NATIONAL WATER RESEARCH INSTITUTE

INSTITUT NATIONAL DE RECHERCHE SUR LES EAUX

VERTICAL DISTRIBUTION OF URANIUM MILL TAILINGS CONTAMINANTS IN THE LANGLEY BAY, LAKE ATHABASCA SEDIMENTS

S.R. Joshi[1], D.T. Waite[2] and R.F. Platford[1]

NWRI Contribution No. 88-46

VERTICAL DISTRIBUTION OF URANIUM MILL TAILINGS CONTAMINANTS IN THE LANGLEY BAY, LAKE ATHABASCA SEDIMENTS

S.R. Joshi[1], D.T. Waite[2] and R.F. Platford[1]

NWRI Contribution No. 88-46

[1] Lakes Research Branch
National Water Research Institute
Canada Centre for Inland Waters
Burlington, Ontario
L7R 4A6

[2] Environmental Protection
Environment Canada
241-1901 Victoria Avenue
Regina, Saskatchewan
S4P 3R4

VERTICAL DISTRIBUTION OF URANIUM MILL TAILINGS CONTAMINANTS IN THE LANGLEY BAY, LAKE ATHABASCA SEDIMENTS

S.R. JOSHI[1], D.T. WAITE[2] and R.F. PLATFORD[1]

[1]Lakes Research Branch, National Water Research Institute, Canada Centre for Inland Waters, Environment Canada, P.O. Box 5050, Burlington, Ontario L7R 4A6 (Canada)

[2]Environmental Protection, Environment Canada, 241-1901 Victoria Avenue, Regina, Saskatchewan S4P 3R4 (Canada)

ABSTRACT

The Gunnar Mill, located on the north shore of Lake Athabasca in the Uranium City area, generated nearly five million tonnes of tailings which were initially discharged into a nearby small lake. Significant quantities of these tailings subsequenty moved into another small water body in the area and then into Langley Bay, a shallow bay opening onto Lake Athabasca. Analyses of several sediment cores for both radioactive and non-radioactive constituents reveal that the contamination, covering the entire bottom of the bay, has

reached Lake Athabasca. It is estimated that about 11% of the total ^{226}Ra activity discharged from the mill resides in the Langley Bay sediments with about 76% still remaining at the original disposal areas. The ^{137}Cs measurements suggest that natural cover on the submerged tailings will develop at a very slow rate. A very small fraction of ^{222}Rn generated in the Langley Bay delta deposit escapes to the atmosphere.

RÉSUMÉ

L'usine Gunnar, située sur la rive nord du lac Athabasca dans la région de Uranium City, a produit près de cinq millions de tonnes de stériles qui avaient été déversés initialement dans un petit lac voisin. Des quantités importantes de ces stériles se sont ensuite transportées dans un autre petit plan d'eau de la région et ensuite dans la baie Langley, une baie peu profonde s'ouvrant sur le lac Athabasca. L'examen de plusieurs carottes sédimentaires pour déterminer la présence de matières radioactives et de matières non radioactives montre que la contamination a atteint tout le fond de la baie et a même atteint le lac Athabasca.

Executive Summary

The Gunnar uranium mine and mill complex generated about five million tonnes of tailings which were discharged into the nearby Mudford Lake during 1955-64. Subsequently, the tailings were allowed to flow into a small basin and then into Langley Bay which connects with Lake Athabasca, Saskatchewan via a narrow channel. The present study was initiated in 1983 in collaboration with Environmental Protection, Western and Northern Region to assess the movement of the tailings contaminants in Langley Bay. A number of sediment cores were collected and analyzed for both radioactive and non-radioactive constituents. The results from these measurements show that the tailings have covered the entire bottom of Langley Bay and have also reached Lake Athabasca. Natural cover on the submerged tailings will develop at a very slow pace as indicated by Cs-137 profiles. It is estimated that about 11% of Ra-226, the most toxic radionuclide released to the environment , produced at the site has been deposited in Langley Bay, while the bulk (about 76%) still remains at the Mudford Lake disposal site. A very small fraction (o.1%) of Rn-222,the toxic noble gas produced by the radioactive decay of Ra-226, escapes to the atmosphere from the Langley Bay deposit.

Résumé

De 1955 à 1964, la mine et l'usine d'uranium de Gunnar ont déversé environ cinq millions de tonnes de stériles dans le lac Mudford, situé à proximité. Par la suite, on a laissé les stériles passer dans un petit bassin et ensuite dans la baie Langley, qui est reliée au lac Athabasca, Saskatchewan, par un étroit chenal. La présente étude a démarré en 1983, avec la collaboration du Service de protection de l'environnement, région de l'Ouest et du Nord; elle vise à évaluer le mouvement des contaminants contenus dans les stériles de la baie Langley. Plusieurs carottes sédimentaires ont été prélevées et leur teneur en matières radioactives et non radioactives a été évaluée. Les résultats montrent que les stériles ont entièrement couvert le fond de la baie Langley et qu'ils ont également atteint le lac Athabasca. Une couverture naturelle sur les stériles submergés va se former très lentement, comme l'indiquent les profils du Cs-137. Il est estimé qu'environ 11 % du Ra-226, le radionucléide le plus toxique produit au complexe à être libéré dans le milieu, est passé dans la baie Langley alors que la majeure partie (environ 76 %) se trouve toujours dans le lieu de rejet du lac Mudford. Une très petite fraction du Rn-222 (0,1 %), le gaz noble toxique produit par décomposition radioactive du Ra-226, passe du point de rejet de la baie Langley à l'atmosphère.

INTRODUCTION

The Gunnar uranium deposit, located on the north shore of Lake Athabasca, was discovered in 1952 (Beck, 1969). The discovery led to the development of the Gunnar uranium mine and mill complex where more than five million tonnes of ore were processed between 1955 and 1964 (Griffith, 1967). The waste rock was piled adjacent to the open pit on a natural surface of bedrock and silty glacial clays. The untreated tailings, resulting from a sulphuric acid leach and the then-new magnesium diuranate precipitation process, were discharged to Mudford Lake, located about 500 m northwest of the mill. Subsequently, the tailings were allowed to flow into a small basin and then into Langley Bay which connects with Lake Athabasca via a narrow channel.

A preliminary Environment Canada survey of area water pollution revealed the presence of elevated levels of some uranium-series radionuclides in waters, sediments and biota of Langley Bay (Ruggles et al., 1981). The present study was launched in 1983 to assess the distribution and movement of tailings in the local aquatic system. Waite et al. (1988) have described the limnological, chemical and biological aspects of the study and have subsequntly also reported (Waite et al., in press) on the migration of dissolved radionuclides from submerged tailings into the Langley Bay surface waters. Platford and Joshi (1988) have assessed the radiation doses to the Langley Bay biota. In the present article, we report on the

vertical distribution of uranium mill tailings contaminants in Langley Bay sediments and derive estimates of pollutant inventories in three distinct sections of this water body. An attempt is also made to reconstruct the historical pattern of contaminant deposition in the area sediments. Finally, the loss of ^{222}Rn from the relatively dry Langley Bay deposit to the air is also estimated.

MATERIALS AND METHODS

Study area

Figure 1 shows the location of the Gunnar Mine and the tailings discharge system around Langley Bay, Lake Athabasca. Initially, the tailings were deposited in Mudford Lake (now known as Gunnar Main). The tailings flowed into the lake until the basin was filled to capacity at which point additional disposal capacity was gained by demolition of a retaining dam on Mudford Lake and allowing the tailings slurry to flow down into a small basin now known as Gunnar Central. Following this initial event, tailings from the main holding areas moved downhill and into Langley Bay forming a delta bisecting the bay. Run-off from the uphill tailings continues to transport various contaminants into Langley Bay to the present time as no decommissioning steps were taken to contain the movement of tailings following site abandonment. The main part of Langley Bay has a surface area of 75.1 ha and a volume of 2.9×10^6 m^3 while the smaller section, referred to as Back Bay, has a surface area of

18.9 ha and a volume of 3.5×10^5 m^3. A recent survey (BBT Geotechnical Consultants Limited, 1986) revealed that the Gunnar Main tailings grade from fine sand to silty clay while those at Gunnar Central are largely silty sand and silt. The Langley Bay tailings were predominantly silt and silt clay with only minor amounts of silty sand. The depth of tailings varied up to greater than 14 m at Gunnar Main, 3 to 4 m at Gunnar Central, and 2 to 4 m in Langley Bay.

Sediment core samples

Sediment cores from stations 1 to 7, shown in Fig. 1, were obtained with a Brown's piston corer and immediately sliced into 1 cm thick sections. The numbering system of the sediment core sampling stations differs from that reported earlier by Waite et al. (1988, in press) and Platford and Joshi (1988). The embayment denoted as station 7 was initially thought to represent a non-contaminated area and has been referred to as such by Platford and Joshi (1988); however, completion of sediment core analyses led us to consider this station under the zone of impact as well. Attempts to retrieve sediment cores at several other locations within about 1 km north or west of station 6 were unsuccessful.

Analytical techniques

Precise sample thicknesses were obtained by dividing the wet mass of the sediment aliquot by its bulk density and area.

The sediment porosity was measured as desribed earlier (Durham and Joshi, 1980). The sediment core samples were assayed for ^{210}Pb, ^{226}Ra and ^{228}Th by direct counting of the freeze-dried, ground material on a calibrated high-resolution hyperpure germanium planar detector as described earlier (Joshi, 1985a, 1987). A calibrated coaxial detector was used to count the 662-keV gamma-ray emission for deriving the ^{137}Cs contents of the samples. The concentrations of all other elements were determined by instrumental neutron activation analysis. These analyses were performed, under contract, by Becquerel Laboratories, Inc., Mississauga, Ontario, Canada.

RESULTS

The principal sources of radiation hazard in the uranium mining and milling industry are those associated with uranium and its progenies. Of particular concern are the radionuclides ^{226}Ra and ^{210}Pb. The former has a conservative chemical behaviour like that of other group IIA elements and is generally capable of migration in aquatic systems as has been observed at this particular site as well (Waite et al., in press). This radionuclide decays with a half-life of 1620 y and, therefore, persists in the environment for a significant period of time. The chemical behaviour of this radioelement also facilitates its uptake and subsequent incorporation in biota where the energetic alpha-particle emissions from ^{226}Ra and three of its immediate, short-lived daughters cause radiation damage to the living

cells. The decay of ^{210}Pb (half-life, 22.3 y), on the other hand, is accompanied by the emission of only low-energy beta particles but two of its immediate daughters, ^{210}Bi (half-life, 5 d) and ^{210}Po (half-life, 138 d), decaying with the emission of strong beta and alpha particles, respectively, are capable of causing significant radiation damage. The slow emission of relatively low-energy alpha particles from the long-lived (half-life, 4.5×10^9 y) principal uranium isotope, ^{238}U, reduces the radiological significance of this radionuclide when compared with that of ^{226}Ra; however, the known chemical toxicity and tendency to migrate in the sediment-pore water subsystem enhance the general environmental significance of uranium.

