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PARADOXICAL SECULAR DISEQUILIBRIUM OF RADIOISOTOPES
IN URANIUM MINE TAILINGS

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PARADOXICAL SECULAR DISEQUILIBRIUM OF RADIOISOTOPES
IN URANIUM MINE TAILINGS

by

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ABSTRACT

The activity ratios of daughter/parent of radioisotope species in groundwater seldom equal 1.0, indicating that isotopic disequilibrium is a common occurrence. The most common expectation of many researchers for the secular disequilibrium parameters is that this activity ratio will be less than 1.0 for parent isotopes with a very long half-life, and daughter isotopes with a very short half-life, since the activity of the daughter isotopes depends on the existence of the parent isotopes.

A review of non-mining related studies shows that activity ratios of the ^{238}U , ^{232}Th and ^{235}U decay series could differ greatly between sites, and may or may not show trends among sites. Some extreme values of activity ratios (from less than 1.0 up to 3000 with most higher than 1.0) have been observed. The review of uranium mining related studies indicates less drastic ratio values ranging from 1.0 to 241 with predominant values between 1.0 and 2.0.

The deviation of the activity ratio from the expected theoretical value of less than 1.0 (for normal secular disequilibrium) is called paradoxical secular disequilibrium.

This thesis presents an in-depth study of interactions between liquid and solid media containing members of radioisotope decay chains, in a uranium tailings deposit. Paradoxical secular disequilibrium is seen in many of the activity ratios obtained with ratios depending on the location of the sample in the tailings pile, the nature of the isotopes involved, and the geochemical processes taking place. Laboratory experiments conducted to demonstrate the existence of paradoxical secular disequilibrium using ^{223}Ra and ^{224}Ra as natural tracers, are also described.

The importance of taking this disequilibrium into account in designing sampling and analysis procedures for systems such as a tailings area is outlined.

RÉSUMÉ

Les rapports de l'activité du précurseur à celle des descendants pour les radioisotopes des eaux souterraines sont rarement égaux à 1,0, ce qui indique que les déséquilibres isotopiques sont fréquents. Pour les paramètres du déséquilibre séculaire, beaucoup de chercheurs s'attendent à ce que le rapport des activités soit inférieur à 1,0 pour les précurseurs à très longue période et les descendants à très courte période, puisque l'activité des descendants dépend de l'existence de précurseurs.

Un examen des études ne portant pas sur les activités minières montre que les rapports des activités pour les séries de désintégration de ^{238}U , ^{232}Th et ^{235}U peuvent varier beaucoup d'un endroit à un autre et qu'il n'y a pas nécessairement de tendances parmi les différents endroits. On a observé des valeurs très divergentes pour ces rapports des activités (depuis moins de 1,0 jusqu'à 3000, mais la plupart du temps plus de 1,0). L'examen des études relatives à des activités minières montre par contre beaucoup moins de divergences (de 1,0 à 241 seulement) et la plupart des valeurs sont entre 1,0 et 2,0.

L'écart entre le rapport des activités et la valeur théorique prévue inférieure à 1,0 (pour le déséquilibre séculaire normal), s'appelle le déséquilibre séculaire paradoxal.

Cette thèse contient une étude détaillée des interactions entre les milieux solides et liquides d'un dépôt de résidus de mine d'uranium contenant des séries isotopiques en désintégration. Le déséquilibre séculaire paradoxal se retrouve dans nombre des rapports des activités obtenus, ceux-ci dépendant de l'endroit où l'on a prélevé l'échantillon dans les résidus, de la nature des isotopes et des processus géochimiques qui se produisent. On décrit les expériences faites au laboratoire pour démontrer l'existence du déséquilibre séculaire paradoxal en utilisant ^{223}Ra et ^{224}Ra comme traceurs.

Enfin, on décrit l'importance de prendre en considération ce déséquilibre lorsque l'on compare les méthodes d'échantillonnage et d'analyse pour les systèmes comme les dépôts de résidus miniers.

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CHAPTER 1

INTRODUCTION

The aqueous migration of radioactive elements of various mobilities is commonly studied. This migration produces geochemical plumes that can be detected by measurements of gamma and beta emissions, and alpha-particle counting, and the differential mobilities leading to decay-series disequilibrium within the plumes.

Many studies have been done in the vicinities of the uranium ore deposits and uranium mill tailings. The migration of the radionuclides within the uranium ore deposit reflects their solubility and aqueous behaviour, and the formation of the plumes, having an analog in the contaminant geochemistry of uranium mill tailings.

Uranium mill tailings can represent an extreme example of radioactive deposits in which the ore is fine grained and unconsolidated. The porewater contains strong leaching chemicals, and since most of the uranium has been removed during milling, the decay equilibrium which initially existed within the ore is disturbed (1). The contaminated porewater, which is commonly replenished by infiltration of precipitation and/or mill effluent, often drains downward into underlying groundwater flow systems ((1) in review). Therefore, radionuclide-laden porewater seepage into the ground water provides opportunities for researchers to examine the radionuclide migration and its deviation from decay series equilibrium.

This thesis presents a study of disequilibrium in the decay series of uranium species in the uranium tailings area within the aqueous and solid tailings media. It shows in-depth studies of interactions between aqueous and solid media, which in turn affect the radionuclide equilibrium. A general review of previous studies of isotope disequilibrium in groundwater,

and some detailed reviews on analytical methods are presented to assist in comprehending the framework of this thesis.

CHAPTER 2

RADIOISOTOPE DECAY

2.1 A QUICK REVIEW OF THE URANIUM DECAY SERIES

All elements found in natural sources with atomic numbers greater than 83 (bismuth) are radioactive. They belong to chains of successive decays, and all the species in one such chain constitute a radioactive family or series (3). Three of these families include all the natural activities in this region of the periodic chart. One of them is ${}_{92}\text{U}^{238}$ with a general formula for the approximate mass of $4n+2$, where n is an integer. Therefore, the uranium series is also known as the $4n+2$ series. Figure 2.1 shows U (with mass 238) as parent substance, and after fourteen transformations (eight of them by α -particle emission, and six by β -particle emission) it reaches a stable end product ${}_{82}\text{Pb}^{206}$ known as Radium B.

The second series is known as the $4n$ Thorium series with its parent ${}_{90}\text{Th}^{232}$ and a stable end product ${}_{82}\text{Pb}^{208}$ (Figure 2.2). The third series is known as the $4n+3$, or actinium series, with ${}_{92}\text{U}^{235}$ as parent and a stable end product ${}_{82}\text{Pb}^{207}$ (Figure 2.3).

Isotopes are referred to an atomic species of the same atomic number, i.e., belonging to the same element, but having a different mass number. For example, a ${}_{17}\text{Cl}^{35}$ nucleus having a mass number of $A=35$ and an atomic number of $Z=17$, contains 17 protons and 18 neutrons. The number of protons in a nucleus equals its atomic number Z , and the total number of neutrons and protons (collectively called nucleons) equal its mass number A . Therefore, the neutron number $N = A - Z$. The difference $N - Z$ or $(A - 2Z)$ between the number of neutrons and protons in a nucleus is referred to as its neutron excess or isotopic number. The symbol used to denote a nuclear species is the chemical

U 92	U^{238}, U_I (uranium I) 4.51×10^9 years		U^{234}, U_{II} (uranium II) 2.48×10^5 years				
Pa 91		α	β	β	α		
			Pa^{234}, UX_2 (99.85%) 1.18 minutes				
			Pa^{234}, UZ 6.7 hours				
Th 90	Th^{234}, UX_1 (uranium X ₁) 24.1 days			Th^{230}, Io (ionium) 7.52×10^4 years			
Ac 89				α			
Ra 88				Ra^{226}, Ra (radium) 1622 years			
Fr 87				α			
Rn 86				Rn^{222}, Rn (radon) 3.825 days			
At 85				α	At^{218} 1.3 seconds		
Po 84				Po^{218}, RaA (radium A) 3.05 minutes	β (0.02%) α	Po^{214}, RaC' (radium C') 1.6×10^{-4} second	Po^{210}, RaF (polonium) 138.4 days
Bi 83				α (99.98%)	Bi^{214}, RaC (radium C) 19.7 minutes	β (99.96%) α	Bi^{210}, RaE (radium E) 5.01 days
Pb 82				β	Pb^{214}, RaB (radium B) 26.8 minutes	α (0.04%)	Pb^{210}, RaD (radium D) 22 years
						α ($5 \times 10^{-6}\%$)	Pb^{206}, RaG (stable lead isotope)
Tl 81					Tl^{210}, RaC'' (radium C'') 1.32 minutes	β α ($1.8 \times 10^{-6}\%$)	Tl^{206}, RaE'' (radium E'') 4.3 minutes
Hg 80						β	Hg^{206} 8.5 minutes

Fig. 2.1 - The uranium (4n+2) series. (From Nuclear and Radiochemistry (3)).

Th 90	Th ²³² , Th (thorium) 1.39 × 10 ¹⁰ years		Th ²²⁸ , RdTh (radiothorium) 1.90 years		
Ac 89		Ac ²²⁸ , MsTh ₂ (mesothorium 2) 6.13 hours			
Ra 88	Ra ²²⁸ , MsTh ₁ (mesothorium 1) 6.7 years		Ra ²²⁴ , ThX (thorium X) 3.64 days		
Fr 87					
Rn 86			Rn ²²⁰ , Tn (thoron) 54.5 seconds		
At 85					
Po 84			Po ²¹⁶ , ThA (thorium A) 0.158 second		Po ²¹² , ThC' (thorium C') 3.0 × 10 ⁻⁷ second
Bi 83			Bi ²¹² , ThC (thorium C) 60.6 minutes		
Pb 82			Pb ²¹² , ThB (thorium B) 10.6 hours		Pb ²⁰⁸ , ThD (stable lead isotope)
Tl 81					Tl ²⁰⁸ , ThC'' (thorium C'') 3.1 minutes

Fig. 2.2 - The thorium (4n) series. (From Nuclear and Radiochemistry (3)).

U 92	U^{235} , AcU (actinouranium) 7.13×10^8 years				
Pa 91	α	Pa^{231} , Pa (protactinium) 3.48×10^4 years			
Th 90	Th^{231} , UY (uranium Y) 25.6 hours	β	α	Th^{227} , RdAc (radioactinium) 18.17 days	
Ac 89		Ac^{227} , Ac (actinium) 22.0 years	β (98.8%)	α	
Ra 88		α (1.2%)	Ra^{223} , AcX (actinium X) 11.7 days		
Fr 87		Fr^{223} , AcK (actinium K) 22 minutes	β	α	
Rn 86		α (4×10^{-3} %)	Rn^{219} , An (actinon) 3.92 seconds		
At 85		At^{219} 0.9 minute	β (3%)	α	At^{215} 10^{-4} second
Po 84		α (97%)	Po^{215} , AcA (actinium A) 1.83×10^{-3} second	β (5×10^{-4} %)	α
Bi 83		Bi^{215} 8 minutes	β	α	Bi^{211} , AcC (actinium C) 2.15 minutes
Pb 82			β	α (99.68%)	Pb^{207} , AcD (stable lead isotope)
Tl 81				β	Tl^{207} , AcC' (actinium C') 4.79 minutes

Fig. 2.3 - The actinium ($4n+3$) series. (From Nuclear and Radiochemistry (3)).

symbol of the element with the atomic number as a left subscript and the mass number as a superscript (in the U.S.A. it is usually written to the right, elsewhere to the left). The atomic number is often omitted because it is uniquely determined by the chemical symbol.

2.2 A QUICK REVIEW OF RADIOACTIVE DECAY AND GROWTH

The decay of a radioactive substance follows the exponential law $N=N_0e^{-\lambda t}$, where N is the number of unchanged atoms at time t , N_0 is the number present when $t=0$, and λ is a constant characteristic of the particular radioactive species.

The decay rate, $-dN/dt$, is set proportional to the number of atoms present: $-dN/dt = \lambda N$. On integration the result is $\ln N = -\lambda t + \alpha$, and the constant α is evaluated from the limit $N=N_0$ when $t=0$: $\alpha = \ln N_0$. Combining these terms we have: $\ln (N/N_0) = -\lambda t$, or $N/N_0 = e^{-\lambda t}$. The characteristic of a radioactive decay may be given in terms of half-life $t_{1/2}$, which is the time required for an initial (large) number of atoms to be reduced to half that number by transformation. Thus, at time $t=t_{1/2}$, $N=N_0/2$ and $\ln(1/2) = -\lambda t_{1/2}$ or $t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.69315}{\lambda}$. However, in practical work the number of atoms N is not directly measured. The usual procedure is through indirect methods, such as measurement of its electric, photographic, or other effect, which yield a quantity proportional to its λN . The term used for this quantity is 'activity' A , with $A = c\lambda N = c(-dN/dt)$, where c is the coefficient for instrumentation efficiency. Care must be taken to keep the efficiency factor constant throughout a series of measurements. The decay law can be written as $A=A_0e^{-\lambda t}$ (3), and is commonly used in that form.

2.2.1 Secular Equilibrium

In an undisturbed sample containing N_1 atoms of a parent isotope, a steady state is established in which the rate of formation of the daughter

isotope N_2 is just equal to their rate of decay. This means $-dN_1/dt = \lambda_2 N_2$, in this case because the formation of daughters is the same rate as the decay rate of the parent. Therefore, using the earlier relationship, it can now be expressed as $\lambda_1 N_1 = \lambda_2 N_2$, where λ_1 and λ_2 are the respective decay constants. This is sometimes more conveniently expressed as $N_1/(t_{1/2})_1 = N_2/(t_{1/2})_2$. This event is known as **secular equilibrium**. The same relationship, $\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3$, etc. may be used when several short-lived daughters arise from successive decays beginning from a long-lived parent. However, secular equilibrium only exists under one strict condition, that the material is **undisturbed**, that is, no daughter substances are removed or allowed to escape for a sufficient length of time for secular equilibrium to be established (Figure 2.4).

2.2.2 Transient Equilibrium

Let us consider the general equation for the decay of radioactive species N_1 and N_2 , where N_1 is the parent isotope and N_2 is the daughter. The behaviour of N_1 is $-(dN_1/dt) = \lambda_1 N_1$, and $N_1 = N_1^0 e^{-\lambda_1 t}$, where N_1^0 denotes the value of N_1 at time $t=0$. Now the daughter is formed at the rate of $\lambda_1 N_1$, which decays at the rate $\lambda_2 N_2$. Thus:

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

$$\frac{dN_2}{dt} + \lambda_2 N_2 - \lambda_1 N_1^0 e^{-\lambda_1 t} = 0 \quad \text{Eq 1}$$

The solution to this equation may be obtained by standard method and gives:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t} \quad \text{Eq 2}$$

where, N_2^0 is the value of N_2 at time $t=0$.

In the case where the parent isotope is longer-lived than the daughter ($\lambda_1 < \lambda_2$), after radioactive equilibrium is reached, the ratio of

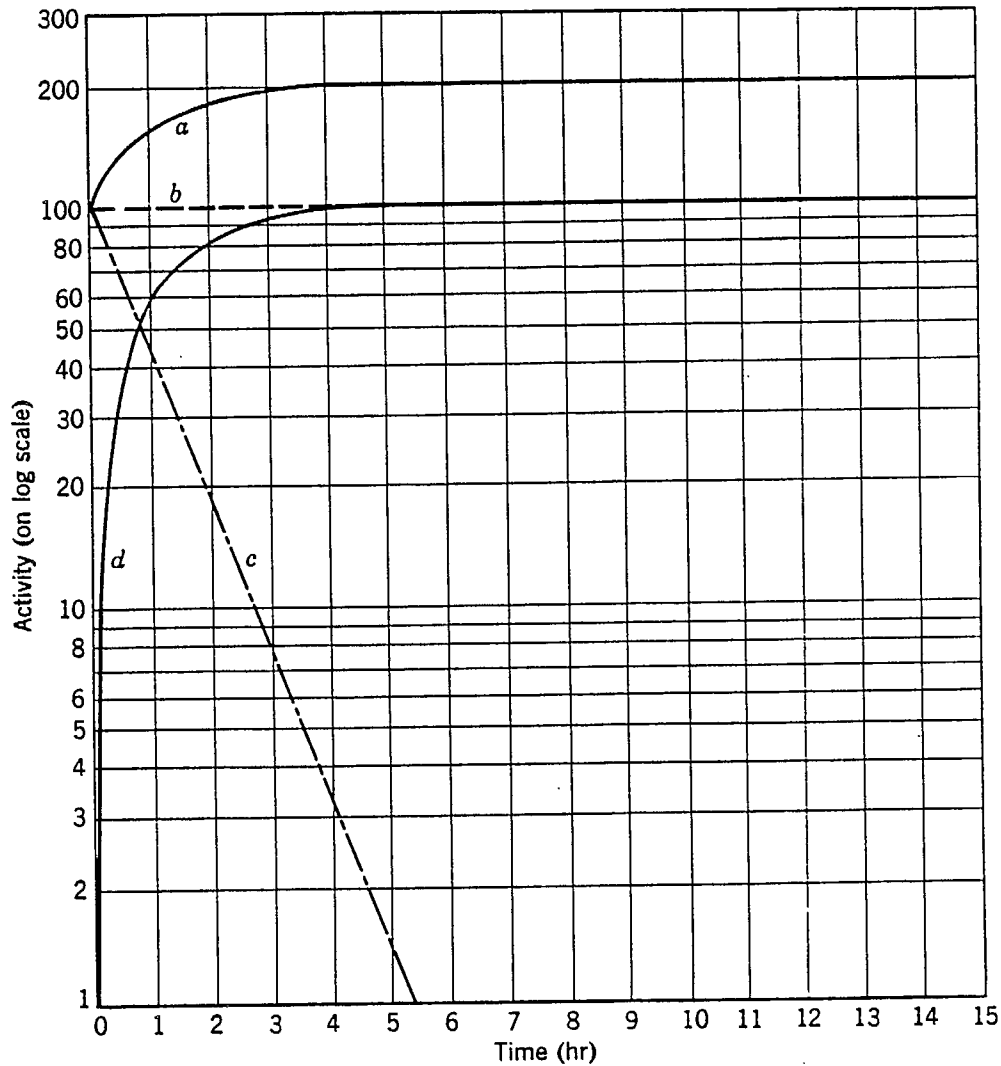


Fig. 2.4 - Secular equilibrium. (From Nuclear and Radiochemistry (3)).

- Note:
- a) total activity of an initially pure parent fraction;
 - b) activity due to parent ($t_{1/2} = \infty$); this is also the total daughter activity in parent-plus-daughter fractions;
 - c) decay of freshly isolated daughter fraction ($t_{1/2} = 0.80$ h);
 - d) daughter activity growing in freshly purified parent fraction.

the number of atoms and the ratio of the integration rates between parent and daughter become constant. From Equation 2 it can be seen that after t becomes sufficiently large, $e^{-\lambda_2 t}$ is negligible in comparison with $e^{-\lambda_1 t}$, and $N_2^0 e^{-\lambda_2 t}$ also becomes negligible, then:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 e^{\lambda_1 t}$$

and since, $N_1 = N_1^0 e^{\lambda_1 t}$, then $\frac{N_1}{N_2} = \frac{\lambda_2 - \lambda_1}{\lambda_1}$. Eq 3

The relation between two activities from $A_1 = c_1 \lambda_1 N_1$ and $A_2 = c_2 \lambda_2 N_2$ is found to be:

$$\frac{A_1}{A_2} = \frac{c_1 (\lambda_2 - \lambda_1)}{c_2 \lambda_2} \quad \text{Eq 4}$$

In most cases $c_1 = c_2$, therefore $A_1/A_2 = 1 - (\lambda_1/\lambda_2)$, and may have a value between 0 and 1, depending on the ratio of λ_1 and λ_2 (when it is in equilibrium the daughter activity will be greater than the parent activity by a factor of $\lambda_2/(\lambda_2 - \lambda_1)$).

In transient equilibrium, the condition is $\lambda_2 \gg \lambda_1$, and the sum of parent and daughter integration rates in an initially pure parent fraction goes through a maximum before transient equilibrium is achieved (Figure 2.5). The general condition for the total measured activity ($A_1 + A_2$) of an initially pure parent fraction to exhibit a maximum is $c_2/c_1 \gg \lambda_1/\lambda_2$. This condition holds regardless of the relative magnitudes of λ_1 and λ_2 . The condition of:

$$(\lambda_1 - \lambda_2)/\lambda_2 \leq c_2/c_1 \leq \lambda_1/\lambda_2$$

will give a maximum in the total measured activity which occurs at a negative time.

2.2.3 The Case of No Equilibrium

If the parent is shorter lived than its daughter ($\lambda_1 \gg \lambda_2$), it is evident that no equilibrium is achieved at any time. If the parent is made initially free of daughters, then as the parent decays the amount of daughters will rise, pass through a maximum, and eventually decay with the

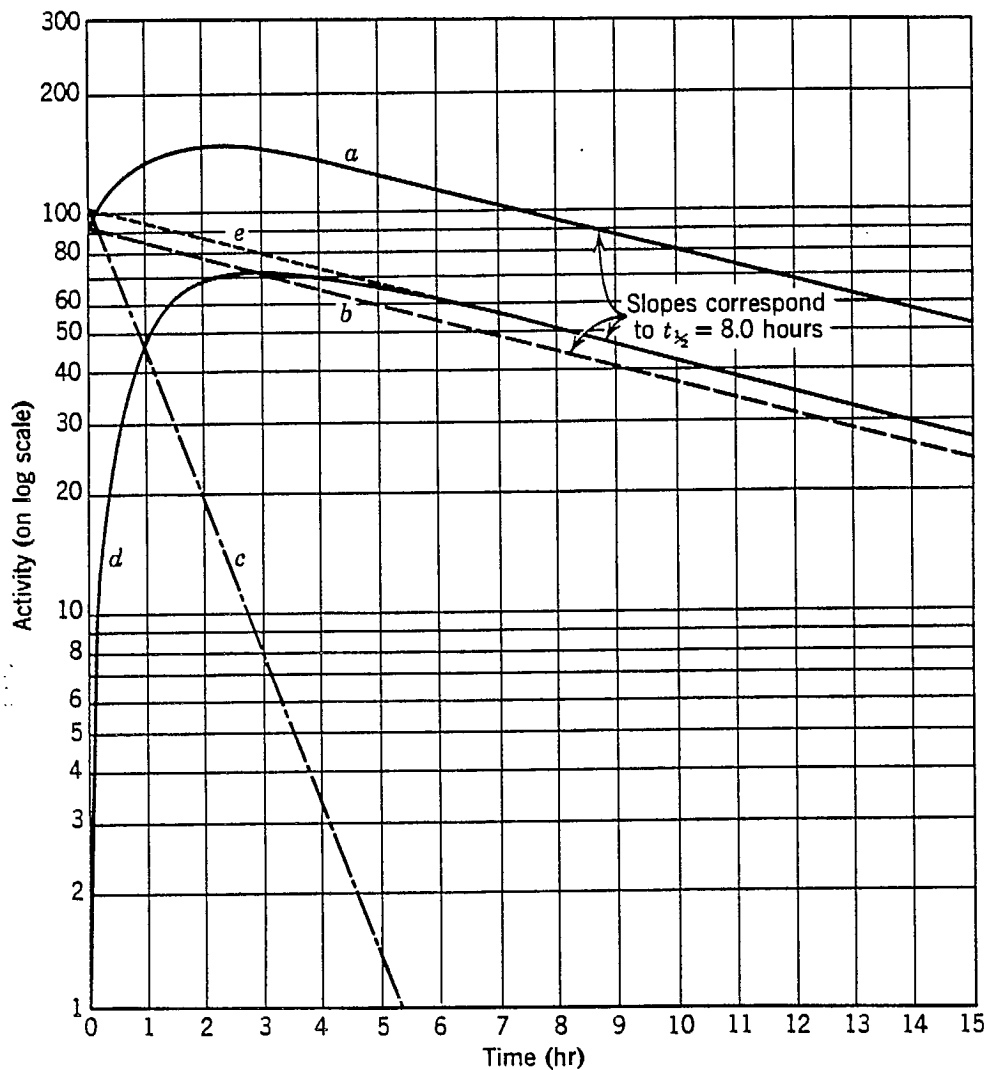


Fig. 2.5 - Transient equilibrium. (From Nuclear and Radiochemistry (3)).

Note: a) total activity of an initially pure parent fraction;
 b) activity due to parent ($t_{1/2} = 8.0$ h);
 c) decay of freshly isolated daughter fraction ($t_{1/2} = 0.80$ h);
 d) daughter activity growing in freshly purified parent fraction;
 e) total daughter activity in parent-plus-daughter fractions.

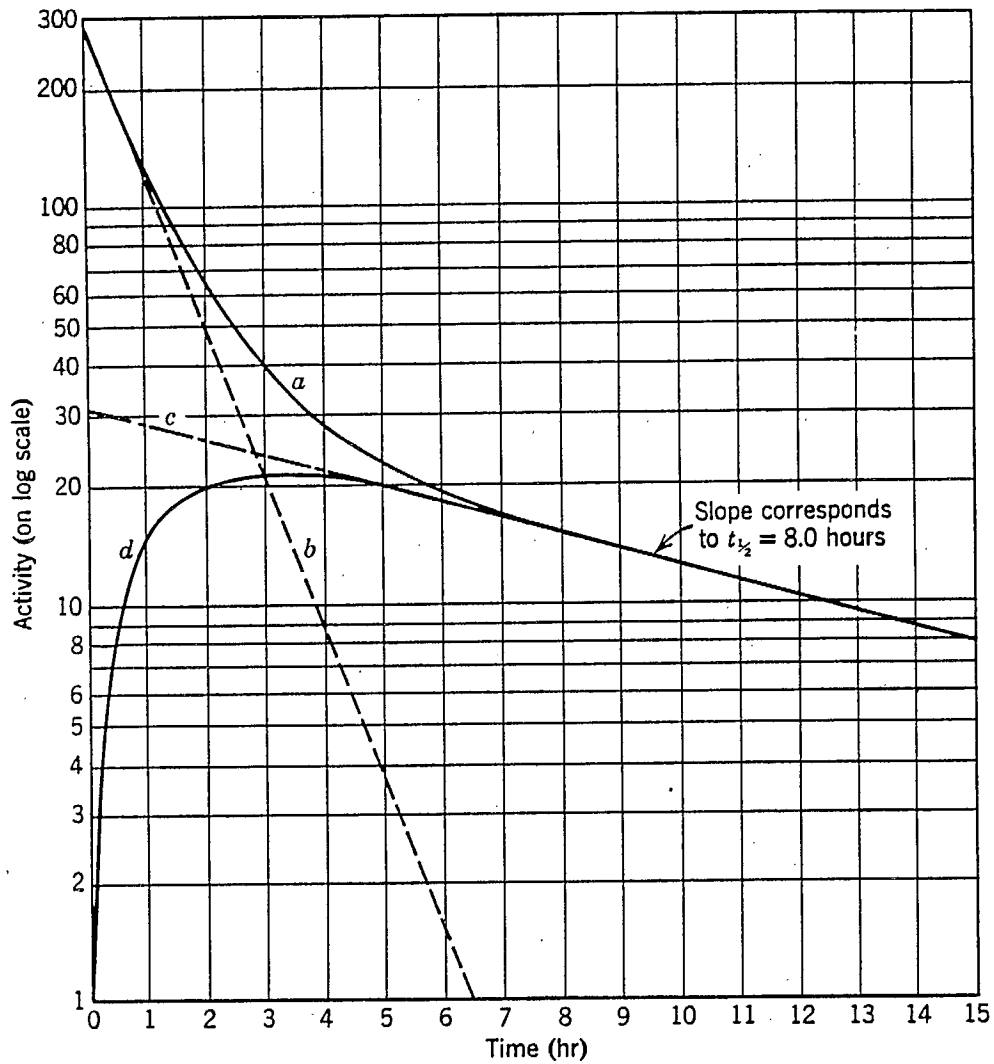


Fig. 2.6 - The case of no equilibrium. (From Nuclear and Radiochemistry (3)).

- Note:
- a) total activity;
 - b) activity due to parent ($t_{1/2} = 0.80$ h);
 - c) extrapolation of final decay curve to time zero;
 - d) daughter activity in initially pure parent.

characteristic half-life of the daughter (Figure 2.6). The final exponential decay of the daughter is extrapolated back to time $t=0$. This method is useful if $\lambda_1 \gg \lambda_2$, when the intercept measures the activity $c_2 \lambda_2 N_1^0$. The N_1^0 atoms give rise to N_2 atoms so early that N_1^0 may be set equal to the extrapolated value of N_2 at time $t=0$. The ratio of the initial activity, $c_1 \lambda_1 N_1^0$, to this extrapolated activity gives the ratio of the half-lives if the relationship between c_1 and c_2 is known:

$$\frac{c_1 \lambda_1 N_1^0}{c_2 \lambda_2 N_1^0} = \frac{c_1}{c_2} \times \frac{\lambda_1}{\lambda_2} = \frac{c_1}{c_2} \times \frac{(t_{1/2})_2}{(t_{1/2})_1} \quad \text{Eq 5}$$

If λ_2 is not negligible compared to λ_1 , then the ratio λ_1/λ_2 can be replaced by $(\lambda_1 - \lambda_2)/\lambda_2$ and the expression involving half-lives changes accordingly.

The transient and no equilibrium cases are often analyzed in terms of time, t_m , for the daughter to reach its maximum activity when growing in a freshly separated parent fraction.

By differentiating Equation 2, it was found that:

$$\frac{dN_2}{dt} = - \frac{(\lambda_1)^2}{\lambda_2 - \lambda_1} N_1^0 e^{-\lambda_1 t} + \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} N_1^0 e^{-\lambda_2 t}$$

and by setting $dN_2/dt=0$ when $t=t_m$, then:

$$\frac{\lambda_2}{\lambda_1} = e^{(\lambda_2 - \lambda_1)t_m} \quad \text{or} \quad t_m = \frac{2.303}{\lambda_2 - \lambda_1} \log \frac{\lambda_2}{\lambda_1}$$

At this time the daughter decay rate, $\lambda_2 N_2$, is equal to the rate of formation, $\lambda_1 N_1$ (Figures 2.4, 2.5 and 2.6).

It should be noted that time, t_m , is infinite for secular equilibrium (3).

2.2.4 Many Successive Decays

If we consider a chain of three or more radioactive products, it is clear that the equations already derived for N_1 and N_2 as functions of time are valid, and N_3 may be found by solving the new differential equation:

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3 \quad \text{Eq 5}$$

This is entirely analogous to the equation for dN_2/dt , but the solution calls for more labour, since N_2 is a much more complicated function than N_1 . The next solution for N_4 is still more tedious. H. Bateman (3) has given the solution for a chain of n members with the special assumption that at $t=0$ the parent substance alone is present, that is, $N_2^0 = N_3^0 = \dots N_n^0 = 0$. This solution is:

$$N_n = C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \dots C_n e^{-\lambda_n t},$$

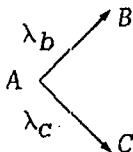
$$C_1 = \frac{\lambda_1 \lambda_2 \dots \lambda_n - 1}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1) \dots (\lambda_n - \lambda_1)} N_1^0,$$

$$C_2 = \frac{\lambda_1 \lambda_2 \dots \lambda_n - 1}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2) \dots (\lambda_n - \lambda_2)} N_1^0. \quad \text{etc.} \quad \text{Eq 6}$$

If we do require a solution to the more general case with $N_2^0, N_3^0 \dots N_n^0 \neq 0$, we may construct it by adding to the Bateman solution for N_n in an n -membered chain, a Bateman solution for N_n in an $(n-1)$ -membered chain with substance 2 as the parent, and, therefore, $N_2 = N_2^0$ at $t=0$, and a Bateman solution for N_n in an $(n-2)$ -membered chain, etc. (3).

2.2.5 Branching Decay

Another variant that is met in general decay schemes is the branching decay, illustrated by:



Here the two partial decay constants, λ_b and λ_c , must be considered when the general relations in either branch are studied because, for example, the substance B is formed at the rate $\lambda_b N_A$, but A is consumed at the rate $(\lambda_b + \lambda_c) N_A$. Notice that A can have but one half-life, in this case $t_{1/2} = 0.693/(\lambda_b + \lambda_c)$. By definition the half-life is related to the total rate of disappearance of a substance, regardless of the mechanism by which it disappears.

If the Bateman solution is to be applied to a decay chain containing branching decays, the λ 's in the numerators of the equations defining C_1 , C_2 , etc., should be replaced by the partial decay constants; that is, λ_i in the numerators should be replaced by λ_i^* , where λ_i^* is the decay constant for the transformation of the i th chain member to the $(i + 1)$ th member. If a decay chain branches, and subsequently the two branches are rejoined as in the natural radioactive series, the two branches are treated by this method as separate chains; the production of a common member beyond the branch point is the sum of the numbers of atoms formed by the two paths (3).

CHAPTER 3

PREVIOUS STUDIES OF AQUEOUS DISEQUILIBRIUM

Since there are many papers published on disequilibrium in soil, rock formations and water, only a small selection of typical studies will be reviewed in this thesis.

The natural decay of uranium series are named for the primary parent radionuclides: ^{238}U , ^{232}Th and ^{235}U . The ^{238}U -series, which is the usual one examined in the natural environments, begins with ^{238}U and ends with the stable isotope ^{206}Pb . The intermediate radionuclides from ^{238}U -series usually examined are ^{234}U , ^{230}Th , ^{226}Ra , ^{222}Rn and ^{210}Pb (Ivanovich et al. (62). The variations in activity ratios between daughter/parent, such as $^{234}\text{U}/^{238}\text{U}$ and $^{226}\text{Ra}/^{230}\text{Th}$, along the groundwater flowpath reflect the effects of radioactive decay and its interaction with groundwater and rock formations (over time and distance). An understanding of the decay series (2), and the complex behaviour of radionuclides in groundwater flow systems is necessary to understand the framework of this thesis (Chapter 2).

3.1 NON-MINING RELATED STUDIES

In a study of the Floridan Aquifer in northern Florida, Kaufman et al. (12) found that $^{234}\text{U}/^{238}\text{U}$ activity ratios ranged from 0.5 to 1.2 in uranium concentrations from 0.21 to 3.42 $\mu\text{g/L}$ with an activity ratio less than 1.0 indicating relative depletion of ^{234}U in groundwater.

Titayeva et al. (25) studied the disequilibrium of the ^{232}Th and the ^{238}U series in Polar URAL groundwater. Six $^{234}\text{U}/^{238}\text{U}$ activity ratios ranging from 0.9 to 1.27, and two $^{230}\text{Th}/^{238}\text{U}$ values ranging from 0.63 to 1.11 were reported. The mean values for uranium and thorium concentrations were 0.08

and 0.11 $\mu\text{g/L}$, respectively.

For the ^{232}Th series, four values of $^{228}\text{Th}/^{232}\text{Th}$ ranging from 7.5 to 17.7 were reported. In addition, the activity ratio of $^{232}\text{Th}/^{230}\text{Th}$ varied from 0.34 to 1.9. Titayeva et al. (25) found that enrichment of the ratio in water (<1) generally corresponded with depleted ratios in rock.

Kronfeld (13) reported sixteen $^{234}\text{U}/^{238}\text{U}$ activity ratios ranging from 0.92 to 12.25 in a study of groundwater from the Trinity Aquifer in central Texas. Uranium concentrations were between 0.02 and 0.12 $\mu\text{g/L}$ with two extreme concentrations of 1.870 and 15.46 $\mu\text{g/L}$. Kronfeld attributed the high activity ratios to alpha-recoil ejection from the solid phase uranium into groundwater during the decay of $^{238}\text{U}/^{234}\text{Th}$, and the subsequent rapid decay of the aqueous ^{234}Th ($t_{1/2} = 24.1$ days) to ^{234}Pa ($t_{1/2} = 1.2$ min) (63).

Surface waters in the recharge area were found to have relatively high uranium concentrations and activity ratios of $^{234}\text{U}/^{238}\text{U}$ around 1.0 to 2.0.

After tens of kilometers of subsurface flow, uranium concentrations were relatively low, and activity ratios rose from 5 to 12, and there appeared to be a trend of increasing activity ratios with greater distance.

Osmond and Cowart (22) described a scenario in the Carrizo Sandstone of southern Texas similar to Kronfeld's study.

Groundwater near a recharge outcrop had $^{234}\text{U}/^{238}\text{U}$ activity ratios less than 1.0, and uranium the concentration of approximately 1.0 ppb. The ratio rose to about 9.0, and the concentration decreased to about 0.01 ppb after flowing downdip for approximately 15 km. These trends were attributed to α -recoil (63), and the transition from relatively oxidizing to reducing conditions, respectively. Further downdip, the activity ratio decreased to about 2.0. This decrease was attributed to geochemical effects rather than decay effects. Frohlich et al. (8) simulated the differential behaviour of the two uranium isotopes at this site through sorption processes.

In Texas, Cowart (6) studied variations of $^{234}\text{U}/^{238}\text{U}$ activity ratios in the Edward Carbonate Aquifer. From the recharge area, and along tens of kilometers of flowpath, the activity ratio remained in the range 1.07 to 1.22 with uranium concentrations of 0.662 to 0.820 $\mu\text{g}/\text{L}$. Parallel to the flowpath, a portion of the aquifer containing slower-moving, higher TDS (Total Dissolved Solids) groundwater, had activity ratios of 1.62 to 2.66, and uranium concentrations of 0.046 to 0.101 $\mu\text{g}/\text{L}$. The mixing zone of the two waters had an anomalous uranium concentration of 0.071 to 4.59 $\mu\text{g}/\text{L}$, and the activity ratio ranged from 0.79 to 1.40 which Cowart attributed to changes in location of the redox boundary rather than simple mixing of the oxidized and reduced groundwater.

Laul et al. (14) studied the Palo Duro Basin groundwater (Texas) and reported radionuclide analyses of ^{238}U and ^{232}Th series in two brines.

The values for ^{238}U activity ratios were:

$$^{234}\text{Th}/^{238}\text{U} = 25-50$$

$$^{234}\text{U}/^{234}\text{Th} = 0.025 \text{ to } 0.050$$

$$^{234}\text{U}/^{238}\text{U} = 1.1 \text{ to } 1.3$$

$$^{230}\text{Th}/^{234}\text{U} = 0.19 \text{ to } 1.11$$

$$^{226}\text{Ra}/^{230}\text{Th} = 2.0 \times 10^4 - 3.0 \times 10^4$$

$$^{222}\text{Rn}/^{226}\text{Ra} = 0.8 \text{ to } 2.4$$

The activity ratios for ^{232}Th series were:

$$^{228}\text{Ra}/^{232}\text{Th} = 800 \text{ to } 2040$$

$$^{228}\text{Th}/^{228}\text{Ra} = 0.009 \text{ to } 0.010$$

$$^{228}\text{Th}/^{232}\text{Th} = 7 \text{ to } 23$$

$$^{224}\text{Ra}/^{228}\text{Th} = 90 \text{ to } 100$$

$$^{224}\text{Ra}/^{228}\text{Ra} = 0.9 \text{ to } 1.0$$

Cowart (5) examined the $^{234}\text{U}/^{238}\text{U}$ activity ratio and ^{226}Ra activities in groundwater in the Cambrian/Ordovician aquifers in the Tri-State region (Missouri, Kansas and Oklahoma) of the U.S.A. In the aquifer's calcium carbonate water, the usual ratio is around 7 to 10, and uranium concentrations around 0.3 to 0.5 $\mu\text{g}/\text{L}$. The flow is westward, and eventually

mixes with sodium chloride, H₂S-bearing (more reducing) groundwater. Within the mixing zone, ²²⁶Ra increases from around 0.5 Bq/L up to 0.32 Bq/L, uranium precipitates from 0.02 to 0.04 µg/L, and notably, uranium activity ratios remain essentially constant.

Although the decrease in uranium concentration is consistent with reduction in Eh (redox potential), Cowart could only speculate on explanations for a steady uranium ratio and an increase in ²²⁶Ra. If the uranium concentrations reported as pCi/L in Cowart's Table II (5) essentially represent ²³⁸U, then the calculated values for the ²²⁶Ra/²³⁴U ratio increase from 0.5 to about 1.5, and there is a sharp increase of 6 to 236 in the mixing zone. Under the assumption made for the calculation, this trend indicates that there is a relatively strong aqueous enrichment of ²²⁶Ra over ²³⁴U during mixing, probably synonymous with the parallel decrease in uranium concentration.

CBCL Limited (58) carried out a detailed study of a small, post glacial, uraniferous peat deposit at the base of a granitic talus slope near Portland Creek, Newfoundland. Piezometer nests were installed in a line perpendicular to the talus slope. The groundwater in the peat was low TDS, calcium-sodium-bicarbonate water with a pH constant near 6. Uranium decreased with distance from the slope by a factor of 1.7 from 415 µg/L. Many ratio values for the ²³⁸U series have been calculated and are shown here (Table B-5 (58)).

The values for ²³⁸U activity ratios were:

$$\begin{aligned}^{234}\text{U}/^{238}\text{U} &= 1.2 \text{ to } 1.04 \\^{230}\text{Th}/^{234}\text{U} &= 2.73 \times 10^{-3} \text{ to } 3.52 \times 10^{-2} \\^{226}\text{Ra}/^{230}\text{Th} &= >1.4 \text{ to } 0.38 \\^{210}\text{Pb}/^{226}\text{Ra} &= 10.15 \text{ to } 26.74 \\^{210}\text{Po}/^{210}\text{Pb} &= 0.50 \text{ to } 0.64\end{aligned}$$

The significant variations in ²³⁰Th/²³⁴U and ²¹⁰Pb/²²⁶Ra may be the result of an increase in the activity of ²³⁰Th. The relatively high values

for $^{210}\text{Pb}/^{226}\text{Ra}$ suggest the accumulation of ^{222}Rn gas in the past.

The activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the uraniferous peat samples varied from 0.92 to 1.26, averaging 1.13. The ^{234}U tends to be more soluble than ^{238}U because of the ejection of the ^{234}U daughter from the uranium bearing mineral by the effect of mass recoil when ^{238}U decays. When the uranium derived from the mineralized source is subsequently precipitated it will retain the same elevated $^{234}\text{U}/^{238}\text{U}$ ratio. Being a recent deposit (i.e., less than 10,000 to 15,000 years old), little time has been available for the ingrowth of radionuclide daughter products of uranium. The deposition of uranium with the peat deposit is considered to be an on-going process.

The activity ratios from the above papers are summarized in Table 3.1. The most obvious observation is that activity ratios spanning a natural decay series are not usually examined along the groundwater flowpath. Nevertheless, the available data indicate that a high degree of variability exists between sites and significant changes in ratio can occur along a flowpath at a site. This variability reflects the numerous processes affecting the migration of radionuclides in groundwater (1).

3.2 URANIUM MINING RELATED STUDIES

Because of the potential of near-surface contamination from uranium tailings impoundments, several detailed studies have been undertaken near the impoundments. There are several studies of major ion chemistry and total concentrations in the vicinity of uranium tailings. However, only a few provide a number of ratios along a flowpath, and most only report ^{226}Ra activities as it is considered the most toxic component in the tailings seepage.

Cherry et al. (4) reviewed several uranium tailings sites and listed chemical analyses for tailings porewater, or pond water, at three sites in

Wyoming, U.S.A. and one site in Canada. At the four sites, tailings water pH was limited to 1.8 to 2.7; TDS ranged from 12,200 to 18,000 mg/L; and major ion composition could generally be described as predominantly iron, hydrogen and sulphate. If the reported 'U=nat. (mg/L)' is assumed to essentially represent ^{238}U , then calculated $^{230}\text{Th}/^{234}\text{U}$ ratios are 12.29 and 23.46 for two Wyoming sites, and 0.01 for the Ontario site (the Nordic Main Impoundment will be discussed later in this thesis). The Wyoming values generally agree with the ratios calculated from 'typical concentrations in tailings solutions' (Table 1a of reference 23): 1.00 for $^{230}\text{Th}/^{238}\text{U}$, and 27.78 for $^{230}\text{Th}/^{234}\text{U}$. Based on available activities, two $^{226}\text{Ra}/^{230}\text{Th}$ ratios for Wyoming sites are 0.011 and 0.058, which are over an order of magnitude higher than Taylor's typical ratio of 0.0027, and the Nordic Main site in Ontario provides a ratio of 7.37. The differences in ratios in the upper portion of the ^{238}U series between the Wyoming sites and the Nordic Main site can mostly be attributed to four orders of magnitude higher ^{230}Th activities at the Wyoming sites. The three available $^{210}\text{Pb}/^{226}\text{Ra}$ ratios in Wyoming are 0.17, 0.57, and 14.57. Taylor's typical ratio is 1.00, and the Nordic Main ratio is 12.14.

Because of the variability in ore characteristics passing through a mill, the variability in mill processing, and the physical and chemical variabilities produced during and after discharging a slurry to an impoundment, the activity ratio can be expected to vary with lateral and vertical distance in an impoundment.

Moffett and Tellier (15) who examined the Nordic West Arm tailings adjacent to the Nordic Main Impoundment, classified one 'high acid' and two 'low acid' lateral regions within the tailings pile. The high acid region displays no definite trends along its length, and assuming that the total uranium represents ^{238}U , then the $^{230}\text{Th}/^{238}\text{U}$ activity ratio was 4 to 22. The

$^{226}\text{Ra}/^{232}\text{Th}$ ratio was around 5.8×10^{-4} to 1.7×10^{-3} , and the $^{210}\text{Pb}/^{226}\text{Ra}$ ratio varied strongly from 2.05 to 240.88. For the ^{232}Th series, the $^{228}\text{Th}/^{232}\text{Th}$ activity ratio was around 0.36 to 2.00, and the ^{227}Th interference was noted for some analyses. For comparison of the magnitude of the two thorium series, the $^{228}\text{Th}/^{230}\text{Th}$ rate was around 0.031 to 0.27.

Highland et al. (10) examined the chemical composition of 'tailings liquor' from one of the Wyoming ponds mentioned previously and the average groundwater from a point approximately 1 km down gradient from the pond. From their data, pH increases from 1.9 in the pond to 6.2 in the distant groundwater. TDS decreases from 15,000 to 1,820 mg/L, $^{230}\text{Th}/^{238}\text{U}$ decreases from 1.28 to 0.012, and $^{210}\text{Pb}/^{226}\text{Ra}$ decreases from 14.27 to 0.79. The change in ratios is caused by the significant decrease in the activities of radionuclides, particularly a nearly three orders of magnitude decrease in ^{230}Th . Hoffman and Playton (11) examined another Wyoming tailings site. The chemical data suggests an intermediate point in the above 'flowpath' of Highland et al. (10): pH = 3.6 to 3.7; TDS 15,984 to 17,970 mg/L; $^{226}\text{Ra}/^{230}\text{Th} = 0.001$ to 0.098; $^{210}\text{Pb}/^{226}\text{Ra} = 8.41$ to 8.73; and $^{210}\text{Po}/^{210}\text{Pb} = 0.0$ to 0.33.

Taylor and Antonmaria (24), Taylor (23) and Haji-Djafari and Antonmaria (9), describe a Wyoming tailings area which is sufficiently instrumented to describe a four point flowpath:

Distance from Piezometer	Tailings (m)	pH	TDS (mg/L)
Tailings pond	0	1.95	11810
WM-6S	0	3.00	4274
WN-14H	390	5.50	4042
WN-1HDA	1070	7.25	1395

The corresponding activity ratios are listed in Table 3.2, and good trends are apparent for decreasing $^{230}\text{Th}/^{238}\text{U}$, generally constant $^{226}\text{Ra}/^{230}\text{Th}$, and decreasing $^{210}\text{Po}/^{226}\text{Ra}$. The sharpest change in ratios occurs between WN-6S and WN-14H where pH increases from 3.00 to 5.50.

Veska studied groundwater seepage emanating from a uranium-mine waste rock pile near Bancroft, Ontario. A set of piezometers were chosen from Veska's Appendix 5.12 (26) to depict general trends. The chosen piezometers are:

Piezometer	Source Distance (m)	pH	Elec. Cond. ($\mu\text{S}/\text{cm}$)
L	0	3.7	900
GR4	9	4.6	400
GR6	12	5.0	304
M3-4.76	20	6.0	442
M4-8.83	30	6.2	305
M5-11.00	39	6.5	320
M6-14.00	47	6.8	320
M7-15.17	56	7.5	290
M8-14.59	76	7.6	300
M9-15.25	94	7.6	320

Variations in activity ratios show trends of increasing $^{234}\text{U}/^{238}\text{U}$, decreasing $^{226}\text{Ra}/^{234}\text{U}$, and initially decreasing then increasing $^{210}\text{Pb}/^{226}\text{Ra}$, Table 3.2. Sharp changes in $^{226}\text{Ra}/^{234}\text{U}$ can be attributed to the factor of five decreases in ^{226}Ra . The sharp rise in $^{234}\text{U}/^{238}\text{U}$ between GR6 and M3, and the continued trend of relative ^{234}U enrichment were attributed to a scenario requiring preferential leaching of ^{234}U from the waste rock in the 6+ oxidation state (UO_2^{2+}), stronger adsorption of ^{238}U (as U^{4+}) over ^{234}U (as UO_2^{2+}) in the sand aquifer, and little redox exchange between the 4+ and 6+ states (26).

Table 3.2 summarizes the activity ratios reviewed in this section. Based on the limited data for $^{234}\text{U}/^{238}\text{U}$, there is near equilibrium (0.86 to 2.07) in tailings water unlike that generally noted in Table 3.1. The $^{234}\text{U}/^{238}\text{U}$ ratios of Veska (26) show a trend of increasing value along the flow path. Below ^{234}U in the ^{238}U -series, notable disequilibrium occurs, but general trends can be seen. Ratios for $^{230}\text{Th}/^{238}\text{U}$ are typically greater than 4 in tailings, but decrease to less than 0.1 in the flow path. The $^{226}\text{Ra}/^{230}\text{Th}$ ratios are variable and usually less than 0.13. The ratio of $^{226}\text{Ra}/^{234}\text{U}$ spans a wider range and data of Veska (26) indicates the ratio decreases along the flow path. The $^{210}\text{Pb}/^{226}\text{Ra}$ and $^{210}\text{Po}/^{226}\text{Ra}$ ratios are variable, but appear to be somewhat less than 1 in most groundwater seepage. There are insufficient data to evaluate the lowest portion of the ^{238}U series. Furthermore, no data could be located on variations in ratios along flow paths for the ^{232}Th and ^{235}U series.

3.3 HYDROGEOCHEMISTRY, NORDIC MAIN TAILINGS, ONTARIO. FIELD STUDY OF RADIOISOTOPE DISEQUILIBRIUM IN THE SEEPAGE AREA.

In studies of groundwater seepage from the Nordic Main tailings, near Elliot Lake, Ontario, Morin (16) identified four seepage areas around the impoundment perimeter. The major seepage area, seepage area 'A', was studied from 1979 to 1984. In particular, Morin et al. examined the effect of the various processes of sorption, mineral precipitation-dissolution, coprecipitation, aqueous speciation, and decay of radionuclide migration in area 'A'. The seepage plume is located just beneath the water table at a general depth of 1-2 metres. For simplicity of discussion, one piezometer in each nest was selected to represent the centre of the plume and the flowpath that passes through each of these piezometers is called the 'centreline' of the plume. Morin also recognized that a high degree of analytical error is

expected when dealing with the measurement of relatively low radionuclide activities in high TDS and geochemically reactive water. The study was concentrated mostly along a groundwater flowpath of 20.8 m. Because the groundwater requires approximately 24 days to travel this length of flowpath, the observed variations during one sampling event are predominantly the result of relatively fast geochemical and radioisotope decay processes and are not closely related to temporal variations in the radioactive source (the solid tailings), which can account for variations along flowpaths in slow moving groundwater systems. However, fluctuation in annually measured values at each piezometer can reflect temporal source fluctuations. The $^{234}\text{U}/^{238}\text{U}$ ratio, for example, increases from 1.23 to 3.92, and $^{227}\text{Th}/^{227}\text{Ac}$ decreases from 1.88 to 1.26 along the flowpath. Also isotopes of an element display different aqueous behaviour, which is dependent on the relative contribution of the isotope to the total element concentration in the molal bases. For example, ^{230}Th steadily decreases along the flowpath from 1370 to <19 mBq/L, whereas ^{228}Th initially increases from 130 to 190 mBq/L then decreases to <19 mBq/L. The variations in radionuclide activity ratios along groundwater flowpaths are controlled by the particular set of geochemical and decay mechanisms that regulate each isotope of each element (Morin et al.)(1).

3.4 SUMMARY

In general, most of the authors observed similar disequilibrium occurrences, decrease or increase of ratios between daughters and their parent isotopes with distance from the source. However, the explanation of the disequilibrium mechanism itself was not very satisfactory.

Titayeva et al. (25) found that enrichment of ratios in water (values greater than 1) generally corresponded with depleted ratios in rock.

Kronfield (13) found that activity ratios rose to 5-12, and there

appeared to be a trend of increasing activity ratio with further distance. The uranium concentrations were relatively low after tens of kilometres of subsurface flow. Kronfield also attributed the high ratios to alpha-recoil ejection from solid phase uranium into groundwater.

Osmond and Cowart (22) attributed the disequilibrium to geochemical effects rather than decay effects.

Cowart (6) attributed the disequilibrium to changes in the location of the redox boundary rather than simple mixing of oxidized and reduced groundwater.

CBCL Ltd. (58) made an interesting observation on the uraniumiferous peat sample, in which the activity ratio of $^{234}\text{U}/^{238}\text{U}$ averaged 1.13. The explanation by CBCL Ltd. was that ^{234}U tends to be more soluble than ^{238}U due to the ejection of the ^{234}U daughter from the uranium bearing mineral by the effect of mass recoil when ^{238}U decays. When the uranium derived from the mineralized source is subsequently precipitated it will retain the same elevated $^{234}\text{U}/^{238}\text{U}$ ratios. The deposition of the uranium with the peat deposit is considered to be an on-going process. Being a recent deposit (i.e., less than 10,000 years old) little time has been available for the ingrowth of radionuclide daughter products of uranium.

Similar relations were found on studying disequilibrium related to uranium mining activities, where the ratios could increase or decrease with distance away from the source (1,19-21). Morin examined the effect of the various processes of sorption, mineral precipitation-dissolution, aqueous speciation and decay on radionuclide migration. This type of pathway analysis study was probably the best approach towards a more reasonable explanation of disequilibrium mechanism.

Taylor (23) listed typical ratios of 1.0 for $^{210}\text{Bi}/^{210}\text{Pb}$ and $^{210}\text{Po}/^{210}\text{Bi}$, indicating that rapidly attained equilibrium is often expected

because of short half-lives in this portion of the series. This is an important statement and will be discussed further elsewhere in this thesis, especially in dealing with sampling, sample treatment, timing and analytical procedures.

Veska (26) found that the sharp rise in $^{234}\text{U}/^{238}\text{U}$, and the continued trend of relative ^{234}U enrichment were attributed to a scenario requiring preferential leaching of ^{234}U from the waste rock in the 6+ oxidation state (UO_2^{2+}), stronger adsorption of ^{238}U (as U^{4+}), over ^{234}U (as UO_2^{2+}) in the sand aquifer, and little redox exchange between the 4+ and 6+ states (26). The validity of this explanation was tested with the aid of the cell model, in which a solution spiked with $^{238}\text{U}(\text{IV})$, $^{238}\text{U}(\text{VI})$, $^{232}\text{Th}(\text{IV})$, $^{226}\text{Ra}(\text{II})$ and $^{210}\text{Pb}(\text{II})$ was added by increments to the groundwater solution.

It is possible that during the decay chain from ^{238}U to ^{234}U , the uranium isotopes lost 2 of their electrons. As an atom progressively decays through a decay series, its chemoelectrical properties, including valence, vary. However, for example, the upper portion of the ^{238}U decay series indicates the following transitions for a ^{238}U atom: $^{238}\text{U}^{4+,5+\text{or}6+} \rightarrow ^{234}\text{Th}^{4+} \rightarrow ^{234}\text{Pa}^{5+} \rightarrow ^{234}\text{U}^{4+,5+\text{or}6+} \rightarrow ^{230}\text{Th}^{4+} \rightarrow ^{226}\text{Ra}^{2+}$ (Morin) (27). Therefore, as was shown by Morin, ^{238}U or ^{234}U could exist as (IV), (V) or (VI) valence and the reasoning as described by Veska as preferential leaching could be misleading and misconstrue the real cause of disequilibrium mechanisms in the field.

In addition, it does not appear feasible to practically detect preferential leaching, as described by Veska, in the groundwater sample itself, without the higher concentration of a standard spiked solution. However, Veska's experiment to solve the mystery of disequilibrium was interesting and it demonstrated the level of difficulties involved in solving these complex problems.

TABLE 3.1

Summary of activity ratios at non-mining-related sites
(From Morin and Cherry (1)).

<u>238U DECAY SERIES</u>							
Site	Place in flow sys.	$\frac{234\text{Th}}{238\text{U}}$	$\frac{234\text{U}}{238\text{U}}$	$\frac{230\text{Th}}{234\text{U}}$	$\frac{226\text{Ra}}{234\text{U}}$	$\frac{226\text{Ra}}{230\text{Th}}$	OTHER
Floridan Aq	gen*		0.5-1.2				
Polar Ural	gen		0.9-1.27	0.90, 0.56			
Trinity Aq	recharge 10's of km		1-2 5-12				
Carrizo Sdst	recharge 15 km >15 km		<1.0 9 approx. 2 approx				
Edwards Aq	gen, oxid gen, red gen, mixed		1.07-1.22 1.62-2.66 0.79-1.40				$\frac{222\text{Rn}}{226\text{Ra}}$
Palo Duro Bas	gen	25-50	1.1-1.3	0.19-1.11		$2-3 \times 10^4$	0.8-2.4
Tri-St. Reg	unmixed mixed		7-10 7-10		0.5-1.5 6-236		$\frac{210\text{Pb}}{226\text{Ra}}$ $\frac{210\text{Po}}{210\text{Pb}}$
Newfoundland	begin. 135 m		1.20 1.04	0.00273 0.0352		>1.4 0.38	10.15 26.74
							0.50 0.64

(* gen = general, no clear flowpath)

232Th DECAY SERIES

Site	$\frac{228\text{Ra}}{232\text{Th}}$	$\frac{228\text{Th}}{232\text{Th}}$	$\frac{224\text{Ra}}{228\text{Th}}$
Polar Ural		7.5-17.7	
Palo Duro Bas	800-2040	7-23	90-100

235U DECAY SERIES

Site	$\frac{231\text{Pa}}{235\text{U}}$
Newfoundland	<0.04

TABLE 3.2

Summary of activity ratios at uranium-mining-related sites
(From Morin and Cherry (1)).

Ref.-Location	$\frac{^{234}\text{U}}{^{238}\text{U}}$	$\frac{^{230}\text{Th}}{^{238}\text{U}}$	$\frac{^{226}\text{Ra}}{^{230}\text{Th}}$	$\frac{^{226}\text{Ra}}{^{234}\text{U}}$	$\frac{^{210}\text{Pb}}{^{226}\text{Ra}}$	OTHER
Cherry et al. (1982)						
Wyo. tailings		12.29- 23.46*	0.011- 0.058		0.17,0.57; 14.57	$\frac{^{210}\text{Bi}}{^{210}\text{Pb}}$ $\frac{^{210}\text{Po}}{^{210}\text{Bi}}$
Nordic Main "Typical"	1.00	0.01* 27.78	7.37 0.0027		12.14 1.00	$\frac{^{210}\text{Pb}}{^{210}\text{Bi}}$ 1.00 1.00
Moffett & Tellier (1978)						
High-acid		4-22*	0.00058- 0.0017		2.05- 240.88	
Low-acid					0.25-0.60	
Highland et al. (1981)						
Tailings		12.29	0.012		14.27	$\frac{^{210}\text{Po}}{^{210}\text{Pb}}$
"Intermed. data"		4.09*	0.001- 0.098		8.41- 8.73	0-0.33
Average groundwater		0.076	1.28		0.79	
Taylor (1980)						
Tailings		11.73*	0.115			$\frac{^{210}\text{Po}}{^{226}\text{Ra}}$
WN-6S		3.73*	0.116			0.196
WN-14H		0.034*	0.0031			0.015
WN-1HDA		0.068*	0.125			0.0
						0.0
Veska (1983)						
L	0.97			0.33	1.15	
GR4	0.87			1.63	0.05	
GR6	0.86			1.23	0.27	
M3	1.20			0.35	0.66	
M4	1.28			0.15	<0.45	
M5	1.27			0.05	<1.00	
M6	1.57			0.08	<0.38	
M7	1.75			0.10	<0.61	
M8	1.88			0.10	<0.67	
M9	2.07			<0.65	-	

* = ^{238}U calculated from total uranium

CHAPTER 4

STUDY OF NORDIC MAIN TAILINGS

In Canada alone there are approximately 120 million tonnes of uranium tailings spread over an area of approximately 635 hectares. It is expected that by the time the present uranium ore reserves are mined there will be about 1 billion tonnes of uranium tailings which will require proper management technology, **without much human involvement, in limiting the release of contaminants to the environment.**

Hydrogeochemical investigations of the Nordic Main Tailings area have been on-going since 1978. Radioisotope and chemical profile studies of solid and liquid phases have been conducted on various sites.

Some studies have also focused on the identification and quantification of the routes of radionuclide escape from the tailings and their entry into the biosphere. However, in this thesis, only a part of the tailings studies related to the disequilibrium of radioisotopes will be discussed.

4.1 FIELD STUDY STRATEGY

The first phase of an investigation of the chemical composition (including radioisotopes) of subsurface water and the hydrochemical processes in and near the abandoned Nordic tailings at Elliot Lake, Ontario, was conducted in 1979. The study had three components which included:

1. Monitoring of the groundwater flow and the chemical composition of water in the tailings;
2. Monitoring the flow pattern and chemical composition of groundwater in the sandy aquifer adjacent to the tailings embankment; and

3. Development of a hydrogeological and hydrochemical framework to account for the observed chemical profiles in the tailings and the contaminant distribution in the aquifer (28-30).

Most of the groundwater research was carried out through team work between the University of Waterloo, Rio Algom Ltd., and the Mining Research Laboratory, Elliot Lake, Canada Centre for Mineral and Energy Technology (CANMET), Energy, Mines and Resources Canada.

However, since solid tailings and porewater exist together, the chemical and radioisotope distribution profile of the solid tailings was also determined in a supplementary study to the groundwater research (31-32).

4.2 MINING HISTORY AND TAILINGS DEPOSITION

The Nordic Mine is one of ten present and past uranium mining and milling operations in the Elliot Lake area, Figure 4.1. It was in continuous production from 1957 up to its closure in 1968. The mine was operated with a milling capacity ranging from 2,500 tonnes to 3,300 tonnes per day, and milled a total of 12,300,000 tonnes of ore before closure (Robertson (33)).

The uranium ore is a quartz pebble conglomerate containing 5-15% pyrite. The principal ore minerals are uraninite (pitchblende), brannerite and minor monazite. The ore contains 0.11% U_3O_8 , 0.020% ThO_2 and 0.056% rare earths (yttrium, cerium and neodymium oxides).

