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Groundwater Chemistry in SKB's Safety Assessment SR-Site: Initial Review

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SSM perspektiv

Bakgrund

Strålsäkerhetsmyndigheten (SSM) granskar Svensk Kärnbränslehantering AB:s (SKB) ansökningar enligt lagen (1984:3) om kärnteknisk verksamhet om uppförande, innehav och drift av ett slutförvar för använt kärnbränsle och av en inkapslingsanläggning. Som en del i granskningen ger SSM konsulter uppdrag för att inhämta information i avgränsade frågor. I SSM:s Technical note-serie rapporteras resultaten från dessa konsultuppdrag.

Projektets syfte

Syftet med detta projekt är att granska SKB's hantering av den långsiktiga utvecklingen av grundvattenkemi som inkluderar perioder med olika klimat som tempererade betingelser, permafrost och glaciala betingelser. Den nu avslutade karakteriseringen av grundvattenkemi vid Forsmark utgör en grund för förståelse av den geokemiska och hydrologiska utvecklingen under långa tidsperioder. Geokemiska processer (och även mikrobiologiska) i närområdet av ett slutförvar som påverkar redox och pH har en särskild betydelse eftersom redox och pH tillståndet påverkar kopparkapselns korrosionsmekanismer och även förutsättningarna för retardation och transport av radionuklider (för grundvatten som kommer i kontakt med kopparkapslar och bränsleelement).

Författarens sammanfattning

Denna preliminära granskning av säkerhetsanalysen SR-Site fokuserar specifikt på hanteringen av grundvattenkemi i platsbeskrivningen av Forsmark och i den långsiktiga säkerheten för ett slutförvar vid Forsmark. Data för kemiska komponenter och isotoper i grundvatten har bidragit till den platsbeskrivande modellen (SDM= ”Site Descriptive Model”) för Forsmark. I denna SDM tolkas grundvattenkemi av flera olika skäl. Huvudsakliga resultat är beskrivningen av dagens kemiska betingelser i berget vid aktuellt djup för ett slutförvar samt en tolkning av hur dagens grundvattensystem har utvecklats långsiktigt beroende på omgivningens påverkan av grundvattnets rörelsemönster.

Grundvattenkemidata och tolkningar i den platsbeskrivande modellen (SDM) har bidragit till utvecklingen av säkerhetsanalysen SR-Site som beskriver den långsiktiga säkerheten för ett framtida slutförvar. Detta kräver en tillförlitlig och tillräckligt bra förståelse av hur kemiska betingelser i närområdet av ett slutförvars tekniska barriärer förändrar sig med tiden och vilka kemiska reaktioner med mineral i berget som kan moderera möjliga störningar och naturliga ändringar. I det fall där radionuklider frigörs från slutförvaret och sprider sig i omgivande grundvatten påverkar sammansättningen av grundvatten också hur radionukliderna transporteras genom berget.

SKB:s tilltro till grundvattenkemiska aspekter av SR-Site är rent generellt underbyggd av tillgängliga data, tolkningar och modeller. Det finns ett antal frågeställningar kring data, tolkningar och modeller med innebörden att framtida grundvattensammansättningar möjligen kan ligga utan-för det parameter intervall som finns beskriven i SR-Site; och om dessa ytterligare osäkerheter är försumbara eller inte för säkerhetsanalysens resultat behöver utvärderas.

Många av de frågor som har identifierats i denna initiala granskning uppstår på grund av de förenklingar och antaganden som har genomförts för tolkningar och modelleringar som är relevanta i säkerhetsanalysen. I vissa fall är användningen av expertbedömningar för att förenkla konceptuella modeller, säkerhetsfunktioner, säkerhetsfunktionskriterier eller andra parametrar inte tillräckligt motiverade.

Två exempel ges som illustrerar den typ av frågor för vilka det behövs klarlägganden eller ytterligare motiveringar; Det första fallet gäller höga koncentrationer av det korroderande ämnet sulfid i grundvattenprover som har blivit borträknade då de överstiger förväntade värden i den biogeokemiska modellen för sulfidbildning och sulfidkontroll. Det andra fallet avser en överdriven förenkling av den hydrokemiska modelleringen av grundvattnets kemiska utveckling beroende på att silkatvittringsreaktioner och jonbytesprocesser har uteslutits utan en utvärdering av den resulterande osäkerheten i utdata.

Projektinformation
Kontaktperson på SSM: Bo Strömberg
Diarienummer ramavtal: SSM2011-3637
Diarienummer avrop: SSM2011-4260
Aktivitetsnummer: 3030007-4012

SSM perspective

Background

The Swedish Radiation Safety Authority (SSM) reviews the Swedish Nuclear Fuel Company's (SKB) applications under the Act on Nuclear Activities (SFS 1984:3) for the construction and operation of a repository for spent nuclear fuel and for an encapsulation facility. As part of the review, SSM commissions consultants to carry out work in order to obtain information on specific issues. The results from the consultants' tasks are reported in SSM's Technical Note series.

Objectives of the project

The objective of this project is to review of SKB's treatment of long-term groundwater chemistry development including various periods of different climate such as temperate, permafrost and glacial conditions. The completed groundwater chemistry characterization at the Forsmark site provides an important basis for understanding the site geochemical and hydrological development during long-time periods. The near-field geochemical (and microbial) processes affecting redox and pH conditions of groundwater have a particular importance, since the redox and pH state affect various canister corrosion mechanisms as well as conditions for radionuclide retardation and transport (groundwater contacting e.g. copper canisters and spent fuel elements).

Summary by the author

This preliminary review of SR-Site is focused specifically on the treatment of groundwater chemistry in the description of the Forsmark site and in the long-term safety of a deep geological repository at Forsmark. Data for the chemical and isotopic compositions of groundwaters have contributed to a site descriptive model (SDM) for Forsmark. In that SDM, groundwater chemistry is interpreted for several purposes. The main outputs are the description of present-day chemical conditions in rocks at the target depth for a repository and an interpretation of how the present-day groundwater system has evolved over the long-term due to the effects of environmental changes on groundwater movements.

Groundwater chemistry data and interpretations in the SDM have contributed to the development of the SR-Site assessment of long-term safety of a future repository. That requires a reliable and adequate understanding of how near-field chemistry around the engineered barriers might change over time and what reactions with rock will moderate potential perturbations and natural changes. For the case that radionuclides are released from the repository into the surrounding groundwaters, the compositions of groundwaters may also affect how radionuclides are transported through the bedrock.

SKB confidence about groundwater chemistry aspects of SR-Site is generally supported by the data, interpretations and models. There are a number of issues about the data, interpretations and models that could mean that future groundwater chemistry conditions would possibly lie outside the parameter range that is described in SR-Site; whether these additional uncertainties are negligible or otherwise for the safety case is yet to be evaluated.

Many of the issues that have been identified in this initial review arise because of the assumptions and simplifications that have been used in processing site data for interpretations and models that are relevant to the safety case. In some cases, the application of expert judgement in simplifying conceptual models, safety function indicator criteria and other parameters are not adequately justified.

Two examples illustrate the types of issues that need clarifications or additional justifications. In the first, high concentrations of corrodant sulphide in some present-day groundwaters are discounted because they exceed expected model values in a biogeochemical model for sulphide production and regulation. In the second example, hydrogeochemical modelling of groundwater chemistry evolution has been over-simplified by omitting silicate mineral dissolution/precipitation and cation exchange reactions, without adequate evaluation of the resulting uncertainties in the model outputs.

