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Geological disposal of nuclear waste: II. From laboratory data to the safety analysis – Addressing societal concerns

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ABSTRACT

After more than 30 years of international research and development, there is a broad technical consensus that geologic disposal of highly-radioactive waste will provide for the safety of humankind and the environment, now, and far into the future. Safety analyses have demonstrated that the risk, as measured by exposure to radiation, will be of little consequence. Still, there is not yet an operating geologic repository for highly-radioactive waste, and there remains substantial public concern about the long-term safety of geologic disposal. In these two linked papers, we argue for a stronger connection between the scientific data (paper I, Grambow et al., 2014) and the safety analysis, particularly in the context of societal expectations (paper II). In this paper (II), we assess the meaning of the technical results and derived models (paper I) for the determination of the long-term safety of a repository. We consider issues of model validity and their credibility in the context of a much broader historical, epistemological and societal context. Safety analysis is treated in its social and temporal dimensions. This perspective provides new insights into the societal dimension of scenarios and risk analysis. Surprisingly, there is certainly no direct link between increased scientific understanding and a public position for or against different strategies of nuclear waste disposal. This is not due to the public being poorly informed, but rather due to cultural cognition of expertise and historical and cultural perception of hazards to regions selected to host a geologic repository. The societal and cultural dimension does not diminish the role of science, as scientific results become even more important in distinguishing between the conflicting views of the risk of geologic disposal of nuclear waste.

1. Introduction

Nuclear waste is one of the key environmental concerns regarding the use of nuclear energy. As early as 1978 it was stated that "Unless a solution can be found for the permanent disposal of radioactive waste and spent fuel, there will not be the consensus needed to go ahead with nuclear power as an energy source. ... We have to convince the public, those who know little about nuclear power but fear radiation" (US House of Representatives, 1978). Geological disposal is considered by most national and international bodies and large parts of the scientific community to be the best strategy to reduce the long-term risk of highly-radioactive wastes arising from nuclear energy production (Blue Ribbon Commission on America's Nuclear Future, 2012; European Council, 2011; Ministry of Trade and Industry (Finland), 1994; OECD/NEA, 1995; Republique Française, 2006). Regarding geological disposal, the NEA Radioactive Waste Management Committee issued a Collective Statement (Nuclear Energy Agency, 2008) noting:

> "*The overwhelming scientific consensus worldwide is that geological disposal is technically feasible. This is supported by extensive experimental data accumulated for different geological formations and engineered materials from surface investigations, underground research facilities and demonstration equipment and facilities; by the current state of the art in modeling techniques; by the experience in operating underground repositories for other classes of waste; and by the advances in best practice for performing safety assessments of potential disposal systems.*"

Nevertheless, serious doubt remains in the public mind and are expressed by Non-Government Organisations, NGOs (Wallace, 2010). Industrial solutions for the storage of low-level waste (ANDRA, 2011) or transuranic radioactive wastes (WIPP, 2010) are operating in various countries. Some radionuclide releases to the local environment were reported, without measurable health effect, but resulting in large public concerns (Arimone et al., 2010; McBride and Grace_Lucas, 2014). No geological disposal site

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exists for most of the radioactivity generated during nuclear energy production. This waste mostly exists as: spent nuclear fuel or nuclear waste glass in countries that reprocess their spent nuclear fuel. There are almost 300,000 tons of spent fuel generated worldwide, largely stored at reactor sides and about 100,000 tons have been reprocessed (IAEA, 2009). Both forms of waste generate a considerable amount of heat during the first few thousands of years and remain radiotoxic for several millions of years.

The question remains whether sufficient societal consensus and confidence (OECD/NEA, 2013) can be established as regards the long-term isolation of radioactive waste by means of passive natural and engineered man-made barriers that will not need later human intervention. Until now, society has never pursued such an enterprise. The debate on the fate of nuclear waste has continued for more than 35 years (Rosa et al., 2010) even though as early as 1982, it was commonly believed and stated by government representatives that the issues were not scientific, but rather political, and that the remaining challenge was for scientists and engineers to convince the public of the safety of a geologic repository. For example, in the United States, the Nuclear Waste Policy Act (NWPA), amended in 1987 – which tied the entire US high-level waste management program to the fate of the Yucca Mountain site – has been derailed and has failed to produce a timely solution for dealing with the US's most hazardous radioactive materials (Blue Ribbon Commission on America's Nuclear Future, 2012).

The evolution of a geological isolation system over many hundreds of thousands of years is not predictable in detail, and various scenarios over time have to be developed (NEA/OECD, 2001) in order to analyze all features, events and processes (FEPs) combinations that might occur in the future and that could compromise the required isolation performance of the geologic repository. Among the key questions that need to be addressed are:

- What is the geological stability of the site, for example against earth quakes?
- What are the risks of human intrusion?
- How long will containers remain 'water tight' in the face of inevitable corrosion by groundwater?
- At what rate will radionuclides be released from the waste into groundwater?
- To what degree will contaminated groundwater be 'decontaminated' by interaction with the surfaces presented by minerals contained within the engineered barrier materials and rocks?
- How long will the migration of radionuclides through the rock take, and when and at which rates will they eventually enter the biosphere?

Any assessment of the long term performance of a waste facility requires a thorough understanding of the physico-chemical mechanisms that govern either retention of radionuclides or their mobilization, of the hydraulic properties of the confinement system, of the spatial and temporal evolution of the geochemical and geological system, and this must be represented in detailed mathematical simulation models constrained by, and coherent with, the best available knowledge. Sensitivity and uncertainty analyses must be performed to assess confidence levels. Without covering all of the questions, an example for the type of necessary data and their technical interpretation is given in paper I.

The only problem, but a major one from a societal point of view, is that the large number of processes, models and parameters involved in the formal demonstration of repository safety does not offer any clearly evident measure of safety that is understandable to politicians and citizens or even to scientists which are not directly involved in the analyses. Furthermore, due to the long times for which estimations of safety must be performed, the overall outcome cannot be verified, as it is inferred from many individual observations made in many laboratories and by field observations, all of which are necessarily simplified in the analysis. This poses a number of important questions that must be addressed:

- How can one deduce from short-term experimental data, measured on centimeter- to decimeter-sized rock samples, the long term behavior of geologic units over a scale of hundreds of meters?
- How can one be assured that a scientific model or theory derived from experimental data is applicable under larger-scale repository conditions?
- What should be done when different models are consistent with the same data set?
- How does one combine simple models for various individual processes into an overall vision of the performance of the waste isolation system?
- How does one understand and evaluate the performance of individual barriers as a function of time?
- What are the appropriate indicators for safety and risk?
- How credible are projections for hundreds of thousands of years?
- Is there any use in carrying out predictions for hundreds of thousands of years, if all data indicate that the risk will be negligible for the next tens of thousands of years?
- Is there a direct impact of an increase in scientific understanding on the perception of risks from nuclear waste disposal?

This paper addresses these issues by examining a specific example: the disposal of high-activity waste in a geologic repository in a clay formation. We have discussed the properties of clay as a repository medium and how these data can be used to assess the mobility of radionuclides in the first paper. In this paper we discuss how this first step is extended to a discussion of safety and risk in order to address societal concerns.

We should here note that the issues raised in this second paper transcend the simple application of the scientific method, as classically described, to the issue of the long-term safety of a geologic repository. The first paper is a typical scientific analysis of experimental results that is intended to validate models used to simulate the key features of a geologic repository. The natural science approach requires that all data, model hypotheses and experimental conditions be thoroughly documented and be self-consistent. However, the link between the technical data, concepts, and derived models with the temporality of environmental and health risks is not addressed, nor is it trivial. Here we show the need to avoid the easy and self-sufficient shortcut, believing that the comparison of model and data assure, by themselves, the credibility and validity of models that are extrapolated for the long term and that temporal scales of hundreds and thousands of years have any self-evident meaning outside of the scientific community. In this second paper, we address the validity, credibility and temporality of the models in the first paper in the much larger historical, epistemological and societal context. Finally, we address risk and safety in its social dimension avoiding considering it as purely technical issue as is normally done.

2. From experimental data and models to safety evaluation

In Grambow et al. (2014), we have presented some critical data for clayrock that are required for an analysis of radionuclide mobility in a geologic repository in clay. The extension of the empirical data to the geologic repository performance, of course, requires a theoretical framework supported by the appropriate models. However, the data and models have inherent uncertainties. The experiments are conducted with the objectives of parameterizing the models and of identification of the associated uncertainties. In turn, the question must be posed whether the experimental conditions and sample geometry are representative and whether these models and the associated parameters (i.e., diffusion law and diffusion coefficients, Darcy's law and permeability values, solubility of minerals, mass action equations of species in solution, etc.) derived from small samples are also valid and applicable to the real situation, and for which we have assumed the rock samples to be representative.

