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On the environmental impact of a repository for spent nuclear fuel

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SUMMARY

An estimate is presented of the contamination of groundwater discharging at the surface with actinides from spent nuclear fuel emplaced at depth in fractured, water-saturated, crystalline rock. No engineered barriers are considered, and the fuel in this case is free to interact with the groundwater and the host rock. The contaminated groundwater is assumed to reach the surface via a highly water-conducting fracture zone in the rock.

A sequence of processes and factors controlling the final concentration of actinides in the water is considered.

First, oxidative dissolution of spent fuel takes place due to radiolysis of the water. Next, reductive precipitation of uranium, with coprecipitation of the other actinides, results from interaction with reductants in the host rock. The maximum uranium concentration then remaining in solution is about 0.2 microgram per litre. The ratio of other actinides to uranium will reflect the proportions in the fuel.

Only 0.1 per cent or less of the groundwater finally reaching the surface at a local spring or well will come from a properly located repository at depth. The contamination from spent fuel in the groundwater brought to the surface will therefore, after natural mixing en route, amount to 0.0002 microgram of

uranium per litre. This uranium is accompanied by a proportional concentration of harmful actinides. Their radiotoxicity is a function of the time between discharge of the fuel from the reactor and their reentry into the surface environment.

Most of this time is spent in transit to the surface from the depth of emplacement. With parallel linear flow in a zone of high conductivity and a retardation of the actinides under reducing conditions, this takes more than 300 000 years. Radioactive decay during this time reduces the radiotoxic contamination of surfacing groundwater with actinides to a level corresponding to a natural uranium content of 0.05 microgram per litre. This corresponds to less than one per cent of the average concentrations of uranium and radium in water from springs and wells in granitic areas uncontaminated by nuclear installations. Even if instantaneous reentry could take place, the environmental impact of the actinides would only be of the same magnitude as that from natural sources of uranium and radium in the average spring, provided reductive precipitation of uranium has taken place before mixing with shallow groundwater.

Introduction

If spent nuclear fuel in a deep-seated repository is exposed to water, traces of radioactive substances may be carried into the surface environment. Engineered barriers have therefore been developed that will completely isolate the fuel until it has lost, by its own radioactive decay, more than 99.5% of its harmfulness.

An attempt is nevertheless made here to outline briefly, and without any regard to such barriers, the main mechanisms which would control a release of radioactive substances to the surface from a repository in fractured crystalline rock. In this case the spent fuel would be free to interact with the surrounding groundwater and host rock. The groundwater is then assumed to reach the surface via a water-conducting fracture zone.

The analysis is presented in the form of a highly simplified calculation of what contamination of natural waters would result from such a situation. In spite of the inherent approximations and extrapolations, it is hoped that this analysis will provide a basically realistic picture of the mechanisms involved. If so, it could be used in the evaluation of different technical concepts and of the benefits obtained from alternative isolation arrangements. It also identifies key areas for additional research and development.

Before embarking on the main subject, the paper presents a brief review for non-specialists of some practical and functional aspects, reflecting in part the repository concept currently being studied in Sweden (1). The main analysis, however, aims at a more general situation, and is not confined to any particular repository concept.

Nuclear fuel

Nuclear fuel, in the present context, consists of cylindrical bodies, pellets, of crystalline uranium dioxide (UO_2), which are assembled into fuel-rods contained in heat- and water-resistant closed tubes of a zirconium alloy. The radioactivity of new fuel is weak enough to permit handling with rather limited radiation protection, see figures 1 and 2. After irradiation in a reactor, however, the radioactivity of the fuel is much increased and requires substantial protective measures for safe handling, see figure 3. This is due to the formation in the reactor of fission products, such as strontium-90 and caesium-137, and of transuranic elements, such as the actinides americium, plutonium and neptunium. It is this great increase in radioactivity and the long time required for its decay to lower levels, that calls for special approaches to the terminal disposal of spent nuclear fuel.

The uranium content of spent fuel is about 80 weight per cent. The main radioactive elements in spent fuel, their radioactivity and decay with time are shown in figure 4. On the right, the figure shows how many times more radioactive the spent fuel and its components are than an equivalent amount of natural uranium (without daughter products).

The radiation from each constituent of the spent fuel has its own specific energy and health effect (2). Figure 5 therefore shows the radiotoxicity or harmfulness of spent fuel, here expressed as how many times more toxic spent fuel and its main constituents are than an equal amount of fresh nuclear fuel (before irradiation).

The toxicity of the latter for practical purposes is equivalent to that of an equal amount of natural uranium. It is seen in figure 5 that spent fuel after removal from the reactor is about 60 000 times more toxic than an equal amount of natural uranium, and that the toxicity decreases rather rapidly after about hundred years. Up to that time strontium-90 and caesium-137 are most important. Thereafter, different isotopes of americium, plutonium and neptunium are predominant for a period of about ten million years. They are therefore given special attention in the following. The toxicity of iodine-129 and technetium-99 is below the range of this diagram.

The expression of radiotoxicity as a ratio versus that of natural uranium is a key element in the following presentation. It facilitates order-of-magnitude assessments and reflects the fact that uranium is chemically the predominant constituent of spent fuel. Furthermore, the chemical behaviour of americium, plutonium and neptunium in many respects closely resembles that of uranium. Finally the behaviour of uranium in the natural environment, notably in shallow and deep natural waters, is fairly well understood.

Practical aspects

When nuclear fuel is taken out from a reactor it is highly radioactive, and also generates considerable heat. It is therefore first stored in a special facility at the reactor site, which allows effective cooling, radiation shielding and close surveillance. In the Swedish concept this stage of monitored storage, which is presently being successfully practiced at reactors in all parts of the world, is foreseen to last about forty years, in order to allow the radioactivity and heat to decline sufficiently to facilitate further handling of the fuel. Thereafter, according to present plans, the spent fuel will be encased in canisters of highly resistant materials, such as metallic copper.

Practical tests have demonstrated the feasibility of this encapsulation of the fuel, and experimental, thermodynamic and geological evidence indicates that such canisters may last millions of years in the protected environment offered by a deep-seated repository (3,4). The canisters thus provide complete containment and protection of the environment, and therefore constitute a highly efficient engineered barrier to the dispersal of radioactive substances.

The canisters with the spent fuel will after appropriate quality checks, be finally disposed of by emplacement in a repository constructed in crystalline bedrock at a depth of several hundred metres. Each canister will there be placed in a separate hole in the rock. These holes, as well as the tunnels and shafts of the repository, will be backfilled by appropriate blends of clays and soil materials, which are stable in the environment. By virtue of their capacity to swell, the clays will provide a tight seal and the backfill will attain a permeability to water that is equal to or less than that of the surrounding rock (5). Large scale practical tests of backfilling techniques and materials are presently being carried out under realistic conditions in the Stripa mine (6). The repository, after any period of control and monitoring, may thus be permanently closed and sealed, with no responsibility for monitoring or surveillance being imposed on future generations.

Functional aspects

The fundamental purpose of the entire system is to protect man and his environment both now and in future. From this point of view, the procedures just outlined represent two different phases. The first phase covers the period of active handling of the spent fuel and the canisters, as well as operation of the repository, including its preparation for final closure. During this stage, all operations have to meet existing national and international regulations on radiation protection and special legislation on occupational hazards and safety.

The second phase begins when a repository has been finally closed and no further direct human involvement with the fuel is required. Effective general shielding to prevent human exposure to radiation from the fuel must then be provided and reentry of radioactive substances from the fuel into the human environment must be restricted to very low levels.

Complete shielding of the radiation coming from the fuel requires less than five metres of intervening solid material such as bedrock. In many flat areas, undisturbed by recent volcanism, violent earthquakes and ongoing mountain-building, rates of erosion are less than ten metres per one million years. A repository located at depth in such areas will not be uncovered for many millions of years. Some portions of the precambrian Shields, where man today can walk on intact erosional surfaces formed more than 500 million years ago, provide perhaps the most impressive evidence of the limited effects of subsequent erosion in such areas. These conditions provide full protection against human injury due to direct exposure to radiation from spent fuel in properly located deep repositories, over periods of time far in excess of those required for the decay of its harmful radioactive elements.

Reentry of radioactive substances from the repository into the human environment through natural or induced groundwater flow rather than erosion therefore constitutes the remaining, and hence crucial, problem in the management of spent fuel.

An environmental impact of a repository would thus require an extensive breach of the engineered barriers, eg perforation of a large fraction of the canisters, and would thereafter be directly controlled by the capacity of the groundwater to transport the radioactive substances to the surface.

It may be interesting to compare a specific repository concept, such as the one outlined above, with a basic reference case not bound by particular features of design, construction and emplacement. The following analysis therefore considers a case where the spent fuel is exposed to the groundwater and free to interact with it and the host rock, as if emplaced in the natural rock unchanged by engineering activities. This degree of exposure defines what in short may be called a leaky repository. More strictly speaking, it should perhaps be called a zero-case leaky repository, since different engineering measures could reduce or increase its leakiness.

Dissolution of the fuel

The groundwater contained in a fractured host rock, will, upon contacting the highly radioactive spent fuel, undergo partial decomposition into its components, hydrogen and oxygen, as a result of radiolysis. Recombination, leading to the formation of both water and hydrogen peroxide, also occurs. The extent of radiolysis is sensitive to variations in the content of dissolved substances, to the extent and geometry of the contact with the fuel and to preferential escape of hydrogen through the fractured rock. The overall effect would be local oxidation. The possible extent of radiolysis has been investigated theoretically (7,8) and on the basis of evidence from the natural reactors at Oklo (9) with reasonable agreement in the results.

The oxidation resulting from radiolysis of the groundwater is an important factor in the dissolution of the spent fuel. It leads to the formation of hexavalent uranium, the pertinent compounds of which are much more soluble than the uranium(IV) oxide of the fuel (10). The americium and plutonium in the fuel are less easily brought into solution by oxidation than uranium, as shown by thermodynamic calculations (11) and leaching experiments (12,13). Although these studies do not quantitatively represent the conditions in a leaky repository, they show clearly that a preferential leaching of uranium is to be expected. The water leaving the repository will therefore carry a lower ratio of the other actinides to uranium than that present in the spent fuel. To make our presentation easy to follow, the proportion of actinides in solution will nevertheless be taken to be the same as in the spent fuel.

Reprecipitation of uranium and coprecipitation of actinides

It is well known that hexavalent uranium, dissolved in oxidizing groundwater, will be reprecipitated upon encountering more reducing conditions along the flowpath of the water. This process in Nature gives rise to special types of uranium ore deposits, and its bearing on the underground disposal of spent nuclear fuel has been discussed repeatedly, eg (14,15,16). Uranium from spent fuel dissolved in groundwater will also be reduced to the tetravalent state by ferrous iron and other reductants in the host rock beyond the realm of radiolysis (17). It will thereby be reprecipitated at a redox front, ie the interface between oxidizing and reducing conditions along the flowpath. This entails a return to natural conditions away from the spent fuel.

