LEACHABILITY OF RADIOACTIVE CONSTITUENTS FROM URANIUM MINE TAILINGS
STATUS REPORT
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by

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ABSTRACT

A project using lysimeters to determine the leaching of radioactive constituents from abandoned uranium mine tailings, and $BaRaSO_4$ sediments from these tailings was initiated at the Wastewater Technology Centre, Burlington, Ontario.

Lime addition to the surface of acidic abandoned tailings did not reduce the level of radioactive constituents in the leachate. Considerable increases in levels of the radionuclides ^{230}Th, ^{232}Th and ^{228}Th, as well as gross alpha and beta activity in the leachate, occurred five months after recycling of $BaRaSO_4$ sediments to the surface layers of abandoned tailings. After nine months of leaching, the levels of ^{226}Ra in the leachate were 30% greater from the tailings + sediment treatment than from tailings only (control).

The project was revised by increasing the high water application rate from 82 mm/month to a continuous flow of about 460 mm/month. From data obtained to date, recycling of $BaRaSO_4$ sediments resulting from the treatment of acid seepage from abandoned tailings would appear to be undesirable because high thorium and other radioactive constituents in drainage water would require continuous treatment indefinitely.

Another experiment compared the quality of effluent flowing over chemically-fixed (solidified) $BaRaSO_4$ sediments with that of non-fixed sediments (control) in simulated sedimentation ponds.

During seven months of monitoring, the release of ^{226}Ra to water from chemically-fixed $BaRaSO_4$ sediments remained <1 pCi/L, compared to $\sim$7 pCi/L from non-chemically-fixed sediments.

A further experiment compared the leaching of radioactive constituents from chemically-fixed fresh tailings (with and without vegetation) to non-fixed (control) tailings. Air-dried digested sewage sludge (with $FeCl_3$ for phosphorus removal) was applied to supply 3% organic matter in the top 15 cm of the revegetated lysimeters.

Chemical fixation of fresh tailings reduced the ^{226}Ra leachate levels from a high of 85 pCi/L for the control to less than 1 pCi/L.

Undiluted effluent and leachate from chemically-fixed $BaRaSO_4$ sediments (Experiment 2) and fresh tailings (Experiment 3) were 100% lethal to Daphnia pulex and rainbow trout (Salmo gairdneri) in static 96-hour bioassay tests. Diluted (50%) effluent samples were non-toxic.

RÉSUMÉ

On a étudié, à l'aide de lysimètres, la percolation des constituants radioactifs des stériles d'une mine d'uranium abandonnée et des dépôts de $BaRaSO_4$ de ces mêmes stériles. Cette étude s'est effectuée au Centre technique des eaux usées de Burlington, en Ontario.

L'épandage de chaux à la surface de stériles acides n'a pas réduit la radioactivité du percolat. Cinq mois après le recyclage des dépôts de $BaRaSO_4$ à la surface des stériles, on a observé une augmentation considérable de la teneur en radionuclides Th-230, Th-232 et Th-228 ainsi qu'une activité alpha et bêta d'ensemble dans le percolat. Après neuf mois de percolation, la concentration de Ra-226 dans le percolat des stériles et des sédiments était de 30 p.100 supérieure à celle des stériles de contrôle.

On a modifié l'expérience en substituant à l'addition importante d'eau (81 mm) un débit continu et encore plus élevé de 460 mm/mois. Les résultats donnent à penser que le recyclage des sédiments de $BaRaSO_4$ provenant du traitement du percolat acide des stériles abandonnés n'est pas souhaitable. La raison en est que la forte teneur en thorium et autres constituants radioactifs des eaux d'égouttement exigerait un traitement ininterrompu et indefini.

Dans une autre expérience, on a comparé la qualité respective des effluents s'écoulant sur des sédiments de $BaRaSO_4$ chimiquement liés (solidifiés) et sur des sédiments de contrôle non liés dans des bassins simulés de décantation. Pendant les sept mois de contrôle, la quantité de Ra-226 libérée dans l'eau par les sédiments liés est démeurée inférieure à 1 pCi/L, comparativement à environ 7 pCi/L pour les non liés.

Une expérience supplémentaire a permis de comparer la percolation des constituants radioactifs provenant des stériles récents, non liés chimiquement (en presence ou en l'absence de végétation), et des stériles de contrôle non liés chimiquement. Des boues digérées et séchées à l'air (additionnées de $FeCl_3$ pour la déphosphatation) ont été afoutées afin d'assurer un contenu de 3 p.100 de matières organiques dans les 15 cm supérieures de dépôts "végétalisés" des lysimètres.

Le percolat des stériles récents chimiquement liés avait une teneur de Ra-226 inférieure à 1 pCi/L, comparativement à 84 pCi/L pour celui des stériles de contrôle.

Les effluents non dilués et les percolats des sédiments de $BaRaSO_4$ chimiquement liés (deuxième expérience) et des stériles récents (troisième expérience) se sont révelés létaux pour 100 p.100 des Daphnia pulex et des truites arc-en-ciel (Salmo gairdneri), au cours d'essais biologiques de 96 heures. Après une dilution de 50 p.100, les échantillons d'effluents n'étaient plus toxiques pour ces mêmes espèces.

TABLE OF CONTENTS

TABLE OF CONTENTS (CONT'D)

LIST OF FIGURES

LIST OF TABLES

CONCLUSIONS AND RECOMMENDATIONS

CONCLUSIONS

1. Recycling $BaRaSO_4$ sediments resulting from the treatment of seepage and runoff from abandoned mine tailing sites back to the surface of these sites is not an acceptable disposal method. Since these tailings are acidic, constituents in the sediments will dissolve and require reprecipitation to avoid a potentially hazardous environmental condition, particularly because of such radionuclides as radium, thorium and lead.
2. Lime applied to the surface of abandoned tailing sites at rates recommended to support revegetation had no effect on the leaching of radioactive constituents of concern.
3. Sediments ($BaRaSO_4$) from abandoned and fresh tailing sites require further characterization. The solubilization, leaching and co-precipitation of various metal and mineral hydroxides make the precipitated sediments a conglomeration of various constituents.
4. Chemical fixation of $BaRaSO_4$ sediments and fresh tailings has effectively reduced the release of ^{226}Ra to effluent and leachate during the first nine months of these experiments. The long-term stability of chemically-fixed sediments and tailings remains to be established.
5. Chemical fixation of fresh tailings appears to improve the media for growing vegetation compared to that of non-fixed tailings. This may be due to the increase in the water holding capacity of the chemically-fixed tailings.
6. Chemical fixation produces a condition in undiluted effluent and leachate which is extremely toxic to Daphnia and rainbow trout. Dilute (50%) effluent samples were non-toxic.

RECOMMENDATIONS

1. Sediments ($BaRaSO_4$) from acid seepage treatment should not be recycled to the top of abandoned tailing sites as they will significantly augment the levels of radioactivity already being leached from the site, hence requiring continual reprecipitation.

2. Tailings seepage and runoff from all abandoned tailing sites should be analyzed for all radioactive constituents of concern, prior to any treatment.
3. Various methods of waste residue solidification should be investigated and evaluated for effectiveness and economic feasibility, including methods not yet commercially available.
4. Improvement of the growing media for revegetation, by chemical fixation, requires further study. Investigations should include thickened discharge to obtain sloping topography depths of fixation required to stabilize abandoned tailing sites, as well as an assessment of economic feasibility.
5. Investigations should be expanded to include the chemical fixation of abandoned tailings, as well as sediments from fresh tailing sites.
6. The present project should continue long enough to determine the long-term stability of the chemical fixation process.
7. The causes of Daphnia and rainbow trout mortality following fixation should be determined and potential corrective measures investigated.

INTRODUCTION

One of the largest uranium ore reserves in the world is located in the Elliot Lake area of Ontario. It is estimated that uranium tailings from previous mining operations amount to about 80 x 10^6 tonnes (Moffett and Tellier, 1976) covering more than 400 hectares (1000 acres). Increased mining of the existing reserves (1000 x 10^6 tonnes) will add considerably to the environmental problems caused by the disposal of these wastes.

