SOME COMMENTS FROM THE INTEGRATED PERFORMANCE ASSESSMENT POINT OF VIEW ON THE BEHAVIOUR OF A DEEP GEOLOGICAL DISPOSAL SYSTEM--CASES OF $^{129}$I AND HEAVY NUCLIDES RELEASES

By:
Didier GAY, François BESNUS, Christophe SERRES, Patrick BAUDOIN, Catherine CERTES
Institute of Protection and Nuclear Safety (IPSN/DES) - 77-83, avenue du général de Gaulle, 92140 Clamart, France

ABSTRACT

Performance assessment of a deep geological disposal requires the definition of a series of assumptions of very different natures. The particular features of the site studied, the amount and the characteristics of the waste to be disposed of, or the definition of the various assumptions and parameters used to build the transfer model must be specifically defined. The multiple sources of differences related to the assumptions made by the teams involved in this international exercise make the interpretation difficult when a broad comparison of results obtained is considered.

The work performed and the results obtained within the recent SPA project (Spent fuel Performance Assessment) however enabled to draw some general conclusions on the main radionuclides and parameters that potentially govern the radiological impact of a spent fuel geological repository. The particular importance of $^{129}$I was notably highlighted as well as the possible role of transuranic elements. For $^{129}$I, matrix degradation rate and dilution were found to have a predominant influence on the calculated dose. For transuranic elements, disposal system was found to delay the arrival of activity into biosphere over a very long period of time. A relatively wide range of breakthrough times were however observed depending on sorption modelling in geosphere in particular. As for transuranic relative dose contribution, a determining influence was played by solubility limit values.

BACKGROUND

The SPA project (Baudoin et al, 1999) was a direct continuation of the efforts made by the European Union (EU) since 1982 to build a common understanding of the methods applicable to performance assessment of a deep geological disposal. Devoted to the case of spent fuel, SPA was a follow-up to the PAGIS (Cadelli et al, 1988), PACOMA (Mobbs et al, 1991) and EVEREST (Cadelli et al, 1996) projects that dealt with the disposal of intermediate level long lived wastes and high level vitrified wastes.

One of the particular interest of these successive projects was to constitute a “practical” framework to develop and implement methods and tools for integrated performance assessment. They also enabled to draw preliminary conclusions on the respective importance of the radionuclides present in the different types of waste, on the influence of some of the main assumptions used in the modelling and helped to specify the role that could be expected from the various disposal system components.

In addition, international participation gave an opportunity to share experiences and practices. Different approaches could be compared and their particular interest and justification better understood.

In the case of SPA, six national research institutions representing implementing as well as technical safety organisations in six member countries of EU (ENRESA for Spain, GRS for Germany, IPSN for France, NRG for The Netherlands, SCK.CEN for Belgium and VTT for Finland) worked together from 1996 to 1999. The project comprised assessments on four different granite sites (by ENRESA, GRS, IPSN and VTT), three different clay sites (by IPSN, NRG and SCK.CEN) and two different rock salt formations (by GRS and NRG). In each assessment, in addition to a « normal evolution » scenario, two to three altered evolution scenarios were studied.

This paper only addresses the assessments made for granitic and clay sites and focuses on the case of a normal evolution scenario for which an overview of the results is proposed. It comprises a comparison of total doses and breakthrough times and the identification of most important contributors. From this comparison, the predominant role played by $^{129}$I in terms of total radiological impact is analysed and different issues arising from the radiological impact associated to heavy nuclides are addressed.
OVERVIEW OF THE PERFORMANCE ASSESSMENTS

Though the performance assessments undertaken within the project were only methodological exercises, they required the definition of a general framework to back up the different assumptions used in the studies. This general framework comprised the definition of the amount and nature of the wastes to be disposed of, the characteristics of the site and the characteristics of the different engineered barriers. Choices were generally made in agreement with the national contexts but did not intend to precisely reflect every technical option prevailing on projects actually developed in each country.

The total amount of activity considered to be disposed of in the repository depended on the starting assumptions used by each participant. It was usually relevant to the importance of nuclear power for electricity production and the waste management strategy in each participating country. This amount was thus relatively high in the case of GRS and IPSN (25,000 and 20,000 tHM respectively) and lower in the case of ENRESA, NRG, SCK.CEN and VTT (1,000 to 6,000 tHM).

Performance assessment entailed the modelling of five distinct components: the waste package, the container, a clayey engineered barrier surrounding the container – also called “buffer”–, the geological medium – sometimes comprising a Damaged Rock Zone (DRZ) – and the biosphere.