A large body of data exists on the uranium content of natural waters, mainly from geochemical prospecting (18). Unfortunately, most of these data are of limited value due to a lack of vital information on the pH, Eh and carbonate content of the analyzed waters, and on their possible interaction with organic matter, (19). Simplifying geochemical assumptions cannot readily serve as a substitute for such information. Significant results are instead available from studies of the isotopic fractionation of uranium found in many waters (20). Figure 6, adapted from the presentation by Osmond and Cowart, summarize these findings. It can be seen that both surface waters and groundwaters show a very wide range of uranium concentrations, from 0.01 to more than 100 micrograms per litre. The diagrams also show that there is a marked difference between surface waters and groundwaters with regard to their activity ratios, ie the ratio of the alpha-activity from uranium-234 to that from uranium-238. In waters with a high activity ratio, the ratio of uranium-234 to uranium-238 is much higher than at radioactive equilibrium. This is most marked in groundwaters with a uranium content of less than 0.2 micrograms per litre. Obviously specific conditions exist in the environment of such waters which favour a displacement of the normal ratio between the two uranium isotopes.

The uranium in the waters originates from the surrounding rocks, but extensive studies (21) show that the activity ratio in rock generally show much less variation. This leads to the conclusion that a preferential release of uranium 234 sometimes takes place in the interaction between the rock and the groundwater. Theoretical considerations, experimental evidence and careful case studies in Nature show that the isotopic fractionation of uranium is due to atomic recoil effects. These occur when a 234 atom is formed by radioactive disintegration, in two steps, from a 238 atom. The recoil effects enhance the mobility of the 234 atoms, and may aid in their transfer from the uranium-bearing solid to the water (20).

The isotopic fractionation effects are negligible as long as uranium is transferred to the groundwater by chemical dissolution of the uranium present in the rock, and as long as the uranium content of the water is relatively high. This situation characteristically prevails under oxidizing conditions in the crust, and reflects the solubility of many compounds of hexavalent uranium, in particular in the presence of complexing agents such as carbonate and sulphate ions. Under reducing conditions, the insolubility of uranium(IV) compounds leads to low uranium contents in the water even at saturation. Only at this point, when the tendency of uranium to enter into solution is practically zero, will the higher mobility of uranium-234 due to recoil give rise to high activity ratios in the water. The actual observation of a low uranium content and a high activity ratio in a groundwater is therefore evidence of saturation with uranium under reducing conditions.

Inspection of figure 6 shows that the concentration of uranium in reducing groundwaters varies between 0.01 and 0.2 microgram per litre. This variation probably reflects differences in other factors, such as pH and the concentration of uranium complexing compounds, in the investigated groundwaters. The large body of available data suggests that the normal chemical variation in reducing groundwaters should be adequately covered. Therefore, 0.2 microgram of uranium per litre would seem to represent a fair estimate of the maximum concentration to be expected in groundwaters that reduce uranium to its tetravalent state under a wide variety of natural conditions. This conclusion is confirmed for groundwater in crystalline rock by recent data from different test sites in Sweden, cf figure 7. It shows that higher uranium contents in shallow sections, representing oxidizing conditions, become drastically reduced to values below 0.2 microgram per litre at greater depths.

As far as the other actinides are concerned, it must be remembered that this entire group of elements shows great similarities between its members with regard to their ionic radii, valence states and reactions with many natural complexing agents and precipitants. They may therefore be expected to be scavenged from solution by coprecipitation with uranium, even if they are not present at their individual saturation concentrations in the groundwater. So far it has been shown experimentally that hexavalent uranium in aqueous solution, when precipitated by reduction, will coprecipitate traces of plutonium present in solution. The ratio of plutonium to uranium in the precipitate was found to be about the same as in solution (22). Extending this to americium and neptunium suggests that the ratio of the three actinides to uranium remaining dissolved in groundwater after reductive precipitation will not differ greatly from that prevailing in the oxidized solution. On this basis the radiotoxicity of groundwater contaminated by spent fuel may be estimated as a function of its uranium content and time. It can be seen, for instance, in figure 5 that spent fuel one thousand years after removal from the reactor is about 10 000 times more harmful than the equivalent amount of natural uranium. Reducing groundwater, which carries the maximum concentration of 0.2 microgram of uranium per litre, coming from such fuel and carrying a proportional concentration of the other actinides will at that point in time be as harmful as water containing 10 000 x 0.2 = 2000 micrograms (2 mg) of natural uranium per litre. These relationships are fully represented in figure 8.

Aspects of groundwater flow

Figure 8 describes the radiotoxicity of groundwater at depth, that has passed the repository and a redox front in the rock. Its impact on the biosphere will be determined by how and when the radioactive substances dissolved in the water eventually reach the surface. The flow and volumes of groundwater that could transport radioactive substances from depth to the surface will therefore be considered next.

The flow of groundwater in fractured crystalline rocks reaching great depths on a regional scale is normally sustained entirely by surface infiltration of rainfall and other forms of precipitation. At first, the water seeps vertically through the superficial layers of the ground, until it joins the groundwater. This constitutes the continuous body of water that fills all the interconnected pores and cracks in the ground and bedrock. Its upper boundary is often called the free groundwater table. If a given point within this body of water is connected to an otherwise isolated tube, open to the atmosphere, the water will rise in the tube to a steady-state level, called the hydraulic head or hydraulic potential at that point. Groundwater will flow, if unconfined by physical barriers, from loci of higher potential to points of lower potential, in direct analogy to the flow of electrical currents. A classical picture of such a flow system (23) is shown in figure 9A. It may be taken to represent flow in a vertical section of the ground between two equal wells, and also through a topographical ridge between two equal valleys. The dashed curves in the diagram join points of equal potential and actually represent intersections of the plane of the diagram with surfaces of equal potential underground. Free flow always takes place perpendicularly to such surfaces, as illustrated by the curved flowlines, marked by arrows, in the diagram.

The diagram illustrates a number of important aspects. It can be seen that all flowlines converge towards the points of minimum potential in the valleys (wells), which actually mark the loci of groundwater discharge to the surface environment. Thus all the water infiltrated through the surface of the land included in the section will here return to the surface, although it arrives there along different flowpaths, which ideally are fully determined by the distribution of hydraulic potentials. The flowlines entering from the left and right sides of the diagram indicate that one half of the groundwater issuing at the surface is derived from infiltration from outside the central section hosting the repository. In a three-dimensional symmetrical model the central area will only provide one quarter of the total groundwater flow. Furthermore, most of the infiltration will be drained by the shallow portions of the section (shaded). Only that small part of the total flow that infiltrates close to the summit of the ridge, near the so-called groundwater divide, will actually reach a repository located in the deeper parts of the central section.

Figure 9A represents a very simple situation, where hydraulic conductivity is the same everywhere in the ground. In reality, however, the uppermost parts, including unconsolidated soils and the weathered and disintegrated top section of the bedrock, show conductivities much higher than those in the deeper sections. The higher conductivities generally prevail to depths of one hundred metres or more even in glacially eroded areas, where fresh rock is present close to the surface. This is thought to reflect various surface influences, notably the decrease in the load imposed by overlying rock sections, on approaching the surface, allowing more fractures to open up and to increase in size. Hydraulic conductivity in crystalline rocks, as measured in 25 m long sections at Swedish sites, covers the full range between 10^{-11} to 10^{-5} m/s (24). This distribution of hydraulic conductivities in the ground channels even more of the groundwater through the shallow sections and further reduces the amount of water flowing through the deeper parts, cf figure 9B.

Crystalline rocks generally contain marked zones of increased fracturing. They are often manifested at the surface as linear depressions. Even at great depth they may occur as narrow zones of increased hydraulic conductivity, comparable to that otherwise found near the surface. They tend to transmit the hydraulic potentials from near the surface into the interior of the bedrock. Fracture zones reaching the surface in areas of infiltration will therefore conduct groundwater into the deeper parts of the rock, and fracture zones reaching loci of groundwater discharge will drain the adjacent less conductive rock mass. Fracture zones may thus considerably alter the distribution of hydraulic potentials and the flow pattern in the deeper parts of the bedrock from the picture given in figure 9A. However, their cross-sectional areas are small in comparison to that of the total flow system. They therefore do not radically change the ratio of deep to shallow groundwater discussed in the preceding section. This is substantiated by recent 3-D model calculations of groundwater conditions at four Swedish test sites, using site-specific data and including a number of conductive fracture zones reaching great depths (25). They show that the overall groundwater flow at depths below 450 m is at all sites less than 0.1 litre per m^2 and year. Groundwater recharge by infiltration at the surface is, on the other hand, known to be around 100 litres or more per m^2 and year. It has already been shown above that the total infiltration area sustaining the local flow of groundwater to the surface is several times larger than the cross-section of the repository itself. The contribution of water that has actually passed a repository to the total flow of groundwater discharging at a point at the surface will therefore be less than one per mil. Figure 10 shows the radiotoxic contamination from the spent fuel of the issuing groundwater, using this proportion of 1:1000, as a function of the time between removal of the fuel from the reactor and reentry of traces of actinides into the surface environment.

It may be added for clarity that the fracture zones discussed above play an entirely different role when they intersect an open tunnel at depth. Such a fracture zone will then conduct water into the tunnel in response to a difference in hydraulic potential (hydraulic head) corresponding to the difference in elevation between the tunnel and the groundwater table, where it is intersected by the fracture zone. Inflow into the tunnel therefore takes place even when the conductive zone reaches a place of normal groundwater discharge at the surface. These conditions lead to substantial localized inflows into a tunnel (26), which after some time of drainage are largely sustained by newly infiltrated water with a high tritium content (27). Such inflows greatly facilitate recognition of waterbearing zones during the construction of a repository, and aid in the selection of suitable disposal areas within a repository site. After terminal closure of a repository, however, conditions slowly revert to normal and the fracture zones lose much of their overall significance.

Time of trace reentry

The lapse of time between removal of fuel from a reactor and the actual reentry into the surface environment of radioactive traces from a deep repository leads to a natural reduction of radiotoxicity by progressive radioactive decay, cf figures 5,8 and 10. Probably around one hundred years will pass between the discharge of fuel and the water-saturation of a repository. This time-span includes all the handling, interim storage and final emplacement of the fuel, as well as backfilling and closing the repository. Extensive contact with the groundwater may perhaps be further delayed for several hundred years by the zircaloy tubing enclosing the fuel. Such lengths of time would be sufficient to eliminate strontium-90 and caesium-137, but they will not significantly affect the actinides in the fuel. Instead, the time required for groundwater transport of the actinides to the surface is of greater interest.