A typical uranium ore also contains significant amounts of another naturally-occurring radioelement, thorium. Very few measurements on the distribution of thorium-series radionuclides in various environmental matrices are available. The long-lived (half-life, 1.39×10^{10} y) principal thorium isotope, ^{232}Th, decays with the emission of an alpha particle of energy slightly lower than that of ^{238}U to the beta-emitting ^{228}Ra (half-life, 6.7 y). The next member of the series, ^{228}Ac, has a half-life of only 6.1 h and, following beta particle emission, transforms to ^{228}Th (half life, 1.9 y), the decay of which is accompanied by the emission of an alpha particle. The seven ^{228}Th progenies, with half-lives ranging between fraction of a second and less than four days, all emit strong alpha or beta

particles. Thus, from the radiological point of view, this segment of the thorium-series is considerably more important.

We have measured both ^{228}Th and total thorium (i.e., principally ^{232}Th) in all samples and the results are shown in Fig. 2 along with those for ^{210}Pb, ^{226}Ra and total uranium (i.e., principally ^{238}U). At some of the stations, notably stations 1, 2 and the tailings pile, the compression of deposited material prevented raising of sufficiently long cores. In nearly all other cases, all the radioactive constituents show almost identical concentration-depth profiles for a given station. The only major exception is the total uranium profile at station 5. This station is most subject to wave action and also mixing of Langley Bay and Lake Athabasca waters and as a result the deposited materials undergo physical and chemical changes. The observed concentration-depth profiles, therefore, represent only frequently redeposited, and possibly chemically remobilized, material. The general absence of distinct contaminant pulses, as are present in profiles from the adjoining stations 4 and 6, also indicate the susceptibility of station 5 sediment to physical disturbance.

The presence of potentially toxic, nonradioactive elements in uramuim ores, leach solutions or mill effluents generally has not been regarded as a significant problem in studies designed to assess the impact of this industry on the aquatic environment. We have attempted to study the distribution of a number of trace elements that are frequently enriched in uranium ore of

epigenetic origin (Dreesen et al., 1982), the predominant ore type in the study area (McMillan, 1978). Following Dreesen et al. (1982) we have chosen V, As, Se and Mo as the uranium analogue elements for their common geochemical behaviour. Cobalt, Ni and Zn were selected as the elements generally associated with sulphide minerals. The concentration-depth profiles of Ag and Au were also measured since precious metals are often found associated with these uranium deposits (McMillan, 1978). In addition, the concentration-depth profiles of the major elements Na, Mg, Al, K, Ca, Fe and Mn were also measured. The latter two elements are solubilized during acid leaching of uranium ore and are also susceptible to interstitial redox reactions in the deposited wet sediment. Magnesium was also used in the process to precipitate uranium as magnesium diuranate, the final product.

The measured concentration-depth profiles of non-radioactive elements are shown in Figs. 3 to 6. The uranium analogue elements (Fig. 3) show trends similar to those observed for total U (Fig. 2) except that Mo shows some departure from the expected trend at stations 3, 5 and 6. Selenium was present in measurable amounts only in the tailings core and at station 1. Of the sulphide mineral elements (Fig. 4), in most cases Ni was present at levels below the detection limit of the technique used. Both Co and Zn generally show behaviour somewhat different from that expected on the basis of radioactive constituent profiles (Fig. 2). This may be attributable to the

acidic and oxidizing conditions which would likely change sulphide minerals to more soluble forms. The levels of Ag and Au in most sediment core segments (Fig. 5) were below the limits of detection. At station 4, the Au concentration-depth profile is significantly different from those for radioactive materials (Fig. 2). As expected, most of the concentration-depth profiles of major elements (Fig. 6) do not show much variation from the apparent average levels. The variations in the Fe and/or Mn concentrations at stations 2 to 6 may be attributable to redox reactions. Upward migration of soluble Mn under reducing conditions in sediment pore waters with surface precipitation by oxidation has been demonstrated (Weiler, 1973). More detailed studies are required to define the role of redox reactions in the mobilization of such elements in sulphate-rich aquatic environments.

DISCUSSION

The fate of uranium mill tailings contaminants depends both upon physical and chemical processes. The disposal of tailings as slurries provides for particle size fractionation as a result of differential settling of sands, silts and clays. Such particle size fractionation in the study area is indicated from a recent survey (BBT Geotechnical Consultants Limited, 1986). The tailings impoundment areas receive tailings solids (sands, clays, salts, hydrous oxide coatings, etc.), mill process waters, spent leach solutions, and other miscellaneous items

such as flocculating agents and filter aids. Soluble components in the tailings may precipitate, form salts, seep into underlying strata or may be carried away with the run-off. All these processes result in heterogeneous distribution of elements within the tailings pile when compared with that in the original ore as has been shown by Dreesen et al. (1982). The Langley Bay tailings are thus likely further distinguished from those at Gunnar Central and Gunnar Main in their chemical composition. Since all these three likely distinct tailings deposits impact the local aquatic system, it may be possible that the observed variations in concentration-depth profiles (Figs. 2 to 6) merely reflect inputs of variable chemical composition. Thus, more work is required to ascertain whether the observed variations in the concentration-depth profiles are due to the internal evolution of the deposited sediments or simply reflect the composition of the material entering the main bay.

The ongoing influence of tailings contaminants on the main bay and Lake Athabasca sediments may be inferred from a consideration of the Sediment Enrichment Factor (SEF). Kemp et al. (1976) have proposed that the Al content of fine-grained clay minerals in Lake Erie bottom sediments may be used to normalize the surface concentrations of elements to pre-colonial levels. The SEF is then a measure of the degree of normalcy of the most recent sediment being deposited. Since, as noted earlier in the text, the Langley Bay tailings and sediments are predominantly comprised of silt and silty clay with only minor

amounts of silty sand and since Na, Mg, Al, K, Ca and Fe are the dominant inorganic elements found in the bottom sediments of Langley Bay and Lake Athabasca, the SEF may be defined as

$$SEF = \frac{\frac{C_s}{Al_s} - \frac{C_b}{Al_b}}{\frac{C_b}{Al_b}} ,$$

where C_s and C_b are the measured concentrations of radionuclide or element in the surface and bottom (background) segments of the sediment core, respectively, and Al_s and Al_b, respectively, are the concentrations of Al in the corresponding segments of the sediment core. This definition of the SEF is the same as that given by Kemp et al. (1976) except that background concentrations are used in place of pre-colonial concentrations used for the Lake Erie sediments. Positive values indicate increased concentration in the surface sediment, the size of the SEF representing the degree of concentration increase. Negative values indicate loss of element or radionuclide.

The computed SEFs at locations where background levels could be discerned (stations 3, 4, 6 and 7) are given in Table 1. The SEFs for Ni, Ag and Au were not computed as the levels were below the limits of detection. Kemp et al. (1976) have designated SEF values ranging between -0.2 and +0.2 as zero, implying constant element 1 concentration in the sediment core. Zero SEF values would be expected for major elements Na,

Mg, K and Ca which have conservative behaviour. The observed fluctuations in the SEF values of these elements (Table 1) are suggestive of nonuniform deposition of these major matrix materials. It should be noted that significant quantities of Na and Mg were also used in the milling process and subsequently released to the tailings impoundments. Iron and Mn show the highest and consistent enrichment among the major elements. As noted earlier in the text, we do not have other pertinent measurements to assess the enrichment and potential subsequent migration of these elements. The SEF values for Co and Zn suggest marginal inputs to and losses from bottom sediments. With the exceptions of Se at station 7 and Mo at station 6, U and its analogue elements show consistent enrichment in all cases. Most significant enrichment is observed at station 3, while the station 6 sediments show the highest U enrichment. By far the most consistent and significant SEF values are obtained for the radioactive constituents, especially for the major tailings contaminants, ^{210}Pb and ^{226}Ra. Most of the surface sediment contamination persists in the main bay and the nearby station 6. The continuing contamination of Lake Athabasca is evident from the contaminant concentration-depth profiles (Figs. 2 to 5) and SEF values (Table 1) obtained at stations 6 and 7.

The continuing contamination of main Langley Bay surface waters has been recently demonstrated by Waite et al. (in press). It was found that nearly 80% of the ^{226}Ra in the

surface water could be accounted for by diffusion of this radionuclide from submerged tailings. Much smaller releases of other radionculides were also noted. It is possible that the released radionuclides may subsequently be deposited elsewhere along with naturally sedimenting particles. Thus, two potential routes are now known for the spread of contaminants in the sediments of main Langley Bay and Lake Athabasca: physical migration of contaminated tailings and leaching and subsequent deposition of contaminants. Of the two potential routes, physical migration of contaminated tailings is likely to provide the type of SEFs noted earlier (Table 1) since radionuclides such as ^{210}Pb and ^{228}Th as well as U showed little migration in the dissolved form (Waite et al., in press).

In order to estimate the recent (i.e., subsequent to the initial, sudden removal of tailings to Langley Bay) inputs of contaminants to the area sediments, the ^{137}Cs profiles in the sediment cores (Fig. 7) were also examined. This radionuclide was introduced into the environment as a result of extensive atmospheric testing of nuclear weapons in the 1950's and 1960's. Large-scale weapons testing in the northern hemisphere started in 1954, increased significantly in 1958-59, and peaked in 1962-63. Since then only a few atmospheric tests have been conducted by China and fallout debris has decreased substantially. A typical ^{137}Cs profile in an undisturbed lake sediment core records at least the 1958-59 horizon and the 1962-63 peak activity provided the prevailing sedimentation rate permits

adequate sampling resolution (Durham and Joshi, 1980; Joshi, 1985b).