Container performance was dealt with through the definition of a mean failure time: before that time, all the containers were considered to be perfectly tight, after they were considered to ensure no role. Typical values for failure times were in the range of some thousands years. In the case of VTT, due to the choice of an iron-copper container, packaging integrity was however expected to last much longer and only a few defective containers were assumed to leak within the period considered in the assessment. For calculation purpose, VTT thus studied two reference situations: one assumed “total disappearing” of one container after 10,000 years, the other an “initial hole” in one container. In the first case modelling was similar to the “failure time” approach adopted by the other participants. In the second case, the geometry of the hole played an effective role on the control of activity release. Results and comments hereafter are based on the first case only.

The performance assessment of the waste form was dealt with by the definition of a source-term model. Because the distribution of radionuclides in spent fuel was considered to be heterogeneous, two distinct parts of a fuel assembly were distinguished. The first part was associated to the metallic parts (the claddings and the structural parts of the fuel assembly); it notably concerned the major part of C, Ni, Zr and Nb inventories. The second part was associated to the fuel pellets. Within the fuel pellets, a distinction was done between the part of activity bounded to UO$_2$ matrix and a fraction of some fission and activation products located in the gaps or at the grain boundaries (see Fig. 1). This latter fraction was essentially constituted of some % of fuel inventory of fission products such as C, Cl or I.

![Fig. 1: Schematic section of an irradiated fuel element](image-url)

In relation to this distribution of activity in the fuel assembly, a common source term model was defined by the participants and used in the assessments. It distinguished three different contributions. The first contribution was
associated with the inventory fraction located in fuel gaps; it was instantaneously released when water comes into contact with the waste form. The second was governed by the progressive degradation of claddings, structural parts and grain boundaries of fuel pellets; it led to a continuous release over 1,000 years. The third was governed the UO$_2$ matrix degradation; it led to a continuous release over one million years.

Performance assessment of buffer and geological medium was based on radionuclides groundwater transport modelling and involved the modelling of processes such as diffusion, advection, matrix diffusion, sorption and dissolution / precipitation.

In granite, the clayey buffer and geosphere were separately modelled by ENRESA, GRS and VTT and considered as two media with distinct properties within one single model by IPSN. In the first case, simplified 1-D radial (ENRESA, GRS) or compartment (VTT) diffusion models were used for buffer. Separate 1-D discrete models were then used to simulate transport in single fractures. In IPSN granite modelling, a single 3-D code was used to model the transport of radionuclides from the waste package up to the natural outlets, integrating both the Near-Field and Far-Field. A Disturbed Rock Zone (DRZ) at the buffer/geosphere interface was considered by all the participants. IPSN model explicitly accounted for the transport of radionuclides across the DRZ by considering that hydraulic conductivity was increased with respect to the rock mass, whereas the other participants dealt with the DRZ through a “mixing tank” boundary condition at the interface buffer/DRZ.

In clay, clayey buffer and clay host rock were assumed to have similar properties and these two media were not differentiated in the modelling.

Main assumptions for the Near-Field modelling are summarised in table 1.

| Table 1: Near-Field models in granite: main features, processes and assumptions. |
|---|---|---|---|---|
| Code | ENRESA | GRS | IPSN | VTT |
| Geometry | 1-D radial | 1-D radial | 3-D | Compartments |
| Near-Field representation | Explicit representation of container and engineered barriers | Explicit representation of container and engineered barriers | Full representation of the repository in geosphere | Explicit representation of container and engineered barriers |
| Contaminant transport in buffer | Diffusion to DRZ (advection neglected) | Diffusion to DRZ (advection neglected) | Diffusion/advection to host rock (equivalent porous medium) | Diffusion |
| Contaminant transport in backfill | Not considered | Neglected | Not considered | Advection and diffusion |
| Transport out of repository | Instantaneously along DRZ to major conducting features | Instantaneously along DRZ to major conducting features | Advection/diffusion in equivalent porous medium (DRZ and geosphere) | Into rock fissures, DRZ and via tunnel backfill |
| Source term for transport | Solubility applied in container (1 m$^3$ of water/container) | Solubility applied in container (0.3 m$^3$ of water/container) | Solubility applied in buffer (=8 m$^3$ of water/container) | Solubility applied in container (0.4 to 0.7 m$^3$ of water/container) |
| Outer boundary condition(s) | "Mixing tank" adective flux = diffusive flux (buffer/DRZ interface) | "Mixing tank" adective flux = diffusive flux (buffer/DRZ interface) | Diffusive flux = 0 at the outlets (geosphere / biosphere interface) | Transfer coefficients host rock/engineered barrier interface |

Geosphere calculation outputs at river or well outlets were converted in dose values using biosphere modelling. In the case of VTT, dose intake was limited to water drinking whereas the other participants also assumed that water was used for irrigation purposes and was transferred to plants and animals. Doses were then calculated as the sum of external exposure, inhalation and ingestion.
GRANITE FORMATIONS

Main features of the sites

Among the four assessments carried out for granite, three were based on hypothetical sites. Only VTT considered an existing site.

