

***Natural activity concentrations  
and fluxes as indicators  
for the safety assessment of  
radioactive waste disposal***

*Results of a coordinated research project*



**IAEA**

International Atomic Energy Agency

October 2005

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## FOREWORD

Plans for disposing of solid radioactive wastes have raised a number of unique problems, mainly due to the very long timescales which have to be considered. While there is general agreement on disposal concepts and on the approach to establishing that disposal facilities are safe, discussion continues on a number of related issues. One of these issues concerns appropriate measures of disposal system performance in the very far future that could be complementary to the standard measures of radiological dose and risk.

To assist in promoting discussion amongst international experts and in developing consensus, the IAEA established a Working Group under the International Radioactive Waste Management Advisory Committee (INWAC). The group started its work in 1991 as the Working Group on Principles and Criteria for Radioactive Waste Disposal. With the reorganization in 1995 of the IAEA Safety Standards committees, a closer linkage has been established between the Working Group and its parent committee, now titled the Waste Safety Standards Committee (WASSC).

The Working Group first examined the use of safety indicators complementary to dose and risk, and published their deliberations in a report entitled *Safety Indicators in Different Time Frames for the Safety Assessment of Underground Radioactive Waste Repositories* (IAEA-TECDOC-767, 1994). The development of complementary safety indicators to dose and risk is an issue that has been discussed repeatedly through the years, particularly the application of radionuclide concentrations and fluxes, but has not gained widespread international recognition largely because of perceived problems with the lack of assessment criteria for these indicators.

To address the issue, an IAEA Coordinated Research Project (CRP) entitled *Natural Safety Indicators (Concentrations and Fluxes)* was begun with the stated objective of contributing, through the development of international consensus, to the assessment of the long term safety of radioactive waste disposal by means of additional safety indicators based on the observation of natural systems. Nine countries participated in the CRP: Argentina, Brazil, China, Cuba, the Czech Republic, Finland, Japan, Sweden and the United Kingdom. The project began in 1999 and was completed in 2003. This report presents the findings of that project. The participation of countries from around the world has meant that the distribution of naturally occurring radionuclides (average concentrations and ranges) on both local, regional and global scales is now better understood. This has enabled the project to define ways in which these abundances may be used to set criteria against which the potential releases from a repository (calculated in a safety assessment) may be evaluated.

In parallel to the CRP, the Working Group published a further report on the subject entitled *Safety Indicators for the Safety Assessment of Radioactive Waste Disposal* (IAEA-TECDOC-1372, 2003) that discusses in some detail the various suggested ways that radionuclide concentrations and fluxes could be applied in a safety case in a manner complementary to dose and risk.

This report, along with others from the Working Group on Principles and Criteria for Radioactive Waste Disposal, contains the developing views of experts within the international community and should be of use to those engaged in producing national and international standards and guidance in this area. They should not, however, be seen as representing a Member State consensus on the subjects being discussed.

The IAEA wishes to thank all those who contributed to this report. The work of W. Miller, of the United Kingdom, who chaired the meetings and was responsible for drafting this report, is acknowledged. The IAEA officer responsible for this report was K. Hioki of the Division of Radiation, Transport and Waste Safety.

#### *EDITORIAL NOTE*

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# 1. INTRODUCTION

## 1.1. BACKGROUND

A primary requirement of disposal systems for radioactive wastes is that they should pose no significant hazard to human health in the short and long term, and should cause no unacceptable harm to the environment. Safety indicators are the characteristics or consequences of a disposal system by which potential hazards or harm can be measured, and may be evaluated in the course of making a safety case for a disposal system. The role and utility of different types of safety indicators over different timescales have been discussed in previous IAEA publications [1, 2].

The most widely used indicators in the context of the long term safety of disposal systems are those of radiological dose and risk. The dose to human beings that are assumed to inhabit a specific location and to be exposed, in some way, to radionuclides migrating from the disposal system is typically evaluated in the course of making a safety case. The dose can be transformed to a corresponding risk level using a suitable dose/risk conversion factor [3], and taking account of the likelihood of the scenario envisaged for exposure. In general, a safety case needs to provide reasonable assurance that the eventual consequences of any migration of radionuclides to the accessible environment are in compliance with the dose and risk standards that are acceptable today [3, 4].

In view of such migration only taking place in the very far future, the use of safety indicators, other than dose and risk, to assess and confirm the safety of disposal systems for radioactive waste, has been suggested repeatedly through the years. The IAEA publications [1, 2] pointed to problems associated with the use of dose and risk as measures of safety, particularly in the far future. The difficulties are mainly related to the uncertainty surrounding their evaluation, to providing direct evidence of assurance and to public communication. It was concluded that a safety case can most effectively be made by the combined use of several safety indicators, including not only dose and risk, but also environmental concentrations and fluxes, and the timescales of relevant processes, while recognising that dose and risk remain the most fundamental indicators of safety.

Since the first of these IAEA publications on the subject [1], various studies into the application of natural safety indicators have been undertaken [e.g. 5–7]. These studies have shown that, in principle, safety assessment predictions of repository releases may be compared with the concentrations and fluxes of naturally occurring chemical species in the environment so as to provide context and an understanding of comparative hazards.

In Finland this idea has been put into practice and the Radiation and Nuclear Safety Authority (STUK) has recently issued a guide for the long term safety of spent fuel disposal which includes constraints based on activity releases to the environment in the long term such that, at their maximum, the radiation impacts arising from disposal can be comparable to those arising from natural radioactive substances [8]. This Finnish guidance builds on a previous document produced by licensing authorities in the Nordic countries where the idea of complementary safety indicators was promoted [9].

In Japan, the recent safety assessment for a generic high level waste (HLW) repository [10] included a section which compared the calculated repository release activity concentrations with measured concentrations of naturally occurring radionuclides in river and groundwaters. This represents the first significant application of natural safety indicators to a major published safety case.

Clearly there is a growing interest in the possibility of using safety indicators other than dose and risk (particularly of environmental concentrations and fluxes). The approach has never, however, been the subject of a comprehensive international evaluation and no broad consensus has yet been developed as to its application. Some of the reasons why consensus has not developed relate to:

- the lack of appropriate data on measured concentrations and fluxes of naturally occurring radionuclides;
- the lack of internationally agreed forms of comparisons between repository and natural radionuclide abundances;
- the absence of internationally agreed hazard indices against which to evaluate the comparisons.

To address these and related issues, the IAEA launched an internationally supported Coordinated Research Project (CRP) entitled Natural Safety Indicators (Concentrations and Fluxes). This report presents the findings of that project.

## 1.2. THE COORDINATED RESEARCH PROJECT

### 1.2.1. Scope and objectives

The stated objective of the CRP was to contribute, through the development of international consensus, to the assessment of the long term safety of radioactive waste disposal by means of additional safety indicators based on the observation of natural systems. The main focus of the programme was on long term safety aspects, including relevant safety criteria and assessment techniques and methodologies.

The project began in 1999 and was completed in 2003.

The participating countries, lead organizations and principal researchers involved in the CRP are given below:

- Argentina: R. Ferreyra, Comisión Nacional de Energía Atómica;
- Brazil: F. Lemos, Comissão Nacional de Energia Nuclear;
- China: Sun Qinghong, China Institute for Radiation Protection;
- Cuba: J. Peralta Vital, Centro de Protección e Higiene;
- Czech Republic: A. Laciok, Nuclear Research Institute;
- Finland: K.-H. Hellmuth, Radiation & Nuclear Safety Authority;

- Japan: K. Ishiguro, Japan Nuclear Cycle Development Institute;
- Sweden: B. Thunholm, A. Lindén, Geological Survey of Sweden;
- UK: S. Norris, P. Degnan, UK Nirex Ltd.

IAEA staff responsible for project management were F. Gera and K. Hioki. CRP meetings were chaired by B. Miller (Enviros Consulting Ltd, UK).

The specific research objectives of the CRP, set at the outset of the project, were:

- to develop a database of measured concentrations of target substances (elements, radioactive isotopes and particular chemical compounds) in different solid and aqueous materials, from different geographical areas, showing the spatial variability in these concentrations from the regional to the site scale;
- to develop a database of estimated fluxes of the target substances between different subsurface and surface compartments, showing the spatial and numerical variations in natural fluxes from the regional to the site scale;
- to assess the measured concentrations and estimated fluxes so as to construct ‘measures’ or ‘ranges’ that can be independently assessed to be safe and against which the outcomes of performance assessments can be evaluated;
- to publish the activities and findings of the CRP.

Of these objectives 1 and 2 were considered to be the most significant because the lack of appropriate data on measured concentrations and fluxes of naturally occurring radionuclides has been seen a key factor limiting the application of complementary safety indicators.

On this basis, the focus of the CRP was directed to characterization of the natural geochemical environment so that it may be used as a comparator against which predicted repository releases may be compared.

In addition to these specific research objectives, the participants agreed at an early research coordination meeting (RCM) a further set of aims for the project:

- to focus on an agreed suite of elements (U, Th, K, Rb, Sn, Cs, Cu, Ra and Rn) so that elemental concentration and flux data from the different participants could be most readily compared;
- to identify and collate any relevant epidemiological data which can be correlated to human health impacts arising from naturally occurring elemental or radionuclide concentrations;
- to identify and collate any relevant safety assessment results, in the form of calculated repository derived contaminant releases, which may be compared with the naturally occurring elemental or radionuclide concentrations and fluxes; and
- to propose and evaluate potential hazard indices which may be appropriate for the evaluation of both repository derived contaminant releases and naturally occurring elemental or radiological concentrations.

These additional aims were intended to provide greater clarity and focus to the project by directing activities directly onto issues of most significance for understanding repository performance.

### **1.2.2. Approach**

As is common in CRPs, the IAEA provided overall direction to the research activities and individual participants undertook tasks within their own countries consistent with the stated project objectives.

Within the resource limitations for this CRP (in terms of money and time), few participants were able to undertake new campaigns of material sampling and analysis (e.g. of rocks, soils or waters). Most participants focussed their activities instead on collating information from existing sources such as information in published reports and papers or in databases held by Government departments and agencies (such as national geological surveys) or commercial organizations.

Early in the project it became clear that very large amounts of relevant data did exist. Many of these data were, however, widely dispersed amongst various private and national organizations, and much of it had never been digitized or catalogued. Much of the CRP effort was, thus, spent in the labour intensive processes of reviewing, assessing and cataloguing information that existed only in paper format. It also became evident that many relevant geochemical data had been acquired by other organizations for commercial reasons (e.g. uranium exploration) and, thus, could not all be made available to the project. Future projects with greater resources (or more detailed country specific projects) may be able to collect and collate even more relevant information that were available to this CRP.

## **1.3. REPORT STRUCTURE**

Section 2 of this report summarizes the main features of safety and performance indicators, focusses on environmental concentrations and fluxes, and considers briefly their role in a safety case. Section 3 provides an overview of the geochemical data collected as part of the CRP, and provides country-by-country summaries of the elemental and radionuclide concentration data derived in a variety of media, and the process rate data used to determine fluxes. Section 4 provides conclusions from the CRP, and a discussion of the utility of the data collected and considers possible ways that the findings of the CRP could be used to progress international consensus on the use of complementary safety indicators. A series of Appendices provide details of the work undertaken in each participating country, the data they acquired and the various applications made of those data in national safety assessments.

## **1.4. OTHER REPORTS OF THE WORKING GROUP OF THE IAEA**

The activities of the CRP, reported here, and the recent report from the Working Group [2] are complementary. The CRP focussed on the collation of data that characterized the natural radiation environment that could be compared to predictions of repository releases calculated in safety assessments. The Working Group focussed on ways that those comparisons could best be made within the structure of a repository safety case. The reader is thus recommended to read this report in conjunction with [2] since that report provides context for much of the work of the CRP.

## 2. PERFORMANCE AND SAFETY INDICATORS

### 2.1. LIMITATIONS OF DOSE AND RISK

Radiological dose and risk are the two standard measures of disposal system performance and are used ubiquitously in all safety cases. Estimates of dose and risk arising in the future from releases from a closed disposal facility are based on calculated values of concentrations of waste derived radionuclides in the accessible environment. In a safety case, these estimates are generally of a bounding nature. To calculate the environmental concentrations of waste derived substances, the fluxes between the disposal facility and the accessible environment also need to be estimated. For the additional conversion of these environmental values to dose or risk (for human beings and other organisms), assumptions have to be made about the evolution of the biosphere, including exposure pathways, the habits of relevant human population groups and the presence and behaviour of other living species.

The use of dose or risk as safety indicators thus has one clear disadvantage, associated with the uncertainty surrounding their estimation. Biosphere modelling is an integral part of dose and risk evaluation, and uncertainties associated with biosphere modelling increase considerably with the timescale under consideration [11], in particular with regard to human habits, but also with regard to the natural evolution of the surface environment and the changing nature and distribution of other organisms (e.g. in response to climate change). The engineered barriers of a disposal system and the geosphere are expected to be characterized, in general, by greater long term stability.

Other limitations of dose and risk arise because neither of these indicators provide much explicit information on the wider potential environmental impacts arising from repository releases and because the mathematical derivation and presentation of dose/risk calculations, and the underlying fundamental radiological concepts, are difficult to understand by non-technical audiences.

Given these limitations, the use of safety indicators, other than dose and risk, to assess and confirm the safety of radioactive waste repositories has been suggested repeatedly through the years. Previous work by the IAEA [1] concluded that a safety case can most effectively be made by the combined use of several safety indicators, while recognising that dose and risk remain the most fundamental indicators of safety. In more recent work by the IAEA's Working Group on Principles and Criteria [2], a more detailed assessment of proposed safety indicators complementary to dose and risk was made, including an evaluation of how they may be applied within a safety case.

### 2.2. TYPES OF INDICATORS

An *indicator* is taken to be any characteristic or consequence of a disposal system that has a bearing on the ability of the system to perform its safety functions. Indicators may be:

- directly measurable characteristics of the disposal system (e.g. radionuclide concentrations in groundwater at different locations and depths);
- characteristics derived from system understanding (e.g. container lifetimes and radionuclide fluxes across different boundaries in the disposal facility system); and

- characteristics derived from calculations of the long term evolution of the disposal system (e.g. dose).

A distinction is also usually drawn between a ‘performance indicator’ and a ‘safety indicator’. A performance indicator provides measures of performance to support the development of system understanding and to assess the quality, reliability or effectiveness of a disposal system as a whole or of particular aspects or components of a disposal system. A safety indicator (which may be regarded as a special type of performance indicator) is used to assess calculated performance in terms of overall safety.

The most widely used safety indicators for assessing the safety of radioactive waste disposal are radiological dose and risk. Other safety indicators that may be used in addition to dose and risk are referred to as *complementary safety indicators*.

Performance and safety indicators are not generally useful to a safety case if taken in isolation. They are usually compared to some reference value, guideline or criteria (sometimes called a “yardstick”) to provide an effective comparative measure. A reference value may provide a direct test of the ability of the overall disposal system, or a system component, to contribute to safety by limiting the radiological impact or attenuating radionuclide releases. Alternatively, it may relate to a property that a system component should fulfil in order either to be effective itself as a barrier, or to provide a suitable environment for the operation of other parts of the system. Reference values may be derived from a number of sources, including legislation or regulation, which typically provide guidelines or limits on dose or risk. Internationally recognized reference values are not available for all performance and safety indicators, and this is one reason why the use of complementary safety indicators has not gained universal support.

Table 1 provides an overview of performance and safety indicators, including the complementary safety indicators discussed in [2]. The table lists individual indicators, their ‘sources’ (i.e. the type of measurement or calculation used to quantify the indicator), the location within the disposal system to which the indicator applies and the reference values against which the value assigned to the indicator may be judged (where no recognized reference values, suggested or generic parameters are indicated). The two indicators of relevance to this CRP, environmental concentrations and fluxes are highlighted in grey. The system of indicators shown in Table 1 it is not to be interpreted as a rigid structure; it implies neither that all indicators must be developed nor that additional indicators should be identified.

TABLE 1. EXAMPLES OF PERFORMANCE AND SAFETY INDICATORS, FROM [2]

<b>General performance indicators</b>			
<b>Indicator</b>	<b>Source</b>	<b>Application</b>	<b>Reference values</b>
Radionuclide transfer times	Quantitative evaluations of parts of total system	Engineered barriers ( <i>e.g.</i> clay backfill) or geosphere	Radionuclide half-lives
Radionuclide concentrations in the near field	Assessment model results	Engineered barrier system	Subsystem criteria derived from sensitivity analyses ("relative" comparisons between different analyses may also be of value - see main text)
Radionuclide fluxes in the near field	Assessment model results	Engineered barrier system	Subsystem criteria derived from sensitivity analyses ("relative" comparisons between different analyses may also be of value - see main text)
Characteristics that control "dilution" in time and space ( <i>e.g.</i> waste-form dissolution or release rates, canister failure rate, porosities)	Experiments, technical specifications, and/or process model calculations	Engineered barriers or geosphere	Criteria derived from total system performance-assessment calculations
Age profile of groundwater	Site characterization, paleohydrogeology	Geosphere	Timescale of assessment
Other physico-chemical properties of the disposal system ( <i>e.g.</i> waste package loading, buffer composition and density, fracture frequency, lack of exploitable mineral resources)	Experiments, technical specifications, and/or process model calculations	Engineered barriers or geosphere	Subsystem criteria developed by the regulatory authority or by the operator
<b>Safety indicators</b>			
Risk	Assessment model results	Human beings	Risk limit or constraint
Dose	Assessment model results	Human beings	Dose limit or constraint
Environmental impact	Assessment model results	Other species	Environmental protection standards
<b>Complementary safety indicators</b>			
Radionuclide concentrations outside the near field	Assessment model results	Accessible environment	Levels of corresponding natural concentrations
Radionuclide fluxes outside the near field	Assessment model results	Accessible environment; geosphere-biosphere interface	Corresponding natural fluxes
Containment times	Experiments, technical specifications, and/or process model calculations	Canisters/ containers Engineered barriers or geosphere	"Crossover times" for hazard indices

The proposed system of indicators in Table 1 is meant to help in placing the various indicators in a logical relationship with the various elements of a safety case. A safety case may use dose or risk or both, together with a number of complementary safety indicators. The most useful

application of the proposed indicators will need to be determined on a case by case basis. The appropriateness of the different indicators is likely to vary depending on the characteristics of the waste, the facility and the assessment context. The exact manner in which complementary safety indicators may be used in safety cases is also likely to vary among countries, depending, for example, on the particular requirements of individual licensing authorities. It is possible that in some cases their use might become mandatory, through inclusion in regulatory requirements. In other cases, it may be up to the disposal system developer to choose from among the full set of potential indicators. It is conceivable that complementary safety indicators could be applied to all assessment time frames, alongside calculations of dose and risk, or they may be applied only to assessment times in the far future when quantitative dose and risk estimates are most uncertain.

### 2.3. APPLICATION TO A SAFETY CASE

This subsection addresses the potential application of complementary safety indicators, particularly environmental concentrations and fluxes, to safety cases for radioactive waste disposal systems.

Complementary safety indicators may go some way to overcome the disadvantages of dose and risk discussed in the previous sections. Their main potential advantage is that they are derived, in most cases, from calculations of radionuclide migration and distribution in the relatively stable medium of the geosphere, eliminating from the assessment the part of the system (the biosphere) characterized by the most intractable uncertainty.

There are, however, other potential advantages to using complementary safety indicators over and above the fact that they avoid the need to make assumptions about future biospheric and demographic conditions in estimating the values of the indicators themselves. The reference values against which complementary indicators are compared may also be based on observations of natural systems generally characterized by long term stability. Complementary safety indicators could also alleviate some of the concerns associated with the application of radiation-protection principles to the impacts of radioactive waste disposal in the remote future, such as uncertainty about the future validity of the assumed relationship between dose and detriment [12].

Complementary indicators provide for flexibility, diversity and transparency for a wide range of stakeholders (technical and non-technical). They provide numbers to compare with items which may be more easily understood by these stakeholders, for example, background radiation levels, fluxes and concentrations of naturally occurring radionuclides and other natural or man-made hazards.

Nonetheless, although complementary safety indicators offer a number of benefits, they also have some disadvantages, the most obvious of which is the lack of international consensus on how to apply them to safety cases. Other practical problems relate to the availability of information on natural radionuclide concentrations and fluxes, and their variability, for use in the development of reference values: this limitation provides the main focus of this CRP.

The application of environmental concentrations and fluxes as complementary safety indicators in a safety case is expected to be of most value when evaluating the consequences of normal or 'expected' disposal system evolution scenarios in which radionuclides released from the near-field are transported to the accessible environment by natural processes (e.g. groundwater movement). The approach is likely to be less useful when evaluating the



consequences of alternative evolution scenarios in which the geological barrier is bypassed or degraded, such as those involving human intrusion or incomplete closure and sealing of the facility.

In the normal evolution scenario, radionuclides and other contaminants will slowly be released from the wasteform and will migrate in groundwater (or in gaseous phase) through the near and far-fields to towards the accessible environment. Their rates of movement (fluxes) and concentrations at various places along the migration path will be controlled, in part, by interactions with the groundwater and rock along the way. The resulting distributions of waste derived radionuclides would be calculated in the safety assessment. These calculated waste derived radionuclide distributions can be compared with the concentrations and fluxes of naturally occurring chemical species in the same or similar geological environments. The concentrations and fluxes of naturally occurring species would constitute the reference values for comparison with the relevant safety indicators (which in this case are the waste derived environmental concentrations and fluxes).

Such comparisons can be valid because both the natural and waste derived species would occur in the same or similar systems and their transport behaviour would effectively be controlled by exactly the same processes, at the same rates.

### **2.3.1. Possible comparisons**

Although simple in concept, there is currently no consensus on the most appropriate comparisons to make between the natural and waste derived concentrations and fluxes, or on the interpretation of such comparisons. The IAEA's Working Group on Principles and Criteria [2] discussed this issue in some detail and considered possible comparisons in terms of:

- What elements or radionuclides to consider?
- What processes (fluxes) to consider?
- What spatial scales to consider (e.g. site scale or larger)?

The answers to these questions were considered to be specific to the disposal system under investigation and the relevant assessment context. As a consequence, no single or generic description of 'the natural system' was considered to be appropriate as a source of reference values for comparison with the safety indicators.

Recommendations for the application of environmental concentrations and fluxes to a safety case were also discussed within the CRP. It was recognized that the actual application must be consistent with the context for each assessment and, therefore, the CRP could not define a single set of detailed guidelines. Nonetheless, it was felt that it would be useful to provide some suggestions to safety assessors for the types of comparisons that are possible between natural and repository systems. Two basic types of comparisons were defined:

***Inventory comparisons*** – these describe the inventory at ' $t = 0$ ' when the waste is first emplaced and before any releases occur. These may be used to support and justify why certain disposal approaches are being adopted.

- comparisons of elemental mass (e.g. mass of U in the waste and mass of U in the rock), comparisons could be made, for example, either for a unit volume of rock (e.g. 1 m<sup>3</sup>) or for the repository equivalent rock volume (RERV);
- comparisons of total activity (e.g. total radioactivity in the waste and in the rock) and comparisons could be made for the same rock volumes as for mass; and
- comparisons of total radiological hazard (e.g. expressing the total radiological hazard by summing the number of Annual Limits of Intake (ALI) units for all contaminants in the waste and in the rock), comparisons could be made for the same rock volumes as for mass.

***Release comparisons*** – these describe the small proportion of the inventory that is released from the engineered barriers to the near-surface environment. They may be used to support the dose/risk calculations and to place the associated hazards in a natural context

- comparisons of concentrations (e.g. concentration of disposal system releases in near-surface materials such as the rock, groundwater or sediments compared to concentration of naturally occurring chemical species in the same materials), the concentrations may be expressed in terms of mass, total activity, activity per nuclide or in equivalent ALI units;
- comparisons of fluxes (e.g. fluxes of repository releases across the geosphere-biosphere interface ‘GBI’ or other relevant boundary compared to fluxes of naturally occurring chemical species at the same boundary), the fluxes may be expressed in terms of mass, total activity, activity per nuclide or in equivalent ALI units occurring in the liquid phase (groundwater discharge) or solid phase (erosion) per km<sup>2</sup>; and
- comparisons of concentrations with relevant limits such as the World Health Organization ‘WHO’ drinking water standards.

The CRP did not have the resources to test these different proposed comparisons and it is hoped that this could be done at a later stage.

### **3. SUMMARY GEOCHEMICAL DATA ACQUIRED IN THE CRP**

#### **3.1. GENERAL OBSERVATIONS ON THE GEOCHEMICAL DATA**

The Earth’s land surface is unique and highly variable in nature, being dependent at any given location on geography, topography, geology and climate. This variability in the land surface gives rise to the variable elemental concentrations, distributions and fluxes reported by each country member under this CRP. The theme of spatial variability recurs in all the detailed accounts (see Appendices A to I), and therefore references to countries and their regions have been maintained in the discussions below.

The first two specific research objectives of the CRP, defined at the outset of the project, were (see Section 1):

- to develop a database of measured concentrations of target substances (elements, radioactive isotopes and particular chemical compounds) in different solid and aqueous materials, from different geographical areas, showing the spatial variability in these concentrations from the regional to the site scale; and
- to develop a database of estimated fluxes of the target substances between different subsurface and surface compartments, showing the spatial and numerical variations in natural fluxes from the regional to the site scale;

As the project progressed through its first year, however, it became evident that it would be neither sensible nor possible to attempt to combine all of the geochemical data acquired by the participants into a single database because the datasets acquired in each country were not directly compatible with each other. There were several reasons for this, the most important being:

- Data from different countries represented widely varying spatial scales, spatial averages and sampling densities (i.e. the spacing between samples). In some cases, data were reported from regional or country sized surveys that used low sample densities when, in other cases, data were reported from small areas using high sampling densities. Statistically, such datasets do not represent the same geochemical information regarding the geochemical variability of different countries and should not be so interpreted;
- Data from different countries were often acquired using different analytical methods or sampling procedures. In some cases, numerous individual samples of particular soil horizons or stream sediment size fractions were analysed using laboratory equipment, in other cases data were acquired using airborne geophysical methods or other methods that averaged over large areas. The different approaches typically reflected the difference both in size and accessibility of countries such as China or Argentina compared to countries such as Finland and the UK. Again, statistically, such datasets do not represent the same geochemical information;
- Not every country reported data for all of the elements and radionuclides considered, and some countries reported ranges (maximum, minimum) whilst others reported only averages (usually mean with standard deviation also reported in some cases); and
- Data were collected from only the 9 participant countries (and not necessarily from all areas in each of those 9) and, thus, cannot be considered to be wholly representative of the true global geochemical variability.

As a consequence, it was decided that it is most appropriate to report the geochemical data separately for each country. This should not detract from the usefulness of the information acquired within the CRP because comparison of the country datasets provides some qualitative understanding of the geochemical variability within and between the regions for which data are available. Furthermore, developing views on how environmental concentrations and fluxes may be used as safety indicators in a safety case [2] suggest that the most appropriate spatial scales for the comparison between the disposal and natural systems is the site or local area scales. Hence, it would not be necessary for the IAEA or other organizations to provide a true ‘global’ database but, rather, to focus more on regional datasets that address the geochemical variability on spatial scales similar to those in which a disposal system would be located.

It follows that understanding the geochemical variability on regional scales appropriate to the siting of a disposal facility is most important for defining appropriate reference values to compare with the environmental concentration and flux safety indicators discussed in Section 2.

### 3.2. OVERVIEW OF THE ACQUIRED CONCENTRATION DATA

This subsection contains a summary description and overview of the elemental concentration-related data acquired in the project. The data include mass concentration ranges and averages for rocks, soils/sediments and waters and an indication of spatial and lithological variations. Some natural radionuclide radioactivity concentrations are also presented. This compilation enables a reader to make comparisons between different rock types and geographical situations in a relatively easy way. See Appendices A to I for further details and fuller explanation of the data sets.

The elements and natural nuclides considered here are mainly those for U, Th, K, Ra, and Rn. In contrast, there are relatively fewer data available to the CRP for some of the other elements of interest namely Rb, Cs, Cu and Sn.

In order to convert mass concentrations to activity concentrations for the natural radioelements U, Th and K, the following expressions were used (to three significant figures):

- Activity of  $^{238}\text{U}$  in 1 g of natural U =  $1.24 \times 10^4$  (Bq)
- Activity of  $^{234}\text{U}$  in 1 g of natural U =  $1.24 \times 10^4$  (Bq)
- Activity of  $^{235}\text{U}$  in 1 g of natural U =  $5.68 \times 10^2$  (Bq)
- Activity of  $^{(238+235+234)}\text{U}$  in 1 g of natural U =  $2.53 \times 10^4$  (Bq)
- Activity of  $^{232}\text{Th}$  in 1 g of natural Th =  $4.06 \times 10^3$  (Bq)
- Activity of  $^{40}\text{K}$  in 1 g of natural K = 30.4 (Bq)

#### 3.2.1. Elemental concentration data for rocks by country

Geoscientific research in China (see Appendix C) has produced some average elemental abundances in rocks belonging to China's main geological areas (see Table 2).

TABLE 2. ELEMENTAL CONCENTRATION DATA FOR THE MAIN GEOLOGICAL AREAS IN CHINA

Element (mg/kg)	Cathaysian craton	West China craton	South Tibet craton	East China
U	1.85	3.10	2.40	1.8
Th	6.21	8.89	7.45	9.5
K	9530	8890	9900	Not available
Rb	29	97	57	95
Cs	1.0	8.2	3.9	3.3
Cu	35	42	44	17
Sn	2.98	2.46	3.08	1.8

These data indicate that the variability of radioelements is within about a factor of 2 on the large geographical areas represented by the different cratons. Somewhat larger variations are seen when comparing the average elemental abundances in different rock types in China [Table 3], of around one order of magnitude.

TABLE 3. ELEMENTAL ABUNDANCES IN VARIOUS ROCKS OF CHINA

Element (mg/kg)	Granite	Granodiorite	Diorite	Basalt	Limestone	Quartzose-sandstone
U	2.9	1.5	1.2	0.73	1.3	2.1
Th	17	9.0	5.4	3.0	1.1	4.0
Rb	160	95	64	23	10	23
Cs	3.6	3.3	1.9	0.83	0.6	0.8
Cu	5.5	18.5	27	52	4.2	4.4
Sn	2.2	1.4	1.3	1.2	0.5	0.6

Cuba has been investigated in terms of its three geographical regions, Western, Central and Eastern. From each, a variety of rock types were subjected to geochemical analysis: in the Western Region limestone, sandstone, shale and lutite; in the Central Region limestone, serpentine, skarn, granite, tuff and shale; and in the Eastern Region limestone, shale, tuff, granite and serpentine. Some results, rounded to three significant figures, are given in Table 4. The values designated as ‘background’ represent average mass concentrations.

TABLE 4. ELEMENTAL ABUNDANCES IN THE REGIONAL ROCKS OF CUBA

Element		West	Centre	East
U (mg/kg)	Minimum	0	1.00	1.25
	Maximum	1180	428	72.5
	Background	80.9	92.1	16.0
Th (mg/kg)	Minimum	0	0	1.91
	Maximum	29.5	47.5	41.0
	Background	8.37	8.95	13.8
Ra (mg/kg)	Minimum	22	2.00	2.00
	Maximum	1070	917	87.5
	Background	71.5	192	13.9
K (mg/kg)	Minimum	0	500	1000
	Maximum	37 500	34 200	53 000
	Background	9900	14 800	22 300

The variations in concentration are associated with different lithologies. Table 5 gives an indication of how the abundances differ between the different rock types in the three regions of Cuba.

TABLE 5. NATURAL RADIONUCLIDE CONCENTRATIONS FOR ROCK TYPES IN CUBA

Element		West			Central			East		
(mg/kg)		Lst	Lutite	Shale	Skarn	Lst	Shale	Granite	Lst	Tuff
U	Min.	1	33	5	2	8	50	5	2	5
	Max.	673	488	229	3445	160	63	29	14	184
Ra	Min.	5	32	5	7	6	26	2	N/A	2
	Max.	577	490	134	2500	50	628	22	N/A	129
Th	Min.	0	1	0.14	0.30	0	2.5	5	0.13	2
	Max.	20	20	28	150	150	16	48	202	203
K	Min.	0	1000	1000	0	0	5000	3000	0	2000
	Max.	40 000	20 000	35 000	50 000	50 000	40 000	84 000	15 000	138 600

The concentrations of the radioelements U, Th and K in selected rocks from regions of the Czech Republic which are being proposed for a deep geological repository in the country are presented in Table 6 below.

TABLE 6. CONCENTRATIONS OF THE MAIN RADIOELEMENTS IN SELECTED ROCK TYPES IN THE CZECH REPUBLIC

	<b>K</b>	<b>eU</b>	<b>eTh</b>
<b>Rock Type (massif)</b>	<b>(mg/kg)</b>	<b>(mg/kg)</b>	<b>(mg/kg)</b>
Porphyric granite (Borsky massif)	41 000	6.1	19.9
Migmatites (sumavske moldanubikum unit)	25 000	4.4	12.0
Granulites (sumavske moldanubikum unit)	29 000	1.9	6.9
Granodiorites (sumavske moldanubikum unit)	33 000	6.1	18.0
Granite (Chvaletice Massif)	35 000	6.9	17.1
Granodiorite (Kladrub Massif)	32 000	3.8	10.6
Porphyric biotitic granite (Sedmihori Massif)	39 000	6.1	30.9
Two-mica adamellite (Sedmihori massif)	35 000	4.3	11.2
Granodiorite - Merklin type (Stod massif)	19 000	3.7	8.2
Granodiorite (Stenovice Massif)	21 000	4.5	13.6
Granite - mountains type (Carlsbad Massif)	36 000	4.0	27.3
Granite – Erzgebirge type (Carlsbad Massif)	42 000	14.2	22.0
Granites (Kynzvalt – Zandov Massif)	38 000	7.2	15.8
Granite (Tis Massif)	15 000–20 000	2–4	6–8
Granodiorite (Cista – Jesenice Massif)	10 000–15 000	3–5	8–14
Granodiorite (Buk site, Central Bohemian Plutonic Complex)	19 000	3.6	13.4
Granodiorite (Konetopy site, Central Bohemian Plutonic Complex)	25 000	4.2	16.4

The following rock data for Finland (Table 7) come from Report no. 4 of the Finnish report series included in the attached CD.

TABLE 7. TYPICAL CONCENTRATIONS OF SOME ELEMENTS IN SELECTED ROCK TYPES IN FINLAND

<b>Element (mg/kg)</b>	<b>Granite</b>	<b>Granodiorite</b>	<b>Diorite</b>	<b>Gabbro</b>
U	2–0	0–12	0.3–4.0	0.1–3.5
Th	4–264	2–28	0–7.1	0.06–2.7
K	6000–75 000	7000–49 000	1700–30 000	100–35 000
Rb	22–1160	<10–206	3–130	1–278
Cu	<10–170	0–50	10–730	16–8230

Some averaged elemental concentrations in crystalline and sedimentary rocks of Japan are presented in Table 8 below.

TABLE 8. ELEMENTAL CONCENTRATION DATA FOR ROCKS IN JAPAN

<b>Element</b>	<b>Average (mg/kg)</b>	<b>Range of Values</b>
<b>Crystalline rocks</b>		
U	1.89	0.01–21
Th	3	0–37
K	14 700	25–67 300
Rb	59	0.12–676
Cs	3.0	0.02–27
Cu	31.4	0–558
Sn	1.31	0.05–17.4
<b>Sedimentary rocks</b>		
U	2.89	0.05–13
Th	7.85	0–51
K	17 300	6.2–50 700
Rb	–	0.12–250
Cs	–	0.005–16
Cu	37.2	0.1–206
Sn	–	1–7.0



Petrophysical measurements have given the following concentration estimates for U, Th and K (see Table 10 below derived from Table H3 in Appendix H), which are thought to be fairly representative for the Swedish Precambrian bedrock.

TABLE 9. ELEMENTAL CONCENTRATION DATA FOR BEDROCK IN SWEDEN

<b>Element</b>	<b>Average (mg/kg)</b>	<b>Standard Deviation</b>	<b>Median (mg/kg)</b>
U	5.06	9.03	3.56
Th	16.1	21.4	11.8
K	32 200	13 400	33 000

Some estimated average elemental mass concentration data for the major UK rock types are presented in Table 10).

TABLE 10. ELEMENTAL ABUNDANCES IN SOME MAJOR ROCK TYPES IN THE UK

<b>Element (mg/kg)</b>	<b>Granite</b>	<b>Gabbro</b>	<b>Gneiss</b>	<b>Shale</b>	<b>Limestone</b>	<b>Sandstone</b>
U	4.8	0.6	3.2	3.5	2.2	0.6
Th	27	2.8	16	12	1.7	3.2
K	40 000	10 000	n/a	32 000	6000	n/a
Rb	150	30	n/a	140	50	46
Cu	10	100	n/a	50	4	n/a
Sn	3	1	2.2	4	0.5	0.5

### 3.2.2. Review of the elemental concentration data from rocks

It is difficult to compare and contrast the elemental concentration data for rocks presented by the different countries, and summarized in the previous subsection, because some data are reported for specific rock types and some as averages for specific regions (that would contain more than one rock type). Similarly, it is not feasible to derive a true mean concentration for each element (or provide other statistical analysis) from the data acquired. Some basic observations can, however, be made, as follows:

- The reported abundances for specific elements are not grossly different between the countries. In most cases, the variations for each element are within one order of magnitude, except where data relate to mineralized regions where enhanced elemental abundances would be expected. For example, the ‘typical’ reported U concentrations are

in the range of ~1 to 15 mg/kg, with a median value of around 4 to 5 mg/kg. The exception are the background U concentration values reported for Cuba which, for the Central and Western regions, are in the region 80 to 90 mg/kg, and a maximum reported concentration of 1180 mg/kg [Table 4]; and

- The highest concentrations reported in all cases is for the element K, which reflects its importance as an essential element in many silicate minerals and a major element in many rock types (as opposed to the other reported elements which typically occur in trace amounts in most rocks).

### 3.2.3. Radionuclide concentration data for rocks by country

Fewer data were reported for radionuclide concentrations (by activity) than were reported for elemental concentrations (by mass). This is not surprising given the greater difficulty of measuring radionuclide abundances. Table 11 contains some activity data for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{226}\text{Ra}$  for a variety of rock types in the UK.

TABLE 11. MEASURED ACTIVITIES FOR SOME NUCLIDES FROM VARIOUS ROCK TYPES IN THE UK

Rock/location	Measured activity (Bq/kg)
<i>Measurements of <math>^{238}\text{U}</math>:</i>	
Caledonian Granite	9–160
Granite	55–770
Rock – Scotland	1–190
Granite – southwest England	4–440
Uraniferous limestone – Scotland	70–3600
Uraniferous vein	2 480 000
Sandstone – Orkney	140–28 000
Sandstone	22–23
<i>Measurements of <math>^{232}\text{Th}</math>:</i>	
Caledonian granites	12–160
Granite	2–280
Sandstone	27–30
Uraniferous limestone – Scotland	7.5–170
<i>Measurements of <math>^{226}\text{Ra}</math>:</i>	
Uraniferous limestone – Scotland	79–1700

The range of activities for each rock given in Table 11 reflects both mineralized and un-mineralized rocks.

### 3.2.4. Elemental and radionuclide concentration data for soils and sediments

Soil data can be very effective in indicating the composition of the underlying rocks, because the soils are largely derived from the rock by physical denudation processes, with an additional input from biological processes. Soil data do not, however, provide exact compositions of the rocks present at the sample point because:

- soils can be transported from their parent rocks;
- soils can be mixed together;
- biological activity preferentially mobilises certain nutrient elements;
- readily soluble chemical species are depleted from the soil and preferentially go into solution; and
- they may be subject to anthropogenic contamination.

Geochemical soil surveys in China (see Appendix C) have resulted in the following nationwide information on the average radioactivity concentrations for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  and the corresponding average elemental mass concentrations in soils for U, Th and K (Table 12). The elemental mass concentrations for U, Th and K were calculated from the quoted  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  nuclide activity concentrations respectively.

TABLE 12. SOIL CONCENTRATION AND ACTIVITY DATA FOR CHINA

Element/Nuclide (units)	Average (area weighted)	Standard Deviation	Range of Values
$^{238}\text{U}$ (Bq/kg)	39.5	34.4	1.8–520
U (mg/kg)	3.19	2.77	0.15–42
$^{232}\text{Th}$ (Bq/kg)	49.1	27.6	1.0–438
Th (mg/kg)	12.1	6.8	0.25–108
$^{40}\text{K}$ (Bq/kg)	580	202	11.5–2185
K (mg/kg)	19 080	6645	378–71 880

The soils from the southern half of China are richer in U and Th than those in the northern half. The geochemical nature of the parent rock is a large determinant for what is present in the soils. For example K is lowest in soils developed on metamorphic rocks.

The average U, Th and K contents of soils in Cuba are presented in Table 13 below. The values are taken from the airborne gamma spectrometric data in Table D4 in Appendix D. The ‘aerial’ data are largely a measure of signals from the upper layers of the ground surface where soils and sediments are residing as overburden.

TABLE 13. SOIL CONCENTRATION DATA FOR THE REGIONS OF CUBA

Element	Western Region	Central Region	Eastern Region
	Average and [range] (mg/kg)	Average and [range] (mg/kg)	Average and [range] (mg/kg)
U	4.3 [0.73–9.6]	3.51 [1.40–9.70]	0.91 [0.93–15.9]
Th	6.86 [0.96–19.1]	2.27 [0.78–7.70]	9.65 [0.37–62.0]
K	5000 [2600–20 300]	5400 [2800–25 100]	11 300 [3100–51 700]

The concentration variations are mainly a product of the different parent-rocks bearing the soils and sediments. This has been noted above for the elemental concentrations in different lithological units of Cuba.

Some averaged elemental concentrations in different soil/sediment compartments of Finland are presented in Table 14 below.

TABLE 14. ELEMENTAL CONCENTRATION DATA FOR SOIL/SEDIMENTS FROM FINLAND

Element	Average (mg/kg)	Standard Deviation	Range of Values
<b>Till</b>			
U	3.42	2.04	<0.3–48
Th	Not calculated	Not calculated	<20–639
K	2470	1855	<1300–24 800
Rb	76.1	25.1	<15.0–239
Cu	28	26	<8–1640
<b>Lake sediments</b>			
U	7.52	59	0.008–5020
Cu	37.5	78	1.0–3280
<b>Stream sediments</b>			
U	4.31	15	<0.01–379
Th	5.69	3.47	<0.04–31.2
K	2220	1999	<200–14 100
Cu	15.9	18	<1–476

Till covers much of the land surface in Finland, and where till is absent peat often occurs. Although 30% of the land area is covered by peat, no data are given for this particular biosphere compartment.

Some averaged elemental concentrations in different soil/sediment compartments of Japan are presented in Table 15 below.

TABLE 15. ELEMENTAL CONCENTRATION DATA FOR SOIL/SEDIMENTS IN JAPAN

Element	Average (mg/kg)	Range of Values
<b>Soils</b>		
U	1.3	0.258–5.51
Th	5.6	2.56–51.3
K	17 500	26–51 100
Rb	166	14.5–296
Cs	–	1.5–3.8
Cu	–	2.5–888
<b>Stream/lake sediments</b>		
U	2.29	1.0–3.87
Th	–	2.33–45.3
K	–	5810–26 600
Rb	–	26.9–285
Cs	–	0.94–30.6
Cu	80.6	5.1–4630
Sn	–	2.77–195
<b>Marine sediments</b>		
U	4.73	1.71–18.4
Th	–	5.66–17.5
Cu	–	1.60–40.0
Sn	–	6.70–15.3

The following Table 16 is a compilation of activity concentration data from soils (0 – 10 cm depth) of the Osaka Prefecture in Japan, and gamma radiometric data for nuclides in rice field soils in Japan.

TABLE 16. ACTIVITY CONCENTRATION DATA FOR SOILS IN JAPAN

Element	Average (Bq/kg)	Range of Values
<b>Soils in Osaka Pref.</b>		
$^{238}\text{U}$	37.0	19–82
$^{228}\text{Ra}$	48.5	21–119
$^{40}\text{K}$	676	384–1104
<b>Rice field soils</b>		
$^{226}\text{Ra}$	67	21–195
$^{40}\text{K}$	425	130–880
$^{137}\text{Cs}$	21	8.1–82

Airborne gamma measurements carried out in Sweden give the following estimates of the U, Th and K contents for the upper part of the overburden i.e. mainly for soils (see Table 17 below derived from Table H2 in Appendix H).

TABLE 17. ELEMENTAL CONCENTRATION DATA FOR SOILS IN SWEDEN

Element	Average (mg/kg)	Standard Deviation	Range of Values
U	1.79	1.48	0.00–50.0
Th	6.40	4.96	0.00–199
K	15 200	9300	0–69 500

The UK contains a wide range of soil types and these can have quite distinct compositions. Soil compositions from across the UK have been determined by the British Geological Survey (BGS) as part of its ongoing G-BASE project. The most recent Geochemical Atlases (for NW England and Wales, and NE England) record minimum, average and maximum measured concentrations for 14 and 30 elements, respectively. Some of these soil data are summarized in Table 18. NM means not measured.

TABLE 18. STATISTICAL DATA ON SOILS FROM THE BGS GEOCHEMICAL ATLASES FOR NE ENGLAND AND NW ENGLAND AND WALES

	<b>NW England and Wales</b>		<b>NE England</b>	
<b>Element</b>	<b>Average (mg/kg)</b>	<b>Range of Values</b>	<b>Average (mg/kg)</b>	<b>Range of Values</b>
U	NM	NM	3	0.5–17.2
K	NM	NM	15 000	3000– 45 000
Rb	NM	NM	79	10–199
Cu	43	7–1316	23	7–543
Sn	8.4	2–880	5	5–85

The soil data given in Table 18 can be considered to be representative of the likely ranges in elemental concentrations in soils over the entire UK. The reason for this assumption is that the NW England and Wales, and NE England regions contain all of the major rock types to be found in the UK, including granite, metamorphic rocks, sandstone, limestone, mudstone etc. Given that a dominant control over soil compositions is the underlying geology, this should mean that the soils in these regions should display much of the chemical characteristics of soils elsewhere in the UK, with the possible exception of soils derived from the Chalk.

Some relevant data are presented in Table 19 for stream sediments for the same two regions of the UK.

TABLE 19. STATISTICAL DATA ON STREAM SEDIMENTS FROM THE BGS GEOCHEMICAL ATLASES FOR NE ENGLAND, AND NW ENGLAND AND WALES

	<b>NW England and Wales</b>		<b>NE England</b>	
<b>Element</b>	<b>Average (mg/kg)</b>	<b>Range of Values</b>	<b>Average (mg/kg)</b>	<b>Range of Values</b>
U	3.3	0.3–67.8	3.8	0.4–22.7
K	21 000	4000–59 000	19000	2000–58 000
Rb	107	15–300	83	6–253
Cu	41	7–1408	24	3–953
Sn	7.2	2–834	6	6–475

Fewer analyses have been made for the radioactivity associated with UK soils and sediments than the elemental compositions. Most of the activity concentration data available up to 1993 are compiled in Table 20 (taken from [7]).

TABLE 20. MEASURED ACTIVITIES FOR NUCLIDES FROM UK SOILS AND SEDIMENTS

Material/location	Measured activity (Bq/kg)
<i>Measurements of <math>^{238}\text{U}</math>:</i>	
Soil	50–330
Soil–Scotland	14–200
Soil–Cornwall	29–2000
Uraniferous soil–Scotland	18–9800
Clay	31
Alluvium	34
Stream sediment	<12–1900
Sediment–Irish Sea	19
<i>Measurements of <math>^{232}\text{Th}</math>:</i>	
Soil–Scotland	9.3–130
Soil–Cornwall	11–57
Soil	8–94
Uraniferous soil–Scotland	5–21
Sediment–Irish Sea	20
Clay	38
<i>Measurements of <math>^{40}\text{K}</math>:</i>	
Soil–Cumbria	410–590
Soil–Exmoor	300–560
Soil–Scottish Islands	0–3200
Soil–Scotland	0–2300

Where no location is given, the value is an average of a number of analyses from different locations. The activity data for soils and sediments are broadly consistent with the activity data for UK rocks.

### 3.2.5. Elemental concentration data for waters

Concentration data for the natural radioelements in water bodies are essential for deriving estimates of natural radioactivity fluxes via water pathways. Large concentration variations



would be expected from region to region and according to the season, sediment loading, salinity and temperature. Running surface waters would be especially prone to elemental concentration variations. Deep aquifers, especially those that are confined, may be the exception to the normal rule of wide variability in concentration. However, even for these aquifers different water-rock interactions can give rise to different salinity and redox zones and consequently spatial heterogeneity in the hydrogeochemical composition of an aquifer.

Table 21 below summarizes some measured information from China (see Tables C1 and C2 in Appendix C). The data in Table 21 indicate:

- the U concentration values in various water bodies, with the exception of saltwater lake, are of the order of  $\mu\text{g/l}$ ;
- Th concentration values are typically in units of  $10^{-1}\mu\text{g/l}$ ;
- and  $^{40}\text{K}$  concentrations range from 50 - 400 mBq/l, with the exception of sea, saltwater lake and hot spring samples.

TABLE 21. URANIUM, THORIUM AND  $^{40}\text{K}$  CONCENTRATIONS IN WATER BODIES IN CHINA

Water Body		U ( $\mu\text{g/l}$ )		Th ( $\mu\text{g/l}$ )		$^{40}\text{K}$ (mBq/l)	
		Range	Average	Range	Average	Range	Average
Rivers		0.02-42.35	1.66*	<0.01-9.07	0.31*	8.0-7149	89.4*
Lake	freshwater	0.04-19.20	2.10**	0.02-0.93	0.24**	4.8-2640	102.4**
	saltwater	1.07-387.0	22.36**	0.04-8.60	0.64	54.8-22410	2108**
Reservoir		0.03-19.50	0.73**	<0.01-2.04	0.09**	3.2-1205	50.2
Spring	hot	0.02-18.60	0.87	0.02-4.85	0.25	5.1-8125	1058
	cold	0.04-14.88	1.37	0.01-1.50	0.20	2.9-1830	254.5
Well	shallow	<0.01-101.6	3.82	<0.01-6.29	0.15	3.3-5867	401.9
	deep	0.02-358.87	2.95	<0.01-1.32	0.16	1.0-923.4	122.1
Offshore		0.07-5.20	2.21	<0.01-5.92	0.53	2500-21600	10320
Tap water		0.01-56.68	2.12	0.01-3.00	0.13	1.8-1580	133.3

\* Runoff-weighted average; \*\* storage-water capacity-weighted average.

In addition, Table C2 in Appendix C indicates that  $^{226}\text{Ra}$  has a mean mass concentration value of  $16.5 \times 10^{-17} \text{ g/l}$  for samples from the main rivers of China. The  $^{226}\text{Ra}$  mass concentration values in these rivers range from  $(1.37 - 272) \times 10^{-17} \text{ g/l}$ .

Some radioelement and radioactivity data for shallow groundwater samples from the most important granitic bodies in the Czech Republic are summarized in Table 22 below.

TABLE 22. RADIOELEMENT CONCENTRATION AND RADIOACTIVITY DATA FOR SHALLOW GROUNDWATER IN GRANITES, CZECH REPUBLIC

	Maximum	Minimum	Median
K (mg/l)	78.3	0.4	2.9
U (mg/l)	0.02	0.0001	0.0015
Ra-226 (Bq/l)	20	0.005	0.05
Rn-222 (Bq/l)	8624	1	175
Gross alpha (Bq/l)	14	0.01	0.1
Gross beta (Bq/l)	4.4	0.05	0.195

Radioactivity data for groundwater from deeper boreholes are summarized in Table 23.

TABLE 23. RADIOACTIVITY DATA FOR DEEPER GROUNDWATERS, CZECH REPUBLIC

Gross alpha (Bq/l)			Gross beta (Bq/l)		
Median	Range	No. of samples	Median	Range	No. of samples
0.16	0.019–615	786	0.32	0.08–11.1	192

Czech radioactivity data for groundwater from shallow boreholes (mainly situated in Quaternary sediments) are summarized in Table 24.