Project information
Contact person at SSM: Bo Strömberg
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1. Scope of review

The scope of my review is defined primarily by SSM's requirements:

- Identify need for complementary information and clarification;
- Detailed analysis of specific issues is not required at this stage;
- Scope includes treatment of long-term groundwater chemistry development including temperate, permafrost and glacial conditions;
- Near-field geochemical and microbial processes affecting redox and pH conditions are important;
- Coupling of hydrogeology and groundwater chemistry as basis for understanding of flow patterns near repository over long timescale;
- Concentrations of corrodants in groundwater (notably HS^- and O_2);
- Evolution of groundwater salinity;
- Methods used to define near-field chemistry – are they appropriate?
- Range of geochemical conditions defined by sampling and modelling – is it reasonable and defensible?
- Detailed review of microbial processes is done in another task.

In addition, I have considered:

- Clarity, credibility and transparency of the explanation of hydrogeochemical data and processes, and of their relationships with long-term repository safety;
- Identification and prioritisation of safety-relevant issues relating to groundwater chemistry;
- Robustness of hydrogeochemical concepts, interpretations and numerical models in the wider geoscientific context.

2. General comments

The purpose of the SR-Site safety assessment is to investigate whether a KBS-3-design spent nuclear fuel geological repository in the identified candidate rock volume at Forsmark will be safe in the long-term, i.e. reported by SKB up to 1 million years. In the context of groundwater chemistry in SR-Site, long-term safety according to the multi-barrier concept concerns the chemical conditioning and stability of the near field that will secure long-term containment by the engineered barrier system plus retardation of any released radionuclides in the groundwater system between the deep repository and the biosphere.

My general impression is that SKB are confident that there are no issues of major concern and no surprises in the groundwater chemistry. In my opinion, that general position is supported by the data, interpretations and models in SR-Site. Nevertheless there are a number of issues where the risks to the safety case in SR-Site are low but are not as well constrained as they should be. In other words, there are possibilities albeit with low likelihood that groundwater chemistry conditions could lie outside the envelope of possibilities that is described in SR-Site. SKB admit to this position by the frequency with which they state that uncertainties in various areas should be reduced and that investigations are still ongoing.

The SR-Site Main Report has been put together with a substantial effort to show the logical construction of the safety case from basic principles through to the integrated safety assessment. Its structure works well considering the complexity of the cross-linking between interpretations and models. In general, the scientific approaches taken by SKB to take account of groundwater chemistry in SR-Site are mostly appropriate and fairly comprehensive. Expert judgements play an important role in the safety case and this is acknowledged and explained (TR-11-01, Vol. 1, p92), though I think that there are various problems with the transparency and justifications of specific cases where expert judgement has been used.

Inevitably, the level of achievement in obtaining groundwater samples and chemical analyses that are sufficient in terms of reliability and spatial distribution is an issue. SKB have applied ‘state of art’ methods for characterising the chemical and isotopic compositions of groundwaters that are representative for repository depth and for the far field geosphere. There are some restrictions of the data set that constrain the completeness of characterisation. One aspect is the paucity of water samples from below repository depth especially below the target volume in the northwest part of the candidate area; the potential significance of this lies in increasing groundwater salinities.

Another aspect of data and interpretative limitations is the characterisation and understanding of water compositions and water-rock reactions in less transmissive fractures and in the bedrock matrix. State of art methods have been used to analyse pore waters in the matrix but the hydrochemical data are restricted to chloride concentrations and stable isotope ratios, for which the reliability is not well established. These issues have a number of implications, one of which is that there are various sources of uncertainties about the compositions of water that would enter deposition holes and the localised water-rock reactions that would buffer those compositions. The uniformity or otherwise of diffusive solute exchange between fracture waters and pore waters, and the rock properties that might affect it, is not well understood.

The interpretations of processes and the hydrogeochemical concepts that SKB uses to understand the initial state of the site and to model the future evolution of groundwater conditions should be supported by references to published and peer-reviewed interpretations of comparable geosphere systems, preferably including geoscientific studies outside the narrow focus of geological disposal. The site-specific conceptual models that underlie the safety functions should be shown as much as possible to be consistent with, and supported by, more general principles and paradigms for long-term evolution and for solute transport in the geosphere.

A large proportion of the citations in SR-Site comprise SKB reports and papers and the hydrogeochemical interpretation of the Forsmark site is not effectively set in the wider context achieved by studies of other granitic groundwater systems, e.g.

- Frape et al 2005 (Deep fluids in the continents: II, crystalline rocks. Chapter 17, Volume 5, Surface and ground water, weathering and soils, Treatise on Geochemistry. H D Holland & K K Turekian, eds);
- Pearson 1987 Models of mineral controls on the composition of saline groundwaters of the Canadian Shield (In: Saline waters and gases in crystalline rocks, Geol Assoc Canada Special Paper 33);
- Michard & Pearson 1996 (Chemical evolution of waters during long term interaction with granitic rocks in northern Switzerland, Applied Geochemistry 11, 757-774);
- Nordstrom et al 1989 (Groundwater chemistry and water-rock interactions at Stripa, Geochimica et Cosmochimica Acta, 53, 1727-1740);
- Oelkers & Schott 2009 (Thermodynamics and kinetics of water-rock interaction, Reviews in Mineralogy & Geochemistry, Vol 70).

3. Main review findings

3.1. Data sources for groundwater chemistry

Measures taken to optimise the reliability of chemical and isotopic data (e.g. drilling water tracing, monitoring during sampling) could not reasonably have been improved upon. Water samples were biased towards higher transmissivity zones and have a low spatial density, especially in some deeper parts of the candidate repository volume.

Variable data accuracy and precision inevitably occurs with the diverse data types that characterise groundwater chemistry and this is exacerbated by the challenges of sampling groundwaters and the varying degrees of 'representativeness' for chemical/isotopic and microbiological entities in deep groundwaters. There is only occasional explanation to give the reader a better insight of these issues and to promote a balanced and transparent understanding of data reliability and of the weight that should be placed on it.

SR-Site provides illustrations of groundwater chemistry that, with few exceptions (e.g. for HS^- data), do not identify and examine the sources of and possible significance of 'anomalous' data values. In this way, spatial heterogeneity of data and other variations such as potential correlations between groundwater compositions and hydraulic conductivities are not fully considered.

An example that illustrates this point is Figure 4-22 (TR-11-01, Vol. 1, p133). Compositions of waters sampled from (inferred) fracture zones are interpolated/extrapolated onto the major deformation zones whereas pore water compositions (Cl^- concentrations only, and for just a few borehole locations) have been interpolated according to an unknown procedure, probably expert judgement. Thus Figure 4-22 is a smoothed representation of present-day groundwater compositions. It is a schematic illustration that does not show the accuracy that would be expected for a diagram that is 'interpreted from hydrogeochemical data' as stated in the caption.

Most of the sampled groundwaters originate in deformation zones (DZs) and a 'limited amount' of groundwater chemistry data represent typical bedrock in the fracture domains (FFM) (p303 in TR-08-05). Therefore there is some uncertainty about the chemistry of water in the FFM fractures that will be in proximity to the deposition holes. The overall uncertainties in composition are particularly of concern for FFM01, the target volume of bedrock, in which the opportunities to sample groundwaters have been especially low and perhaps unrepresentative because of the low fracture density.

Figure 9-6 (p313 in TR-08-05) is a 3D visualisation of chloride (Cl^-) concentrations through the candidate volume. The paucity of groundwater samples from the target volume of FFM01 rock in the NW part of the site is evident, with only two data points below 600 m depth in the region where higher salinities are encountered. The adequacy of these sparse data in relation to potential uncertainties in groundwater chemistry, 'initial state' and boundary conditions needs to be considered in more detail.

Data for rock matrix pore waters are available from a few boreholes (Figure 6-17, TR-08-05, p329). Interpretation of the depth profiles suggest that pore water compositions are distinct between FFM01 and FFM03/02, the latter having higher fracture density. Extrapolation of the few fracture water compositions according to the gradient of pore water compositions in FFM01 suggests higher salinity below 800 m depth than is envisaged in SR-Site. This reinforces the case for alternative models to be considered for initial state and boundary compositions.