2.1. Model validity

How do we know whether a model derived from short-term experimental data of cm-sized rock samples is correct and applicable to the long-term assessment of the barrier function of a geological repository? Models need to remain applicable and credible during scale up in space by a factor of 10^2–10^3 and in time by a factor of 10^5–10^6.

In the nuclear waste management literature one often finds claims that a model has been "validated". This simple word can have many meanings and the "validation" may be serious and thoughtful or superficial and useless. In particular as far as public policies are at stake, scientist and agencies are eager to propose or to use "validated" scientific models to allow for rational decisions. In this particular discussion the question is focused on whether models are "valid" over rather extension of spatial and temporal scales. Oreskes (Oreskes et al., 1994) argue that verification and validation of numerical models of natural systems is, in principle, impossible and only impartial confirmation by the demonstration of agreement between observation and prediction is possible. The reasons are that natural systems are never closed, that there are always incomplete and hardly accessible input parameters, non-verifiable assumptions and often one is confronted with the non-uniqueness of models. This is evident for models describing nuclear waste repository safety, as validation would require a conclusion that never in the future will there be an observation that disagrees with the model. In the absence of future observations, a full validation cannot be made but efforts for validation of procedures of reasoning or of the use of indirect means (application of a natural law, comparison to natural analog systems...) need to be pursued.

The question of model validation has been addressed in the larger context of science and society and the meaning of truth (Nordstrom, 2012). Based on correspondence theory of truth, scientific laws, theories, models are more certain (more valid?) the more their consequences correspond to reality. But how can we measure correspondence and how to compare it with a reality in the very far future? Alternatively, coherence theory of truth refers to the consistency of a theory with the large body of scientific truth. But such a theory does not only limit any new discovery in conflict with previous knowledge, it also does not avoid the risk of incompleteness and contingency of model descriptions. Even if a model is consistent with all scientific data, its application to long-term predictions will remain uncertain. Nordstrom finally suggests thinking in terms of the usefulness of models. It becomes clear: the quest for validated models asks too much from science and creates expectations that cannot be satisfied. It is not a failure but the nature of science that it cannot provide fully validated answers to the urgent societal problem of nuclear waste management. However, it is also part of the nature of the scientific method that it requests trying to validate (or falsify) models and their parameterization. The orientation on usefulness of models cannot replace the need of exposing them to validation procedures. The dynamics between ever continuing efforts in validating models and the impossibility for full validation constitutes a balance wheel driving scientific credibility in the assessment of the safety of a nuclear waste repository concept. This process of partial validation can establish a rational base for understanding and reducing risk and uncertainties. As such it can be used in support of decision making, trying to define bounding values for barrier performance and to take into account best estimate understanding of the disposal system at a given moment.

Taking the geochemical clay pore water model from (Grambow et al., 2014) as an example: can one say that it is validated since it corresponds to the data presented in the paper? Indeed great progress has been made in the geochemical literature as regards the prediction of groundwater composition and water–rock interaction based on thermodynamic data (Glynn and Plummer, 2005). This is not to say that there may not be inherent short-comings that may affect the application to these data and the ability to predict, such as incomplete data bases, extrapolation of short term laboratory data to the long-term, missing kinetic constants of mineral reactions or incapacity to describe future groundwater flow and associated uncertainties correctly (Ewing et al., 1999). Still, while admitting the inherent partial character of model confirmation, characterized by incompleteness (not all trace elements are described) and non-uniqueness (different mineral phase assemblages and solid solutions may explain the same water composition), this geochemical pore water model adequately describes experimentally observed non-trivial evolution of solution concentrations, indicating the usefulness of the model. The problem arises in defining what is adequate. Even scientists sometimes do not agree on this. The answer depends on the purpose of the pore water model. If it were only to explain observed water compositions or the ionic strength, the model may be satisfactory. If on the other hand it would have to be used to assess the none-measured carbonate concentrations, one can postulate that the model assuming cacite equilibrium may provide a reasonable estimation since (1) water residence times are long, (2) equilibria between calcite and water at high rock/water ratios in pore space are fast and (3) measured Ca and pH values agree to model predictions. Considering these limitations, the model can be used to represent pore water compositions in more complex water/rock interaction models, such as those involved in the transport of radionuclides. Depending on the sensitivity of radionuclide migration on carbonate concentrations (low for ^{135}Cs and ^{129}I, high for Uranium) some more extensive validation effort (measurement of carbonate) would be necessary or not.

Another example from Grambow et al. (2014) is the development of the water transport model and the hydrodynamic parameters derived from the data. In the 1990s, there was a very passionate debate on whether groundwater transport models can be validated at all. According to some authors (Konikow and Bredehoeft, 1992) the predictive capacity of such models can be improved but even agreement of model and reality is no proof of model validity, and the accuracy of prediction can only be assessed once the predicted period of time has passed. More practically, others (de Marsily et al., 1992) have argued that, since groundwater transport models are based on mass balance and Darcy's law, both of which have shown applicability in a huge amount of cases, if a model based on these principles reproduces the observed behavior it can also be used for predictions. It is not always clear whether mass balance is assured in complex overall systems performance assessments, but because the data in Grambow et al. (2014) can be used to show that Darcy's law is applicable under the conditions of the applied high hydrodynamic gradients, it is evident that the law can be used in principal to predict the rate of water transport driven by realistic low hydraulic gradients (i.e., the same parameter applies for all gradients) in more complex models. It has been concluded from the data in Grambow et al. (2014) that diffusion is the principal transport mechanism for water in the Callovo-Oxfordian clay formation.

With the limit of partial validation, which is true for all empirical science, we can deduce from the correspondence between the model and experimental reality that the models parameterized by the percolation test results from Grambow et al. (2014) can be applied "with caution" to the large scale repository conditions since they are describing consistently the observations based "on the large body of scientific truth" (diffusion law, Darcy's law, solubility of minerals, and mass action equations of species in solution).

2.2. Credibility of predictions

Application of a "partially validated model" (=a model which did not fail in a number of attempts of validation) to large time and space scales has the character of a prediction. One may prefer the term "long term assessment" since one does not intend to predict a real future evolution and many evolutions are possible; one only tries to assess the consequences for wide range of possible evolutions. The question is how credible predictions for nuclear waste disposal performance can be? This is not only a question of the availability of a number of partially validated models; it is the question of coherent coupling of these models, of their adequate parameterization, of correct boundary conditions and of many, often ill-defined supplementary assumptions, such as the representation of heterogeneities on various scales with respect to porosity, mineral surface area, and mineral composition. It is also the question of an exhaustive assessment of the evolution scenarios of the disposal system, of the interrelationship in the performance of the various geological and engineered barriers, of the radioactivity inventories of the waste and of the associated uncertainties. The use of a valid model with wrong boundary conditions or overly optimistic scenarios will certainly not allow credible predictions. Before discussing some safety assessment approaches in more detail, some general considerations on the credibility of predictions are addressed.

2.2.1. General concerns on credible predictions

According to Nordstrom (2012), we can distinguish between phenomenological and chronological predictions. The first applies to time-independent phenomena, such as thermodynamic solubility. The prediction of the migration of radionuclides in a repository is a chronological prediction.

Predictions (or assessments) of evolutions of natural systems are faced by inherent uncertainties, incomplete information and flaws, but despite the impossibility of full model validation, many predictions remain credible. Examples are the prediction of the long term stability of geologic formations based on geological records or the prediction of heat transfer in a repository. The mere notion of "credibility" implies that we do not know the truth or have completely validated models. Scientific understanding can increase the credibility of predictions, despite the empirical character of the laboratory data (Grambow et al., 2014) for the assessment of the long term migration behavior of radionuclides in clay cores. With respect to credibility, one may distinguish between two types of predictions which we may call strong and weak predictions: strong predictions are very credible. They are for example based on natural laws (Fick's law, Darcy's law...), on deduction based on the quantitative understanding of the reasons of a certain observation. Or they are based on interpolation between known states considering very well defined boundary conditions. The use of certain sets of polynomial expressions (SIT theory, Pitzers equations...) fitting large series of experimental data on activity coefficients at various ionic strength, ionic media, temperatures and pressures allow for example very accurate prediction of activity coefficients, provided that the predicted value lies in the field covered by the measurements (interpolation). In this case, full understanding of the experimental data is not necessarily a prerequisite for accurate predictions. Extrapolation to conditions not covered by experimental data on the other hand yield highly uncertain, weak predictions. Weak predictions are those based on extrapolation and analogy without understanding of the underlying processes or without being able to base the predictions on conditions covered by a very large field of experimental observations.

2.2.2. Strong predictions and interpolation

Strong predictions are applicable for simple systems and for simple aspects of complex systems. The confidence in science since the Greeks, and even more evident since the renaissance has strongly benefited from successful strong predictions even though false predictions probably outweigh correct ones. An example is the prediction of the solar eclipse of 28th May 585 BC or the prediction of the return of the comet observed by Halley in 1682.

