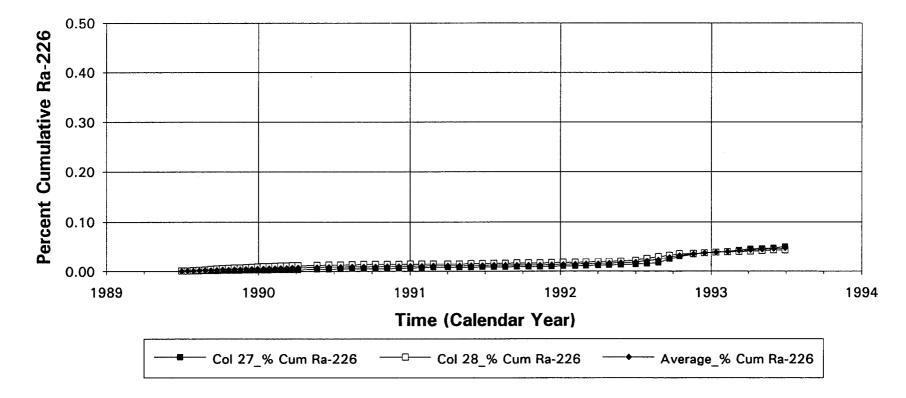
# High Acid Generating Crushed Waste Rock Cumulative Lead Removal vs. Time

Fig 254 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Pb removal with time.



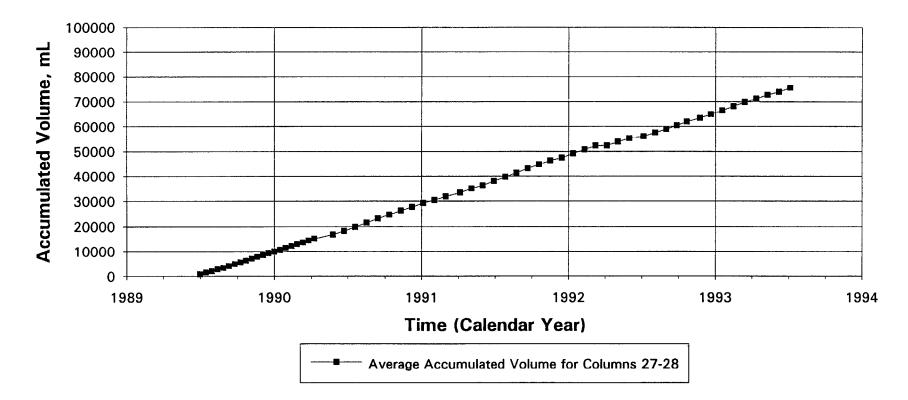
#### High Acid Generating Crushed Waste Rock Ra-226 Loading vs. Time

Fig. 255 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), variation of dissolved Ra-226 loading rate with time.



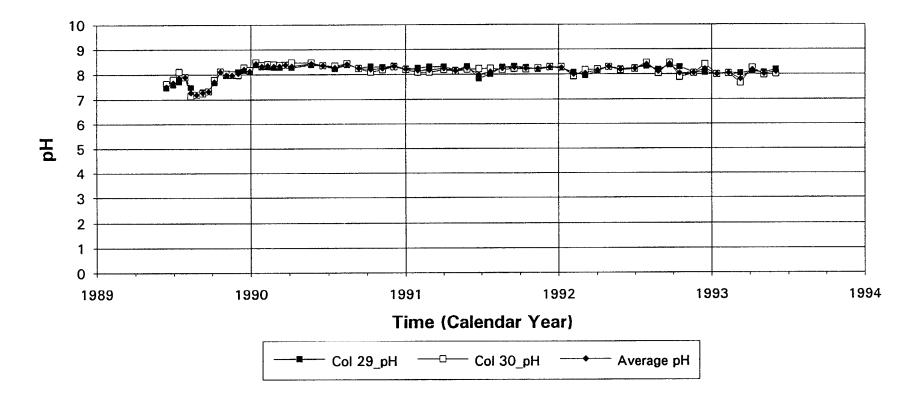
### High Acid Generating Crushed Waste Rock Cumulative Ra-226 Removal vs. Time

Fig. 256 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), cumulative Ra-226 removal with time.



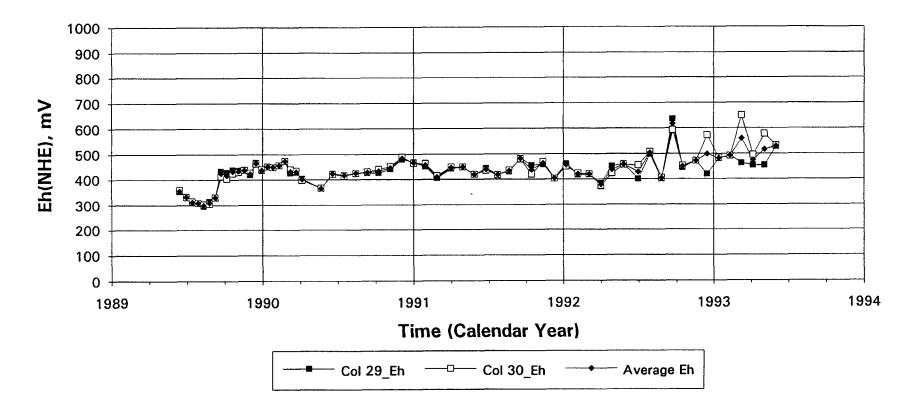
#### High Acid Generating Crushed Waste Rock Accumulated Volume vs. Time

Fig. 257 Leaching profiles of crushed waste rock having high acid generation potential (NNP = -17.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #27 and 28), average cumulative drainage volume collected with time.



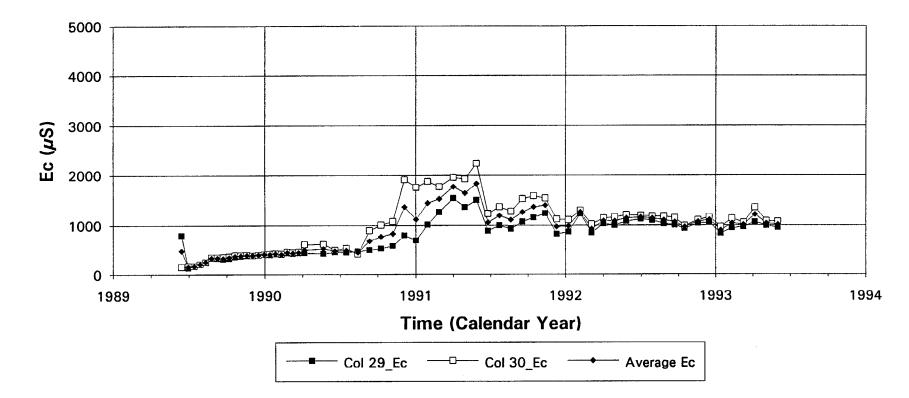
### Low Acid Generating Crushed Waste Rock pH vs. Time

Fig. 258 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of pH with time.