The ore milling process at Nordic Mine, and at currently operating mines, involves grinding the ore to 50% minus 200 mesh followed by acid digestion of oxide ore minerals with dilute sulphuric acid. The resulting pregnant liquor is passed through ion-exchange columns to remove the uranium. The uranium is eluted from the ion exchange columns using nitric acid, and then reacted with ammonia to precipitate ammonium diuranate, known as yellow cake. The process effluent water containing SO_4^{+2} , NO_3^{-1} , NH_4^{+1} , Th^{2+} and

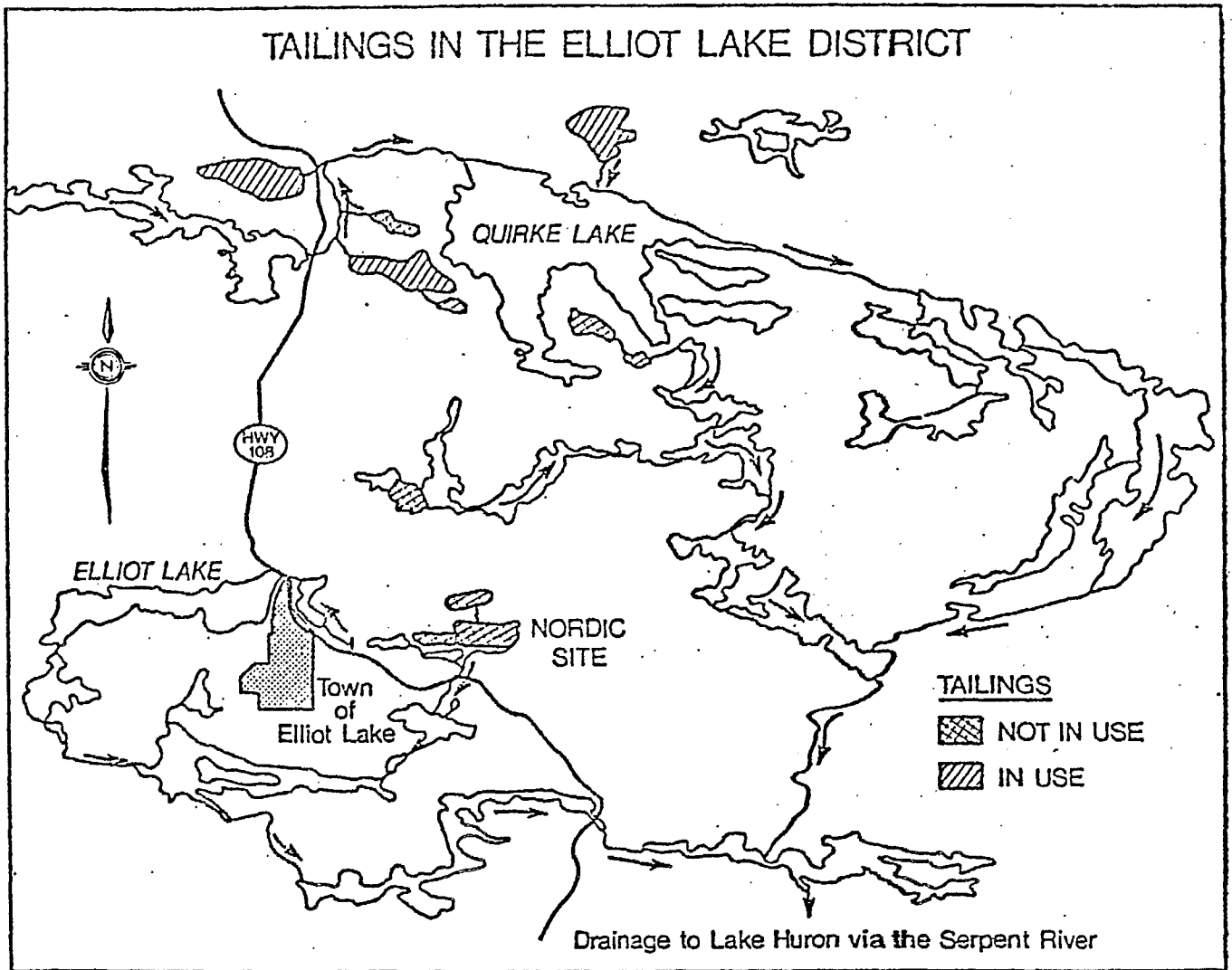


Fig. 4.1 - General location and plan.
(From Blair et al. (28)).

^{226}Ra is collected at the end of the mill circuit along with the leached ore pulp. The effluent water and pulp were neutralized with limestone and lime and discharged to the tailings pond. The average U_3O_8 recovery was 95%, and to a certain extent, ThO_2 and rare earths were recovered from the leach liquor.

The tailings consists of a mixture of sand and silt, 50% passing the 200 mesh size, and is comprised of quartz, feldspar, approximately 5% pyrite and minor sericite. Gypsum formed from precipitation following lime neutralization of the mill effluent was also present. The tailings were slurried with neutralized mill effluent and discharged to the tailings pond where settling took place in broad low angle alluvial fans. The coarse sand size fraction settled close to the point of discharge, and the fine silt-size or slime fraction settled in the distal area of the impoundment. The coarse sized fraction progressively advanced out over the fine distal fraction in a deltaic fashion (28).

The Nordic Main Tailings Pond occupies an area of approximately 85 hectares, measuring 1460 m east-west, and 550 m north-south, to a depth of 10 m, Figure 4.2. The tailings were deposited in a level valley on an area formerly occupied by a spruce bog. The tailings pond is confined along the north and west perimeter by prominent bedrock ridges. The eastern and southern perimeter of the pond are contained by a 10 to 12 m high rockfill dam.

Deposition of the tailings began in 1957 at the southwest corner of the basin and extended outward to the north and east. The Nordic tailings pond was filled to its capacity and became inactive with closure of the mine in 1968. In 1975, a rehabilitation program by revegetation on the surface of the tailings was started. The surface of the tailings was first treated with lime and crushed limestone and regrowth of grasses was established over the

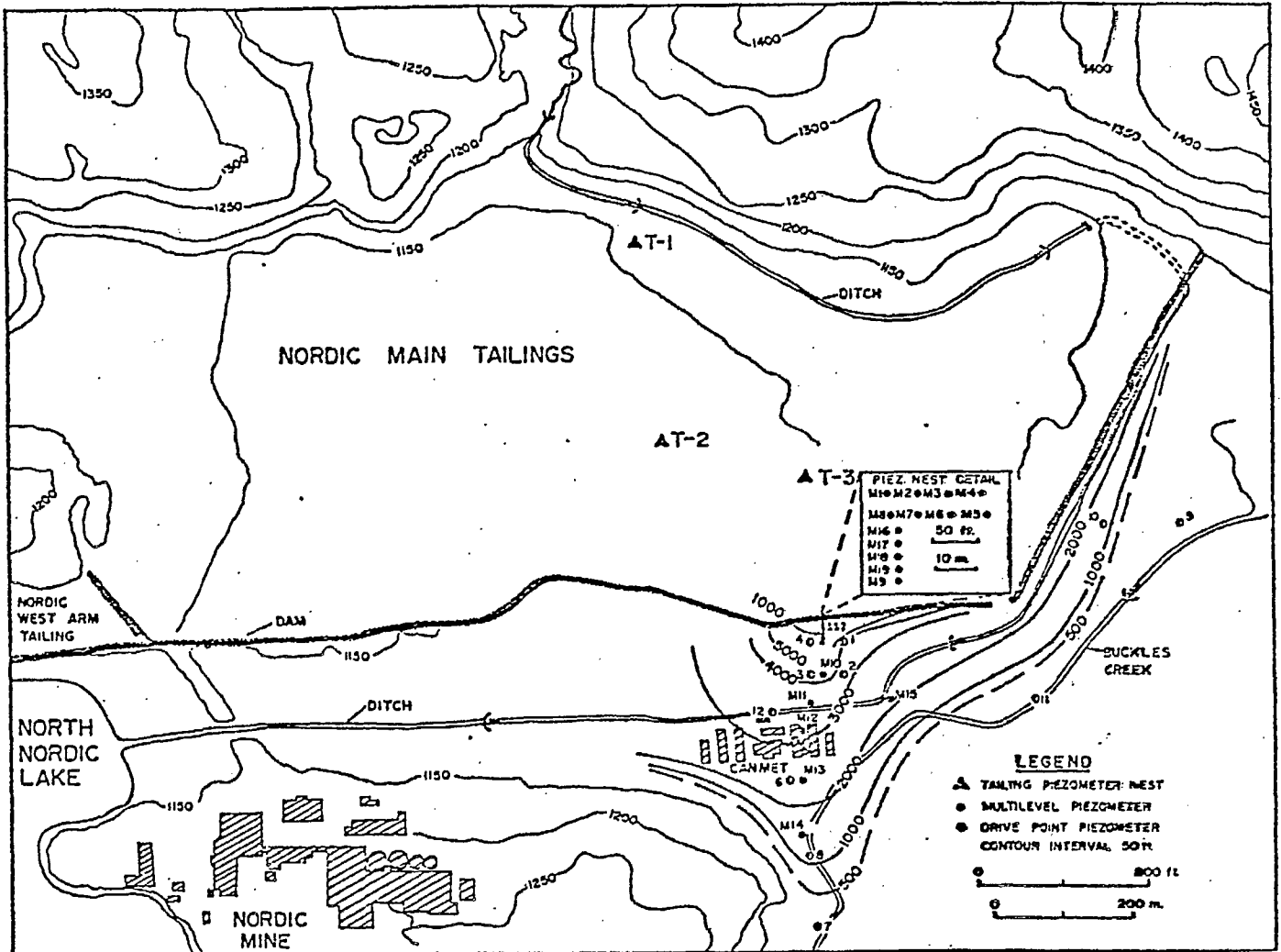


Fig. 4.2 - Nordic mine hydrogeochemistry, piezometer instrumentation and sulphate plume. (From Blair et al. (28)).

period 1975 to 1979 until the vegetation was self-sustaining.

4.3 SITE GEOLOGY

The Nordic mine tailings site is located in an east-west trending valley flanked by parallel trending bedrock ridges. The ridges are formed from lower Proterozoic arenaceous meta-sedimentary rocks that strike east-west and dip 10 to 15° north forming a dip and scarp topography. The surface elevations range from 340 to 345 m ASL.

The Nordic Valley is infilled with Pleistocene glacial deposits comprised of basal till overlain by glaciofluvial outwash sands and gravel with a combined thickness varying up to 30 m. The glaciofluvial deposits of the Nordic Mine Valley extend to Buckles Mine Valley in the south.

The glaciofluvial sands form a fairly uniform deposit of fine to coarse-grained micaceous quartz-feldspathic sand with occasional pebble-gravel interbeds. Drilling for piezometer installation indicated that the depth of sand varied from 10 m to greater than 15 m (Blair et al. (28)). The glaciofluvial deposits are overlain throughout much of the valley area by a layer of black peat 0.5 to 1.0 m thick which was formed under a recent spruce bog that existed in the area south of the tailings dam and in the vicinity of the old CANMET Laboratory, Figure 4.3.

The area receives approximately 1 m of precipitation annually. The peak stream flow periods are experienced during the spring snow melt in April and May, and during the fall rains in October and November. The stream flow varies seasonally from 7.5×10^{-3} to 2.7×10^{-1} m³/sec.

4.4 METHODS OF INVESTIGATION: WATER SAMPLING INSTRUMENTATION

In the Tailings Pile

Piezometer nests were installed in the tailings area approximately 250 m apart. The piezometers at each nest had tips at intervals of 1.5 m and

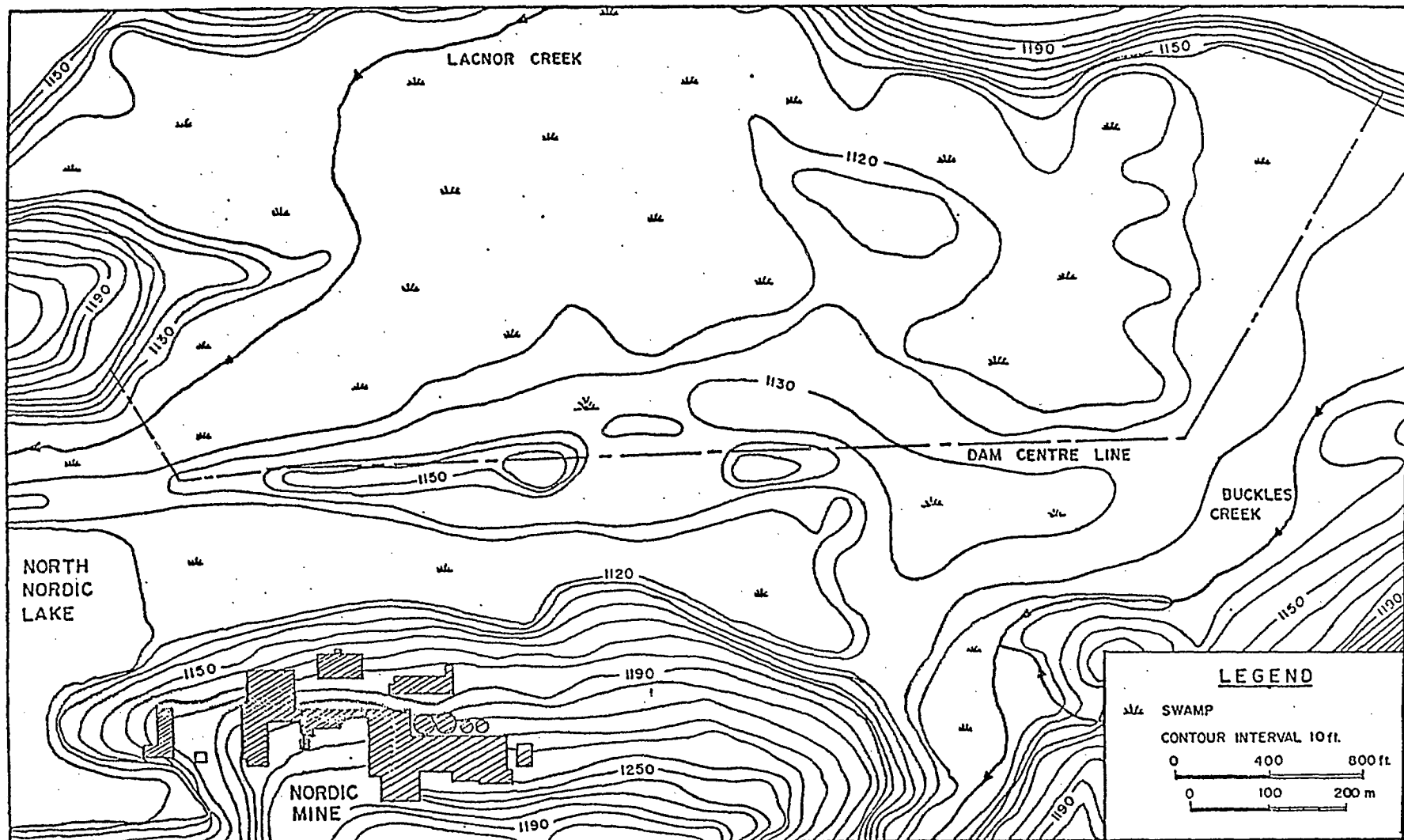


Fig. 4.3 - Nordic mine tailings basin original topography (1956).
 (From Blair et al. (28)).

were spaced vertically from the water table to about 1 m below the tailings. To obtain a representative porewater sample from a desired depth, the piezometer tip was placed in contact with the tailings causing as little disturbance as possible during the installation.

A modified Casagrande drive-point stand-pipe piezometer constructed entirely of 1.125 in (2.86 cm) schedule 80 PVC pipe (Figure 4.4) was used in an area of fine-grained tailings. A porous polyethylene cartridge (VYON) with a 50 μ pore size was used for the piezometer screen. The porous Vyon cartridge was sandwiched between the drive point tip and a straight female coupling and sealed with caulking compound. This was found to be very useful in preventing silt-sized tailings particles from entering the piezometer.

A vehicle-mounted hollow-stem auger drill was used to install the piezometer in and beneath the tailings where gravel was encountered. The hollow-stem auger was used to drill into the tailings at 1 m above the desired depth. A hand-operated gasoline powered rock drill was used to advance the piezometer point to its desired position. The tailings below the water table liquified around the piezometer pipe allowing the piezometer to advance easily. The tailings then settled back tightly around the piezometer. Bentonite was poured in to fill the void between the piezometer and the hollow-stem auger and the auger was then removed allowing the tailings to collapse around the assembly.

In the Sand Aquifer

Based on the reconnaissance investigation, a detailed groundwater monitoring network was designed along the toe of the dam to intercept chemicals and radioisotopes contaminating the groundwater migrating from the tailings. The monitoring was accomplished with multi-level bundle-type piezometers that provided individual water samples from many different depths over the thickness of the aquifer. The multi-level assemblies were

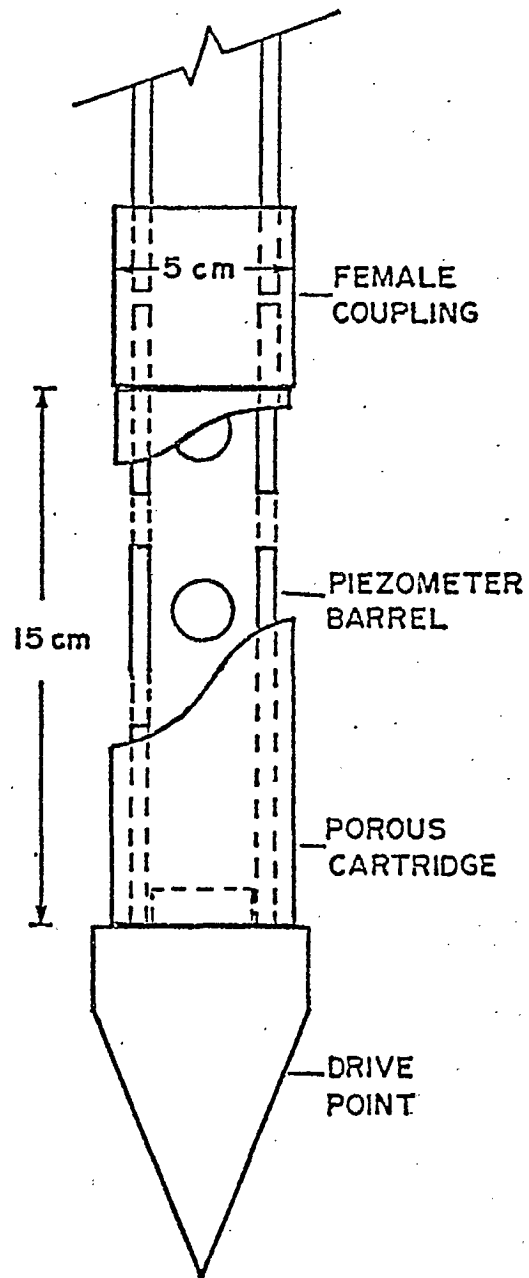


Fig. 4.4 - Design of piezometer drive point tips used in tailings (From Blair et al. (28)).

constructed of a bundle of 12 mm (0.5 in) O.D. polyethylene tubes, each tube with a 10 cm screened interval at the end, fastened at different intervals to a central 12 mm I.D. PVC pipe. The multi-level assemblies were installed using a hollow-stem auger to a maximum depth of 15 m. The cohesionless sand collapsed around the assembly immediately upon withdrawal of the auger (Morin et al. (18-21)).

4.5 TAILINGS SOLID SAMPLING METHOD

Solid cores were taken during the hydrogeological investigation. Samples were extracted by drilling or augering a hole down to the required depth and collecting the samples from the undisturbed formation with a split spoon sampler, or short Shelby tube. During the drilling and sampling at the West Arm tailings area, drilling mud (a suspension of bentonite in water) was used for lubricant and washing, and it is suspected that by using this method some sample contamination and ionic imbalance could occur because of the churning action of the drill (31,34,35).

Later, a new method was introduced and used for sampling tailings solids in the Nordic Main tailings area. A vehicle mounted hollow-stem auger was used, and at a shallow depth above the saturated zone, the sampling was done by using a short Shelby tube or split spoon sampler. The hollow-stem auger with centre plug was advanced to a desired depth and the split spoon sampler was lowered through the centre of the hollow stem-auger after the plug has been removed. After the sample was obtained the centre plug was placed back into the hollow stem auger and advanced further into the next desired position. From the water table downward a 30 ft (9.1 m) long tube of thin walled aluminum pipe 3 in (7.62 mm) O.D. was used. The long tube was slowly pushed down and hammered slightly at the same time until it penetrated

the black peat layer and hit the gravel formation beneath. The bottom of the thin aluminum pipe was crimped closed after a few hammer blows against the gravel formation to form a plug.

The pipe was lifted and placed against the drilling tower where it was cut in sections starting at the top in 6 in (15 cm) intervals.

During the drilling and sampling operation all important stratification data, such as starting depth, length of sample, compaction, sample interval, colour, and odour such as H₂S gas, etc. were recorded.

Piezometer nests were installed at the same location where the solid tailings were sampled in such a way that the water samples at certain depths could be directly correlated with tailings solids of the same location and depth.

Locations of piezometer nests T-1, T-2, T-3, T-7, and T-8 are shown in Figure 4.5 and a general cross-section of the Nordic Main Tailings in Figure 4.6.

4.6 FIELD PREPARATION

Water Sampling and Field Preparation

Before the ground water samples were collected, the piezometer well was first pumped nearly dry to clear the stagnant water in the well and allow fresh water from the formation to enter. The water sample was collected following the recovery of the piezometer so that the water retrieval was representative of the tailings formation water in the immediate vicinity of the piezometer screen.

The height of the hydraulic head was monitored before and after pumping to test the response of the piezometer well and to collect hydrology data.

Groundwater samples were collected from the piezometers in the

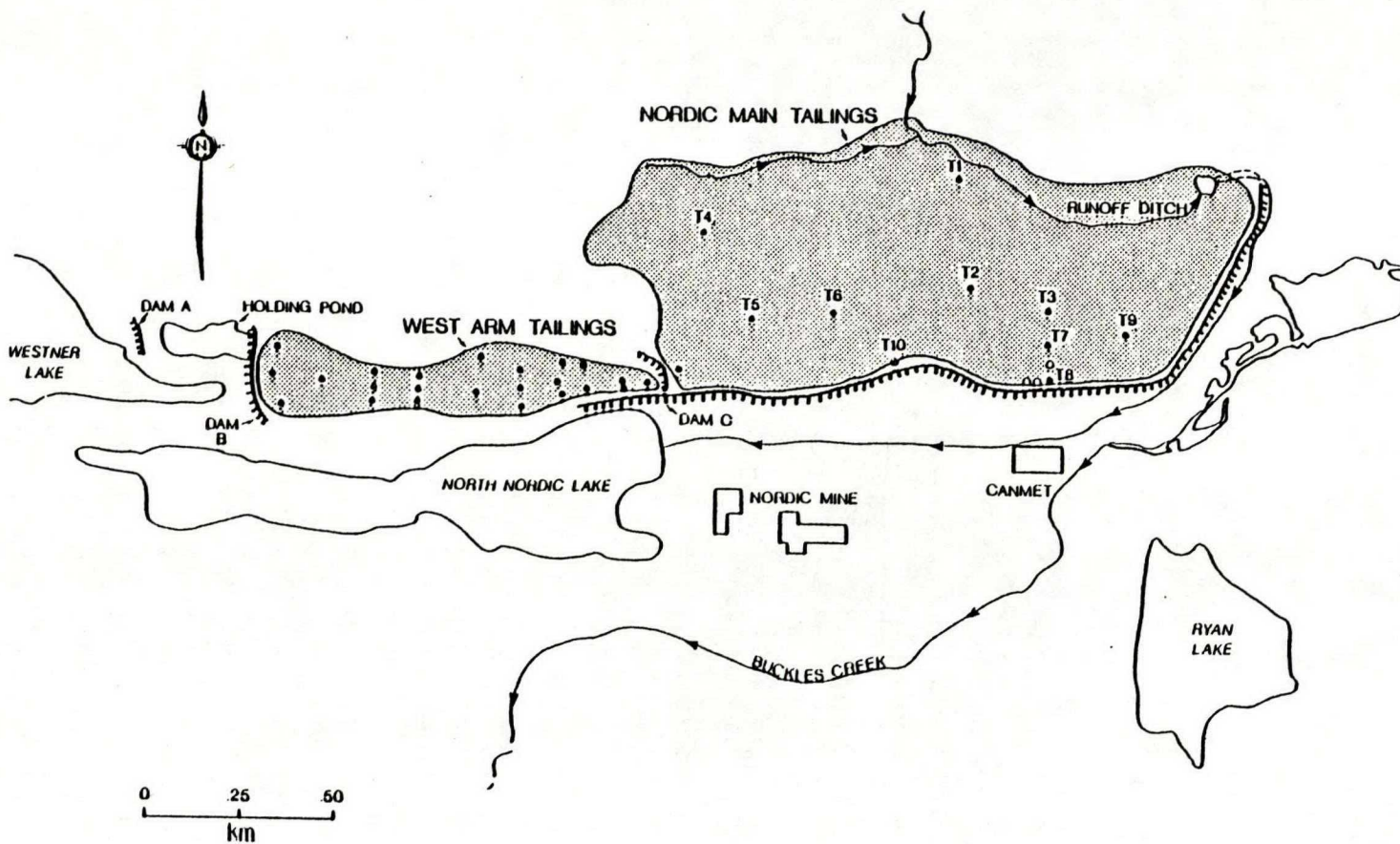


Fig. 4.5 - Location map and monitoring sites at the Nordic main and Nordic west arm tailings. (From Cherry et al. (29)).

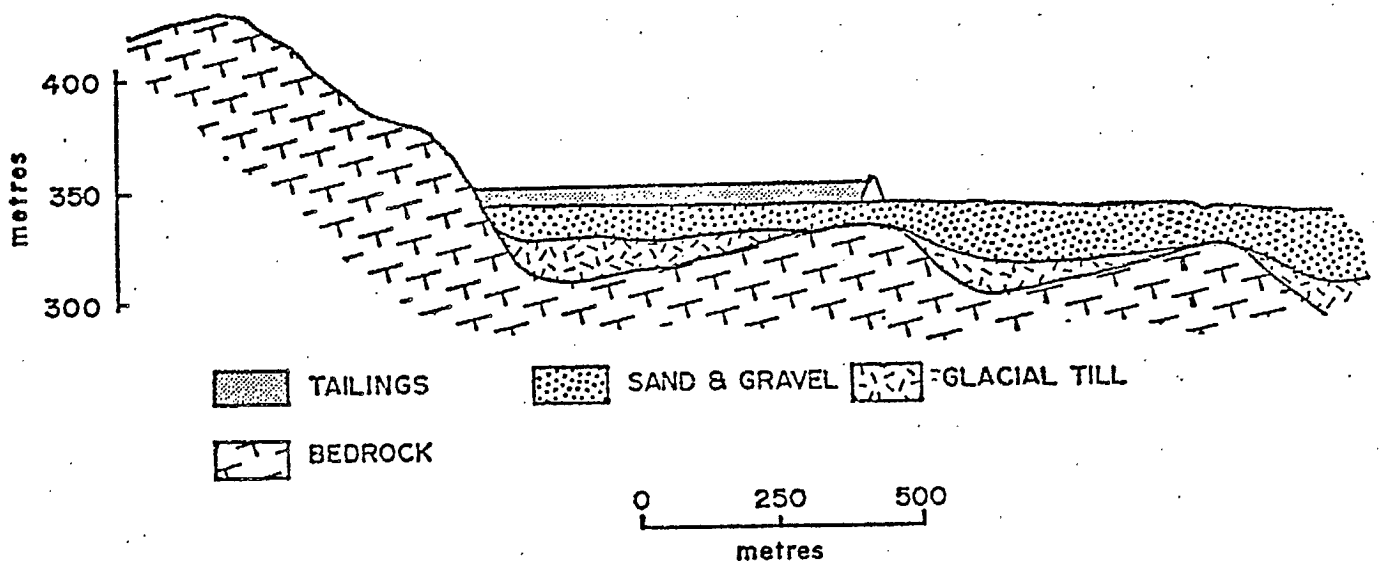


Fig. 4.6 - Geological cross-section of the Nordic main tailings pond and surficial deposits. (From Blair et al. (28)).

tailings and in the sand aquifer using a peristaltic pump and passed through an 0.45 μm , 127 mm diameter, filtration assembly at the end of the pressure side of the peristaltic pump.

The clear pre-filtered water was then analyzed as soon as possible for formation temperature, pH, electrical conductivity, redox potential, iron (II), alkalinity and acidity by titration methods. Other elements such as sulphate, heavy metals, and some radioisotopes such as ^{226}Ra , ^{232}Th , ^{228}Th , ^{230}Th and ^{210}Pb were analyzed later in the laboratory after the sample had been preserved with hydrochloric acid to pH = 1.

All the sample bottles were labelled with sample identification such as location, depth of sample, date and time of sampling, etc.

4.7 HYDROGEOLOGY OF THE TAILINGS

The depth of most of the Nordic Main tailings area, where the piezometer nests were located was approximately 10 m. A layer of compact black peat, approximately 1 m thick, forms an intermittent layer under most of the tailings pond area, separating the tailings from the underlying sand and gravel. A peat layer was not present at T-3 location which corresponded to the elevated areas in the original basin topography, Figure 4.3.

Water Table

The water table in the tailings has a fairly constant slope from the northside of the tailings area, where the water table is 1 m below surface, to the south side of the tailings adjacent to the dam where the water table is estimated to be 10 m below the surface of the tailings. The average water table fluctuation observed during the period August to December was 0.5 m. The water table elevation declined through the dry summer and early fall, then rose in response to heavy rainfall in November. The response at piezometer nest T-1 location was very rapid, where the water table was within

1.5 m. The rapid response suggested that the tension-saturated zone above the shallow water table was near the surface of the tailings. This rapid response was not observed at piezometer nests T-2, T-3, T-7 or T-8 where the water table was greater than 4 m.

Hydraulic Gradient

The vertical hydraulic gradient in the Nordic Main tailings is strongly downward with an average of 0.2 m/m, and is affected by the presence of the peat layer at locations T-1 and T-2 which causes a sharp increase in gradient. The average gradient across the tailings at T-1 and T-2 is fairly constant at 0.06 m/m, then increases sharply to 1.45 m/m across the peat layer in response to lower hydraulic conductivity in the peat layer compared to that for the overlying tailings. At T-3, where there is no peat layer, the gradient is constant at 0.2 m/m through the tailings into the gravel.

The horizontal hydraulic gradient in the tailings and underlying gravel aquifer is 0.0065 m/m towards the south, approximately 3 orders of magnitude less than the vertical hydraulic gradients. The horizontal gradient increases slightly towards the toe of the dam (Blair et al. (28)).

Hydraulic Conductivity

Each piezometer located below the water table in the tailings was bailed dry and the water levels recorded as recovery occurred. Hydraulic conductivity in the tailings was then calculated by the rising head method of Hvorslev (59), and was found to range from 1×10^{-4} cm/s to 3.1×10^{-6} cm/s. The individual hydraulic conductivity and the corresponding depth locations within the tailings are represented in Table 2 of Blair (28).

The range of hydraulic conductivity values obtained reflect stratification within the tailings of layers with different grain sizes. In general, the lower hydraulic conductivities are located near the bottom of the tailings and are overlain by tailings of higher hydraulic conductivity.

Groundwater Flux and Velocity

The Darcy equation (28) was used to obtain estimates of the groundwater flux in the tailings. The average groundwater velocity was obtained by dividing the Darcy flux by the porosity, which is approximately 0.30. The observed gradients and bulk average hydraulic conductivity of 1×10^{-5} cm/s were used in the calculations. The flux values obtained at the piezometer nests are listed in Table 1 of Blair et al. (28).

The average downward groundwater flux at piezometer nests T-1 and T-2 was approximately $0.14 \text{ m}^3/\text{m}^2/\text{year}$ with a downward velocity of 0.5 m/year. The corresponding value at T-3 was greater by a factor of 3.5. The downward flux was approximately $0.5 \text{ m}^3/\text{m}^2/\text{year}$ with a downward velocity of 1.6 m/year.

The higher ground water flux at T-3 corresponds to the absence of flow restraints caused by the low hydraulic conductivity of the peat layer.

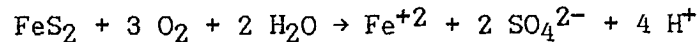
The groundwater seepage fluxes and velocities for the surficial aquifer are determined from the Darcy equation using measured hydraulic gradients and a representative hydraulic conductivity of 1×10^{-2} cm/s. The average lateral groundwater seepage flux in the surficial aquifer is in the order of $20 \text{ m}^3/\text{m}^2/\text{year}$ with an average velocity of 70 m/year.

These values increase towards the toe of the tailings dam where the hydraulic gradient is larger. Between piezometers M-1 to M-8, the specific discharge from beneath the toe of the tailings dam attains an average value of $167 \text{ m}^3/\text{m}^2/\text{year}$ with a seepage velocity of 550 m/year. The specific discharge and velocity diminish rapidly southward from the point of discharge as the seepage flowpaths diverge outward away from the bedrock ridge restriction, located in the dam west of the piezometers.

4.8 HYDROGEOCHEMICAL PROCESSES

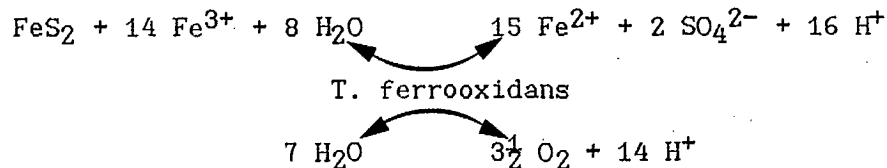
The hydrogeochemical processes that control the chemical composition of the porewater in the tailings and the chemical composition of contaminated groundwater in the sandy aquifer are the subject of on-going research. In this thesis only a brief description of the major hydrogeochemical processes will be discussed.

The most important influence on the chemistry of porewater in the tailings, and subsequently in the groundwater in the surficial aquifer near the tailings, has been the oxidation of pyrite (FeS_2) in the tailings. This process can be expressed as:



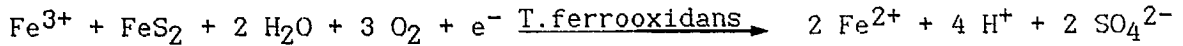
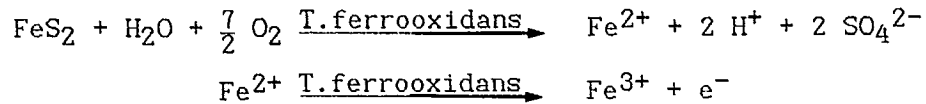
The production of hydrogen ions causes a progressive decline in the pH of the porewater from the initial pH of about 8, which existed when the tailings discharged from the mill, to a pH that is currently about 3 in the groundwater zone just below the water table. At a pH of 3 or lower, ferric ion rather than oxygen becomes the most dominant agent in pyrite oxidation (Nordstrom (37)).

The bacterium Thiobacillus ferrooxidans in the presence of oxygen and water oxidizes ferrous ions previously released from the pyrite to ferric ion, and then the ferric ion oxidizes in a manner that can be represented as:

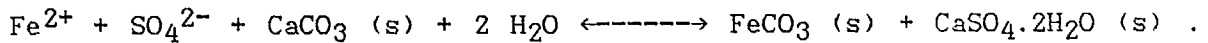
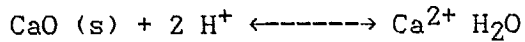
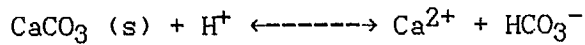


A similar explanation was introduced by Dave et al. (31) as follows:

In the presence of moisture and oxygen, pyrite is being oxidized by Thiobacilli ferrooxidans in the uppermost layer of the tailings producing H^+ ions (acidity) and high TDS porewater, represented by:



The acid produced further leaches out other trace metals and radionuclides. With the downward infiltration of this porewater during recharge events, neutralization and precipitation take place in the neutralized buffer zone in the presence of residual alkalinity in the form of calcite (CaCO_3), or lime (CaO), precipitating gypsum and siderite.



The solutes are transferred back to the solid phase in a lower zone which continuously progresses downwards with the consumption of the alkaline buffer and replacement of the original porewater by highly acidic, high TDS porewater.

An understanding of the hydrogeochemical aspects of the tailings and aquifer system is an essential prerequisite for the development of predictive models of the long-term evolution of water quality in the drainage basins in which tailings exist (Blair et al. (28)).

Oxidation of pyrite is the major cause of acidification of the uranium tailings porewater, where the original neutralized porewater is gradually replaced by acidic porewater that leaches and mobilizes the radionuclides and heavy metals in the tailings (Cherry et al. (29)).

The hydrogeochemical interactions within the tailings itself produce a dynamic mixture of oxidation, leaching, desorption, solute transportation, neutralization, precipitation, coprecipitation and sorption processes which gradually contribute to the evolution of a solid phase profile (Dave et al. (31)).

CHAPTER 5

CHEMICAL ANALYSES

Fractions of the water samples taken in the field were immediately analyzed for temperature, electrical conductance, pH, redox potential, acidity, alkalinity and ferrous ion content before the characteristics of the samples were changed by an oxidation reaction to the atmospheric conditions.

This analysis is a very important aspect of the field investigation, because it ensures the precision and accuracy of the data collected from every sampling point at various depths within the tailings pile.

After the basic field data were analyzed, the samples were stabilized and preserved by adding hydrochloric acid to pH = 1 for further laboratory analyses.

Ferrous ion was determined in the field by potassium dichromate titration as described in the manual "Standard Methods of Chemical Analysis", (36). Acidity and alkalinity were also determined using the titration method.

The pH, redox potential and ferric/ferrous ions ratio are indicators used to determine the extent of oxidation of the tailings with depth. The ferric content was obtained by subtracting the ferrous ions from the amount of total iron determined in the laboratory.

Electrical conductance is a quick indicator used in the field to determine the amount of dissolved electrolytes in the sample, which could also be used to gauge the amount of dissolved sulphate from the porewater of pyritic tailings. Therefore, this was also used as a useful indicator to determine dilution factors or sample cuts for sulphate analyses in the laboratory. The correlation test of electrical conductance versus concentration of sulphate is shown in Figure 5.1.

Correlation Study

Sulphate vs Electrical Conductance

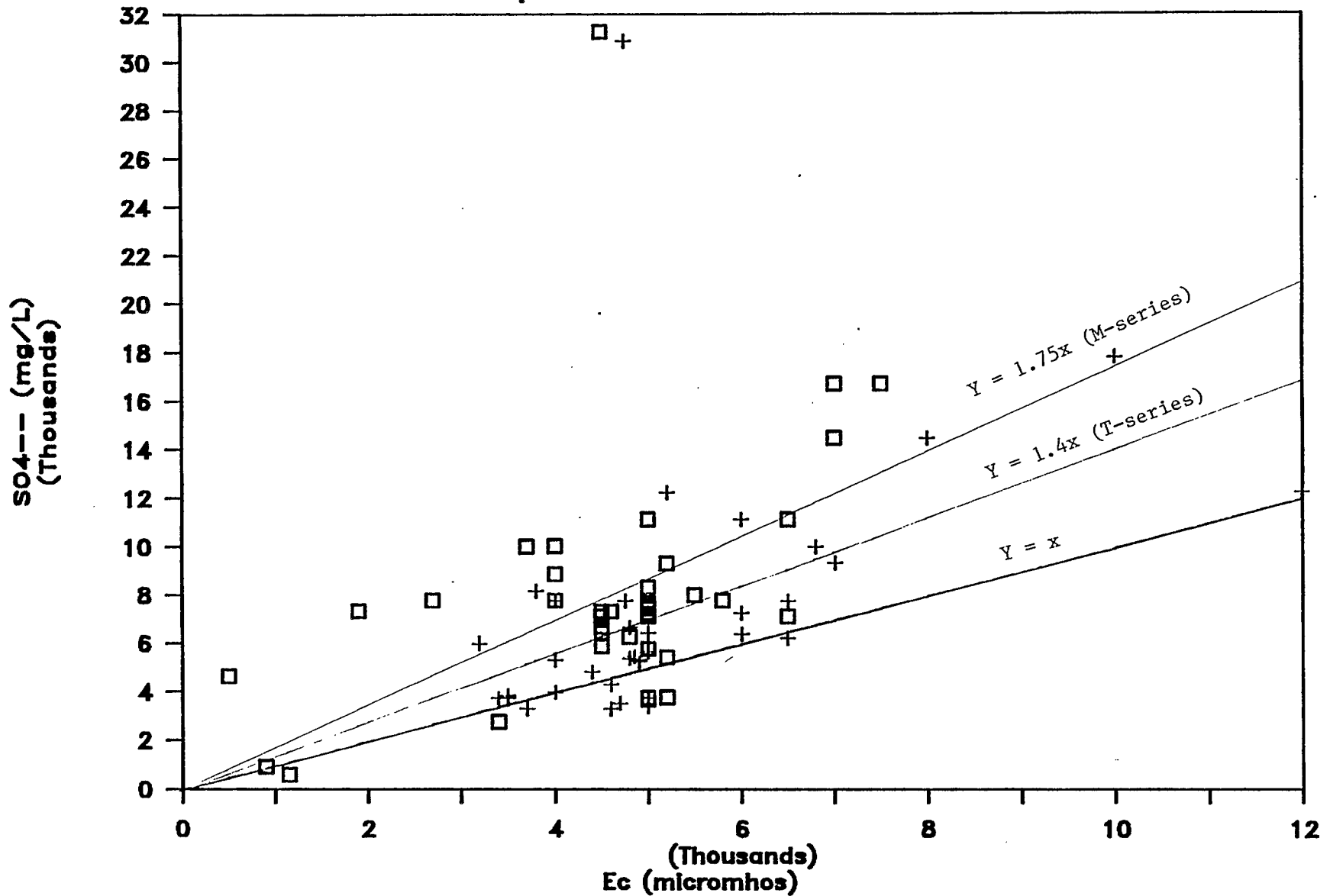


Fig. 5.1 - Correlation study of sulphate versus electrical conductance. Linear regression for M-series \square , and T-series $+$.

Heavy metals and earth alkali metals, and other inorganic elements from porewater and solid tailings, such as total Fe, B, U, Th, Mn, Ca, Mg, etc., were analyzed at the Canada Centre for Mineral and Energy Technology (CANMET), Elliot Lake, Ontario. Some of these samples were analyzed by contractors such as Guelph Chemical Laboratory Ltd., Guelph, Ontario, and Chemex Laboratories Ltd., Vancouver, B.C.

5.1 MEASUREMENT OF SAMPLE pH

The solid samples were prepared using freshly made distilled water in 1:1 ratio by weight, and the pH was measured using a glass electrode with a saturated calomel electrode as a reference electrode. The pH meter used for the measurement was a research grade instrument capable of measuring pH to ± 0.002 units, and was equipped with automatic temperature compensation. The glass electrode system had the capability of measuring the entire pH range from 0 to 14.

The reference electrode was dipped into a potassium chloride solution. The glass electrode consisted of a glass membrane filled with saturated potassium chloride. The potential of the electrode system was tested to ensure that it followed the Nernst equation (55):

$$E_g = E_0 + \frac{RT}{F} \ln C_{H^+}$$

where, R , T and F represent the gas constant, the absolute temperature, and the Faraday, respectively. E_0 is the constant of the electrode system, and C_{H^+} is the concentration of H^+ ions.

In the presence of extremely high alkaline solutions, especially when Na^+ is also present, negative deviations from the true pH may be caused by the migration of Na^+ to the glass membrane. A special glass electrode was used to eliminate error due to the salt effects and to give highly accurate pH values up to pH 13.

To ensure the precision and accuracy of the pH measurement, the instrument was calibrated first using a NBS buffer standard solution such as potassium hydrogen-phthalate (pH = 4.00 at 25°C), and a tetraborate buffer (pH = 9.00 at 25°C).

Precautions were taken to ensure that the electrodes were adequately washed on completion of each determination to avoid cross-contamination.

5.2 PRETREATMENT FOR SOLID TAILINGS ANALYSIS

Prior to the chemical analysis, the solid tailings were first weighed accurately, 1 g net weight (solid sample cut on dry basis) and placed in a platinum crucible. Drops of water and reagent grade HClO_4 (perchloric acid) were added, and the crucible was agitated slightly to ensure that the sample was thoroughly wetted. Then 10-20 mL of 48% HF (hydrofluoric acid) was added to the crucible and it was put on a hot plate with the temperature set at approximately 200°C to allow the solution to evaporate to dryness (2 to 3 hours). White fumes appeared before the solution evaporated to a complete dryness which indicated that an excess of perchloric acid was present. (Note: precautions should always be taken when working with perchloric acid. A fumehood should be especially designed for this purpose with a washout system in the output stack to prevent explosive powder from building up.)

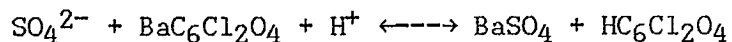
The crucible was then allowed to cool, and a mixture of 30% borax flux and 70% sodium carbonate were added. The mixture was then heated using a blast burner at a high temperature to bring it to the melting point. While heating it was continuously whirled in a gentle motion for approximately 15 minutes. The melt was allowed to cool off and a solution was then made up by dissolving the melt in 1 N HCl into a litre volume.

This solution was used in the analysis for most of the chemicals and radioisotope components in the solid tailings sample.

5.3 SULPHATE

After complete dissolution of solid samples, using the method described in 5.2, or the stabilizing and preserving of prefiltered porewater from the field, soluble sulphate ion concentration was determined by Guelph Chemical Laboratories Ltd. using a spectrophotometric technique.

This sulphate determination involved the reaction between barium chloranilate with sulphate in acid solution to give barium sulphate and acid-chloranilate ion:



The amount of acid-chloranilate liberated is proportional to the sulphate ion concentration. The reaction was carried out in 50% aqueous ethanol, buffered at pH 4.

To eliminate interferences from other cations, the sample containing sulphate ions was first filtered through an ion-exchanger column in hydrogen form. The pH of the effluent was adjusted to 4 with dilute hydrochloric acid or ammonia solution. The eluted sulphate was then made up to a known volume in a volumetric flask for further stoichiometric calculations.

At pH 4, the acid-chloranilate ion gives a broad peak of 530 nm. This wavelength was used for sulphate determination in the range between 2 to 400 ppm. A much more intense absorption occurs in the ultraviolet: a sharp band at 332 nm will enable the limit of detection of sulphate ions to be extended to 0.06 ppm with accuracy and precision to $5 \mu\text{g/g} \pm 3\%$.

Calibration

An aliquot containing freshly made standard sulphate ions was prepared. The pH 4 buffer (a 0.04 M solution of analytical reagent grade potassium hydrogenphthalate) and ethanol were added. A weighed quantity of barium chloranilate was added and the flask was shaken for 10 minutes. The excess barium chloranilate and the precipitated barium sulphate were filtered. The absorbance of the filtrate was then measured at 530 nm against

a blank which was prepared in the same manner. A calibration curve using standard potassium sulphate solution was thus prepared from the A.R. grade salt.

For a sample solution containing less than 2 ppm sulphate, the measurement was carried out at a shorter wavelength as described above with an appropriate calibration curve.

Later in this thesis, another new method of sulphate analysis will be discussed. This method involves the radioisotope Ba-133 as a tracer to determine the amount of sulphate present in the solution.

5.4 SILICA

Determination of Silica as SiO₂

Insoluble silicate was fused with sodium carbonate, and the melt which contains the silicate in acid-decomposable form was then treated with hydrochloric acid. The acid solution of decomposed silicate was evaporated to dryness in the water bath to separate the gelatinous silicic acid as insoluble silica SiO₂.yH₂O; the residue was heated at 110-120°C to partially dehydrate the silica and render it as insoluble as possible. The residue was then extracted with hot dilute hydrochloric acid to remove salts of iron, aluminum and other metals which might be present as interferences. The greater portion of the silica remained undissolved, and was therefore filtered off next. The filtrate was evaporated to dryness, and the residue was heated to dryness at 110-120°C as before which rendered insoluble the small amount of silicic acid that escaped dehydration. The residue was then treated again with dilute hydrochloric acid as before and the second portion of silica was filtered off on a fresh filter.

The two washed precipitates were combined and ignited in a platinum crucible at about 1050°C to produce silicon oxide and then weighed. The

ignited residue is not usually a pure silicon oxide, generally containing a small amount of impurities as determined by treating the weighed residue in the platinum crucible with an excess of hydrofluoric acid and a little concentrated sulphuric acid.

The silica will be expelled as the volatile silicon tetrafluoride; the impurities (e.g., Al_2O_3 and Fe_2O_3) will first be converted into fluorides which pass into the sulphates in contact with the less volatile sulphuric acid following the subsequent brief ignition (at 1050-1100°C for a few minutes).

The loss of weight therefore represents the amount of pure silicon dioxide present.

The determination of silica in a 'soluble' silicate was carried out by the same procedure (Guelph Chemical Laboratories Ltd.). The accuracy and precision of this method was checked against the NBS standard and found to be excellent with a coefficient variance of less than 2%.

Dalton (38-40), using the X-ray diffraction method, reported the silica content as quartz.

5.5 METALS

Atomic Absorption

Fe, Ca, Al, Pb, Mg and K were analyzed using an atomic absorption spectrophotometer, dual beam Varian CR-90, equipped with automatic background correction and digital readout. The equipment is suitable for both flame and flameless (graphite furnace) spectrophotometry. The detection limit for flame spectrophotometry for various elements required to be analyzed are given in Table 5.1.

Sequential Atomic Emission Spectrophotometer

Routine analysis of Fe, Ca, Mg, Al, Pb, Mn, U, Th, Si, K, Ni, Co, Cu,

TABLE 5.1

Lower limit of detection of metals to be analyzed

Element	Conc. in Solution ppm (mg/L)	Conc. in Solids 1 gm in 10 mL soln ($\mu\text{g/g}$)
Al	0.5	5
Ca	0.01	0.1
Fe	0.02	0.2
Pb	0.1	1.0
Mg	0.003	0.03
K	0.003	0.03

Note: In calcium and magnesium determination, trivalent lanthanum chloride was added as a releasing agent to suppress the Na and K enhanced reading.

In potassium determination, CsCl was added to the sample to attain a high degree of accuracy in A.A. measurement.

etc., was carried out with a high energy plasma excitation source (DCP), at the Elliot Lake Laboratory.

The direct current plasma (DCP) emission spectrophotometer employs a three electrode configuration as a plasma excitation source and an echelle grating monochromator. The grating is used in combination with a quartz prism for order separation and provides a two directional spectral pattern covering 190 nm to 800 nm. With the diffraction of collimated incident radiation the peak efficiency of all wavelengths occurs in a succession of different orders, which are vertically stacked, and within these orders the wavelengths are dispersed horizontally. The echelle system has a focal length of only 0.75 m and provides an average reciprocal linear dispersion of 0.12 nm/mm, and an average resolution of 0.003 nm. The detection system on the sequential DCP spectrophotometer uses a single photomultiplier tube and a wavelength is selected by moving the grating and prism within a calibrated scale. The two dimensional spectrum can also be photographed using a Polaroid camera attachment to permit spectrographic qualitative analysis.

During routine operation of the DCP spectrophotometer, it was observed that the overall stability of the instrument was quite short-lived, sometimes as short as 10 to 15 minutes.

The lowest limit of detection, and the ranges of some metals analyzed routinely at the Elliot Lake Laboratory are shown in Table 5.2.

5.6 THORIUM AND URANIUM ANALYSES

Chemical thorium and uranium analyses were also done by Neutron Activation Analysis (NAA). This is a very sensitive and accurate method which analyzes thorium and uranium with LLD in the range of microgram and picogram, depending on the form in which the sample is available. The thorium was done by NAA-Gamma Spectroscopy, and the uranium by Delayed

TABLE 5.2

Lower limit of detection of metals analyzed by
DCP Sequential Spectrophotometer

Element	Wavelength (nm)	LLD (mg/L)	LDR (mg/L)	BEC (mg/L)	Precision (RSD) at...xBEC...%
Al	236.705	1	10-1000	40	2 xBEC 0.6%
	308.215	0.08	0.8-100	2	2 xBEC 0.9%
	396.152	0.02	0.02-100	0.04	25 xBEC 1.3%
Ca	315.887	0.03	0.3-100	1	10 xBEC 0.8%
	445.478	0.09	0.9-100	2	5 xBEC 0.7%
Co	236.379	0.02	0.2-1000	0.8	1 xBEC 0.7%
Cu	224.7	0.03	0.3-1000	2	10 xBEC 1.5%
Fe	259.94	0.007	0.07-100	0.2	5 xBEC 0.6%
	371.994	0.005	0.05-1000	0.1	10 xBEC 1.0%
Pb	220.353	0.2	2-1000	10	1 xBEC 1.3%
	368.348	0.01	0.1-600	0.4	20 xBEC 0.8%
Mg	279.553	0.0002	0.002-60	0.006	2 xBEC 0.8%
Mn	259.373	0.002	0.02-100	0.06	5 xBEC 0.6%
Ni	243.789	0.09	0.9-1000	3	10 xBEC 0.8%
K	404.414	0.9	9-100	20	1 xBEC 9.0%
Th	401.913	0.02	1-10	2	?
U	424.167	0.04	0.4-1000	1	10 xBEC 0.4%

Note: LLD = Lowest limit of detection.
LDR = Linear dynamic range.
BEC = Background equivalent concentration.
RSD = Relative standard deviation.

Neutron Counting.

One of the most important advantages of the NAA method is its unique freedom from the effects of accidental cross-contamination of the sample with the element being determined. The maximum pretreatment necessary for most samples is grinding and homogenization before bombardment with neutrons.

The disadvantage of this technique is that only a few companies have the necessary reactor facilities. In this case, most of the work was contracted out to Atomic Energy of Canada Ltd.

5.7 DISCUSSION

Most of the chemical analyses carried out, as described in Chapter 5, were an essential prerequisite to the understanding of the hydrogeochemical processes which may have a connection with the mechanism of radioisotope disequilibrium in the system.

CHAPTER 6

RADIOCHEMICAL ANALYSES

A review of recent literature pertaining to the radiochemical measurement of certain, naturally occurring, radioactive elements was prepared in the form of a laboratory manual by Smithson et al. (42), and Chiu et al. (43). It gave detailed analytical methodology for the measurement of gross alpha, gross beta, gross gamma, uranium, ^{226}Ra , ^{228}Ra , ^{210}Pb , ^{232}Th , ^{230}Th , ^{228}Th , total thorium, ^{222}Rn , ^{220}Rn and ^{219}Rn .

A summary of the ^{238}U , ^{235}U and ^{232}Th decay series is shown in Figure 6.1. However, in this thesis only a few radiochemical analyses will be discussed in relation to the subject of disequilibrium of radioisotopes from the vicinity of uranium tailings.

6.1 RADIUM-226 (^{226}Ra)

^{226}Ra is the most frequently determined radionuclide because of its potential health hazard as a bone seeking element and its long half-life of 1602 years. Therefore, it is used as an indicator of the radioactive contamination level in the environment surrounding the uranium mining and milling operation.

^{226}Ra can be determined by five different methods, namely, the gross alpha measurement, the radon emanation method, the alpha-spectroscopy method, by gamma ray spectroscopy, or by coincidence spectroscopy.

6.1.1 Gross Alpha Counting

This is the simplest and most economical method. It involves the separation of radium as sulphate from the sample solution with a lead sulphate carrier. Radium sulphate is then isolated by dissolving the radium-

	URANIUM 238 SERIES		URANIUM 235 SERIES			THORIUM 232 SERIES		
URANIUM 92	^{238}U 4.5-10 ⁹ y	^{234}U 2.5-10 ⁵ y		^{235}U 7.1-10 ⁸ y				
PROTACTINIUM 91		^{234}Po 1.2 m		^{231}Pa 3.2-10 ⁴ y				
THORIUM 90	^{234}Th 24 d	^{230}Th 8.0-10 ⁴ y		^{231}Th 26 h	^{227}Th 18 d	^{232}Th 1.4-10 ¹⁰ y	^{228}Th 1.9 y	
ACTINIUM 89				^{227}Ac 22 y		^{228}Ac 6.1 h		
RADIUM 88		^{226}Ra 1.6-10 ³ y		^{223}Ra 11 d		^{228}Ra 6.7 y	^{224}Ra 3.6 d	
FRANCIUM 87								
RADON 86		^{222}Rn 3.8 d		^{219}Rn 4.0 s			^{220}Rn 55 s	
ASTATINE 85								
POLONIUM 84		^{218}Po 3.1 m	^{214}Po 1.6-10 ⁻⁴ s	^{210}Po 140 d	^{215}Po 1.8-10 ⁻³ s		^{216}Po 0.14 s	^{212}Po 3-10 ⁻⁷ s
BISMUTH 83		^{214}Bi 20 m	^{214}Bi 5 d	^{210}Bi 5 d	^{211}Bi 2.2 m		^{212}Bi 61 m	^{212}Po 65%
LEAD 82		^{214}Pb 27 m	^{210}Pb 20 y	^{206}Pb (STABLE)	^{211}Pb 36 m	^{207}Pb (STABLE)	^{212}Pb 11 h	^{208}Pb (STABLE)
THALLIUM 81					^{207}Tl 4.8 m		^{208}Tl 3.1 m	

Fig. 6.1 - Some important characteristics of the ^{238}U , ^{235}U and ^{232}Th series decay chains (taken from Chiu et al. 1986 (43)).

lead sulphate in alkaline EDTA or DTPA and coprecipitating with a barium sulphate carrier at pH 4.8. The radium-barium sulphate carrier is then counted on a gross alpha counter.

To obtain accurate ^{226}Ra activities, correction should be made for the presence of ^{223}Ra and ^{224}Ra by differential analysis. However, this is not an easy correction to do since gross alpha counting cannot differentiate the partitioning between ^{223}Ra and ^{224}Ra activities.

The simplest way to eliminate this uncertainty is to wait and count 66 days after the time of separation, that is after the ^{223}Ra and ^{224}Ra have decayed (Chiu et al. (43)).

6.1.2 Radon Emanation Method

The radon emanation method is an indirect method based on the relationship between the activity of the ^{222}Rn daughter measured against the ^{226}Ra parent present.

Radium, as pure as possible, is separated from the sample solution (as described in the gross alpha counting 6.1.1). and the radium-barium sulphate is redissolved in alkaline EDTA solution.

The redissolved solution is then placed in a bubbler from which the gaseous radon is stripped from the solution to zero concentration of radon in the bubbler.

The bubbler is then sealed off and isolated to allow radon gas to grow in the solution. After a suitable ingrowth period has elapsed, the radon gas is again stripped from the solution and is transferred to a ZnS scintillation (Lucas) cell.

With this method, there may be a considerable delay between sealing the bubbler and stripping the gaseous radon from the solution. For example, if a 50% equilibrium between ^{226}Ra and ^{222}Rn is desired, a waiting period of one half-life is required, i.e., about four days (Chiu et al. (43)).

Later ^{226}Ra can be indirectly determined by counting the activity on a scintillation counter after allowing time (3.5 hours) for ^{222}Rn to come to equilibrium with its daughters, namely, ^{218}Po and ^{214}Po .

In the presence of ^{223}Ra and ^{224}Ra , some of their daughters, namely ^{219}Rn and ^{220}Rn , respectively, interfere with ^{226}Ra determination. Correction can be made by using a long transfer tube from the bubbler to the Lucas cell to prevent ^{219}Rn and ^{220}Rn arriving in the Lucas cell, as they will decay along the tube before their arrival in the cell. However, if the concentrations of ^{223}Ra and ^{224}Ra are very high, this method will become ineffective, as a long tube can cause many changes in the total volume and more errors could be introduced into the system. Furthermore, it is difficult to determine the length of the tubing required, as the system is not designed to differentiate between the isotopes.

6.1.3 High-Resolution Gamma Spectroscopy

High-resolution gamma-spectroscopy requires the use of Ge(Li) or high-purity N-type Ge detectors. This technique is based on the simultaneous determination of radionuclides by measurement of emitted gamma photons from 40-2000 KeV. In certain instances it can be complicated and time consuming because of the presence of large numbers of gamma photopeaks, but simplification is possible if the radionuclide(s) of interest is (are) physically separated from the others (Chiu et al. (43)). For example, Michel et al. (44) successfully determined the activities of ^{226}Ra and ^{228}Ra by separating radium with barium carrier from the sample.

For determination of ^{226}Ra , photopeaks of its daughters ^{214}Pb (295 KeV, 352 KeV), and ^{214}Bi (609 KeV, 1120 KeV) were used. In the absence of ^{235}U , the 185 KeV line of ^{226}Ra can be used, especially after extraction of the radium as previously mentioned, or if dealing with samples from uranium tailings, where the uranium has been extracted during the milling process.

Similarly, for ^{228}Ra , the photopeaks of its daughters ^{228}Ac at 338 KeV, 911 KeV, and 965 KeV were used.

Other investigators have reported the use of gamma-spectroscopy to determine uranium (45), thorium (45), and ^{210}Pb (46).

However, high-resolution gamma-detectors usually have a very low detector efficiency. Therefore, to achieve good counting statistics, a large sample and a long counting time are needed.

6.1.4 Coincidence Spectroscopy

Coincidence spectroscopy has recently been reported for the determination of ^{224}Ra , ^{226}Ra and ^{228}Ra . The coincidence spectrometer consists of a NaI(Tl) gamma-detector, and an alpha-beta-detector which are connected to a coincidence analyzer and associated electronics (Chiu et al. (43)).

This technique is based on separate measurement of the coincident alpha-gamma transitions of ^{226}Ra and ^{224}Ra , and the coincident beta-gamma transitions of ^{228}Ac , the daughter of ^{228}Ra . Chemical separation and purification of radium into a source is necessary. ^{224}Ra and ^{226}Ra are measured directly, and ^{228}Ra is measured indirectly via its daughter ^{228}Ac (6.13 hours).

6.1.5 Alpha-Spectroscopy

Alpha-spectroscopy involves the direct measurement of the alpha-emission from ^{226}Ra . Source preparation usually involves coprecipitation of radium sulphate with barium sulphate. The spectral resolutions obtained depend on the quantity of barium carrier, the size of the barium sulphate crystals, and the presence of other insoluble impurities. However, when the quantity of the barium carrier is reduced to improve the resolution, the chemical yield decreases.

Lim and Dave, (47), reported alpha-resolutions of 229 KeV to 496 KeV (FWHM) for 100 µg to 1 mg of barium carrier, coupled with a chemical yield of 65% to 85%. The main advantage of this approach is that ^{133}Ba , a gamma-emitter and a proven tracer for radium, is used for the chemical yield calculation.

Dean and Chiu, (48), reported alpha-resolutions of 120, 156 and 221 KeV (FWHM) for 68, 138 and 276 µg of barium carrier, respectively.

Sill (49) indicated recently that with complete sample dissolution, a modification of barium precipitation procedure, and removal of insoluble impurities from all reagent solutions, a high alpha-resolution (60 KeV) and a high chemical yield can be obtained. Electron microscopy has shown that barium sulphate crystals obtained by normal precipitation from hot DTPA solution are angular and relatively large, at least 1 µm on a side. They tend to form a cluster on the membrane filter so that the fraction of the total available filter area actually covered is small. Consequently, the effective thickness of the deposit is much larger than for a uniform distribution of smaller crystals. On the other hand, barium sulphate crystals obtained by precipitation from DTPA solution at room temperature, using a barium sulphate seeding suspension, are almost amorphous (<1 µm), and cover the entire surface area of the filter.

The high resolution and low background of the solid state surface barrier detectors provide specificity and high sensitivity.

Based on these electronic properties and the simplicity of the chemical separation method for ^{226}Ra analysis, a field laboratory was set up in Elliot Lake, specializing in ^{226}Ra analysis. This field laboratory was designed especially to handle field investigation and research related to environmental studies surrounding uranium mining and milling activities across Canada.

For a laboratory manual of ^{226}Ra analysis used by the Elliot Lake Laboratory, see reference 47. Data pertaining to ^{226}Ra from Nordic tailings area were obtained using this manual. Some analyses were contracted out to Monenco Ltd. who used the technique described by Chiu et al. (43).

6.2 RADIUM-228 (^{228}Ra)

Because of the difficulty in measuring the very low energy beta emission of ^{228}Ra (0.055 MeV), all techniques reported to date are based on the indirect measurement of its daughters, ^{228}Ac (6.1 h half-life), ^{228}Th (1.9 y half-life), or ^{212}Pb (10.6 h half-life). The application of gamma spectroscopy and beta-gamma coincidence spectroscopy has been discussed earlier.

The most widely used technique is beta counting of ^{228}Ac , which requires less than two days for equilibration between ^{228}Ra and ^{228}Ac .