2D cross-section schematic diagrams of groundwater compositions and mixtures (Figures 9-22 and 9-23 in TR-08-05; Figure 4-22 in TR-11-01, Vol. 1) suggest that Littorina water penetrated to around 600 m depth in the gently-dipping DZs (FFM03/02) and to only 150-300 m in the FFM01 footwall bedrock unit. In general, the heterogeneous spatial distribution of groundwater compositions, influenced by both gently-dipping DZs and the internal hydraulic characters of the fracture domain bedrock units, suggests compartmentalisation of groundwater in a way that is not simulated adequately by the ConnectFlow model. SR-Site has no discussion of the detailed comparison of hydraulic and hydrochemical heterogeneities and the implications for uncertainties and alternative interpretations of groundwater movements (this issue is raised in Section 4.8).

Traceability of hydrochemical data from the site descriptive model (R-08-47, Bedrock Hydrogeochemistry, Forsmark, SDM-Site, Laaksoharju et al 2008) into SR-Site is difficult to follow and in a few specific aspects is poor or at least seems to oversimplify the observed parameter ranges and discount anomalous data values. Specific examples of this in the Hydrogeochemical Evolution report for SR-Site (TR-10-58, Salas et al 2010) are: (i) Near-surface waters in the depth range 0-20 m are said to be fresh, i.e. <200 mg/L Cl^- (TR-10-58, p19) which is mostly the case, but Fig 4-1 in R-08-47, p62) shows at least one sample with about 4000 mg/L (this may be significant because high salinity indicates potential upward flow); (ii) Shallow ‘mixed brackish’ groundwaters in the depth range 20-200 m are said to have Cl^- in the range 200-2000 mg/L (TR-10-58, p19) whereas Fig 4-1 in R-08-47 shows that Cl^- concentrations are mostly between 3000-6000 mg/L; (iii) The scatter of data points for pH in Fig 6-10 (TR-10-58, p53) is rather different, though generally covering the same range, compared with the scatter shown in Fig 4-6 in R-08-47 (p67); (iv) The number of data points and the range covered (>0.095 mg/L PO_4^{3-}) in Fig 4-4 (TR-10-58, p39) are greater than the data shown in Fig 4-11a in R-08-47 (p73) for which the maximum of the range is <0.07 mg/L.

These differences may just be due to an updated data set being used for SR-Site, but those data are only traceable in SKB’s spreadsheet which has been provided on request but is not user-friendly. A thorough QC-type check is advised, especially to ensure that ‘anomalous’ measurements have not been discounted without justification.

Transparency and traceability of data would be aided by tables of data used being included in SR-Site, e.g. as appendices. In general, tracing how data in SDM-Site have been used in SR-Site is made difficult by the high density of narrative description and discussion versus the paucity of accessible measured data in SR-Site. The SR-Site Data Report (TR-10-52) summarises the locations of hydrochemical data sets in Table 6-2 (p189) and identifies the primary source of groundwater analyses data as the ChemNet spreadsheet (as provided to me on request by SKB). TR-10-58 (Salas et al 2010, Hydrogeochemical Evolution report) has appendices containing summaries of modelled data for the various evolution scenarios, but does not have the measured data that are essentially the ‘primary data’.

3.2. Description of initial state for groundwater chemistry

The full interpretation of the initial state is contained in the SDM report (TR-08-05, SDM-Site Forsmark); groundwater chemistry interpretation in that report is abstracted for the initial state description in SR-Site (TR-11-01, Vol. 2, p337). Both the undisturbed natural groundwater condition and the early-stage perturbations due to excavation and construction are presented.

The main perturbation effects are those influencing salinity (upconing of deeper saline water and drawdown of shallow dilute water), redox (oxygen ingress), pH and rock alteration (alkaline grout reactions), organic materials ingress, and colloids generation. Chemical evolution in and around the repository is dealt with mostly by scoping modelling. However it is not clear whether the resulting alterations, for example the effect of alkaline alteration on the sorption and diffusion properties of rock, has been taken into account in the model of radionuclide transport and retardation. Secondary CSH minerals, clays, iron oxide and calcite may have significant effects on retardation around the tunnels and deposition holes (noting that this would depend on the spatial relationship between grouting effects and radionuclide transport pathways). The possibility of dilute water being drawn down into the open tunnels has been simulated with an analytical transport model, for which details are not provided or referenced.

Descriptions of compositional variations for water in the fracture domain bedrock at repository depth in terms of a simple binary system comprising groundwater in transmissive fractures and porewater in the diffusive rock matrix (TR-11-01, pp132-133) are probably over-simplistic. Assuming that deposition holes will be positioned remote from significant fractures, it seems that water that will eventually resaturate emplaced buffer could derive from fractures with very low transmissivity. This type of water may have composition and chemical buffering intermediate between fracture water and pore water. SKB has attempted to sample groundwater in less transmissive fractures but with limited success (see section 4.1). SKB could clarify their reasoning concerning the overall uncertainty in composition of initial state water that will enter the deposition holes and resaturate the buffer.

3.3. Conceptual models for hydrogeochemical processes

The interpretation of measured compositions of groundwaters and the derivation of conceptual models for hydrogeochemical processes are the foundation for modelling the future evolution of groundwater chemistry. In particular, the understanding of presently–active processes and how they might change over time should constrain the variability of the key chemical parameters, i.e. safety function indicators pH, redox, (including dissolved O_2, HS^-, Fe), ionic strength, cation concentrations (M^{2+} versus M^+).

The geochemical equilibria between the major and minor matrix minerals and secondary fracture-filling minerals and groundwaters is not interpreted to a level of detail that is adequate to underpin modelling of future evolution in the system. The omission of silicate alteration reactions such as incongruent dissolution and equilibrium with secondary minerals, and also of cation exchange, means that alternative hydrogeochemical models for the evolution of pH, alkalinity and cation concentrations have not been considered. SKB state that ‘cation exchange processes have not been included……. due to two reasons…… the available CEC values for fracture-filling minerals are very scarce and uncertain….. and the thermodynamic database doesn't have the possibility to deal with cation exchange (TR-10-58, p117; though it is noted on pp31-32 that some scoping calculations with 'deduced' CEC values have been carried out). SKB need to provide more reasoning and evidence that omission of these reactions has negligible significance.

SKB's model for water-rock reaction is limited to the mineral phases calcite, quartz, hydroxyapatite, iron oxide, and iron sulphide as reacting minerals (TR-11-01, p355). The statement (p355) that 'buffer capacity of saline waters (to Na^+ and Ca^{2+}) is such that these elements behave almost as conservative' is inconsistent with the statement (p132) that 'these deep waters show an increase in Ca^{2+} with depth, which is a well-recognised trend and indicative of water/rock interactions that occur under increasingly low flow to stagnant groundwater conditions with increasing depth'. Moreover the statement (p356) '…minerals chosen (for modelling) are a limited subset of those present……and effectively represent the chemical effects observed in reactive components' misrepresents the hydrogeochemistry.

The omission of silicate dissolution from the conceptual model neglects its significance, e.g. silicate reactions are generally understood to be involved in the general long-term evolution of groundwater compositions in deep crystalline bedrock (Nordstrom et al 1989 Groundwater chemistry and water-rock interactions at Stripa, Geochimica et Cosmochimica Acta, 53, 1727-1740; Grimaud et al 1990 Modelling of the evolution of ground waters in a granite system at low temperatures: the Stripa ground waters, Applied Geochemistry, 5, 515-525; Trotignon et al 1999 Equilibrium modelling of Äspö groundwaters, Applied Geochemistry, 14, 907-916), for example being the cause of the evolution from Na-Cl to Ca-Cl compositions in saline waters and brines in some Shield rocks (Pearson 1987 Models of mineral controls on the composition of saline waters of the Canadian Shield, GAC Special Paper 33; Gascoyne 2004 Hydrogeochemistry, groundwater ages and sources of salts in a granitic batholith on the Canadian Shield, Applied Geochemistry, 19, 519-560). SKB seems to acknowledge this in interpreting the trend of increasing Ca^{2+} with depth (TR-11-01, p132).