TABLE 24. RADIOACTIVITY DATA FOR SHALLOW BOREHOLE WATERS IN QUATERNARY SEDIMENTS, CZECH REPUBLIC

Gross alpha (Bq/l)			Gross beta (Bq/l)		
Median	Range	No. of samples	Median	Range	No. of samples
0.14	0.03–8.3	632	0.39	0.05–3	120

Radioactivity data for Czech spring waters are summarized in Table 25.

TABLE 25. RADIOACTIVITY DATA FOR SPRING WATERS, CZECH REPUBLIC

Gross alpha (Bq/l)			Gross beta (Bq/l)			Rn-222 (Bq/l)		
Median	Range	No. of samples	Median	Range	No. of samples	Median	Range	No. of samples
0.09	0.02–3.2	490	0.13	0.05–1.27	81	72	6–1330	62

Data for waters in Czech rivers and streams are summarized in Table 26.

TABLE 26. RADIOACTIVITY AND RADIOELEMENT DATA FOR SURFACE WATERS IN THE CZECH REPUBLIC

	Median	No. of samples
Gross alpha-total (Bq/l)	0.237	2044
Gross alpha - soluble (Bq/l)	0.152	2138
Gross alpha - suspended (Bq/l)	0.059	2113
U - total (µg/l)	6.0 (range of 0.2–2000)	
U - soluble (µg/l)	4.0 (range of 0.1–2000)	
U - suspended (µg/l)	1.0 (range of 0.1–184.4)	
Ra-226 - soluble (Bq/l)	0.018	6634
Ra-226 - suspended (Bq/l)	0.001	1804
K-40 total (Bq/l)	0.137	1164
K-40 - soluble (Bq/l)	0.130	7189
Gross beta (Bq/l)	0.231	2521
Gross beta - soluble (Bq/l)	0.203	9953
Gross beta - particulate (Bq/l)	0.049	5173
Gross beta after correction for K-40 (Bq/l)	0.025	9041

Czech spa mineral waters contain from <1 to 1000 µg/l of U; from <0.05 to 12.3 Bq/l of Ra-226; and from <0.5 to 1865 Bq/l of Rn-222.

In Finland, country-wide hydrogeochemical mapping of shallow groundwater at a density of 1 sample per 50 km<sup>2</sup> was carried out in 1978-1982, followed up by denser sampling programmes in 1982-1998. In 1999, 1000 new groundwater samples were collected nationwide from springs, dug wells (representing groundwater of the overburden) and wells drilled into bedrock (representing bedrock groundwater from an average depth of about 50 m but with 5% of the depths at over 100 m). Table 27 summarizes some data from Table F1 in Appendix F.

TABLE 27. ELEMENTAL CONCENTRATION DATA FOR WATERS IN FINLAND

Element	Average (µg/l)	Standard Deviation	Range of Values
<b>Stream water</b>			
U	0.186	0.437	<0.01–6.28
K	1.18	1.38	0.068–17.1
Cu	0.932	1.32	0.059–24.5
<b>Groundwater: bedrock wells</b>			
U	13.7	55.1	<0.01–643
Th	Not calculated	Not calculated	<0.02–1.41
Rn (Bq/l)	311	593	1.00–4880
K	4.42	4.93	0.23–40.2
Rb	3.19	4.71	0.03–42.7
Cu	32.3	78.3	<0.04–917
<b>Groundwater: overburden wells</b>			
U	0.846	3.03	<0.01–36.6
Th	Not calculated	Not calculated	<0.02–1.50
Rn (Bq/l)	37.8	82.8	0.10–893
K	4.97	7.36	0.19–92.3
Rb	4.98	7.45	0.04–73.3
Cu	14.1	33.3	<0.04–410

It is estimated that the WHO limit of 2 µg/l for U is exceeded in about 100 000 springs and drilled wells, which means that a population of about 800 000 may be exposed to high U levels. The value of these concentration data in the evaluation of possible harmful effects is clearly evident in these circumstances.

Some averaged elemental concentrations in two different water compartments of Japan are presented in Table 28 below.

TABLE 28. ELEMENTAL CONCENTRATION DATA FOR WATERS IN JAPAN

Element	Average (µg/l)	Range of Values
<b>Surface water</b>		
U	0.0254	0.00087–0.169
Th	0.0035	0.00005–0.0763
Ra (Bq/l)	–	0.00011–0.06
Rn (Bq/l)	–	0.07–157
K	–	250–5500
Rb	1.66	0.07–5.88
Cs	0.0391	0.0031–0.252
Cu	1.96	0.0194–33.0
<b>Groundwater</b>		
U	–	0.0004–1340
Th	–	0.060–0.076
Ra (Bq/l)	–	0.000074–4.10
Rn (Bq/l)	–	0.03–29 500
K	–	80– $3.02 \times 10^6$
Rb	–	0.62–323
Cs	–	0.010–0.52
Cu	–	0.028–32

The concentration values in Table 28 come from a mix of countrywide and local water surveys in Japan.

A countrywide investigation of uranium and radium in groundwater in Sweden showed that U ranges from 0.1 to 43.5 µg/l and that the Ra-226 activity ranges from 0.0005 to 1.90 Bq/l with generally higher values in the Precambrian bedrock. This investigation was based on 42 samples from representative Swedish aquifers. In addition there were samples analysed for Ra from 492 drilled wells. Data on radon were more extensive with about 2500 measurements in springs, dug wells and drilled wells (see summary of results in Table 29 below based on Tables H9-11 in Appendix H).

TABLE 29. CONCENTRATION DATA FOR WATERS IN SWEDEN

Element	Median (Bq/l)	Range of Values
<b>Sedimentary bedrock aquifers</b>		
Quaternary deposits aquifer U (µg/l)	1.0	—
Bedrock aquifer U (µg/l)	0.3	—
Quaternary deposits aquifer Ra	0.006	—
Bedrock aquifer Ra	0.0025	—
<b>Precambrian bedrock aquifers</b>		
Quaternary deposits aquifer U (µg/l)	0.6	—
Bedrock aquifer U (µg/l)	2.3	—
Quaternary deposits aquifer <sup>226</sup> Ra	0.0035	—
Bedrock aquifer <sup>226</sup> Ra	0.097	—
<b>Groundwater – drilled wells</b>		
<sup>226</sup> Ra	0.012	0.0002–2.455
<b>Groundwater – springs, wells</b>		
Springs Rn	39	1–845
Dug wells Rn	20	1–947
Drilled wells Rn	85	0–9289

Another survey of U in groundwater from 269 drilled wells in the Uppsala municipality showed a median of 12 µg/l, with an approximate range of between 0.2 and 246 µg/l. These values can be considered as fairly typical for U in groundwater in Swedish Precambrian bedrock. This could be compared with earlier estimates of about 3 µg/l for U in springs,

which are considered to be similar to the U content of groundwater in the overburden. The U content in surface waters has been estimated to be less than 1 µg/l.

A small number of analyses on Th concentrations in groundwater have indicated low concentrations i.e. < 0.1 µg/l, which is in line with other studies in Sweden.

The median concentrations of U, K, Rb and Cu in groundwater from some major aquifers in the United Kingdom are shown in Table 30 (from [7]).

TABLE 30. ELEMENTAL ABUNDANCES IN SOME IMPORTANT UK AQUIFERS

Element (µg/l)	Aquifer						
	<i>1</i>	<i>2</i>	<i>3</i>	<i>4</i>	<i>5</i>	<i>6</i>	<i>7</i>
U	0.11	1.3	–	<0.1	1.5	0.4	<0.1
K	3500	7800	2600	2000	690	2300	1400
Cu	0.3	0.4	<0.3	0.2	0.4	0.6	0.3
	<i>8</i>	<i>9</i>	<i>10</i>	<i>11</i>	<i>12</i>	<i>13</i>	<i>8</i>
U	5.8	–	0.6	0.5	1.5	0.01	U
K	2400	4600	2000	2700	2900	1400	K
Rb	–	–	–	–	1.2	3.3	Rb
Cu	3.2	2.1	1.6	0.5	4	3.8	Cu

1 = Berkshire chalk; 2 = Central London Basin chalk; 3 = Lincolnshire limestone; 4 = Derbyshire millstone grit; 5 = Derbyshire carboniferous limestone; 6 = Lower greensand; 7 = Ashdown Beds (Wealden); 8 = Shropshire Triassic sandstones; 9 = Shropshire carboniferous limestone; 10 = Lancashire Permo-Triassic sandstones; 11 = Moray Basin Old Red sandstone and Permo-Triassic sediments; 12 = Birmingham Permo-Triassic sandstones; 13 = West Devon upper carboniferous sediments.

The radioelement activities of U and K in groundwater from some major UK aquifers are presented below in Table 31.

TABLE 31. RADIOELEMENT ABUNDANCES IN SOME MAJOR UK AQUIFERS

<b>Aquifer</b>	<b>Maximum U content (µg/l)</b>	<b>Median K content (µg/l)</b>	<b>Activity from U (Bq/l)</b>	<b>Activity from K (Bq/l)</b>
Moray Basin Old Red Sandstone	2.3	2700	0.06	0.07
Central London Basin Chalk	2.1	7800	0.05	0.19
Derbyshire Carboniferous Limestone	2.0	690	0.05	0.02
Lancashire Permo-Triassic Sandstones	2.2	2000	0.06	0.05
Shropshire Triassic Sandstones	8.3	2400	0.21	0.06

The activities of individual nuclides have been measured in a number of UK groundwaters and these are reported in [7] and from this source is derived Table 32 below. Table 32 shows measured activity concentration ranges of  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$  in different types of UK aquifer groundwater.

TABLE 32. RADIUM AND RADON ACTIVITIES IN DIFFERENT UK AQUIFER TYPES

<b>Aquifer type</b>	<b><math>^{226}\text{Ra}</math> range (Bq/l)</b>	<b><math>^{222}\text{Rn}</math> range (Bq/l)</b>
Granites	0.004–0.9	7–1100
Limestones	0.003–0.013	1.4–16
Sandstones	0.002–0.08	3.3–370

The G-BASE database system of the British Geological Survey houses detailed elemental concentrations for UK stream waters on a regional basis. There are other sources too, but there is no easy way of obtaining information on averages and ranges for relevant elements. Activities for some individual nuclides measured in a number of surface waters in the UK are given in Table 33 (derived from [7]). Where no location is cited, the value is an average of a number of analyses from different locations.



TABLE 33. ACTIVITIES FOR SOME INDIVIDUAL NUCLIDES IN DIFFERENT SURFACE WATERS IN THE UK

Surface water/Location	Activity (Bq/l)
<i>Measurements of <math>^{238}\text{U}</math>:</i>	
Stream water - northern Scotland	$1.2 \times 10^{-3}$ –0.59
Stream water – Cornwall	$2.5 \times 10^{-2}$ –2
River water	$<5.5 \times 10^{-3}$ –0.40
River water – Cornwall	$<5.5 \times 10^{-3}$ – $2 \times 10^{-2}$
Seawater - Forth Estuary	$4.9 \times 10^{-4}$ – $4.3 \times 10^{-2}$
<i>Measurements of <math>^{232}\text{Th}</math>:</i>	
River water – drinking	$1 \times 10^{-2}$ – $5 \times 10^{-2}$
Stream water - Cornwall	$1.8 \times 10^{-3}$ – $4.3 \times 10^{-3}$
River water – Cornwall	$1.5 \times 10^{-3}$
<i>Measurements of <math>^{226}\text{Ra}</math>:</i>	
Surface water	$4 \times 10^{-3}$
Surface water – granite Devon	0–0.16
Surface water - sandstone Devon	$4 \times 10^{-3}$ – $1.9 \times 10^{-2}$
Surface water – granite Cornwall	$7 \times 10^{-3}$ – $6 \times 10^{-2}$
Surface water - sandstone Cornwall	$1 \times 10^{-2}$ – $6 \times 10^{-2}$
River water	$1 \times 10^{-2}$ – $4 \times 10^{-2}$
<i>Measurements of <math>^{222}\text{Rn}</math>:</i>	
Surface water - granite Cornwall	5.2–26
Surface water - sandstone Cornwall	0–3.9
Surface water - granite Devon	0–11
Surface water - sandstone Devon	0–1.9
Surface water - Wales	1.9
Surface water	0–15
River water - Cornwall	1.5–4.4
<i>Measurements of <math>^{40}\text{K}</math>:</i>	
Surface water	$5.3 \times 10^{-2}$ –0.23
River water	$<2.0 \times 10^{-2}$ –0.2

### 3.3. OVERVIEW OF THE ACQUIRED PROCESS RATE DATA

#### 3.3.1. Rates of processes

A variety of processes and process-rate data have been used in this project. The processes include, for example, groundwater discharge, denudation and river erosion. Many of these processes have been assigned rates that are specific to the repository areas being considered in different countries. Full details of each country's contribution on this topic are given in the individual Appendices. Table 34 below may serve as a useful guide to the details.

A natural groundwater flow rate and discharge rate in the setting of a repository is dependent on the regional and local hydrogeological conditions. If an underlying aquifer occurs beneath a near-surface repository, knowledge of the groundwater flow rate and the naturally occurring element concentrations in the saturated zone would provide a sound basis for estimating natural element fluxes and developing these as natural safety indicators for the system. This information would be site specific.

If the setting is related to a deep geological repository, the hydraulic heads and gradients may be low and therefore the flow rates and discharge rates would be slow. By itself, this situation is a strong point in support of a safety case, and comparisons between the groundwater release rates of repository nuclides and natural radioelements may be of only secondary importance for supporting the safety case. However, some long term scenarios may envisage changes to the hydrogeological regime, and therefore some estimates of higher future groundwater flow and discharge rates may be necessary. These flow and discharge rates would be site specific.

Measurements of the flow rates of nearby rivers and streams would allow the estimation of dissolved and suspended naturally occurring radioelement loads being transported away from a repository site area. Data on these chemical fluxes by water erosion would provide potentially useful natural safety indicators. River and stream flow rate data would be specific to the site area, and would be dependent on the hydrological characteristics of the catchment area in which the repository is located. Climate would have a large role in determining the masses of dissolved and suspended soil and rock materials being transported by the water courses.

Similarly, measurements of the rates of soil erosion or deposition by wind would provide useful data for the estimation of fluxes of natural U, Th and K and other relevant elements being released from or added to the soils of a repository site area. Again, site specific wind erosion/deposition data would be required. The ultimate fate of wind-borne material carried away from a site would be the rivers and the sea via atmospheric deposition and surface runoff.

Denudation rates of a relevant geographical area are not only useful for estimations of when a deep repository would be expected to approach the near-surface environment, but for deriving flux information for naturally occurring elements being removed from the land surface. Denudation rates would be a complex result of the dynamics of uplift, weathering and erosion by water and wind.

Table 34 below gives an indication of those participant countries which have considered the rates of these different processes. Details can be pursued by referring to the relevant Appendix for that country.

TABLE 34. PROCESS RATES CONSIDERED BY PARTICIPANT COUNTRIES

<b>Process</b>	<b>Countries which have considered the rate of the process</b>
Groundwater flow	Cuba; Finland; UK
Groundwater discharge	China; Finland; Japan; Sweden; UK
River flow and erosion	China; Finland; Japan; Sweden
Wind erosion of soil	China
Denudation	Cuba; Japan; UK

At the site-scale, water infiltration and surface runoff are important processes that contribute to the larger scale erosion processes observed in groundwater discharge and river erosion. For near-surface repositories, data on water infiltration rates through the soil into the unsaturated zone and to the water table would be valuable for estimating natural radioelement transport rates to the saturated zone for direct comparison with rates for repository derived nuclides. This information would be site specific and local scale.

Surface runoff erosion rates for natural dissolved and suspended elements being transported into a surface water body or soil compartment are useful parameters to measure for both near-surface and deep repositories. Surface runoff rate data would be site specific and local scale.

The surface runoff and water infiltration processes have not been illustrated by rate data under this CRP, although they have been described in qualitative terms e.g. see the discussion in Section F1.2.2 in Finland's Appendix F. Rates for these processes may require further future investigation in the development of natural safety indicators, particularly for those countries with repository site(s) already selected and being characterized.

### **3.3.2. Fluxes of natural elements**

The fluxes of naturally occurring elements considered in this project are specific to the sites which have been investigated by participant countries. These flux values can be difficult to estimate either because of the complexity of the different processes contributing to a net chemical flux (as experienced in Finland for uranium) or because of the non-steady state conditions that might prevail at a site (as experienced in Sweden). The Appendices contain the site specific details and explanation of the derivations of relevant fluxes and their uncertainties.

Table 35 below indicates the fluxes which have been estimated for particular natural elements at sites considered by different participant countries. Details can be pursued by referring to the relevant Appendix for that country.

TABLE 35. FLUXES CONSIDERED BY PARTICIPANT COUNTRIES

<b>Flux</b>	<b>Countries which have considered the flux (elements)</b>
River flux	China (U, Th, Rb, Cs, Cu, Sn); Finland (U, Th, Rn, K, Rb, Cu); Sweden (K)
Groundwater flux	Cuba (U, Th, K); Finland (U); Japan (U); Sweden (U, K); United Kingdom (U, Rn, K, Rb, Cs, Cu, I)
Denudation	Japan (U); United Kingdom (U, Th, K, Cs, Sn)

The applications made of these flux data by the various participants are discussed in the next section.

#### 4. CONCLUSIONS AND UTILITY OF THE CRP DATA

##### 4.1. MEETING THE OBJECTIVES OF THE CRP

In the following text, the extent to which the objectives outlined in Section 1 have been met is discussed.

##### 4.1.1. A database of measured concentrations

It is clear from the experiences of the CRP participants, that very large amounts of relevant elemental and radionuclide concentration data exist. These data are, however, often widely dispersed amongst the open and restricted literature, and various private and national organizations, and much of it has never been digitized or catalogued, which makes it a labour-intensive processes to convert into appropriate forms for use in defining reference values for complementary safety indicators. Furthermore, much of these geochemical data have been acquired for commercial reasons (e.g. uranium exploration) and, thus, cannot all be made available to safety assessors. That said, a considerable amount of geochemical data were collated for this study. The most frequently reported data were for elemental concentrations (by mass, mg/kg) of U, Th, K and Rb in rocks and soils. Data for other elements (e.g. Ra) were less frequently reported, and radionuclide concentrations (by activity, Bq/kg) were also less abundant. Nonetheless, several hundred geochemical data entries are reported in the Appendices and the supporting or background reports.

As discussed in Section 3, it was decided that it would be neither sensible nor possible to attempt to combine all of these concentration data into a single database because the datasets acquired in each country were not directly compatible with each other. This was due to the different sampling methods, sampling densities and statistical methods for reporting the data employed by the participants. In the event, this could not have been avoided because all participants were collating and reporting existing geochemical data and, thus, had no control over the methods used to acquire it at source. Only by initiating a new sampling programme and enforcing a standard sampling methodology could a uniform, quality-controlled set of data be developed.

Nonetheless, the very large amount of geochemical data acquired in the project are extremely valuable to the application of environmental concentrations and fluxes as complementary safety indicators, even if all of these data are not presented in a simple ‘look up’ table format. As such, it is concluded that Objective 1 of the CRP has been met but with some limitations. The potential application of these elemental and radionuclide concentration data to a safety case is discussed below.

#### **4.1.2. A database of estimated fluxes**

Fluxes are estimated from measured concentrations of particular chemical species and measured or inferred rates of the processes that cause their mobility. For example, from the concentration of U in groundwater and the rate of groundwater flow.

Fewer data overall were acquired for process rates than for elemental or radionuclide concentrations. Most data of this type related to groundwater flow or discharge, and river flow. Considerably less information was available for the rates of other processes causing mass movement, such as denudation and erosion. This is not surprising because it is very difficult directly to measure rates for these processes or to derive appropriate spatial or temporal average data by indirect methods. As a consequence, the literature and data sources available to the participants held few such explicit process rate data.

Where process rate data were available, they have been used to calculate a number of different elemental or radionuclide fluxes (as was done by China, Finland, Sweden, Cuba, Japan and the UK). The fluxes that have been calculated were summarized in Table 35.

It is difficult directly to compare these calculated fluxes with each other because they often relate to processes operating over widely different spatial and temporal scales. This is particularly the case for groundwater flow and discharge because these fluxes are calculated for a diverse range of flow systems that are represented by a similarly diverse range of conceptual models. This raises an issue as to appropriate methods to present or calculate such values but, nonetheless, it is similar to the situation that arises when trying to compare the calculated releases to the accessible environment from different safety assessments because these intermediate results are also often presented in a variety of incompatible formats and from different conceptual systems (e.g. from a single well, to a river, to an aquifer etc).

As with the concentration data, therefore, it was not considered appropriate to combine all of these flux data into a single database. Note that because the flux data are, in part, derived from the concentration data, the same limitations apply with regard to collating the data. That said, it is evident that, where appropriate data are available, it has proved possible to calculate fluxes of natural chemical species, driven by natural processes that are comparable to the fluxes that are expected to occur from a radioactive waste disposal facility. As such, it is concluded that Objective 2 of the CRP has also been met but with some limitations. The potential application of these elemental flux data to a safety case is discussed below.

#### **4.1.3. Concentration and flux ranges that can be independently assessed to be safe**

It is well known that in some localities around the world, very high elemental and radionuclide concentrations occur due to natural process that have caused their concentration over time. These are often, but not always, regarded as economic orebodies. The best known example is perhaps the Poços de Caldas region in Brazil.

Anecdotal evidence suggests that there are no widespread chronic health effects within the human or animal populations at any of these high concentration sites that would suggest they would be regarded as 'unsafe'. The exception to this is where anthropogenic action (e.g. mining) has caused reworking or remobilization of the orebody which has caused enhanced exposures to the local population where the health effects may be of a chemotoxic nature.

It had been hoped that epidemiological evidence from the participant countries would have been available to confirm or refute the anecdotal evidence and quantify concentration thresholds that could be independently assessed to be safe. In the event, no epidemiological data was reported by the participants. This is believed to relate to the lack of health specialists within the CRP with direct knowledge of or access to the relevant data. As such it is concluded that Objective 3 has not been met. It is probable that some relevant epidemiological data does exist and this objective could possibly be met in future in another study, if it were structured so as to include health specialists with appropriate knowledge.

#### **4.1.4. Publication of the activities and findings of the CRP**

This TECDOC contains the summary presentation of the activities and findings of the CRP. Other related work of the IAEA that considers the application of complementary safety indicators to a safety case is presented in [2]. As such it is concluded that Objective 4 has been met.

## **4.2. UTILITY AND APPLICATION OF THE CRP DATA**

An underlying objective of the CRP was to provide information on the abundances of natural radioelement distributions (concentrations and fluxes) that could be compared to the calculated distributions of waste derived contaminants. This objective has been achieved to a large degree but some thought needs to be given to the utility and application of the data contained in this report.

Essentially there are two types of safety case: generic and site specific. Usually generic safety cases are undertaken to test the feasibility of disposal system design concepts or types of potential host geological environments, or both. Site specific safety cases follow later, and are usually undertaken when one or more short-listed host sites have been identified, and sufficient site characterization data are available to be used in the safety assessment.

The application of environmental concentrations and fluxes as safety indicators to these two types of safety case differs only so far as to the source of the natural geochemical data used to derive reference values for comparison with the safety assessment calculation results (see Section 2). The data provided in this report is most likely to be useful for the generic safety cases but can also be used to provide further input to site specific safety cases.

In the case of generic safety cases, the information in this report can be used to provide 'proxy' reference values for comparison with safety assessment calculation results. The safety assessor needs, however, to choose appropriate sets of data from this report on the basis of which country, region or locality considered within is most similar to the geological environment considered for the generic study. For example, if the generic disposal system is considered to be located in crystalline rock in a northerly latitude, then the data from Finland or Sweden may be most appropriate to use as proxy reference values.

In the case of site specific safety cases, it is likely that the actual site characterization programme could be structured so as to provide all the necessary data to derive the site

specific reference values (such as compositions of the rocks, soils and sediments, and rates of groundwater flow, denudation etc.), including information from the immediate locality of the site and in the wider regional area. In this case, the information in this report may have less obvious utility but, nonetheless, can provide additional context to the safety case by providing information on the wider range of natural concentrations and fluxes that can occur in nature. This may be useful to show, for example, that the natural concentrations and fluxes expected to occur at a particular disposal system site are no different to those that may be found in other countries or regions and, thus, no unexpectedly different performance of the disposal system should be expected.

Although not a specific objective of the CRP, a number of participants have used the geochemical data they have collected to test the concept of using environmental concentrations and fluxes as complementary safety indicators. These approaches are summarized in Table 36.

**TABLE 36. COMPARISONS OF NATURAL SYSTEMS WITH REPOSITORY SYSTEMS BY PARTICIPANT COUNTRIES**

<b>Country</b>	<b>Approach</b>
China (Appendix C)	Comparison of calculated 3H and 14C releases from the Beilong low and intermediate level waste (L/ILW) disposal facility with concentrations of these nuclides in surface waters and WHO drinking water standards, and comparison of radionuclide release rates from the near-field with ALI units for the same nuclides. The analysis indicated that the waste derived 3H and 14C concentrations in surface waters would be higher than the natural concentrations of these elements but are below the WHO limits, and that the radionuclide release rates from the near-field are, in most cases, below the respective ALIs.
Cuba (Appendix D)	Comparison of total nuclide activity in the proposed Cuban low level waste (LLW) disposal facility with the natural radioactivity in the repository equivalent rock volume (RERV), and comparison of the calculated disposal facility release rates with the natural discharge from the host granodiorite rock for the U chain, the Th chain and 40K. The analysis shows that although the repository contains a much higher radionuclide inventory than the rock, the repository release rates to the accessible environment are many orders of magnitude lower than from the rock.

TABLE 36. COMPARISONS OF NATURAL SYSTEMS WITH REPOSITORY SYSTEMS BY PARTICIPANT COUNTRIES (contd.).

Country	Approach
Finland (Appendix F)	Determination of natural U inventories and fluxes within the bedrock and to the surface for the Palmottu U orebody site, and determination of natural U fluxes at different depths at four potential spent fuel repository sites. Comparison of these with current drinking water standards as well as estimated U concentrations in surface and groundwaters arising from a spent fuel disposal system with breached canisters. The analyses show that the natural groundwaters in some locations in Finland exceed the drinking water standard for U and that these concentrations are 3 to 6 orders of magnitude higher than the estimated releases from the spent fuel disposal system.
Japan (Appendix G)	Comparison of calculated repository radionuclide releases and fluxes in and between different rock and water compartments, and comparison with measurements of natural $4N+2$ decay series radionuclide abundances. Including comparisons for river waters and groundwaters for the normal disposal system evolution scenario. The analyses show that for river water, the waste derived radionuclide concentrations are always lower than the natural component, and in the groundwater the waste derived radionuclide concentrations may reach the natural concentration because both are limited by the same U solubility limit.
U.K. (Appendix I)	Determination of concentrations and fluxes (U, Th, K, Cs, Sn and natural series radionuclides) in and between different rock and water compartments at 4 different geological systems, including a granite, chalk aquifer, clay and geothermal spring. Comparison of these with calculated release rates from a hypothetical disposal system for intermediate level radioactive waste. The analysis shows that the waste derived fluxes to the surface are a small fraction of those that occur from the natural rocks and groundwaters.

Although most of the analyses summarized in the table above are of a simple, scoping nature (and none are fully site specific comparisons), some do involve extensive calculations and numerical analysis, and at least one (Japan) has been used to support a published safety assessment [10]. Two fundamental observations can be made from these analyses:

- it is clearly possible to make sensible comparisons between the environmental concentrations and fluxes of naturally occurring chemical species and calculated releases from a disposal system; and
- in most cases, the concentrations and fluxes of waste derived species are substantially lower than the equivalent natural concentrations and fluxes.

These observations require further interpretation and must await more fully site specific comparisons and calculations before final conclusions could be reached but they do indicate the utility of the CRP data and the feasibility of applying environmental concentrations and fluxes as complementary safety indicators to a safety case.



#### 4.3. FINAL CONCLUSIONS AND RECOMMENDATIONS

This CRP has partially met most of the objectives that were set by the IAEA, in so far as geochemical data on elemental and radionuclide concentrations and fluxes have been collated in a form that facilitates their use as reference values for comparison with safety indicators based on environmental concentration and flux.

Additional work undertaken within the CRP has demonstrated that the geochemical data acquired by the CRP have real utility and that safety indicators based on environmental concentration and flux can readily be applied to a safety case.

On this basis, it is possible to conclude that this CRP has moved forward the debate on complementary safety indicators to the point that they need no longer be considered as hypothetical but as a practicable component of a safety case. It should be recognized, however, that the work of this CRP ran in parallel to that of the Working Group on Principles and Criteria as published in [2]. As such, it is to be recommended that the suggestions of the Working Group from [2] should be combined with the data acquired in this CRP, so as further to test the utility of the CRP data and to assess the suggested approach set out in [2] for including complementary safety indicators in a safety case. A further CRP may prove to be an appropriate mechanism for doing this and another international team could be assembled for the task. It is recommended that, if such a further project is considered, that it should involve both safety assessors and geochemists from both disposal organizations and regulatory bodies, as well as people who understand stakeholder communication issues, since one of the suggested advantages of complementary safety indicators is their utility in presenting complex themes.

A number of specific recommendations to develop the complementary safety indicators approach further have been identified during the course of this CRP. These could be used to form the scope of a further international study. They are:

- It would be useful to define a set of ‘standard’ comparisons to be made between natural systems and disposal systems. The suggestions set out in Section 1.4.1 could be used as a starting point, and may be tested for utility and feasibility;
- Safety assessment methodologies would need to be adapted so as to generate appropriate intermediate outputs for comparison with the natural concentration and flux data used to define reference values. The European Commission’s recent project entitled “Safety and Performance Indicators (SPIN)” could provide a starting point for determining what are realistic expectations from the safety assessments;
- The use of complementary safety indicators could benefit from being included within other ongoing or proposed international programmes such as the Coordinated Research Project on Application of Safety Assessment Methodologies for Near-Surface Waste Disposal Facilities (ASAM), so that they can be considered fully in the development of recommendations for future safety cases; and
- Further attempts may be made to define limits to the concentrations of natural species that may be considered to be safe. This would require the involvement of health specialists with access to appropriate epidemiological data.



## APPENDIX A

### ARGENTINA

R. Ferreyra, Comisión Nacional de Energía Atómica

#### Abstract

The airborne geophysical surveys developed in Argentina are described. They have got more than 500.000 km<sup>2</sup> acquiring data for U, Th, K and total background activity. Other types of published data are also mentioned (satellite imagery, seismic hazard, climate, soil distributions, etc.). The availability of maps with the abundances of elements analyzed at the country and also at laboratories from Canada is described, as well as data of analysis of several elements at two study areas proposed at the outset of the project. The availability of process rate data and epidemiological data is also explained. Argentina intended fully to participate in the CRP at the outset of the project. Due, however, to external resource constraints imposed on the participating organization (Comisión Nacional de Energía Atómica, CNEA), the level of participation had to be significantly reduced.

Nonetheless, in the first period of the CRP, Argentina undertook to collate existing geological and geochemical information within the country, and began to define potential areas for site specific natural systems safety indicator studies.

#### Mapping data

In Argentina several airborne geophysical surveys have been undertaken, acquiring data for U, Th, K and total background activity. CNEA has acquired survey data from between 1978 and 1981 for an area of 140 000 km<sup>2</sup> (Figure A1). Furthermore, from 1995 up to 1999, the Geological Survey of Argentina have performed several additional surveys covering more than 400 000 km<sup>2</sup>, providing U, Th, K and total background activity. All these data have at present time an important economic value, and it is possible to purchase them, but it is not allowed for publication. It is possible to ask for permission for printing some data of special areas, as in case of a basin not studied for commercial purposes.

There are also many geological maps, scale 1:2 500 000 for all the country, as well as maps for rainfall distribution, population, seismic hazard, climate, average annual temperatures, soil distributions, vegetation types, etc. All the country is covered with satellite imagery, available at scales of 1:500 000–250 000, which can be processed to work at higher scales.

The availability of maps with the abundances of elements is limited to some publications that are being planned, covering certain areas of the country. These regions have analyses performed on samples carried out, in some cases, thirty years ago. These analysis were performed at laboratories from Canada, as part of previous international projects unrelated to radioactive waste management. The data is published for area sizes 1.5 longitude degrees and 1 degree of latitude. At the present there are some ten maps of this kind already published, and several more will be available within the next few years. These geochemical maps show the distribution of more than forty elements.

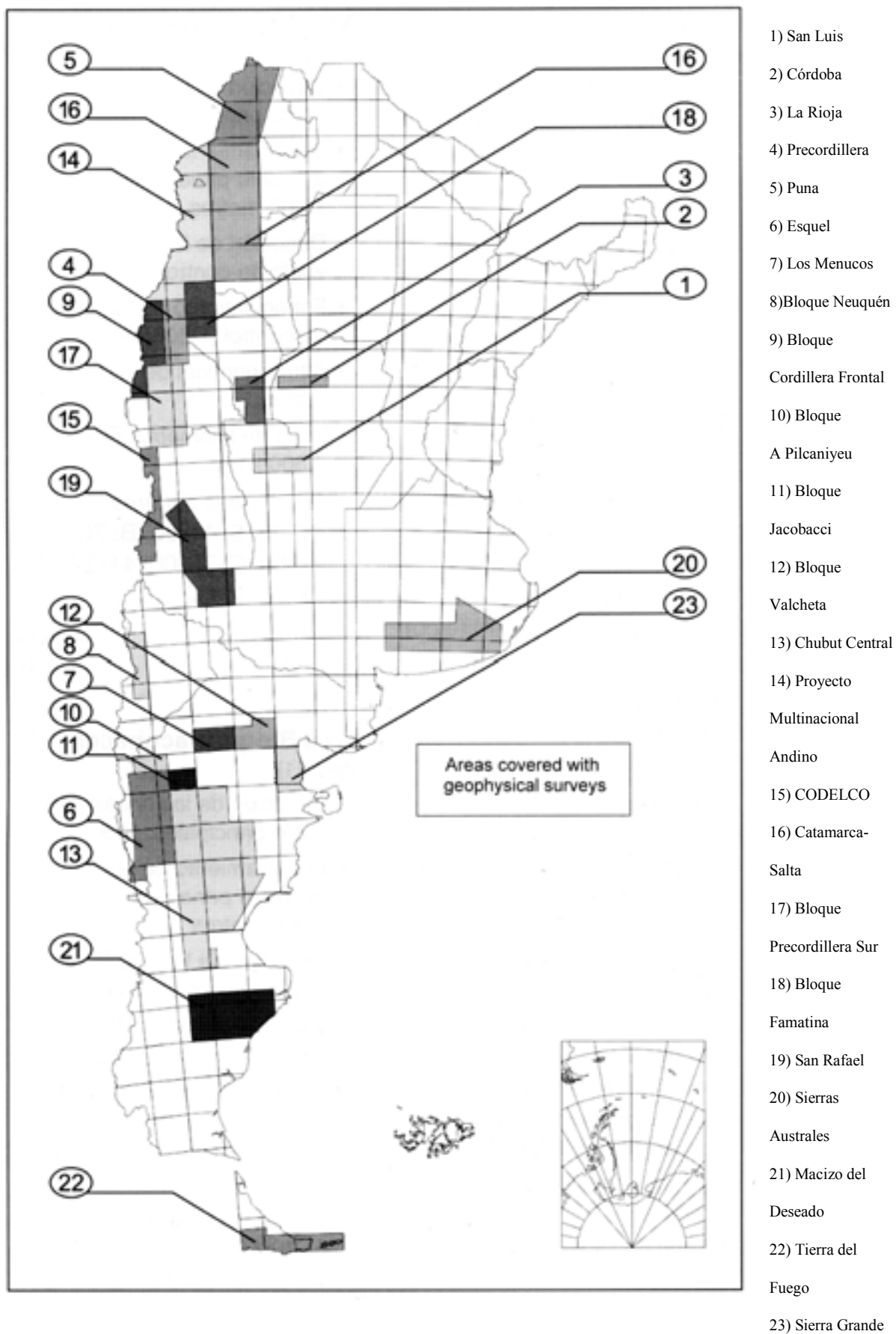


FIG. A1. Areas covered by the geophysical surveys.

## **Study areas**

Some evaluations were performed to define an area for further studies for the CRP. The proposed area included a granitic batholith. The area had been covered with a geophysical survey and data about U, Th and K equivalent values. The batholith has several U anomalies, and one low grade U ore deposit was mined by the 1980s. This body has some structural, petrological and hydrogeochemical studies. It was considered an area where it could be possible to develop some characterization of an area, having enough data already available to focus future work.

A second area was also considered, located in a small basin in volcanoclastic and sedimentary rocks. At the lower part of the basin a U ore deposit was partially mined. At the present time mining work has stopped. There are a lot of analytical data for rocks and waters of the basin, downstream and upstream of the mine. These data could be used to understand the hydrogeochemistry of the waters of the basin. Control samples are systematically taken at the present time, in order to understand any contamination of the water of the system.

Both areas were considered taking account of that they have an interesting amount of information.

## **Process rate data**

The availability of process rate data is very variable. Some data for big basins are obtainable. No maps for erosion or denudation were obtained. The river flow of all rivers of the country is available, including sediment loads.

## **Epidemiological data**

Some projects for the elemental distribution of As and Cr in underground waters are being carried out within Argentina, unrelated to radioactive waste management. According to the data from the country Health Department, there are no maps of epidemiologic data which can be used in comparison with hydrogeochemical data. There are data of As, Se, Cr in waters of some large areas where the contamination related to natural and anthropogenic processes is known.



## **APPENDIX B**

### **BRAZIL**

F. Lemos, Comissão Nacional de Energia Nuclear

#### **Abstract**

Brazil has a very large and diversified territory. During the last 20 years innumerable mineral deposits have been discovered. However, most of the research and prospections, regarding these mineral deposits, is related to their commercial use and, therefore, these data may need further analysis and interpretation.

Literature data are presented. Other publicly available data have, however, been acquired and compiled that relate to abundance of U, Th, K, Cu and Sn, from a number of mineralized regions. Other relevant information has been acquired on rock type, mineralogy, soil type and some climate data. These data are being used to determine elemental concentration ranges and fluxes in rivers crossing the mineralized regions.

An approach based on fuzzy logic for data analysis and interpretation is presented. Fuzzy logic offers a potential method for answering difficult questions such as ‘What is the country average concentration of a particular element?’ and ‘What is the degree of impact on the environment?’ when the available data comprise only information collected from specific sites without good geographical coverage. It is thought that not only can the approach be useful for technical studies but also for public communication because it allows the use of language expressions for description of site conditions.

#### **Introduction**

Brazil has a very large and diverse territory. During the last 20 years numerous mineral deposits have been discovered. However, most of the research and prospections, regarding these mineral deposits, is related to their commercial use and, therefore, these data may not have the right format needed by this CRP.

Each sub-section corresponds to an element and is sub-divided by the mineral deposits.

For each deposit, there is a brief description of some local geology, pedology and other information related to its characterization.

#### **Mineral deposits in Brazil**

In South America there are three large tectonic regions according to their origin, age and structural evolution. The South America platform is one of them and Brazil is entirely contained in this platform. It has a very complex history and it was originated in the Archean. Its cycle ended at the beginning of the Paleozoic. Its structure is based on metamorphic rocks of amphibolite facies and granitoids of Archean age. This basement outcrops in three Cratons: Central Brazil, Atlantic and Guyana.

Sedimentary and volcanic overlays were developed over this platform and spread over three major basins: Amazon, Parnaíba and Paraná. These basins contain very thick sedimentary rocks of the Paleozoic and Mesozoic ages. Some important mineral deposits are associated with the metamorphic, ultrametamorphic, and magmatic rocks. These are:

Iron: State of Minas Gerais.

Mn: states of Minas Gerais, Amapá and Bahia.

Au: Morro do Vellho mine (State of Minas Gerais).

There are some Cu, Ni and Cr deposits associated with ultrabasic rocks in the states of Bahia and Goiás. Uranium is associated with the magmatism that begun during Mesozoic, near the city of Poços de Caldas. Other geologic formation along with important mineral concentration are shown in the following Figures B1 through B6. [B2].

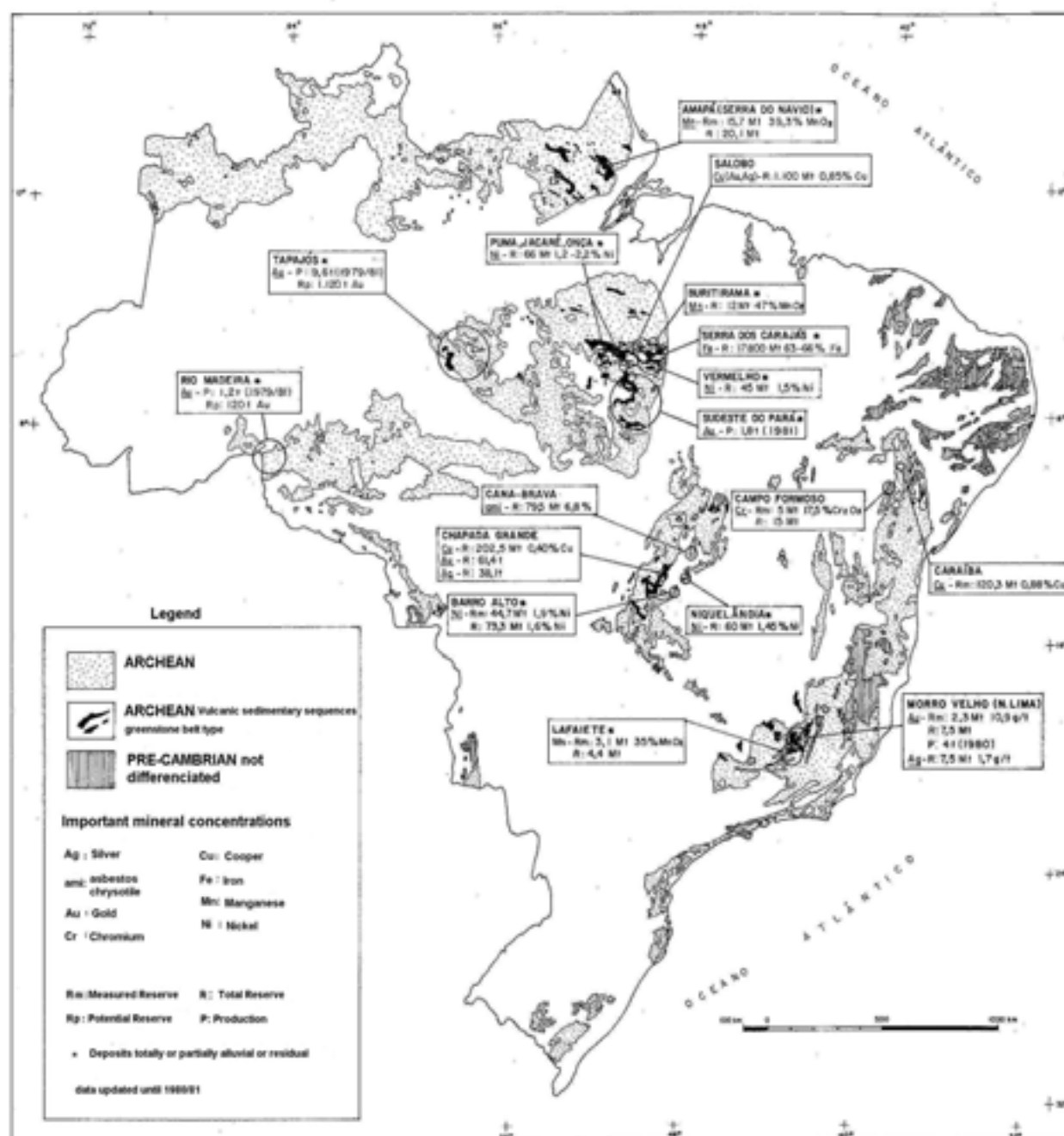


FIG. B1. Distribution of Archean and Pre-Cambrian units in Brazil along with some related mineral concentrations [B2].



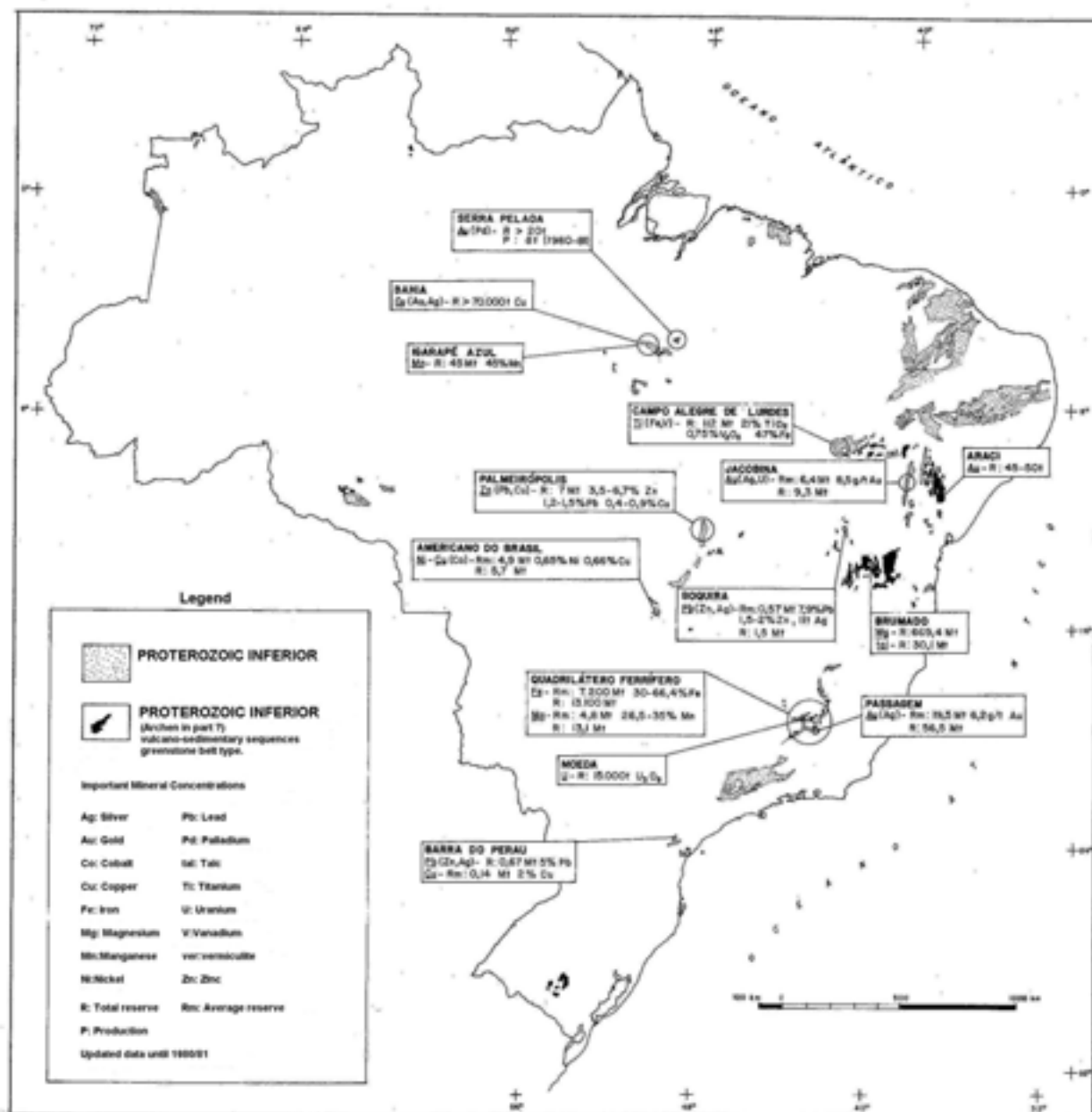


FIG. B2. Distribution in Brazil of Proterozoic Inferior along with some related mineral concentration [B2].

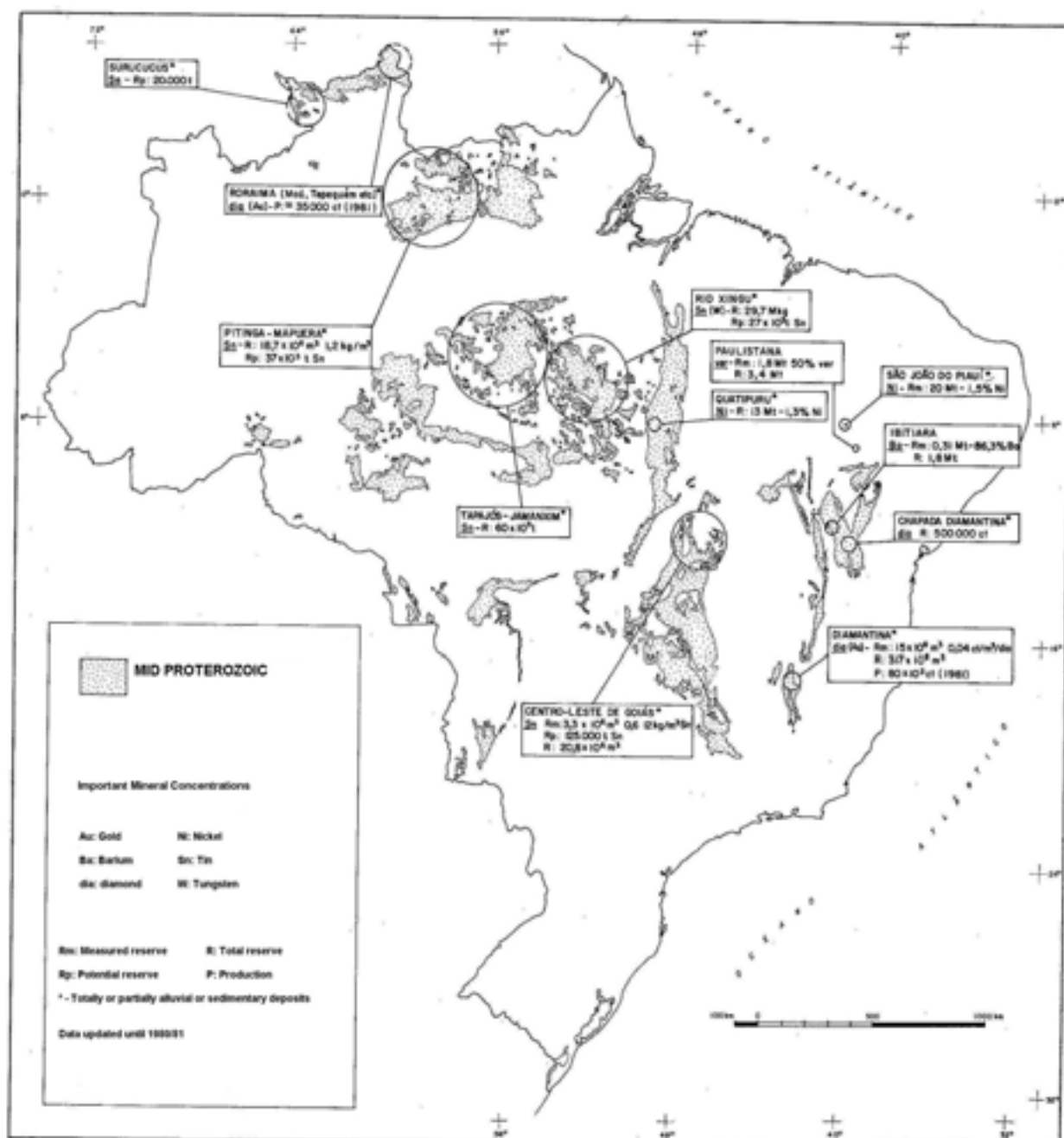


FIG. B3. Distribution in Brazil of Middle Proterozoic units along with some related mineral concentrations [B2].