A few procedures use alpha-spectroscopy to measure the long-lived daughter ^{228}Th or beta-counting of the short-lived daughter ^{212}Pb . Both of these procedures require 6-12 months for the ingrowth of daughters which is unacceptable for routine measurement.

^{228}Ra must be separated from other radionuclides before its daughter ^{228}Ac is allowed to grow in. A faster and more efficient method is used by Chiu and Dean (50). Radium is separated and purified in the form of barium-radium sulphate. After the ingrowth of ^{228}Ac , the sulphate is redissolved in alkaline EDTA or DTPA and the ^{228}Ac is precipitated as hydroxide with cerous hydroxide as a carrier and collected on a membrane filter for immediate beta-counting. The entire procedure takes less than 30 min, which permits the preparation of one sample to be completed just as the beta-counting of another sample is finished, thereby reducing the risk of decreased

sensitivity while an ^{228}Ac source is waiting to be counted (Chiu et al. (43)).

6.3 LEAD-210 (^{210}Pb)

The radioactivity of ^{210}Pb is normally measured indirectly by alpha-counting of its daughter ^{210}Po (5.3 MeV, 138 d half-life), or by beta-counting of its daughter ^{210}Bi (1.17 MeV, 5 d half-life) because of the low energy beta emission of ^{210}Pb (0.018 MeV).

The appropriate methodology depends on the state of equilibrium between ^{210}Pb , ^{210}Po and ^{210}Bi (Chiu et al. (43)).

6.3.1 Lead-210 (^{210}Pb) in Equilibrium With Polonium-210 (^{210}Po)

If radioactive equilibrium is known to exist, the measurement of ^{210}Po by alpha-spectroscopy is preferred. ^{210}Po in aqueous solution can be extracted by spontaneous deposition as the free element on a disc of a less noble metal, such as copper, nickel or silver. Copper or nickel are better than silver for the extraction of ^{210}Po , although nickel also extracts ^{210}Bi and ^{210}Pb (up to 20%). However, since ^{210}Po is an alpha-emitter, and ^{210}Bi and ^{210}Pb are beta-emitters, this method is specific for the indirect determination of ^{210}Pb . It is very sensitive because of the high-resolution alpha source obtained. The chemical procedure is relatively short and the plating can be done in two hours at 60-65°C, and ^{208}Po which is also an alpha-emitter is recommended as a tracer.

6.3.2. Lead-210 (^{210}Pb) in Disequilibrium

In most of the uranium tailings studies, the degree of radioactive equilibrium is unknown, however, in this case three techniques are available.

The first method is to measure ^{210}Po in a sample three to six months after the removal of initial ^{210}Po by spontaneous deposition on a nickel

disc. The second method is to measure the ^{210}Bi ingrowth five or more days after separating ^{210}Pb from the sample, and the third method is to allow the sample to age for 30 days after separation of ^{210}Pb from the sample, during which time complete equilibrium between ^{210}Pb and ^{210}Bi will be ensured to be greater than 98%. The ^{210}Bi is separated from ^{210}Pb and determined by beta-counting.

6.3.3 Lead-210 (^{210}Pb) Separation

^{210}Pb can be separated by several techniques such as electroplating, solvent extraction, ion exchange chromatography and electrochromatography. All these techniques are labour-intensive and very time-consuming.

The simplest method involves a single extraction of ^{210}Bi with an 0.1% solution of DDTC in chloroform from a 2 N hydrochloric acid solution. ^{210}Bi is subsequently precipitated as bismuth oxychloride and collected on a membrane filter for beta-counting, and ^{207}Bi gamma-emitting tracer is used for chemical recovery measurement (Chiu et al. (43)).

A recent report by Schery (51) indicates that the direct measurement of a 47 KeV photon emitted by ^{210}Pb has not been successful in the past because of the self-absorption in the sample and the low sensitivity to low-energy photons.

However, a new generation of high purity N-type germanium detectors now available are characterized by improved sensitivity to low energy photons and increased active surface area. Therefore, direct assay of ^{210}Pb in moderately sized environmental samples may be possible by measuring the 47 KeV photon.

6.4 THORIUM ISOTOPES

The long-lived thorium isotopes from the uranium and thorium series, such as ^{230}Th (8×10^4 y half-life), ^{232}Th (1.4×10^{10} y half-life) and its

daughter ^{228}Th (1.91 y half-life, emit alpha-particles with energies of 4.6 MeV, 4.0 MeV and 5.3 MeV, respectively. They can be determined simultaneously after separation from all other radionuclides and deposited as a uniform and thin source for the best alpha spectrometric resolution.

The purification of thorium by solvent extraction has been found to be effective using bis(2-ethyl-hexyl)phosphoric acid, Aliquot-336, thenoyltrifluoroacetone, trilaurylamine, etc., and is also used in combination with ion exchange. Electrodeposition on a polished stainless steel disc has been found to be the best method of preparing a uniform thin source for the best resolution of alpha-spectroscopy. However, this method is time-consuming and requires elaborate electrodeposition apparatus.

Sill and Williams (52), have developed an alternative method. The purified thorium isotopes are precipitated as hydroxides with 50 μg of cerium carrier from a strongly alkaline solution of EDTA or DPTA and is collected on an 0.1 μm membrane filter for high resolution alpha-spectroscopy. For chemical recovery, ^{234}Th tracer is commonly used.

Another method uses a combination of high resolution gamma-spectroscopy and alpha-spectroscopy. ^{228}Th is measured on the original sample by gamma-spectroscopy, and the isotopic ratios of ^{228}Th , ^{230}Th and ^{232}Th are determined by alpha-spectroscopy on the thorium source after extraction. By means of the ^{228}Th activity and the isotopic ratios so obtained, the activity of ^{230}Th and ^{232}Th are calculated (Durham et al. (53)).

Dean and Chiu (48) measured ^{228}Th indirectly via its ^{224}Ra daughter using alpha-spectroscopy. After ensuring equilibrium between ^{228}Th and ^{224}Ra (20 d half-life), ^{224}Ra is co-precipitated with barium sulphate for alpha-spectroscopy.

6.5 RADON

Radon isotopes occur in all three naturally occurring radioactive decay series. ^{222}Rn (3.82 d half-life) from the ^{238}U series, ^{219}Rn (4.0 s half-life) from the ^{235}U series, and ^{220}Rn (55.3 s half-life) from the ^{232}Th series.

Radon is an inert gas and the distance each isotope can travel from its origin depends on its half-life, and therefore ^{222}Rn , which has the longest half-life (3.82 d) can be transported further.

The most common and simplest measurement technique is the counting of alpha-disintegrations in a Lucas cell as previously mentioned in ^{226}Ra determinations (Chapter 6.1.2).

There are other methods of radon determination, such as a pulse-type ionization counting chamber, track-etch detection, and thermoluminescence. The most appropriate technique largely depends on the application of the measured data.

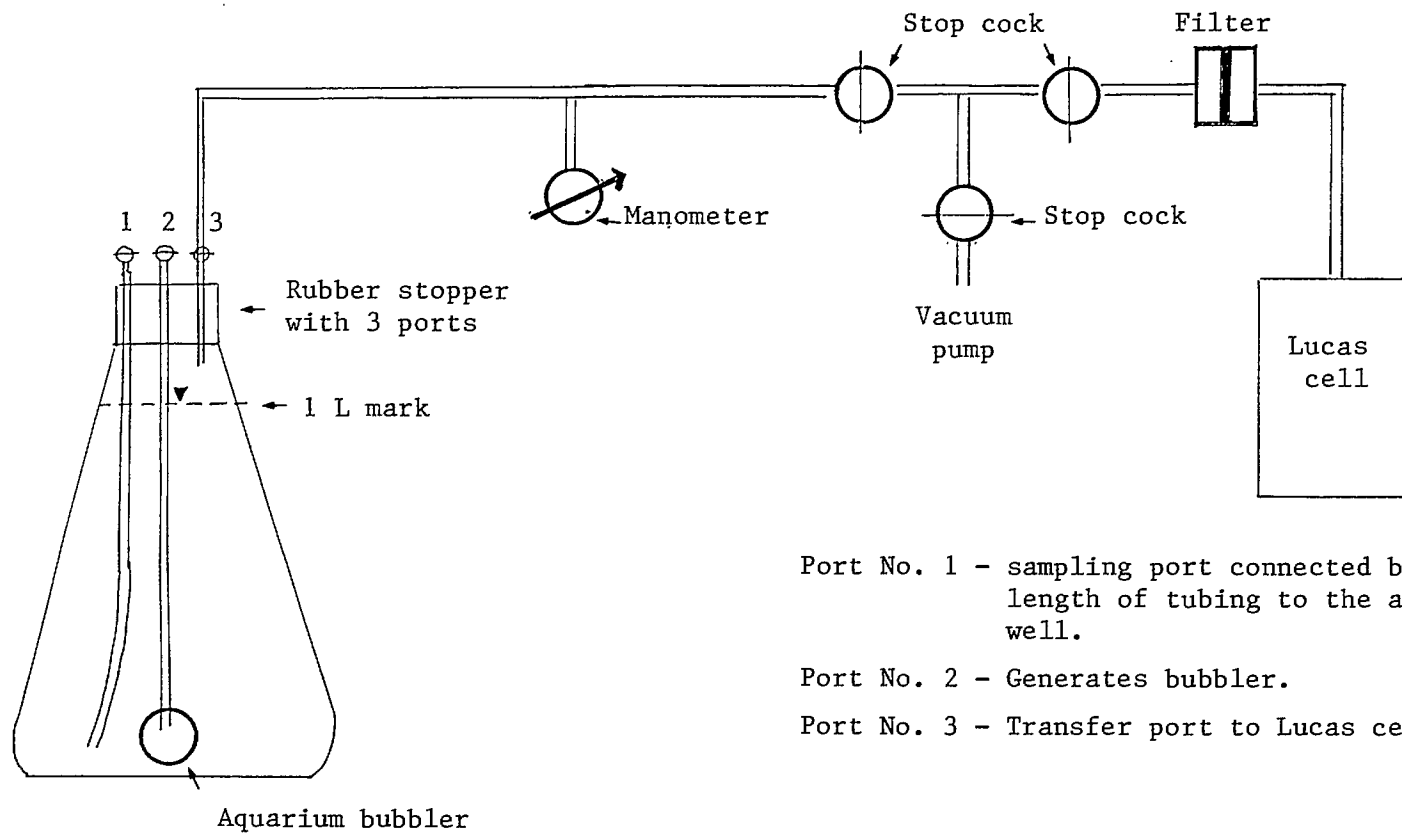
However, in this thesis the discussion will be limited to the subject of disequilibrium of radioisotopes in groundwater, therefore, only sampling and radon determinations related to this subject will be mentioned. For instance the determination of dissolved ^{222}Rn in a water sample originating from an artesian well near Elliot Lake was as follows:

A water sample was collected in a 1 litre bottle. A rubber stopper with three port holes was used to seal the bottle, and a long length of tubing was installed far enough into one of the port holes to reach the bottom of the bottle. This port hole was used to sample the water by flooding the bottle from the bottom upward, thus preventing turbulence and degassing while sampling. Another long length of tubing attached to a bubbler was inserted into the second hole until it rested on the bottom of the bottle. This port hole was used to bubble radon gas out of the water.

The third port hole was connected to a short length of tubing as an outlet to transfer radon gas into a Lucas cell (2.5 mL) (Figure 6.2).

A water sample was taken by completely immersing a long length of tubing into an artesian well. A rubber cork with a hole in it was used to access the other end of the tubing, thus sealing the well and as such restricting the water flow through the tubing and preventing diffusion of radon gas before sampling. The tubing from the well was then connected to the first port hole in the sample bottle and the water sample was allowed to flow into the bottle by flooding it from the bottom upward. The water sample was allowed to overflow through port holes No. 2 and 3 for 5 min, after which it reached a steady state condition and the inlet tubing was disconnected. The excess water was decanted to a litre mark and all three port holes were plugged with clamps.

Radon gas was transferred in the laboratory using the vacuum pump transfer system and the alpha-disintegrations were counted after ^{222}Rn reached equilibrium with its daughters.



- Port No. 1 - sampling port connected by a long length of tubing to the artesian well.
- Port No. 2 - Generates bubbler.
- Port No. 3 - Transfer port to Lucas cell

Fig. 6.2 - Radon bubbler and transfer system to Lucas cell.

CHAPTER 7**NORDIC MAIN TAILINGS - RESULTS AND DISCUSSION**

Parts of the data pertaining to the Nordic Main uranium tailings project, which are necessary to form a general cross-section of the tailings pile, are displayed in this Chapter. The data for the solid samples are shown in Tables 7.1 to 7.8, and from 7.9 to 7.12 for the liquid phase porewater samples, respectively. Data for the radioisotope ratios in the solid tailings samples are displayed in Tables 7.13 to 7.16.

Additional data from location T-5 within the Nordic Main tailings are summarized in Tables 7.17 to 7.18. A general cross-sectional view of the Nordic Main tailings is shown in Figure 7.1, and a general flow pattern within the tailings pile is shown in Figure 7.2. The flow net patterns were drawn from interpretation of data from the hydraulic head measurements, and were translated into equipotential line contour mapping using the Kriging technique (53,54).

The Kriging technique is a method used to create a grid pattern using a regional variable theory. The Kriging algorithm assumes an underlying linear variogram, which produces a more accurate contour in comparison to the conventional inverse distance algorithm (Surfer TM version 3.0) (54).

Results for the chemical and radioisotope stratification at various depths for each station are presented in units of $\mu\text{g/g}$ or mBq/g for solid samples, and in mg/L or mBq/L for liquid phase porewater samples, respectively, and are plotted in Figures 7.3 to 7.55. Only the parts of the stratification figures, which are related to the subject of radioisotope disequilibrium are presented here. For example, the stratification figures for thorium and uranium in the solid tailings originating from locations T-1, T-2, T-3, T-7 and T-8 are presented in Figures 7.3 to 7.12, respectively, and

for the liquid phase porewater samples, are presented in Figures 7.13 to 7.17, respectively.

Results for the stratification of lead content in the solid and liquid phases are presented in Figures 7.18 to 7.25, respectively. ^{210}Pb content is presented in Figures 7.26 to 7.30. ^{226}Ra content is presented in Figures 7.31 to 7.40. The stratification of ^{228}Th , ^{230}Th and ^{232}Th of solid tailings samples from locations T-1, T-2, T-3, T-7 and T-8 are presented in Figures 7.41 to 7.55, respectively. No thorium isotope data were obtained from the liquid phase porewater samples.

Contour maps outlining the concentration intervals of the tailings cross-section, from North to South, are presented in Figures 7.56 to 7.90. The stratification figures and the contour maps were derived from data obtained from Tables 7.1 to 7.12.

The distribution coefficient K_d is a widely used parameter in studies of groundwater contamination, which reflect the partitioning of solutes between liquid and solid phases in a porous medium, and can be expressed as:

$$K_d = \frac{\text{mass of solute on the solid phase/unit mass of solid phase}}{\text{concentration of solute in solution}}$$

The measured K_d values are normally reported in mL/g (Freeze and Cherry, Chapter 9 (53)).

The distribution coefficients for sulphate are presented in Figures 7.62 and 7.63 (Table 7.14). The first figure is based on the partitioning of the liquid phase and the bulk mass of sulphate in the solid phase. The second figure reflects the partitioning between the liquid phase and the mass of sulphate adsorbed, or precipitated on the solids per gram of bulk dry tailings.

The major difference between the two sets of sulphate data is the procedure for the pretreatment of samples prior to sulphate analyses. The

bulk mass of sulphate in the solid phase was obtained by fusing the solid tailings sample using borax flux, which resulted in a complete dissolution of the solids. This pretreatment method gave an abnormally high reading of the measured sulphate level in the solid phase which did not reflect the true value of the adsorbed or precipitated sulphate on the solid. This method is simpler and more economical to perform. The second data sets were obtained by leaching the adsorbed and precipitated sulphate out of the solid. This pretreatment method gave a more accurate set of data regarding the partitioning between the liquid phase and the adsorbed or precipitated sulphate on the solid, however, it is time consuming and results vary from sample to sample unrepeatably since the measurement depends on the size distribution, i.e., the sample cannot be homogenized by pulverizing it prior to analysis.

Comparison of the contour shapes between Figures, 7.62 and 7.63, indicate that both show the same trend, i.e., enrichment of sulphate in the liquid phase as the porewater moves downward (Figure 7.2), and the calcium distribution coefficient seems to follow the same trend as sulphate (Figure 7.69). This probably occurred because these constituents exist in the form of calcium sulphate (Dalton) (38-40). The magnesium distribution coefficient, Figure 7.72, also follows the same trend as calcium, i.e., enrichment of magnesium in the liquid phase as the porewater moves downward (Figure 7.2).

The distribution coefficients for Fe, Al, Th and U, show reverse trends to those for the sulphate, i.e., enrichment of Fe, Al, Th and U into the solid phase either by adsorption or reprecipitation as the porewater moves downward. The results are shown in Figure 7.66, 7.75 and 7.78, respectively.

These events are very well known, since these species, especially Fe,

Al, U, Th and Pb are pH dependent. As the porewater moves downward from the low pH zone within the upper layer of the water table, it carries leachable products, such as heavy metals and other trace elements. The porewater gradually reacts with a buffering agent within the tailings pile, such as calcium carbonate and calcium hydroxide (lime), which were added as a neutralizing agents in the mill circuit before discharge into the tailings pond. Therefore, the pH is gradually increased as the porewater moves downward, and at the same time Al, Fe, U, Th and Pb are reprecipitated with the increase of pH. Most of the trace elements, such as radium, and probably protactinium, actinium, polonium, bismuth and thalium are dependent on other scavenger elements, such as aluminium, iron and manganese as hydroxide to coprecipitate with, or barium if present, in the solution (47.57). However, the study of scavenging is beyond the scope of this work.

As these elements move downward from the upper layer of the water table, their concentration in the water phase diminishes, and their concentration in the solid phase shows enrichment as a product of reprecipitation, coprecipitation and adsorption (1,4,16-21,27-32).

The activity ratios of daughter/parent isotopes within the solid tailings are shown in Figures 7.91 to 7.93 (Tables 7.13 to 7.16). The activity ratios for $^{210}\text{Pb}/^{226}\text{Ra}$ show an expected secular disequilibrium pattern with a ratio of less than 1.0 (Figure 7.91). However, the activity ratios for $^{226}\text{Ra}/^{230}\text{Th}$ and $^{228}\text{Th}/^{232}\text{Th}$ show an unusual pattern with a high ratio of approximately 20:1 close to the surface of the tailings which decreases with depth on approaching 1.0 at the peat layer.

Unfortunately, no analyses of ^{210}Pb , ^{224}Ra , ^{228}Th , ^{230}Th or ^{232}Th on the liquid phase were performed, therefore, there are no activity ratio data available for comparison with liquid samples from T-1, T-2, T-3, T-7 or T-8

cross-sections. However, limited data are available for the T-5 location (Tables 7.17 and 7.18) and the results show that the activity ratio between daughter/parent, especially for $^{210}\text{Pb}/^{226}\text{Ra}$ ratio in the liquid phase is as high as 164, and is approximately 200 times larger than the ratio in the solid phase. These ratios are very puzzling since, in general, it is expected that secular equilibrium should exist between the short-lived daughters and long-lived parents, and at least, if disequilibrium should exist, the ratio of daughter to parent should be less than 1.0, or within the permissible statistical limit of the analytical method used. This type of secular disequilibrium should be called paradoxical secular disequilibrium since its behaviour is very irregular and beyond the boundary of the expected ratio for secular equilibrium. There were a number of arguments in the past caused by misunderstanding, since most of the time researchers are concentrating their efforts on finding the solution to the problems by investigating only the liquid phase. This paradoxical secular disequilibrium behaviour of radioisotopes has some similarity with most of the data observed by other investigators in Chapter 3 (1-26).

However, if one reviews the other basic rule of secular equilibrium, that the sample should be undisturbed, and no parent or daughter substances removed or allowed to escape for a time long enough for secular equilibrium to be established, then it is expected, that if parts of the daughters or the parents are removed, disequilibrium will exist in the total bulk mass. Also, in the solid phase, if the parent is removed by a leaching process, or a daughter is gained by reprecipitation and adsorption at a rate faster than the time required to gain equilibrium, then a paradoxical secular disequilibrium may exist. A paradoxical secular disequilibrium also exists in the liquid phase, if the daughter activity is gained by preferential leaching, or if the parent isotopes reprecipitate at a faster rate than the

time needed to reach equilibrium.

As an example, let us compare the solubilities of the three main elements in Figure 7.91 to 7.93, i.e., radium, lead, uranium and thorium with the majority of the anion as sulphate. The solubilities of the sulphate salts are obtained from the Handbook of Chemistry and Physics (55) and tabulated in Table 7.19. The solubility table shows that radium sulphate is almost insoluble, followed by lead sulphate, thorium sulphate, and uranium sulphate which is very soluble. Therefore, it can be shown and concluded here, that the behaviour of the activity ratio distribution of $^{210}\text{Pb}/^{226}\text{Ra}$ (less than 1.0) in the solid tailings cross-section is caused by preferential leaching or removal of ^{210}Pb from the solid phase over ^{226}Ra , and at a faster rate than ^{226}Ra , decays into ^{210}Pb to reach secular equilibrium (Figure 7.91).

The activity ratio in the liquid phase is controlled more by the pH, solubility or chemical equilibrium constant of $^{210}\text{Pb}(\text{SO}_4)$, and $^{226}\text{Ra}(\text{SO}_4)$, and the residence time of the porewater (Chapter 7, Freeze and Cherry (53)). This ratio is sometimes extremely high, caused by the higher availability of ^{210}Pb with the increase of ^{226}Ra activity present in the bulk mass, and a higher solubility with decreases in pH.

It can also be shown that a similar process occurs for the $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio in the solid phase, where thorium sulphate is more soluble, and therefore is more easily leached than the radium sulphate (Figure 7.92).

The differences in the activity ratios of $^{226}\text{Ra}/^{230}\text{Th}$ from $^{210}\text{Pb}/^{226}\text{Ra}$, i.e., $^{226}\text{Ra}/^{230}\text{Th}$ values, are mostly between 1 to 10, caused by the long half-life of ^{230}Th (half-life = 8×10^4 y), and therefore, long production time to produce ^{226}Ra . Because of the high solubility of thorium sulphate, the ^{230}Th was removed faster by the leaching process than its production rate for ^{226}Ra .

Whatever the initial ^{226}Ra activity was, this isotope was assumed to be in equilibrium with ^{230}Th at the time of the initial tailings disposal from the milling process. ^{226}Ra is leached at a much slower rate than the ^{230}Th , and therefore, reflects a higher activity ratio, larger than 1.0 in the solid sample.

^{210}Pb is a product of a much faster rate of decay from ^{226}Ra (1.6×10^3 y), and since the lead sulphate solubility is much lower than ^{230}Th sulphate, but much higher than radium sulphate, it is therefore, possible for ^{210}Pb to build up its activity to near secular equilibrium as the geochemical dynamic leaching process of the tailings continues to leach lead sulphate faster than the radium sulphate. This results in an activity ratio of less than 1.0 in the solid phase.

Another interesting case is the activity ratio of $^{228}\text{Th}/^{232}\text{Th}$, in which the activity ratio shows a similar trend to that in $^{226}\text{Ra}/^{230}\text{Th}$, although ^{228}Th and ^{232}Th belong to the same species and have the same chemical property (solubility). The ^{232}Th decay series (Figure 6.1) shows that ^{228}Th is a product of a decay series from $^{232}\text{Th} \rightarrow ^{228}\text{Ra} \rightarrow ^{228}\text{Ac} \rightarrow ^{228}\text{Th}$, and therefore, ^{228}Th is not a direct product of ^{232}Th . It is assumed that the activity ratio of $^{228}\text{Ra}/^{232}\text{Th}$ was initially at, or near, secular equilibrium for the tailings deposited, and as the dynamic geochemistry process continued, the ^{232}Th was leached at a much faster rate than the ^{228}Ra .

On taking into account that the half-lives of ^{232}Th (1.4×10^{10} y), and ^{228}Ra (6.7 y), to ^{228}Ac (6.1 h), and to ^{228}Th , it can be concluded that ^{228}Ra is the controlling factor causing the segregation of ^{232}Th and ^{228}Th .

It has been shown that in the low pH water carrying the leached product of ^{232}Th , the ^{228}Ra movement is retarded by the low solubility of radium sulphate, and ^{228}Th is continuously generated by ^{228}Ra , resulting in a

trend of activity ratio similar to $^{226}\text{Ra}/^{230}\text{Th}$, which has a high ratio near the surface of the tailings pile and decreases downward towards a ratio of 1.0 near the peat layer.

According to Veska (26), similar cases with a sharp rise of $^{234}\text{U}/^{238}\text{U}$ in the liquid phase and a continued trend of relative ^{234}U enrichment were attributed to a scenario requiring preferential leaching of ^{234}U in the 6+ oxidation state (UO_2^+), a stronger absorption of ^{238}U (as U^{4+}), over ^{234}U (as UO_2^+) in the sand aquifer, and little redox exchange between the 4+ and 6+ state. This explanation is not entirely valid, as was mentioned by Morin (27), since ^{238}U or ^{234}U can exist in a 4+, 5+ or 6+ state.

However, preferential leaching for $^{234}\text{U}/^{238}\text{U}$ exists, and can also be caused differently, for example, in a scenario where the initial activity ratios of ^{238}U , ^{234}Th , ^{234}Pa and ^{234}U in the solid phase are in secular equilibrium. As the solid phase is leached by low pH water, the ^{238}U and ^{234}U leach out at an equal ratio, since they both behave as chemical uranium.

Our results show that ^{234}Th and ^{234}Pa were leached out at a lower rate than the uranium (uranium sulphate is more soluble than thorium sulphate, see Table 7.19). We expect that the movement of ^{234}Th could be retarded, resulting in enrichment of ^{234}Th in the solid phase, and at the same time ^{234}Th decays and generates its daughter ^{234}Pa , which also decays into ^{234}U . Since the half-life of ^{234}Th and ^{234}Pa is very short in comparison to ^{238}U and ^{234}U , the increase of ^{234}U activity is also very rapid. Thus, the actual rapid increase of $^{234}\text{U}/^{238}\text{U}$ activity ratio in the liquid phase can be attributed to the additional activity of ^{234}U in the liquid phase through the retardation of ^{234}Th movement in the solid phase. Therefore, the $^{234}\text{U}/^{238}\text{U}$ activity ratio in the liquid phase reflects the enrichment of ^{234}Th in the solid phase and it increases with distance from the source as the ^{234}Th generates ^{234}U .

CBCL Ltd. (58), claimed that $^{234}\text{U}/^{238}\text{U}$ activity ratio in the uraniferous peat samples, averaging 1.13, was attributed to the mass recoil and ejection of ^{234}U from ^{238}U in the uranium ore bearing mineral when ^{238}U decays, resulting in higher solubility of ^{234}U than ^{238}U . Assuming that:

- all uranium originally dissolved in the groundwater was precipitated or adsorbed within the peat (supported by $^{234}\text{U}/^{238}\text{U}$ ratios);
- ^{230}Th has low solubility (i.e., no ^{230}Th was transported off site);
- all of the ^{232}Th produced remained in the deposit; and
- ^{238}U and ^{234}U are in equilibrium, then ^{238}U activity could be estimated and adjusted from the measured total uranium concentration. Therefore, CBCL Ltd. concluded that the peat layer is a recent deposit (i.e., less than 10,000 to 15,000 years) since little time has been available for the ingrowth of radionuclide daughter products of uranium (Table 21 of CBCL report (58)). The deposition of uranium within the peat deposit is considered to be an on-going process.

The explanation given for $^{234}\text{U}/^{238}\text{U}$ ratio of 1.13 average as being caused by mass recoil and ejection of ^{234}U from ^{238}U is probably inaccurate. As mentioned earlier, ^{234}U is not a direct daughter of ^{238}U . Mass recoil and ejection of daughters can also happen to ^{234}Th and ^{234}Pa during their decay chains.

^{234}Th (24.1 d) and ^{234}Pa (1.2 m) could disappear quickly from the solution because of its short half-life and transform through decay chains into ^{234}U (2.5×10^5 y), therefore resulting in a higher activity ratio of $^{234}\text{U}/^{238}\text{U}$ in groundwater, consistent with those of soil samples. In Table 21 of the CBCL Ltd. report (58), the age of the uranium mineralization within the deposit was estimated from the ratios of measured ^{230}Th activities and calculated ^{238}U activities and the decay rate of uranium. As was mentioned by CBCL, the uranium deposition is considered to be an on-going process,

i.e., the hydrogeochemical process is very active. Thus it can be concluded that the age measurement, as reported by CBCL Ltd., could be seriously in error since ^{234}U could be added to the peat layer as the dynamic hydrogeochemical process is actively on-going. Thus, the more ^{234}U added to the peat layer, the more ^{230}Th will be produced, and hence the $^{230}\text{Th}/^{238}\text{U}$ ratio could be higher than expected. Maybe it would be better if the age dating was determined using a $^{230}\text{Th}/^{234}\text{U}$ activity ratio instead of ^{238}U , assuming that ^{230}Th is not in the peat layer to begin with.

Therefore, one must be very careful when interpreting data for the dynamic decay chain, especially when dealing with radiochemistry and related subjects, such as radioactive dating, uptake of radioactive materials into biological matter as part of the food chain, etc. within the hydrogeochemical dynamic environmental conditions.

It can be concluded here, that the solid phase system is acting as an indicator of the long term results of hydrogeochemical evolution, which appears to be a very slow-moving process. Solid phase results are extremely valuable in the study of the evolution of radioisotope species from the uranium tailings. In comparison, the liquid phase system is a much faster short term indicator of what happens from day to day, and sometimes the changes are so rapid, such as the oxidation mechanism of iron species, that complex problems arise in the design of mathematical models for the long term tailings events. There are many more parameters to deal with in the liquid phase.

On the basis of our studies, we conclude that when dealing with the solid phase, especially with the radioisotope species, a non-destructive analytical method, such as the use of borehole spectral gamma-logging could be more economical, even though it will probably require a longer time interval for sampling. The mathematical modelling could probably be

simplified by using only an empirical model.

However, both solid and liquid phases exist together, and at the present time interest is concentrated in the effluent discharge, which is directly related to environmental pollution control. The liquid phase is still the major issue in this tailings study. Studies of the solid phase serve to enhance our knowledge of the hydrogeochemical evolution.

Our study has shown that, when dealing with hydrogeochemical processes and field data interpretation for radioisotope migration, one must have an in-depth understanding of the physics of radioisotope decay, the chemical behaviour of the isotopes, and the groundwater hydrology of the system.

A laboratory test to demonstrate the paradoxical secular disequilibrium and hydrogeochemical processes will be discussed in Chapter 8.

Table 7.1 - Nordic main tailings data. Solid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

||Solid Sample Study

SAMPLE	Northing	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	Al	Ca	Fe
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)			
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	(mg/100g)	(mg/g)	(mg/g)
T - 1 - 1.00	544.80	350.79	13764.0		17760.0	3219.0	1258.0	185.0	3840	13.65	9.30
T - 1 - 1.50	544.80	350.79	7955.0		9768.0	703.0	5106.0	851.0			
T - 1 - 2.00	544.80	350.79	15873.0		21201.0	1813.0	22126.0	2738.0			
T - 1 - 2.00	544.80	350.79	15947.0		17131.0	2220.0	23199.0	3034.0			
T - 1 - 2.00	544.80	350.79	17760.0		21423.0	2775.0	23680.0	3034.0	3919	35.30	24.50
T - 1 - 2.50	544.80	350.79	13653.0		14171.0	1073.0	8991.0	1332.0			
T - 1 - 3.00	544.80	350.79	11803.0		14726.0	888.0	13209.0	1332.0	3806	21.58	26.80
T - 1 - 4.00	544.80	350.79	15799.0		14245.0	407.0	2035.0	999.0			
T - 1 - 4.00	544.80	350.79	18537.0		18056.0	2368.0	9805.0	2627.0			
T - 1 - 4.00	544.80	350.79	15799.0		18167.0	1443.0	6327.0	1147.0			
T - 1 - 4.25	544.80	350.79	12950.0		12506.0	888.0	2775.0	962.0			
T - 1 - 4.50	544.80	350.79	5846.0		6105.0	333.0	851.0	148.0			
T - 1 - 4.60	544.80	350.79	10471.0		11840.0	814.0	8325.0	962.0			
T - 1 - 4.75	544.80	350.79	32264.0		21830.0	3330.0	6660.0	2035.0			
T - 1 - 5.00	544.80	350.79	14615.0		15540.0	2183.0	6919.0	1517.0			
T - 1 - 5.25	544.80	350.79	31968.0		25752.0	1036.0	3182.0	999.0			
T - 1 - 5.50	544.80	350.79	12950.0		13653.0	1036.0	1924.0	888.0			
T - 1 - 5.75	544.80	350.79	13135.0		16132.0	666.0	2701.0	851.0			
T - 1 - 6.00	544.80	350.79	15651.0		15836.0	407.0	3885.0	1221.0	3053	14.65	32.50
T - 1 - 6.00	544.80	350.79	16502.0		18500.0	1480.0	5106.0	1332.0	3022	15.15	32.50
T - 1 - 6.00	544.80	350.79	18796.0		18537.0	1517.0	6031.0	1295.0			
T - 1 - 6.00	544.80	350.79	12950.0		14208.0	1147.0	11729.0	1332.0			
T - 1 - 6.25	544.80	350.79	10175.0		9398.0	1036.0	3145.0	925.0			
T - 1 - 6.50	544.80	350.79	13542.0		14134.0	592.0	2664.0	592.0			
T - 1 - 6.75	544.80	350.79	8806.0		8954.0	370.0	2146.0	888.0			
T - 1 - 7.00	544.80	350.79	19277.0		14430.0	481.0	3885.0	814.0			
T - 1 - 7.00	544.80	350.79	12247.0		13468.0	740.0	4847.0	592.0	2890	9.55	33.50
T - 1 - 7.25	544.80	350.79	5883.0		4810.0	74.0	259.0	74.0			
T - 1 - 7.40	544.80	350.79	17057.0		20609.0	2331.0	9398.0	1036.0			
T - 1 - 7.50	544.80	350.79	8954.0		10989.0	296.0	2516.0	481.0			
T - 1 - 7.60	544.80	350.79	10619.0		12728.0	962.0	6031.0	703.0			
T - 1 - 7.60	544.80	350.79	10619.0		12469.0	2627.0	6697.0	777.0			
T - 1 - 7.60	544.80	350.79	10360.0		11507.0	666.0	8695.0	1332.0	3810	17.25	34.00
T - 1 - 7.75	544.80	350.79	16539.0		15096.0	555.0	2590.0	703.0			
T - 1 - 8.00	544.80	350.79	3885.0		6068.0	2109.0	14393.0	1036.0	2326	7.90	37.00
T - 1 - 8.00	544.80	350.79	23754.0		19610.0	1813.0	6105.0	2035.0			
T - 1 - 8.25	544.80	350.79	35853.0		24494.0	2627.0	6364.0	1554.0			
T - 1 - 8.50	544.80	350.79	19388.0		22459.0	925.0	7770.0	1036.0			
T - 1 - 8.50	544.80	350.79	24901.0		19092.0	666.0	2960.0	1036.0			
T - 1 - 8.50	544.80	350.79	26307.0		20239.0	3145.0	10360.0	2775.0			
T - 1 - 9.00	544.80	350.79	18759.0		20239.0	1924.0	14948.0	1554.0			
T - 1 - 9.00	544.80	350.79	18685.0		21497.0	1665.0	14652.0	1480.0			
T - 1 - 9.00	544.80	350.79	15725.0		18241.0	2738.0	16428.0	1887.0	5053	45.15	33.30
T - 1 - 9.60	544.80	350.79	16761.0	1295.0	13579.0	1476.3	18107.8	1587.3	5785	41.60	39.50
T - 1 - 9.60	544.80	350.79	17908.0	1480.0	17782.2	1968.4	22618.1	2234.8	6475	43.70	37.00
T - 1 -10.20	544.80	350.79	20646.0	2849.0	20398.1	2886.0	26551.2	2667.7	6000	42.60	37.00
T - 1 -10.60	544.80	350.79	621.6	74.0	3307.8	59.2	292.3	25.9	946	22.00	4.07
T - 1 -10.70	544.80	350.79	218.3	74.0	266.4	55.5	310.8	77.7	1676	17.45	2.32

Table 7.2 - Nordic main tailings data. Solid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

||Solid Sample Study

SAMPLE	Northing	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	Al	Ca	Fe
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)			
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	(mg/100g)	(mg/g)	(mg/g)
T - 1 -10.77	544.80	350.79	12062.0	2072.0	13908.3	1857.4	18396.4	1713.1	4125	43.00	22.32
T - 1 -10.80	544.80	350.79	225.7	70.3	336.7	85.1	188.7	51.8	2454	8.85	4.54
T - 1 -11.18	544.80	350.79	484.7	122.1	588.3	129.5	762.2	136.9	5837	12.20	16.80
T - 1 -12.57	544.80	350.79	18796.0		17168.0	2146.0	25345.0	2442.0			
T - 2 - 1.00	518.74	351.58	14689.0		16391.0	370.0	444.0	22.2	3779	24.25	6.30
T - 2 - 1.40	518.74	351.58	10138.0		13764.0	666.0	1480.0	185.0			
T - 2 - 2.00	518.74	351.58	12987.0		17575.0	2886.0	18907.0	3293.0	3604	21.15	29.50
T - 2 - 3.00	518.74	351.58	8140.0		9805.0	1480.0	11211.0	1961.0			
T - 2 - 4.00	518.74	351.58	14504.0		12506.0	925.0	11211.0	1369.0			
T - 2 - 4.00	518.74	351.58	18130.0		16909.0	2220.0	10582.0	1221.0			
T - 2 - 4.00	518.74	351.58	17834.0		15762.0	1184.0	10841.0	1258.0			
T - 2 - 4.60	518.74	351.58	15096.0		15836.0	666.0	4921.0	629.0	3022	65.60	31.80
T - 2 - 5.00	518.74	351.58	10693.0		13061.0	2294.0	21090.0	2146.0			
T - 2 - 5.79	518.74	351.58	22163.0		18796.0	1332.0	11914.0	925.0			
T - 2 - 6.02	518.74	351.58	25197.0		23495.0	1628.0	17834.0	1776.0			
T - 2 - 6.93	518.74	351.58	21571.0		22829.0	1147.0	16465.0	1739.0			
T - 2 - 7.24	518.74	351.58	32745.0		27824.0	2479.0	22977.0	2442.0			
T - 2 - 8.00	518.74	351.58	23495.0		21978.0	2812.0	22718.0	2812.0	3984	44.80	39.50
T - 2 - 8.31	518.74	351.58	14319.0		14726.0	1184.0	10804.0	1591.0			
T - 2 - 8.76	518.74	351.58	22089.0		22459.0	2146.0	23569.0	2590.0			
T - 2 - 8.76	518.74	351.58	21867.0		25937.0	1887.0	21608.0	2849.0	5437	43.00	37.30
T - 2 - 8.76	518.74	351.58	23902.0		22866.0	1961.0	22903.0	2516.0			
T - 2 - 9.07	518.74	351.58	33522.0		26566.0	3626.0	29267.0	3256.0			
T - 2 - 9.07	518.74	351.58	31968.0		28897.0	3108.0	29822.0	3996.0			
T - 2 - 9.07	518.74	351.58	30414.0		26862.0	2997.0	28934.0	3737.0	6074	55.00	47.50
T - 2 - 9.61	518.74	351.58	27491.0		31931.0	3552.0	27306.0	3034.0	6026	60.40	46.00
T - 2 - 9.77	518.74	351.58	37259.0		42180.0	3996.0	42624.0	4810.0	5497	71.50	49.00
T - 2 - 9.79	518.74	351.58	27972.0	2664.0	35786.4	2934.1	48421.9	2941.5	6558	73.20	51.00
T - 2 - 9.87	518.74	351.58	33633.0	5254.0	35989.9	4632.4	51788.9	5820.1	6050	49.50	50.00
T - 2 - 9.94	518.74	351.58	27750.0	3034.0	29326.2	2856.4	35353.5	5220.7	5215	58.50	46.00
T - 2 -10.15	518.74	351.58	321.9	7.4	492.1	14.8	236.8	37.0	898	18.20	9.93
T - 2 -10.25	518.74	351.58	181.3	92.5	858.4	85.1	159.1	40.7	4032	7.80	5.00
T - 2 -10.33	518.74	351.58	233.1	44.4	318.2	96.2	103.6	18.5	6144	17.05	18.00
T - 3 - 1.00	509.62	350.83	15540.0		17538.0	888.0	925.0	114.7			
T - 3 - 1.30	509.62	350.83	12876.0		13061.0	1221.0	6512.0	777.0			
T - 3 - 2.00	509.62	350.83	9583.0		9842.0	666.0	2516.0	222.0			
T - 3 - 2.60	509.62	350.83	24383.0		19055.0	1332.0	14800.0	1332.0			
T - 3 - 3.00	509.62	350.83	11803.0		10582.0	703.0	6549.0	703.0			
T - 3 - 3.00	509.62	350.83	11655.0		11618.0	555.0	7400.0	814.0			
T - 3 - 3.00	509.62	350.83	11433.0		11211.0	592.0	5846.0	666.0			
T - 3 - 4.00	509.62	350.83	5476.0		7511.0	407.0	1628.0	444.0	4435	39.40	30.50
T - 3 - 4.30	509.62	350.83	26011.0		27084.0	2183.0	23051.0	2553.0			
T - 3 - 4.30	509.62	350.83	27232.0		24457.0	1998.0	21904.0	2368.0			
T - 3 - 4.30	509.62	350.83	27528.0		23828.0	2072.0	21460.0	2257.0			
T - 3 - 5.00	509.62	350.83	11359.0		14874.0	1443.0	9250.0	1295.0	2787	14.25	28.50
T - 3 - 6.93	509.62	350.83	48285.0		49136.0	6549.0	41773.0	6808.0			
T - 3 - 6.93	509.62	350.83	44474.0		53465.0	4884.0	40959.0	6660.0	5619	61.40	38.50
T - 3 - 6.93	509.62	350.83	48248.0		44067.0	4514.0	39849.0	5661.0			

Table 7.3 - Nordic main tailings data. Solid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

Solid Sample Study											
SAMPLE	Northing	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	Al	Ca	Fe
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)			
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	(mg/100g)	(mg/g)	(mg/g)
T - 3 - 7.25	509.62	350.83	20868.0		19166.0	777.0	4551.0	1036.0			
T - 3 - 7.25	509.62	350.83	22089.0		17131.0	1924.0	4662.0	851.0			
T - 3 - 7.25	509.62	350.83	19869.0		22792.0	1887.0	5550.0	1295.0			
T - 3 - 7.50	509.62	350.83	22089.0		17427.0	1850.0	4810.0	1332.0			
T - 3 - 7.54	509.62	350.83	22422.0		24679.0	2294.0	24198.0	2664.0	4771	19.60	34.30
T - 3 - 7.75	509.62	350.83	12839.0		12617.0	851.0	2257.0	592.0			
T - 3 - 7.85	509.62	350.83	28194.0		32116.0	2849.0	41329.0	3848.0			
T - 3 - 7.85	509.62	350.83	29489.0		34891.0	4810.0	42328.0	4995.0			
T - 3 - 7.85	509.62	350.83	28194.0		37481.0	3071.0	39257.0	3515.0	4634	66.40	40.00
T - 3 - 7.96	509.62	350.83	2479.0		3071.0	296.0	1110.0	185.0			
T - 3 - 7.97	509.62	350.83	29970.0		27972.0	1184.0	3330.0	666.0			
T - 3 - 8.01	509.62	350.83	1406.0		1813.0	148.0	74.0	18.5			
T - 3 - 8.06	509.62	350.83	518.0		518.0	74.0	222.0	74.0	7243	18.85	42.50
T - 3 - 8.29	509.62	350.83	418.1	74.0	710.4	88.8	1098.9	107.3			
T - 3 - 8.53	509.62	350.83	1554.0	218.3	1872.2	203.5	1491.1	244.2	2938	14.25	34.00
T - 7 - 1.00	496.41	351.95	10027.0		12506.0	481.0	1184.0	148.0			
T - 7 - 1.30	496.41	351.95	13949.0		17094.0	888.0	2183.0	296.0			
T - 7 - 2.00	496.41	351.95	8103.0		12136.0	629.0	740.0	66.6			
T - 7 - 3.00	496.41	351.95	16095.0		19610.0	888.0	18870.0	1628.0			
T - 7 - 3.00	496.41	351.95	10841.0		11803.0	1147.0	17020.0	2516.0			
T - 7 - 4.00	496.41	351.95	14615.0		14615.0	1850.0	8436.0	1110.0			
T - 7 - 5.00	496.41	351.95	12432.0		17242.0	1480.0	9879.0	1332.0			
T - 7 - 5.00	496.41	351.95	12506.0		15096.0	1406.0	8658.0	999.0			
T - 7 - 5.00	496.41	351.95	12247.0		19425.0	2183.0	9805.0	1517.0			
T - 7 - 6.00	496.41	351.95	11914.0		15244.0	1406.0	8547.0	1184.0			
T - 7 - 6.40	496.41	351.95	14245.0	1480.0	18437.1	1420.8	14448.5	1139.6	3293	15.43	32.80
T - 7 - 6.90	496.41	351.95	30081.0	2960.0	29511.2	3204.2	28915.5	3193.1	4734	36.00	39.50
T - 7 - 6.90	496.41	351.95	27972.0	2775.0	28342.0	2516.0	26880.5	3178.3	4531	33.95	34.30
T - 7 - 7.00	496.41	351.95	26344.0		31043.0	2072.0	17760.0	2886.0			
T - 7 - 7.20	496.41	351.95	28194.0	2960.0	27539.1	3204.2	32844.9	3470.6	4463	40.00	39.50
T - 7 - 7.70	496.41	351.95	14578.0	1517.0	14037.8	1716.8	15273.6	2604.8	3262	21.40	32.80
T - 7 - 7.81	496.41	351.95	7770.0	444.0	5846.0	532.8	4181.0	377.4	2760	14.30	29.00
T - 7 - 7.86	496.41	351.95	15355.0	2183.0	15318.0	1920.3	16265.2	2379.1	4550	22.05	37.00
T - 7 - 7.91	496.41	351.95	24198.0	2886.0	25382.0	2941.5	28893.3	4699.0	4881	58.00	42.30
T - 7 - 7.96	496.41	351.95	16021.0		16354.0	1332.0	10952.0	1369.0			
T - 7 - 8.38	496.41	351.95	2812.0	333.0	2797.2	355.2	1938.8	270.1	6255	12.00	37.00
T - 7 - 8.99	496.41	351.95	814.0	111.0	888.0	122.1	362.6	96.2	5856	16.55	38.00
T - 8 - 1.00	490.78	353.47	7474.0		8954.0	703.0	407.0	44.4			
T - 8 - 2.00	490.78	353.47	5920.0		6438.0	259.0	296.0	48.1			
T - 8 - 3.00	490.78	353.47	5402.0		7178.0		2220.0	2405.0			
T - 8 - 3.00	490.78	353.47	6068.0		7918.0	333.0	1739.0	148.0			
T - 8 - 4.00	490.78	353.47	7178.0		10989.0	296.0	481.0	77.7			
T - 8 - 5.00	490.78	353.47	6697.0		8991.0	666.0	10656.0	851.0			
T - 8 - 6.00	490.78	353.47	7733.0		8695.0	518.0	8325.0	703.0			
T - 8 - 6.00	490.78	353.47	7622.0		9324.0	629.0	8066.0	703.0			
T - 8 - 6.00	490.78	353.47	8399.0		8880.0	592.0	8140.0	888.0			
T - 8 - 7.00	490.78	353.47	19499.0		21497.0	1665.0	14837.0	1739.0			
T - 8 - 7.60	490.78	353.47	18056.0	1591.0	14474.4	1539.2	17131.0	1502.2	3634	20.50	36.50

Table 7.4 - Nordic main tailings data. Solid phase sample results of chemicals and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

||Solid Sample Study

SAMPLE	Northing	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	Al	Ca	Fe
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)			
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	(mg/100g)	(mg/g)	(mg/g)
T - 8 - 8.40	490.78	353.47	8991.0	1961.0	7437.0	1879.6	8376.8	703.0	2487	7.35	35.00
T - 8 - 8.40	490.78	353.47	7511.0	851.0	8658.0	1172.9	7440.7	1354.2	2625	7.95	34.30
T - 8 - 8.76	490.78	353.47	14652.0	1628.0	14999.8	1572.5	13149.8	1609.5	3310	10.80	36.30
T - 8 - 9.07	490.78	353.47	26640.0		28823.0	5254.0	19055.0	2664.0			
T - 8 - 9.37	490.78	353.47	16687.0	1406.0	15351.3	973.1	10855.8	862.1	5540	16.95	42.50
T - 8 - 9.98	490.78	353.47	41736.0		42809.0	5624.0	40145.0	3811.0			
T - 8 -10.29	490.78	353.47	24790.0		26973.0						
T - 8 -10.29	490.78	353.47	30081.0		26196.0	3293.0	31302.0	3700.0			
T - 8 -10.29	490.78	353.47	29045.0		31524.0	1924.0	21201.0	2109.0			
T - 8 -11.05	490.78	353.47	23347.0		24013.0	1665.0	25530.0	2664.0			
T - 8 -11.20	490.78	353.47	22385.0		22200.0	1776.0	21127.0	2257.0			
T - 8 -11.44	490.78	353.47	33781.0		32893.0	2553.0	28379.0	2627.0			
T - 8 -11.49	490.78	353.47	12543.0		12432.0	518.0	15170.0	962.0			
T - 8 -11.60	490.78	353.47	5328.0		6956.0	1184.0	2183.0	222.0			
T - 8 -11.65	490.78	353.47	9324.0	1110.0	9224.1	865.8	10145.4	965.7			
T - 8 -11.65	490.78	353.47	9805.0		10434.0	925.0	9287.0	888.0	2988	12.90	32.80
T - 8 -12.24	490.78	353.47	6845.0		7422.2	740.0	7248.3	825.1	3566	12.05	30.50

Table 7.5 - Nordic main tailings data. Solid phase sample results of chemicals and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram

SAMPLE	K	Mg	Pb	Silica	Silica Fused	Soluble	Total	Th	U
DESCRIPTION				Acid extract	w/ NaHCO3	Sulphate	Sulphate	(ppm)	(ppm)
DEPTH (m)	(mg/g)	(mg/g)	(mg/100g)	(mg/g)	(mg/g)	(mg/g)	(mg/g)		
T - 1 - 1.00	25.42	1.90	33.15			16.11	17.85	28	18
T - 1 - 1.50								114	38
T - 1 - 2.00								513	43
T - 1 - 2.00								550	45
T - 1 - 2.00	31.16	1.92	31.90	1.53	23.00	49.57	57.07	519	41
T - 1 - 2.50								180	102
T - 1 - 3.00	33.15	1.35	27.10			35.96	46.54	234	72
T - 1 - 4.00								304	49
T - 1 - 4.00									51
T - 1 - 4.00									55
T - 1 - 4.25								194	45
T - 1 - 4.50								54	50
T - 1 - 4.60								157	46
T - 1 - 4.75								482	86
T - 1 - 5.00								272	77
T - 1 - 5.25								522	89
T - 1 - 5.50								212	80
T - 1 - 5.75								220	70
T - 1 - 6.00	30.69	1.13	27.45			25.02	72.37	286	62
T - 1 - 6.00	22.13	1.04	26.55			24.61	61.85	316	70
T - 1 - 6.00									46
T - 1 - 6.00								214	61
T - 1 - 6.25								126	58
T - 1 - 6.50								164	63
T - 1 - 6.75								100	67
T - 1 - 7.00								286	94
T - 1 - 7.00	29.40	1.07	24.40			16.89	63.44	106	85
T - 1 - 7.25								196	58
T - 1 - 7.40								176	87
T - 1 - 7.50								106	75
T - 1 - 7.60								128	64
T - 1 - 7.60								129	62
T - 1 - 7.60	23.38	0.86	21.60			23.48	81.61	131	64
T - 1 - 7.75								172	76
T - 1 - 8.00	19.36	0.57	9.10			10.20	87.35	40	25
T - 1 - 8.00								348	86
T - 1 - 8.25								618	61
T - 1 - 8.50									93
T - 1 - 8.50								532	104
T - 1 - 8.50								582	85
T - 1 - 9.00								323	118
T - 1 - 9.00								354	127
T - 1 - 9.00	25.99	1.92	26.75			58.64	81.50	309	119
T - 1 - 9.60		1.58	28.00	1.03	53.50	49.57	67.50		
T - 1 - 9.60	27.98	1.52	30.85			51.84	81.50		
T - 1 - 10.20		1.86	34.15	2.53	22.00	65.45	43.68		
T - 1 - 10.60	2.64	2.56	2.20			0.87	6.20		
T - 1 - 10.70	2.88	1.90	28.00			0.76	6.64		

Solid Sample Data

Table 7.6 - Nordic main tailings data. Solid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

SAMPLE	K	Mg	Pb	Silica	Silica Fused	Soluble	Total	Th	U
DESCRIPTION				Acid extract	w/ NaHCO3	Sulphate	Sulphate	(ppm)	(ppm)
DEPTH (m)	(mg/g)	(mg/g)	(mg/100g)	(mg/g)	(mg/g)	(mg/g)	(mg/g)		
T - 1 -10.77	17.25	2.80	14.00			5.86	51.35		
T - 1 -10.80	3.12	3.39	1.10			2.27	7.72		
T - 1 -11.18	14.98	2.99	1.30			0.52	0.56		
T - 1 -12.57								425	77
T - 2 - 1.00	27.64	0.90	37.60	0.51	31.50	40.50	42.73	21	16
T - 2 - 1.40								26	27
T - 2 - 2.00	24.88	0.94	27.30			31.42	66.42	470	58
T - 2 - 3.00								110	50
T - 2 - 4.00								211	42
T - 2 - 4.00								196	40
T - 2 - 4.00								216	41
T - 2 - 4.60	24.20	0.47	27.25			22.35	62.12	89	71
T - 2 - 5.00								269	55
T - 2 - 5.79								193	74
T - 2 - 6.02								287	63
T - 2 - 6.93								261	69
T - 2 - 7.24								347	74
T - 2 - 8.00	24.23	1.65	37.25			54.11	73.96	480	78
T - 2 - 8.31								213	22
T - 2 - 8.76								407	149
T - 2 - 8.76	32.63	2.13	40.20			45.03	42.73	421	153
T - 2 - 8.76								408	150
T - 2 - 9.07								629	100
T - 2 - 9.07								598	95
T - 2 - 9.07		2.84	53.40	3.55	29.00	85.87	105.19	623	118
T - 2 - 9.61	36.53	1.23	47.15			76.79	107.35	587	141
T - 2 - 9.77	37.40	2.14	45.00			10.29	78.11	756	73
T - 2 - 9.79	25.90	1.89	44.40			10.06	94.42		
T - 2 - 9.87	40.72	4.55	47.60			76.79	93.09		
T - 2 - 9.94	26.99	1.20	42.85			7.23	70.73		
T - 2 -10.15	1.34	3.28	15.40			2.27	6.64		
T - 2 -10.25	15.79	2.56	0.75	0.51	14.00	1.93	0.44		
T - 2 -10.33	23.10	6.47	1.20			1.76	0.00		
T - 3 - 1.00								34	15
T - 3 - 1.30								118	36
T - 3 - 2.00								80	72
T - 3 - 2.60								248	77
T - 3 - 3.00								150	66
T - 3 - 3.00								146	57
T - 3 - 3.00								139	44
T - 3 - 4.00	27.50	1.49	45.00			55.24	59.96	394	70
T - 3 - 4.30								342	55
T - 3 - 4.30								356	51
T - 3 - 4.30								354	52
T - 3 - 5.00	22.78	0.59	27.40			19.51	62.12	163	64
T - 3 - 6.93								814	325
T - 3 - 6.93	38.75	2.84	8.00			8.81	71.41	870	296
T - 3 - 6.93								818	305

Table 7.7 - Nordic main tailings data. Solid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

SAMPLE	K	Mg	Pb	Silica	Silica Fused	Soluble	Total	Th	U
DESCRIPTION				Acid extract	w/ NaHCO3	Sulphate	Sulphate	(ppm)	(ppm)
DEPTH (m)	(mg/g)	(mg/g)	(mg/100g)	(mg/g)	(mg/g)	(mg/g)	(mg/g)		
T - 3 - 7.25								185	97
T - 3 - 7.25									93
T - 3 - 7.25								426	85
T - 3 - 7.50								400	61
T - 3 - 7.54		3.27	35.95	3.55	26.50	60.91	94.42	394	59
T - 3 - 7.75								218	37
T - 3 - 7.85								728	51
T - 3 - 7.85								707	62
T - 3 - 7.85	30.78	3.62	43.50			91.54	107.35	695	58
T - 3 - 7.96								20	37
T - 3 - 7.97								716	53
T - 3 - 8.01								12	9
T - 3 - 8.06		6.90	1.55	7.60	31.00	0.28	1.65	23	6
T - 3 - 8.29									
T - 3 - 8.53	36.79	0.71	30.15			2.38	57.07		
T - 7 - 1.00								20	12
T - 7 - 1.30								28	11
T - 7 - 2.00								27	42
T - 7 - 3.00								370	49
T - 7 - 3.00								270	62
T - 7 - 4.00								183	48
T - 7 - 5.00								181	71
T - 7 - 5.00								186	72
T - 7 - 5.00								173	76
T - 7 - 6.00								183	68
T - 7 - 6.40	29.60	0.90	32.20	1.53	18.50	31.42	61.85		
T - 7 - 6.90	24.44	1.18	53.00			46.17	49.19		
T - 7 - 6.90	27.18	1.34	56.25			47.30	56.73		
T - 7 - 7.00								340	261
T - 7 - 7.20	33.66	2.15	49.10			58.64	76.20		
T - 7 - 7.70	25.07	1.05	25.50	2.03	24.00	3.82	77.19		
T - 7 - 7.81	29.44	1.01	15.40			9.07	54.83		
T - 7 - 7.86	34.89	1.67	25.50	2.03	18.50	40.50	87.68		
T - 7 - 7.91	26.69	1.85	37.15			65.45	93.35		
T - 7 - 7.96								201	30
T - 7 - 8.38	15.49	2.84	4.00			3.40	14.73		
T - 7 - 8.99	14.09	8.33	1.88			2.09	2.02		
T - 8 - 1.00								16	13
T - 8 - 2.00								15	13
T - 8 - 3.00								27	35
T - 8 - 3.00								26	31
T - 8 - 4.00								24	37
T - 8 - 5.00								184	42
T - 8 - 6.00								138	46
T - 8 - 6.00								133	48
T - 8 - 6.00								135	48
T - 8 - 7.00								215	74
T - 8 - 7.60		0.66	36.65			37.09	68.54		

Solid Sample Data

Table 7.8 - Nordic main tailings data. Solid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/gram.

SAMPLE	K	Mg	Pb	Silica	Silica Fused	Soluble	Total	Th	U
DESCRIPTION				Acid extract	w/ NaHCO3	Sulphate	Sulphate	(ppm)	(ppm)
DEPTH (m)	(mg/g)	(mg/g)	(mg/100g)	(mg/g)	(mg/g)	(mg/g)	(mg/g)		
T - 8 - 8.40	21.34	0.20	21.40	0.51	23.00	12.14	64.27		
T - 8 - 8.40	19.30	0.50	21.00	0.51	14.00	14.97	76.12		
T - 8 - 8.76		0.90	30.35	2.03	20.50	21.78	70.46		
T - 8 - 9.07								516	75
T - 8 - 9.37		2.18	4.80	2.03	21.00	23.48	9.24		
T - 8 - 9.98								532	272
T - 8 - 10.29								398	79
T - 8 - 10.29								519	83
T - 8 - 10.29								505	86
T - 8 - 11.05								492	71
T - 8 - 11.20								367	74
T - 8 - 11.44								524	78
T - 8 - 11.49								232	51
T - 8 - 11.60								65	35
T - 8 - 11.65									
T - 8 - 11.65	18.57	1.14	18.00			20.08	59.96	147	33
T - 8 - 12.24	21.02	1.47	14.90			12.14	42.19		

Table 7.9 - Nordic main tailings data. Liquid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/litre.

||Water Sample Study

SAMPLE DESCRIPTION	NORTHING	ELEV.TOP	TOTAL	WATER	PRESSURE	HYDRIC	TEMP	Eh	Ec	pH	Al
	(M)	PIEZO.	DEPTH	LEVEL	POINT	HEAD					
	(x10)	(M)	(M)	(M)	(M)	(M)	'C	(mV)	(uS)		(mg/L)
T1-P1 10-09-84	544.80	351.86	12.99	6.10	339.02	345.76		230	3400	6.11	0.30
T1-P2 10-09-84	544.80	351.86	8.36	4.45	343.65	347.41		200	5200	5.96	0.50
T1-P3 10-09-84	544.80	351.86	8.42	4.45	343.59	347.41		170	6800	5.90	0.00
T1-P4 10-09-84	544.80	351.86	6.95	4.34	345.06	347.52		200	3800	6.02	0.00
T1-P5 10-09-84	544.80	351.86	5.43	4.23	346.58	347.63		270	3500	5.74	0.00
T1-P6 10-09-84	544.80	351.86	3.86	DRY	348.15	DRY	DRY	DRY	DRY	DRY	DRY
T2-P1 06-09-84	518.74	351.71	12.65	7.63	339.21	344.08	12.5	270	4750	5.83	4.95
T2-P2 06-09-84	518.74	351.65	10.94	7.09	340.86	344.56	12.3	100	6500	6.23	0.00
T2-P3 06-09-84	518.74	352.01	9.82	6.38	342.34	345.63					0.20
T2-P4 06-09-84	518.74	352.01	8.30	6.20	343.86	345.81	19.0	400	4000	4.90	0.00
T2-P5 06-09-84	518.74	352.04	6.86	6.01	345.33	346.03	22.0	450	3700	4.65	2.60
T2-P6 06-09-84	518.74	352.04	5.82	DRY	346.37	DRY	DRY	DRY	DRY	DRY	DRY
T2-P7 06-09-84	518.74	352.04	5.46	DRY	346.73	DRY	DRY	DRY	DRY	DRY	DRY
T2-P8 06-09-84	518.74	352.01	3.99	DRY	348.17	DRY	DRY	DRY	DRY	DRY	DRY
T2-P9 06-09-84	518.74	352.04	2.50	DRY	349.69	DRY	DRY	DRY	DRY	DRY	DRY
T2-P10 06-09-84	518.74	352.04	10.75	5.75	341.44	346.29		365	6000	5.12	0.00
T2-P11 06-09-84	518.74	352.04	8.70	7.70	343.49	344.34	20.0	440	6000	4.62	0.87
T3-CENTER 07-09-84	509.62	351.37	17.30	7.70	334.22	343.67	16.0	265	5000	5.90	0.50
T3-1 07-09-84	509.62	351.37	16.11	7.73	335.41	343.64	12.0	245	4000	5.97	0.30
T3-2 07-09-84	509.62	351.37	15.10	7.75	336.42	343.62	10.5	270	5000	6.12	0.25
T3-3 07-09-84	509.62	351.37	14.20	7.70	337.32	343.67	10.5	240	4700	6.22	0.60
T3-4 07-09-84	509.62	351.37	13.07	7.70	338.45	343.67	10.5	260	4600	6.15	0.20
T3-5 07-09-84	509.62	351.37	12.00	7.70	339.52	343.67	11.0	265	4800	6.14	0.40
T3-6 07-09-84	509.62	351.37	11.20	7.70	340.32	343.67	11.0	290	4900	5.96	0.31
T3-P3 07-09-84	509.62	351.37	8.57	7.73	342.95	343.64	18.0	410	4750	4.95	1.50
T7-CENTER 07-09-84	496.41	351.95	23.20	8.60	328.90	343.35	11.0	240	5000	6.10	2.84
T7-1 07-09-84	496.41	351.95	19.08	8.55	333.02	343.40	11.0	230	4400	6.30	0.00
T7-3 07-09-84	496.41	351.95	17.25	8.58	334.85	343.37	11.5	190	4600	6.40	0.00
T7-4 07-09-84	496.41	351.95	15.10	8.53	337.00	343.42	11.5	210	4900	6.20	0.00
T7-5 07-09-84	496.41	351.95	13.10	8.55	339.00	343.40	11.5	30	4800	6.70	0.00
T7-6 07-09-84	496.41	351.95	11.08	8.55	341.02	343.40	12.5	240	4850	6.10	0.00
T7-7 07-09-84	496.41	351.95	9.25	8.55	342.85	343.40	13.0	440	3500	5.10	26.49
T7-P1 07-09-84	496.41	351.95	10.16	8.79	341.94	343.16	14.0	310	5000	5.88	0.30
T7-P2 07-09-84	496.41	351.95	9.16	8.77	342.94	343.18	15.0	550	3200	3.23	56.90
T7-P5 07-09-84	496.41	351.95	11.06	9.89	341.04	342.06	16.0	440	10000	4.16	0.40
T7-PVCV SMALL TUBE 07-09-84	496.41	351.95	10.50	9.25	341.60	342.70	19.0	520	9000	4.51	171.23
T8-P1 10/09/84	490.78	353.47	14.54	11.45	339.08	342.02	16.0	240	8000	5.89	0.20
T8-P2 10/09/84	490.78	353.47	13.02	11.45	340.60	342.02	16.5	230	12000	5.73	0.15
T8-P3 10/09/84	490.78	353.47	11.44	11.10	342.18	342.37	16.5	280	4000	5.87	0.00
T8-P4 10/09/84	490.78	353.47	11.27	10.05	342.35	343.42	20.0	290	6000	5.55	0.15
T8-P5 10/09/84	490.78	353.47	17.03	10.10	336.59	343.37	16.5	250	6500	6.00	2.41
T8-P6 10/09/84	490.78	353.47	17.50	9.55	336.12	343.92	15.5	260	7000	5.78	2.73

Table 7.10 - Nordic main tailings data. Liquid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/litre.