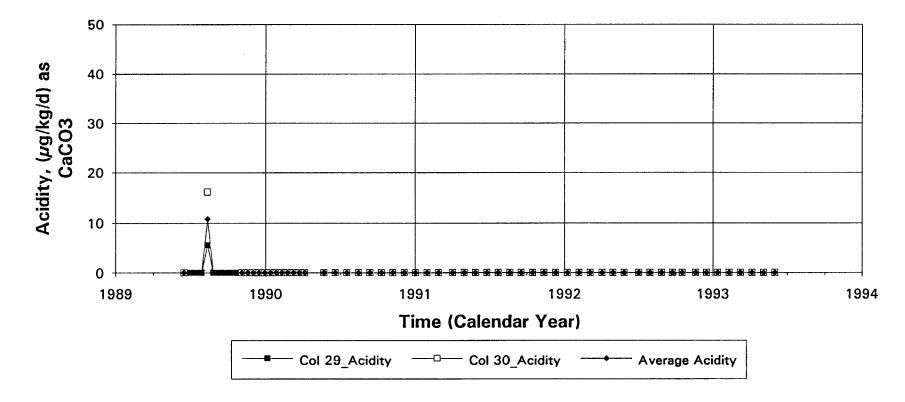
# Low Acid Generating Crushed Waste Rock Eh(NHE) vs. Time

Fig. 259 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of Eh with time.



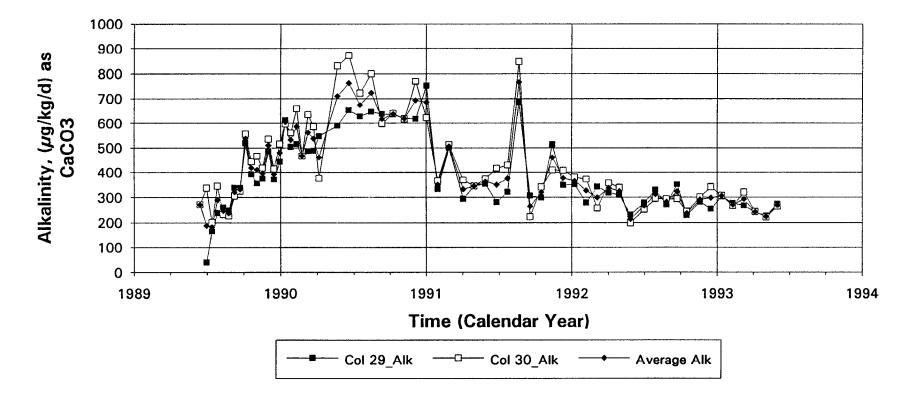
### Low Acid Generating Crushed Waste Rock Electrical Conductivity vs. Time

Fig. 260 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of Ec with time.



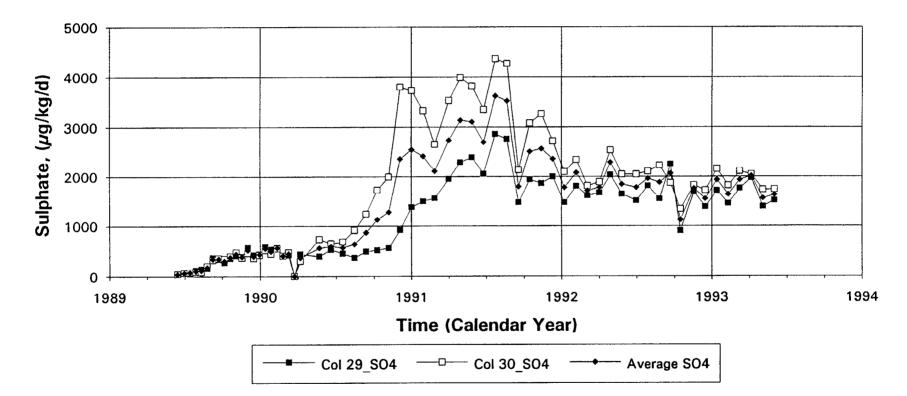
# Low Acid Generating Crushed Waste Rock Acidity Loading vs. Time

Fig. 261 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of total acidity loading rate with time.



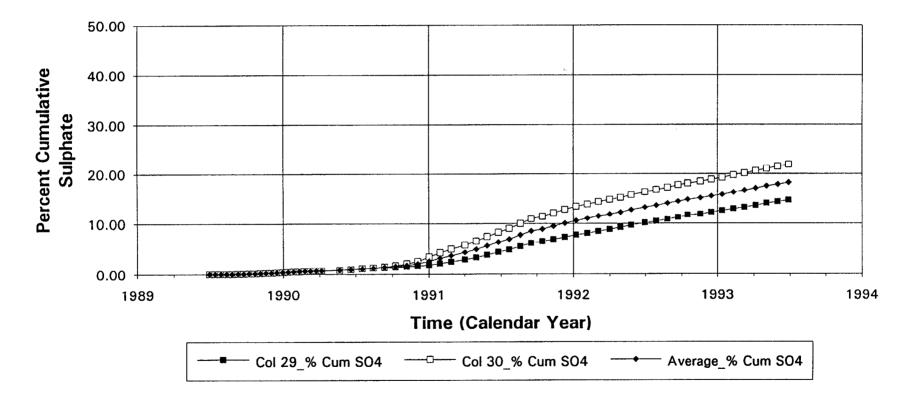
# Low Acid Generating Crushed Waste Rock Alkalinity Loading vs. Time

Fig. 262 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of total alkalinity loading rate with time.



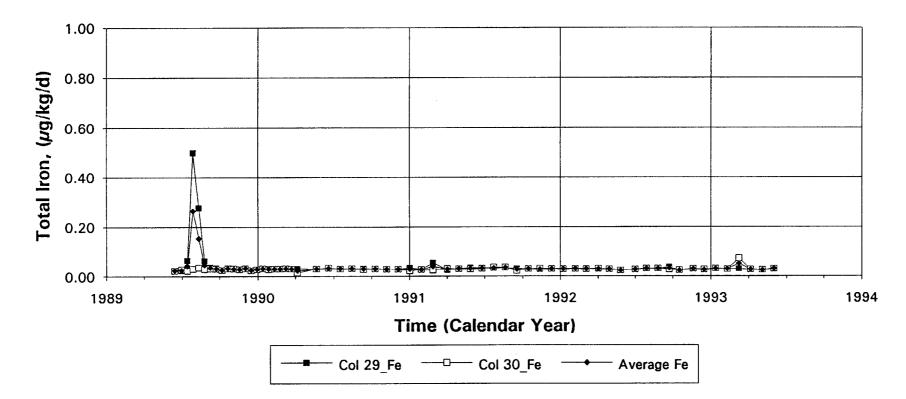
### Low Acid Generating Crushed Waste Rock Sulphate Loading vs. Time

Fig. 263 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved SO<sub>4</sub><sup>-2</sup> loading rate with time.