Experimental study of groundwater transport involves the introduction some tracer substances at one point along the flowpath and observation of their time of arrival and concentration at another point (28,29). Pumping is often performed to ensure recovery of the tracers and to shorten the duration of the experiments. Results of such tests in crystalline rocks are illustrated in figure 11. It is found that different tracers, although introduced at the same time and moving along the same flowpath, arrive at different times. The slower tracers also arrive at greatly reduced concentrations, cf figure 11D. It can in fact be shown that their retardation is due to sorption by the rock. The passage of dissolved substances through the bedrock may thus be seen as a kind of chromatography, where the rock along the flowpath takes the place of the sorption column.

Such tests have been performed in fractured zones in granitic rocks and gneiss, with hydraulic conductivities of between 10^{-5} and 10^{-7} m/s, over distances of between 10 and 51 m (30-34). The flow porosity in the zones is around 10^{-3}, as obtained from tests with tracers such as tritium, iodide and bromide, which show no or insignificant retention. Strontium and caesium were used as tracers to study retardation. The former, at concentrations of 10^{-4} M and at hydraulic conductivities around 10^{-7} m/s, showed transit times about 20 times longer than bromide and iodide. Caesium did not arrive within the duration of the experiment (5000 h). Supplementary laboratory studies concern the migration of actinides of different oxidation states in natural rock fractures and the determination of distribution coefficients between the groundwater and crystalline rocks and their minerals for different radioactive nuclides (35,36). They show that in reducing groundwater environments, the actinides will travel more than a thousand times slower than strontium.

These results may be used for simple estimates of travel times for actinides migrating from a repository to the surface. Consider for instance a fracture zone with a hydraulic conductivity of 10^{-7} m/s and a flow porosity of 10^{-3} that connects a repository to a point of groundwater discharge at the surface. Let the distance be 500 m and the difference in hydraulic head 5 m (a gradient of 0.01). The average velocity of groundwater flow in this zone, according to Darcy's law, would be 30 m/year. Strontium would thus migrate 1.5 m/year. The velocity of the actinides, in a reducing groundwater, would be 1.5 mm/year or less. The time of reentry of actinides along this zone therefore would be around 300 000 years or more.

No comparable data are available for fractured crystalline rocks outside such conductive fracture zones. Here the hydraulic conductivity is below 10^{-10} m/s (24) but the flow porosity is poorly known. Tracer experiments in such rocks are underway. It has been pointed out, however, that the speed of migration of substances being retarded in a given medium should be directly proportional to the hydraulic conductivity and independent of porosity (37). If the ratio of porosity available for flow to porosity available for diffusion does not change radically, the migration of actinides in the rock surrounding a fracture zone under the same hydraulic gradient would be a thousand times slower. A retardation of three million years would then result over a distance of 5 m of such rock.

This last statement is speculative, and also disregards the rate of diffusion, which, over such short distances and under appropriate concentration gradients, would be greater than the rate of migration due to the hydraulic gradient (38). On the other hand, the time of reentry along the fracture zone is based on considerable evidence. The validity of such transport calculations has recently been demonstrated for groundwater flow in a sandstone (39). A time of reentry of 300 000 years for the actinides therefore appears realistic or conservative. It corresponds to a travel time of the groundwater of only 17 years. It can be seen in figure 10 that a time to reentry of this length reduces the radiotoxicity contributed by the repository to the discharging groundwater to an equivalent uranium concentration of about 0.05 microgram/litre.

Comparison with uncontaminated groundwaters

The content of radon-222, radium-226 and uranium in water from 41 springs and wells in Sweden has been investigated (40). The data are summarized in table 1. All these waters are uncontaminated by nuclear installations. Their radioactivity is instead derived from the naturally occurring radioactive substances found practically everywhere in the bedrock and its overburden.

To illustrate characteristic variations, median values and averages are given here for the complete set as well as for samples from drilled wells in bedrock and from all wells and springs in areas underlain by granitic rocks. It can be seen that the averages are much higher than the median values. This indicates that a majority of samples show low concentrations and that the averages reflect the influence of high concentrations found in a small number of samples. To reduce this influence somewhat, averaging has been carried out after omission of the single highest values for uranium, radium and radon from the complete set.

For the present purpose the median values may be taken to indicate roughly what concentration levels may most frequently be expected in individual springs and wells, since half of the investigated cases show lower and the other half higher values than the median. The median values in these data are close to the most frequent ones.

The averages, on the other hand, indicate what concentrations at equal flows should be present in each well to maintain the overall concentration levels in the water brought to the surface environment. In this respect, the contributions from the few wells with high concentrations are, of course, rather important.

The different levels of radioactivity from radium and uranium can be translated into conventional weight-volume-based concentrations and further be expressed as equivalent concentrations of natural uranium, allowing for differences in their radiotoxicity. This has been done for each group of samples in table 1 to express for each of them averages and medians and the difference between these two values. This difference should represent an interesting aspect of the variation in these waters, indicating how much the concentration of uranium might increase in a modal well or spring before reaching the average. This difference is seen to correspond to about 10 micrograms of uranium per litre.

The radioactivity of radon-222 alone in these waters is usually about one thousand times higher than that of either uranium or radium. Natural radon therefore provides the main contribution of radioactivity to the surface environment from underground via the groundwater. Being a gas, however, its environmental impact differs radically from that of the other, non-volatile radioactive substances dissolved in groundwater. Radon levels are therefore not simply expressed as equivalent uranium concentrations.

Only the concentrations of radium and uranium are compared here with the release from spent fuel. Their concentration in the average spring is equivalent to about 10 micrograms of uranium per litre, see table 1. The radiotoxicity of actinides from spent fuel arriving at the surface after about 300 000 years would thus at most represent less than one per cent of that of the radium and uranium constantly brought into the surface environment from natural sources by the average uncontaminated spring or well.

Discussion

This brief outline deals with a complex subject in highly simplified terms. Already the use of standard radiotoxicities is a substitute, far from perfect, for detailed consideration of the biological and physiological fate and effects of the individual radioactive nuclides. On the other hand, it greatly facilitates comparison and evaluation of the concentration of the different nuclides in the groundwater discharging at the surface. This discharge represents the last focal point on their way back into the surface environment. It therefore constitutes a natural point for an estimate of their environmental impact. The approximations inherent in the use of standard radiotoxicities may even be seen as matching the range of natural variations in the other input data used in the analysis.

The effects of radioactive heating of the bedrock and groundwater have so far been disregarded completely. These effects reflect technical parameters, such as the duration of interim storage and the pattern of emplacement in the repository, which are not part of our central subject. It may be stated, however, that most crystalline rocks are rather stable at elevated temperatures. Low-temperature minerals, such as fracture-filling clays, may be affected, but this would not substantially change the present calculations. The high sorption of such minerals has not been allowed for in the estimate of the retardation of actinides in transit to the surface.

Heating of the groundwater would lead to a decrease in its density and hence to a buoyancy that could lead to a reduction of the gravity-controlled groundwater circulation or even to a reversal or other changes in the flow pattern. These conditions may become rather complex, cf (41,42). The buoyancy would be counteracted, however, by an increase in groundwater salinity with depth, which seems to be a general feature in crystalline rocks (43). It has also been suggested that the heat-induced rise of groundwater in fractured rock is a threshold effect (44). Above all, the heating could be controlled to give hydraulic gradients that would only locally exceed the value of 0.01 used in the present calculation of the time of trace reentry. The overall effects of heating therefore do not jeopardize the validity of our estimates.

Rockmechanics also has been disregarded in the present analysis. The primary effect of the heating would be a tendency for the rock surrounding a repository to expand. The ensuing compression of the existing fractures would lower the hydraulic conductivity of the rock and thus further reduce the proportion of groundwater passing the heated portion of the bedrock hosting the spent fuel.

The effects of the construction process itself do not have to be considered for our zero-case repository. They would reflect the design and techniques used in an actual case. It is difficult to envision any reasonable practical arrangement that would seriously affect rock quality beyond a limited distance from the actual emplacement. The functional role of a fracture zone and the far-field host rock, which in the present analysis are the crucial natural barriers, would not be impaired. It should be recalled at this point that the time to trace reentry is calculated by considering transport along a highly conductive 500 m long fracture zone and not through the normal host rock surrounding the fuel.

In view of the uncertainty and safety margins related to the factors that have been considered, the following comments may be in order.

Preferential release of uranium by a factor of ten over americium and plutonium in connection with the oxidative dissolution of the spent fuel is suggested by the available evidence. This would also reduce their environmental impact by a factor of ten. Further experimentation on the ratios between the actinides brought into solution under conditions approaching those prevailing in a repository would seem to be most desirable.

There is little reason to expect any great changes in the maximum saturation concentration of uranium in reducing groundwater, or in the proportion of actinides remaining in solution after reductive precipitation of uranium and coprecipitation. No significant upward changes in our estimate are therefore expected due to variations in these factors. It should be pointed out, however, that only limited experimental evidence is as yet available on the latter point. It is furthermore restricted to coprecipitation of plutonium. Additional experimental evidence on the coprecipitation of actinides is called for.

The ratio of 1:1000 for the volume of groundwater passing a repository to the total volume discharging at the surface represents a rather rough, and perhaps unduly pessimistic, estimate. Little change would be expected in the underlying distribution of hydraulic conductivities, but consideration of reasonable proportions between the total area of infiltration and the cross-sectional area of a repository may suggest that perhaps ten to a hundred times more groundwater is actually bypassing the repository. This is further supported by recent calculations of the influx to a well drilled right on top of a repository (45). Such changes in the proportions of flows would imply a corresponding relative reduction in the influx of radionuclides to the surface environment. Instrumentation for direct measurement of groundwater flows at depth would provide most valuable evidence on this question.

Finally,our estimate of the time of trace reentry is in itself complex. Considering first the hydrodynamic properties of the fracture zone, they appear to be well founded. In fact, the recent data from site investigations in Sweden show that a hydraulic conductivity of 10^{-7} m/s was found at depths below 200 m in only three sections out of a total of about one thousand. It follows that only in extreme cases would a repository be drained by such a fracture zone, and this zone would only drain a limited part of it. A major part of the repository would be surrounded by less conductive rock, unless adversely affected by construction activities.

The calculated groundwater velocity, 30 m/year, and the total transit time for the water of only 17 years reflect the values chosen for the hydraulic conductivity of the zone and for the hydraulic gradient. The calculation assumes the simplest case of parallel linear flow, whereas in reality more intricate, partly convergent, flow patterns are more likely. This could easily be improved on by more site-specific calculations; our example serves only to illustrate principles and the type of input at hand.