SAMPLE DESCRIPTION	Ca (mg/L)	Fe (mg/L)	K (mg/L)	Mg (mg/L)	Mn (mg/L)	D.C.P. Na (mg/L)	Pb (mg/L)	Ra-226 (pCi/L)	Ra-226 (mBq/L)	SiO2 (mg/L)
T1-P1 10-09-84	457.0	295.0	38.25	450.00	20.00	21.920	0.000	1.80	66.60	34.40
T1-P2 10-09-84	437.5	4625.0	73.25	633.60	17.40	6.560	0.000	147.45	5455.65	47.70
T1-P3 10-09-84	385.0	3975.0	92.50	895.00	28.90	6.700	0.000	153.00	5661.00	55.40
T1-P4 10-09-84	415.0	3612.0	36.00	182.50	6.24	8.160	0.000	144.20	5335.40	43.90
T1-P5 10-09-84	435.0	630.0	19.00	75.00	4.82	7.330	0.000	114.95	4253.15	36.30
T1-P6 10-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P1 06-09-84	417.5	175.0	50.25	260.00	38.50	19.300	0.000	4.91	181.67	24.80
T2-P2 06-09-84	417.5	500.0	147.50	1160.00	127.00	25.910	0.000			26.70
T2-P3 06-09-84	437.5	5550.0	110.50	168.96			0.000	168.54	6235.98	49.60
T2-P4 06-09-84	430.5	2100.0	24.25	75.00	1.67	3.469	0.000	231.30	8558.10	38.20
T2-P5 06-09-84	417.5	925.0	22.50	12.80	1.22	4.020	0.000	218.40	8080.80	43.90
T2-P6 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P7 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P8 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P9 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P10 06-09-84	696.0	4750.0		114.00	9.04	13.690	0.000			12.02
T2-P11 06-09-84	666.0	3620.0		193.00	10.20	22.400	0.000			10.51
T3-CENTER 07-09-84	515.0	440.0	82.50	352.50	42.00	25.200	0.000			26.70
T3-1 07-09-84	495.0	175.0	82.50	350.00	42.40	27.510	0.000			26.70
T3-2 07-09-84	495.0	450.0	81.50	383.50	42.90	27.210	0.000	131.76	4875.12	24.80
T3-3 07-09-84	465.0	385.0	68.75	300.00	41.40	25.850	0.000	167.31	6190.47	22.90
T3-4 07-09-84	447.5	305.0	72.50	260.00	39.30	27.500	0.000	73.50	2719.50	22.90
T3-5 07-09-84	495.0	1475.0	85.50	331.00	39.10	60.160	0.000	1.90	70.30	22.90
T3-6 07-09-84	487.5	2430.0		268.00	36.60	25.280	0.000			
T3-P3 07-09-84	457.5	2260.0		175.00	10.20	6.010	0.322			64.90
T7-CENTER 07-09-84	712.0	2380.0		293.00	25.90	40.100	<0.100			
T7-1 07-09-84	679.0	1650.0		311.00	23.60	48.800	0.000	24.37	901.69	
T7-3 07-09-84	664.0	1640.0		315.00	23.50	47.800	<0.100			
T7-4 07-09-84	704.0	1970.0		288.00	25.40	43.290	<0.100			
T7-5 07-09-84	662.0	2020.0		279.00	25.60	39.500	<0.100			
T7-6 07-09-84	682.0	1960.0		273.00	26.70	41.030	<0.100			
T7-7 07-09-84	567.0	1560.0		55.50	8.82	4.700	<0.100			
T7-P1 07-09-84	462.5	2838.0	78.00	495.00	24.50		0.000			34.50
T7-P2 07-09-84	382.5	1975.0	42.00	72.50	6.47	38.200	0.000	61.22	2265.14	99.30
T7-P5 07-09-84	402.5	8040.0	83.00	265.00	9.47	12.780	0.000			95.50
T7-PVCV SMALL TUBE 07-09-84	653.0	9140.0		287.00	30.30	20.550	0.392			31.96
T8-P1 10/09/84	380.0	5200.0	100.25	440.00	52.20		0.000			45.80
T8-P2 10/09/84	437.5	4990.0	117.25	790.00	95.70		0.000	98.09	3629.33	34.40
T8-P3 10/09/84	488.0	2840.0	59.00	200.00	9.29		0.000			57.30
T8-P4 10/09/84	425.0	4300.0	80.25	140.45	5.81		0.000			47.70
T8-P5 10/09/84	666.0	3660.0		231.00	31.40		0.124			11.20
T8-P6 10/09/84	741.0	4660.0		305.00	46.90		0.000			8.17

Table 7.11 - Nordic main tailings data. Liquid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/litre.

SAMPLE DESCRIPTION	Charge Balance										
	Sulphate	Th	U	Al	Ca	Fe	K	Hg	Mn	Na	
	(mg/L)	(mg/L)	(mg/L)	(mmol/L)	(mmol/L)	(mmol/L)	(mmol/L)	(mmol/L)	(mmol/L)	(mmol/L)	
T1-P1 10-09-84	3760.13	<0.100	0.604	0.03	22.8	10.6	0.98	37.02	1.09	0.954	
T1-P2 10-09-84	12260.13	<0.100	0.765	0.06	21.8	165.6	1.87	52.12	0.95	0.285	
T1-P3 10-09-84	10023.29	0.142	0.849	0.00	19.2	142.4	2.37	73.63	1.58	0.292	
T1-P4 10-09-84	8173.42	<0.100	0.678	0.00	20.7	129.4	0.92	15.01	0.34	0.355	
T1-P5 10-09-84	3760.13	<0.100	0.624	0.00	21.7	22.6	0.49	6.17	0.26	0.319	
T1-P6 10-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	
T2-P1 06-09-84	30897.00	<0.100	0.710	0.55	20.8	6.3	1.29	21.39	2.10	0.840	
T2-P2 06-09-84	6220.65	<0.100	1.140	0.00	20.8	17.9	3.77	95.43	6.94	1.128	
T2-P3 06-09-84	10023.29	0.100	0.100	0.02	21.8	198.8	3.03	13.90	0.00	0.000	
T2-P4 06-09-84	5325.92	<0.100	0.642	0.00	21.5	75.2	0.62	6.17	0.09	0.151	
T2-P5 06-09-84	3312.75	2.211	6.540	0.29	20.8	33.1	0.58	1.05	0.07	0.175	
T2-P6 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	
T2-P7 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	
T2-P8 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	
T2-P9 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	
T2-P10 06-09-84	7272.00	0.000	0.839	0.00	34.7	170.1	0.00	9.38	0.49	0.596	
T2-P11 06-09-84	6392.00	<0.100	0.000	0.10	33.2	129.6	0.00	15.88	0.56	0.975	
T3-CENTER 07-09-84	3760.13	<0.100	0.155	0.06	25.7	15.8	2.11	29.00	2.29	1.097	
T3-1 07-09-84	3983.80	0.000	<0.100	0.03	24.7	6.3	2.11	28.79	2.32	1.197	
T3-2 07-09-84	3401.48	0.000	0.230	0.03	24.7	16.1	2.08	31.55	2.34	1.184	
T3-3 07-09-84	3536.45	0.000	0.000	0.07	23.2	13.8	1.76	24.68	2.26	1.125	
T3-4 07-09-84	3312.76	<0.100	0.279	0.02	22.3	10.9	1.85	21.39	2.15	1.197	
T3-5 07-09-84	6668.02	0.000	0.244	0.04	24.7	52.8	2.19	27.23	2.14	2.618	
T3-6 07-09-84	5306.00	<0.100	0.221	0.03	24.3	87.0	0.00	22.05	2.00	1.100	
T3-P3 07-09-84	7786.44	0.000	0.164	0.17	22.8	80.9	0.00	14.40	0.56	0.262	
T7-CENTER 07-09-84	5655.00	0.131	0.410	0.32	35.5	85.2	0.00	24.10	1.41	1.745	
T7-1 07-09-84	4832.00	0.000	0.406	0.00	33.9	59.1	0.00	25.58	1.29	2.124	
T7-3 07-09-84	4320.00	<0.100	0.446	0.00	33.1	58.7	0.00	25.91	1.28	2.080	
T7-4 07-09-84	5270.00	0.000	0.471	0.00	35.1	70.5	0.00	23.69	1.39	1.884	
T7-5 07-09-84	5396.00	0.111	0.118	0.00	33.0	72.3	0.00	22.95	1.40	1.719	
T7-6 07-09-84	5458.00	<0.100	0.128	0.00	34.0	70.2	0.00	22.46	1.46	1.785	
T7-7 07-09-84	3846.00	0.521	2.440	2.95	28.3	55.9	0.00	4.57	0.48	0.205	
T7-P1 07-09-84	6444.35	<0.100	0.100	0.03	23.1	101.6	2.00	40.72	1.34	0.000	
T7-P2 07-09-84	5996.97	3.800	4.080	6.33	19.1	70.7	1.07	5.96	0.35	1.662	
T7-P5 07-09-84	17852.23	7.340	4.750	0.04	20.1	287.9	2.12	21.80	0.52	0.556	
T7-PVCV SMALL TUBE 07-09-84		2.710	1.480	19.04	32.6	327.3	0.00	23.61	1.65	0.894	
T8-P1 10/09/84	14496.97	0.262	0.000	0.02	19.0	186.2	2.56	36.20	2.85	0.000	
T8-P2 10/09/84	12260.12	0.468	0.000	0.02	21.8	178.7	3.00	64.99	5.23	0.000	
T8-P3 10/09/84	7786.44	0.144	0.100	0.00	24.4	101.7	1.51	16.45	0.51	0.000	
T8-P4 10/09/84	11141.71	0.184	0.696	0.02	21.2	154.0	2.05	11.55	0.32	0.000	
T8-P5 10/09/84	7765.00	0.322	0.434	0.27	33.2	131.1	0.00	19.00	1.71	0.000	
T8-P6 10/09/84	9370.00	0.299	0.259	0.30	37.0	166.9	0.00	25.09	2.56	0.000	

Table 7.12 - Nordic main tailings data. Liquid phase sample results of chemical and radioisotope analyses from locations T-1, T-2, T-3, T-7 and T-8 at various depths in concentration/litre.

SAMPLE DESCRIPTION	Pb (mmol/L)	Ra-226 (mmol/L)	SiO2 (mmol/L)	Sulphate (Anions) (mmol/L)	Th (mmol/L)	U (mmol/L)	Total Cations (mmol/L)	Ion Balance (mmol/L)	Ion Balance Error
T1-P1 10-09-84	0.000000	1.61E-11	2.290	78.285	0.00086	0.0101	75.75	-2.54	-3.30%
T1-P2 10-09-84	0.000000	1.32E-09	3.176	255.255	0.00086	0.0129	245.94	-9.32	-3.72%
T1-P3 10-09-84	0.000000	1.37E-09	3.688	208.684	0.00245	0.0143	243.13	34.45	15.25%
T1-P4 10-09-84	0.000000	1.29E-09	2.923	170.170	0.00086	0.0114	169.63	-0.54	-0.32%
T1-P5 10-09-84	0.000000	1.03E-09	2.417	78.285	0.00086	0.0105	53.93	-24.35	-36.83%
T1-P6 10-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P1 06-09-84	0.000000	4.39E-11	1.651	643.272	0.00086	0.0119	54.93	-588.34	-168.53%
T2-P2 06-09-84	0.000000	0.00E+00	1.777	129.513	0.00086	0.0192	147.80	18.29	13.19%
T2-P3 06-09-84	0.000000	1.51E-09	3.302	208.684	0.00172	0.0017	240.85	32.16	14.31%
T2-P4 06-09-84	0.000000	2.07E-09	2.543	110.885	0.00086	0.0108	106.27	-4.61	-4.25%
T2-P5 06-09-84	0.000000	1.95E-09	2.923	68.971	0.03811	0.1099	59.19	-9.78	-15.26%
T2-P6 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P7 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P8 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P9 06-09-84	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY	DRY
T2-P10 06-09-84	0.000000	0.00E+00	0.800	151.402	0.00000	0.0141	216.12	64.72	35.22%
T2-P11 06-09-84	0.000000	0.00E+00	0.700	133.081	0.00086	0.0000	181.08	48.00	30.56%
T3-CENTER 07-09-84	0.000000	0.00E+00	1.777	78.285	0.00086	0.0026	77.79	-0.49	-0.63%
T3-1 07-09-84	0.000000	0.00E+00	1.777	82.942	0.00000	0.0084	67.20	-15.74	-20.97%
T3-2 07-09-84	0.000000	1.18E-09	1.651	70.818	0.00000	0.0039	79.66	8.84	11.75%
T3-3 07-09-84	0.000000	1.50E-09	1.525	73.628	0.00000	0.0000	68.41	-5.22	-7.35%
T3-4 07-09-84	0.000000	6.58E-10	1.525	68.971	0.00086	0.0047	61.39	-7.58	-11.63%
T3-5 07-09-84	0.000000	1.70E-11	1.525	138.827	0.00000	0.0041	113.27	-25.56	-20.28%
T3-6 07-09-84	0.000000	0.00E+00	0.000	110.470	0.00086	0.0037	136.53	26.06	21.10%
T3-P3 07-09-84	0.003108	0.00E+00	4.321	162.113	0.00000	0.0028	123.47	-38.64	-27.06%
T7-CENTER 07-09-84	0.000483	0.00E+00	0.000	117.736	0.00226	0.0069	148.35	30.61	23.01%
T7-1 07-09-84	0.000000	2.18E-10	0.000	100.602	0.00000	0.0068	121.98	21.37	19.21%
T7-3 07-09-84	0.000483	0.00E+00	0.000	89.942	0.00086	0.0075	121.15	31.21	29.57%
T7-4 07-09-84	0.000483	0.00E+00	0.000	109.721	0.00000	0.0079	132.65	22.93	18.92%
T7-5 07-09-84	0.000483	0.00E+00	0.000	112.344	0.00191	0.0020	131.45	19.10	15.67%
T7-6 07-09-84	0.000483	0.00E+00	0.000	113.635	0.00086	0.0022	129.93	16.29	13.38%
T7-7 07-09-84	0.000483	0.00E+00	0.000	80.073	0.00898	0.0410	92.41	12.33	14.30%
T7-P1 07-09-84	0.000000	0.00E+00	2.297	134.171	0.00086	0.0017	171.10	36.93	24.19%
T7-P2 07-09-84	0.000000	5.48E-10	6.611	124.856	0.06551	0.0686	111.94	-12.91	-10.91%
T7-P5 07-09-84	0.000000	0.00E+00	6.358	371.681	0.12653	0.0798	339.62	-32.06	-9.02%
T7-PVCV SMALL TUBE 07-09-84	0.003784	0.00E+00	2.128		0.04672	0.0249	407.31		
T8-P1 10/09/84	0.000000	0.00E+00	3.049	301.825	0.00452	0.0000	249.87	-51.95	-18.83%
T8-P2 10/09/84	0.000000	8.78E-10	2.290	255.254	0.00807	0.0000	276.06	20.81	7.83%
T8-P3 10/09/84	0.000000	0.00E+00	3.815	162.113	0.00248	0.0017	148.35	-13.77	-8.87%
T8-P4 10/09/84	0.000000	0.00E+00	3.176	231.969	0.00317	0.0117	192.33	-39.64	-18.68%
T8-P5 10/09/84	0.001197	0.00E+00	0.746	161.666	0.00555	0.0073	186.05	24.38	14.03%
T8-P6 10/09/84	0.000000	0.00E+00	0.544	195.082	0.00515	0.0044	232.37	37.29	17.45%

Table 7.13 - Nordic main tailings data. Solid phase samples. Summary of results from radioisotope analyses and activity ratios from locations T-1, T-2, T-3, T-7 and T-8 at various depths.

Solid Sample Study				ACTIVITY RATIOS								
SAMPLE	Northing	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	RATIO	RATIO	RATIO	RATIO
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	Pb-210/	Ra-226/	Th-228/	Ra-224/
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	Ra-226	Th-230	Th-232	Th-228
T - 1 - 1.00	544.80	350.79	13764.0		17760.0	3219.0	1258.0	185.0	0.78	14.12	17.40	
T - 1 - 1.50	544.80	350.79	7955.0		9768.0	703.0	5106.0	851.0	0.81	1.91	0.83	
T - 1 - 2.00	544.80	350.79	15873.0		21201.0	1813.0	22126.0	2738.0	0.75	0.96	0.66	
T - 1 - 2.00	544.80	350.79	15947.0		17131.0	2220.0	23199.0	3034.0	0.93	0.74	0.73	
T - 1 - 2.00	544.80	350.79	17760.0		21423.0	2775.0	23680.0	3034.0	0.83	0.90	0.91	
T - 1 - 2.50	544.80	350.79	13653.0		14171.0	1073.0	8991.0	1332.0	0.96	1.58	0.81	
T - 1 - 3.00	544.80	350.79	11803.0		14726.0	888.0	13209.0	1332.0	0.80	1.11	0.67	
T - 1 - 4.00	544.80	350.79	15799.0		14245.0	407.0	2035.0	999.0	1.11	7.00	0.41	
T - 1 - 4.00	544.80	350.79	18537.0		18056.0	2368.0	9805.0	2627.0	1.03	1.84	0.90	
T - 1 - 4.00	544.80	350.79	15799.0		18167.0	1443.0	6327.0	1147.0	0.87	2.87	1.26	
T - 1 - 4.25	544.80	350.79	12950.0		12506.0	888.0	2775.0	962.0	1.04	4.51	0.92	
T - 1 - 4.50	544.80	350.79	5846.0		6105.0	333.0	851.0	148.0	0.96	7.17	2.25	
T - 1 - 4.60	544.80	350.79	10471.0		11840.0	814.0	8325.0	962.0	0.88	1.42	0.85	
T - 1 - 4.75	544.80	350.79	32264.0		21830.0	3330.0	6660.0	2035.0	1.48	3.28	1.64	
T - 1 - 5.00	544.80	350.79	14615.0		15540.0	2183.0	6919.0	1517.0	0.94	2.25	1.44	
T - 1 - 5.25	544.80	350.79	31968.0		25752.0	1036.0	3182.0	999.0	1.24	8.09	1.04	
T - 1 - 5.50	544.80	350.79	12950.0		13653.0	1036.0	1924.0	888.0	0.95	7.10	1.17	
T - 1 - 5.75	544.80	350.79	13135.0		16132.0	666.0	2701.0	851.0	0.81	5.97	0.78	
T - 1 - 6.00	544.80	350.79	15651.0		15836.0	407.0	3885.0	1221.0	0.99	4.08	0.33	
T - 1 - 6.00	544.80	350.79	16502.0		18500.0	1480.0	5106.0	1332.0	0.89	3.62	1.11	
T - 1 - 6.00	544.80	350.79	18796.0		18537.0	1517.0	6031.0	1295.0	1.01	3.07	1.17	
T - 1 - 6.00	544.80	350.79	12950.0		14208.0	1147.0	11729.0	1332.0	0.91	1.21	0.86	
T - 1 - 6.25	544.80	350.79	10175.0		9398.0	1036.0	3145.0	925.0	1.08	2.99	1.12	
T - 1 - 6.50	544.80	350.79	13542.0		14134.0	592.0	2664.0	592.0	0.96	5.31	1.00	
T - 1 - 6.75	544.80	350.79	8806.0		8954.0	370.0	2146.0	888.0	0.98	4.17	0.42	
T - 1 - 7.00	544.80	350.79	19277.0		14430.0	481.0	3885.0	814.0	1.34	3.71	0.59	
T - 1 - 7.00	544.80	350.79	12247.0		13468.0	740.0	4847.0	592.0	0.91	2.78	1.25	
T - 1 - 7.25	544.80	350.79	5883.0		4810.0	74.0	259.0	74.0	1.22	18.57	1.00	
T - 1 - 7.40	544.80	350.79	17057.0		20609.0	2331.0	9398.0	1036.0	0.83	2.19	2.25	
T - 1 - 7.50	544.80	350.79	8954.0		10989.0	296.0	2516.0	481.0	0.81	4.37	0.62	
T - 1 - 7.60	544.80	350.79	10619.0		12728.0	962.0	6031.0	703.0	0.83	2.11	1.37	
T - 1 - 7.60	544.80	350.79	10619.0		12469.0	2627.0	6697.0	777.0	0.85	1.86	3.38	
T - 1 - 7.60	544.80	350.79	10360.0		11507.0	666.0	8695.0	1332.0	0.90	1.32	0.50	
T - 1 - 7.75	544.80	350.79	16539.0		15096.0	555.0	2590.0	703.0	1.10	5.83	0.79	
T - 1 - 8.00	544.80	350.79	3885.0		6068.0	2109.0	14393.0	1036.0	0.64	0.42	2.04	
T - 1 - 8.00	544.80	350.79	23754.0		19610.0	1813.0	6105.0	2035.0	1.21	3.21	0.89	
T - 1 - 8.25	544.80	350.79	35853.0		24494.0	2627.0	6364.0	1554.0	1.46	3.85	1.69	
T - 1 - 8.50	544.80	350.79	19388.0		22459.0	925.0	7770.0	1036.0	0.86	2.89	0.89	
T - 1 - 8.50	544.80	350.79	24901.0		19092.0	666.0	2960.0	1036.0	1.30	6.45	0.64	
T - 1 - 8.50	544.80	350.79	26307.0		20239.0	3145.0	10360.0	2775.0	1.30	1.95	1.13	
T - 1 - 9.00	544.80	350.79	18759.0		20239.0	1924.0	14948.0	1554.0	0.93	1.35	1.24	
T - 1 - 9.00	544.80	350.79	18685.0		21497.0	1665.0	14652.0	1480.0	0.87	1.47	1.13	
T - 1 - 9.00	544.80	350.79	15725.0		18241.0	2738.0	16428.0	1887.0	0.86	1.11	1.45	
T - 1 - 9.60	544.80	350.79	16761.0	1295.0	13579.0	1476.3	18107.8	1587.3	1.23	0.75	0.93	0.88
T - 1 - 9.60	544.80	350.79	17908.0	1480.0	17782.2	1968.4	22618.1	2234.8	1.01	0.79	0.88	0.75
T - 1 - 10.20	544.80	350.79	20646.0	2849.0	20398.1	2886.0	26551.2	2667.7	1.01	0.77	1.08	0.99
T - 1 - 10.60	544.80	350.79	621.6	74.0	3307.8	59.2	292.3	25.9	0.19	11.32	2.29	1.25
T - 1 - 10.70	544.80	350.79	218.3	74.0	266.4	55.5	310.8	77.7	0.82	0.86	0.71	1.33

Radioisotopes Ratio Solid Sample Data

Table 7.14 - Nordic main tailings data. Solid phase samples. Summary of results from radioisotope analyses and activity ratios from locations T-1, T-2, T-3, T-7 and T-8 at various depths.

Solid Sample Study			ACTIVITY RATIOS										
SAMPLE	Northing	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	RATIO				
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	Pb-210/	Ra-226/	Th-228/	Ra-224/	
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	Ra-226	Th-230	Th-232	Th-228	
T - 1	-10.77	544.80	350.79	12062.0	2072.0	13908.3	1857.4	18396.4	1713.1	0.87	0.76	1.08	1.12
T - 1	-10.80	544.80	350.79	225.7	70.3	336.7	85.1	188.7	51.8	0.67	1.78	1.64	0.83
T - 1	-11.18	544.80	350.79	484.7	122.1	588.3	129.5	762.2	136.9	0.82	0.77	0.95	0.94
T - 1	-12.57	544.80	350.79	18796.0		17168.0	2146.0	25345.0	2442.0	1.09	0.68	0.88	
T - 2	-1.00	518.74	351.58	14689.0		16391.0	370.0	444.0	22.2	0.90	36.92	16.67	
T - 2	-1.40	518.74	351.58	10138.0		13764.0	666.0	1480.0	185.0	0.74	9.30	3.60	
T - 2	-2.00	518.74	351.58	12987.0		17575.0	2886.0	18907.0	3293.0	0.74	0.93	0.88	
T - 2	-3.00	518.74	351.58	8140.0		9805.0	1480.0	11211.0	1961.0	0.83	0.87	0.75	
T - 2	-4.00	518.74	351.58	14504.0		12506.0	925.0	11211.0	1369.0	1.16	1.12	0.68	
T - 2	-4.00	518.74	351.58	18130.0		16909.0	2220.0	10582.0	1221.0	1.07	1.60	1.82	
T - 2	-4.00	518.74	351.58	17834.0		15762.0	1184.0	10841.0	1258.0	1.13	1.45	0.94	
T - 2	-4.60	518.74	351.58	15096.0		15836.0	666.0	4921.0	629.0	0.95	3.22	1.06	
T - 2	-5.00	518.74	351.58	10693.0		13061.0	2294.0	21090.0	2146.0	0.82	0.62	1.07	
T - 2	-5.79	518.74	351.58	22163.0		18796.0	1332.0	11914.0	925.0	1.18	1.58	1.44	
T - 2	-6.02	518.74	351.58	25197.0		23495.0	1628.0	17834.0	1776.0	1.07	1.32	0.92	
T - 2	-6.93	518.74	351.58	21571.0		22829.0	1147.0	16465.0	1739.0	0.94	1.39	0.66	
T - 2	-7.24	518.74	351.58	32745.0		27824.0	2479.0	22977.0	2442.0	1.18	1.21	1.02	
T - 2	-8.00	518.74	351.58	23495.0		21978.0	2812.0	22718.0	2812.0	1.07	0.97	1.00	
T - 2	-8.31	518.74	351.58	14319.0		14726.0	1184.0	10804.0	1591.0	0.97	1.36	0.74	
T - 2	-8.76	518.74	351.58	22089.0		22459.0	2146.0	23569.0	2590.0	0.98	0.95	0.83	
T - 2	-8.76	518.74	351.58	21867.0		25937.0	1887.0	21608.0	2849.0	0.84	1.20	0.66	
T - 2	-8.76	518.74	351.58	23902.0		22866.0	1961.0	22903.0	2516.0	1.05	1.00	0.78	
T - 2	-9.07	518.74	351.58	33522.0		26566.0	3626.0	29267.0	3256.0	1.26	0.91	1.11	
T - 2	-9.07	518.74	351.58	31968.0		28897.0	3108.0	29822.0	3996.0	1.11	0.97	0.78	
T - 2	-9.07	518.74	351.58	30414.0		26862.0	2997.0	28934.0	3737.0	1.13	0.93	0.80	
T - 2	-9.61	518.74	351.58	27491.0		31931.0	3552.0	27306.0	3034.0	0.86	1.17	1.17	
T - 2	-9.77	518.74	351.58	37259.0		42180.0	3996.0	42624.0	4810.0	0.88	0.99	0.83	
T - 2	-9.79	518.74	351.58	27972.0	2664.0	35786.4	2934.1	48421.9	2941.5	0.78	0.74	1.00	0.91
T - 2	-9.87	518.74	351.58	33633.0	5254.0	35989.9	4632.4	51788.9	5820.1	0.93	0.69	0.80	1.13
T - 2	-9.94	518.74	351.58	27750.0	3034.0	29326.2	2856.4	35353.5	5220.7	0.95	0.83	0.55	1.06
T - 2	-10.15	518.74	351.58	321.9	7.4	492.1	14.8	236.8	37.0	0.65	2.08	0.40	0.50
T - 2	-10.25	518.74	351.58	181.3	92.5	858.4	85.1	159.1	40.7	0.21	5.40	2.09	1.09
T - 2	-10.33	518.74	351.58	233.1	44.4	318.2	96.2	103.6	18.5	0.73	3.07	5.20	0.46
T - 3	-1.00	509.62	350.83	15540.0		17538.0	888.0	925.0	114.7	0.89	18.96	7.74	
T - 3	-1.30	509.62	350.83	12876.0		13061.0	1221.0	6512.0	777.0	0.99	2.01	1.57	
T - 3	-2.00	509.62	350.83	9583.0		9842.0	666.0	2516.0	222.0	0.97	3.91	3.00	
T - 3	-2.60	509.62	350.83	24383.0		19055.0	1332.0	14800.0	1332.0	1.28	1.29	1.00	
T - 3	-3.00	509.62	350.83	11803.0		10582.0	703.0	6549.0	703.0	1.12	1.62	1.00	
T - 3	-3.00	509.62	350.83	11655.0		11618.0	555.0	7400.0	814.0	1.00	1.57	0.68	
T - 3	-3.00	509.62	350.83	11433.0		11211.0	592.0	5846.0	666.0	1.02	1.92	0.89	
T - 3	-4.00	509.62	350.83	5476.0		7511.0	407.0	1628.0	444.0	0.73	4.61	0.92	
T - 3	-4.30	509.62	350.83	26011.0		27084.0	2183.0	23051.0	2553.0	0.96	1.17	0.86	
T - 3	-4.30	509.62	350.83	27232.0		24457.0	1998.0	21904.0	2368.0	1.11	1.12	0.84	
T - 3	-4.30	509.62	350.83	27528.0		23828.0	2072.0	21460.0	2257.0	1.16	1.11	0.92	
T - 3	-5.00	509.62	350.83	11359.0		14874.0	1443.0	9250.0	1295.0	0.76	1.61	1.11	
T - 3	-6.93	509.62	350.83	48285.0		49136.0	6549.0	41773.0	6808.0	0.98	1.18	0.96	
T - 3	-6.93	509.62	350.83	44474.0		53465.0	4884.0	40959.0	6660.0	0.83	1.31	0.73	
T - 3	-6.93	509.62	350.83	48248.0		44067.0	4514.0	39849.0	5661.0	1.09	1.11	0.80	

Radioisotopes Ratio Solid Sample Data

Table 7.15 - Nordic main tailings data. Solid phase samples. Summary of results from radioisotope analyses and activity ratios from locations T-1, T-2, T-3, T-7 and T-8 at various depths.

Solid Sample Study				ACTIVITY RATIOS									
[SAMPLE	[Northing	[Elevation	[Pb-210	[Ra-224	[Ra-226	[Th-228	[Th-230	[Th-232	[RATIO	[RATIO	[RATIO	[RATIO	
[DESCRIPTION	[(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	Pb-210/	Ra-226/	Th-228/	Ra-224/	
[DEPTH (m)	[(x10)		[+/-10%		[+/-10%	[+/-20%	[+/-20%	[+/-20%	Ra-226	Th-230	Th-232	Th-228	
T - 3 - 7.25	509.62	350.83	20868.0		19166.0	777.0	4551.0	1036.0	1.09	4.21	0.75		
T - 3 - 7.25	509.62	350.83	22089.0		17131.0	1924.0	4662.0	851.0	1.29	3.67	2.26		
T - 3 - 7.25	509.62	350.83	19869.0		22792.0	1887.0	5550.0	1295.0	0.87	4.11	1.46		
T - 3 - 7.50	509.62	350.83	22089.0		17427.0	1850.0	4810.0	1332.0	1.27	3.62	1.39		
T - 3 - 7.54	509.62	350.83	22422.0		24679.0	2294.0	24198.0	2664.0	0.91	1.02	0.86		
T - 3 - 7.75	509.62	350.83	12839.0		12617.0	851.0	2257.0	592.0	1.02	5.59	1.44		
T - 3 - 7.85	509.62	350.83	28194.0		32116.0	2849.0	41329.0	3848.0	0.88	0.78	0.74		
T - 3 - 7.85	509.62	350.83	29489.0		34891.0	4810.0	42328.0	4995.0	0.85	0.82	0.96		
T - 3 - 7.85	509.62	350.83	28194.0		37481.0	3071.0	39257.0	3515.0	0.75	0.95	0.87		
T - 3 - 7.96	509.62	350.83	2479.0		3071.0	296.0	1110.0	185.0	0.81	2.77	1.60		
T - 3 - 7.97	509.62	350.83	29970.0		27972.0	1184.0	3330.0	666.0	1.07	8.40	1.78		
T - 3 - 8.01	509.62	350.83	1406.0		1813.0	148.0	74.0	18.5	0.78	24.50	8.00		
T - 3 - 8.06	509.62	350.83	518.0		518.0	74.0	222.0	74.0	1.00	2.33	1.00		
T - 3 - 8.29	509.62	350.83	418.1	74.0	710.4	88.8	1098.9	107.3	0.59	0.65	0.83	0.83	
T - 3 - 8.53	509.62	350.83	1554.0	218.3	1872.2	203.5	1491.1	244.2	0.83	1.26	0.83	1.07	
T - 7 - 1.00	496.41	351.95	10027.0		12506.0	481.0	1184.0	148.0	0.80	10.56	3.25		
T - 7 - 1.30	496.41	351.95	13949.0		17094.0	888.0	2183.0	296.0	0.82	7.83	3.00		
T - 7 - 2.00	496.41	351.95	8103.0		12136.0	629.0	740.0	66.6	0.67	16.40	9.44		
T - 7 - 3.00	496.41	351.95	16095.0		19610.0	888.0	18870.0	1628.0	0.82	1.04	0.55		
T - 7 - 3.00	496.41	351.95	10841.0		11803.0	1147.0	17020.0	2516.0	0.92	0.69	0.46		
T - 7 - 4.00	496.41	351.95	14615.0		14615.0	1850.0	8436.0	1110.0	1.00	1.73	1.67		
T - 7 - 5.00	496.41	351.95	12432.0		17242.0	1480.0	9879.0	1332.0	0.72	1.75	1.11		
T - 7 - 5.00	496.41	351.95	12506.0		15096.0	1406.0	8658.0	999.0	0.83	1.74	1.41		
T - 7 - 5.00	496.41	351.95	12247.0		19425.0	2183.0	9805.0	1517.0	0.63	1.98	1.44		
T - 7 - 6.00	496.41	351.95	11914.0		15244.0	1406.0	8547.0	1184.0	0.78	1.78	1.19		
T - 7 - 6.40	496.41	351.95	14245.0	1480.0	18437.1	1420.8	14448.5	1139.6	0.77	1.28	1.25	1.04	
T - 7 - 6.90	496.41	351.95	30081.0	2960.0	29511.2	3204.2	28915.5	3193.1	1.02	1.02	1.00	0.92	
T - 7 - 6.90	496.41	351.95	27972.0	2775.0	28342.0	2516.0	26880.5	3178.3	0.99	1.05	0.79	1.10	
T - 7 - 7.00	496.41	351.95	26344.0		31043.0	2072.0	17760.0	2886.0	0.85	1.75	0.72	0.00	
T - 7 - 7.20	496.41	351.95	28194.0	2960.0	27539.1	3204.2	32844.9	3470.6	1.02	0.84	0.92	0.92	
T - 7 - 7.70	496.41	351.95	14578.0	1517.0	14037.8	1716.8	15273.6	2604.8	1.04	0.92	0.66	0.88	
T - 7 - 7.81	496.41	351.95	7770.0	444.0	5846.0	532.8	4181.0	377.4	1.33	1.40	1.41	0.83	
T - 7 - 7.86	496.41	351.95	15355.0	2183.0	15318.0	1920.3	16265.2	2379.1	1.00	0.94	0.81	1.14	
T - 7 - 7.91	496.41	351.95	24198.0	2886.0	25382.0	2941.5	28893.3	4699.0	0.95	0.88	0.63	0.98	
T - 7 - 7.96	496.41	351.95	16021.0		16354.0	1332.0	10952.0	1369.0	0.98	1.49	0.97		
T - 7 - 8.38	496.41	351.95	2812.0	333.0	2797.2	355.2	1938.8	270.1	1.01	1.44	1.32	0.94	
T - 7 - 8.99	496.41	351.95	814.0	111.0	888.0	122.1	362.6	96.2	0.92	2.45	1.27	0.91	
T - 8 - 1.00	490.78	353.47	7474.0		8954.0	703.0	407.0	44.4	0.83	22.00	15.83		
T - 8 - 2.00	490.78	353.47	5920.0		6438.0	259.0	296.0	48.1	0.92	21.75	5.38		
T - 8 - 3.00	490.78	353.47	5402.0		7178.0		2220.0	2405.0	0.75	3.23	0.00		
T - 8 - 3.00	490.78	353.47	6068.0		7918.0	333.0	1739.0	148.0	0.77	4.55	2.25		
T - 8 - 4.00	490.78	353.47	7178.0		10989.0	296.0	481.0	77.7	0.65	22.85	3.81		
T - 8 - 5.00	490.78	353.47	6697.0		8991.0	666.0	10656.0	851.0	0.74	0.84	0.78		
T - 8 - 6.00	490.78	353.47	7733.0		8695.0	518.0	8325.0	703.0	0.89	1.04	0.74		
T - 8 - 6.00	490.78	353.47	7622.0		9324.0	629.0	8066.0	703.0	0.82	1.16	0.89		
T - 8 - 6.00	490.78	353.47	8399.0		8880.0	592.0	8140.0	888.0	0.95	1.09	0.67		
T - 8 - 7.00	490.78	353.47	19499.0		21497.0	1665.0	14837.0	1739.0	0.91	1.45	0.96		
T - 8 - 7.60	490.78	353.47	18056.0	1591.0	14474.4	1539.2	17131.0	1502.2	1.25	0.84	1.02	1.03	

Radioisotopes Ratio Solid Sample Data

Table 7.16 - Nordic main tailings data. Solid phase samples. Summary of results from radioisotope analyses and activity ratios from locations T-1, T-2, T-3, T-7 and T-8 at various depths.

Solid Sample Study			ACTIVITY RATIOS									
SAMPLE	Northng	Elevation	Pb-210	Ra-224	Ra-226	Th-228	Th-230	Th-232	RATIO	RATIO	RATIO	RATIO
DESCRIPTION	(m)	(m)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	(mBq/g)	Pb-210/	Ra-226/	Th-228/	Ra-224/
DEPTH (m)	(x10)		+/-10%		+/-10%	+/-20%	+/-20%	+/-20%	Ra-226	Th-230	Th-232	Th-228
T - 8 - 8.40	490.78	353.47	8991.0	1961.0	7437.0	1879.6	8376.8	703.0	1.21	0.89	2.67	1.04
T - 8 - 8.40	490.78	353.47	7511.0	851.0	8658.0	1172.9	7440.7	1354.2	0.87	1.16	0.87	0.73
T - 8 - 8.76	490.78	353.47	14652.0	1628.0	14999.8	1572.5	13149.8	1609.5	0.98	1.14	0.98	1.04
T - 8 - 9.07	490.78	353.47	26640.0		28823.0	5254.0	19055.0	2664.0	0.92	1.51	1.97	
T - 8 - 9.37	490.78	353.47	16687.0	1406.0	15351.3	973.1	10855.8	862.1	1.09	1.41	1.13	1.44
T - 8 - 9.98	490.78	353.47	41736.0		42809.0	5624.0	40145.0	3811.0	0.97	1.07	1.48	
T - 8 -10.29	490.78	353.47	24790.0		26973.0				0.92			
T - 8 -10.29	490.78	353.47	30081.0		26196.0	3293.0	31302.0	3700.0	1.15	0.84	0.89	
T - 8 -10.29	490.78	353.47	29045.0		31524.0	1924.0	21201.0	2109.0	0.92	1.49	0.91	
T - 8 -11.05	490.78	353.47	23347.0		24013.0	1665.0	25530.0	2664.0	0.97	0.94	0.63	
T - 8 -11.20	490.78	353.47	22385.0		22200.0	1776.0	21127.0	2257.0	1.01	1.05	0.79	
T - 8 -11.44	490.78	353.47	33781.0		32893.0	2553.0	28379.0	2627.0	1.03	1.16	0.97	
T - 8 -11.49	490.78	353.47	12543.0		12432.0	518.0	15170.0	962.0	1.01	0.82	0.54	
T - 8 -11.60	490.78	353.47	5328.0		6956.0	1184.0	2183.0	222.0	0.77	3.19	5.33	
T - 8 -11.65	490.78	353.47	9324.0	1110.0	9224.1	865.8	10145.4	965.7	1.01	0.91	0.90	1.28
T - 8 -11.65	490.78	353.47	9805.0		10434.0	925.0	9287.0	888.0	0.94	1.12	1.04	
T - 8 -12.24	490.78	353.47	6845.0		7422.2	740.0	7248.3	825.1	0.92	1.02	0.90	

Table 7.17 - Nordic main tailings data. Additional data from location T-5. Solid and liquid phase samples for comparison of radioisotope activities at various depths.

Depth m	Piezometer No.	Solid Sample					Liquid Sample						Remarks	
		²²⁸ Th pCi/g	²³² Th pCi/g	²³⁰ Th pCi/g	²²⁶ Ra pCi/g	²¹⁰ Pb pCi/g	²²⁸ Th pCi/L	²³² Th pCi/L	²³⁰ Th pCi/L	²²⁶ Ra pCi/L	²¹⁰ Pb pCi/L	²²³ Ra pCi/L		
4.70		11	11	32	105	125								
4.80		10	7	42	110	107								
5.00		33	11	64	164	206								
5.40		8	7	28	162	115								
5.58	P5	24	21	66	303	287	160±33	0.9±0.6	40±1.4	86±6	1951±87	439±18		
5.75		26	40	152	296	353								
6.00		9	8	52	125	159								
6.25		24	13	52	111	163								
6.50		14	15	44	182	176								
6.60	P7	20.8±1.9	49.7±1.8	242.8±13.4	250±11.2	304±14	104±11	5.8±1.2	8.9±1.5	29±3	2114±94	460±18		
6.75	P3	21	15	58	331	314	128±14	3.9±0.9	14.1±1.7	81±4	2421±103	694±24		
7.00		8	17	55	281	332								
7.25	P1	25	6	43	176	223	4±2	<0.5	<0.5	137±5	31±10	444±14	Water table	
7.50		63	65	238	556	653								
7.75		11	13	36	130	164								
8.00	P2	14	10	31	292	312	<0.5	<0.5	1.1±0.6	239±10	288±10	2360±71		
8.10	P8	17.5±2	10.8±2	110.1±7	182.2±8.6	154±7	41±7	0.5	0.5	17±2	2785±121	492±17		
8.50		15	17	60	180	183								
8.75	P9	8	5	24	141	167	5±2	0.5	0.5	186±8	382±25	317±14		
9.00	P4	40	16	65	349	323	123±18	0.8±0.4	0.9±0.7	52±4	3473±142	1007±38		
9.25		11	6	23	156	129								
9.75		6	3	36	144	158								
10.00	P11	37	31	104	338	280	3±2	0.6±0.5	0.7±0.4	126±6	62±9	160±8		
11.70	P12	6.1±1.2	7.6±1.2	67.4±4.8	171.8±8.2	164±8	10±4	1.2±0.2	3.7±1.2	97±6	639±32	761±26		
13.40	P13	26.2±2.6	25.2±2.5	299.5±17	343.9±15.2	336±15	112±18	15±2	16±2	112±18	3430±18	670±15		
15.10		16.3±1.5	12.2±1.2	112.5±6.6	141.3±7.3	126±6	0.5	0.5	0.5	139±7	55±12	52±5		
16.00		9.1±1.5	8.9±1.5	131.4±8.5	206±10.4	136±7								
16.20		10.2±1.3	11.8±1.4	123.7±7.5	200.3±9.5	163±7								
16.30	Bundle 5	11.9±1.4	17.7±1.8	139.8±8.2	195.5±9.2	170±8	20±11	<0.5	<0.5	104±8	16±8	62±8	Peat zone	
24							7±5	10±2	67±6	30±3	109±12	72±8		

Table 7.18 - Nordic main tailings data. Additional data from location T-5. Solid and liquid phase comparison of radioisotope activity ratios. Activity ratio = daughter activity/parent activity at various depths.

SOLID PHASE				LIQUID PHASE		
SAMPLE	RATIO	RATIO	RATIO	RATIO	RATIO	RATIO
DESCRIPTION	Pb-210/ Ra-226	Ra-226/ Th-230	Th-228/ Th-232	Pb-210/ Ra-226	Ra-226/ Th-230	Th-228/ Th-232
DEPTH (m)						
IP - 5 - 5.58	0.95	4.59	1.14	22.69	2.15	177.78
IP - 7 - 6.60	1.22	1.03	1.02	72.90	3.26	17.93
IP - 3 - 6.75	0.95	5.71	1.40	29.89	5.74	32.82
IP - 1 - 7.25	1.27	4.09	4.17	0.23	274.00	8.00
IP - 2 - 8.00	1.07	9.42	1.40	1.21	217.27	1.00
IP - 8 - 8.10	0.85	1.65	1.62	163.82	34.00	82.00
IP - 9 - 8.75	1.18	5.88	1.60	2.05	372.00	10.00
IP - 4 - 9.00	0.93	5.37	2.50	66.79	57.78	153.75
IP - 11 - 10.00	0.83	3.25	1.19	0.49	180.00	5.00
IP - 12 - 11.70	0.95	2.55	0.80	6.59	26.22	8.33
IP - 13 - 13.40	0.98	1.15	1.04	30.63	7.00	7.47
I 15.10	0.89	1.26	1.34	0.40	278.00	1.00
IB - 5 - 16.30	0.87	1.40	0.67	0.15	208.00	40.00

Table 7.19 - Solubility of major radioisotopes and related sulphate salts in uranium tailings

Salts	Solubility (g/100 mL)	Temperature °C
RaSO ₄	0.000002	25
PbSO ₄	0.00425	25
Th(SO ₄) . 4 H ₂ O	9.41	17
Th(SO ₄) . 6 H ₂ O	1.63	16
Th(SO ₄) . 8 H ₂ O	1.88	25
Th(SO ₄) . 9 H ₂ O	1.57	20
U(SO ₄) . 4 H ₂ O	23.0	11
U(SO ₄) . 8 H ₂ O	11.3	18
Pb-sulphide	0.000008	24
Po-sulphide	Insoluble	-
Bi-sulphide	0.000018	18

Stratification of Nordic Tailings Area

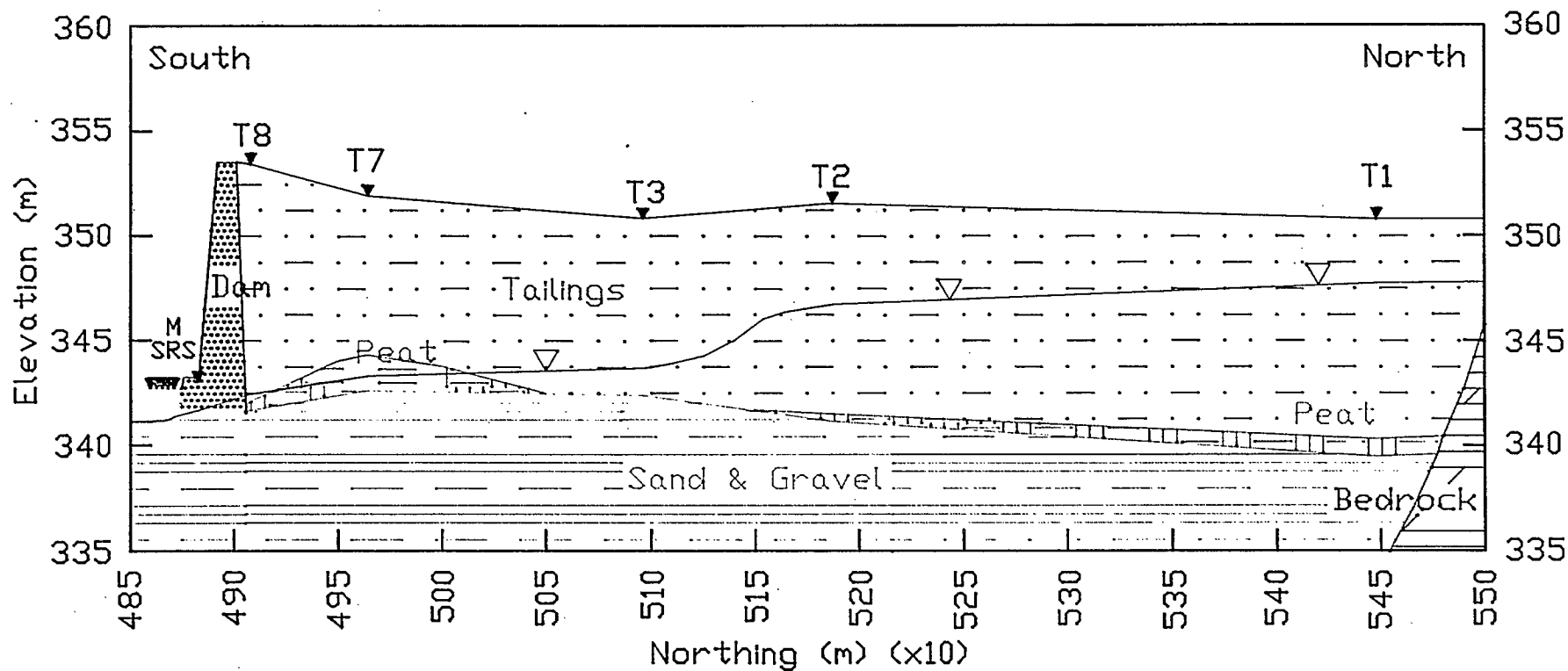


Fig. 7.1 - General stratification of Nordic main tailings area. Vertical north-south profile. Elevation (m) ASL versus Northing (m) (x10).

Piezometer Hydraulic Head (m)

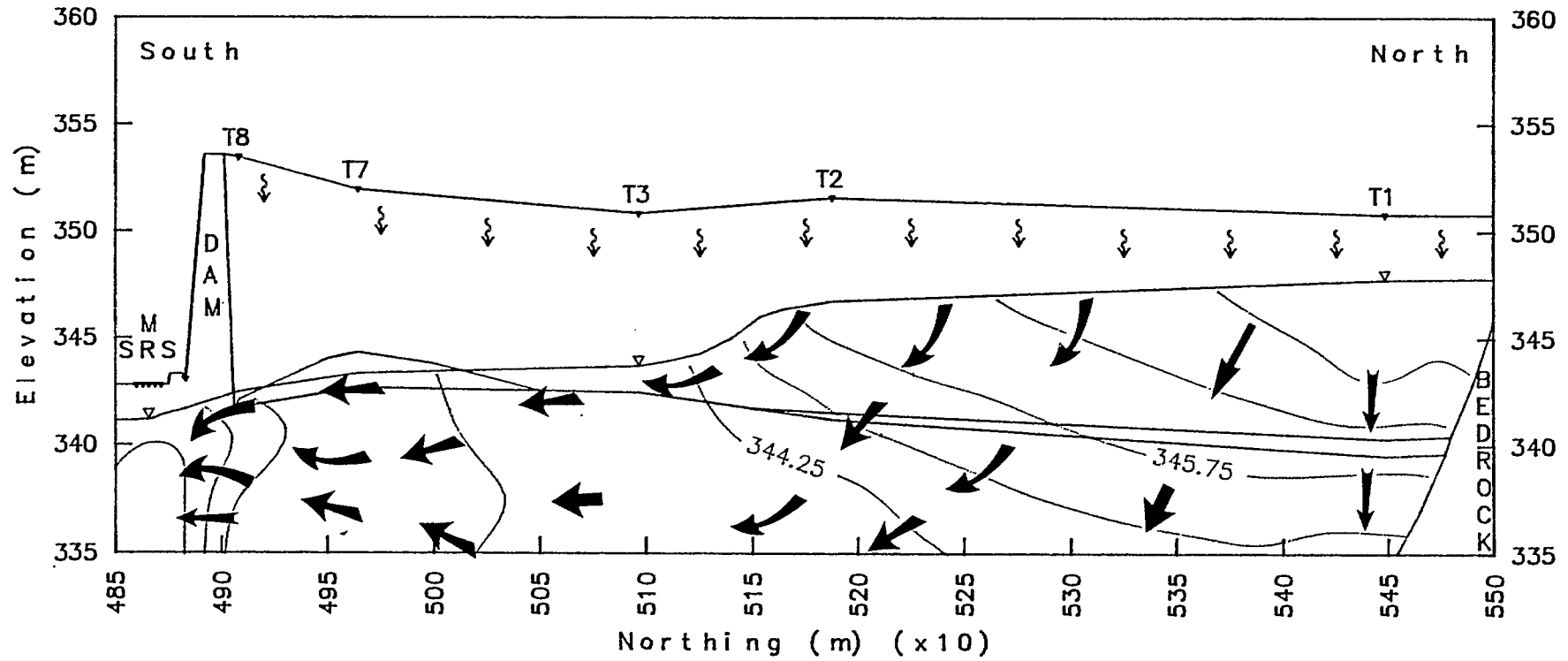


Fig. 7.2 - General groundwater flow pattern of Nordic main tailings. Vertical north-south profile. Based on data interpretation of hydraulic head into equipotential line contour mapping.

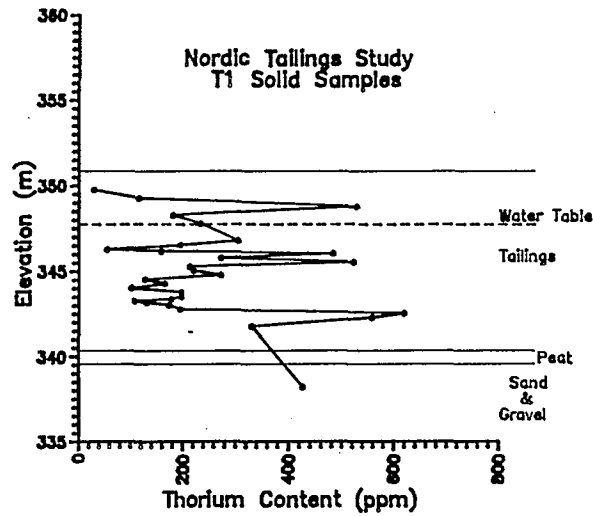


Fig. 7.3 - Nordic main tailings data. Location T-1, solid phase concentration of thorium at various depths.

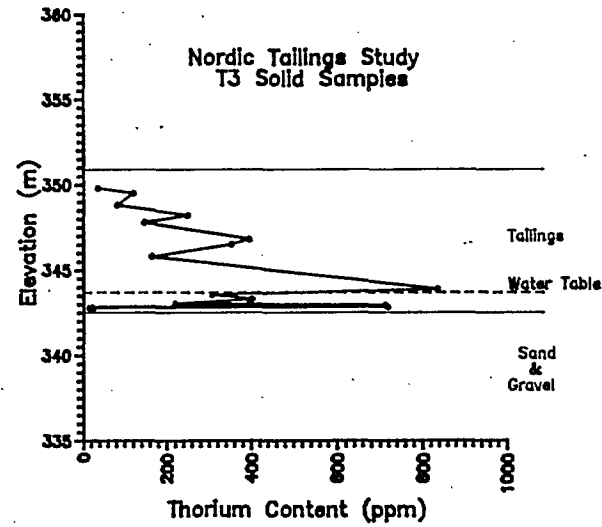


Fig. 7.5 - Nordic main tailings data. Location T-3, solid phase concentration of thorium at various depths.

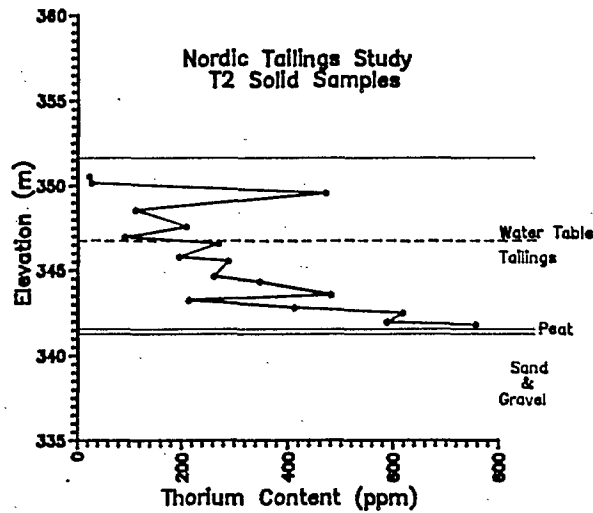


Fig. 7.4 - Nordic main tailings data. Location T-2, solid phase concentration of thorium at various depths.

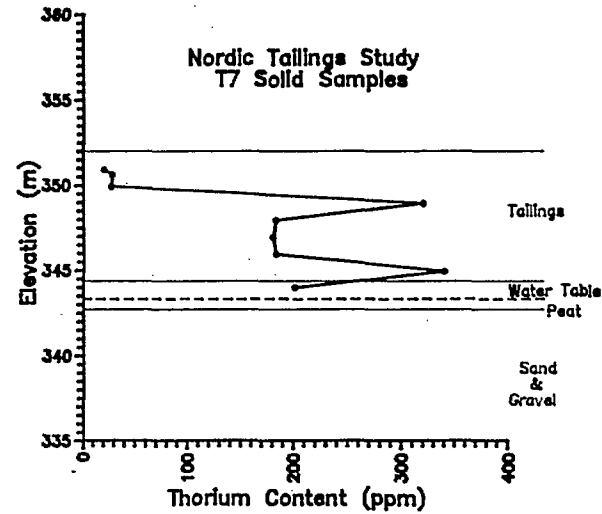


Fig. 7.6 - Nordic main tailings data. Location T-7, solid phase concentration of thorium at various depths.

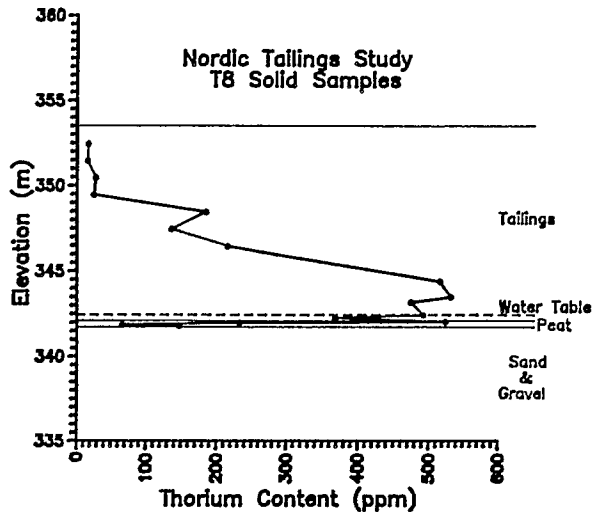


Fig. 7.7 - Nordic main tailings data. Location T-8, solid phase concentration of thorium at various depths.

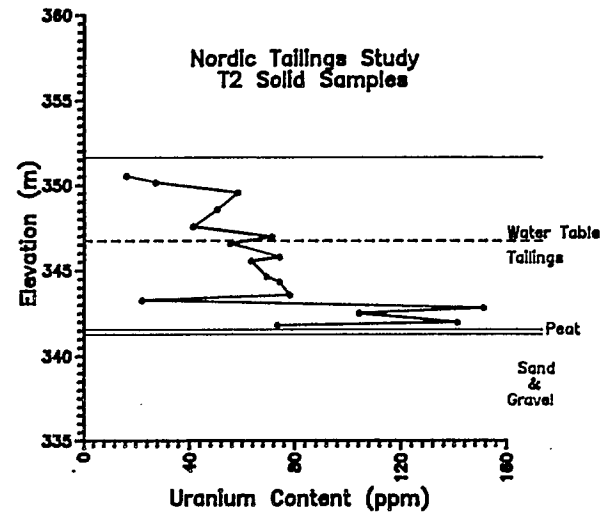


Fig. 7.9 - Nordic main tailings. Location T-2, solid phase concentration of uranium at various depths.

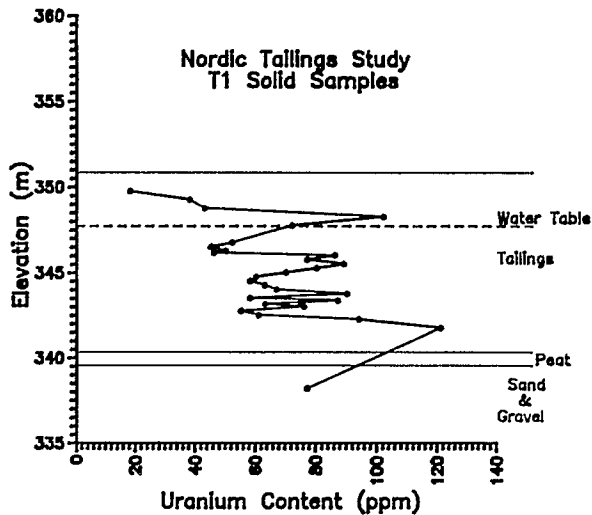


Fig. 7.8 - Nordic main tailings. Location T-1, solid phase concentration of uranium at various depths.

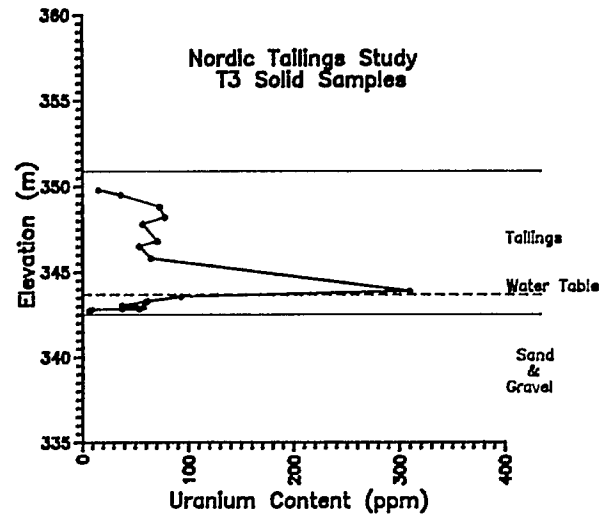


Fig. 7-10 - Nordic main tailings data. Location T-3, solid phase concentration of uranium at various depths.

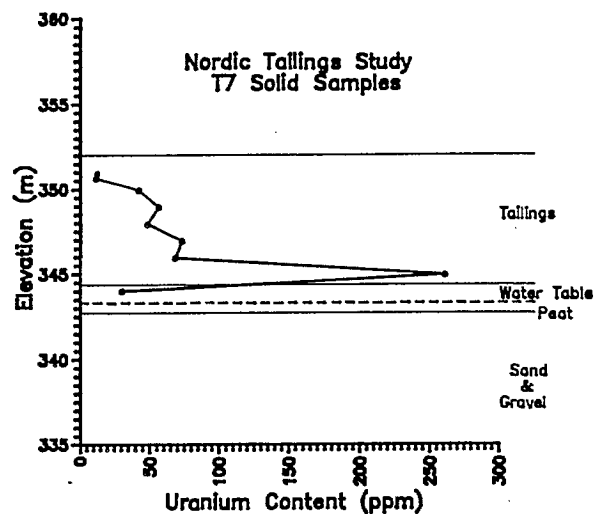


Fig. 7.11 - Nordic main tailings data. Location T-7, solid phase concentration of uranium at various depths.