A comparison of the profiles shown in Figs. 2 and 7 reveals that both ^{137}Cs and radioactive contaminants have similar trends at most stations, the only definitive exception being station 4 where peak ^{137}Cs activity is observed closer to the sediment/water interface rather than at the 10 to 15 cm depth as is the case with tailings contaminants. The suggested incompatibility between the ^{137}Cs and other radioactive contaminant profiles at station 2 is not definitive since no concentration gradient could be established for any of the tailings contaminants in this short core. The observation of this general similarity between the trends exhibited by these radionuclides leads to the reckoning that the deposition of ^{137}Cs and tailings constituents occurred simultaneously. The absence of the 1958-59 ^{137}Cs activity horizon and of ^{137}Cs signals from prior atmospheric inputs suggests the prevalence of very low natural sedimentation rates at these locations. Low deposition of natural sediment is also indicated by the observation that the cessation of the atmospheric nuclear weapons testing since about 1963 is also not reflected in the ^{137}Cs profiles (Fig. 7). The ^{137}Cs inventories (Table 2) also indicate that the inputs and deposition of this radionculide at study locations are controlled by factors other than natural sedimentation of direct fallout ^{137}Cs. At all stations, most notably at stations 3, 4, 6 and 7 where nearly complete ^{137}Cs profiles are available, the measured ^{137}Cs

activity inventory is significantly lower than that expected from direct fallout. It is also worth noting that when the direct fallout on the entire Langley Bay drainage basin and its subsequent partial removal to the bay are considered, the total ^{137}Cs available for deposition in the bay would be much higher than that provided by direct fallout alone. Thus it is reasonable to infer that little natural sediment is available to remove ^{137}Cs to the bottom sediments. A corollary to this observation provides that natural cover on tailings submerged in Langley Bay will develop very slowly.

The measured inventory of ^{137}Cs in the Langley Bay tailings deposit suggests that most of the atmospheric ^{137}Cs impinging on the Langley Bay drainage basin was trapped by the finer particles which are more numerous in this deposit. Thus it may be hypothesized that ^{137}Cs acts as a coincidental tracer for these contaminants. From the foregoing observations, it may be postulated that the initial, sudden removal of tailings to Langley Bay and Lake Athabasca around 1959-60. Most of the contamination stayed in the delta bisecting the bay, but significant amounts were dispersed in the bay covering its entire bottom as can be inferred from the concentration-depth profiles (Figs. 2-6) and the radioactive contaminant inventories (Fig. 8) inferred from analytical data. Undoubtedly, tailings were also deposited in the nearby deeper sections of Lake Athabasca as well where, as noted earlier in the text, we were unable to extract any sediment cores. The inventory estimates (Fig. 8 and

Table 2) also show that, as expected, next to the tailings delta the highest ^{137}Cs and tailings contaminants levels are observed in Back Bay, now an enclosed water body.

Radioactive materials are by far the most significant contaminants residing in the Langley Bay tailings/sediments. Of these, ^{226}Ra is radiologically the most toxic contaminant. It is estimated (Ruggles et al., 1981) that about 10^{14} Bq of this radionuclide were discharged from the Gunnar operations. The average depth of the Langley Bay delta tailings is about 3 m (BBT Geotechnical Consultants Limted, 1986), while the surface area appears to be about 0.1 km^2. Using the average ^{226}Ra concentration of 25 Bq/g and the average dry mass per unit volume of about 1.17×10^6 g m^{-3}, we estimate that about 8.8×10^{12} Bq ^{226}Ra resides at this location. Similar calculations using analytical data from the present study, and assuming uniform deposition to a depth of 0.25 m, show that Langley Bay and Back Bay bottom sediments contain about 0.7×10^{12} Bq and 1.3×10^{12} Bq, respectively, of ^{226}Ra. Taken together, the three distinct sections of this water body account for about 11% of the total ^{226}Ra discharged from the mill.

Preliminary estimates of ^{226}Ra inventories at Gunnar Main and Gunnar Central sites may be derived assuming the tailings at these two locations have ^{226}Ra concentrations and dry mass distributions similar to those prevailing in Langley Bay delta tailings. These assumptions may be somewhat unrealistic in view of the known fractionation of tailings as mentioned earlier in

the text. However, in the absence of the availability of any pertinent data, these assumptions may be deemed adequate for arriving at preliminary estimates. The Gunnar Main and Gunnar Central tailings deposits are considered to be about 14 and 3.5 m deep, respectively (BBT Geotechnical Consultants Limited, 1986). The surface area of the Gunnar Main deposit is taken to be 16 ha (Kalin, 1981), while that of the Gunnar Central site is assumed to be 10 ha. Using these parameters, we estimate that the Gunnar Main site contains over 65.5×10^{12} Bq, or about 66% of the total ^{226}Ra released from the mill. The Gunnar Central site contains about 10.3×10^{12} Bq, or over 10% of the total ^{226}Ra released from the mill. Taken together, the three major tailings sites account for about 87% of the ^{226}Ra released from the mill. The balance of the contamination may have thus escaped to Lake Athabasca or spread to areas adjacent to main tailings impoundments. It should be noted that this estimate is based on the measured surface ^{226}Ra concentration, which is then assumed to be constant throughout the depth of the deposit. In actual practice, the tailings surface is flushed out on a seasonal basis (spring runoff and summer rains) with the possibility that the deeper tailings may contain higher levels of ^{226}Ra.

As shown in Fig. 9, the contaminant concentration-depth profiles may reflect up to six distinct phases of uranium mining and milling operations in the area. Phase 1 essentially covers the pre-operational time period. The uranium ore was discovered

in 1952; the exploration activities did not result in any extraneous inputs of contaminants to Langley Bay and, therefore, only natural background levels of radionuclides prevailed in the sediments. Phase 2 spans the time period involving start-up and pilot plant activities which continued until commencement of production in 1955. This phase should reflect little, if any, input of contaminants in the sediment core profiles. Phase 3 is characterized by the discharge of tailings to Mudford Lake (Gunnar Main). The highly acidic liquid effluent consisted primarily of dissolved radionuclides and some suspended materials. Undoubtedly, some of this material was removed to and subsequently deposited in Langley Bay where it should appear as a distinct signal above natural background levels in sediments. The emergence of this signal is particularly noticeable at stations 3 and 4 (Fig. 2). The demolition of a retaining dam on Mudford Lake, and consequential sudden release of slurried tailings to the Langley Bay aquatic ecosystem, is the main feature of Phase 4. Large amounts of tailings were released to the bay over a rather short period of time during the pendency of this phase. The broad peaks appearing in the contaminant profiles (Fig. 2) for stations 3 and 4 likely include contribution from this phase. Phases 3 and 4 coincided with the onset and peak ^{137}Cs fallout period. The duration of Phase 5 is characterized by likely further inputs of subsequent discharges from the mill as well as settling of particulates. This phase is not reflected in the contaminant profiles (Fig. 2) for

station 3, indicating that from about this phase onward the tailings migrated largely along the southern shore of Langley Bay. A significant portion of the contaminant profiles for station 4 (Fig. 2) is undoubtedly derived from this phase though the suggested increase (due to settling of fines) in signal intensity (Fig. 9) is not discerned. Phase 6 refers to the conditions likely to prevail following the 1964 closure of mining and milling operations. These could involve return to natural dynamic equilibrium or continuous limited discharges; fluctuations could be caused by natural events such as runoff, storm activity, etc. This phase is not readily distinguished from Phase 5 in the contaminant concentration-depth profiles.

The ^{226}Rn emissions from uranium mill tailings have long been recognized as a potential health hazard. This noble gas can diffuse from the tailings and be transported from its point of origin by prevailing winds before decaying to a longer-lived progency, ^{210}Pb. The diffusion of ^{222}Rn is considerably affected by the water content of the porous medium. The measured porosities of sediment cores are shown in Fig. 10. The porosities in the Langley Bay cores average about 0.8, indicating that we can approximate any diffusion process by molecular diffusion in water. The Langley Bay delta tailings, however, are drier with a porosity of about 0.52. To calculate a limiting rate from these tailings, we have used the value of 10^{-5} cm^2 s^{-1} for the radon diffusion coefficient of pure water and a tortuosity of 0.66 (Nielson and Rogers, 1982). The

effective value of pore radon diffusion coefficient in the presence of water is then $0.52 \times 0.66 \times 10^{-5}$ cm^2 s^{-1} or 0.34×10^{-5} cm^2 s^{-1}. Using this value and analytical data as applicable, we derive from Fick's law a ^{222}Rn flux of about 2×10^{-5} Bq $cm^{-2} \cdot s$. The annual ^{222}Rn loss to the air from the 0.1 km^2 surface area of the Langley Bay delta tailings is then about 6×10^{11} Bq. The yearly production rate of ^{222}Rn at this deposit is estimated to be about 6×10^{14} Bq from the ^{226}Ra inventory of 8.8×10^{12} Bq. Thus, only about 0.1% of the total ^{222}Rn generated at this site escapes to the atmosphere, the rest remaining trapped or dissolved in the tailings-pore water subsystem. Furthermore, in support of this estimate, it can be shown that ^{222}Rn will migrate only about 2.5 cm during its mean life of 5.5 d. The mean life of ^{220}Rn, derived from ^{228}Th, is only about a minute, so its migration is even less significant.

In summary, the movement of uranium mine tailings from the Gunnar Main and Gunnar Central disposal sites has deposited substantial amounts of both radioactive and non-radioactive pollutants in the bottom sediments of Langley Bay. The results from the present study show that smaller amounts of these contaminants have also reached Lake Athabasca. It is estimated that the Langley Bay sediments contain about 11% of the total ^{226}Ra activity discharged from the mill. Some of this ^{226}Ra diffuses into the overlying waters, as has been demonstrated by Waite et al. (in press), and is the prime contributor to the degradation of radiological water quality of Langley Bay. The

development of natural cover on the submerged tailings appears to be a very slow process. Only about 0.1% of the ^{222}Rn produced in the Langley Bay delta deposit escapes to the atmosphere.

ACKNOWLEDGEMENTS

The competent technical assistance provided by J.E. Tozer, S.P. Thompson and J.A. FitzGerald is gratefully acknowledged.

REFERENCES

BBT Geotechnical Consultants Limited, 1986. Gunnar field study summary report. Supply and Services Canada, Hull, Québec, Contract 15SQ.2341-4-1674X.

Beck, L.S., 1969. Uranium deposits of the Athabasca Region, Saskatchewan. Province of Saskatchewan, Department of Mineral Resources, Regina, Report 126.

Dreesen, D.R., J.M. Williams, M.L. Marple, E.S. Gladney and D.R. Perrin, 1982. Mobility and bioavailability of uranium mill tailings contaminants. Environ. Sci. Technol., 16: 702-709.