Uranium tailings contain about 85% of the initial radioactivity of the source ore. It is estimated that 1 tonne of tailings has a radioactivity of at least 5 millicuries (Moffett and Tellier, 1976). About 90% of this activity is due to the ^{238}uranium series. The most troublesome daughter isotopes of ^{238}uranium are ^{230}thorium, ^{226}radium and ^{210}lead. The transport of this radioactivity into surrounding watercourses has been identified as a major cause of pollution in the Serpent River basin (Roy and Keller, 1976). The solid tailings at Elliot Lake are made up of quartz, pyrite, calcium sulphate (from the neutralized reaction caused by lime addition at discharge), metal hydroxides, precipitated thorium and rare earths (Vivyurka, 1975). The liquid portion of the tailings is a neutral to alkaline saturated calcium sulphate solution, containing radium, ammonia and nitrate. Fresh tailing areas have a pH of 6.5 to 9.0, whereas seepage and runoff from abandoned tailings have a pH of 2 to 3.

Treatment of the effluent from fresh tailings consists of barium chloride addition to co-precipitate barium, radium and sulphate in lagoons. Treatment of seepage and runoff from abandoned tailings areas requires lime addition to pH 9, along with the addition of barium chloride to co-precipitate radium in sedimentation ponds.

The effectiveness of the currently practised precipitation treatments is dependent upon liquid flow rate, system residence time, influent concentrations, chemical dosage and point of chemical addition.

Limiting the percolation of water through both freshly-disposed and abandoned tailings would greatly reduce the amount of seepage requiring treatment in associated lagoons or ponds. It may also considerably reduce the bacterial oxidation process which causes a tailing pH decrease, and hence less solubilization of radioactive constituents. Revegetation with various crops (Murray and Moffett, 1976), and discharge with added thickeners to form cone-shaped piles as proposed by Rubinsky (1975), are two possible methods of limiting water percolation through tailing piles. The long-term stability of the precipitated sediments is not known. Until the long-term leaching characteristics of precipitated sediments are established, or alternate treatment process technology is developed, a reduction in sediment volume contained in readily accessible locations is desirable.

A Radioactivity Sub-Group was formed in 1974 by the Federal Mining Task Force to assist in the development of effluent guidelines and regulations for the uranium mining industry. This sub-group defined knowledge gaps related to the treatment of uranium mining and milling effluents and the dissolution of various radionuclides from tailings and precipitates. The current practice of $BaRaSO_4$ sediment storage in lagoons was considered to be an unsuitable long-term disposal solution since re-dissolution will probably occur after the abandonment period and cause contamination of receiving waters. The sub-group established that insufficient information was available to set effluent standards, or even to ascertain if such standards are warranted. As a result, a program was initiated at the Wastewater Technology Centre, Burlington, Ontario, in cooperation with the Environmental Management Service, to investigate the leachability of radioactive constituents from various uranium mining wastes.

2 OBJECTIVES

The initial lysimeter project commenced in 1974 with the primary objective of determining the amount of radioactive constituents released to receiving waters from abandoned tailings and tailings + $BaRaSO_4$ sediments mixtures. During 1976, the initial experiment was altered (Experiment 1, Phase 2) and two additional experiments added as shown in Figure 1.

The objective of Experiment 1, Phase 2, was to establish the influence of time and water flow on the rate of release of radioactive constituents from abandoned tailings and a tailings + sediment mixture.

Experiment 2 was initiated to investigate the release of radioactive constituents to effluent from co-precipitated $BaRaSO_4$ sediments under simulated field (pond) conditions with and without chemical fixation.

The objective of Experiment 3 was to compare levels of radioactive constituents in leachate from fresh tailings with chemically and biologically stabilized fresh tailings.

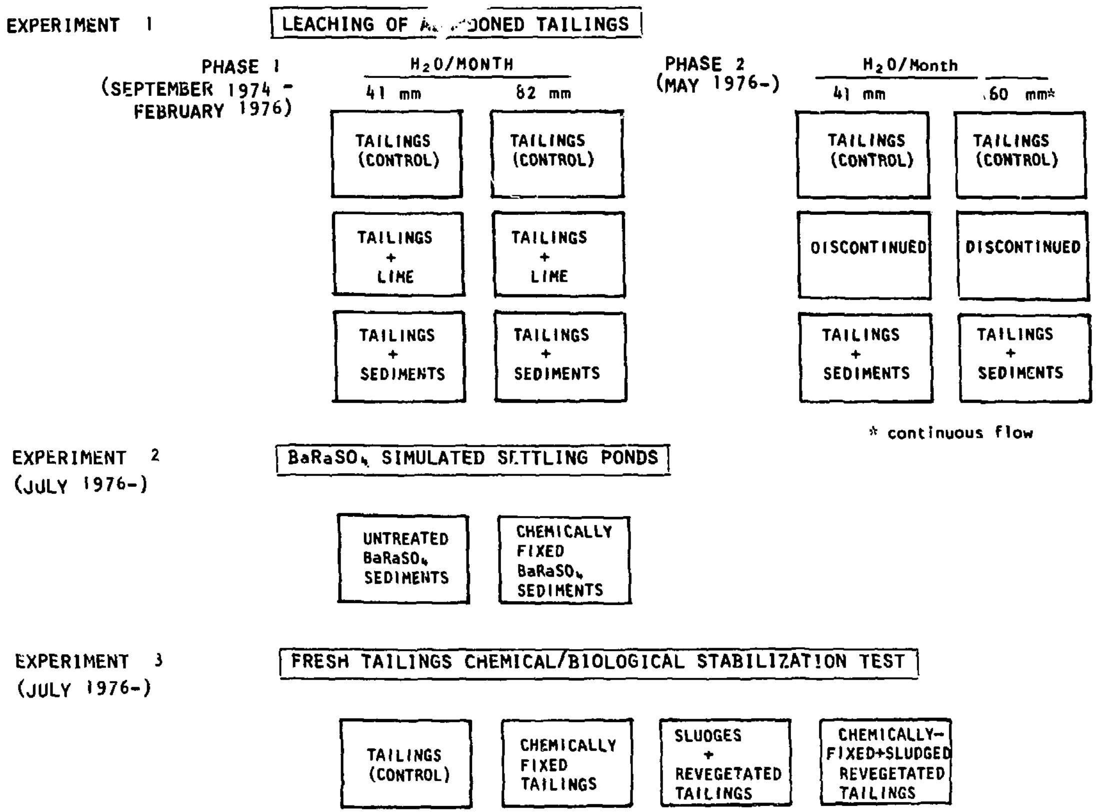


FIGURE 1 RADIOACTIVE LEACHABILITY PROJECT EXPERIMENT PHASING AND CHRONOLOGY

3 METHODS AND MATERIALS

3.1 Experimental Design and Treatments

All three experiments were randomized blocks with two replications. The six treatments in Experiment 1, Phase 1, as shown in Figure 1, consisted of tailings only; tailings with lime neutralized surface; and 89% tailings by weight plus 11% $BaRaSO_4$ sediments. Two water rates (41 mm and 82 mm per month) were applied to each of the three compositions every 10 days. At the suggestion of the Canada Centre for Mineral and Energy Technology (CANMET) Staff, Elliot Lake, the Experiment 1, Phase 2, high water rate was increased to a constant flow of ≃460 mm/mth to accelerate the leaching process.

Experiment 2 compared effluent from untreated $BaRaSO_4$ sediments to chemically-fixed (CF) sediments.

The four treatments to fresh tailing in Experiment 3 consisted of:

1) tailings only (control);
2) CF tailings;
3) tailings sludged to 3% organic matter (150 tonnes/hectare) and revegetated with a grass mixture of creeping fescue and red top; and
4) CF tailings + sludge + revegetation.

3.2 Lysimeters

Cylindrical acrylic lysimeters, 305 mm in diameter and 120 cm deep, were used as tailings containers in Experiment 1 (Figure 2).

Rectangular steel supported polyethylene lysimeters, 61 cm wide, 91 cm long and 81 cm deep, were used as containers for both Experiments 2 and 3 (Figure 3).