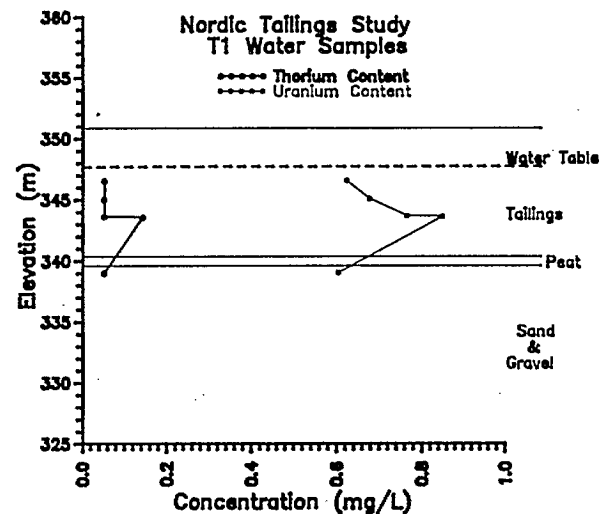


Fig. 7.13 - Nordic main tailings data. Location T-1, liquid phase concentration of thorium and uranium at various depths.

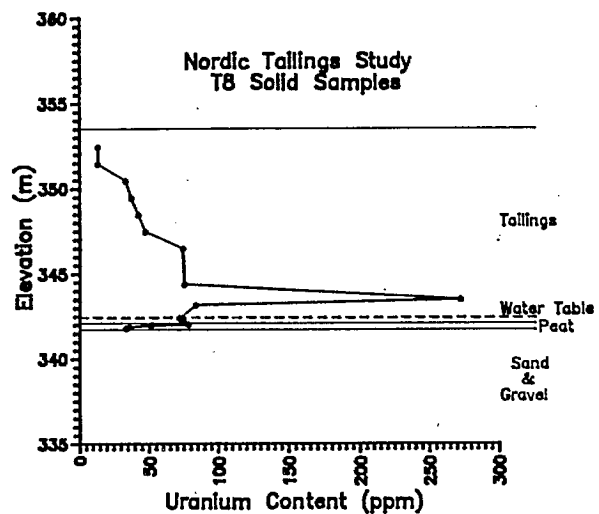


Fig. 7.12 - Nordic main tailings data. Location T-8, solid phase concentration of uranium at various depths.

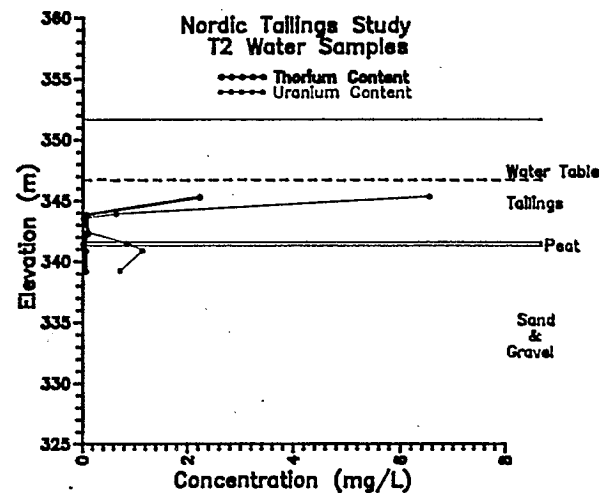


Fig. 7.14 - Nordic main tailings data. Location T-2, liquid phase concentration of thorium and uranium at various depths.

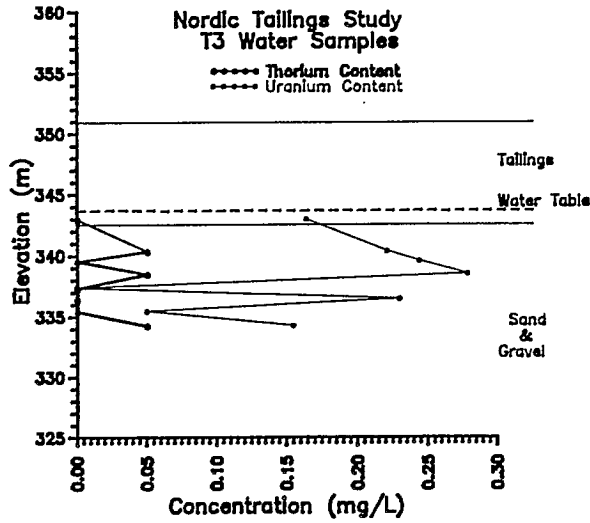


Fig. 7.15 - Nordic main tailings data. Location T-3, liquid phase concentration of thorium and uranium at various depths.

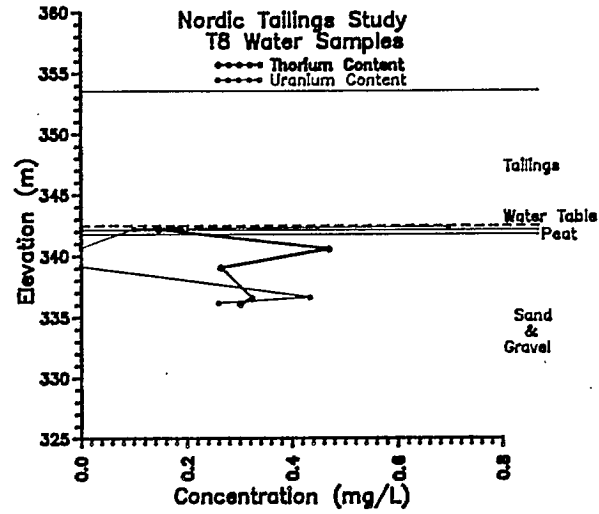


Fig. 7.17 - Nordic main tailings data. Location T-8, liquid phase concentration of thorium and uranium at various depths.

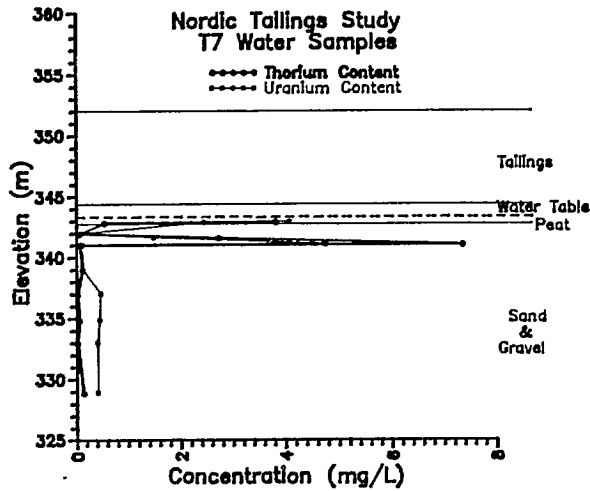


Fig. 7.16 - Nordic main tailings data. Location T-7, liquid phase concentration of thorium and uranium at various depths.

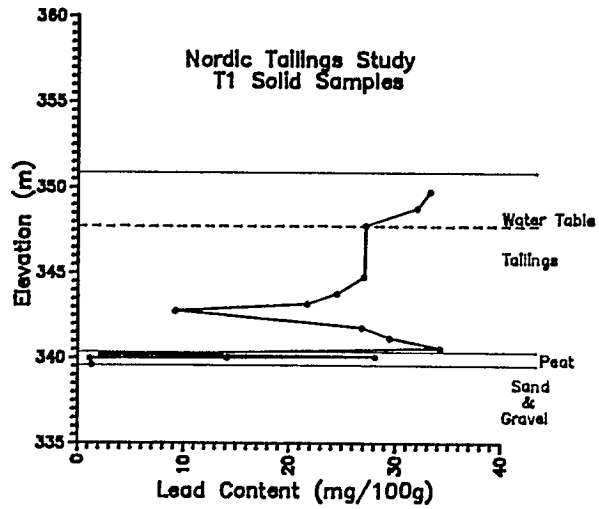


Fig. 7.18 - Nordic main tailings data. Location T-1, solid phase concentration of lead at various depths.

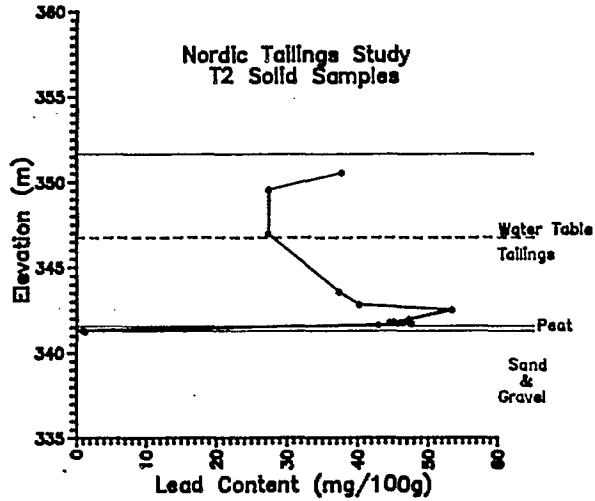


Fig. 7.19 - Nordic main tailings data. Location T-2, solid phase concentration of lead at various depths.

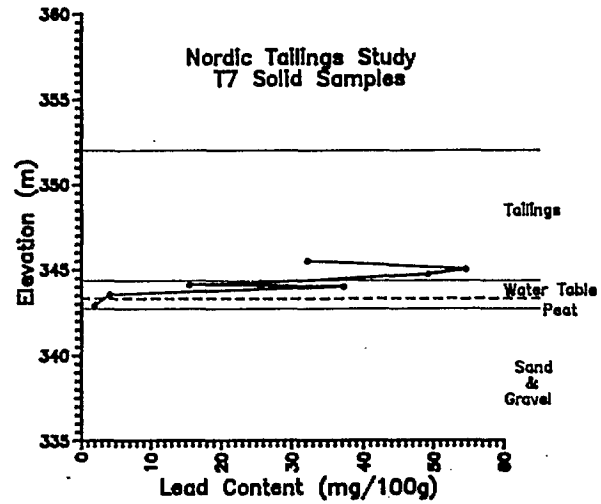


Fig. 7.21 - Nordic main tailings data. Location T-7, solid phase concentration of lead at various depths.

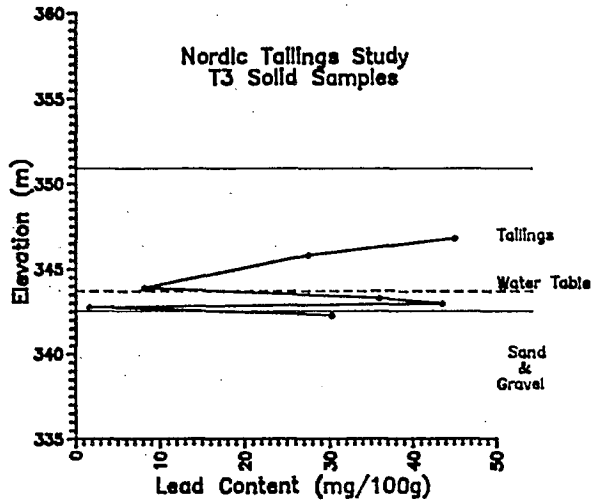


Fig. 7.20 - Nordic main tailings data. Location T-3, solid phase concentration of lead at various depths.

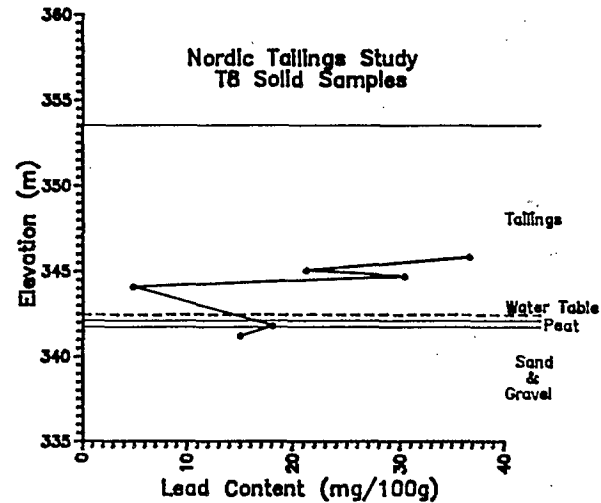


Fig. 7.22 - Nordic main tailings data. Location T-8, solid phase concentration of lead at various depths.

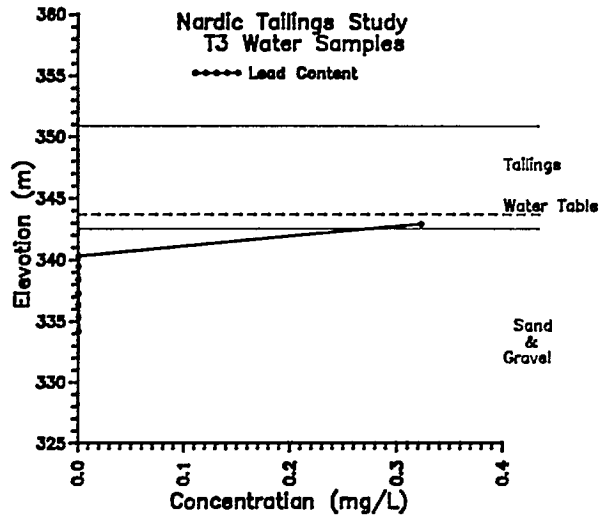


Fig. 7.23 - Nordic main tailings data. Location T-3, liquid phase concentration of lead at various depths.

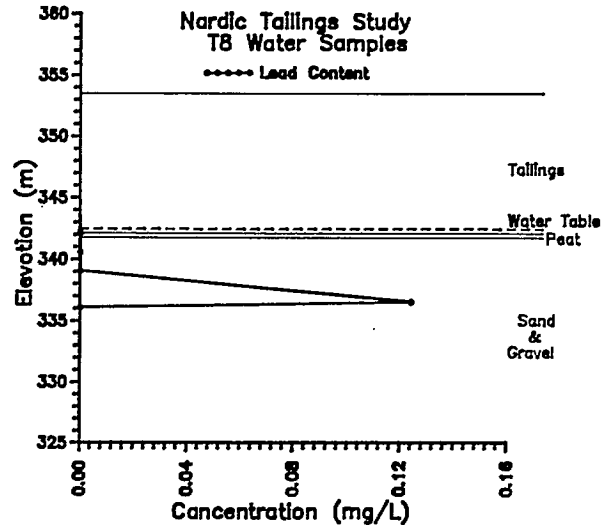


Fig. 7.25 - Nordic main tailings data. Location T-8, liquid phase concentration of lead at various depths.

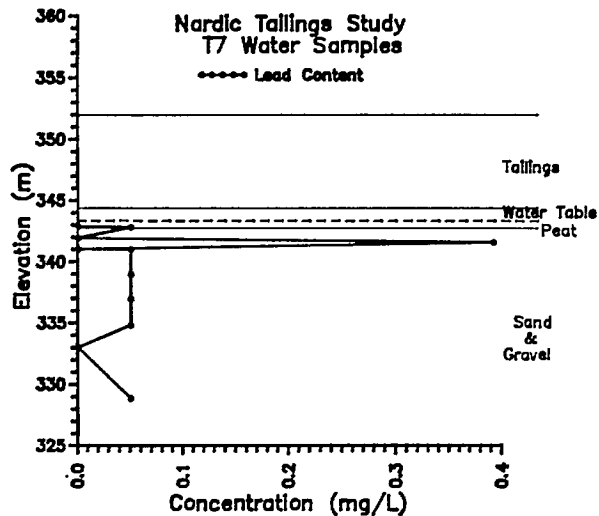


Fig. 7.24 - Nordic main tailings data. Location T-7, liquid phase concentration of lead at various depths.

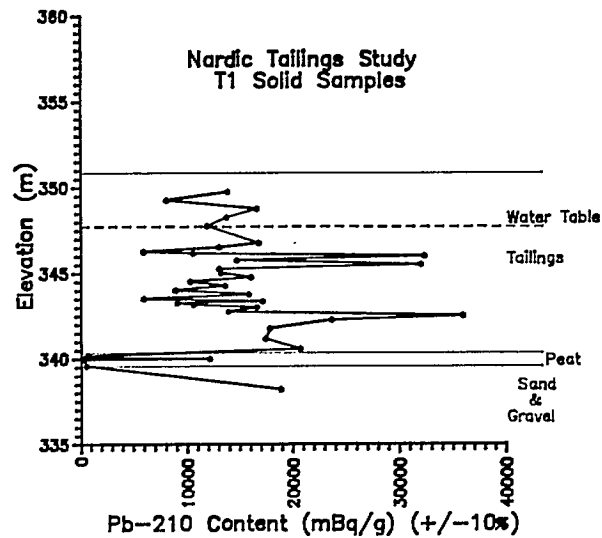


Fig. 7.26 - Nordic main tailings data. Location T-1, solid phase concentration of ^{210}Pb at various depths.

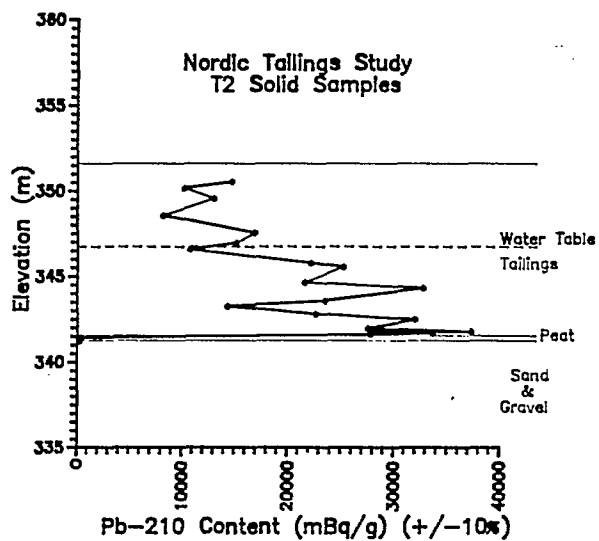


Fig. 7.27 - Nordic main tailings data. Location T-2, solid phase concentration of ^{210}Pb at various depths.

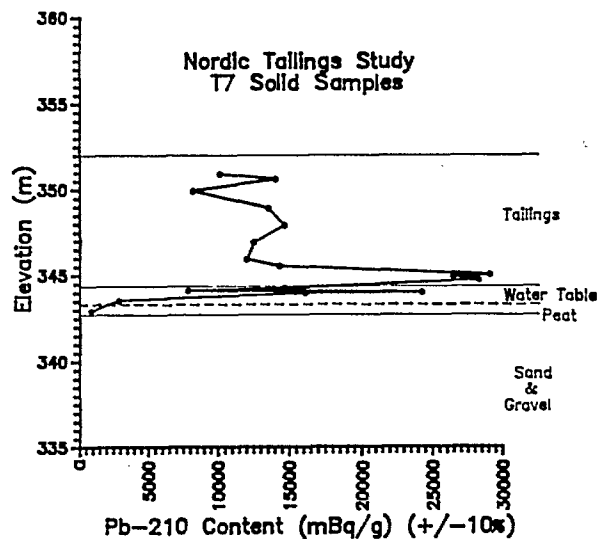


Fig. 7.29 - Nordic main tailings data. Location T-7, solid phase concentration of ^{210}Pb at various depths.

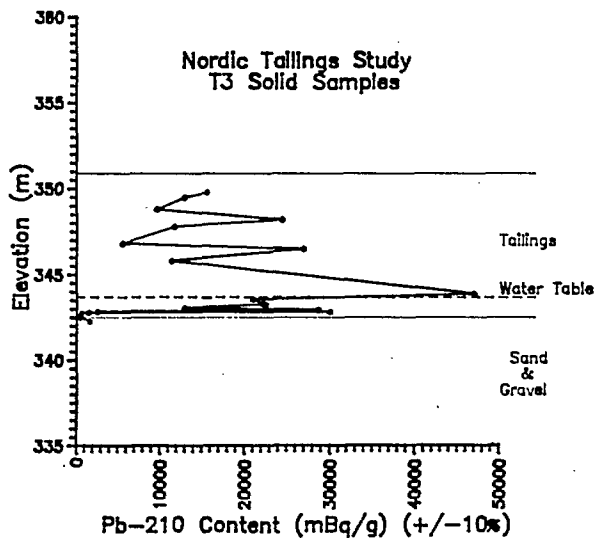


Fig. 7.28 - Nordic main tailings data. Location T-3, solid phase concentration of ^{210}Pb at various depths.

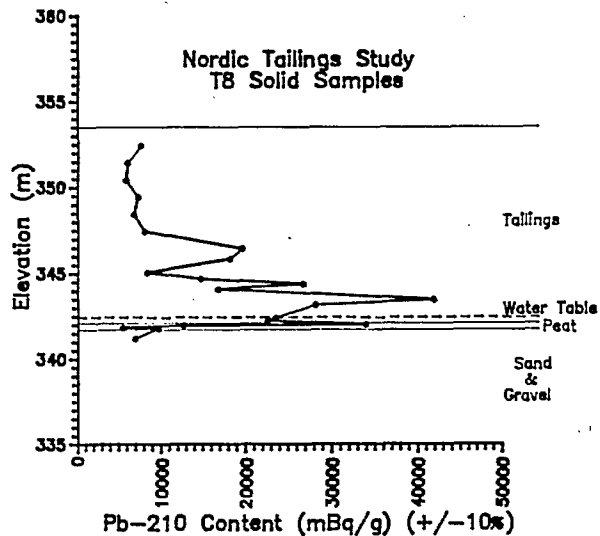


Fig. 7.30 - Nordic main tailings data. Location T-8, solid phase concentration of ^{210}Pb at various depths.

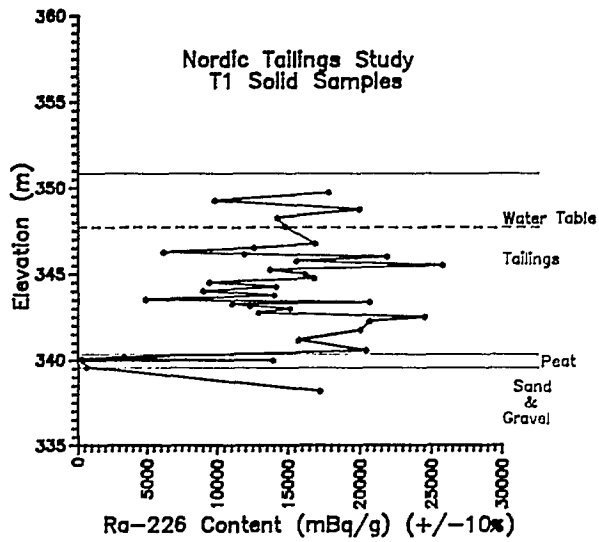


Fig. 7.31 - Nordic main tailings data. Location T-1, solid phase concentration of ^{226}Ra at various depths.

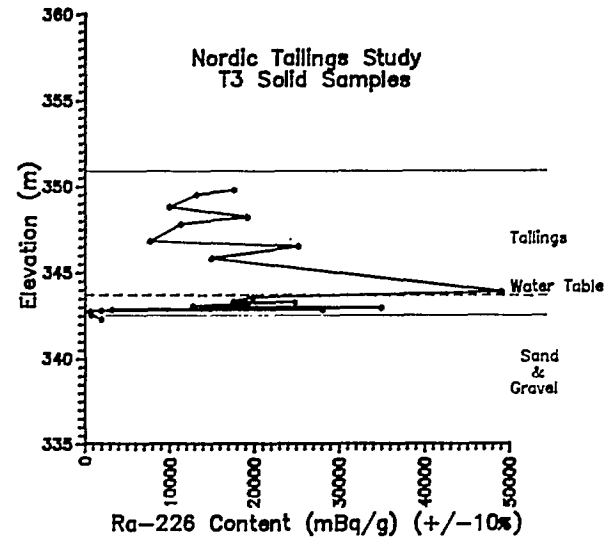


Fig. 7.33 - Nordic main tailings data. Location T-3, solid phase concentration of ^{226}Ra at various depths.

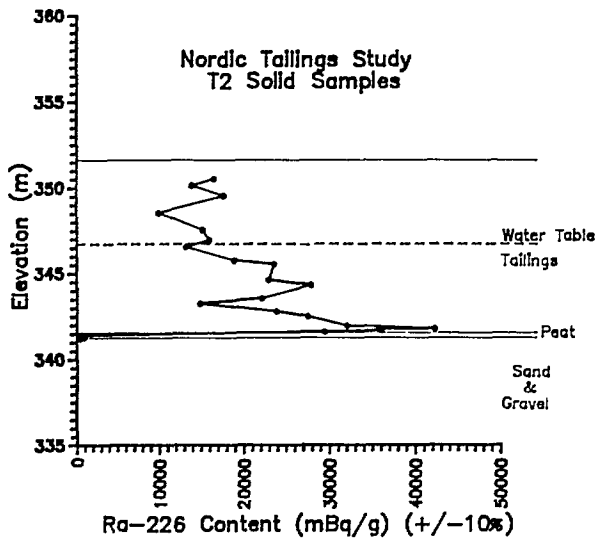


Fig. 7.32 - Nordic main tailings data. Location T-2, solid phase concentration of ^{226}Ra at various depths.

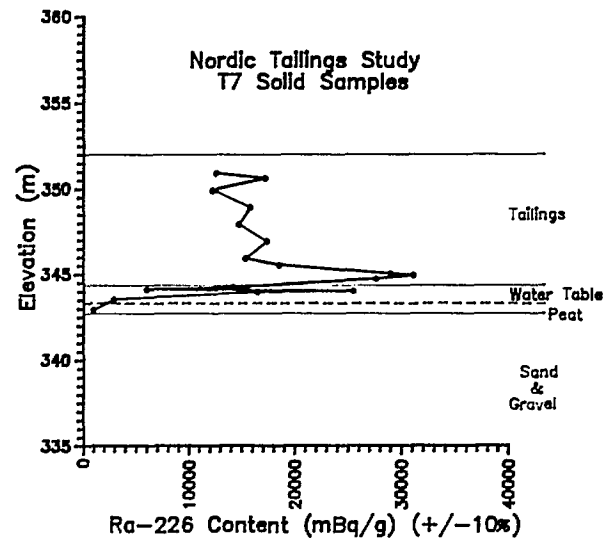


Fig. 7.34 - Nordic main tailings data. Location T-7, solid phase concentration of ^{226}Ra at various depths.

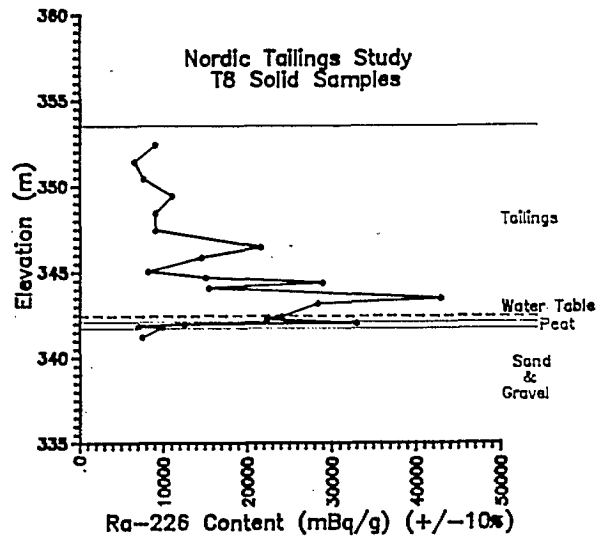


Fig. 7.35 - Nordic main tailings data. Location T-8, solid phase concentration of ^{226}Ra at various depths.

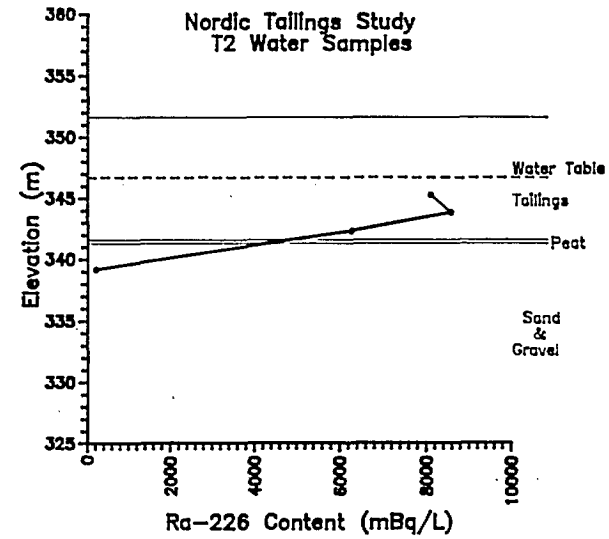


Fig. 7.37 - Nordic main tailings data. Location T-2, liquid phase concentration of ^{226}Ra at various depths.

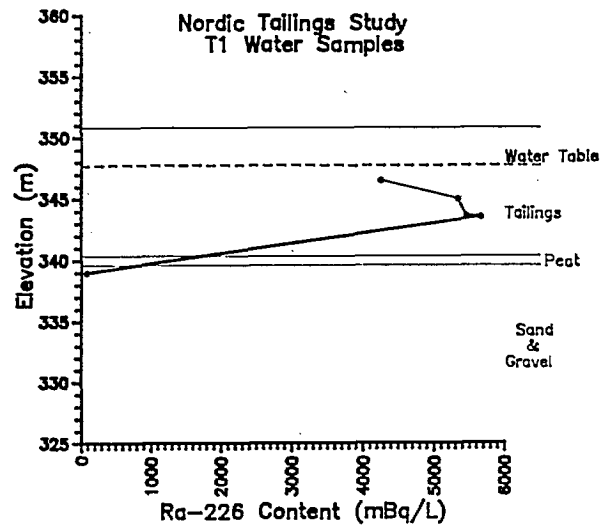


Fig. 7.36 - Nordic main tailings data. Location T-1, liquid phase concentration of ^{226}Ra at various depths.

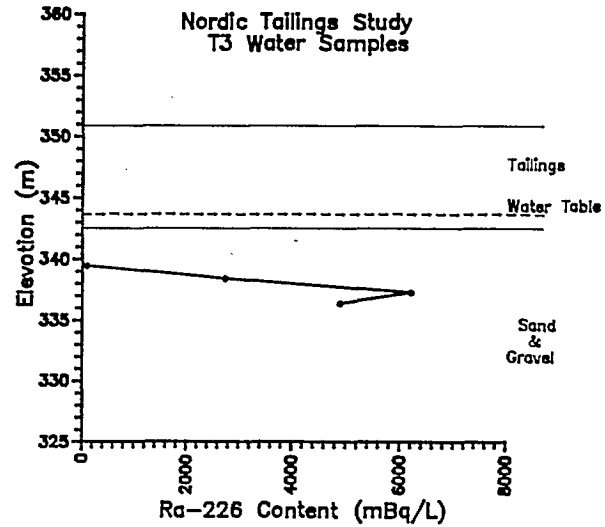


Fig. 7.38 - Nordic main tailings data. Location T-3, liquid phase concentration of ^{226}Ra at various depths.

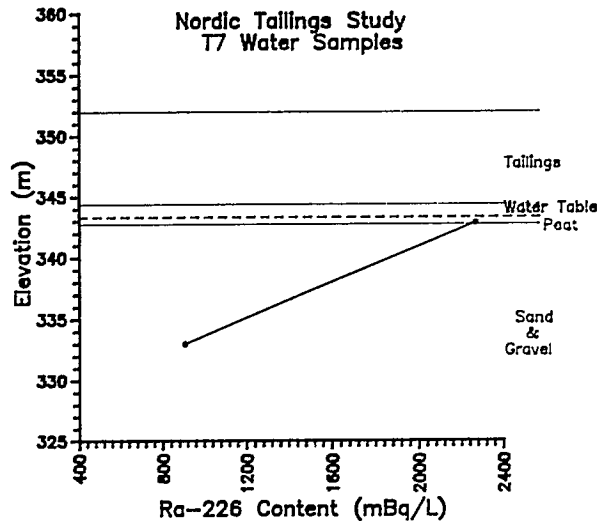


Fig. 7.39 - Nordic main tailings data. Location T-7, liquid phase concentration of ^{226}Ra at various depths.

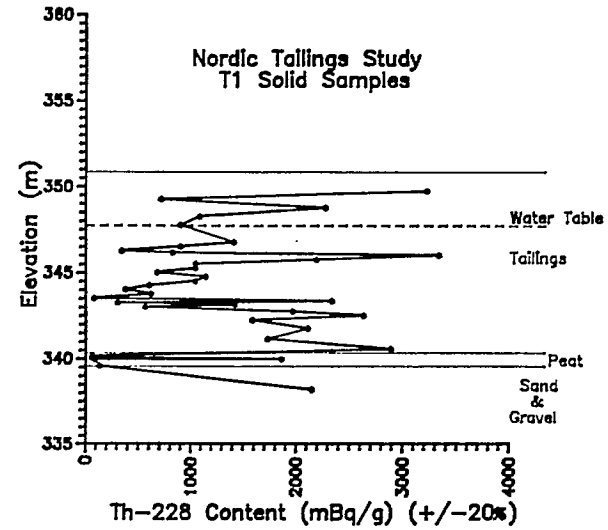


Fig. 7.41 - Nordic main tailings data. Location T-1, solid phase concentration of ^{228}Th at various depths.

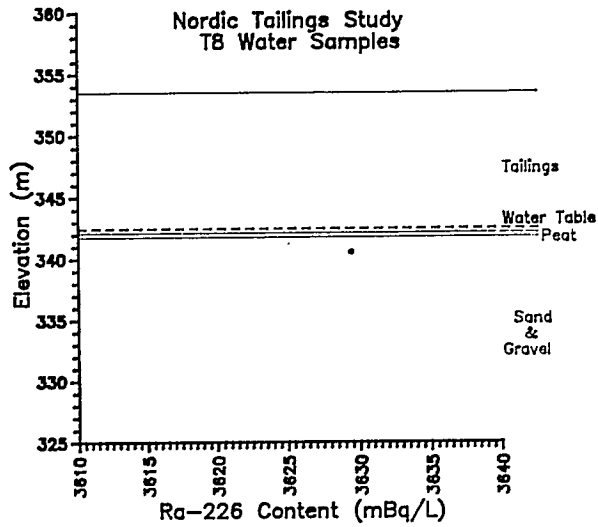


Fig. 7.40 - Nordic main tailings data. Location T-8, liquid phase concentration of ^{226}Ra at various depths.

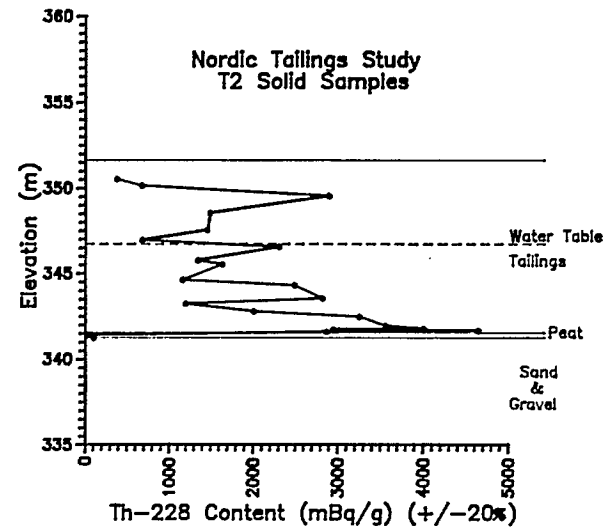


Fig. 7.42 - Nordic main tailings data. Location T-2, solid phase concentration of ^{228}Th at various depths.

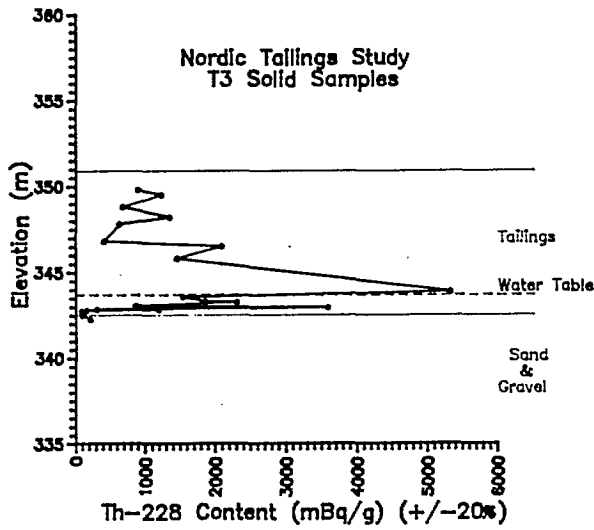


Fig. 7.43 - Nordic main tailings data. Location T-3, solid phase concentration of ^{228}Th at various depths.

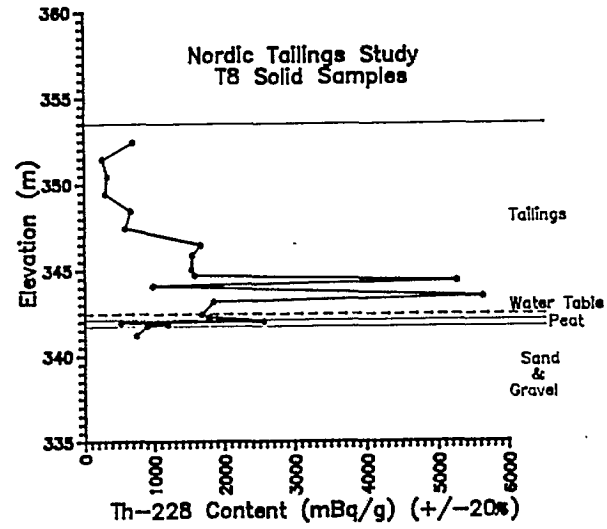


Fig. 7.45 - Nordic main tailings data. Location T-8, solid phase concentration of ^{228}Th at various depths.

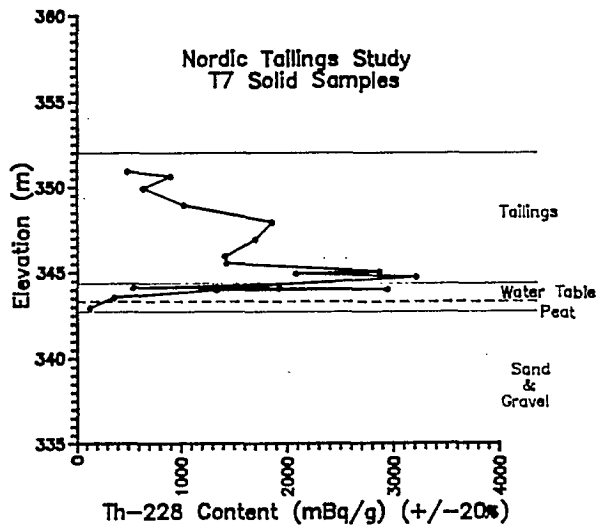


Fig. 7.44 - Nordic main tailings data. Location T-7, solid phase concentration of ^{228}Th at various depths.

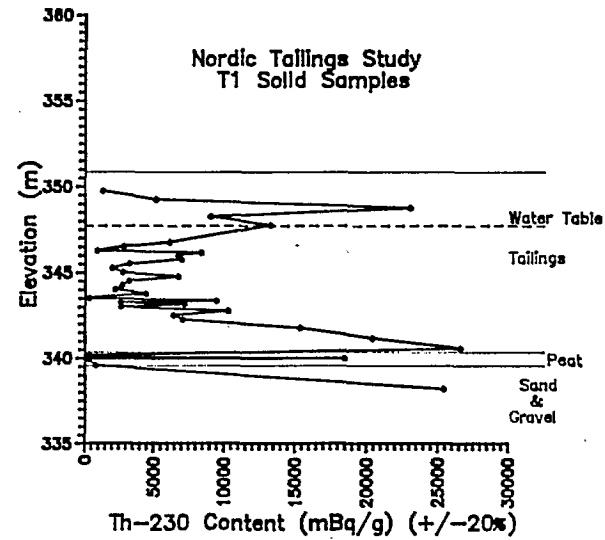


Fig. 7.46 - Nordic main tailings data. Location T-1, solid phase concentration of ^{230}Th at various depths.

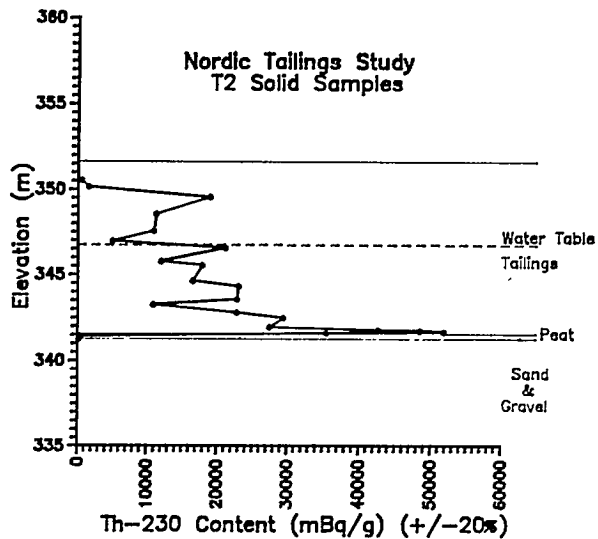


Fig. 7.47 - Nordic main tailings data. Location T-2, solid phase concentration of ^{230}Th at various depths.

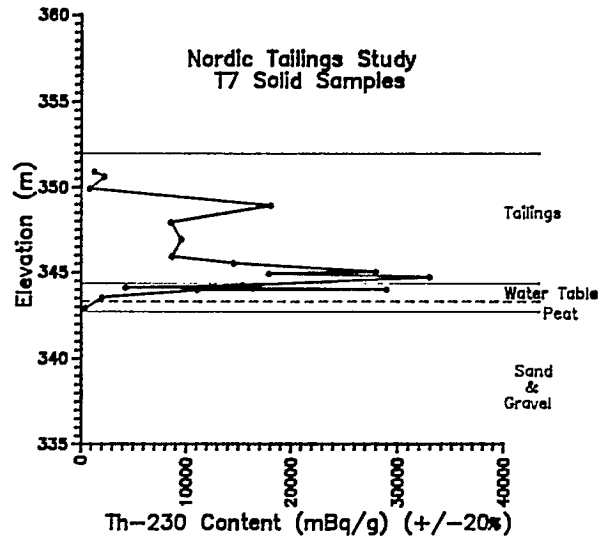


Fig. 7.49 - Nordic main tailings data. Location T-7, solid phase concentration of ^{230}Th at various depths.

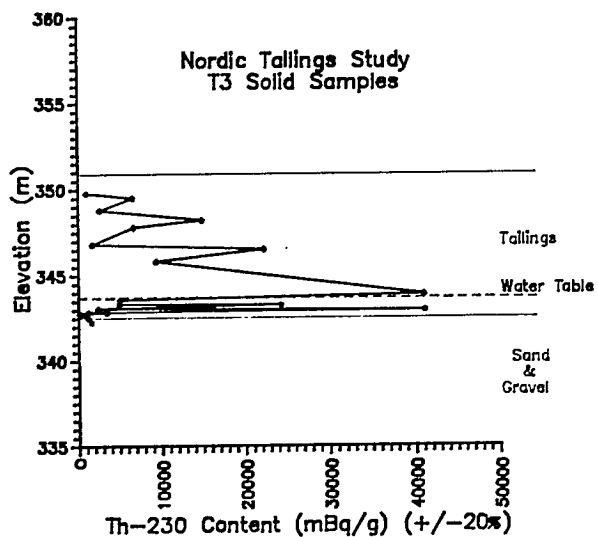


Fig. 7.48 - Nordic main tailings data. Location T-3, solid phase concentration of ^{230}Th at various depths.

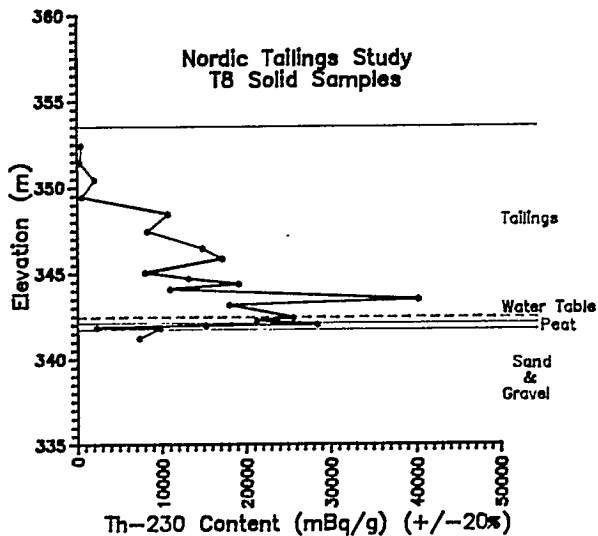


Fig. 7.50 - Nordic main tailings data. Location T-8, solid phase concentration of ^{230}Th at various depths.

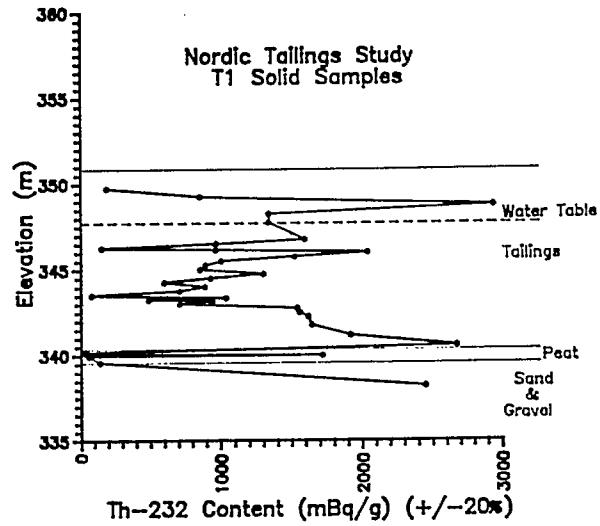


Fig. 7.51 - Nordic main tailings data. Location T-1, solid phase concentration of ^{232}Th at various depths.

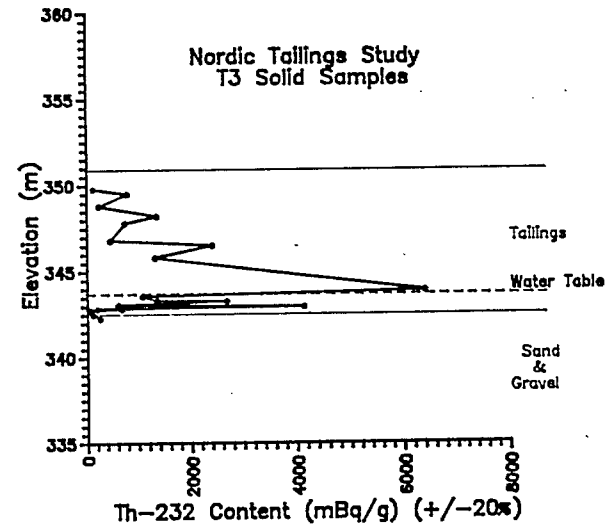


Fig. 7.53 - Nordic main tailings data. Location T-3, solid phase concentration of ^{232}Th at various depths.

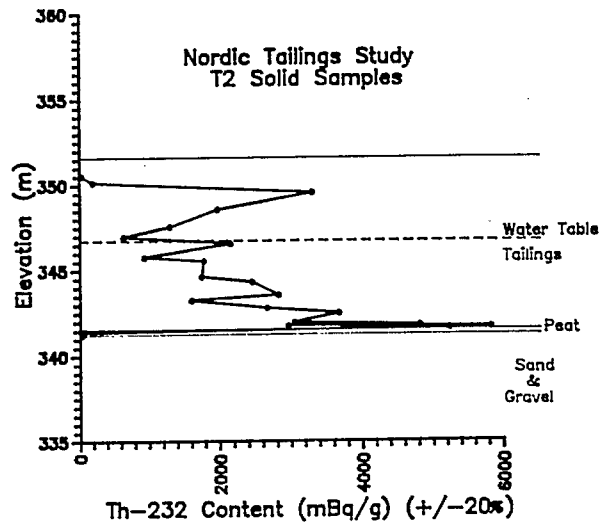


Fig. 7.52 - Nordic main tailings data. Location T-2, solid phase concentration of ^{232}Th at various depths.

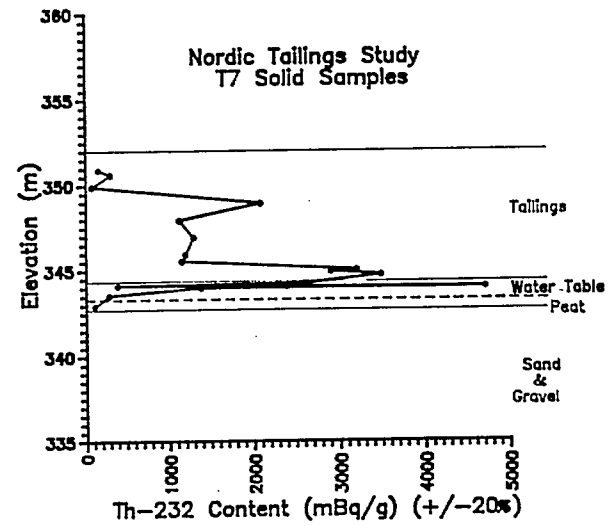


Fig. 7.54 - Nordic main tailings data. Location T-7, solid phase concentration of ^{232}Th at various depths.

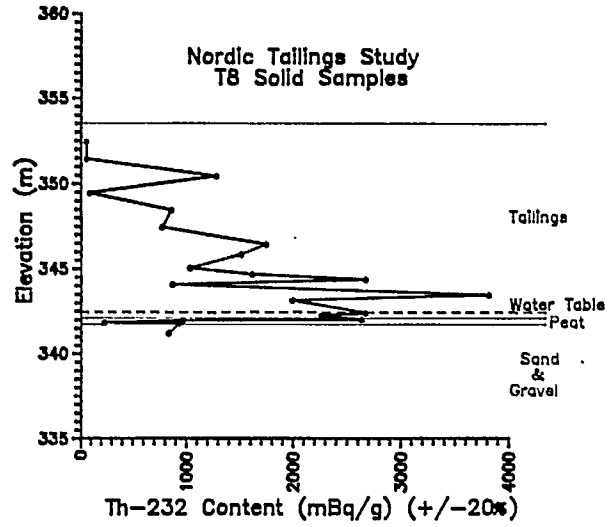


Fig. 7.55 - Nordic main tailings data. Location T-8, solid phase concentration of ^{232}Th at various depths.

Water Sample pH

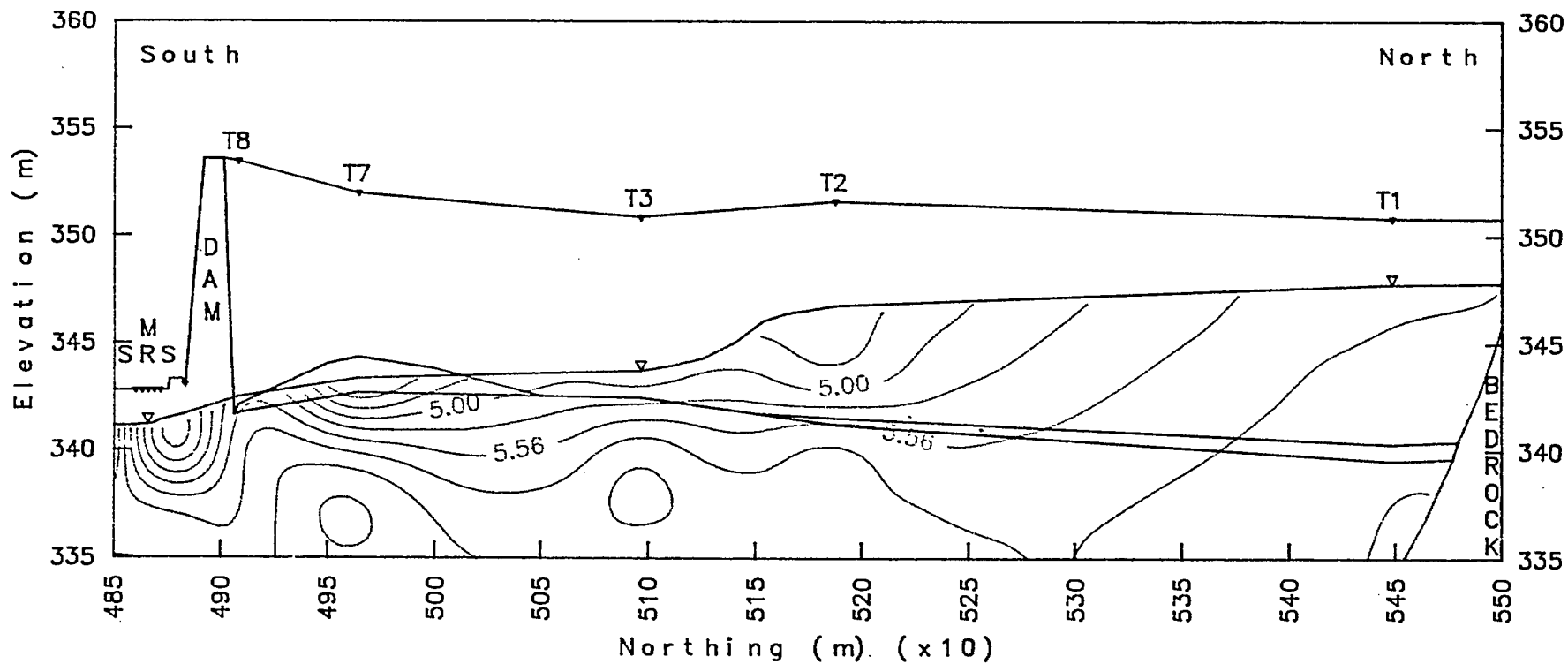


Fig. 7.56 - Nordic main tailings. Vertical north-south profile liquid phase, pH, contour map at various depths.

Water Sample Eh (mV)

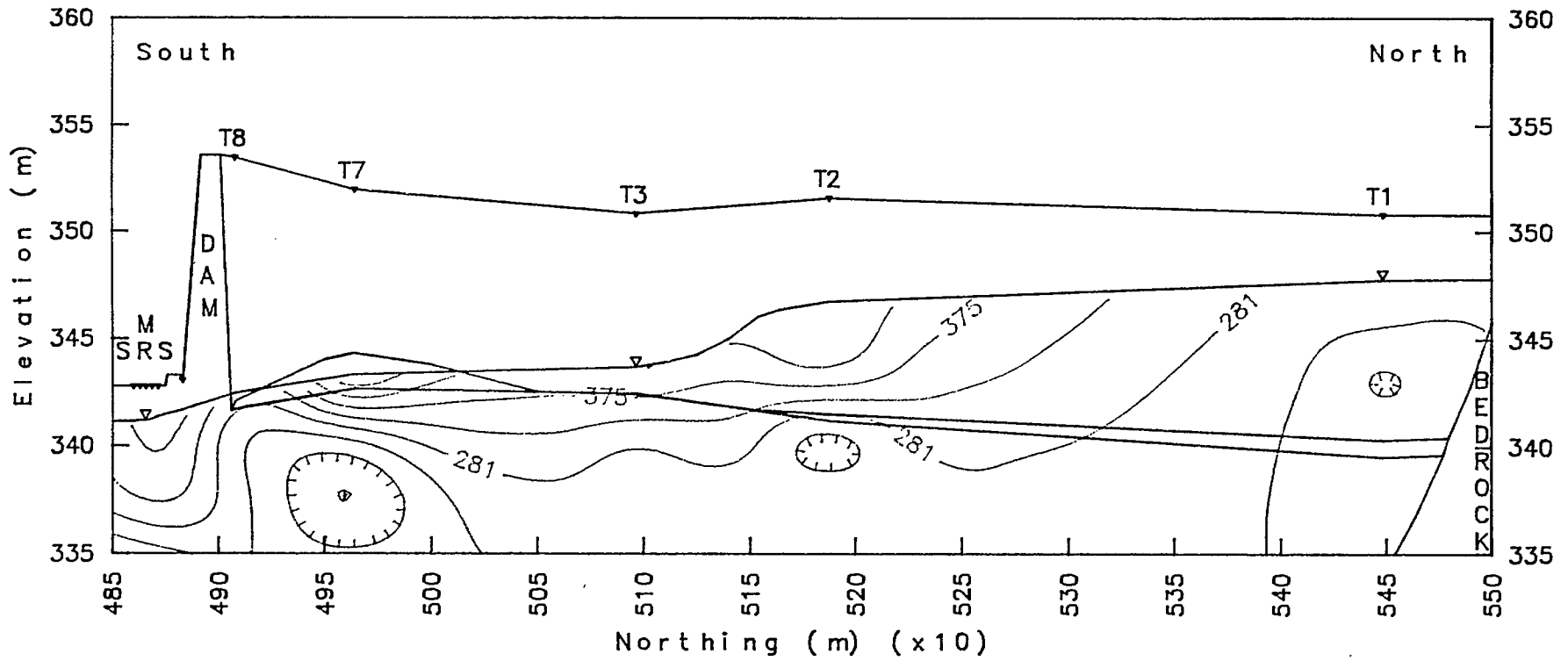


Fig. 7.57 - Nordic main tailings. Vertical north-south profile liquid phase, Eh (mV), contour map at various depths.

Water Sample Ec ($\mu\text{S}/\text{cm}$)

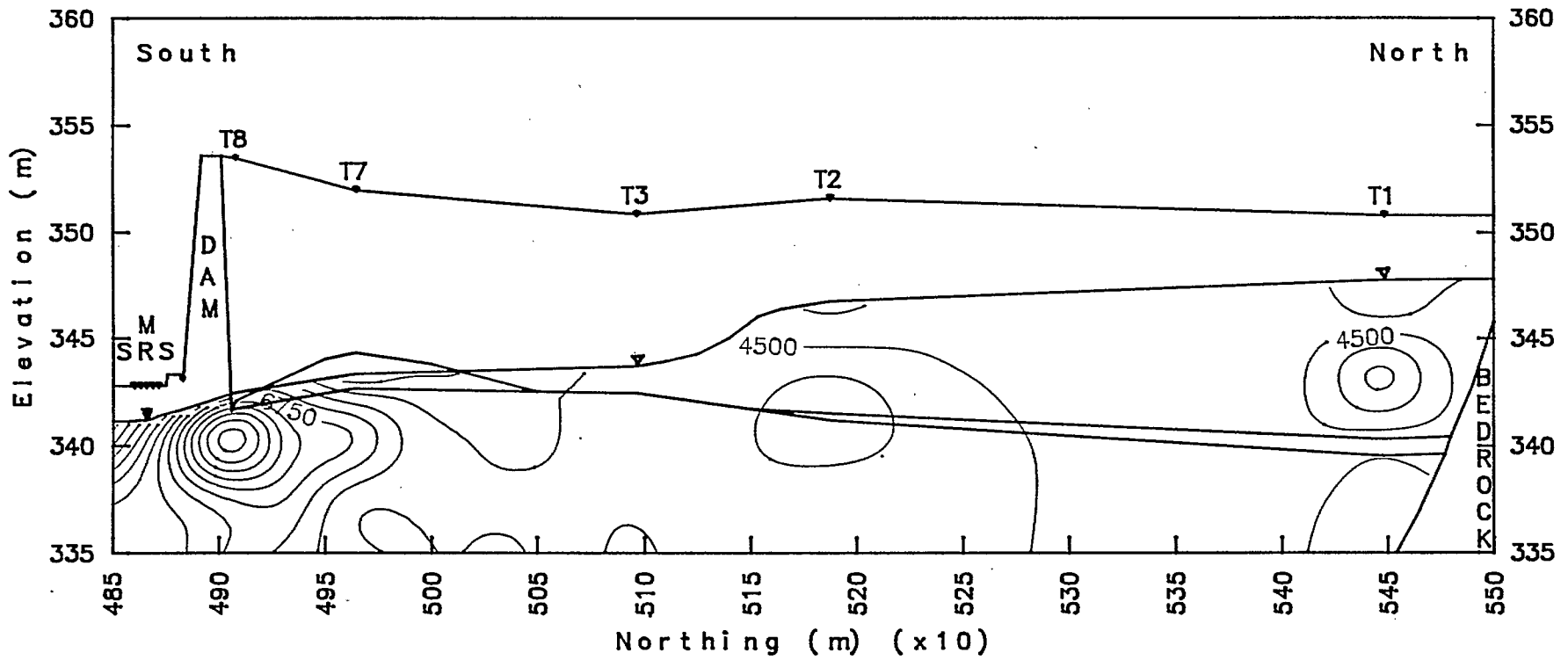


Fig. 7.58 - Nordic main tailings. Vertical north-south profile of liquid sample Ec ($\mu\text{S}/\text{cm}$), contour map at various depths.

Solid Sample Total Sulphate Content (mg/g)

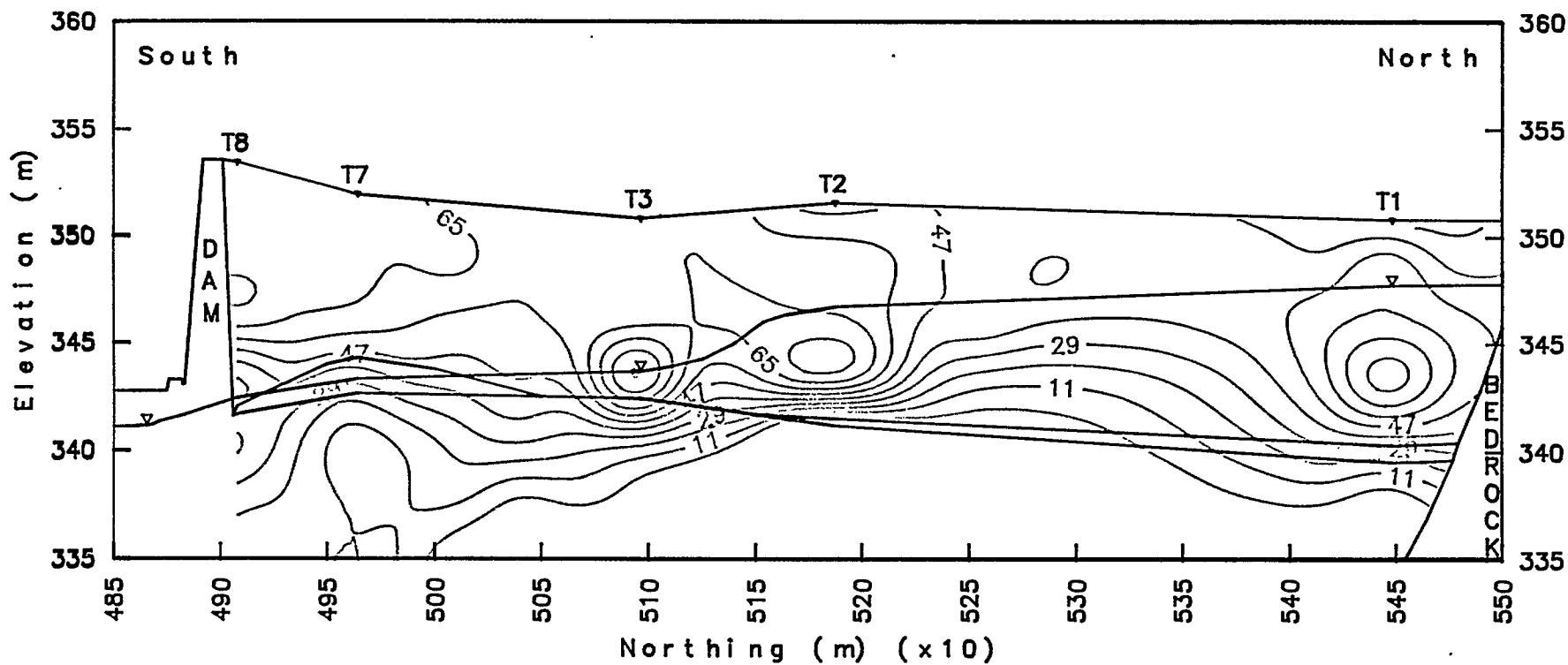


Fig. 7.59 - Nordic main tailings. Vertical north-south profile solid sample of total sulphate content (mg/g), contour map at various depths.