Durham, R.W. and S.R. Joshi, 1980. Recent sedimentation rates, ^{210}Pb fluxes, and particle settling velocities in Lake Huron. Chem. Geol., 31: 53-66.

Griffith, J.W., 1967. The uranium industry - its history, technology and prospects. Energy, Mines and Resources Canada, Ottawa, Queen's Printer Cat M38-5/11.

Griffith, J.W., 1967. The uranium industry - its history, technology and prospects. Energy, Mines and Resources Canada, Ottawa, Queen's Printer Cat M38-5/11.

Joshi, S.R., 1985a. Determination of thorium-228, thorium-230 and thorium-232 in sediments by anion exchange and nuclear spectrometry. Anal. Chem., 57: 1023-1026.

Joshi, S.R., 1985b. Recent sedimentation rates and ^{210}Pb fluxes in Georgian Bay and Lake Huron. Sci. Total Environ., 41: 219-233.

Joshi, S.R., 1987. Nondestructive determination of lead-210 and radium-226 in sediments by direct photon analysis. J. Radioanal. Nucl. Chem., Articles, 116: 169-182.

Kachanoski, R.G. and E. de Jong, 1984. Predicting the temporal relationship between soil cesium-137 and erosion rate. J. Environ. Qual., 13: 301-304.

Kalin, M., 1981. A preliminary assessment of the environmental conditions of two abandoned uranium mill tailings sites in Saskatchewan. Environment Canada, Edmonton, Report EPS-WNR-5-81-1.

Kemp, A.L.W., R.L. Thomas, C.I. Dell and J.-M. Jaquet, 1976. Cultural impact on the goechemistry of sediments in Lake Erie. J. Fish. Res. Board Can., 33: 440-462.

McMillan, R.H., 1978. Genetic aspects and classification of important Canadian uranium deposits. CIM Bull., 71: 61-67.

Nielson, K.K. and V.C. Rogers, 1982. A mathematical model for radon diffusion in earthen materials. U.S. Nuclear Regulatory Commission, Washington, Report NUREG/CR-2765.

Platford, R.F. and S.R. Joshi, 1988. Dose rates to aquatic life near a U waste site. Health Phys., 54: 63-68.

Ruggles, R.G., D.J. Robinson and A. Zaidi, 1981. A study of water pollution in the vicinity of two abandoned uranium mills in northern Saskatchewan, 1978. Environment Canada, Edmonton, Report EPS-WNR-5-81-2.

Waite, D.T., S.R. Joshi and H. Sommerstad, 1988. The effect of uranium mine tailings on radionuclide concentrations in Langley Bay, Saskatchewan, Canada. Arch. Environ. Contam. Toxicol., 17: 373-380.

Waite, D.T., S.R. Joshi and H. Sommerstad, in press. Movement of dissolved radionuclides from submerged uranium mine tailings into the surface water of Langley Bay, Saskatchewan, Canada. Arch. Environ. Contam. Toxicol.

Weiler, R.R., 1973. The interstitial water composition in the sediments of the Great Lakes. I. Lake Ontario. Limnol. Oceanogr., 18: 918-931.

FIGURE CAPTIONS

Fig. 1 Map of study area showing the Gunnar Mine and associated tailings discharge system along with sediment core sampling locations.

Fig. 2 The concentration-depth profiles of radioactive constituents in sediment cores.

Fig. 3 The concentration-depth profiles of uranium analogue elements in sediment cores.

Fig. 4 The concentration-depth profiles of Co, Ni and Zn in sediment cores.

Fig. 5 The concentration-depth profiles of Ag and Au in sediment cores.

Fig. 6 The concentration-depth profiles of major elements in sediment cores.

Fig. 7 The concentration-depth profiles of ^{137}Cs in sediment cores.

Fig. 8 Inventories of radioactive contaminants in the study area.

Fig. 9 Postulated records of various phases of uranium mining and milling activities in the bottom sediments.

Fig. 10 Porosity profiles in sediment cores.

TABLE 1

Sediment enrichment factors for radioactive and nonradioactive constituents

Radionuclide/element	Station number			
	3	4	6	7
Radioactive constituents				
^{210}Pb	64.4	100.8	181.6	11.3
^{226}Ra	105.9	84.4	25.6	9.2
^{228}Th	6.2	3.0	3.7	0.7
Th	9.9	3.9	5.9	0.5
U	20.7	0.5	53.4	1.1
Uranium analogue elements				
V	5.7	0.1	0.3	0.4
As	44.6	2.1	0.8	1.5
Se	0.4	-	-	-0.5
Mo	1.0	-	-1.0	3.1
Sulphide mineral metals				
Co	-0.2	0.1	-0.1	0.1
Zn	1.3	-0.4	-0.1	2.4
Major elements				
Na	1.0	1.2	-0.4	2.3
Mg	1.0	-0.4	-0.4	0.2
K	0.5	-0.5	-0.2	-0.2
Ca	-0.5	-0.6	-0.3	0.5
Fe	3.2	1.8	0.1	0.4
Mn	0.3	2.5	1.0	0.2

TABLE 2

Inventories of ^{137}Cs at study locations

Station number	^{137}Cs (Bq m^{-2})
1	>1320
2	>400
3	385
4	≥1080
5	>515
6	940
7	660
Tailings	>4500
Direct fallout[a]	~2800

[a] Based on the estimated accumulation of decay-corrected ^{137}Cs in noneroded soil in Saskatchewan reported by Kachanoski and de Jong (1984).

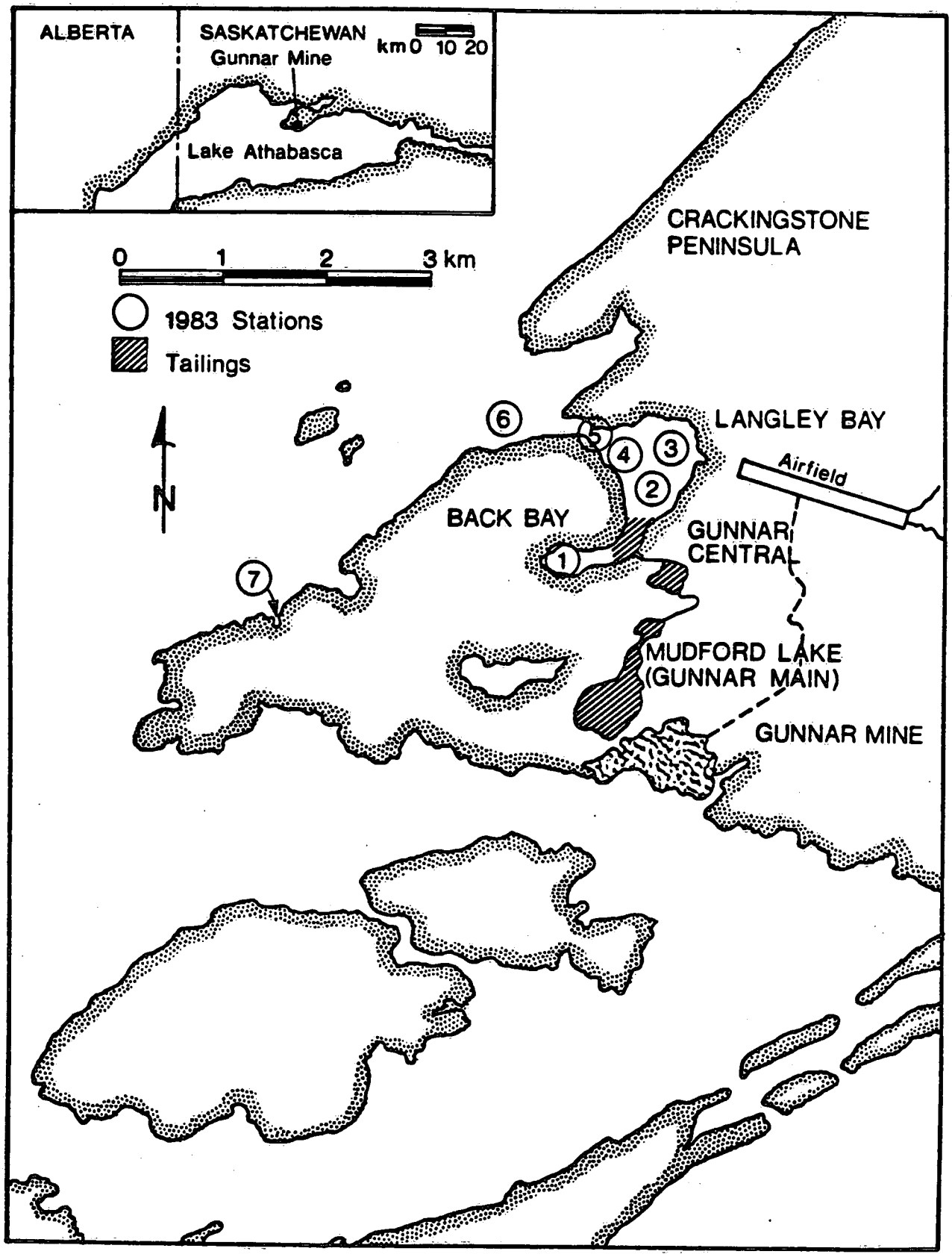

ALBERTA
SASKATCHEWAN
Gunnar Mine
km 0 10 20
Lake Athabasca
0 1 2 3 km
1983 Stations
Tailings
N
CRACKINGSTONE PENINSULA
LANGLEY BAY
Airfield
BACK BAY
GUNNAR CENTRAL
MUDFORD LAKE (GUNNAR MAIN)
GUNNAR MINE
1
2
3
4
5
6
7

