SKBF TECHNICAL
KBS REPORT
83-75

The geochemical behavior of protactinium 231 and its chosen geochemical analogue thorium in the biosphere

Marianne Gillberg-Wickman

National Defence Research Institute
Umeå, Sweden March 1983

SVENSK KÄRNBRÄNSLEFÖRSÖRJNING AB / AVDELNING KBS
Swedish Nuclear Fuel Supply Co/Division KBS
MAILING ADDRESS: SKBF/KBS, Box 5864, S-102 48 Stockholm, Sweden
Telephone 08-67 95 40

THE GEOCHEMICAL BEHAVIOR OF PROTACTINIUM 231 AND ITS CHOSEN GEOCHEMICAL ANALOGUE THORIUM IN THE BIOSPHERE

Marianne Gillberg-Wickman

National Defence Research Institute
Umeå, Sweden March 1983

A list of other reports published in this series during 1983 is attached at the end of this report. Information on KBS technical reports from 1977-1978 (TR 121), 1979 (TR 79-28), 1980 (TR 80-26), 1981 (TR 81-17) and 1982 (TR 82-28) is available through SKBF/KBS.

SUMMARY

To be able to judge whether protactinium 231 might represent a major contribution to the human radiation risk from high level radioactive waste a literature study of the geochemical behavior of protactinium has been made. The interest in protactinium determinations has, as far, been in the field of marine geochemistry and geochronology. These investigations show that thorium may be used as a chemical analogue.

The content of protactinium 231 is determined by the ^{235}U content and consequently the occurrence of protactinium in nature is directly associated to the geochemistry of uranium.

The pronounced hydrolytic tendency of protactinium and its great sorption and coprecipitation capacity ought to prevent or at least appreciably delay its transport from a back-filled nuclear waste vault to the uppermost surface of the earth. It also has a tendency to form colloids or particulates which may be strongly fixed on a rock surface. In adsorption and desorption processes kinetics must play an important role. Our knowledge in this field is quite limited.

Under the physico-chemical conditions in the sea, protactinium is rapidly scavenged from the water column by particulates. It accumulates in the sediments.

THE GEOCHEMICAL BEHAVIOR OF PROTACTINIUM 231 AND ITS CHOSEN GEOCHEMICAL ANALOGUE THORIUM IN THE BIOSPHERE

by

Marianne Gillberg-Wickman

Contents

INTRODUCTION

The question has been raised whether protactinium 231 with a half life of 32.8 ka, might represent a major contribution to the human radiation risk long time after the burial of high level radioactive waste. In order to do this it must reach the biosphere in such a form and concentration that it could be harmful.

To be able to judge this possibility a literature study of the geochemical behavior of protactinium is presented in this report. However, the sole interest in protactinium determinations has, as far, been in the field of marine geochemistry and geochronology. These investigations show that thorium may be used as a chemical analogue.

The content of protactinium 231 is determined by the ^{235}U content and consequently the occurrence of protactinium in nature is directly associated to the geochemistry of uranium. Figure 1 shows the radionuclides in the upper parts of the three naturally-occurring radioactive chains. However, it is worth-while to have a closer look at the isotope fractionation phenomena in the ^{238}U series. For example, assuming we have a chemical uranium analysis of a groundwater sample and calculate ^{231}Pa using an activity ratio of 1.00 for $^{234}U/^{238}U$ and instead this ratio is 12, our calculation will give an overestimate of ^{231}Pa by 6.5 times.

The safety requirement of a geological repository comprises very long periods of time of the order of a million years. The concept of what is regarded as a long or a short period of time in geology, is quite different from the homo-centred scale we ordinarily use. For example in plate tectonics one million year is a very short period of time. Here we are talking mainly about the ingestion hazard (water and food-chains) and consequently our interest is centered to the uppermost surface of the earth and then one million years is a long time. Especially the drastic changes which have occurred during the last few million years with glaciations, changing climates, changing sea-levels and changing sediment supply to the sea, make this perfectly clear. Sediments are being formed by erosion and weathering and carried away by the transport agents and sooner or later deposited again. The soil may be regarded as a transient phase through which a large part of the rock debris from the continents must pass before its final removal. Most rivers reach the sea and pour into it their dissolved and suspended loads; making the sea the ultimate destination for most of the different kinds of surface water.

With all these dynamic processes going on all the time, the important thing, in this respect is trying to grasp the geochemical behavior of protactinium and thorium in the different relevant stages of their journey from the crustal rock to the sea and seafloor. For analytical reason the start is made at the endstation.

1 THE OCCURRENCE OF PROTACTINIUM IN NATURE AND ITS CHEMICAL PROPERTIES

1.1 Occurrence

Of the many protactinium nuclides known only ^{231}Pa and ^{234}Pa occur naturally. However, the half-life of the latter is short.

Protactinium 231 is a daughter of uranium 235, and has according to the latest accepted value a half-life of 32.8 ka. Of the three naturally occurring uranium isotopes ^{238}U and ^{235}U are extremely long-lived, whereas ^{234}U is far too short-lived ($t_{1/2}$= 245 ka) to be remanent from the formation of the earth. At equilibrium 1 g of natural uranium should contain 0.322µg ^{231}Pa. This calculation is based on the following values: ^{231}Pa: $t_{1/2}$ = 32.53 ± 0.25 ka, ^{235}U: $t_{1/2}$ = 0.704 Ga, 0.711 weight per cent of ^{235}U in natural uranium. The average value for crustal abundance of protactinium is not agreed upon, since there is a disagreement about the average crustal abundance of uranium.

In ordinary igneous rocks the trace elements very seldom become concentrated enough to form discrete minerals. In granitoid igneous rocks the accessory minerals zircon, sphene and apatite are likely hostminerals for uranium and thorium. The remaining amount is distributed in very low concentration in rock-forming minerals; as isomorphous substitution, in lattice imperfections and in dislocations. Grain-boundaries and fractures in a mineral grain are also likely sites and they also occur in fluid inclusions.

There are very few reliable experimental data on the concentration of protactinium in specific natural uranium ores.

1.2 Chemical properties of protactinium

Protactinium has a great tendency to adsorb on all kinds of solid-liquid interfaces; a tendency which complicates the analytical procedure. It also adsorbs on colloidal precipitates.

In aqueous solution the most stable oxidation state is +5. It also occurs as +4, whereas the oxidation state +3 does not exist in solution. Pentavalent protactinium is difficult to reduce, and Pa(IV) oxidizes rapidly by air to Pa(V). Pa(V) has, similar to thorium, a very pronounced hydrolytic tendency. This may be the reason for the extreme low thorium/uranium ratio in natural water. There is less than 70 pg/l of thorium in surface sea water. Much of the published data concerning actinide hydrolysis, a process which is important in nature, is made fairly long ago. According to Kuznetsov et al, 1966, if microamounts of protactinium occur in aqueous solution it undergoes hydrolysis which at pH >3 leads to ionic species of type $Pa(OH)^{4+}$,$Pa(OH)^{3+}{}_2$, whereas at pH > 5 colloids are formed. According to Allard, 1983 (pers. comm.) Pa(V)-species are: $PaO(OH)^{2+}$, $PaO(OH)^{+}{}_2$ and maybe $PaO_2(OH)$. I have not been able to find anything about the colloid stability in solutions with higher ionic-strength. Protactinium also forms mixed ligand complex like $(Pa(OH)_nCl_m)^{5-n-m}$. It also seems to be able to form metal-organic complexes with fulvic acid.

In trace amounts pentavalent protactinium is coprecipitated almost quantitatively by many hydroxides (cf the hydrolysates), it is also carried by some carbonates. (Coprecipitation and sorption are two phenomena which are often hard to distinguish from each other. In both cases a microcomponent is separated from a solution by reactions on a solid surface.)