Equally, omission of cation exchange neglects the role of the sorbing solid phase as a reservoir/buffer of cations that will moderate water compositional changes as the system evolves, for example when dilute water infiltrates and displaces brackish water. Both silicate dissolution and cation exchange will affect the evaluation of the M^{2+}/M^+ (e.g. Ca^{2+}/Na^+) ratio that controls colloid stability.

SKB acknowledges the potential relevance of silicate reactions by stating (TR-11-01, p359) that 'Ca^{2+}….may be released by weathering of feldspar' and 'Mg^{2+}….. is regulated by precipitation and dissolution of chlorite' and acknowledges the potential modelling difficulty 'because of low solubility of chlorite and uncertainty in composition, modelling of Mg is much more uncertain that that of Ca'. SKB also states that 'Reaction modelling in SR-Site is not well suited to constrain K^+ concentration because there is not enough information on possible controlling reactions' (TR-11-01, Vol. 2, p360).

The reason given by SKB for not simulating ion exchange is that CEC data for fracture-fillings are 'very scarce' and have 'important uncertainties' i.e. insufficient data. It also states that the thermodynamic database 'imposed by SKB does not have the 'capacity' for calculations of ion exchange equilibria. SKB should clarify the reason for simplifying the conceptual model and show with scoping modelling that the omitted reactions are negligible with regard to the safety function indicators.

There is an apparent contradiction in assuming that iron-containing silicate minerals (such as biotite and chlorite) react rapidly to release Fe^{2+} in the model for oxygen ingress and attenuation (TR-10-57; see Section 4.7.3 in this report) whereas dissolution of silicate minerals (such as feldspars, micas, etc.) is discounted as having a significant effect on general hydrogeochemical evolution because of slow kinetics.

In this respect, it is noted that SSM commissioned a geochemical modelling study 'Infiltration of dilute groundwaters and resulting groundwater compositions at repository depth' (SSM Research 2011:22, Bath 2011 Infiltration of Dilute Groundwaters and Resulting Groundwater Compositions at Repository Depth). It simulated geochemical reactions of dilute water, focussing on relative concentrations of alkali and alkaline earth cations. Two approaches were studied – with reactions being controlled by kinetics or by local equilibrium. Whilst noting the various limitations of both approaches and not having the scope for full analyses of uncertainties and sensitivities to assumptions and parameters, the SSM modelling study found that the most realistic and likely hydrogeochemical reaction path involving silicate reactions and ion exchange would result in increases of $[Ca^{2+}]$ relative to $[Na^+]$. However it also identified specific hydrogeochemical conditions, albeit unusual, that in theory could result in groundwater compositions that are strongly depleted in divalent cations Ca^{2+} and Mg^{2+}. In some model runs where incongruent dissolution of Na and K feldspars and local equilibrium with various secondary minerals was simulated with pH rising to exceptionally high values, Ca^{2+} and Mg^{2+} decreased sharply whilst Na^+ increased.

3.4. Handling of uncertainties in data and processes

Handling of uncertainties in the reliability and representativeness of groundwater chemistry data and of hydrogeochemical process interpretations is a substantial issue. It is especially pertinent in groundwater chemistry because of the technical difficulties of obtaining representative water samples and because of the complexity of processes which leads to the need for simplification of interpretations and models. There are two main areas of uncertainties: errors in data and model parameters that should be quantifiable, and validity of expert judgement that should be considered in terms of alternative concepts and models.

As a generalisation, the quantification of uncertainties and consideration of alternative interpretations and models is incomplete in some aspects of the hydrogeochemical modelling. An example is in the hydrogeochemical evolution simulation which is done by coupling the transient hydrogeological model of mixing between end-member waters with the geochemical equilibrium model (TR-10-58, Salas et al 2010, Hydrogeochemical evolution of the Forsmark site). Simulations of mixing have been done with various hydrogeological codes, and in terms of either end-member waters (for the open repository and temperate climate stages) or simply dilution of pre-existing salinity (for the remaining parts of the reference glacial cycle). Geochemical reaction simulations have been done with an equilibrium model with alternative controls for redox, iron and sulphide although for sulphide only values from the iron oxide equilibrium variant have been selected because those from FeS equilibrium were judged to be too high (Fig 4-5 & p40, TR-10-58).

The simulation results have been reported in terms of box-and-whisker plots showing statistics of spatial variations of parameters within the candidate repository volume (Figs 10-39 to 10-47, pp358-365, TR-11-01, Vol. 2). These plots show the uncertainties in terms of spatial variability but do not necessarily include other uncertainties due for example to selected values for end-member water compositions and to the restricted nature of the selected geochemical equilibrium model (i.e. iron oxide equilibrium, silicate reactions and cation exchange neglected). Uncertainties due to boundary compositions (e.g. unknown maximum salinity in the deep saline end-member water and the single composition used for altered meteoric end-member water) and due to simplifications in the modelling concept (e.g. water-rock reactions restricted only to calcite, iron oxide, quartz, FeS, hydroxyapatite equilibria) are not taken into account in the box-and-whisker diagrams (as discussed above in section 4.4).

Expert judgement has justified these simplifications in the modelling and there is some discussion of uncertainties (pp116-119), but the possibilities of the alternative models, even though unlikely, and their implications for key parameters have not been fully considered.

SKB has commented that details of expert judgements, i.e. where they have been made and by whom, have been documented on file (TR-11-01, p92). This information should be scrutinised for SSM in future detailed review. The extent to which alternative interpretations and models have been considered should be examined. Examples of where relatively high conceptual uncertainties could be represented by alternative models are: (i) the forecasts of groundwater conditions during the periglacial and glacial stages of the reference scenario, and (ii) the composition and modelled response of the deep highly saline groundwater to a future prolonged period of fresh water infiltration (the ‘global warming variant’), i.e. upconing or dilution.

3.5. Safety functions and groundwater chemistry

Groundwater chemistry data and processes that have direct impacts on the long-term safety assessment are taken into account by the safety functions, and more specifically are evaluated as safety function indicators. Safety functions are subdivided into those relating to containment and those relating to retardation in the geosphere (TR-11-01, Vol. 1, pp252-261). Safety functions for containment concern performance of the engineered barriers (EBS) and the rate of fuel dissolution, and might therefore be considered to be outside the scope of this review except that the chemical conditions in the EBS will be controlled by groundwater chemistry in the long term. Safety function indicators in this context have chemical criteria attributed for:

- Corrodants O_2 and HS^- should be “low”,
- Chemically “reducing” conditions for exclusion of O_2 and also for slow fuel dissolution and low radionuclide solubilities,
- Ionic strength, I, below upper limits for maintaining buffer and backfill properties,
- pH within a central range that would not promote corrosion,
- Cl^- below an upper limit so that chloride-assisted corrosion will not be promoted,
- Sum of cations, $\sum q[M^{q+}]$ above a lower limit to destabilise colloids originating from erosion of buffer, and thus to prevent colloid-facilitated radionuclide transport,

- Concentrations of certain 'detrimental agents' (in addition to O_2 and HS^-) should be "low", i.e. nutrients or energy sources (H_2, CH_4, DOC) that would be used by sulphate-reducing bacteria (SRB) to produce HS^-, and cations that might reduce alteration of buffer and backfill (K^+, Fe^{2+}).