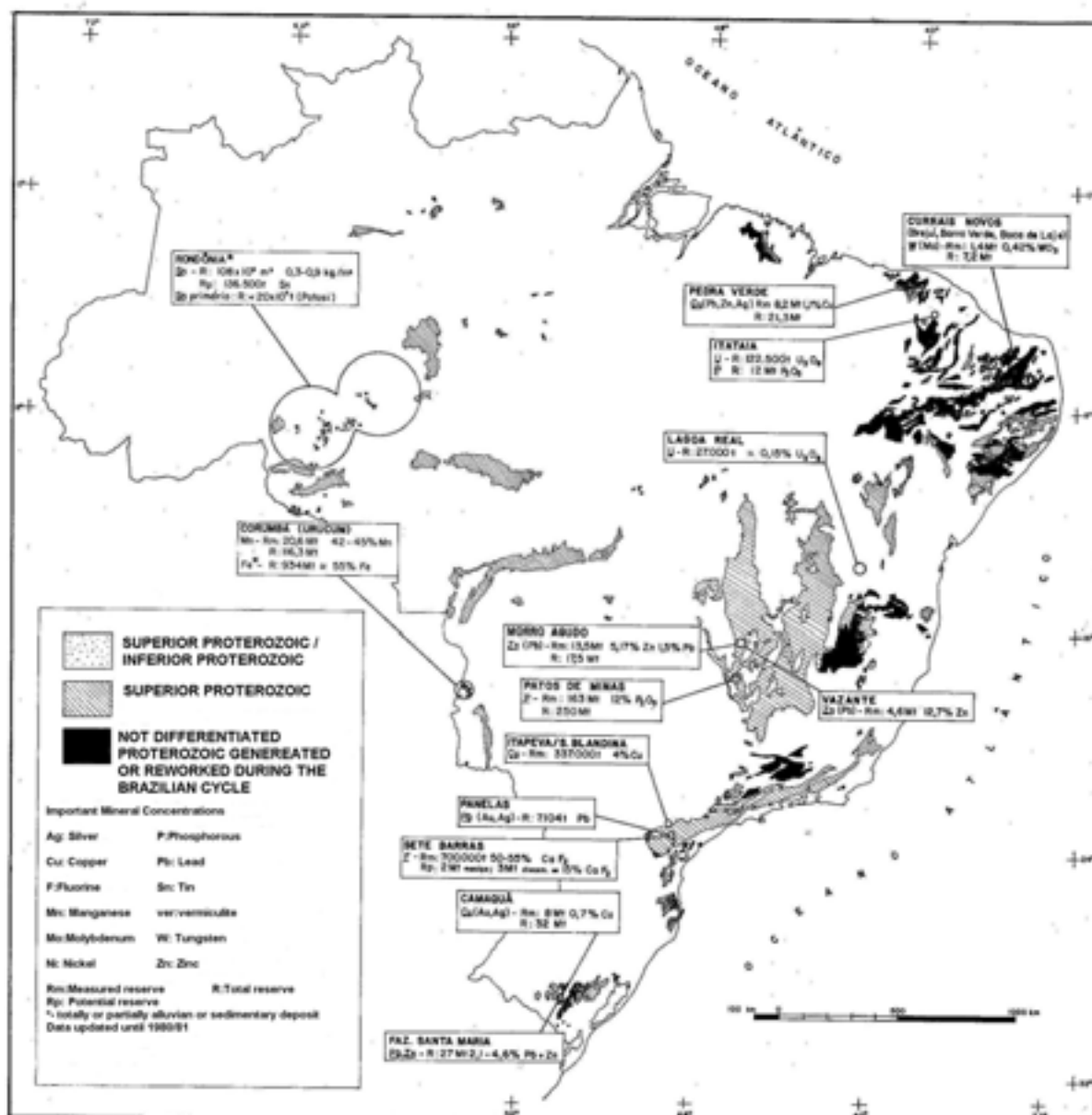


FIG. B4. Distribution of units from non-differentiated Proterozoic, generated or reworked during the Brazilian cycle, from Superior Proterozoic and from the limit Superior Proterozoic-Inferior Paleozoic (Cambrian-Ordovician), along with some important mineral concentrations [B2].

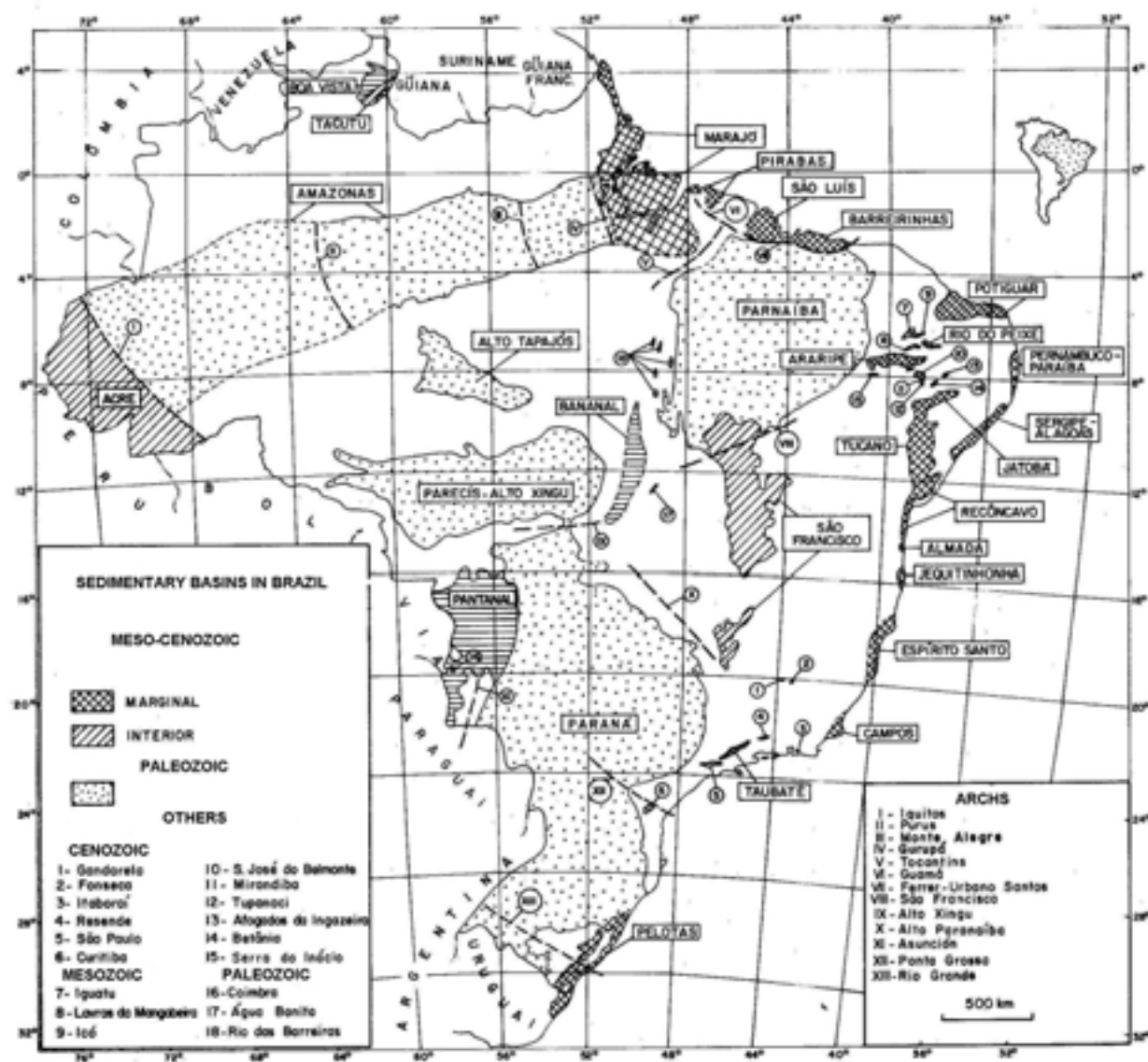


FIG. B5. Sedimentary Basins in Brazil [B2].

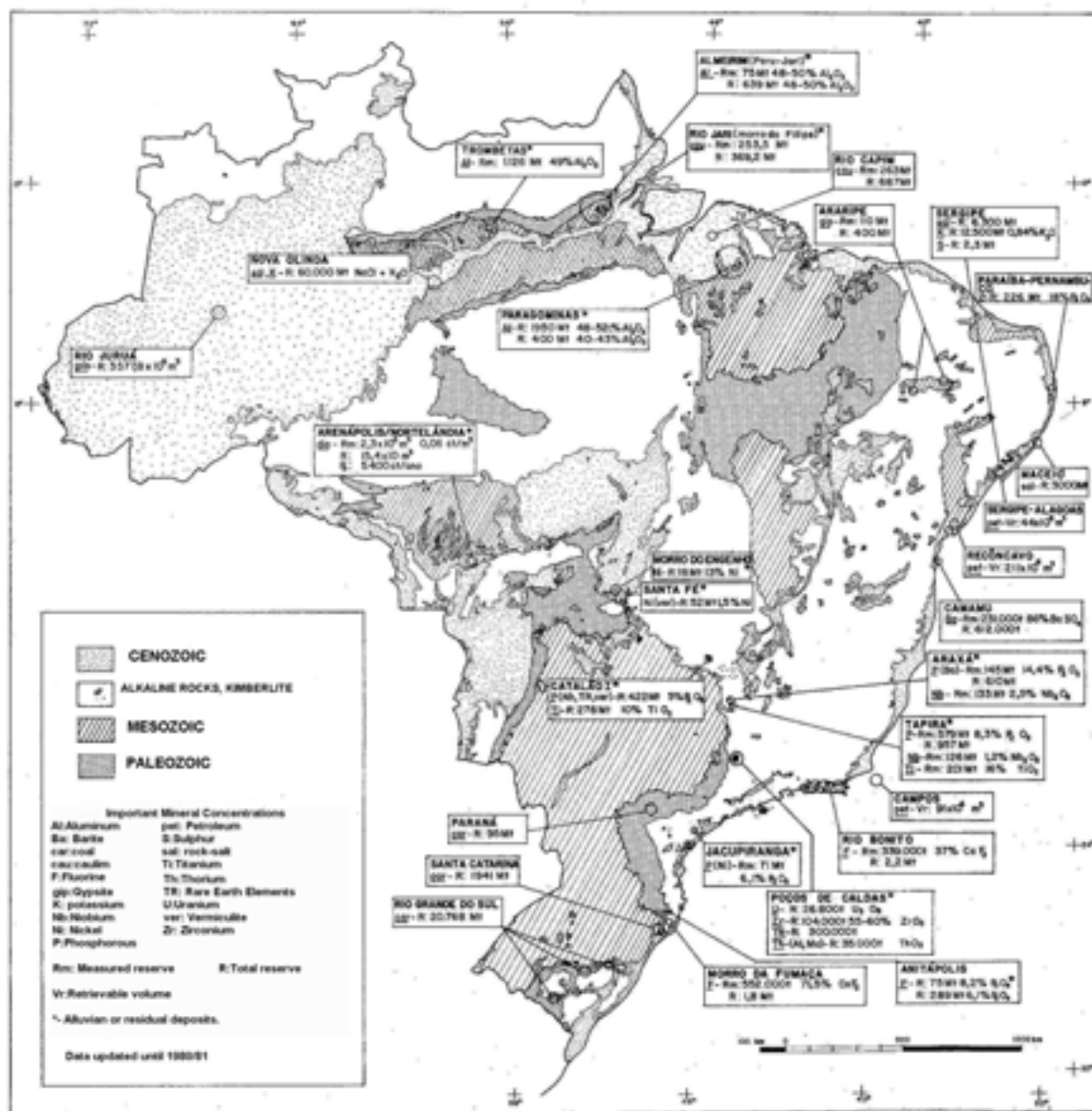


FIG. B6. Distribution of units from Paleozoic (post-Ordovician), Mesozoic and Cenozoic along with related mineral concentrations [B2].

## **Copper**

### **Camaquã Deposit**

Location: Caçapava do Sul in the state of Rio Grande do Sul

Coordinates: 30°7S and 52°24W

Climate: temperate, pluviometric rate: 1500 mm/a and annual average temperature: 17–18°C

Topography: moderate altitudes: 340–1200 m above sea level. Dendritic drainage pattern

Regional Geology: The Camaquã mines lie in sedimentary rocks of the Bom Jardim Group (Proterozoic to Mid-Paleozoic age).

The copper mineralization occurs as disseminated-type ore in conglomerated and sandstone and as vein type ore. The disseminated sulfides vary from isolated fine-grained to massive concentrations that constitute the rocks cement. The vein-type ore, lies along NW faults, and is interpreted as remobilization of the disseminated sulfides of diagenic origin. The total reserves are about 31 Mt, with an average content of 1.06% Cu. There are four lithologic units:

- base: fine to medium size sand, detrital quartz fragments, feldspar and rocks.
- inferior conglomerate: 12 m thick, sandy matrix.
- intermediate: 20–25 m thick, fine and medium sand grains, locally conglomerates.
- superior conglomerate: 200 m thick, rough to medium conglomerates, clastics with various sizes ( up to 1.5 m diameter).

The lithologic composition of clastics is very variable, mainly comprised of granites, quartz, feldspar, and sediment rock fragments.

### **Chapada Deposit**

Location: central Goiás state and central Brazil

Climate: humid tropical, with two distinguished seasons: dry (May to September) and rainy (October to April).

Average annual temperature is 24°C, and average pluviometric annual rate is 21.9 mm (dry season) and 210 mm (rainy season).

Vegetation: grass and bushes.

Hydrography : Low density. Dois Bois River at south of the region east to west direction.

Topography: Over archean rocks, peneplain partially lateritic (with a summit at 400 m above sea level).

Geology: Host rocks are volcano-sedimentary of the Mara Rosa sequence of archean age.  
Lithology: schists, from clay and silt origin.

Mineralization: there are sulfates, pyrite and calcopirit disseminated in feldspar biotit-shists with silicate matrix of fine and medium granulation. Copper concentration: 0.4–1.25%. Typically this copper is associated to Gold and Silver.

### **Salobo Deposit**

Location: Southeast of Pará State

Geology: Salobo-Pojuca sequence comprised of, from bottom to top: Quartzite, gnaisses, ferrito zone and shists. This ore is considered to be formed under volcano-sedimentary environment.

The deposits are associated to the denominated Salobo-Pojuca sequence whose relationships with the granite-gneissic basement (Xingu Complex) are not well defined yet.

This sequence is made up, from base to top, of quartzites, upper gneisses, banded iron formation (oxide facies) schists (iron formation belonging to oxide and silicate facies) that are the host rocks of the mineralizatin, and lower gneisses metamorphosed to amphibolite facies.

The copper mineralization occurs as sulfide and alteration ore mineral. The first type is made up of bornite-chalcocite-minor chalcopyrite association occurring in a disseminated, massive sulfide and fractured filling form, mainly confined within magnetite schists, being associated with subordinate molybdenite, gold and silver. The alteration ore mineral is associated with iron hydroxide, micas and clay minerals. The total reserves are divided in sulfide and alteration ores. The reserve of sulfides is about 1.0 billion tons at 0.84% Cu in average. The reserve of alteration ore is of about 106 Mt with 0.75% Cu. Mineralization: association of  $\text{Cu}_5\text{FeS}_4$  –  $\text{CuFeS}_2$  –  $\text{Cu}_2\text{S}$  in a disseminated form.

### **Pedra Verde Deposit**

Location: northeast of Ceará State. 3° 31' 30" S and 41° 08'00" W

Geology: Graben type structure controlled by a system of regional faults, in layed basement gnaisses and quatzites and fulfilled with a sequence of carbonaceous metapelitic rocks and areanaceous intercalations of the proterozoic Mambrina Formation. These units are overlaid discordantly by cambro-ortovician conglomerates (Ubari Formation) and siluro-devonian sandstones (Sera Grande Formation).

The mineralization is disseminated and restricted to an upper phyllitic unit of the Mambira Unit (Pedra Verde Member), just bellow to an enriched zone of iron oxides ("red zone") related to the basal level of the upper conglomerate unit. It follows a well defined zoning represented by calcosine, bornite, chalcopyrite, from hanging wall to a foot wall. The thickness and grade of the mineralized zone are very irregular. The reserve has an average of 1.0% Cu and 10g/ton silver. A silver mineralization occurs subordinately on the foot wall of the copper ore. The deposit is considered to be originated by supergenic enrichment of singenetic copper in a paleo-regolith.

Mineralogical characteristics: disseminated type and restricted to phyllites, underneath a red zone (iron oxide). Average concentration of copper is 0.65%. There is also Silver at 10g/ton.

## **Arapiraca Deposit**

Location: state of Alagoas, north of Brazil

Geology: Related to a mafic-ultramafic metamorphosed rock, with 400 m depth.

Mineralogical characteristics: The primary phase of mineralization characterized by paragenesis oxides-sulfates from magnetites + ilmenites + calcopyrites + bornites + pentlandites +/- Gold, and some less abundant minerals such as : melonite, vioralite, and Nickel teluretos, Bismuth, Lead and Gold.

Concentration : 0.8% Cu and 0.3 ppm Gold in average. In some cases there are associations with Nickel, Vanadium, Cobalt, Chromium, Zinc, Tellurium, Silver, and traces of Platinum and Palladium.

## **Caraiba And Curaçá Deposits**

Location: Bahia State, between 9°30' and 10°00' S and 39°45' and 40° 00' W.

Geology: Igneous origin occurring as mafic –ultramafic sills.

Mineralogical characteristics: Basically comprised of calcopyrite and bornite  
Concentrations: 0.85% in average.

The copper district of Curaçá is situated in the high grade metamorphic terrain of eastern Bahia state which is a late Archean or Early Proterozoic mobile belt of N-S trending. At least three hundred mafic-ultramafic sills derived from differentiated tholeiitic magma have been identified as intruded pre-tectonic intrusions in a supracrustal pile.

The supracrustal sequence is interbedded with tonalitic-granodiorite sheets during two tectonic-magmatic events in thrust –underthrusting regime producing a structural thickening. A third event was a strong E-W compressive stress field resulting in a up-right folding phase with tight to open D3 folds. The mafic-ultramafic bodies are showing internal stratigraphic layering from hypersthene in the base to norite-gabbro in the top. The ore is closely related to the sills stratigraphy, mainly in the hypersthene rocks as well to hinge zones of D2 folds. The remaining geologic reserves in the district amount to 120 Mt grading 1% Cu of which 92.5 Mt are in Cariba deposit.

## **Serrote Da Laje Deposit**

Location: Alagoas State, north of Brazil

Characteristics: This is a small hill sustained by two thick massive magnetite layers enveloped in weathered magnetite-biotite schists.

The massive magnetite rock is rich in Cr<sub>2</sub>O<sub>3</sub> (1.85%), TiO<sub>2</sub> (1.16%), Zn (0.5%) and Cu (0.03%) while additional chemical and petrographic data pointed out that the magnetite horizons were of segregational nature and related to hyperstenites retro-metamorphosed to magnetite-bearing biotite schists.

The complex presents the same deformational and metamorphic history of its enclosing metasedimentary rocks, mainly sillimanite-feldspar-quartz gneisses, porphyroblastic biotite-quartz-cordierite-garnet gneisses and anhydrite-bearing calc-silicate rocks, and shows a



disseminated copper mineralization hosted in the hyperstenites and their metamorphic counterparts (magnetite-biotite schists), in the norites and in the massive magnetite layers.

This primary mineralization can be described as an oxide-sulfide paragenesis made-up mainly by magnetite + ilmenite + hercynite + borinite + pentlandite +/- gold. The concentration of copper is estimated to be 0.8% and 0.3 ppm of gold in average. This deposit is very similar to the Caraiba in Bahia State. Both deposits are hosted by mafic-ultramafic complexes with the hyperstenite-gabbro compositional range, both have the same lithological controls of their similar mineralizations and both have undergone a very similar metamorphic and deformational history. Also, the ores of both deposits have the same remarkably high Cu:Ni ratio, only equal to that found in the ores of O'okiep copper district in South Africa.

Whereas the mineralized "noritoids" of O'okiep have an uncertain magma chemistry and are discordant, postdating the deformation of their wall-rocks, the hosts of the Serrote da Laje and Caraiba deposits, with tholeiitic nature, are pre-tectonic and intruded the sedimentary pile prior to deformation in a distensional tectonic setting

## **Tin**

### **Pitinga Deposit**

Location: Amazon State

Characteristics: The oldest rocks of the region are granitoids of a granodiorite composition, named Ademilto Água Branca. The main stratigraphic unit in the region is the Uatumã, which is comprised of volcanic and pyroclastic rocks of the group Irecoumé and granites of the intrusive suite Mapuera. Over the former unit, in a erosive discordance, there are sedimentary rocks of the Urupi formation. These sediments show immature character and are comprised of polymitic conglomerate, feldspatic sandstone, siltstones and argillites.

The ores: The Pitinga deposits are genetically related to two granitic bodies, called Água Branca and Madeira which are 1 Km apart from each other. They are comagmatic. Both seem to be one batolite, linked at great depth. The facies are typical alkali-feldspar granite, comprised mainly of potassic feldspar and quartz and biotite as the main accessory. The placers are of small to medium size width 40-600 m and depth from 2 to 3 m. The alluvium from the granite present a great amount of cassiterite associated with zircon, columbite-tantalite, pyrochlore and xenotime. The mineralization is genetically related with the Água Boa and Madeira granites, which intrude Early Protozoic Platform covers comprising volcanic and pyroclastic rocks of the Iricoume Group and sedimentary rocks and tuffs of the Urupi Formation .

Several phases of magma injection and crystallization as well as late and post-magmatic alteration have been distinguished . In the Água Boa granite the characteristic mineral association is of the greisen vein type: quartz + Li-bearing mica + topaz + cassiterite + columbite - tantalite. In the Madeira granite the ore is cassiterite, zircon, columbite – tantalite, pyrochlore and xenotime, with strong Hf, REE, Pb and Bi enrichment disseminated within the central albitized granite (apogranite) facies. Rich alluvial fan concentrations of these minerals are near their source rocks, commonly overlying directly the granites.

## **Central Rondonia State Deposit**

Location: Rondonia State

Characteristics: Two tin bearing quaternary sedimentary sequences. They are mainly based on erosional unconformities as a result of climate changes. The sequence I is tentatively correlated to Middle Pleistocene age and related to the Illinoian glaciation while sequence II of Upper Pleistocene-Holocene age is related to the Wisconsinian glaciation. The sedimentation for this two unites happened under semi-arid climate conditions through extensive mass movement. This transportation allowed enrichment of cassiterite to form fanglomeratic placer deposits.

Sequence I was followed by an important phase of lateritisation and braided stream entrenchment of the previous pediments with development of residual-elutriation and alluvial placers correlated to the interglacial Sangamon stage. The Flandrian transgression (Pleistocene-Holocene) was followed by a deep meander incision with further placer reworking and redeposition forming poor tin grade placers of little economic importance.

There are four main districts: São Lourenço, Oriente Novo, Monte Negro and Cachoeirinho. Concentrations vary from 200 g Sn/m<sup>3</sup> to 8 Kg Sn/m<sup>3</sup>, depending on the deposit.

### **Deposits of Cassiterite In Santa Barbara, Jacunda And Alto Candeias, Rondonia State**

The different types of occurrences are defined as mines of primary origin and as mines of secondary origin.

Eluvio-colluvials; 14 de Abril deposit. Also bearing cassiterite directly associated with the west edge of the granitic intrusion Novo Mundo. Concentration : 300 gSn/ m<sup>3</sup>.

Serra da Onça deposit : dispersion of cassiterite from greisens. Concentration : 300 g Sn/ m<sup>3</sup>.

Serra do Cacimba deposit : a concentration of cassiterite scattered on elluvial and colluvials over greisen veins. Concentration: 300 gSn/ m<sup>3</sup>. Colluvio-alluvials; Alto Taboquinha deposit

### **São Pedro Do Irii Deposit**

Location: Pará State, north of Brazil

Characteristics: The cassiterite deposit is associated with anarogenic granites of the Amazon Craton. The basal is comprised of granitic and granodiorite rocks (from the Xingu Complex) that occupy the north and west portion of the area. The Xingu Complex is overlain by the Uatumã Super group, represented by tuffs, dactites and riolites. Some works show the association of the cassiterite mineralization with the São Pedro do Irii granite, that are considered the responsible for the mineralization of cassiterite through quartz and quartz-greisens veins, formed by post- magmatic hydrothermal activities.

The erosion of this primary mineralized source and the transport of the desegregated matters, allowed the formation of the concentration of cassiterite in colluvials and alluvials, and therefore building secondary deposits. The erosion processes started the transport of these secondary deposits causing rare-concentration in the current creeks. Therefore, there are three types of deposits: (i) Recent alluvials, (ii) Alluvial terraces, (iii) Colluvials. Concentrations: the average concentration is 700g Sn/m<sup>3</sup>. The cassiterite concentrations occurs in recent

alluvials, alluvial terraces and colluvial deposits, derived from greisen and greisenized granites of the so called “São Pedro do Iriiri Massif”.

The regional rock association belongs to the Amazon Craton and encompasses the following geological units: Xingu Complex, Uatumã Supergroup, Bala Massif, Triunfo Sandstones, Basic Intrusives, São Pedro do Iriiri Massif, and Recent Deposits.

### **Pedra Branca Deposit**

Location: Goiás State

Characteristics: This tin deposit is hosted both in the pluton and in the country rocks in different rock associations. Placer deposits of cassiterite occur near or at the base of the Pedra Branca range. Tin deposits are distributed in four main zones, associated with albitized and greisenized granites, making the “endogranitic orebodies” characteristic of metasomatic granitic cupolas.

The Pedra Branca Granite Massif carries an important tin deposit. It is one of the granitic bodies with major metallogenetic potential in the Goiás tin Province. The tin ore (cassiterite) is hosted both in the pluton and in the country rocks in different rock associations. Placer deposits of cassiterite occur near at the base of the Pedra Branca range.

Tin deposits are distributed in four main zones, associated with albitized and greisenized granites and greisen, making the “endogranitic ore bodies”, characteristic of metasomatic granitic cupolas.

Cassiterite also occurs in greisen and quartz veins hosted in country rocks, forming the “exogranitic orebodies”. Fluorite appears as disseminations in granitic rocks and greisen and may form important concentrations in greisenized zones like quartz-fluorite veins. Cassiterite – sulfide associations are common in the S-SW part of the massif; sphalerite, followed by chalcopyrite and pyrite, are the dominant sulfide minerals.

The main control of the mineralization is represented by joint sets probably developed during the emplacement of the pluton. Late tectonic events deformed primitive tin orebodies.

Tin deposits of the Pedra Branca Granite are very similar to those of the Serra Branca Granite, in Goiás. The known reserves of primary tin ore are in the order of 15000 tons of tin-metal, only in the Faixa Placa deposit.

Placer deposits are in the order of 1 000 000 m<sup>3</sup> with a mean content of 550 g Sn/m<sup>3</sup>.

### **Potassium**

#### **Fazendinha Deposit**

Location: Mid Amazon Basin, by the margin of the Madeira river

Characteristics: The Amazon evaporitic sequence has regressive characteristics. From this sequence one formation has reached the conditions for the precipitation of the more soluble salts, K<sup>+</sup> and Mg<sup>++</sup>. The ore is divided into three distinct intervals (i) inferior: white sylvinite, (ii) middle: sulfate zone, (iii) superior: red sylvinite.

The deposition sequence suggests that it might have been generated by the gradual increase of in and constant concentration of existing brine in the hypersalt lake of Fazendinha. The sulfated interval indicates an invasion of brines rich in  $\text{Ca}^{++}$  and  $\text{SO}_4$  in the sylvinitic basin with the resultant co-precipitation of chlorides and sulfates of  $\text{K}^+$ ,  $\text{Mg}^{++}$ ,  $\text{Ca}^{++}$ . As for the red sylvinite, a secondary genesis it is assumed by means of total leaching of  $\text{Mg}^{++}$  of carnallites originally deposited. The middle area is made up of an assorted zone of sulfates of  $\text{K}^+$ ,  $\text{Ca}^{++}$  and  $\text{Mg}^{++}$ , of relatively rare occurrence. Total deposit area is approximately  $131 \text{ km}^2$  and mineral is found in depths from 980 to 1130 m. The thickness ranging from 0.8 to 4.4m and content is between 14.3% and 38.7% KCl, with weight average of 28.8%.

The Amazon evaporitic sequence, of regressive characteristics, is made up of eleven cycles, of which only Cycle VII (Nova Olinda Formation) reached conditions of considerable restriction., making it possible the precipitation of the more soluble of  $\text{K}^+$  and  $\text{Mg}^{++}$ . The ore has been subdivided into three distinct intervals, informally referred to as inferior (white sylvinite), middle (sulfate zone) and superior (red sylvinite). The deposition sequence and macroscopic characteristics presented by inferior sylvinite suggest that it might be an ore of primary origin, generated by the gradual increase in and constant concentration of existing brine in the hypersalty lake of Fazendinha.

The sulfated interval indicates an invasion of brines rich in  $\text{Ca}^+$  and  $\text{SO}_4$  in the sylvinite basin, with the resultant co-precipitation of chlorides and sulfates of  $\text{K}^+$ ,  $\text{Mg}^{++}$ ,  $\text{Ca}^{++}$ . As for the red sylvinite, a secondary genesis is assumed by means of total leaching of  $\text{Mg}^{++}$  of carnallites originally deposited (sylvinization of carnallites).

The middle area is made up of an assorted zone of sulfates of  $\text{K}^+$ ,  $\text{Ca}^{++}$  and  $\text{Mg}^{++}$ , of relatively rare occurrence and of high commercial value. This is the first occurrence of these minerals found in Brazil. The total deposit area is approximately  $131 \text{ km}^2$ , and the thickness ranges from 0.8 m to 4.4 m (average of 2.7 m) and the content ranges from 14.3% to 38.7 % KCl, with a weighted average of 28.8%.

### **Taquari / Vassouras Deposits**

Location : State of Sergipe north of Brazil

Characteristics: Its origin is associated with the rift phase of the South Atlantic opening. The precipitation conditions during the VII evaporitic cycle of the Taquari-Vassouras deposit were such that several relatively thick sylvinite bodies were formed with small carnallite associated.

The evaporitic sequence of the Cretaceous Basin of Sergipe has its origin associated with the rift phase of the South Atlantic opening. This sequence is distributed in many sub-basins, from which the Taquaril-Vassouras and Santa Rosa de Lima are of economical potential because of its potassium (mainly sylvinite) and magnesium salt contents.

This complex is located in the northeastern portion of the Sergipe State, between the Carmopolis and Siririzinho petroleum fields, near the coast and energetic and hydric resources, about 40 km away from Aracaju, capital of Sergipe.

The evaporitic sequence of the Sergipe Basin is divided into nine secondary cycles, the first six cycles are represented by a section of soluble salts, with zones of halite, carnallite and tachydrate, the last ones being composed, from bottom to top, by a section of carnallite, sylvinites and halites, underlain by anhydrites, halites, limestones and shales.

These evaporites generally show structural deformations related to dissolution, slippage and flow processes, partly associated to epirogenetic tectonic reactivations. The upper part of the evaporitic sequences is missing in many parts of almost all sub-basins, because of erosion soluble salts.

The precipitation conditions during the VII evaporite cycle of the Taquari-Vassouras deposit were such that several relatively thick sylvenite (KCl + NaCl) bodies were formed, with small carnallite layers associated. These bodies were grouped in two sets of layers, informally denominated Superior Basal Sylvinitic and Inferior Basal Sylvinitic, generally separated by an halite layer with thickness varying between 3 and 6 m.

The inferior basal Sylvinitic lays directly over tachydrite rocks ( $\text{CaCl}_2$ , 2  $\text{MgCl}_2$ , 12  $\text{H}_2\text{O}$ ) the mechanical properties of which present low resistance to compression and high plasticity, besides being highly hygroscopic. The Superior Basal Sylvinitic is approximately 26 Km<sup>2</sup>, depth varying from 324 to 836 m; thickness varying from 0.20 to 11.59 meters, and average content of 24.05% KCl.

## **Uranium**

### **Pocos de Caldas Deposit**

Location: East of Minas Gerais State, Southeast of Brazil.

Deposits are associated with molybdenum and zirconium. The main host rocks of the uranium mineralization are tinguaites, phonolites, and pyroclastic rocks. There are four distinct types of mineralization in the main bodies:

- primary vein types,
- secondary oxidation reduction types,
- diffuse mineralization
- pockets of reduced soil preserved in oxidized rocks.

### **Lagoa Real Deposit**

Location: Bahia State. 13° 56' 36" S and 42° 15' 32" W

Climate: AW in the Köppen classification, tropical warm and humid. Annual average precipitation is 800 mm, with two distinct seasons: dry from May to September and rainy from October to April.

Soils: pedology in this region is highly influenced by the climate and by the lithology. In many parts, the altered layer of the rock is thick and present lateritic and argillaceous soils.

Hydrography: the region is characterized by seasonal creeks, dried during winter and torrential during summer.

The uranium mineralization occurs in albitites which are characterized by the presence of sodic plagioclase, aegirine-augite and andradite. The rocks are a product of intensive Na-metassomatism of Archean granitic –gneissic and migmatitic rocks. The deposits are

monomirelaltic. The uranium mineralization occurs in elongated, lenticular albitite bodies which have roughly a N-S strike and steep dip.

### **Itatiaia Deposit**

Location: Ceará State, north of Brazil. 04° 31' 40" – 04° 36' 08" S 39° 43' 25" – 39° 54' 05" W

Climate: Bsh in the Köppen classification. Two definite seasons, dry and rainy. Annual average pluviometric rate is 700mm, and annual average temperature is 27°C.

Vegetation: The region can be divided into three sections: (i) Caatinga, kind of vegetation resistant to dry seasons; (ii) Tropical forest; and (iii) Anthropic areas.

Soils: mainly non-karstic, plansols, cambisols and some associations of non-euthrophic soils of low permeability and podizolic soils, red-yellow.

Hydrography: characterized by a rapid runoff, intense evaporation.

The main deposits are paragneisses and large carbonate lenses. Both gneisses and carbonate rocks are cut by several granitic and pegmatitic apophyses. Two types of ores: (i) uniform masses of collophane and the another of collophane veinlet and stockwork ores in marbles, gneisses and in "episyenites"; (ii) in cryptocrystalline hydroxy-apatite.

Mineralization: In the collophanes the mineralization has 26.35% P<sub>2</sub>O<sub>5</sub> and 0.19 U<sub>3</sub>O<sub>8</sub>, and P<sub>2</sub>O<sub>5</sub> : U<sub>3</sub>O<sub>8</sub> = 138:1 In the other ore type: P<sub>2</sub>O<sub>5</sub> = 11.23% and U<sub>3</sub>O<sub>8</sub> = 0.63%, and P<sub>2</sub>O<sub>5</sub> : U<sub>3</sub>O<sub>8</sub> = 18:1

### **Figueira Deposit**

Location: Paraná State, south of Brazil. 23° 48' 00" S, 23° 50' 10" S and 50° 26' 52" W, 50° 27' 30" W

Climate: Cfb in the Köppen classification, sub-tropical , humid, mesothermic, mild summers, severe chilly rains and no dry season. The annual average temperature is 34.6 °C maximum and 5°C minimum. The pluviometric rate is 1199.7 mm/ a .

Vegetation: some cerrado ( grasses, small trees and bushes) and coffee.

Soil: shallow soils, red-yellow, argillaceous, and poor drainage. The soil is acid, low base saturation, high Aluminum saturation and clay of high activity.

Topography: the topography is smooth, with some hills and the highest altitude is 600 m.

Located in the Middle Permian sediments of the Rio Bonito formation which is composed by sandstones and carbonaceous sediments. The mineralization which occurred in the basal sediments of Rio Bonito was the result of both syngenetic and epigenetic processes. The main ore body is lenticular in shape and follows a north-south trending paleochannel. The uranium occurs in as uraninite in sandstones or associated with organic matter in carbonaceous clays and coals.

## **Espinhares Deposit**

Location: Paraíba State, north of Brazil. 6° 51' S and 37° 20' W

Climate: the climate is semi-arid (Bsh in the Köppen rank), characterized by alternations between dry and rainy seasons. The annual average precipitation is 450 to 550 mm. The annual average temperature is 25–27 °C.

Vegetation: the vegetation is very adapted to the water shortage.

Soils: the main soils are the litholitic and brune. The brune soils are shallow, with a textural B horizon, a sudden change from A to B horizons. There is a high concentration of primary minerals and clays of high activity. The litholitic soils are shallow and poorly developed. Both soils are associated with stony surfaces or with the so called desertic pavement. Both soils are very sensible to erosion.

Hydrography: The region is characterized by rivers of intermittent regime and of small sizes. The strong rain storms together with the low permeability of the terrain, create a regime of high velocities runoffs at relatively short period of time.

Topography: the uranium deposit is located on a depression, with a surface characterized by surface strongly weathered.

There are three formations: (i) Jucurutu: many types of gneisses with intercalated amphibolites, amphibolite-schists, marbles, itabirites, and sheelite bearing rocks. (ii) Equador: predominance of quartzites, with intercalated biotite-schists and metaconglomerates. (iii) Seridó: predominantly comprised of schists (biotite and chlorite) in a thick sequence, with some intercalated limestone in the base.

Geology: There are some signs of a process of metassomatic alteration to which the uranium mineralization is associated. This event is posterior to all rock types including the granites. Those altered rocks have a Uranium content higher than the regional background. This deposit occurs in Precambrian gneisses and schists with intrusive granites associated, that had been altered by metassomatic processes as albitization and hematitization, with silica leaching and phosphate enrichment. It is an epigenetic uranium type deposit, of metassomatic origin, similar to several occurrences scattered over the Brazilian north-east.

## **Amorinópolis Deposit**

Location: 16° 23' to 16° 54' South    50° 50' to 51° 10' West.

Climate: There is a lack of data regarding to climate. The annual mean temperature is 20 °C (in high elevations) and 28 °C at north. The pluviometric rate is not much influenced by the topography being more related to atmospheric circulation. It can go up to 1800 mm / a.

Vegetation: it is characterized by the “cerrado”, a type of vegetation characterized by grasses and small trees and bushes.

Soils: It is characterized by ferruginous concretions starting at 40 to 60 m from the surface. These are developed from sediments containing sand, silt or clay.

Hydrography: The region belongs to the Araguaia river system, with a sub-rectangular pattern, oriented according to tectonic faults, or detritic in sedimentary regions and less disturbed by

tectonic movements. The main rivers are Rio Claro, Caiapó tributaries of the Araguaia river. Rio Claro is the main river in this mineralization area, which occur mainly in pre-Cambrian rocks.

**Topography:** The region has a smooth surface with geomorphologic facies with a structural mesas and hills. The region altitudes vary from 450 to 950m. The deposit areas are at heights from 450 to 650 m.

The uranium mineralization occurs in pelezozoic sediments of the Lower Devonian Ponta Grossa Formation of Paraná Basin. The host rock of uranium mineralization is a banket arkose with 2 to 8 m thickness. Three types of uranium mineralization: (i) Geochemical cells (ii) Associated with the residual cangas (iii) Fractures. The main type of mineralization is epigenetic in the form of geochemical cells, and forming bodies in the shape of inverted C (roll front) as was found in the Colorado Plateau and Wyoming, USA. The uranium minerals occurs in two forms:  $U^{6+}$  (autunite and sabugalite) and the primary  $U^{4+}$  (pitchblende and coffinite).

### **Campos Belos And Rio Preto Deposits**

**Location:** 46° 45' W – 47° 00' W and 13° 00' S-13° 10' S (Campos Belos) and 47° 50' W-48° 05' W and 13° 47' S- 14° 00' S (Rio Preto).

**Climate:** there are two main seasons, i. e., rain, from October to March; and dry from April to September. The annual average temperature is of the order of 24 °C at low altitudes and 20 °C at high elevations. The annual average pluviometric rate is 1750mm.

**Vegetation:** The typical vegetation is cerrado, i. e., grasses and small trees and bushes.

**Hydrography:** The main river is the Bezerra river. The region is characterized by a phase of transformations with a high rate of erosion, as it can be seen by the lack of alluvial deposits.

The Campos Belos deposit is associated with quartz-sericite schists with graphite and granite-gneissic rocks which belong to the Goiano Complex. So far, all the uranium mineralization detected are secondary, composed mainly by autinite, torbernite and renardite. The Rio Preto deposits are found in the Ticunzal Formation of lower proterozoic age. The lower part of this formation consists of alternations of biotite schists and quartz feldspar gneiss, while the upper portion is composed of garnet-micaschists and graphitic schists. Primary uranium mineralization (uraninite) was detected in drill cores of the Ticunzal Formation. The mineralization occurs in fractures along the graphitic schists containing pyrite. Uranium concentrations vary from 100 ppma to 0.5%  $U_3O_8$ .

### **Moeda Formation Deposit**

This area is located in south of Minas Gerais State, and is composed predominately of quartzite, conglomerate, and phyllite. As at the Witwatersrand (South Africa) Au and Uranium deposits, the distribution of these elements is controlled by sedimentological factors. There is some similarity between the characteristics of the South African deposits and the mineralization features found in the Moeda Formation.



## **Thorium**

Thorium deposits in Brazil are distributed as follows:

- Pegmatites under the form of monazites crystals or as niobium – tantalites complexes (samarskites, euxenite, etc.).
- In secondary deposits derived from magmatic rocks and from pegmatites, forming elluvials and alluvials that contain residual monazite under the form of small fragments and round grains , together with other weathering resistant minerals.
- Under the form of thorium minerals originated from intrusions of nefelinic sienites and carbonatites, making up pirocloro deposits and thorium containing rocks.

In Brazil there are important thorium deposits: (i) State of Maranhão : low proportion of shore sands between Mearin river and Parnaíba. (ii) State of Rio Grande do Norte.

### **Additional information sources**

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### **REFERENCE TO APPENDIX B**

[B1] DEPARTAMENTO NACIONAL DE PRODUÇÃO MINAROLÓGICA, Geologia do Brasil, Brasília, 1984.



## APPENDIX C

### CHINA

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#### Abstract

Through surveying various relevant literatures, data on nationwide element abundance or concentration in soil, rock and water in China is collected. Based on these data, natural fluxes due to some natural processes in some major water bodies are calculated. We also made comparison between concentration and fluxes due to natural process and those derived from LILW repository in South China.

The results indicate that there are a number of useful data that can be used potential in the CRP, including element abundance in the sediments, rocks and surface water bodies. The results indicate also that the element concentration and flux derived from south China LILW repository is several order magnitude less than that due to natural process as well as other risk indices.

This shows very clearly that natural safety indicators are useful supplement to dose and risk indicators and can play important role in safety assessment of radioactive waste disposal. But it should also be noted that there exist some difficulties in applying natural safety indicators to safety assessment of LILW disposal because the dominant nuclides in LILW are not natural ones. So further work should be carried out so that those natural safety indicators can play more important roles to public confidence building in radioactive waste disposal.

#### Background

In China, the history of nuclear applications spans some fifty years. At present, there are two nuclear power stations in operation and another one in the process of construction. In addition, there are many isotope users in China. The government of China has made a continuous effort to improve radioactive waste management. Presently, two repositories for low and intermediate radioactive waste have been built and one of them has been put into operation. The national repository for high level radioactive waste is in the process of siting and two boreholes have been drilled in one of candidate sites.

It is well known that radioactive waste disposal requires waste to be isolated from humans and the environment in the long term. Considering the factor of uncertainty involved with dose and risk indicators, the use of natural safety indicators in the assessment of radioactive waste disposal has been paid more attention to in recent years in China and other countries and some natural safety indicators have been successfully used in safety assessment.

Regarding that HLW disposal programme in China is in primary stage and the data on candidate site is in the process of collection; Beilong LILW disposal site is selected as a specific site to study whether or not the natural safety indicators can be successfully used in safety assessment.

## Elemental and radionuclide concentration data

A lot of literature has been reviewed from various sources and a considerable amount of useful data has been obtained in China in the CRP. The data involves nationwide U, Th,  $^{226}\text{Ra}$  and  $^{40}\text{K}$  distribution in soil, rock, water, [C1] nationwide soil erosion rate as well as element abundance in various rocks in east of China [C2].

## Nationwide element abundance in soils and water bodies

Organized by the China National Environment Protection Agency, a nationwide survey of environmental natural radioactivity level was conducted in China during the time period from 1983 through to 1990. The main aim was to ascertain the current level, distribution and pattern of radioactivity in China. The survey area covers 29 provinces, autonomous regions and municipalities directly under the central government (Hainan province is included in Guangdong province).

## Nationwide $^{238}\text{U}$ , $^{232}\text{Th}$ and $^{40}\text{K}$ concentrations in soil

### (1) Sampling method

Basically, sampling points are uniformly set up in grid size of  $25\times 25$  km nationwide and are also added on the basis of different soil-forming parent rock and material and the types of soil. The added points are also set up in following areas: the areas in which the natural  $\gamma$  radiation dose rate is locally abnormal, the area close to city and radioactive mines which might result in radioactive contamination. In some areas where population density is extremely small, the grid size is enlarged to  $50\times 50$  km.

According to the above sampling principle, 13 866 soil samples were collected and 11 216 samples were measured, of which 9114 were grid-point samples 2102 additional ones. Analysis and statistics were conducted based on 7777 basic grid in  $25\times 25$  km and  $50\times 50$  km samples.

### (2) Results

- The averaged concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  (area weighted) and its standard deviation for single measurement are 39.5 and 34.4, 49.1 and 27.6, 580.0 and 202.0 Bq/kg within the range of (1.8-520.0), (1.0-437.8), (11.5-2185.2) Bq/kg, respectively.
- The geographical distribution of natural radionuclide concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in soil appears apparently zoning. For  $^{238}\text{U}$  and  $^{232}\text{Th}$ , their concentration in the south part of China is higher than that in north part. But, the concentration change of  $^{40}\text{K}$  in soil is more complicated.
- The concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are apparently correlated to the types of parent-rock. The analysis results from 1552 soil samples with different parent-rock show: the concentrations of  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in magmatite is highest,  $^{238}\text{U}$  and  $^{232}\text{Th}$  in metamorphic rock type higher, in sedimentary rock type the lowest. However, the concentration of  $^{40}\text{K}$  in sedimentary rock type is higher, in metamorphic rock type the lowest.

## Nationwide U, Th and <sup>40</sup>K concentration in water bodies

### (1) Sampling collection

First, all water bodies were classified in seven different classes. They are river water, lake water, reservoir water, offshore water, spring water, well water and tap water, separately. In additional, less cellar water were also involved.

In the survey process, a total of 4021 sampling sites were laid out and a total of 6611 samples were collected in ordinary and dry seasons. The samples were taken from 282 outward rivers, 29 continental rivers, 101 freshwater lakes, 27 saltwater lakes, 279 reservoirs, 130 hot springs, 159 cold springs, 713 shallow water wells, 76 deep water wells, 55 coastal waters, 702 waterworks in cities and 13 cellars in countryside, separately.

### (2) Results

Measurement results indicate: U concentration in various water bodies except of saltwater lake is in the order of magnitude of µg/l; Th concentration in 10<sup>-1</sup>µg/l and <sup>40</sup>K concentration range from 60-200mBq/l except of sea, saltwater lake and hot spring. Summary of measurement results is listed in Table C1. In additional, element concentration in main rivers is measured and the results are listed in Table C2.

TABLE C1. SUMMARY OF MEASUREMENT RESULTS OF U, TH AND <sup>40</sup>K CONCENTRATION IN WATER BODIES IN CHINA

Type		U, µg/l		Th, µg/l		<sup>40</sup> K, mBq/l	
		range	average	range	average	range	average
river		0.02-42.35	1.77*	<0.01-9.07	0.31*	8.0-7149	89.4*
lake	freshwater	0.04-19.20	2.10**	0.02-0.93	0.24**	4.8-2640	102.4**
	saltwater	1.07-387.0	22.36**	0.04-8.60	0.64	54.8-22 410	2108**
reservoir		0.03-19.50	0.73**	<0.01-2.04	0.09**	3.2-1205	50.2
spring	hot	0.02-18.60	0.87	0.02-4.85	0.25	5.1-8125	1058
	cold	0.04-14.88	1.37	0.01-1.50	0.20	2.9-1830	254.5
well	shallow	<0.01-101.6	3.82	<0.01-6.29	0.15	3.3-5867	401.9
	deep	0.02-358.87	2.95	<0.01-1.32	0.16	1.0-923.4	122.1
offshore		0.07-5.20	2.21	<0.01-5.92	0.53	2500-21600	10320
tap water		0.01-56.68	2.12	0.01-3.00	0.13	1.8-1580	133.3

\* Runoff weighted average \*\* Storage water capacity weighted average

TABLE C2. ELEMENT CONCENTRATIONS IN MAIN RIVERS IN CHINA

River	U (ppb)		Th (ppb)		$^{226}\text{Ra}$ ( $10^{-17}$ )		$^{40}\text{K}$ (ppt)	
	Range	mean	Range	mean	Range	mean	Range	mean
Rivers in northeast of China	0.02–14.90	1.00	<0.01–2.60	0.21	1.42–271.74	27.82	51.15–32 64.74	422.55
Hailuan river	0.14–12.00	2.19	0.02–2.73	0.26	1.37–73.25	18.29	163.00–34 172.22	2124.23
Yellow river	0.03–42.35	4.60	0.04–3.05	0.36	1.69–148.79	24.35	109.94–11 921.32	763.84
Huai river and river in Shandong	0.03–10.00	2.11	0.03–9.07	0.64	1.58–23.07	12.07	108.98–28 010.8	1151.50
Yingzi river	0.05--8.62	0.83	<0.01–5.60	0.32	2.29–158.34	11.71	38.24–9579.12	416.34
Rivers in Zhejiang and Fujian	0.04--1.36	0.25	0.02–2.50	0.16	2.46–33.579	10.24	119.5–2495.64	417.77
Zhu river and other river in Guangdong and Guangxi	0.03-3.68	0.55	0.02–0.61	0.13	2.35–84.28	13.35	60.23–1120.91	250.95
Rivers in southwest of China	0.16–5.85	1.53	0.05–6.97	0.55	3.41–162.57	21.51	52.58–1032.48	298.27
Continental rivers	0.76–12.21	4.30	0.05–1.87	0.35	1.99–83.27	15.10	109.46--2706.44	778.66
Total	0.02-42.35	1.66	<0.01–9.07	0.31	1.37–271.74	16.52	38.24–34 172.22	686.86

### Average element abundance in crust and rocks

The continental crust of China is composed of three regional crusts, the Cathaysian crust, the West China crust and the South Tibet crust. Based on research results by Li Tong etc, the Table C3 lists the abundance of element interest to the CRP in three regional crusts.

TABLE C3. ELEMENT ABUNDANCES OF CONTINENTAL CRUSTS IN CHINA (UNIT:PPM)

	K	Cu	Rb	Sn	Cs	Th	U
Cathaysian crust	9530	35	29	2.98	1.0	6.21	1.85
West China crust	8890	42	97	2.46	8.2	8.89	3.10
South Tibet crust	9900	44	57	3.08	3.9	7.45	2.40

Sponsored by the Ministry of Geology and Mineral Resources, the regional abundance of elements in the upper crust in eastern China was investigated in detail in the 1990's. In the project, 28253 rock samples are systematically collected from 500 standard stratum profiles and 800 intrusive and metamorphic rock bodies in the area of 3.3 million km<sup>2</sup> of eastern China, which is more than one third of total area in China. Some of results are listed in Tables C4-6.

