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Performance Assessment Methodologies in Application to Guide the Development of the Safety Case

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Expert judgement review and exercise (estimation of the solubility limit distributions for five elements in the near field in a repository in granite)

MILESTONE (M-N°:2.2.A.12)

Author(s):**R. Bolado, L. Duro, J.L. Cormenzana, M.A. Cuñado, M. Grivé & D.G. Bennett**

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Dissemination Level		
PU	Public	X
RE	Restricted to a group specified by the partners of the [PAMINA] project	
CO	Confidential, only for partners of the [PAMINA] project	

Foreword

The work presented in this report was developed within the Integrated Project PAMINA: **P**erformance **A**ssessment **M**ethodologies **IN** **A**pplication to Guide the Development of the Safety Case. This project is part of the Sixth Framework Programme of the European Commission. It brings together 25 organisations from ten European countries and one EC Joint Research Centre in order to improve and harmonise methodologies and tools for demonstrating the safety of deep geological disposal of long-lived radioactive waste for different waste types, repository designs and geological environments. The results will be of interest to national waste management organisations, regulators and lay stakeholders.

The work is organised in four Research and Technology Development Components (RTDCs) and one additional component dealing with knowledge management and dissemination of knowledge:

- In RTDC 1 the aim is to evaluate the state of the art of methodologies and approaches needed for assessing the safety of deep geological disposal, on the basis of comprehensive review of international practice. This work includes the identification of any deficiencies in methods and tools.
- In RTDC 2 the aim is to establish a framework and methodology for the treatment of uncertainty during PA and safety case development. Guidance on, and examples of, good practice will be provided on the communication and treatment of different types of uncertainty, spatial variability, the development of probabilistic safety assessment tools, and techniques for sensitivity and uncertainty analysis.
- In RTDC 3 the aim is to develop methodologies and tools for integrated PA for various geological disposal concepts. This work includes the development of PA scenarios, of the PA approach to gas migration processes, of the PA approach to radionuclide source term modelling, and of safety and performance indicators.
- In RTDC 4 the aim is to conduct several benchmark exercises on specific processes, in which quantitative comparisons are made between approaches that rely on simplifying assumptions and models, and those that rely on complex models that take into account a more complete process conceptualization in space and time.

The work presented in this report was performed in the scope of RTDC 2.

All PAMINA reports can be downloaded from http://www.ip-pamina.eu.

1. Introduction

Expert Judgement (EJ) has been used during roughly the last seventy years in different areas of science, technology, weather forecasting, strategic planning, economy and many other fields as a reasonable way to assess uncertainties about events and variables when the source of uncertainty is lack of knowledge (epistemic uncertainty). Since the design of the pioneering Delphi method, several structured protocols have been proposed and improved thanks to the experience acquired in many applications. Nuclear Safety has been an extremely fertile field for the application and improvement of these protocols. During the mid 1980's, researchers from Sandia National Laboratories (SNL) in collaboration with experts in the area of EJ developed the best known protocol to provide information in large scale risk studies, namely Probabilistic Safety Analyses (PSA) of Nuclear Power Plants (NPPs) and Performance Assessments (PA) of Spent Nuclear Fuel (SNF) and/or Radioactive High Level Waste (HLW) repositories, see Bonano et al. (1990) and Gorham-Bergeron et al. (1991). This protocol is described in Bolado et al. (2009) and is referred to as the SNL/NUREG-1150 protocol.

PAMINA Work Package 2.2 (WP2.2) has been dedicated to test and develop approaches for the treatment of uncertainties. Within WP2.2, task 2.2.A (topic 5) has dealt with the use of EJ for assigning PDFs. This activity has been divided in two tasks. The first one was a review of EJ literature, while the second one was an EJ application in the area of PA. The review of EJ literature, was done originally by JRC and peer reviewed by NDA, delivering as a result a final joint document, see Bolado et al. (2009).

This report is a summary of the second task, the EJ application case developed within PAMINA to assess the distributions that characterise the uncertainty about the solubility limit of five chemical elements (Ra, Sn, Se, U and Pu) under the expected conditions in the near field of the Spanish reference concept for SNF disposal in granite. In order to do this, a protocol was designed, Bolado (2008), based on the SNL/NUREG-1150 protocol, and applied during the second half of 2008. In the next pages this application is described in detail. In fact this report is the actual implementation of the last step of the protocol used: the documentation phase. This work has been done by JRC, ENRESA and AMPHOS 21 in collaboration with two experts.

2. The protocol applied

The protocol applied in this EJ exercise is an adaptation of the SNL/NUREG-1150 protocol to a small scale application and consists of the following steps

1. Selection of the project team.
2. Preparation of supporting material and definition of the questions to be studied.
3. Selection of experts.
4. Training sessions.
5. Refinement of the questions to be studied.
6. First individual work period.
7. Presentation of individual approaches adopted by the experts.
8. Second individual work period.
9. Elicitation sessions.
10. Analysis and aggregation of results.
11. Review.
12. Documentation.

These steps may be grouped into three major phases:

1. pre-process (steps 1 to 3, developed before the actual participation of the experts, where the key players are the organisation interested in getting the opinions of experts and the project team),
2. process (steps 4 to 9, where experts participate actively) and
3. post-process (steps 10 to 12, where the project team plays the key role, post-processing the opinions of experts and delivering the application final report).

The development of this protocol involves three meetings that must be attended by all experts and the project team: 1) the training sessions (step 4) and the refinement of the questions to be studied (step 5) are performed in the first meeting, which takes two days and involves the participation of the project team and all the experts simultaneously, 2) the session to describe individual approaches (step 7) requires again the concurrence of the project team and all the experts simultaneously in a second meeting, which takes one day, and 3) the elicitation sessions require a third meeting, which involves the concurrence of the project team and the experts, in this case one by one since opinions are elicited individually in this protocol. These sessions require one day per expert.

Three main innovations are introduced in this protocol with respect to the SNL/NUREG-1150 protocol: 1) The addition of extra training sessions, 2) the division of the individual work in two phases separated by a meeting where each expert shows the approach adopted to address the questions under study, and 3) the flexibility in the selection of techniques to aggregate the opinions of the different experts.

The following pages describe how the protocol was actually implemented to characterise the uncertainty about the solubility limit of five elements (Ra, Sn, Se, U and Pu) under the expected conditions in the near field of the Spanish reference concept for Spent Nuclear Fuel disposal in granite.

2.1 Selection of the project team

The project team consisted of one analyst and two generalists. The analyst has a wide experience in the application of expert judgement protocols in nuclear safety issues. The first generalist is a specialist in PA studies, familiar with both the development of PA models and with the probabilistic framework of the PA. The second one is an expert in radionuclide geochemistry that could herself have participated in this application as an expert, as in fact she has done in a similar study developed by NDA. This is not a frequent situation, but the project PAMINA has given us the possibility of counting on two generalists, which has actually improved the development of the whole process and the quality of the results obtained. The first one has been in charge of focusing on the questions under study from the point of view of the PA, while the second one has dealt with information sources and with the definition of the questions from a scientific point of view. Both of them have actively collaborated in other supporting activities.

The project team consists of the following persons:

- Analyst: Ricardo Bolado (IE, EC DG-JRC)
- Generalist 1: José Luis Cormenzana (Empresarios Agrupados)
- Generalist 2: Lara Duro (AMPHOS 21)

In addition to the project team, Miguel Ángel Cuñado (ENRESA) also participated in the project as a representative of the PA owner (ENRESA) and he provided key inputs particularly at some of the later steps of the protocol. In the early stages of the project, the project team also benefited from interactions with Jesús Alonso a former representative from ENRESA.

2.2 Preparation of supporting material and definition of the questions to be studied

Four documents were prepared during 2008 as supporting material for this exercise. The intention behind the generation of these reports was to provide experts key information about the exercise where they were going to participate. The first one is the description of the EJ protocol designed to be applied in this exercise; see Bolado (2008). The second one is a review of expert judgement methods and protocols; see Bolado et al. (2009). The third one is a description of the disposal system for which the parameter estimates are needed; see Cormenzana (2008). The last one (the Datapack) contains the scientific description of the problem to be solved and a collection of data / scientific literature concerning relevant information about the solubility of the chemical elements under study, see Duro (2008). These documents contain the most relevant information and training material for the first protocol meeting.

In parallel with the edition of these documents the generalists, interacting with the PA owner, made a decision about the chemical elements to be addressed in the EJ exercise and the actual questions to be posed to the experts. The following criteria were set in order to proceed with the chemical elements selection:

- The elements must be of interest for the Safety Case.
- The elements are expected to precipitate as solid phases under the conditions of interest in the assessment. This excludes those elements not likely to form solid phases, such as I or Cl, although they can present active isotopes of interest for the Safety Case.
- The list of elements whose solubility will be evaluated must include different behaviour with regards to the redox state of the system. This implies that both types of elements, redox-sensitive and non redox-sensitive, must be considered.
- The list of elements must be as comprehensive as possible in terms of redox states. This means that the selection of two elements presenting different redox states will prevail over the selection of two elements presenting the same redox state.
- The list of radioelements to consider must be short enough as to allow a proper expert judgement application within the time frame and resource constraints of the project.

On the basis of the criteria presented above, Ra, Sn, Se, U and Pu were selected for the assessment of their solubility limits.