In ENRESA assessment, the generic site studied was developed from the data available for different granites already investigated in Spain.

In GRS assessment, the work achieved within the national project GEISHA (Papp, 1997) was used as a basis to define the generic site characteristics. Within the GEISHA project, the possible German granite sites were compiled and roughly characterised. Main granite formations were located in the Southern and Eastern parts of Germany. They were sited in a geological active region and might be intensively faulted or disturbed. Within the SPA project, the properties of the generic site were based on the data used in the Swiss study Kristallin-I (NAGRA, 1994) too.

In IPSN assessment, the hypothetical generic site was the same as the one defined within the framework of EVEREST. The topography and general geological and structural context were derived from investigation data available at a granitic site in Western part of France. These data were complemented by additional data judged to be representative of a granitic context. The massif reached a height of 320 m, intruding a surrounding schistose formation.

In the case of VTT, the assessment was based on the four candidate sites for a spent fuel repository currently investigated in Finland: the Hästholmen nuclear power plant (NPP) site in Loviisa, Kivetty in Äänekoski, the Olkiluoto NPP site in Eurajoki, and Romuvaara in Kuhmo. The data collected since 1997 within the programme of detailed site investigations were thus largely used to back the different assumptions done in the assessments.

Conceptual modelling of radionuclides transport

ENRESA, GRS and VTT used 1 D discrete approach to model the migration of contaminants through a single fracture taking into account matrix diffusion. These calculations were complemented with dedicated aquifer modelling. IPSN used a 3 D continuum approach based on a single porosity modelling transport up to the natural outlets. No complementary aquifer calculations were thus required. A schematic description is shown in Fig. 2 and main modelling assumptions are summarised in table 2.

IPSN modelled the groundwater flow as well as Near-Field and Far-Field transport of radionuclides with the same 3 D code based on a continuum approach (single porosity). The IPSN modelling assumed that small and intermediate scales of discontinuities might be homogenised. The larger faulted zones were explicitly represented as continuous 2 D planes included in the 3 D finite element mesh. The main structures outlined by the regional geology and taken into account in the modelling were grouped into three porous media: the faulted zones (described as continuous planes), the swarms associated with the faulted zones, and the rock mass. In each of the three entities, the nature of the migration phenomena was the same and differences simply resulted from differences in hydrogeological properties. Sorption phenomena were dealt with through the use of a retardation factor. Retardation was assumed to be essentially due to sorption on clay minerals filling the fractures. These clay minerals were considered to represent about 1% of the rock mass. Migration of radionuclides was performed from the entire set of galleries.

For ENRESA generic site, the structures or principal discontinuities were assumed to be classified in three groups depending on their extension. In addition to these three groups of structures with a hydrogeological role, a last one characterised by a lower hydraulic conductivity was considered. In this latter unit were grouped all the smaller fractures, such as smaller faults and joints. In the modelling approach, effective 1 D pathways were quantified from 3 D flow modelling taking into account these different structures. Migration of radionuclides was performed through these pathways from different parts of the repository to the outlets. The mathematical modelling was based on a 1 D discrete approach to simulate transport in a fracture accounting for matrix diffusion in the rock mass adjacent to the fracture plane. In addition to diffusion, sorption in the matrix was also taken into account. Both processes were not explicitly modelled but dealt with by means of effective retardation factors.
Assessments done by ENRESA and IPSN reflected a range of transit times that could realistically be associated with the heterogeneity of a crystalline rock at the scale of a potential repository (several km$^2$).

VTT and GRS considered that the radiological impact was dominated by the part of activity releases associated to the fastest channels in the geosphere. The complexity of the hydraulic patterns of the geosphere was reduced to one preferential pathway and the behaviour of the repository system was assessed with respect to this route. The modelling of the radionuclides migration was based on a 1 D discrete approach. The retardation was caused by diffusion from the water-conducting fracture into the adjacent rock matrix and sorption into the matrix (sorption on fracture surface and fillings was conservatively neglected). A key transport parameter was the combination between the flow rate in the fracture and the flow wetted fracture surface.

As a result from the previous differences, the transit times sometimes drastically differed from one assessment to the other. The shortest travel times from the repository to the biosphere performed by IPSN and ENRESA were thus in the order of some 1,000 years against some years for GRS and VTT. These results strongly depended on the parameterisation of the pathway(s) which was of prime importance to reflect the heterogeneity of the site properties of the bedrock at large scale.
Table 2: Far-Field models in granite: main features, processes and assumptions.