1.3 Isotope fractionation effects in the uranium 238 and thorium 232 series

Uranium 234 is generated in nature by decay of its parent ^{238}U and consequently, at secular equilibrium their activity ratio should be 1.00. However, about 25 years ago it was discovered that large deviations in this activity ratio commonly occur in nature. It has been demonstrated that disequilibria can be found in most groundwaters, rivers, lakes, the ocean, in some rocks, minerals and uranium ores. Usually, all natural waters have an amount of ^{234}U which is higher than the equilibrium amount. It is inferred that the rocks from which the uranium is derived have a deficiency of ^{234}U. This loss of ^{234}U relative to ^{238}U has long been attributed to recoiling alpha-emitting nuclei. Experimental evidence on the mechanisms have been largely lacking until recently (cf Fleischer, 1980). Significant isotope fractionation effect between ^{238}U and ^{235} U does not occur in nature, the only exception is the Oklo reactor Gabon ore-body, West Africa. Other pairs that exhibit fractionation effects in nature are $^{228}Th/^{232}Th$ and $^{224}Ra/^{228}Ra$.

2. THE DISTRIBUTION OF PROTACTINIUM AND THORIUM IN THE OCEAN

The importance of thorium and also protactinium in marine geochemistry has been recognized since the early 1950s. Since the oceans are the ultimate destination for most of the different kinds of surface water, the interest has been centred on the geochemical balance of the isotopes in the ocean and the Pleistocene geochronology of the deep-sea sediments. Another important topic in Pleistocene geochronology is dating of coral reefs and sea level stands by the study of **deficiencies** of ^{230}Th and ^{231}Pa relative to uranium in coral reefs.

In 1957, Koczy et al, were able to positively prove the disequilibrium of thorium 230 in sediments and water. The thorium isotopes 234, 230 (^{238}U) and 228 (^{232}Th) are produced in situ in sea-water whereas ^{232}Th is only supplied to the sea by weathering of crustal rocks (terrigenous material). By the development of alpha spectrometric methods and better separation techniques it was possible to determine protactinium 231, which is a daughter isotope of uranium 235. The study of ^{230}Th and ^{231}Pa in deep-sea sediments has generally two objectives: age determination of buried sediment horizons and the rates of sediment accumulation. This is possible since both ^{230}Th and ^{231}Pa are daughter isotopes of uranium. Changes in sedimentation rates are usually interpreted due to changing glacial conditions on the continents.

Unfortunately, there exist much more data for thorium contents in sea water and sediment cores than for protactinium. The reason is the difficulties involved in determining small amounts of protactinium. Sackett, 1960, reports that the ^{231}Pa content in ocean water is less than 3 per cent of the amount which should be in equilibrium with a total uranium content of 3 μg/l in ocean water. This deficiency of ^{231}Pa in the ocean water indicates that ^{231}Pa is removed as it is formed from the radioactive decay of ^{235}U in a time which is much shorter than its half-life ($t_{1/2}$ = 32.8 ka). This behavior is very similar to that of ^{230}Th. As in the case of ^{230}Th he found unsupported (meaning the amount of daughter isotope not supported by its parent, $A_d/A_p > 1$, by some authors called excess or XRD) ^{231}Pa in the top layers of Equatorial Pacific Ocean deep-sea sediments (6 samples).

In the paper by Moore and Sackett, 1964, the initial purpose was to set reasonable bounds about the amounts of ^{230}Th and ^{231}Pa that remain in solution. The residence time for both the isotopes was also calculated. Their water samples were from the North Atlantic and the Caribbean (cf Table 1). Their conclusions are that ^{231}Pa and ^{230}Th are quantitatively removed from sea-water as they are formed. Only 0.2 per cent of protactinium and 0.05 per cent thorium of the equilibrium amount were detected in sea-water (3.7 μg/l uranium in sea-water, ^{234}U/^{238}U activity ratio 1.14, no fractionation effect between ^{235}U/^{238}U). Assuming steady state conditions, they obtained a residence time less than 50 years for ^{230}Th and less than 100 years for ^{231}Pa.

Kuznetsov et al, 1966, have in detail and from different aspects investigated 19 deep-sea sediment cores from the southern part of the Indian Ocean (the majority of the samples) and the Equatorial part of the Pacific. The isotopes determined are ^{231}Pa, ^{230}Th and ^{238}U. They have also determined the granulometric composition of the samples and the total surface area of the samples. They found that protactinium and thorium behave practically indentical; the correlation coefficient being 0.95 (cf Table 2). In the open oceans there are three main types of sediments; those which contain calcium carbonate (foraminiferal sediments), those containing silica (diatomaceous and radiolarian sediments) and the red clay of the deep-sea mainly consisting of aluminum silicates and iron compounds. In their material the contents of protactinium and thorium varies from 1 to 8 pg/g and from 0.3 to 6.6 ng/g respectively. The lowest concentrations were found near the coast of the southern part of the Indian Ocean, which also contains an ice zone. Low concentrations were also found in some foraminiferal and diatomaceous sediments from the same general area. Sediments from the deeper parts of the ocean show increasing contents of protactinium and thorium. The maximum values were found in red clay from the Pacific. Table 2 shows the correlation coefficients between respectively Pa and Th and different components in the sediments and size fractions. **The most likely mechanism for their accumulation in the sediments is sorption.**

Osmond, 1979, concludes from data of deep-sea sediments that ^{231}Pa and ^{230}Th appear to be responding differently to compositional variables of the sediments and/or to the different accumulation rates. Due to the difficulties involved in protactinium analyses it is hard to prove.

Ku and Broecker, 1967, report in an investigation of a manganese nodule from the North Pacific that most of ^{230}Th and ^{231}Pa produced in the water column are apparently incorporated in the adjacent sediments rather than into the Mn-nodules (cf Kutznetsov et al, 1966, Table 2).

There are many thorium analyses by different laboratories of surface water from the open ocean. Kaufman, 1969, gives a value of less than 70 pg/l of ^{232}Th. The results of Bhat et al, 1969, show an average of 0.12 year removal time of ^{234}Th from surface water.

Imai and Sakanoue, 1973, found a remarkable disequilibrium between ^{230}Th and ^{234}U (a precursor of ^{230}Th) in surface seawater from North Pacific and East China Sea. They found only 0.020 per cent of the equilibrium amount in the samples from North Pacific and 0.078 per cent in those from the East China Sea. They have only 4 surface water samples in which ^{231}Pa is determined. These values are in accord with Kuznetsov et al, 1966, from the Equatorial Pacific and slightly higher than those in the North Atlantic obtained by Moore and Sacket, 1964.

Broecker et al, 1973, suggest that the thorium produced within the open surface ocean is effeciently removed by each generation of plant matter. The removal time for ^{228}Th, 0.7 year, (activity ratio ^{228}Th/^{228}Ra = 0.21 average "open" ocean sample) is roughly the time plant matter is cycled within the open surface ocean. Unlike phosphorus atoms which are reused about 10 times, thorium is carried below the base of the mixed layers after its first incorporation into plant matter.

Aller et al, 1976, have made a study of the behavior of ^{234}Th ($t_{1/2}$ = 24.1 d) in the Long Island Sound, USA. The sediment-water interface is the site of intense interaction between physical and chemical processes in the marine environment. Because of the complexity and rapid rates of these interactions in the upper 10-30 cm of marine deposits, this sedimentary zone is poorly understood.

The area chosen in the Long Island Sound is undisturbed in any direct way by large scale human activity as dumping. It has been studied biologically and physically during 1972 to 1975. The water samples collected in the fall 1974 showed a ^{234}Th/^{238}U activity ratio of 0.04! They report a residence time for ^{234}Th of 1.4 days, and point out that this value may be a maximum value. Their investigation shows that ^{234}Th is rapidly scavenged from the water column by particles. Upon falling to the sea floor these particles are reworked in the sediments by both biogenic and physical activity. The rate of rapid particle reworking varies seasonally and is highest in the fall.

Summarizing the behavior of protactinium and thorium in the oceans the following conclusions can be drawn:

Protactinium and thorium behave practically identical both in sea-water and deep-sea sediments.

The content of ^{231}Pa in sea-water is less than 5 per cent of the equilibrium amount. Corresponding value for ^{230}Th is less than 1 per cent.

Surface sea-water contains less than 70 pg/l of ^{232}Th. By comparison ^{238}U, which is three times less abundant in terrestrial rocks, has a concentration of 3 μg/l in sea-water.