After selecting the elements to analyse, the generalists proposed a preliminary version of the question to ask to the experts. This was the first version of the question posed to the experts:

‘*Provide probability distribution functions that represent the solubility of 5 different chemical elements (Selenium, Uranium, Plutonium, Tin and Radium) under near field conditions for*

- *the long term water compositions (≥10^5 years) presented in table 7.2 of Cormenzana (2008) (reproduced as table 2.2.1 of this report).*
- *a range of pH between 7.5 and 9.5 (long term pH ±1 pH unit).*
- *a range of redox potential between -360 and -180 mV*’.

Table 2.2.1 presents the expected evolution of bentonite porewater characteristics at the central point of the bentonite buffer in the reference disposal concept. Additionally, a H_2 gas pressure of about 2 MPa can build up in the near field as a result of canister materials corrosion under anaerobic conditions, and should be considered in the estimation of the solubilities. The evolution of pH, Eh (pe) and bentonite porewater composition shown in table 2.2.1 was determined without taking into account the development of any H_2 gas pressure. The effect of the H_2 gas pressure on near field conditions should be considered.

Although co-precipitation can control the concentration of some of the selected elements (Ra in particular), in the present exercise, the solubility will be assumed to be controlled by pure phases of each chemical element.

This original question definition was regarded as needing refinement with the collaboration of the experts participating in the exercise.

Table 2.2.1.- Expected evolution of the bentonite porewater characteristics at the central point of the bentonite buffer in the Spanish reference disposal concept in granite Cormenzana (2008).

Concentration (Mol/l)										pH	pe	T (ºC)
Time (y)	**HCO_3^-**	**Cl^-**	**SO_4^{-2}**	**Ca^{+2}**	**Mg^{+2}**	**Na^+**	**K^+**	**Fe^{+2}**	**SiO_2(aq)**			
1.000	1.75E-03	9.28E-02	2.67E-02	1.39E-02	1.38E-02	9.10E-02	8.26E-04	7.66E-06	6.96E-04	6.71	-2.830	66.88
2.500	2.09E-03	8.11E-02	3.22E-02	1.44E-02	1.38E-02	8.96E-02	8.27E-04	1.33E-05	5.61E-04	6.740	-2.770	58.24
5.000	2.55E-03	6.48E-02	3.96E-02	1.51E-02	1.37E-02	8.73E-02	8.27E-04	2.49E-05	4.25E-04	6.780	-2.700	48.48
10.000	3.21E-03	4.15E-02	4.38E-02	1.40E-02	1.21E-02	7.85E-02	7.78E-04	4.07E-05	3.37E-04	6.820	-2.650	41.10
25.000	4.48E-03	1.11E-02	1.23E-02	2.15E-03	1.67E-03	3.16E-02	3.19E-04	7.07E-06	2.84E-04	7.290	-3.210	36.00
50.000	5.14E-03	1.52E-03	1.31E-03	1.28E-04	8.15E-05	8.46E-03	8.49E-05	1.84E-07	2.85E-04	8.180	-4.460	36.00
100.000	5.28E-03	4.07E-04	3.02E-05	5.16E-05	2.88E-05	5.31E-03	5.37E-05	8.35E-08	2.83E-04	8.530	-5.120	35.74
500.000	5.28E-03	3.95E-04	1.57E-05	5.45E-05	3.06E-05	5.26E-03	5.53E-05	1.35E-07	2.43E-04	8.560	-5.140	31.02
1.000.000	5.27E-03	3.95E-04	1.57E-05	5.93E-05	3.36E-05	5.22E-03	5.77E-05	1.76E-07	2.27E-04	8.550	-5.090	29.99

2.3 Experts' selection

As it is widely known, experts qualified to participate in an EJ exercise must have the necessary knowledge and experience to perform the required assessments/estimations, must be willing and available to participate in the assessment and ought not to have important motivational biases. The main criteria to decide if a subject has the necessary knowledge and experience are: reputation, experimental experience, number and quality of scientific publications, awards received and balance of views. Additionally, it is always desirable to gather a set of experts with an as diverse as possible background. In particular, this EJ exercise requires the following specific knowledge from each expert:

- Deep knowledge about the general chemical behaviour of radium, tin, selenium, uranium and plutonium.
- Deep knowledge about the possible solid phases that the above-mentioned elements may form.
- Good general understanding about the use of solubility limits in PA studies.
- Capability to provide estimates in probabilistic terms for the quantities under study.

The target number of expert was 3, in accordance with normal practice in this area. The process started with a list of 8 experts, all of them fulfilling the knowledge aforementioned requirements. Experts working for national radioactive waste management agencies were excluded in order to avoid motivational biases. After that exclusion, the selection process was based essentially on availability. The experts eventually selected were:

- Dr. M. Grivé (AMPHOS 21), from now on referred to as expert 1
- Dr. D.G. Bennett (TerraSalus), from now on referred to as expert 2
- A third expert, from now on referred to as expert 3.

After a first contact and acceptance of contractual conditions, they were formally invited to participate in this exercise. Together with the invitation letter each expert was provided with the four supporting documents. They were also informed about the most likely dates for the three meetings considered in the protocol. Experts were given one month for examining the documentation before the first meeting, which was held on September 15th and 16th 2008. AMPHOS 21 (Spain) provided the meeting facilities for the whole exercise.

2.4 First meeting

The first meeting started with the welcome from generalist 2 (meeting host) to the entire group, and especially to the three experts. After that formality, the analyst gave a brief overview of the whole process and then gave the floor to generalist 1, who gave a presentation about the Spanish reference disposal concept in granite. This presentation included the following topics:

- A system description, focusing especially on the near field,
- A description of the expected repository evolution, stressing the bentonite saturation transient, the thermal transient and the evolution of bentonite porewater,
- A summary of the results obtained in the PA of the reference repository, focusing on the main contributors (radionuclides) to risk, and an analysis of the reasons for the importance of radionuclide solubility in PA results,

- The criteria to select the elements to study (already shown in section 2.2),
- A description of the way the PDFs obtained in this exercise could be used,
- And the preliminary question definition (already shown in section 2.2).

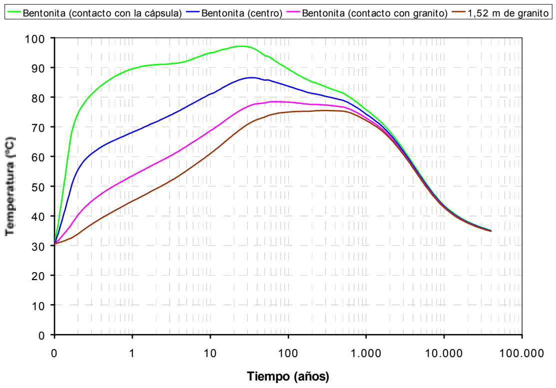

Figure 2.4.1.- Temperature evolution at different positions within the bentonite buffer (capsule/bentonite interface – green line, bentonite middle point – blue line, bentonite/granite interface – magenta line and 1, 52 m into the granite – brown line).

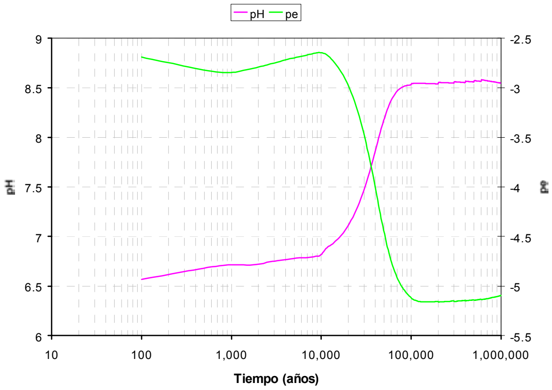

Figure 2.4.2.- Evolution of pH and Eh (pe) over time in the bentonite buffer.

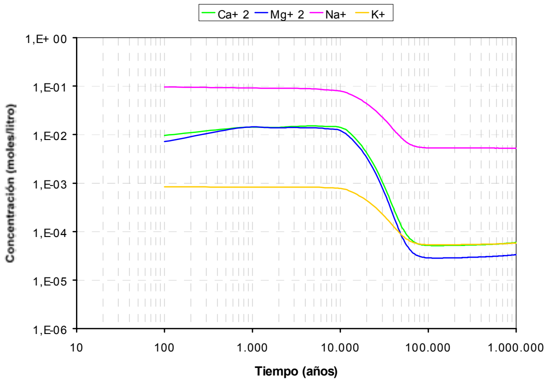

Figure 2.4.3.- Evolution of main cations' concentration (Mol/dm^3) over time in the bentonite buffer.

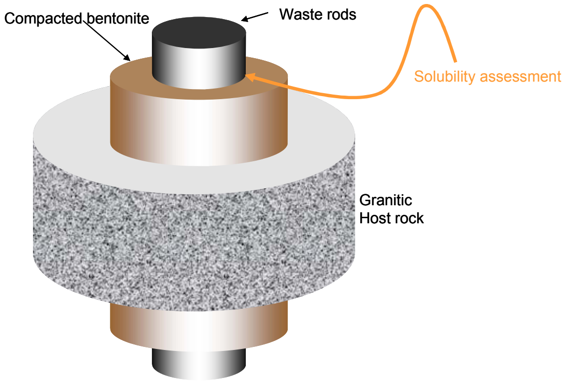

Figure 2.4.4.- Illustration of the near field. The solubility limits should be estimated at the interface between the iron canister and the bentonite buffer (orange arrow).