The safety function indicator criteria that relate to groundwater chemistry are partly well-defined, e.g. for pH, Cl^- and $\sum q[M^{q+}]$, and partly loosely-defined, e.g. for O_2 and HS^-, I, H_2, CH_4 and DOC, and K^+ and Fe^{2+}, most of which are specified only to be "low". The requirement for 'chemically reducing' conditions rather than defining a threshold Eh or some other redox indicator is another loosely-defined criterion. SKB discusses the rationale for these criteria (TR-11-01, Vol. 1, pp252-260) and the arguments are compelling, for example "the presence of reducing agents (e.g. Fe^{II} and HS^-) is sufficient to indicate reducing conditions" and "negative Eh is not always well-defined and thus less useful as a basis". It is unclear how these qualitative criteria are applied transparently, i.e. what is a 'low' value and what is an unacceptably 'high' value (this raises the question of how anomalous data have been handled).

An additional category of detrimental agents that are not mentioned in the Main Report of SR-Site (TR-11-01) is the N and organic compounds (nitrite, ammonia, acetate) that would potentially enhance some copper corrosion processes, specifically stress corrosion cracking (SCC). These SCC-promoting compounds are by-products of microbial processes on nitrogen compounds and organic carbon in near-field groundwater. N solutes and organics potentially have natural and anthropogenic sources at the ground surface, natural sources in deep rocks and also are likely to derive in a future repository from introduced materials. SKB has not assigned quantitative criteria for these species but has sparse analyses for existing quantities and has modelled possible amounts during future evolution (TR-10-58: p15, pp92-96, Fig 8-2 & pp102-103, Fig 8-5).

In deep reducing groundwater, the most abundant of the N species is likely to be ammonium (NH_4^+) and this is reflected in the measured data (TR-10-58, Fig 8-5) which are between 10-1 and 10-4 mM, decreasing with depth down to 1000m. SKB report anomalous concentrations of NH_4^+ up to 33.2 mM at Laxemar (TR-10-58, p102), though this value is not shown on Fig 8-5. SKB should clarify whether this high value is credible and, if so, whether a deep natural source of N, e.g. from NH_4 contained in mica minerals, is possible for Forsmark. In SR-Site, SKB attributes NH_4 primarily to the Littorina marine water source on the basis of the decrease with depth. Evidence of a potential natural geological source in rocks would indicate the need to survey N contents of rock and specific mineral phases, and to consider future evolution paths for N in near-field groundwaters.

In addition to reducing conditions in the near field being a safety function indicator because of the necessity to lower radionuclide solubilities, concentrations of hydrochemical species that form complexes with some radionuclides also need to be low. These species are primarily phosphate and carbonate. This is not mentioned specifically in the SR-Site Main Report (TR-11-01) but both phosphate and carbonate (as inorganic carbon) are explicitly considered in present-day groundwaters and modeled on the basis of hydroxyapatite and calcite equilibrium respectively for future groundwaters in the Hydrogeochemical Evolution report (TR-10-58: pp38-40, Fig 4-4; p52, Fig 6-9; and Figs 6-13 & 6-14, p55).

The safety function indicator criterion relevant to destabilising colloids formed by buffer erosion is $\sum q[M^{q+}]$. This parameter and the quantified criterion ($\geq$ 2-4 mM) is assumed by SKB to be equivalent to a criterion based divalent cations only, $\sum[M^{2+}]$. The reasoning for the criterion is set out in the Buffer, Backfill and Closure Process report (TR-10-47, pp151-160) and originates in DLVO theory for colloidal stability and the concept of a critical coagulation concentration (CCC) for colloids with monovalent cations which predicts that colloids would be stable up to Na^+ concentration of about 25 mM. Incorporation of divalent cations into the criterion is done by incorporating experimental evidence that relatively low concentrations of Ca^{2+} have a strong destabilising effect.

The cited experimental evidence suggests that colloids are likely to be destabilised if $[Ca^{2+}]$ is above about 4 mM, and if $[Ca^{2+}]$ is below that there are $[Na^+]/[Ca^{2+}]$ ratios (roughly exceeding 4-5 as a molar ratio, for which Ca fraction in the clay will be lower than 0.9) for which colloids will be stable as long as $[Na^+]$ is <25 mM (TR-10-47, Fig 3-29, p153). It is unclear why SKB has simplified the safety function indicator criterion concerning colloid stability to the sum of all cations, i.e. $\sum q[M^{q+}] > 4$ mM in place of considering separately the sum of divalent cations $\sum[M^{2+}]$ and the sum of monovalent cations $\sum[M^+]$. The threshold for $\sum[M^{2+}]$ suggested by the empirical evidence seems to be > 4 mM, but > 1 mM is quoted in TR-10-58 (p13). The threshold for $\sum[M^+]$ which relates directly to the DLVO theoretical basis is > 25 mM, though >100 mM is quoted in TR-10-58.

The safety function indicator relevant to maintaining swelling pressure of the buffer is ionic strength, I. The basis for using I as the indicator is the variation of osmotic pressure across the pore-water/groundwater interface of the compacted bentonite. At the specified compaction density, the osmotic pressure drop that can be tolerated for maintaining a swelling pressure of 1 MPa is stated by SKB to be equivalent to a salinity step of 1.7M NaCl (ca 100 mg/L NaCl; Buffer, backfill and closure process report for SR-Site, TR-10-47, p20). However SKB does not give a specific criterion, stating that 'the limit of tolerable ionic strength is highly dependent on the material properties of the buffer' (TR-11-01, Vol 1, p259). An upper limit of 0.6M NaCl (ca 35 mg/L) is similarly derived for maintaining backfill swelling, but is also not given as a specific criterion. The calculated pressures appear to be consistent with experimental observations on bentonite samples (TR-10-47: Figs 3-12 & 3-13, p88 and Fig 3-21, p137; Karnland et al, 2005, 2006) which also reflect qualitatively the decreasing swelling pressure as salinity of external solution is increased. The uncertainties in all of this, as SKB admits in stating 'uncertainties still remain as to how this activity should be treated in a relevant way' (TR-10-47, p139), are unclear although it can be noted that SKB's approach appears to be pessimistic because the compacted buffer is assumed to behave as an ideal osmotic membrane.

The safety function indicators concerning groundwater chemistry for retardation are (TR-11-01, Vol. 1, p261):

- Reducing conditions,
- Ionic strength,
- Matrix diffusion and sorption (D_e and K_d),
- Low colloid concentration.

SKB do not give quantitative criteria for these safety function indicators. It is therefore unclear how SKB make consistent assessments against these indicators without explicit criteria. One of the main hydrochemical factors affecting these indicators is the complexation and speciation of radionuclides and this should be reflected in the safety function indicators. For example, bicarbonate concentration, and also other anions such as sulphate and phosphate, will affect the ionic speciation of radionuclides and thus potentially matrix diffusion and sorption properties also.

3.6. Near-field geochemical and microbial processes

3.6.1. Redox conditions, dissolved oxygen, sulphide

Groundwater chemistry data and the underlying processes in the near field for redox conditions, including concentrations of the corrodants oxygen and sulphide are key elements of SR-Site. The two corrodants could exist only in contrasting redox conditions – oxidising and reducing respectively. Consumption of residual oxygen in the repository in the early post-closure stage and attenuation of dissolved oxygen in infiltrating groundwater before it reaches repository depth are the two processes that will sustain ‘reducing conditions’ (safety function R1a).

Interpretation and understanding of measurements in the present-day system underpin confidence in forecasting of future evolution. Actual measurements of dissolved O_2 (DO) data are not presented in SR-Site because, as I understand it, of the difficulty of making measurements of low or zero DO consistently without contamination by O_2 introduced as an artefact of drilling and sampling. However there are a few ‘selected’ zero or near-zero DO data in SKB’s hydrochemical data spreadsheet (Forsmark_ 2.3_ updated Dec_3_2007.). SKB should be asked to clarify the position on DO data, i.e. the reliability of DO data in the database and the reason for excluding discussion of these.