Predictions based on radioactive decay or of heat conduction in a repository are typical examples for strong predictions. There is general agreement on using the law of radioactive decay or Fourrier's Law for predicting future inventories of radioactive waste or temperature evolutions in a repository, provided that decay constants and thermal conductivity and the initial conditions (radioactivity inventory...) are known with the necessary accuracy.

Another example is the migration of radionuclides in the repository rock. Radionuclides migrate in pore water by diffusion driven by chemical gradients and by water movement due to hydraulic gradients. Fick's and Darcy's laws governing these processes have been shown applicable (Grambow et al., 2014) considering distinction between anion and cation accessible porosity as well as distribution coefficients for the radionuclides between water and mineral surfaces. Migration times across the Callovo-Oxfordian Clay layer of 500,000 and 100 million of years have been estimated for ^{129}I and ^{135}Cs, leading to disappearance of ^{135}Cs by radioactive decay prior to completion of the migration path. Strong predictions would require that the variation of porosities, pore water compositions, permeabilities and of distribution coefficients are known at each position along the 70 m long transport paths in the Callovo-Oxfordian clay formation and that these properties remain either constant or that its temporal evolution is known over the period of interest. Constancy of conditions may be considered in the far field but in the near field close to the waste, the conditions may change due to reactions of pore water with engineered barrier materials (iron, concrete...). There are two ways to account for these variations: (a) to study a number of representative samples allowing for exhaustive interpolation between them or (b) to build a model which relates mineralogical variation and retention properties (see for example for Cs in Montavon et al. (2014)). The number of samples in the study of Grambow et al., 2014 being limited to four (Table 3, paper I), the migration parameters will have relatively important uncertainties. Within this range of uncertainty, interpolation between these samples allows for credible consideration of spatial variations since the rock samples studied are representative for some extreme cases in carbonate and clay mineral contents and for hydrodynamic characteristics. As far as temporal variations in the vicinity of the waste are concerned, principal modifications of rock properties are imposed by interaction with high level waste, leading to the formation of iron rich clays and of iron oxides caused by release of iron from corroding containers. Iron oxides are known to be very strong adsorbents for radionuclides. When this temporal evolution is ignored, this beneficial effect will not be taken into account. This ignorance decreases the credibility of the model to predict actual behavior but it increases the credibility to predict conservative bounding values for radionuclide migration. The term "conservative" means calculated migration rates are faster (higher risk) than the rates expected in presence of iron oxides.

2.2.3. Weak predictions: extrapolations, mechanism and analogy

The situation is more complicated with extrapolations: an inductive reasoning from a laboratory experiment conducted for few days to years to an unknown situation (repository in far future...) is less credible than the prediction of experimental results at time steps less than a year. Similarly, in the context of radionuclide migration in clayrock, the interpolation between two known extreme states (samples of high/low clay contents...) to an unknown intermediate one appears to be more convincing than extrapolation from two known intermediate states to an unknown extreme one, where the terms "extreme" or "intermediate" refer to properties (clay content) proven to determine radionuclide retention or hydrodynamic constraints. Another case is the kinetics of nuclear waste glass dissolution: direct extrapolation of trends of glass dissolution rates made 30 years ago (Altenhein et al., 1982, 1983; Altenhein et al., 1984) has led to predicted glass life times of hundreds of thousands of years. Current predictions based on much more sophisticated models give rather similar life times. From the viewpoint of current knowledge, one can certainly qualify the previous prediction as weak since experimental data collected for 1 year where fitted by an empirical polynomial expression of ever decreasing rates extrapolated to an unknown situation millions of year ahead. If we would have used the type of data as form the glass dissolution experiment in paper I for such an extrapolation, it would as well be a weak prediction even though repository rock was present.

We argue in the following that today's predictions are stronger (more credible) since they are based on mechanistic understanding of glass dissolution. A physical chemical mechanism and the environmental factors capable to interfere with these mechanisms intend to provide a sound scientific base according to which events are causally related when there is a mechanism that connects them (Glennan, 1996). The weakness of the predictions of glass dissolution 30 years ago stems from the extrapolation of trends without a clear causal framework relating glass dissolution rates with the environmental conditions to which the glass is exposed. For the problem of extrapolations from laboratory to repository, the idea is that the same physico-chemical causes shall produce the same effects even in one million years. However, there always remains a difference between the laboratory and the target situation. Guala (2005) suggested as criteria the existence of some empirical knowledge of model situation and target, but such empirical knowledge is impossible to gather for situations in the far future. Some fundamental problems of the mechanistic approach were recently addressed (Howick et al., 2013), in particular the fact that knowledge of the mechanism is always incomplete.

2.2.3.1. Comparative process tracing. To assess whether a mechanism can be used as a basis for an extrapolation, Steel (2008) has proposed a procedure called "comparative processes tracing". Applied to nuclear waste glasses, this would require one to study all important mechanism that may occur in the model system (mm- to cm-sized simulated nuclear waste glass exposed to various simulated environments in the laboratory). The reliability of comparative process tracing then depends on correctly identifying the points at which significant differences between the laboratory studies and the canistered nuclear waste glass in a future repository are likely to occur. Significant differences are those that would make a difference as to whether the causal generalization to be extrapolated remains valid in all evolved futures that might occur to the glass and its environment during disposal.

In this sense, large experimental research programs have been conducted worldwide for more than 30 years that have investigated the effects of glass composition, surface area, solution volume and flow rate, temperature and pressure and solution composition, and all in the presence of repository materials such as iron, cement, rock samples, and in radiation fields. In general, the results showed very low long-term glass dissolution rates in water in a confined space such as is expected in the repository near-field. General mechanism were developed from model systems to explain the impact of all these variables on the model system by fundamental physico-chemical processes and parameters like diffusion, solubility, transition state rate laws, and activation energies. This extensive and compelling data base supports a strong prediction. There was some debate whether affinity terms or protective surface layers where responsible for the observed decreasing reaction rates, but today there is agreement that both processes are operative (Frugier et al., 2006; Grambow and Muller, 2001; Van_Iseghem et al., 2007). These studies allow one to address the differences between laboratory models and the repository target: geometric constraints like size and fracturing of the industrially produced highly radioactive glass bloc, the hydrodynamic conditions, the radiation fields, and the contact of iron from canister. In this sense the glass dissolution experiments by Grambow et al. (2014) were conducted to "validate" the applicability of current mechanisms to the quantitative understanding of glass corrosion in the clay–water system.

2.2.3.2. Natural analog systems. The largest difference between model and target is the exposure history to an aqueous environment. This cannot be studied in the laboratory, but only by studying so called "natural analog" glasses. Concerning analogy, it certainly plays an essential role in conceptualization of experimental data, in developing evolution and exposure scenarios for repository performance in a qualitative way. Many natural analog studies were performed (Alexander and Van Luik, 1990; Brandberg et al., 1993) for example using volcanic basalt glasses (Crovisier et al., 2003, 1987; Ewing, 1979; Ewing and Jercinovic, 1987; Lutze et al., 1985; Mazer et al., 1992) as an analog to study the long term performance of nuclear glasses in natural environment, and it certainly increases credibility of arguments in favor of repository safety. For example it can be shown that (1) natural basalt glasses behave under similar conditions similar to nuclear waste glasses, (2) natural basalt glasses can survive in natural water conditions for millions of years (Malow et al., 1984), (3) that complete transformation of glass in clays and zeolites will occur after sufficient long time and (4) that low rates observed under laboratory conditions in confined silica saturated environments are also observed with natural glasses (Grambow et al., 1986). While not allowing strong predictions, natural analog studies provide a strong qualitative support to the safety case (IAEA, 2012). Additionally, one can move beyond the analogy and actually study natural systems in order to understand and quantify their complexity and determine what parameters matter.

2.3. Bounding case predictions

Detailed prediction of the release of radionuclides from a nuclear waste repository cannot be considered a strong prediction. The prediction of the evolution of a geological system over geological time is not possible, due to variations in the initial and above all, boundary, conditions, since the system is open to the exchange of mass and energy. Also, there is the possibility of rapid unforeseen events. In the case of a nuclear waste repository *it is neither possible nor necessary to do an exact and quantitative prediction of the transfer of radionuclides from the disposal location to the biosphere in space and time*. Also the ICRP states (Weiss et al., 2013) that the scientific basis becomes very uncertain for assessments of health impacts over very long times into the future indicating that *the strict application of numerical criteria may be inappropriate*. It is sufficient to demonstrate that dose and risk thresholds are not exceeded at any given time for during the evolution of the disposal system and its environment. The prediction of the 'non-exceeding a bounding dose value' is not a prediction of a future evolution since many future evolutions may be compatible with such a bounding value. An absurd example may illustrate this type of bounding case prediction: it is certainly impossible to predict tomorrow's exact temperature, but most scientists (and laymen) will agree that one can predict that the average temperature in Europe over the next 10,000 years will not exceed 50 °C. If this type of information is sufficient for a given purpose, a credible prediction can be made, despite the open and empirical character of the system. Further, this is a strong prediction, since substantial scientific understanding and data are the foundation of the assessment of this bounding case. Certainly, such a prediction is not free of potential error. A large asteroid may strike the earth, invalidating the 50 °C bounding value. Applied to the case of a nuclear waste repository, one can try to do strong prediction of a bounding dose value for an evolution under a range of normal (no asteroid) expected circumstances.