This presentation was very important for the experts to understand the system under study and the relevance of the proposed questions. After describing main features of the repository, Generalist 1 described in detail the chemical composition of the bentonite and the expected repository evolution. Special attention was paid to the thermal transient, see figure 2.4.1, showing that bentonite temperature never reaches 100 °C and that after 20,000 y (minimum canister duration) this transient is over (less than 10 °C above natural temperature at repository depth). Regarding the evolution of bentonite porewater characteristics, he showed that the transition between bentonite controlled conditions and granitic groundwater controlled conditions happens between 10,000 y and 100,000 y, see figures 2.4.2 and 2.4.3 and table 2.2.1, and he stressed that in this exercise the interest is focused on long term conditions (after 100,000 y).

Then, generalist 1 explained that a few highly soluble radionuclides dominate the risk associated with the reference system; while low solubility radionuclides are very efficiently retarded by repository barriers. Radionuclide transport through the bentonite is controlled by diffusion, and the radionuclide concentration in the canister cavity water is the boundary condition for transport at the bentonite innermost surface (contact surface between canister and bentonite). If the concentration of a given radionuclide is limited, this has a direct (large) impact on its transport through the bentonite; in fact radionuclide transport rates through the different barriers and doses in the biosphere are calculated in the PA to be proportional to the solubility limit.

Finally, in addition to explaining the criteria used to select the elements to study and showing the preliminary definition of the question, he showed how the PDFs eventually obtained could be used in a PA as a part of the Monte Carlo uncertainty propagation method to estimate the consequences and risk associated to the repository.

Most of the rest of the first day of this meeting was dedicated to training. The analyst started with an introduction to EJ, why it is needed in many areas of science and technology, including PA, the different roles played in an EJ exercise (analyst, generalists, experts and PA owner). Then he recalled the development of EJ protocols since the end of World War II and their main features. After that, he did a short review of key concepts in probability theory, with some incursions in statistics, which included

- Random experiments
- Dependent and independent events
- Probability axioms
- Main interpretations of probability (classic, frequentistic and Bayesian)
- Total probability theorem
- Bayes' theorem as a tool to update information
- Random variables (continuous and discrete), probability density functions, probability mass functions and cumulative distribution functions
- Different probability models of interest (Poisson, Bernouilli, binomial, geometric, negative binomial, uniform, normal, log-normal, exponential, Weibull, gamma, etc.) and main relations among them.

The next training sessions were dedicated to inform experts about basic issues related to cognitive biases and the potential consequences on their assessments, and to help them avoiding such problems. Experts were informed about the difficulties that human beings in general, and experts in particular, encounter when they have to deal with different sources of information and they have to make statements in terms of probability. They were also informed about the main simplifying strategies (heuristics) they usually adopt when they have to make judgements, the problems they find when they have to deal with statistical information and the main biases that this introduces in their judgements. Special attention was dedicated to the most widespread and feared bias: overconfidence. This training session was illustrated with many examples taken from the literature in the area of knowledge psychology, which helps to understand the origin of biases. At the end of this session, the analyst invited experts to participate next morning in a calibration test based on using general culture questions to see in practical terms the high risk of falling into overconfidence.

At the end of the first day, the original question definition, as posed by generalist 1 in the early morning was recalled. Experts were invited to think about this definition and all factors that should be taken into account in order to reach a consensus definition, accurate enough to pass the 'clairvoyant test' (see Bolado et al. (2009), chapter 5) and relevant to the PA.

The second day of the first meeting started with the calibration test. All meeting attendants, excluding the analyst, were given a paper containing a set of twelve general culture questions whose corresponding answers were numeric. They were asked to provide 90% probability intervals[1] as answers. They were reminded that the target was not to give a very narrow interval, but to catch within the interval limits the right answer with a high probability (0.90). The results of the calibration exercise were as expected. Most of the participants delivered excessively narrow intervals. The numbers of scores were 6, 4, 6, 5, 4 and 9. A well calibrated expert should have delivered at least 8 intervals containing the right answer. The probability that a well calibrated expert delivers 4 or more wrong intervals is approximately 2.5%; the probability of delivering 5 or more wrong intervals being well calibrated is less than 1%, which is a probability that brings the decision maker to reject the null hypothesis in common statistical hypothesis test. Broadly speaking, it could be said that experts were providing 50% rather than 90% probability intervals. This test was found useful by the participants to note the natural tendency to provide too narrow distribution functions and try to avoid it.

After this calibration exercise, generalist 2 gave a presentation that consisted of two parts, firstly she recalled some hypotheses that should be taken into account in the problem resolution, and secondly she introduced the Datapack.

In the first part of the presentation she stressed that the estimates to be provided by the experts regarding the solubility of the different radioelements should be made for a specific location: the interface between the waste canister and the bentonite buffer and for a time when the canister has already lost its isolating properties, see figure 2.4.4. Then she listed

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[1] A 90% probability interval is an interval that the probability that it contains the right answer is 0.90. In the case of a well calibrated expert, approximately 90% of the 90% probability intervals should contain the right answer.

the following additional hypotheses (to be added, after discussion, to the ones presented in the preliminary definition of the questions shown in section 2.2)

- The radionuclides are not limited at the source; they are controlled by the precipitation of secondary phases.
- Water from the granitic formation intrudes the bentonite and interacts with it and the carbon steel canister prior to contacting the waste.
- The solid phases most likely to form are those assumed to readily precipitate under the conditions of interest according to Ostwald's rule.
- No co-precipitation or formation of mixed solid phases will be considered.
- No kinetic constraints are considered, except those derived from the consideration of the formation of amorphous solid phases over the more crystalline ones.
- No microbial activity is considered.
- Corrosion products of the canister can be present in the system.
- The redox conditions are in the range reducing to anoxic.

She also stressed that, along the whole exercise, whenever participants refer to 'solubility of a given element or radioelement or radionuclide', they should understand it as the solubility of the sum of all active and non-active isotopes of the same chemical element, e.g. the parameter to assess via EJ will not be the solubility of ^{235}U, but the solubility of U, including all its isotopes.

The second part of the presentation was dedicated to an overview of the information contained in the Datapack. Firstly, a short description of each element was given, highlighting factors that may affect their respective solubilities and main sources of uncertainty. Records of contents of each element in natural waters were also shown in this part of the presentation. Secondly, a summary of experimental measures obtained for each element was given, which included experimental conditions. Thirdly, a summary of solubility limits used in different performance assessments was also presented. This analysis was split in two parts, a country-based analysis, and an element-based analysis. The country-based analysis included elements considered, PA conditions (pH, pe, temperature, consideration or not of co-precipitation, solid phases, etc.) and range of values used for each element (in most of the cases a best estimate or proposed value and a conservative one). The element-based analysis consisted of a set of graphs comparing the ranges used in different countries. One graph per element was also provided to show all the spread of solubility values obtained for all sources of information (experiments, natural waters and PA's). The last part of the presentation was dedicated to describing the contents of the five elements under study in rocks and minerals. This presentation and the supporting document was a very important source of information for the experts along the whole EJ exercise.

The presentation provided a good starting point to start discussing and refining the question definition, in order to develop a more precise and definitive statement accepted by all participants and interpreted by all in the same manner. All experts acknowledged the importance of reaching an accurate question definition and a common understanding. They also acknowledged the relevance of all the hypotheses proposed by the generalists when assessing the question under study and the need for debate about the problem. One by one, all hypotheses were discussed in a very interactive way.

This debate was followed by some further training sessions. Some training about heuristics and biases not finished on the first day was finished in the first training session of the second day. Then, the analyst informed experts about the main techniques to assess probabilities, paying special attention to the two most popular ones: the interval technique and the quantile technique. In the first case the analyst sets intervals, either closed or open, in the support of the parameter under study and asks the expert to provide the probability of such intervals. In the second case the analyst chooses a given number of probabilities and asks the expert to provide an estimate of the values that could be exceeded by the parameter under study with such probability.

The last session of the meeting was devoted to further discussion about the question definition. Eventually the following question definition was agreed:

‘*Provide probability distribution functions that represent the concentration (solubility) limit of 5 chemical elements (selenium, uranium, plutonium, tin and radium) under near field conditions for*’

- The long term Groundwater composition (time $\geq 10^5$ years) presented in table 2.2.1 in this report except for $[Fe]_{aq}$ and Eh.
- Calculate $[Fe]_{aq}$ and Eh according to the following assumptions:
    - pH range between 7 and 9.
    - Hydrostatic pressure around 50 atm.
    - $H_2(g)$ gas pressures of 20 atm (Fe_3O_4 is the product formed from iron corrosion) and 80 atm ($Fe(OH)_2$ is the product formed from iron corrosion).
- Temperature between 30 and 40°C.
- No microbial activity considered; neither sulphate nor carbonate reduction happens.
- FeSe / $FeSe_2$ might be formed.
- Calcite will be allowed to form if oversaturated
- No colloidal form is considered, only true dissolved elements should be considered.
- No co-precipitation of a given radionuclide with a major element is considered.