<table>
<thead>
<tr>
<th>Modelling approach</th>
<th>ENRESA</th>
<th>GRS</th>
<th>IPSN</th>
<th>VTT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Transport processes</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rock matrix</td>
<td>Diffusion+ sorption</td>
<td>Diffusion+ sorption</td>
<td>Rock matrix</td>
<td>Diffusion+ sorption</td>
</tr>
<tr>
<td>Rock matrix</td>
<td>Advection</td>
<td>Advection + dispersion</td>
<td>Equivalent porous medium</td>
<td>Advection</td>
</tr>
<tr>
<td>Open fractures</td>
<td>Advection</td>
<td>Advection + dispersion</td>
<td>Equivalent porous medium</td>
<td>Advection</td>
</tr>
<tr>
<td>Open fractures</td>
<td>Advection</td>
<td>Advection + dispersion</td>
<td>Equivalent porous medium</td>
<td>Advection</td>
</tr>
<tr>
<td>Infilled fractures</td>
<td>(channeling)</td>
<td>(channeling)</td>
<td>(channeling)</td>
<td>(channeling)</td>
</tr>
</tbody>
</table>

CLAY FORMATIONS

Main features of the sites

For CEN.SCK, the Boom clay of the Mol site was selected. The clay layer was more than 100 m thick, homogeneous, with a low hydraulic conductivity, good retention capacity and adequate plasticity. Its top was 180 m below the surface and no faults were detected at the site. Observations on the aquifers over and underlying the Boom clay indicated a general groundwater flow from East to West. At Mol, a downward gradient between the over and underlying aquifers was about 2 m over the 100 m thick clay layer. West from the Mol site, the hydraulic gradient over the Boom clay was upward.

NRG assumed that the disposal facility was located in the middle of a 100 m thick layer of Boom clay too but at an hypothetical site. The top of the clay layer was considered to be at a depth of 500 m. The main properties of the clay layer were analogue to those mentioned above.

For IPSN, the repository site reflected the main features of the Paris basin but did not correspond to a specific location. It was only assumed that the hypothetical repository area was located on the southern edge of the basin in the 167 m thick Toarcian/Domerian clay layer, at a depth of about 400 m. From a tectonic point of view, the area was considered as stable and located at some distance from the major faults affecting the Paris basin. In this zone, a vertical drainage of about $2 \times 10^{-5}$ m.year$^{-1}$ as well as a very small horizontal gradient in the overlying aquifer (velocity of about 0.03 m.year$^{-1}$) were considered. The resulting transport pattern through the Toarcian/Domerian host clay formation was essentially upward from the underlying Trias formations to the overlying Bajociam formation.

Conceptual modelling of radionuclides transport

For Belgian and Dutch sites, because advection was negligible in the Boom Clay, transport in the clay layer only involved omnidirectional diffusion. It made possible a model reduction based on symmetries in the disposal concept. The model reduction applied by SCK.CEN was a detailed 2-D model representing a fourth of a disposal gallery. In addition to diffusive transport in the clay layer, a separate aquifer modelling was carried out.

For the French site, advection and diffusion were both considered significant notably because of the higher permeability of the chosen clay layer. A fairly detailed, variable resolution 3-D mesh was used by IPSN to integrate the repository description in the 3-D geosphere model. Host clay layer and the surrounding aquifers were integrated in the same 3-D mesh. As for granite, no complementary aquifer calculations were required.

Table 3 and Fig.3 summarise the main features and assumptions of the conceptual modelling performed by IPSN and NRG/SCK.CEN.

Table 3: Comparison of the main features of the conceptual modelling in Clay.

<table>
<thead>
<tr>
<th>IPSN</th>
<th>NRG/SCKCEN</th>
</tr>
</thead>
<tbody>
<tr>
<td>Modelling approach</td>
<td>3-D continuum</td>
</tr>
<tr>
<td>Contaminant transport in the host rock</td>
<td>Diffusion/advection</td>
</tr>
<tr>
<td>Dilution in the overlying aquifers</td>
<td>$10^6$ [m$^3$/year]</td>
</tr>
<tr>
<td>Dilution in the overlying aquifers</td>
<td>$10^3$ [m$^3$/year] NRG</td>
</tr>
<tr>
<td>Dilution in the overlying aquifers</td>
<td>$6.66 \times 10^3$ [m$^3$/year] SCK/CEN</td>
</tr>
</tbody>
</table>
Fig. 3: Schematic representation of transport and dilution processes from the repository to a potential outlet.

- **C**: concentration [Bq.m⁻³]
- **Φ**: Activity flux at the interface Clay/Aquifer

### RESULTS

Figure 4 shows the time evolution of effective dose rates obtained by the different participants in the case of a normal evolution for granite and clay formations respectively.