Fig. 1

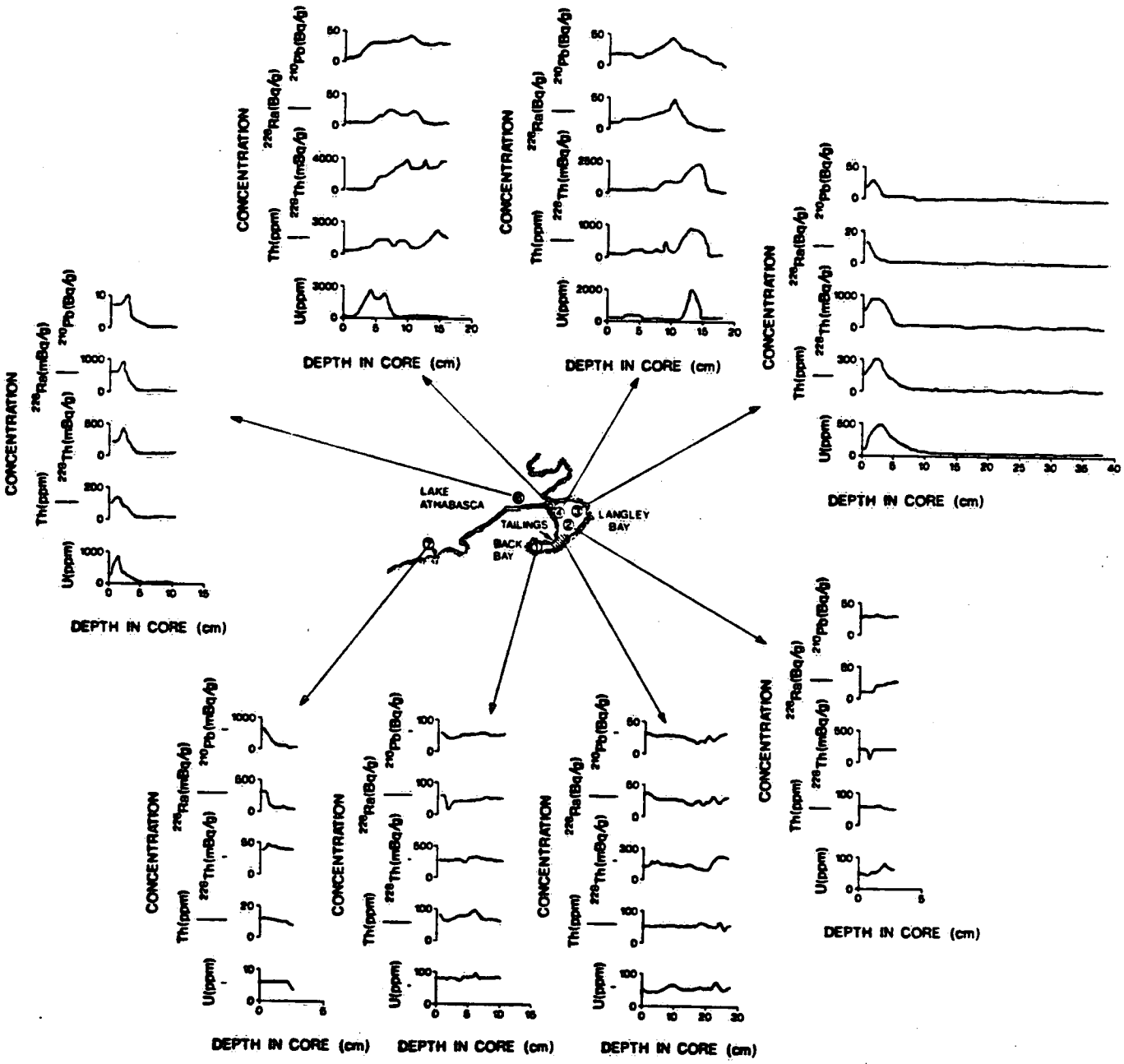

CONCENTRATION
^{210}Pb(Bq/g)
^{226}Ra(Bq/g)
^{226}Th(mBq/g)
Th(ppm)
U(ppm)
DEPTH IN CORE (cm)
LAKE ATHABASCA
TAILINGS
BACK BAY
LANGLEY BAY