Regarding the retardation factors, it seems fair to state that they are based on extensive experimental evidence supported by basic physicochemical analysis. Recent work (46) further indicates that dissolved nuclides, on prolonged contact with the host rock, will diffuse into its micropores and thus be retarded even stronger. On the other hand, site-specific effects are poorly known and field experiments at each repository site seem called for. Perhaps they should ideally follow a sequence such as simple push-pull tests for orientation (47) - interpretation and prediction - extended tracer tests for verification. It may be added that in the situation considered here, the existence of reducing conditions along the flowpaths draining a repository is perhaps the single most important aspect of a repository site. It will control both precipitation and retardation of the actinides. In fact, as long as reducing conditions prevail, the time for trace reentry is not really critical. Comparison of figure 10 with table 1 shows that there is no point in time where the radiotoxicity from the actinides in the groundwater would exceed that from natural sources of radioactivity in the average well or spring.

Conclusions

The main points of the present study are tentatively summarized in the following conclusions:

- The harmfulness of spent nuclear fuel is dominated, for a time span from one hundred to ten million years after removal from the reactor, by the three actinides americium, plutonium and neptunium.

- Upon contact with the groundwater, spent fuel will gradually pass into solution due to oxidation caused by radiolysis.

- The ratios of the actinides to uranium in solution will resumble those in the spent fuel.

- The actinides will be coprecipitated with uranium upon interaction of the solutes with reductants in the host rock, leading to a maximum concentration of uranium in the groundwater of about 0.2 microgram per litre. The proportions between the actinides will not be radically changed by this process.

- A properly located, deep-seated repository in fractured crystalline rock will be affected by less than 0.1 per cent of the total groundwater discharging locally at the surface.

- The time between removal of fuel from a reactor and the trace reentry into the surface environment of the actinides from such a repository at a depth of 500 m will be about 300 000 years or more.

- The factors listed so far lead to an estimate of the environmental impact of actinides from spent fuel emplaced in fractured and water-saturated crystalline rock. Unless some major factor has been overlooked, this impact would, at most, be equivalent to that of a small fraction of the radium and uranium that is constantly being brought into the local surface environment from natural sources by the average spring or well in areas underlain by similar rocks. This overall conclusion is rather insensitive to changes in the time to trace reentry of actinides. Key factors are the extent of radiolysis, the restoration of natural redox conditions, and the small fraction of deep groundwater in the water reaching the surface environment.

Acknowledgements

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Table 1. Radioactivity from Radon (Rn), Radium (Ra) and Uranium (U) in water from springs and wells in Sweden

Samples	Number	Rn-222		Ra-226		Uranium		Ra + U		Difference
		Ave.	Med.	Ave.	Med.	Ave.	Med.	Ave.	Med.	Ave.-Med
	(N)	(Bq/l)		(mBq/l)		(mBq/l)		(eq.U µg/l)		(eq.U µg/l)
All	40	99	55	22	3.9	79	14	8	1.5	6.5
Wells in bedrock	19	109	29	40	3.7	86	14	13	1.5	11.5
Wells and springs in granite areas	13	213	92	41	6.8	150	35	15	3	12

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Analysis of groundwater from deep boreholes in Svartboberget. KBS TR 83-41. SKBF 1983.

FIGURE TEXTS

1 Fresh nuclear fuel - a cylindrical pellet of crystalline uranium dioxide - can be handled with very modest protection. Photo courtesy C-E Wikdahl, M Witt/OKG.

2 Assembly of fresh nuclear fuel rods of uranium dioxide pellets within zircaloy tubing being emplaced in the reactor. The radioactivity is low enough to allow handling without extra shielding. Photo courtesy C-E Wikdahl, M Witt/OKG.

3 Container, 70 tonnes of steel and water, required for adequate cooling and radiation protection when spent fuel is taken out of the reactor. Photo courtesy C-E Wikdahl, M Witt/OKG.

4 The radioactivity of spent nuclear fuel and its main constituents, and its decay with time.

5 The radiotoxicity, on oral intake, of spent nuclear fuel and its main constituents expressed as a ratio versus the radiotoxicity of natural uranium (without daughter elements).

6 Left: Uranium content and activity ratio for uranium-234 to uranium-238 in surface waters. Right: Uranium content and activity ratio for uranium-234 to uranium-238 in groundwaters. Diagram after Osmond and Cowart, ref. 20. Approximate position of border line between oxidizing and reducing conditions indicated by present writer.

7 Uranium content versus depth in three boreholes at three different Swedish sites, G = Gideå, F = Fjällveden, S = Svartboberget. Range of HCO_3-concentrations in the samples is also indicated. Data from Laurent, rrs. 48,49,50.

8 The radiotoxicity of groundwater after contamination by spent nuclear fuel and reaching reducing conditions. The toxicity is here expressed as the corresponding concentration of natural uranium (without daughters), that would give rise to an equivalent radiotoxicity. The diagram is not valid for strontium-90, caesium-137 and radium-226, which would not be coprecipitated with uranium in the same way as the actinides.

9A Diagram of groundwater flow in a vertical section through the ground, assuming uniform hydraulic conductivity. After Hubbert, ref. 23. Shallow portions, carrying groundwater not reaching a deepseated repository are indicated by present writer.

9B Order-of-magnitude estimates of hydraulic head and flow volumes per year in a one metre thick vertical section through ground with a schematic, yet semirealistic, distribution of hydraulic conductivities. A horizontal fracture-zone, as indicated in the diagram, would channel all groundwater contacting spent fuel above it, to the surface. An inclined or vertical fracture-zone would only affect part of the inventory.

10 The radiotoxicity of groundwater reaching the surface environment after contamination by spent fuel at depth and passing reducing conditions on its way. Not valid for strontium-90, Caesium-137 and radium-226.

11 Examples of tracer tests illustrating groundwater transport of nuclides in fracture zones in granitic and gneissic rocks.

A
Breakthrough curve for iodide (nonsorbing) in a pulse test. Distance between points of injection and observation 30 m. Horizontal axis is time (hours) after injection, vertical axis observed concentration as fraction of the injected concentration. Transport is found to occur along two main channels in the rock. After ref. 31, figure 6.2.3a.

B
Breakthrough curve for iodide at continuous injection, which more closely represents conditions pertaining to contamination from emplaced fuel. Same condition as in A, same reference, fig 6.2.3b.

C
Breakthrough curve for a pulse test with bromide (Br-82), nonsorbing, corrected for radioactive decay. Distance between points of injection and observation 52 m. From ref 30, redrawn after figure 13.

D
Breakthrough curve for strontium-85, corrected for radioactive decay. A nuclide susceptible to sorption injected simultaneously with the bromide shown in C. Retardation as well as reduction in concentration is observed. From ref 30, redrawn after figure 12.