^{231}Pa and ^{230}Th are quantitatively and rapidly removed from sea-water as they are formed from the radioactive decay of uranium. They accumulate on the sea-floor, supposedly adsorbed on sediment particles.

Some deep-sea sediments contain very large excess of ^{231}Pa and ^{230}Th in comparison with the equilibrium amounts. High content of especially iron but also manganese in the size fraction less than 0.001 mm and a large surface area of the sediment together with a low content of biogenous components, give an increasing content of ^{231}Pa and ^{230}Th in the sediments.

3 SURFACE RUN-OFF WATER, SEDIMENT AND EROSIONAL FLUXES

The elemental composition of river-water varies with climate and local geology. Also the presence of large cities, industrial plants and intense agriculture influence the composition. By convention dissolved material in rivers is defined as the material which is not removed by a 0.45 μm filter. Since the average residence time of river water is very short, (range in estimates a few days to 12 days) the rivers return their own volume of water to the sea many times a year. For this reason river composition can change rather rapidly in response to external conditions. The old saying by Heraclitus of Ephesus (about 500 B.C.) "You cannot step twice in the same river, for fresh waters are ever flowing upon you" is pertinent. In comparison with especially deep groundwater but also sea-water, river-water usually contains much larger amount of organic material, some of which can act as a ligand. Other complicating factors are that hydrolysis reactions and formation of chelates may be quite slow processes, and consequently equilibrium is not established before they enter the sea.

The average concentration of uranium in rivers is not agreed upon. The values may vary widely from river to river and from season to season. Since the activity ratio $^{234}U/^{238}U$ in rivers is better known than the uranium concentration Osmond and Cowart, 1976, calculated the residence time of uranium in the sea based on uranium isotopic data. They obtained 280 000 years, which is in the lower range of estimated residence times (up to 725 000 years) based on river data. The estimated residence times for thorium and protactinium in the sea are less than 50 and 100 years, respectively (cf p 4).

Garrels and MacKenzie, 1971, have estimated the dissolved and suspended load of rivers and concluded that the world's landmasses are being eroded by chemical versus mechanical processes in the ratio of 4.7:1.0. Similar results have been obtained by some other geoscientists. Relatively insoluble elements like iron and manganese are concentrated in or on the surface of hydrolysate minerals, and are being transported in pathway proportions (chemical to mechanical) which are less than 4.7:1.0. Since the ^{232}Th content in sea-water is extremely small, some even claim that the values obtained are due to pollution, one can assume that the pathway proportions for protactinium <<4.7:1.0.

Osmond and Cowart, 1976, calculated these pathway proportions for uranium in a very elegant way, using activity ratio data. Their calculation showed that there is **5 times as much uranium transported with sediments as in the dissolved state**. In massbalance calculations involving the hydrologic cycle, underground water seepage into the sea is disregarded. The flux of neither ^{238}U nor ^{234}U could be very significant along this pathway. An important assumption of their balance calculation is that the escaping components are in balance at equilibrium. The low activity ratio in soil will tend to revert towards equilibrium by the decay function. This will lead to an underestimation of the deficiency of ^{234}U in the sediments and make their ratio an upper limit. Also changes in Eh conditions may lead to adsorption of dissolved uranium on particulate material in the river.

With respect to regional run-off water a high (> 1.5) ^{234}U/^{238}U activity ratio is characteristic of run-off where the ratio of chemical to mechanical weathering is low. Medium or low activity ratios are characteristic of regions where chemical weathering is more important (Osmond and Cowart, 1976).

4 THORIUM AND URANIUM IN SOIL AND SOIL-FORMING PROCESSES

The soil is a phase through which a large part of the rock debris from the continent must pass before it is ultimately removed. Soil develops not only on bedrock but also on a great variety of loose deposits transported into their present position by gravity, running water and moving ice.

In a soil the physical and chemical processes of weathering cooperate in intimate association with biological processes. All these processes depend on climate but also other factors are involved, the nature of the bedrock or deposit on which the soil is formed (the parent material), the relief of the land, the age of the soil and the superimposed effects of cultivation.

As a soil gets older, especially if the climate is of a more extreme type, the influence of long continued weathering and organic growth and decay get more and more important. The composition of the soil develops gradually into a certain type which is different for each climate region implying that the influence of the composition of the parent material gets smaller. Differences due to the type of bedrock are mostly marked in young soils and in temperate regions, where the mechanical weathering processes are more important than the chemical ones. For example a granite gives podzol (gray soil) in a temperate region, chernosem (black earth) in the steppes and lateritic soil (reddish earth) in tropical regions of seasonal rainfall.

The principal constituents in a soil are: coarse inorganic particles, colloidal inorganic particles, organic matter, living organisms, soil solution (pore water) and a gas phase. In a well-drained soil the gas is air of nearly normal composition, whereas in permanently frozen soil the oxygen content in air is replaced by CO_2 (Bowen, 1979).

The three main groups of organic compounds, which interact with the inorganic material in soil, are small molecules (different amino- and other acids), fulvate and humates (polymers) and enzymes. A review of the composition of these compounds and their interaction with rock fragments and inorganic colloids is given by Bowen, 1979. He also compiled data about the role of micro-organisms in soil-forming processes. The enrichment of carbon, nitrogen, phosphorous and sulphur commonly found in the upper layers of soils can be correlated with a large amount of organic matter.

A normal soil profile in a temperate climate consists of three different layers called A, B and C, where the C-layer is little modified by weathering or addition of organic material. Even a specific layer is not homogeneous. According to Bowen, 1979, a crumb of soil with a diameter of 3 mm or more may have an oxygenic outer layer and an anoxic core.

The rates of soil formation is a topic which, to my knowledge, is seldom treated. Soils in a temperate climate appear to have ages in the range 10^3-10^6 years (Bowen, 1979).

4.1 Migration and redistribution of thorium and uranium in soil

In general, soil systems as such are very complicated. Thorium and uranium distribution in soil is very intricate. Thorium isotopes occur in all three series (cf Fig 1) and furthermore in a lot of radionuclides of different elements with different chemical properties. It is essential to realize that the behavior of a certain isotope in a soil depends on the local environment on an atomic scale, for example whether the atom is inside a mineral grain, or in solution, adsorbed on the surface of a mineral grain or adsorbed by organic compounds.

To my knowledge there do not exist any protactinium analyses from undisturbed glacial derived soil or soil profiles in a temperate climate. In the following, thorium is used as a geochemical analogue to protactinium, which is based on their practically identical behavior in the ocean and its sediments. However, the content of ^{231}Pa is determined by ^{235}U and, since fractionation effects do not occur between $^{235}U/^{238}U$, by the ^{238}U content.

In unweathered (fresh) rocks, where secular equilibria are maintained, the activity ratios for parent-daughter nuclides in the uranium and thorium series should be 1.00. In weathered rocks (rock-water system) the wellknown acitivity ratio will be $^{234}U/^{238}U < 1$, $^{228}Th/^{232}Th < 1$ and $^{238}U/^{230}Th < 1$. The observed fractionation effects show that the daughter nuclides are leached faster than the parent nuclides. The ability for migration falls in the general order U > Ra > Th. However, the redox-potential plays an important role in the uranium migration-fixation processes.

Rosholt et al, 1966, have determined ^{238}U, ^{235}U, ^{234}U, ^{232}Th and ^{230}Th in glacial derived soils from Minnesota and Utah, USA. Of the three Minnesota deep soil-profiles, two consist of well-drained gray-brown podzolic Fayette soil developed on loess (a fine-grained sediment deposited by the wind) of the Late Wisconsin age (FF and WF), and the third soil-profile consists of moderately drained Bruizen, Kenvon soil developed on till of supposedly Early Wisconsin age (K). They also have two thin soils from Utah; one which was produced from till of estimated pre-Wisconsin age (SI) and the other one from till of Early Wisconsin age (S2). The three Minnesota profiles are very similar (cf Table 3) and the over-all calculated deficiency of ^{234}U is 8.3, 8.9 and 8.2 per cent, respectively. The two thin soild from La Sal Mountains, Utah, indicate a position in the isotopic evolution in accord with their relative ages. One shows a slight excess of $^{234}U/^{238}U$ (1.06) whereas the other one has a ratio of 1.00. This ratio arises from the competition between two processes leaching and subsequent sorption. The A-horizon is a very black brown forest soil with a very high content of organic material. The most extreme ^{234}U deficiency found in their material was 42 per cent and occurred in the C-horizon of a desert type soil profile (brown podzol with a low organic content) from Arizona.