Soluble Sulphate in Solid Sample (mg/g)

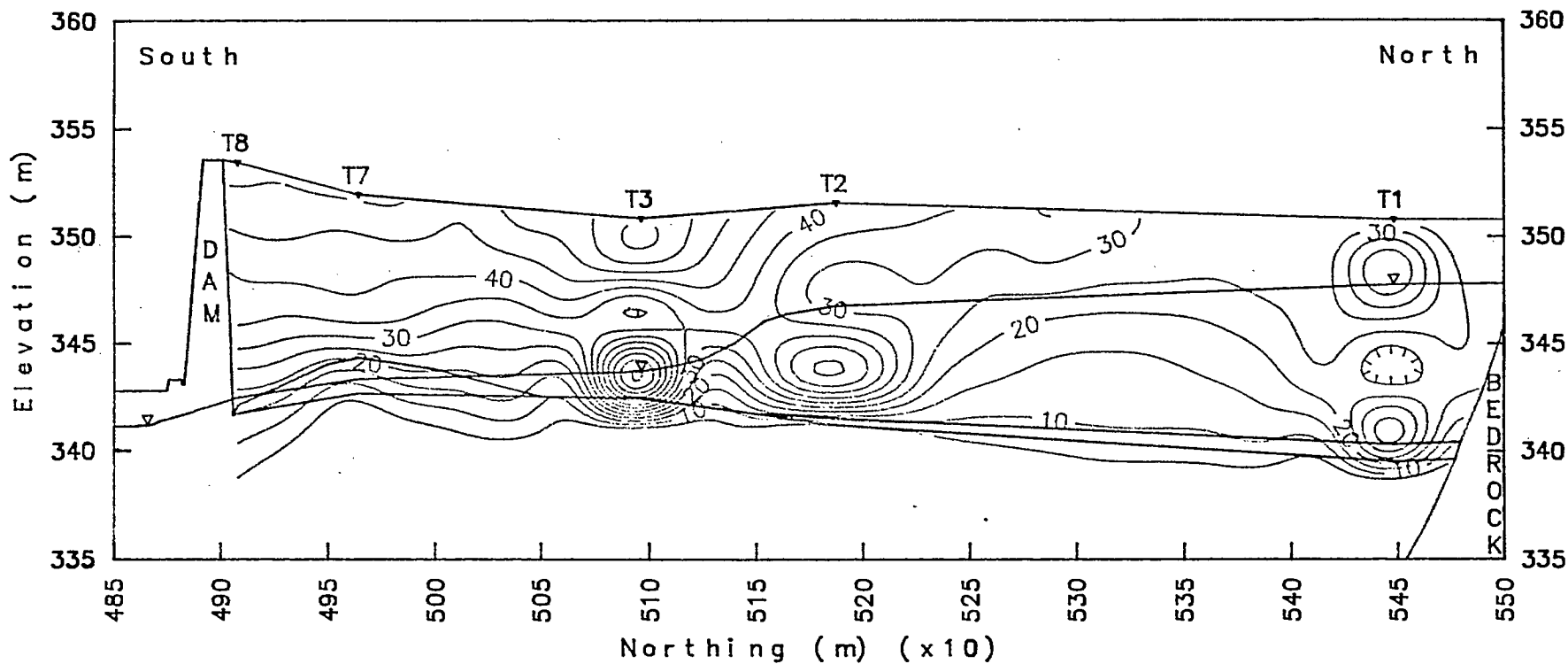


Fig. 7.60 - Nordic main tailings. Vertical north-south profile of soluble sulphate in solid sample (mg/g), contour map at various depths.

Water Sample Sulphate Content (mg/L)

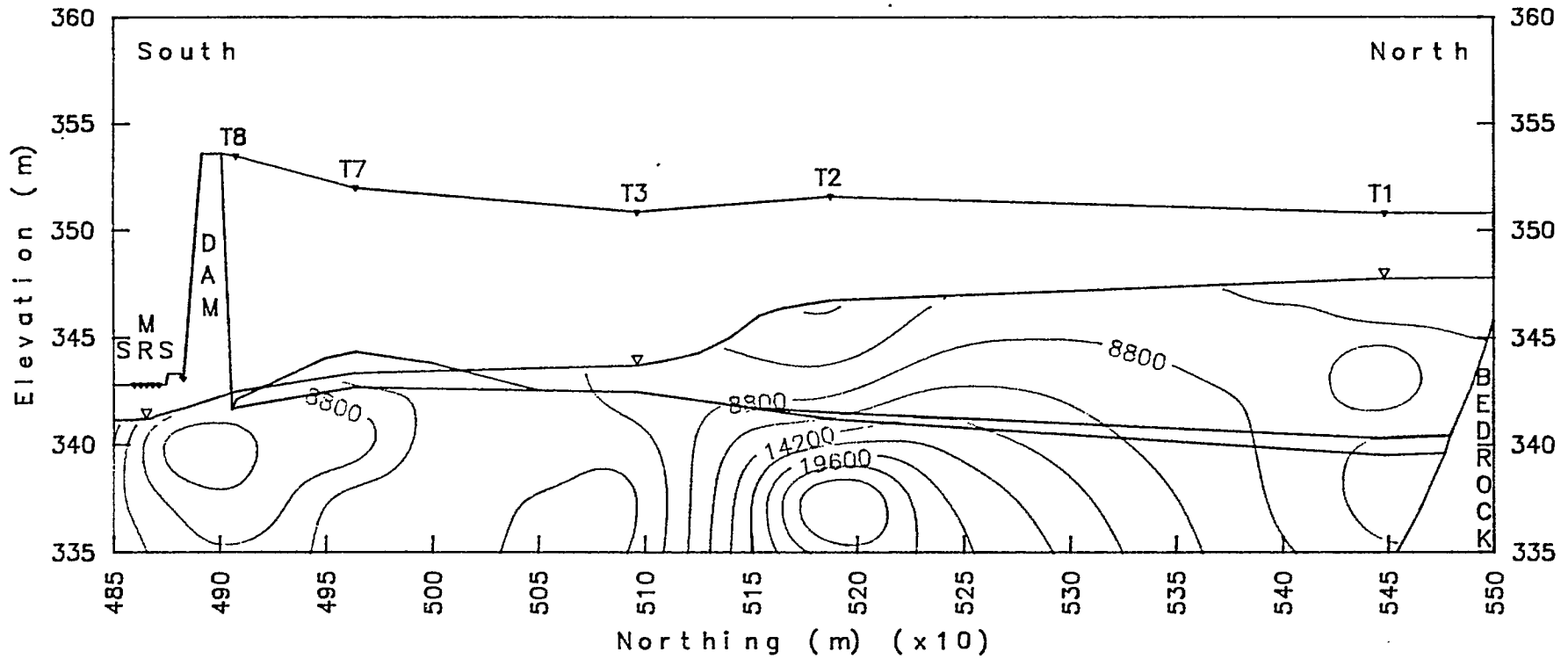


Fig. 7.61 - Nordic main tailings. Vertical north-south profile of liquid sample, sulphate content (mg/L), contour map at various depths.

Total Sulphate Distribution Coefficient (mL/g)

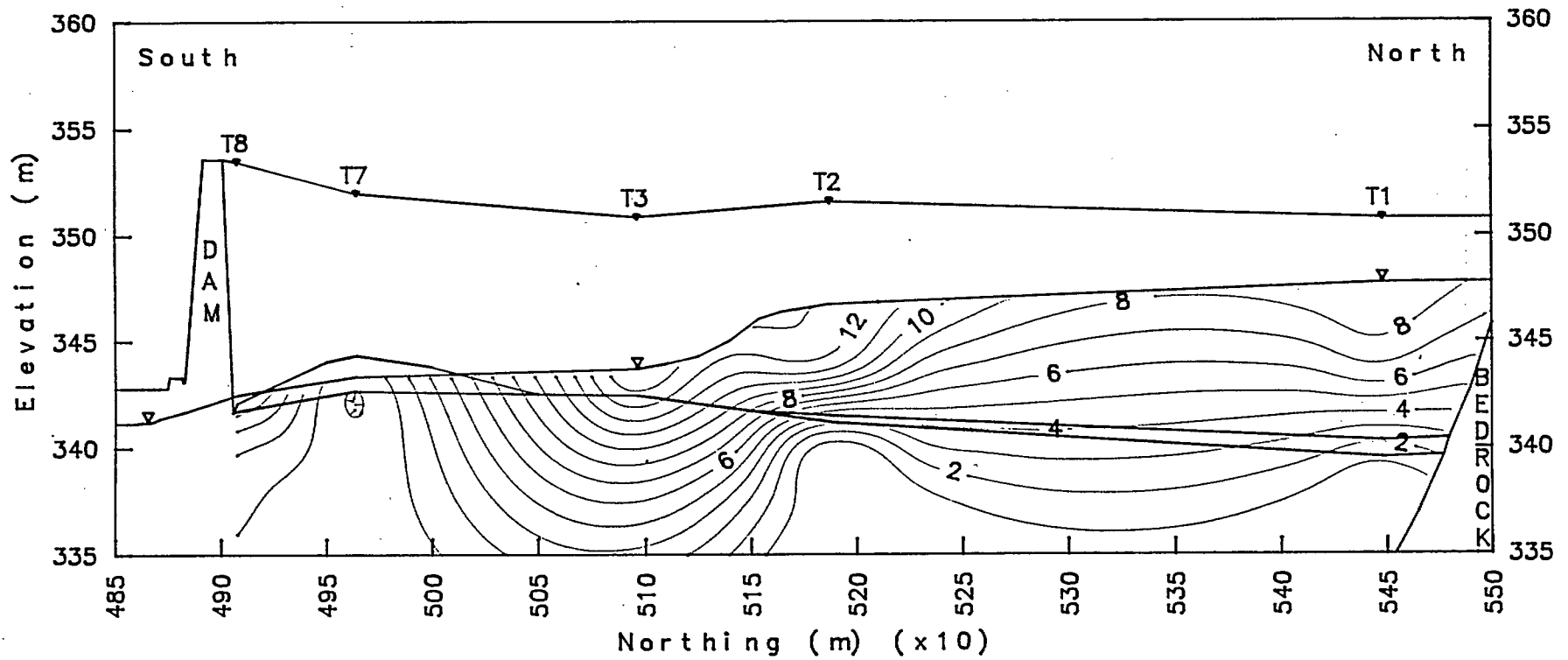


Fig. 7.62 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient (mL/g), contour map at various depths.
(Based on total sulphate in bulk mass.)

Soluble Sulphate Distribution Coefficient (mL/g)

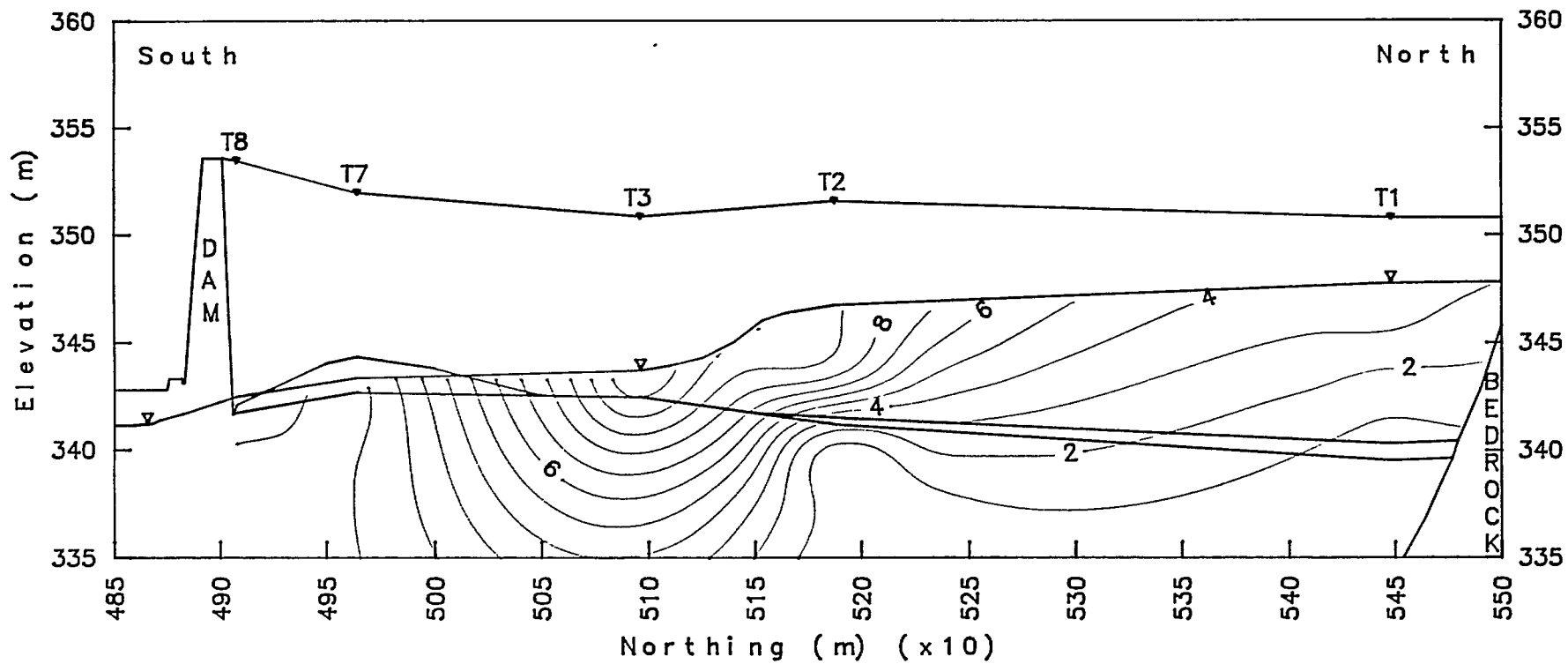


Fig. 7.63 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of soluble sulphate (mL/g), contour map at various depths. (Based on total soluble sulphate on bulk mass.)

Solid Sample Iron Content (mg/g)

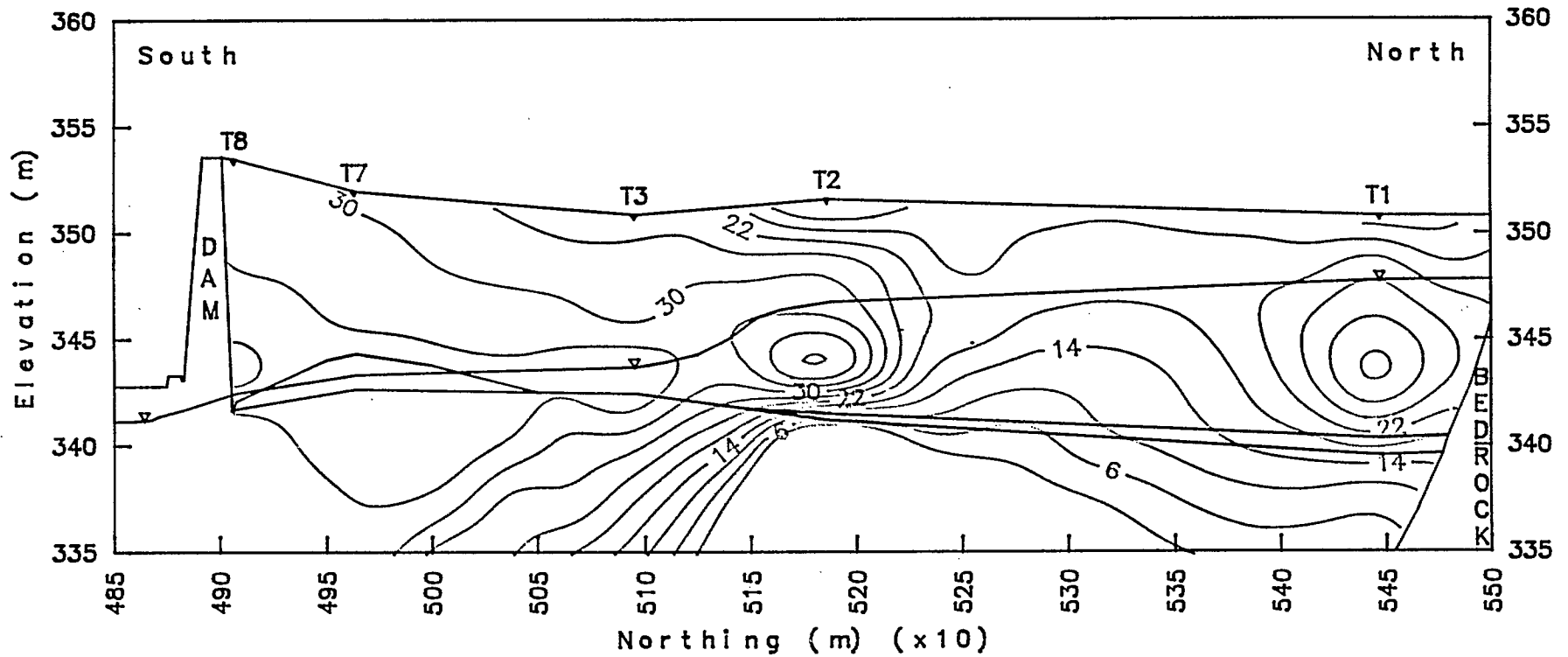


Fig. 7.64 - Nordic main tailings. Vertical north-south profile of solid sample, iron content (mg/g), contour map at various depths.

Water Sample Iron Content (mg/L)

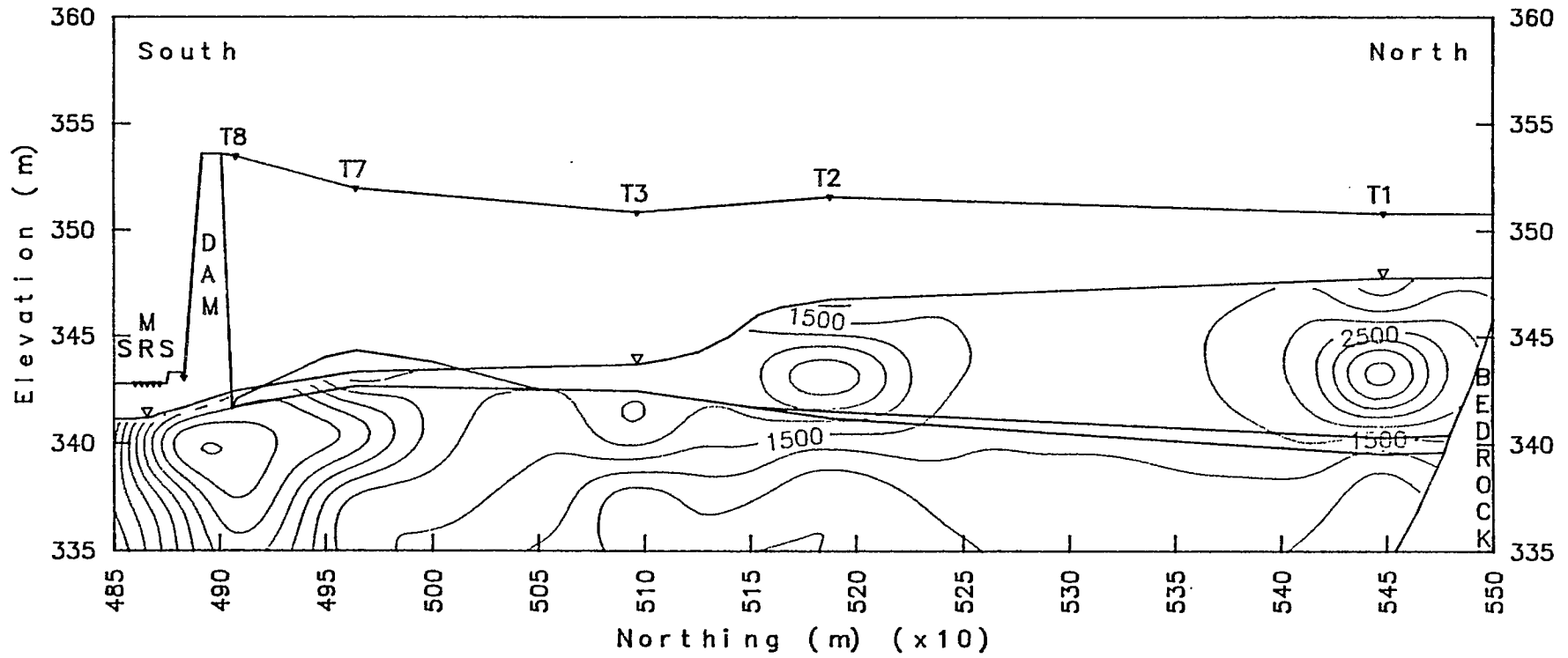


Fig. 7.65 - Nordic main tailings. Vertical north-south profile of liquid sample, iron content (mg/L), contour map at various depths.

Iron Distribution Coefficient: (mL/g)

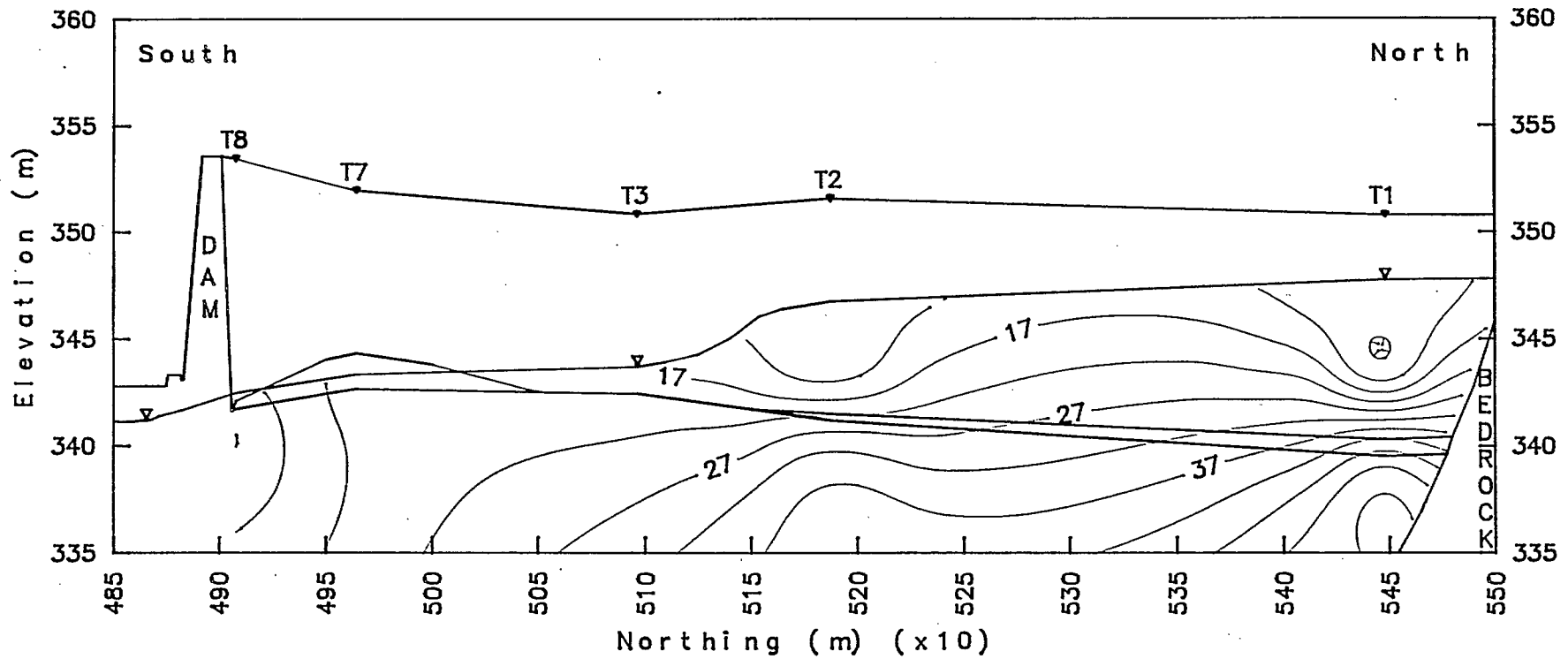


Fig. 7.66 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of iron (mL/g), contour map at various depths. (Based on iron in bulk mass.)

Solid Sample Calcium Content (mg/g)

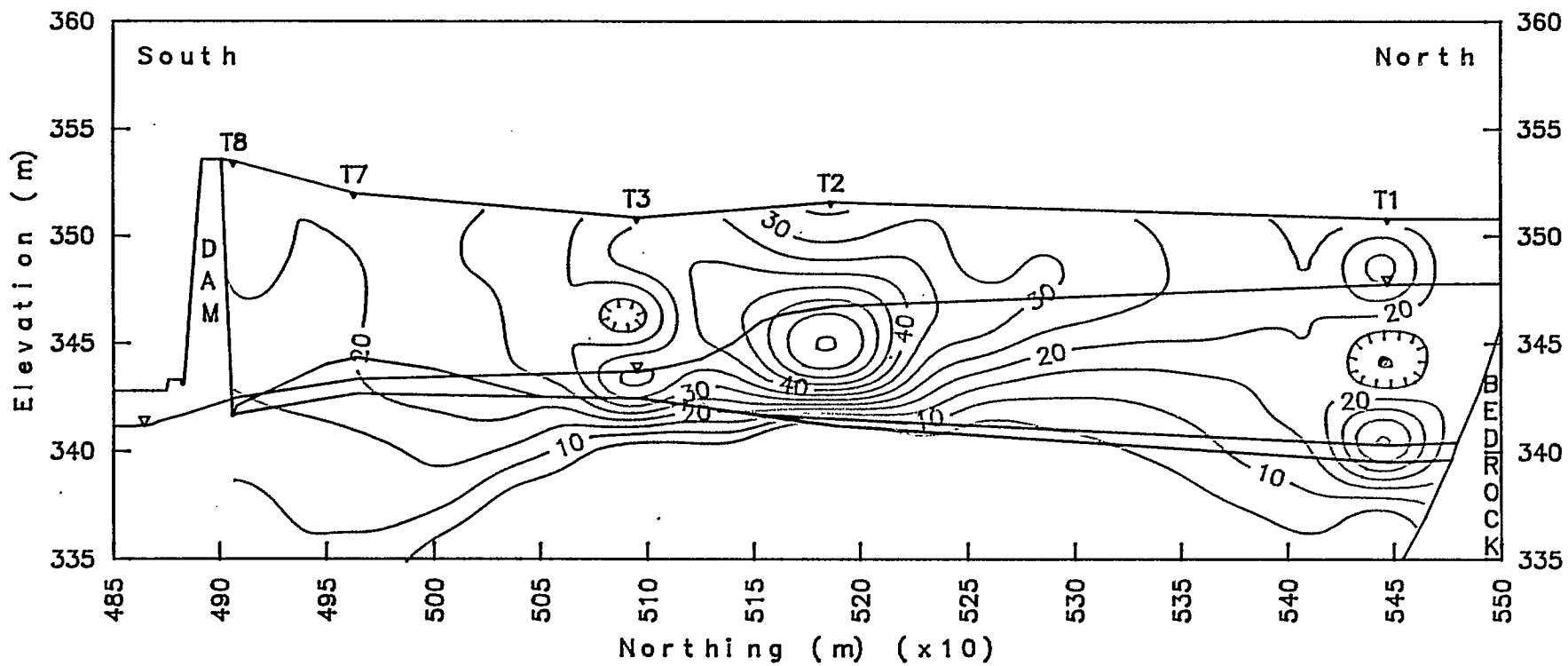


Fig. 7.67 - Nordic main tailings. Vertical north-south profile of solid sample, calcium content (mg/g), contour map at various depths.

Water Sample Calcium Content (mg/L)

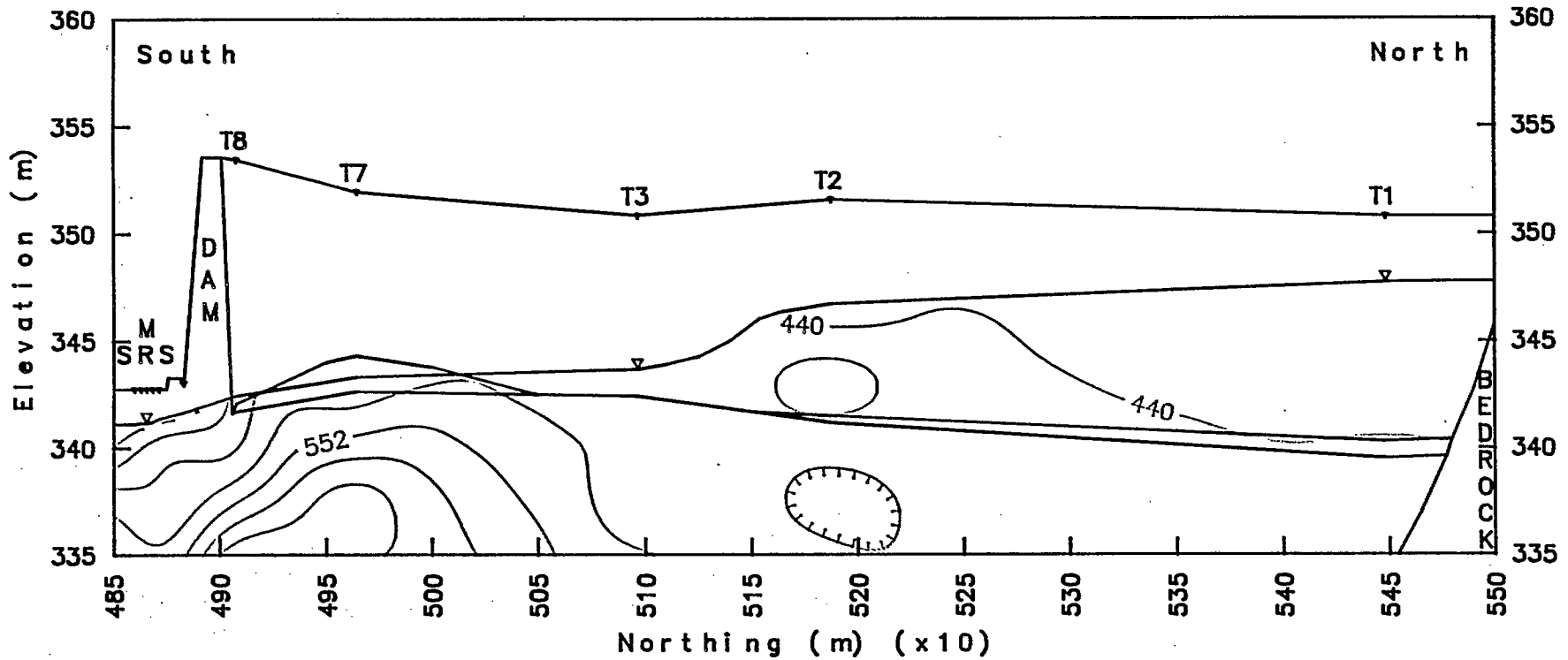


Fig. 7.68 - Nordic main tailings. Vertical north-south profile of liquid sample, calcium content (mg/L), contour map at various depths.

Calcium Distribution Coefficient (mL/g)

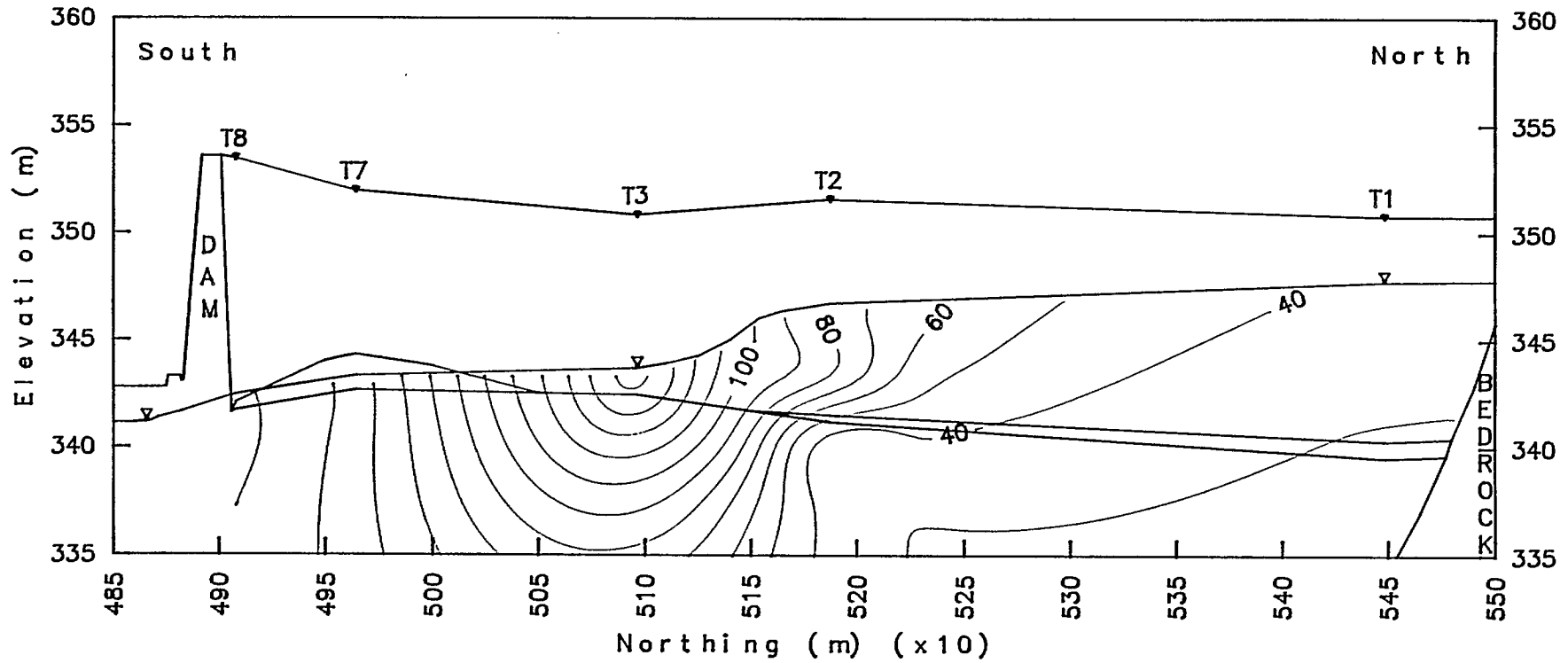


Fig. 7.69 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of calcium (mL/g), contour map at various depths. (Based on total calcium in bulk mass.)

Solid Sample Magnesium Content (mg/g)

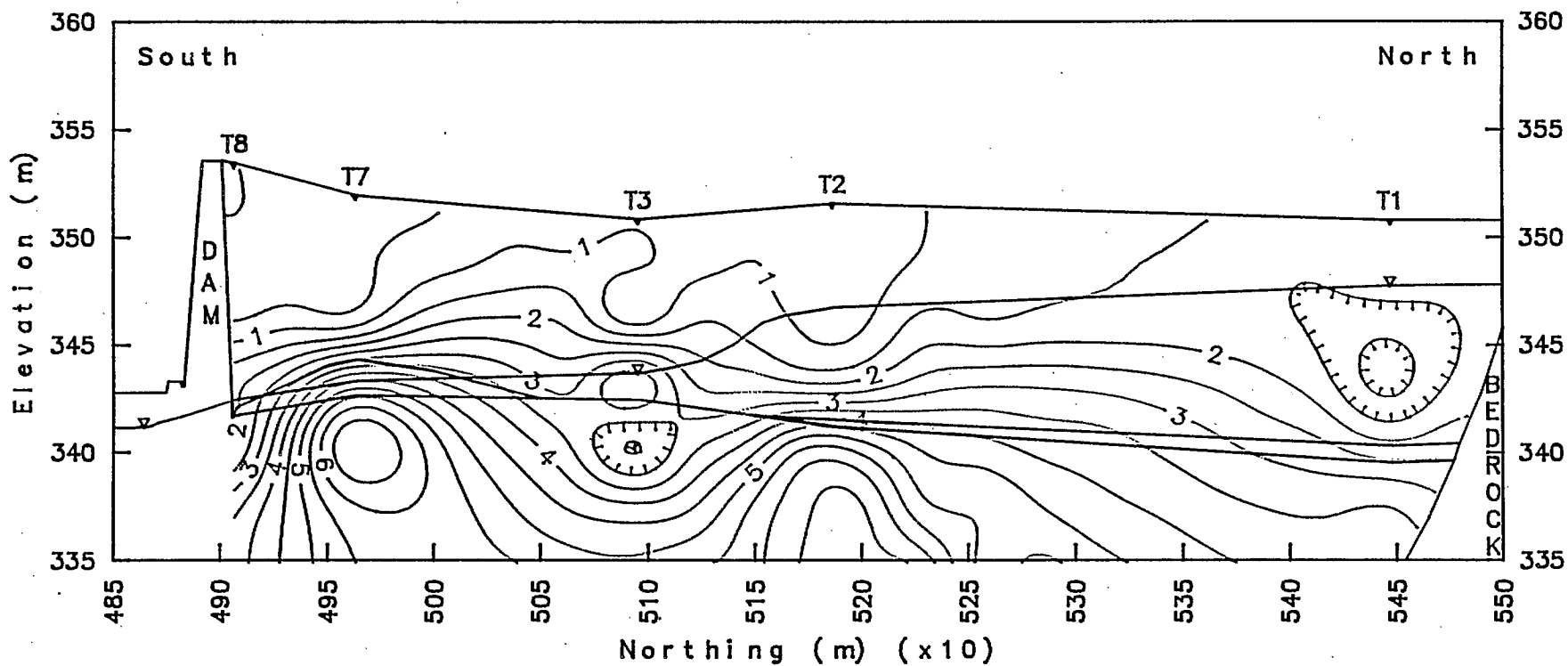


Fig. 7.70 - Nordic main tailings. Vertical north-south profile of solid sample magnesium content (mg/g), contour map at various depths.

Water Sample Magnesium Content (mg/L)

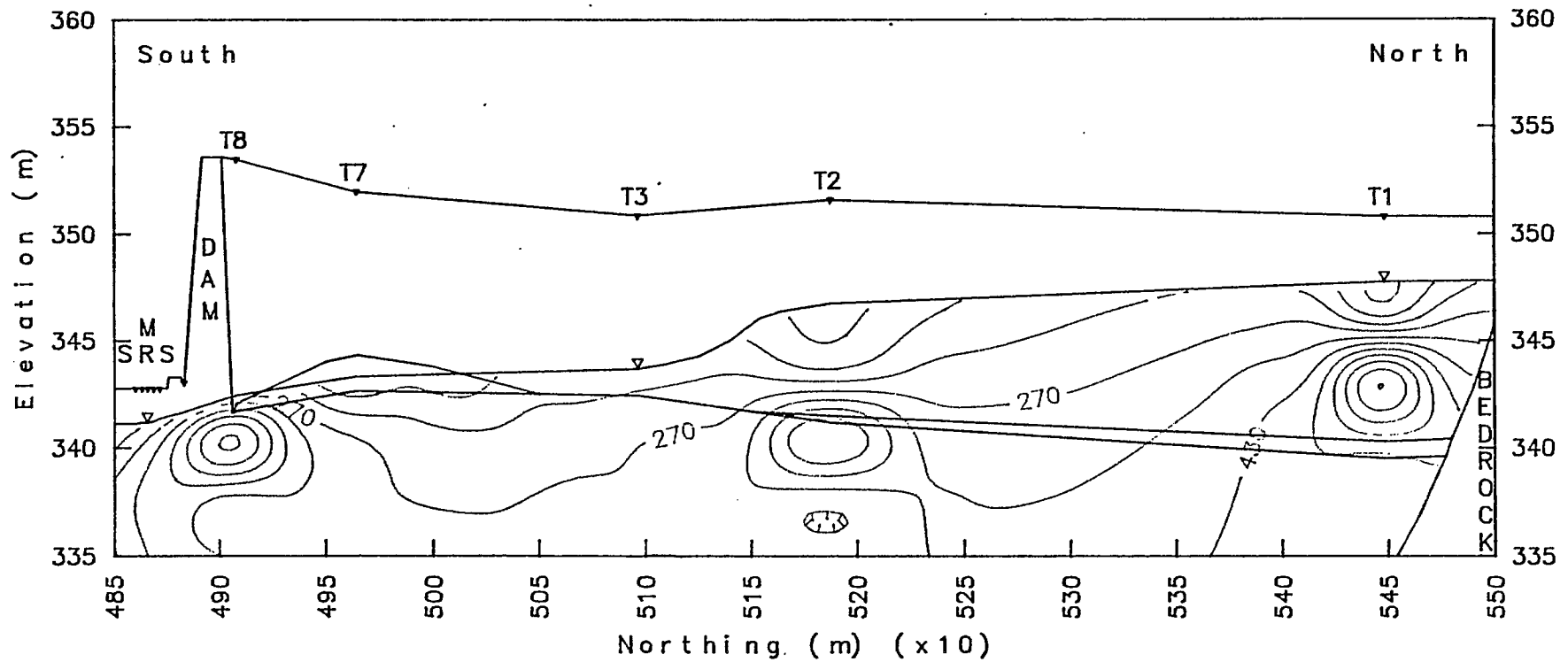


Fig. 7.71 - Nordic main tailings. Vertical north-south profile of liquid sample magnesium content (mg/L), contour map at various depths.

Magnesium Distribution Coefficient (mL/g)

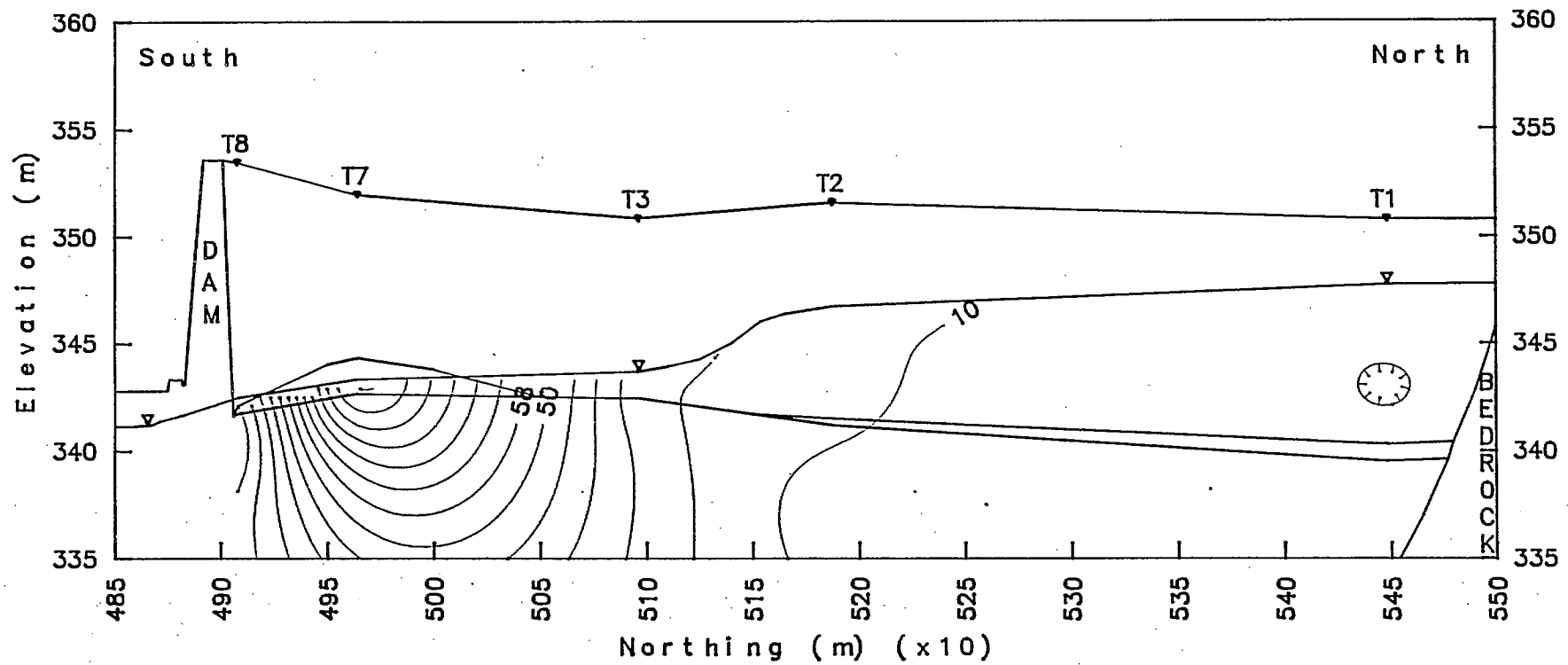


Fig. 7.72 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of magnesium (mL/g), contour map at various depths. (Based on total magnesium in bulk mass.)

Solid Sample Aluminum Content (mg/100g)

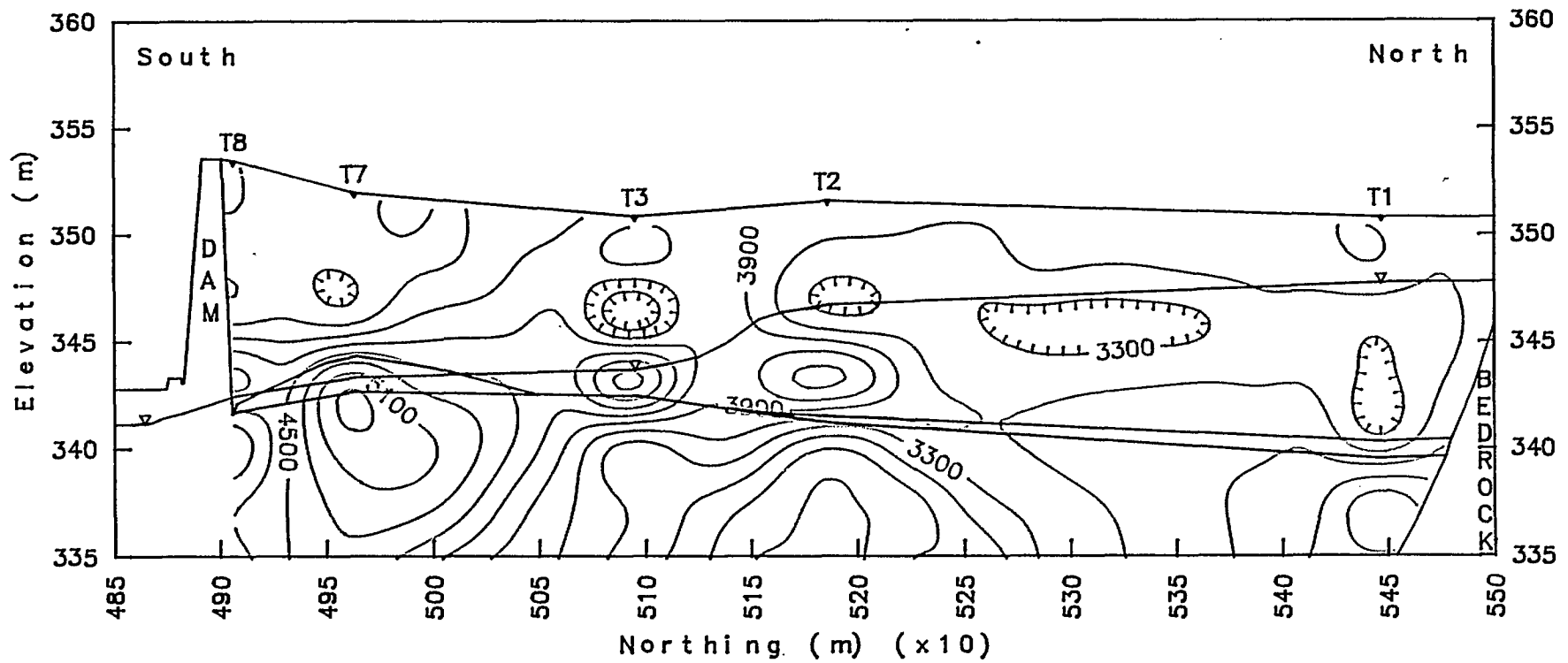


Fig. 7.73 - Nordic main tailings. Vertical north-south profile of solid sample aluminum content (mg/100 g), contour map at various depths.

Water Sample Aluminum Content (mg/L)

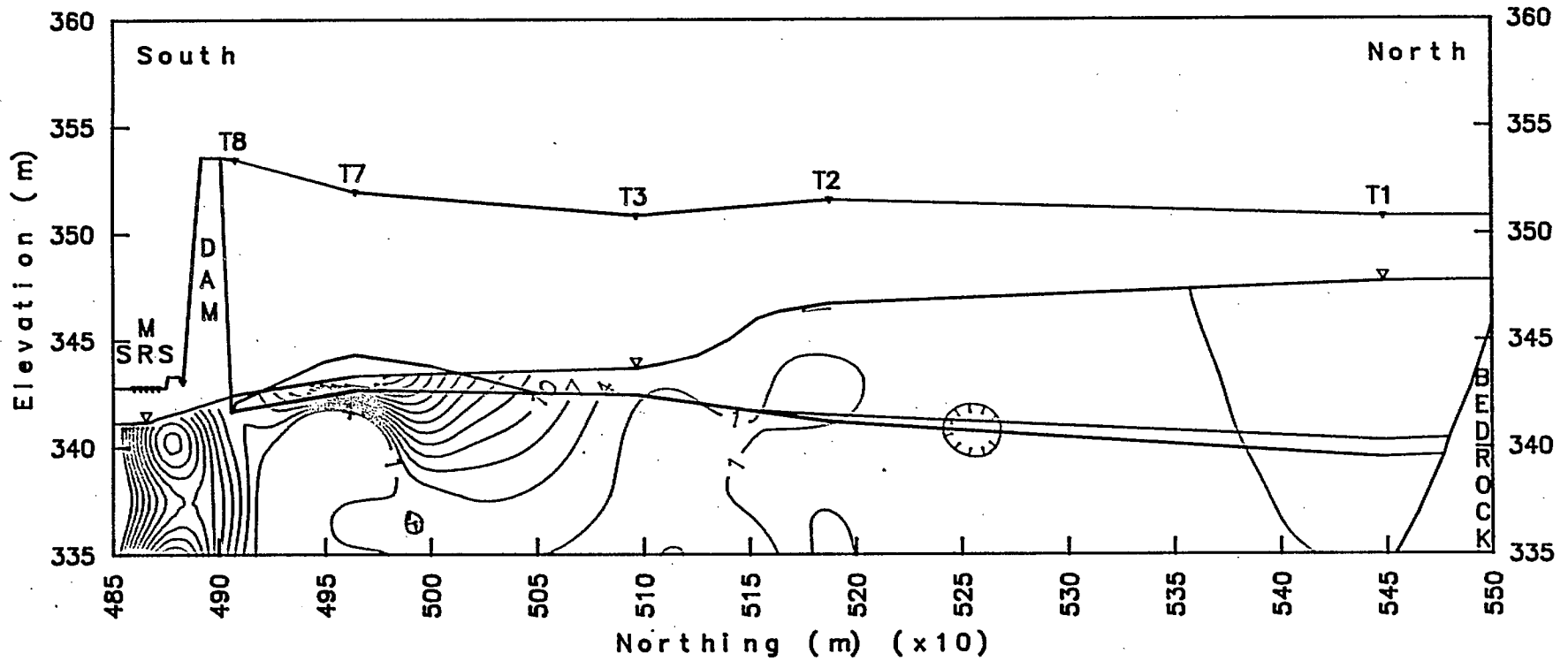


Fig. 7.74 - Nordic main tailings. Vertical north-south profile of liquid sample aluminum content (mg/L), contour map at various depths.

Aluminum Distribution Coefficient (mL/g)

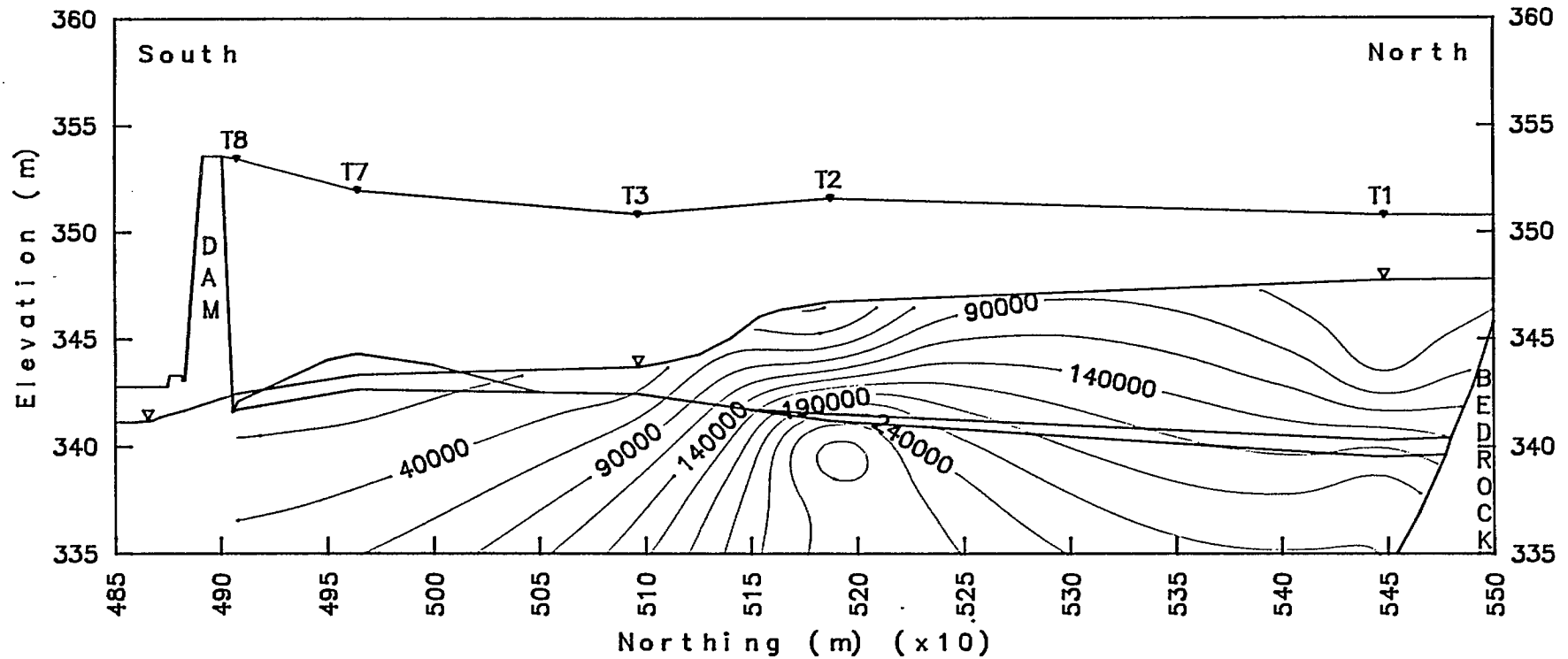


Fig. 7.75 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of aluminum (mL/g), contour map at various depths. (Based on aluminum in bulk mass.)

Solid Sample Thorium Content (ppm)

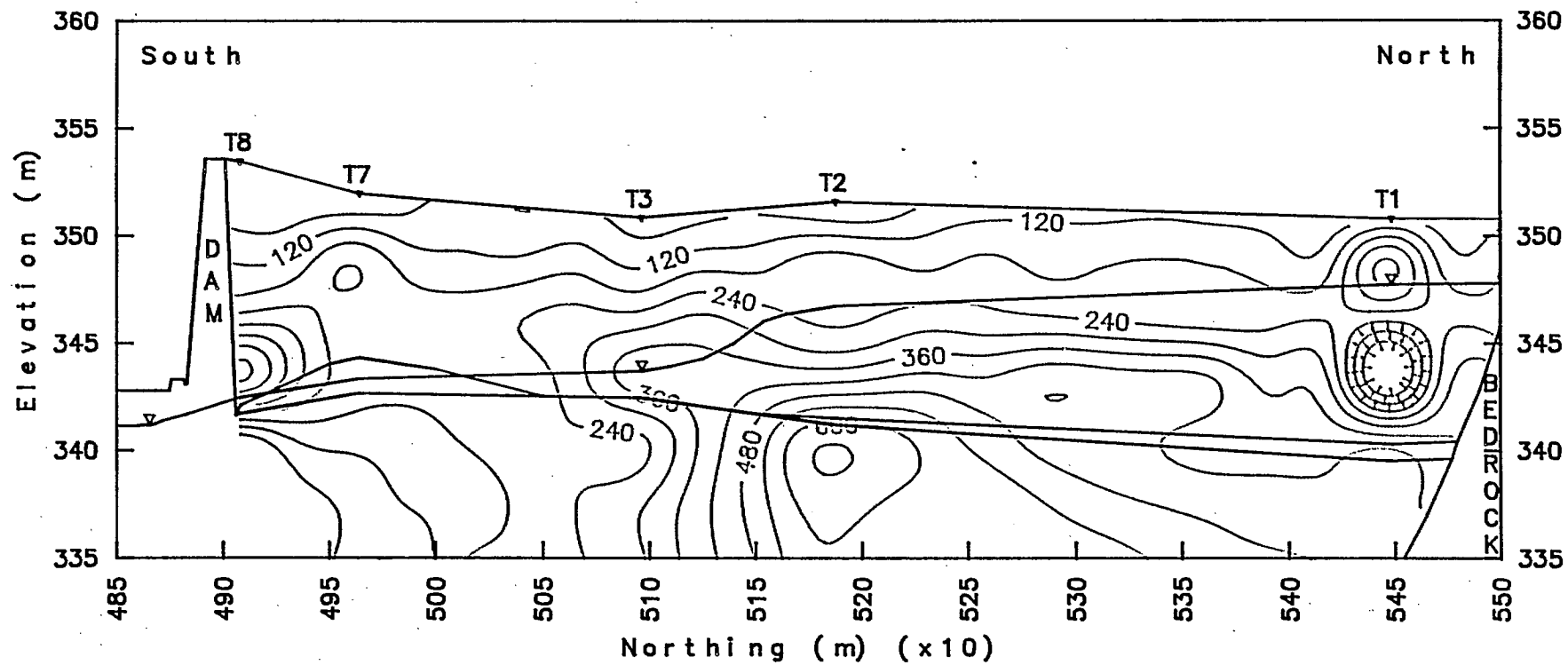


Fig. 7.76 - Nordic main tailings. Vertical north-south profile of solid sample thorium content (ppm), contour map at various depths.

Water Sample Thorium Content (mg/L)

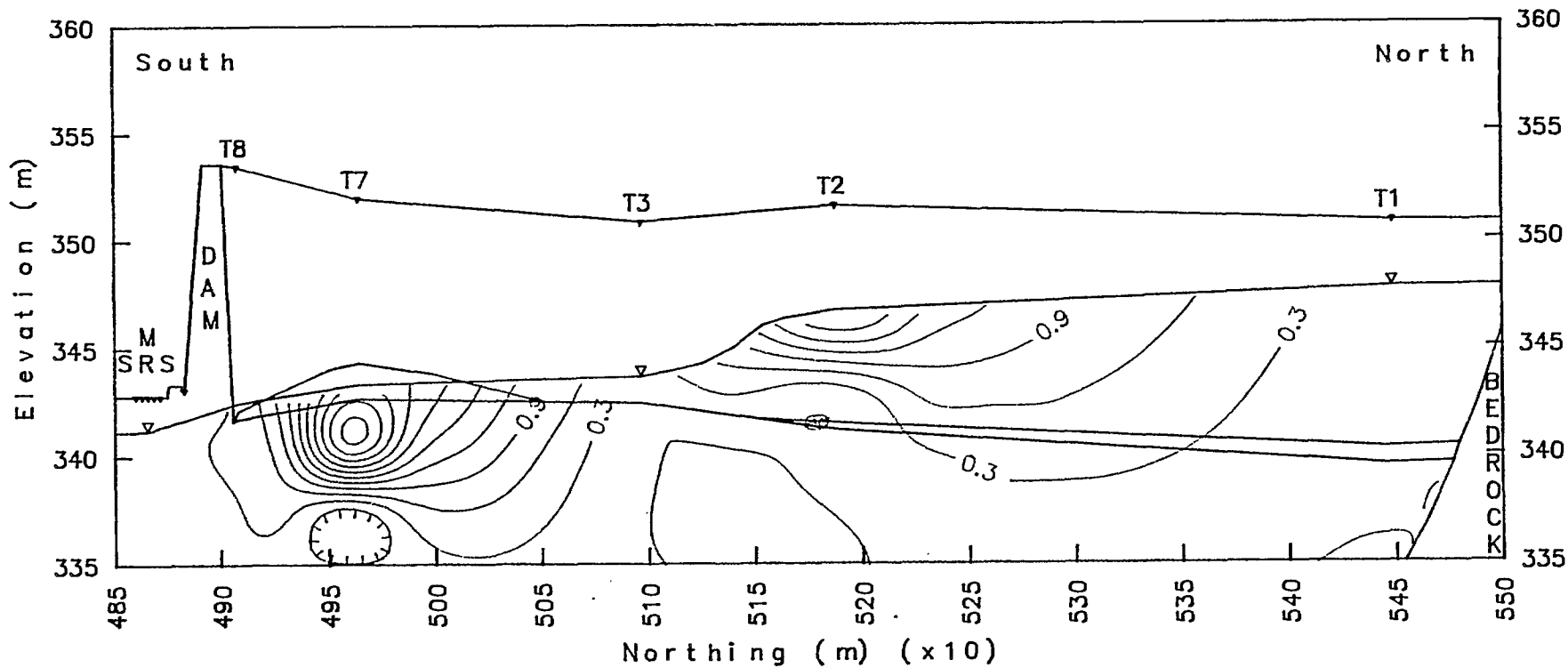


Fig. 7.77 - Nordic main tailings. Vertical north-south profile of liquid sample thorium content (mg/L), contour map at various depths.

Thorium Distribution Coefficient (mL/g)

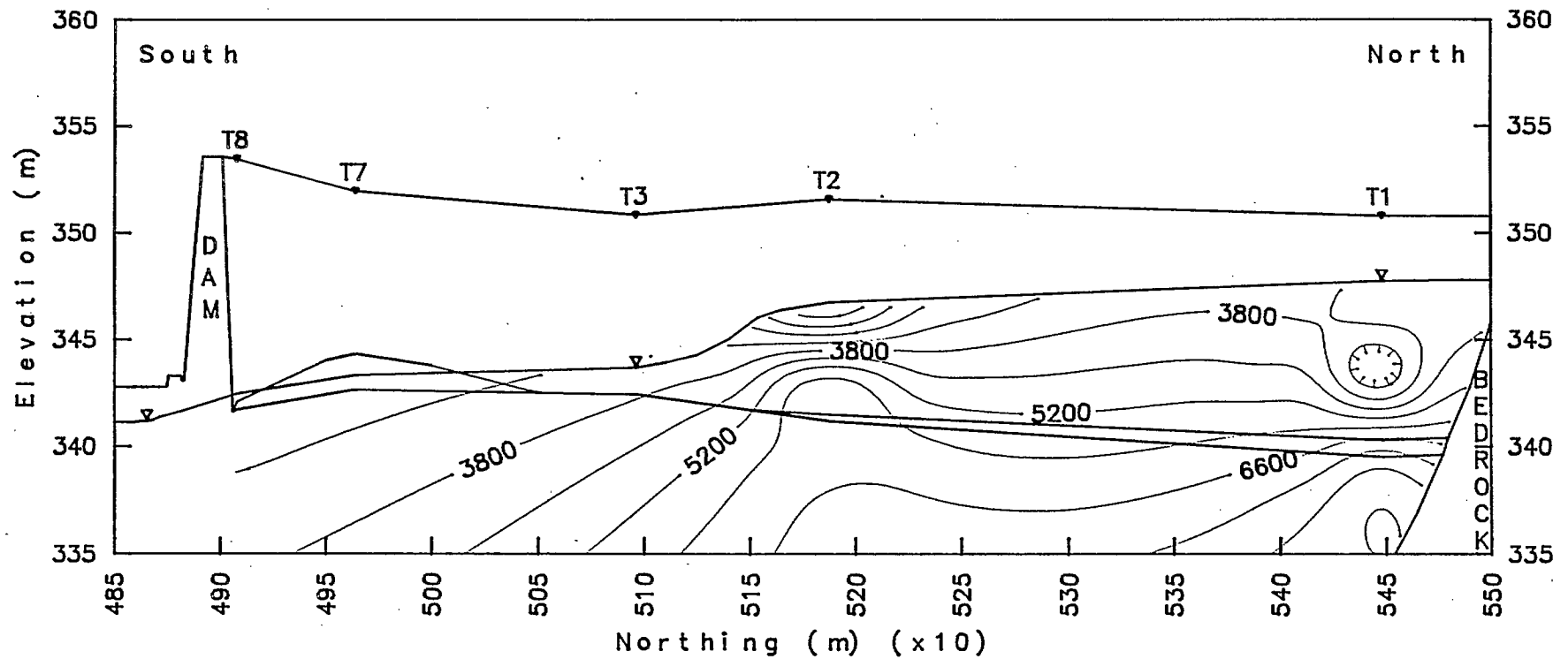


Fig. 7.78 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of thorium (mL/g), contour map at various depths. (Based on thorium in bulk mass.)