3.3 Source of Radioactive Materials

The abandoned tailings used in Experiment 1 were collected in 1974 from the Nordic tailing site. The $BaRaSO_4$ sediments collected in 1974 (Experiment 1) and 1976 (Experiment 2) were from the North Nordic Lake settling area used for treating acid seepage. The fresh tailings

Abandoned Tailing Compositions

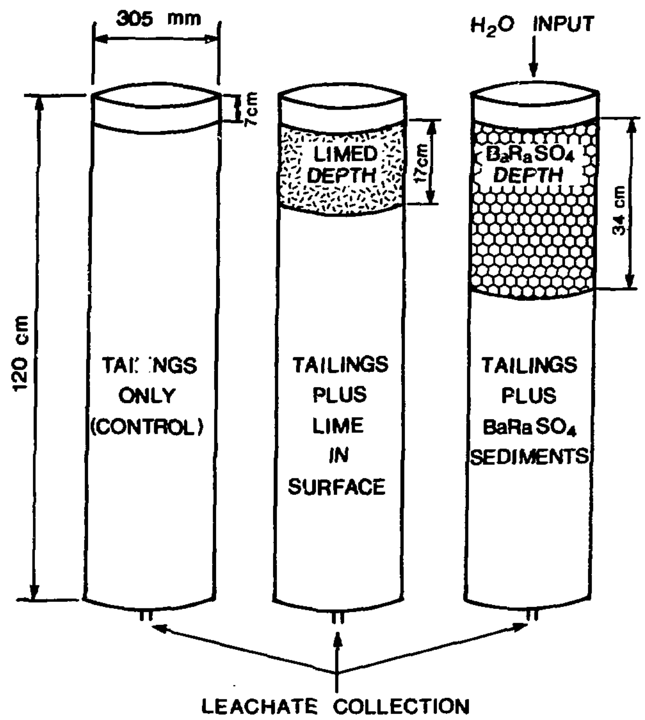
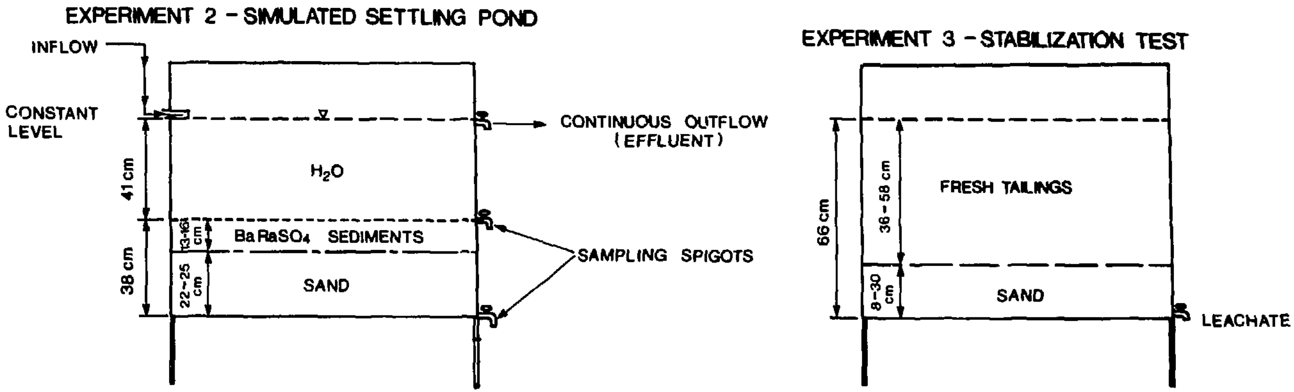


FIGURE 2 TYPICAL ACRYLIC LYSIMETER USED IN EXPERIMENT 1



Rectangular Polyethylene Lysimeters
(61 cm Wide x 91 cm Long x 81 cm Deep)

FIGURE 3 TYPICAL POLYETHYLENE LYSIMETERS USED IN EXPERIMENTS 2 AND 3 used in Experiment 3 were collected in 1976 from the current discharge site of an operational mine (Quirke) at Elliot Lake (Figure 4).

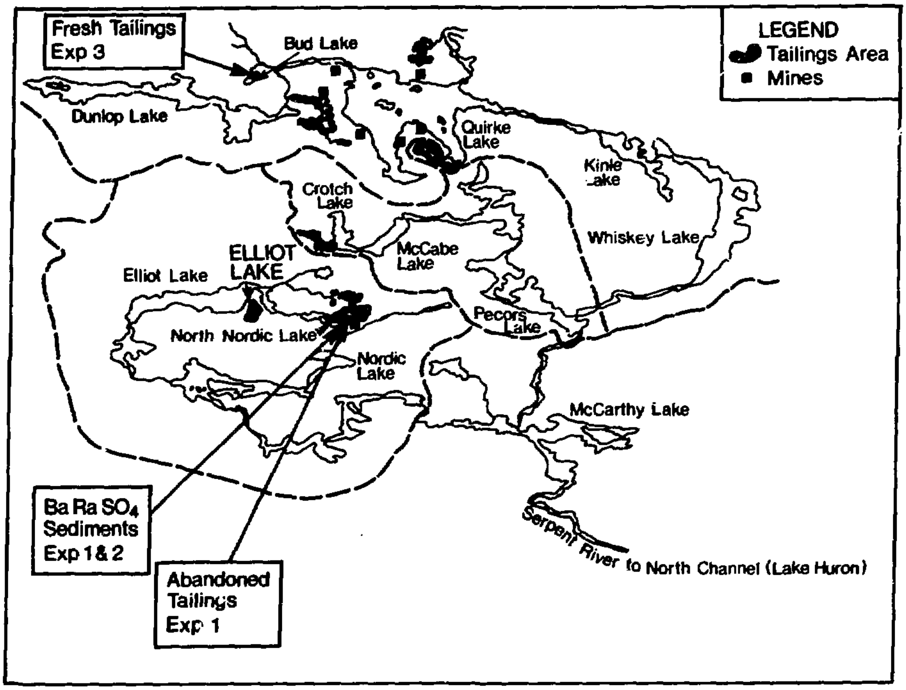


FIGURE 4 SOURCES OF RADIOACTIVE MATERIAL- Elliot Lake Area

3.4 Description of Materials in Lysimeters

The control treatment lysimeters used in Experiment 1 were packed with abandoned tailings by applying and tamping six lots of 15 kg each. The tailings + lime treatment lysimeters had five lots of 15 kg tailings applied and tamped, with the top layer consisting of a mixture of 5 kg of tailings and 10 kg of lime. The tailings + sediment treatment lysimeters had four lots of 15 kg tailings applied and tamped, followed by the top two layers which were a mixture of 10 kg of tailings and 5 kg of $BaRaSO_4$ sediment. The total weight of material in each lysimeter was 90 kg.

The simulated settling ponds in Experiment 2 had a silica sand base. For the non-fixed treatment, the sand base was 25 cm deep followed by 13 cm of $BaRaSO_4$ sediments. The CF treatment lysimeter had a sand base of only 22 cm to compensate for the increased volume resulting from the CF treatment. The chemically-treated sediment (16 cm) was poured over the sand base and levelled by trowelling. The overall depth for both treatments was 38 cm. The CF sediments were sprinked with water bi-weekly over a two-week curing period. Distilled water was applied to all treatments to a depth of 41 cm at the end of the curing period.

The fresh tailings used in Experiment 3 were homogenized in a commercial cement mixer. The chemical fixation process required a series of pumps and portable mixers. The control and tailings + sludge + vegetation treatments were applied as a slurry (85% dry matter by weight) to a depth of 36 cm over 30.5 cm of silica sand. The increased volume after chemical fixation required 58 cm of fixed tailings over 8 cm of silica sand to ensure equivalent amounts of tailings in each lysimeter. Overall sand and tailings depths of all treatments was 66 cm.

3.5 Sludge Application and Grass Seeding

Air-dried (90% dry matter) anaerobically digested sewage sludge, treated with $FeCl_3$ from a phosphorus removal system, was applied August 24, 1976 to the top 15 cm of the growing media to increase the organic matter content to 3%. Sludge constituent concentrations and loading rates to the tailings appear in Table 1.

A grass seed mixture (40% fescue and 60% red top by weight) was broadcast seeded on August 31, 1976.

3.6 Sampling Periods

Sampling of leachate from Experiment 1, Phase 1, was monthly from September, 1974 until February, 1976. From April, 1976 (Phase 2), leachate was collected bi-weekly and a portion of each sample composited to represent three-month leaching periods.

Sampling of the effluent from Experiment 2 (simulated settling ponds) was bi-weekly with a portion of each sample composited to represent monthly flow periods.

Sampling of leachate from Experiment 3 is dependent on climatic conditions. Approximately seven periods per year are anticipated.