Fig. 2

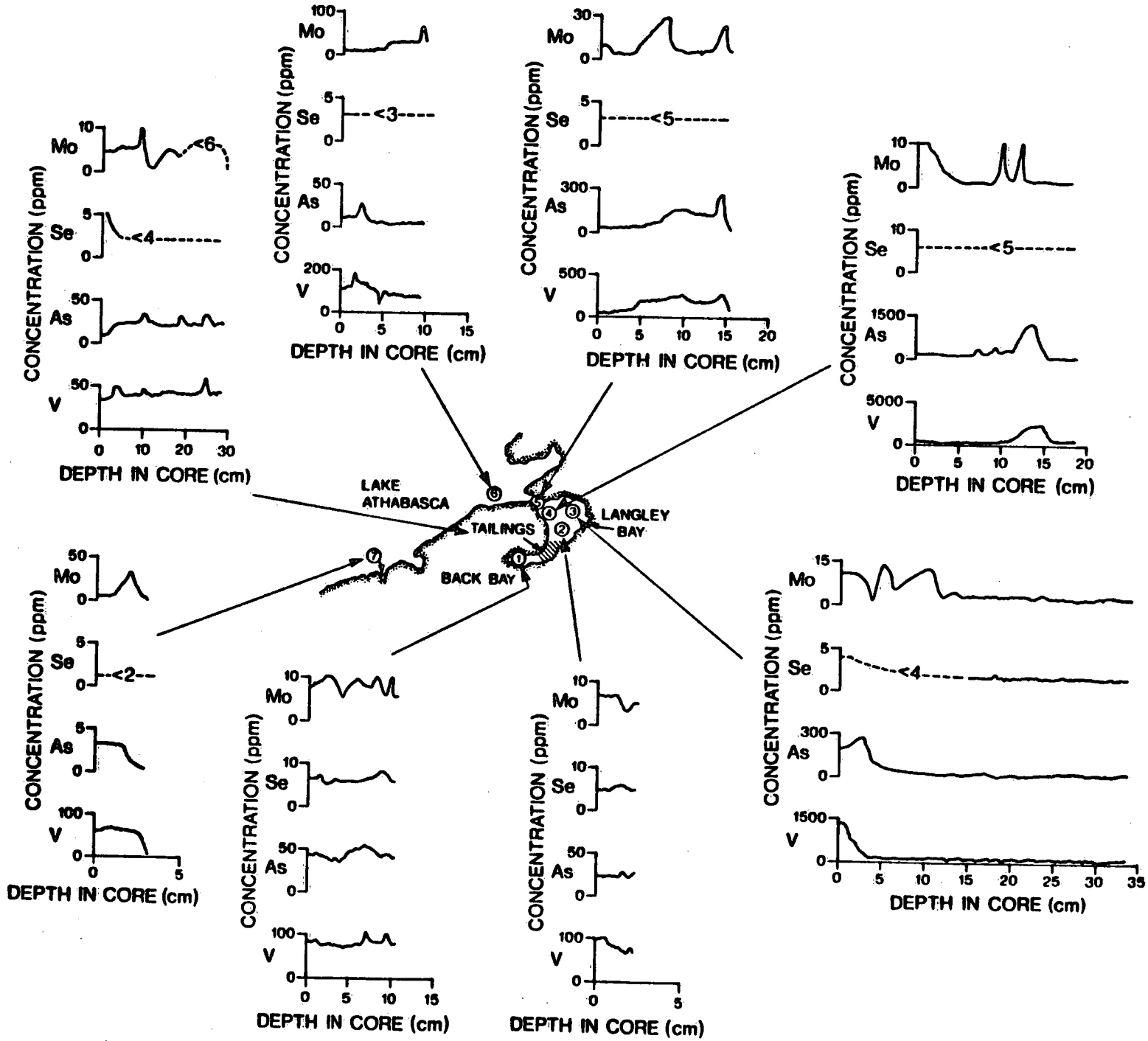

LAKE ATHABASCA
TAILINGS
LANGLEY BAY
BACK BAY
CONCENTRATION (ppm)
Mo
Se
As
V
DEPTH IN CORE (cm)
<6
<4
<3
<5
<5
<2
<4

Fig. 3

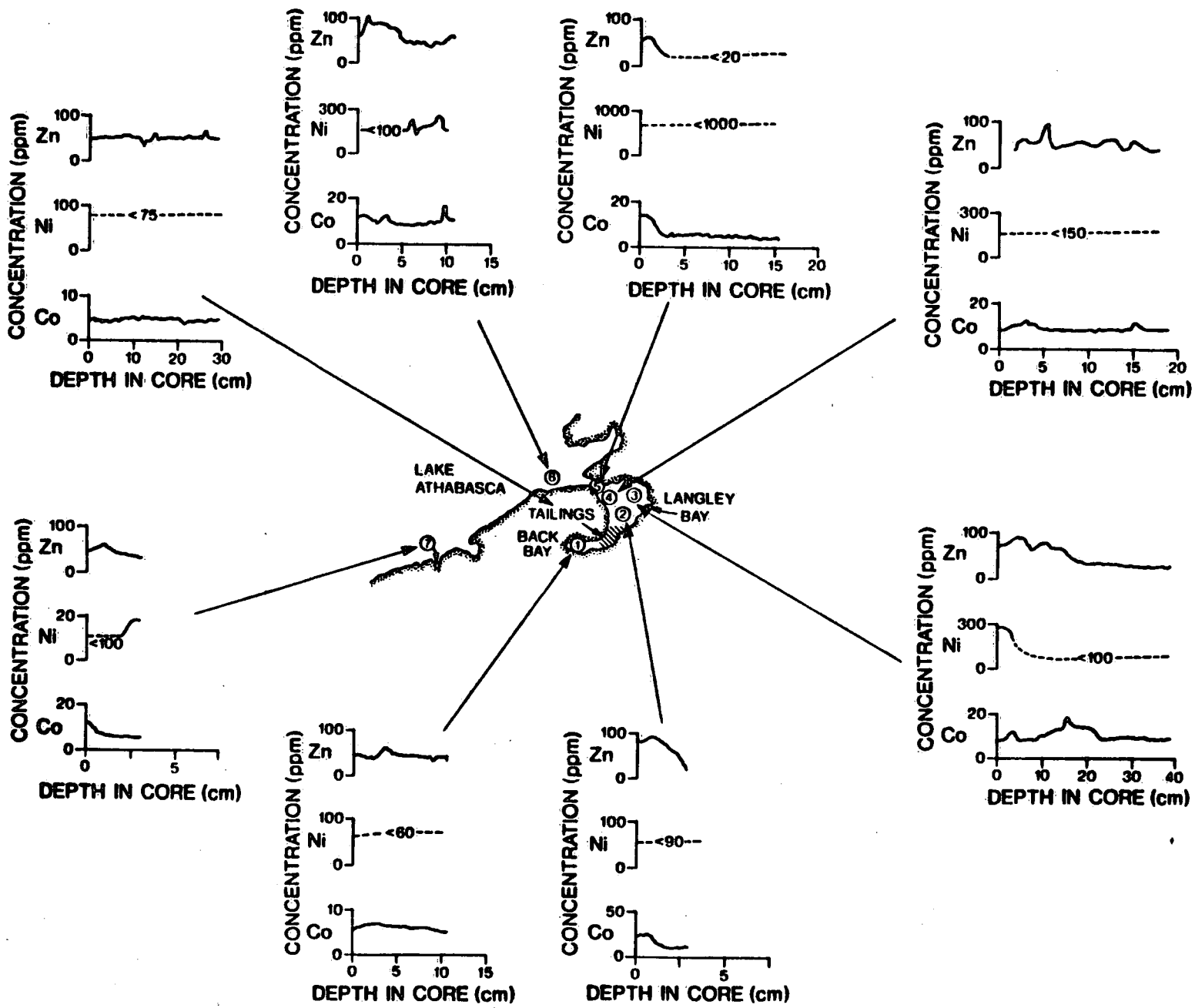
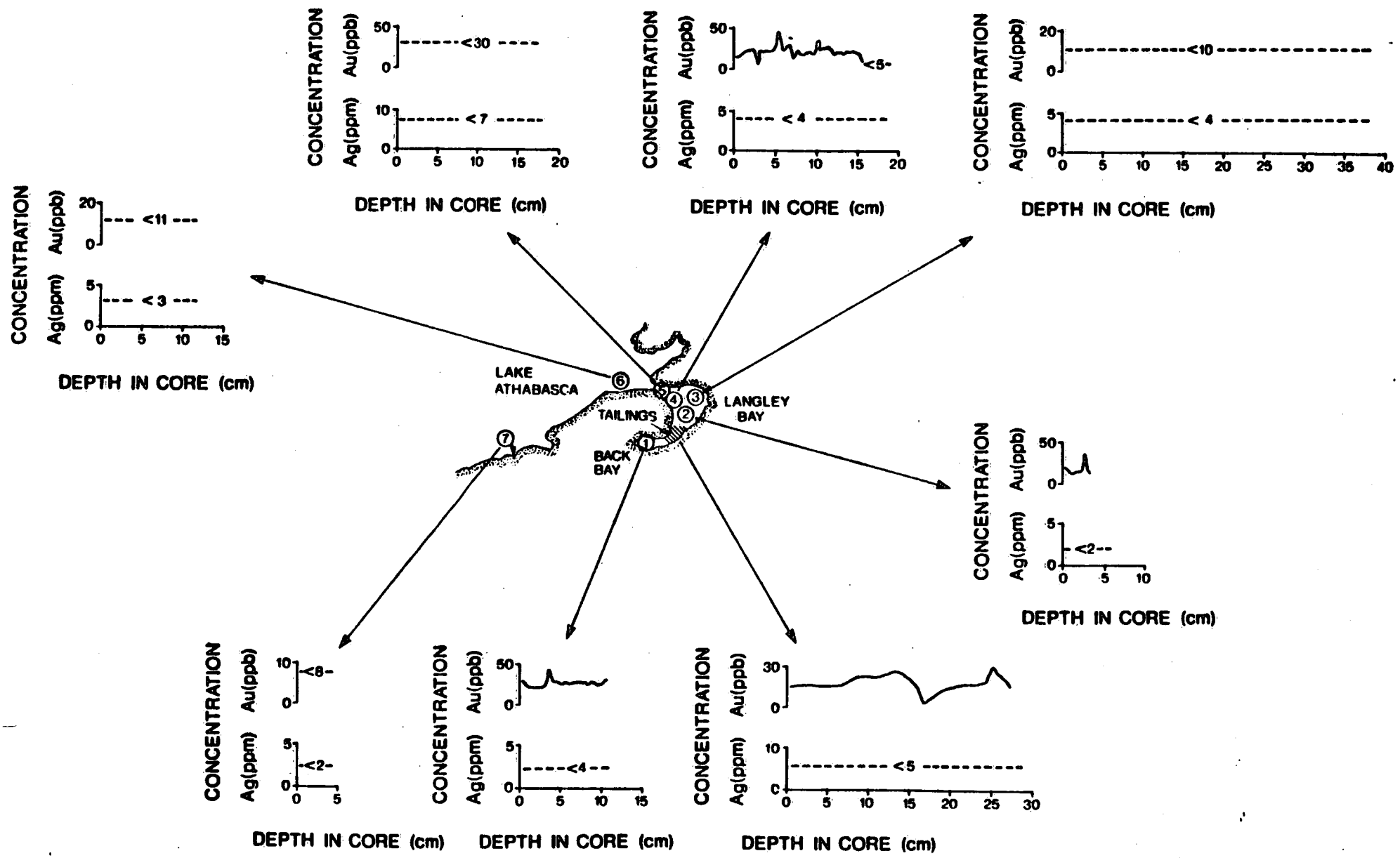

LAKE ATHABASCA
TAILINGS
BACK BAY
LANGLEY BAY
CONCENTRATION (ppm)
Zn
Ni
Co
<75
<100
<20
<1000
<150
<100
<100
<60
<90
DEPTH IN CORE (cm)

Fig. 7


CONCENTRATION
Au(ppb)
Ag(ppm)
<30
<7
DEPTH IN CORE (cm)
<5
<4
<10
<4
<11
<3
LAKE ATHABASCA
TAILINGS
BACK BAY
LANGLEY BAY
<2
<8
<2
<4
<5

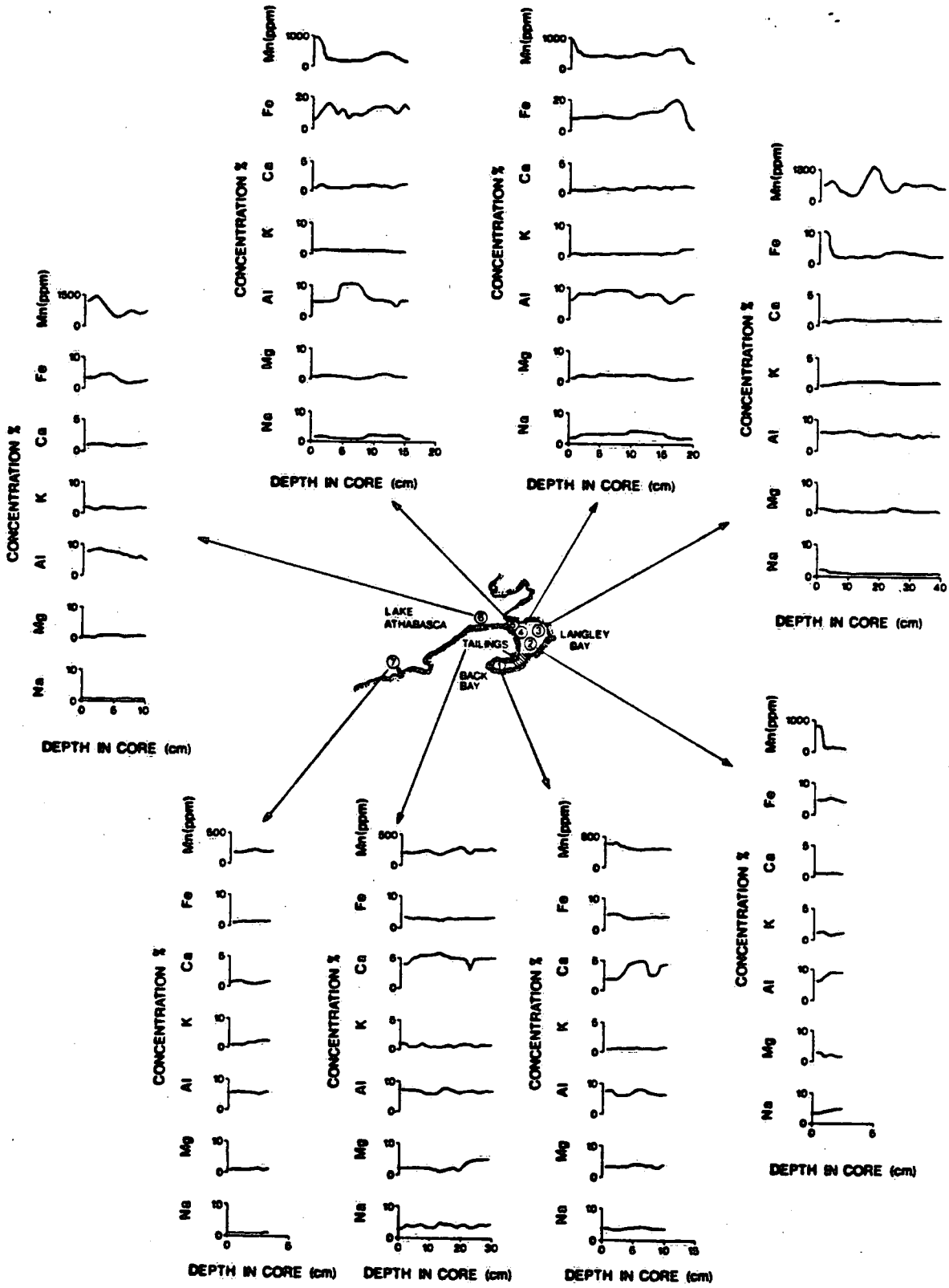

LAKE ATHABASCA
TAILINGS
BACK BAY
LANGLEY BAY
CONCENTRATION %
Mn(ppm)
Fe
Ca
K
Al
Mg
Na
DEPTH IN CORE (cm)