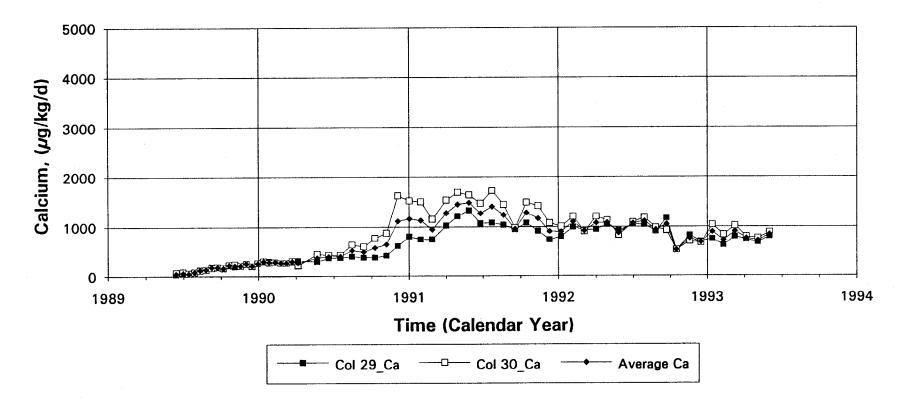
### Low Acid Generating Crushed Waste Rock Cumulative Sulphate Removal vs. Time

Fig. 264 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), cumulative  $SO_4^{-2}$  removal (as % total S) with time.



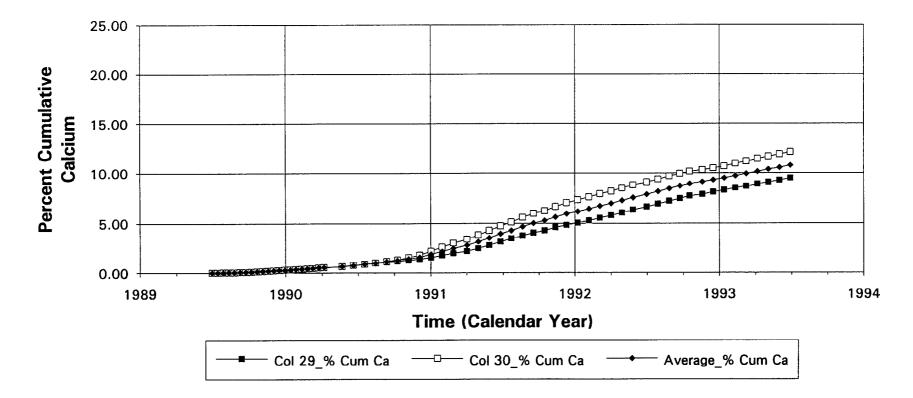
Low Acid Generating Crushed Waste Rock Total Iron Loading vs. Time

Fig. 265 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved total Fe loading rate with time.



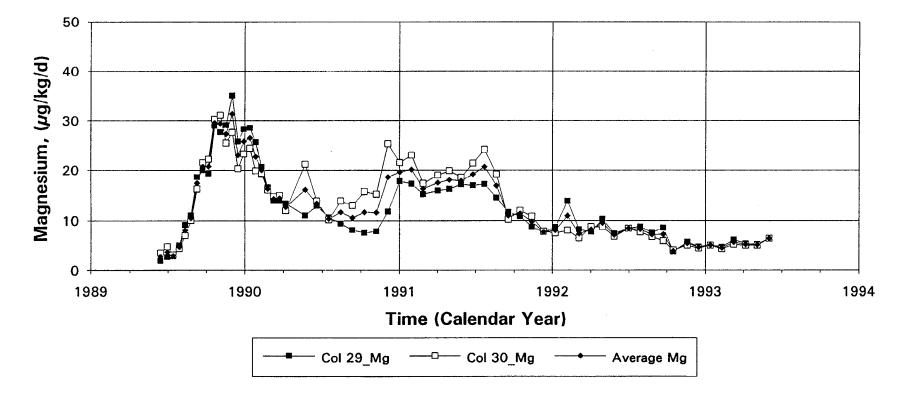
### Low Acid Generating Crushed Waste Rock Calcium Loading vs. Time

Fig. 266 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Ca loading rate with time.



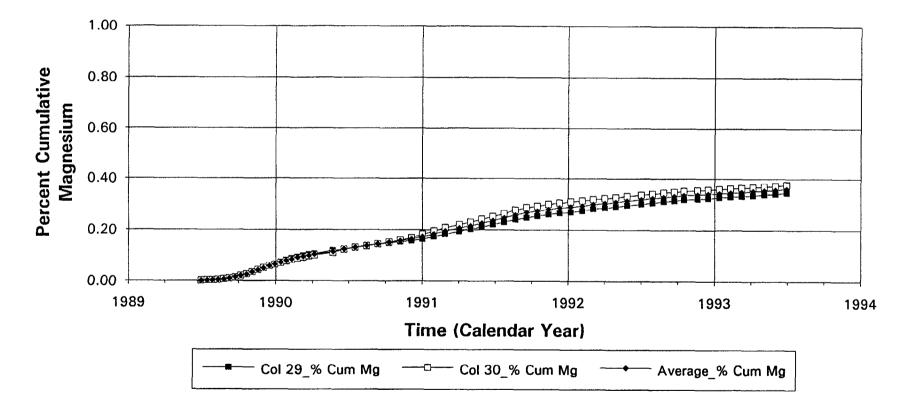
#### Low Acid Generating Crushed Waste Rock Cumulative Calcium Removal vs. Time

Fig. 267 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), cumulative Ca removal with time.



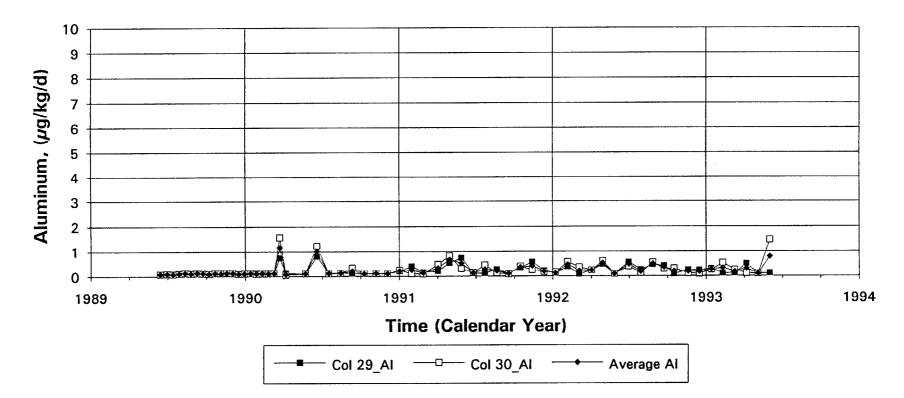
### Low Acid Generating Crushed Waste Rock Magnesium Loading vs. Time

Fig. 268 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Mg loading rate with time.



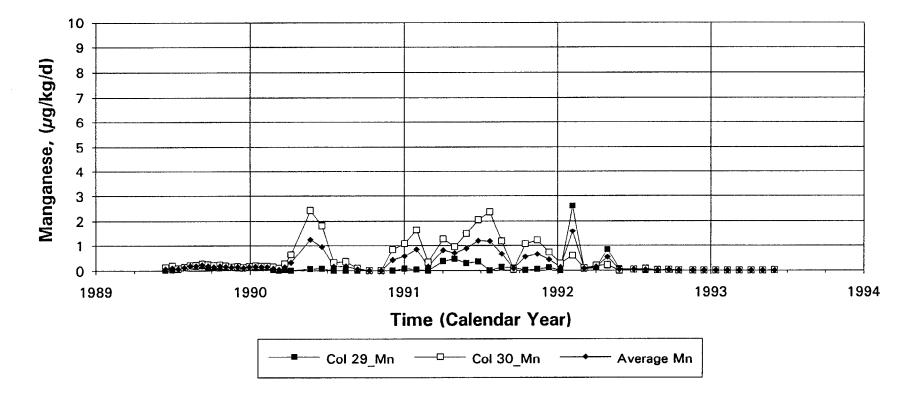
Low Acid Generating Crushed Waste Rock Cumulative Magnesium Removal vs. Time

Fig. 269 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), cumulative Mg removal with time.