TABLE C4. AVERAGE ELEMENT ABUNDANCE IN EARTH CRUST IN EAST CHINA (UNIT:PPM)

	Cs	Cu	Rb	Sn	Th	U
Earth crust	2.0	26	70	1.4	6.0	1.3
Upper earth crust	3.3	17	95	1.8	9.5	1.8
Middle and lower earth crust	1.4	31	50	1.1	4	0.7

TABLE C5. AVERAGE ELEMENT ABUNDANCE IN VARIOUS ROCKS IN CHINA (UNIT:PPM)

	Cs	Cu	Rb	Sn	Th	U
Granite	3.6	5.5	160	2.2	17	2.9
Rhyolite	3.9	4.5	142	2.2	14.5	3.0
Granodiorite	3.3	18.5	95	1.4	9.0	1.5
Tonalite	1.7	23	43	1.1	3.8	0.8
Diorite	1.9	27	64	1.3	5.4	1.2
Basalt	0.83	52	23	1.2	3.0	0.73
Quartzose sandstone	0.8	4.4	23	0.6	4.0	2.1
Limestone	0.6	4.2	10	0.5	1.1	1.3

TABLE C6. ELEMENT ABUNDANCE IN GRANITE IN VARIOUS TECTONIC UNITS IN EAST CHINA (UNIT: PPM)

	Cs	Cu	Rb	Sn	Th	U
East China	4.1	5.4	145	2.2	14.9	2.8
Xingan-jilhei orogenic belt	4.4	4.5	130	2.1	13.1	2.4
North China platform	2.0	5.5	135	1.4	13.6	1.7
Qinling-dabie orogenic belt	4.0	8.7	145	1.8	17.8	2.9
Yingzi platform	6.3	9.9	153	3.3	16.2	3.7
South China fold system	5.6	6.5	223	3.9	23.2	5.6

## Elemental and radionuclide flux data

### Element fluxes in main rivers

Based on flow rate and element concentration, element mass fluxes and element activity fluxes in main surface drainage systems in China are calculated and results are listed in Table C7 and Table C8, respectively.

TABLE C7. ELEMENT MASS FLUXES IN MAIN SURFACE DRAINAGE SYSTEMS IN CHINA

	Catchment area ( $10^4 \text{ km}^2$ )	Flow rate ( $10^8 \text{ m}^3/\text{y}$ )	U (kg/y)	Th (kg/y)	$^{226}\text{Ra}$ (g/y)	$^{40}\text{K}$ (kg/y)
Rivers in northeast of China	116.6	1731.2	1.7E5	3.6E4	48.2	7.3E4
Hailuan river	31.9	283.5	6.2E4	7.0E3	5.2	6.0E4
Yellow river	75.2	574.5	2.6E5	2.1E4	14.0	4.4E4
Huai river and river in Shandong	32.6	597.5	1.3E5	3.8E4	7.2	6.9E4
Yingzi river	180.7	9793.5	8.1E5	3.1E5	114.7	4.1E5
Rivers in Zhejiang and Fujian	21.3	2001.3	5E5	3.2E4	20.5	8.4E4
Zhu river and others in Guangdong and Guangxi	55.3	4466.3	2.5E5	5.8E4	59.6	1.12E5
Rivers in southwest of China	86.4	4428.6	6.8E5	2.4E5	95.3	1.32E5
Continental rivers	348.0	1130.7	4.9E5	4.0E4	17.1	8.8E4



TABLE C6. ELEMENT ABUNDANCE IN GRANITE IN VARIOUS TECTONIC UNITS IN EAST CHINA (UNIT: PPM)

	Cs	Cu	Rb	Sn	Th	U
East China	4.1	5.4	145	2.2	14.9	2.8
Xingan-jilhei orogenic belt	4.4	4.5	130	2.1	13.1	2.4
North China platform	2.0	5.5	135	1.4	13.6	1.7
Qinling-dabie orogenic belt	4.0	8.7	145	1.8	17.8	2.9
Yingzi platform	6.3	9.9	153	3.3	16.2	3.7
South China fold system	5.6	6.5	223	3.9	23.2	5.6

## Elemental and radionuclide flux data

### Element fluxes in main rivers

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	Catchment area ( $10^4 \text{ km}^2$ )	Flow rate ( $10^8 \text{ m}^3/\text{y}$ )	U (kg/y)	Th (kg/y)	$^{226}\text{Ra}$ (g/y)	$^{40}\text{K}$ (kg/y)
Rivers in northeast of China	116.6	1731.2	1.7E5	3.6E4	48.2	7.3E4
Hailuan river	31.9	283.5	6.2E4	7.0E3	5.2	6.0E4
Yellow river	75.2	574.5	2.6E5	2.1E4	14.0	4.4E4
Huai river and river in Shandong	32.6	597.5	1.3E5	3.8E4	7.2	6.9E4
Yingzi river	180.7	9793.5	8.1E5	3.1E5	114.7	4.1E5
Rivers in Zhejiang and Fujian	21.3	2001.3	5E5	3.2E4	20.5	8.4E4
Zhu river and others in Guangdong and Guangxi	55.3	4466.3	2.5E5	5.8E4	59.6	1.12E5
Rivers in southwest of China	86.4	4428.6	6.8E5	2.4E5	95.3	1.32E5
Continental rivers	348.0	1130.7	4.9E5	4.0E4	17.1	8.8E4

TABLE C8. ELEMENT ACTIVITY FLUXES IN MAIN SURFACE DRAINAGE SYSTEMS IN CHINA

	Catchment area (10 <sup>4</sup> km <sup>2</sup> )	Flow rate (10 <sup>8</sup> m <sup>3</sup> /y)	<sup>238</sup> U (TBq)	<sup>242</sup> Th (TBq)	<sup>226</sup> Ra (TBq)	<sup>40</sup> K (TBq)
Rivers in northeast of China	116.6	1731.2	2.09	1.48	1.76	15.26
Hailuan river	31.9	283.5	0.76	0.29	0.19	12.54
Yellow river	75.2	574.5	3.20	0.86	0.51	9.20
Huai river and river in Shandong	32.6	597.5	1.60	1.56	0.26	14.42
Yingzi river	180.7	9793.5	9.97	12.71	4.20	85.28
Rivers in Zhejiang and Fujian	21.3	2001.3	0.62	1.31	0.75	17.56
Zhu river and others in Guangdong and Guangxi	55.3	4466.3	3.08	2.38	2.18	23.41
Rivers in southwest of China	86.4	4428.6	8.37	9.84	3.49	27.59
Continental rivers	348.0	1130.7	6.03	1.64	0.63	18.39

#### Element mass fluxes due to erosion

On the basis of a second nationwide survey (1999-2001) employing remote sensing technology, the most recent data of erosion rate by water and wind in China are obtained [C3]. The survey indicates that total erosion area in China is 3 560 000 km<sup>2</sup> and water erosion area and wind erosion area is 1 650 000 km<sup>2</sup> and 1 910 000 km<sup>2</sup>, respectively. The survey also indicates that the total amount soil eroded is about 5 billion tons per year in China and amount in Yingzi river drainage area is 2.4 billion tonnes per year and amounts eroded in middle reaches of Yellow river is 1.6 billion tonnes per year. Erosion area regarding erosion module is listed in Table C9.

TABLE C9. WIND EROSION AREA AND WATER EROSION AREA WITH RESPECT TO EROSION MODULE IN CHINA

	Wind erosion area (10 <sup>4</sup> km <sup>2</sup> )	Water erosion area (10 <sup>4</sup> km <sup>2</sup> )
<2500 t/km <sup>2</sup> y	79	83
2500–5000 t/km <sup>2</sup> y	25	55
5000–8000 t/km <sup>2</sup> y	25	18
8000–15 000 t/km <sup>2</sup> y	27	6
>15 000 t/km <sup>2</sup> y	35	3

Element mass fluxes associated with erosion in drainage areas of the Yangzi River and the middle reaches of the Yellow River are also calculated and results can be found in Table C10. It is based on assuming that all the soil eroded will finally reach into the rivers and take element concentrations in soil eroded in the Yangzi drainage region as corresponding to the ones of soil in China and that in the middle reach of the Yellow river as corresponding to the ones of loess in China.

TABLE C10. ELEMENT MASS FLUXES ASSOCIATED WITH SOIL EROSION

	Soil eroded (10 <sup>12</sup> kg)	Cs (10 <sup>6</sup> kg)	Cu (10 <sup>6</sup> kg)	Rb (10 <sup>6</sup> kg)	Sn (10 <sup>6</sup> kg)	Th (10 <sup>6</sup> kg)	U (10 <sup>6</sup> kg)
Yangzi drainage region	2.4	16.8	57.6	240	6.0	30.0	6.48
Middle reach of Yellow river	1.6	12	38.88	153.6	4.48	18.88	4.32

### Interpretation and application of the concentration and flux data

As mentioned previously, plans for a HLW repository is still at an initial stage in China and insufficient data is available for the CRP at this time; therefore, one of LILW disposal facilities, Beilong LILW repository, is selected as a research site.

### Introduction to Beilong LILW repository [C4]

Beilong disposal site is located at the north-east of the Dapun peninsula in Guangdong Province, about 5 km far away from the Daya Bay NPP. It is situated with the Paiya hill to the rear and the Daya Bay to the front. It is a seaside and lower hilly land area. As the same as most regions in south China, it is warm and humid with an average 1900 mm precipitation and 1600 mm evaporation per year. Beside the disposal site, there are two rivulets flowing to

the Daya Bay, which are main discharge channels for surface water and groundwater in the site area. One of them is the Longzi rivulet with a minimum flow rate of  $0.015 \text{ m}^3/\text{s}$ , another is the Lingao rivulet and will disappear after the Lingao reservoir located to the north of site is built. The groundwater table is between 6 m to 15 m and can change extensively after rainfall. Figure C1 is sketch map of the site.

Beilong LILW repository is composed of 70 dispose vaults with total capacity of  $80\,000 \text{ m}^3$  and total area of  $0.205 \text{ km}^2$ , which will be used to dispose LILW arising mainly from NPPs in the south of China. All disposal vaults are above surface and minimize 6 m depth to groundwater table.

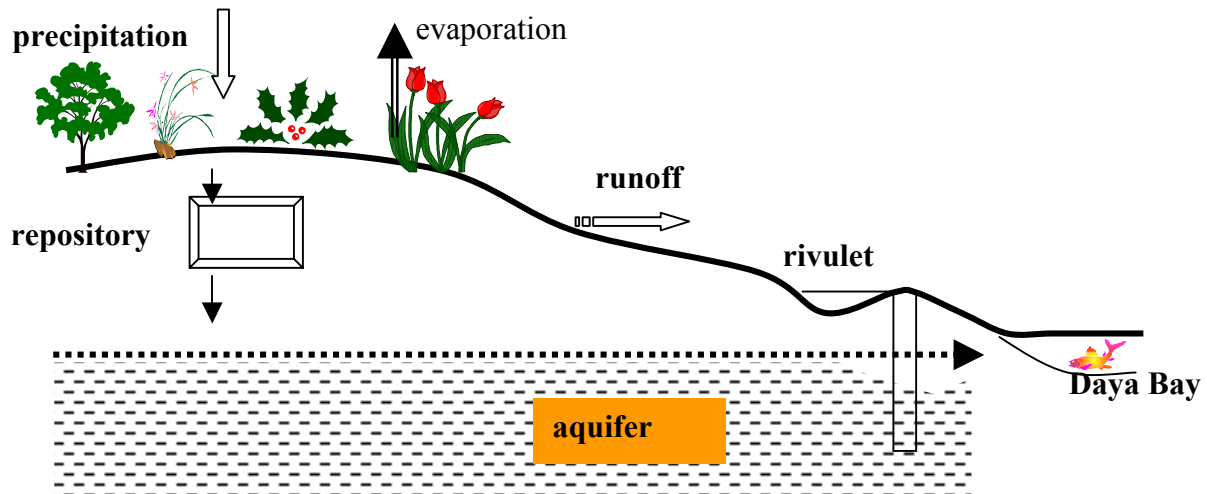


FIG. C1. Schematic representation of the Beilong LILW disposal site.

### Comparison of total activity

As mentioned above, the waste in the repository is mainly from NPPs so dominant radionuclides are fission products and activated productions. Seven radionuclides are selected as dominant radionuclides and Table C11 gives their inventory when repository is closed (abstracted from Environment Assessment Statement (EIS) by Cui etc.). EIS's results also indicate that nuclides released from the repository will be contained within the unsaturated zone except  $^3\text{H}$  and  $^{14}\text{C}$ . Considering that the disposal site's area is  $0.205 \text{ km}^2$  and the minimum thickness of the unsaturated zone is 6m and soil density is  $1.66 \times 10^3 \text{ kg/m}^3$ , this means that the volume of the unsaturated zone is  $0.205 \times 10^6 \times 6 = 1.23 \times 10^6 \text{ m}^3$  and the total mass is  $2.04 \times 10^9 \text{ kg}$ . On the basis of the radioactive specific activity, radionuclide activities in the unsaturated zone are calculated and are listed in Table C12.

TABLE C11. THE RADIOACTIVITY IN REPOSITORY AT CLOSURE

Nuclide	<sup>137</sup> Cs	<sup>60</sup> Co	<sup>90</sup> Sr	<sup>63</sup> Ni	<sup>14</sup> C	<sup>239</sup> Pu	<sup>3</sup> H	Total
Activity (Bq)	1.1E15	3.3E15	3.7E11	9.6E14	3.3E13	3.4E11	4.2E12	5.4E15

TABLE C12. RADIONUCLIDE ACTIVITIES IN UNSATURATED ZONE

Nuclide	<sup>238</sup> U	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	Total
Activity (Bq)	8.77E10	6.94E10	1.47E11	1.15E12	1.45E12

A comparison between the total activity in the repository and that in the unsaturated zone shows that the former is three orders of magnitude more than the latter. Of course, this does not mean that the repository is not safe because it takes no account of the isolation function of engineer barriers.

#### Comparison of concentration in water

Only <sup>3</sup>H and <sup>14</sup>C are considered due to the fact that only they can reach aquifer and surface water body within assessment period. Table C13 provides the maximum concentrations of them in discharge point of underground water, natural radioactivity concentrations in Longzi rivulet, natural radionuclide concentrations in natural groundwater, radioactivity concentration in Daya Bay and maximum permissible concentration of WHO's drinking quality standard (which corresponds 0.1 mSv/y).

TABLE C13. COMPARISON BETWEEN CONCENTRATION DERIVED FROM REPOSITORY AND NATURAL CONCENTRATION IN WATER (UNIT: BQ/M<sup>3</sup>)

Nuclide	Discharge point	Longzi rivulet	Groundwater	Sea water	Drinking water quality
<sup>3</sup> H	1.03E4	710	664	1000	7.8E6
<sup>14</sup> C	2.12E2			16	2.5E5
<sup>238</sup> U			26	4.5	
<sup>226</sup> Ra		4	34	7.2	
<sup>232</sup> Th			25	8900	
<sup>40</sup> K		93			
total		807	749	9928	

Based on the Table C13, the maximum concentrations of  $^3\text{H}$  and  $^{14}\text{C}$  in the groundwater discharge point is one to two orders of magnitude larger than that in surface water bodies and groundwater but it is several orders of magnitude less than the maximum permissible concentrations of WHO's drinking quality standard. This means that concentrations in the groundwater discharge point are acceptable and the repository will not bring significant harm to the environment and humans from the point of view of radionuclides released through underground water.

## Comparison of fluxes

### Fluxes released from repository

On the basis of EIS, Figure C2 shows radionuclide release rate curves from the bottom of disposal vaults versus time after closure. Table C14 lists the maximum release rates and corresponding ALI values for the public in basic health standards for radiological protection in China.

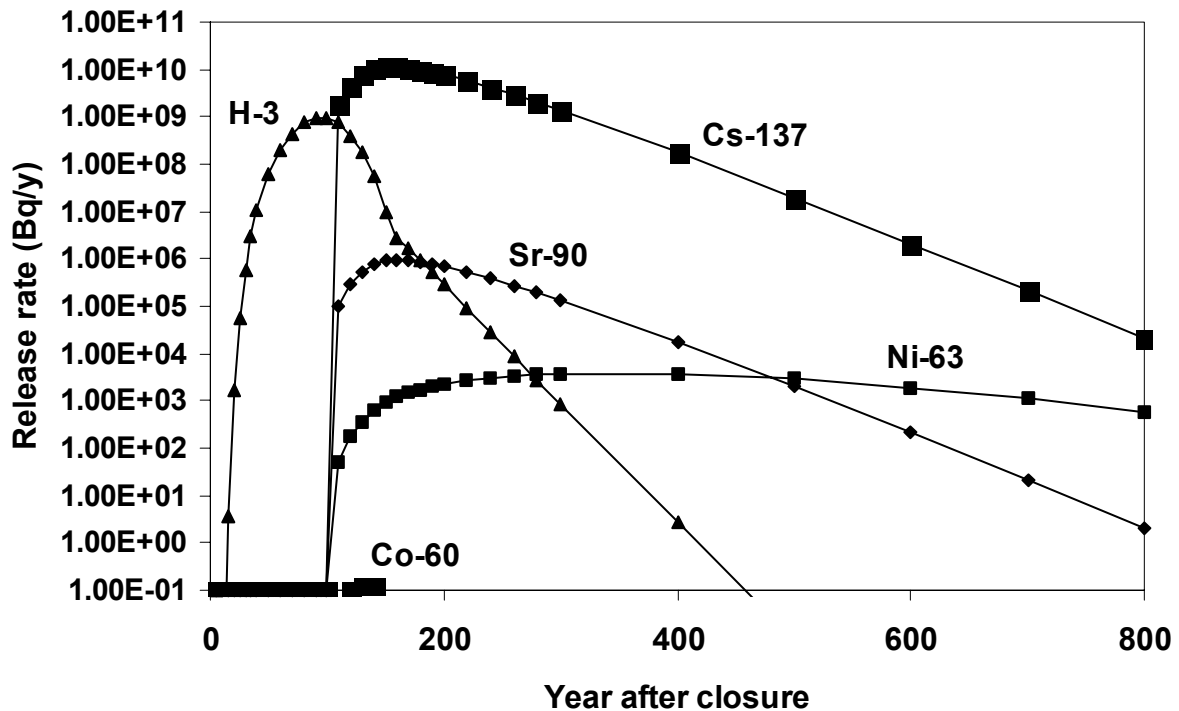


FIG. C2. Nuclide release rate from bottom of disposal vaults.

TABLE C14. COMPARISON BETWEEN MAXIMUM RELEASE RATE FROM VAULTS AND ALI

nuclide	<sup>137</sup> Cs	<sup>60</sup> Co	<sup>90</sup> Sr	<sup>63</sup> Ni	<sup>14</sup> C	<sup>3</sup> H	<sup>239</sup> Pu
Max. release rate (Bq/yr)	9.44E9	9.68E-2	7.46E6	3.02E3	1.07E7	7.75E8	1.6E3
ALI for public (Bq/yr)	3.7E5	1.9E6	1.6E6	3.4E7	8.8E6	2.9E8	2.4E5

Table C14 shows very clearly that the maximum release rate of all concerning nuclides except for <sup>137</sup>Cs are several orders of magnitude lower than ALI for the public or are of the same order of magnitude as ALI for the public. EIS results indicate that <sup>137</sup>Cs released from the repository is contained in 1 m thickness soil below the disposal vaults. Assuming <sup>137</sup>Cs is released from repository distributes homogenous in soil, then its specific activity is 27.8 Bq/kg, which is one order of magnitude lower than DIC (derived intake concentration) for the public (460 Bq/kg) in the Basic health standards for radiological protection in China.

#### Activity fluxes due to groundwater discharge

The water balance analysis shows that the flow rate of groundwater discharging into the Longzi rivulet is  $1.61 \times 10^4 \text{ m}^3/\text{y}$ . Given that the concentration of <sup>3</sup>H in the groundwater retains its peak values in discharge point, activity fluxes derived from the repository due to groundwater discharge to the Longzi rivulet are calculated and listed in Table C15.

TABLE C15. <sup>3</sup>H ACTIVITY FLUXES DUE TO GROUNDWATER DISCHARGE

	<sup>3</sup> H activity flux (Bq/y)
Groundwater with nuclides released from repository	1.66E8
Natural groundwater	1.14E7
Longzi rivulet	3.34E8

Although the <sup>3</sup>H activity flux due to the groundwater discharge is one order of magnitude higher than that of natural one in the disposal site, it is of the same order of magnitude as that in the Longzi rivulet and several orders of magnitude lower than the total activity fluxes in large-scale water bodies (see Table C8). This means that activity fluxes in the Longzi rivulet will not increase significantly and still remain within the range of natural levels after the repository is put into operation.

## Conclusions

(1) The literature from various sources is reviewed and much useful data is obtained. The nationwide data consists of comprehensive element abundance or concentration in soils, surface water bodies and groundwater. In addition, element abundance in crustobodies and in various rocks as well as that in granite in different tectonic units in East China is also collected. The newest erosion rate data in China is also obtained. Some of it has still not been applied to safety cases in research yet, but it is vital to establishing an international database on element abundance in various media.

(2) Comparison between concentration and fluxes due to natural process as well as relevant hazard indices such as WHO's drinking water quality standard and ALI value and that derived from LILW repository in South China indicates that concentration and fluxes derived from a LILW repository are of the same order of magnitude as natural ones or several orders of magnitude lower than natural ones. This shows very clearly that natural safety indicators are a useful supplement to dose and risk indicators and can play an important role in the safety assessment of radioactive waste disposal. On the other hand, it should also be noted that there some difficulties exist in applying natural safety indicators to safety assessment of LILW disposal because the dominant nuclides in LILW are not natural ones. Further work should be carried out so that those natural safety indicators can play a more important role to building up public confidence in radioactive waste disposal.

## REFERENCES TO APPENDIX C

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## APPENDIX D

### CUBA

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#### Abstract

Cuba has been committed to the peaceful use of ionising radiation for benefits of the society, this comprises the safety management of all radioactive waste generated from the nuclear applications. The Center for Radiation Protection and Hygiene (CPHR) is responsible for the radioactive wastes management in the country, including the safety assessment (SA) of disposal facilities. Into the SA methodologies, the associated impact to the environment and the human health from the radioactive waste disposal is measured through of safety indicators. The radiological dose and risk are the most common endpoints adopted, but when in the SA the long-term impact is evaluated, the former indicators can be affected by different assumptions about the critical group, assessment period and parameter uncertainties. With the inclusion of others safety indicators besides dose and risk a reasonable assurance can be achievable and may facilitate the defence of the safety assessment results, according to the radiation protection principles. The concentrations of naturally occurring radionuclides (NOR) and the natural fluxes of radionuclides were selected as safety indicators complementary to dose and risk for the carried out assessment. Both indicators can provide a measure of the potential effect on the human health and the environment, they are less affected for the time and also, it is easy to implement in the safety assessment methodology. The NOR concentrations for Cuba were obtained from a database based in the results of geological and geophysical surveys in the country, the natural concentrations of U, Ra, Th, K were associated with different lithologies for 3 geographical areas of the Country. The site selected for the disposal facility is located in a massif granodiorite in the central part of the country, all relevant information about the geosphere was obtained during the site selection process where geologic, hydrogeologic and geophysical investigation were carried out in order to obtain the most important parameters of the host medium. The repository concentrations were calculated from radionuclide inventory adopted for a high level disposal facility. In order to make a proper comparison between the NORM and repository concentrations was adopted the Reference Equivalent Rock Volume (RERV), a volume of rock with dimensions according to our repository conceptual design. For the natural activity fluxes two processes were evaluated, the erosion and the groundwater fluxes, and the values were obtained from the site investigations and international references. For calculation of the flux data from the Repository, have been carried out a safety assessment for the Cuban radioactive waste disposal system following the Improvement of Safety Assessment Methodologies for Near Surface Disposal Facilities (ISAM) methodology. According the assessment context, have been defined a normal scenario (release scenario), which takes into account only the natural occurrence process in the disposal system, corrosion, degradation, infiltration and so on. A compartments model was adopted in order to simulate the complete radioactive waste disposal system. The final objective was to obtain the specific activity flux from the repository for each radionuclide, allowing preliminary release comparisons against the natural-occurring species.

The preliminary comparison showed that the calculated concentrations and fluxes in the conceptual repository are not higher than the natural values in the host rock. According the carried out assessment using the complementary safety indicators, the reference disposal facility does not increase the natural activity concentration and fluxes in the environment. In Cuba, it is the first application of other safety indicators for safety assessment of radioactive

waste disposal facilities, evidencing that those endpoints could be a useful tool to support the safety assessment for the radioactive waste disposal system. In next studies, should be evaluated other relevant elements and radionuclides present in the specific Cuban inventories and carry out more specific site investigation in order to reduce the data uncertainty.

## **1. Background**

### **1.1 Nuclear programmes**

With the creation, in 1979 of the Cuban Atomic Energy Commission (CEAC) and subsequently the Executive Secretariat for Nuclear Affairs (SEAN) the first steps for the control of the radioactive waste in the country were given up. In 1982 the inventories of all radiation sources and users of radioactive materials in the country were carried out. By the year 1984 the inventories were revised and updated. A Circular, which was addressed to all the users of radioactive materials, prohibiting the direct discharges of radioactive wastes into the environment, was issued during 1985. The construction and implementation of adequate storage facilities at the institutions was demanded.

Since 1994 the Cuban integral policy of nuclear development is entrusted to the Nuclear Energy Agency of the Ministry of Science, Technology and Environment (CITMA). This Ministry assumes the functions of the former CEAC. The National Centre for Nuclear Safety (CNSN) is responsible for the licensing and supervision of radioactive and nuclear installations. The Centre for Radiation Protection and Hygiene (CPHR), is responsible for developing and implementing a national strategy for collection, handling, treatment, conditioning and storage of low and intermediate level radioactive wastes.

The management and disposal of radioactive wastes in Cuba are controlled by a Decree that regulates the work with radioactive materials and a Practice Guide that establishes the methods for segregation, registration and control of radioactive waste. The CPHR is responsible for centralized collection, transportation, treatment, conditioning, long term storage, and disposal of radioactive waste, as well as for developing new waste conditioning and containment methods.

Cuba has been firmly committed to the peaceful applications of ionizing radiations in medicine, industry, agriculture and research in order to achieve socioeconomic development in diverse sectors. The radioactive wastes management from the Program Nuclear Cuban undertakes the wastes generated by the Nuclear Applications (medicine, agriculture, research, etc.) and the possible to generate for the Nuclear Power Plant, which is temporally paralyzed the construction. Consequently, the use of radioactive materials and radiation sources as well as the production of radioisotopes and labeled compounds may always produce radioactive wastes, warranting safe and proper management.

The current national strategy involves the use and development of storage and conditioning facilities and a continuous program of research and assessment to ensure that each type of waste is managed in the most appropriate way.

The Cuban Nuclear Programme will generate approximately 120 m<sup>3</sup> of conditioned wastes (waste packages including matrix and container for disposal) at the end of 2030. According to the storage capacity and an optimal distribution of these waste packages, the Storage Facility will be fulfilled at that year. Most conditioned wastes, in terms of activity content and volume

will be spent sealed sources. Keeping in mind the status of the Nuclear Power Plant, and foreseeing that this installation will generate the major volume of radioactive wastes in the Cuba, the authorities are working to prolong the useful life of the Facility temporal storage.

The existing storage facility will operate as a ‘long term’ storage facility of conditioned wastes. Because of that, the long term safety of this facility has to be evaluated. For this reason, a new research project will begin next year. Some aspects relating with the site characteristics (geography, meteorology, climatology, geology, hydrology), facility design, construction, operational aspects, radionuclide release under normal and unusual operation conditions, the assessment of impacts and the long term stability will be detailed studied and evaluated. Finally, a Safety Analysis Report (SAR) will be prepared according to IAEA recommendations.

The former Nuclear Technology Centre (CTN) carried out a site selection process in regional scale focused to selection of favourable geological environments containing appropriate host geological materials. This selection process considered a screening since national scale to more specific areas. The studies included investigations in topics such as geology, hydrogeology, seismic, geophysics, environment, socioeconomics and other, finally a favourable site has been selected.

The site is located in the central region of the country, its area is about 3 km<sup>2</sup>, the population density of 30 to 40 inhabitants per km<sup>2</sup>, the principal economical resources are the agricultural activity (sugar cane, oranges, etc). The site climate is tropical type, according to the country characteristics (island, cyclones, etc) and topography influence, the site has a warm climate, high humidity, and intensive precipitation (see details Table D1). There are only two seasonal periods, summer and winter with marketable differences.

TABLE D1. METEOROLOGICAL PARAMETERS

<b>Meteorological parameters</b>	<b>Values</b>
Annual temperature	24.7°C
Maximal annual temperature	35.8°C
Minimal annual temperature	7.4°C
Annual relative humidity	78%
Annual precipitation	1275.3 mm
Wind direction	EN
Annual wind velocity	10.9 km/h
Atmospheric pressure	1014.9 Hpa

Seismic investigations have also been carried out on a regional scale, taking into account the geology of the area, presence of important faults, the historic records, geophysics information, and so on. The principal seismic characteristics were also defined; the design basis earthquake (100 years), is V in MSK scale (MSK, intensity scale of earthquake, it has 12 levels similar to Mercalli scale) and the maximal calculated earthquake (10 000 years) is VII in the same scale.

The geologic medium is characterized by a massive granodioritic with several lithologic types such as: Diorites, quartzdiorite of the Upper Cretassic. The tectonics are simple; the site is away from the principal tectonic structures of the area and there are not importance faults near of the site. The geologic profile is:

- 1<sup>st</sup> layer, loamy sand 0 to 15m;
- 2<sup>nd</sup> layer, cracked granodiorite 10 to 15m;
- 3<sup>rd</sup> layer, unaltered granodiorite more 15m.

The groundwater presence is poor, according the rock type (igneous), the permeability of the order of  $10^{-2}$  to  $10^{-4}$  m/d and the aquifer is associated to fissures and the principal source of water is the infiltration from rainfall.

## **2. Repository conceptual design**

The design adopted for the disposal facility include a multibarrier concept, which consist of a system of engineered and natural barriers placed between the wastes and the environment, those barriers are independent and redundant [D1]. The repository is located at 20 m under ground surface, in a stable geologic host. The facility design has eight horizontal disposal chamber (mined) connected to surface through three vertical shafts. The wastes will be disposal conditioned in metal drums of 200 litres (LLW) and adding a concrete container for ILW.

The repository has capacity to disposal all radioactive waste (low and intermediate) generated during 100 years for two blocks of “Juragua” nuclear power plant and the wastes arising from users of nuclear applications in Cuba.

## **3. Radioactive wastes sources**

In Cuba the radioactive wastes arrive from:

- The applications of radioisotopes in medical field as well as in clinical medicine, these wastes are mainly liquids and solid materials contaminated with short lived radionuclides and sealed sources used in radiotherapy and for sterilization of medical materials.
- Radioactive wastes from industrial applications are generally spent sealed sources, which were used in level detection, quality control, smoke detection and non-destructive testing. The current inventory of spent sealed sources stored in CPHR facility is presented in Table D2.
- From research centres, these wastes are miscellaneous liquids, trash, biological wastes, scintillation vials, sealed sources and targets.

The CPHR also gives special attention to the users of  $^3\text{H}$  and  $^{14}\text{C}$  contained in liquid scintillation vials. Solid radioactive wastes are produced mainly during cleaning and decontamination activities and consist of rags, paper, cellulose, plastics, gloves, clothing, overshoes, etc. Laboratory materials such as cans, polyethylene bags and glass bottles also contribute to the solid waste inventory. Small quantities of non-compactable wastes are also collected and received for treatment. They include wood pieces, metal scrap, defective components and tools.

TABLE D2. SPENT SEALED SOURCE INVENTORY

Nuclide	Activity	Quantity	Nuclide	Activity	Quantity
<sup>241</sup> Am	12 GBq	1138	<sup>22</sup> Na	0.2 GBq	9
Am-Be	102 GBq	4	<sup>63</sup> Ni	0.4 GBq	1
<sup>14</sup> C	0.2 MBq	5	<sup>147</sup> Pm	3.7 GBq	3
<sup>107</sup> Cd	0.4 MBq	1	<sup>210</sup> Po	1.5 MBq	1
<sup>109</sup> Cd	67 MBq	3	Po-Be	0.4 GBq	5
<sup>252</sup> Cf	-	1	<sup>238</sup> Pu	9.0 GBq	12
<sup>57</sup> Co	16 MBq	4	<sup>239</sup> Pu	1.2 MBq	18
<sup>60</sup> Co	1.5 TBq	110	Pu-Be	0.9 TBq	5
<sup>137</sup> Cs	2.8 TBq	425	<sup>226</sup> Ra	0.2 TBq	400
<sup>152</sup> Eu	-	1	<sup>106</sup> Ru	-	3
<sup>55</sup> Fe	0.9 GBq	5	<sup>35</sup> S	37 kBq	1
<sup>3</sup> H	3.4 GBq	4	<sup>119m</sup> Sn	-	1
<sup>3</sup> H / Ti	-	10	<sup>89</sup> Sr	0.2 MBq	2
<sup>192</sup> Ir	2.4 GBq	103	<sup>90</sup> Sr	16 GBq	138
<sup>85</sup> Kr	11.3 GBq	6	Sr-Y	37 kBq	62
<sup>54</sup> Mn	(a)	1	<sup>204</sup> Tl	0.2 GBq	22

Note: (a)sources of unknown activity.

The collection of wastes consists in their transference from the place of origin to the place (WSTF) where they will be treated and/or stored. Collection is made periodically and transportation is carried out in accordance with the actual Regulation.

#### 4. Topography, geology and climate

The Cuban geology is very complex, due to its physical form and geographical position. In addition, Cuba is an island. The country is geological constituted, in a general approach, for sedimentary formations of carbonated rocks (more 70%), which are represented for different lithologic transitions, the most representative rocks in this formation are the limestone, clay and sandstone. The 30% remaining, is mainly formed, for metamorphic and igneous geologic formations, conformed for skarn, shale, granite and serpentine. For this reason, Cuba is represented by three regions in Figure D1.

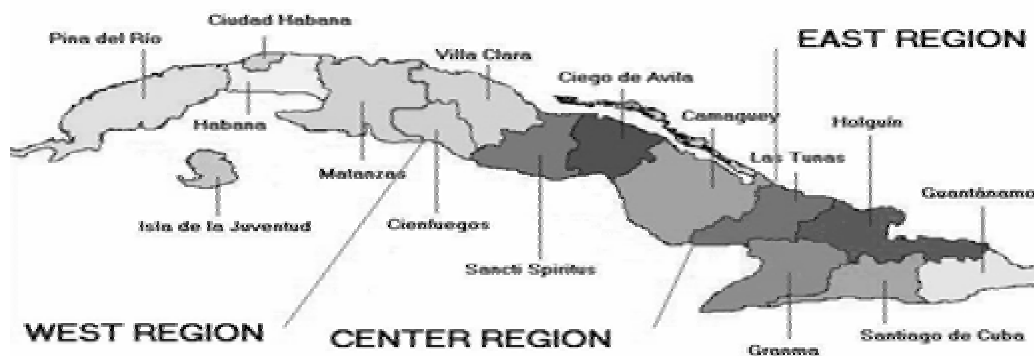


FIG. D1. Regions surveyed in Cuba.

## Notes for Figure D1:

*West region:* this region includes the provinces of Pinar del Río, Havana City and Matanzas. The mentioned area is geologically conformed by Limestone, Sandstone, Shale and Lutites.

*Central region:* this region includes the provinces of Villa Clara, Sancti Spiritus, Ciego de Avila and Camaguey. This area is geologically conformed by Limestone, Serpentine, Skarn, Granite, Tuff, and Shale.

*East region:* this region includes the provinces of Holguín, Gramma, Guantánamo, and Santiago de Cuba. This area is geologically conformed by Limestone, Shale, Tuff, Granite and Serpentine.

## 5. Study areas

In order to define the disposal system for low, intermediate and possible high level radioactive waste, Cuba has been carried out site investigations and adopted a Repository conceptual design. A favourable site, with granodiorite rock as host, has been selected in the central part of the country. The proposal facility will be constructed underground, a few metres below the surface and it including galleries and disposal chambers mined in the rock. This design should be fulfil with the main objective for the waste disposal, to achieve the safety isolation of radioactive waste to environment and the man. For this purpose the safety assessment (SA) is a useful tool in order to estimate the future behaviour for different components of the isolation system.

The IAEA Coordinated Research Project “ISAM” experience in the implementation of the approaches and tools of safety assessment for near surface radioactive waste disposal facility was applied, and as a part of the SA process, in the scenario generation stage.

On the basis of the accepted selection requirement and adopted, a methodology has been created that conforms with the guide for the gradual definition of the safety location [D2, D3]. This methodology includes a national study to regional scale; selection and characterization of the most favourable areas; obtaining the detailed location of the area to confirm and to license.

### (a) National study

According to the scheme of site selection and keeping in mind the signal requirement, the best international practices, and the geographical and geological conditions of Cuba, a national search of the most favourable geologic formations has been compiled - National Inventory of Geologic Formations.

Formations, being evaluated are:

1. Carbonated formations.
2. Intrusive and extrusive formations.
3. Saline formations.
4. Vulcanogen-sedimentary formations.

## **(b) Regional study**

The geological formations were evaluated at the regional scale (technically E-1:100 000). The most favourable regions are selected after a study and evaluation of each one of the selected geological formations. The following types of regional geology were considered further:

### 1. Carbonated formations:

- 8 areas limestone (from the Km 13 of the national Freeway to Matanzas).
- 11 hollows limestone (caverns).
- 1 place limestone of Bolondrón (Matanzas).

### 2. Formations intrusive and extrusive:

- Solid ultrabasic San Miguel de los baños (Matanzas).
- Belt ofiolitic of the North of the Rosario's Sierra (Cajalbana), (Pinar del Río).
- Belt ofiolitic of the North of the Central Region (Villa Clara).
- Belt ofiolitic of the North (Camagüey).
- Belt granodioritic of the North of the Escambray (Cienfuegos).
- Belt granodioritic of Sancti Spiritus-The Tunas (Tunas).

### 3. Saline formations:

- Dome saline of Punta Alegre (Ciego de Avila).
- Dome saline of Turiguanó-Cunagua (Ciego de Avila).

### 4. Vulcanogen-sedimentary formations:

- zeolite ore of Piojillo-Tasajera (Villa Clara).
- South of Camaguey.
- Guaos mines (Cienfuegos).
- Matahambre mines (Pinar del Río).

## **(c) Selection of favourable regions**

The selected regions are evaluated according to their socio-economic development, and their environmental and geological state to smaller scale (E-1:50 000). On this analysis the most favourable regions are selected for the location.

On the basis of the study and analysis of the geological-geophysical investigations executed in the identified regions and after a regional geological consideration, the following favourable sites have been evaluated:

1. Of the carbonated formations:

- calcareous rocks of the region Bolondrón (2).

2. Of the formations intrusive and extrusive:

- rocks ofiolitic of the region of Cajálbanas (1).
- rocks granodioritic and granitoide of the region of Manicaragua (3).
- rocks granodioritic and granitoide of the Sibanicú region - Victoria of the Tunas (5).

3. Of the saline formations:

- saline rocks of the Punta Alegre region (4).

The most favourable sites are shown in Figure D2.



*FIG. D2. Sites that are favourable for the emplacement of nuclear facilities.*

**(d) Selection of favourable sectors**

In these areas selected as the most favourable for the location of nuclear facilities, a series of complex geological investigations were carried out to detailed scale (E-1: 10 000) (geologic, engineer-geologic, geomorphology, hydrological, geophysical, etc.). The probability of extreme events is also evaluated (aeroplane fall, meteorites fall, fall free of an explosive load, sabotages, hurricanes, floods, etc.).

**6. Regions selected (Fig. D2)**

On the base of the study in detail of the geologist-geophysical conditions of the selected geologic formations and after an analysis of the current socio-economic development and perspective of each region, we have selected the following sectors:

- Rocks granodioritic and granitoides of the Manicaragua region;
- Rocks granodioritic and granitoides of the Sibanicú-Victoria Tunas region.



In both areas complex geologist-geophysical investigations have been carried out, sufficient to choose the most promising sections. For fundamental economic reasons, it was decided to continue the investigations in the sector corresponding to the rocks granodioritic and granitoid of the Manicaragua region (Cumanayagua), this site is located in a centralized area and nearer to the main supplying future of radioactive waste in the country (Nuclear Power Plant of Juragua).

#### **(e) Licensing proposal**

In accordance with the objectives and activities showed in the scheme of site selection, different investigation projects have been executed (geologic, engineer-geologic, hydrogeology and geophysical), to scales: of 1-10 000 and 1-5 000. This investigations have allowed proceeded according to the “Proposal of Area to Licence” with extension to 1×1 km. After that the scale is focussed to 1: 5 000, and then onto “Licence of the site”, which is the current status of the general investigation. The results of these investigations have, at the present time, focussed the area of investigations to 1.2 km<sup>2</sup>, as the most favourable sector.

#### **(f) General characteristics of the selected sector**

- **Geology.** The area is located in the granodiorite of the Cumanayagua place, belonging to the granitoid texture of the structure-facial area Manicaragua, where the granodiorite, like the main lithology appear, crossed by a complex of dikes whose thickness does not surpass 2m; these dikes are constituted by aplites and lamprofid. Inside the granodiorites appear small bodies of microdiorites, andesites and rocks anfibolitic, the directional textures little developed are and the rocks have a massive and granular aspect, although a system of cracks cut it in different addresses.
- **Tectonic.** The belt of granitoid of Manicaragua place in its group constitutes an independent unit separated from the neighboring units (anfibolites mabujina, complex vulcanogen-efusive of the Cretasic) for flaws of abrupt dip of stratum whose activity showed until the Superior Eocene. The tectonic intern is relatively simple, formed mainly by vertical normal flaws or subverticals that divide to the solid one in irregular polygons of variable surface (2-20 km<sup>2</sup>). Prevailing the tectonic alternative, motivated by the rigidity of the present rocks in the solid one, the cracking possesses subordinate addresses to the systems of main flaws (NNW and NE), also associating the complex of dikes that you/they cut the granodiorites, in depth the cracking diminishes until disappearing. A study seismologic of the central area of the country was made, in which was obtained for the selected sector with the use of historical data, engineer-geologic, geophysical investigations and seismologic, the project earthquake (100 years) of V degrees in the scale MSK and the maximum earthquake of calculation (10 000 years) of VII in the same scale.
- **Geomorphology.** The territory possesses a subjected complex relief to a long evolutionary process, a high difference geomorphologic evidenced by the presence of different exists you stagger morphostructurals with genesis of tectonic character. Due to the high impermeability of the solid the net hydrogeology is ramified with encased valleys and marked declines that exceed the 15°, the drainage this subordinate fundamentally to the net of existent cracking.
- **Topography.** The site is located in an area with heights between 50 and 70m on the level of the sea which eliminates the risk of possible floods or another event type, the

area also possesses favourable conditions of accessibility. The following units were defined in the area: – surfaces very low, plane, accumulative of alluvial origin with sand-muddy silts in the pleasures coming from the destruction of the granitic rocks. Bench marks among 30-50m; – surfaces low and wavy, erosive accumulative strongly drained for gulches of character intermittent, bench marks 50–60m; – surfaces of half levelling to high with character erosive. Bench marks 60–70m; – half slopes to strong and it constitutes a step morphostructural of character erosive-denudative. Bench marks 70-90m; – Surface high where the activity denudative is more intense, it diminishes thickness of the bark eluvio-deluvial. Bench marks 90–100 m.

- **Geophysics.** According to the results of the SEV the maximum depth to that can be the roof of the compact granodiorites or little fractured is of 15-30m, depth in which the resistive is of the order of 500 ohm/m or bigger. The structures in the area follow a main address with direction NW-SE.
- **Hydrogeology.** In the whole area of the investigations the phreatic complex of the igneous rocks (granodiorites) is developed of the Superior Cretasic, the aqueousness of the complex is poor (<1 l/s) and its permeability coefficient is of the order of  $10^{-2}$ - $10^{-3}$  m/d. The phreatic levels depend on the relief and they are to depths of 0.0-6.6m, the underground and superficial waters are not aggressive. The hydrogeology is not complex, the feeding is by means of the infiltration of the waters from precipitation; this aquifer does not become extensive for the whole area. The superficial waters run off into the intermittent streams until the lowest bench marks, that is to say, to the area receptor, the river Arima.
- **Geologic engineering.** There are five types of elements that contain the present lithology: (i) Sand without clay particles and fine grain; (ii) Loamy sand of grains means, with variable power until 5 m; (iii) Sand of thick grain with wide content of gravels, not possessing plasticity and reaches thickness of up to 20 m in areas of intense cracking or flaws; (iv) Fractured area and alteration of the rocks, composed by fragments of rocks and their power can end up reaching values of 10m; (v) Groups the solid one rocky, granodiorites of half grain appears with microdiorites inclusions and anfibolites. Their mechanical parameters are good (resistance to the compression of 620–682 kg/cm<sup>2</sup> and their depth is variable coming closer to the surface in the high areas <5 m, being deepened until more than 30 m.
- **Socio-economic factors.** The area is located in the municipality of Cumanayagua (27 km from Cienfuegos and approximately 3 km to the SW of the town of Cumanayagua)The economy is fundamentally agricultural, with prevalence of the citric fruit, being also of importance the cattle raising, appearing some smaller cultivations and cane of sugar.

## 7. Elemental and radionuclide concentration data

### 7.1 Sources of data

The distribution information of the radioelements (geochemistry maps) and the I<sub>γ</sub>, U(Ra), Th and K in form of iso-line maps for the whole country, are in the files of the National Centre of Mineral Resources, this information is to scale 1: 50 0000, annexed to the reports of the different gamma spectrometric airborne, but is not stored electronically. As such, a homogeneous map of the natural elements distribution does not exist in the Republic of Cuba.

Nonetheless there is information of the anomalous gamma spectrometric airborne data contained in each one of the reconnaissance carried out in the country and some data of the radioactive anomalies detected in surface and wells. An initial data survey of U, Ra, Th, K distributions and  $I_\gamma$ , which to characterize the values of the mentioned elements, giving the minimum, maximum values and the average of the total anomalous measurement for each county are available. The maximum and minimum values have been referred to terrestrial and logging well anomalies. The minimum values of  $I_\gamma$  (they correspond with the values of the normal bottom for each county), taken directly of measurement carried out in different field itineraries (using a scintillometer).

For the purpose of obtaining safety indicators, only limited information is available about the concentration of elemental and naturally occurring radionuclide NORM in the sites we selected for the repositories. For that reason, we used data from the specialized international literature taking into account the specific local characteristics.

Our host rock, is a massif granodiorite in the central part of the country in the province of Cienfuegos, all relevant information about the geosphere was obtained during the site selection process. We carried out geologic, hydro-geologic and geophysical investigation in order to obtain the most important parameter associate to the characteristics of the host medium. Those studies started in the early 1990s and were concluded in the stage of the site characterization and all they are mentioned in the document references.

For the repository radionuclide inventory, we use information about our real radioactive waste inventory, but according our nuclear policy, we will not dispose of high level waste in Cuba at the moment, therefore all radionuclide information is associated to low and intermediate level radioactive waste from nuclear applications in Cuba. In a first approach only the U, Th and K inventories in the repository and in the host rock, have been considered.

## **7.2 Summary of data**

### **(a) Principal site data**

The principal site is located in the Manicaragua granodiorite belt, in relation with sedimentary and other volcanic rocks, its surface area is about 225 km<sup>2</sup> and some regional studies suggest a depth of around 5 km, therefore taking into account a square shape the total volume of Manicaragua granodiorite is the order of 1125 km<sup>3</sup> and according its average density of 2650 kg/m<sup>3</sup> the total mass is the  $2.98 \times 10^{15}$  kg.

In the first stage of the study, only the use of NOR was evaluated for comparison purposes; therefore, all calculation are associated to uranium (total activity), <sup>232</sup>Th and <sup>40</sup>K; other elemental or radionuclide data will be used in next stages.

According to national and international investigations the average elemental concentrations in granodiorite rock are: U, 3.5 mg/kg; Th, 16.0 mg/kg; and K, 2000 mg/kg.

Taking into account the previous information, the total mass, for each element, in the granodiorite rock is: U,  $1.04 \times 10^{10}$  kg; Th,  $4.78 \times 10^{10}$  kg; and K,  $5.93 \times 10^{12}$  kg.

According to these data the largest amount is the K in the associated minerals of the granodiorite rock.

The next stage was to obtain the activities for the radionuclides in the selected environment; we used the data from [D4] in Table D3.

TABLE D3. CHARACTERISTICS OF THE RADIONUCLIDES

Radionuclide	Isotopic abundance	Specific activity Bq/g
$^{238}\text{U}$	0.9927	$1.24 \times 10^4$
$^{235}\text{U}$	0.00720	$7.11 \times 10^4$ <sup>(b)</sup>
$^{234}\text{U}$	0.000056	$2.30 \times 10^8$
$^{232}\text{Th}$	1 <sup>(a)</sup>	$4.1 \times 10^3$
$^{40}\text{K}$	0.000118	$2.09 \times 10^5$ <sup>(c)</sup>

(a) All Th is assumed to be  $^{232}\text{Th}$ .

(b) A more accurate value is  $8.00 \times 10^4$  Bq/g.

(c) A more accurate value is  $2.60 \times 10^5$  Bq/g.

According to this information the activity in the granodiorite massif is:  $(^{238}+^{235}+^{234})\text{U}$ , 90 Bq/kg;  $^{232}\text{Th}$ , 65.6 Bq/kg;  $^{40}\text{K}$ , 51.83 Bq/kg.

A useful alternative approach in order to avoid inappropriate comparison is to evaluate the total activity contained in a uniform volume of rock, the RERV. For the Cuban repository conceptual design this corresponds to a volume of 385,152 m<sup>3</sup>. For this equivalent rock volume, we also make calculations of activities.

The total mass for each element in the RERV is: U, 3570 kg; Th, 16300 kg; and K,  $2.04 \times 10^6$  kg.

And the total activity in the RERV is:  $(^{238}+^{235}+^{234})\text{U}$ ,  $1.55 \times 10^5$  Bq;  $^{232}\text{Th}$ ,  $1.07 \times 10^6$  Bq; and  $^{40}\text{K}$ ,  $1.06 \times 10^8$  Bq.

By comparison, the repository inventory data are:  $^{235}\text{U}$ ,  $1.1 \times 10^{10}$  Bq;  $^{238}\text{U}$ ,  $2.0 \times 10^9$  Bq;  $^{234}\text{U}$ ,  $3.5 \times 10^8$  Bq;  $^{232}\text{Th}$ ,  $1.2 \times 10^{10}$  Bq;  $^{40}\text{K}$ ,  $2.3 \times 10^8$  Bq.

According the repository volume, the radionuclide activities are:  $^{238}\text{U}$ ,  $5.1 \times 10^3$  Bq/m<sup>3</sup>;  $^{235}\text{U}$ ,  $2.86 \times 10^4$  Bq/m<sup>3</sup>;  $^{234}\text{U}$ ,  $9.0 \times 10^2$  Bq/m<sup>3</sup>;  $^{232}\text{Th}$ ,  $3.10 \times 10^4$  Bq/m<sup>3</sup>;  $^{40}\text{K}$ ,  $5.97 \times 10^2$  Bq/m<sup>3</sup>.

## (b) General data

The results show the concentrations (U, Ra, Th, K) ranges of the natural radionuclides associated fundamentally to the variation in the lithology and geographical area of the country. In Cuba, the higher concentration (ppm) of U and Ra are referenced to the Central region associated to Skarn, while for Thorium (ppm) and Potassium (%), in the East region has been found the concentration peaks in Tuffs. The concentrations ranges obtained are preliminary, they characterise the behaviour of this parameter for the Cuban geology, but they do not represent limits for safety assessment purpose, yet. Also, should take into account other factors as the assessment context, time scales and others assumptions before to establish the final concentration limits for the natural radionuclides as a radiological and nuclear safety performance indicator complementary dose and risk for safety assessment for radiological and nuclear facilities.

Table D4 shows the data collected in all Cuba, wide U, Ra, Th, K (ppm and %) distributions and  $I\gamma$  ( $\mu\text{R/h}$ ), giving the minimum, maximum values and the background of the total anomalies measured in the different region (West, Central and East) of the Republic of Cuba, see Figure D1. The maximum and minimum values have been referred to terrestrial, logging well, samples and anomalous gamma spectrometric airborne, carried out in the different region of the Republic of Cuba. The minimum values of  $I\gamma$  correspond with the normal background for each region, taken directly of measurement carried out in different field itineraries.