From the results obtained, it could be concluded that (1) calculated doses were usually very low; (2) the contribution from fission and activation products to doses clearly differentiated from that of transuranic isotopes in terms of time of occurrence; (3) only a few radionuclides significantly contributed to the total radiological impact.

Looking at the time dependent dose rates resulting from the different calculations, a relatively clear distinction between three successive periods could be made: the first one was characterised by the absence of radiological impact, the second was dominated by the contribution of fission and activation products and the third by the contribution of heavy nuclides. The starting times for these successive periods varied from one site to another and from one calculation case to another.

According to the calculations performed, total dose rates were dominantly caused, in a first period, by the contributions of activation and fission products (¹⁴C, ³⁶Cl, ¹²⁹I, ⁷⁹Se and ¹²⁶Sn essentially), then by the contributions of heavy nuclides (the most important being ²²⁶Ra, ²³⁰Th and ²³⁹Th). For the fission and activation products considered to be non-sorbed or weakly-sorbed (notably ¹⁴C, ³⁶Cl and ¹²⁹I), arrival in the biosphere occurred relatively early in time. For all the participants but GRS, the major dose contributor in the first ¹⁰⁵ years was ¹²⁹I.

In clay, the release of fission and activation products into biosphere began after some ¹⁰,000 years for IPSN and NRG, after some ¹⁰⁰,000 years for SCK.CEN. Release of isotopes of transuranic elements occurred much later and never before several millions years. In granite, the release of fission and activation products began shortly after container failure for GRS and VTT and only after some ¹⁰,000 years for ENRESA and IPSN. Transuranic elements reached the biosphere only after some ¹⁰⁰,000 years in the case of IPSN and beyond one million years in the case of ENRESA, GRS and VTT.

A total of 28 fission and activation products and 37 isotopes of transuranic elements were initially considered in the assessments. A screening of the radionuclides having an individual contribution greater than ¹⁄₅₀₀₀ to the total dose at some time in the period covered by the calculations, brought to the following short-list: ¹⁴C, ³⁶Cl, ⁷⁹Se, ¹⁰⁷Pd, ¹²⁶Sn, ¹²⁹I, ¹³⁵Cs, ²³⁶U, ²³²Th, ²³⁷Np, ²³³U, ²³⁹Th, ²³⁶U, ²³⁴U, ²³⁰Th, ²²⁶Ra, ²³⁵U, ²³¹Pa. Within the first million years, ¹²⁹I was usually found to be responsible for more than ¾ of the maximum dose and often dominated the impact over the whole period. In granite, maximum dose obtained by GRS was however mostly due to ¹⁴C and ¹²⁹I was only one of the successive leading contributors along with ³⁶Cl and ¹³⁵Cs. For ENRESA and VTT, ¹²⁹I governed the maximum dose over the considered period but at some point in time, ³⁶Cl and ¹²⁶Sn respectively, were found to lead the impact. Lastly, for IPSN, after some ¹⁰⁰,000 years, isotopes of transuranic elements (mainly ²²⁶Ra and ²³⁰Th) became predominant and even caused the maximum dose level. A relative predominance of transuranic elements isotopes in the very long-term was also observed by participants who continued calculations beyond one million years. Thus, for GRS and SCK.CEN, the very-long term dose was dominated by ²²⁹Th and ²²⁶Ra respectively.
Figure 4: annual dose rates results in the case of normal evolution scenario – Granite formations

(4N: $^{236}\text{U} \rightarrow ^{232}\text{Th}$; 4N+1: $^{237}\text{Np} \rightarrow ^{233}\text{U} \rightarrow ^{229}\text{Th} \rightarrow ^{225}\text{Ra}$; 4N+2: $^{238}\text{U} \rightarrow ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$; 4N+3: $^{235}\text{U} \rightarrow ^{231}\text{Pa}$)
Figure 4 (con’t): annual dose rates results in the case of normal evolution scenario – Clay formations

(4N: $^{236}\text{U} \rightarrow ^{232}\text{Th}$; 4N+1: $^{237}\text{Np} \rightarrow ^{233}\text{U} \rightarrow ^{229}\text{Th} \rightarrow ^{223}\text{Ra}$; 4N+2: $^{238}\text{U} \rightarrow ^{234}\text{U} \rightarrow ^{230}\text{Th} \rightarrow ^{226}\text{Ra}$; 4N+3: $^{235}\text{U} \rightarrow ^{231}\text{Pa}$)
MAIN FACTORS GOVERNING $^{129}\text{I}$ CONTRIBUTION

In spite of some significant differences in the assumptions used by the participants to the SPA project, the particular importance of $^{129}\text{I}$ arose from the previous results as a clear common conclusion. $^{129}\text{I}$ was particular, firstly because it was always among the first radionuclides that reach the biosphere, secondly because it was always found to provide a significant contribution to the calculated dose.