TABLE 1. SLUDGE CONSTITUENT CONCENTRATION AND LOADING RATE

Constituents	Concentration (%)	Loading Rate (kg/ha)
TKN-N	2.31	3 465
Total P	4.98	7 470
Total C	21.30	31 950
Total S	0.94	1 410
Ca	3.90	5 850
Mg	0.4033	605
Na	0.0572	86
K	0.0792	119
Fe	7.245	10 870
Mn	0.0475	71
Al	0.9390	1 409
Cu	0.1210	182
Zn	0.2130	320
Ni	0.0020	3
Cr	0.0489	73
Pb	0.1700	255
Cd	0.0026	4
Mo	0.0033	5
Total Solids	90.0	135 000
Volatile Solids (600°C)	40.0	60 000

3.7 Radiochemical Analyses

Samples collected from September, 1974 to August, 1975 were analyzed for dissolved (filtered 0.45 micron) and undissolved ^{226}Ra, ^{232}Th, ^{230}Th, ^{228}Th, ^{210}Pb, gross alpha and beta by the Radiation Protection Laboratory, Ontario Ministry of Labour. All other radiochemical analyses were done at the Radiochemical Laboratory, Canada Centre for Inland Waters, according to methods of Joshi and Durham (1976).

3.8 Bioassay Tests

Daphnia pulex bioassays were 96-hour static tests in which eight juvenile Daphnia (less than 72 hours old) were individually exposed to 20 mL of undiluted leachate. Sample pH was adjusted to 7.5 with sulphuric acid before the test began. Leachate dissolved oxygen concentrations ranged from 7.9 to 8.7 mg/L (non-aerated).

Rainbow trout (*Salmo gairdneri*) bioassays were 96-hour single concentration static tests in which groups of five juvenile rainbow trout (0.9 to 1.0 g) were exposed to 40 litres of 25%, 50% and undiluted leachate.

Samples were aerated at 100 cc/min to maintain dissolved oxygen concentrations above 8 mg/L. Leachate from the chemically-fixed treatments required pH adjustment to 7.5 with sulphuric acid.

4 RESULTS AND DISCUSSION

4.1 Experiment I, Phase I - Leaching from Abandoned Tailings (September, 1974 to February, 1976)

Analyses for ^{232}Th, ^{230}Th, ^{228}Th, and dissolved beta were completed for the period September, 1974 to August, 1975. Dissolved (filtered 0.45 micron) and undissolved alpha emmission, ^{226}Ra, ^{210}Pb, and undissolved beta emmission, were completed for the period September, 1974 to February, 1977.

The results from control and tailings + lime treatments were similar for all radionuclides. Comparisons were therefore limited to controls, and tailings plus $BaRaSO_4$ sediments.

^{226}Ra. Initial levels of ^{226}Ra in leachate from all treatments ranged from 20 to 100 pCi/L, remaining relatively constant for the first 12 months. During the next five months, the levels increased for all treatments to range from 100 to 175 pCi/L. From May, 1975 until February, 1976, ^{226}Ra in leachate from the tailings + sediment mixture was about 30% higher than from the control (Figure 5). The quantity of ^{226}Ra leached per lysimeter was similar for all treatments from October, 1974 to September, 1975. From October, 1975 on, considerably more ^{226}Ra was leached at the higher water rate than at the low rate (Figure 6), indicating the possibility of solubilization of ^{226}Ra previously fixed in the $BaRaSO_4$ sediments.

^{210}Pb. Levels of ^{210}Pb in the leachate tended to increase slightly with time from October, 1974 to August, 1975 for all treatments except the high water rate to the tailings + sediment (Figure 7). From August, 1975 to February, 1976, the levels remained fairly constant (≈1500 pCi/L) with the high water rate to tailings + sediment treatment overcoming the previously mentioned lag to equal the other treatments. Levels of ^{210}Pb were generally higher from the control than from the tailings + sediment treatments regardless of the rate of water applied. The quantity of ^{210}Pb leached per lysimeter was considerably greater from the high water rate (Figure 8). During the summer of 1975, all treatments received similar amounts of precipitation. There was little difference in ^{210}Pb loss due to treatments during this period.