On the basis of the data obtained for the whole material Rosholt and co-workers developed a vector model from the disequilibrium trends. The plane of reference is the relative amount of ^{230}Th compared to ^{234}U and ^{238}U. The model describes the migration of uranium in the complex geochemical processes of the soil-forming environment. They take into account three different processes:

1) Leaching of ^{234}U generated in place from the parent material during insipient weathering before soil-forming processes started and also leaching of isotopically undifferentiated uranium from parent material.

2) Preferential leaching of ^{234}U during the soil-forming processes.

3) Assimilation of transported uranium-234 with high $^{234}U/^{238}U$ ratio and exchange of this uranium with preexisting uranium in the soil. This assimilation and isotopic exchange occurs by capillary migration of a small but significant fraction of the uranium in the soil environment.

The requirements for these processes are:

a) Some thorium (230) may be leached from the system.

b) The dual behavior of ^{234}U, only atoms generated in situ can be preferentially leached.

c) Assimilation of transported uranium with above normal $^{234}U/^{238}U$ ratios in the organic phases of the upper soil horizons, and isotopic exchange in the soil environment.

All the soils investigated show how time-dependent these processes are. Well-developed soil environment and a sufficient period of time would produce an isotopic composition* (activity ratio), where $^{234}U/^{230}Th > 1$ and $^{238}U/^{230}Th < 1$. Thus the equivalent isotope composition* would approach $^{234}U > ^{230}Th > ^{238}U$ (cf rock-water system).

A very interesting investigation by Titaeva et al, 1978, shows that the **migration and plant uptake behavior differs between the natural radionuclides and those** belonging to the same series **added to the soil as contaminants a long time ago.**

They have studied the migration patterns of eight naturally occurring heavy radionuclides through the rock-water-soil-plant system in a large humid climatic zone in the USSR. In the same zone they also had the opportunity to study three soil profiles which were artificially contaminated with uranium ore-processing products. One soil profile was modified by long-term discharge of deep-layer waters on the diurnal surface of radioactive edge-water. To the other two soil profiles were added solid ore-processing waste free from any mobile components. The discharge ended 20 years prior to their investigation.

Of all the investigated nuclides (^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th, ^{226}Ra, ^{224}Ra, ^{210}Po,) only the ^{232}Th content was the same as in soil of the same type outside the contaminated area. The different thorium isotopes behave differently because of their origin. They conclude the following: ^{232}Th is combined with the soil genetically, occurring primarily in the soil matrix. ^{228}Th, which is short-lived ($t_{1/2}$= 1.9d) accumulates as a result of radioactive decay after the contamination was ended. It penetrates the soil, but it may be fixed by the organic-mineral complex, when transported with the pore-water. (In a humid climate there is reason to believe that besides uranium also thorium may migrate as water soluble fulvatic complex.) ^{230}Th enters the soil with the contaminants and should be less mobile than ^{228}Th and is firmly fixed in the organic-mineral complex. It penetrates deeper into soddy podzolic soil in comparison with soddy meadow soil, which is rich in organic material and colloids. Their investigation of the three plots shows that radioactive equilibrium is established neither in the uranium nor in the thorium series. The degree of equilibrium changes strongly with depth. It is associated both with the disequilibrium of the source of contamination and the different migration abilities of the radionuclides.

The biological accumulation coefficient, defined as the radionuclide concentration in plant ash, divided by the concentration in dried soil, for ^{238}U, ^{234}U, ^{230}Th are three times higher in the plot contaminated by the edgewater than in those contaminated by the solid waste. By comparing the normalized isotopic activity ratios in soil and plants from the same plot, they concluded that plants absorb radionuclides better from the mineral part of soil or pore water than from the organic-mineral complex.

---

*The expressions used are those by Rosholt et al, 1966. They are confusing, but common in mass spectrometrical work. For explanations of units and calculations see text below Table 3.

Megumi and Mamuro, 1977, have determined uranium series nuclides in soil samples originating from weathering of Japanese granites. They started their investigation by determining the particle size distribution in each soil sample. For each size-fraction the contents of ^{238}U, ^{230}Th, ^{226}Ra and Pb-210 were determined by gamma-spectrometry. They found that the concentration of these nuclides increased with decrease in the particle size < 0.1 mm (cf p 4 and Table 2). Since the interactions in the rock-soil system occur on the surface of particles and the surface area per gram of soil particles increases with decrease in particle size, surface adsorption plays an important role in the enrichment. However, one has to keep in mind that the mineralogical composition varies with each size-fraction because different minerals have different resistance to weathering. The activity ratio $^{230}Th/^{238}U$ is high (3:5), in the whole size-range which can be explained by much larger mobility of uranium than thorium. Compare the very low thorium content in ocean water (p 5).

For each size-fraction they also determined the contents of Ca, K, Na, Cu, Mn and Zn. The over-all correlation coefficients between some of these elements and ^{230}Th are 0.78 (Fe), 0.57 (Mn) and -0.11 (Ca). They show the same trend as those for the deep sea sediments (cf Table 2).

The complexity of the migrations of heavy radionuclides in the soil-forming environment is well formulated by Titaeva et al, 1978: "Biogeochemical pecularities of the migrations of heavy radionuclides in the environment are determined by their chemical properties and half-lives, the physico-chemical properties of the environment, and the peculiarities of the sources and environmental pathways of the radionuclides (and, thus, by their forms)."

Like Rosholt et al, 1966, Titaeva et al, 1978, points out that isotopic activity ratios provide a useful tool for studying the direction of radionuclide migration and its influence on observed distribution patterns. They also stress the different behavior of isotopes of the same element generated in place i.e. basically belonging to the mineral matrix and those transported, and supplied with a contaminant to the soil.

5 SUBSURFACE WATER; GROUNDWATER

It is generally agreed that transport by groundwater constitutes the only likely means of returning buried radionuclides to the biosphere. Many uranium analyses of groundwaters have been performed in connection with geochemical prospecting, but information about pH, Eh and important constituents are often lacking.

Since protactinium analyses have been performed only in ocean waters and deep-sea sediments, information of the mobility of the chosen geochemical analogue, thorium, in other natural waters are valuable. According to Langmuir and Herman, 1980, who have made a study of the mobility of thorium in natural waters, the chemical and isotopic analyses have lacked sufficient geochemical, mineralogical, geologic and/or hydrologic data to allow quantitative predictions of the behavior of thorium.

They conclude that although complexing increases the solubility of thorium-bearing minerals below pH 8, the total thorium concentration, which rarely exceeds 1 ng/g in natural waters, is probably limited by the scarcity and slow solution rate of these minerals and by sorption processes rather than by mineral-solution equilibria.

Groundwater originates from precipitation by infiltration through soils and weathered zones in crustal rocks. These weathered zones are the principal place for uranium isotope fractionation phenomena.

The main different pathways for uranium in the weathering-erosion part of its geochemical cycle are (Osmond and Cowart, 1976 p 648):

a) Weathered rock fragments where ^{234}U is deficient relative to equilibrium.

b) Dissolved in oxidized surface and shallow groundwater where ^{234}U is in excess.

c) Dissolved in deep reduced groundwater where the concentration of dissolved uranium is very low but ^{234}U shows very high excess.

d) Trapped in the recycling system by downward percolating oxidized water and a reducing barrier, up-dip the dissolved uranium concentration may be high and ^{234}U may show a very large deficiency.

The first two pathways may be the normal routes to the final destination i.e. the sea, whereas the contribution of the last two must be small, since process d) will cease if the recycling uranium escapes.