$^{129}\text{I}$ importance was before all the result of the combination of three characteristics: a long half-life (16 millions years), a very high solubility (iodine was considered to be soluble by every participant) and a weak sorption (no sorption for IPSN, SCK.CEN, VTT and NRG and $K_d$ values of $5 \times 10^{-4}$ and $10^{-3}$ m$^3$.kg$^{-1}$ for ENRESA and GRS respectively).

The differences observed in Fig. 4 was thus to be explained by differences (1) in the amount of activity reaching the considered outlet, (2) in the capacity of the different barriers to delay and lower the released flux, (3) in the dilution capacity of the system and (4) in the assumptions made to take account for environmental transfer and exposure pathways.

With regard to the amount of activity reaching a specific outlet, both the hydrogeological characteristics of the site and the corresponding modelling assumptions were of particular concern. They could indeed entail the existence of different potential outlets and determine the distribution of the released activity between them.

For a granitic site, this distribution was closely linked to the assumption of well differentiated preferential pathways. In the case of GRS and ENRESA, a series of connected fractures were assumed to exist and to constitute potential pathways that drained part of the repository content. The main outlet was thus associated to only 25 to 30 % of the total flux released. For VTT, in addition to the existence of preferential pathways, the use of iron-copper containers further restricted the fraction of the inventory that effectively contributed to the impact: for the considered outlet, a single container was assumed to leak and thus contributed to activity release. This difference largely explained the lower values obtained by VTT.

In the case of clay site, the distribution of the contaminated plume in the geological medium only led to minor differences mainly linked with the assumption of purely diffusive transfer. In SCK.CEN and NRG assessments, only 50 % of the flux released from the repository thus migrated upward and eventually reached the top of the clay layer and the overlying aquifers whereas in IPSN assessment, because of the effective role played by leakage, the entire released flux reached the overlying aquifer. A schematic illustration of these two possible situations is shown in Fig.3.

With regard to the capacity of the different barriers to lower the released flux, a distinction was done between the two components of the source-term, namely the instantaneous release from the gaps and the continuous release from the fuel matrix. As suggested by the flat shape of $^{129}\text{I}$ curves in Fig. 4, the activity flux of $^{129}\text{I}$ that reached the considered outlet was remarkably steady for most of the participants. This suggested a predominant influence of the fuel matrix degradation on $^{129}\text{I}$ contribution.

The instantaneously released fraction was usually efficiently smoothed during its transfer through the successive barriers and did eventually not contribute significantly to the total dose (see Fig.4). One exception however existed for VTT, because of the relatively high conductive properties of the fracture constituting the preferential pathway and the resultantly very short transfer time through the geosphere ($\approx 10$ years). The pulse associated to the instant release fraction was then only weakly spread in time when reaching the outlet and thus significantly contributed to the total dose.

Contrarily to the case of instant release fraction, the second component of the source term (the continuous release from the spent fuel matrix) was usually not affected by the transfer through the successive barriers. The flux of $^{129}\text{I}$ reached a plateau equal to the flux released at the considered outlet. Because of the long half-life of $^{129}\text{I}$ and the matrix degradation time on the one hand, the transfer time through the disposal system on the other hand, a steady state was usually reached. The transfer through the disposal system did not enable to reduce the flux level. The efficiency of the system to control the activity release then relied on the fuel matrix performance. The geological barrier only delayed the release. The delay was more or less important depending on the hydrogeological properties of the barrier.

Only in the particular case of ENRESA, the delay brought by the geosphere was such that the previous comments were contradicted: no steady state was reached and the $^{129}\text{I}$ curve displayed a “peak” instead of a “plateau”. In ENRESA modelling, the retardation was provided by matrix diffusion and sorption onto the rock matrix. In spite of a
relatively weak sorption coefficient for iodine \( (K_d = 5 \times 10^{-4} \text{ m}^3\text{.kg}^{-1}) \), these two processes led to a high effective retardation factor \( (R \approx 1,200) \). Because of the resulting long transfer time, the source release stopped before the steady state was reached. In this particular case, \(^{129}\text{I}\) flux was not controlled by matrix degradation rate but by the hydrogeological properties of the site.

With regard to the dilution through the geological barrier, two phenomena were of concern. The first one was linked to the dilution properties of the geosphere itself and its ability to spread the plume, the second one was linked to a complementary dilution provided by surface water when a river was considered as the natural outlet. Practically, every participant (but IPSN for its granite site) considered a pumping well as the main outlet. In this case, only the first kind of dilution was effective. The dilution values corresponding to the results previously shown were summarised in table 4. These values corresponded to the annual volume of water in which the annual activity flux reaching the outlet was diluted.