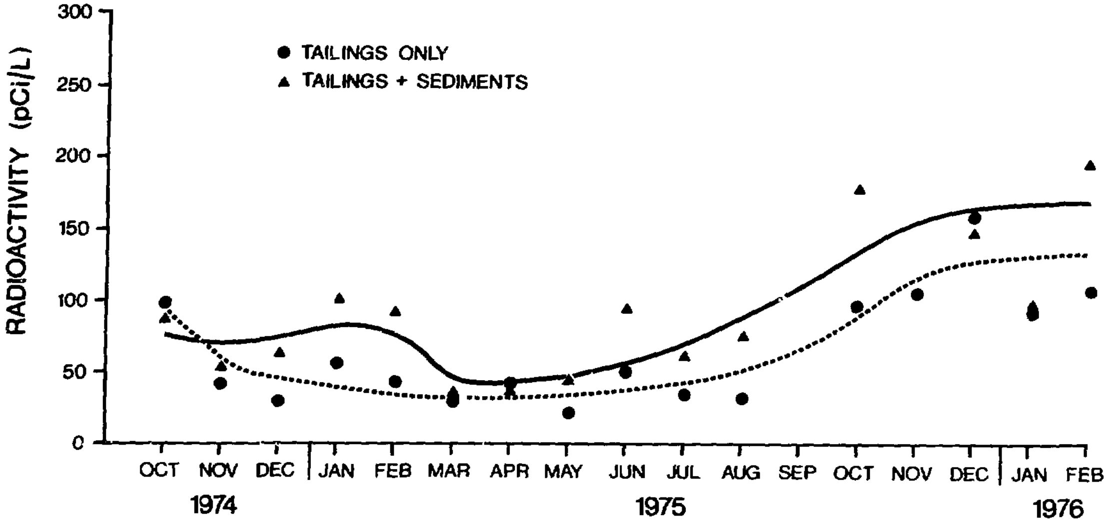


FIGURE 5 LEVELS OF ^{226}Ra IN LEACHATE FROM ABANDONED TAILINGS vs TIME

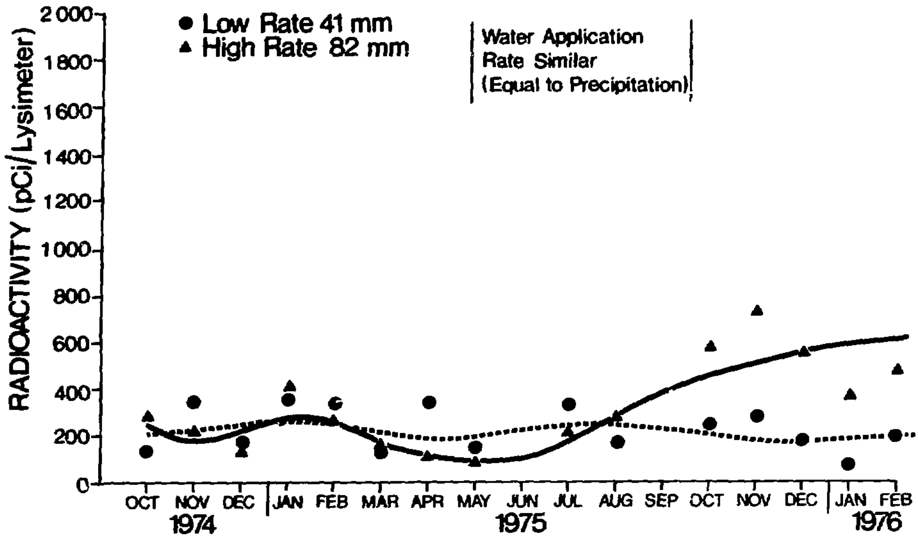


FIGURE 6 ^{226}Ra LEACHED FROM ABANDONED TAILINGS vs TIME

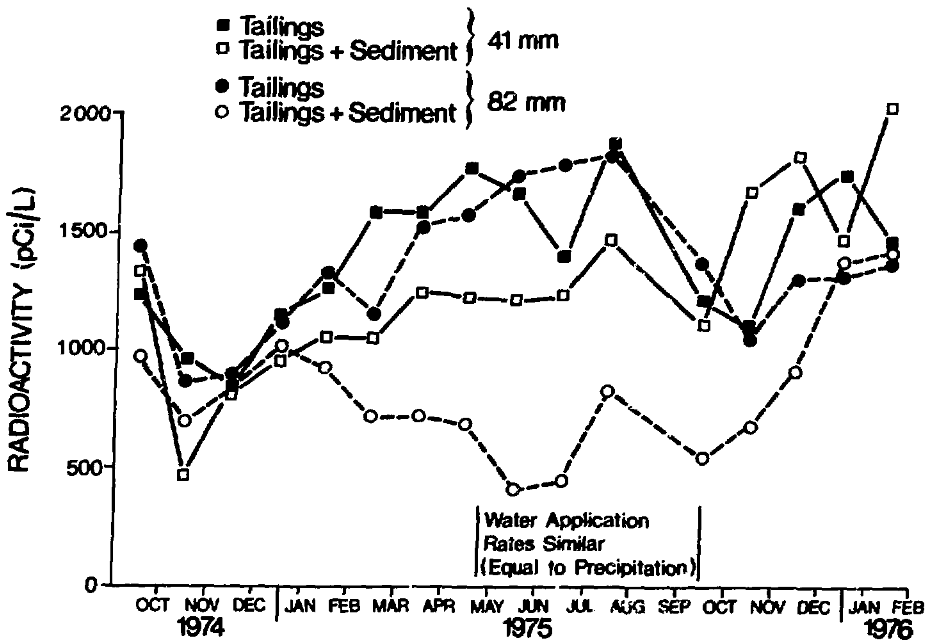


FIGURE 7 LEVELS OF ^{210}Pb IN LEACHATE FROM ABANDONED TAILING vs Time

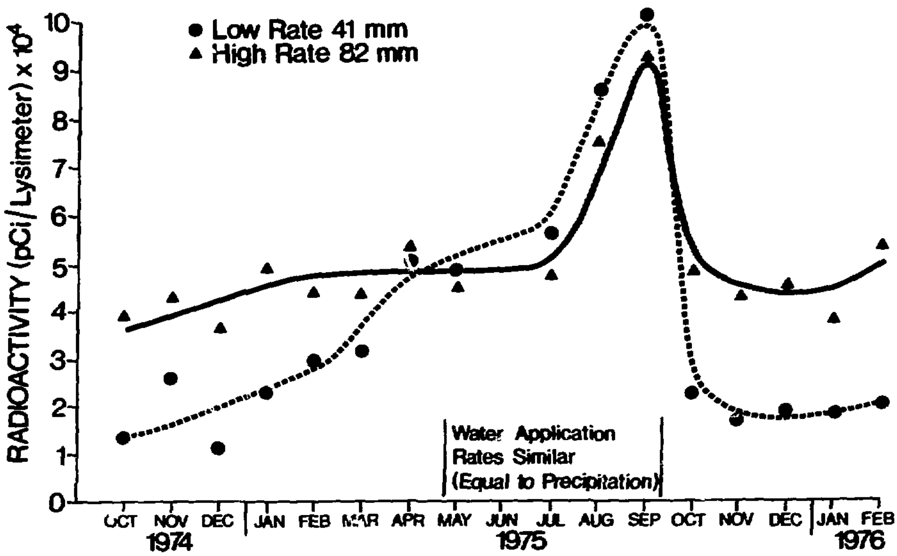


FIGURE 8 ^{210}Pb LEACHED FROM ABANDONED TAILINGS vs TIME

^{230}Th, ^{232}Th and ^{228}Th, gross alpha and beta. The activity of all these constituents showed similar time-radioactivity trends. Leachate radioactivity from tailings + sediment increased during the initial four months by several orders of magnitude compared to the control, remaining at these high levels until March, 1975. The gross alpha radioactivity levels increased in five months from an initial 10^3 pCi/L to 10^5 pCi/L from the tailings + sediment treatment, but remained in the 10^3 range for the control. A decrease of one order of magnitude in gross alpha activity from the tailings + sediment treatment occurred between March, 1975 and February, 1976 (Figure 9). Radiochemical analyses for thorium isotopes were delayed due to technical difficulties, but alpha spectrometry has shown that most gross alpha activity in these samples is from ^{230}Th. After five months of continuous leaching, ^{230}Th levels reached a high of 1×10^5 pCi/L (Figure 10) compared to 1×10^4 for ^{232}Th and 5×10^3 for ^{228}Th after which a slight decrease in activity occurred. The ^{230}Th leached (pCi/lysimeter) was less than gross alpha, but the time trends were similar for both constituents (Figure 11).

The $BaRaSO_4$ sediments used in this experiment contained considerable amounts of thorium, as evidenced by the high release of its radioisotopes to the leachate from the tailings + sediment treatment. Bacteriological oxidation of the 5 to 10% pyrite in the abandoned tailings results in an acidic condition (Figure 12). The low pH causes many metals and radioactive constituents to be leached from the tailings. The acidification process occurring in lysimeters containing tailings + sediment mixtures lowers the $BaRaSO_4$ sediment pH below the initial 8.0, and results in dissolution of the more soluble sediment constituents such as thorium and lead.

4.1.1 Experiment 1, Phase 2 - Leaching from Abandoned Tailings (May, 1976 to March 1977)

The one-month sampling frequency was altered to obtain composite samples representing three-month leaching periods. The radiochemical analyses completed for the first three leaching periods include ^{226}Ra and ^{210}Pb. Conductivity and pH were also monitored.