Fig. 6

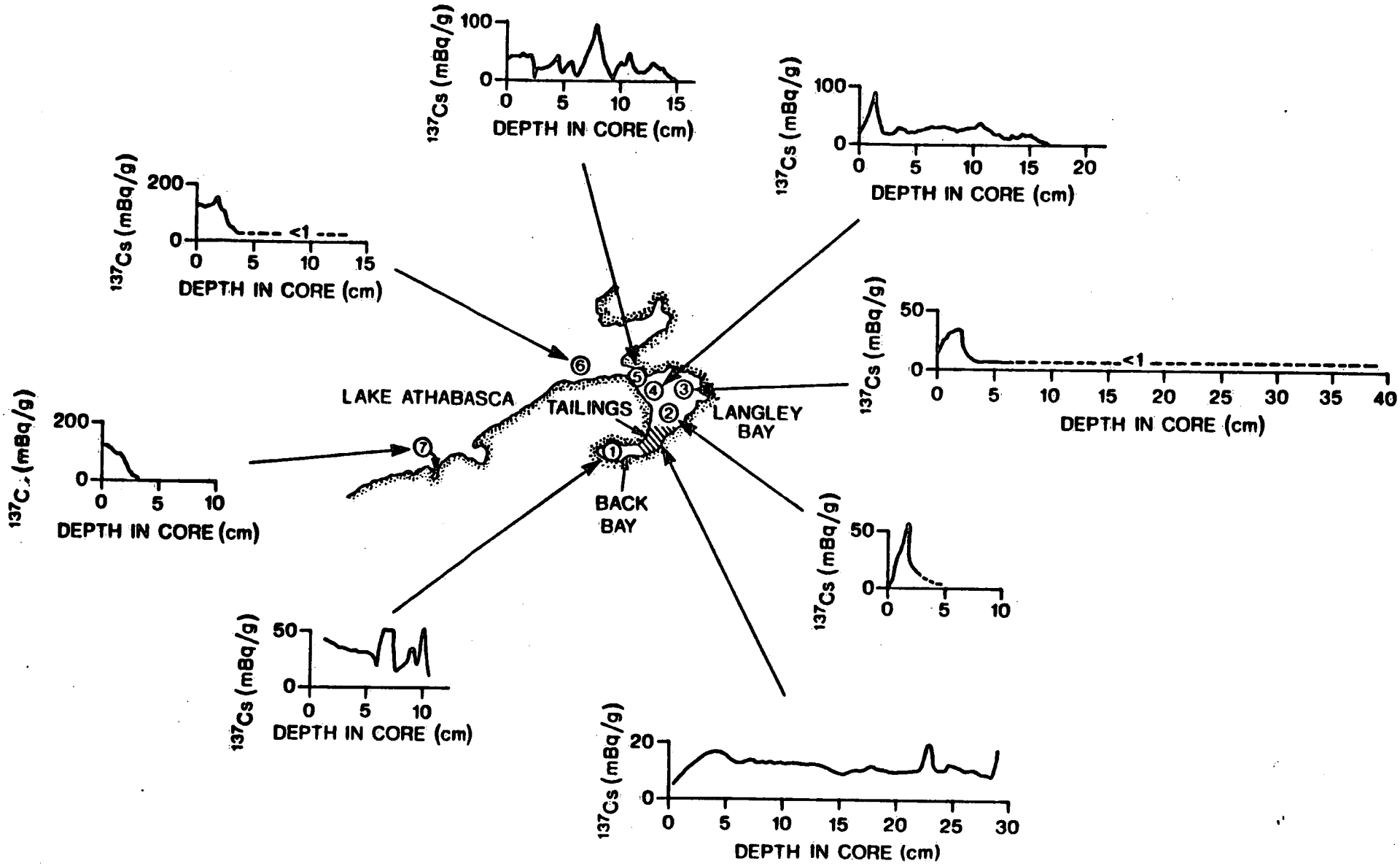

LAKE ATHABASCA
TAILINGS
LANGLEY BAY
BACK BAY
^{137}Cs (mBq/g)
DEPTH IN CORE (cm)
<1

Fig. 7

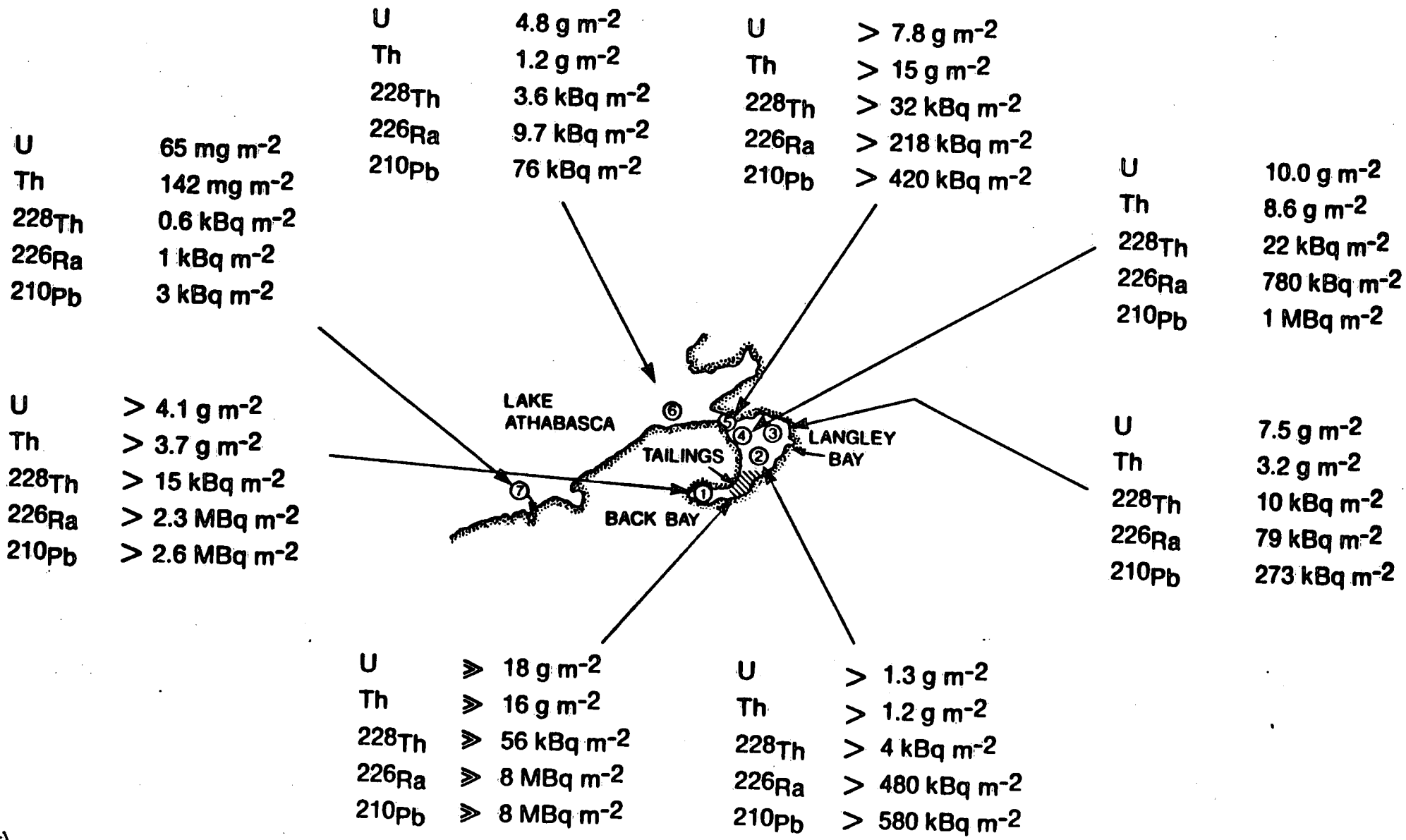

U 65 mg m-2
Th 142 mg m-2
228Th 0.6 kBq m-2
226Ra 1 kBq m-2
210Pb 3 kBq m-2
U 4.8 g m-2
Th 1.2 g m-2
228Th 3.6 kBq m-2
226Ra 9.7 kBq m-2
210Pb 76 kBq m-2
U > 7.8 g m-2
Th > 15 g m-2
228Th > 32 kBq m-2
226Ra > 218 kBq m-2
210Pb > 420 kBq m-2
U 10.0 g m-2
Th 8.6 g m-2
228Th 22 kBq m-2
226Ra 780 kBq m-2
210Pb 1 MBq m-2
U > 4.1 g m-2
Th > 3.7 g m-2
228Th > 15 kBq m-2
226Ra > 2.3 MBq m-2
210Pb > 2.6 MBq m-2
LAKE ATHABASCA
TAILINGS
BACK BAY
LANGLEY BAY
1
2
3
4
5
6
7
U 7.5 g m-2
Th 3.2 g m-2
228Th 10 kBq m-2
226Ra 79 kBq m-2
210Pb 273 kBq m-2
U ≫ 18 g m-2
Th ≫ 16 g m-2
228Th ≫ 56 kBq m-2
226Ra ≫ 8 MBq m-2
210Pb ≫ 8 MBq m-2
U > 1.3 g m-2
Th > 1.2 g m-2
228Th > 4 kBq m-2
226Ra > 480 kBq m-2
210Pb > 580 kBq m-2