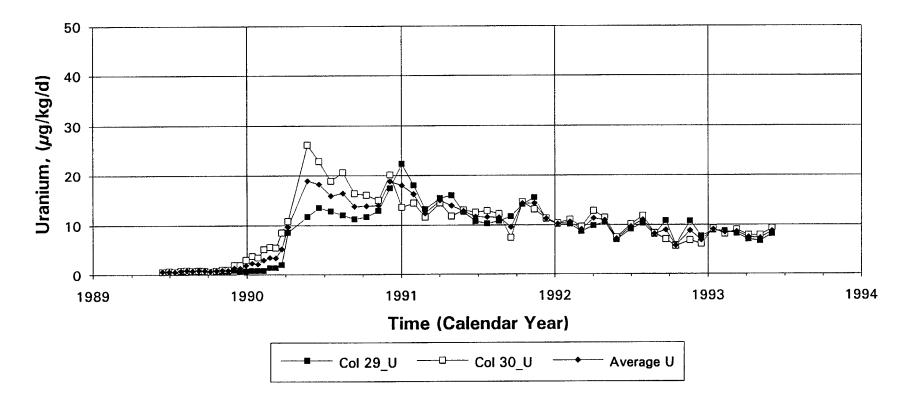
## Low Acid Generating Crushed Waste Rock Aluminum Loading vs. Time

Fig. 270 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Al loading rate with time.



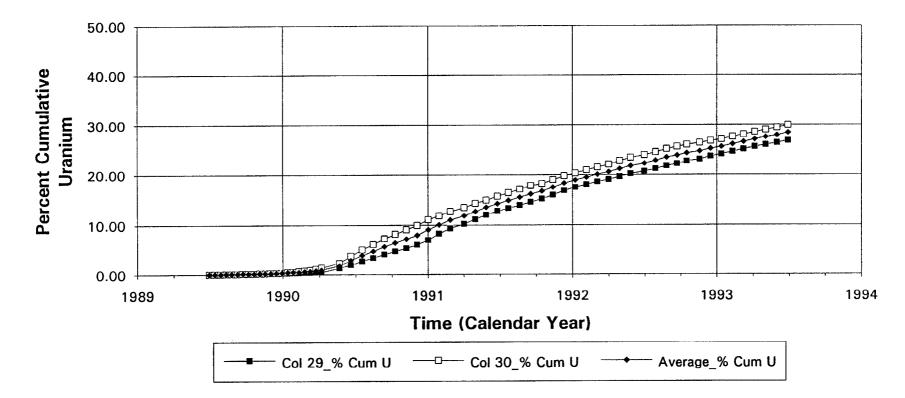
### Low Acid Generating Crushed Waste Rock Manganese Loading vs. Time

Fig. 271 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Mn loading rate with time.



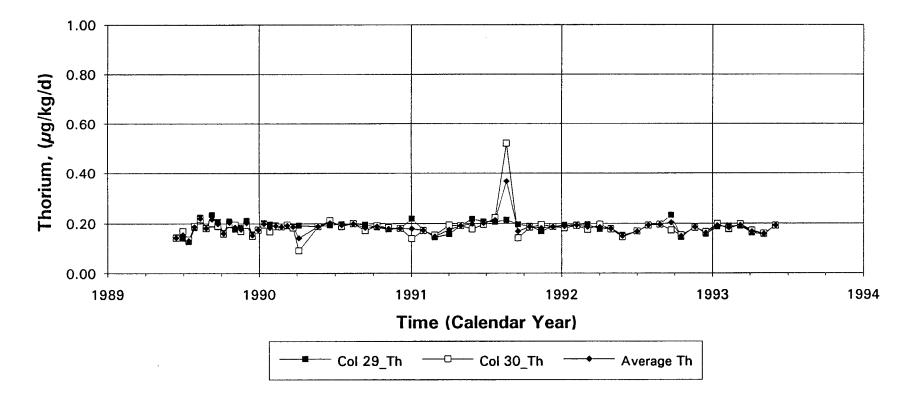
# Low Acid Generating Crushed Waste Rock Uranium Loading vs. Time

Fig. 272 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved U loading rate with time.



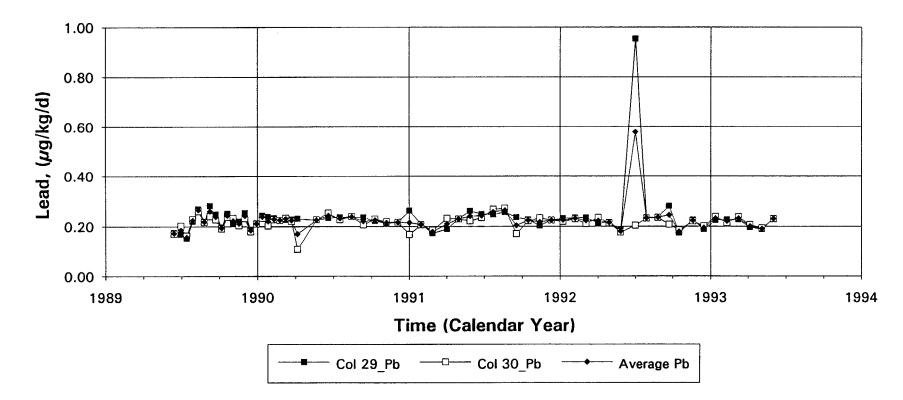
#### Low Acid Generating Crushed Waste Rock Cumulative Uranium Removal vs. Time

Fig. 273 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), cumulative U removal with time.



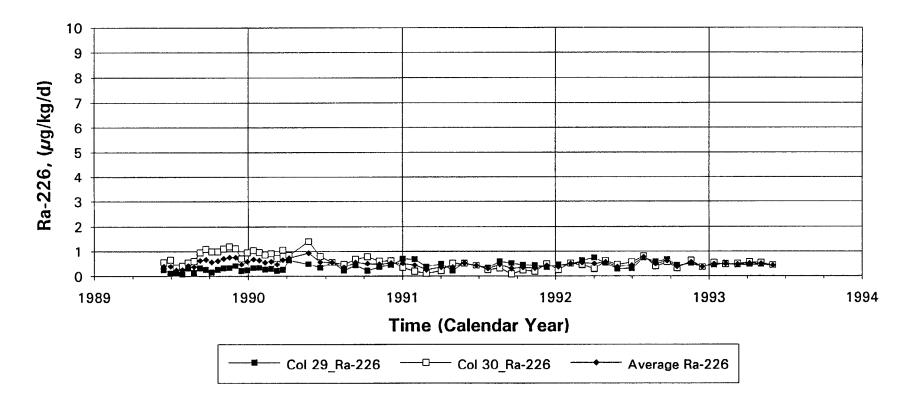
## Low Acid Generating Crushed Waste Rock Thorium Loading vs. Time

Fig. 274 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Th loading rate with time.