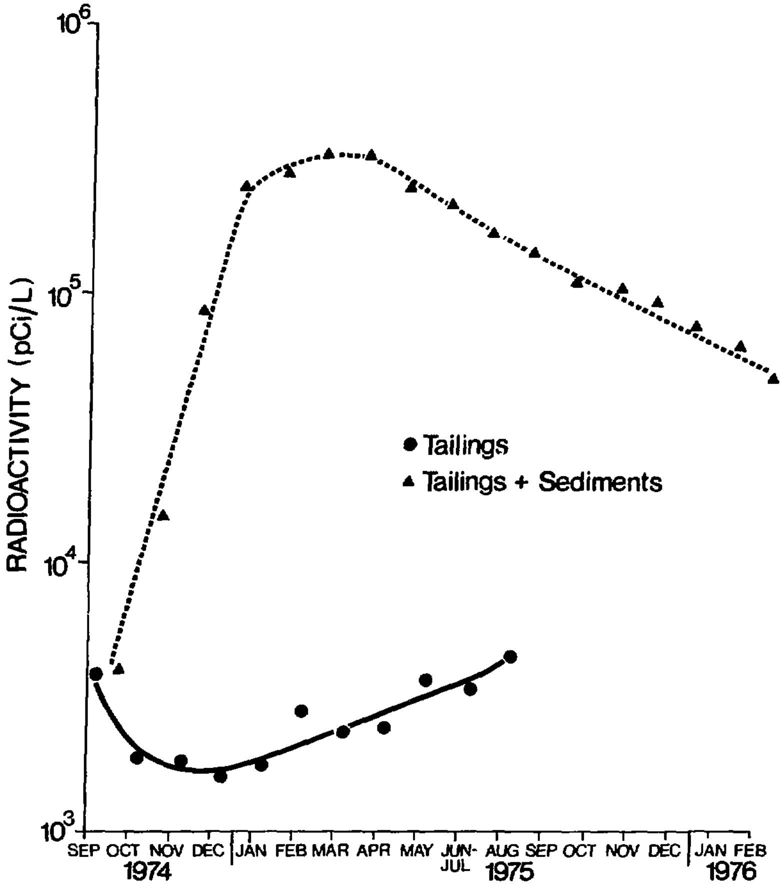


FIGURE 9 LEVELS OF GROSS ALPHA IN LEACHATE FROM ABANDONED TAILING vs TIME

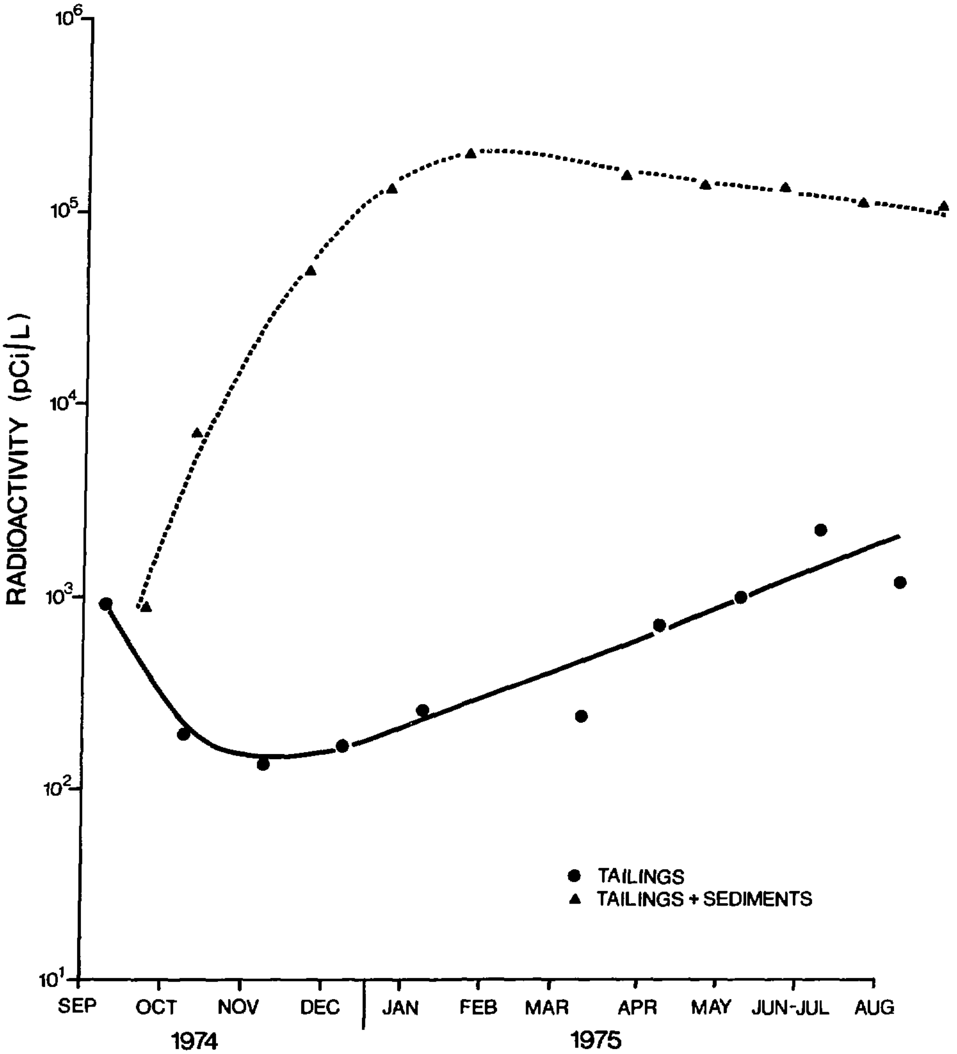


FIGURE 10 LEVELS OF ^{230}Th IN LEACHATE FROM ABANDONED TAILINGS vs TIME

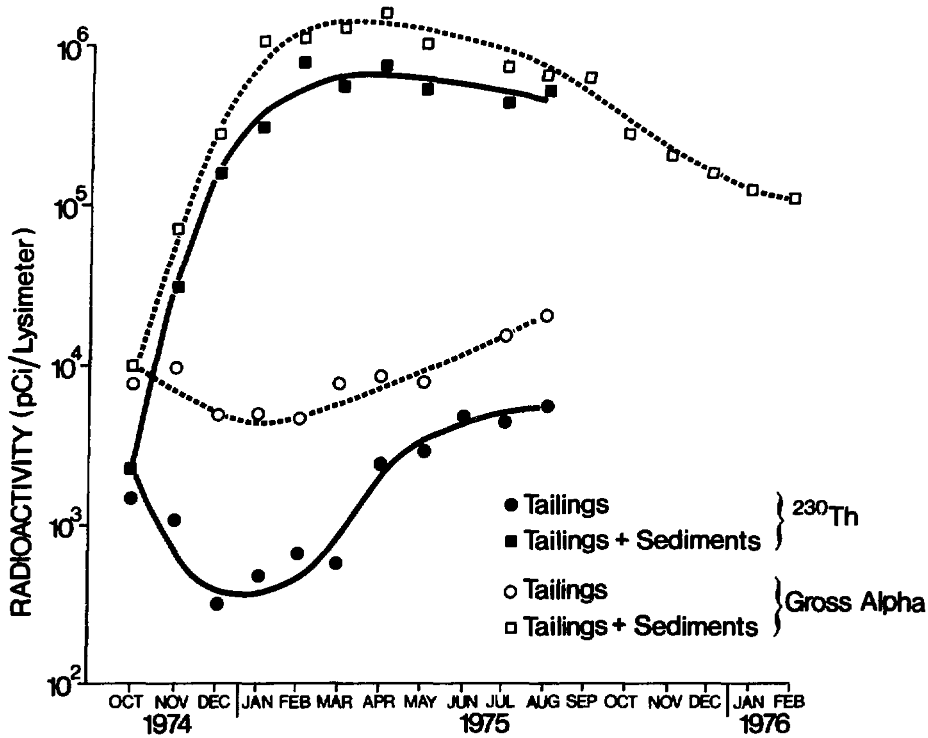


FIGURE 11 ^{230}Th LEACHED COMPARED TO GROSS ALPHA FROM ABANDONED TAILINGS vs TIME

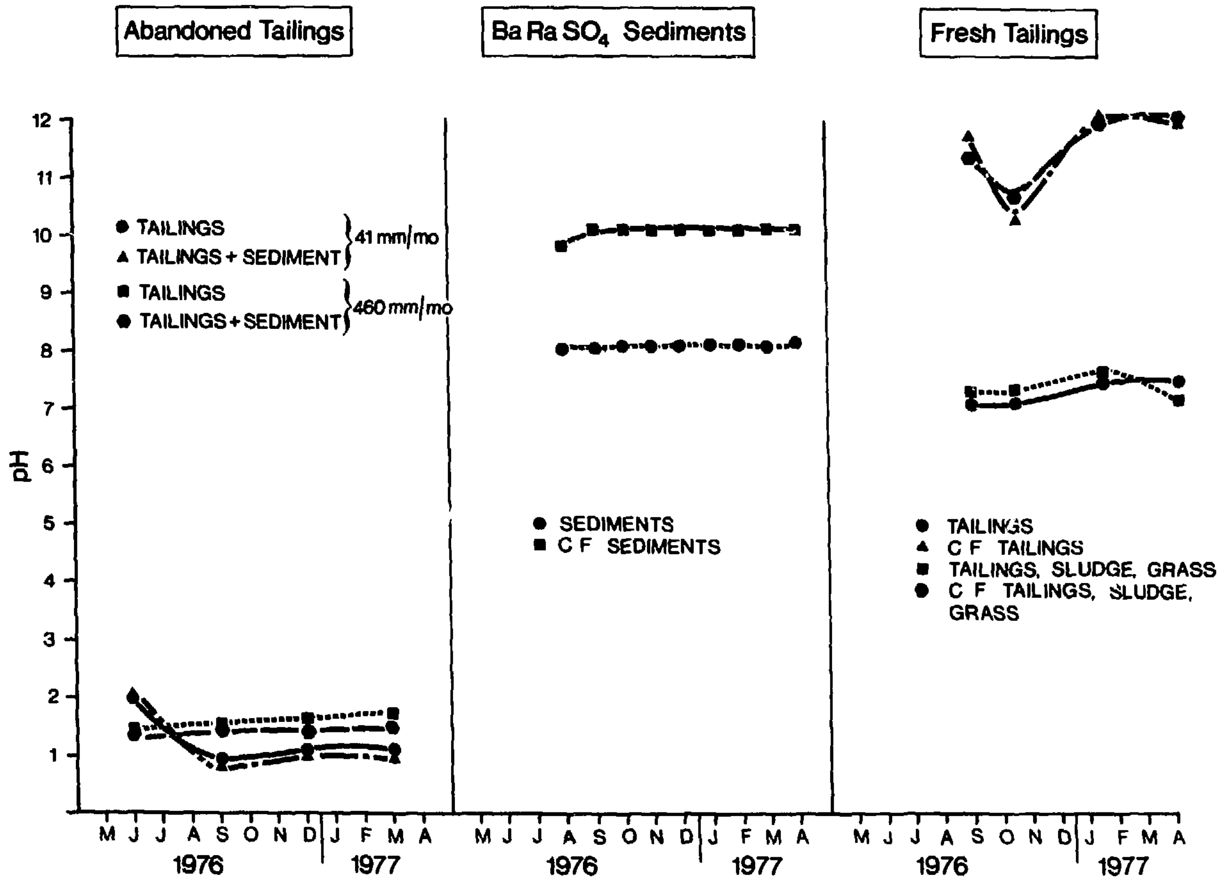


FIGURE 12 pH OF LEACHATE FROM ABANDONED TAILINGS, Ba Ra SO_4 SEDIMENT EFFLUENT AND FRESH TAILINGS LEACHATE vs TIME

Leachate. The first leaching period was limited to one month to obtain initial background information. Thereafter, leaching periods were of three months duration. Leachate volumes were similar for both tailing compositions at the high water application rate (Table 2). Leachate volume at the low water rate was dependent on local environmental conditions.