In view of redox being the ‘proxy’ for DO in safety function R1a, there needs to be a clear and quantifiable set of criteria for judging whether chemical conditions, now and in the future, are and will be ‘reducing’. If measured and/or modelled Eh is not sufficient because it is an operational parameter with complex electrochemistry, then it is unclear how the ‘reducing’ character is quantified by other redox parameter such as HS^- and Fe^{2+}. Measured Fe^{2+} concentrations generally decrease with depth from 10^{-1} to $<10^{-2}$ mmol/L (Fig 5-6, TR-10-39, p47) so the measured range is rather narrow and subject to various analytical and geochemical factors. SR-Site mentions potential variability of the strength of reducing conditions, but an explanation of what might vary, i.e. Fe^{2+}, HS^-, DOC, is needed, and what will be the significance of those variations in terms of near-field hydrogeochemistry and behaviour of radionuclides.

Measurements of HS^- at around repository depth are reported and interpreted in TR-11-01 (Vol.2, p360) and in TR-10-39 (Sulphide Content in the Groundwater at Forsmark). It infers that varying HS^- concentrations in water samples originate from biogeochemical reactions in stagnant borehole water that is progressively flushed as pumping proceeds (TR-10-39, pp24-25 and Fig.3-5). This interpretation is a compelling explanation of anomalous HS^- values as an artefact of within-borehole biogeochemistry. Some analyses of HS^- exceed the values corresponding to FeS equilibrium (Fig 3-22, R-08-86, Gimeno et al 2008 Water-rock interaction modelling and uncertainties of mixing modelling). It is noted that the more recent high HS^- analyses (in TR-10-39) were not available for R-08-86.

SKB assume that ‘precipitation of amorphous monosulphide is very rapid and a groundwater showing significant supersaturation of this phase cannot exist…… it therefore indicates a serious analytical error in either the sulphide or ferrous iron analyses, or both’ (TR-10-39, p33). This assumption underlies the ‘selected set of sulphide concentrations’ that is shown in Fig 6-15 (TR-10-58, Salas et al 2010, p56). However, discussion in R-08-86 suggests that ‘if Fe^{2+} is not available, S^{2-} increases in solution and the monosulphide precipitation is inhibited in spite of the SRB activity' and 'active precipitation of iron monosulphide seems to be much less important in Forsmark than in Laxemar, and it can be due to the absence of a source of iron….'. SKB also state that "actual precipitation of FeS(am) has not been possible to demonstrate " (TR-10-39, p46).

SKB's discounting of some high HS^- concentrations, on the basis that they exceed FeS equilibrium, is not sufficiently justified. SKB seems to infer that HS^- analyses or corresponding Fe^{2+} concentrations are unreliable. More information is required about the number of analyses that have been discounted in this way, and whether there are independent reasons to reject the analyses in these cases. SKB should provide additional evidence to justify the assumption that FeS is the mineral phase controlling HS^-.

SKB's conceptual model for HS^- production implies that DOC (dissolved organic carbon) is the energy source for the relatively rapid biogeochemical reduction of SO_4. Information is needed on potential sources of the DOC in the sampling and monitoring installations, to underpin a model for the potential production of anomalously high HS^- concentrations during the initial stage and long-term evolution. Some open issues are: (a) whether a source of labile DOC could be either 'introduced' substances (i.e. construction/operation contaminants in tunnels) or natural DOC, (b) whether there are particular types of natural DOC that would be particularly labile/reactive, (c) whether there is a threshold concentration of labile DOC below which biogeochemical HS^- production will be negligible, and (d) whether existing or future levels of dissolved methane, CH_4 (presumably abiogenic), will act as the energy source for SO_4 reduction (i.e. anaerobic methane oxidation).

Most of the DOC is said to be large molecule organic carbon that is not biogeochemically available to SRB (TR-11-01, Vol, 2, p361 and Fig. 10-43). Is this assertion supported by specific data from the SDM? SKB states in TR-10-58 (Chap. 8, p89) that "important uncertainties remain in the degradation rate of organic C" and also that the acetate component of DOC (which is important as an intermediate in microbial processes) has been analysed only in groundwater at Laxemar. Acetate data and DOC characterisation would improve the biogeochemical interpretation of Forsmark groundwaters at repository depth. This are potentially significant issues because future variations of DOC may be the controlling factor in HS^- production (unless CH_4 or H_2 are energy sources for reduction).

SKB notes that "DOC, SO_4, HS and $\delta^{34}S(SO_4)$ are poorly correlated with the numbers of SRB microbes" (TR-10-39, p54), and SRB numbers scatter from 0.1 to 1000 MPN cell/mL (Fig 5-13, p54). It seems that not much is learned about the relationship between SRB and HS^- production that will increase confidence in predictability of HS^- concentrations. Discussion of microbial processes in TR-10-48 (Geosphere Process Report, p169) is rather general and is not focused on the relevant processes. Constraints due to CH_4 and/or H_2 are mentioned only as a 'model simplification' (TR-10-48, p18). The potential role of DOC as the dominant constraint on microbial activity appears to be discounted.

In situ SO_4 reduction rates are estimated to be 10^{-6} to 10^{-8} mM/year (p55 and Appendix 3, TR-10-39). These look reasonable values by comparison with the quoted higher (by 2-3 orders of magnitude) reduction rates estimated in studies of deep sedimentary aquifers, but their reliability needs to be assessed.

The reasoning that future concentrations of HS^- in the near field and geosphere will remain around present levels (i.e. slightly higher for marine water influx and lower for glacial melt water influx) seems to involve both biogeochemical kinetics and mineral equilibrium (TR-10-39). Detailed review of this by SSM, and also of predictions of future CH_4 and DOC is suggested (Table 6-1, p60, TR-10-39).

3.6.2. pH and alkalinity buffering

The model proposed in TR-11-01 (Vol. 1, p133) is that pH and HCO_3^- are buffered by calcite equilibrium. As discussed above in the section on conceptual models for hydrogeochemical processes, silicate dissolution reactions are omitted from the hydrogeochemical model. pH and HCO_3^- are influenced by silicate dissolution, so their buffering is not comprehensively represented by the present model.

3.6.3. Hydrogeochemistry of major cations

Concentrations of major cations, Na^+, Ca^{2+} and Mg^{2+} are an implicit constraint for safety function R1c, for which the safety function indicator criterion is $\sum q[M^{q+}] \leq 2$-4 mM (see discussion in section 4.5).

SKB's hydrogeochemical modelling simulates long-term temporal and spatial evolution of the major cations by simple mixing of end-member waters with a limited range of mineral equilibrium constraints, primarily that of calcite equilibrium on Ca^{2+}. A more complete range of water-rock reactions affecting Na^+, Mg^{2+} and K^+ as well as Ca^{2+}, i.e. dissolution and precipitation of silicate minerals and cation exchange, are not simulated (see discussion in section 4.3).

The hydrogeochemical conceptual model in TR-10-58 (section 2, pp17-21) has a rather cursory interpretation that focuses on mixing of different water masses and neglects the water-rock equilibria other than the simplest representation of calcite, iron oxide, iron sulphide and hydroxyapatite equilibrium. Section 4 of the same report states that "chemical reactions are needed to obtain information about non-linear parameters and behaviour of other parameters mainly controlled by equilibrium reactions".

3.7. Long-term evolution of groundwater chemistry

The spatial discretisation of modelling of the hydrogeochemical evolution of the varying groundwater mixtures in future climate conditions (TR-10-58, Hydrogeochemical Evolution report, Salas et al 2010) is determined by the discretisation of the hydrogeological model which is disproportionately detailed in relation to the inherent uncertainties in forward modelling of hydrogeochemistry. Uncertainties in the evolution modelling are considered in Section 9.5 of TR-10-58. SKB identify the assumed compositions of end-member water compositions for the mixing model as a substantial source of uncertainty. That issue has been addressed in R-08-86 (SDM-Site, Water-rock interaction modelling and uncertainties of mixing modelling; Gimeno et al 2008). In particular, alternative compositions for the deep saline end-member water plus an additional 'old meteoric' end-member water have been considered and a sensitivity analysis for hydrogeochemical model results has been carried out by Monte Carlo modelling.