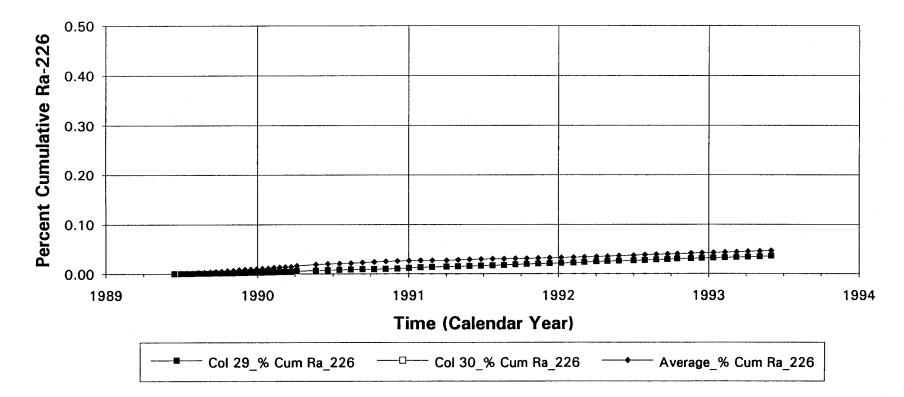
## Low Acid Generating Crushed Waste Rock Lead Loading vs. Time

Fig. 275 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Pb loading rate with time.



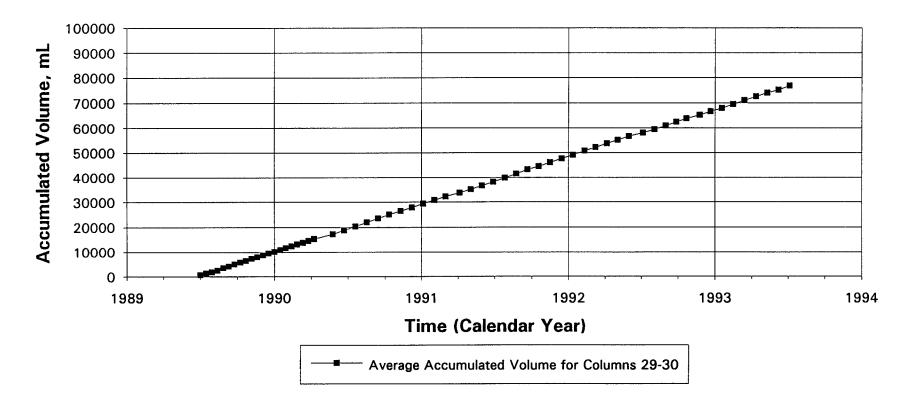
#### Low Acid Generating Crushed Waste Rock Ra-226 Loading vs. Time

Fig. 276 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), variation of dissolved Ra-226 loading rate with time.



Low Acid Generating Crushed Waste Rock Cumulative Radium-226 Removal vs. Time

Fig. 277 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), cumulative Ra-226 removal with time.



#### Low Acid Generating Crushed Waste Rock Accumulated Volume vs. Time

Fig. 278 Leaching profiles of crushed waste rock having low acid generation potential (NNP = +13.8 Kg CaCO<sub>3</sub> / tonne) and under unsaturated conditions (columns #29 and 30), average cumulative drainage volume collected with time.

TABLES

Element/Ra-226	Method	Lowest Limit of Detection (LLD), mg/l
Al	ICAP-AES	0.3
Ba	ICAP-AES	0.001
Ca	ICAP-AES	0.002
Ce	ICAP-AES	0.06
Cu	ICAP-AES	0.04
Fe	ICAP-AES	0.008
K	ICAP-AES	1.2
Mg	ICAP-AES	0.001
Mn	ICAP-AES	0.005
Na	ICAP-AES	0.08
Ni	ICAP-AES	0.02
Р	ICAP-AES	0.02
Pb	ICAP-AES	0.06
Si	ICAP-AES	0.1
Ti	ICAP-AES	0.006
Th	ICAP-AES	0.05
U	ICAP-AES	0.2
Z	ICAP-AES	0.005
S	Radiochemical	0.3
S	ICAP-AES	0.05
Ra-226	Radiochemical	4 mBq/l

 Table 2 - Minimum detection limits for various elements and Ra-226 using Inductively

 Coupled Argon Plasma Spectrophotometer and Radiochemical methods.

Table 3 - Comparison of analytical results for QA/QC Program: Measured and<br/>certified dissolved metal concentrations of Certified Reference<br/>Materials: Digested (Dissolved) River Sediment (CRM-RS-A) and Soil<br/>(CRM-Soil-B) Samples.

Element	Measured Value mg/l	Certified Reference Material Value, mg/l
Al	255	225
Ca	305	300
Fe	1211	1200
Mg	71.5	75
Mn	7.7	8.0
Ni	0.43	0.5
Pb	5.7	7
Zn	15.0	15
Ba*	6.5	7.0
Cu*	2.98	3.0
P*	9.92	10.0
Pb*	59.5	60.0
V*	0.71	0.8

\*Certified Reference Standard CRM-Soil-B solution samples.

Table 4 - Comparison of analytical results for QA/QC Program: Measured and certified<br/>dissolved metal concentrations of a Certified Reference Material: Trace Metal<br/>Drinking Water Sample (CRM-TMDW).

Element	Measured Value µg/l	Reference Material Certified Value, µg/l
Al	112	115
Ca	35380	35000
Fe	101	100
Mg	8700	9000
Mn	34	35
Ni	60	60
Pb	37	35
Zn	68	70
Ba	47	50
Cu	20	20
К	2431	2500
V	33	30

Table 5 - Results for an inter laboratory comparison study for QA/QC Program at the Elliot Lake Laboratory: Analytical results for Certified Reference Materials, Uranium Ore Standard (Soil A, BL-5) and Soil Standard (Soil B, SO-2), Vegetation Standards (CLV-1) and (CLV-2).

		······································	ELRFS (S	oil A) <sup>1</sup>		ELR	RFS and Ca	ANMET (S	Soil B) <sup>2</sup>
Parameters	Unit	Approximate Composition	ELRFS <sup>4</sup>	L.U. <sup>5</sup>	CANMET <sup>3</sup>	Certified Value	ELRFS	L.U.	CANMET
Aluminum (Al)	%	6.00	6.30	6.09	5.99	8.07	8.41	8.01	7.93
Barium (Ba)	%	Unknown	n/a	0.0190	0.0244	Unknown	n/a	0.1090	0.0978
Calcium (Ca)	%	4.00	4.36	3.83	3.97	1.96	2.31	1.95	1.98
Copper (Cu)	%	Unknown	0.019	0.0135	0.0292	0.0007	<0.0004	0.0012	0.0005
Iron (Fe)	%	5.90	5.27	5.66	6.20	5.56	4.74	5.64	5.62
Lead (Pb)	%	1.50	1.12	0.87	1.5309	3.0021	0.3060	0.0016	<0.0140
Magnesium (Mg)	%	1.50	1.13	1.23	0.82	0.54.	0.48	0.50	0.52
Manganese (Mn)	%	0.05	0.048	0.045	0.046	0.072	0.066	0.074	0.065
Nickel (Ni)	%	Unknown	0.007	0.0149	<0.0045	0.0008	0.0010	0.0007	<0.0045
Potassium (K)	%	0.40	0.35	0.37	0.43	2.45	2.29	2.41	2.44
Sodium (Na)	%	3.60	3.25	3.50	3.59	1.90	1.72	1.74	2.03
Zinc (Zn)	%	Unknown	0.010	0.0916	0.0091	0.0124	0.0120	0.0104	0.0124
LOI Ra-226	% mBq/g (CLV-1)	700 <u>+</u> 30	n/a	7.743	n/a 689. <u>+</u> 70	Unknown 38 <u>+</u> 12 (CLV-2)	n/a	14.521	n/a 44.5 <u>+</u> 18

<sup>1</sup>ELRFS (Soil - A) - Canadian Certified Reference Material (BL-5) for Uranium, Ra-226 and Pb-210, Chemical compositions of others elements are taken from (CCRMP 90-1E) and are reported to be approximate.