TABLE 2. LEACHATE VOLUMES FROM ABANDONED TAILINGS (L/lysimeter)

Leaching Periods 1976-77	Months	Tailings		Tailings + Sediments	
		H_2O Rate/Month		H_2O Rate/Month	
		41 mm*	460 mm**	41 mm*	460 mm**
May - June	1	2.0	41	1.4	39
June - Sept.	3	7.3	86	7.4	86
Sept. - Dec.	3	4.0	80	3.2	77
Dec. - Mar.	3	2.5	101	1.5	91
Cumulative	10	15.8	308	13.5	293

* Applied in 10-day intervals.
** Continuous flow.

pH. The pH of the leachate from all treatments during Phase 2 remained less than 2.0 (Figure 12).

Conductivity. The conductivity from both treatments remained stable with time at $\sim 1 \times 10^4$ micromhos/cm at the high water rate compared to $\sim 8.5 \times 10^4$ at the low water rate.

^{226}Ra. Leachate ^{226}Ra levels at the low water rate increased with time, particularly from the tailings + sediment treatment (Table 3). At the high water application rate, ^{226}Ra in the leachate stabilized after one month with levels about 39% lower from tailings + sediments than from the control. The quantity of ^{226}Ra leached per lysimeter was considerably higher from the high water application rate (Table 3). The quantity leached at the low water rate from the tailings + sediment treatment was about twice that from the control during both three-month leaching periods, unlike the situation at the high water application rate. The increased quantity of ^{226}Ra leached from tailings + sediments

TABLE 3. LEVELS OF ^{226}Ra IN LEACHATE AND QUANTITY LEACHED FROM ABANDONED TAILINGS

Leaching Periods 1976	Months	Tailings				Tailings + Sediments			
		H_2O Rate/Month				H_2O Rate/Month			
		41 mm*		460 mm**		41 mm*		460 mm**	
		pCi/L	pCi/lysimeter	pCi/L	pCi/lysimeter	pCi/L	pCi/lysimeter	pCi/L	pCi/lysimeter
May - June	1	74	148	108	4 428	137	192	121	4 719
June - Sept.	3	105	767	130	11 180	180	1 332	80	6 880
Sept. - Dec.	3	127	508	133	10 640	340	1 088	77	5 929
Cumulative	7	-	1 423	-	26 248	-	2 612	-	17 528

* Applied in 10-day intervals.
** Continuous flow.

at the low water rate corresponds with the results obtained from Phase 1 (Figure 4). The lower release of ^{226}Ra at the high water application rate from the tailings + sediment cannot be explained at this time. Barium leached from sediments at the top of the lysimeter may reprecipitate as $BaSO_4$ when in contact with $SO_4^=$ ions lower in the lysimeter, tying up soluble ^{226}Ra from both the tailings and the sediment.

^{210}Pb. Levels of ^{210}Pb in leachate from tailings + sediment at the high water rate decrease with time. After seven months of leaching, the low water rates had ^{210}Pb levels of 2730 and 3015 pCi/L for control and tailings + sediment treatments, respectively (Table 4), compared to the high water rate levels of 858 and 973 pCi/L. The ^{210}Pb leached was considerably greater at the high water rates for both treatments (Table 4). The total loss after seven months was 28% greater from the tailings + sediment treatment than from the control at the low water rate, and 14% greater at the higher water rate.

4.1.2 Experiment 1 - Summary

From the data obtained to date, recycling of $BaRaSO_4$ from acid seepage treatment to abandoned tailing piles would appear to be an undesirable disposal practice. The increasing radioactivity with time in the leachate from tailings + sediment, compared to the control, strongly indicates degradation of the $BaRaSO_4$ sediments. Recycling this sediment would require continuous treatment of the effluent (seepage and runoff).

4.2 Experiment 2 - Simulated Settling Ponds (August, 1976 to April, 1977)

Quality of effluent (flow) over chemically-fixed $BaRaSO_4$ sediments was compared to non-fixed sediments commencing in August, 1976. Radiochemical analysis for ^{226}Ra and ^{210}Pb was completed for six leaching periods (August, 1976 until February, 1977). Conductivity and pH were monitored from August, 1976 to April, 1977. Static 96-hour bioassay tests using Daphnia were performed on effluent in December, 1976, and tests using rainbow trout were performed in April, 1977.

TABLE 4. LEVELS OF ^{210}Pb IN LEACHATE AND QUANTITY LEACHED FROM ABANDONED TAILINGS

Leaching Periods 1976	Months	Tailings				Tailings + Sediments			
		H_2O Rate/Month				H_2O Rate/Month			
		41 mm*		460 mm**		41 mm*		460 mm**	
		pCi/L	pCi/ lysimeter	pCi/L	pCi/ lysimeter	pCi/L	pCi/ lysimeter	pCi/L	pCi/ lysimeter
May - June	1	1 980	3 960	1 305	53 505	2 100	2 940	1 425	55 575
June - Sept.	3	2 200	16 060	860	73 960	2 600	19 240	1 250	107 500
Sept. - Dec.	3	2 730	10 920	858	67 399	3 015	9 648	973	69 601
Cumulative	7	-	30 940	-	194 864	-	31 828	-	232 676

* Applied in 10-day intervals.
** Continuous flow.

pH. The pH of the effluent from chemically-fixed sediments remained constant at $\sim$10.0 compared to pH $\sim$8.0 from non-fixed sediments during nine months (Figure 12).

Conductivity. The conductivity of the effluent from non-fixed sediments remained stable at 650 micromhos/cm. The conductivity of the chemically-fixed sediments increased during the initial four months to $\sim$3000 micromhos/cm after which a sl[illegible]ecrease occurred to about 2700 micromhos/cm (Figure 13).

^{226}Ra. Levels of ^{226}Ra in the effluent from non-fixed sediments ranged from 3 to 9 pCi/L compared to <1 pCi/L from the chemically fixed sediments.

^{210}Pb. Levels of ^{210}Pb in the effluent from all treatments has remained <15 pCi/L.

Static Bioassay Tests. In undiluted effluent from chemically fixed tailings, 100% mortality (96-hour test) occurred to both Daphnia and rainbow trout (Table 5). In a 50% effluent concentration from chemically fixed sediments (pH adjusted) no mortality occurred to rainbow trout. Initially, it was suspected that the high pH of the effluent from the chemically-fixed sediments was responsible for the toxicity, but 100% mortality occurred following pH adjustment to 7.0. No mortality of these species occurred in tests using undiluted effluent from non-fixed tailings.

4.2.1 Experiment 2 - Summary

It is apparent that chemical fixation of $BaRaSO_4$ sediments substantially decreases the release of ^{226}Ra to the effluent. Several years of monitoring may be required to determine the long-term stability of chemically-fixed sediments. The fixation chemicals used have resulted in leachate which is toxic to both Daphnia and rainbow trout when undiluted. The identity of the toxic substances released, as well as the dilution required to eliminate this toxicity, will be investigated as the project continues.