Before applying this concept in the use the type of experimental data in Grambow et al. (2014), we must recall some very useful recommendations by Nordstrom (2012):

(1) Model computations must be explicable to non-modelers. For example explaining geochemical model computations to non-modelers and especially to non-technical audiences is a test of how well the modeler understands the computations.
(2) Nordstrom emphasized that the main conclusions or arguments of a complex computational simulation should be reproducible by a parallel and simpler 'back-of-the-envelope' calculation.

We emphasize that Nordstrom's recommendations are even more important when deriving the safety implications from experimental data, since safety is the main issue with the public.

2.4. From laboratory data to safety assessment

2.4.1. Identification of subsystems

We may divide the overall system of nuclear waste disposal in clayrock into subsystems that represent certain barrier function, for which one can try to develop strong bounding-case predictions based on physical and chemical laws and well known boundary conditions Individual subsystems can be described in a sufficiently simple manner allowing 'back-of-the-envelope' calculations as suggested by Nordstrom (2012). Subsystems can represent parts of the real system, or they can represent bounding cases (worst case conditions…) for radionuclide behavior. To illustrate this approach with an example: suppose, one would have waste forms dissolving so slow in groundwater that radionuclide release would not cause health risk, it might be sufficient to describe the subsystem "waste form dissolution" with sufficient scientific foundation to demonstrate compliance of the repository with regulatory constraints. This concept may further be discussed with some more detailed examples.

2.4.2. Subsystem "solubility in groundwater close to waste"

For example: a first subsystem may be the groundwater or the pore water in the vicinity of the waste, and its subsequent contamination by radionuclides when the canister is breached. The degree of potential contamination will depend on detailed scenarios of waste/water interactions, the time of container failure, the distance of the water from the waste and the time after disposal. Maximum levels of contamination can be predicted by predicting the solubility in groundwater. Knowing solubility constraints and the water transport rate, one could then determine a bounding constraint for a maximum flux of radionuclides at a given position in the isolation system. Solubility constraints are independent of time, except if the geochemical conditions change. Hence, taking the terminology of Nordstrom, prediction of solubility controlled transport of radionuclides is a phenomenological prediction. Thermodynamically well-founded solubility predictions for given chemical boundary conditions are only strong predictions if the temperature, pressure, the nature of the solubility controlling solid phases and their composition and the solution concentrations of all complexing substances and corresponding stability constants are known, including those for organic species. Uncertainties in solution concentrations and stoichiometry of complexing substances and in the nature and the composition of solid phases mean that only bounding values can be predicted in a strong manner. In order to determine a bounding solubility value, one needs a bounding model for the nature of solution complexes including representative organic complexes, as well as of the solubility controlling solid. We may take as an example the solubility of radionuclides in the pore water in clayrock. Highest radionuclide concentrations are expected nearest the nuclear waste glass surface, but it is not the glass that is the solubility-controlling solid phase. Instead, solubility may be controlled by one or more secondary solid alteration phases, which form on the glass surface, often in an ill-defined amorphous state. A theoretical assessment of empirical solubility constraints of actinides at the glass/water interface is given by Rai et al. (2011) (Rai et al., 2011) for pH and redox conditions, is also relevant for clayrock pore water compositions.

For example for tetravalent actinides uncertainties due to unknown solubility controlling phases may lead to uncertainties in maximum degrees of water contamination of many orders of magnitude. Normally, in a series of potentially solubility controlling phases, the phases with lowest solubility should control solution concentrations. But this is only true if the solubility controlling phase forms rapidly. For example crystalline oxides like PuO_2 have much lower solubility (are more stable) than amorphous hydrated oxides like PuO_2(am), but precipitation of the crystalline oxide is very slow. Hydrous amorphous phases can then be used to assess bounding solubility constraints. In the pH range of clay pore water and its potential modification by glass or steel corrosion (pH 7–9) the solubility of $Pu(OH)_4$ is about 10^{-9} M. In the literature, examples are given where solution concentrations of radionuclides are higher than solubility values due to colloid formation. This is not possible in clayrock: colloids may form, but they cannot be transported since the pore space acts as a very effective filter. Hence predictions of thermodynamic solubility of fast-forming phases like amorphous hydrous oxides, hydroxides or carbonates in clayrock pore water can be considered as strong predictions of a bounding maximum solution concentrations of contaminants.

2.4.3. The subsystem "sorption of radionuclides on clay rock" and the problem of non-uniqueness of models

Another subsystem is the sorption of radionuclides on clayrock surfaces. Once released from the emplaced waste, the most toxic radionuclides, the actinides, contaminate the contacting clay pore water, the maximum concentration possible being given by the solubility constraints discussed above. The dissolved radionuclide species will diffuse away from the waste and enter into contact with hitherto non-contaminated clayrock surfaces. A smaller or larger fraction of these radionuclides will then interact with the clayrock surface of the pore space and become strongly attached to these surfaces so that they become retarded in their migration to the biosphere, in most cases to an extent that radioactive decay occurs well before the radionuclide in question reaches the biosphere. Large sorption databases have been created involving various radionuclides in contact with a clay powder or clay blocks in the presence of clay pore-water.

Sorption models are non-unique. Different models like the K_d model or various models for surface complexation and ion exchange are frequently used. A key hypothesis is that there is a thermodynamic equilibrium between the concentrations of radionuclides sorbed on the solid and the concentrations of the same radionuclide in the contacting aqueous solution. This idea of equilibrium implies reversible exchange, a hypothesis which is not always tested and which is not always true since there may be irreversible entrapment of radionuclides on the solid, for example of Cs on frayed edge sites of illite (Poinssot et al., 1999). If there is irreversible fixation of radionuclides on the solid, there will be a smaller quantity of radionuclides available for migration to the biosphere. Hence, the non-consideration of irreversible entrapment allows for a bounding case calculation, providing as a result a quantity of migrating radionuclides higher than the real case. Nordstrom stated that predictions of complex events are ambiguous because of model non-uniqueness. However, this statement is only correct if the wrong model is used. Since various sorption models are correct (describe correctly the observations for a given frame of conditions), model non-uniqueness does not necessarily diminish the credibility in terms of a strong prediction. As long as a given model describes sorption in agreement with the experimental data quantitatively and independent of time one may use it for strong predictions of bounding effects. Sorption models may vary largely in their range of operational validity. A phenomenological K_d model may only be able to describe sorption for disposal of nuclear waste if the conditions of the disposal system are identical to the set of experimental conditions used to determine this K_d, while a model based on mechanistic description of surface complexation and ion exchange may be able to describe the variation of sorption as a consequence of the variation of initial conditions. A K_d model may become incorrect if mineral or water composition changes, while a surface complexation model may be able to capture such differences. In some cases, molecular modeling and spectroscopic characterization supports the development of an understanding of the formation of surface complexes. However, surface complexation models require many more parameters and therefore are much harder to "validate", even though these may apply to broader conditions.

A geological host formation like a clay formation of hundreds of meters varies little with time but shows some spatial variability. Hence, it is sufficient to measure the K_d in a sufficiently large quantity of statistically representative samples. In the near field of the waste, this approach may not be feasible, and a full surface complexation approach may be necessary, since chemistry of pore waters may vary largely due to release of waste package materials (i.e., organic).

2.4.4. Subsystem "water and radionuclide movement in clay rock"

One can predict for clayrock and for natural hydraulic gradients of Callovo Oxfordian clay that diffusion dominates over advective water flow. Taking the French clayrock of the proposed disposal site as an example, the ages of water in the formation above the Callovo-Oxfordian clay layer are dated to be between 300,000 and 1.5 million years (Fourré et al., 2011). To know a maximum value for the amount of radionuclides that can be transported by pore water in clayrock, one needs to know the radionuclide concentration at the source (potentially solubility controlled), the distribution ratio (K_d) of the radionuclide between the clayrock and the pore water, the diffusion coefficient and accessible porosity. This means that we need to couple the subsystem "solubility" with the subsystem "sorption" and both with subsystem "diffusion". The minimum time necessary (the "breakthrough time") for radionuclides to migrate from the waste via the clayrock to overlaying geological formations, were calculated in paper I for different radionuclides assuming diffusion via the shortest pathway (70 m): 250,000 years for the fastest moving nuclides (iodine-129) up to complete retention prior to radioactive decay for uranium, the transuranic elements and for cesium-135. Even large uncertainties in retention parameters do not change these findings. For example for iodine-129 transport through clay the use of the experimentally observed K_d values would allow one to calculate breakthrough times of about 500,000 years. Due to uncertainties on this K_d value one can define a bounding case value a K_d of zero leading to the previously proposed 250,000 years for breakthrough. Considering that iodine is the fastest moving radionuclide we can conclude that for times shorter than this value all radionuclides of the waste remain confined in the 140 m thick Callovo-Oxfordian clay formation, about 400 m beneath the earth surface.