The concluding section of R-08-86 outlines the preferred conceptual model for hydrogeochemical evolution and seems to have been carried forward into TR-10-58.

One of the most significant uncertainties in the evolution lies in the composition assumed for the deep saline end-member water. The composition of the most saline sample from borehole KLX02 at Laxemar has been used, but with a low SO_4^{2-} content (TR-10-58, Table 4-2, p37) based on the observed low SO_4^{2-} found in deep saline waters at Olkiluoto. Whether SO_4^{2-} in future deep saline water will have low SO_4^{2-} as assumed or higher SO_4^{2-} as presently observed is a significant uncertainty because, with a diminishing content of Littorina water, the deep saline end member will increasingly be the dominant source of SO_4^{2-} and thus of HS^- at repository depth.

It can be argued that the deep saline water at Olkiluoto is the better analogue on which to base the deep saline end-member composition for Forsmark, so there would be additional uncertainty arising from the Olkiluoto water having higher Cl^- than the deep saline end-member Cl^- that is presently assumed. There is thus potentially more significant uncertainty in the future water compositions arising from these sources than from spatial variability that has been modelled in such detail in TR-10-58. SKB could be asked to clarify their position and their lines of argument to justify that the present modelling is adequate.

3.7.1. Temperate climate conditions

Evolution of near-field groundwater compositions in the initial period after closure is simulated with an analytical model for dilute water infiltration using flow-related migration properties (TR-11-01, p339). Additional information is required on whether the model has been verified, and on how it has been tested to be adequate for this purpose.

General evolution of near-field groundwater compositions in the temperate period is modelled up to 7000 or 5000 years in the future (TR-11-01, Vol. 2, p343 & p347). The simulation is run forward from 10000 years ago. Why the forward modelling for the reference evolution goes to only 7000 years is not explained, though it is noted that forward modelling of the ‘global warming’ variant scenario is extended to 60000 years of temperate conditions.

There are various assumptions and simplifications in the modelling of dilute water infiltration. For example, the assumption that matrix and fracture waters have equilibrated salinities at the start is a simplification of the present-day heterogeneity between matrix and fracture waters (TR-11-01, Vol.2, p347). The output from the model is the proportion of deposition holes for which adjacent fracture water goes to $\leq$ 10% of initial salinity over the modelled period. The modelling simulates the hydraulic constraints on long-term dilute water penetration in the system, i.e. the stratification of water salinities and the ‘fingering’ of dilute water downwards in transmissive zones. The model indicates that only ~8% of deposition holes would be diluted to $\leq$ 10% (i.e. $\leq$ 1 g/L TDS) after 1 million years, i.e. that the maximum penetration depth of dilute water is ~500 m regardless of time (Fig. 10-32, p348, TR-11-01, Vol. 2). The sensitivity of this model to parameter values, geometrical assumptions and the simplification of compositional heterogeneity between fracture waters and matrix waters should be tested and understood comprehensively, because the most likely scenario is that there will be a prolonged period of fresh water infiltration.

Evolution of groundwater chemistry in the near field is stated to be modelled by “loose coupling” which “allows description of geochemical heterogeneity” (p355, Vol. 2, TR-11-01). To what extent are heterogeneous water-rock reactions modelled, or is it simply calcite, FeS and Fe-oxide equilibria? SKB has not included dissolution of silicate minerals and cation exchange in its hydrogeochemical modelling, as discussed in sections 4.3 and 4.6.3.

Concerning the evolution of redox, the model assumes redox control either by Fe-oxide or by Fe-sulphide mineral phases (TR-11-01, Vol. 2, p364). Modelled Eh values are given for both alternatives, without discussion of which redox regulation reaction is likely. SKB reports that Eh will change as groundwater at repository depth is progressively diluted from ‘a mean value’ of -230 mV to -190 mV (TR-11-02, Vol. 2, p365). Additional information would clarify what data have been averaged to obtain these values and whether the change of modelled Eh is primarily dependent on pH change (i.e. because Eh and pH are coupled via the redox half-reactions) or is reflecting modelled changes of other redox variables.

3.7.2. Periglacial climate with permafrost ground conditions

There is quite large conceptual uncertainty of how the groundwater system under permafrost will evolve. This is significant because periglacial conditions may persist for relatively long periods in the next 10^4-10^5 years. SKB asserts that the small number of studies of groundwater chemistry underneath permafrost tend to show that it is not much affected by permafrost (TR-11-01, Vol. 2, p511). More information is required about what was found in those studies and SSM could assess how meaningful and relevant it is for long-term evolution at Forsmark.

3.7.3. Glacial climate with ice sheet advance and retreat

SKB reports that evolution of groundwater through periglacial and glacial conditions is modelled for a 19000 year period (TR-11-01, Vol.2, p489). It is unclear why that duration was selected for modelling. The modelling shows saline water being flushed upwards by the intrusion of glacial melt water so that higher salinity occurs near the ground surface under permafrost (p493 and Fig. 10-130, TR-00-01, Vol. 2). SKB could clarify whether this is based only on modelling or whether there is field evidence of this having occurred in the last glaciation (the topic is referenced to King-Clayton et al., 1997). It is unclear whether saline upconing has been taken into account in the hydrochemical evolution modelling (p502, TR-11-01, Vol. 2).

The model assumes that melt water infiltrates under an enhanced hydraulic gradient only for the short period when an advancing or retreating ice front passes over the repository location, assumed to be 20 or 100 years. If that is the case, then more information is needed on what modelling indicates about evolution of the groundwater system during the remainder, major part, of the glaciation which could be several tens of thousands years, i.e. >19000 years.

SSM commissioned its own modelling study of possible evolution pathways for the composition of dilute melt water infiltrating towards repository depth. A fairly wide range of cation proportions, pH values and alkalinity concentrations were found to be possible. SKB have assumed that melt water will have relatively high pH (~9) due to “weathering of bedrock minerals”, though this has not been modelled. SSM’s modelling suggests that there are various possible hydrochemical evolutions that are outside the variations assumed in SR-Site, as discussed in section 4.3 (SSM

Research 2011:22, Bath 2011 Infiltration of dilute groundwaters and resulting groundwater compositions at repository depth).

SKB states that there is a "large degree of uncertainty in the geochemical modelling results of Salas et al (2010; TR-10-58)" which are represented in Figs. 10-151 to 10-153 in TR-11-01. These simulated groundwater compositions are dependent on the assumptions and simplifications for modelling. For example, $\sum$(cations) in Figs. 10-151 and 10-152 goes as low as 10 mM, and the modelling might indicate lower values if alternative assumptions are used.

SR-Site states that it "cannot be excluded that colloids may be generated and transported" in dilute waters during glacial periods. A low limit on viable colloid concentrations is based on what has been reported for groundwaters at the Grimsel site in Switzerland (TR-11-01, Vol. 2, p520). The conditions that control the low concentrations of colloids are not explained and, in general, there is insufficient information to underpin a claim that colloid concentrations would therefore always be low and relatively immobile in Forsmark groundwaters.

The oxygen infiltration scenario is dealt with in SR-Site (TR-11-01, Vol. 2, p. 521-524 & p533 for the reference evolution, and TR-11-01, Vol.3, p603-605 for variant scenarios). Processes and modelling of oxygen attenuation in the geosphere is covered in detail in TR-10-57 (Sidborn et al, 2010, Oxygen Ingress in the Rock at Forsmark During a Glacial Cycle). SKB concludes, based on the geochemical model for O_2 attenuation and the groundwater flow model for subglacial conditions, that "corrosion due to O_2 penetration with diffusive conditions in the deposition hole can be neglected" and for advective conditions in the deposition hole that "effects of O_2 penetration can be excluded from the corrosion scenario" (p605-606, TR-11-01, Vol. 3).