Solid Sample Uranium Content (ppm)

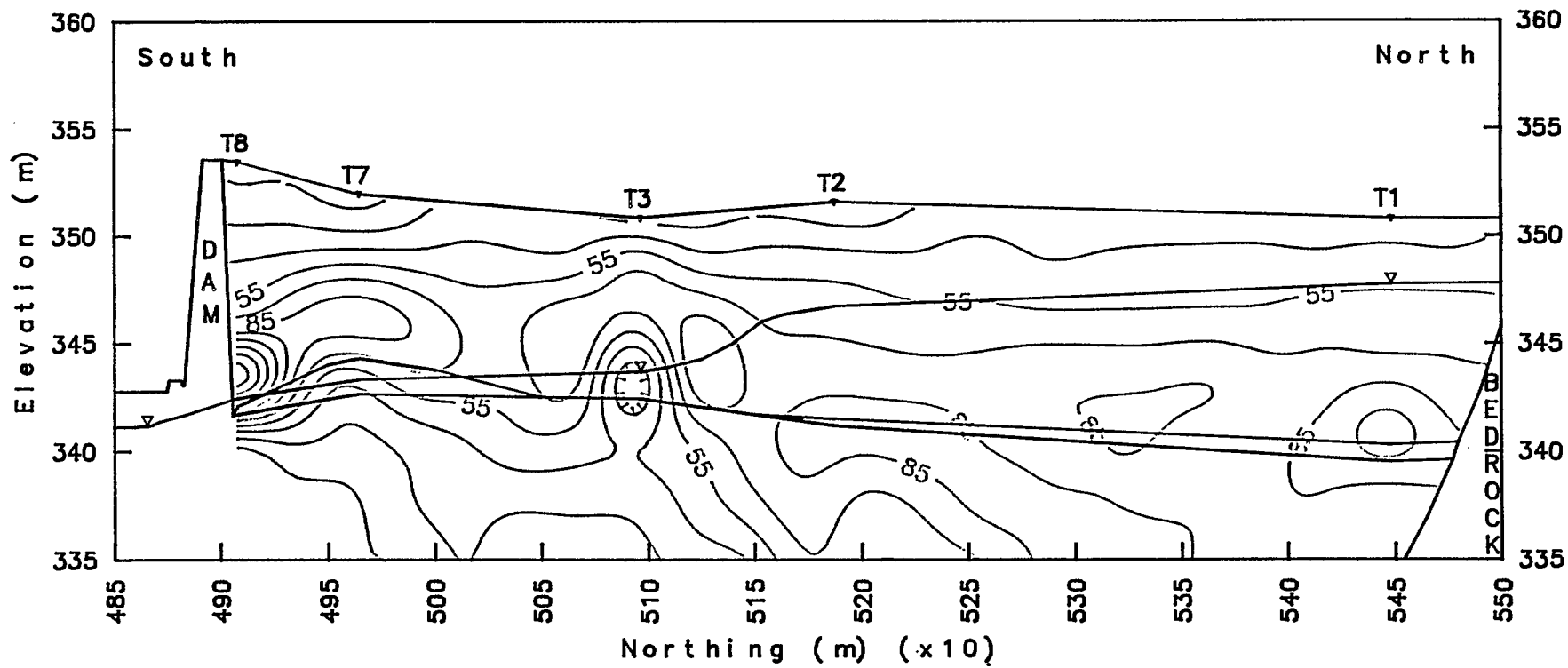


Fig. 7.79 - Nordic main tailings. Vertical north-south profile of solid sample uranium content (ppm), contour map at various depths.

Water Sample Uranium Content (mg/L)

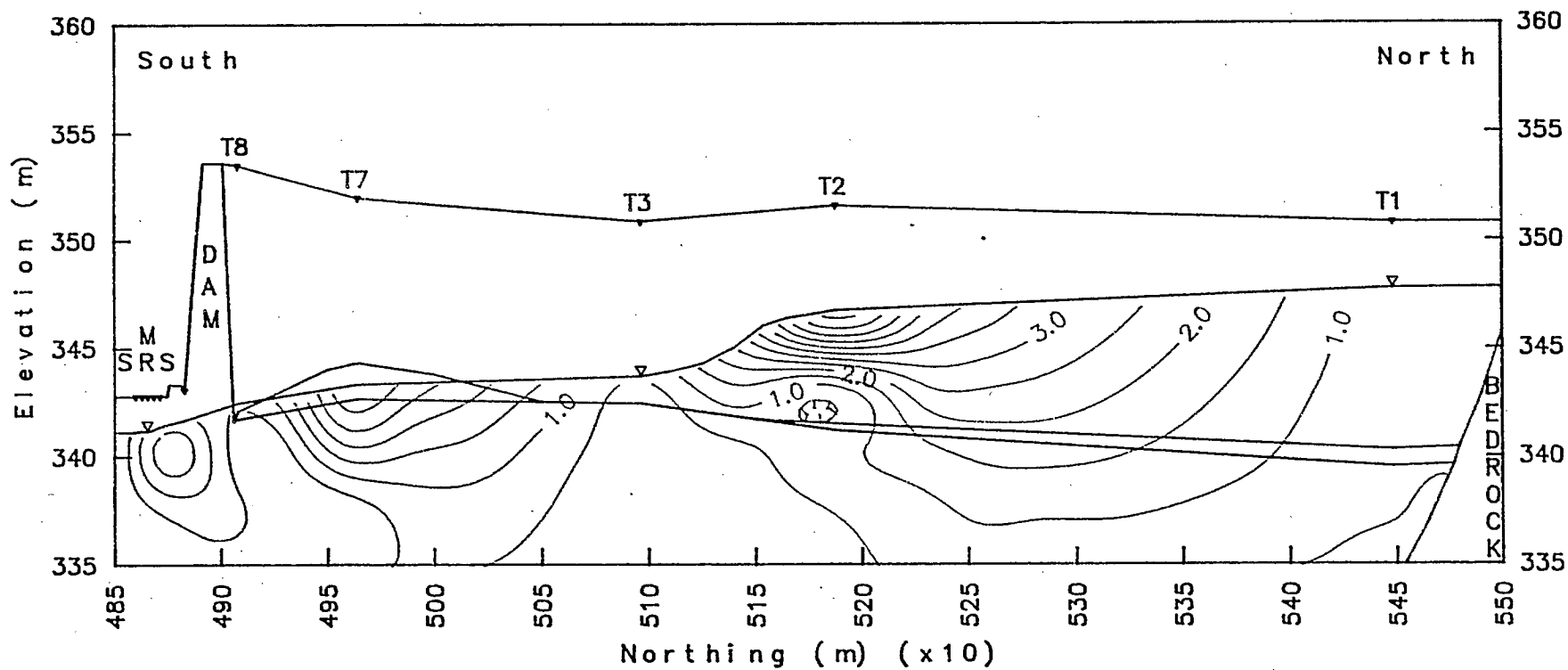


Fig. 7.80 - Nordic main tailings. Vertical north-south profile of liquid sample uranium content (mg/L), contour map at various depths.

Uranium Distribution Coefficient (mL/g)

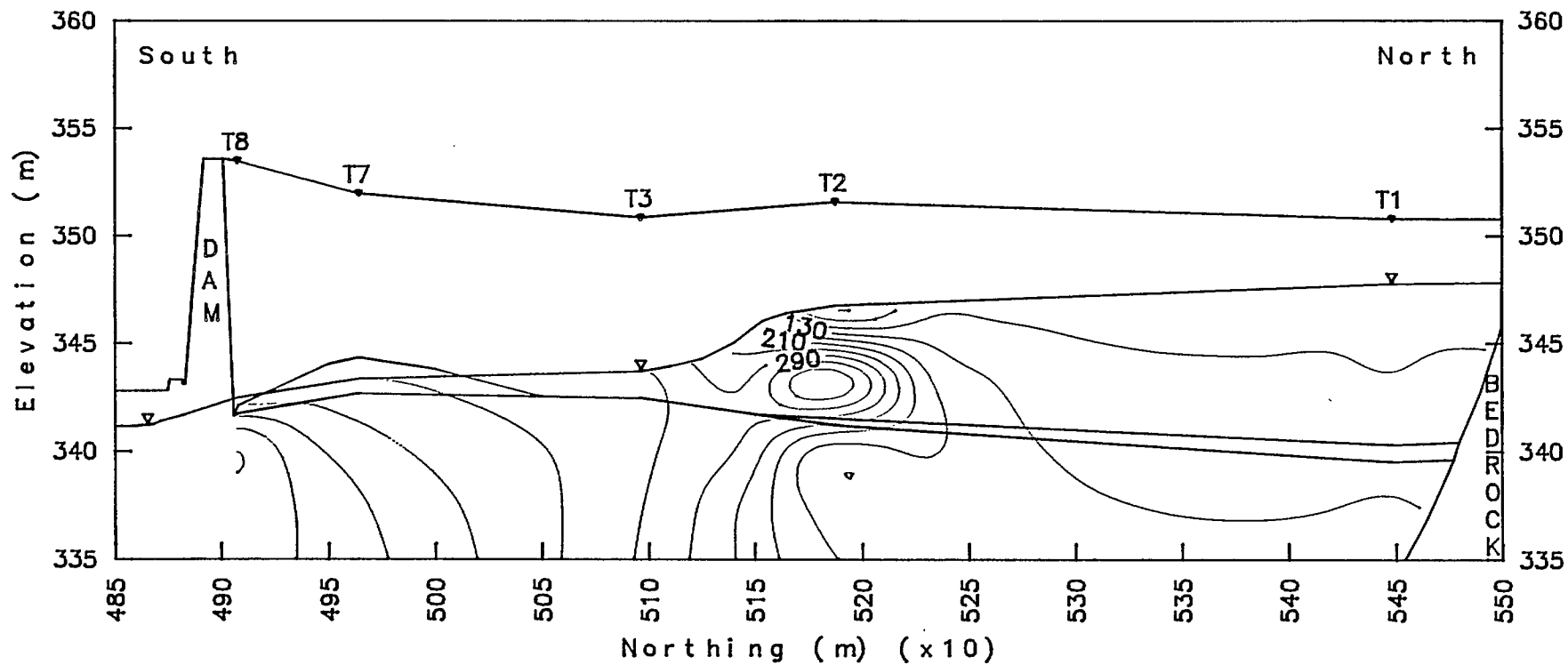


Fig. 7.81 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of uranium (mL/g), contour map at various depths.
(Based on uranium in bulk mass.)

Solid Sample Lead Content (mg/100g)

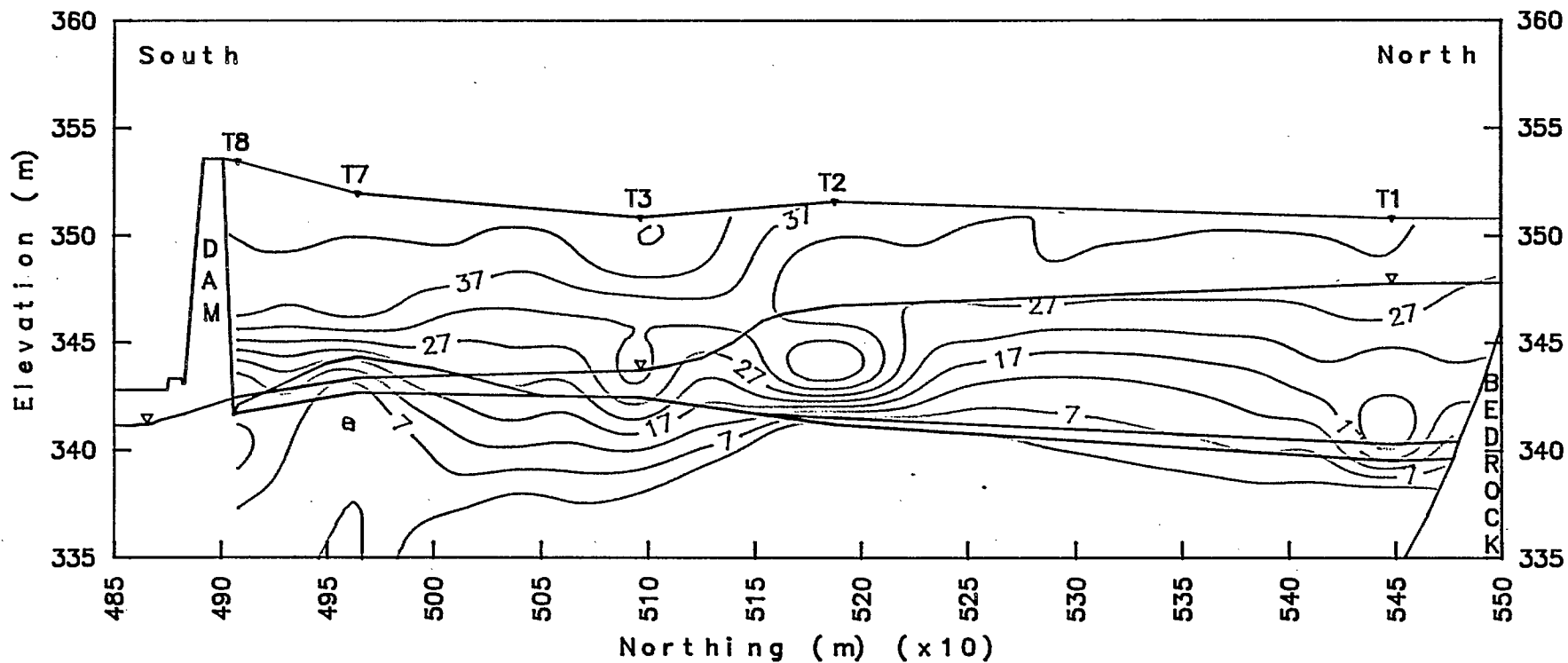


Fig. 7.82 - Nordic main tailings. Vertical north-south profile of solid sample lead content (mg/100 g), contour map at various depths.

Solid Sample Pb-210 Content (mBq/g) ($\pm 10\%$)

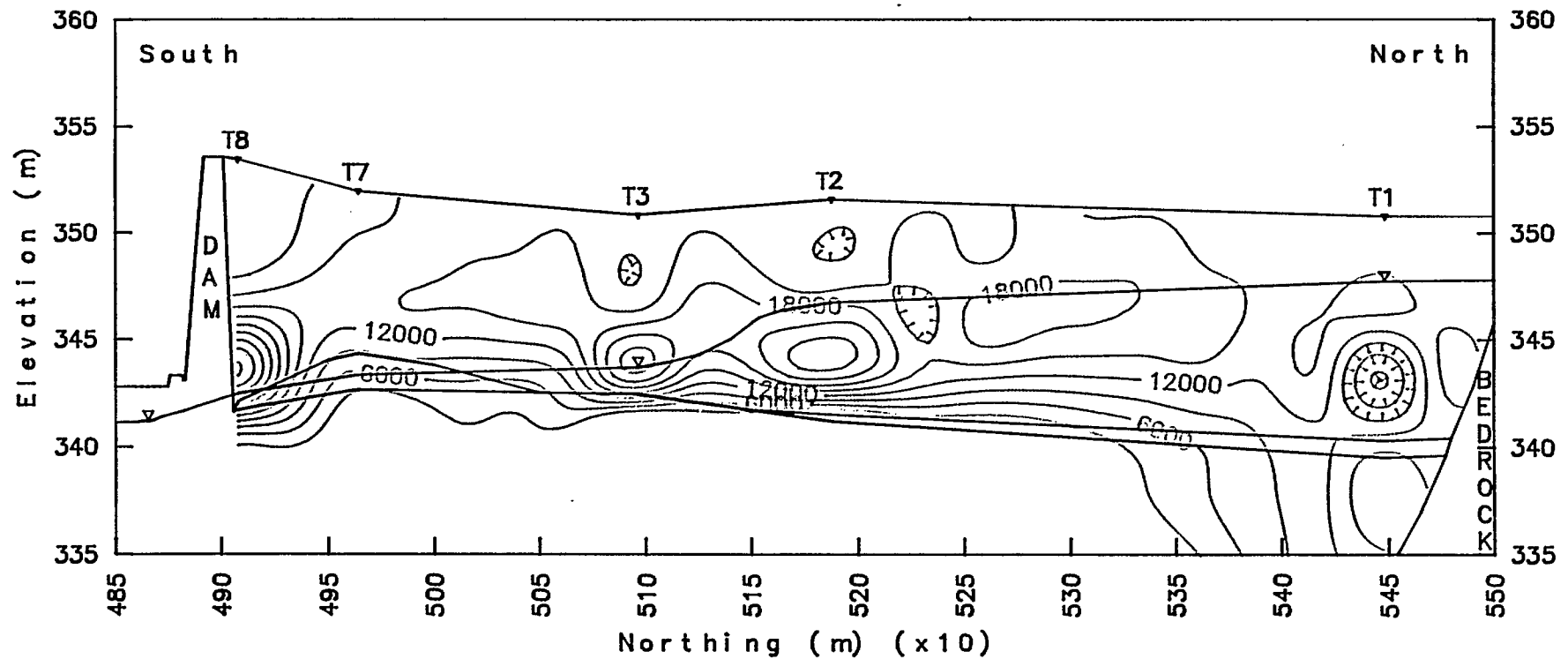


Fig. 7.83 - Nordic main tailings. Vertical north-south profile of solid sample ^{210}Pb content (mBq/g) ($\pm 10\%$), contour map at various depths.

Solid Sample Ra-226 Content (mBq/g) ($\pm 10\%$)

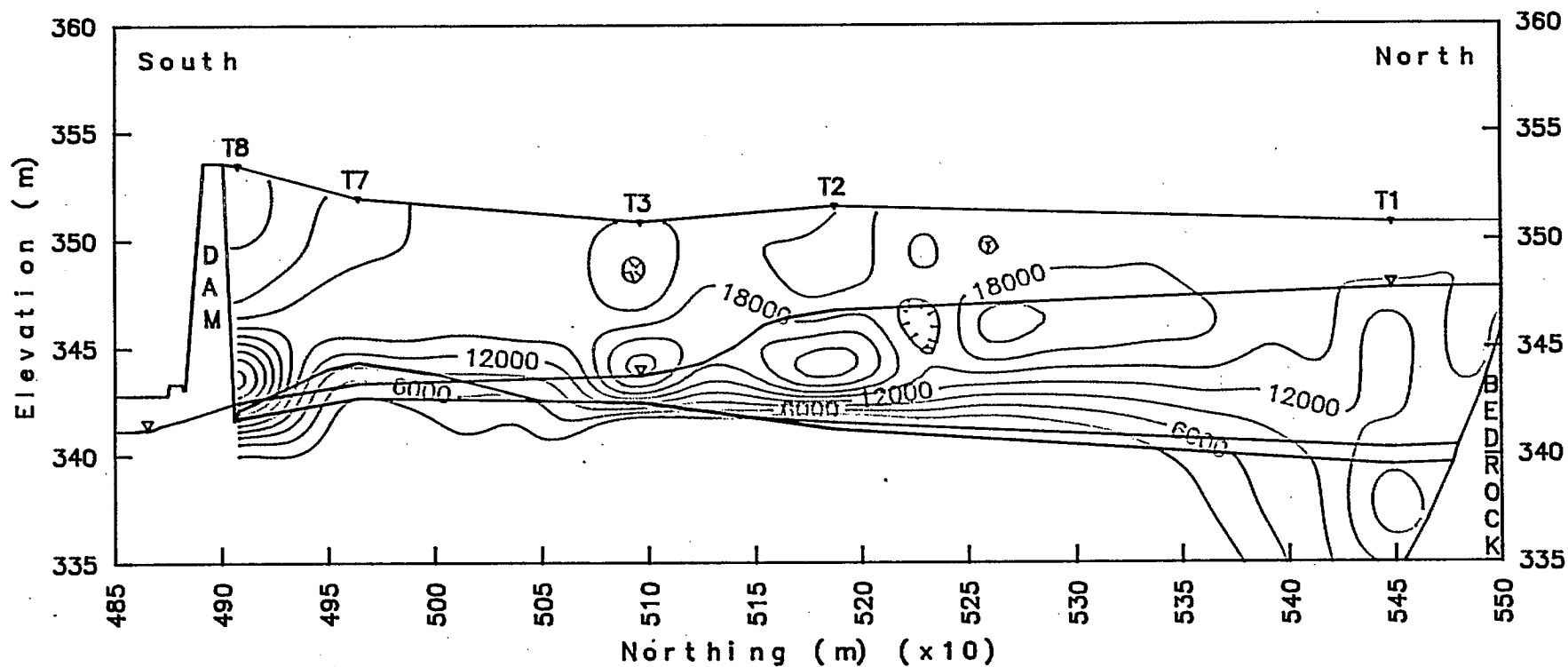


Fig. 7.84 - Nordic main tailings. Vertical north-south profile of solid sample ^{226}Ra content (mBq/g) ($\pm 10\%$), contour map at various depths.

Water Sample Ra-226 Content (mBq/L)

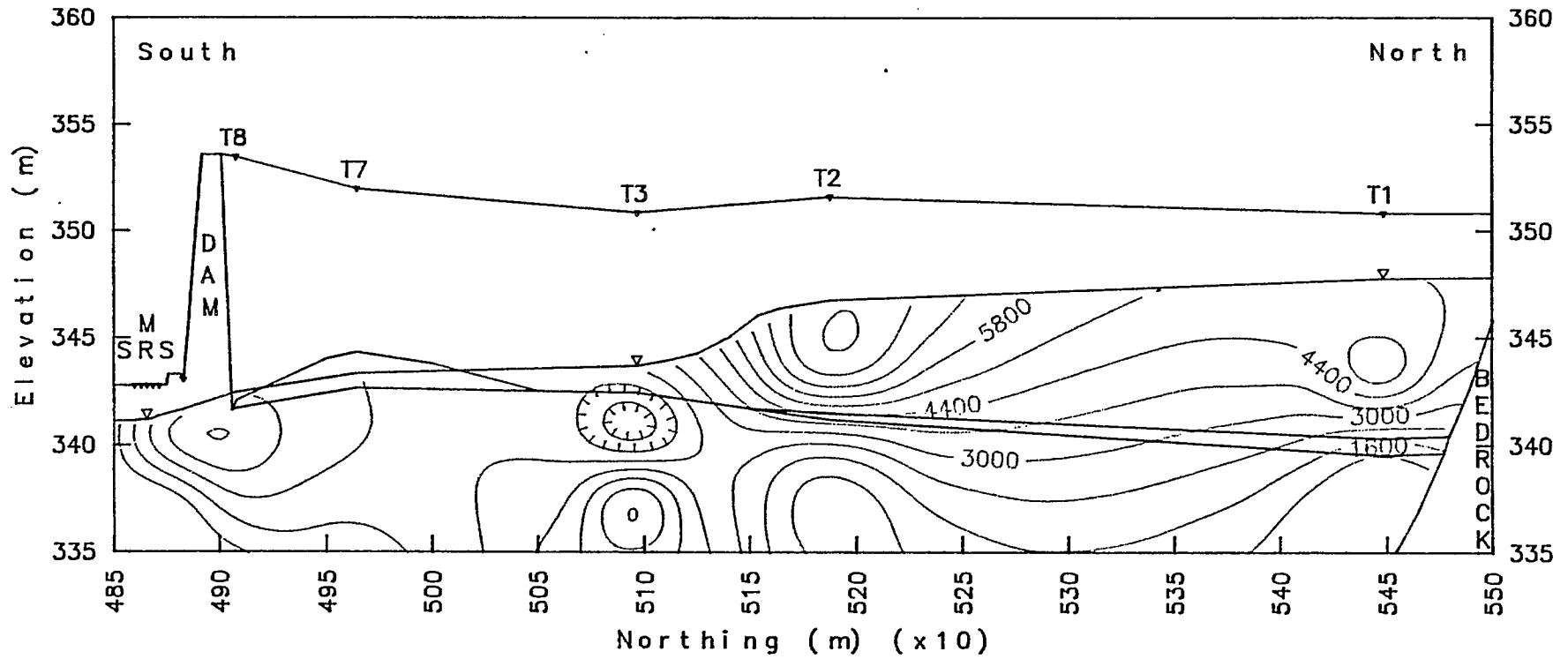


Fig. 7.85 - Nordic main tailings. Vertical north-south profile of liquid sample ^{226}Ra content (mBq/L), contour map at various depths.

Ra-226 Distribution Coefficient (mL/g)

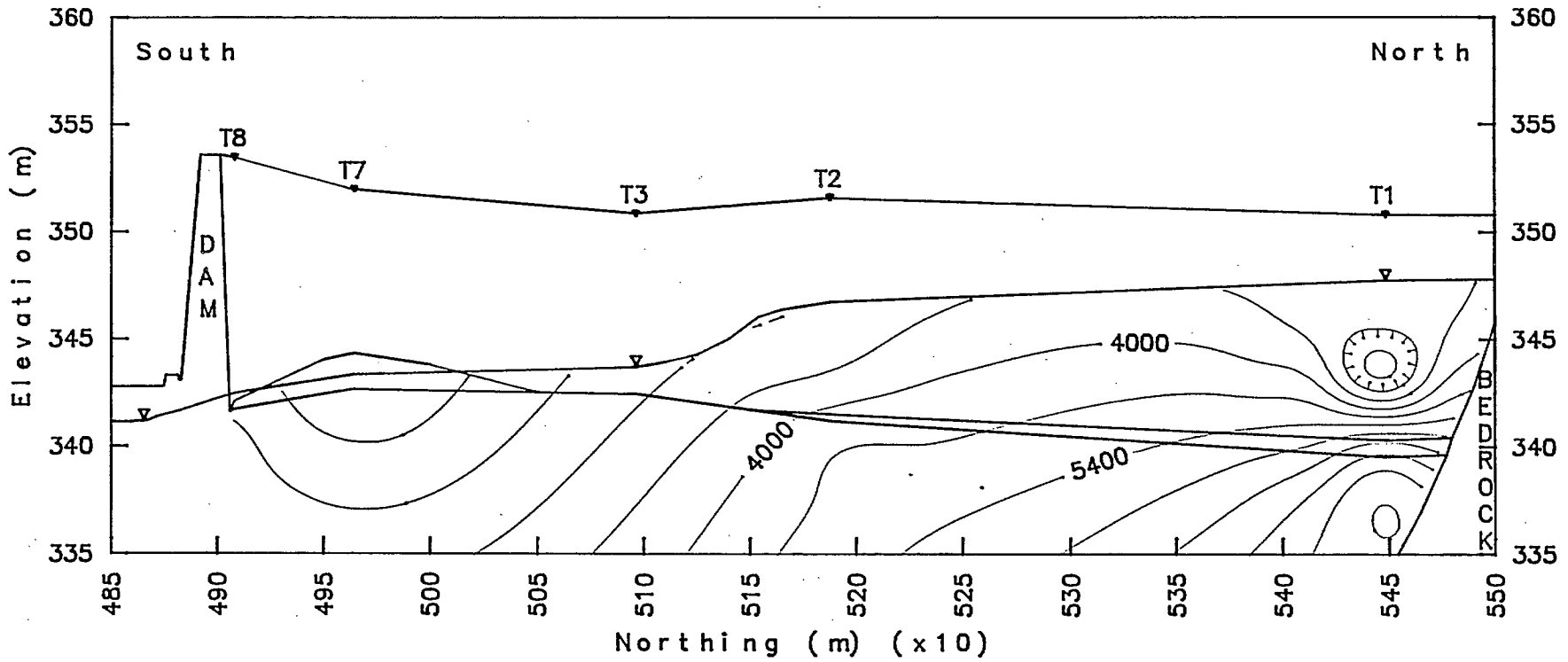


Fig. 7.86 - Nordic main tailings. Vertical north-south profile solid/liquid distribution coefficient of ^{226}Ra (mL/g), contour map at various depths.
(Based on ^{226}Ra in bulk mass.)

Solid Sample Th-228 Content (mBq/g) ($\pm 20\%$)

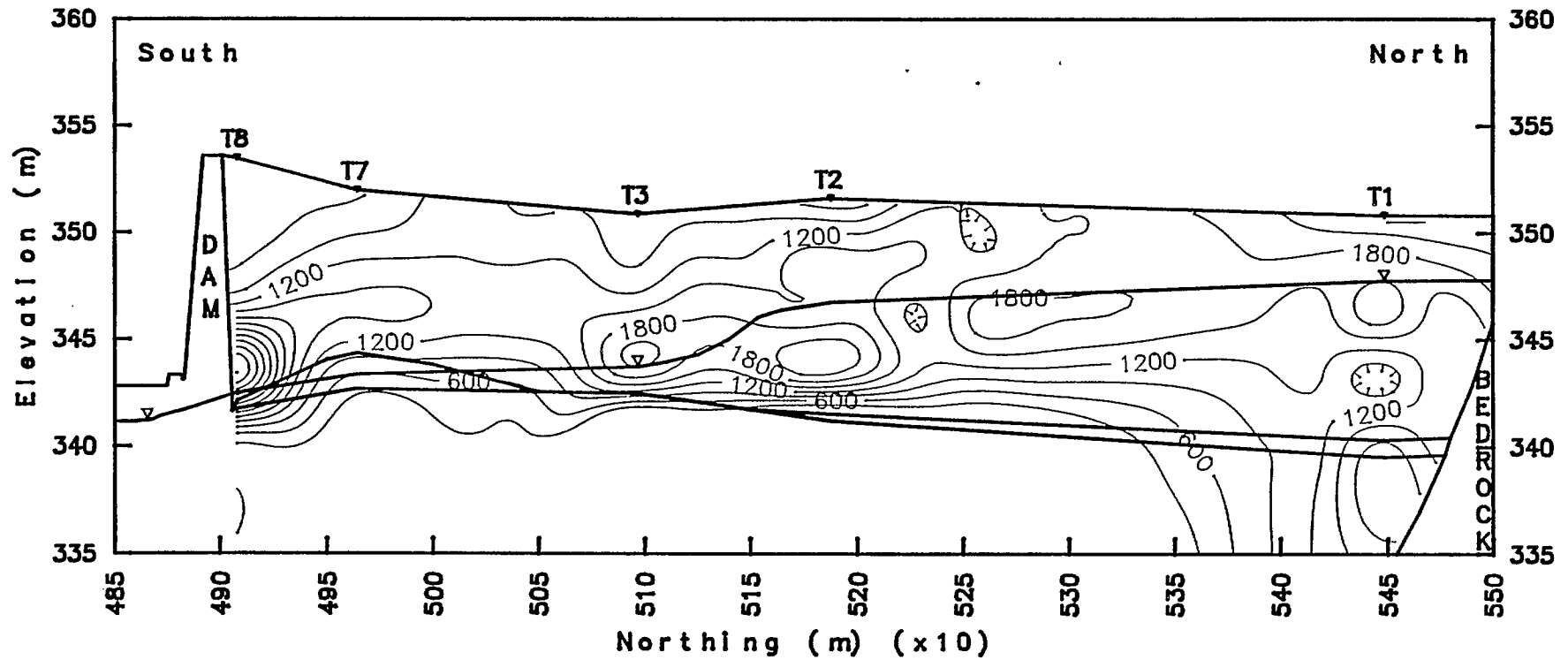


Fig. 7.87 - Nordic main tailings. Vertical north-south profile of solid sample ^{228}Th content (mBq/g) ($\pm 20\%$), contour map at various depths.

Solid Sample Th-230 Content (mBq/g) ($\pm 20\%$)

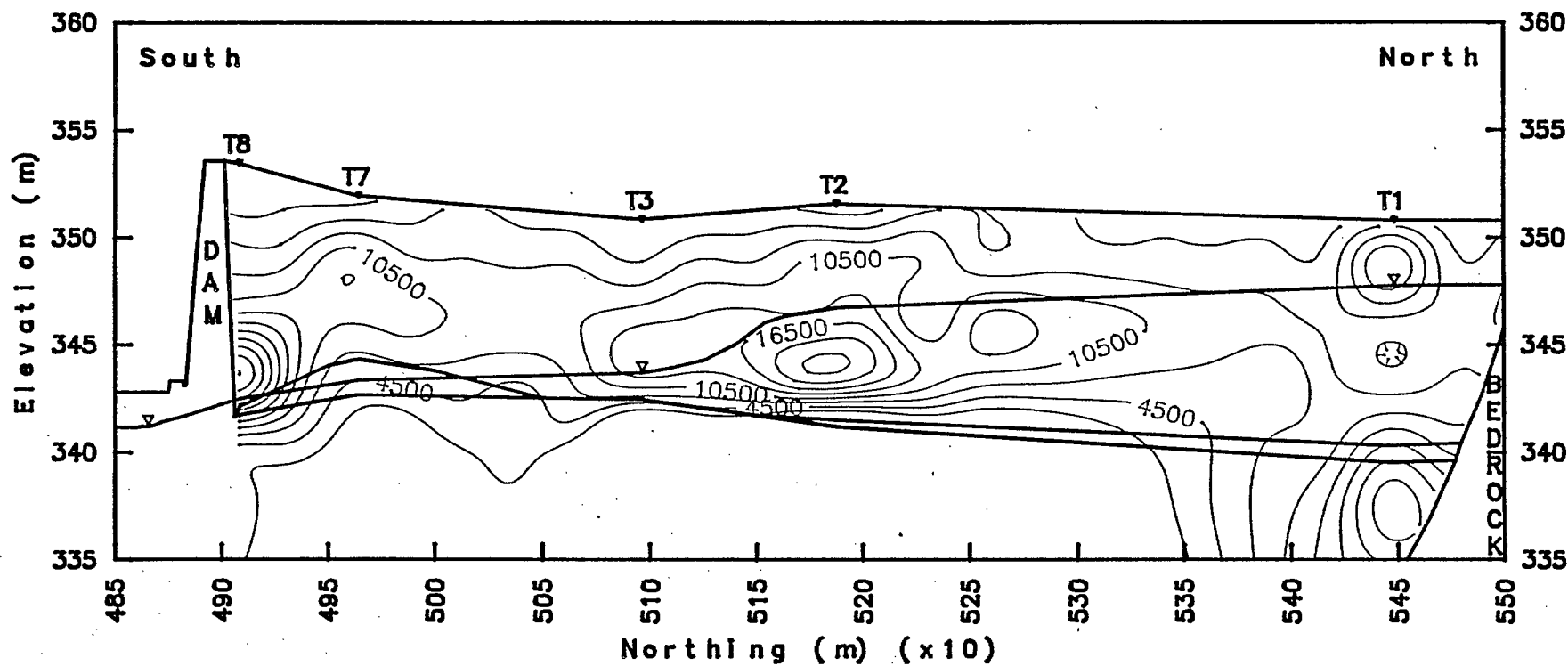


Fig. 7.88 - Nordic main tailings. Vertical north-south profile of solid sample ^{230}Th content (mBq/g) ($\pm 20\%$), contour map at various depths.

Solid Sample Th-232 Content (mBq/g) ($\pm 20\%$)

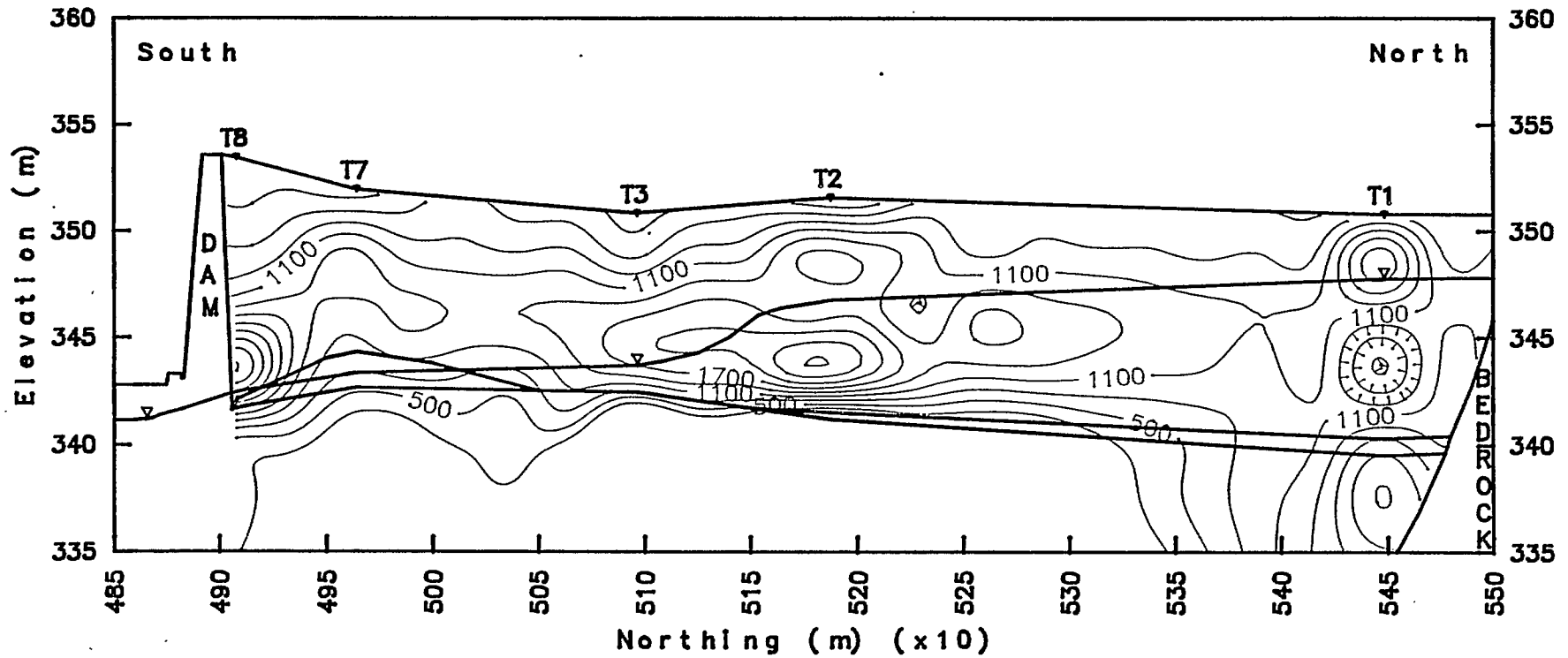


Fig. 7.89 - Nordic main tailings. Vertical north-south profile of solid sample ^{232}Th content (mBq/g) ($\pm 20\%$), contour map at various depths.

Ion Balance Error in Water Samples (%)

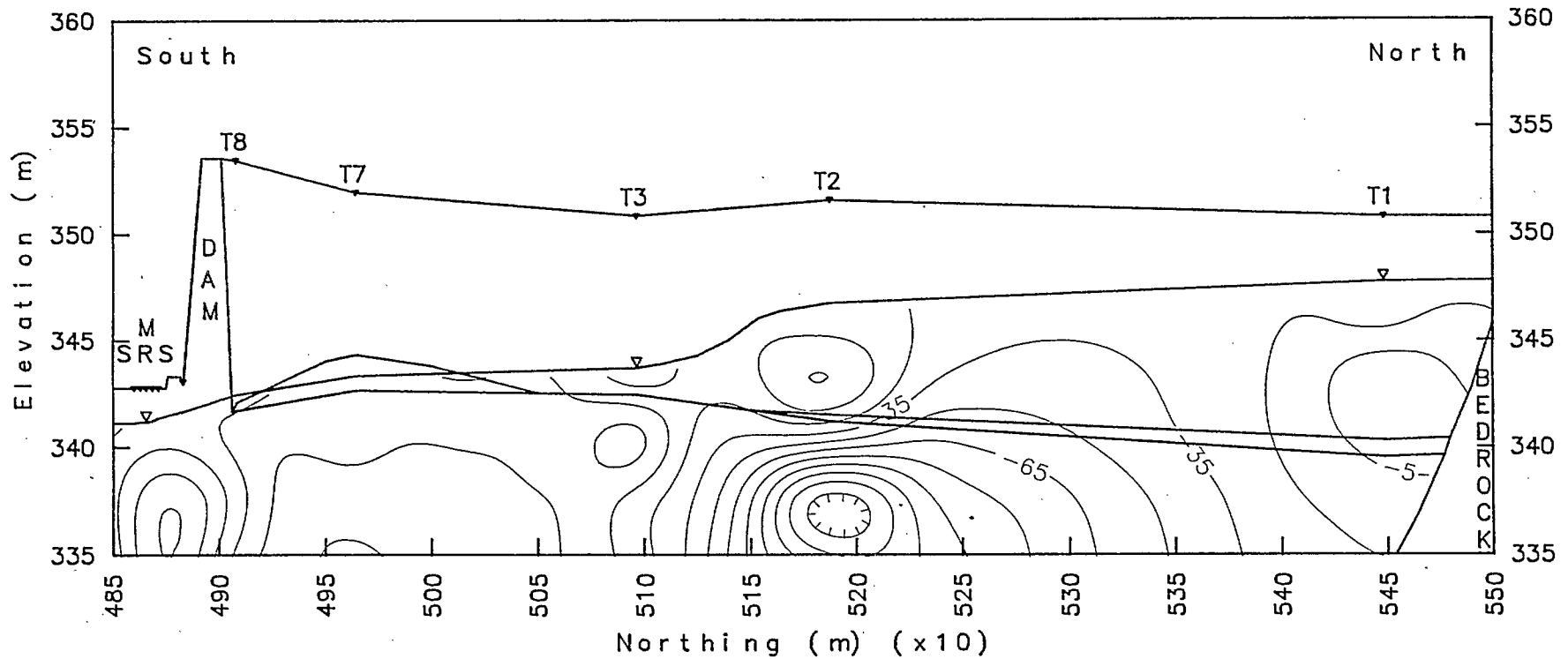


Fig. 7.90 - Nordic main tailings. Vertical north-south profile of ion balance error in water samples (%), contour map at various depths.

Activity Ratios Pb-210/Ra-226

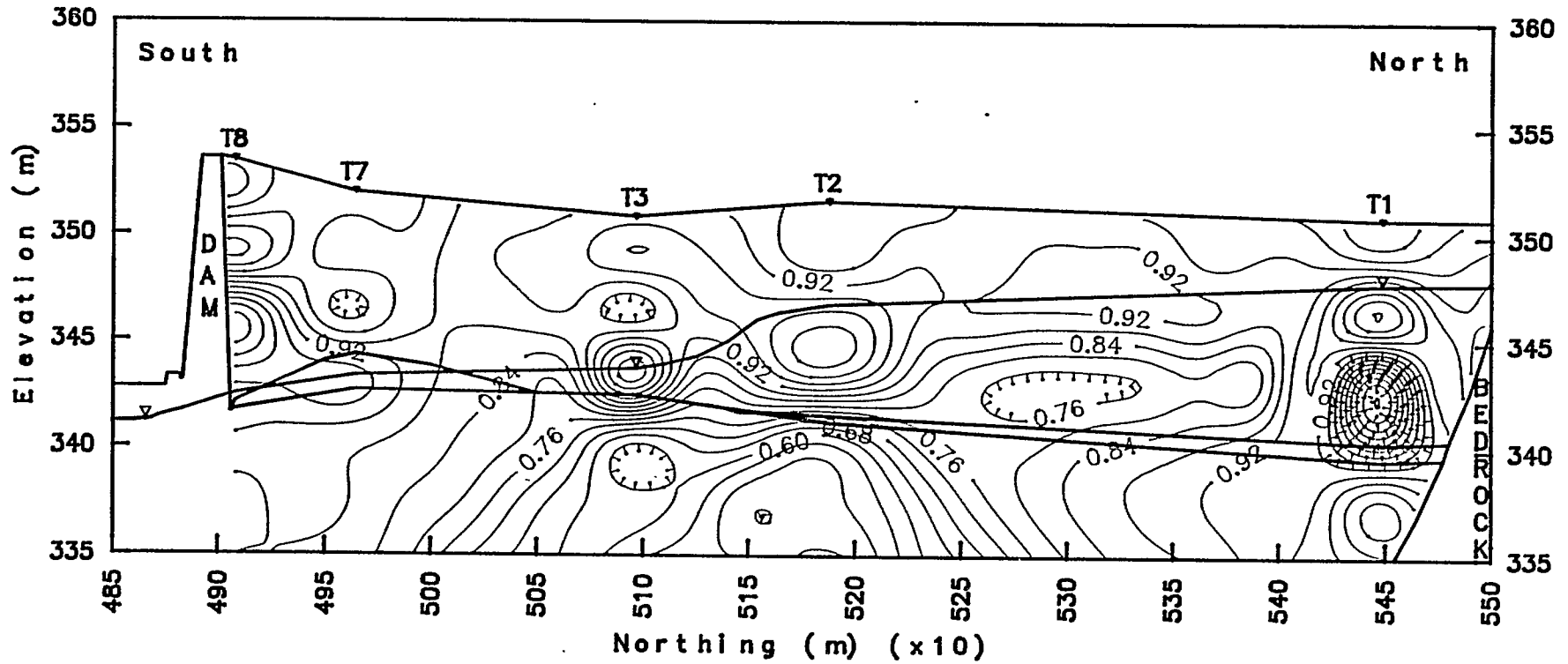


Fig. 7.91 - Nordic main tailings. Vertical north-south profile of solid phase activity ratios $^{210}\text{Pb}/^{226}\text{Ra}$, contour map with depth.

Activity Ratios Ra-226/Th-230

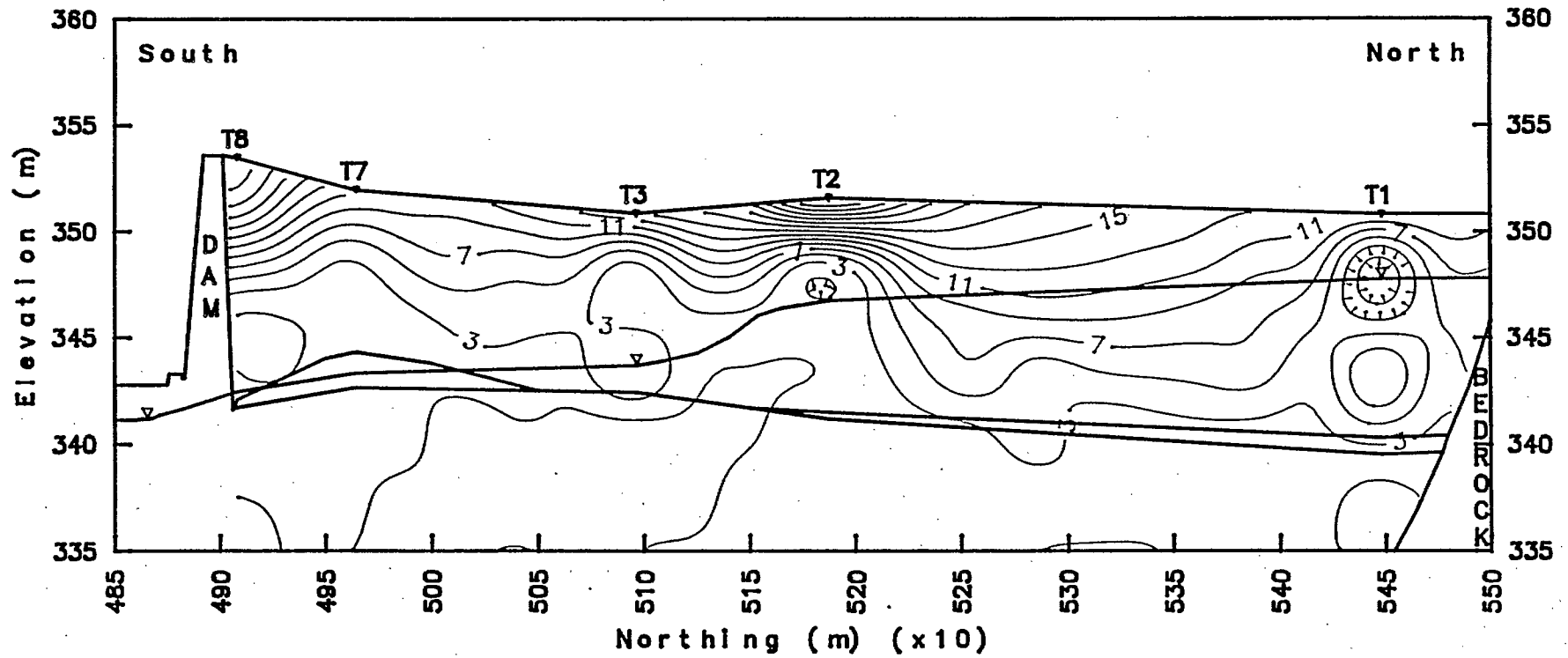


Fig. 7.92 - Nordic main tailings. Vertical north-south profile of solid phase activity ratios $^{226}\text{Ra}/^{230}\text{Th}$, contour map with depth.

Activity Ratios Th-228/Th-232

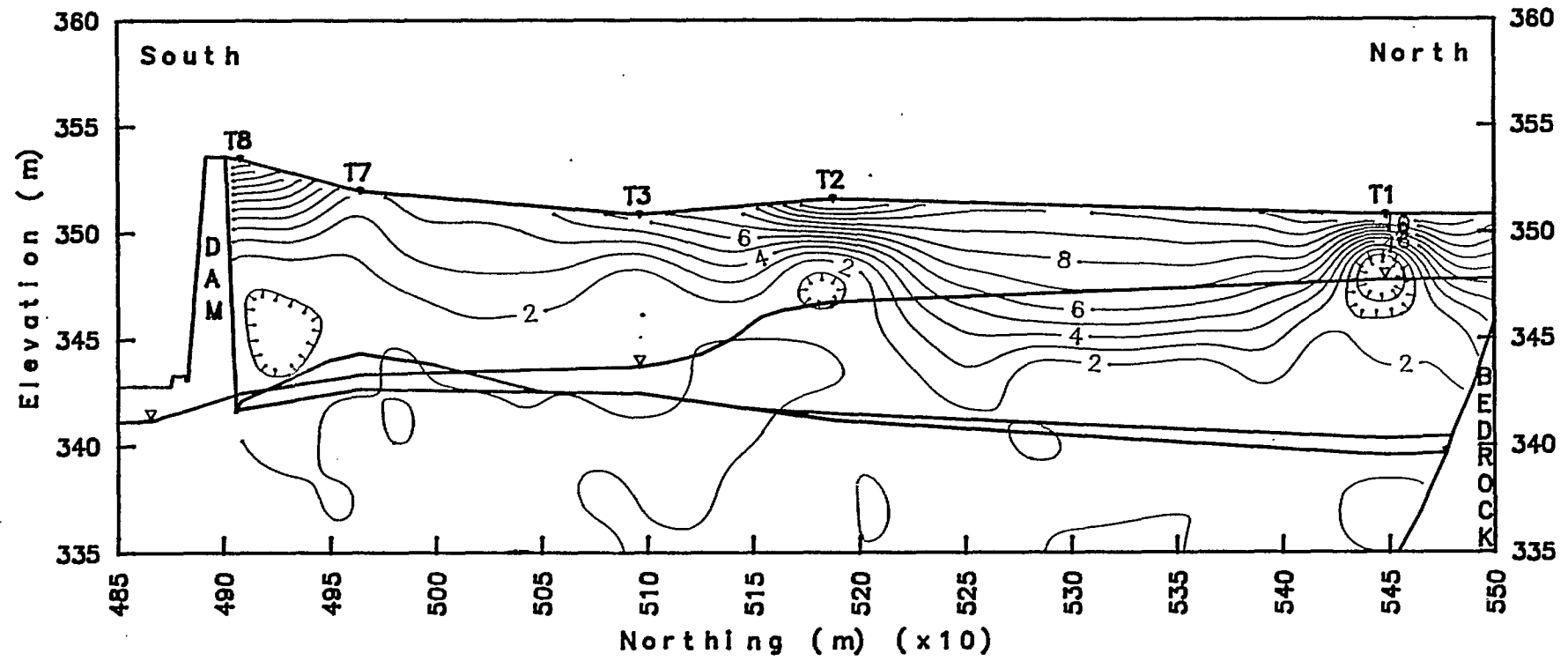


Fig. 7.93 - Nordic main tailings. Vertical north-south profile of solid phase activity ratios $^{228}\text{Th}/^{232}\text{Th}$, contour map with depth.

CHAPTER 8

SAMPLING METHOD AND TIMING SEQUENCE - LABORATORY EXPERIMENT

The existence of paradoxical secular disequilibrium has been shown in Chapter 7. The behaviour of this paradoxical secular disequilibrium is time-dependent as a result of the dynamics of nuclear decay chains and geochemistry processes. This Chapter will be devoted to the important role of sampling techniques and the timing sequence in identifying and characterizing this disequilibrium.

8.1 LABORATORY EXPERIMENT

A laboratory experiment was designed to exhibit the dynamic changes in the activity of the liquid phase. The design was based on related experiences in the field investigation, especially on radium analyses using alpha-spectroscopy (reference 47, Figures 16 to 18), in which some abnormality in the radium spectrum was observed. It was concluded that this abnormality was caused by the ingrowth of ^{233}Ra and ^{224}Ra isotopes within the sampling bottle a few weeks after sampling. Since the resolution of the technique for radium analyses at that time was very low, no further investigation was planned to test for the mechanism.

The laboratory facility at Elliot Lake Laboratory, at the present time is still very limited for doing analyses on the trans-uranium series, therefore, only alpha-particle emitting radium species were used in this experimental design. However, suitable radium alpha-emitters with short half-lives (11 d for ^{223}Ra , and 3.6 d for ^{224}Ra , respectively) could be used, and therefore rapid changes were expected. A new technique with high resolution alpha-spectroscopy for radium analysis, recently published, was used in this

experiment in order to identify the ^{223}Ra and ^{224}Ra (56).

8.2 EXPERIMENTAL DESIGN

In this experimental design, only natural radioisotopes from uranium tailings were used, and no spiked standard (such as that designed by Veska (26)) was added to enhance the limit of detection so only true paradoxical secular disequilibrium events that occurred naturally were observed.

A 100 L acid washed plastic container was used, and approximately 100 kg of the top layer of the tailings sample from Nordic main tailings was put into it and flooded with water.

A half inch pipe, 50 cm high, with side holes surrounded by a glass wool filter, and a rubber plug on the bottom, was installed in the centre of the tailings as a sampling port, Figure 8.1.

The sample was put aside for two years, because of other priorities. From time to time air was blown into the tube to keep the oxidation and low pH condition in effect. Since the sample had no porewater removed, it was expected that the solid and liquid phase would come to chemical equilibrium, as well as radioisotope equilibrium on some short-lived radioisotope species such as ^{227}Th (18 d), ^{228}Th (1.9 y), ^{223}Ra (11 d) and ^{224}Ra (3.6 d).

A 4 L liquid phase sample was extracted from the mixture using a peristaltic pump and passed through a $0.45\ \mu\text{m}$ filter. The sample bottle was labelled 'RDE', for Radioisotope Disequilibrium Experiment, and some of the standard chemical parameters were measured, such as pH, Eh, Ec and temperature. Then 10 mL concentrated hydrochloric acid was added to the liquid phase sample to preserve and stabilize it. The time of liquid and solid phase separation was noted, and a 250 mL sample cut was taken for radium analysis. On three separate occasions liquid samples were extracted and analyzed for radium content. The radium planchets (56) were labelled

RDE-1, RDE-2 and RDE-3, respectively.

The separation times for $\text{Ba}(\text{Ra})\text{SO}_4$ from the EDTA solution were noted for RDE-1, RDE-2 and RDE-3. These data were used later to determine the amount of ^{227}Th and ^{228}Th in the liquid phase at time of separation from the solid phase.

The radium samples on each planchet were periodically counted at 6000 sec intervals up to 600 hours from the liquid separation time. Several radium spectra were obtained from each planchet, and the integration of ^{226}Ra , ($^{223}\text{Ra}+^{224}\text{Ra}$), ^{215}Po , ^{214}Po and ^{212}Po peaks were recorded every 6000 sec. The data were used later on ^{226}Ra , ^{223}Ra and ^{224}Ra determinations.

It should be noted here that this is the first time 'High Resolution Alpha-Spectroscopy for Radium Analysis' (56) has been employed and tested for field use.

8.3 RESULTS AND DISCUSSION

The radioisotope disequilibrium experimental data are shown in Tables 8.1 to 8.11. Some typical spectra of mixed radium species are shown in Figures 8.2 to 8.4. These three Figures were obtained from the same planchet and show a gradual transformation of spectra with time as the radium decays into its daughters.

An example of the decay curve of mixed radium species is shown in Figures 8.5 and 8.6.

The raw data for RDE-1 are shown in Table 8.1, and the standard information, such as time of sample collection, time of radium species separation from the liquid phase sample, and the ^{133}Ba recovery factor to correct the radium activity are also shown.

Table 8.2 shows the processed data based on the \ln (natural logarithm) function of corrected radium counts to 100% yield.

In Figure 8.6, an example using RDE-3 data shows the decay curve of mixed radium species of \ln (count rate/6000 sec) versus time in hours. The ^{212}Po (8.78 MeV) daughter of ^{224}Ra (3.6 d) shows a rapid increase in activity caused by the short half-life of its parent. After reaching T_{max} in approximately 40 hours, the ^{212}Po (daughter) came to secular equilibrium with ^{224}Ra (parent), and the decay curve was parallel with its parent.

Therefore, the antilog of the projection of the linear regression of ^{212}Po decay curve, between T_{max} and 700 hours to $t=0$, in which radium was separated from the liquid phase sample, determined the activity of pure ^{224}Ra (Table 8.3).

From Tables 8.1 to 8.10, the activity of ^{223}Ra (5.71 MeV) and ^{224}Ra (5.68 MeV), as unresolvable twin peaks, was determined from the first data obtained before too much ^{222}Rn and other daughter isotopes interfered with the twin peaks.

By subtracting the activity of the determined ^{224}Ra from the corrected twin peaks of ^{223}Ra and ^{224}Ra , the activity of ^{223}Ra was obtained. The same routine was carried out for RDE-2 and RDE-3 samples to obtain the activities of ^{223}Ra and ^{224}Ra .

Compiled data for RDE-1, RDE-2 and RDE-3 are shown in Table 8.11. The activities of ^{223}Ra and ^{224}Ra are arranged as a function of time after liquid phase separation from the solid.

Using Equation 2, (Chapter 2.2.2), ^{227}Th and ^{228}Th can be calculated as follows:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) + N_2^0 e^{-\lambda_2 t} \quad \text{Eq 2}$$

where, N_1 = number of parent atoms, N_2 = number of daughter atoms. Since, $N = \frac{A}{\lambda}$, where A_1 is the activity of the parent and A_2 is the activity of the daughter, we have:

$$\frac{A_2}{\lambda_2} = \frac{\lambda_1}{\lambda_2 - \lambda_1} \left(\frac{A_1^0}{\lambda_1} \right) \left(e^{-\lambda_1 t} - e^{-\lambda_2 t} \right) + \left(\frac{A_1^0}{\lambda_2} \right) e^{-\lambda_2 t}$$

By multiplying it with $\lambda_2 e^{\lambda_2 t}$, the above equation becomes:

$$A_2 e^{\lambda_2 t} = \frac{\lambda_2}{\lambda_2 - \lambda_1} A_1^0 \left(e^{(\lambda_2 - \lambda_1)t} - 1 \right) + A_2^0$$

A plot of:

$$A_2 e^{\lambda_2 t} \text{ vs. } \frac{\lambda_2}{\lambda_2 - \lambda_1} \left(e^{(\lambda_2 - \lambda_1)t} - 1 \right)$$

will be linear with slope A_1^0 and intercept A_2^0 , where A_1^0 and A_2^0 are the activities of parent and daughter isotopes at time $t = 0$, when the liquid phase was separated from the solid, respectively.

Table 8.11 displays the calculated results for ^{227}Th , ^{223}Ra , ^{228}Th and ^{224}Ra . The ratio of daughter/parent activities was calculated as follows:

$$\frac{^{224}\text{Ra}}{^{228}\text{Th}} = \frac{50 \text{ (Bq/L)}}{338 \text{ (Bq/L)}} = 0.148, \text{ and}$$

$$\frac{^{223}\text{Ra}}{^{227}\text{Th}} = \frac{50 \text{ (Bq/L)}}{690 \text{ (Bq/L)}} = 0.073$$

It can be seen from these ratios that the daughter/parent isotopes, $^{223}\text{Ra}/^{227}\text{Th}$ and $^{224}\text{Ra}/^{228}\text{Th}$ are not in equilibrium, and yet it was expected that they should be, since the sample had been stored for two years without any of the liquid phase being removed. Thus the type of secular disequilibrium observed here can also be classified as paradoxical.

It is also important to mention here that, because of the dynamic growth of ^{223}Ra (11.7 d), and ^{224}Ra (3.6 d), and the dynamic decay of ^{227}Th (18.2 d), and ^{228}Th (1.98 y), secular equilibrium is reached in the sample container within days after the liquid phase separation. Consequently the timing sequence for sampling and geochemical mechanism is assuming an important role in the field study (see Table 8.11).

For the sampling technique of the future, the timing sequence should be included in the design to synchronize the the time required for field sampling with the time needed to analyze the short-lived radionuclides in either solid or liquid phase samples before secular equilibrium is reached in

the isolated sample container.

Liquid or solid samples that have been stored for too long (years) might lose their integrity as a tool for hydrogeochemical evolution study of radioisotope species as most short-lived radioisotopes would have reached their secular equilibrium. It would then be impossible to evaluate the condition of the sample or the formation of these radioisotopes in the tailings pile at the time of sampling.

8.4 APPLICATION TO RADIOISOTOPES IN A GROUNDWATER SPRING

Based on the above data and experiment, a similar mechanism can also be applied to the enrichment of some radioisotopes from the sample obtained from an active groundwater spring, as the spring water can be classified as liquid extraction from the main bulk mass of the geological formation. This can be explained by evaluating the previous experimental data.

For example, as shown in Table 8.11, ^{228}Th at the time of liquid separation had an activity of 338 ± 8 Bq/L, and ^{224}Ra activity was only 50 ± 80 Bq/L. After it had been stored in the bottle for $T = 390$ hours, the ^{224}Ra activity grew and reached 320.7 Bq/L. This could only be explained by the fact that, in isolation, the Ra-224 isotope which grew from its parent ^{228}Th did not have the solid phase to attach to, adsorb to, or coprecipitate with, and probably did not have the right condition to precipitate. (In this case hydrochloric acid was added to prevent it precipitating.)

Therefore, in the natural groundwater spring, a particular daughter isotope can grow from its parent with time and distance from the source, and reflect an enrichment, or increase in activity ratio, between daughter/parent in combination with geochemical effects (1,5-14,22-26).

The important role of the radon species in radioisotope disequilibrium should be noted in this thesis, especially as radon gas is classified as an

inert gas. Inert gases do not react with other chemical species and remain static in the liquid phase as dissolved gas.

Fleischer (Figure 1, (60)), discussed the mechanism of ^{222}Rn release from solids, which is an ejection of ^{222}Rn into the liquid phase, especially in the saturation zone, due to alpha-recoil as ^{226}Ra decays.