<sup>2</sup>ELRFS and CANMET (Soil - B) - Canadian Certified Reference Material (SO-2).

<sup>3</sup>CANMET, Elliot Lake Laboratory, <sup>4</sup>Elliot Lake Research Field Station, Elliot Lake, <sup>5</sup>Laurentian University, Sudbury, Ontario.

Column	Material			Permeabi	lity Test		- · · · · · · · · · · · · · · · · · · ·
#	Total Mill	Depth	Volume	Time	Permeability	Average	Standard
	Tailings	cm	ml	S	cm/s	cm/s	Error
1	Control - 1	34.0	811.9	172800	1.04E-05		
1	Control - 1	34.0	396.8	86400	1.02E-05		
2	Control - 1	35.0	739.8	172800	9.78E-06	8.67E-06	21.71
2 2 3	Control - 1	35.0	360.5	86400	9.53E-06		
	Control - 1	35.0	225.3	86400	5.96E-06	-	
3	Control - 1	35.0	462.9	172800	6.12E-06		
4	T + L, -6.3mm	36.0	719.8	172800	9.79E-06		
4	T + L, -6.3mm	36.0	354.6	86400	9.64E-06		
5	T + L, -6.3mm	34.5	238.8	86400	6.22E-06	7.66E-06	19.38%
5	T + L, -6.3mm	34.5	485.1	172800	6.32E-06		
6	T + L, -6.3mm	36.0	253.1	86400	6.88E-06		
6	T + L, -6.3mm	36.0	522.4	172800	7.10E-06		
7	T + L, -2.4mm	34.5	249.4	86400	6.50E-06		
7	T + L, -2.4mm	34.5	468.8	172800	6.11E-06		
8	T + L, -2.4mm	34.0	451.6	172800	5.80E-06	6.25E-06	11.18%
8	T + L, -2.4mm	34.0	196.5	86400	5.05E-06		
9	T + L, -2.4mm	34.5	268.2	86400	6.99E-06	l [	
9	T + L, -2.4mm	34.5	541.4	172800	7.05E-06		
10	T + L, -0.85mm	35.0	430.5	172800	5.69E-06		
10	T + L, -0.85mm	35.0	203.9	86400	5.39E-06		
11	T + L, -0.85mm	36.0	583.2	172800	7.93E-06	6.73E-06	14.31%
11	T + L, -0.85mm	36.0	287.8	86400	7.83E-06		
12	T + L, -0.85mm	35.0	522.1	172800	6.90E-06		
12	T + L, -0.85mm	35.0	250.2	86400	6.61E-06		
13	T + WGL	37.0	195.9	86400	5.47E-06	1	
13	T + WGL	37.0	432.9	172800	6.05E-06		
14	T + WGL	37.0	402.5	172800	5.62E-06	5.85E-06	7.05%
14	T + WGL	37.0	189.2	86400	5.29E-06		
15	15 T + WGL		222.5	86400	6.39E-06		
15	T + WGL	38.0	437.6	172800	6.28E-06	$03E-06+22^{4}$	

Table 6 -Column permeability (saturated hydraulic conductivity) results for Group 1 - UnclassifiedTotal Mill Tailings obtained from the Quirke mill, Columns #1-15.

Overall Mean:

 $7.03E-06 \pm 22.5\%$ 

T = Tailings

L = Limestone, Screen Size

WGL = Wet Ground Limestone

Table 7 - Column permeability (saturated hydraulic conductivity) results for Group 2-<br/>Coarse Tailings obtained from the Quirke Waste Management Area, West Arm<br/>Tailings Basin, Columns #16-21.

Column	Material		······	Perr	neability Test		<u> </u>
#	Coarse	Depth	Volume	Time	Permeability	Average	Standard
	Tailings	cm	ml	S	cm/s	cm/s	Error
16	Control - 2	62.0	5270.6	5220	4.09E-03		
16	Control - 2	62.0	1845.9	1920	3.89E-03		
16	Control - 2	62.0	3513.2	3720	3.82E-03	4.21E-03	6.94%
17	Control - 2	59.9	3481.7	3060	4.45E-03		
17	Control - 2	59.9	5294.2	4560	4.54E-03		
17	Control - 2	59.9	1723.4	1500	4.49E-03		
18	T + L, -6.3mm	60.7	1809.3	2160	3.32E-03		
18	T + L, -6.3mm	60.7	5522.7	6540	3.35E-03		
18	T + L, -6.3mm	60.7	3674.0	4320	3.37E-03	3.12E-03	7.05%
19	T + L, -6.3mm	58.5	3499.4	4560	2.93E-03		
19	T + L, -6.3mm	58.5	1720.7	2280	2.88E-03		
19	T + L, -6.3mm	58.5	5295.4	6960	2.90E-03		
20	T + WGL	61.2	3449.5	5160	2.67E-03		
20	T + WGL	61.2	1763.2	2400	2.93E-03		
20	T + WGL	61.2	5174.3	7620	2.71E-03	2.94E-03	6.99%
21	T + WGL	69.1	1740.9	2400	3.27E-03		
21	T + WGL	69.1	3447.8	5100	3.05E-03		
21	T + WGL	69.1	5196.8	7740	3.03E-03		

Overall Mean

3.43E-03 ±17.3%

T = Tailings L = Limestone, Screen Size WGL = Wet Ground (Pulverized) Limestone Table 8 - Column permeability (saturated hydraulic conductivity) results for Group 3-<br/>Submerged Coarse Tailings, obtained from the Quirke Waste Management Area<br/>West Arm Tailings Basin, Columns #22-24.