2.5. Safety assessment

What do the model estimates of radionuclide behavior in subsystems of the disposal system tell us about the safety of a future repository in clayrock? Whether, when and in which quantity (is it safety relevant?) radionuclides from the waste may migrate toward the biosphere? Does the reasoning based on the data and models in the context of subsystems of the disposal system provide already a procedure to evaluate the effectiveness of nuclear waste isolation systems in preventing adverse radiological effects to present and future human beings and their environments?

The knowledge collected to allow long term predictions respond to a question of the type "what if?". Answers are always given for a given set of boundary conditions and scenarios. Safety relevant phenomena and boundary conditions of natural, human and waste/repository origin must be studied all together in safety assessments. Safety assessment, the quantification of radiation dose and risks that may arise from the disposal facility, is an integral part of a general safety case that has to be made in order to demonstrate safety of any geological disposal concept for radioactive waste: "*The safety case is the collection of scientific, technical, administrative and managerial arguments and evidence in support of the safety of a disposal facility, covering the suitability of the site and the design, construction and operation of the facility, the assessment of radiation risks and assurance of the adequacy and quality of all of the safety related work associated with the disposal facility*" (IAEA, 2012).

2.5.1. Safety assessment approaches

The risk assessment approach combines classical risk analyses (events leading to release of radionuclides multiplied by the probability of the event) with systems analysis (description of processes leading to a dose to humans). Probabilistic systems analyses (PSA) is used in some cases to represent combined natural and engineered barrier systems whose parameters are not entirely characterized (Ewing et al., 1999). PSA includes 6 essential steps: (1) identify performance measures, (2) characterize the way how the system affects performance, (3) build mathematical models describing performance, (4) quantify uncertainties of model parameters by probability distribution functions, (5) propagate uncertainty through the system (both steps are often neglected) and (6) compare performance with the real system. The latter step cannot be conducted for the necessary long time intervals. Probabilities can be assigned explicitly e.g., by probability density functions or implicitly through the selection of likely and less likely scenarios. However, if probabilities of expected behavior are not known, probabilistic PA becomes meaningless (Konikow and

Ewing, 1999). Probabilities are essentially obtained using available data (always limited), expert views (usually varying widely), and guess work (mostly). If the probability distribution functions are too broad, we get risk dilution and if they are too narrow, safety assessment can also be done in a deterministic manner based on expected (best estimate) evolution of the repository system or on worst-case scenarios.

Assessment may be divided in the analyses of scenarios (how are containment barriers breached?...) and of consequence (which dose?...). Scenario analyses determines systematically (NEA/OECD, 2001) by which chain of features, events and processes (FEPs) is it possible that radionuclides may migrate into the environment. The analyses of safety functions over time concern any barrier against water circulation including the geological environment and the disposal architecture, any barrier that immobilizes radionuclides in packages and containers in the disposal cells, and any barrier that delays and reduces the migration of radionuclides that were released outside the boundary of the disposal cell.

One needs to consider the groundwater access to the waste (*when?*, *how much?*, *vapor or liquid?*), and the waste matrix (*thermal effects*, *which rate of corrosion of the matrix in water?*), the evolution of the engineered barriers in the near-field (*are all voids filled to avoid accumulation of large gas or water volumes?*, *is there retention of radionuclides on engineered barrier materials?*, *what are the physical and chemical interactions at materials interfaces?*), the transport of solutes (*diffusion or advection control?*, *which impact of preexisting fractures and fractures created by repository construction?*, *which variation of hydrodynamic properties in space?*), the geology (*what is the frequency of tectonic events?*, *which geomechanical properties?*), the behavior of radionuclides (*which radionuclides are released?*, *which radionuclides are retained by solubility constraints and sorption?*) and the transfer to the biosphere, considering the impact of natural phenomena, human activities and the effect of the waste on the repository.

2.5.2. Performance indicators: radiotoxicity, risk, dose

There are various performance indicators that can be used to assess the effectiveness of a geological disposal concept to protect future generations against radiological risks. There is a vast literature that compares the radiotoxicity of the radionuclide inventories of the waste with the toxicity of the ore body that was used to generate the corresponding nuclear fuel. With reprocessing it takes some 20,000 years, without reprocessing some millions of years, until toxicity decreases to that of the ore body (US Congress, 1985). But toxicity inventories are not a measure of risk since they are not related to exposure scenarios. For a given exposure scenario it is common to distinguish predicted risk and predicted dose (in dose predictions there is no reference to the probability to be exposed to a given dose). Dose thresholds are given in mSv year^{-1} for potential exposure by ingestion or external irradiation. Recently, for the exposure from a geological disposal site a dose limit of 0.3 mSv year^{-1} and a risk value of 10^{-5} year^{-1} were recommended as "planned exposure" (Weiss et al., 2013).

Radiological dose thresholds are in some sense also risk thresholds as there is a conversion factor of 5.5×10^{-5} cancer Sv^{-1} (ICRP, 2007) between dose and risk of cancer using the typical linear-no-threshold LNT hypothesis. Only if the probability of a given exposure scenario is also taken into account, can one do a risk assessment.

2.5.3. A short non-exhaustive history of assessments

As early as 1979, there was the belief that for "reasonable" isolation systems expected doses would be lower than natural background levels (Burkholder, 1979). In the European Community, performance assessment started in the early 1980s with examples being the PAGIS project (1982–1989), PACOMA (1989–1991), EVEREST (1992–1996) and SPA (1996–1999) using a common methodology applied to repositories in clay, granite and salt, considering data and process uncertainties by sensitivity analyses and uncertainties in the future evolution of the geological isolation system by scenario analyses (Storck, 2000). In the PAGIS project for radionuclide inventories for 30 years of nuclear power generation, calculated dose rates were in all cases lower than national regulatory limits. Resulting doses calculated in the PACOMA project on alpha-bearing waste were not much lower even though radionuclide inventories were orders of magnitude lower, indicating that there is no direct link between radionuclide inventory and expected dose. The SPA project has shown that differences between calculations by different organizations for a given repository type, such as granite, can easily amount to more than one order of magnitude, corresponding to different hypotheses taken regarding container failure rates and groundwater travel times. A much higher actinide inventory in a deep repository does not lead to higher dose rates since flux to the biosphere is controlled by solubility constraints at the waste surface independent of actinide inventory. Hypotheses for transfer to biosphere are important: water taken from wells gives higher doses than from rivers due to dilution effects, the food chain is also important: eating fish gives higher doses than drinking the water. Comparing various international approaches the EVEREST project showed large effects of conceptual and model uncertainties on the final results (Orres et al., 1997).

The recent European Community Project PAMINA brought together 27 organizations from ten European countries (Bailey et al., 2011) and studied the impact of uncertainties on the safety case (Galson and Bailey, 2012). Uncertainties were classified depending on whether they are model, data or scenario related, all of which can be of random or epistemic (knowledge-based) nature. Some uncertainties are not important to safety since safety is controlled by other processes or since the probability of an event is extremely low. One can reduce uncertainties in some cases by adding additional barriers. Other uncertainties need to be included explicitly in the analyses or one needs to develop a bounding case and demonstrate that safety remains acceptable.

Safety assessment is now a routine exercise used at various stages in the development of repository projects, to compare alternatives: designs, layouts, materials, geological environments and repository sites. It offers a means of obtaining an overall view of the robustness of a given disposal concepts. The quantitative results from safety assessment are exceedingly important for demonstration of compliance with regulations, but the principal role of safety analyses is not so much to obtain the numerical results (dose, risk) *but the identification of key factors influencing safety*.

2.5.4. Role of data in safety analysis

If we conclude, as previously discussed, from the type of data and models presented by paper I (Grambow et al., 2014) that it takes more than 250,000 years for transport of the most mobile radionuclides across the Callovo-Oxfordian clayrock barrier, does this mean, if the waste is placed in this geological formation, that we can already demonstrate that the biosphere is protected from the nuclear waste for at least this time period? Certainly, these results are an indicator for safety or a piece of evidence. But we can easily imagine scenarios with faster radionuclide transport. Mobile anionic radionuclides may migrate through plugs and seals or through damaged clayrock if these are not tight. Large gas pressures generated for example by anoxic iron container corrosion or by radiolysis may create new pathways for groundwater. These alternative evolution scenarios are typically studied one by one, whether or not these possibilities are realistic. This leads to studies on the mechanism how fissures form and how they heal, for example during excavation. Large research projects (Johnson et al., 2013; Shaw, 2013) deal with these questions, but this subject is outside of the scope of the present communication. Also external forces might compromise the isolation system such as earth quakes (one can select geological formations with a low probability of seismic events) and mineral exploitation might occur or geothermal resources might be looked for in the future (one has to look for sites with little potential as an economic or energy resource). We will discuss below that even societal-based scenarios can have an important impact.