TABLE D4. DISTRIBUTION OF ANOMALIES OF  $I\gamma$ , U, RA, TH AND K IN THE SURFACE OF THE EARTH; LOGGING WELL, BOREHOLE AND AIRBORNE GAMMA SPECTROMETRIC DATA

Radioactive elements		Region					
		West		Centre		East	
		Aerial	Terrest.	Aerial	Terrest.	Aerial	Terrest.
$I\gamma$ ( $\mu\text{R/h}$ )	Minimum	0.66	5	0.54	4.6	0.60	5.75
	Maximum	5.25	643.3	3.31	268.00	5.59	11.25
U (ppm)	Minimum	0.73	0	1.40	1.00	0.93	1.25
	Maximum	9.6	1182	9.70	428.2	15.89	72.5
	Background	4.30	80.88	3.51	92.1	0.91	15.95
Ra (ppm)	Minimum	-	22	-	2.00	-	2.00
	Maximum	-	1067	-	917.00	-	87.5
	Background	-	71.53	-	191.53	-	13.89
Th (ppm)	Minimum	0.96	0	0.78	0	0.37	1.91
	Maximum	19.1	29.50	7.70	47.5	61.98	41.00
	Background	6.86	8.37	2.27	8.95	9.65	13.83
K (%)	Minimum	0.26	0	0.28	0.05	0.31	0.1
	Maximum	2.03	3.75	2.51	3.42	5.17	5.3
	Background	0.50	0.99	0.54	1.48	1.13	2.23
Studied lithologies		Limestone, Sandstone, Shale and Lutites		Limestone, Serpentine, Skarn, Granite, Tuff, and Shale		Limestone, Shale, Tuff, Granite and Serpentine	

In the West Cuba, the biggest contribution is associated to the sedimentary rocks. Also, the different regions of the country differ in their results, for example, for the Uranium and the Ra, of the West and Central region had the highest values in the country, quite differentiated from the values of the East region, associated fundamentally to the limestone and skarn respectively.

For Th and K there are not important variations among the data of the three regions, those results are an image of the different geology present in the areas. As a result of the studies, in the whole country, the concentration distribution for the natural radioactive elements was obtained, associated to the main present lithologies. The variation of the concentrations is associated to the changes in the different lithologies, genesis, etc.

TABLE D5. NATURAL RADIONUCLIDE CONCENTRATIONS FOR ROCK TYPES

Radioactive elements		Main lithologies								
		Regions								
		West			Central		East			
		Limestone	Lutite	Shale	Skarn	Limestone	Shale	Granite	Limestone	Tuff
<i>U (ppm)</i>	<i>Min.</i>	1	33	5	2	8	50	5	2	5
	<i>Max.</i>	673	488	229	3445	160	63	29	14	184
<i>Ra (ppm)</i>	<i>Min.</i>	5	32	5	7	6	26	2	N/A	2
	<i>Max.</i>	577	490	134	2500	50	628	22	N/A	129
<i>Th (ppm)</i>	<i>Min.</i>	0	1	0.14	0.30	0	2.5	5	0.13	2
	<i>Max.</i>	20	20	28	150	150	16	48	202	202.7
<i>K (%)</i>	<i>Min.</i>	0	0.10	0.10	0	0	0.50	0.30	0	0.2
	<i>Max.</i>	4	2	3.50	5	5	4	8.40	1.5	13.86

Table D5 shows the results according the different types of lithologies present in the country and the ranges of variation for the concentration for each element. The concentrations obtained characterise for regions of the country, the main rock associate to this indicator, which are the main existent types in the Cuban geology.

According the Cuban geology where the sedimentary rocks are majority, the limestone is included in the three regions, in the West it possesses the maximum concentration values for U and Ra; for Th and K the relative peaks are located in the East and Central respectively. All these variations respond to the different geologic histories of the evaluated regions. For the igneous rocks, the granites possess relatively low values of natural radioelements concentration; the tuffs have interesting maximal values for Th and K. The metamorphic rocks included the Skarn. The skarn rock has the higher concentration of U and Ra of natural occurrence in Cuba, which are associated to an anomalous area of the Central of the country (taking account this U concentration high values, have been done 10 new assessment in order to prove it and the result was the same, see Table D6). The shale, in the West and Central regions possesses important values of concentration in Ra and U.

### (c) Next stages

After this preliminary stage of data collection, analysis of the results has been initiated in order to establish the concentration of natural radioelements as a safety indicator. It will be evaluated if the obtained values of concentrations (fundamentally the upper limits), can be defined as limits of concentration environment for a region or area, taking into account in a qualitative and quantitative way the occurrence of a negative impact in the environment and man.

The obtained data will be incorporated into the methodology for radioactive waste disposal facility in Cuba. These safety indicators will be the assessment endpoints supporting the dose and risk in the methodology. The assessment should take account the specific characteristics of the geological conditions, design of the nuclear facilities, etc. Finally, it will be established, as a result of the assessment, the ranges of variation of the concentration of the natural radioactive elements by lithologic types in the Cuban environment. The obtained indicators will allow their incorporation inside the methodology of the safety assessment, in the stage of evaluation of the results as comparison measure to evaluate the possible impact of a nuclear practice. These indicators will establish the limits (concentration) of the natural occurrence of radioelements for different rock types; which should be fulfilled, in certain ranges to guarantee that the nuclear practice has not negative impact in the environment and the man.

TABLE D6. NEW ASSESSMENT ABOUT HIGHER VALUES

<i>Sample number</i>	<i>U Concentration (ppm)</i>	<i>Standard deviation</i>	<i>Confidence interval</i>
1	3390.00	112.00	4.00
2	3560.00		
3	3280.00		
4	3260.00		
5	3600.00		
6	3460.00		
7	3530.00		
8	3450.00		
9	3420.00		
10	3500.00		
<i>Average 3445.00</i>			

## 8. Elemental and radionuclide flux data

### 8.1 Sources of data

There is a national database about the fluxes data, which take into account river flow rates, and the aquifer fluxes. The National Research Institute of Water Resource operates a comprehensive monitoring system for both flow and water quality using a network of over 73 gauging stations. This flux data, together with information from other sources, is held in the National Office for Mineral Resource, which stores all information about the geology and hydrology investigation. The archive contains a broad range about this important information in order to can obtain relevant evaluation of the fluxes.

The main data sources are the materials from the site selection process for the disposal facility and from other relevant national organizations, some of them are institutions associated to geosciences, water resources, and agricultural topics. There are some lack of information for certain elemental and specific site parameters, in this case, we used data from international references, previous a technical review.

## 8.2 Summary of data

There are no specific investigations about erosion rate in granodiorite in Cuba, therefore generic international data are applied. According to the Cuban geology, the climate and the topographical aspects an average erosion rate of 5 m/My seems appropriate. For 80 km<sup>2</sup> of exposed rock, the total mass of material eroded is  $1.06 \times 10^{12}$  kg/year, that means a flux due to erosion of  $1.325 \times 10^4$  kg/km<sup>2</sup>/year. The main fluxes associated to rock erosion are summarized in the Table D7.

TABLE D7. FLUXES DUE TO GRANODIORITE EROSION

Species	Mass flux for total kg/yr	Specific mass flux kg/km <sup>2</sup> /yr	Activity flux for total Bq/yr	Specific activity flux Bq/km <sup>2</sup> /yr
<b>U</b>	3.71	0.05	—	—
<b>Th</b>	16.9	0.21	—	—
<b>K</b>	2120	26.6	—	—
<b>U chain</b>	—	—	$4.6 \times 10^7$	$5.75 \times 10^5$
<b>Th chain</b>	—	—	$8.6 \times 10^5$	$1.07 \times 10^4$
<b><sup>40</sup>K</b>	—	—	$6.67 \times 10^5$	$8.3 \times 10^3$

The other important natural process causing elemental fluxes that are taken into account is the groundwater flow, according to the site specific data and some parameters from international references. The hydraulic parameters are defined in Table D8.

TABLE D8. GRANODIORITE HYDRAULIC PARAMETERS

Rock	Hydraulic conductivity (m/s)	Porosity	Gradient	Groundwater flux (l/m <sup>2</sup> /yr)	Groundwater velocity (m/yr)
<b>Granodiorite</b>	$1.1 \times 10^{-7}$	0.1	0.02	$4.1 \times 10^{-4}$	7.2

Elemental concentrations in the groundwater in the site are:

U, 3.0 mg/l; Th, 2.1 mg/l; K, 4.0 mg/l.

The specific groundwater flux is:

U,  $1.23 \times 10^{-3}$  kg/km<sup>2</sup>/yr; Th,  $8.61 \times 10^{-4}$  kg/km<sup>2</sup>/yr; K,  $1.64 \times 10^{-3}$  kg/km<sup>2</sup>/yr.

The activity flux from groundwater discharge is:

U,  $15.25 \times 10^3$  Bq/km<sup>2</sup>/yr; Th,  $14.12 \times 10^3$  Bq/km<sup>2</sup>/yr; K,  $34.28 \times 10^4$  Bq/km<sup>2</sup>/yr.

For calculation of the flux data from the repository, the ISAM methodology [D5] was followed. The calculated specific activity flux is shown in Table D9.



TABLE D9. RESULTS CALCULATION OF SPECIFIC ACTIVITY FLUX

Radionuclide	Specific activity flux Bq/km <sup>2</sup> /yr
<sup>238</sup> U	$9.0 \times 10^4$
<sup>235</sup> U	$4.8 \times 10^2$
<sup>234</sup> U	43
<sup>232</sup> Th	39
<sup>40</sup> K	$5.6 \times 10^{-2}$

## 9. Interpretation and application of the concentration and flux data

Simple comparisons of mass and activities for the rock and the repository and the same for the flux associated to natural process in the granodiorite and the normal release scenario in the repository are made in Table D10.

TABLE D10. ACTIVITY COMPARISON

Radionuclides	Total activity RERV, Bq	Total activity repository, Bq
U chain	$1.55 \times 10^5$	$1.30 \times 10^{10}$
<sup>232</sup> Th	$1.07 \times 10^6$	$1.20 \times 10^{10}$
<sup>40</sup> K	$1.06 \times 10^8$	$2.30 \times 10^8$

The simple activity comparison between the granodiorite and the repository, taking into account the RERV, showed that the total activities in the repository are several orders bigger than the granodiorite activities. The main reason of this difference is associated, that the repository is designed for low and intermediate level and the adopted inventory (in this preliminary approach) is for high waste level repository. In the next stage, it is planned to carry out an assessment with a more realistic inventory according to the repository size.

The comparison between activity flux from granodiorite and the repository flux, the last one was obtained by calculations of the model for normal release scenarios, which only included natural process relevant for the repository performance; Table D11 summarizes these results.

TABLE D11. FLUX COMPARISON

Species	Activity flux, Bq/yr		Specific activity flux, Bq/km <sup>2</sup> /yr	
	Granodiorite	Repository	Granodiorite	Repository
U chain	$4.6 \times 10^7$	$1.1 \times 10^{-1}$	$5.75 \times 10^5$	$9.0 \times 10^4$
Th chain	$8.6 \times 10^5$	$4.2 \times 10^{-5}$	$1.1 \times 10^4$	$3.9 \times 10^2$
<sup>40</sup> K	$6.7 \times 10^5$	$4.8 \times 10^{-8}$	$8.3 \times 10^3$	$5.6 \times 10^{-2}$

The comparison showed that the values of activity fluxes from the repository are lower than the flux from the host rock. In this case, the main difference is the activity comparison; the repository barrier plays an important role as a flux controller, despite, the fact that the disposal concept is not designed for high level waste. The wasteform, the engineered barriers, and the important role of the rock (granodiorite) with poor hydrogeology conditions, water movement associated to fracture and faults, was the principal reason of these results.

The preliminary results showed the possibility to use other safety indicator to support the safety assessment activities, these complementary endpoints are useful for long term assessment and are less influenced by human factors. In next stages, we will evaluate other elements and radionuclides present in our specific inventories, we will carry out more specific site investigation in order to reduce the data uncertainty and we use the safety assessment methodology with dose and other safety indicators to derive acceptance disposal criteria.

## **10. Conclusions**

The main conclusions reached are:

- A wide-ranging database was obtained in three Cuban regions, which characterise the natural elemental radioactive concentration and the range of variation for the lithologic types studied as a preliminary stage in order to use it in the safety assessment.
- In Cuba the main lithologies relevant to the study are limestone, lutites, skarn, shale, granite and tuff.
- The concentrations of U vary in a wide range from 0.5 to 3445 ppm; the Ra from 2 to 2500 ppm; the Th from 0 to 203 ppm; and the K from 0 to 14%. For all regions, the Central region has the rock (skarn) with the highest concentration values for U and Ra.
- The studies carried out, evaluated for the first time in Cuba, the concentrations as a safety indicator for safety assessment for radioactive waste disposal facilities.
- The inventories used in the activity comparison are for high level waste, while the facility design is for low and intermediate level waste, therefore, their volume is smaller than the repository mentioned. Thus, the activity values to be compared for RERV are bigger than the obtained from the granodiorite rock. On the other hand when the activity flux from the granodiorite rock and the repository are compared, the values of the latter are smaller, because, in this case the different engineered barriers used and the favourable site begin to play their useful role, which reduces the radionuclides discharges from the repository.

## **Recommendations**

- Introduce a methodology, in order to establish the validity of the results obtained in the data collecting stage and so to establish the variation limits of the concentration of the natural radioactive elements in the environment as safety indicators.
- At these stages, consideration will need to be given to the presentation of natural safety indicators in a manner which is scientifically credible, but also understandable by non-technical audiences.
- Natural safety indicators are considered to be complementary to dose and risk; but they do not replace dose and risk with natural safety indicators.
- Complete the database with information of Havana area, in order to cover the whole country.
- Include the evaluation of other elements and radionuclides present in the specific inventories. More specific site investigation in order to reduce the data uncertainty and the use of safety assessment methodology with dose and other safety indicators to derive acceptance disposal criteria.

### **Additional information sources**

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- [D2] PERALTA VITAL, J.L., “Solución segura al Sistema nacional de aislamiento de desechos radiactivos. Reducción de su impacto en el medio ambiente Cubano”, Master’s Thesis. ISPJAE, CPHR, Havana (2000).
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- [D5] INTERNATIONAL ATOMIC ENERGY AGENCY, IAEA. “International Atomic Energy Agency. Safety Assessment Methodologies for Near Surface Disposal Facilities. Results of a Coordinated Research Project. Volume I: Review and enhancement of safety assessment approaches and tools; Volume 2: Test cases.



## APPENDIX E

### CZECH REPUBLIC

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#### Abstract

The safety indicators can provide valuable information concerning long-term safety of deep geological repository. Therefore, the research focused on safety indicators forms a part of the Czech deep disposal programme. It is supposed that around 3,720 t of spent nuclear fuel will be produced in the Czech Republic. The concept of deep disposal is based on granitic host rock using bentonites as buffer/backfill material. Brief geology of the Czech Republic is presented in the article. Three main sources were used for collection of reference data on concentration and fluxes of natural radionuclides - Geofond (hydrogeological objects), State Office for Nuclear Safety (potable water monitoring system - radionuclides) and Czech Hydrometeorological Institute (groundwater and surface water quality system). Large dataset was collected demonstrating existence of many rock bodies with radioactive element concentration above average values. This phenomenon also generally results in higher content of radionuclides in groundwater, radon in air and finally in radiation exposure of population (3.67 mSv/year). Extraordinary concentration of radioactivity is illustrated at the example of Joachimstahl radon spa waters (units of kBq/l). The most portion of data are available in a form of gross alpha and beta activities; data about individual radionuclides are much rare. Relevant data about past uranium production are specified. Finally, information about legislatively-determined limits of radionuclides in building materials and potable water are presented.

#### Background

##### Radioactive wastes

The radioactive waste streams can be divided into two main groups:

1. Waste from nuclear power plants (NPPs)
2. Waste from non-nuclear industry, medicine, research and universities – institutional area

In relation to the production of materials and waste containing radionuclides, waste can also be added from the production of uranium (mining and milling) and NORM (naturally occurring radioactive materials) produced in various plants and industries.

Two nuclear power plants are in operation in the Czech Republic. Four reactors of Dukovany NPP (WWER 440 MW) were put in operation in 1985 – 1987. Dukovany NPP with 2 reactors (WWER 1000 MW) is being put into operation now. The total installed capacity of electricity production is 15.3 GWe (from which 2.7 GWe are nuclear). Liquid radioactive waste is solidified by bitumenization and solid waste is (super) compressed to reduce volume. Standard disposal unit is 200 l steel barrel. Produced spent nuclear fuel at NPPs (Dukovny NPP – 43 t HM/y, Temelin NPP – 85 t HM/y) is stored in casks. Projections of total production of radioactive waste based on 40 years of operation of NPPs are summarized in Table E1.

TABLE E1. PROJECTIONS OF RADIOACTIVE WASTES IN THE CZECH REPUBLIC

Nuclear Power Plant	Processed operational radioactive waste (m <sup>3</sup> )	Spent nuclear fuel (t HM)	Processed radioactive waste from decommissioning (m <sup>3</sup> )	
			LLW/ILW	HLW
Dukovany NPP	10 250	1937	6000	2000
Temelin NPP	12 000	1787	4600	620
TOTAL	22 250	ca. 3720	10600	2620

Now there are approximately 150 smaller radioactive waste producers in the Czech Republic (most of waste is produced in NRI Rez). Radioactive waste of small producers is centrally treated and processed in NRI. Liquid radioactive waste (approx. 150 m<sup>3</sup>/y) is evaporated to reduce volume and cemented. Solid radioactive waste (20–30 m<sup>3</sup>/y) is mainly compressed by low pressure facility. Several units of spent sealed sources (mainly Cs, Co, neutron and radium sources) are also produced in a year along with hundreds of fire alarm sources (Am) that are embedded in cement matrix. Standard disposal unit of institutional radioactive waste is 200 l steel barrel. There are also three research nuclear reactors in the Czech Republic, two of which are in the Nuclear Research Institute Rez. Spent nuclear fuel from these reactors is stored in wet storage facility constructed in NRI Rez.

### Legal and institutional background

The radioactive waste management system is based on four main cornerstones:

- Legal system is consisting of Atomic Energy Act (1997, as amended in 2002) and many Regulations of SONS. Legislative norms are fully compatible with ICRP 60, IAEA Radiation Protection Standards and European Directive 96/29/Euratom.
- Independent regulatory body in the area of nuclear safety and radiation protection - State Office for Nuclear Safety (SONS).
- State organization responsible for final disposal of spent nuclear fuel and radioactive waste on the territory of the Czech Republic - Radioactive Waste Repository Authority (RAWRA). RAWRA is controlled by Board of RAWRA consisting of representative ministries, producers and municipalities impacted by disposal.
- Nuclear Account for covering all liabilities connected with disposal spent nuclear fuel and radioactive waste.

### Disposal facilities

Excluding the Hostim subsurface repository operated in 1950s and 1960s (currently finally sealed), there are 3 near-surface repositories in operation:

- Dukovany surface repository (operated since 1994) situated at the Dukovany NPP site used for operational radwaste from NPPs. The repository is composed of 2 double-rows of ferro-concrete vaults (6 × 6 × 18 m) with the total capacity 55 000 m<sup>3</sup> (130 000 drums).

- Richard and Bratrstvi subsurface repositories situated in abandoned limestone and uranium mines, respectively, used for disposal of institutional radioactive waste. The basic data are summarized in Table E2.

TABLE E2. DATA ON THE CZECH DISPOSAL FACILITIES

Repository	Commencement of operation	Disposed waste		
		Activity (Bq)	Volume (m <sup>3</sup> )	Typical radionuclides
Richard	1964	2x10 <sup>16</sup>	2,700	<sup>137</sup> Cs, <sup>60</sup> Co, <sup>90</sup> Sr, <sup>3</sup> H, <sup>14</sup> C,
Bratrstvi	1972	1x10 <sup>12</sup>	250	<sup>226</sup> Ra, <sup>232</sup> Th

Deep geological repository is in development. The current concept is based on direct disposal of spent nuclear fuel in steel disposal containers to granitic host rock. Bentonitic materials would be used as a buffer, backfill and sealing materials. Currently, 8 regions in granitic bodies were selected for future geological investigations. Deep repository would be put in operation in 2065 according to the generally defined time schedule of development (approved by the Government in the State Policy in 2002).

## Geology

### Geology of the Bohemian massive

The territory of the Czech Republic of 78 863 km<sup>2</sup> area consists of two regional geological units. The Bohemian Massif builds predominant part of the Czech Republic; to its eastern part extend the Western Carpathians (Figure E1). The Bohemian Massif belongs to the Meso-Europe zone, which was consolidated in the late Palaeozoic period during the Hercynian (Variscan) orogeny. The Bohemian Massif represents the easternmost known part of the Hercynian branch.

The Bohemian Massif consists basically of the following three units:

1. Precambrian (Cadomian) basement
2. Crystalline and Palaeozoic rocks affected by Hercynian orogeny
3. Post-Hercynian platform cover.

### Precambrian (Cadomian) basement

Precambrian basement not affected by the later Hercynian orogeny occurs in the form of various metamorphosed rocks on the eastern territory of the Czech Republic (Brunovistulicum unit).

### Crystalline and Palaeozoic rocks affected by Hercynian orogeny

This unit includes metamorphosed pre-Cambrian and Palaeozoic rocks and non-metamorphosed rocks of Cambrian to Middle Devonian age.

Complexes of metamorphosed rocks (phyllites, schists, gneisses, migmatites) situated mainly in S part of the Czech Republic prevail in this unit. Elements of Cadomian metamorphism appear in most of these units. The Hercynian regional metamorphism is the most significant feature on the territory of the Bohemian Massif.

Non-metamorphosed formations of Cambrian to Middle Devonian age occur in several separated basins (Prague area). Shales, limestones, sandstones and conglomerates, often accompanied by volcanism, predominantly build the formations.

Cadomian and Hercynian plutonic rocks are numerous. These plutonic complexes can be distinguished to the following types:

- metaplutonites of uncertain age, which occur predominantly in the form of orthogneiss predominantly in marginal parts of the Bohemian Massif;
- plutonites associated with Cadomian orogeny, which forms several granitoid and gabbroid bodies in the W and N parts of the Czech Republic;
- plutonites (almost only granitoids) associated with Hercynian orogeny, which are the most frequent on the territory of the Bohemian Massif. Granitoids form 2 main bodies: the Central Bohemian Plutonic Complex (area of 3200 km<sup>2</sup>) and the Moldanubian Plutonic Complex (an area of approximately 8000 km<sup>2</sup>).

#### **Post-Hercynian platform cover.**

Post-Hercynian platform rock bodies can be divided according to age of formation into the following units:

- Permo-Carboniferous sediments occurring in 2 different formations (paralic and limnic).
- Mesozoic sediments — The absolute majority of Mesozoic sediments in the territory of the Czech Republic are of Upper Cretaceous age occurring in 2 separate basins (Bohemian Cretaceous Basin is the most important with the area of 14 000 km<sup>2</sup>).
- Tertiary sediments are mostly of lacustrine origin. The sediments occur in the NW part of the Czech Republic (where they are accompanied by alkaline volcanism of Tertiary and Quaternary age) and also in the NW part where there are exploited lignite seams, which are bound to these sediments.



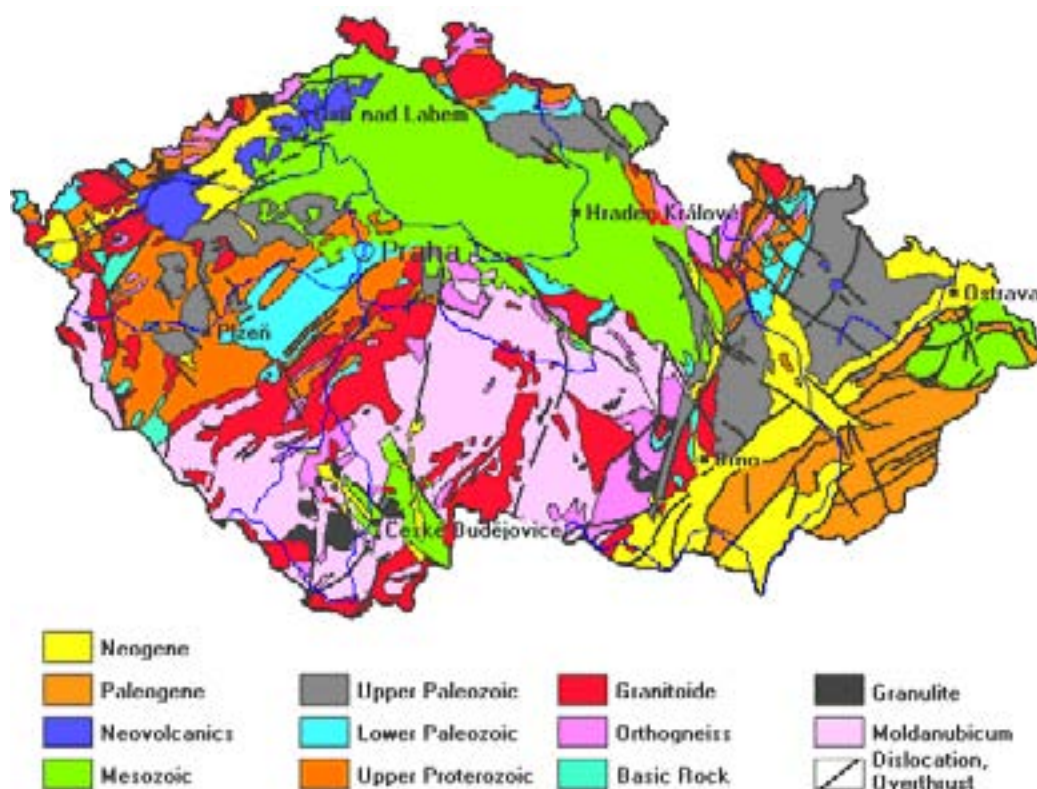


FIG. E1. Simplified geological map of the Czech Republic.

## Elemental and radionuclide concentration data

### Sources of data

Radioactive elements distribution in various geological units and rock formations were analysed by various geological companies, research and university institutes. Some of these results were published and are available in open literature. Data mainly from surveillance surveys (surface surveillance, boreholes) were centrally collected by Geofond (state institution responsible to Ministry of Environment) and are available in the forms of reports and databases.

Compilation of information about concentration of radioactive elements and radionuclides in water for the Czech Republic was done on the basis of three data sources:

1. Data from hydrogeological objects (boreholes, wells) collected by Geofond
2. Data finally collected by State Radiation Protection Institute (SONS) according to requirements of Atomic Energy Act — water (both groundwater and surface water) used for drinking purposes [E1]. Quality of water supplied to public system or packed water is observed from radiological point of view. Water is in many cases treated.
3. Data collected by Czech Hydrometeorological Institute (CHMI) as indicator of general quality of water in the Czech Republic. All main geological units are covered by monitoring system with special focus on Cretaceous Basin as a main water reserve. Deeper, shallow (mainly located in Quaternary sediments), springs and surface waters are monitored along with river sediments [E2-E4].

## Summary of data

### Radionuclides in the rocks

Concentrations of radioactive elements in the selected rocks are summarized in the following Table E3 (selected data focused on regions proposed for a deep repository) [E5].

TABLE E3. RADIOELEMENT CONCENTRATIONS IN ROCKS RELEVANT TO A DEEP REPOSITORY

Rock Type (massif)	K	U	Th
	% K	ppm eU	ppm eTh
Porphyric granite (Borsky massif)	4.1	6.1	19.9
Migmatites (sumavske moldanubikum unit)	2.5	4.4	12.0
Granulites (sumavske moldanubikum unit)	2.9	1.9	6.9
Granodiorites (sumavske moldanubikum unit)	3.3	6.1	18.0
Granite (Chvaletice Massif)	3.5	6.9	17.1
Granodiorite (Kladrub Massif)	3.2	3.8	10.6
Porphyric biotitic granite (Sedmihori Massif)	3.9	6.1	30.9
Two-mica adamellite (Sedmihori massif)	3.5	4.3	11.2
Granodiorite - Merklin type (Stod massif)	1.9	3.7	8.2
Granodiorite (Stenovice Massif)	2.1	4.5	13.6
Granite - mountains type (Carlsbad Massif)	3.6	4.0	27.3
Granite – Erzgebirge type (Carlsbad Massif)	4.2	14.2	22.0
Granites (Kynzvalt –Zandov Massif)	3.8	7.2	15.8
Granite (Tis Massif)	1.5–2	2-4	6-8
Granodiorite (Cista – Jesenice Massif)	1–1.5	3-5	8-14
Granodiorite (Buk site, Central Bohemian Plutonic Complex)	1.9	3.6	13.4
Granodiorite (Konetopy site, Central Bohemian Plutonic Complex)	2.5	4.2	16.4

### Radionuclides in water compartments

Data for shallow groundwater (Geofond source) from the most important granite bodies are summarized in the following Table E4.

TABLE E4. RADIOELEMENT CONCENTRATION DATA FOR SHALLOW GROUNDWATER IN GRANITES

	Maximum	Minimum	Median
K (mg/l)	78.3	0.4	2.9
U (mg/l)	0.02	0.0001	0.0015
Ra-226 (Bq/l)	20	0.005	0.05
Rn-222 (Bq/l)	8624	1	175
Gross alpha (Bq/l)	14	0.01	0.1
Gross beta (Bq/l)	4.4	0.05	0.195

Data for water used in public supply system (SONS source) are summarized in the following Table E5 (Bq/l if not indicated otherwise).

TABLE E5. RADIOELEMENT DATA FOR PUBLIC SUPPLY WATERS

	No. of samples	Geom. average	Mean	Maximum
gross alpha	2,611	0.046	0.0770	3.020
gross beta	2,580	0.087	0.114	2.150
Rn-222	2,666	15.5	37.0	2214
U (mg/l)	177	0.003	0.0058	0.120
K-40	195	0.041	0.0563	0.850
Pb-210	6	0.025	0.0281	0.035
Po-210	8	0.016	0.0213	0.071
Ra-224	4	0.079	0.0858	0.140
Ra-226	169	0.023	0.0405	0.270
U-234	13	0.162	0.482	1.560
U-235	13	0.006	0.0162	0.038
U-238	31	0.053	0.162	1.080

Data for groundwater from deeper boreholes (CHMI source) are summarized in the following Table E6.

TABLE E6. RADIOACTIVITY DATA FOR DEEPER GROUNDWATERS

Gross alpha (Bq/l)			Gross beta (Bq/l)		
Median	Range	No. of samples	Median	Range	No. of samples
0.16	0.019–615	786	0.32	0.08–11.1	192

Data for groundwater from shallow boreholes (mainly situated in Quaternary sediments) are summarized (CHMI source) in the following Table E7.

TABLE E7. RADIOACTIVITY DATA FOR SHALLOW BOREHOLE WATERS IN QUATERNARY SEDIMENTS

Gross alpha (Bq/l)			Gross beta (Bq/l)		
Median	Range	No. of samples	Median	Range	No. of samples
0.14	0.03–8.3	632	0.39	0.05–3	120

Data for water from springs (CHMI source) are summarized in the following Table E8.

TABLE E8. RADIOACTIVITY DATA FOR SPRING WATERS

Gross alpha (Bq/l)			Gross beta (Bq/l)			Rn-222 (Bq/l)		
Median	Range	No. of samples	Median	Range	No. of samples	Median	Range	No. of samples
0.09	0.02–3.2	490	0.13	0.05–1.27	81	72	6–1330	62

Data for surface water — rivers and streams — (CHMI source) are summarized in the following Table E9 (the results are not fully representative due to statistical treatment).

TABLE E9. RADIOACTIVITY AND RADIOELEMENT DATA FOR SURFACE WATERS

	<b>Median</b>	<b>No. of samples</b>
Gross alpha-total (Bq/l)	0.237	2044
Gross alpha - soluble (Bq/l)	0.152	2138
Gross alpha - suspended (Bq/l)	0.059	2113
U - total (µg/l)	6.0 (range of 0.2 - 2000)	
U - soluble (µg/l)	4.0 (range of 0.1 - 2000)	
U - suspended (µg/l)	1.0 (range of 0.1 - 184.4)	
Ra-226 - soluble (Bq/l)	0.018	6634
Ra-226 - suspended (Bq/l)	0.001	1804
K-40 total (Bq/l)	0.137	1164
K-40 - soluble (Bq/l)	0.130	7189
Gross beta (Bq/l)	0.231	2521
Gross beta - soluble (Bq/l)	0.203	9953
Gross beta - particulate (Bq/l)	0.049	5173
Gross beta after correction for K-40 (Bq/l)	0.025	9041

Figure E2 shows the quality of Czech Republic river waters according to the content of radioactive elements (blue: the best; red: the worst quality).

Data for selected representative samples of mineral and balneological waters (data source from Czech Inspectorate of Spa and Springs — an organization within the responsibility of the Ministry of Health) are summarized in the following Table E10.

TABLE E10. RADIOACTIVITY DATA FOR MINERAL AND BALNEOLOGICAL WATERS

Spring	Rn-222 (Bq/l)	Ra-226 (Bq/l)	U (mg/l)
Matttoni	4	0.25	0.009
Podebradka	< 0.5	0.31	< 0.002
Hanacka kyselka	19	0.47	0.0056
Korunni	5.8	< 0.05	< 0.001
Rudolfuv pramen (curing)	14.4	0.05	< 0.001
Ida	47.6	< 0.05	< 0.002
Teplice spa			
Pravridlo	258	0.33	0.008
Hynie	65	0.15	0.002
Horsky	1865	0.53	0.6 - 1

Mineral water (balneological use) data from Joachimstahl radon spa, as an example of groundwater with very high radioactive content, are shown in Table E11.

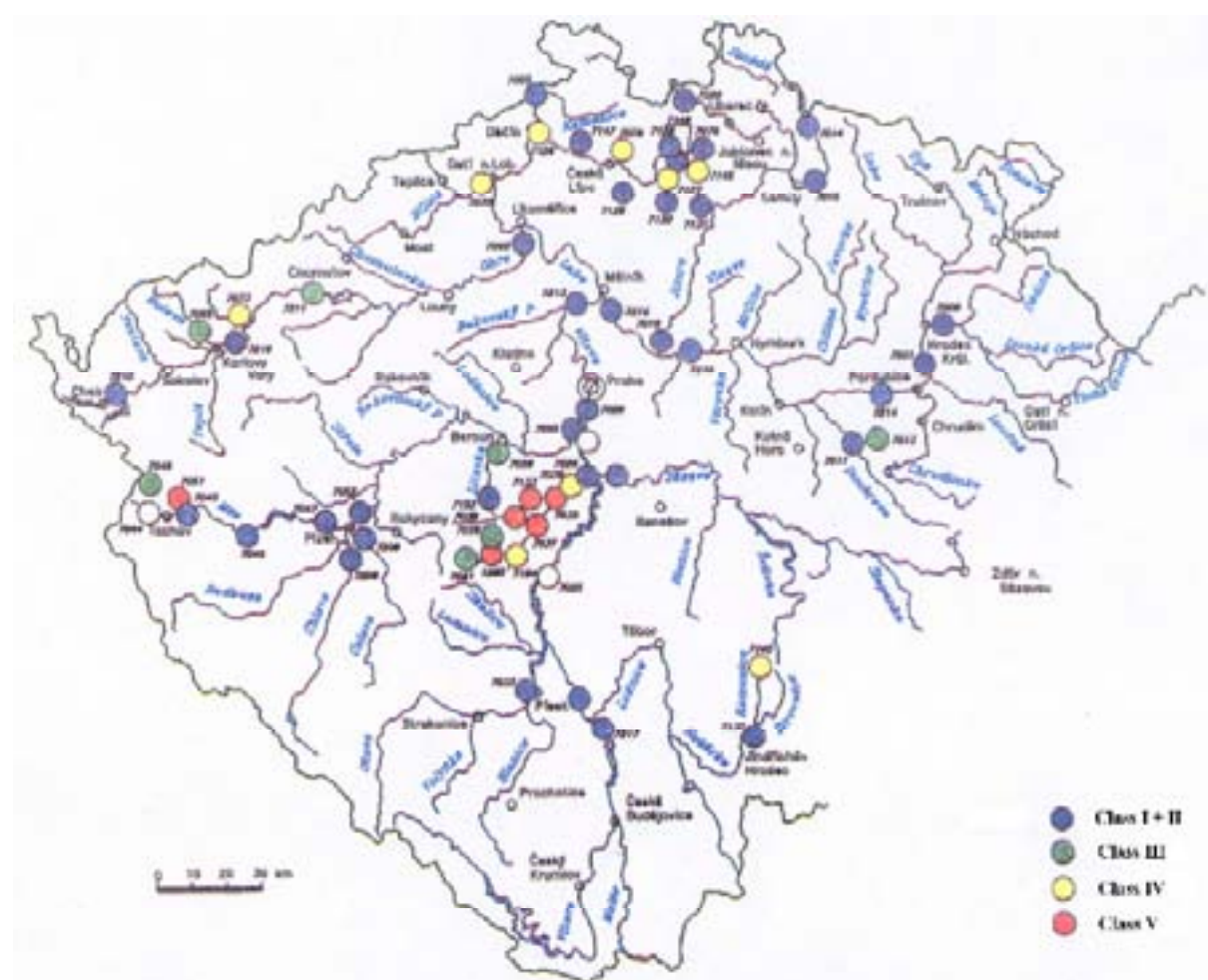


FIG. E2. The quality of Czech Republic river waters according to the content of radioactive elements (blue: the best; red: the worst quality).

TABLE E11. RADIOELEMENT DATA FOR BALNEOLOGICAL WATERS, JOACHIMSTAHL SPA

Mine spring	Ra-226 (Bq/l)	Rn-222 (kBq/l)	U (mg/l)	Pb-210 (Bq/l)	Po-210 (Bq/l)
Behounek	12.3	6.5–12.8	0.084	0.542	0.045
Curie	2.4	3.3–6.3	0.011	0.825	0.077
C1	3.4	4.3–14.4	0.013	0.946	0.241

### Radionuclides technologically-enhanced in the environment

Uranium has been used for various purposes (dyes, colouring of glass) since half of the 19<sup>th</sup> century. Joachimstahl tailings was the material from which Mr. Skłodowska-Curie separated radium and polonium. Uranium was intensively excavated in the period of 1945 – 1990, when totally approximately 105 000 t of U was exploited. Uranium was mined mainly by deep shafts from vein deposits of various ages (90 000 t of U) – mining was concentrated mainly to 3 areas (Joachimstahl – 7%, Příbram – 37%, Rozinka – 17%). Sandstone-hosted deposit in NW part (Straz pod Ralskem site) was also excavated by acid (using sulphuric acid) in-situ leaching (14 500 t U).

The extensive uranium mining in the Czech Republic has led to large environmental burdens, which remediation is expected to last next fifty years. The main burden is at the site of the former application of in-situ leaching in Cenomanic sandstones of Bohemian Cretaceous Basin where approx. 186 mil. m<sup>3</sup> of groundwater were contaminated by sulphates and nitrates of various cations (mainly Fe, Al, Ca), by some toxic elements like Be, As and V, and also by the full spectrum of natural radioactive elements (5 mil. t of dissolved solids). Characteristics of tailings and heaps of barren rocks as a result of deep mining are summarized in the following Table E12.

TABLE E12. RADIOELEMENT DATA FOR TAILINGS AND HEAPS

	Area (m <sup>2</sup> )	Volume (mil. m <sup>3</sup> )	Uranium content (t)	Activity of Ra-226 (Bq)
Tailings	6 310 000	46.9	7100	$5.8 \times 10^{14}$
Heaps of barren rocks	3 806 000	56.1	2743	$1.2 \times 10^{12}$

Impacts of mining activities can be distinguished at monitoring net where the composition of bottom sediments is monitored.

### Technologically-enhanced naturally occurring materials

Other sources of radionuclides in the environment are products of dumping and landfilling of technologically-enhanced naturally occurring materials (TENORMs) – materials and wastes that were not previously monitored for their potential radioactivity. These materials have to be evaluated according to amendment of Atomic Energy Act (2002).

In the Czech Republic mainly the following activities and materials will be checked:

- phosphate fertilisers production
- titanium dioxide pigments production
- fossil fuel burning (resulting ash and slag)
- transportation and processing of oil and gas (resulting slag)
- zirconium refractory materials
- Th electrodes
- treatment of water
- metallurgy (slag).

### **Radiation Exposure in the Czech Population**

The Czech Republic belongs to countries where the population receives higher doses due to the occurrence of many granitic geological bodies with higher content of uranium and thorium and then higher emission rates of radon. Average natural background radiation in the Czech Republic is summarized in Table E13.

TABLE E13. AVERAGE NATURAL BACKGROUND RADIATION

Source	mSv/y
Internal exposure from cosmogenic radionuclides	0.01
External exposure from cosmic radiation indoor and outdoor	0.3
External exposure from terrestrial radionuclides – outdoor	0.08
External exposure from terrestrial radionuclides – indoor	0.42
Internal exposure from terrestrial radionuclides (excluding radon)	0.3
Internal exposure from radon – outdoor	0.06
Internal exposure from radon – indoor	2.5
TOTAL	3.67

As a consequence of higher exposures from radon, Government established Radon surveillance programme, and also simultaneously the limits of concentrations of natural radionuclides where incorporated in the legislative norms.

### **Exposure rate from terrestrial radionuclides**

The radiometric map of the Czech Republic (Figure E3) was compiled on the basis of data on terrestrial radiation on the territory of the Czech Republic available by 1995 [E6]. Procedure of the map compilation corresponds to the present knowledge on gamma radiation



measurements and presentation of results in dose rate. The map data illustrate the distribution of radionuclides in rocks of the territory and above-average radioactivity of the rocks of the Czech Republic as a whole. The map of the gamma dose rate of rocks on scale 1:500 000 gives values between 6 and 245 nGy.h<sup>-1</sup>, with a mean value of 65.6 nGy.h<sup>-1</sup>. This value is comparable to the estimate of average global terrestrial radiation of 55 nGy.h<sup>-1</sup> [E7].

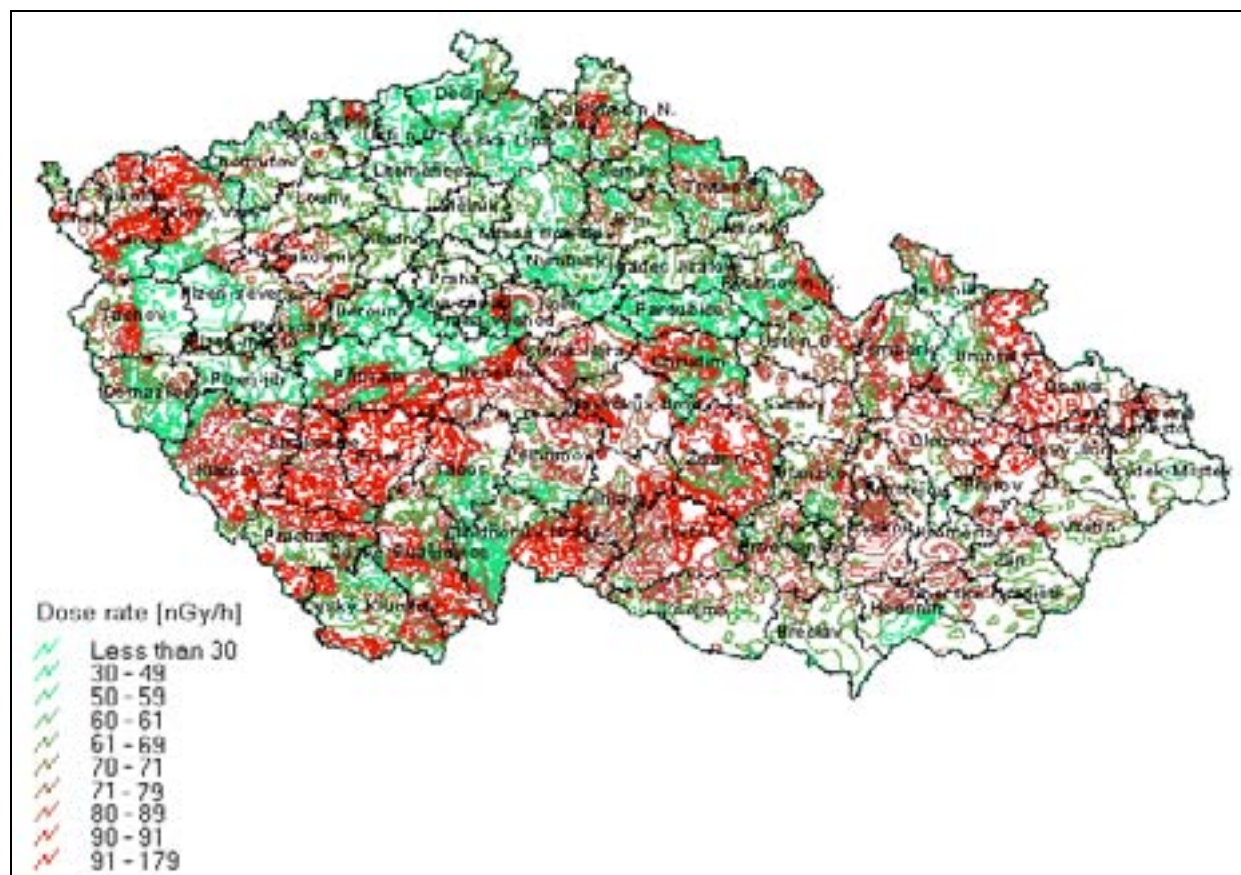


FIG. E3. Radiometric map of the Czech Republic.

### Radon distribution and limitation of natural radionuclides and radiation

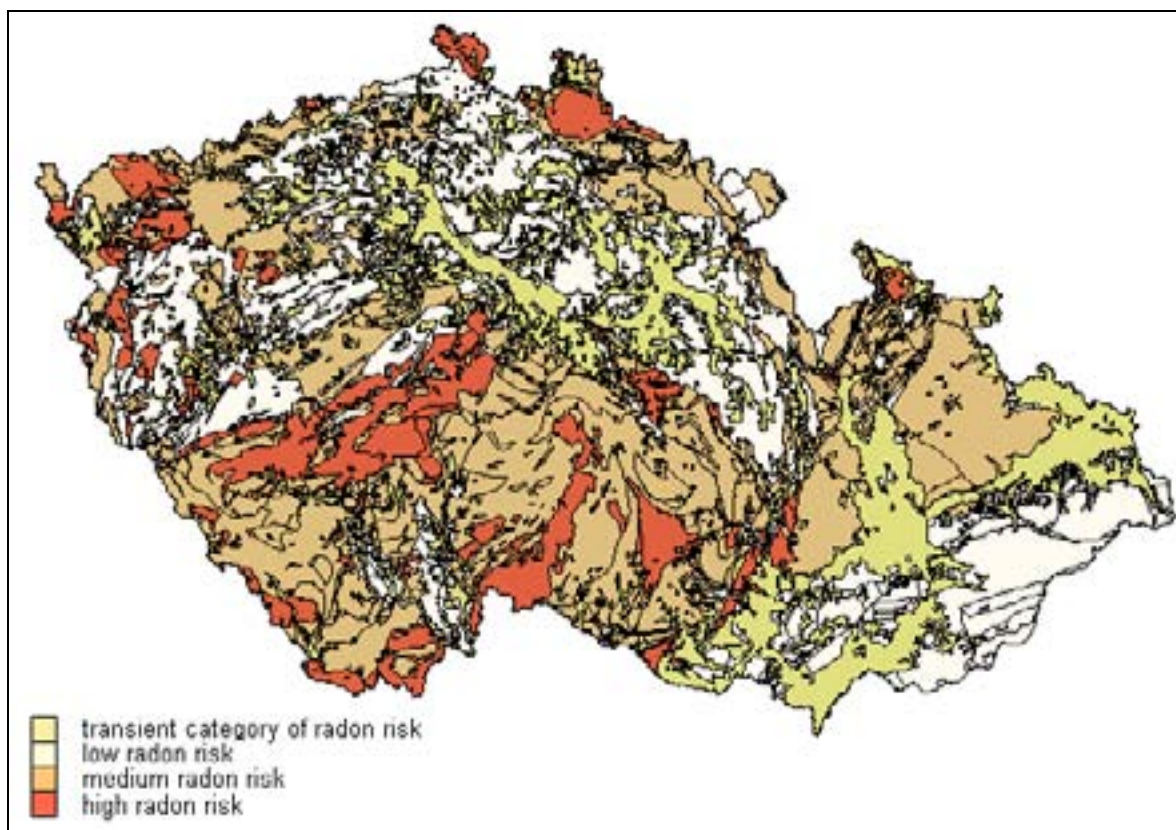
As mentioned previously, the Government established a National survey programme for dwellings with higher radon concentrations at the beginning of the 1990s. The goal of the programme is to identify the maximum houses with higher radon concentrations so that inhabitants of dwellings can be protected from exposure to radon. Radon protection measures are usually supported by state. The programme is coordinated by the State Radiation Protection Institute — SONS (organization within the responsibility of the State Office for Nuclear Safety) in close cooperation with district authorities.

The measurements of radon are carried out within one year by track detectors that are installed in the residential rooms of houses.

Identification of areas for measurement of buildings is derived mainly from maps of radon risks composed by the Czech Geological Survey in the scale 1:50 000. The maps were composed on the bases of compilation of geological and tectonic information and on radon measurement in the field. The main granitic units can be distinguished on these maps (see Figure E4).

More than 100 000 houses have been measured in the Czech Republic so far, that is rating the Czech Republic to the first positions in the world. Radon measurements are also accompanied by appropriate epidemiological studies.

It is presumed that mainly Rn-222 is attributing to the health consequences, although in some rock bodies with high concentration of Th also Rn-220 can be relevant (Trebic Massif).



*FIG. E4. Radon risk map of the Czech Republic.*

Natural radioactivity and content of natural radionuclides are limited in defined compartments of environment and products according to Atomic Energy Act and related Regulations of SONS. Range of guiding levels for preparation and realization of interventions with the intent of diversion or reduction of exposure from natural radiation sources is 5 – 50 mSv in terms of average effective dose rate of individuals from the critical group. The following matters are limited:

- Radon content in soil of land intended for construction of buildings in terms of radon index. Land with low radon index is defined for soil with volume radon activity less than 10 kBq/m<sup>3</sup> for highly permeable soil, 20 kBq/m<sup>3</sup> for medium and 30 kBq/m<sup>3</sup> for low permeable soil.
- Guidance and limiting levels in buildings with rooms declared for living purposes (Table E14).

TABLE E14. GUIDANCE AND LIMITING LEVELS IN BUILDINGS WITH ROOMS DECLARED FOR LIVING PURPOSES

	<b>Volume activity of radon in inner air (Bq/m<sup>3</sup>)</b>	<b>Photon dose rate equivalent (μSv/h)</b>
Guidance levels for existing buildings	400	1
Limiting levels for existing buildings	4000	10
Guidance levels for designed or constructed buildings	200	0.5

- Guidance levels in selected building materials are established in terms of index of weight activity and limiting levels in volume activity of Ra-226, for example see Table E15.

TABLE E15. GUIDANCE LEVELS IN SELECTED BUILDING MATERIALS

<b>Building material</b>	<b>Index of weight activity</b>	<b>Limiting levels: Ra-226 (Bq/kg)</b>	
		<b>Living rooms</b>	<b>Other use</b>
Building rock	1	300	1000
Sand, gravel, clay	1	300	1000
Ash, slag, artificial rock	1	300	1000
Tiles and veneer	2	300	1000
Bricks	0.5	150	500
Cement, plaster, porous concrete	1	300	1000

- Guidance and limiting levels in public supply potable water systems (Tables E16 and E17 below).