Groundwaters show much greater variation in the activity ratio $^{234}U/^{238}U$ than surface waters do. Osmond and Cowart, 1976, have made a compilation of published isotopic analyses of undergroundwater. Especially at low concentration (< 0.1 µg/l uranium) the activity ratio varies between 1-12 whereas in the concentration range of 0.1-10 µg/l they cluster in the interval 0.8-2.

The uranium isotopes have been used as natural tracers in ground water systems and the relative volume proportions of mixing water sources have been calculated (cf Figs. 8 and 9, Osmond and Cowart, 1976). They can also be used to classify different underground artesian water systems. These two disequilibria applications are important tools in the selection of sites for a deep-seated repository.

However, mixing of dissimilar groundwaters may cause dissolution or precipitation of minerals (Runnels, 1969).

5.1 Solubilities of uranium in groundwater under reducing conditions

The solubility of uranium in deep natural groundwater, under reducing conditions is low. How low the uranium content will be under different conditions is certainly difficult to estimate.

In nature we are dealing with multicomponent systems, while in the laboratory the minimum number of intensive variables (solubility itself is an intensive variable) required by the Gibb's phase rule for invariancy are either fixed, measured or controlled. Such experiments may sound relatively simple but usually it isn't.

Goodwin et al, 1981, have calculated uranium solubilities in two synthetic groundwaters (granite- and brine-water) for a range of conditions, which have been chosen to represent natural conditions for a Canadian fuel disposal vault. Their calculations showed uranium solubilities in the order of 10^{-10} M (20 ng/l) during equilibrium conditions. Similar concentration levels have also been calculated by Allard, 1983, for reducing deep groundwaters with low carbonate concentrations. However, one must always remember that thermodynamic models depend on the quality of the experimentally determined data used.

We know that water-rock interactions are complex and furthermore a nuclear waste vault introduces still more components and species into the system. Also the radiolysis of the water may have strong influence on the chemical equilibria in the zone of elevated temperature.

In the natural fission reactor Oklo, Gabon, ore-body W Africa, the elements Zr, Nb, Tc, Te, the lanthanide series, U, and Pu, have only been redistributed to some extent without major migration. No separation occurred between uranium and plutonium (Cowan, 1977).

6 CONCLUSIONS

The solubility of uranium in deep reducing groundwater is very small. ^{235}U, which is a parent to ^{231}Pa, only constitutes a small part of the total uranium content in the waste.

The pronounced hydrolytic tendency of protactinium and its great sorption and coprecipitation capacity ought to prevent or at least appreciably delay its transport from a back-filled nuclear waste vault to the uppermost surface of the earth. It also shows a tendency to form colloids or particulates which may be strongly fixed on a rock surface. In adsorption and desorption processes kinetics must play an important role. Our knowledge in this field is quite limited.

Under the physico-chemical conditions in the sea, protactinium is rapidly scavenged from the water column by particulates. It accumulates in the sediments which have low biogenous content.

Table 1. Protactinium 231 and thorium 230 in sea water (after Moore and Sackett, 1964)

Sample	Location	Pa-231 dpm/1000 l	Location	Th-230 dpm/1000 l
Blank		0.020±0.008		0.043±0.011
N. Atlantic Surface	14°20'N 60°54'W	0.13±0.13	13°31'N 60°39'W	0.44±0.11
N. Atlantic 4500 m	-	-	32°15'N 74°47.5'W	1.6±0.2
Caribbean Surface	13°31'N 60°39'W		14°14'N 64°40'W	0.43±0.1
Caribbean 800 m	17°36'N 65°16'W	0.22±0.10	the same	0.31±0.12
Caribbean 800 m	13°21'N 64°19.5'W	0.24±0.10	-	-

Table 2. Correlation coefficients (r) for different components in the sediments (after Kuznetsov et al, 1966)

	r	
Component	Pa-231	Th-230
Th-230	0.95	-
Fe	0.80	0.93
Mn	0.48	0.48
CaCO	-0.58	-0.53
SiO_2 amf	-0.36	-0.44
Fraction >0.1 mm	-0.08	-0.05
	-0.44	-0.11
Fraction <0.001 mm	0.62	0.53
Total sediment surface cm^2 Depth 2 cm	0.69	0.60

Table 3. Contents of uranium and thorium, isotopic ratios and variation of Th-230 with U-238 and U-234 parent isotopes in soil profiles (after Rosholt et al, 1966).

Sample	Horizon	U (ppm)	Th (ppm)	$\frac{U}{Th}$	$\frac{^{234}U}{^{238}U}$ 1)	$\frac{^{238}U}{^{232}Th}$ 3)	$\frac{^{230}Th}{^{232}Th}$ 3)	$\frac{^{230}Th}{^{238}U}$ 2)	$\frac{^{230}Th}{^{234}U}$
					(radioactive equivalent ratio)				
FF-1	A	3.17	9.30	0.341	0.947	0.338	0.437	1.29	1.36
FF-2	B	3.22	10.2	.316	.922	.313	.441	1.41	1.53
FF-3	B	3.27	10.6	.318	.908	.306	.434	1.42	1.56
FF-4	B	3.07	10.4	.295	.891	.293	.418	1.43	1.60
FF-5	C	3.03	10.5	.289	.928	.286	.419	1.46	1.58
FF-6	C	2.57	8.13	.316	.926	.314	.403	1.28	1.38
WF-1	A	2.33	8.46	.275	.937	.273	.364	1.33	1.42
WF-2	B	2.66	9.91	.268	.903	.266	.381	1.43	1.58
WF-3	B	2.49	9.61	.259	.909	.257	.377	1.47	1.61
WF-4	B	2.37	9.37	.253	.902	.251	.380	1.51	1.68
WF-5	C	2.21	8.26	.267	.906	.266	.377	1.42	1.56
WF-6	C	2.20	8.06	.275	.915	.273	.381	1.40	1.52
WF-7	C	2.04	7.58	.269	.909	.267	.361	1.35	1.48
K-1	A	3.24	8.30	.390	.993	.388	.438	1.13	1.14
K-2	B	3.45	8.61	.401	.977	.398	.463	1.16	1.19
K-3	B	2.18	8.13	.268	.869	.266	.317	1.19	1.37
K-4	C	1.92	7.94	.242	.867	.240	.405	1.69	1.95
K-5	C	2.06	7.59	.271	.880	.269	.442	1.64	1.86
SIA	A	3.12	9.70	.322	1.06	.319	.357	1.12	1.05
SIB	B	2.87	10.0	.287	1.00	.285	.328	1.15	1.15
S2A	A	3.46	9.10	.380	0.980	.377	.402	1.07	1.09
S2B	B	3.18	8.34	.381	.946	.379	.418	1.10	1.17

For soil type, age and location cf p 9.

Three fundamental measurements have been made, which are combined for calculating different ratios

Method used: isotope dilution U/Th ratio (weight concentration ratio of unity).

Method used: mass spectrometry

$$\frac{^{235}U/^{234}U(\text{reference})}{^{235}U/^{234}U\ (\text{sample})} = \frac{^{234}U}{^{238}U}$$

Dimension: radioactive equivalent units. The numerical value is the same as for "normal" activity ratio.

Method used: alpha-spectrometry. The calculation of the activity-ratio reference for thorium isotopic composition is:

$$^{230}Th/^{232}Th\ (\text{ref.}) = \frac{^{230}Th/^{232}Th(\text{Th-isotope ref.}) - ^{230}Th/^{232}Th(^{232}Th\ \text{subreferences})}{\mu g\,^{238}U/\mu g\,^{232}Th(\text{in subreferences})}.$$

$$\frac{^{230}Th/^{232}Th(\text{sample})}{^{230}Th/^{232}Th(\text{ref.})} = \text{equivalent}\ \frac{\text{ppm}\ ^{230}Th}{\text{ppm}\ ^{232}Th\ (\text{sample})}$$

The last term can also be defined as the amount of ^{238}U in ppm that would be required to support the measured amount of ^{230}Th for each ppm in the sample at secular equilibrium.