Another case: the results show that glass is an important barrier against radionuclide release also in compact clayrock environments. Times for complete alteration of more than 200,000 years should then also be valid under such conditions if the radioactive glass is immersed in clay pore-water. But the question of the scenario of the temporality of water ingress to the waste is also important. We know that glass dissolution rates in fast flowing water are much higher, but the scenario of fast water flow is incompatible with the compact low permeability (see Table 2 in paper I) of the clayrock. To recall from (Grambow et al., 2014): pressures of tens to hundreds bars are required in order to move just a few milliliters of water through the rock. Also one needs to develop a scenario on how water will come into contact with the nuclear waste glass. For the first thousands of years, there will be no water access to the glass and hence, no glass corrosion and no radionuclide release since the container is expected to remain intact. The container may fail only by corrosion combined with geomechanical pressure, and this process may take thousands of years. Indeed slow iron corrosion rates even in presence or microorganism under repository conditions have been confirmed, among many other data from other research groups, by the S experiment (steel corrosion) in (Grambow et al., 2014). But even after container failure, water may still have difficulties to enter into the container since, due to anoxic corrosion of steel containers, an overpressure by hydrogen gas is produced, potentially blocking the inflow of water to the glass inside of the corroding container. Under such conditions, glass corrosion occurs under vapor phase conditions. The present data are not directly applicable to this scenario, but previous data (Abrajano et al., 1989; Bates et al., 1984; Gong et al., 1998) show that corrosion continues in vapor and complementary research is underway to study the consequences of this scenario, as a function of relative humidity and temperature and first results show that corrosion rates in vapor are at least as fast (if not faster) as those in water (Neeway et al., 2012).

3. Safety and society

One of the astonishing results of more than 30 years of safety analyses is that despite important scientific progress and work on stakeholder confidence (OECD/NEA, 2013), the public confidence in the results have not significantly increased. If a repository concept is convincing, as in Sweden or Finland, where the safety relays strongly on the very robust copper container, there is no major problem with safety analyses. Indeed, the subsystem "container" carries in these countries a large part of the burden of proof for repository safety for hundreds of thousands of years.

Safety assessment is today commonly considered by research organizations, agencies for nuclear waste management and by technical support organizations for regulators as a purely technical issue of compliance with respect to regulatory criteria. Social sciences are rarely, if ever, involved. This separation ignores that safety and risk are societal concepts, that regulatory criteria are related to social choice and in the absence of a proper consideration of societal issues the safety assessment suffers from a lack of compelling credibility. In this sense, the OECD/NEA Forum on Stakeholder Confidence, argues that safety in the broader civil-society sense means also "feeling safe" and that the safety case should serve as a link in the system of actors comprising the implementer, the technical regulators, specialist groups in various advisory roles and the public (Pescatore, 2013). The public debate on nuclear waste is a typical example of the fact that risks are social constructions with conflicting views on the definition of risk (Beck, 2007). Even though most safety analyses have shown that there is only a small risk, opposition to disposal remains strong. Often this problem is presented as a problem of communication, or of a difference between risk and risk perception. Indeed, despite the fact that geological disposal provides an additional barrier (the geological barrier) against radiological exposure of populations, when compared to current interim storage of the waste at the earth's surface, the perceived risk of geological disposal may still be higher since disposal may be perceived as an abandonment of the wastes with no further possibility of control. A detailed discussion of the issues involved (attitudes and beliefs (Sjöberg, 2004), cultural cognition of environmental risks (Kahan et al., 2011) or historical and cultural of hazards to territories destined for disposal) is outside of the scope of the present paper. Only two questions related to the significance of our experimental data and their interpretation in the context of safety are discussed in the following: (1) What happens to the radionuclide migration scenarios if future actors do not behave as planned? (2) What is the societal meaning for safety of the calculated very long migration times?

3.1. What happens to the radionuclide migration scenarios if future actors do not behave as planned?

The impact of human activity on repository safety is often considered as the "human intrusion scenario" that is actions that compromise the isolation system by direct or inadvertent actions, such as the exploration for mineral or geothermal resources. There is also much debate on the needs of long-term monitoring or as to whether humans will preserve knowledge of the location and characteristic of the geologic repository. These questions are dealt with in the safety case in a qualitative manner without reference to quantitative performance criteria (Pescatore, 2013). Not considered at all is the inverse case: what happens, if humans do not act at all as planned and abandon the disposal site before closing it permanently?

Let us take another look at the scenario of migration of radionuclides from the waste through the clayrock to the biosphere. The very long isolation time of more than 250,000 year for the fastest moving anions is only valid if solute diffusion through the clayrock is the principal transport mechanism. This requires that plugs and seals be tight and that galleries are backfilled all the way to the entry shaft from the surface down to repository level in a manner so as to leave only very small void spaces. The just described scenario requires human action: after waste emplacement, particularly in those cases that provide for reversibility after some hundreds of years, future decision makers will have to decide to mobilize large financial, human and material resources to close the site correctly. However, if the society has already lived for more than hundred years with an open and reversible disposal site, what will motivate decision makers to invest billions of euros? In the US, there is a similar situation. The license application filed with the NRC takes the presence of Ti (with 2% palladium) drip shields into consideration. These drip shields figure prominently in the safety analysis, but there is no evidence (not even a prototype) that the 10s of billions of dollars will be spent to install them. One may want to use the financial resources for the ever pressing, urgent needs to win elections? This leads us to suggest another scenario that merits a careful technical analysis: the abandonment of backfilling and closing of the disposal site. One may only speculate about the consequence since a detailed assessment is missing: it may be that galleries will become very difficult to access with time, fissures may become larger and water from overlaying geological formations may eventually infiltrate. The migration distance of about 70 m for radionuclides from the confined location of waste emplaced in the center of the Callovo-Oxfordian clayrock formation to the overlaying geological strata with faster ground water circulation may then not be the preferential migration path from the waste to the environment, but the path way from the waste through plugs and seals toward the galleries. Still, many tens of thousands of years of isolation are probably to be expected for the fastest moving anionic radionuclides, as well as complete isolation of actinides until their decay, but it would have to be confirmed whether the very large isolation times of >250,000 year remain still realistic.

3.2. What is the societal meaning for safety of the very long migration times?

Predictions for hundreds of thousands of years are unprecedented in human technological history. Can one reduce the time for which a quantitative evaluation is necessary? The US-EPA requested for WIPP predictions that shall be quantitative for the first 10,000 years and qualitative thereafter (US EPA, 1985). The US Board of Radioactive Waste Management stated (National Research Council, 1990) that predicting "accurately the response of a complex mass of rock and groundwater as it reacts over thousands of years to the insertion of highly radioactive materials is not possible". (Ramspott, 1993) stated that there is no technical basis for setting a time limit of 10,000 years on the regulated performance of a nuclear waste repository. Total system performance assessment (TSPA) of the Yucca Mountain repository project gives maximum doses to public well beyond 10,000 year, actually stretching to one million years. Already in 1999 a review panel (Peer Review Panel, 1999) stated that it is unlikely that the TSPA viability assessment, taken as a whole, describes the long-term probable behavior of the proposed repository. Also the international joint evaluation (NEA-IAEA, 2001) stated that while compliance to performance criteria in the 10,000 year period was attained, work should continue beyond this regulatory period. In 2004 the D.C. Circuit Court of Appeals ruled that the 10,000-year compliance period was not "based upon and consistent with" the recommendation of the National Academy of Science that compliance be assessed at the time of maximum risk, within the period of geologic stability of the site. Few years later Yucca Mountain was abandoned as repository project.

But still we are confronted with the question whether risk prediction has any meaning for hundreds of thousands of years. The physical concept of time, more or less in the sense of Newton as a time in which things happen, serves as a universal temporal framework to quantify and to communicate the risk associated with the future release of radionuclides to future man. The physicists, chemists and geologists working on geological disposal projects are convinced that decisions on disposal should be made on the basis of this concept of time. In the tradition of safety analyses no distinction is made between the significance of exposure risks which occur early or late in the disposal time. For example, even if safety analyses or the above described "back of the envelope calculation" shows that after repository closure there is essentially zero risk for the first tens or even hundred-thousand years, waste management and technical support (TSO) organizations tend to focus on the maximum dose that may occur after hundreds of thousands of years. The reason for the indifference to the times for risks to occur is probably that the time in question is so long that it is meaning for today's decisions remains unclear. The citizen is not interested in predictions for millions of years. Risks are perceived by civil-society as typically related to a time scale for a few months to years. The mere fact that one is obliged to assess performance for such a long time increases the perceived risk.