Column	Condition			Per	meability Test		·····
#	Coarse	Depth	Volume	Time	Permeability	Average	Standard
	Tailings	cm	ml	s	cm/s	cm./s	Error
22	Submerged	55.5	3317.5	2520	4.77E-03		
22	Submerged	55.5	3309.7	2700	4.44E-03		
22	Submerged	55.5	1576.1	1200	4.76E-03		
22	Submerged	55.5	5414.7	4620	4.25E-03		
22	Submerged	55.5	1692.5	1320	4.64E-03	4.55E-03	4.75%
22	Submerged	55.5	5383.5	4080	4.78E-03		
22	Submerged	55.5	1706.8	1440	4.29E-03		
22	Submerged	55.5	4875.9	3720	4.75E-03		
22	Submerged	55.5	3566.9	3000	4.31E-03		
23	Submerged	55.0	3586.5	2940	4.38E-03		
23	Submerged	55.0	1820.9	1440	4.54E-03		
23	Submerged	55.0	5366.2	4380	4.40E-03		
23	Submerged	55.0	1685.0	1440	4.20E-03		
23	Submerged	55.0	3280.4	2100	5.61E-03	4.70E-03	10.79%
23	Submerged	55.0	1603.5	1200	4.80E-03		
23	Submerged	55.0	3364.7	2880	4.19E-03		
23	Submerged	55.0	5023.5	3240	5.57E-03		
23	Submerged	55.0	5636.3	4380	4.62E-03		
24	Submerged	55.0	3248.4	2880	4.05E-03		
24	Submerged	55.0	5081.5	4620	3.95E-03		
24	Submerged	55.0	1601.1	1080	5.32E-03		
24	Submerged	55.0	1656.9	1500	3.96E-03		
24	Submerged	55.0	1675.8	1500	4.01E-03	4.30E-03	10.88%
24	Submerged	55.0	5063.7	4440	4.09E-03		
24	Submerged	55.0	3304.8	2520	4.71E-03		
24	Submerged	55.0	3374.4	3120	3.88E-03		
24	Submerged	55.0	4870.1	3720	4.70E-03		

Overall Mean

 $4.52\text{E-}03\pm10\%$ 

Table 9 -	· Colum	in perm	neability	(satur	ated	hydrau	ılic c	onductiv	vity) resul	ts for	Grou	ıp 4 - C	rushed
	Waste	Rock	(screen	size	-2.0	mm,	-10	mesh)	obtained	from	the	Quirke	Waste
	Manag	ement .	Area, We	est Ari	n Tai	lings H	Basin	(Interna	ll Dyke Ce	:11 #14)	, Col	umns # 2	25-30.

Column	Material			Peri	neability Test			
#	Waste	Depth	Volume	Time	Permeability	Average	Standard	
	Rock	cm	ml	s	cm/s	cm/s	Error	
			Medium	Acid Prod	ucing		*	
25	QWR-A	50.0	1752.0	3000	1.91E-03			
25	QWR-A	50.0	3496.0	5640	2.02E-03			
25	QWR-A	50.0	5574.0	8520	2.13E-03	3.34E-03	39.88%	
26	QWR-A	53.6	1782.0	1260	4.95E-03			
26	QWR-A	53.6	3556.0	2820	4.41E-03			
26	QWR-A	53.6	5334.0	4020	4.64E-03			
	• • • • • • • • • • • • • • • • • • •		High Ac	cid Produc	ing	•	•	
27	QWR-B	50.2	1918.0	2820	2.23E-03			
27	QWR-B	50.2	3744.9	5460	2.25E-03	2.83E-03	36.11%	
27	QWR-B	50.2	5798.7	8220	2.31E-03			
28	QWR-B	55.5	1692.5	1320	4.64E-03			
			Low Ac	id Produc	ing	· · · · · · · · · · · · · · · · · · ·	<u></u>	
29	QWR-C	55.0	1361.5	5460	8.95E-04			
29	QWR-C	55.0	2688.1	10800	8.93E-04			
29	QWR-C	55.0	4130.4	16680	8.89E-04	7.97E-04	11.98%	
30	QWR-C	55.0	1064.8	5460	7.00E-04			
30	QWR-C	55.0	2482.7	12660	7.04E-04			
30	QWR-C	55.0	3842.5	19680	7.01E-04			

Overall Mean

2.27E-03 ± 66.4%

 Table 10 - Analysis of variance of column permeability results between control tailings and limed tailings sets for total mill tailings (Group 1) and coarse tailings (Group 2).

	Control Column #	Limed Column #	Calculated F - Test Value	df(1)	df(2)	Tabulated F - Ratio	Significant Difference at 5% Level
Group - 1 Total Mill Tailings	1 to 3	4 to 15	10.25	1	28	4.2	Yes
Group - 2 Coarse Tailings	16 to 17	18 to 21	76.84	1	16	4.49	Yes

#### F - Ratio Comparison at 5% Level of Significance

F - Ratio Comparison at 10% Level of Significance

	Control Column #	Limed Column #	Calculated F - Test Value	df(1)	df(2)	Tabulated F - Ratio	Significant Difference at 10% Level
Group - 1 Total Mill Tailings	1 to 3	4 to 15	10.25	1	28	7.64	Marginal
Group - 2 Coarse Tailings	16 to 17	18 to 21	76.84	1	16	8.53	Yes

df = degree of freedom

3-9

Table 11 - Analysis of variance of column permeability results within columns of the same group for submerged coarse tailings (Group 3) and crushed waste rock (Group 4).

Group # Material	Column #	Calculated F - Test Value	df(1)	df(2)	Tabulated F - Ratio	Significant Difference at 5% Level
Group - 3 Submerged Coarse Tailings	22 to 24	1.91	2	24	3.40	No
Group 4 Crushed Waste Rock	25 to 30	9.25	2	13	3.81	Yes

#### F - Ratio Comparison at 5% Level of Significance

F - Ratio Comparison at 10% Level of Significance

Group # Material	Column #	Calculated F - Test Value	df(1)	df(2)	Tabulated F - Ratio	Significant Difference at 10% Level
Group 4 Crushed Waste Rock	25 to 30	9.25	2	13	6.70	Marginal

df = degree of freedom

Element / Parameter	Measured Concentration	Standard Deviation
Al	2.89	0.03
As, (µg/g)	<50	-
Ba	0.103	.0008
Ca	1.71	0.01
Ce	0.08	0.001
Cu, (µg/g)	59	5
Fe	2.49	0.03
К	2.03	0.07
Mg	0.085	0.001
Mn, (μg/g)	24	0.6
Na	0.075	0.003
Ni, (µg/g)	<20	-
P, (µg/g)	<30	-
Pb	0.035	0.001
Sb, (µg/g)	<30	-
Si	34.82	-
Th	0.031	0.006
Ti	0.23	0.004
U	< 0.003	-
Zn (μg/g)	32	1.0
S - Soluble, (%)	1.31	-
S - Total, (%)	3.48	-
S - sulphide, (%)	2.17	-
Ra-226, (mBq/g)	7506	150
Total Acid Generation	67.81	-
Potential, kg CaCO <sub>3</sub> /tonne		
Total Neutralization	6.40	-

- 61.41

Potential, kg CaCO<sub>3</sub>/tonne

Potential, kg CaCO<sub>3</sub>/tonne

Net Neutralization

Table 12 - Solid phase compositions of total mill tailings without additional limestone, Control - 1, (columns #1 - 3). All values are in (%), unless noted otherwise.

Element/Parameter	Measured Concentration	Standard Deviation
Al	2.89	0.1
As, (μg/g)	315	26
Ba	0.095	0.004
Ca	4.91	0.3
Ce	0.082	0.001
Cu, (µg/g)	45	3
Fe	2.46	0.1
K	2.24	0.18
Mg	0.113	0.006
Mn, (μg/g)	25	2
Na	0.07	0.01
Ni, (µg/g)	26	11
P, (µg/g)	464	40
Pb	0.024	0.001
Sb, (µg/g)	-	-
Si	32.51	1.0
Th	0.05	0.003
Ti	0.21	0.01
U	<0.004	-
$Zn, (\mu g/g)$	29	5
S - Soluble, (%)	1.02	-
S - Total, (%)	3.0	0.13
S - Total Sulphide, (%)	2.0	-
Ra-226, (mBq/g)	7577	326
Total Acid Generation	61.9	-
Potential, kg CaCO <sub>3</sub> /tonne		
Total Neutralization Potential, kg CaCO <sub>3</sub> /tonne	82.32	-
Net Neutralization Potential, kg CaCO <sub>3</sub> /tonne	+20.44	-

Table 13 - Solid phase compositions of total mill tailings mixed with 7.5% (w/w) limestone, columns #4-15. All values are in (%), unless noted otherwise.