Fig. 8

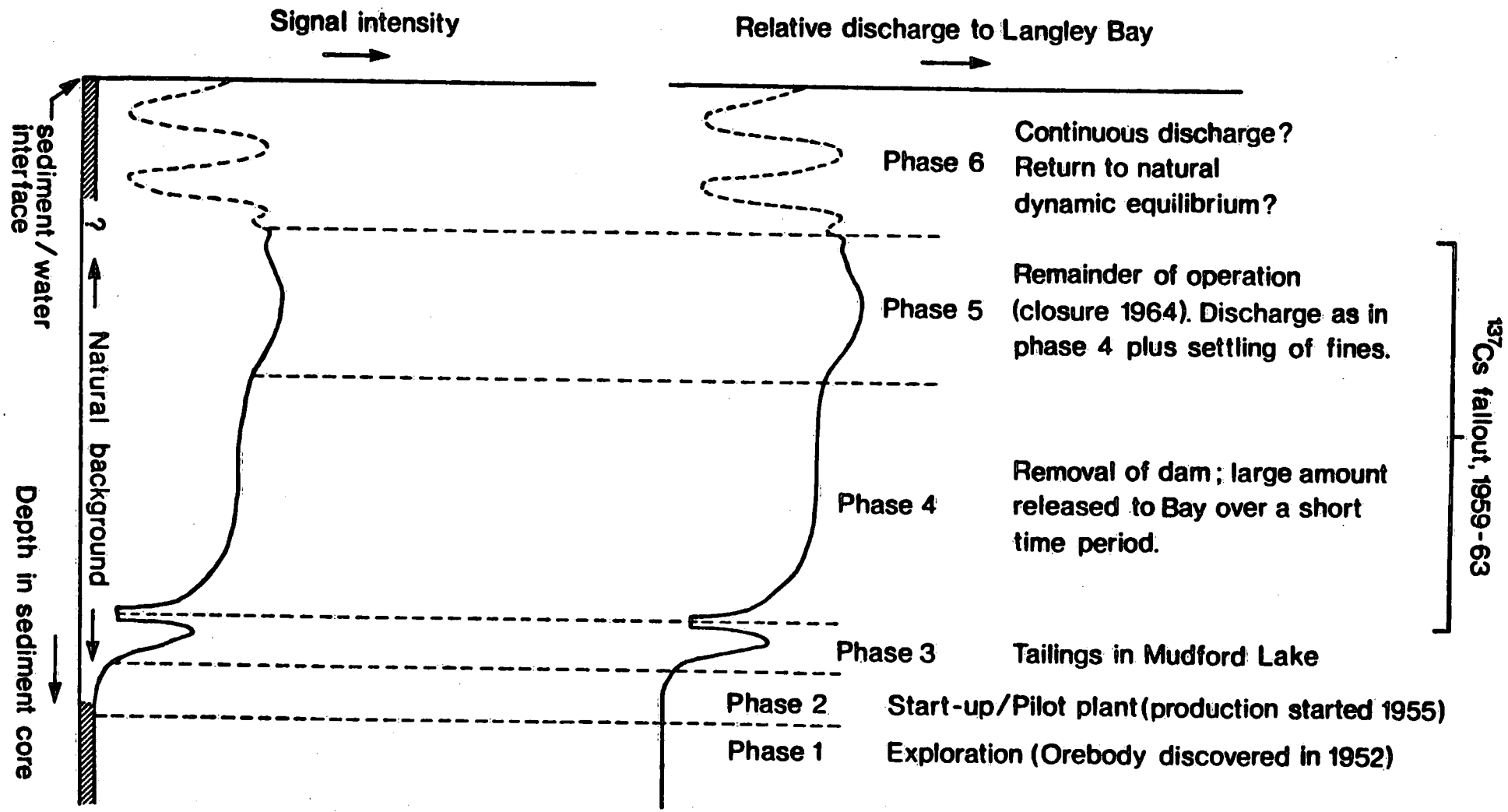

Signal intensity
Relative discharge to Langley Bay
sediment/water interface
?
Natural background
Depth in sediment core
Phase 6
Continuous discharge? Return to natural dynamic equilibrium?
Phase 5
Remainder of operation (closure 1964). Discharge as in phase 4 plus settling of fines.
Phase 4
Removal of dam; large amount released to Bay over a short time period.
Phase 3
Tailings in Mudford Lake
Phase 2
Start-up/Pilot plant (production started 1955)
Phase 1
Exploration (Orebody discovered in 1952)
137Cs fallout, 1959-63

Fig 9

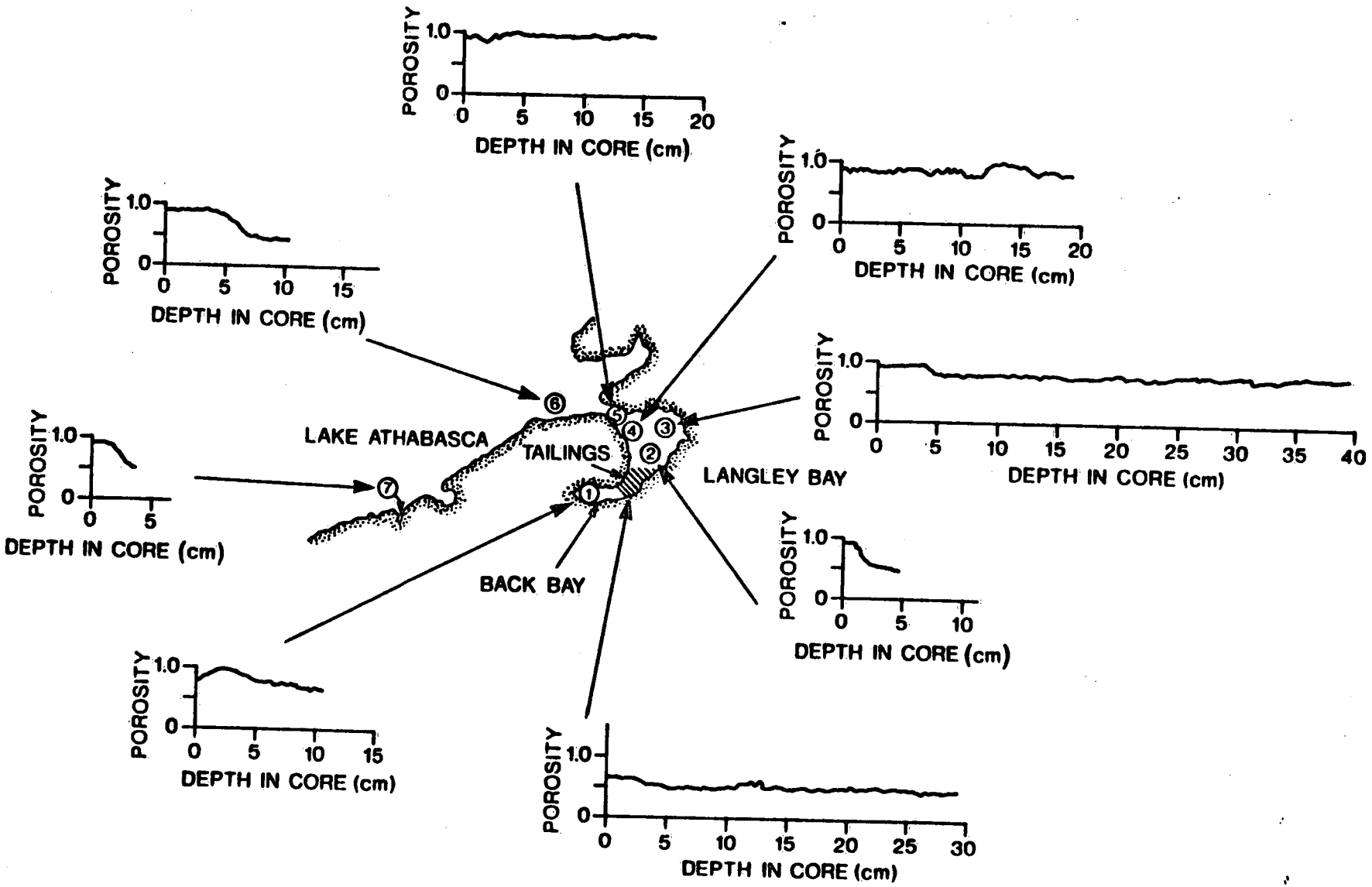
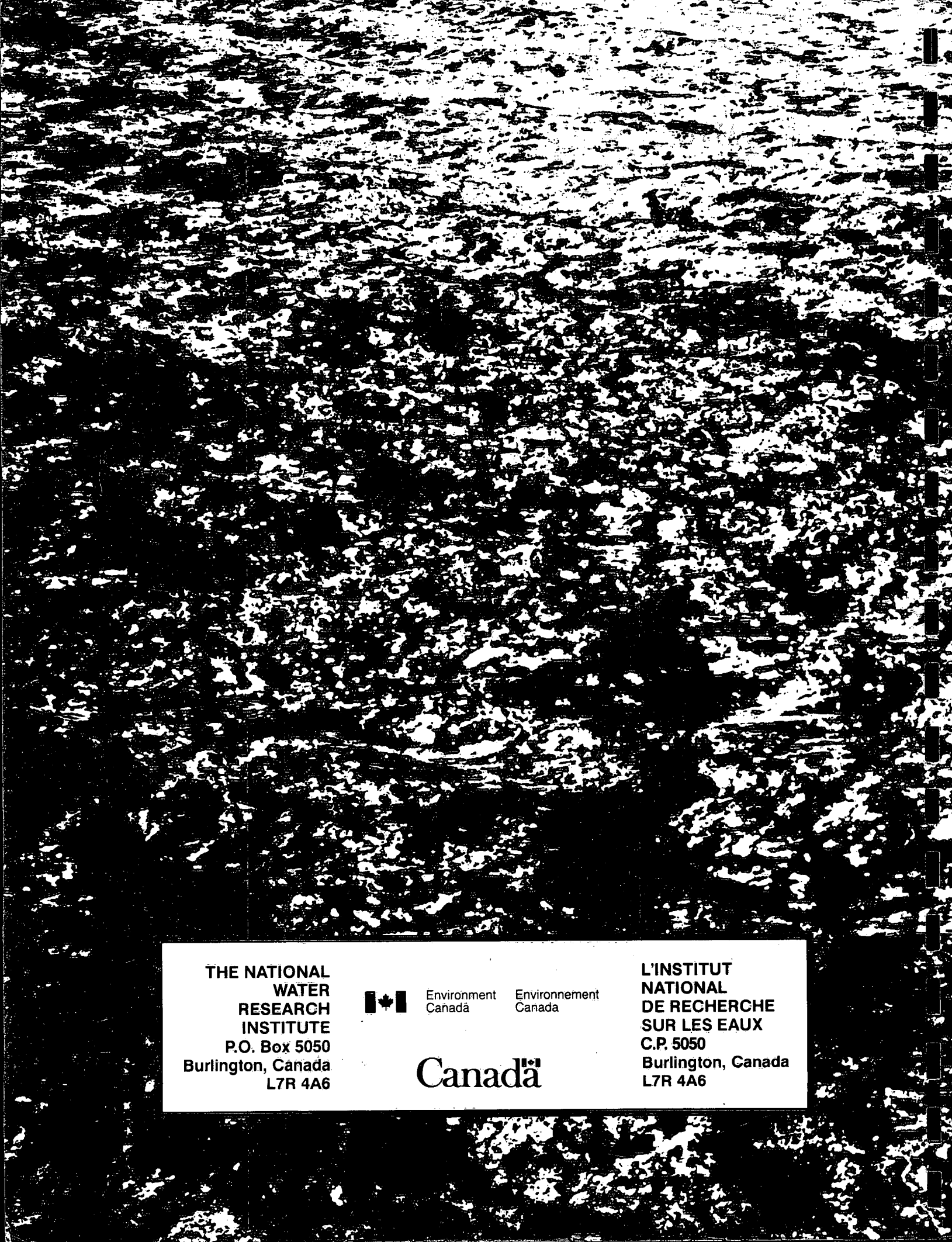

POROSITY
1.0
0
DEPTH IN CORE (cm)
LAKE ATHABASCA
TAILINGS
LANGLEY BAY
BACK BAY
1
2
3
4
5
6
7

Fig. 10



THE NATIONAL
WATER
RESEARCH
INSTITUTE
P.O. Box 5050
Burlington, Canada
L7R 4A6

Environment Canada | Environnement Canada

Canada

L'INSTITUT
NATIONAL
DE RECHERCHE
SUR LES EAUX
C.P. 5050
Burlington, Canada
L7R 4A6