The concept of "risk" implies always a projection into future, and it cannot be separated from the intrinsic vulnerability of man, beyond our own existence. The temporality of risk refers on the one hand to physical time (which may extend to millions of years) and societal time. The first temporality provides the frame for the capacity to predict and the second governs the public's concerns. Decisions may take both concepts of time into account since these are not concepts between which we must choose. Both perspectives exist in parallel and must be addressed. Although the ability to predict and to understand predictions is limited, responsibility for safety is not. To the degree to which we are able to predict risk, we have the duty to provide protection for future generations. Radioactive waste disposal risk assessment pushes the conflict between physical and human temporality to an extreme level, deprived of any unifying horizon.

4. Conclusions

Despite large public interest, there is rather little effort to communicate the scientific results gained in the last 30 years on nuclear waste disposal so that these results are still very poorly known among non-technical stakeholders. Very few scientists actually understand how their results from experiments and field investigations will be used in a safety assessment. An effort has been made in the papers I and II to overcome this situation.

The experimental simulation of repository performance by percolation tests (paper I) has provided important indicators for the safety for clayrock. Yet, due to the complexity of interlinked long-term processes operating in the barrier system of a nuclear waste repository, the translation of laboratory observations to conclusions on safety remains difficult to understand to stakeholders not directly involved in the analyses. Simplified "back of the envelope" models for subsystems indicate safety for more than hundreds of thousands of years, but for whatever degree of sophistication, models can only partially be validated. Strong (credible) predictions for hundreds of thousands of years of the detailed (quantitative) evolution of the engineered barrier system in its deep geological surrounding are not feasible, neither today nor in the near future. Instead, only bounding case predictions (upper limits of impacts) can be made in credible manner. Principal result of such assessments is not the compliance to a numerical performance indicator (dose to human beings living some hundreds of thousands of years from now...) but the degree of confidence created. Repository safety is not a purely technical but as well a societal issue ("feeling safe"). As a consequence, there is no direct link between increased scientific understanding of long term risks and a public position on nuclear waste disposal. Risks framed in physical temporality over many thousands of years needs to be translated in the context of societal temporalities of public's concerns and decision making. Social sciences need to be more strongly involved into safety assessment since safety it not purely technical, but as well it is societal.

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References

Abrajano Jr., T.A., Bates, J.K., Mazer, J.J., 1989. Aqueous corrosion of natural and nuclear waste glasses II. Mechanisms of vapor hydration of nuclear waste glasses. J. Non-Cryst. Solids 108, 269–288.

Alexander, D.H., Van Luik, A.E., 1990. Natural Analogue Studies Useful in Validating Regulatory Compliance Analyses. GEOVAL'90. Swedish Nuclear Power Inspectorate (SKI), OECD Nuclear Energy Agency, Stockholm, Sweden.

Altenhein, F.K., Lutze, W., Ewing, R.C., 1982. Long-term radioactivity release from solidified high-level waste. Part I: an approach to evaluating experimental data. In: Lutze, W. (Ed.), Scientific Basis for Nuclear Waste Management V. North-Holland, New York, NY, USA, pp. 45–56.

Altenhein, F.K., Lutze, W., Ewing, R.C., 1983. Long-term radioactivity release from solidified high-level waste. Part II: parametric study of waste form properties, temperature and time. In: Brookins, D.G. (Ed.), Scientific Basis for Nuclear Waste Management VI. North-Holland, New York, NY, USA, pp. 269–280.

Altenhein, F.K., Lutze, W., Ewing, R.C., 1984. Long-term radioactivity release from solidified high-level waste. III: the effect of canister lifetime. In: Wicks, G.G., Ross, W.A. (Eds.), Nuclear Waste Management. The American Ceramic Society, Columbus, OH, USA, pp. 636–644.

ANDRA, 2011. Rapport d'information sur la sûreté nucléaire et la radioprotection du Centre de stockage des déchets 2011 radioactifs de faible et moyenne activité de l'Aube.

Arimone, Y., Dinant, S., Alain André, Legrand, L., Alain Delaplanche, Vervialle, J.-P., Chino, P., 2010. Chapitre 2: Le stockage de déchets radioactifs contenant du tritium: impact des rejets sur l'environnement. In: ASN (Ed.), Livre Blanc du Tritium. ASN, Paris, pp. 157–171.

Bailey, L., Becker, D., Beuth, T., Capouet, M., Cormenzana, J.L., Cuñado, M., Galson, D.A., Griffault, L., Marivoet, J., Serres, C., 2011. European Handbook of the State-of-the-Art of Safety Assessments of Geological Repositories – Part 1. Pamina deliverable D.1.1.4. European Commission.

Bates, J.K., Seitz, M.G., Steindler, M.J., 1984. The relevance of vapor phase hydration aging to nuclear waste isolation. Nucl. Chem. Waste Manage. 5, 63–73.

Beck, U., 2007. Weltrisikogesellschaft: Auf der Suche nach der verlorenen Sicherheit [World at Risk: The Search for Lost Security]. Frankfurt am Main, Suhrkamp Verlag KG.

Blue Ribbon Commission on America's Nuclear Future, 2012. Report to the Secretary of Energy, Washington, D.C., p. 180.

Brandberg, F., Grundfelt, B., Höglund, L.O., Karlsson, F., Skagius, K., Smellie, J., 1993. Studies of Natural Analogues and Geological Systems. Their Importance to Performance Assessment. Swedish Nuclear Fuel and Waste Management Company, Stockholm, Sweden, p. 175.

Burkholder, H.C., 1979. The Waste Isolation Performance Assessment – A Status Report. Battelle Memorial Institute, Columbus, OH, USA, p. 17.

Crovisier, J.L., Honnorez, J., Eberhart, J.P., 1987. Dissolution of basaltic glass in seawater: mechanism and rate. Geochim. Cosmochim. Acta 51, 2977–2990.

Crovisier, J.-L., Advocat, T., Dussossoy, J.-L., 2003. Nature and role of natural alteration gels formed on the surface of ancient volcanic glasses (natural analogs of waste containment glasses). J. Nucl. Mater. 321, 91–109.

de Marsily, G., Combes, P., Goblet, P., 1992. Comment on "Ground-water models cannot be validated", by L.F. Konikow & J.D. Bredehoeft. Adv. Water Resour. 15, 367–369.

European Council, 2011. Directive 2011/70/EURATOM du Conseil du 19 juillet 2011 établissant un cadre communautaire pour la gestion responsable et sûre du combustible usé et des déchets radioactifs. J. Off. Union Eur.

Ewing, R.C., 1979. Natural glasses: analogues for radioactive waste forms. In: McCarthy, G.J. (Ed.), Scientific Basis for Nuclear Waste Management. Plenum Press, New-York, NY, USA, pp. 57–68.

Ewing, R.C., Jercinovic, M.J., 1987. Natural analogues: their application to the prediction of the long-term behavior of nuclear waste forms. In: Bates, J.K., Seefeldt, W.B. (Eds.), Scientific Basis for Nuclear Waste Management X. Materials Research Society, Pittsburgh, PA, USA, pp. 67–83.

Ewing, R.C., Tierney, M.S., Konikow, L.F., Rechard, R.P., 1999. Performance assessments of nuclear waste repositories: a dialogues on their value and limitations. Risk Anal. 19, 933–958.

Fourré, E., Jean-Baptiste, P., Dapoigny, A., Lavielle, B., Smith, T., Thomas, B., Vinsot, A., 2011. Dissolved helium distribution in the Oxfordian and Dogger deep aquifers of the Meuse/Haute-Marne area. Phys. Chem. Earth 36, 1511–1520.

Frugier, P., Gin, S., Lartigue, J.E., Deloule, E., 2006. SON68 glass dissolution kinetics at high reaction progress: mechanisms accounting for the residual alteration rate. Mater. Res. Soc. Symp. Proc. 932, 305–311.

Galson, D.A., Bailey, L.E.F., 2012. Performance assessment and the safety case: lessons from the European Commission PAMINA project. Mineral. Mag. 76, 3483–3489.

Glennan, S.S., 1996. Mechanisms and the nature of causation. Erkenntnis 44, 49–71.

Glynn, P.D., Plummer, L.N., 2005. Geochemistry and the understanding of groundwater systems. Hydrogeol. J. 13, 263–287.

Gong, W.L., Wang, L.M., Ewing, R.C., Vernaz, E., Bates, J.K., Ebert, W.L., 1998. Analytical electron microscopy study of surface layers formed on the French SON68 nuclear waste glass during vapor hydration at 200 °C. J. Nucl. Mater. 254, 249.

Grambow, B., Muller, R., 2001. First-order dissolution rate law and the role of surface layers in glass performance assessment. J. Nucl. Mater. 298, 112–124.

Grambow, B., Jercinovic, M.J., Ewing, R.C., Byers, C.D., 1986. Weathered basalt glass: a natural analogue for the effects of reaction progress on nuclear waste glass alteration. Mater. Res. Soc. Symp. Proc. 50, 263–272.