Figure 1

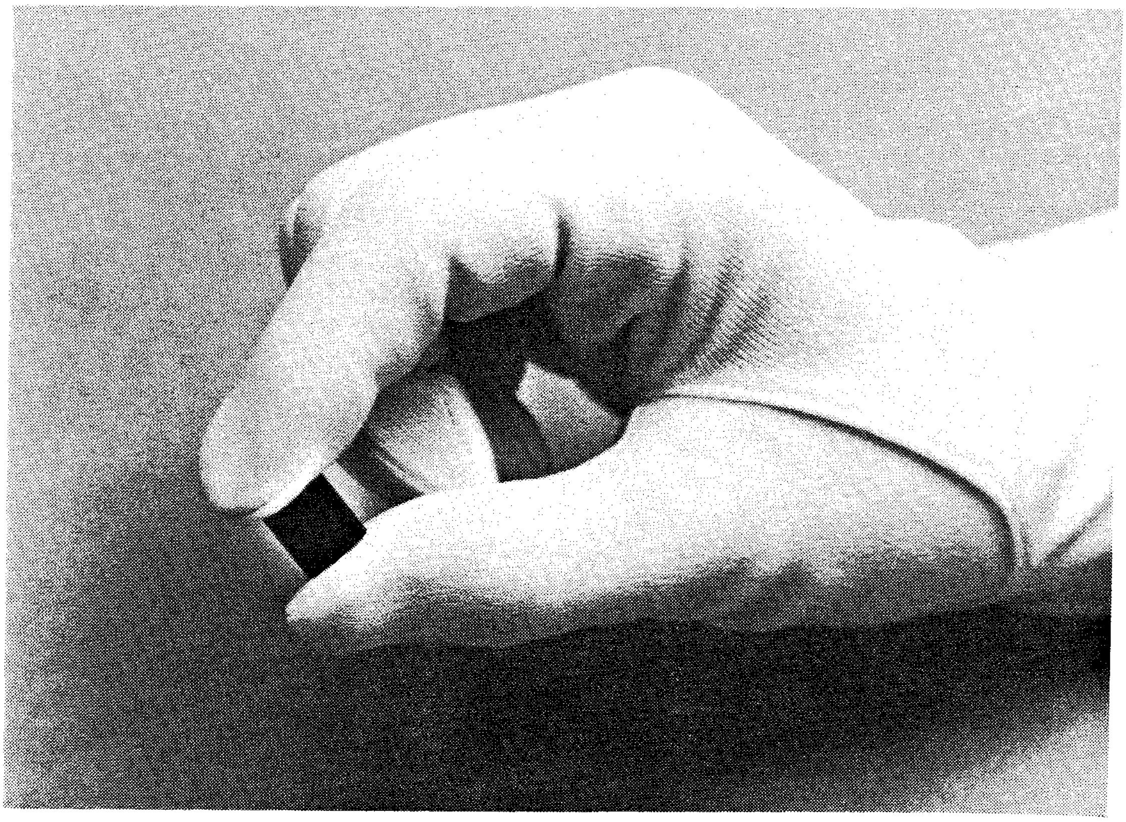


Figure 2

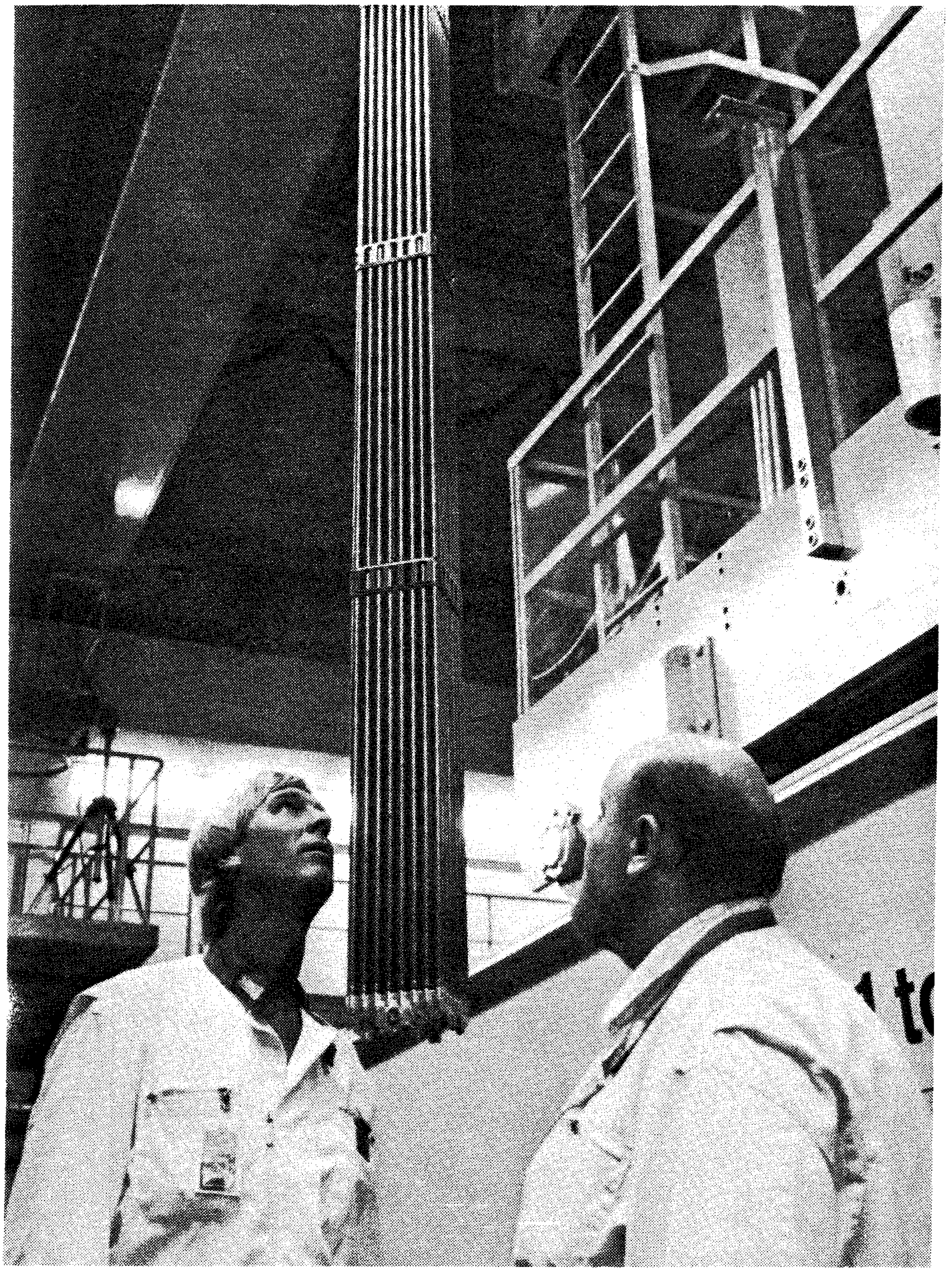


Figure 3

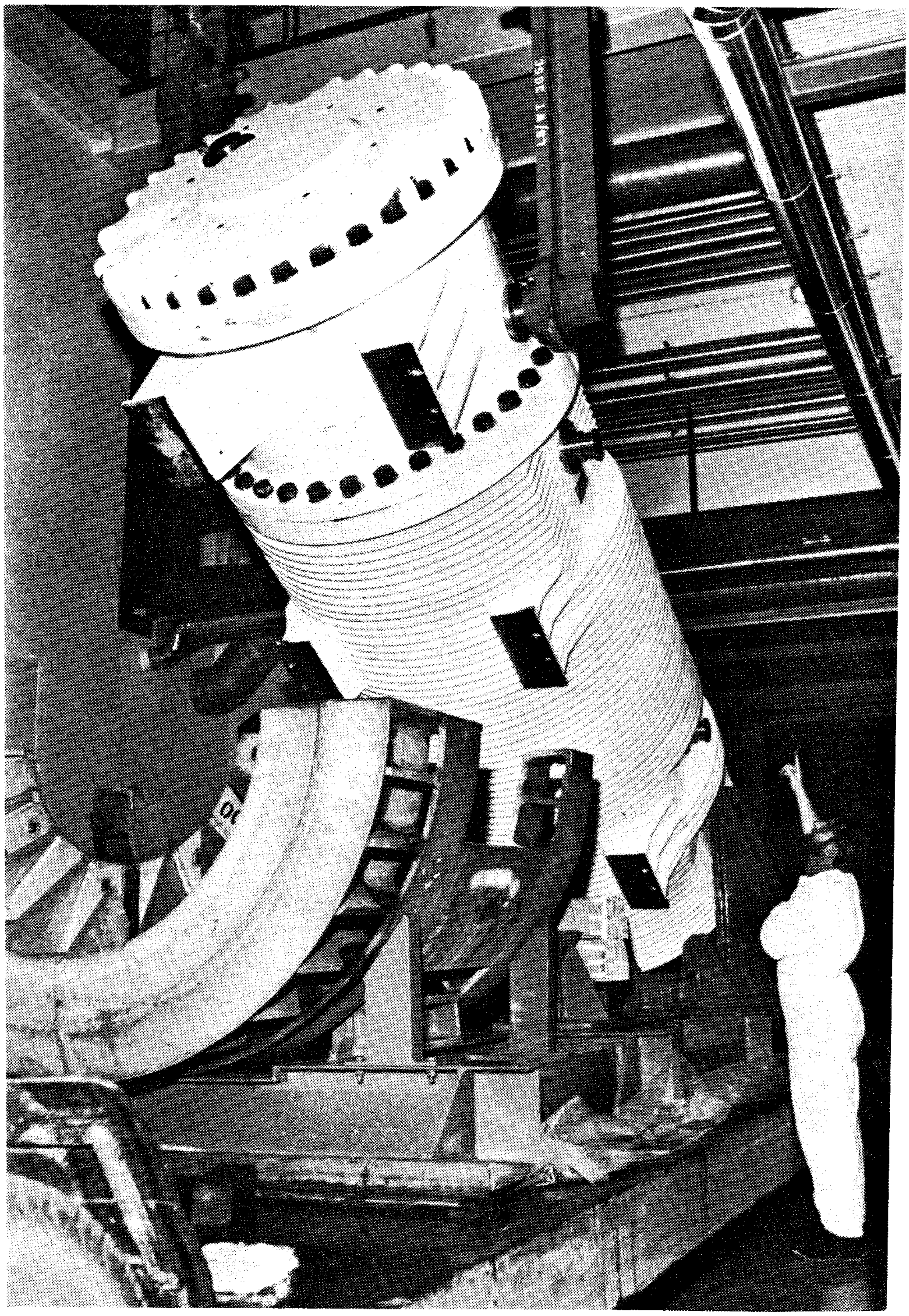
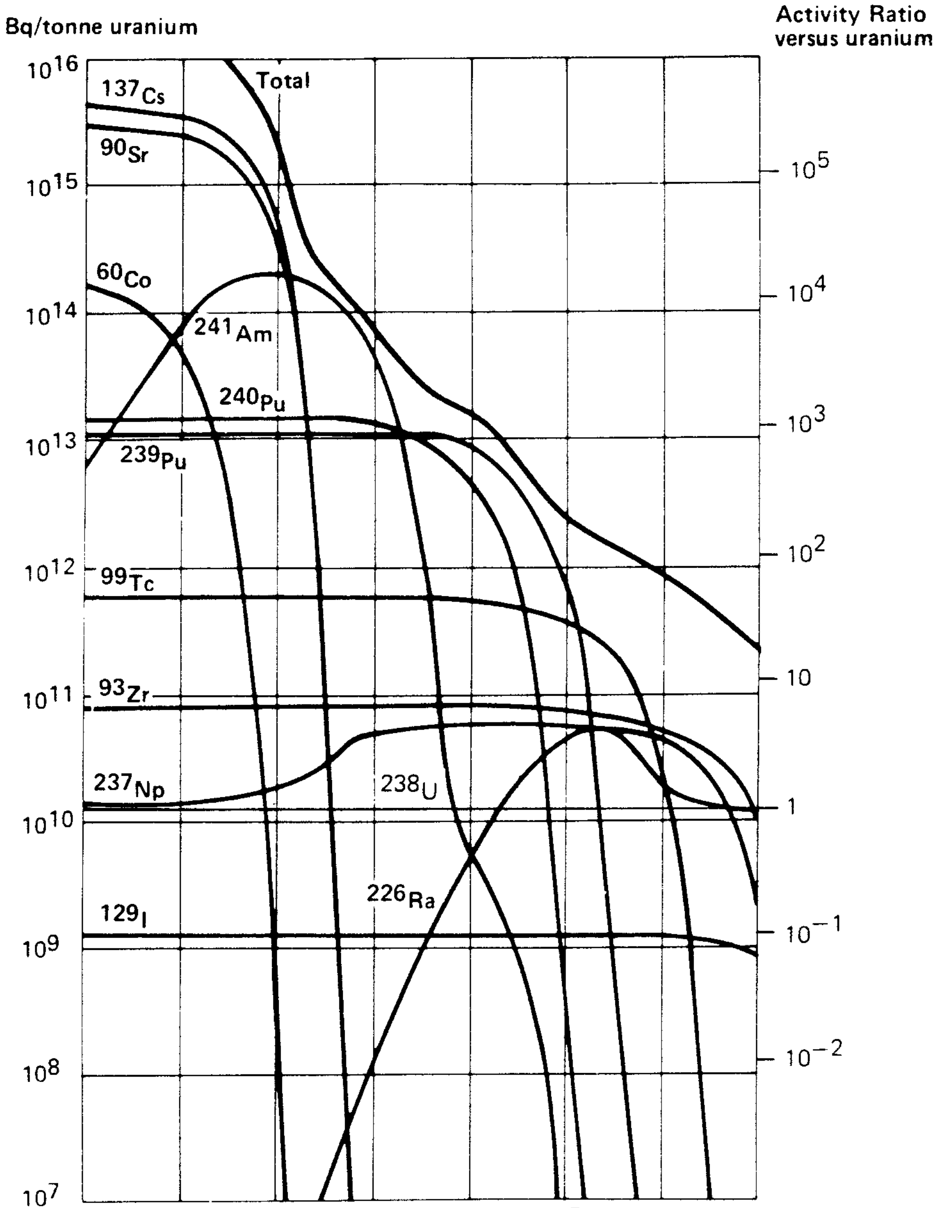


Figure 4



Bq/tonne uranium
Activity Ratio versus uranium
10^{16}
10^{15}
10^{14}
10^{13}
10^{12}
10^{11}
10^{10}
10^{9}
10^{8}
10^{7}
10^{5}
10^{4}
10^{3}
10^{2}
10
1
10^{-1}
10^{-2}
^{137}Cs
^{90}Sr
Total
^{60}Co
^{241}Am
^{240}Pu
^{239}Pu
^{99}Tc
^{93}Zr
^{237}Np
^{238}U
^{226}Ra
^{129}I
1
10
10^{2}
10^{3}
10^{4}
10^{5}
10^{6}
10^{7}
Time after discharge from reactor, years