TABLE E16. GUIDANCE LEVELS IN PUBLIC SUPPLY POTABLE WATER SYSTEMS

Parameter	Guidance level – volume activity (Bq/l)		
	Bottled infant water	Potable water in public supply system, bottled potable water	Bottled natural mineral water
Volume activity of Rn-222	20	50	100
Gross alpha activity	0.1	0.2	0.5
Gross beta activity after subtraction of K-40	0.1	0.5	1

Procedure for required sequence of additional analyses in the case of exceeding guidance levels:

- Content of U in the case of exceedance of gross alpha activity level
- Volume activity of Ra-226 in the case when gross alpha activity after subtraction of contribution of U is exceeding guidance level
- Volume activity of Ra-228 in the case of exceedance of gross alpha activity guidance level by Ra-226
- Determination of additional alpha radionuclides specified in the following table in the case of exceedance of gross alpha activity guidance level after subtraction of contribution of uranium and Ra-226
- Content of K in the case of exceedance of gross beta activity guidance level
- Determination of additional beta radionuclides specified in the following table in the case of exceedance of gross beta activity guidance level after subtraction of contribution of K-40

TABLE E17. LIMITING LEVELS IN PUBLIC SUPPLY POTABLE WATER SYSTEMS

Radionuclide	Limiting level – volume activity (Bq/l)		
	Bottled infant water	Potable water in public supply system, bottled potable water	Bottled natural mineral water
Pb-210	0.2	0.7	1.4
Po-210	0.1	0.4	0.8
Rn-222	100	300	600
Ra-224	0.7	6	12
Ra-226	0.4	1.5	3
Ra-228	0.1	0.5	1
Th-228	0.5	6	12
Th-230	0.4	3	6
Th-232	0.4	3	6

Measurement of produced building materials and potable water is within the responsibility of producers. The Regulation of SONS prescribes the building rules for frequency of analyses, quality assurance and recording of results.

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## APPENDIX F

### FINLAND

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#### Abstract

A short introduction is given into the Finnish nuclear programme which is at an advanced stage, preparing for the licensing of the final disposal of spent fuel in a repository deep in crystalline rock.

Initially, a brief characterization of the topography, geology and hydrology of Finland is given with emphasis on special features affecting natural geochemical cycles in glaciated terrain. Countrywide elemental and radionuclide concentration data are collected and displayed as concentration maps and box plot diagrams covering the most relevant compartments. Special attention is given to concentrations in groundwaters in different depth ranges. The implications of the geochemical concentration data are evaluated with respect to rates and directions of geochemical fluxes. Elemental and radionuclide flux data are extracted mainly from specific investigations focussing on headwater stream systems and well-defined small catchments which had been followed over longer time periods. The measured chemical erosion (weathering) rates are compared with estimated rates from till erosion. Site-specific uranium fluxes at a natural uranium anomaly (Palmottu U ore deposit) are derived from detailed site investigations and are presented together with inventory and mass balance data. U fluxes in the oxidizing zone are orders of magnitude larger than those in the reducing zone below about 100 m depth. In addition, site-specific fluxes of some elements through repository reference planes of 1 km<sup>2</sup> at various disposal depths as well as near the surface at four repository investigation sites are estimated based on modelled groundwater flow data and chemical groundwater analyses. From the evaluation of the complete data set and all available complementary information it becomes evident, that in Finland natural geochemical cycles of elements and radionuclides in crystalline rock environment under the prevailing conditions are strongly affected primarily by hydraulic and geochemical factors. Low hydraulic gradients and conductivities in combination with chemical fronts (in particular, in redox conditions) limit geochemical fluxes mediated by groundwater from the deep geosphere to the surface environment. It is shown that a better mechanistic and quantitative understanding of natural geochemical fluxes (mobilization and fixation of elements) is beneficial for the development of realistic radionuclide migration models and performance assessment tools and that the application of natural fluxes for comparison with calculated repository-derived radionuclide fluxes can support a safety case.

#### Background

##### Nuclear programmes

The Finnish power companies operate four nuclear power reactors with a total capacity of 2 656 MWe which generated in 2001 27% of all electricity consumed in Finland. Teollisuuden Voima Oy (TVO) owns two BWR units ( $2 \times 840$  MWe) at the Olkiluoto power plant. Fortum Power and Heat Oy has two PWR units ( $2 \times 488$  MWe) at the Loviisa power plant. A positive political decision for the construction of a fifth unit has been conducted during the year 2002. The producers of nuclear waste are responsible for the safe management

of the waste. The preparatory work and implementation of the final disposal of spent fuel is done by Posiva, jointly owned by TVO and Fortum. The waste management programme is supervised by the Ministry of Trade and Industry and the Radiation and Nuclear Safety Authority (STUK).

Spent fuel management and disposal site selection:

- The amount of spent fuel from the Loviisa plant during its planned lifetime of 40 years, will be about 1070 tU (of this amount, about 330 t U has been shipped to the Russia) while that from the Olkiluoto plant is estimated to be 1 870 t U.
- At Olkiluoto and Loviisa repositories of low and intermediate level reactor wastes are already in operation since 1998 and 1993, respectively.
- The selection of a disposal site for spent fuel disposal was launched by an area screening reported in 1985, continued by preliminary investigations at five sites (including the Olkiluoto NPP site), detailed investigations at three sites, additional investigations at the Loviisa NPP site and has been concluded by the proposal for the disposal site, made at the end of 1999 (selection of one (Olkiluoto) out of four sites investigated)
- The environmental impact assessment (EIA) process for spent fuel disposal was initiated in 1997 and was completed in 1999
- The first formal licence application (Decision in Principle) which was supported by a safety analyses (TILA-99) has been submitted to the authorities in spring 1999, and the positive decisions by government and parliament were made until May 2001.
- STUK has made its own safety evaluation supported by external expertise. The result was that no safety issues were found that would speak against the implicability of the final disposal concept at the proposed site.
- A programme for research, development and technical design for the pre-construction phase of the spent fuel repository was published in early 2001 and then submitted for review by the authorities
- The results of baseline studies ongoing at Olkiluoto will be reviewed in 2003 and the construction of an underground research facility at the site will begin in 2005.
- Development of performance assessment methodologies is continued and will be reported and reviewed in 2006; Finland participated in the EU project "Testing of Safety and Performance Indicators"
- The construction licence process is scheduled to start in 2010 and the operating licence process in 2020.

In the safety regulations given by STUK nuclide-specific constraints are defined. These constraints are defined so that at their maximum, the radiation impacts arising from disposal can be comparable to those arising from natural radioactive substances.

### **Topography, geology and climate**

The bedrock in Finland is the result of two main orogenic phases in the Precambrian. Sedimentary and volcanic rocks formed in connection with these orogenic phases were buried



about 20 km deep where they were recrystallized and their original mineral content was changed. The present erosional level represents rocks which were at about 15 km depth 1800 Ma ago. Later, really significant rock types were formed only by intrusion of the rapakivi granites about 1500–1700 Ma ago. The present erosional level of the rapakivi granites represents rock from about 5–8 km depth. Due to the old age and deep burial sedimentary and volcanogenic rocks are highly metamorphosed, recrystallized and partly mixed with other rocks. As a result, most of the Finnish bedrock presently exposed belongs to plutonic and metamorphic rock types. About 53% of all bedrock in Finland is granitic in composition (granite, granodiorite, quartzdiorite), 22% is migmatitic, 9% are schists and only 8% are basic rock types. The present bedrock surface is a peneplane quite near to that which was exposed about 650 Ma ago. During the period after about 500–600 Ma ago obviously only weathering of the exposed rock surface occurred. During the last 1 Ma about ten glaciations occurred which were interrupted of warm interglacials of only short duration (about 10 000 a each). The first glaciations removed most of the previously weathered layers and exposed the fresh bedrock surface. The periods of the interglacials most probably were too short for formation of significant weathering layers.

Areas of glaciated terrain differ in many respect from areas unaffected by glaciations. The glacial (till) and glacio-fluviatile deposits forming the present overburden mainly consist of varying mixtures of crushed fresh bedrock, older glacial sediments and some preglacial weathering crust. In comparison to old shields in non-glaciated regions where a continuous development of a weathering layer in a quasi-equilibrium state with the erosion rate could take place, in glaciated terrain a time gap and clear discontinuity exists between the young overburden which is about 10 000–60 000 a old and the Cambrian bedrock surface. The median of the thickness of the till cover in Finland is only 6.7 m, the most common thickness is 3–4 m and 95% is below 30 m. About 3% of the surface is exposed bedrock. The rock surface is exposed more generally along the south coast, in the lake district and in most northern parts of Lapland.

In contrast to surface boulders which have been transported further, the bulk material in till has travelled shorter distances and has not been sorted during transport. Some separation effects, however, are due to preferential crushing or partial outwashing of fine fractions. Regionally, the concentrations of elements in till are generally representative of the underlying bedrock, in particular, in the deepest layers, whereas in the upper layers more mixing has occurred. Compared to mechanical transport chemical migration is a much more limited phenomenon in till. Due to the short time period since the last glaciation weathering has usually affected only the till surface, while at the rock-till interface even sulphide grains may still be preserved.

Bedrock geochemistry and conditions of boreal and arctic climate (precipitation: 400–700 mm/a; low evaporation: 100–350 mm/a (smaller Figures for the north)) favour the formation of acid podzolic soils with low adsorption and acid buffering capacity and organic soils (histosols). Leaching of elements and downward transport leads to a pale A-horizon which is covered by a thin humus layer. Precipitation of iron and other metal ions occurs in the brown B-horizon. The C-horizon consists mostly of unweathered till material. Due to the humid and cool climate and the flat, poorly drained topography, organic postglacial sediments (peat) are common. In the average about 30% of the country are covered by peat, while in the northern parts locally 70% are reached.

The hydrology of Finland is characterized by a flat and partly hilly topography with a height above sea level of mostly not more than 100–200 m (in Lapland 300–600 m) and small height variations (peneplane). This leads to complex drainage systems with many lakes in the central

parts. Free-flowing rivers, with a few exceptions, are often relatively small and concentrated along coastal regions and in Lapland. The removal of all preglacial sediments by repeated glaciations has led to a situation where groundwater aquifers in the classical sense are not existing. Under these conditions near-surface groundwater flow is a more local phenomenon within orders of magnitude of usually a few km. Driven by minor local gradients, groundwater in fractures in the bedrock is connected to the near-surface groundwater regime. Water-conducting fractures are more common within the upper tens of meters of the bedrock. Groundwater flow at greater depth on a regional scale is generally very slow and could be measured only in rare cases. A peculiarity of the Fennoscandian Shield is the wide-spread presence of saline groundwaters at depth. Along the coast these saline waters can be found at quite shallow depths from about 50 m on, while in the inland they usually occur below about 200-400 m of fresh water. There is evidence that saline waters have been practically isolated from surface-close hydrological cycles for time periods often exceeding 10 000 a. The various postglacial fresh and saline phases of the development of the Baltic Sea had influenced groundwaters which is still today clearly visible in the groundwater chemistry in coastal regions. Glacial rebound in Finland has exceeded 500 m and is still going on at rates up to 9 mm/a which means that presently very slow displacement of saline by fresh water is going on and no equilibrium state is reached.

### **Study areas**

A look at the groundwater flow systems at the four different repository candidate sites in Finland studied in detail by Posiva is indispensable for achieving a deeper understanding of geochemical flow systems in the country. These four sites are unique in that respect that there is no other place in Finland which has been studied by a comparable comprehensive use of advanced characterization methods, except that a comparable level of detailed knowledge is also available at the Palmottu natural analogue study site, the location of a uranium mineralization which is also included into the CRP. A detailed characterization of the flow system, the groundwater chemistry and evolution including water-rock interaction modelling forms the basis for a quantification of elemental fluxes.

Two of the investigation sites are coastal sites (Olkiluoto, Loviisa (Hästhölm)) and two are inland sites (Romuvaara, Kivetty). The site characterization proceeded from the rock structure model to the creation of a groundwater flow model with calculations going from the regional to the site and waste canister scale. The site scale flow reports contain highly detailed and illustrative images showing different cross-sections through the 3-D rock fracture zone model and numerous computed velocity fields in cross-sections. The modelling takes into account future changes in the flow field by modelling the present state, 100 a, 1000 a and 10 000 a A.P. (after present). Also the displacement of saline by fresh water is modelled in the same time frame. The result is a fracture model in which the dominant flow routes from the repository to the biosphere at various times are indicated.

The Palmottu U-Th mineralisation in SW Finland, which is not disturbed by mining, consists of narrow pegmatite veins (2–10 m) cutting granite and mica gneiss. The mineralisation extends to depths of at least 400 m and total resources are estimated at around 1000 tonnes. The deposit provided an excellent opportunity for studying radionuclide transport along well-identified groundwater pathways in the fractured crystalline rock of the Fennoscandian Shield. Natural analogue studies have been in progress at Palmottu since 1987, aimed at characterising the geology, hydrology and hydrochemical setting of the uranium mineralisation. The present groundwater flow system dates back to the last deglaciation and the subsequent emergence of the landmass some 10 000 years ago. The uranium deposit extends from near the bedrock surface through a redox transition zone into a strongly reducing

groundwater environment. This has allowed the full cycle of uranium redox chemistry under natural conditions to be studied. In many respects, the geological history of the Palmottu deposit resembles the postulated evolution of the repository proposed for spent U fuel at Olkiluoto, including the inventory, host rock and geographical setting. The deposit is limited in size, contains uranium mainly in the form of  $\text{UO}_2$  and has been subjected to a series of perturbations since its emplacement.

## **Elemental and radionuclide concentration data**

### **Sources of data**

Glacial till has been the most important sample medium for geochemical mapping in Finland which is available throughout most of the country, and the chemical composition of till samples reflects the composition of the underlying bedrock better than stream sediment samples. Reconnaissance scale till geochemical mapping of Finland was followed by mapping at the regional scale. Both these mapping programmes were based on analyses of the fine fraction ( $<0.06$  mm) of till samples. All data was collected and made available by the Geological Survey of Finland (GSF). Radionuclides were measured by STUK and the data were partly included into the GSF data base and own ones.

In the reconnaissance scale, a total of 1057 glacial till samples were collected from the whole country. Composite samples of 3 to 5 subsamples were collected from a rectangular area of  $300 \text{ m} \times 1000 \text{ m}$ . The average sampling density was one sample per  $300 \text{ km}^2$ . The samples were collected from the slightly weathered C-horizon, at a depth of 0.5–2 m, under podzolic soil. About 40 elements found in the fine fraction of till were included in maps in the Geochemical Atlas of Finland. Two extraction methods were used: 1) total dissolution with strong concentrated mineral acids; and 2) partial dissolution using aqua regia. As rock geochemical mapping in reconnaissance scale is currently being carried out in the whole country, rock concentration data had to be taken from the literature. More detailed geochemical mapping has been done in exploration target areas.

The till geochemical mapping in the regional scale was carried out during 1982–1994. Altogether 82 062 till samples were collected. The sampling density was 1 sample/ $4 \text{ km}^2$ . The country was divided into  $2 \times 2 \text{ km}^2$  squares and from each square a composite of 3–5 subsamples were taken. Only till was used as sampling material, and all samples were to be taken from beneath the groundwater level.

9347 groundwater samples were taken during the hydrogeochemical mapping of shallow groundwater in reconnaissance scale covering the whole country at a density of 1 sample per  $50 \text{ km}^2$  carried out in 1978–1982, and during subsequent denser sampling programmes carried out in 1982–1998. In 1999, 1000 new samples were taken from the whole country. The samples were collected from springs, captured springs, dug wells (representing groundwater of the overburden) and wells drilled into bedrock (representing bedrock groundwater from an average depth of about 50 m and with 5% of the depths  $>100$  m).

Stream water and organic stream sediment samples were collected from 1165 sampling points throughout Finland in August–September 1990. The mean sampling density was one sample per  $300 \text{ km}^2$ . Each sampling point represents a drainage area of ca.  $30 \text{ km}^2$ . Organic stream sediment samples were collected by using a scoop net with mesh diameter of 0.06 mm. During 1973–1984 GSF carried out geochemical mapping based on lake sediments. About 16 000 organic-rich sediment samples were collected from an area of  $80\,000 \text{ km}^2$  at an average sampling density of 0.2 samples/ $\text{km}^2$ .

The sampling programme of the geochemical mappings were conducted according to quality standards of the international programme FOREGS (FORum of European Geological Surveys Geochemical Baseline Programme). Almost all analysis have been carried out in the Chemical Laboratory of GTK which is accredited according to the EN 45001 standard and ISO Guide 25 to perform chemical analyses of geological and environmental samples.

### **Summary of data**

The selected countrywide concentration data from the various compartments (till, stream sediments, stream water, groundwater from dug and drilled wells and lake sediments) (summarized in Table F1) have been processed and displayed in box-plot Figures divided into sets for pronounced geochemical provinces (a typical example is shown in Figure F1). In addition, maps showing the area distributions are given (in Figure F2 the results for U in till are displayed). Depending on sampling density and method of analysis (total analysis or partial extraction) the distributions, ranges and mean values show certain variation which is expected. Significant variation is caused by differences in the elemental content of the parent rocks. Concentrations of As, Cu, K, Rb, Th, U are given for till, while for other materials the list of analysed elements is smaller. The missing data are: for stream sediments: Rb; for lake sediments: As, K, Rb and Th; for dug and drilled well groundwaters: Th. Ra values are only available from well waters. Cs concentrations were not analysed and of the analysed Sn concentrations in bedrock groundwater only 10 of 1000 were above the detection limit. Element-specific results are given and discussed according to their relation to geological provinces. The country-wide rock geochemistry project is still going on, so that only rock data from the literature is available now. Detailed, comprehensive results are given as an appendix to this short summary.

The regional differences in metal contents and the enrichments such as ore deposits encountered in Finland are of endogeneous origin and related to the Precambrian. The location of many ore zones and single deposits is bound to very old structural units such as joints and fracture zones or certain lithologies, mostly shists and basic intrusions. Migmatitic and granitic units are almost devoid of ore enrichments. Even very low density sampling produced coherent geochemical information, for example large scale geological units such as major schist belts or even smaller rapakivi intrusions are identified. Although anomalies caused by discrete ore deposits usually are not recognized at this scale, wider zones where mineralizations are more common than in others are well reflected. Not only large lithological units, but also tectonic features such as fracture and shear zones in these extending across contacts of lithological units and even geological formations of different age were reflected. In these active weakness zones geochemical dispersion leading to anomalies was favoured in the past and may even continue as secondary migration caused by postglacial weathering.

As Finland is only sparsely populated, private drinking water supply by two types of wells, dug in the overburden or drilled in the bedrock, is very common. It has been found that groundwater chemistry depends on many factors which often camouflage the rock type influence. The waters in each rock type contained least ions below sand and gravel cover and most ions below clay sediments. The dominant influence of the overburden over the rock type might be due to the more intense contact with the fine-grained material as compared to fracture surfaces in the bedrock, but the overall influence of the mineralic solids seems to be low, because of short contact times in small flow systems. Water from depths greater than 100 m had enhanced cation content which was interpreted as a consequence of increased water-rock interaction. The influence of tectonic weakness zones (fractures) was reflected in a lower pH, higher bicarbonate content and higher temperature at a depth greater than 60 m. Obviously, intrusion of surface water more deeper into the rock is enhanced by fractures.

The regional distribution of the groundwater properties of springs and dug wells shows a few distinct exceptions on a fairly homogeneous background. The concentrations of the most abundant heavy metals, Fe and Mn are depending more on conditions such as redox potential and dissolved organic material and less on the lithological environment. Thus, mainly the western coastal regions deviate from a fairly homogeneous background (data for dug wells). The patterns of heavy trace metals are more irregular and only partly correlated with local bedrock conditions in deeper waters. U concentration is enhanced particularly in drilled wells in granitic areas mainly in southern Finland and some corresponding enhancements are observable in dug wells and springs. Statistically U correlates moderately with bicarbonate in drilled wells, otherwise there is no clear correlation.

Comparison of area elemental distributions in different materials does not give direct information on natural geochemical fluxes, but allows some conclusions about the sources and dominating transport processes. On the larger scale glacial till and often the groundwater in drilled wells represents fairly well the composition of the underlying bedrock. The correlations between concentrations in different compartments from till to sediments and different water bodies are strong only for relatively mobile elements such as U. Thus the correlations could be indicators of fluxes from the geo- to the biosphere, but also for fixation in sinks. For example, the precipitation of As in stream water is indicated by strong correlation with As concentrations in stream sediments. For elements whose contents are controlled mostly by water-solid interactions such as K and Rb and for low solubility elements such as Th, influences of chemical transport from the geosphere are concealed. The non-systematic dependence of the correlation coefficients from the distance of the sampling points could have its origin in the very small-scale local groundwater flow patterns caused by the low relief topography and the absence of large connected aquifers. Obviously, as is most evident in the U data, the concentrations in ground- and surface waters are dominated by local variations in till and rock geochemistry and regional signatures are less pronounced.

Considering the present state of geochemical elemental transport cycles working between bedrock, bedrock groundwater and the surface environment, it seems that due to geologic, topographic and climatic reasons, the main direction of transport is downwards within the overburden profile and horizontally along the surface layers. Most of the fluxes seem to occur in the upper soil layers where the intruding rain water is soon buffered and becomes less aggressive when reaching the C-horizon or the bedrock surface and the rock fracture system. In a larger global context these observations are to be expected, as the prerequisites for active elemental transport by deep groundwaters to the biosphere which can be observed for example in large parts of central Europe — high relief, high thermal gradients, deep-reaching conductive tectonic structures and/or release of gases from great depth — are not fulfilled in Finland.

Chemical elements which are mobilized by weathering usually move downwards within the soil profile, are eventually demobilized and remobilized again, move with shallow groundwater to discharge locations and end finally in headwater streams and rivers. If there are secondary sinks (e.g. peat bogs, lake sediments) in the discharge areas, then again immobilization can occur. Under the prevailing hydrological conditions only a very small fraction of the groundwater infiltrates to greater depths and, respectively, at discharges the fraction of groundwater which comes from greater depth is very small and not detectable in geochemical surveys. Only very sophisticated and therefore only locally applicable techniques such as component analysis and isotope techniques should be able to identify contributions of deeper groundwaters. Correspondingly, selective leaching and subsequent analysis of various mobile fractions of elements in solid materials might be useful.

The key to the successful application of concentration data and derivation of realistic geochemical fluxes is to thoroughly understand the processes that give rise to the overburden and the exchange processes between bedrock and overburden. Compared to mechanical (glacial) transport, recent chemical migration is a much more limited phenomenon in till. Due to the short time period since the last glaciation weathering has usually affected only the till surface. The usually low hydraulic conductivity and the predominance of unweathered crushed rock material in a thin glacial till cover on mostly unweathered bedrock and the abundant presence of secondary sinks such as peat and sediments have pronounced influence on geochemical fluxes of elements. Due to these facts in connection with the action of chemical barriers near the ground surface elemental fluxes are mostly restricted locally and to the upper horizons of the overburden and contributions of transport by deep groundwater are hardly detectable. Numerous examples of case studies on geochemical anomalies (element enrichments, ores) at various depths in various geological settings illustrate these conclusions. There are indications, however, that results of selective extractions of only mobile forms of elements can give more sensitive measures of migration of elements from more deeply buried anomalies to the biosphere. At the moment such information is available only from a few local case studies, but availability from larger scale surveys seems to be increasing in the future.

Detailed site specific concentration data from Posiva's investigation sites as well as from the Palmottu study site are available mainly from earlier studies. These are from boreholes and focus on rock and groundwater. In baseline studies radioactivity and concentration of radioactive elements at the nuclear waste repository candidate sites have been compared with average values for the whole country. Uranium concentrations in groundwater often show large scatter with depth, and it is of dubious value to calculate average concentrations without understanding the processes that influence these concentrations. The observed values depend on the groundwater flow pattern in the rock fracture system, groundwater chemistry and the geochemistry of the flow paths. The primary value of this data is its use in understanding and modelling the geochemistry of a site, but also for mass balance estimations. As an example the concentration distribution of uranium in groundwater at Palmottu is given (Figure F3); there can be clearly distinguished between oxidizing and reducing types of groundwater. There are two types of oxidizing waters with enhanced U contents which are practically limited to the upper 150 m of the system.

TABLE F1. STATISTICAL VALUES OF ELEMENT CONCENTRATIONS IN DIFFERENT COMPARTMENTS IN FINLAND

		Count	Min	5 %	Median	Mean	S.D.	95 %	99 %	Max
Lake sediments										
Cu	mg/kg	15304	1.00	2.00	18.0	37.5	78.2	129	284	3280
U	mg/kg	15304	0.008	0.824	3.15	7.52	58.9	18.3	63.9	5020
Till: Reconnaissance scale										
As	mg/kg	1054	0.13	0.257	2.56	3.59	4.60	11.5	25.8	44.0
Rb	mg/kg	1054	<15.0	40.9	72.6	76.1	25.1	123	152	239
U	mg/kg	1054	<0.3	1.61	3.08	3.42	2.04	6.37	8.86	48.2
Till: Regional scale										
As	mg/kg	82065	<40.0	<40.0	<40.0	.	.	<40.0	63.2	2550
Cu	mg/kg	82065	<8	<8	21.9	28.4	25.5	69.4	111	1640
K	mg/kg	82065	<1300	<1300	1950	2470	1855	6090	9450	24800
Th	mg/kg	82065	<20	<20	<20	.	.	26.1	42.4	639
Stream sediments										
As	mg/kg	1166	<0.05	0.751	2.89	5.41	14.2	14.7	69.1	268
Th	mg/kg	1166	<0.04	1.52	4.94	5.69	3.47	12.7	17.6	31.2
U	mg/kg	1166	<0.01	0.611	2.01	4.31	14.9	11.8	41.7	379
Cu	mg/kg	1166	<1	5.03	12.4	15.9	18.4	35.6	59.9	476
K	mg/kg	1166	<200	502	1550	2220	1999	6647	10562	14100
Stream water										
As	µg/l	1154	<0.05	0.059	0.359	0.551	0.627	1.58	3.85	6.5
Cu	µg/l	1154	0.059	0.17	0.638	0.932	1.32	2.36	6.04	24.5
K	mg/l	1154	0.068	0.203	0.695	1.18	1.38	3.83	7.2	17.1
U	µg/l	1154	<0.01	<0.01	0.073	0.186	0.437	0.728	2.35	6.28
Groundwater: Drilled bedrock wells										
As	µg/l	263	<0.05	<0.05	0.16	0.999	2.82	6.74	15.0	23.6
Cu	µg/l	263	<0.04	<0.04	9.14	32.3	78.3	146	357	917
K	mg/l	263	0.23	0.719	3.00	4.42	4.93	14	23.9	40.2
Rb	µg/l	263	0.03	0.153	1.87	3.19	4.71	10.6	27.6	42.7
Th	µg/l	263	<0.02	<0.02	<0.02	.	.	0.096	0.154	1.41
U	µg/l	263	<0.01	0.01	0.68	13.7	55.1	54.5	309	643
Rn	Bq/l	263	1.00	8.87	142	311	593	1390	2980	4880
Groundwater: Dug wells										
As	µg/l	739	<0.05	<0.05	0.14	0.353	1.00	1.33	4.74	19.7
Cu	µg/l	739	<0.04	0.136	2.53	14.1	33.3	76.2	176	410
K	mg/l	739	0.19	0.587	2.79	4.97	7.36	17.3	43.9	92.3
Rb	µg/l	739	0.04	0.227	2.73	4.98	7.45	15.9	41.1	73.3
Th	µg/l	739	<0.02	<0.02	<0.02	.	.	0.140	0.583	1.50
U	µg/l	739	<0.01	0.01	0.09	0.846	3.03	3.83	21.5	36.6
Rn	Bq/l	739	0.10	2.00	12.0	37.8	82.8	194	526	893





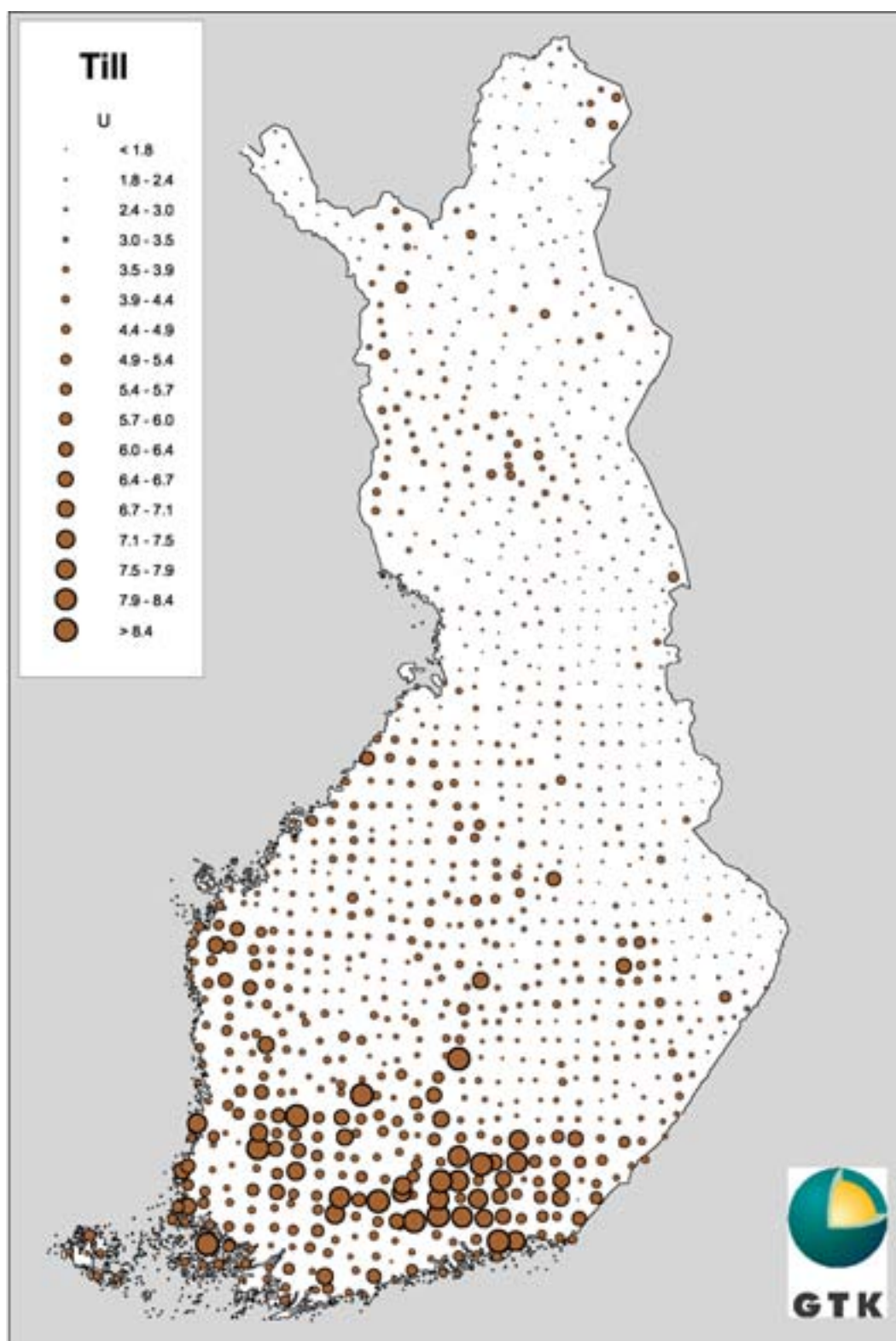


FIG. F2. Uranium (U) concentration (mg/kg) in till in Finland.

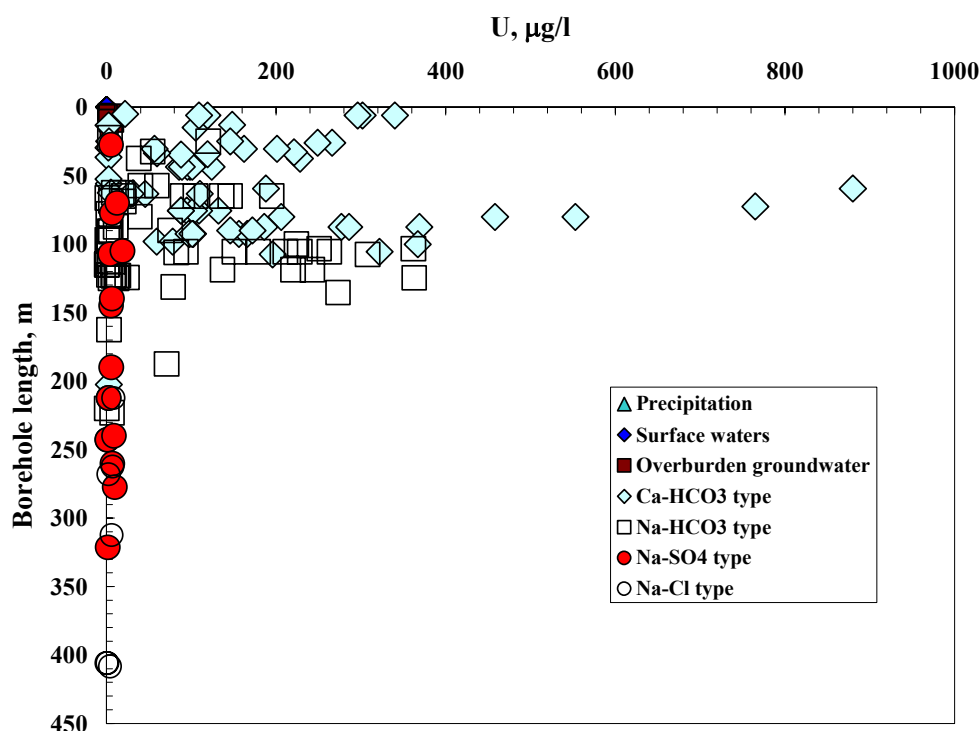


FIG. F3. Uranium concentration in groundwater versus borehole length, Palmottu.

## Elemental and radionuclide flux data

### Sources of data

From an investigation programme by GSF on geologically and hydrogeologically well-defined small catchments monitored over a longer time period, where groundwater is discharging in springs, a number of representative sites from southern and central Finland were selected for determination of elemental fluxes by groundwater (groundwater erosion).

Estimations of heavy metal weathering rates for forest soils in the southern and central parts of Finland were conducted by GSF. The estimations are based on base metal weathering rates and heavy metal concentrations in till with a data base of 638 reanalysed till samples (results are given for Cu and U). For more representative rates of till chemical erosion and comparison with measured groundwater erosion rates in well-defined catchments the study was extended within the CRP to the analysis of soil profiles including a weathering-resistant component (Zr) within three of the catchments.

Natural fluxes of Cu and U from Finnish headwater streams during one summer period (1990) from 30 basins with a drainage area of about 30 km<sup>2</sup> were determined by GSF by measuring concentrations of the elements in stream water and the rate of stream flow. The mean sampling density was 1 sample/300 km<sup>2</sup>. Each sampling point represented a drainage area of 30 km<sup>2</sup>.

The site specific data from the Palmottu uranium mineralization were mainly gained within the EU project "The Palmottu natural analogue project", contract FI4W-CT95-0010 (1994-1998) which was coordinated by GSF; data from groundwater discharge areas were gained

within the CRP. The estimations of U mass transport are based on analyses of U concentrations in all relevant materials and calculations of groundwater flow rates based on hydraulic testing.

The site specific data from the four repository investigation sites were from the data bases of Posiva. Groundwater flow data and groundwater composition data for defined areas at certain depths were extracted and used for calculation of elemental fluxes.

### **Summary of data**

There are numerous approaches to measure or estimate elemental fluxes caused by weathering and chemical erosion, but they often require certain assumptions. Therefore, more certainty in the results can be gained when rates obtained by various independent methods are compared. Some methods are more reliable in regions of short regolith weathering histories such as glaciated terrain, as in Finland. The direct use of abundant and versatile till geochemical data which is available in Finland improves weathering rate estimates by giving more reliable information concerning weatherable base cations than bedrock data.

From an investigation programme on geologically and hydrogeologically well-defined small catchments over a longer time period, a number of representative sites were selected. Groundwater chemistry and the observed large and rapid variations of the chemistry lead to the conclusion that groundwaters flow mostly in the Quaternary in all five catchments and represent shallow groundwaters characterized by short contact times with the geological substrate. Geochemical fluxes worth mentioning seem to be constrained to the surficial geological layers which include overburden and the most fractured uppermost layers of bedrock. Influences of fluxes which might bring substances from greater depth cannot be distinguished in these catchments. The results show significant scatter of groundwater chemical erosion rates in the different catchments. From the average amount of K removed in the catchments since the end of the latest glaciation a theoretical weathering depth of about 40 mm can be derived for average granitic composition with about 3% K under the assumption that weathering is limited to the surface layer only, a number which seems reasonable. Thicker overburden seem to lead to higher trace metal concentrations in water due to more intense water-rock interaction, but the presence of rock fractures does not find unambiguous expression in element contents or fluxes. It is clear that observed differences depend on the geochemistry of all branches of the relevant flow paths and not only on the properties of the parent material and conclusive explanations would require very detailed site investigations. In most cases the chemical weathering rate estimates give at least some insight in the order of magnitudes in areas where the anthropogenic input is low. Normalized median chemical weathering rates are summarized in Table F2 for easy comparison. For the future it can be expected that weathering rate estimates will be improved by increasing availability of selectively leached, mobile metal ion data.

It is emphasized here that the comparison of fluxes can hide the problem of high concentrations of harmful elements in small streams with low runoff. It has been recommended to compare the concentrations of metals in regional maps with the lowest concentrations likely to cause injurious effects to aquatic organisms the s.c. "lowest known levels of effect". These values were exceeded for Cu in four streams of this study, although the estimated Cu fluxes for these four headwater streams were lower than the mean value of the 30 studied streams.

TABLE F2. SUMMARY OF MEASURED AND ESTIMATED CHEMICAL EROSION RATES (TRANSPORT IN DISSOLVED FORM); UNITS: KG/KM<sup>2</sup> A (RN: GBQ/KM<sup>2</sup> A)

<b>Chemical erosion rates (5 catchments: rates measured (median) or estimated from till erosion (in brackets))</b>	<b>As</b>	<b>Cu</b>	<b>K</b>	<b>Rb</b>	<b>Th</b>	<b>U</b>	<b>Rn</b>
Catchment 1: Onkalolähde	0.006 (0.003)	0.023 (0.041)	96.3 (217)	0.19 (0.30)	0.003 (0.02)	0.003 (0.003)	1.7
Catchment 2: Kiilan lähde	0.057 (0.34)	0.46 (0.41)	980 (532)	0.048 (10.7)	0.005 (1.5)	0.43 (0.40)	3.3
Catchment 3: Käikälän lähde	0.035 (1.1)	0.24 (1.4)	567 (259)	0.38 (5.22)	0.006 (0.9)	0.025 (0.28)	13
Catchment 4: Oinaslähde	0.013 (0.49)	0.067 (0.43)	195 (185)	0.25 (3.76)	0.005 (0.7)	0.11 (0.16)	51
Catchment 5: Isolähde	0.033 (0.16)	0.081 (0.038)	260 (142)	0.36 (0.54)	0.001 (0.05)	0.011 (0.02)	27
Estimated till mean chemical erosion rates (Finland)		0.21		1.2	0.17	0.054	
Headwater stream median chemical erosion rates (Finland, except Lapland)		0.17				0.017	

The uranium mass balance estimation for the Palmottu mineralization (about 1000-1500 t of U) showed that very little U was lost from the system during its long geological history. When considering a block of rock containing most of the ore, it is interesting to note that in the rock matrix itself actually the same order of magnitude of U is contained (about 1500 t). Most of the small fraction of the inventory that has been mobilized is found at present absorbed in fractures or fixed in fracture coatings in the oxidizing zone comprising the upper 100 m (8 respectively 23 t of U). The respective values in the reducing zone (lower 150 m) are 6 and 0.2 t. The amounts of U in groundwater are much lower: flowing bedrock groundwater in fractures in oxidizing zone: 0.15 kg, in reducing zone: 0.002 kg; stagnant pore water in rock matrix in oxidizing zone: 3 kg, in reducing zone: 0.045 kg. The measured U concentrations in lake and river water are very low; in the groundwater of the overburden the U inventory is also low, about 0.72 kg. The major sinks for U in the biosphere are: a lake (0.08 kg U), lake sediment (130 kg U), two peat bogs (84 + 17 kg U). In the overburden are found 11 t U, but only 0.2% of that amount are mobile. The peat bogs do not seem to be at the main groundwater discharge locations. The time scale for accumulation of U in the secondary sinks is about 10 000 a.

The U mass balance and the fluxes to the biosphere are displayed in Figure F4. The key conclusion is that the U flux from the upper, oxidizing zone is much higher (160 g/a) than that from the lower, reducing zone (0.002 g/a), but in the same order as the U flux caused by surficial bedrock erosion and chemical weathering (70 g/a). Natural uranium fluxes are very limited under reducing conditions, but also under oxidizing conditions mineralization processes and organic complexation limit or delay transport of U. Considering long term perspectives, there are indications that during past glaciations melt waters may have intruded deeper into the flow system at Palmottu, but reducing conditions were largely maintained by the redox buffering capacity of the rock.

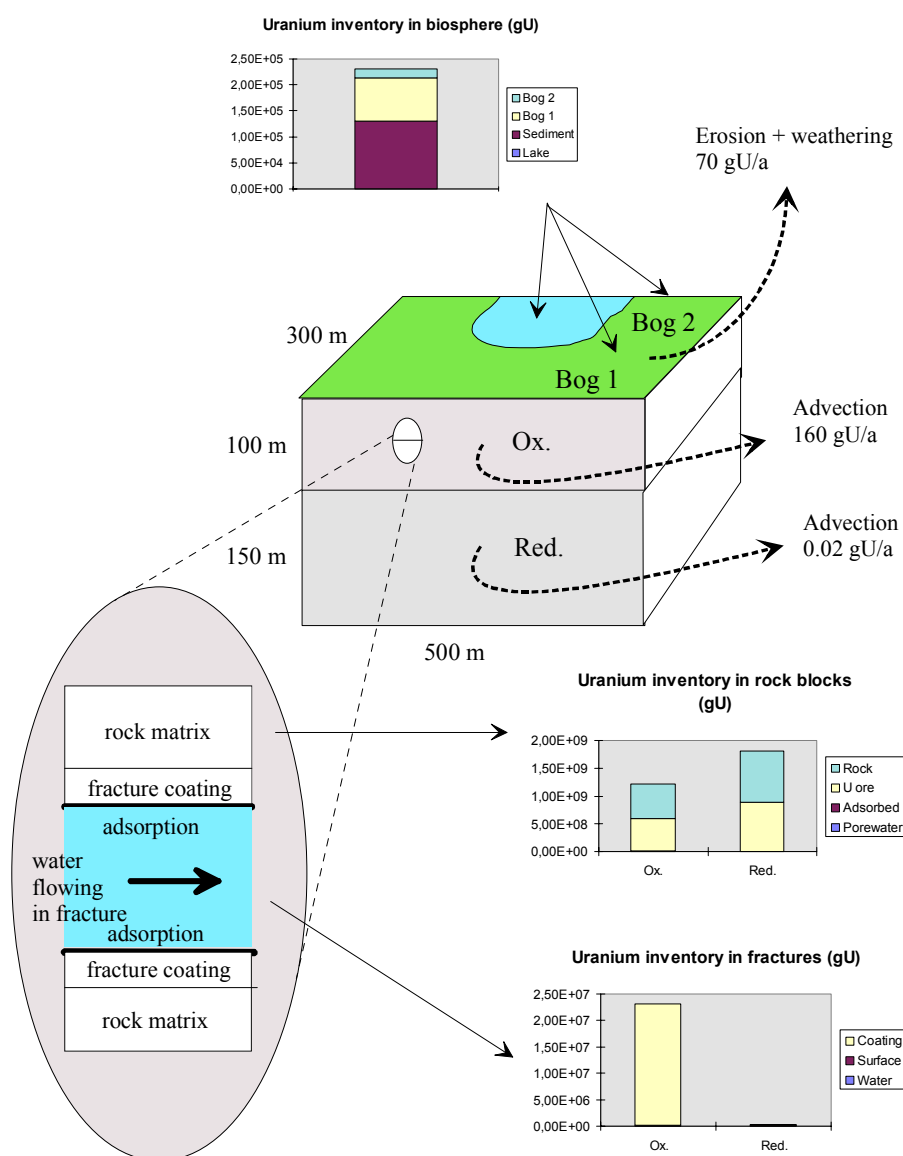


FIG. F4. U inventories and fluxes for a bedrock block at Palmottu.

Geochemical fluxes from potential repository depth at each of four candidate sites were calculated through 1 km × 1 km reference planes which are located at about 500 m depth; the planes were divided into subregions of size 25 m × 25 m. As the hydraulic properties at the sites show most pronounced changes at about that level, sensitivity calculations also at about 400 m and 600 m depth were conducted. Natural geochemical concentrations, however, were calculated from depth zones, which are approximately reference plane depth ± 100 m

(Hästholmen:  $\pm 170$  m), depending on the availability of analytical data. The relative variation of salinity can be significant near the reference plane. The fluxes computed for this work were based on the site-scale groundwater flow analyses carried out for the latest safety analysis TILA-99. The analyses for the two inland sites, Kivetty and Romuvaara, comprised of steady state simulations of groundwater flow, while a transient and coupled flow and solute transport was simulated for the two coastal sites, Olkiluoto and Hästholmen (Loviisa).

The computed fluxes through the planes (fluxes are decreasing significantly with increasing depth) show that a major part of water flows through the subregions intersected by the fracture zones. This is natural because these zones have higher hydraulic conductivity than the surrounding intact rock (i.e. sparsely fractured rock). The flow directions relative to the planes are mainly downwards in the intact rock of all the four sites. On the other hand, along the fracture zones water flows both upwards and downwards depending on the hydraulic connections and properties of the zones. The calculated fluxes at repository depth at each site are not perfectly consistent with hydrogeochemical results; for example, groundwater chemistry often indicates significantly longer mean residence times. The longer mean residence times in the coastal sites than in the inland sites can be partly explained by higher head fields at the inland sites, but also by the high density gradient in groundwater at the coastal sites may have decreased the descend of younger groundwater types during post-glacial period.

Contents of radioactive elements vary significantly between the sites in groundwaters at about 500 m depth. Uranium concentrations correlate relatively with bedrock contents except at Olkiluoto where U content in deep groundwater is particularly low. In shallow groundwaters, in contrast, U contents in rock and water seem to correlate. These waters generally originate from oxic conditions dissolving U evenly and reflecting the U content of the environment. It seems that the U concentrations near the surface are dominated by variations of water-rock interaction times (low values at the inland sites, higher values at the coastal sites). Deep groundwaters are from reducing environment where U is typically precipitating. At the four sites the U concentration decreases downwards. However, uranium measured from samples may have been released in completely different location than where sampling has occurred. Therefore, the whole flow path is important in evaluating U concentrations in groundwaters.

The mean contents of U in deep groundwaters are systematically lower than in shallow groundwaters, particularly in Olkiluoto. Variation of geochemical fluxes depends significantly on groundwater fluxes in addition to geochemical concentrations. Variations of groundwater fluxes between the corresponding reference planes of different sites are less than one order of magnitude, but the range between the deep and discharge planes is about two orders of magnitude at a single site, except at Kivetty where it is one. There are several reasons to assume that hardly any of the U flux at the surface is derived from the reference plane level; chemical and isotopic parameters of the water support this conclusion. Hydrogeochemical interpretations indicate that the salinity level has been preserved since the last glaciation, thus the flux from the depth may be at the maximum at the per mille level in shallow flux at both coastal sites. Mixing calculations and salinity distributions suggest even that meteoric water circulation in these sites has been limited to the upper 100m depth. The estimated U fluxes are summarized in Table F3 (rounded to two decimals).

TABLE F3. ESTIMATED U FLUXES THROUGH REFERENCE PLANES AT FOUR REPOSITORY INVESTIGATION SITES

U flux (g/km <sup>2</sup> a) (range)	Olkiluoto (coastal site)	Hästhölm (coastal site)	Romuvaa (inland site)	Kivetty (inland site)
Repository depth: upper level	0.35 (0.027–1.2)	3.6 (0.96–9.6)	3.0 (0.10–14)	32 (6.3–96)
middle level	0.028 (0.005–0.060)	1.9 (0.3–6.1)	0.32 (0.03–0.70)	30 (6.0–81)
lower level	0.0093 (0.0062–0.016)	0.55 (0.035–1.9)	0.16 (0.014–0.34)	11 (2.6–35)
Surface discharge	73 + 740 (71–74) + (740–750)	680 (270–990)	58 (20–160)	33 (4.7–80)

At the end a summary of uranium fluxes is given and the values, measured and estimated by different methods and in different scales, are compared (Table F4). It becomes evident from the results that the geochemistry of the groundwater-rock system and the chemical properties of the element are very prominent factors determining natural geochemical fluxes; the redox conditions are ranging at the top of the list for U. It becomes also clear that U is the most suitable element that is reasonable to examine between the source area and possible discharge area within this type of study. In analogy to spent nuclear fuel in a repository also the U occurring in nature is dissolved from a primary source, is migrating in the groundwater-rock system and, finally, eventually repeatedly re-enriched and remobilized or dispersed. Also arsenic, originating from dissolution of sulphides, seems to be a suitable element, although there are less data available; indeed, in areas where most prerequisites for deep groundwater circulation are fulfilled (high topographic relief; deep-reaching, conductive faults; heat or gas flux from depth) As has been identified as a typical element from deep circulation systems in crystalline rocks (see: Report 3 in the reference list). Ra would be suitable too, if more data would be available. The suitability of Th is hampered by very low solubilities near analytical detection limits. Transition elements such as Cu typically show low concentrations in deep flow systems due to reducing conditions and pH buffering by water-rock interaction; only surface-weathering conditions in the presence of sulphides result in enhanced concentrations due to low pH values. The use of the elements K and the very similar Rb does not make sense in this natural flux study: in the Finnish bedrock groundwater they are not dissolved from a source in the rock, but fluxes come from mixing processes with water bodies having high contents such as fossil or recent seawater or old brines.

TABLE F4. U FLUXES (IN DISSOLVED FORM) MEASURED OR ESTIMATED BY DIFFERENT METHODS AND IN DIFFERENT SCALES (SITE SPECIFIC TO COUNTRY-WIDE) NEAR THE SURFACE (OXIDIZING CONDITIONS) AND AT DEPTH (REDUCING CONDITIONS)

U flux, g/km <sup>2</sup> a	oxidizing zone	reducing zone
Small catchments, measured	3–43	
Headwater streams, measured	17	
Chemical erosion, (till), estimated	54	
Palmottu U-mineralization, estimated	3200	0.3
Repository investigation sites, estimated	33–990	0.01–32

### Interpretation and application of the concentration and flux data

Geochemical mapping data of Finland has been used for mapping of areas where concentrations of harmful elements exceed permissible limits and for epidemiological studies. The focus was on F, As and U, but also on some other heavy metals. The limit for As was in about 1% of the drilled wells exceeded and very high values were found in single cases. There seemed to be an association between high As values and bladder cancer. In baseline studies at repository candidate sites it was found that U contents in drinking water from drilled wells in bedrock were roughly one order of magnitude higher than in municipal water supplies and the significant role of U in the radiation exposure of the population was emphasized. In drilled wells the local value for Loviisa was about twice the average for the whole country. In Finland it is estimated that in about 100 000 springs and drilled wells the limit for U of 2 µg/l is exceeded which means that a population of about 800 000 may be exposed to high U levels. The maximum U content measured in a drilled well in bedrock was 12 400 µg/l. The role of concentrations as a measure to evaluate possible harmful effects is emphasized here, as the comparison of fluxes alone can hide the problem of high concentrations of harmful elements under certain circumstances.

In the Finnish literature results of performance assessment simulations for chemical long term environmental effects of the substances released from the materials in a nuclear waste repository are reported. The models used were partly simplified from the recent TILA-99 performance assessment exercise and thus gave rough overestimations of releases which were accompanied by large uncertainties. The resulting concentrations in drilled well water were compared with current drinking water standards. Although the solubility limits of many of the substances are higher than the permissible concentrations in drinking water, due to the low water flow the calculated release rates are very low and the dilution factor is high, so that the resulting concentrations in the well water are at least four orders of magnitude below permissible limits.