**Table 4: comparison of the dilution values \( (\text{m}^3\text{.year}^{-1}) \).**

<table>
<thead>
<tr>
<th>SCK.CEN</th>
<th>VTT</th>
<th>NRG</th>
<th>IPSN granite</th>
<th>IPSN clay</th>
<th>ENRESA</th>
<th>GRS</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.67 \times 10^5</td>
<td>10^5</td>
<td>10^5</td>
<td>10^8 (river)</td>
<td>10^5</td>
<td>4.30 \times 10^5</td>
<td>1.25 \times 10^5</td>
</tr>
</tbody>
</table>

At last, with regard to the assumptions made to take account for environmental transfer and exposure pathways, the main factors of concern were on the one hand, the choice of exposure pathways, on the other hand, the choice of parameters values to model the radionuclide transfer in these pathways. As a results of differences in these factors, for a given radionuclide, a same concentration in water led to different dose values. For \(^{129}\text{I}\), the doses calculated by every participant for a unit concentration are shown in table 5. Though discrepancies were very limited between VTT and SCK.CEN, the calculated doses were 6 to 8 times higher for ENRESA, GRS and IPSN. The lower values obtained by VTT were, for instance, due to the fact that the only exposure pathway taken into account was the consumption of drinking water, whereas the other participants considered the use of water for irrigation and took into account the dose intake by ingestion of agricultural crops and animal produce.

**Table 5: Comparison of the effective doses calculated for a unit concentration \([\text{(Sv.} \text{year}^{-1})/\text{(Bq.m}^{-3})]\).**

<table>
<thead>
<tr>
<th>VTT/NRG</th>
<th>SCK.CEN</th>
<th>ENRESA</th>
<th>GRS</th>
<th>IPSN</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.5 \times 10^{-8}</td>
<td>6.11 \times 10^{-8}</td>
<td>3.82 \times 10^{-7}</td>
<td>3.70 \times 10^{-7}</td>
<td>4.80 \times 10^{-7}</td>
</tr>
</tbody>
</table>

To conclude, beyond the many differences that existed from an assessment to an other, the interpretation of the results obtained showed that the radiological impact due to \(^{129}\text{I}\) was governed by:

- the initial activity associated with the considered outlet (especially influent in the case of VTT),
- the degradation rate of the spent fuel matrix,
- the dilution at the outlet.

In addition to the previous elements, biosphere transfer and choice of exposure pathways had an influence too. However, according to the comparison made, this influence appeared to be relatively limited for \(^{129}\text{I}\).

Finally, as far as \(^{129}\text{I}\) was concerned, the performance of the spent fuel disposal system could usually be boiled down to an efficient smoothing of the instant released fraction associated to gap inventory and a delay of the continuously released fraction associated to matrix inventory. For the considered matrix degradation rate \( (10^{-6} \text{ year}^{-1}) \), the geological barrier did usually not level down the flux migrating from the source toward the considered outlet. The main parameter governing the dose was then dilution.

**MAIN FACTORS GOVERNING TRANSURANIC ELEMENTS CONTRIBUTION**

Whereas a good agreement existed between the participants to recognise \(^{129}\text{I}\) as one of the leading dose contributors, Fig. 4 shows that results for transuranic elements were much more contrasted. Significant discrepancies were thus found with regard to their relative importance in terms of dose and with regard to their breakthrough time at the considered outlet.

As compared to \(^{129}\text{I}\), the behaviour of transuranic elements radioactive in the geological disposal system was a priori more complex notably because it implied a combined effect of radioactive decay and filiation, sorption and solubility.
According to the assumptions adopted by the participants, transuranic elements were usually characterised by a high sorption ($K_d$) and a low solubility.

High sorption was first responsible for long transit times through the system. Because of these long transfer times, the medium half-life radionuclides in the head of the radioactive chains decayed before reaching the system outlets. Meanwhile, secular equilibrium established in the rest of the chains. Finally, at the system outlet, the decay chains were usually roughly reduced to the following reduced radioactive chains:

- **4N chain:** $^{236}\text{U} \rightarrow ^{232}\text{Th}$
- **4N+1 chain:** $^{237}\text{Np} \Rightarrow ^{233}\text{U} \Rightarrow ^{239}\text{Th}$
- **4N+2 chain:** $^{238}\text{U} \Rightarrow ^{234}\text{U} \Rightarrow ^{230}\text{Th} \Rightarrow ^{226}\text{Ra}$
- **4N+3 chain:** $^{235}\text{U} \Rightarrow ^{231}\text{Pa}$

(“$\Rightarrow$” means that a secular equilibrium usually established; in bold: radionuclides that could be considered to roughly govern the activity of their daughter nuclides)

Once the secular equilibrium was reached, the relative importance of a radionuclide in a given chain was mainly determined by its radiotoxicity. Because of their higher radiotoxicity, daughter nuclides in the tail of the decay chains (notably $^{226}\text{Ra}, ^{229}\text{Th}, ^{230}\text{Th}$ and $^{231}\text{Pa}$) thus usually played a leading role in terms of dose. Among them, $^{226}\text{Ra}$ was often recognised as one of the most important dose contributor.