At the end of the meeting the project team members informed the experts of what was expected from them in the next two protocol steps (first individual work period and presentation of individual approaches adopted by experts). They were informed that the target of the first individual work period was to start studying the problem, designing a method to address it and identifying tools and sources of information to use (computational tools, databases, sets of data, publications, etc.). That was not the phase to perform the final analysis of the problem and prepare all needed material for the elicitation session. They were informed that they would be asked in the second meeting to present their approach to the problem and the tools they intended to use in order to solve it. They were also informed that the first individual work period was the right time to analyze the hypotheses considered so far and their implications, identify possible gaps in the question definition and alternative hypotheses of interest. Any doubt, any implicit hypothesis not explicitly mentioned in the first meeting should be brought for discussion in the second meeting. The project team told them that the main objective of the second project meeting was to confirm the final question definition to study, making sure that this definition was in agreement with ENRESA PA's objectives and that all participants, project team and experts, had a common understanding of the problem. Finally, experts were informed that the analyst would be fully available during the first individual work period to help them defining the space of calculations and for any other support in the area of Statistics. Generalist 2 would also be available to solve any question regarding sources of information gathered in the Datapack. The meeting finished after setting the date of the second meeting: October 14th, 2008.

2.5 Second meeting (question definition refinement)

Unfortunately, during the first individual work period expert 3 decided to abandon the project. Experts 1 and 2 did what was expected from them. Each expert adopted a strategy to solve the problem and both worked on the problem hypotheses in order to refine the question definition. Since the expected strategy adopted by each expert did not change substantially between the second meeting and the third meeting, both will be described in the section dedicated to the third meeting

Most of the second meeting was dedicated to discussing the question definition. All the phenomena involved were discussed again, as for example the expected H_2 pressure due to the generation of this gas via anoxic corrosion of the steel canister and the pressure relief through the bentonite when this magnitude exceeds 80 atm. The discussion helped setting definitively the $H_2(g)$ pressure in the range 20 to 80 atm. A range of temperature between 30 and 40°C, and a hydrostatic pressure of 50 atm were selected as representative of long term repository conditions. Due to the lack of reaction rates under these conditions, it was agreed to perform calculations at 25°C and 1 atm, after appropriate justification of the validity of this approximation. The discussion was also very much focused on the groundwater composition in the long term and how to fix the conditions in order to estimate the radionuclide solubilities. The debate also focused on the importance of properly identifying the solubility limiting phases, the expert judgement that this task involves, and the differences in estimates that disagreements in this area could produce. The use of activity corrections in computations was also addressed. The group agreed to use, after appropriate justification provided by experts in the next meeting, the extended Debye-Hückel (D-H) approach. Other hypotheses already set in the first meeting were simply confirmed, as for example the exclusion of co-precipitation, microbial activity and colloidal forms. Finally, the definitive question definition agreed by the whole group was the following one:

‘*Provide probability distribution functions (PDFs) that represent the concentration (solubility) limit of five different chemical elements (radium, tin, selenium, uranium and plutonium) under the following near field (iron canister / bentonite buffer interface) conditions* ’

- The radionuclides are not limited at the source (i.e., by the rate of spent fuel dissolution); they are controlled by the precipitation of secondary phases.
- T between 30-40°C and hydrostatic pressure of 50 atm, although calculations done at T= 25ºC and 1 atm (justify the validity of this approach).
- $H_2(g)$ pressure between 20 and 80 atm.
- I corrections: Extended D-H (justify the validity of this approach).
- The concentration of carbonate given in the table of groundwater composition (Cormenzana (2008)) refers to total carbonate concentration.
- The groundwater used during the assessment should be the one resulting from equilibrating the 100,000 year old bentonite porewater with a $H_2(g)$ pressure of 20 or 80 atm and magnetite.
- Take as the central pH value the one for the resulting GW from the previous point and consider an uncertainty of ±1 unit.
- No microbial activity considered, neither sulphate nor carbonate reduction happens.
- Calculations will be run with the 5 elements at a time but it will be shown that the inclusion of other elements is not important.
- No degradation of bentonite is considered.
- No co-precipitation of a given radionuclide with a major element is considered.
- $FeSe/FeSe_2$ might be formed.
- Calcite is allowed to form if oversaturated.
- No colloidal forms are considered, only true dissolved elements.
- Iron canister corrosion products are present in the system.
- Amorphous solids are more likely to form and control solubility than crystalline solids, in accordance with Ostwald ripening.

It is important to realise how the question definition evolved from the very preliminary one set by generalist 1 to this more definitive one, increasing the number of conditions and the level of detail in some conditions / hypotheses.

A short time was devoted to the last training session. This session was dedicated to inform experts about the type of statistical support that they could get from the analyst in the second individual work period. Conditional on the fact that both were going to use computer codes (speciation calculations) within their respective approaches, the analyst focused specially on the way to select points in the input parameter space in order to optimise the information obtained from a few computer code runs. He showed strategies to identify possible interactions among input parameters and lack of model linearity.

Before the end of the meeting the project team informed experts about what was expected from them in the next two protocol steps (second individual work period and elicitation sessions). During the next 6 weeks they should do all necessary computations and literature review needed to solve the problem. They were not asked to bring any probability distribution estimate to the elicitation session, generating the distributions is the work to do in the elicitation sessions, but they were allowed to do so if they wanted. Not later than November 21st, 2008, each expert should send to the project team a short report summarising the way to approach the problem, including assumptions considered. Finally, they were informed that they could bring to the elicitation sessions any supporting material (scientific articles, books, laptops to make computations, etc.) that they deemed of interest to support their opinions. The meeting finished after agreeing about the third and final meeting dates: December 2nd to 4th, 2008. Expert 1 was invited to the elicitation sessions on December 2nd, expert 2 was invited on December 3rd, and both were invited to participate in the third day meeting to discuss results and to participate in the reconciliation sessions if these were eventually needed, as it was in fact for 4 out of the 5 elements studied.

2.6 Third meeting (elicitation sessions and reconciliation session)

The approach followed to describe what happened in the third meeting and the results of the whole process is not chronologic. In this section firstly we describe the approach adopted by each expert to solve the problem, and then we show, element by element, each expert's rationale and the corresponding distributions elicited. Finally the reconciliation session is described, again element by element, and showing the final distributions provided by each expert and the combined distribution.

2.6.1 Experts' approaches

Expert 1

Expert 1 used the geochemical codes PHREEQC, Parkhurst and Appelo (2001), and HYDRA-MEDUSA, Puichdomènech (2002), for performing computations. The

Thermodynamic database used was ThermoChimie (official Thermodynamic database of the Agence Nationale pour la Gestion des Déchets radioactifs – ANDRA, Duro (2010)).

The procedure followed by expert 1 to assess solubility limits was the following one. She obtained the reference porewater using the hypotheses considered in the question definition for partial pressures of H_2 of 20 and 80 atm. She analysed two different cases related to carbonate and sulphate reduction: Case A (neither carbonate nor sulphate reduction) and case B (carbonate and sulphate reduction allowed). In total she got four sets of results corresponding to the 4 possible combinations of 20 and 80 atm of $p(H_2)$ and cases A and B. No relevant difference was found between the cases with $p(H_2)$ of 20 and 80 atm, while important differences were found between cases A and B (case A is the one considered in the hypotheses of the problem definition). Then, speciation and solubility calculations were done with HYDRA-MEDUSA assuming a temperature of 25°C and an Ionic strength of 0.006 mol/l (from the obtained reference porewater). Predominance diagrams were generated when deemed necessary. The uncertainty ranges considered in those cases are shown in table 2.6.1.1. Then, using this information and the information contained in the datapack, including experimental studies and data from natural analogues, she selected the solid phases. Once this had been done, solubility limits were estimated. These estimations were used as best estimates. Then, further analysis of all the available information helped estimating the full distribution function for each element.

Table 2.6.1.1.- Uncertainty ranges considered by expert 1 to obtain predominance diagrams in both cases (A and B) considered. Case A corresponds to the question definition.

	CASE A	CASE B
pH	$8 \rightarrow 10$	$10 \rightarrow 13$
Eh (V)	$-0.9 \rightarrow -0.4$	$-0.9 \rightarrow -0.4$
$[C]_{tot}$	$10^{-6} \rightarrow 10^{-2} / 10^{-1}$	$10^{-6} \rightarrow 10^{-2} / 10^{-1}$
$[Fe]_{tot}$	$10^{-6} \rightarrow 10^{-2}$	$10^{-9} \rightarrow 10^{-3}$
$[S]_{tot}$	$10^{-6} \rightarrow 10^{-2}$	$10^{-6} \rightarrow 10^{-2}$

Expert 1 provided the following justifications for the hypotheses considered in the question definition that required justification

- Computations were done at 25°C, although the actual temperatures expected are between 30 and 40°C: Solubility computations at temperatures other than 25°C need data on reaction enthalpy, which are not always available for the aqueous complexes and solids relevant in our study. In her opinion, the uncertainty associated to the extrapolation to the range 30 to 40°C of results obtained at 25°C is probably less than the uncertainty associated to computations obtained at 30°C, taking into account the gaps in the enthalpy tables.

- Hydrostatic pressure of 50 atm and p(H_2) between 20 and 80 atm, although calculations done at P = 1 atm: For temperatures below 200°C small differences are expected when changing pressure. References consulted for the conditions of interest show that the maximum expected variation of reaction constants in aqueous species is ½ order of magnitude. Nevertheless, she acknowledged that pressure can affect the degree of crystallinity of the selected solid phase.
- I corrections: Extended D-H: This approach is valid for ionic strengths below 0.03 mol/kg. The ionic strength in the study porewater is 0.006 mol/kg.
- Calculations will be run with the 5 elements at a time but it will be shown that the inclusion of other elements is not important: Computations performed did not show important differences.

Though not required, expert 1 did also justify the lack of reduction of carbonates under the conditions of interest, in the absence of microbial activity. Under abiotic conditions carbonate reduction is only possible at pressures between 2 and 11 GPa and temperatures between 200 and 1,500°C (metamorphic conditions far from our problem conditions).