Natural concentrations and fluxes from Finland have been reported to the EU project "Testing of safety and performance indicators" and were included in the list of reference values, but any detailed comparisons with performance assessment results obviously could not be conducted within the frame of the project. Recently site specific data from Palmottu has been



compared with performance assessment predictions (Read et al., 2002). The TILA-99 performance assessment was designed to be transparent and is readily amenable to comparison with alternative approaches. Dose calculations relate to a well for drinking water that is assumed to be located in the vicinity of the repository or in the groundwater discharge zone. This is the only exposure pathway considered. The model employed assumes that the annual releases from the repository into the biosphere are diluted in 100 000 m<sup>3</sup> of water and that an individual drinks 500 dm<sup>3</sup> of this water per year. For cases where the canister is assumed to ‘disappear’, the maximum release rate for U from the geosphere is given as only 1.2 Bq y<sup>-1</sup> (and then after 1 million years). This equates to a concentration of 4 · 10<sup>-15</sup> mol dm<sup>-3</sup>. In comparison, uranium concentrations measured at Palmottu range from around 4 · 10<sup>-9</sup> mol dm<sup>-3</sup> for the most reducing waters to >10<sup>-6</sup> mol dm<sup>-3</sup> nearer the surface. The safety case considered release from only one canister; however scaling up to match the total inventory at Palmottu (~1000 t), is insufficient to account for such a large discrepancy. If all the canisters were assumed to disappear there would still be 10<sup>3</sup>–10<sup>6</sup> times less uranium in the simulated well water than is actually observed in the geologically similar Palmottu system (Figure F5). The difference is entirely due to assumptions made regarding the geochemical behaviour of uranium. This is not only a feature of TILA-99 but of all current performance assessment calculations.

An interesting issue is to see nuclide-specific constraints for release of activities to the biosphere according to the Finnish regulations (Guide YVL 8.4) when they are converted to mass units in relation to natural fluxes; a limit of 0.3 GBq/a for U isotopes (averaged over <1000 a) is equivalent to as much as 30 kg/a.

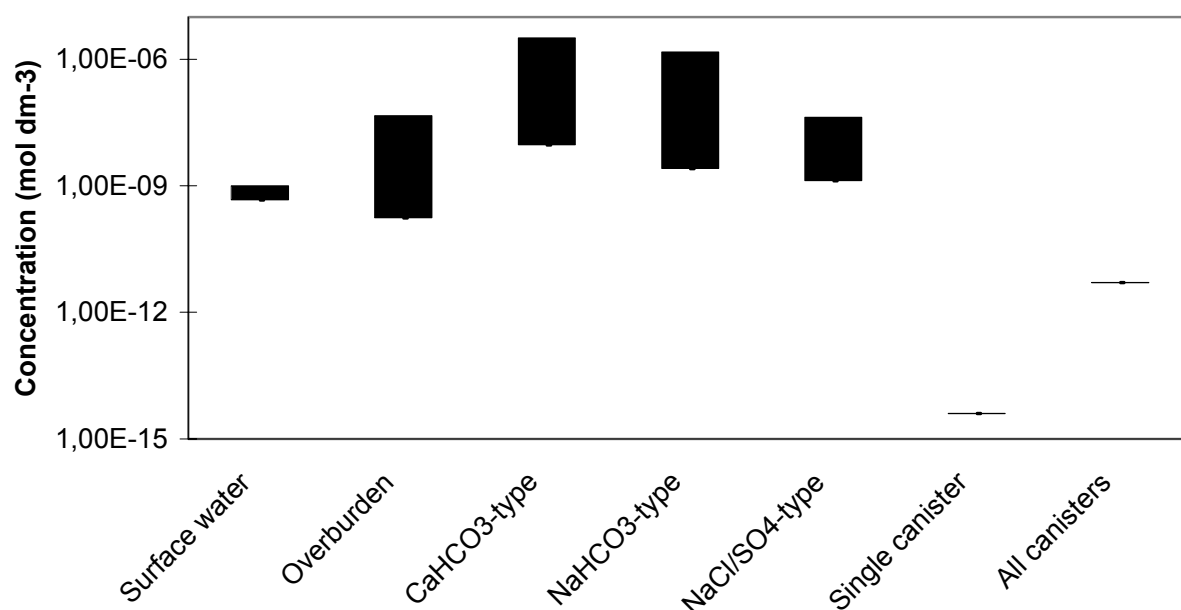


FIG. F5. Uranium concentrations in Palmottu surface and groundwaters compared to performance assessment predictions for a drinking water well.

In the future it is expected that based on a better understanding of natural U fluxes more realistic models for radionuclide migration in the geosphere can be developed and the development of performance assessment tools can benefit from these. As a first step preliminary results of these alternative models can be compared with results obtained by conventional performance assessment tools. When the behaviour of U at the Palmottu site can

be simulated satisfactorily, then also the behaviour of spent fuel at the same site can be tried to simulate and thereafter the alternative modelling approach can be tested in the Olkiluoto system.

## Conclusions

The benefits of the CRP for Finland can be divided into three main topics:

- a comprehensive data base of natural concentrations and fluxes has been compiled for derivation of reference values in different scales (for example, country-wide, according to geochemical provinces and site specific) as well as for various compartments or materials.
- The achieved basic understanding of the processes leading to mobilization and fixation (dispersion and enrichment) of elements in geochemical cycles and the consideration of the global context in terms of geology, topography and climate implies a significant increase of confidence in the ability to understand and quantify the long term behaviour of a nuclear waste repository. The only available geological option in Finland which is characterized by water-saturated, fractured crystalline rock, glaciated terrain, peneplane, secondary sinks etc. can now be better evaluated against alternative geological options all over the world. For example, the geological conditions in Finland do not favour large geochemical fluxes mediated by groundwater from deep formations to the surface. In addition, more detailed knowledge of the complexity of natural processes facilitates drawing simplifying conclusions which can be beneficial in the communication with non-geoscientific and non-technical audiences.
- Detailed and quantitative knowledge of local, element-specific natural geochemical fluxes is being used to develop better modelling tools to be integrated into a safety case, that take account of interactions between geochemical and hydrological processes and that fundamentally do not misrepresent the known geochemical behaviour of the elements.

The assumption in repository performance assessments of simple dispersion and dilution of ions released from the waste, transported through the geosphere by groundwater and finally reaching the biosphere is not supported by observations from natural anomalies. In nature complex mobilization, migration and, in particular, reconcentration (followed eventually by remobilization) processes prevail. The more near to the top soil layers metal ions migrate the more complex becomes their behaviour and the more difficult predictions can be made.

The major weakness of current performance assessment exercises which is significantly decreasing credibility is that safety is expressed in a few numbers giving levels of doses or risks. When selecting and expressing reference values of natural concentrations and fluxes as single numbers there is the same risk that the underlying complex geochemistry of interactions with the geosphere is not taken into account and possibly wrong numbers are applied. Concentrations and fluxes of elements are no natural constants and cannot be derived in a straight-forward way, but are the result of the influence of chemical conditions, gradients, fronts in the source materials and substrates and along transport paths.

Experiences from Finland show that under the influence of the presence of mostly unweathered, largely crushed bedrock, low hydraulic conductivity in till and groundwater saturation with groundwater levels often relatively near to the top level of the regolith profile, in connection with a low relief, leading often to less oxygen-rich or partly reducing conditions already below less than a metres depth due to the high buffer capacity of the fresh, fine-grained rock material and the low groundwater flow rate, chemical fronts of pH, redox

conditions, anion and complexant concentrations etc. can act as effective chemical barriers limiting the movement of elements and leading to secondary enrichments depending on their respective chemical properties. When these natural geochemical characteristics are taken into account in future repository performance assessments then the comparison with natural elemental concentrations and fluxes makes sense, can support a safety case, and is according to the protection goals of the Finnish regulations giving nuclide-specific release limits.

### **Additional information sources**

In this appendix only a summary of the most prominent results contributed to the CRP are given. For detailed references (original citations) we give a list of reports which contain all the information from the literature, on the methods and data, as well as results of supporting studies putting the Finnish situation in a more global context. These reports have been published all before the end of 2003.

- [F1] VAGANOV, P., IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 1, Geochemical Cycles and the Dispersion and Concentration of Elements in the Earth's Crust — Global, Regional and Local Scale —  
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- [F6] KAIJA, J., RASILAINEN, K., BLOMQVIST, R., IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 6, Site Specific Natural Geochemical Concentrations and Fluxes at the Palmottu U-Th-mineralization (Finland) for Use as Indicators of Nuclear Waste Repository Safety. Rep. YST-114, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland (2003).
- [F7] PITKÄNEN, P., LÖFMAN, J., LUUKKUNEN, A., PARTAMIES, S., IAEA Coordinated Research Project, The Use of Selected Safety Indicators (Concentrations, Fluxes) in the Assessment of Radioactive Waste Disposal, Report 7, Site Specific Natural Geochemical Concentrations and Fluxes at Four Repository Investigation Sites in Finland for Use as Indicators of Nuclear Waste Repository Safety., Rep. YST-115, Geological Survey of Finland, Nuclear Waste Disposal Research, GSF, Espoo, Finland (2003).
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## APPENDIX G

### JAPAN

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#### Abstract

The geochemical data survey has been conducted based on domestic literatures and provided countrywide concentration ranges and averages for a suite of elements and radionuclides, covering many different rock types. Chemical species data of U, Th, K, Rb, Sn, Cs and Cu are searched and summarized for the elemental concentration of water (surface and subsurface), rock (sedimentary and crystalline) and fragments (soil, stream/lake sediment and marine sediment), and those of U, Th, K, Cs, Ra and Rn for the radionuclide concentration data of the same materials. Process data such as denudation rate and river flow rate to evaluate elemental and radionuclide fluxes were also summarized based on literatures survey. A large effort has been made to develop and test methodologies for the application of natural safety indicators to safety assessments. The repository-derived nuclide concentrations and fluxes evaluated in the geosphere and biosphere in the H12 repository system were compared with measurements of naturally occurring radionuclides or their evaluated fluxes.

#### Background

##### Nuclear programmes

Since the first commercial operation of a nuclear power plant in 1966, the use of nuclear energy in Japan has expanded steadily and currently makes up around one third of the total electricity production. Fifty-three commercial nuclear power units with a total installed capacity of 45,907 MW are now in operation. In addition, four units are under construction and six units are in preparation to construction at the end of January 2002.

Nuclear fuel cycle technologies have been developed in order to continue the nuclear energy supply over a longer period of time by effective use of such materials as plutonium and uranium recovered from spent fuel. Nuclear energy policy in Japan calls for reprocessing of all spent fuels. Japan Nuclear Fuel Limited (JNFL) is constructing a commercial reprocessing plant at Rokkasho-mura, which is planned to start operation around 2005.

In Japan, radioactive waste is categorized into two basic types: HLW and LLW. HLW is the high-level liquid waste separated at spent fuel reprocessing or in its final vitrified form. All other wastes are generally regarded as LLW and further categorized into several types primarily based on their origin and nature: nuclear power plant (NPP) waste, transuranium (TRU) contaminated waste, uranium waste and radioisotope and institute (RI/Institute) waste.

The management policy and regulations of these HLW and LLW have been established step-wise by Atomic Energy Commission (AEC) and Nuclear Safety Commission (NSC) of Japan, and some NPP wastes have been disposed in Rokkasho Center operated by JNFL since 1992. The overall waste disposal policy by AEC was established by December 2000. Especially, for HLW, the government and related organizations should actively push forward with R&D activities, including scientific studies of deep geological environment, and with development of technologies to enhance reliability of geological disposal technology.

Based on progress to date, in the year 2000 the geological disposal program of HLW in Japan moved from the phase of generic R&D into the phase of implementation. The “Law on Final Disposal of Specified Radioactive Waste” came into force in June 2000, taking into account the technical achievements documented in H12 report published by Japan Nuclear Cycle Development Institute (JNC). This report shows that sufficiently stable geological environment exists commonly in Japan and indicates that the evaluated disposal systems can be secured of safety having sufficient margin for natural radiation exposure level taking account alteration of natural phenomena and uncertainties in models and data. Pursuant to the overall HLW management program, and in accordance with the new Law, the Nuclear Waste Management Organization of Japan (NUMO) was established in October 2000 with responsibility for implementing geological disposal of HLW. The assigned activities of NUMO include selection of the repository site, demonstration of disposal technology at the site, relevant licensing applications and construction, operation and closure of the repository. According to the present time schedule, repository operation will start in the decade of the 2030s.

A generic performance assessment (PA) was conducted in the H12 report. An assessment method was developed and applied in the H12 to evaluate the safety functions and the level of robustness of the proposed system under various conditions taking account of a wide range of potentially suitable Japanese geological environments. The assessment included a comprehensive evaluation of uncertainty and potentially detrimental factors, including perturbations due to external events and processes.

In the framework of H12 performance analysis and subsequent performance assessment studies, consequences have been evaluated and discussed in terms of supplementary safety indicators such as concentrations and fluxes as well as doses.

### **Topography, geology and climate**

Japan is situated on the eastern margin of the Eurasian Continent and composed of several island arcs which have been formed by the subduction of the Pacific Plate and the Philippine Sea Plate to under the Eurasian Plate and so on. As Japan is characterized by such Circum-Pacific Orogeny, there exist a variety of topographies from mountainous regions, with more than three thousands of meters of an altitude, which have continued to uplift, to plain regions that have continued to subside to a depth of more than one thousand of meters in the Quaternary period. Volcanic or steep erosional topographies can be seen widely in Japan while gently eroded topography due to weathering is also admitted in some areas of Japan. It has 59% of mountainous areas, 6% of volcanoes, 11% of hills, 11% of heights and 13% of lowlands. Such a variety of topography is due to crustal movement and a mild climate with a plenty of precipitation in the middle latitude.

The bedrock of Japan is mainly formed by an accretionary complex and igneous rocks such as volcanic rocks and granitic rocks which intruded the complex. Less consolidated sedimentary rocks and unconsolidated sediments of Late Neogene to Quaternary periods are also distributed in subsidence regions that have been locally formed in some of plate margins. Quaternary volcanoes form the volcanic fronts that are situated parallel to trenches and no volcano exists in the ‘forearc’ regions of the fronts. Earthquakes have frequently occurred all over Japan because it is situated on the subduction zones.

Japan is situated in a mid-latitude region of 25°N to 45°N, and surrounded by two warm currents, the Black Current in the southern side, and the Tsushima Current in the western and northern sides, and a cold current of the Oyashio Current in the eastern side. Present Japan belongs to the temperate zone, in particular the warm and humid climate zone, and has a

relatively large amount of precipitation with an annual mean value of about 1700 mm. Since Japan is especially influenced by monsoons from the tropical belt in summer and from Siberia in winter, each season has distinct characteristics. Also, as there exist high mountains along the centre part of the island arcs, there are differences in climate between inland and coastal regions, and between the Pacific side and the Japan Sea side even at the same latitude.

### **Study areas**

No study areas have yet been determined in Japan.

### **Elemental and radionuclide concentration data**

#### **Sources of data**

The sources of the elemental and radionuclide concentration data acquired in Japan are all those which have been published and are now available by approaching several database such as GEOLIS-JP [G1], which is a bibliographic database on earth sciences, compiled by the Library of the Geological Survey of Japan, AIST, and the publication of nuclear-related academic societies and institutes. The data acquired are available for different materials (sedimentary and crystalline rocks, fragments such as soils, stream/lake sediments and marine sediments, and water such as surface and subsurface water) depending on each of original literatures.

#### **Summary of data**

Chemical species data of U, Th, K, Rb, Sn, Cs and Cu are available for the elemental concentration of water (surface and subsurface), rock (sedimentary and crystalline) and fragments (soil, stream/lake sediment and marine sediment), and those of U, Th, K, Ra and Rn for the radionuclide concentration of the same materials. Those data are principally countrywide, but some of site specific data are also compiled. These data are collected from the literatures in which the analytical methods or the geological information of the targeted area have been appended clearly. Tables G1 and G2 show the elemental and radionuclide concentration data collected from the literature.

The statistical data, the log-normal distribution and the other information (the range of data and remarks) of the concentration data are shown in the additional information on CD. Among all of the statistical data which were widely collected and treated based on a variety of literatures, the following datasets are well naturally distributed; elemental concentrations of Th (crystalline rock;  $n = 410$ ), K (subsurface water, sedimentary rock and crystalline rock;  $n = 328, 418$  and  $1663$ , respectively), Rb (crystalline rock;  $n = 1110$ ), Cs (crystalline rock;  $n = 319$ ), and Cu (sedimentary rock, crystalline rock and stream/lake sediment;  $n = 210, 873$  and  $278$ , respectively), and radionuclide concentration of  $^{222}\text{Rn}$  (subsurface water;  $n = 309$ ).

Some other site specific datasets among all of the literatures surveyed are as follows. The datasets (as average values of  $0.0254 \mu\text{g/l}$  of U and  $0.0035 \mu\text{g/l}$  of Th) of 109 specimens from 82 river water [G2] and that (as the same of  $0.000271 \text{ Bq/l}$  of U) of 27 specimens from 23 river water [G3] are some of the well-documented results which have been widely obtained in Japan. The U data of river waters near Lake Biwa [G4], subsurface waters at Okinawa [G5] and San-in [G6], sedimentary rocks at Kitakami, [G7] and soils at Osaka [G8] are also site specifically documented well. As to radioactive concentration data of Ra, the datasets of  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$  in soils were obtained from Osaka [G8] and several regions in Japan [G9], respectively. Regarding the elemental concentration data of Sn, a countrywide dataset of crystalline rock obtained as GSJ Geochemical Reference samples [G1] can be shown.

TABLE G1. ELEMENTAL AND RADIOACTIVE CONCENTRATION DATA ON U, TH, K, Rb and Sn

Chemical species	Materials		Elemental concentration (mg/kg or µg/l)		Radioactive concentration (Bq/kg or Bq/l)		Scale
			Number	Average	Number	Average	
U	Water	Surface	42	-	51	7.95E-03	countrywide
			109	2.54E-02	0	0.00E+00	countrywide
		Subsurface	25	-	29	-	countrywide & local
	Rock	Sedimentary	168	2.89E+00	36	-	local
		Crystalline	188	1.89E+00	0	-	countrywide
	Fragment	Soil	68	-	60	3.53E+01	countrywide & local
		Stream / lake sediment	5	2.29E+00	5	-	local
		Marine sediment	97	4.73	0	-	local
Th	Water	Surface	26	-	0	-	countrywide
			109	3.50E-03	0	0.00E+00	countrywide
		Subsurface	2	-	10	3.23E-01	local
	Rock	Sedimentary	202	7.85E+00	0	-	local
		Crystalline	410	2.97E-02	0	-	countrywide
	Fragment	Soil	104	-	18	-	countrywide
		Stream / lake sediment	7	-	0	-	local
		Marine sediment	50	-	0	-	local
K	Water	Surface	53	-	19	-	countrywide
		Subsurface	328	-	0	-	countrywide
	Rock	Sedimentary	418	1.73E+04	1	-	countrywide
		Crystalline	1,663	1.47E+04	0	-	countrywide
	Fragment	Soil	115	1.75E+04	160	5.35E+02	countrywide
		Stream / lake sediment	8	-	39	-	local
		Marine sediment	1	-	9	-	local
Rb	Water	Surface	19	-	-		countrywide
			28	1.66E+00			countrywide
		Subsurface	12	-			local
	Rock	Sedimentary	97	-			countrywide
		Crystalline	1,110	5.89E+01			countrywide
	Fragment	Soil	21	1.66E+02			local
		Stream / lake sediment	5	-			local
		Marine sediment	0	-			-
Sn	Water	Surface	0	-	-		-
		Subsurface	0	-			-
	Rock	Sedimentary	10	-			local
		Crystalline	121	1.31E+00			countrywide
	Fragment	Soil	0	-			-
		Stream / lake sediment	5	-			local
		Marine sediment	12	-			local

Note : mg/kg and Bq/kg in solid materials , and µg/l and Bq/l in water



TABLE G2. ELEMENTAL AND RADIOACTIVE CONCENTRATION DATA ON Cs, Cu, Rn and Ra

Chemical species	Materials		Elemental concentration (mg/kg or µg/l)		Radioactive concentration (Bq/kg or Bq/l)		Scale
			Number	Average	Number	Average	
Cs	Water	Surface	3	-	33	2.82E-04	countrywide
			28	3.91E-02	0	-	countrywide
		Subsurface	5	-	0	-	local
	Rock	Sedimentary	15	-	0	-	local
		Crystalline	319	3.00E+00	0	-	countrywide
	Fragment	Soil	2	-	0	-	local
		Stream / lake sediment	7	-	0	-	local
		Marine sediment	0	-	0	-	-
Cu	Water	Surface	51	-	-		countrywide
			38	1.96E+00			countrywide
		Subsurface	5	-			local
	Rock	Sedimentary	210	3.72E+01			local
		Crystalline	873	3.14E+01			countrywide
	Fragment	Soil	25	-			local
		Stream / lake sediment	278	8.06E+01			local
		Marine sediment	12	-			local
Rn	Water	Surface	0	-	31	-	local
		Subsurface	0	-	309	-	countrywide
	Rock	Sedimentary	0	-	0	-	-
		Crystalline	0	-	0	-	-
	Fragment	Soil	0	-	0	-	-
		Stream / lake sediment	0	-	0	-	-
Ra	Water	Surface	0	-	4	-	local
		Subsurface	0	-	47	-	local
	Rock	Sedimentary	0	-	0	-	-
		Crystalline	2	-	8	-	local
	Fragment	Soil	0	-	67	-	countrywide & local
		Stream / lake sediment	0	-	0	-	-
		Marine sediment	0	-	0	-	-

Note : mg/kg and Bq/kg in solid materials , and µg/l and Bq/l in water

## **Elemental and radionuclide flux data**

### **Sources of data**

The sources of the rates of geological processes acquired in Japan are all those which have been published and are now available by approaching e.g. JNC's H12 report [G10], Japanese statistical information [G11] and GEOLIS-JP [G2].

### **Summary of data**

#### **(1) Distribution of denudation rate in Japan**

The countrywide denudation rate map is shown in Figure G1 [G10]. The rate was introduced from the relationship between the standard dispersion of altitude and the denudation rate, and is shown as an average denudation on the area of  $6 \times 6$  km. The denudation rate is less than 0.5 mm per year in most areas of Japan, except in some high mountain regions and at the tips of peninsulas, where the rate exceeds more than 1 mm per year in the central part.

#### **(2) River flow rate in Japan**

The river flow rate of the first-grade river in Japan is shown in the additional information on CD. The range of river flow rate is from 300 000 to 3 000 000  $\text{m}^3 \text{y}^{-1} \text{km}^{-2}$  [G11].

#### **(3) Others**

The data acquired are precipitation, eolian dust accumulation rate, stream load and recharge. They are principally country-wide and analysed according to each degree of data acquisition on original literatures.

## **Interpretation and application of the concentration and flux data**

Using the H12 model chain for the Reference Case [G10], concentrations and fluxes of radionuclides and radiotoxicity can be calculated at various points in the disposal system and compared with those of naturally occurring radionuclides or other indices.

### **Radionuclide concentrations in river water**

Assuming an uncontaminated river water flow rate of  $10^8 \text{ m}^3 \text{y}^{-1}$ , which is the same as in the H12 biosphere model, derives concentrations in river water due to releases from repository. Figure G2 shows a comparison of the calculated concentrations in a river of 4N+2 chain members released to the biosphere with the ranges of naturally occurring concentrations [G12]. The Figure also shows the maximum permissible uranium concentration according to the Drinking Water Quality Guidelines of the World Health Organization [G13]. The comparison shows that the concentrations of radionuclides originating from the repository would be several orders of magnitude lower than those of naturally occurring radionuclides and the WHO guideline.

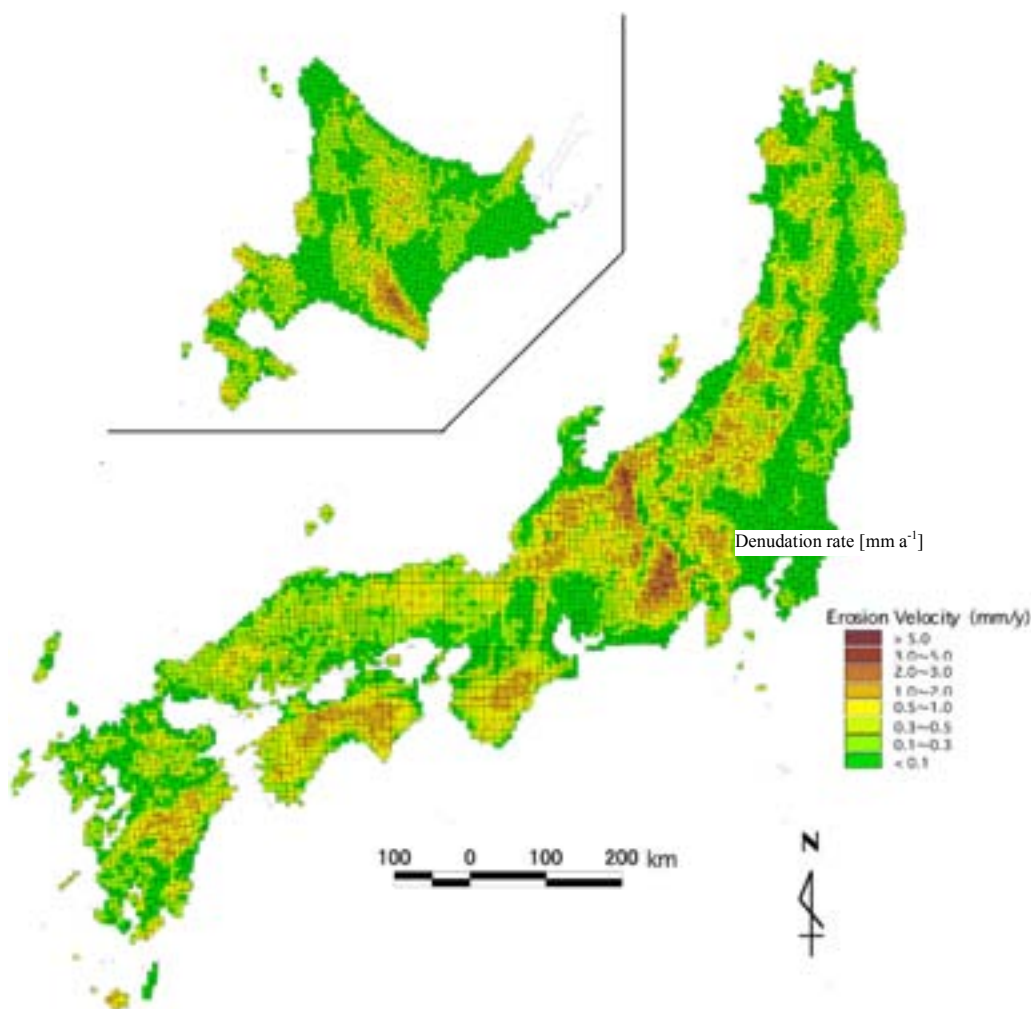


FIG. G1. Distribution of denudation rates [G10]. The denudation rates were calculated for  $6 \text{ m}^2$  areas based on topographic relief according to a relationship estimated from the results of a survey at 82 dams throughout Japan.

### Radionuclide concentrations in groundwater

Figure G3 illustrates the results of a comparison made between the concentration of U-238 in the geosphere due to EBS release (as a function of distance from the EBS and for different times and the concentration of U-238 naturally present in groundwater, measured at various locations in Japan [G12]. For natural U-238 concentrations in groundwater, available data measured in both crystalline (Tono area and Kamaishi mine) and sedimentary rock (Tono area) are indicated. In addition, the Figure shows measured natural U-238 concentrations in hot spring waters sampled in the San-in district, which are relatively high since these waters are located in near surface under oxidizing conditions.

From this Figure, it is apparent that the predicted concentration of U-238 in groundwater increases for up to  $10^8$  years after disposal, and then decreases. The comparison suggests that the concentration of U-238 released from a repository would only reach the same level as that of natural radionuclides. This is reasonable as, in both the EBS and in natural waters, the uranium concentration is limited by its low solubility. In the calculation, uranium solubility has conservatively been set as  $8 \times 10^{-9} \text{ mol m}^{-3}$ , which is larger than field measurements.

## **Equivalent fluxes of U-238**

Direct comparisons such as those above are possible for radionuclides that exist in nature, but arguments based on analogy are required for those nuclides that do not occur naturally. To complement these, the radionuclide flux from the geosphere (i.e. for the MWCF) in the H12 Reference Case can be converted to equivalent fluxes of U-238 using the Annual Limit of Intake (ALI) [G14]. These results are shown in Figure G4 [G15].

It can be considered that there are many types of (surface) water systems that could receive radionuclides released from the geosphere. River water is one of the most likely. In Figure G4, the equivalent flux of U-238 from the Reference Case is compared with that of natural radionuclides fluxes based on measured U and Th concentration in eight rivers of around  $10^8$  to  $10^9 \text{ m}^3 \text{ y}^{-1}$  flow rate [G16], which are comparable with the flow rate  $10^8 \text{ m}^3 \text{ y}^{-1}$  for the H12 biosphere model in the Reference Case. It is apparent that the equivalent flux due to the repository is around two orders of magnitude lower than that of naturally occurring uranium. It is still lower than the range of natural radionuclides fluxes of groundwater based on measured U and Th concentration in groundwater [G10] and an average pumping rate ( $2 \times 10^6 \text{ m}^3 \text{ y}^{-1}$ ) of deep well water [G17].

## **Qualitative “what if?” calculations for natural disruptive events**

For isolation failure scenarios due to natural disruptive events, such as uplift and erosion, fluxes in solid material are relevant indicators for comparison with naturally occurring radionuclide fluxes due to these events.

Equivalent fluxes of U-238 due to the repository are calculated as “what if?” analyses for two uplift/erosion rates and two initial repository depths: 1000m (crystalline rock and Pre-Neogene sedimentary rock: hard rock) and 500m (Neogene sedimentary rock: soft rock). The earlier the repository reaches the ground surface, the greater the calculated fluxes (see Figure G5). Results are relatively insensitive to initial depth for the faster erosion rate, but more sensitive for the slower rate. This is because, for the slower erosion rate, Np-237 (which is a key nuclide with a half-life of about two million years) begins to decay significantly before the repository is exposed at the surface [G15].

## **The methodology of natural flux assessment**

A hypothetical methodology of natural flux assessment has been discussed in consideration of specific conditions regarding topographical, geological and climatic features and existing matter cycles as well as social aspects. For the adoption of the catchment area concept for this methodology, how to determine fluxes and assessment points and how to deal with uncertainties within fluxes and related process rates are identified as key factors.

## **Future tasks**

Above studies illustrate the examples of application of supplementary safety indicators for long term safety assessment of geological disposal. In the characterization phase at a specific site, it will be possible to measure site specific concentrations and fluxes of naturally occurring radionuclides and to compare them with the results of performance analyses.

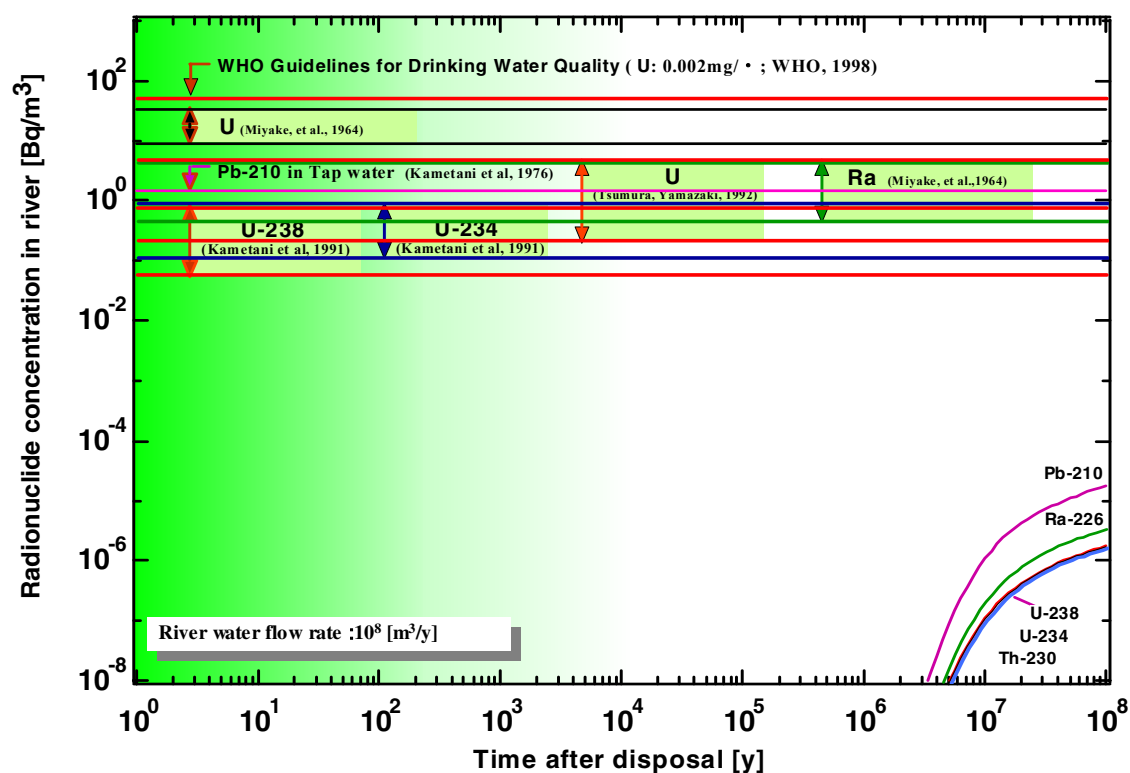


FIG. G2. Comparison of calculation results (Reference Case) and natural radionuclides: concentration of 4N+2 natural decay series nuclides in river water [G12].

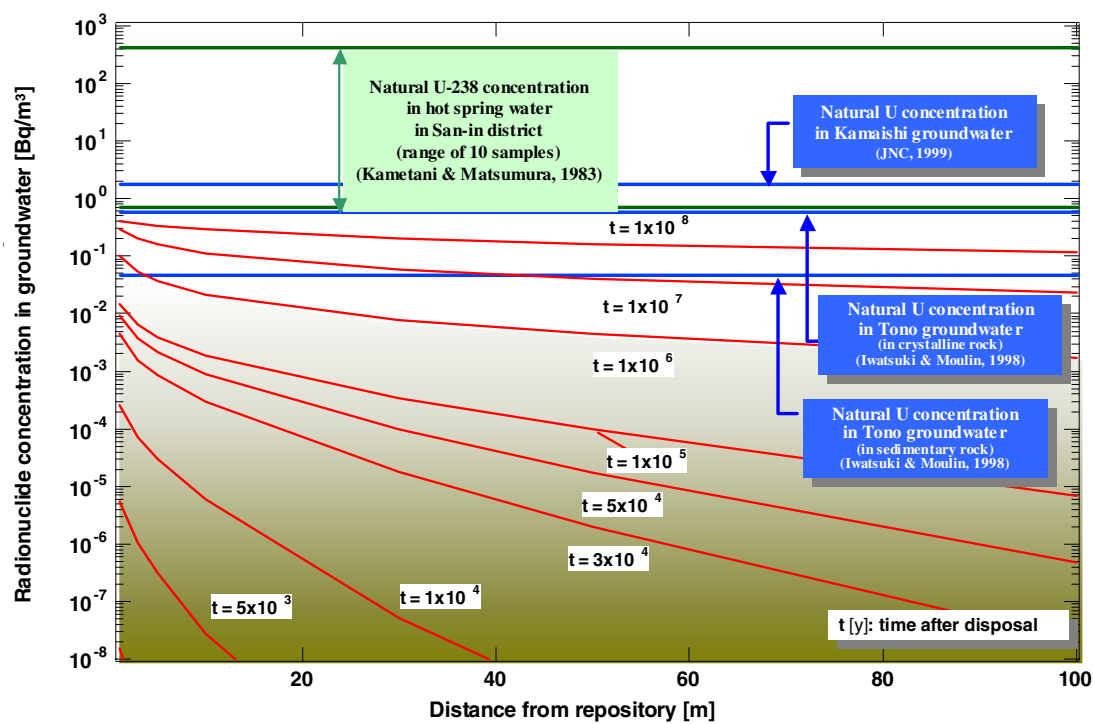


FIG. G3. Comparison of calculation results (Reference Case) and natural radionuclides: U-238 concentration in groundwater [G12].

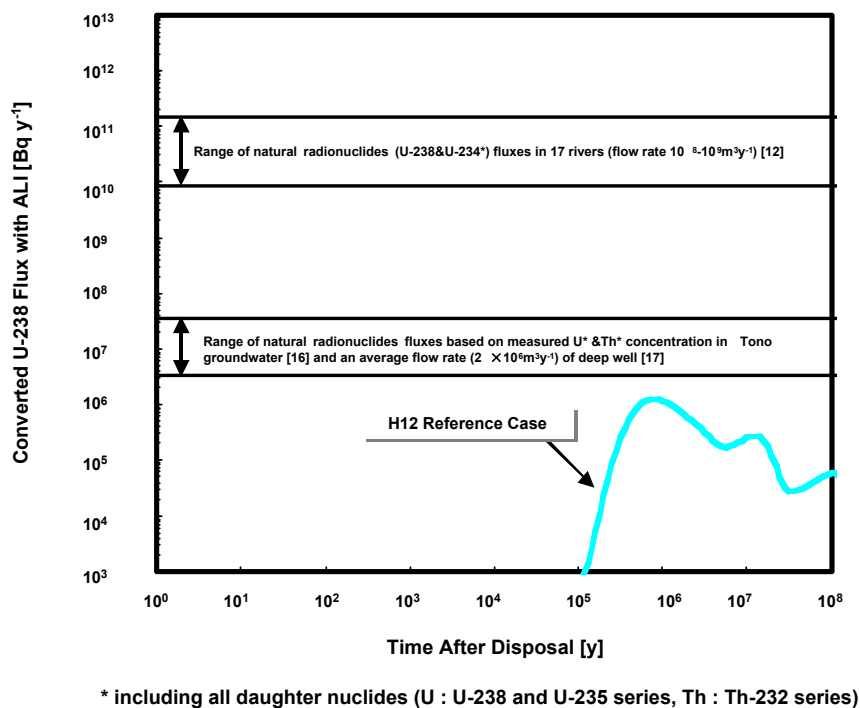


FIG. G4. Assessment of the impact from non-naturally occurring radionuclides in the repository using the converted flux of U-238 [G15].

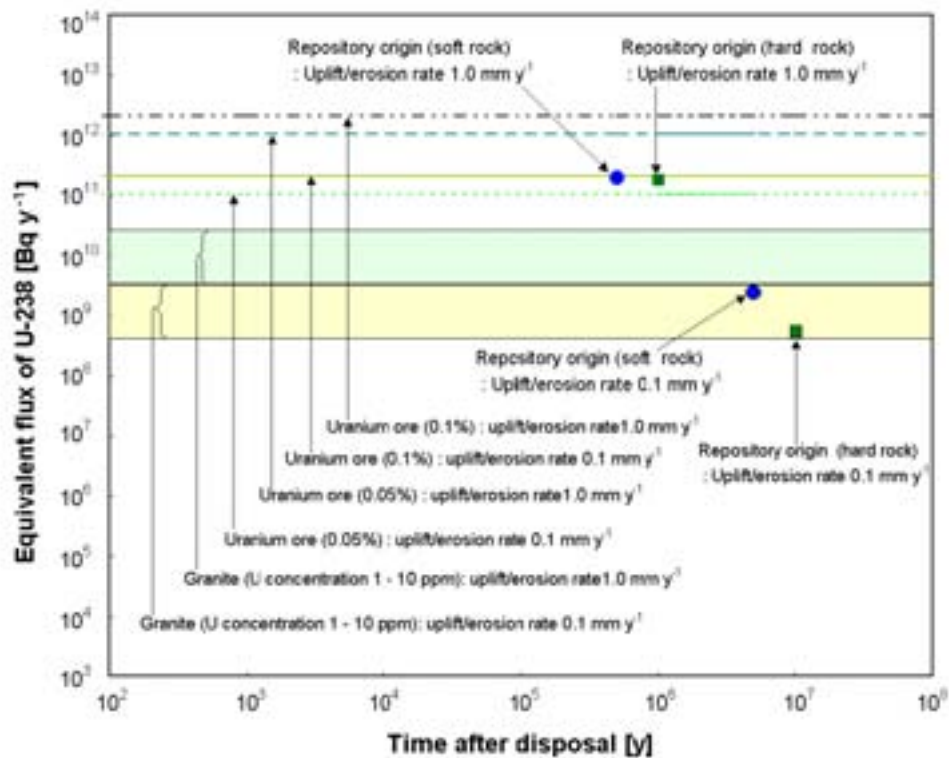


FIG. G5. Comparison of fluxes of nuclides due to the repository with fluxes of natural origin for the uplift/erosion scenario [G15].

## Conclusions

The geochemical data survey has been conducted on domestic literatures and using the Geological Literature Search System of the Geological Survey of Japan (GSJ). Chemical species data of U, Th, K, Rb, Sn, Cs and Cu are searched and summarized for the elemental concentration of water (surface and subsurface), rock (sedimentary and crystalline) and fragments (soil, stream/lake sediment and marine sediment), and those of U, Th, K, Ra and Rn for the radionuclide concentration data of the same materials. Process data such as denudation rate and river flow rate to evaluate elemental and radionuclide fluxes were also summarized based on literatures survey.

The application of safety indicators has been evaluated in the H12 performance assessment and subsequent studies. The repository derived nuclide concentrations and fluxes evaluated in the geosphere and biosphere in the H12 repository system were compared with measurements of naturally occurring radionuclides or their evaluated fluxes. This suggests that application of supplementary safety indicators could increase the reliability of long term safety by more rigorous measurements of concentrations or evaluated fluxes of naturally occurring radionuclides in the characterization phase at a specific site, which will be compared with concentrations or fluxes of relevant nuclides predicted by site specific performance assessment.

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## APPENDIX H

### SWEDEN

B. Thunholm, A. Lindén, Geological Survey of Sweden

#### Abstract

A major part of the work was focused on collecting data on geophysics as well as geochemistry and groundwater chemistry; mainly uranium (U), thorium (Th) and potassium (K). Data were interpreted resulting in maps and statistical description. The concentrations for the elements U, Th and K have the following median values:

Media	U (ppm)	Th (ppm)	K (ppm)
Overburden, bedrock, wetlands (air-borne measurements)	1.6	5.8	16200
Overburden (chemical analysis)	<5.0	<10.0	24100
Bedrock (petrophysical analysis)	3.6	11.8	33000
Bedrock (chemical analysis)	2.0	6.7	27000
Groundwater (overburden)	0.0006	-	2.2
Groundwater (crystalline bedrock)	0.0023 – 0.0073	0.00004	2.5

The data on concentrations in groundwater are in general somewhat uncertain for U. The amount of data on Th in groundwater was very limited thereby the estimate is even more uncertain. However the few analyses made indicates very low concentrations in groundwater.

Fluxes have been roughly estimated by multiplying data on concentrations with data on runoff. Flux of potassium is estimated to range between 100–400 kg/km<sup>2</sup>/year, while a very rough estimate of U flux range between 0.5–2 kg/km<sup>2</sup>/year. No relevant data on flux of Th have been found. This is partly due to problems to estimate the weathering of Th.

The Uranium archive contains data on about 2000 objects and eight of these have been selected to show natural extreme values.

#### Background

##### Nuclear programmes

Sweden has 11 nuclear power reactors (at four sites) and one material testing and research reactor. The radioactive waste originates from the nuclear power industry as well as medical use, industry, research and consumer products.

The legislation in Sweden forms the basis for a system for management of spent fuel and other radioactive waste generated within the nuclear fuel cycle. The Act on Nuclear Activities (1984:3) requires that the waste producers take the full responsibility for research and development (R&D) and funding of future disposal facilities. This task is fulfilled by the Swedish Nuclear Fuel and Waste Management Company (SKB), jointly owned by the nuclear power utilities.

SKB reports R&D programmes to the Government every third year and they shall include a comprehensive description of the measures taken to ensure a safe handling and disposal of spent fuel and nuclear waste. In 1986 SKB presented the first R&D programme. Since then five such programmes (with supplements) have been produced by SKB and reviewed by SKI (Swedish Nuclear Power Inspectorate) and SSI (Swedish Radiation Protection Authority).

At present Sweden has a central interim storage facility for spent nuclear fuel (CLAB) and a repository for low and intermediate level waste (SFR). Planned facilities are an encapsulation plant for spent fuel and repositories for spent fuel, other long-lived radioactive waste and decommissioning waste.

At the beginning of the 2000s the siting process of a repository for spent fuel became more intense. At present SKB is conducting site investigations in two municipalities. SKI and SSI are involved in the Environmental Impact Assessment (EIA) in connection with this siting. According to SKB's plan an application to construct the repository will be submitted to the Government in 2007.

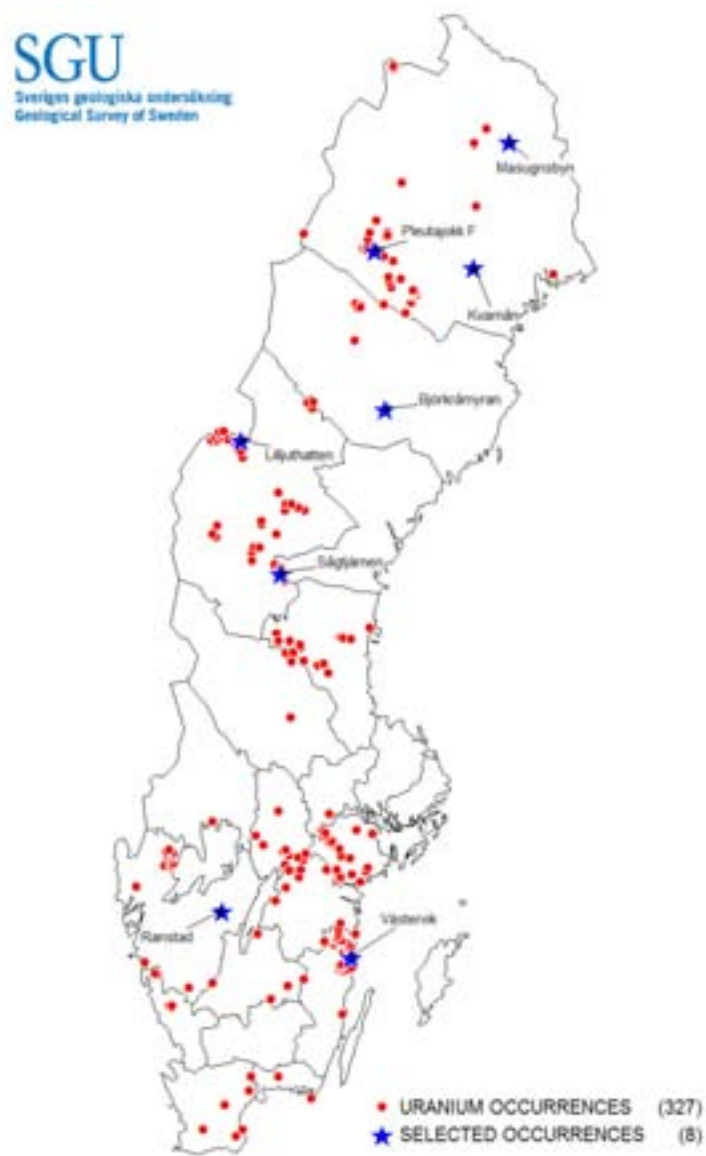
Sweden has previously been involved in several activities for development of non-dose/risk safety indicators. For example, the Swedish authorities participated in the work on the so-called Nordic Flagbook on *Disposal of high-level radioactive waste, consideration of some basic criteria* [H1]. SKI and SSI were also two of the funding organizations for the report *Natural elemental concentrations and fluxes: their use as indicators of repository safety* [H2].

### **Topography, geology and climate [H3]**

The level of the ground surface in Sweden varies between the sea level and 2111 m in the north-western parts of Sweden. Most of the large-scale groundwater flow is directed from the high-level areas in the western parts of Sweden to the Baltic Sea in the East. The bedrock in Sweden largely consists of crystalline rock types with an age of about 1.5–2 Ga. Some parts of Sweden have sedimentary rock types with an age of about 400–500 Ma. The overburden is characterized by the glaciation that ended about 10 000 years ago. About 80% of the overburden consist of glacial till whereas other parts of the bedrock are covered with peat as well as both glacial and post-glacial clay and silt sediments. The annual average ground water recharge (approximately precipitation minus evapo-transpiration) vary between about 150 mm per year in the south-eastern parts of Sweden to about 1500 mm in the north-western parts. The mean value is about 400 mm per year. The average ground temperature ranges between about 10°C in the southern parts to the freezing point in the northern parts. The geothermal gradient in the crystalline bedrock is about 1°C/100 m.

### **Study areas**

A lot of information in the former Uranium archives is of general exploration character such as geophysical, geochemical and geological surveys and as such better stored together with that type of information in the main archives of the Mineral Resources Information Office (MINKO). The Uranium archives in Sweden have a total amount of 2000 objects. Many objects have a lot of data, which could be of interest for this CRP. Eight objects have been chosen (Figure H1, Table H1) for presentation. Different amounts of data exist from airborne gamma measurements, geochemistry and drillings.



*FIG. H1. Selected objects from the uranium archives.*

TABLE H1. DATA FROM SELECTED OBJECTS FROM THE URANIUM ARCHIVES

Name	Lat/Long	Level (m)	Type	Mineral	Tonnage	Grade U (%)
Masugnsbyn	67°28'/20°57'	375	Secondary in peat	U	Unknown	0.06
Pleutajökk	66°20'/17°39'	550	Vein	Uraninite	5100	0.1
Kvarnån	65°58'/20°36'	250	Impregnation	Uraninite	1500	0.08
Björkråmyran	64°22'/17°44'	410	Cataclastic hydrothermal	U, Zr	> 1500	0.1-0.5
Liljuthatten	63°59'/13°28'	700	Impregnation and veins	Pechblende	2000	0.2
Sågtjärn	62°21'/14°56'	300	Pegmatoid	Pechblende	600	0.06
Ranstad	58°18'/13°15'	120	Sediment	Uraninite	> 300 000	0.03
Västervik	57°46'/16°37'	50	Impregnation	Different	Unknown	0.05-1.0

## Elemental and radionuclide concentration data

### Sources of data

The data on concentration of elements are stored in databases at the Geological Survey of Sweden [H4]. All data are publicly available, although there is a policy to cover the cost for usage and distribution of data.

Airborne gamma measurements of uranium, thorium and potassium have been made since the late 1940's. Digital data with spectral resolution are available from 1968. About 70 % of Sweden is covered with a spatial resolution of  $200 \times 40 \text{ m}^2$ . However, the resolution has been improved and the present value is  $200 \times 16 \text{ m}^2$ . Data from a total number of more than 40 million points are available in digital format.

Petrophysical data is based on measurements from about 6000 outcrops with a total of about 20 000 observations. Measurements have been made for uranium, thorium and potassium and other elements.

Petrochemical sampling and analyses have been made in connection with the regular mapping of the bedrock that is made by the Geological Survey of Sweden. A total amount of about 8000 observations is available, while the amount of data on uranium and thorium is limited to about 300 observations.

Geochemical analysis of uranium and other elements in glacial till has been made since 1982. Sampling is made with 1 sample per about  $7 \text{ km}^2$ . During the period 1987-1997 a low density sampling of one sample per  $40 \text{ km}^2$  was used. Data before 1995 have a detection limit of 5 ppm, which should be taken into account. After 1995 the detection limit has been 0.1 ppm. A total amount of 26 000 observations are available. High-density data covers about 40% of Sweden, whereas the low-density data covers about 50%.