The relative importance between the different decay chains was mainly influenced by the solubility limit of three elements (Np, U and Th) and the isotopic content in U. The higher contribution of 4N+2 chain as compared to the contribution of 4N+3 was thus governed by the higher content in $^{238}\text{U}$ than in $^{235}\text{U}$. The relative contribution of 4N+1 chain as compared to 4N+2 depended on the relative solubility of Np and U. As most of the participants used solubility limit values several orders of magnitude lower for Np than for U (some $10^{-6}$ mol.m$^{-3}$ against $10^{-1}$ to $10^{-4}$ mol.m$^{-3}$), 4N+2 chain usually dominated. For VTT and GRS, solubility limit values for U were however lower and rather similar as the values for Np. The importance of 4N+1 chain was thus increased.

These differences in solubility limit values also explained why transuranic elements contribution varied so widely from one participant to another. For the higher values of U solubility ($10^{-1}$ to $10^{-2}$ mol.m$^{-3}$ for SCK.CEN and IPSN), the dose obtained were similar or even higher than those obtained for fission and activation products whereas they were nearly negligible for the lower values of U solubility.

Another essential factor determining the results obtained for transuranic elements was the retardation by the geological medium. For this factor, the assumptions made to model far-field transfer appeared to have a very significant influence. According to the description made earlier in the text, three different situations were considered. In clay, the whole porous medium was involved in sorption; in granite, when preferential pathways through connected fractures were considered, retardation was due to matrix diffusion and sorption onto rock matrix; lastly, for IPSN granite site, no differentiated preferential pathway was assumed and the host rock was dealt with through an assumption of equivalent porous medium. In this latter case, sorption was assumed to occur onto the clay particles filling the small fracture network only.

In the first two cases, the sorption coefficient considered for transuranic elements ($K_d$ usually close to 1 m$^3$.kg$^{-1}$) led to a very strong retardation: breakthrough time was thus delayed beyond a million year. Retardation appeared to be particularly efficient for transfer in a fracture: in spite of relatively rapid transit time for water, matrix diffusion and sorption cause the transuranic elements to reach the outlets only after very long time. Only in the third case (for IPSN in granite) an “early” contribution of transuranic elements (at some hundreds thousands years!) was obtained.

Results obtained thus suggested that, at least in the case of normal evolution scenarios, the contribution from transuranic elements could be delayed up to significantly long periods of time. But at the same time, results did not clearly showed that the level of this contribution could be considered as definitely insignificant, especially as compared to the contribution from fission products. Because quantitative assessment tools clearly lose a great part of their credibility for the considered time periods, the need for an appropriate approach to demonstrate that a sufficient level of protection can be reasonably reached even at very long time scale was questioned. In this respect, natural analogues such as uranium ores was considered of a particular potential interest. It could in particular enabled to put the long term risk associated to a deep geological disposal of high level wastes into perspective and could therefore contribute to enhance the overall level of confidence.
CONCLUSION

Although the SPA project involved very different features (different geological sites, different conceptual modelling and mathematical approaches), general lessons could be drawn from this study.

The geological system first appeared to be an efficient filter for most of the radionuclides present in spent fuel assemblies. In most of the case, the radiological impact was dominated by $^{129}$I in a first period of time, then by the possible contribution from transuranic elements.

For $^{129}$I, activity flux was likely to be controlled by the degradation rate of the spent fuel matrix, the geological barrier providing only a delay in the arrival of the flux at the outlet. In this case, dilution was a key parameter governing the radiological impact. As a consequence, adequate identification and characterisation of the water resources potentially affected by the radioactive plume seemed to be of primary importance.

Because of their high sorption onto geological medium, heavy nuclides were notably delayed before reaching the biosphere. This delay that depended on modelling assumptions made by each participants, was always strong but differed from one assessment to the other. Relative dose contribution varied over a relatively large range mainly as a result from differences in solubility limit values. In some cases, they were of similar importance as contribution for fission and activation products. Because of the very late breakthrough times obtained for transuranic elements and because of their possibly significant contribution, the question of modelling reliability at very long time scales was raised.

REFERENCES