Expert 2

Expert 2 used the geochemical code PHREEQC for performing computations. The Thermodynamic database used was Hatches 18 (Thermodynamic database originally developed by Serco Assurance for Nirex, Bond et al. (1997)).

In order to tackle the problem proposed, he considered all available sources of information but, he adopted systematically the following importance rank

1. Speciation calculations (most emphasis).
2. Experimental solubilities.
3. Data from natural waters and natural analogues.
4. Data coming from previous performances assessments (least emphasis).

In fact, in most of the cases, data coming from previous PA's were noted but not taken as primary evidence.

In order to estimate the solubility limits for each element, expert 2 firstly tried to identify the solubility limiting phases under the established problem conditions. He acknowledged that this was one of the most important phases of the study, for which he used all available sources of information. Once these had been identified, he performed speciation calculations under different experimental conditions, varying the pH and the hydrogen partial pressure. As in the case of expert 1, further considerations about the whole set of available information helped estimating the distribution function for each element.

At this stage of the meeting the whole group, project team and experts, seemed to agree completely about the question definition, including all hypotheses. Nevertheless, on the second day, and more evidently the third day, it became obvious that both experts had a disagreement about one of the hypotheses, the one about the pH. Expert 1 considered along the whole exercise that the pH range was between 8 and 10, while expert 2 took a range between 7.5 and 9.5. This was the main source of disagreement for the estimates about one of the elements.

2.6.2 Elicitation sessions: experts' estimates

It may worth to remind a few ideas. Firstly, as it was already mentioned, experts were not asked to bring any kind of distribution to the elicitation sessions, although they were allowed to do so if they wanted. Nevertheless, expert 2 brought preliminary cumulative distribution functions. Secondly, it became evident after the first interactions with the experts that both felt much more comfortable using the interval technique to give their estimates than using any other method. Moreover, they preferred to think in terms of closed intervals except for setting absolute limits to the distributions. Most of the times, the rationale was based on powers of 10 and half powers of 10. Only in a few cases (intervals), were probabilities assessed for intervals smaller in size than ½ an order of magnitude (a factor 3.16 approximately). Thirdly, experts were allowed to follow the order they preferred to give their estimates. Nevertheless, in the next pages the order followed to show the results of the exercise is the following one: radium, tin, selenium, uranium and plutonium.

<u>Elicitations for Radium</u>

Expert 1

Regarding radium, Expert 1's rationale was as follows

- The solubility of this element is not affected by either pH or Eh, but it may be very much affected by $[SO_4^{-2}]$ and $[CO_3^{-2}]$. In order to check the effect of these concentrations, both were varied ½ order of magnitude above and below their respective best estimate values.
- $RaSO_4$ was identified as the solubility limiting phase.
- Computations resulting from the previous hypotheses spread in the range 10^{-6} to $10^{-3.3}$ mol/dm^3, but the range 10^{-6} to 10^{-5} mol/dm^3 was considered the most likely one.
- The only experimental datum available (approx. $7 \cdot 10^{-6}$ mol/dm^3) was within the range of computations.
- The lower limit, considering all uncertainties was set to 10^{-7} mol/dm^3.

Taking all this into account, expert 1 delivered the distribution for the radium solubility limit shown in figure 2.6.2.1 (dashed red line).

Expert 2

Expert 2's rationale was as follows

- Almost no experimental data are available.
- Groundwater concentrations are de-emphasized. They are too low; probably they reflect co-precipitation effects.
- $RaSO_4$ was identified as the solubility limiting phase.
- $[SO_4^{-2}]$ arises as the main source of uncertainty. Expert 2 expects a high impact of this concentration on the radium solubility limit. Approximately an order of magnitude uncertainty in this magnitude produces an order of magnitude uncertainty in the Radium solubility limit.
- His computations provided a central estimate of approximately $10^{-5.5}$ mol/dm^3.

All this, together with an assessment of the uncertainty in $[SO_4^{-2}]$, brought him to deliver an almost symmetric distribution around the best estimate value, setting absolute limits in 10^{-8} mol/dm^3 (lower limit) and 10^{-3} mol/dm^3 (upper limit), and giving very little weight to the upper and lower orders of magnitude (2% and 5% respectively). Eventually, expert 2 delivered the distribution for the radium solubility limit shown in figure 2.6.2.1 (dashed blue line).

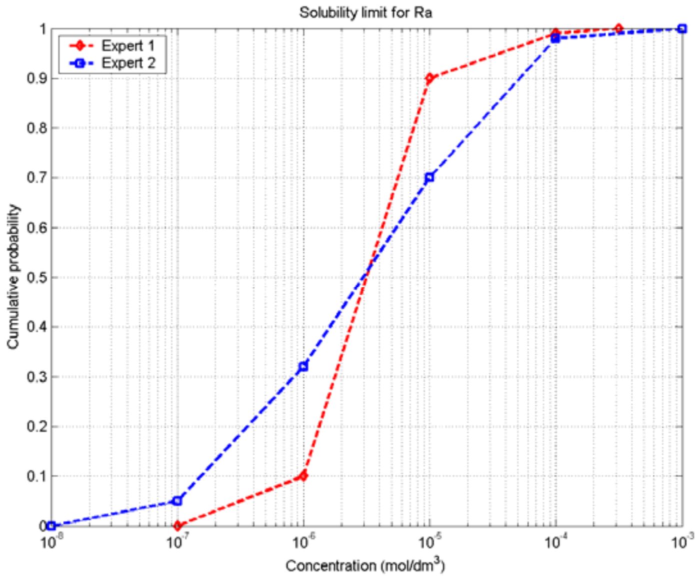

Figure 2.6.2.1.- Original distributions estimated by both experts for radium solubility limit.

Elicitations for Tin

Expert 1

Expert 1's rationale was based on the following ideas

- The solubility limit of tin is sensitive to changes in pH and Eh. This fact suggests to perform sensitivity computations in the specified assumed pH and Eh ranges (between 8 and 10, and between -0.9 and -0.4 V)
- Under these conditions, SnO_2(am) was identified as the solubility limiting phase, except for Eh<-0.7 V; then SnO(s) becomes the solubility limiting phase.
- Computations performed provided results in the range $10^{-6.5} – 10^{-4.5}$ mol/dm^3, which were set as absolute limits for the distribution.

After setting those limits, expert 1 considered the range $10^{-6.5} – 10^{-5.5}$ mol/dm^3 the most likely one, which was attributed a 90% weight. Eventually, she delivered an asymmetric distribution for the tin solubility limit, which is shown in figure 2.6.2.2 (dashed red line).

Expert 2

Expert 2's rationale was based on the following points

- No experimental data are available.
- Data about groundwater are de-emphasized, most likely they reflect co-precipitation effects, but they are not completely disregarded.
- The previous two facts brought him to rely more on speciation calculations.
- SnO_2(am) was identified as the solubility limiting phase.
- Estimates based on speciation calculations generated results in the range $10^{-7.4}$ – $10^{-6.2}$ mol/dm^3.
- The possibility of different degrees of crystallinity of the solid phase brought him to shift the lower limit to 10^{-9} mol/dm^3 (more crystalline) and to set the upper limit to 10^{-5} mol/dm^3 (more amorphous).
- Finally, giving some credibility to available groundwater data, he finally set the lower limit to 10^{-10} mol/dm^3.

Expert 2 finally delivered the distribution shown in figure 2.6.2.2 (dashed blue line).

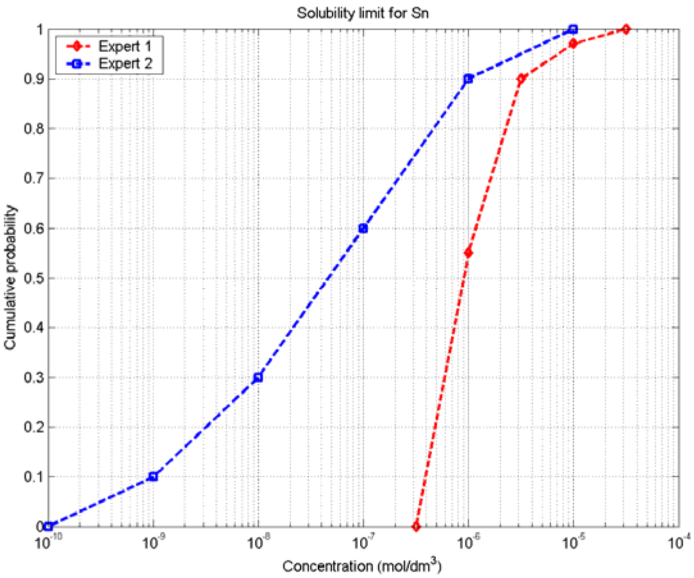

Figure 2.6.2.2.- Original distributions estimated by both experts for tin solubility limit.

Elicitations for Selenium

Expert 1

Expert 1 took into consideration the following facts

- The solubility limit of Se is sensitive to pH, Eh and [Fe].
- $Fe_{1.04}Se$ and $FeSe_2$ were identified as two possible solid phases.
- In order to assess uncertainties correctly, the uncertainty in the solubility constants of both solid phases have to be taken into account (½ order of magnitude for $Fe_{1.04}Se$ and 0.8 orders of magnitude for $FeSe_2$).
- Computations were done taking into account the variation range of pH, Eh and [Fe]. [Fe] was varied in the range [$10^{-5.7}$, $10^{-3.3}$] mol/dm^3.
- Experimental data available were considered pertinent for the problem under study and were used to set the upper limit.