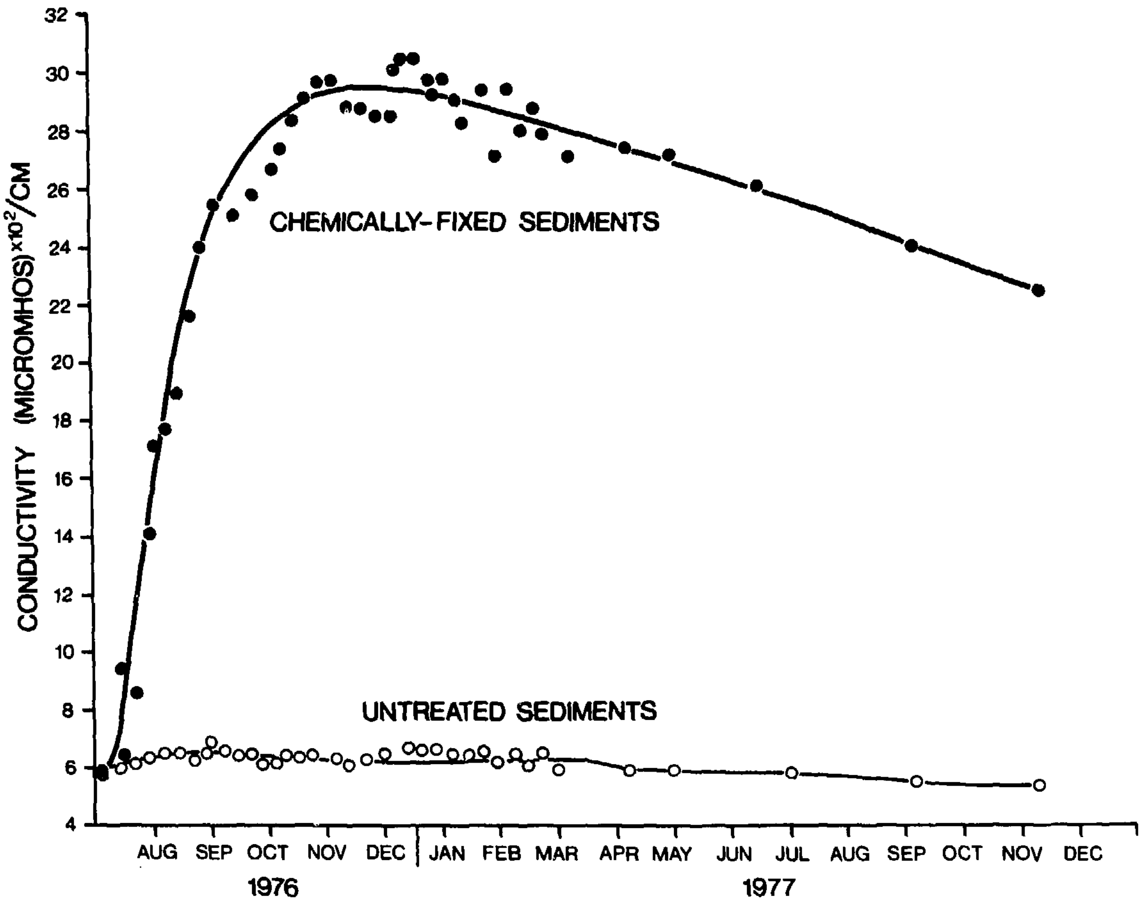


FIGURE 13 CONDUCTIVITY OF Ba Ra SO_4 SEDIMENT EFFLUENT vs TIME

TABLE 5. MORTALITY RATE OF DAPHNIA AND TROUT IN EFFLUENT FROM SIMULATED SETTLING PONDS

Treatments	Effluent Concentration %	Species Mortality %	
		Daphnia	Rainbow Trout
Non-fixed sediments	100	0	0
pH 8.0	50	NT	0
	25	NT	0
Chemically-fixed sediments	100	100	NT
pH 10.0	50	NT	NT
	25	NT	NT
Chemically-fixed sediments	100	100	100
pH adjusted to 7.0	50	NT	0
	25	NT	0

NT = Not tested.

4.3 Experiment 3 - Leaching from Fresh Tailings (August, 1976 to January, 1977)

Leachate radioactivity from chemically-fixed fresh tailings (with and without vegetation) was compared to that of non-fixed tailings.

Radiochemical analyses for ^{226}Ra and ^{210}Pb were completed for three leaching periods from September, 1976 to January, 1977. Conductivity and pH were monitored from September, 1976 to April, 1977. Static 96-hour bioassay tests using Daphnia were conducted on leachate for the period November, 1976 to January, 1977.

Volume. The volume of leachate per lysimeter appears to be affected by chemical fixation. The third leaching period produced twice as much leachate from non-fixed tailings as from chemically-fixed treatments (Table 6). This may be attributed to the increase in the water holding capacity of the chemically-fixed tailings, resulting in considerably more surface evaporation.

pH. The pH of leachate from chemically-fixed tailings stabilized at ∿12.0 compared to ∿7.5 from non-fixed tailings after five months (Figure 12).

TABLE 6. LEACHATE VOLUMES FROM FRESH TAILINGS*

Leaching Periods 1976-77	Months	Tailings (Control)	CF Tailings	Tailings + Sludge + Vegetation	CF Tailings + Sludge + Vegetation
Aug. - Sept.	1	9.5	9.8	11.1	13.8
Sept. - Nov.	2	3.1	3.4	4.2	6.2
Nov. - Jan.	2	10.2	4.0	9.9	4.7
Cumulative	5	22.8	17.2	25.2	24.7

* Litres/lysimeter.

Conductivity. After nine months of monitoring, the conductivity of leachate from chemically-fixed treatments was $\sim 14 \times 10^3$ micromhos/cm compared to $\sim 3 \times 10^3$ micromhos/cm from non-fixed tailings. The application of sludge prior to seeding had no effect on leachate conductivity.

^{226}Ra. Levels of ^{226}Ra in the leachate from chemically-fixed treatments were considerably lower than from non-fixed treatments (Table 7). Tailings receiving sludge treatments had increased ^{226}Ra levels compared to those of the non-sludged treatments. The increase in ^{226}Ra leached following sludge application requires further investigation. Control tailings leached a total of 1558 pCi ^{226}Ra compared to 3071 pCi from sludged tailings. This may be a short term phenomenon related to initial degradation of the organics in the sludge. The vegetative cover was established in late 1976 when growth was limited and should not have affected the amounts of ^{226}Ra leached.

^{210}Pb. Levels of ^{210}Pb in the leachate remained at <15 pCi/L for all treatments.

Static bioassay tests. All treatments having chemical fixation resulted in 100% mortality to Daphnia in undiluted leachate. There was 37.5% mortality in undiluted leachate from the non-chemically-fixed sludged tailings. Bioassay tests results are shown in Table 8. The cause of the toxic condition is presently under investigation.

TABLE 7. LEVELS OF ^{226}Ra IN LEACHATE AND QUANTITY LEACHED FROM FRESH TAILINGS

Leaching Periods 1976-77	Months	Tailings (Control)		CF Tailings		Tailings + Sludge + Vegetation		CF Tailings + Sludge + Vegetation	
		pCi/L	pCi/ lysimeter	pCi/L	pCi/ lysimeter	pCi/L	pCi/ lysimeter	pCi/L	pCi/ lysimeter
Aug. - Sept.	1	85	808	<1	≈5	80	888	13	179
Sept. - Nov.	2	84	260	2	7	225	945	9	56
Nov. - Jan.	2	48	490	2	8	125	1 238	10	47
Cumulative	5	-	1 558	-	≈20	-	3 071	-	282

TABLE 8. MORTALITY RATE OF DAPHNIA IN UNDILUTED LEACHATE FROM FRESH TAILINGS

Leachate pH	Tailings (Control)	CF Tailings	Tailings + Sludge + Vegetation	CF Tailings + Sludge + Vegetation
	% Mortality	% Mortality	% Mortality	% Mortality
Non-Adjusted	0	100	37.5	100
Adjusted to 7.0	NT	100	NT	100

NT = Not tested.

4.3.1 Experiment 3 - Summary

The leaching of ^{226}Ra from fresh tailings is significantly reduced by chemical fixation, but it may take several years to determine the stability of the chemically-fixed product. It is imperative that the cause of Daphnia mortality be identified because chemical fixation could result in large volumes of toxic effluent being discharged to sedimentation ponds or directly into receiving waters. The effect of revegetation cannot be assessed at this time. It is expected that vegetative cover may benefit from the higher water holding capacity of the chemically-fixed tailings.

5 SUMMARY

Any treatment which will maintain a neutral to alkaline pH in tailings or precipitated residues appears desirable, especially pertaining to the solubilization of Pb and Th radionuclides. Levels of ^{210}Pb in effluent or leachate having a pH greater than 7 have never exceeded the detection level of 15 pCi/L, compared to levels ranging from 500 to 2000 pCi/L from acidic leachates. Thorium analyses are not complete to date. However, ^{230}Th radioactivity levels were up to 1×10^5 pCi/L in acidic leachate. It is suspected that this isotope would remain bound in the solid fraction, provided a neutral to alkaline pH is maintained.

Chemical fixation (solidification) of mine waste residues has decreased the release of ^{226}Ra to effluent and leachate. The determination of the long-term stability of these treatments to various residues requires long-term monitoring to develop a satisfactory data base and thereby allow the development and unification of predictive models.

Chemical fixation has resulted in effluent and leachate which is toxic to both Daphnia and rainbow trout. The reason for this toxicity must be determined and corrective treatment investigated.

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