Groundwater data can be divided into two groups of digital data: One study was made in 1981 where about 50 samples were collected in various types of groundwater bodies in different parts of Sweden. The objective was to find average values of uranium, radium and radon. The other group of data consists of about 2000 analyses of radium and radon, which are fairly evenly distributed over Sweden. This group of data have been collected within the countywide mapping of groundwater with some additional data from various investigations.

### Summary of data

Airborne gamma-measurements of uranium, thorium and potassium show differences in concentrations with fairly high values in some areas of Sweden. (Table H2, Figure H2). The data represents mainly the upper part of the overburden with minor influence of outcrops.

The petrophysical measurement values should be fairly representative for the Swedish Precambrian bedrock (Table H3). The values are somewhat higher than the corresponding airborne data that represents mainly the upper part of the overburden.

Another source of data, a database on petrochemistry (i.e. chemical analyses of the bedrock) gives similar results as the petrophysical database for the minimum, median and maximum values (Table H4). This database has about 300 observations for uranium and thorium whereas about 5000 observations are included for potassium. The mean value and standard deviation are fairly high owing to a few samples with extremely high values.

TABLE H2. STATISTICAL DATA FOR AIRBORNE GAMMA MEASUREMENTS ON URANIUM (U), THORIUM (TH) AND POTASSIUM (K)

	<i>U (ppm)</i>	<i>Th (ppm)</i>	<i>K (ppm)</i>
Mean	1.79	6.40	15 200
Standard Deviaion	1.48	4.96	9300
Minimum	0.00	0.00	0
Median	1.61	5.81	16 200
Maximum	50.00	199.30	69 500

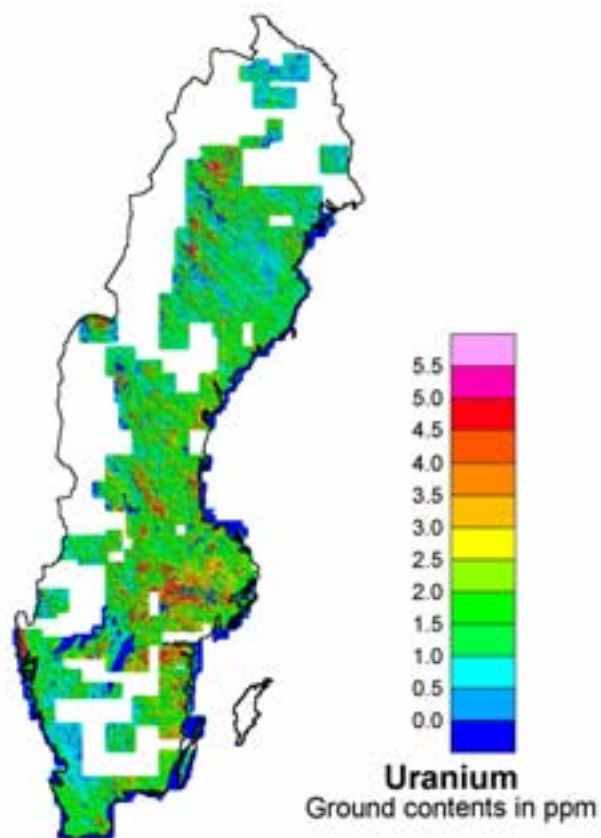
TABLE H3. STATISTICAL DATA FOR PETROPHYSICAL MEASUREMENTS ON URANIUM (U), THORIUM (TH) AND POTASSIUM (K)

	<i>U (ppm)</i>	<i>Th (ppm)</i>	<i>K( ppm)</i>
Mean	5.06	16.10	32 200
Standard Deviaion	9.03	21.43	13 400
Minimum	0.00	0.00	0
Median	3.56	11.82	33 000
Maximum	460.40	596.41	79 000

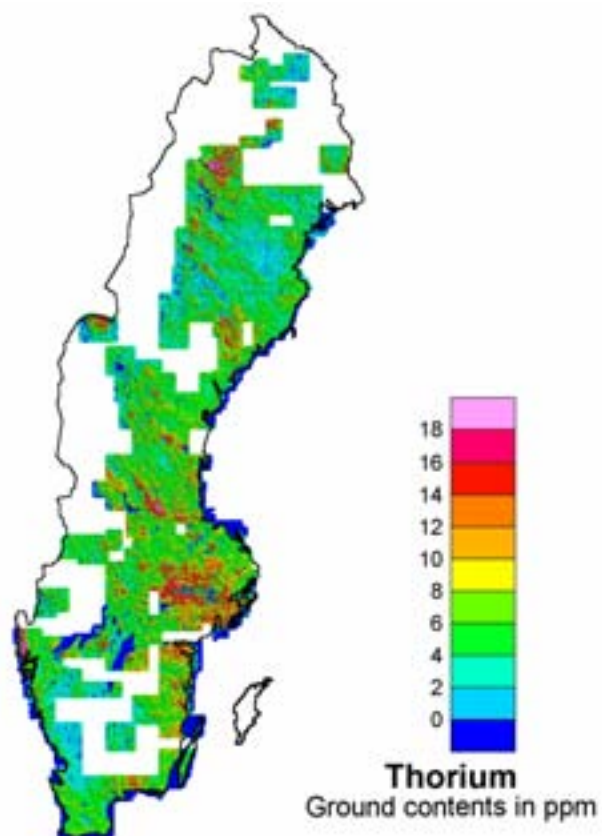
TABLE H4. STATISTICAL DATA FOR PETROCHEMICAL MEASUREMENTS ON URANIUM (U), THORIUM (TH) AND POTASSIUM (K)

	<i>U (ppm)</i>	<i>Th (ppm)</i>	<i>K( ppm)</i>
Mean	129.6	24.1	27 000
Standard Deviaion	1384.9	215.0	19 000
Minimum	0.0	0.0	0
Median	2.0	6.7	27 000
Maximum	18 807.0	3603.1	120 000

a)



b)



c)

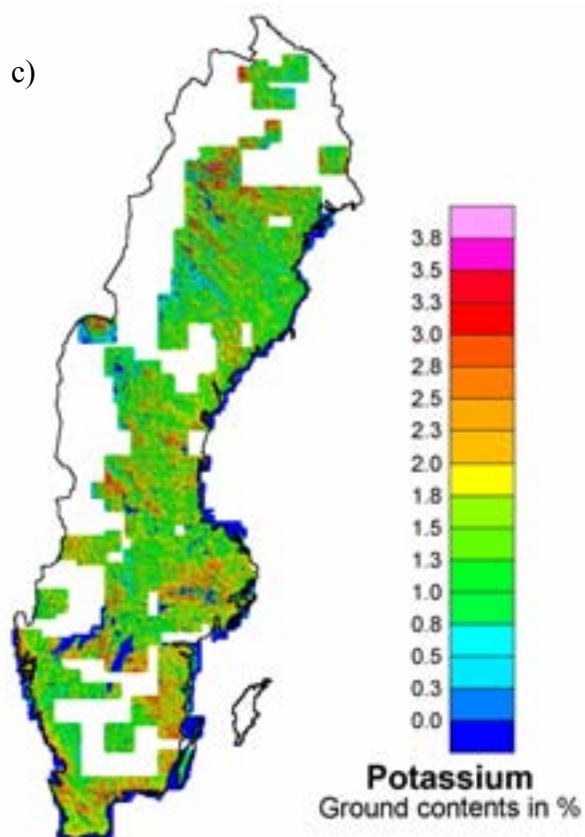
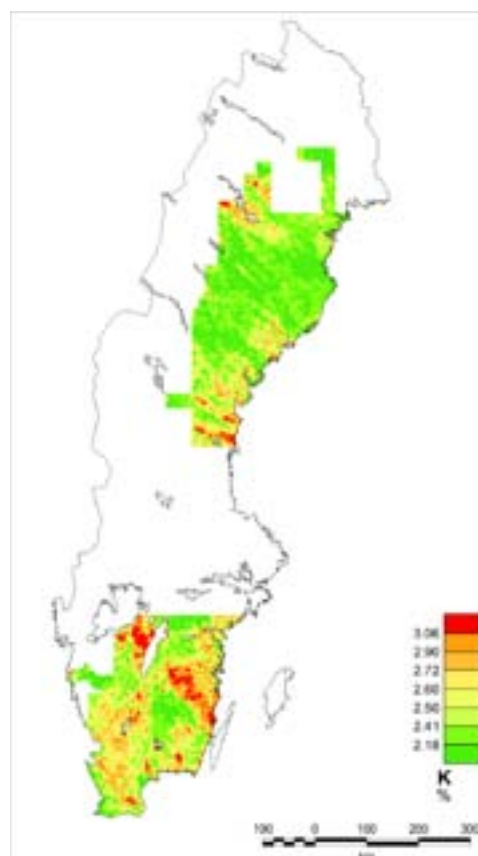


FIG. H2. Ground contents of uranium (a), thorium (b) and potassium (c) based on airborne measurements.

a)



b)



*FIG. H3. Geochemical contents in the C-horizon of U(ppm) and K(%).*

Geochemical characterization of the C-horizon (Table H5-8, Figure H3) has been made with different analysis methods. The quality of bulk analysis (made by XRF = X-Ray Fluorescence) is considered to be good for potassium while not quite satisfactory for uranium and thorium (Table H5 – 6). For those elements the analysis made by ICP-MS (Inductively Coupled Plasma Mass Spectrometry) after acid extraction is far better but does not provide the total content. However, there is a strong correlation between the values after extraction and the total content, which means that the total content may be estimated from the values of the extracted samples.

The data in Table H7 – 8 could be compared with airborne data (Table H2) which largely have the same origin as the geochemical analysis (i.e. data from the upper part of the overburden). A countrywide investigation of uranium and radium in groundwater shows that uranium ranges from 0.1 to 43.5 ppb and that the radium-226 activity ranges from 0.5 to 1900 mBq/l with generally higher values in the Precambrian bedrock. This investigation was based on 42 samples in representative Swedish aquifers (Table H9). In addition there are samples from 492 drilled wells with analyses of radium (Table H10, Figure H4). Data on radon are more extensive with about 2500 measurements in springs, dug wells and drilled wells (Table H11, Figure H4).



TABLE H5. COMPARISON BETWEEN METHODS FOR ANALYSIS OF URANIUM (U, PPM)

<i>Percentile</i>	<i>Total content (XRF)</i>	<i>Acid extraction (ICP-MS)</i>
0	<5	0.20
10	<5	0.89
20	<5	1.05
25	<5	1.11
30	<5	1.17
40	<5	1.30
50	<5	1.44
60	<5	1.62
70	<5	1.87
75	5	2.04
80	5	2.25
90	6	2.97
95	7	3.92
100	44	31.70

TABLE H6. COMPARISON BETWEEN METHODS FOR ANALYSIS OF THORIUM (TH, PPM)

<i>Percentile</i>	<i>Total content (XRF)</i>	<i>Acid extraction (ICP-MS)</i>
0	<10	1.1
10	<10	4.3
20	<10	5.0
25	<10	5.4
30	<10	5.7
40	<10	6.3
50	<10	7.0
60	<10	7.7
70	11	8.5
75	12	9.0
80	12	9.6
90	16	11.8
95	19	14.0
100	57	49.4

TABLE H7. BULK ANALYSIS ON URANIUM (U), THORIUM (TH) AND POTASSIUM (K). XRF

	<i>U (ppm)</i>	<i>Th (ppm)</i>	<i>K (ppm)</i>
Minimum	<5	<10	3100
P10	<5	<10	19800
Q1	<5	<10	21800
Median	<5	<10	24100
Q3	5	12	27200
P90	6	16	29000
Maximum	44	57	56200

TABLE H8. CONCENTRATIONS AFTER ACID EXTRACTION FOR URANIUM (U) AND THORIUM (TH). ICP-MS

	<i>U (ppm)</i>	<i>Th (ppm)</i>
Minimum	0.20	1.1
P10	0.89	4.3
Q1	1.11	5.4
Median	1.44	7.0
Q3	2.04	9.0
P90	2.97	11.8
Maximum	31.70	49.4

TABLE H9. MEDIAN VALUES OF URANIUM (U, PPB) AND RADIUM-226 (RA, MBQ/L) IN DIFFERENT ROCK TYPE AQUIFERS. BASED ON 42 SAMPLES

	<i>Aquifer</i>	<i>U (ppb)</i>	<i>Ra-226(mBq/l)</i>
Sedimentary bedrock	Quaternary deposits	1.0	6.0
	Bedrock	0.3	2.5
Precambrian bedrock	Quaternary deposits	0.6	3.5
	Bedrock	2.3	97.0

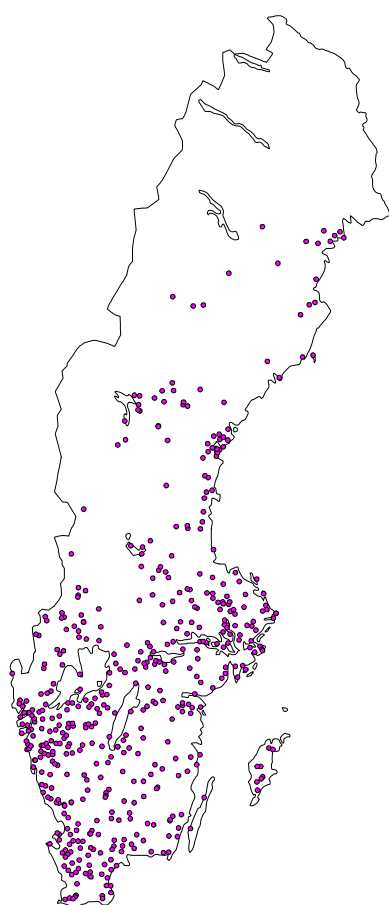
TABLE H10. STATISTICAL DATA FOR RADIUM –226 (RA, MBQ/L) IN DRILLED WELLS. BASED ON 492 SAMPLES

	<i>Drilled wells</i>
Minimum	0.2
P10	2.0
Q1	5.0
Median	12.0
Q3	35.0
P90	117.0
Maximum	2455.0

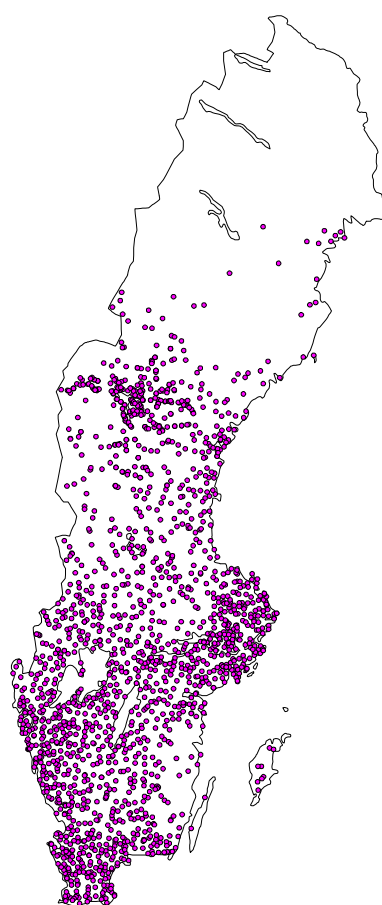
TABLE H11. STATISTICAL DATA FOR RADON (RN, BQ/L) IN SPRINGS, DUG WELLS AND DRILLED WELLS. BASED ON 2500 SAMPLES

	<i>Springs</i>	<i>Dug wells</i>	<i>Drilled wells</i>
Minimum	1	1	0
p10	10	4	10
Q1	20	8	30
Median	39	20	85
Q3	85	49	220
P90	153	96	565
Maximum	845	947	9289

a)



b)



*FIG. H4. Locations of wells with analysis of radium (a) and radon (b) [H5] .*

Another investigation of uranium in groundwater in 269 drilled wells in Uppsala municipality showed a median of 12 ppb with an approximate range between 0.2 and 246 ppb. These values could be considered as fairly typical for uranium in groundwater in Swedish Precambrian bedrock. This could be compared with earlier estimations of uranium content of about 3 ppb in springs, which should be similar to uranium content of the groundwater in the loose deposits. Uranium content in surface waters has been estimated to be lower than 1 ppb.

A small number of analyses on thorium concentrations in groundwater have been made. The results indicate low concentrations, <0.1 ppb. This is uncertain, although studies in various reports indicate low concentrations of similar magnitude.

### **Elemental and radionuclide flux data**

#### **Sources of data**

The estimations of fluxes are based on data from environmental monitoring funded by the Swedish Environmental Protection Agency. Data are publicly available. Data from monitoring in rivers were used which were given as fluxes for various elements, although data on U and Th were not found. In addition data on groundwater runoff together with data on

concentrations in groundwater were used to estimate fluxes. These were estimated by multiplying concentrations with runoff. Data in both surface water and groundwater are based on the dissolved part, which is the dominating part in both cases.

### **Summary of data**

Data on potassium concentration in rivers indicate fluxes in the magnitude of 200–500 kg/km<sup>2</sup>/year. The flux of potassium by groundwater is estimated to range between 100 and 400 kg/km<sup>2</sup>/year. This estimate is based on measured concentrations and runoff together with studies on weathering rates. Values are generally higher in the southwestern parts of Sweden mainly due to higher values on runoff. It should be noted that potassium is an important plant nutrient, which means that the flux is influenced by land use and vegetation.

A rough estimate of uranium fluxes by using data on concentration together with data on groundwater runoff gives values of 0.2–2 kg/km<sup>2</sup>/year. This is based on typical values on concentration of uranium in groundwater (Table H9) and data on runoff. Fluxes of thorium have not been estimated due to a very limited amount of data, which indicates very low values, below 0.1 ppb. In general attempts to estimate fluxes proved to be difficult for various reasons. One difficulty is uncertainties owing to processes that are not in a steady state condition. This is largely due to the glaciation that ended about 10 000 years ago which means that the weathering processes are in an early stage compared with other parts of the world with no glaciation. Other difficulties are associated with the spatial scale. The fluxes of elements within the unsaturated zone could differ considerably from the corresponding catchment area. In addition, fluxes are supposed to vary between small catchments whereas a large river basin provides an average value for all catchments within the river basin as well as the contribution from deep flow paths within the basin.

### **Interpretation and application of the concentration and flux data**

The data presented within this CRP, and the overall project, are intended to be used for reference purposes and to support the development of indicators.

### **Conclusions**

The existing databases at SGU [H4] have a lot of data on concentrations that could be used for different purposes. Data on concentration of U, Th and K are extensive and may be used as complementary indicators to dose and risk. However, the application as indicators need be tested in future studies. Data on fluxes seem to be more difficult to handle. One reason is limitations in the amount of data but also difficulties to interpret flux data. This is largely related to difficulties to deal with spatial scale and to estimate the non-steady state condition of the processes. Additional difficulties could be found in the approach for comparing natural fluxes with possible fluxes from a leaching repository.

### **Additional information sources**

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OLSSON, M., ROSÉN, K., MELKERUD, P.-A., Regional modelling of base cation losses from Swedish forest soils due to whole-tree harvesting. Applied Geochemistry, Suppl. Issue No 2. Pp (1993) 189-194, (1993).

RAAB, B., VEDIN, H. (Eds.), “National Atlas of Sweden – Climate, Lakes and Rivers”, SNA Publishing, Stockholm (1995).

SWEDISH ENVIRONMENTAL PROTECTION AGENCY, <http://www.naturvardsverket.se/> for information on environmental monitoring.

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## **APPENDIX I**

### **UNITED KINGDOM**

S. Norris, P. Degnan, UK Nirex Ltd

#### **Abstract**

The UK has a wide range of rock types, including igneous, metamorphic and sedimentary varieties, and this has a direct affect on the topography of the country. This study examined a number of different geological regions including the granites of Cornwall, the Chalk aquifer of the London Basin, the thermal springs of Bath, and the Oxford Clay. These were chosen because they are geologically diverse and, therefore, may provide some estimation of the potential variation in natural fluxes across the country. Geological and geochemical data for the study were drawn from numerous sources, notably from the databases held by the British Geological Survey. For each of the study areas, information on the rates of processes (such as erosion and groundwater flow) were combined with information on the concentrations of elements and radionuclides in different materials (such as rocks, soils and riverwaters) to establish the elemental and radioactivity fluxes associated with the different processes. These flux data provide the raw material for the natural safety indicators methodology in that they potentially can be compared with the abundances and distributions of repository derived chemical species calculated in a performance assessment and, thus, can be used to assess the hazard posed by them. To test this approach, comparisons were made between the fluxes from some of the study sites (natural source) and calculated fluxes from a repository (estimated in Nirex's Generic Performance Assessment). In general, it was shown that the repository radioactivity fluxes were small in comparison with the natural radioactivity fluxes and, on this basis, it may be argued that the repository releases would not significantly alter the natural radioactive environment. It would not, however, be sensible to conclude on the basis of this comparison that the potential radiological hazard associated with the repository releases are proportionately small because some of the dominant nuclides in the repository releases have higher radiotoxicities than those in natural groundwater discharge. These trial calculations illustrate that appropriate concentrations and fluxes can be quantified for natural systems which are suitable for comparison with performance assessment predictions of repository releases, and that safety comparisons can thus be made relatively quickly and without recourse to complex mathematics. Such comparisons have potential use in both a formal, quantitative performance assessment and in less formal, qualitative demonstrations of safety that could be provided to non technical audiences in a wider safety case.

#### **Background**

##### **Nuclear programmes**

The United Kingdom currently operates 17 Magnox Reactors, 14 Advanced Gas Cooled Reactors (AGR) and one PWR as part of its nuclear power programme. Some of the older Magnox and AGRs are coming to the end of their design lives, and a few have already been taken out of service and are awaiting decommissioning. No new reactors are planned, although the UK energy policy is under review. In addition, as part of the UK nuclear power programme, a number of large fuel reprocessing and experimental facilities are operational. All of these plants and sites generate operational radioactive wastes and, in the future, large amounts of decommissioning wastes will also arise.

At present, there is one operational repository for low-level radioactive wastes, at Drigg near Sellafield, which is of a surface, engineered vault design. Plans to develop a deep geological repository for intermediate level radioactive wastes near Sellafield were shelved in 1997, when the Government rejected UK Nirex's planning application to develop an underground rock characterization facility at the site. All intermediate and high level radioactive wastes are currently in storage pending Government decision on their long term management.

As part of UK Nirex's programme to develop a concept for the deep geological disposal of intermediate level radioactive wastes, they undertook and published a number of performance assessments, the latest being the Generic Performance Assessment [11]. This was a risk-based assessment, although intended future iterations of this assessment are expected also to include the use of natural safety indicators.

### **Topography, geology and climate**

The UK has a wide range of different rock types, including igneous, metamorphic and sedimentary varieties. A simple list of the most common rock types to be found exposed at the ground surface in the UK would include granite, gabbro, chalk, limestone, sandstone, mudrock, schist and gneiss. The distribution of the major rock types in the UK is given in the simplified geological map in Figure 11. The compositional variations between the main rock types in the UK are quite wide, especially for the major elements (those which are an essential component of the major rock forming minerals, such as Si, K, Ca, Na, Fe etc.) because the mineralogical make-up of these rocks varies considerably from rock type to rock type. The abundances of trace elements (those which are not essential and occur in very low concentrations) in the different rock types are somewhat less variable with most trace elements having a general abundance in the few parts per million ( $1 \text{ ppm} \equiv 1 \text{ mg/kg}$ ) range.

The geology has a direct affect on the topography of the country. The majority of the southern parts of England are characterized by low lying sedimentary sequences of chalk, sandstones and clay rocks which provide flat lands and low hills. The northern parts of England, much of Scotland and Wales contain harder, crystalline rocks which provide more mountainous terrain.

Most of the UK was glaciated around 20 000 years ago and this has resulted in erosional features in the northern part of the islands (glacial valleys in the mountains) and depositional features in the south, such as glacial tills and moraines. Today, the climate is temperate. Annual rainfall varies from around 600 mm/yr in the southern parts of England to around 2000 mm/yr in the mountains of Scotland. Snow lies in the mountains of Scotland during the winter but there are no permanent ice caps or areas of permafrost.





FIG. 11. Simplified geological map of the UK.

## Study areas

The study areas examined in this project were all from southern England and were the granites of Cornwall, the Chalk aquifer of the London Basin, the thermal springs of Bath, and the Oxford Clay. These locations were partly chosen because they are geologically diverse and, therefore, may provide some estimation of the potential variation in natural fluxes across the country. The Cornish granites are representative of fractured, crystalline rocks; the Chalk aquifer of the London Basin represents a porous, permeable sedimentary system bounded by less permeable clay-rich horizons; the springs at Bath represent an active thermal system; and the Oxford Clay represent a low permeability, diffusion controlled sedimentary system.

## **Elemental and radionuclide concentration data**

### **Sources of data**

In the UK, considerable amounts of both geochemical data are acquired by a broad range of organizations and much, but not all, is published or otherwise made publicly available.

The principal source of geochemical data is the British Geological Survey (BGS) which is the national organization responsible for undertaking basic earth science studies in the UK. Through various mapping and surveying programmes at national, regional and local scales, they acquire and make available information on a number of geoscientific topics. Of particular note, the BGS is undertaking a comprehensive survey of the surface occurrence and distribution of a wide range of elements for a variety of economic and environmental applications: the Geochemical Baseline Survey of the Environment (G-BASE). This survey is associated with the United Nations sponsored International Geological Correlation Programme - number 360 'Global Geochemical Baselines'. The G-BASE survey includes information on the analysis of up to 35 elements from in excess of 100 000 stream sediment, panned concentrate, soil and surface water samples from England, Scotland and Wales. The G-BASE datasets provide some of the most detailed geochemical data of relevance to the determination of natural safety indicators. Since the elemental compositions in the soils and sediments reflect (but are not identical to) compositions in the underlying rock, they are valuable in the assessment of elemental fluxes from the basement rock to the surface environment. However, the G-BASE survey is an ongoing programme and data are not yet available for the entire country. To date, sampling and analysis is complete for Scotland, Wales and northern England; is ongoing for Midlands and East Anglia; and a sampling programme is planned for Southern England.

Other sources of geochemical data include: the UK Soil Surveys, which provide information on the characteristics of soils and sediments; the National Radiological Protection Board (NRPB), which provides information on distributions of natural and man-made sources of radiation; the Institute of Hydrology, which provides information on surface and groundwaters; and universities and research organizations.

### **Summary of data**

The availability of data for soils and sediments, and some surface waters is very good. Most notably, this is provided by the G-BASE datasets which include data on (for soils and sediments): Ag, As, B, Ba, Be, Bi, CaO, Cd, Co, Cr, Cu, Fe<sub>2</sub>O<sub>3</sub>, Ga, K<sub>2</sub>O, La, Li, MgO, Mn, Mo, Nb, Ni, P, Pb, Rb, Sb, Sn, Sr, TiO<sub>2</sub>, U, V, Y, Zn, Zr, pH and (for surface waters) Al, B, Ba, Ca, Cd, F, Fe, K, La, Li, Mg, Mn, Mo, Na, Ni, P, Pb, SO<sub>4</sub>, Si, Sr, U, Zn, pH, electrical conductivity and alkalinity.

Information on the geochemistry of rocks is harder to obtain and there is no national database of information. Data have to be acquired from individual studies and, therefore, the availability is site dependent. Estimated average elemental abundances in the UK rocks are provided in Table II.

TABLE II. ESTIMATED AVERAGE ELEMENTAL ABUNDANCES IN THE UK ROCKS

Element	Upper crust (mg/kg)	Granite (mg/kg)	Gabbro (mg/kg)	Gneiss (mg/kg)	Shale (mg/kg)	Limestone (mg/kg)	Sandstone (mg/kg)
Cu	19	10	100	n/a	50	4	n/a
Fe	n/a	39000	122000	n/a	67000	5000	n/a
K	28650	40000	10000	n/a	32000	6000	n/a
Pb	n/a	20	5	n/a	20	8	n/a
Rb	111	150	30	n/a	140	50	46
Sn	4	3	1	2.2	4	0.5	0.5
Sr	n/a	285	465	n/a	300	100	n/a
Th	10.5	27	2.8	16	12	1.7	3.2
U	2.7	4.8	0.6	3.2	3.5	2.2	0.6
Zn	62	75	100	95	100	25	30
Zr	n/a	180	150	n/a	160	20	n/a

Information on radioactivity is less available and there is no national database of this information. Estimated averages (from individual studies on a range of rock types) are in the range of 1 to  $10^3$  Bq/kg with higher values from mineralized rocks. In groundwater, the dominant activity source is from  $^{222}\text{Rn}$ , with typical values being in the region of 1 to  $10^2$  Bq/l, with higher values possible in mineralized regions.

### Elemental and radionuclide flux data

#### Sources of data

Information on fluxes is much harder to come across than for elemental abundances, and there is no national database of this information, with the exception of river flow rates. For rivers, the Institute of Hydrology operates a comprehensive monitoring system for both river flow and water quality using a network of over 1400 gauging stations. This relatively dense network of monitoring stations is a necessary response to the heterogeneity of the UK in terms of its climate, geology, land use and patterns of water utilisation. Monitoring by the IH is supplemented by flow measurements undertaken by the Environment Agency in England and Wales, and by the Scottish Environment Protection Agency, and the recently formed Rivers Agency in Northern Ireland.

This river and aquifer data, together with information from other sources, is held in the National Water Archive, which is operated by the Institute of Hydrology. The archive contains a broad range of hydrological and related data from the catchment scale (e.g. detailed climatological and hydrological data for a network of representative catchments) to the national scale (flood event data) and includes some international coverage. Included in the NWA is information relevant to the determination of natural safety indicators associated with UK rivers including river water fluxes, suspended and dissolved sediment yields for catchments, and basic (major element) analyses for river water quality.

Other relevant process rate information, such as groundwater flow and erosion comes from individual study areas, and there are no national maps or databases of these information.

## Interpretation and application of the concentration and flux data

In this study, geochemical flux calculations are performed for the four sites listed in the Introduction, using information available from site specific studies and other relevant data. The results of the study on the Cornish granites are described below, together with an explanation of the approach. The results of the studies on the other three locations are summarized for comparison with the Cornish granites.

### Cornish granites

The Cornish granites form part of a large (~ 200 km long) igneous intrusion which intruded into Devonian sediments around 290 million years ago. The granite contains a high concentration of radioelements (U, Th, K) which generate the largest crustal geothermal gradient in the UK (35°C/km) and a surface heat flow (120 mW/m<sup>2</sup>) which is around double the UK average [12]. As a consequence of this heat flow, the potential to exploit Hot Dry Rock (HDR) geothermal energy from the Cornish granites has been investigated over a 15 year period (1980 - 1995) at the Rosemanowas Quarry in the Carnmenellis Granite, which is an exposed part of the Cornubian Batholith. The HDR project at Rosemanowas involved considerable hydrogeological and hydrochemical research activities both on the site and regional scales, and so provides a substantial amount of information relevant to the determination of natural safety indicators for a fractured, crystalline rock environment. The availability of this information makes the Cornish granites, and the Carnmenellis Granite in particular, a sensible choice for this study.

The enriched abundances of radioelements in the granite cause a high local heat flow and are responsible for the present-day convection of thermal waters with meteoric origin through fractures in the granite. On the basis of homogenous distribution, the total masses of radioelements in the granite and the activity of some specific nuclides have been calculated, and are given in Table I2, which also gives the calculated activities associated with the U and Th decay chains, considering only the longer-lived daughters and assuming secular equilibrium in the chains.

TABLE I2. MASSES OF RADIOELEMENTS AND ACTIVITIES IN THE CARMENELLIS GRANITE

Species/activity	Average concentration (mg/kg)	Mass in total granite (kg)	Activity (Bq/kg)
U in granite	13.4	$2.9 \times 10^{10}$	
Th in granite	9.8	$2.2 \times 10^{10}$	
K in granite	3.9 %	$8.6 \times 10^{13}$	
<sup>238</sup> U activity			160
<sup>235</sup> U activity			6.9
<sup>232</sup> Th activity			40
<sup>40</sup> K activity			960
U chain activity			1580
Th chain activity			161

The radioelements held in the granite in solid form can be mobilized (to generate fluxes) by denudation of the rock surface and by dissolution involving ground and surface waters. These processes are important because they can increase the ‘availability’ of the radioelements to biota at the land surface and, thus, the radiological hazard associated with them. Denudation of the granite occurs directly by physical and chemical processes at the surface because the granite is largely exposed with only a thin, discontinuous soil cover in places. Taking a typical erosion rate of 5 m/Ma, an exposed granite surface area of 80 km<sup>2</sup> and a rock density of 2750 kg/m<sup>3</sup>, this means that 1.1×10<sup>6</sup> kg of rock are eroded each year from the Carnmenellis Granite. Assuming the erosion rate is constant over the granite surface, this is equivalent to a specific mass (rock) erosion rate of 1.4×10<sup>4</sup> kg/km<sup>2</sup>/yr. In reality, it is more probable that the erosion rate would vary over the granite, partly in response to the topography and the surface water runoff and infiltration patterns. Nonetheless, on this basis, and assuming that elements are released by erosion in proportion to their abundance in the granite, the total masses of elements and the activity of some nuclides associated with this erosion have been calculated for the granite surface as a whole and per km<sup>2</sup>, and are given in Table I3, together with the calculated activities due to erosion associated with the U and Th decay chains.

TABLE I3. ELEMENTAL MASS AND ACTIVITY FLUXES DUE TO EROSION OF THE CARNMENELLIS GRANITE

Species/activity	Mass flux for total granite (kg/yr)	Specific mass flux (kg/km <sup>2</sup> /yr)	Activity flux for total granite (Bq/yr)	Specific activity flux (Bq/km <sup>2</sup> /yr)
U	14.7	0.18		
Th	10.8	0.13		
K	4.3×10 <sup>4</sup>	536		
Cs	49.5	0.62		
Sn	19.8	0.24		
<sup>238</sup> U activity			1.8×10 <sup>8</sup>	2.3×10 <sup>6</sup>
<sup>235</sup> U activity			7.5×10 <sup>6</sup>	9.4×10 <sup>4</sup>
<sup>232</sup> Th activity			4.4×10 <sup>7</sup>	5.5×10 <sup>5</sup>
<sup>40</sup> K activity			1.1×10 <sup>9</sup>	1.4×10 <sup>7</sup>
U chain activity			1.7×10 <sup>9</sup>	2.1×10 <sup>7</sup>
Th chain activity			1.8×10 <sup>8</sup>	2.3×10 <sup>6</sup>

This table shows that the largest elemental mass flux is for K by a few orders of magnitude. This is a direct consequence of the high K content (a major element) in the granite relative to the other elements which occur in trace amounts. The largest nuclide-specific activity flux associated with erosion is also associated with K (<sup>40</sup>K) but its dominance over <sup>238</sup>U is reduced because of the small isotopic abundance of <sup>40</sup>K. The activity from <sup>238</sup>U is roughly 1 to 2 orders of magnitude larger than that due to <sup>235</sup>U for a similar reason.

Elements are also lost from the granite through rock-water interaction and groundwater discharge. It has been estimated that the volume of groundwater discharged from the granite annually is 1.06×10<sup>10</sup> l/yr. The measured elemental abundances in the groundwater are given in Table I4, together with the calculated elemental and activity fluxes from the granite, assuming this discharge rate.

TABLE I4. ELEMENTAL ABUNDANCES, PLUS FLUXES OF ELEMENTS AND ACTIVITY DUE TO GROUNDWATER DISCHARGE FROM THE CARMENELLIS GRANITE

Species/activity	Concentration (mg/l)	Mass flux from groundwater discharge (kg/yr)	Activity flux from groundwater discharge (Bq/yr)
U	0.00008	0.81	
K	2.8	$2.0 \times 10^5$	
Rb	< 0.02	2790	
I	0.028	357	
Cs	0.031	4532	
Cu	< 0.0004	24	
$^{238}\text{U}$ activity			$1.0 \times 10^7$
$^{235}\text{U}$ activity			$4.1 \times 10^5$
$^{40}\text{K}$ activity			$4.9 \times 10^9$
$^{87}\text{Rb}$ activity			$2.5 \times 10^8$
U chain activity			$9.6 \times 10^7$

This table shows that the largest elemental mass fluxes are for K, Rb and Cs. This is a direct consequence of their high contents in the groundwater, relative to the other elements which occur in trace amounts (higher elemental solubilities). The largest activity fluxes are also associated with K ( $^{40}\text{K}$ ) and Rb ( $^{87}\text{Rb}$ ). The activity from  $^{238}\text{U}$  is roughly one to two orders of magnitude larger than that due to  $^{235}\text{U}$  for a similar reason.

Additional studies have examined the  $^{222}\text{Rn}$  contents of discharging groundwaters in the Carnmenellis Granite and surrounding rocks and shown them to be locally very high. The comprehensive study by Gregory and Durrance [I3] showed that discharging waters on the granite contained  $^{222}\text{Rn}$  activities in the range 0.6–453 Bq/l, with a mean value of 24.9 Bq/l. Compared to groundwater activities given in Table I4, this mean value is some four orders of magnitude greater than that for  $^{238}\text{U}$  and one order of magnitude larger than that for  $^{40}\text{K}$  and  $^{87}\text{Rb}$ . Applying the average  $^{222}\text{Rn}$  activity concentration for the granitic waters to the total groundwater discharge volume from the granite gives a  $^{222}\text{Rn}$  flux of  $2.6 \times 10^{11}$  Bq/yr, which substantially dominates over all the other radionuclide fluxes.

By pulling together the information on elemental abundances and fluxes for the Carnmenellis Granite, a simple model of the elemental behaviour in the system can be compiled, which is as follows. The U concentration in the granite is above average, leading to a very large total U mass in mineral form and a high  $^{238}\text{U}$  activity. Erosion leads to several kilograms of U released in solid form each year. The U concentration in the groundwater is low because U is poorly soluble in the reducing groundwater. This, in turn, leads to small annual U mass and  $^{238}\text{U}$  activity discharges in the groundwater, even though quite large volumes of water flow through the granite annually. However, Rn is not in equilibrium with U and the  $^{222}\text{Rn}$  activity discharge in groundwater is the largest activity flux in the system. The model for U and Rn fluxes is indicated in Figure I2.



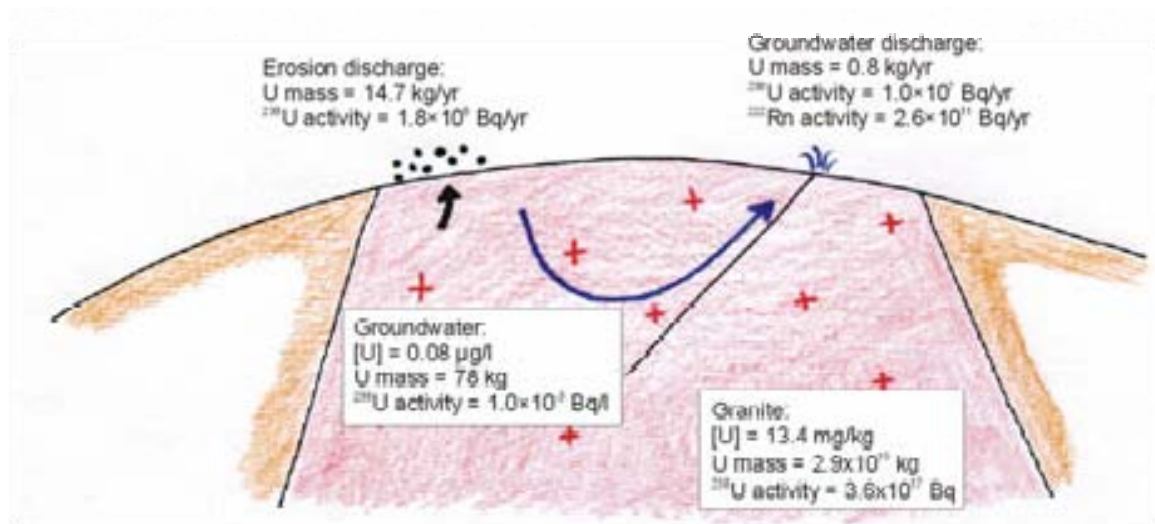


FIG. 12. Model of U and Rn distribution and fluxes in the Carnmenellis Granite.

### London Basin Chalk Aquifer

The Chalk Aquifer of the London Basin and the Berkshire Basin is one of the most important groundwater sources for public water supply in the UK. The Chalk Aquifer and associated lithologies have been studied in detail because of their importance as a supply of drinking water. In particular, the baseline elemental geochemistry of the Chalk aquifer has been investigated [14] to provide the basis for groundwater quality management. The following text summarized the results of the flux calculations for U and Rn.

The U concentration in the Chalk is low but, given that the volume of the Chalk is large, there is a large total U mass in the rock and a high total  $^{238}\text{U}$  activity. Erosion leads to several kilograms of U released in solid form each year. The U concentration in the groundwater is moderate. This, in turn, leads to large annual U mass and  $^{238}\text{U}$  activity fluxes in the groundwater discharging naturally to the Thames river system and via pumped extraction. The U mass flux due to groundwater discharge and due to Chalk erosion are of similar magnitudes. The Rn is not in equilibrium with U and the  $^{222}\text{Rn}$  activity from natural discharge in groundwater is the largest activity flux in the system. The model for U and Rn fluxes is indicated in Figure 13.

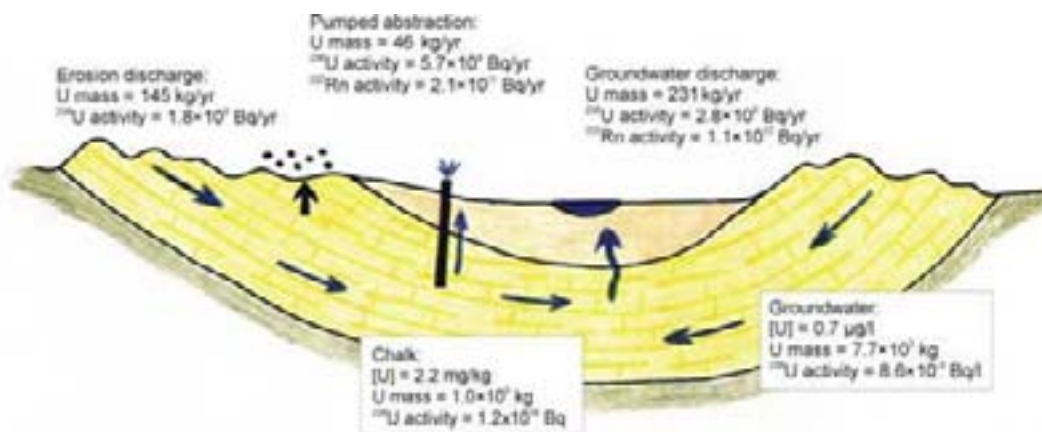


FIG. 13. Model of the U and Rn distribution and fluxes in the Western London Chalk.

## Bath hot springs

The hot springs at Bath are the surface expression of the most active natural hydrothermal system in the UK. These hot springs are known to have been exploited and made famous by the Romans who constructed the original thermae two thousand years ago, during the reign of Emperor Claudius. There are three separate, large springs at Bath situated on the alluvial plain of the River Avon. The springs are artesian and the thermal waters flow freely at a rate of  $1.25 \times 10^6$  l/day, with a temperature at the spring head of between 41 and 47°C. The following text summarizes the results of the flux calculations for U and Rn.

The U concentration in the Carboniferous Limestone is relatively low, but distributed throughout the entire mass of limestone involved in the flow system, the total U mass and total  $^{238}\text{U}$  activity are significant. Infiltrating groundwaters are oxidising and dissolve U from the rock. Continued rock-water interactions along the flow path drive the groundwater to more reducing conditions causing most of the dissolved U to precipitate at depth and the remaining U concentration in the reducing groundwater to be low. This leads to small annual U mass and  $^{238}\text{U}$  activity discharges in the groundwater. The rate of U discharge in the groundwater is less than the rate of U precipitation from the groundwater at depth. However, Rn is not in equilibrium with the U and the  $^{222}\text{Rn}$  activity discharge in groundwater is the largest activity flux in the system. The model for U and Rn fluxes is indicated in Figure I4.

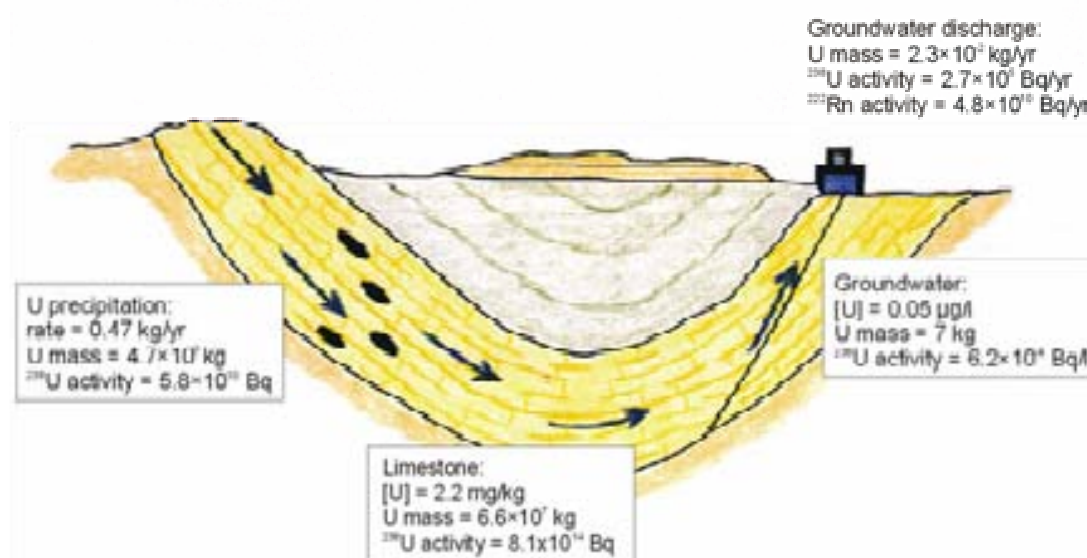


FIG. I4. Model of the U and Rn distribution and fluxes in the Bath Hot Springs.

## The Oxford Clay

The Oxford Clay is a Jurassic (145–215 Ma) mudstone formed by deposition in a low energy, partly restricted marine basin. The Oxford Clay is significant because it comprises a thick, low permeability, generally homogeneous formation. Such formations could potentially be suitable host rocks for a radioactive waste repository in the UK. Consequently, the Oxford Clay and other mudstones with similar mineralogy and physical properties, such as the London Clay, were considered as possible host rocks for shallow repositories in the mid-1980s. In addition, the Oxford Clay was considered in the generic Dry Run 3 assessment of a hypothetical ILW repository located in the vicinity of Harwell [15]. In this region, the clay crops out in the low lying land between the Cotswolds and the Berkshire Downs, where



it is crossed by the upper reaches of the River Thames. The following text summarized the results of the flux calculations for U and Rn.

The U concentration in the clay is moderate but, given that the volume of the clay is large, there is a large total U mass in the rock and a high  $^{238}\text{U}$  activity. Erosion leads to several kilograms of U released in solid form each year. The U concentration in the groundwater is also moderate but the rate of vertical groundwater movement through the low permeability clay is very slow and, thus, the annual U mass and  $^{238}\text{U}$  activity fluxes in the groundwater discharging to the Corallian aquifer are low. The model for U and Rn fluxes is indicated in Figure I5.

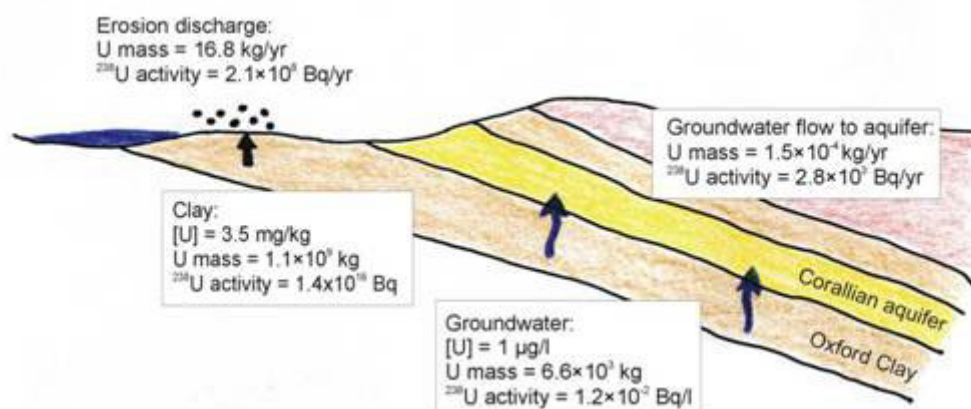


FIG. I5. Model of the U and Rn distribution and fluxes in the Oxford Clay.

## Conclusions

In this report, elemental and isotopic concentration data from a range of rocks, sediments and waters from the UK are presented. In addition, elemental and activity fluxes have been calculated for a range of UK specific geological environments. These concentration and flux data provide the raw material for the natural safety indicators methodology, in that they potentially can be compared with the abundances and distributions of repository derived chemical species calculated in a PA and, thus, can be used to assess the hazard posed by them. To test this approach a simple comparison has been made between the  $^{238}\text{U}$  fluxes from some of some of study sites (natural source) compared to the calculated  $^{238}\text{U}$  flux from a repository (calculated in Nirex's GPA). The results are given in Table I5.

TABLE I5.  $^{238}\text{U}$  ACTIVITY FLUXES FROM THE CARMENELLIS GRANITE AND THE LONDON CHALK, AND DUE TO PREDICTED REPOSITORY RELEASES TO SOIL CALCULATED IN THE GPA

Species	$^{238}\text{U}$ activity flux due to erosion (Bq/m <sup>2</sup> /yr)	$^{238}\text{U}$ activity flux due to groundwater discharge (Bq/m <sup>2</sup> /yr)
Carmmenellis Granite	2.3	0.13
London Chalk	2.5	4.7
Flux to soil derived from GPA output	-	0.017

Table I5 shows that the  $^{238}\text{U}$  activity flux to the soil based on determinations from the GPA output is essentially one tenth of the  $^{238}\text{U}$  activity flux due to groundwater discharge from the Carnmenellis Granite and less than 1% of that from the London Chalk, assuming groundwater was to be discharged equally over the exposed rock surface areas. On this basis it may be argued that the repository releases would not significantly alter the natural radioactive environment, at least for the sites considered here. In fact, it could be expected that the actual difference between the natural and repository total activity fluxes would be even larger because conservative assumptions were made in the repository release calculations. However, it would not be sensible to conclude on the basis of this comparison that the potential radiological hazard associated with the repository releases are proportionately small because some of the dominant nuclides in the repository releases have higher radiotoxicities than those in natural groundwater discharge.

These trial calculations illustrate that appropriate concentrations and fluxes can be quantified for natural systems which are suitable for comparison with performance assessment predictions of repository releases and that safety comparisons can thus be made relatively quickly and without recourse to complex mathematics. Such comparisons have potential use in both a formal, quantitative performance assessment and in less formal, qualitative demonstrations of safety that could be provided to non-technical audiences in a wider safety case.

The lack of a consistent database of natural geochemical and process rate data, and accurate determinations of average values, means that a number of assumptions have had to be made when quantifying concentrations and fluxes for the UK. More precise determinations would be possible, however, between natural and predicted waste concentrations and fluxes where a potential repository site is under investigation and site specific geochemical information has been obtained from the site characterization programme for use in the calculations.

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## DEFINITIONS

The following acronyms and abbreviations are used throughout this report.

ALI	Annual limits on intake by ingestion or by inhalation
ASAM	Application of Safety Assessment Methodologies for Near-Surface Waste Disposal Facilities, Coordinated Research Project
CRP	Coordinated research project
GBI	Geosphere-biosphere interface
ISAM	Improvement of Safety Assessment Methodologies for Near Surface Disposal Facilities, Coordinated Research Project
MPBBs	Maximum permissible body burdens
MPCW	Maximum permissible concentration in potable water
ppb	Parts per billion (e.g. $\mu\text{g}$ of solute per litre of water solvent)
ppm	Parts per million (e.g. $\text{mg}$ of solute per litre of water solvent)
RCM	Research coordination meeting
RERV	Repository equivalent rock volume