Grambow, B., Landesman, C., Ribet, S., 2014. Nuclear waste: I. Laboratory simulation of repository performance. Appl. Geochem. 49, 237–246.

Guala, F., 2005. The methodology of experimental economics. Cambridge University Press, Cambridge.

Howick, J., Glasziou, P., Aronson, J.K., 2013. Problems with using mechanisms to solve the problem of extrapolation. Theor. Med. Bioeth. 34, 275–291.

IAEA, 2009. Costing of Spent Nuclear Fuel Storage. Nuclear Energy Series. IAEA, Vienna, p. 69.

IAEA, 2012. The Safety Case and Safety Assessment for the Disposal of Radioactive Waste: Specific Safety Guide. IAEA Safety Standards Series, Vienna.

ICRP, 2007. The 2007 Recommendations of the International Commission on Radiological Protection. ICRP Publication 103. Annals of the ICRP 37.

Johnson, L., Sellin, P., Mayor, J.-C., Wieczorek, K., Gaus, I., Mente, M., 2013. PEBS: Long-term Performance of the Engineered Barrier System, EURADWASTE'13, European Commission..

Kahan, D.M., Jenkins-Smith, H.J., Braman, D., 2011. Cultural cognition of scientific consensus. J. Risk Res. 14, 147–174.

Konikow, L.F., Bredehoeft, J.D., 1992. Ground-water models cannot be validated. Adv. Water Resour. 15, 75–83.

Konikow, L.F., Ewing, R.C., 1999. Is a probabilistic performance assessment enough? Ground Water 37, 481–482.

Lutze, W., Malow, G., Ewing, R.C., Jercinovic, M.J., Keil, K., 1985. Alteration of basalt glasses: implications for modeling the long-term stability of nuclear waste glasses. Nature 314, 252–255.

Malow, G., Lutze, W., Ewing, R.C., 1984. Alteration effects and leach rates of basaltic glasses: implications for the long-term stability of nuclear waste form borosilicate glasses. J. Non-Cryst. Solids 67, 305–321.

Mazer, J.J., Bates, J.K., Bradley, C.R., Stevenson, C.M., 1992. Water diffusion in tektites: an example of the use of natural analogues in evaluating the long-term reaction of glass with water. J. Nucl. Mater. 190, 277–284.

McBride, B., Grace_Lucas, M., 2014. Radiation Alarm at New Mexico Nuclear Disposal Plant Spurs Air Shutoff. CNN.

Ministry of Trade and Industry (Finland), 1994. Nuclear Energy Act.

Montavon, G., Sabatié-Gogova, A., Ribet, S., Bailly, C., Bessaguet, N., Durce, D., Giffaut, E., Landesman, C., Grambow, B., 2014. Retention of iodide by the Callovo-Oxfordian formation: An experimental study. Applied Clay Science 87, 142–149.

National Research Council, 1990. Rethinking High-Level Radioactive Waste Disposal: A Position Statement of the Board on Radioactive Waste Management. National Academy Press, Washington.

NEA/OECD, 2001. Scenario Development Methods and Practice. An Evaluation based on the NEA Workshop on Scenario Development, Madrid, May 1999. Nuclear Energy Agency, Paris.

NEA-IAEA, 2001. Joint NEA-IAEA International Peer Review of the Yucca Mountain Site Characterization Project's Total System Performance Assessment Supporting the Site Recommendation Process.

Neeway, J., Abdelouas, A., Grambow, B., Schumacher, S., Martin, C., Kogawa, M., Utsunomiya, S., Gin, S., Frugier, P., 2012. Vapor hydration of SON68 glass from 90 °C to 200 °C: a kinetic study and corrosion products investigation. J. Non-Cryst. Solids 358, 2894–2905.

Nordstrom, D.K., 2012. Models, validation, and applied geochemistry: issues in science, communication, and philosophy. Appl. Geochem. 27, 1899–1919.

Nuclear Energy Agency, 2008. Moving Forward with Geological Disposal of Radioactive Waste. A Collective Statement by the NEA Radioactive Waste Management Committee (RWMC). In: OECD/NEA (Ed.), NEA/OECD Report. NEA/OECD, Paris, p. 23.

OECD/NEA, 1995. The Environmental and Ethical Basis of Geological Disposal of Long-lived Radioactive Wastes – A Collective Opinion of the Radioactive Waste Management Committee of the OECD/NEA.

OECD/NEA, 2013. Stakeholder Confidence in Radioactive Waste Management. In: Publishing, OECD/NEA (Ed.) NEA 6988.

Oreskes, N., Shrader-Frechette, K., Belitz, K., 1994. Verification, validation, and confirmation of numerical models in the earth sciences. Science 263, 641–646.

Orres, P.E.d., Marivoet, J., Martens, K.H., Prij, J.I., Cadelli, N., 1997. Sensitivity analysis in performance assessment of geological systems: the EVEREST project. In: McMenamin, T. (Ed.), Fourth European Conference on Management and Disposal of Radioactive Waste. European Commission, Luxembourg, pp. 663–680.

Peer Review Panel, 1999. FINAL REPORT TOTAL SYSTEM PERFORMANCE ASSESSMENT PEER REVIEW PANEL. Total System Performance Assessment (TSPA) Peer Review Panel for Predicting the Performance of a Repository at Yucca Mountain.

Pescatore, C., 2013. Safety, Safety Case and Society – Lessons from the Experience of the Forum on Stakeholder Confidence and other NEA Initiatives, 'Integration Group for the Safety Case' (IGSC) Symposium "The Safety Case for Deep Geological Disposal of Radioactive Waste: 2013 State-of-the-Art". OECD NEA RWMC, Paris, France.

Poinssot, C., Baeyens, B., Bradbury, M.H., 1999. Experimental and modelling studies of caesium sorption on illite. Geochim. Cosmochim. Acta 63, 3217.

Rai, D., Yui, M., Kitamura, A., Grambow, B., 2011. Thermodynamic approach for predicting actinide and rare earth concentrations in leachates from radioactive waste glasses. J. Solution Chem. 40, 1473–1504.

Ramspott, L.D., 1993. The Lack of Technical Basis for Requiring a Ten Thousand Year Prediction for Nuclear Waste Management. Lawrence Livermore National Laboratory, Livermore, USA, p. 9.

Republique Française, 2006. Loi de programme relative à la gestion durable des matières et déchets radioactifs.

Rosa, E.A., Tuler, S.P., Fischhoff, B., Webler, T., Friedman, S.M., Sclove, R.E., Shrader-Frechette, K., English, M.R., Kasperson, R.E., Goble, R.L., Leschine, T.M., Freudenburg, W., Chess, C., Perrow, C., Erikson, K., Short, J.F., 2010. Nuclear waste: knowledge waste? Science 329, 762–763.

Shaw, R., 2013. FORGE – Fate of Repository Gases, EURADWASTE'13, Vilnus, European Commission.
Sjöberg, L., 2004. Risk Perception as a Factor in Policy and Decision Making. Management of Uncertainty in Safety Cases and the Role of Risk – Workshop Proceedings. OECD–NEA, Stockholm, Sweden.
Steel, D., 2008. Across the Boundaries: Extrapolation in Biology and Social Science. Oxford University Press, Oxford.
Storck, R., 2000. Lessons learnt from various performance assessment exercises (from PAGIS to SPA) and their limitation. In: Davis, C. (Ed.), Euradwaste 1999 "Radioactive Waste Management Strategies and Issues". European Commission, pp. 194–203.
US Congress, 1985. Managing the Nation's Commercial High-Level Radioactive Waste. Office of Technology Assessment, p. 347.
US EPA, 1985. CFR Part 191, Federal Register. In: Environmental Protection Agency, (Ed.). vol. 50 (182), pp. 38066 (9/19/1985).
US House of Representatives, 1978. Interagency Review Group on Nuclear Waste Management. Oversight hearing before the Subcommittee on Energy and the Environment of the Committee on Interior and Insular Affairs, House of Representatives In: Environment, U.H.o.R.-S.o.E.a.T. (Ed.).
Van_Iseghem, P., Aertsens, M., Gin, S., Deneele, D., Grambow, B., McGrail, P., Strachan, D., Wicks, G., 2007. A Critical Evaluation of the Dissolution Mechanisms of High-level Waste Glasses in Conditions of Relevance for Geological Disposal (GLAMOR) – Final Report. Nuclear Science and Technology. European Commission, Brussels.
Wallace, H., 2010. Rock Solid? A Scientific Review of Geological Disposal of High-level Radioactive Waste. GeneWatch UK Consultancy Report.
Weiss, W., Larsson, C.-M., McKenney, C., Minon, J.-P., Mobbs, S., Schneider, T., Umeki, H., Hilden, W., Pescatore, C., Vesterlind, M., 2013. Radiological Protection in Geological Disposal of Long-lived Solid Radioactive Waste. Annals of the ICRP – ICRP PUBLICATION 122.
WIPP, 2010. Waste Isolation Pilot Plant Hazardous Waste Permit.