Figure 5

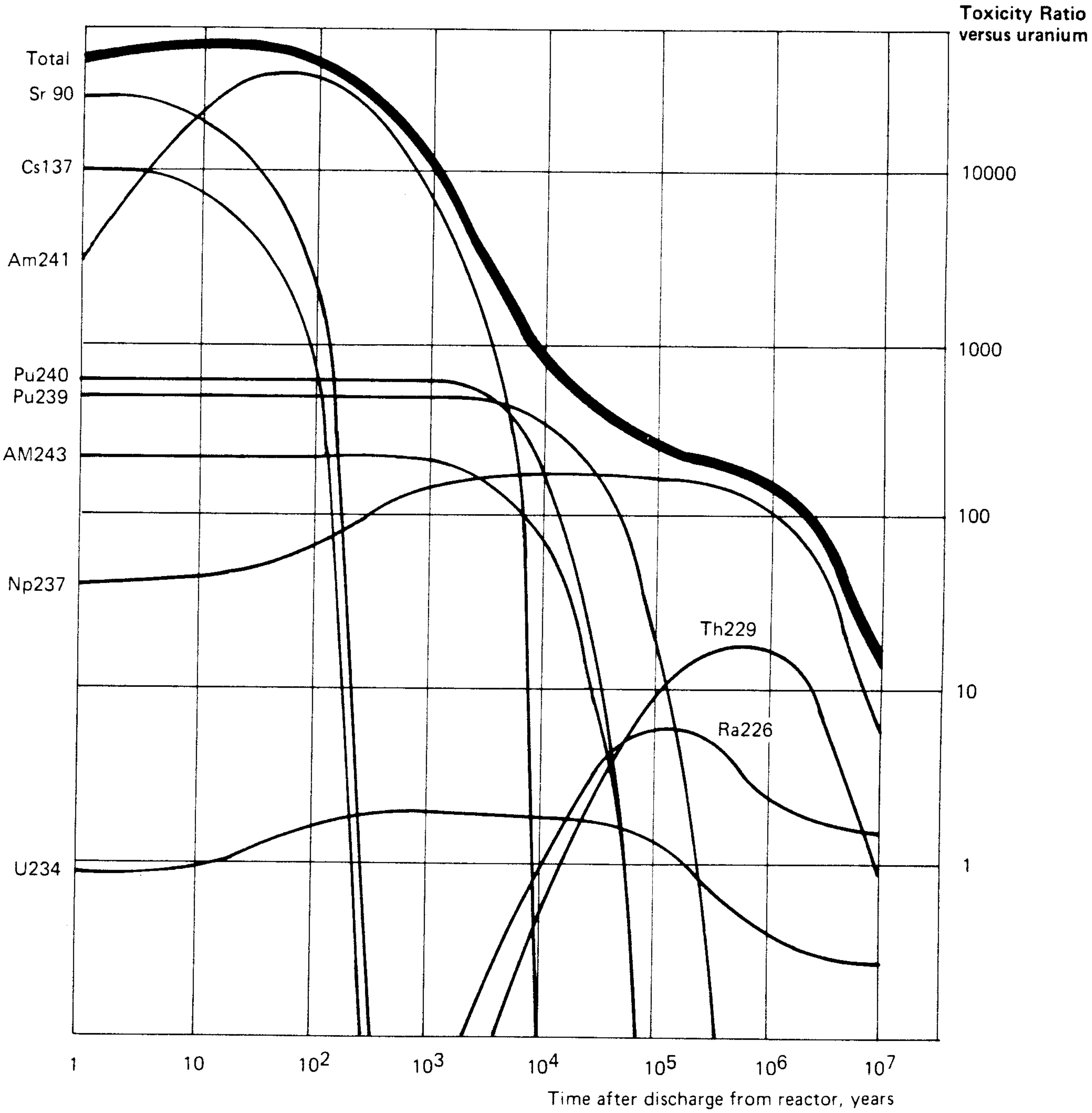
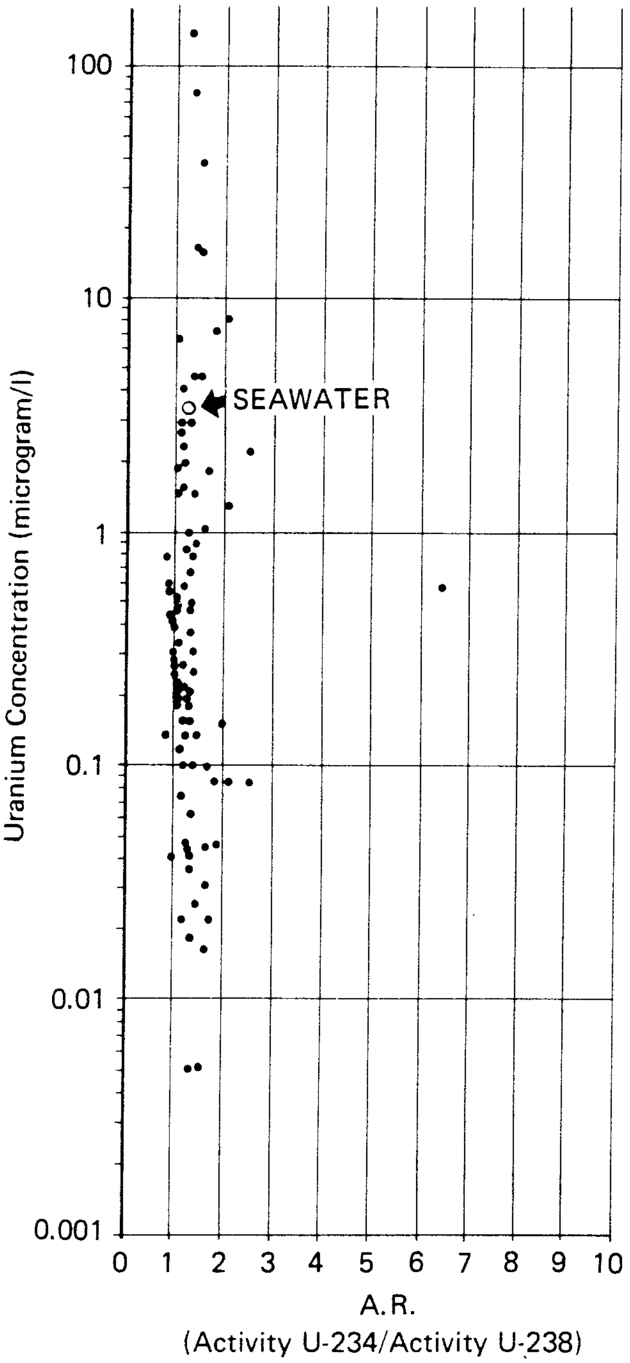
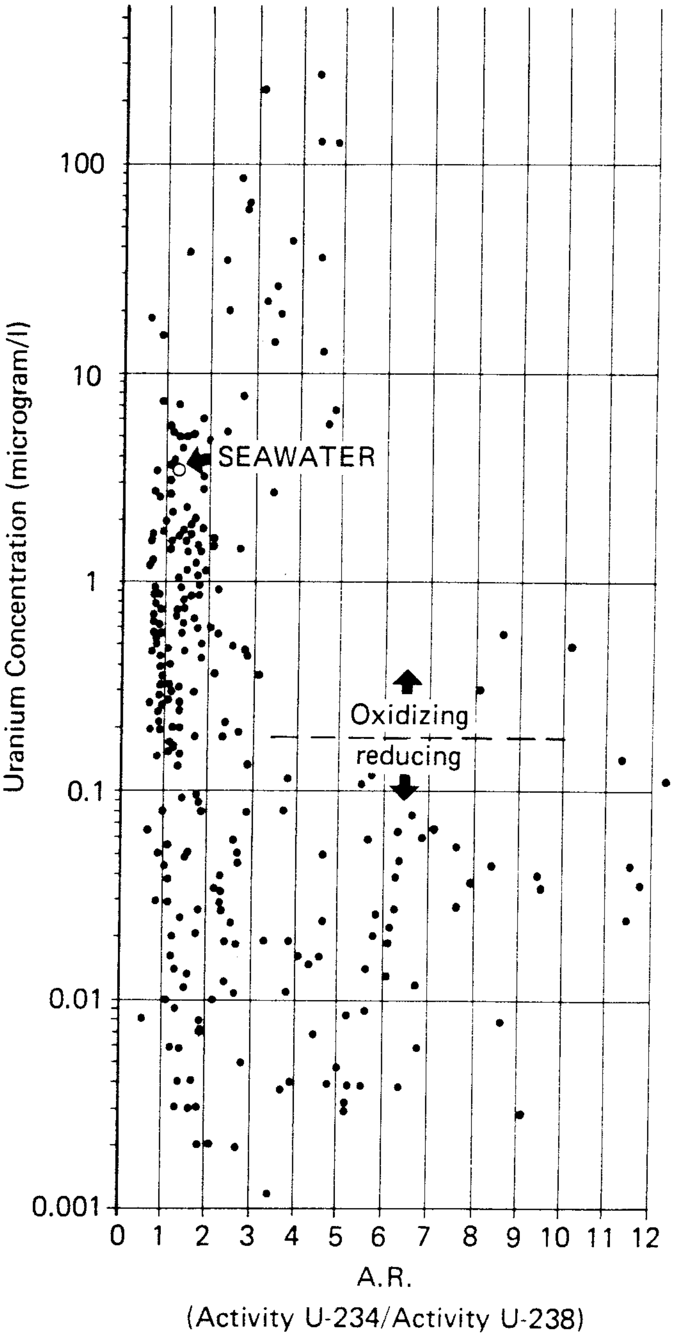

Toxicity Ratio versus uranium
Total
Sr 90
Cs137
Am241
Pu240
Pu239
AM243
Np237
U234
Th229
Ra226
10000
1000
100
10
1
1
10
10^2
10^3
10^4
10^5
10^6
10^7
Time after discharge from reactor, years

Figure 6



Uranium Concentration (microgram/l)
100
10
1
0.1
0.01
0.001
SEAWATER
0
1
2
3
4
5
6
7
8
9
10
A.R.
(Activity U-234/Activity U-238)



Uranium Concentration (microgram/l)
100
10
1
0.1
0.01
0.001
SEAWATER
Oxidizing
reducing
0
1
2
3
4
5
6
7
8
9
10
11
12
A.R.
(Activity U-234/Activity U-238)