---

1) cf above uranium

2) $$^{230}Th/^{238}U = \frac{\left[^{230}Th/^{232}Th(\text{sample})\right]\left[^{230}Th/^{232}Th(\text{reference})\right]}{^{238}U/^{232}Th}$$

3) If the value for $^{230}Th/^{232}Th$ is larger than $^{238}U/^{232}Th$ there is an excess of ^{230}Th. Further details about standards and measurements, cf Rosholt et al, 1966, pp 988-994.

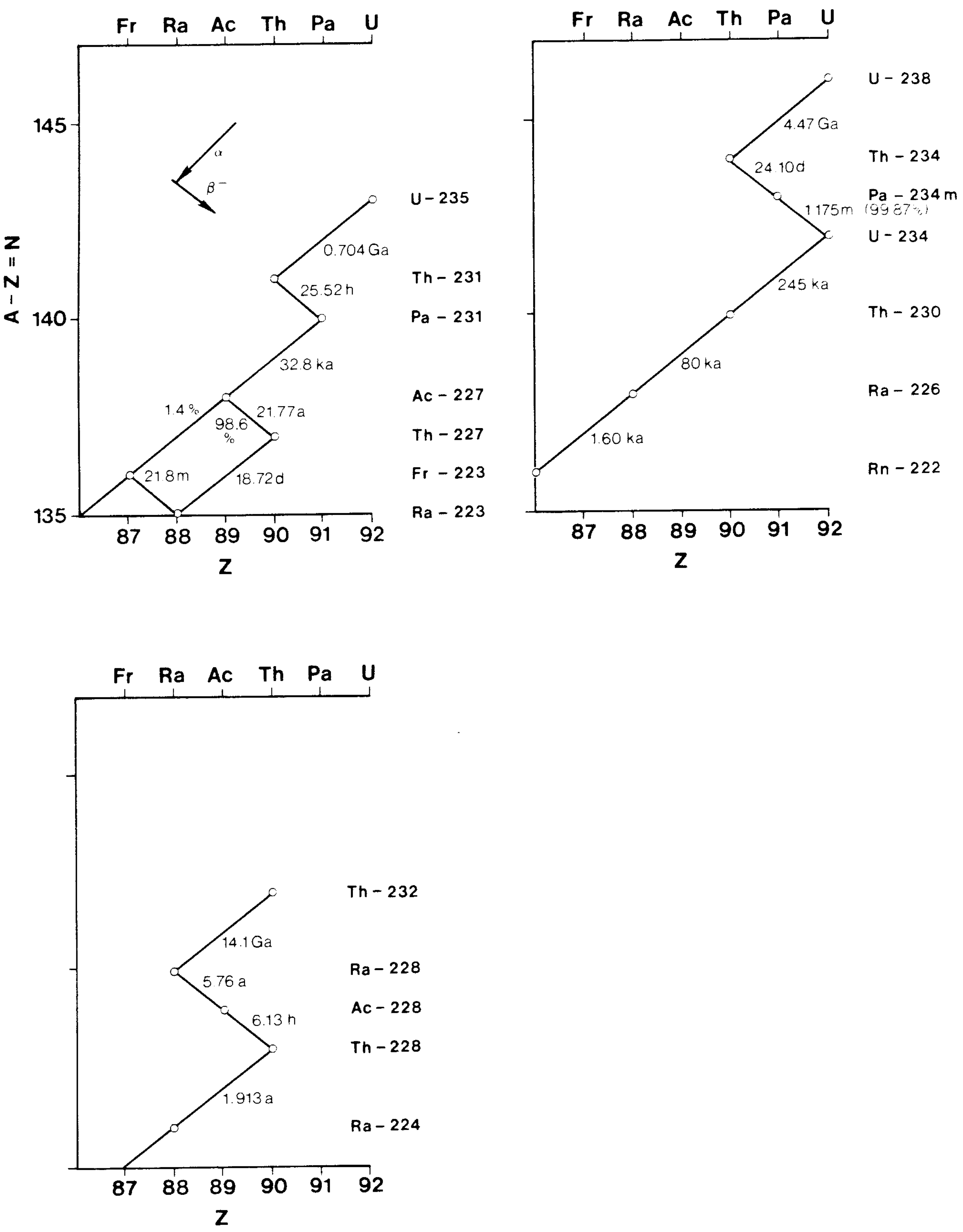

Figure 1.

The radionuclides in the upper parts of the naturally-occurring series: actinium (4n + 3), uranium-radium (4n + 2) and thorium (4n). Half-lives are shown in years (a), days (d), hours (h) and minutes (m).

REFERENCES

Allard, B., 1983, Actinide solution equilibria and solubilities in geologic systems.
KBS TR 83-35, Svensk Kärnbränsleförsörjning AB, Stockholm, 1983.

Aller, R.C., Cochran, J.K., 1976, ^{234}Th/^{238}U disequilibrium in near-shore sediment: Particle reworking and diagenetic time scales. Earth Planet. Sci. Lett. **29**, 37.

Bhat, S.G., Krishnaswamy, D. Lal, Rama, Moore, W.S., 1969, ^{234}Th/^{238}U ratios in the ocean. Earth Planet. Sci. Lett. **5**, 483.

Bowen, H.J.M., 1979, Environmental Chemistry of the elements. Academic Press.

Broecker, W.S., Kaufman, A., Trier, R.M., 1973, The residence time of thorium in surface sea water and its implications regarding the rate of reactive pollutants. Earth Planet. Sci. Lett. **20**, 35.

Covan, G.A., 1977, Migration paths for Oclo reactor products and applications to the problem of geological storage of nuclear wastes. Proc. IAEA Symp. IAEA-TC **119/26**, 693.

Fleisher, R.F., 1980, Isotopic disequilibrium of uranium: Alpha-recoil damage and preferential solution effects. Science **207**, p. 979.

Garrels, R.M., MacKenzie, F.T., 1971, Evolution of sedimentary rocks. W.W. Norton, New York.

Goodwin, B., 1982, Calculated uranium solubilities in groundwater: implications for nuclear fuel waste disposal. Can. J. Chem. **60**, 1759.

Imai, T., Sakanoue, M., 1973, Content of plutonium, thorium and protactinium in sea water and recent corals in the North Pacific. J. Ocean. Soc. Japan **29**, 76.

Kaufman, A., 1969, The ^{232}Th concentration of surface ocean water. Geochim. Cosmochim. Acta **33**, 717.

Koczy, F.F., Picciotto, E., Poulart, G., Wilgain, S., 1957, Determination of thorium isotopes in sea water. Geochim. Cosmochim. Acta **11**. 103.

Ku, T-L., Broecker, W.S., 1967, Uranium, thorium and protactinium in a manganese nodule. Earth Planet. Sci. Lett. **2**, 317.

Kuznetsov, Y.V., Elizarova, A.N., Frenklikh, M.S., 1966, Investigation of the mechanism of precipitation of ^{231}Pa and ^{230}Th from ocean water. (In Russian) Radiokliniya **8(4)**, 459.

Langmuir, D., Herman, J.S., 1980, The mobility of thorium in natural waters at low temperatures. Geochim. Cosmochim. Acta **44**, p. 1753.

Megumi, K., Mamuro, T., 1977, Concentration of uranium series nuclides in soil particles in relation to their size. J. Geophys. Res. 2, 353.

Moore, W.S., Sackett, W.M., 1964, Uranium and thorium series inequilibrium in sea water. J. Geophys. Res. **69**, 5401.

Osmond, J.K., Cowart, J.B., 1976, The theory and uses of natural uranium isotopic variations in hydrology. Atom. Energy Rev. **144**, 621.

Osmond, J.K., 1979, Accumulation models of ^{230}Th and ^{231}Pa in deep sea sediments. Earth Sci. Rev. **15**, 95.

Rosholt, J.N., Doe, B.R., Tatsumoto, M., 1966, Evolution of the isotopic composition of uranium and thorium in soil particles. Geol. Soc. Am. Bull. **77**, 987.

Runnells, D.D., 1969, Diagenesis, chemical sediments and the mixing of natural waters. J. Sed. Petr. **39**, 1188.

Sackett, W.M., 1960, Protactinium-231 content of ocean water and sediments. Science **132**, 1761.