Taking into account the results of her computations, all the sources of uncertainty mentioned and experimental data, expert 1 set the absolute distribution limits to $10^{-8.5}$ and $10^{-5.5}$ mol/dm^3. The full distribution is shown in figure 2.6.2.3 (dashed red line).

Expert 2

Expert 2's rationale was as follows

- No experimental data was found in the range of interest.

- Data from different groundwater sources are considered relevant except in the case of the hyper-alkaline waters of Maqarin, which delivered the highest solubilities, in the range 10^{-6} –10^{-5} mol/dm^3).
- FeSe was identified as the solubility limiting phase.
- Computations performed delivered results in the range 10^{-12} – 10^{-10} mol/dm^3.
- The results of the aforementioned computations and the solubilities observed in sources of groundwater with comparable conditions (range 10^{-10} – 10^{-8} mol/dm^3) provide the range of the bulk of the distribution for selenium.
- The lower limit was set to 10^{-13} mol/dm^3, considering that a more crystalline solid could be formed, and the upper limit was set to 10^{-7} mol/dm^3.

The final distribution delivered by expert 2 is shown in figure 2.6.2.3 (dashed blue line).

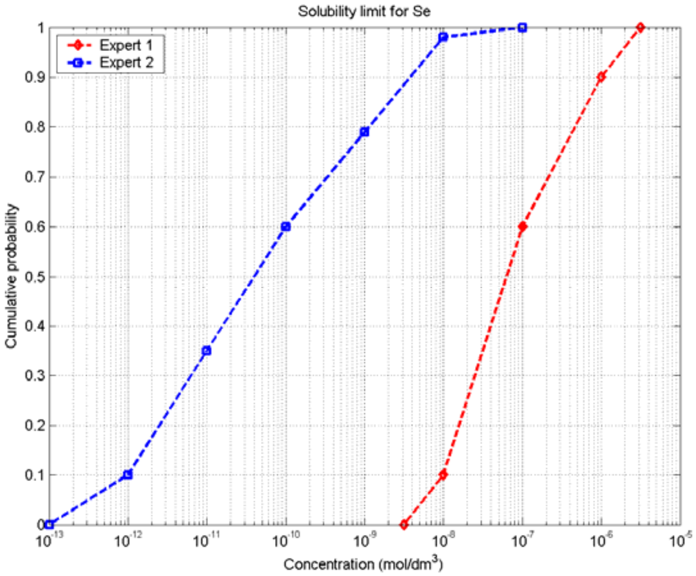

Figure 2.6.2.3.- Original distributions estimated by both experts for selenium solubility limit.

Elicitations for Uranium

Expert 1

Expert 1’s rationale was based on the following ideas

- UO_2(am) was identified as the solubility limiting phase.
- She used NEA’s stability constant in her computations, and she acknowledged that some ±1.7 orders of magnitude uncertainty could be introduced in the results as a consequence of the uncertainty in the assessment of this constant.

- Albeit in principle some sensitivity to pH and Eh variations could be expected, no sensitivity to either pH or Eh was detected in the computations, obtaining a solubility limit single estimate of $10^{-8.5}$ mol/dm^3. Adding the uncertainty mentioned in the previous bullet, she considered a first estimate for the range of variability of the solubility limit between $10^{-10.2}$ and $10^{-6.8}$ mol/dm^3.
- She did also give more credit to experimental data and data obtained from PA's to give more weight to the lower part of the distribution and to shift the lower limit down to 10^{-12} mol/dm^3, though a small weight of only 1% was given to values below $10^{-10.2}$.
- Little credit was given to the highest experimental data because they could be most likely affected by oxygen intrusion. That is why the upper limit was shifted only up to 10^{-6} mol/dm^3 and a weight of only 0.003 was given to the values in the range $10^{-6.8}$ – 10^{-6} mol/dm^3.

The distribution elicited from expert 1 for the Uranium solubility limit is shown in figure 2.6.2.4 (red dashed line).

Expert 2

Expert 2 considered the following ideas

- UO_2(am) was identified as the solubility limiting phase.
- Within the question definition, computations generated the same single solubility estimate of $10^{-8.5}$ mol/dm^3 as calculated by Expert 1.
- As expert 1 did, he considered that the main source of uncertainties was the uncertainty in the estimation of the UO_2 stability constant. In principle he considered that it could introduce an uncertainty in the results between ±1 and ±2 orders of magnitude.
- Data from experiment (ranging from approximately 10^{-9} to $5 \cdot 10^{-4}$ mol/dm^3) were given credit, except the most extreme ones (low and high). These data helped setting the upper limit to 10^{-5} mol/dm^3.
- The lower limit was set to 10^{-11} mol/dm^3 based on data from groundwater.

Finally, based on all these ideas, he delivered the uranium solubility limit distribution shown in figure 2.6.2.4 (dashed blue line).

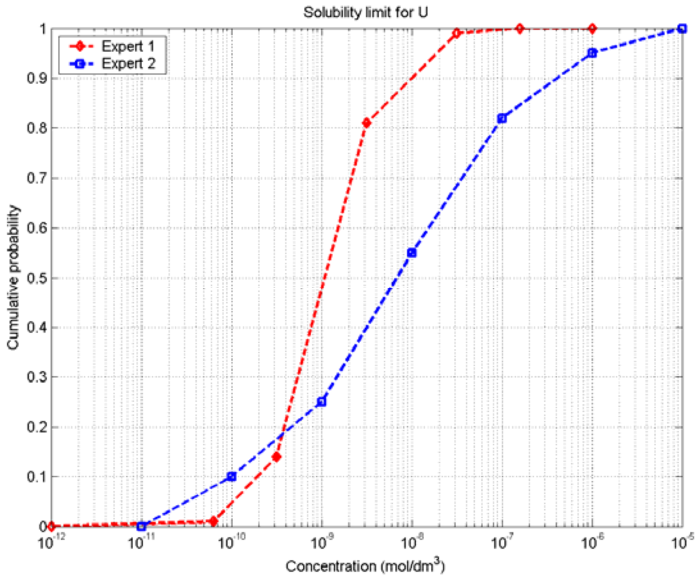

Figure 2.6.2.4.- Original distributions estimated by both experts for uranium solubility limit.

<u>Elicitations for Plutonium</u>

Expert 1

Expert 1 took in consideration the following ideas in her rationale

- The solubility of plutonium is sensitive to changes in pH and Eh in the question hypotheses' range.
- $Pu(OH)_3(s)$, $PuOHCO_3(s)$ and $PuO_2(s)$ were identified as possible solubility limiting phases for this element.
- Computations were done within the range of variation of pH and Eh. Within this range, a central best estimate of $10^{-8.6}$ mol/dm^3 was obtained. Considering that an uncertainty of ±0.6 orders of magnitude could be introduced by the stability constant, the interval $10^{-9.2} – 10^{-8}$ mol/dm^3 was considered to be the core of the distribution, concentrating 70% of the probability.
- The whole computational spread (approximate range $10^{-10} – 10^{-7.5}$ mol/dm^3), was used to set lower and upper limit, but both were shifted ½ order of magnitude (downwards and upwards respectively) in order to account for uncertainties in constants.

According to these ideas, the elicitation of expert 1 produced the plutonium solubility limit distribution shown in figure 2.6.2.5 (dashed red line).

Expert 2

Expert 2's rationale was based on the following ideas

- There is a wide range of experimental data values on the solubility of plutonium in the pH range of interest. The most relevant data lie in the range $5 \cdot 10^{-11}$ - $2 \cdot 10^{-8}$ mol/dm^3.

- Much uncertainty is acknowledged about the solubility limiting phase, which may change along the pH range considered. The most likely phase is an amorphous form of $Pu(OH)_3$, but the crystalline form has to be considered also, which has a stability constant 1.5 orders of magnitude smaller. Other potential, less likely limiting phased, may be $Pu(OH)_2$. $Pu(OH)_2CO_3$ is also considered as a potential solubility limiting phase, though an unlikely one. PuO_2 and $Pu(OH)_4$ were also taken into account.

- Based on the speciation calculations done for most likely solubility limiting phase, the core of the distribution was set in the range 10^{-8} – $10^{-4.5}$ mol/dm^3.

- The upper limit was set to $10^{-3.5}$ mol/dm^3, based on experimental data and some speciation computations, though not giving much credibility to the highest values (experimental data obtained in very saline conditions, and speciation computations obtained at the very lower limit of the pH range).

- Most relevant experimental data indicated that some important weight should be given to the range $5 \cdot 10^{-11}$ - $2 \cdot 10^{-8}$ mol/dm^3, and solubilities obtained when PuO_2 was used as the solubility limiting phase were used to set the lower limit to 10^{-11} mol/dm^3.

Taking into account all these ideas, expert 2 generated the distribution shown in figure 2.6.2.5 (dashed blue line).