Table 14 - Solid phase compositions of coarse tailings without limestone, Control - 1 (columns #16 and 17), and submerged coarse tailings (columns #22-24). All values are in (%), unless noted otherwise.

Element/Parameter	Measured	Standard
	Concentration	Deviation
Al	2.60	0.2
As	-	-
Ba	0.078	0.0001
Ca	0.4	0.03
Ce	0.076	0.002
Cu, ((µg/g))	27	2
Fe	3.99	0.08
K	1.8	0.007
Mg	0.06	0.001
Mn, (μg/g)	29	2
Na	0.078	0.001
Ni, (µg/g)	28	5
P, $(\mu g/g)$	-	-
Pb	0.021	0.0004
Sb, (µg/g)	-	-
Si	36.6	-
Th	0.02	0.0006
Ti	0.24	0.007
U	< 0.004	-
Zn	21	2
S - Soluble (%)	0.14	-
S - Total (%)	4.36	-
S - Total Sulphide (%)	4.22	-
Ra-226, (mBq/g)	3899	260
Total Acid Generation	131.9	-
Potential, kg CaCO <sub>3</sub> /tonne		
Total Neutralization	2.7	-
Potential, kg CaCO <sub>3</sub> /tonne		
Net Neutralization Potential, kg CaCO <sub>3</sub> /tonne	-129.2	-

Element/Parameter	Measured Concentration	Standard Deviation
Al	2.59	0.2
As	-	-
Ba	0.076	0.0001
Ca	3.50	0.03
Ce	0.074	0.002
Cu, (µg/g)	25	1
Fe	3.99	0.08
K	1.58	0.007
Mg	0.064	0.0007
Mn, (μg/g)	9.3	0.7
Na	0.074	0.0008
Ni, (μg/g)	29	5
P, $(\mu g/g)$	-	-
Pb	0.025	0.0002
Sb, (μg/g)	-	-
Si	35.08	1.3
Th	0.019	0.0006
Ti	0.219	0.007
U	< 0.004	-
Zn, (µg/g)	16	2
S - Soluble, (%)	0.117	-
S - Total, (%)	3.92	-
S - Total Sulphide, (%)	3.80	-
Ra-226, (mBq/g)	3693	200
Total Acid Generation Potential, kg CaCO <sub>3</sub> /tonne	118.75	-
Total Neutralization Potential, kg CaCO <sub>3</sub> /tonne	75.9	-
Net Neutralization Potential, kg CaCO <sub>3</sub> /tonne	- 42.8	

Table 15 - Solid phase compositions of coarse tailings mixed with 7.5% (w/w) limestone (columns #18-21). All values are in (%), unless noted otherwise.

Table 16 - Solid phase composition of Quirke Mine waste rock (QWR-A), medium acid generation potential, (columns #25 and 26). All values are in (%), unless noted otherwise.

Element/Parameter	Measured	Standard
·	Concentration	Deviation
Al	4.12	0.04
As, (μg/g)	-	-
Ba	0.175	0.003
Ca	0.47	0.01
Ce	0.039	0.001
Cu, (µg/g)	23	6
Fe	1.91	0.03
K	-	-
Mg	0.166	0.002
Mn, (µg/g)	152	1.5
Na	0.22	0.005
Ni, (μg/g)	-	-
P, (µg/g)	-	-
Pb	0.008	0.0006
Sb, (µg/g)	-	-
Si	38.03	-
Th	0.025	0.01
Ti	0.267	0.003
U	0.008	0.003
Zn, (µg/g)	27	1.0
S - Soluble, (%)	0.02	-
S - Total, (%)	0.82	0.23
S - Total Sulphide, (%)	0.80	-
-		
Ra,-226 (mBq/g)	2758	128
Total Acid Generation	25.17	-
Potential, kg CaCO <sub>3</sub> /tonne		
-		
Total Neutralization	15.06	-
Potential, kg CaCO <sub>3</sub> /tonne		
Net Neutralization	-10.7	_
Potential, kg CaCO <sub>3</sub> /tonne	10.7	
a otomina, kg CaCO3/tomine	<u> </u>	

Table 17 - Solid phase composition of Quirke Mine waste rock (QWR - B), high acid generation potential (columns #27 and 28). All values are in (%), unless noted otherwise.

Element/Parameter	Measured	Standard	
	Concentration	Deviation	
Al	3.94	0.1	
As, (μg/g)	-	-	
Ba	0.207	0.01	
Ca	0.265	0.0005	
Ce	0.037	0.001	
Cu, (µg/g)	18.7	1.5	
Fe	1.46	0.07	
К	2.62	0.15	
Mg	0.123	0.004	
Mn, (μg/g)	92.0	3	
Na	0.133	0.003	
Ni, (μg/g)	-	-	
P, (µg/g)	-	-	
Pb	0.01	0.001	
Sb, (µg/g)	-	-	
Si	38.41	-	
Th	0.022	0.006	
Ti	0.161	0.004	
U	0.0113	0.0008	
Zn, (μg/g)	18	0.3	
S - Soluble, (%)	0.02	-	
S - Total, (%)	0.86	0.07	
S - Total Sulphide, (%)	0.84	-	
Ra-226, (mBq/g)	2769	123	
Total Acid Generation	26.34	-	
Potential, kg CaCO <sub>3</sub> /tonne			
Total Neutralization	8.5	-	
Potential, kg CaCO <sub>3</sub> /tonne			
Net Neutralization	-17.8	-	
Potential, kg CaCO <sub>3</sub> /tonne			

Table 18 - Solid phase composition of Quirke Mine waste rock (QWR-C), low acid generation potential, (columns #29-30). All values are in (%), unless noted otherwise.

Element/Parameter	Measured	Standard
	Concentration	Deviation
Al	5.04	0.04
As, (µg/g)	-	-
Ba	0.159	0.003
Ca	1.035	0.02
Ce	0.0282	.002
Cu, (µg/g)	45.0	0.1
Fe	3.80	0.02
K	2.39	0.05
Mg	0.505	0.001
Mn, (µg/g)	489	5
Na	0.70	0.002
Ni, (µg/g)	-	-
P, (µg/g)	-	-
Pb	0.006	0.0006
Sb, (µg/g)	-	-
Si	34.32	-
Th	0.01	0.0001
Ti	0.425	0.004
U	0.005	0.001
Zn, (μg/g)	94.5	2
S - Soluble, (%)	0.01	-
S - Total, (%)	0.42	0.16
S - Total Sulphide, (%)	0.41	-
		-
Ra-226, (mBq/g)	1750	110
Total Acid Generation	13.01	-
Potential, kg CaCO <sub>3</sub> /tonne		
Total Neutralization	26.82	-
Potential, kg CaCO <sub>3</sub> /tonne		
Net Neutralization	+13.81	-
Potential, kg CaCO <sub>3</sub> /tonne		