Titaeva, N.A., Alexakhin, R.M., Taskaev, A.J., Maslov, V.J., 1978, Migration of heavy natural radionuclides in a humid climatic zone. Natural Radiation Environment III, 1, 724. Proc. Sym. Houston, Texas, April 23-28, 1978.
Publ. 1980 U.S. Dept. of Energy.

List of KBS's Technical Reports

1977–78

TR 121

KBS Technical Reports 1 – 120.

Summaries. Stockholm, May 1979.

1979

TR 79–28

The KBS Annual Report 1979.

KBS Technical Reports 79-01 – 79-27.
Summaries. Stockholm, March 1980.

1980

TR 80–26

The KBS Annual Report 1980.

KBS Technical Reports 80-01 – 80-25.
Summaries. Stockholm, March 1981.

1981

TR 81–17

The KBS Annual Report 1981.

KBS Technical Reports 81-01 – 81-16.
Summaries. Stockholm, April 1982.

TR 82–28

The KBS Annual Report 1982.

KBS Technical Reports 82-01 – 82-27.

1983

TR 83–01

Radionuclide transport in a single fissure A laboratory study

Trygve E Eriksen
Department of Nuclear Chemistry
The Royal Institute of Technology
Stockholm, Sweden 1983-01-19

TR 83–02

The possible effects of alfa and beta radiolysis on the matrix dissolution of spent nuclear fuel

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

TR 83–03

Smectite alteration
Proceedings of a colloquium at State University of New York at Buffalo, May 26–27, 1982

Compiled by Duwayne M Anderson
State University of New York at Buffalo
February 15, 1983

TR 83–04

Stability of bentonite gels in crystalline rock – Physical aspects

Roland Pusch
Division Soil Mechanics, Univeristy of Luleå
Luleå, Sweden, 1983-02-20

TR 83–05

Studies in pitting corrosion on archeological bronzes – Copper

Åke Bresle
Jozef Saers
Birgit Arrhenius
Archaeological Research Laboratory
University of Stockholm
Stockholm, Sweden 1983-01-02

TR 83–06

Investigation of the stress corrosion cracking of pure copper

L A Benjamin
D Hardie
R N Parkins
University of Newcastle upon Tyne
Department of Metallurgy and engineering Materials
Newcastle upon Tyne, Great Britain, April 1983

TR 83–07

Sorption of radionuclides on geologic media – A literature survey.
I: Fission Products

K Andersson
B Allard
Department of Nuclear Chemistry
Chalmers University of Technology
Göteborg, Sweden 1983-01-31

TR 83–08

Formation and properties of actinide colloids

U Olofsson
B Allard
M Bengtsson
B Torstenfelt
K Andersson
Department of Nuclear Chemistry
Chalmers University of Technology
Göteborg, Sweden 1983-01-30

TR 83–09

Complexes of actinides with naturally occurring organic substances – Literature survey

U Olofsson
B Allard
Department of Nucluear Chemistry
Chalmers University of Technology
Göteborg, Sweden 1983-02-15

TR 83–10
Radilysis in nature: Evidence from the Oklo natural reactors
David B Curtis
Alexander J Gancarz
New Mexico, USA February 1983

TR 83–11
Description of recipient areas related to final storage of unreprocessed spent nuclear fuel
Björn Sundblad
Ulla Bergström
Studsvik Energiteknik AB
Nyköping, Sweden 1983-02-07

TR 83–12
Calculation of activity content and related propeties in PWR and BWR fuel using ORIGEN 2
Ove Edlund
Studsvik Energiteknik AB
Nyköping, Sweden 1983-03-07

TR 83–13
Sorption and diffusion studies of Cs and I in concrete
K Andersson
B Torstenfelt
B Allard
Department of Nuclear Chemistry
Chalmers University of Technology
Göteborg, Sweden 1983-01-15

TR 83–14
The complexation of Eu (III) by fulvic acid
J A Marinsky
State University of New York at Buffalo
Buffalo, NY 1983-03-31

TR 83–15
Diffusion measurements in crystalline rocks
Kristina Skagius
Ivars Neretnieks
Royal Institute of Technology
Stockholm, Sweden 1983-03-11

TR 83–16
Stability of deep-sited smectite minerals in crystalline rock – chemical aspects
Roland Pusch
Division of Soil Mechanics, University of Luleå
Luleå 1983-03-30

TR 83–17
Analysis of groundwater from deep boreholes in Gideå
Sif Laurent
Swedish Environmental Research Institute
Stockholm, Sweden 1983-03-09

TR 83–18
Migration experiments in Studsvik
O Landström
Studsvik Energiteknik AB
C-E Klockars
O Persson
E-L Tullborg
S Å Larson
Swedish Geological
K Andersson
B Allard
B Torstenfelt
Chalmers University of Technology
1983-01-31

TR 83–19
Analysis of groundwater from deep boreholes in Fjällveden
Sif Laurent
Swedish Environmental Research Institute
Stockholm, Sweden 1983-03-29

TR 83–20
Encapsulation and handling of spent nuclear fuel for final disposal
1 Welded copper canisters
2 Pressed copper canisters (HIPOW)
3 BWR Channels in Concrete
B Lönnerbeg, ASEA-ATOM
H Larker, ASEA
L Ageskog, VBB
May 1983

TR 83–21
An analysis of the conditions of gas migration from a low-level radioactive waste repository
C Braester
Israel Institute of Technology, Haifa, Israel
R Thunvik
Royal Institute of Technology
Stockholm, Sweden November 1982

TR 83–22
Calculated temperature field in and around a repository for spent nuclear fuel
Taivo Tarandi, VBB
Stockholm, Sweden April 1983

TR 83–23
Preparation of titanates and zeolites and their uses in radioactive waste management, particularly in the treatment of spent resins
Å Hultgren, editor
C Airola
Studsvik Energiteknik AB
S Forberg, Royal Institute of Technology
L Fälth, University of Lund
May 1983

TR 83–24
Corrosion resistance of a copper canister for spent nuclear fuel
The Swedish Corrosion Research Institute
and its reference group
Stockholm, Sweden April 1983

TR 83–25
Feasibility study of electron beam welding of spent nuclear fuel canisters
A Sanderson, T F Szluha, J L Turner, R H Leggatt
The Welding Institute Cambridge
The United Kingdom April 1983

TR 83–26
The KBS UO_2 leaching program
Summary Report 1983-02-01
Ronald Forsyth, Studsvik Energiteknik AB
Nyköping, Sweden February 1983

TR 83–27
Radiation effects on the chemical environment in a radioactive waste repository
Trygve Eriksen
Royal Institute of Technology, Stockholm
Arvid Jacobsson
Univerisity of Luleå
Luleå, Sweden 1983-07-01

TR 83–28
An analysis of selected parameters for the BIOPATH-program
U Bergström
A-B Wilkens
Studsvik Energiteknik AB
Nyköping, Sweden 1983-06-08

TR 83–29
On the environmental impact of a repository for spent nuclear fuel
Otto Brotzen
Stockholm, Sweden april 1983

TR 83–30
Encapsulation of spent nucler fuel – Safety Analysis
ES-konsult AB
Stockholm, Sweden April 1983

TR 83–31
Final disposal of spent nuclear fuel – Standard programme for site investigations
Compiled by
Ulf Thoregren
Swedish Geological
April 1983

TR 83–32
Feasibility study of detection of defects in thick welded copper
Tekniska Röntgencentralen AB
Stockholm, Sweden April 1983

TR 83–33
The interaction of bentonite and glass with aqueous media
M Mosslehi
A Lambrosa
J A Marinsky
State University of New York
Buffalo, NY, USA April 1983

TR 83–34
Radionuclide diffusion and mobilities in compacted bentonite
B Torstenfelt
B Allard
K Andersson
H Kipatsi
L Eliasson
U Olofsson
H Persson
Chalmers University of Technology
Göteborg, Sweden 1983-12-15

TR 83–35
Actinide solution equilibria and solubilities in geologic systems
B Allard
Chalmers University of Technology
Göteborg, Sweden 1983-04-10