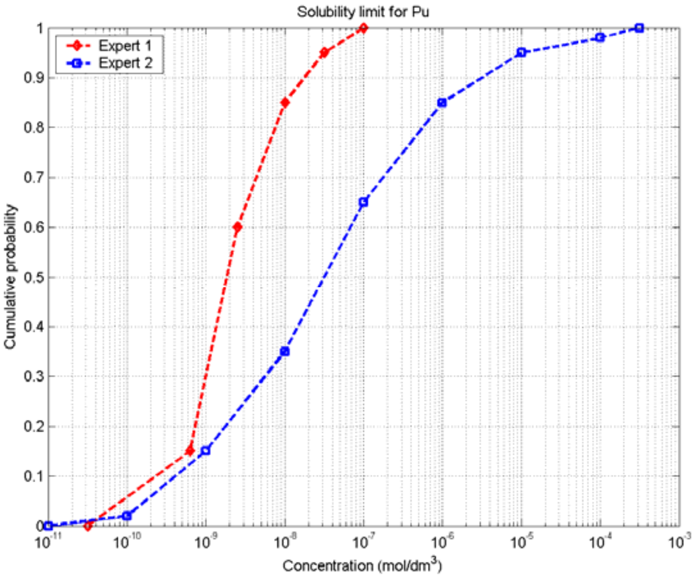

Figure 2.6.2.5.- Original distributions estimated by both experts for plutonium solubility limit.

2.6.3 *Reconciliation sessions*

At the end of the second day the project team discussed areas of disagreement between the experts. Reconciliation sessions were held on the last day of the meeting. The whole project team, including the representative of the PA owner and the two experts participated in these reconciliation sessions. The following sections provide a summary of the reconciliation/ discussion sessions.

Reconciliation/discussion session for Radium

This was not a proper reconciliation session; it was more a results discussion session. The elicitation sessions had revealed a high degree of agreement between the experts. Although differences between both distributions are evident (see either figure 2.6.2.1 or 2.6.3.1), expert 2 delivered a more spread distribution than expert 1, it is also evident that the ranges of both distributions are quite similar. The distribution delivered by expert 2 contains completely the one delivered by expert 1, and the latter covers more than 90% of the former's range (in probability). Taking this into account and the fact that the rationale of both experts did not differ much, no reconciliation is really needed. Under these circumstances the mathematical combination (linear pool) may be obtained, see Bolado et al. (2009). The result of this combination is shown in figure 2.6.3.1 (solid black line).

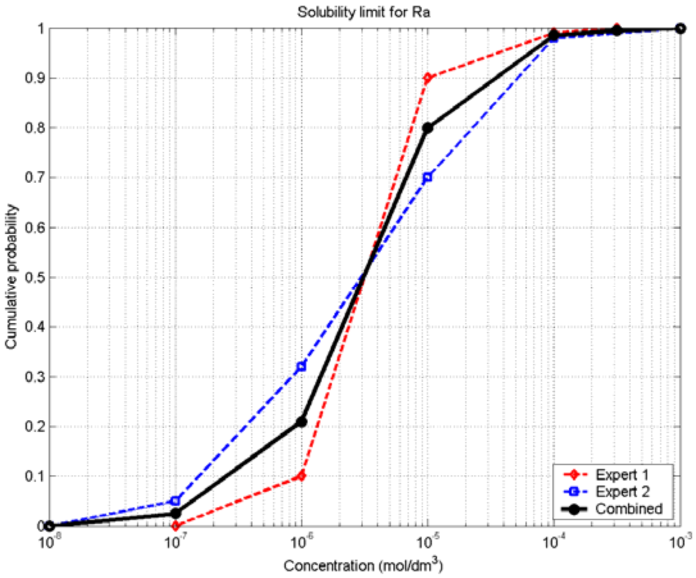

Figure 2.6.3.1.- Original distributions estimated by both experts for radium solubility limit and combined distribution.

Reconciliation session for Tin

The case of Tin is a clear case of disagreement between experts. Results are shown in figures 2.6.2.2 (original estimates) and 2.6.3.2 (original, final and combined estimates). In figure 2.6.3.2, when dashed and solid curves of the same colour overlap, only the solid line can be seen (final opinion of the expert). The original distribution provided by expert 2 covers more than 95% of expert 1's distribution range, but the distribution given by expert 1 covers approximately only 25% of expert 2's distribution range. The differences between both distributions increase monotonically as we move to lower values; while the differences between upper limits is only ½ order of magnitude, the medians differ more than one order of magnitude and the lower limits differ 3.5 orders of magnitude. Basically, expert 1 considers impossible the values at the core of the distribution provided by expert 2. A reconciliation session is unavoidable under these circumstances.

Three main reasons of disagreement may be identified to explain the differences between both distributions. The first one is related to the computational results obtained. Albeit both identified the same most relevant solubility limiting phase, the uncertainty computational ranges overlapped only a little (expert 2 obtained smaller values than expert 1). Secondly, expert 2 gave more weight to the possibility of getting a more crystalline solid, which produced a shift of the distribution towards smaller values. Thirdly, he gave more credit to data from groundwater, which were used as a key reference to set the lower limit.

After a review of the data available in the datapack, Expert 1 changed her mind, especially about the lower part of her distribution. Firstly, under different experimental conditions than the ones assumed in the exercise, but within the same pH range, experimental data as low as $10^{-7.5}$ mol/dm^3 were found. She decided also to give some more credit to the lower values used in similar PA studies. She also acknowledged the low values present in natural waters, but did not give much credit to them because they can reflect a large degree of crystallinity. Taking all this information in consideration, she decided to shift the whole distribution, excluding the upper limit, which was kept unaltered, towards lower values. The new core of her distribution was the interval 10^{-7} – 10^{-6} mol/dm^3, which was given a weight of 80%, and the lower limit was shifted to $10^{-8.5}$ mol/dm^3.

Expert 2 also reviewed the information available in the Datapack. Moving his upper limit for the pH up to 10, he found pertinent experimental data around $5{\cdot}10^{-5}$ mol/dm^3. This information, together with the acknowledgement of uncertainty, brought him to increase the upper limit of the distribution to 10^{-4} mol/dm^3, while the old upper limit became the 97th percentile of the distribution. In the lower part of the distribution, the probability of the interval 10^{-10} – 10^{-9} was reduced by ½. This was based on accepting the relevance of some previous PA data, which delivered not so low solubility limits, and acknowledging that tin concentration in most natural waters are probably below solubility limitation.

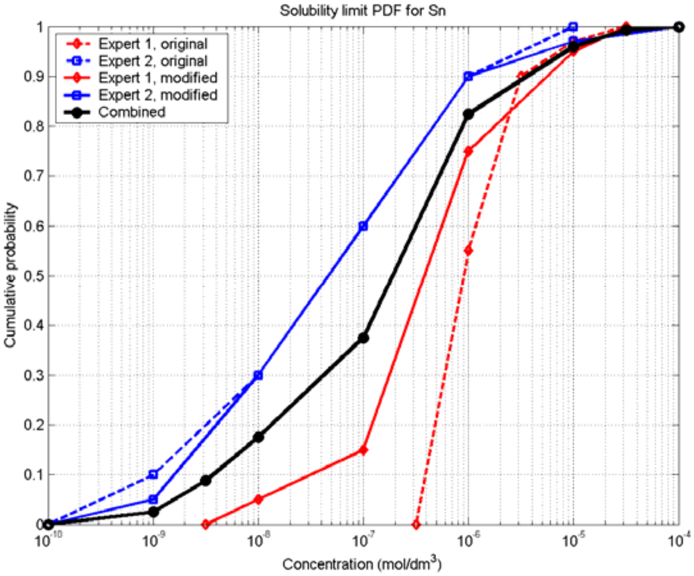

Figure 2.6.3.2.- Original distributions estimated by both experts for tin solubility limit, modified distributions after the reconciliation session and combined distribution.

The distributions delivered by both experts as a result of the reconciliation session are shown in figure 2.6.3.2 (solid red and blue lines). Although an average shift of approximately one order of magnitude between both distributions can still be seen, an acceptable degree of agreement was achieved after the reconciliation session. The distribution range delivered by expert 1 is contained in the distribution range of expert 2, and the former covers roughly 80% of the latter (in probability). Under these circumstances the mathematical combination of both distributions is feasible and defendable. The black solid line in figure 2.6.3.2 is the combined final distribution.

Reconciliation session for Selenium

This element produced the largest disagreement between the experts. Figures 2.6.2.3 and 2.6.3.3 show the original distributions elicited from both experts (dashed lines). An average shift of three orders of magnitude between both distributions may be observed (1.5 orders of magnitude between upper limits, 3 between medians and 4.5 between lower limits). The distribution delivered by expert 1 covers less than 15% (in probability) of the distribution range given by expert 2. This indicates that, in the view of expert 1, most of the distribution delivered by expert 2 lay in an impossible range. On the other hand, Expert 2's view was that Expert 1 was over confident and had not fully captured the uncertainty.

The main source of disagreement became evident very fast. The solubility limiting phases used by both experts were different. In fact, on average, FeSe (solubility limiting phase used by expert 2) is approximately 2.7 orders of magnitude less soluble than $Fe_{1.04}Se$ (one of the two solubility limiting phases considered by expert 1). The different credit given to experimental data became important to set the upper limit.