The dissolved ^{222}Rn , which is highly mobile, moves with the liquid phase at the same velocity downward in the tailings pile, and is the main cause of enrichment of ^{210}Pb in the liquid phase at T-3 site, as observed by Dalton (40).

In the groundwater spring, the dissolved radon concentration is expected to be very high in comparison with its parent, the radium species. With time and distance, because of the agitation and turbulence as the spring water flows away from the source, the radon gas escapes into the air. Therefore, this again causes separation of gas from the liquid phase and may result in rapid decreases of ^{210}Pb with time and distance from the source.

Unpublished data from an artesian spring water sample from a nearby Elliot Lake area (April 1978), showed ^{222}Rn concentrations of 421 pCi/L (15.6 Bq/L), and ^{226}Ra concentrations as high as 65 pCi/L (2.4 Bq/L).

The ^{222}Rn measurement could be approximately 20 times lower than expected as the spring water was sampled directly from the mouth of the spring where there was a lot of turbulence and gas bubbles escaping into the air before the sample bottle could be closed. Other measurements carried out by the Ontario Ministry of the Environment, between 1978 and 1982, from the same well, showed values of 10,000 to 12,000 pCi/L.

Hess et al. (61) reported concentration values for Rn-222 of up to 30,000 pCi/L (1,110 Bq/L) in the United States.

The follow-up study on this particular artesian well near Elliot Lake, where it was believed to be located on the borderline of low grade

uranium ore, was never carried out.

However, based on the cell model by Morin (30), assuming a one cubic meter cell of tailings with 45% porosity and grain density of 2.7 g/mL, and an average ^{226}Ra concentration of 300 pCi/g (11.1 Bq/g), the partition of solid and void space are 450 litres void and 550 litres solid. Thus, the total solid mass is 1,485 kg, and the total ^{226}Ra available in the solid mass is 445.5×10^6 pCi (16.5×10^6 Bq).

If the void space is saturated with water, and if the ^{226}Ra and ^{222}Rn are in secular equilibrium and homogeneously mixed, then the concentration of ^{222}Rn in the steady state water phase can be expected to be as high as (total available ^{222}Rn in secular equilibrium with ^{226}Ra /the volume of void space =) 0.99×10^6 pCi/L, or 36,630 Bq/L.

Under dynamic conditions, where the liquid phase is always being replaced (depending on the flow rate), and taking into consideration that some of the ^{222}Rn will be absorbed by the solid, secular disequilibrium is anticipated, and therefore, lower concentration than 0.99×10^6 pCi/L is expected.

In the rock formation, the porosity and permeability of the rock may be less than in the tailings material, however, the magnitude of the radon release will not differ much, varying only between 400×10^6 pCi/L to 0.99×10^6 pCi/L.

8.5 CONCLUSIONS AND RECOMMENDATIONS

It is clear from the laboratory experiment described above that the timing of sampling and analytical techniques for the investigation of disequilibrium phenomena is critically important. Broad assumptions concerning equilibrium states for radioisotopes in an apparently isolated system must be carefully examined. Factors which can affect the accuracy of

isotope measurements and models, as seen in the laboratory experiment, include:

1. Careful sampling, sample preservation and preparation.
2. Planning for timing sequences starting with
 - liquid phase separation;
 - radium-species separation; and
 - time interval in alpha-counting.
3. The use of a good high resolution alpha-spectroscopy system is essential to obtain a clear spectrum separation of the radium species and its daughters.
4. The limited time shelf-life of a stored sample.

In Table 8.11, when the initial ^{223}Ra and ^{224}Ra peaks were counted, a large statistical error was observed caused by technical difficulties during alpha-counting, when a timing error occurred from vacuum breakdown at a critical moment resulting in twin peaks.

For the future analyses of radium species and their daughters it is recommended that the high resolution alpha-spectroscopy should be combined with a convoluted software program to evaluate complex alpha-spectra with greater accuracy, and to eliminate doubt in separating twin peaks. The thorium species could be analyzed directly as mentioned in Chapter 6.4 and compared with the method described in Chapter 8.2 for greater precision and accuracy.

Table 8.1 - Radioisotope disequilibrium experimental data, RDE-1.

RAW DATA						
REMARK CHN. FROM-TO	TIME (Hrs.) FROM T=0	Ra-226 790-970	Ra-(223+224) 981-1161	Po-215 1394-1494	Po-214 1496-1578	Po-212 1660-1840
MeV. FROM-TO		4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
1 PROBE 1	1.83	4381	34147	2073	45	1403
2	100.08	3525	18147	1796	1131	8178
3 t(collect sample):	148.00	3450	13537	1627	1522	6279
4 10:00, Oct. 7, 1987	174.33	3286	11945	1548	1494	4747
5 t(separation):	241.83	3158	8587	1379	1841	2778
6 10:50, Oct. 9, 1987	291.83	3103	7435	1373	2058	1959
7	311.58	2996	5909	1075	1924	1442
8 pH: 2.0	484.33	3050	4449	930	2184	1085
9 Eh: +434.2 mV	676.03	2761	3448	637	2059	140
10 Ec: 15650	1180.03	2670	3001	396	2113	25
11 temp: 68.9 F	1185.13	2660	3045	393	2082	28
12 BA-133 RECOVERY:	1186.98	2611	3009	344	2096	19
13 67.21%	1212.85	2779	3071	404	2133	22
14	1190.72	2690	2991	372	2036	19
15	1192.58	2651	3064	386	2122	18
16	1194.45	2584	3032	382	2087	21
17	1196.30	2627	2986	414	2062	19
18	1198.17	2666	3020	384	2082	13
19	1200.02	2664	2988	376	2169	18
20	1201.88	2655	2997	395	2135	21
21	1203.75	2664	2929	350	2080	20
22	1205.60	2798	2918	339	2065	20
23	1207.47	2749	2964	386	2103	17
24	1209.33	2521	2955	360	2111	22
25	1211.20	2742	3060	366	2126	13
26	1213.07	2747	3037	370	2079	21
27	1214.93	2634	2921	375	2088	15
28	1216.82	2610	3056	358	2068	25
29	1218.68	2715	2991	367	2052	16
30	1220.55	2591	3036	410	2106	12
31	1222.40	2758	3027	388	2081	21
32	1224.27	2636	2998	399	2121	22
33	1226.12	2759	3007	405	2105	26
34	1227.98	2707	3061	390	2115	24
35	1229.83	2671	3085	433	2055	15

Table 8.2 - Radioisotope disequilibrium experimental data, RDE-1.
 Processed data $\ln(A_{ic})$, derived from Table 8.1,
 where $A_{ic} = A_1 \times (100\% / {}^{133}\text{Ba recovery})$.

PROCESSED DATA						
REMARK	TIME (Hrs.)	Ra-226	Ra-(223+224)	Po-215	Po-214	Po-212
CHN.FROM-TO	FROM T=0	2848-3028	3039-3219	3452-3552	3554-3638	3718-3898
MeV.FROM-TO		4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
1 PROBE 1	1.83	8.78	10.84	8.03	4.20	8.09
2	100.08	8.57	10.20	7.89	7.43	9.85
3 t(collect sample):	148.00	8.54	9.91	7.79	7.73	9.59
4 10:00, Oct. 7, 1987	174.33	8.49	9.79	7.74	7.71	9.31
5 t(separation):	241.83	8.46	9.46	7.63	7.92	8.77
6 10:50, Oct. 9, 1987	291.83	8.44	9.31	7.62	8.03	8.42
7	311.58	8.40	9.08	7.38	7.96	8.12
8 pH: 2.0	484.33	8.42	8.80	7.23	8.09	7.83
9 Eh: +434.2 mV	676.03	8.32	8.54	6.85	8.03	5.79
10 Ec: 15650	1180.03	8.29	8.40	6.38	8.05	4.06
11 temp: 68.9 F	1185.13	8.28	8.42	6.37	8.04	4.18
12 BA-133 RECOVERY:	1186.98	8.26	8.41	6.24	8.05	3.79
13 67.21%	1212.85	8.33	8.43	6.40	8.06	3.93
14	1190.72	8.29	8.40	6.32	8.02	3.79
15	1192.58	8.28	8.42	6.35	8.06	3.73
16	1194.45	8.25	8.41	6.34	8.04	3.89
17	1196.30	8.27	8.40	6.42	8.03	3.79
18	1198.17	8.29	8.41	6.35	8.04	3.41
19	1200.02	8.28	8.40	6.33	8.08	3.73
20	1201.88	8.28	8.40	6.38	8.06	3.89
21	1203.75	8.28	8.38	6.26	8.04	3.84
22	1205.60	8.33	8.38	6.22	8.03	3.84
23	1207.47	8.32	8.39	6.35	8.05	3.68
24	1209.33	8.23	8.39	6.28	8.05	3.93
25	1211.20	8.31	8.42	6.30	8.06	3.41
26	1213.07	8.32	8.42	6.31	8.04	3.89
27	1214.93	8.27	8.38	6.32	8.04	3.55
28	1216.82	8.26	8.42	6.28	8.03	4.06
29	1218.68	8.30	8.40	6.30	8.02	3.62
30	1220.55	8.26	8.42	6.41	8.05	3.33
31	1222.40	8.32	8.41	6.36	8.04	3.89
32	1224.27	8.27	8.40	6.39	8.06	3.93
33	1226.12	8.32	8.41	6.40	8.05	4.10
34	1227.98	8.30	8.42	6.36	8.05	4.02
35	1229.85	8.29	8.43	6.47	8.03	3.55

Table 8.3 - Linear regression data, RDE-1.
Evaluation for ^{226}Ra , ^{223}Ra and ^{224}Ra .

```

=====
REGRESSION DATA AND      Linear regression
FINAL COUNTS FOR T=0     of ln ( $^{212}\text{Po}$ )
                          at 100% yield
=====
      Regression Output:
Constant                    10.09
Std Err of Y Est            0.26
R Squared                   0.98
No. of Observations        34
Degrees of Freedom          32

X Coefficient(s)  -0.00522
Std Err of Coef.  0.000115

Ra-226 (100% Recovery):      3974
Ra-224 (100% Recovery):      24159
Ra-223 (100% Recovery):      26650

```

^{226}Ra (100% recovery) was obtained from the last ^{226}Ra raw data after $t = 1000$ hours, and corrected using ^{133}Ba recovery to a 100% yield.

^{224}Ra (100% recovery) was obtained by means of linear regression interpolation of $\text{antilog ln } A(^{212}\text{Po})$ at $t=0$.

^{223}Ra (100% recovery) was obtained by means of subtracting the activity $A(^{224}\text{Ra})$ at 100% yield from the first data of $A(^{233}\text{Ra} + ^{224}\text{Ra})$ at 100% yield at $t=0$.

Table 8.4 - Radioisotope disequilibrium experimental data, RDE-2

RAW DATA							
RDE	REMARK CHN.FROM-TO	TIME (Hrs.) FROM T=0	Ra-226 790-970	Ra-(223+224) 981-1161	Po-215 1394-1494	Po-214 1496-1578	Po-212 1660-1840
MeV.FROM-TO			4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
1	PROBE 1	1.83	5007	65295	4422	61	1943
2	1987 10 14 - 16:20	18.08	5182	63524	4687	535	21799
3		41.92	4971	52933	4354	768	25060
4	pH: 2.0	113.92	3976	31425	3545	1247	15082
5		163.33	3765	22959	3373	1767	10306
6	Eh: +434.2 mV	377.83	3002	7828	2000	1919	2242
7		721.00	2655	3471	952	1931	156
8		786.45	2704	3585	861	2019	88
9	Ec: 15650	1050.02	2717	3044	557	2006	46

ln A _{1c} versus time PROCESSED DATA							
RDE	REMARK CHN.FROM-TO	TIME (Hrs.) FROM T=0	Ra-226 2848-3028	Ra-(223+224) 3039-3219	Po-215 3452-3552	Po-214 3554-3638	Po-212 3718-3898
MeV.FROM-TO			4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
1	PROBE 1	1.83	8.91	11.48	8.78	4.50	8.41
2	1987 10 14 - 16:20	18.08	8.94	11.45	8.84	6.67	10.82
3		41.92	8.90	11.27	8.77	7.03	10.96
4	pH: 2.0	113.92	8.68	10.74	8.56	7.52	10.46
5		163.33	8.62	10.43	8.51	7.87	10.08
6	Eh: +434.2 mV	377.83	8.40	9.35	7.99	7.95	8.55
7		721.00	8.27	8.54	7.25	7.95	5.88
8		786.45	8.29	8.57	7.15	8.00	5.31
9	Ec: 15650	1050.02	8.30	8.41	6.71	7.99	4.66

where $A_{1c} = A_1 \times (100\% / {}^{133}\text{Ba recovery})$

Table 8.5 - Linear regression data, RDE-2.
Evaluation for ^{226}Ra , ^{223}Ra and ^{224}Ra

```

=====
REGRESSION DATA AND          Linear regression
FINAL COUNTS FOR T=0        of ln ( $^{212}\text{Po}$ )
                             at 100% yield
=====
      Regression Output:
Constant                      11.32
Std Err of Y Est              0.06
R Squared                     1.00
No. of Observations           6
Degrees of Freedom             4

X Coefficient(s)  -0.00756
Std Err of Coef.  0.000080

Ra-226 (100% Recovery):      4008
Ra-224 (100% Recovery):      82562
Ra-223 (100% Recovery):      13767

```

^{226}Ra (100% recovery) was obtained from the last ^{226}Ra raw data after $t = 1000$ hours, and corrected using ^{133}Ba recovery to a 100% yield.

^{224}Ra (100% recovery) was obtained by means of linear regression interpolation of $\text{antilog ln } A(^{212}\text{Po})$ at $t=0$.

^{223}Ra (100% recovery) was obtained by means of subtracting the activity $A(^{224}\text{Ra})$ at 100% yield from the first data of $A(^{223}\text{Ra} + ^{224}\text{Ra})$ at 100% yield at $t=0$.

Table 8.6 - Radioisotope disequilibrium experimental data, RDE-3.

RAW DATA							
#	REMARK	TIME (Hrs.)	Ra-226	Ra-(223+224)	Po-215	Po-214	Po-212
	CHN.FROM-TO	FROM T=0	2848-3028	3039-3219	3452-3552	3554-3638	3718-3898
	MeV.FROM-TO		4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
1	PROBE #1	1.67	8505	87994	6616	234	2704
2		3.53	8567	88322	6691	331	6458
3		5.38	8646	86239	6718	387	10194
4		7.23	8685	86230	6795	490	13453
5	16:00 87,10,23	9.08	8746	85454	6732	589	16420
6	23 Oct 87	10.95	8707	84868	6815	650	19111
7		12.80	8622	82911	6700	737	21455
8		14.65	8561	82610	6777	828	23412
9		16.50	8353	81956	6874	917	25075
10		18.35	8538	81028	6661	970	26704
11	Sample:	20.22	8413	80036	6648	978	27684
12	RAMIXPR1	22.07	8297	79708	6778	992	28517
13	TRIAL 1	23.92	8225	78616	6700	1070	29672
14		25.77	8410	76974	6608	1096	29748
15	time of separation:	27.63	8062	75720	6637	1143	30212
16	15:50	29.48	7994	74965	6643	1179	31014
17		31.33	8087	73773	6531	1150	30973
18	pH: 2.0	33.18	7935	73350	6376	1291	31462
19	Eh: +434.2 mV	35.05	7892	71836	6431	1263	31256
20	Ec: 15650	36.90	7897	71094	6545	1298	31264
21	temp: 68.9 F	38.75	7643	69908	6363	1303	31791
22	Ba-133 RECOVERY	40.60	7727	69605	6353	1360	31749
23	68.78%	42.45	7608	68845	6297	1330	31486
24		44.32	7766	68479	6479	1316	31023
25		46.17	7557	67400	6320	1366	31138
26		48.02	7407	66525	6265	1373	30525
27		49.87	7486	65869	6356	1378	30791
28		51.73	7562	65089	6176	1390	31004
29		53.58	7414	64513	6224	1476	30638
30		55.43	7226	63612	6345	1456	30028
31		73.75	6803	55670	5891	1525	26603
32		75.62	6743	55185	5869	1522	26779
33		77.47	6853	54752	5905	1567	26261
34		79.32	6614	53488	6040	1477	25897
35		81.17	6560	52253	5694	1524	24827
36		83.03	6369	51960	5786	1616	24576
37		84.88	6442	51137	5702	1567	24520
38		86.75	6333	50671	5657	1601	24278
39		88.60	6404	50963	5733	1617	24082
40		90.45	6442	50008	5724	1609	23676
41		96.58	6269	48151	5566	1606	22666
42		98.43	6202	47823	5690	1656	22250
43		100.30	6165	46715	5534	1700	21953
44		102.15	6091	46193	5653	1824	21838
45		104.00	5980	45437	5494	1643	21088

Table 8.7 - Radioisotope disequilibrium experimental data, RDE-3.

RAW DATA							
#	REMARK	TIME (Hrs.)	Ra-226	Ra-(223+224)	Po-215	Po-214	Po-212
	CHN.FROM-TO	FROM T=0	2848-3028	3039-3219	3452-3552	3554-3638	3718-3898
	MeV.FROM-TO		4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
46		105.87	5919	44426	5666	1729	20892
47		107.72	5906	44087	5455	1702	20767
48		109.57	5916	44340	5487	1737	20428
49		111.42	6071	43520	5514	1674	20173
50		113.28	5894	43261	5603	1732	19979
51		673.98	2834	3763	1394	1503	294
52		675.83	2899	3769	1441	1486	252
53		677.68	2947	3798	1328	1526	279
54		679.70	2962	3806	1351	1565	254
55		681.38	2814	3667	1283	1638	270
56		683.25	2823	3869	1364	1571	250
57		685.10	2985	3634	1283	1617	220
58		686.95	2934	3755	1289	1587	243
59		688.80	2937	3727	1328	1611	285
60		690.65	2955	3622	1302	1618	210
61		692.50	2944	3843	1269	1651	223
62		694.37	2999	3881	1271	1607	207
63		696.22	2916	3769	1299	1654	176
64		698.07	2878 ^a	3712	1229	1609	201
65		699.92	2911	3798	1279	1572	199
66		701.77	2986	3845	1249	1750	205
67		703.63	3016	3813	1258	1717	223
68		705.48	2863	3726	1243	1698	206
69		707.33	2835	3707	1230	1683	209
70		709.18	3964	3737	1285	1736	193
71		711.03	2980	3668	1208	1721	211
72		712.88	2882	3809	1231	1753	164
73		714.75	2897	3730	1242	1670	174
74		716.60	2938	3863	1274	1732	165
75		718.45	2896	3788	1255	1731	185
76		720.57	2945	3730	1193	1725	179
77		722.15	2874	3919	1246	1719	175
78		724.02	2892	3882	1246	1725	170
79		725.87	3013	3909	1243	1793	171
80		727.72	2867	3859	1282	1742	179
81		761.20	3142	3744	1272	1667	152
82		1085.48	2996	2964	687	1780	48
83		1087.35	3023	2943	675	1701	46
84		1089.20	2991	3025	703	1659	54
85		1091.07	2924	3054	697	1720	38
86		1092.92	3017	3167	702	1738	42
87		1483.92	2890	2634	411	1628	81
88		1485.65	2832	2648	411	1565	95
89		1487.58	2954	2663	431	1601	81
90		1489.50	2933	2626	398	1607	69

Table 8.8 - Radioisotope disequilibrium experimental data, RDE-3.

PROCESSED DATA							
#	REMARK	TIME (Hrs.)	Ra-226	Ra-(223+224)	Po-215	Po-214	Po-212
	CHN.FROM-TO	FROM T=0	2848-3028	3039-3219	3452-3552	3554-3638	3718-3898
	MeV.FROM-TO		4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
1	PROBE #1	1.67	9.42	11.76	9.17	5.83	8.72
2		3.33	9.43	11.76	9.18	6.18	9.59
3		5.38	9.44	11.74	9.19	6.33	10.05
4		7.23	9.44	11.74	9.20	6.57	10.33
5	16:00 87,10,23	9.08	9.45	11.73	9.19	6.75	10.53
6	23 Oct 87	10.95	9.45	11.72	9.20	6.85	10.68
7		12.80	9.44	11.70	9.18	6.98	10.79
8		14.65	9.43	11.70	9.20	7.09	10.88
9		16.50	9.40	11.69	9.21	7.20	10.95
10		18.35	9.43	11.68	9.18	7.25	11.01
11	Sample:	20.22	9.41	11.66	9.18	7.26	11.05
12	RAMIYPR1	22.07	9.40	11.66	9.20	7.27	11.08
13	TRIAL 1	23.92	9.39	11.65	9.18	7.35	11.12
14		25.77	9.41	11.63	9.17	7.37	11.12
15	time of separation:	27.63	9.37	11.61	9.17	7.42	11.14
16	15:50	29.48	9.36	11.60	9.18	7.45	11.16
17		31.33	9.37	11.58	9.16	7.42	11.16
18	pH: 2.0	33.18	9.35	11.58	9.13	7.54	11.18
19	Eh: +434.2 mV	35.05	9.35	11.56	9.14	7.52	11.17
20	Ec: 15650	36.90	9.35	11.55	9.16	7.54	11.17
21	temp: 68.9 F	38.75	9.32	11.53	9.13	7.55	11.19
22	Ba-133 RECOVERY	40.60	9.33	11.52	9.13	7.59	11.19
23	68.78%	42.45	9.31	11.51	9.12	7.57	11.18
24		44.32	9.33	11.51	9.15	7.56	11.16
25		46.17	9.30	11.49	9.13	7.59	11.17
26		48.02	9.28	11.48	9.12	7.60	11.15
27		49.87	9.30	11.47	9.13	7.60	11.16
28		51.73	9.31	11.46	9.10	7.61	11.16
29		53.58	9.29	11.45	9.11	7.67	11.15
30		55.43	9.26	11.43	9.13	7.66	11.13
31		57.28	9.20	11.30	9.06	7.70	11.01
32		59.13	9.19	11.29	9.05	7.70	11.02
33		60.98	9.21	11.28	9.06	7.73	11.00
34		62.83	9.17	11.26	9.08	7.67	10.98
35		64.68	9.16	11.24	9.02	7.70	10.94
36		66.53	9.13	11.23	9.04	7.76	10.93
37		68.38	9.14	11.22	9.02	7.73	10.93
38		70.23	9.13	11.21	9.01	7.75	10.92
39		72.08	9.14	11.21	9.03	7.76	10.91
40		73.93	9.14	11.19	9.03	7.76	10.89
41		75.78	9.12	11.16	9.00	7.76	10.85
42		77.63	9.11	11.15	9.02	7.79	10.83
43		79.48	9.10	11.13	8.99	7.81	10.82
44		81.33	9.09	11.11	9.01	7.88	10.81
45		83.18	9.07	11.10	8.99	7.78	10.78

Processed data $\ln(A_{1c})$, derived from Table 8.6, where $A_{1c} = A_1 \times$
 (100%/ % ^{133}Ba recovery).

Table 8.9 - Radioisotope disequilibrium experimental data, RDE-3.

PROCESSED DATA							
#	REMARK	TIME (Hrs.)	Ra-226	Ra-(223+224)	Po-215	Po-214	Po-212
	CHN.FROM-TO	FROM T=0	2848-3028	3039-3219	3452-3552	3554-3638	3718-3898
	MeV.FROM-TO		4.03-4.91	4.97-5.85	6.98-7.47	7.48-7.88	8.28-9.16
46		105.87	9.06	11.08	9.02	7.83	10.77
47		107.72	9.06	11.07	8.98	7.81	10.76
48		109.57	9.06	11.07	8.98	7.83	10.75
49		111.42	9.09	11.06	8.99	7.80	10.73
50		113.28	9.06	11.05	9.01	7.83	10.72
51		673.98	8.32	8.61	7.61	7.69	6.50
52		675.83	8.35	8.61	7.65	7.68	6.35
53		677.68	8.36	8.62	7.57	7.70	6.45
54		679.70	8.37	8.62	7.58	7.73	6.36
55		681.38	8.32	8.58	7.53	7.78	6.42
56		683.25	8.32	8.64	7.59	7.73	6.34
57		685.10	8.38	8.57	7.53	7.76	6.21
58		686.95	8.36	8.61	7.54	7.74	6.31
59		688.80	8.36	8.60	7.57	7.76	6.47
60		690.65	8.37	8.57	7.55	7.76	6.17
61		692.50	8.36	8.63	7.52	7.78	6.23
62		694.37	8.38	8.64	7.52	7.76	6.15
63		696.22	8.35	8.61	7.54	7.79	5.99
64		698.07	8.34	8.59	7.49	7.76	6.12
65		699.92	8.35	8.62	7.53	7.73	6.11
66		701.77	8.38	8.63	7.50	7.84	6.14
67		703.63	8.39	8.62	7.51	7.82	6.23
68		705.48	8.33	8.60	7.50	7.81	6.15
69		707.33	8.32	8.59	7.49	7.80	6.16
70		709.18	8.66	8.60	7.53	7.83	6.08
71		711.03	8.37	8.58	7.47	7.82	6.17
72		712.88	8.34	8.62	7.49	7.84	5.92
73		714.75	8.35	8.60	7.50	7.79	5.98
74		716.60	8.36	8.63	7.52	7.83	5.93
75		718.45	8.35	8.61	7.51	7.83	6.04
76		720.57	8.36	8.60	7.46	7.83	6.01
77		722.15	8.34	8.65	7.50	7.82	5.99
78		724.02	8.34	8.64	7.50	7.83	5.96
79		725.87	8.39	8.65	7.50	7.87	5.96
80		727.72	8.34	8.63	7.53	7.84	6.01
81		761.20	8.43	8.60	7.52	7.79	5.84
82		1085.48	8.38	8.37	6.91	7.86	4.69
83		1087.33	8.39	8.36	6.89	7.81	4.65
84		1089.20	8.38	8.39	6.93	7.79	4.81
85		1091.07	8.36	8.40	6.92	7.82	4.46
86		1092.92	8.39	8.43	6.93	7.83	4.56
87		1483.92	8.34	8.25	6.39	7.77	5.22
88		1485.65	8.32	8.26	6.39	7.73	5.37
89		1487.58	8.37	8.26	6.44	7.75	5.22
90		1489.50	8.36	8.25	6.36	7.76	5.05

Processed data $\ln(A_{ic})$, derived from Table 8.7, where $A_{ic} = A_i \times (100\%/ \% \text{ } ^{133}\text{Ba recovery})$.

Table 8.10 - Linear regression data RDE-3.
Evaluation for ^{226}Ra , ^{223}Ra and ^{224}Ra

REGRESSION DATA AND FINAL COUNTS FOR T=0		Linear regression of ln (^{212}Po) at 100% yield

Regression Output:		
Constant		11.42
Std Err of Y Est		0.34
R Squared		0.98
No. of Observations		66
Degrees of Freedom		64
X Coefficient(s)	-0.00713	
Std Err of Coef.	0.00012	
Ra-226 (100% Recovery):		4375
Ra-224 (100% Recovery):		91230
Ra-223 (100% Recovery):		36712

^{226}Ra (100% recovery) was obtained from the last ^{226}Ra raw data after $t = 1000$ hours, and corrected using ^{133}Ba recovery to a 100% yield.

^{224}Ra (100% recovery) was obtained by means of linear regression interpolation of antilog ln $A(^{212}\text{Po})$ at $t=0$.

^{223}Ra (100% recovery) was obtained by means of subtracting the activity $A(^{224}\text{Ra})$ at 100% yield from the first data of $A(^{223}\text{Ra} + ^{224}\text{Ra})$ at 100% yield at $t=0$.

Table 8.11 - Compiled data for RDE-1, RDE-2 and RDE-3. ^{228}Th , ^{223}Ra and ^{224}Ra at time 0 were evaluated based on the concentration of ^{223}Ra and ^{224}Ra observed as a function of time after liquid phase extraction from the solid phase.

SAMPLE DESCRIPTION	TIME OF LIQUID EXTRACTION FROM SOLID PHASE	TIME OF SEPERATION FOR RADIUM SPECIES	TIME FROM EXTRACTION = (hrs)
RDE-1	07-Oct-87 -10 : 0	09-Oct-87 -10 :50	48.83
RDE-2	07-Oct-87 -10 : 0	14-Oct-87 -16 :20	174.33
RDE-3	07-Oct-87 -10 : 0	23-Oct-87 -16 : 0	390.00

SAMPLE DESCRIPTION	COUNTS AT 100% RECOVERY			CONTENT (pCi/L)			CONTENT (mBq/L)		
	RA-226	Ra-224	Ra-223	Ra-226	Ra-224	Ra-223	Ra-226	Ra-224	Ra-223
RDE-1	3974	24159	26650	378	2295	2532	13970	84930	93686
RDE-2	4008	82562	13766	381	7844	1308	14690	290242	48394
RDE-3	4375	91230	36712	416	8668	3488	15380	320714	129059
^{223}Ra CONTENT AT LIQUID SEPARATION FROM SOLID PHASE (Bq/L) :							50 +/- 80		
^{228}Th CONTENT AT LIQUID SEPARATION FROM SOLID PHASE (Bq/L) :							690 +/- 100		
^{224}Ra CONTENT AT LIQUID SEPARATION FROM SOLID PHASE (Bq/L) :							50 +/- 130		
^{228}Th CONTENT AT LIQUID SEPARATION FROM SOLID PHASE (Bq/L) :							338 +/- 8		

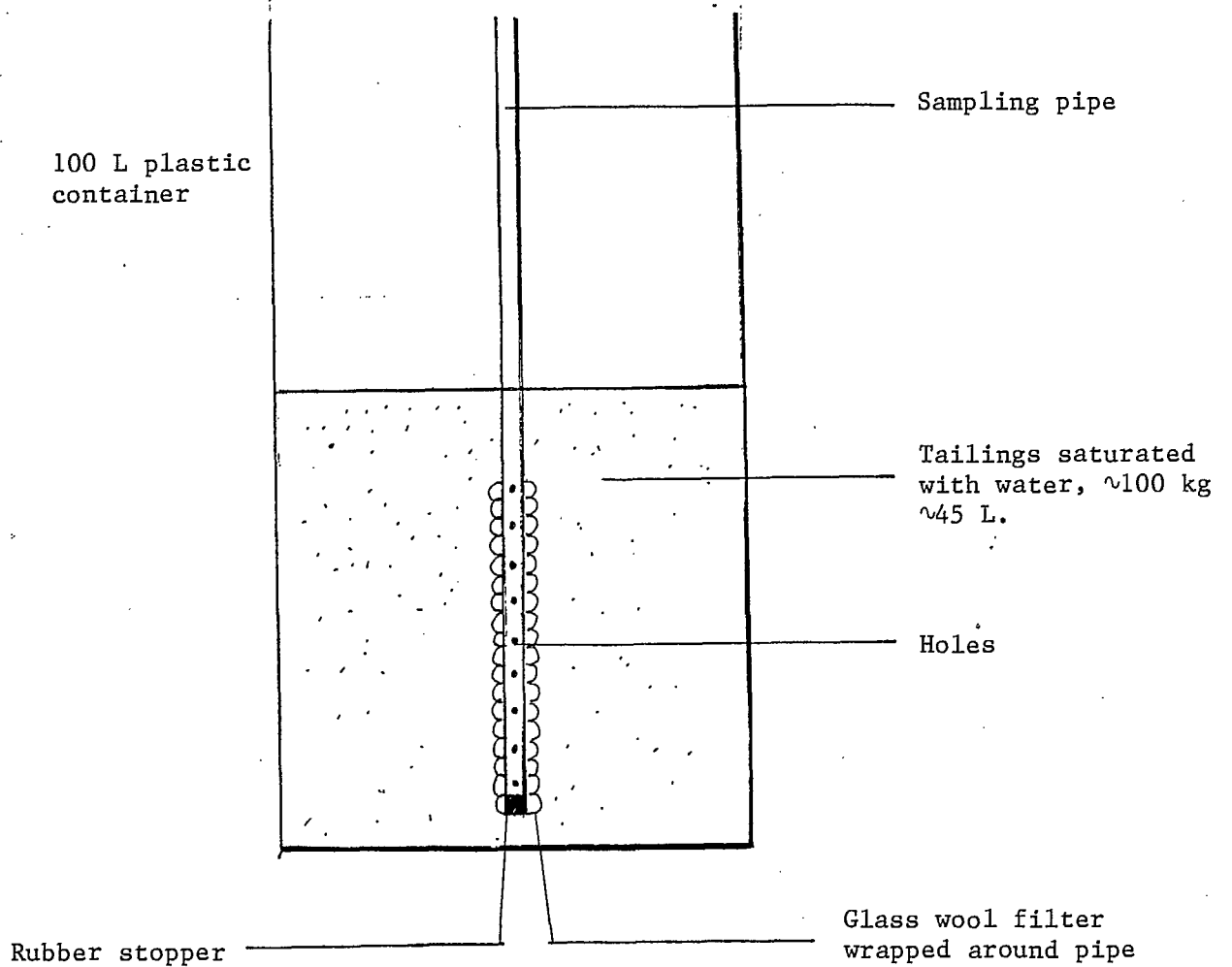


Fig. 8.1 - Laboratory experimental design for testing paradoxical secular disequilibrium in uranium tailings.

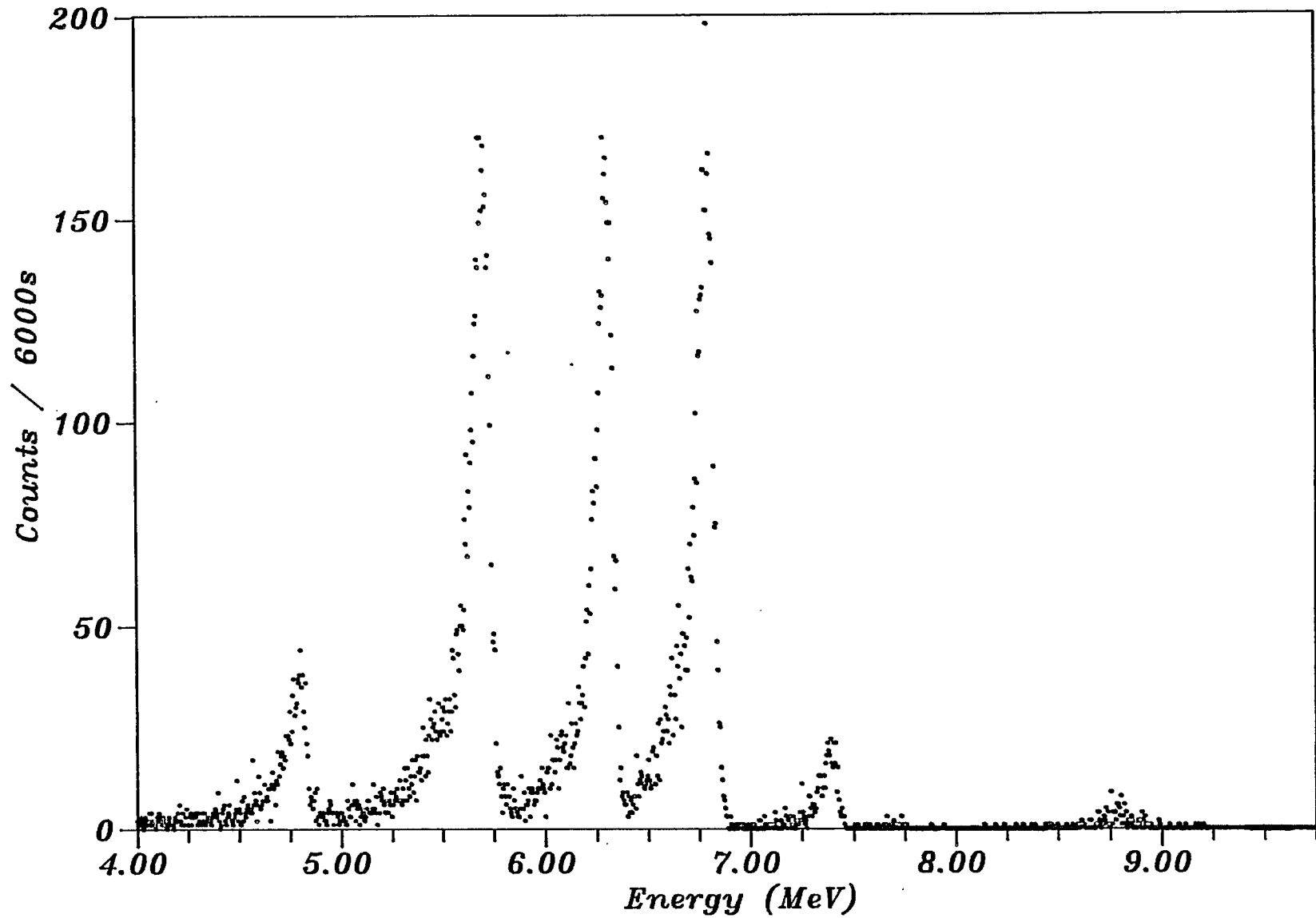


Fig. 8.2 - Mixed radium alpha spectrum. T=0 : 2.10 hours.

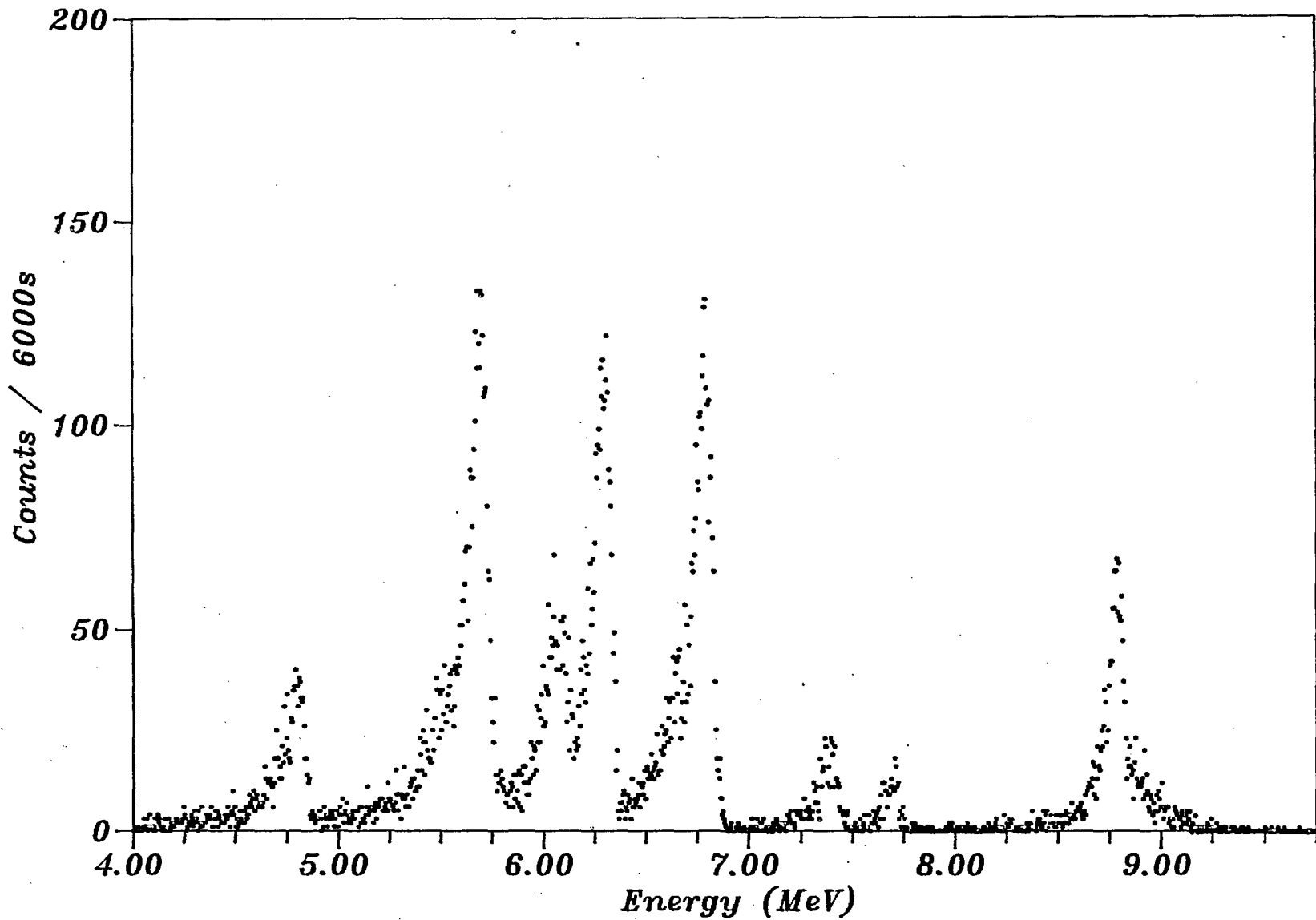


Fig. 8.3 - Mixed radium alpha spectrum. Time from T=0 : 30.80 hours.

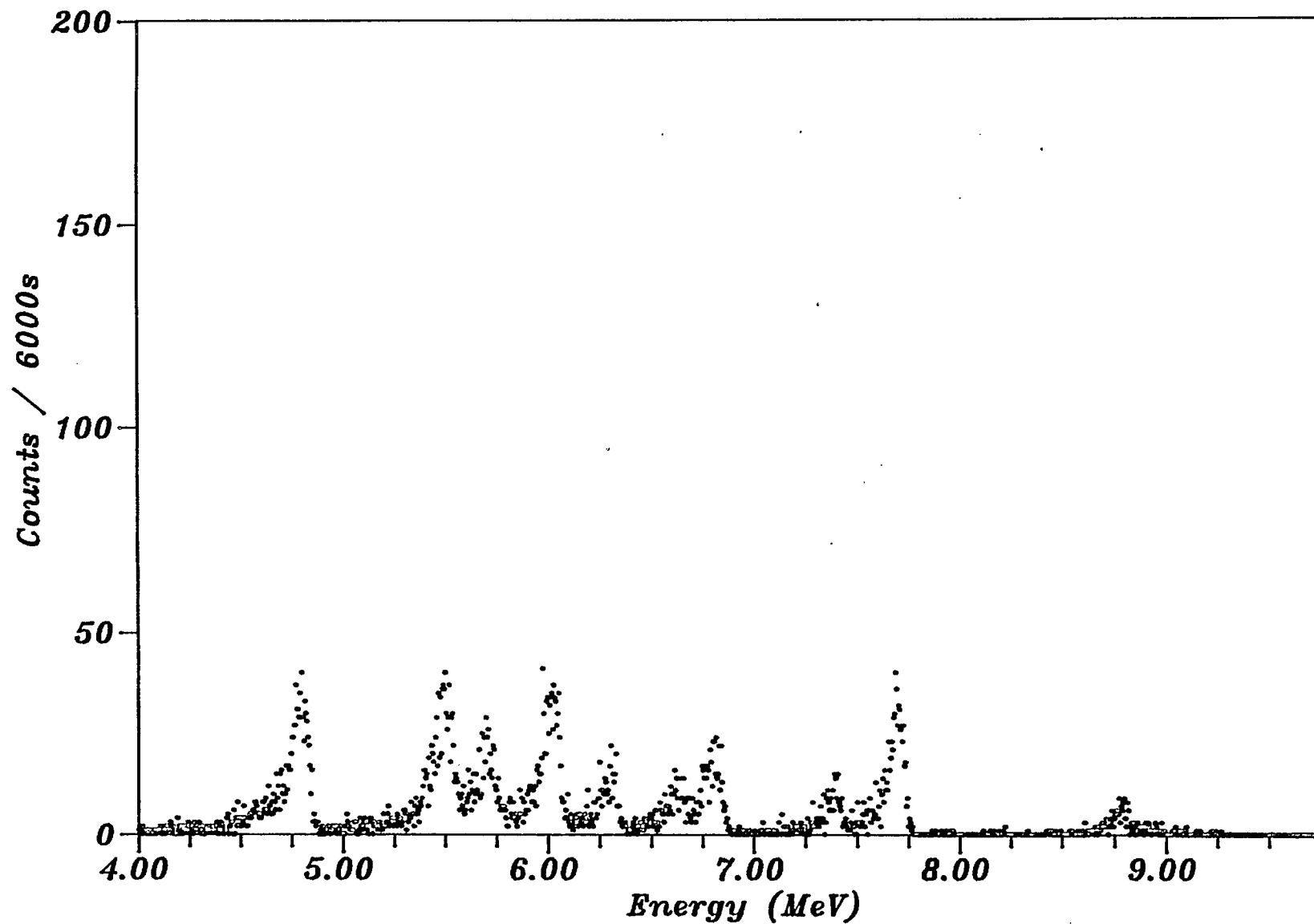


Fig. 8.4 - Mixed radium alpha spectrum. Time from T=0 : 303.72 hours.

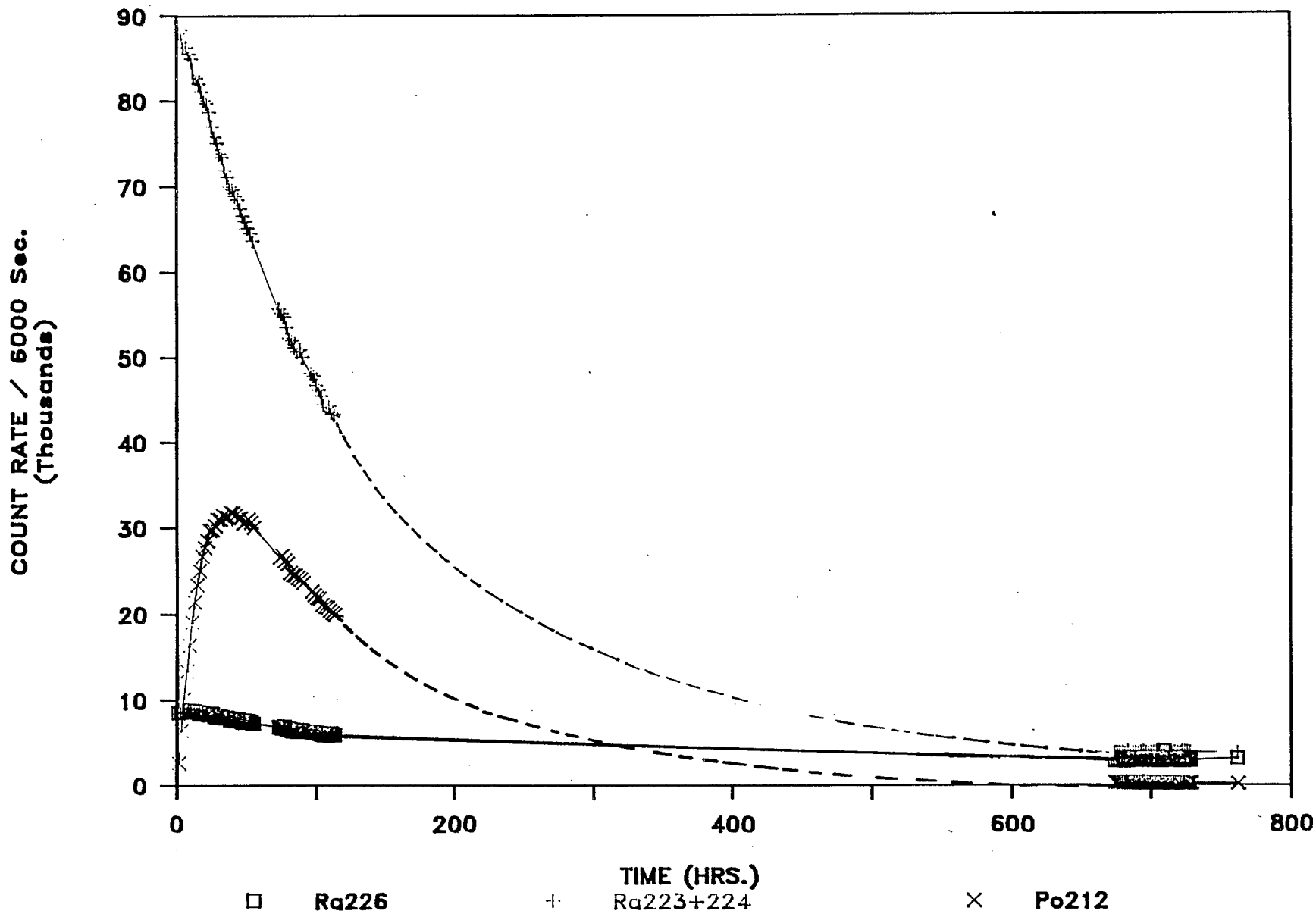


Fig. 8.5 - Decay curve of mixed radium species. (RDE-3) count rate versus time.

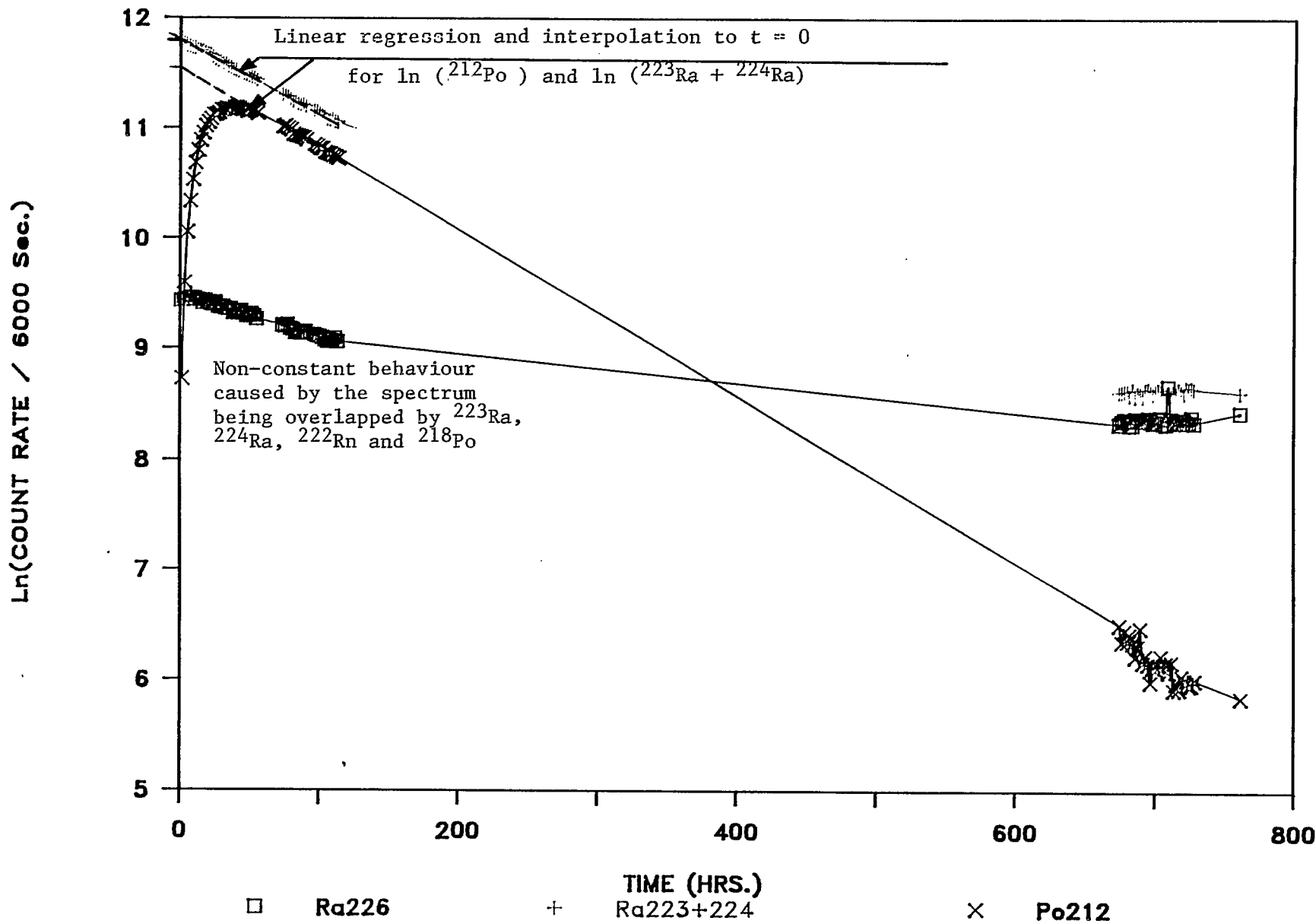


Fig. 8.6 - Decay curve of mixed radium species. (RDE-3) Ln(count rate) versus time.

CHAPTER 9

CONCLUSIONS

At the time when the tailings are placed in its containment, the ratios between daughter/parent isotopes should have been near 1.0 (equilibrium), with the exception of those for the uranium-species which were already extracted in the mill.

The data obtained from the T-5 area in this study showed that the activity ratio between daughter/parent, especially for $^{210}\text{Pb}/^{226}\text{Ra}$ in the liquid phase is as high as 164. This was approximately 200 times larger than the ratio in the solid phase. This anomalous secular disequilibrium should be called "Paradoxical secular disequilibrium" since its behaviour is very irregular and beyond the boundary of the expected ratio for secular equilibrium, which is less than 1.0.

In this study, data from the solid phase were also obtained to demonstrate the hydrogeochemical interaction between liquid and solid phase in the uranium tailings bulk mass. In some cases, these data show the existence of paradoxical secular disequilibrium in the solid phase.

The activity ratios of daughter/parent isotopes for $^{210}\text{Pb}/^{226}\text{Ra}$ isotopes in the solid tailings had values of less than 1.0. However the activity ratios for $^{226}\text{Ra}/^{230}\text{Th}$ and $^{228}\text{Th}/^{232}\text{Th}$ showed an unusual pattern with a high ratio of approximately 20:1 close to the surface of the tailings which then decreased with depth and approached 1.0 at the peat layer.

The mechanism of paradoxical secular disequilibrium in liquid and solid phases can be explained with reference to the basic rule of secular equilibrium, which has often been ignored, that the sample should be undisturbed, and no parent or daughter substances removed or allowed to

escape for a period of time long enough for secular equilibrium to be established. Therefore, in a disturbed and non-isolated system, paradoxical secular disequilibrium can exist:

1. In the solid phase, if the parent is removed by a leaching process, or a daughter is gained by reprecipitation and adsorption at a faster rate than the time required to reach equilibrium, and vice versa.
2. In the liquid phase, if the daughter activity is gained by preferential leaching, or if the parent isotopes reprecipitate at a faster rate than the time needed to reach equilibrium.

The solubilities of the sulphates which increase in value progressively from radium through lead, thorium and uranium (VI) species, is the major controlling factor in radioisotope disequilibrium.

It may be concluded that the behaviour of $^{210}\text{Pb}/^{226}\text{Ra}$ ratio (less than 1.0) in the solid phase is caused by preferential leaching of ^{210}Pb over ^{226}Ra and at a faster rate than the ^{226}Ra production of its daughter ^{210}Pb to reach secular equilibrium. The activity in the liquid phase is controlled by the pH, solubility or chemical equilibrium constant of $^{210}\text{Pb}(\text{SO}_4)$, and the residence time of porewater. This ratio in the liquid phase is sometimes very high, caused by the higher availability of ^{210}Pb with the increased ^{226}Ra activity present in the bulk mass, and higher solubility of lead sulphate with decreases in pH (acidic condition).

A similar process occurs for the $^{226}\text{Ra}/^{230}\text{Th}$ activity ratio in the solid phase, where the thorium sulphate is more soluble than the radium sulphate.

Important differences between the activity ratios of $^{226}\text{Ra}/^{230}\text{Th}$ and $^{210}\text{Pb}/^{226}\text{Ra}$ were observed. The $^{226}\text{Ra}/^{230}\text{Th}$ values are mostly between 1 and 10. These values were a result of the higher solubility of thorium sulphate compared to the radium sulphate, and the slow rate of ^{226}Ra production from

its parent ^{230}Th .

$^{210}\text{Pb}(\text{SO}_4)$, a product of a much faster rate of decay from ^{226}Ra (1.6×10^3 y), has a solubility higher than radium sulphate, but less than thorium sulphate. Thus the $^{210}\text{Pb}/^{226}\text{Ra}$ ratio was close to or less than 1.0 in the solid phase.

In the case of $^{228}\text{Th}/^{232}\text{Th}$ activity ratio, although ^{228}Th and ^{230}Th supposedly behave the same as chemical thorium (same solubility), the result showed a ratio between 20:1 which indicates a chemical segregation to two different elements. A review of the ^{232}Th decay series shows that the ^{228}Th is not a direct daughter of ^{232}Th , but results from several decay chain steps, i.e., ^{232}Th (1.4×10^{10} y) \rightarrow ^{228}Ra (6.7 y) \rightarrow ^{228}Ac (6.1 h) \rightarrow ^{228}Th .

For thorium which undergoes dynamic hydrogeochemical processes, the ^{232}Th (which is more soluble) is leached at a faster rate than ^{228}Ra , while the ^{228}Th is produced indirectly, at a faster rate from ^{228}Ra . It can then be concluded that the decay events, and the solubility of thorium and radium are the major controlling factors in the hydrogeochemical processes that caused the paradoxical disequilibrium between ^{228}Th and ^{232}Th .

This study gives more complete evidence of disequilibrium than studies by other investigators in this field (1,4 to 26,57,58), as it outlines more fully the comprehensive interaction between the solid and liquid phases.

For example, Veska (26) who extensively studied the $^{234}\text{U}/^{238}\text{U}$ ratio in the liquid phase, recognized a continuous trend of relative ^{234}U enrichment in the isolated system, and believed this was caused by preferential leaching of ^{234}U in the 6+ oxidation state, and a stronger absorption of ^{238}U in the 4+ state. This explanation, however, was contradicted by Morin (27) who mentioned that ^{238}U or ^{234}U can exist in a 4+, 5+ or 6+ state.

Based on this study, a scenario can be deduced to explain how the $^{234}\text{U}/^{238}\text{U}$ ratio can result from the ^{238}U chain of decay in the solid phase

where ^{238}U , ^{234}Th , ^{234}Pa and ^{234}U are assumed to be at or near secular equilibrium. As the solid phase is leached by low pH water, the ^{238}U and ^{234}U are leached out at an equal rate as they both behave as chemical uranium. However, ^{234}Th and ^{234}Pa were leached out at a slower rate than the uranium, resulting in retardation of these species as the ^{234}U was being continuously produced. This causes an increase in the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the solid. The increased activity ratio in the solid phase is, of course, reversed in the liquid phase.

Through a simple laboratory experiment (Chapter 8) the impact on sampling, timing and analyses has indicated the parameters required for planning of future work and for evaluating work done in the past.

For example, the explanation given by CBCL Ltd. (58) in regard to $^{234}\text{U}/^{238}\text{U}$ activity ratio of 1.13 average, believed to be caused by mass recoil and ejection of ^{234}U was inaccurate.

As discussed previously in this thesis, ^{234}U is not a direct daughter of ^{238}U because ^{234}Th and ^{234}Pa are produced in between (63). Both ^{234}Th and ^{234}Pa could also be ejected and recoiled by their parents into solution and later adsorbed, precipitated and coprecipitated as the dynamic processes of hydrogeochemistry progress. It was also mentioned by CBCL Ltd. that the uranium deposition is considered to be an on-going process, i.e., the hydrogeochemical process is still very active. Therefore, it can be concluded that the age measurement as reported by CBCL Ltd could be seriously in error as ^{238}U and ^{234}U could be continuously added to the peat layer as the hydrogeochemical processes continue. Thus, caution should be taken in radioactive dating of geological strata and serious consideration should be given to the hydrogeochemical processes.

This study can also be utilized to explain the enrichment of radon gas species in groundwater and to give some information on the pathway to the

surface.

From the results of this study, it is clear that interpretation of data in dynamic situations must be done very carefully, especially when dealing with radiochemistry and related subjects, such as radioisotope dating, pathway analysis of radioisotopes in the food chain of biological matter (biomonitoring).

It is also seen in this study that the solid phase system acts as an indicator of the long term effects of the slow processes of dynamic hydrogeochemical evolution. Solid phase results are extremely valuable, in particular, in the study of the evolution of radioisotope species from uranium tailings. By comparison, the liquid phase system is a much faster, short term indicator of the happenings on a day to day basis. Sometimes the changes are so rapid (such as those connected with the oxidation mechanism of iron species), that complex problems arise in the design of mathematical models for the long term tailings events. The long term modelling could probably be simplified and improved by using only an empirical model based on the solid phase system.

When dealing with hydrogeochemical processes and field data interpretation of radioisotope migration, one must have an in-depth understanding of the physics of radioisotope decay, the chemical behaviour of the isotopes, and the groundwater hydrology of the system. It is clear too, that the timing of sampling, analytical techniques and sample storage for the investigation of disequilibrium phenomena is extremely important.

In conclusion, because of the many ways in which separation or removal of daughter isotopes can occur (hydrogeochemical or gas separation), even in a very tight rock formation with a very low water permeability (e.g., 10^{-9} to 10^{-12} cm/s), true secular equilibrium probably never exists. Many determinations of such equilibrium are limited by the counting statistics of

the instrument.

Most of the paradoxical secular disequilibrium shown in this study and in the data obtained by many other researchers was found by isolating the liquid phase and solid phase (either by natural separation such as in groundwater springs or by manual liquid sample extraction using piezometers). The possibility of paradoxical disequilibrium is often ignored by field investigators in their data interpretation.

Although paradoxical secular disequilibrium actually has nothing to do with the existence of secular equilibrium for series members, it does tell much about the pathways and processes for these isotopes.

RECOMMENDATIONS

Future disequilibrium studies would be aided by more improvements in high resolution alpha-spectroscopy including a convoluted software program to evaluate complex alpha spectra. Methods of direct analyses for ^{238}U , ^{235}U and ^{232}Th series decay chains should also be upgraded for field use (for liquid and solid phases). In situ measurements of gamma spectra using borehole logging techniques as a non-destructive method for field measurement would also give valuable data.

Furthermore, in future research, it is imperative to repeat this study every five years, especially on the Nordic Main tailings area, and include the study of the activity ratio within the liquid phase and the solid phase at the same time. The information will be extremely useful as an aid in the development of mathematical models for the long-term prediction of tailings behaviour.

Variations of sites and types of tailings should also be considered such as Agnew Lake Mine's tailings, Ontario, and a few others in Saskatchewan.

The results of this type of study will also help in decision making

for designing of:

1. future tailings - such as deep water disposal, wet barrier etc.
2. remedial action to correct problems from and past and present tailings deposition using different types of engineering or biological covers such as: consolidated tailings, wet barriers or wet land reducing bacteria, etc.

The objective of the design is to limit the release of contaminants to the environment without much human involvement in the future.

For instance, research involving a wet barrier on top of the tailings is planned for the near future. An impervious dam will be built to contain the porewater, and it is expected that sulphide reducing bacteria will slowly colonize the saturated zone. Under such conditions, it is expected that the contaminant migration will be drastically reduced and thus allow secular equilibrium to exist in the solid phase. Therefore, paradoxical secular disequilibrium can also be used as an indicator of contaminant migration within the uranium tailings deposition.

Another interesting area of study is the scavenging elements and their behaviour, such as the hydroxides of iron, aluminum and manganese as there are possibilities that these elements could be used in the future as scrubbers, and if later on were further processed by nature into insoluble solid minerals (oxidation into hardpan etc.).

Caution should be taken in radioisotope dating of geological strata (or archeological) as there are numerous possibilities where serious errors can vastly mislead the conclusion. One of them is hydrogeochemical interaction between liquid and solid phases. Even in a very tight formation with a permeability of 10^{-9} to 10^{-12} cm/sec, and taking into account the geological time of many thousands of years, significant porewater must have been transported from one place to another, i.e., parent and daughter

isotopes could have been removed or added during that time.

Furthermore, the type of rock could be very important since some types are more soluble than others. However, the chance of making errors is smaller when dealing with the solid phase sample in general, as the solid phase is a much larger sample in mass ratio in comparison to the liquid phase sample. Also, as previously mentioned, the solid phase reacts much more slowly to change of time in comparison to the liquid phase.

The radioisotope chosen should be chemically insoluble and have a half life suitable for the time scale range required. Several activity ratios of the radioisotope members should be done to test the logic.

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