TR 83–36
Iron content and reducing capacity of granites and bentonite
B Torstenfelt
B Allard
W Johansson
T Ittner
Chalmers University of Technology
Göteborg, Sweden April 1983

TR 83–37
Surface migration in sorption processes
A Rasmuson
I Neretnieks
Royal Institute of Technology
Stockholm, Sweden March 1983

TR 83–38
Evaluation of some tracer tests in the granitic rock at Finnsjön
L Moreno
I Neretnieks
Royal Institute of Technology, Stockholm
C-E Klockars
Swedish Geological
Uppsala April 1983

TR 83–39
Diffusion in the matrix of granitic rock Field test in the Stripa mine. Part 2
L Birgersson
I Neretnieks
Royal Institute of Technology
Stockholm, Sweden March 1983

TR 83–40
Redox conditions in groundwaters from Svartboberget, Gideå, Fjällveden and Kamlunge
P Wikberg
I Grenthe
K Axelsen
Royal Institute of Technology
Stockholm, Sweden 1983-05-10

TR 83–41
Analysis of groundwater from deep boreholes in Svartboberget
Sif Laurent
Swedish Environmental Research Institute
Stockholm, Sweden 1983-06-10

TR 83–42
Final disposal of high-levels waste and spent nuclear fuel – foreign activities
R Gelin
Studsvik Energiteknik AB
Nyköping, Sweden May 1983

TR 83–43
Final disposal of spent nuclear fuel – geological, hydrogeological and geophysical methods for site characterization
K Ahlbom
L Carlsson
O Olsson
Swedish Geological
Sweden May 1983

TR 83–44
Final disposal of spent nuclear fuel – equipment for site characterization
K Almén, K Hansson, B-E Johansson, G Nilsson
Swedish Geological
O Andersson, IPA-Konsult
P Wikberg, Royal Institute of Technology
H Åhagen, SKBF/KBS
May 1983

TR 83–45
Model calculations of the groundwater flow at Finnsjön, Fjällveden, Gideå and Kamlunge
L Carlsson
A Winberg
Swedish Geological, Göteborg
B Grundfelt
Kemakta Consultant Company,
Stockholm May 1983

TR 83–46
Use of clays as buffers in radioactive repositories
Roland Pusch
University of Luleå
Luleå May 25 1983

TR 83–47
Stress/strain/time properties of highly compacted bentonite
Roland Pusch
University of Luleå
Luleå May 1983

TR 83–48
Model calculations of the migration of radio-nuclides from a respository for spent nuclear fuel
A Bengtsson
Kemakta Consultant Company, Stockholm
M Magnusson
I Neretnieks
A Rasmuson
Royal Institute of Technology, Stockholm
May 1983

TR 83–49
Dose and dose commitment calculations from groundwaterborne radioactive elements released from a repository for spent nuclear fuel
U Bergström
Studsvik Energiteknik AB
Nyköping, Sweden May 1983

TR 83–50
Calculation of fluxes through a repository caused by a local well
R Thunvik
Royal Institute of Technology
Stockholm, Sweden May 1983

TR 83–51
GWHRT – A finite element solution to the coupled ground water flow and heat transport problem in three dimensions
B Grundfelt
Kemakta Consultant Company
Stockholm, Sweden May 1983

TR 83–52
Evaluation of the geological, geophysical and hydrogeological conditions at Fjällveden
K Ahlbom
L Carlsson
L-E Carlsten
O Duran
N-Å Larsson
O Olsson
Swedish Geological
May 1983

TR 83–53
Evaluation of the geological, geophysical and hydrogeological conditions at Gideå
K Ahlbom
B Albino
L Carlsson
G Nilsson
O Olsson
L Stenberg
H Timje
Swedish Geological
May 1983

TR 83–54
Evaluation of the geological, geophysical and hydrogeological conditions at Kamlunge
K Ahlbom
B Albino
L Carlsson
J Danielsson
G Nilsson
O Olsson
S Sehlstedt
V Stejskal
L Stenberg
Swedish Geological
May 1983

TR 83–55
Evaluation of the geological, geophysical and hydrogeological conditions at Svartboberget
K Ahlbom
L Carlsson
B Gentzschein
A Jämtlid
O Olsson
S Tirén
Swedish Geological
May 1983

TR 83–56
I: Evaluation of the hydrogeological conditions at Finnsjön
L Carlsson
G Gidlund
II: Supplementary geophysical investigations of the Stärnö peninsula
B Hesselström
Swedish Geological
May 1983

TR 83–57
Neotectonics in northern Sweden – geophysical investigations
H Henkel
K Hult
L Eriksson
Geological Survey of Sweden
L Johansson
Swedish Geological
May 1983

TR 83–58
Neotectonics in northern Sweden – geological investigations
R Lagerbäck
F Witschard
Geological Survey of Sweden
May 1983

TR 83–59
Chemistry of deep groundwaters from granitic bedrock
B Allard
Chalmers University of Technology
S Å Larson
E-L Tullborg
Swedish Geological
P Wikberg
Royal Institute of Technology
May 1983

TR 83–60
On the solubility of technetium in geochemical systems
B Allard
B Torstenfelt
Chalmers University of Technology
Göteborg, Sweden 1983-05-05

TR 83–61
Sorption behaviour of well-defined oxidation states
B Allard
U Olofsson
B Torstenfelt
H Kipatsi
Chalmers University of Technology
Göteborg, Sweden 1983-05-15

TR 83–62
The distribution coefficient concept and aspects on experimental distribution studies
B Allard
K Andersson
B Torstenfelt
Chalmers University of Technology
Göteborg, Sweden May 1983

TR 83–63
Sorption of radionuclides in geologic systems
K Andersson
B Torstenfelt
B Allard
Chalmers University of Technology
Göteborg, Sweden 1983-06-15

TR 83–64
Ion exchange capacities and surface areas of some major components and common fracture filling materials of igneous rocks
B Allard
M Karlsson
Chalmers University of Technology
E-L Tullborg
S Å Larson
Swedish Geological
Göteborg, Sweden May 1983

TR 83–65
Sorption of actinides on uranium dioxide and zirconium dioxide in connection with leaching of uranium dioxide fuel
B Allard
N Berner
K Andersson
U Olofsson
B Torstenfelt
Chalmars University of Technology
R Forsyth
Studsvik Energiteknik AB
May 1983

TR 83–66
The movement of radionuclides past a redox front
I Neretnieks
B Åslund
Royal Institute of Technology
Stockholm, Sweden 1983-04-22

TR 83–67
Some notes in connection with the studies of final disposal of spent fuel. Part 2
I Neretnieks
Royal Institute of Technology
Stockholm, Sweden May 1983

TR 83–68
Two dimensional movements of a redox front downstream from a repository for nuclear waste
I Neretnieks
B Åslund
Royal Institute of Technology
Stockholm, Sweden 1983-06-03

TR 83–69
An approach to modelling radionuclide migration in a medium with strongly varying velocity and block sizes along the flow path
I Neretnieks
A Rasmuson
Royal Institute of Technology
Stockholm, Sweden May 1983

TR 83–70
Analysis of groundwater from deep boreholes in Kamlunge
S Laurent
Swedish Environmental Research Institute
Stockholm, Sweden May 1983

TR 83–71
Gas migration through bentonite clay
Roland Pusch
Thomas Forsberg
University of Luleå
Luleå, Sweden May 31, 1983

TR 83–72
On the thermal conductivity and thermal diffusivity of highly compacted bentonite
Sven Knutsson
University of Luleå
Luleå, Sweden October 1983

TR 83–73
Uranium, thorium and radium in soil and crops – Calculations of transfer factors
Sverker Evans
Studsvik Energiteknik AB
Åke Eriksson
Swedish University of Agricultural Sciences
Sweden June 1983

TR 83–74
Fissure fillings from Gideå, central Sweden
Eva-Lena Tullborg
Swedish Geological
Sven Åke Larson
Geological Survey of Sweden
Gothenburg, Sweden August 1983

TR 83–75
The geochemical behavior of protactinium 231 and its chosen geochemical analogue thorium in the bisphere
Marianne Gillberg-Wickman
National Defence Research Institute
Umeå, Sweden March 1983