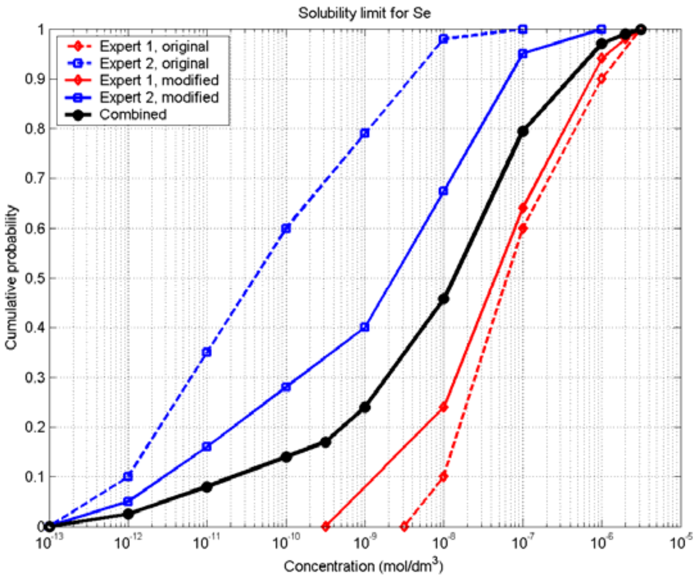

Figure 2.6.3.3.- Original distributions estimated by both experts for selenium solubility limit, modified distributions after the reconciliation session and combined distribution.

In the discussion held during the reconciliation session, expert 2 agreed to consider $Fe_{1.04}Se$ as an additional possible solubility limiting phase. Nevertheless, he considered that the other solubility limiting phase used by expert 1, $FeSe_2$, was unlikely to form in our system; consequently he discarded it. Regarding the credit given to high experimental values, he held his original opinion that they were not relevant for the problem under study because either they had been obtained in too saline systems or under a too high pH value. Eventually he decided to run further speciation calculations considering $Fe_{1.04}Se$ as the solubility limiting phase. He got results in the range 10^{-9} – 10^{-7} mol/dm^3. He also decided to give a weight of 60% to the computations done with $Fe_{1.04}Se$ and a weight of 40% to the computations done with FeSe. Including all possible sources of uncertainty, finally he shifted the upper limit up to 10^{-6} mol/dm^3. He did not see any good reason to modify the distribution lower limit.

Expert 1 decided to shift the lower limit of her distribution one order of magnitude downwards from $10^{-8.5}$ to $10^{-9.5}$ in order to account for more crystallinity in the solid phase, as was done in other PA studies. The rest of the distribution was slightly shifted to lower values, except the upper limit, which was kept.

The distributions finally delivered by both experts after the reconciliation session are shown in figure 2.6.3.3 (solid red and blue lines). Though the convergence of both distributions is obvious, important differences remain (an average difference between 1.5 and 2 orders of magnitude). From the point of view of coverage, The distribution generated by expert 2 covers 95% of the distribution range of expert 1, while the distribution range of expert 1 covers roughly 2/3 of the distribution range of expert 2. In the opinion of the analyst, this is the minimum degree of agreement needed to allow a mathematical combination, which is provided in figure 2.6.3.3 (solid black line).

Reconciliation session for Uranium

The original distributions obtained from the elicitation sessions did not show a large disagreement. Each distribution covers a very high percentage of the range of the other distribution. Nevertheless, it is also obvious the disagreement in the upper part of the distribution, especially in the very highest quantiles (an average shift of approximately one order of magnitude between both distributions).

Both experts identified the same solubility limiting phase (UO_2(am)), used the same best estimate value of the stability constant (NEA) and delivered the same best estimate value for the solubility. They also agreed about the uncertainty in the stability constant. As a consequence of this, both experts obtained similar ranges. The experts disagreed, however, when they had to attribute weights to different parts of the distribution. Expert 1 gave more weight to the lower part of the distribution, giving credit to low experimental values and data from previous PA studies, while she did not give much credit to high experimental values, which she considered could be due to the presence of oxygen in the experiments. Expert 2 gave similar credit to high and low values (excluding very large and very small values), delivering a more symmetric distribution. The reconciliation session did not result in very much closer distributions. Only expert 2 modified slightly his distribution, shifting some weight from the upper part of the distribution to the intermediate values (giving a bit less credit to high experimental values in favour of the intermediate ones). Under these conditions (acceptable degree of agreement) the mathematical combination was obtained. It is shown in figure 2.6.3.4 (solid black line).

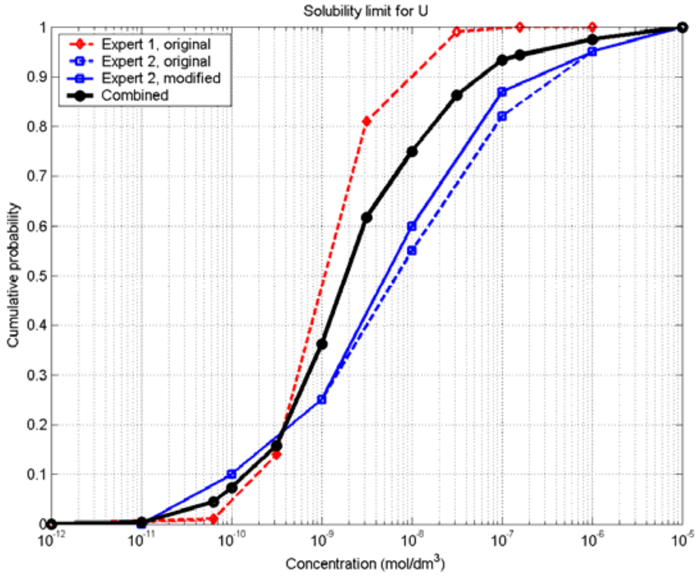

Figure 2.6.3.4.- Original distributions estimated by both experts for selenium solubility limit, modified distributions after the reconciliation session and combined distribution.

Reconciliation sessions for Plutonium

The difference between the distributions generated by the experts for the plutonium solubility limit is evident. The difference increases as we move to higher values. The largest difference is achieved when both maxima are compared (3.5 orders of magnitude). A reconciliation session was certainly needed.

During the reconciliation session it became evident that the main source of differences was the consideration of different pH ranges in their estimations. Expert 1 took 8 as a lower limit, while expert 2 set the limit for his estimations to 7.5. Such a change in the lower limit of the pH produced a change of three orders of magnitude in the solubility. The whole group realised the different interpretation of one of the assumptions of the problem under study by both experts. Moreover, both experts remained firm in their opinions. At this point of the reconciliation session the project team had to make a decision about which limit should be considered, otherwise experts would provide solutions to different problems. The PA owner representative made the decision of taking 7.5 as the pH lower limit, in order to include all possibilities (8 may be kept for sensitivity analyses studies). After this decision, expert 1 decided to change her distribution, converging strongly to the distribution estimated by expert 2 (see the blue and red solid lines in figure 2.6.3.5). In fact, after the reconciliation session, this was the case where both experts most closely agreed, together with the case of Radium. The degree of agreement obtained was really remarkable. The final combined distribution is also shown in figure 2.6.3.5 (solid black line).

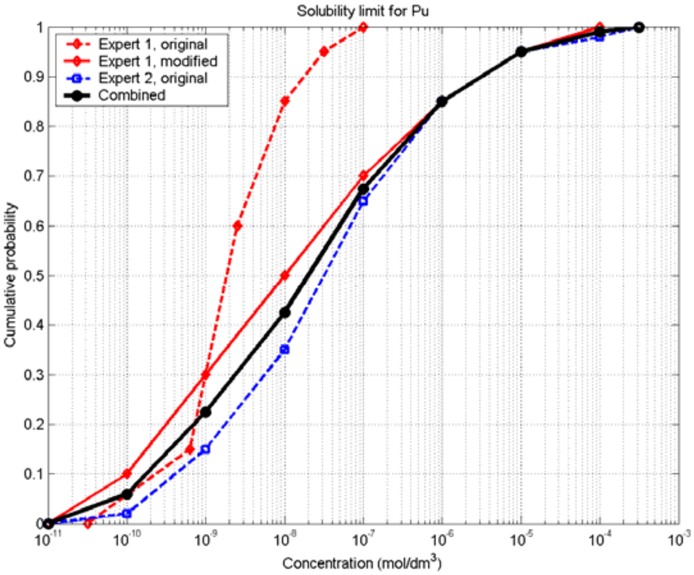

Figure 2.6.3.5.- Original distributions estimated by both experts for plutonium solubility limit, modified distributions after the reconciliation session and combined distribution.

3. Lessons learnt

From a methodological point of view the following lessons were learnt during this study:

- The experts found it very helpful to receive the datapack and supporting documents before the actual start of the process.
- The experts found the training and calibration sessions, where they could experience the risk of overconfidence, interesting. Further improvement of these sessions could be made to make them more attractive and challenging for the experts.
- The joint refinement of the problem definition (involving the PA owner, the project team and the experts) was extremely important to avoid implicit hypotheses and misunderstandings. Even being aware of this and dealing with it explicitly in two protocol sessions, a real hypothesis disagreement arose in the reconciliation session.
- The experts found the interval technique most useful. In fact, after the first questions in the elicitation sessions, both experts chose this technique to give their opinions. They found it most useful to think about concentrations in terms of powers of 10.
- The time schedule in the elicitation sessions was really tight. The whole group worked under time pressure. This suggests that we should have been a bit less ambitious, eliciting not more than 3 or 4 solubility distributions per day.
- It was found useful to elicit several similar parameters in the same session. The time required to elicit the solubility limit of each element decreased monotonically as the process advanced. Experts quickly became familiar with the elicitation process and the last elicitations run smoothly.
- At the beginning of the reconciliation session each expert had no information about the distributions provided by his/her colleague. This made the schedule of the reconciliation session also very tight. An issue to study in the future is whether the reconciliation session (if needed) deserves a specific meeting. This would of course have clear effects on the budget for the elicitation process although the outcome of the elicitation would clearly benefit from the extra time.
- Including three experts in the process is probably the optimum number, in order to get the right balance between diversity of opinions/background and work load.

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