Figure 7

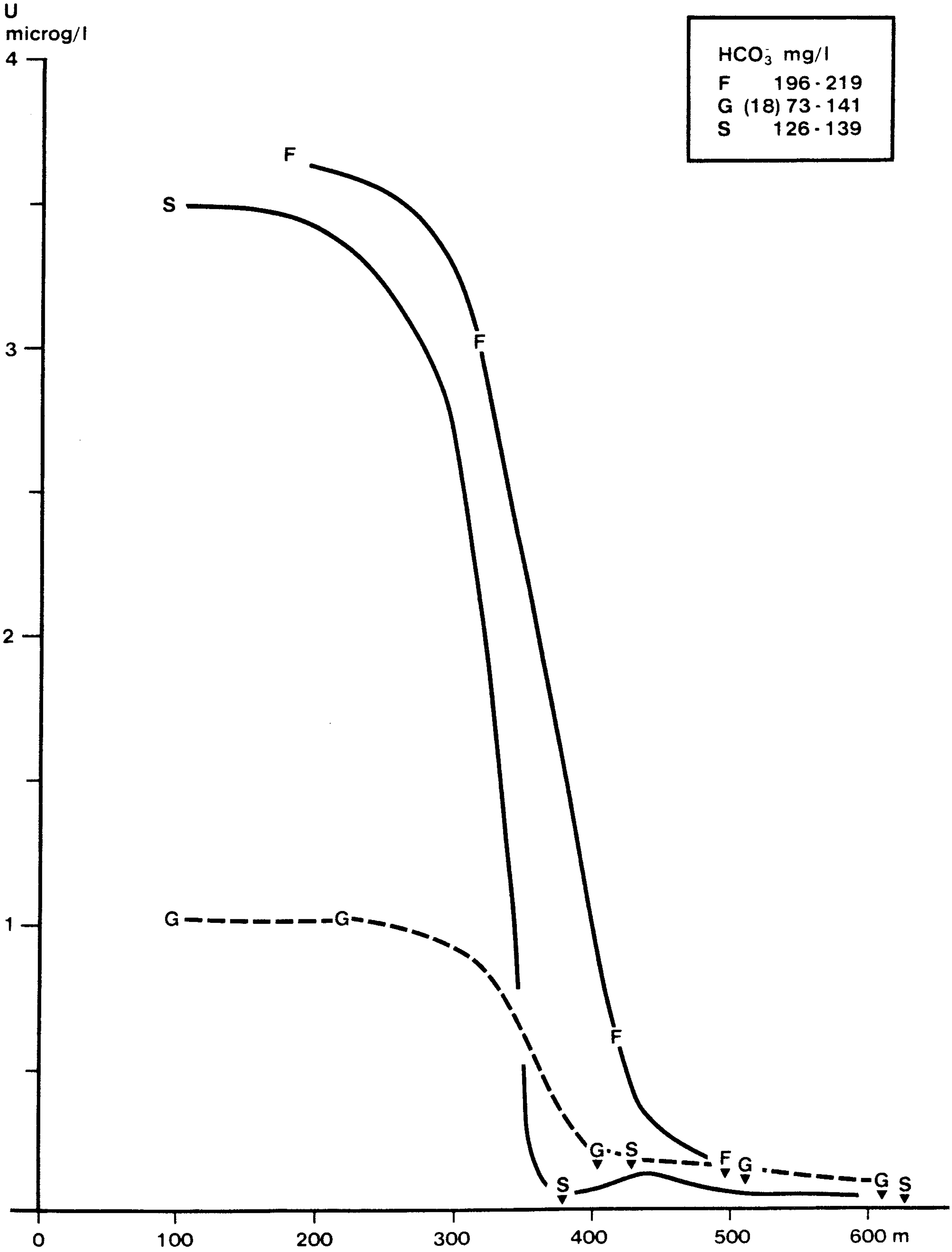

U
microg/l
4
3
2
1
0
100
200
300
400
500
600 m
HCO$_3^-$ mg/l
F 196-219
G (18) 73-141
S 126-139
F
S
F
G
G
F
G
S
S
F
G
G
S

Figure 8

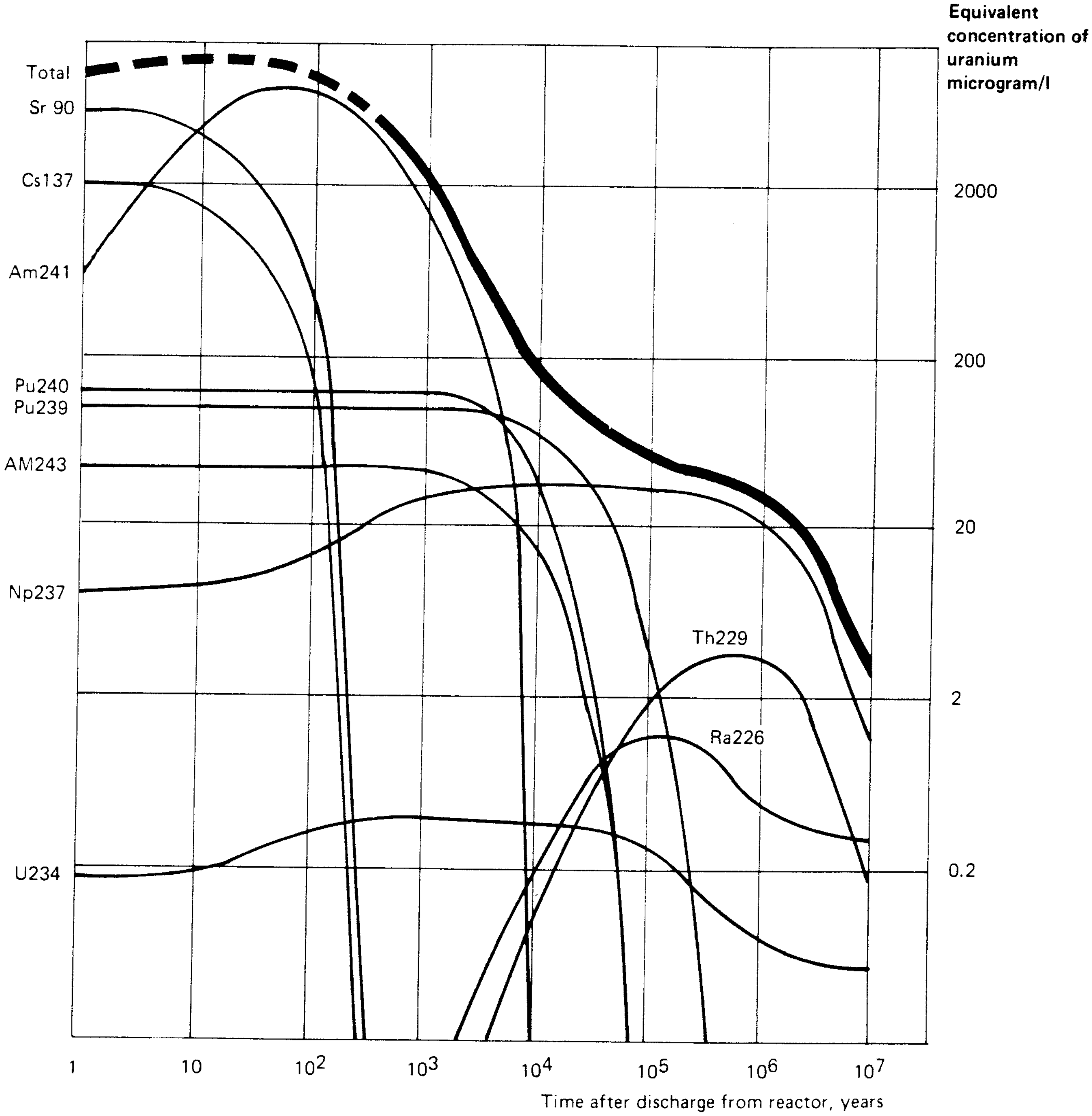

Equivalent concentration of uranium microgram/l
Total
Sr 90
Cs137
Am241
Pu240
Pu239
AM243
Np237
U234
Th229
Ra226
2000
200
20
2
0.2
1
10
10^2
10^3
10^4
10^5
10^6
10^7
Time after discharge from reactor, years

Figure 9

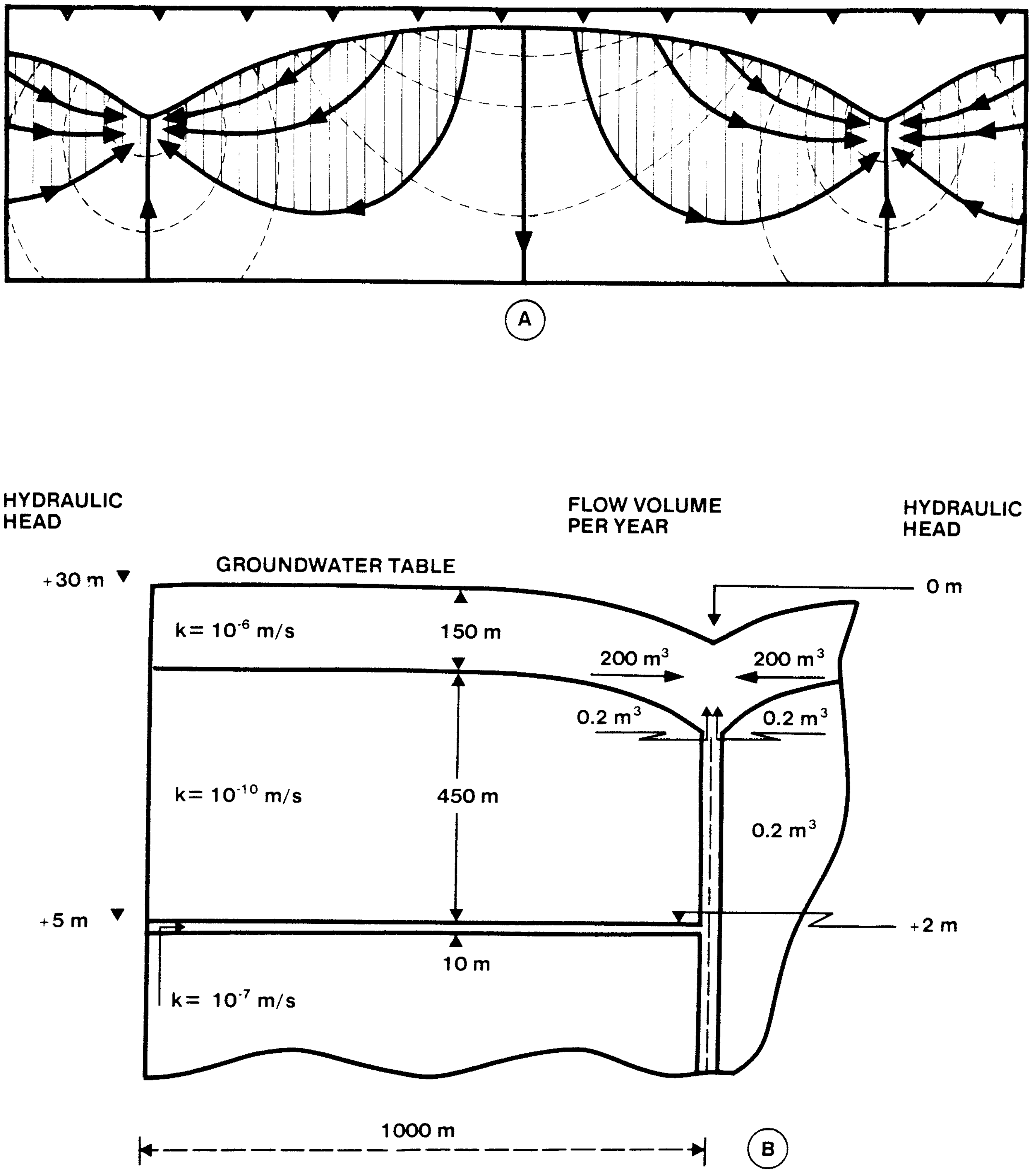
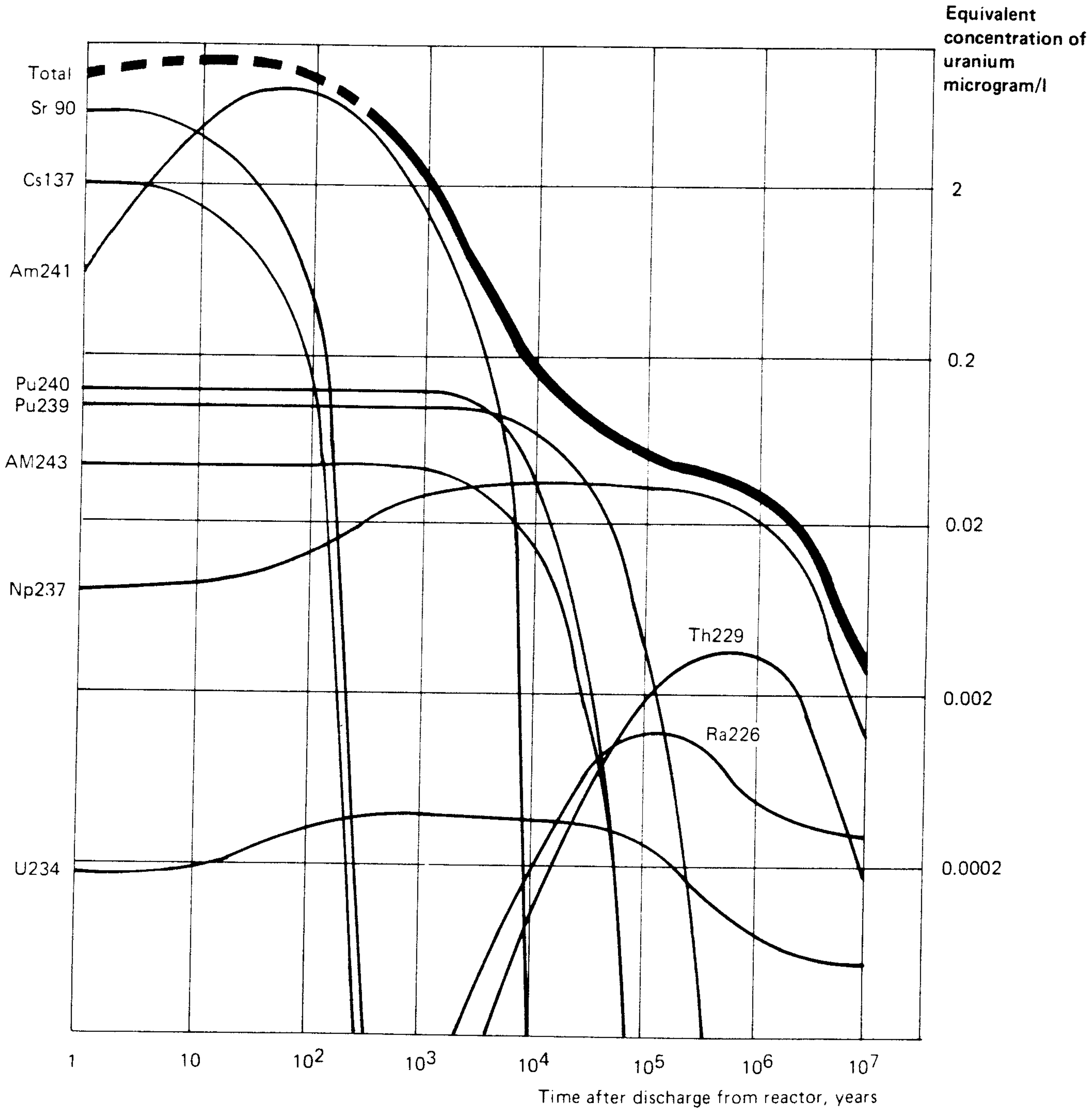

A
HYDRAULIC HEAD
FLOW VOLUME PER YEAR
HYDRAULIC HEAD
GROUNDWATER TABLE
+30 m
0 m
k= 10⁻⁶ m/s
150 m
200 m³
200 m³
0.2 m³
0.2 m³
k= 10⁻¹⁰ m/s
450 m
0.2 m³
+5 m
+2 m
10 m
k= 10⁻⁷ m/s
1000 m
B

Figure 10


Equivalent concentration of uranium microgram/l
Total
Sr 90
Cs137
Am241
Pu240
Pu239
AM243
Np237
U234
Th229
Ra226
2
0.2
0.02
0.002
0.0002
1
10
10^2
10^3
10^4
10^5
10^6
10^7
Time after discharge from reactor, years

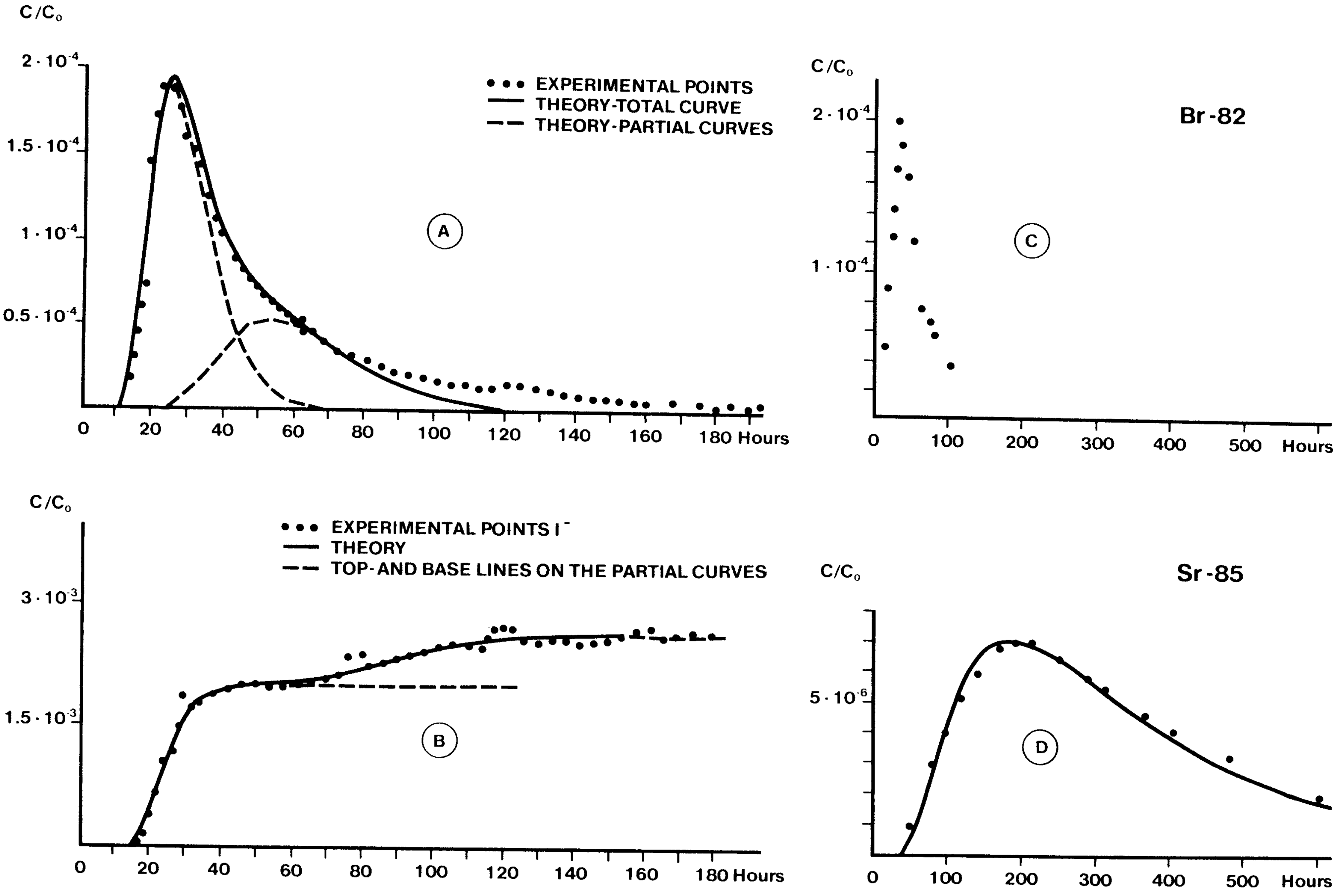

C/C$_o$
2 · 10^{-4}
1.5 · 10^{-4}
1 · 10^{-4}
0.5 · 10^{-4}
EXPERIMENTAL POINTS
THEORY-TOTAL CURVE
THEORY-PARTIAL CURVES
A
0
20
40
60
80
100
120
140
160
180 Hours
C/C$_o$
2 · 10^{-4}
1 · 10^{-4}
Br-82
C
0
100
200
300
400
500
Hours
C/C$_o$
EXPERIMENTAL POINTS I$^-$
THEORY
TOP- AND BASE LINES ON THE PARTIAL CURVES
3 · 10^{-3}
1.5 · 10^{-3}
B
0
20
40
60
80
100
120
140
160
180 Hours
C/C$_o$
Sr-85
5 · 10^{-6}
D
0
100
200
300
400
500
Hours

Figure 11

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I Grenthe
I Puigdomènech
J Bruno
Department of Inorganic Chemistry
Royal Institute of Technology
Stockholm, Sweden, January 1983

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February 15, 1983

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Roland Pusch
Division Soil Mechanics, Univeristy of Luleå
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Åke Bresle
Jozef Saers
Birgit Arrhenius
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D Hardie
R N Parkins
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Department of Metallurgy and engineering Materials
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I: Fission Products
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B Allard
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Chalmers University of Technology
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U Olofsson
B Allard
M Bengtsson
B Torstenfelt
K Andersson
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Chalmers University of Technology
Göteborg, Sweden 1983-01-30

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Göteborg, Sweden 1983-02-15

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Evidence from the Oklo natural reactors**
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