SKB's conclusion regarding oxygen ingress is compelling, dependent on the adequacy (as judged by corrosion experts) of the simple mass balance approach to oxygen corrosion of the canister (p315, TR-11-01, Vol.2) and on the outcome of detailed review of the groundwater modelling and the conservatism in the number of canisters affected. SKB should have modelled a number of variant models for groundwater flow and O_2 transport under glacial conditions instead of presenting what seems to be a single pessimistic model. In view of the importance of the O_2 ingress scenario, SKB should provide more information about the groundwater flow model to supplement the otherwise well-documented hydrogeochemical modelling of O_2 attenuation in TR-10-57.

The 'F factor' (flow-related transport resistance parameter, incorporating flow-wetted fracture surface area and advective travel time) is used as a proxy for accessible fracture surface and rock matrix in the kinetics formulation. This approach assumes that the physical properties incorporated by the F factor are as representative for the consumption of oxygen as for radionuclide retardation, i.e. that both processes are dependent on simple matrix diffusion. That is a reasonable assumption. The assumed equivalence of the two types of matrix diffusion and reaction is likely to be valid if it is being applied to modelling the same type of rock domain, i.e. the bedrock between the deposition holes and the nearest major hydraulic conductor such as a deformation zone. The modelling illustrates a pessimistic set of assumptions for F and for the geochemistry of O_2 consumption reactions, whilst only doing a limited sensitivity analysis.

The O_2-ingress issue was assessed by SSM's more limited modelling study (Chapters 6 & 8 in SKI Report 2008:16 Review of SKB's Safety Assessment SR-Can: Contributions in Support of SKI's and SSI's Review by External Consultants). The outcome was generally similar.

3.8. Integration with hydrogeology and palaeohydrogeology

This section is concerned with the use of hydrochemical information as a semi-quantitative or at least qualitative consistency test of the interpretations and models of present and past groundwater movement in the system. The main approach uses hydrochemical indicators of mixing between different end-member water masses, from different sources and with different ages, to calibrate the transient or 'palaeohydrogeological' model of groundwater flow. This has been done by resolving the measured groundwater compositions into the component waters with the 'M3' multivariate analysis tool.

SKB have documented the method and the M3 tool and have studied sensitivity to various assumptions and parameters such as compositions of end-member reference waters in TR-09-09 (Laaksoharju et al 2009, M3 User's Manual, v3.0), TR-09-05 (Gomez et al, 2009, M3 v3.0, Verification and Validation), and R-08-86 (Gimeno et al 2008, Water-Rock Interaction Modelling and Uncertainties of Mixing Modelling, SDM-Site Forsmark). Not much of this detailed background is provided in SR-Site, even though the M3 analyses and choice of end-member water compositions are substantial sources of uncertainty in the integrated modelling. The validity and robustness of the M3 method should be checked by SSM along with the propagation of uncertainties from hydrochemistry through M3 to the calibration of the flow-transport model.

The palaeohydrogeological model has been calibrated by simulating the hydrodynamic mixing of water masses (i.e. the end-member waters) by forward modelling from hypothetical initial conditions up to the present day, and then comparing the modelled compositions with observed compositions. The original work is reported in R-08-23 (Follin et al 2008 Conceptual Model Development and Numerical Modelling using ConnectFlow, Forsmark, Stage 2.3) and summarised in TR-08-05 (Site Description of Forsmark at Completion of Site Investigation Phase, SDM-Site). Comparison of modelled and observed data is not facilitated by Figs. 4-22 and 4-23 in TR-11-01 (Vol 1, pp 133 & 136) as SKB suggests. Neither figure seems to show the palaeohydrogeological modelling output and both figures, at differing scales, seem too schematic to represent adequately either measured data or modelled output. There is a lot of 'illustrative' detail in the variations of pore water compositions in Fig. 4-22 that is not supported by actual data. SKB should clarify what these cross-sections are intended to show and should provide additional information on the palaeohydrogeological calibration as used in SR-Site.

A significant aspect of the modelling is the contrast between hydrochemical evolution of fracture waters and pore waters (the model includes diffusive exchange between the two types of porosity). The contrast is most marked in the NW part of the cross-section where dilute fracture waters occur only to < 100 m, whereas dilute (< 1500 mg/L) pore waters occur to much greater depth. There are several uncertainties that need to be taken into account in the model interpretation: (i) uncertainty in the composition of present-day fracture water below 600 m in the NW area of the site, (ii) uncertainty in the distribution of compositions for the initial state, i.e. 10000 years ago, (iii) the paucity of pore water compositions data and the uncertainties in those data, and (iv) the transient changes of boundary water compositions through the modelled period.

3.9. Integration with radionuclide transport and retardation

Interpretations of groundwater chemistry in terms of solute transport and of retardation processes should support the conceptual models that underlie the model for radionuclide transport and retardation. Of particular interest in this respect is the evidence that solute diffusion into the rock matrix is occurring.

The first line of evidence is the distribution of pore water compositions and the relationship to fracture water compositions. Cl^- concentrations in pore waters to at least 640m depth are evidently not in diffusive exchange equilibrium with adjacent fracture waters (TR-11-01, Vol. 1, p132). Another observation is that pore water compositions in footwall bedrock, i.e. in FFM01 fracture domain, have a different character from those in hanging-wall bedrock. The concentrations of the natural isotopic solutes ^4He, ^{14}C and ^{36}Cl in fracture waters are also potentially pertinent as indicators of matrix diffusion as a solute retardation mechanism.

The section in SR-Site on ‘bedrock transport properties’ (TR-11-01, Vol. 1, p136) contains a brief description and justification of the conceptual model for solute retardation. The Radionuclide Transport report (TR-10-50) discusses processes and site characteristics affecting retardation in section 2 and has a comprehensive justification of the assumption of linear sorption, i.e. use of the K_d parameter and discussion of the conditions under which that assumption may be inadequate.

Of relevance to the scope of this review, it identifies that potential non-linearities of sorption might occur if groundwater compositions change, for example with respect to redox conditions or complexing solutes (pp25-27, TR-10-50). The potential impact of changing groundwater chemistry explicitly associated with a glacial climate stage on various processes of radionuclide retardation, i.e. ion exchange, surface complexation and co-precipitation, is considered in detail (pp27-32 & Appendix B, TR-10-50). This seems to be an authoritative review of processes and hydrogeochemical factors that might affect the robustness of the assumption that the use of the K_d simplification in the radionuclide transport model will always be a pessimistic estimation of retardation.

Processes such as sorption and irreversible retention on secondary minerals in fractures are apparently not taken into account in the retardation model for transport in the geosphere (although it is noted that co-precipitation of Ra with $BaSO_4$ is taken into account in modelling of transport of Ra out of the canisters; Data report for SR-Site, TR-10-52, p103; Assessment of Ra-Ba co-precipitation and its potential influence on the solubility of Ra in the near field, TR-08-07, Grandia et al 2008). Neglecting these processes is assumed to be ‘cautious’, though it is noted that there may be future hydrochemical conditions in which radionuclides that have been sorbed or co-precipitated with fracture minerals would be remobilised (R-10-48, Crawford 2010, p55).

SKB discusses this potential complexity in the radionuclide transport model. Scenarios can be envisaged for which the simplified retardation model might not invariably be conservative. For example, step changes in hydrochemical conditions (e.g. dilute water influx dissolving secondary carbonates or sulphates, or a redox change causing iron oxides to dissolve) could cause a ‘bulk’ release of radionuclides that had been retained in this way (TR-10-52, R-10-48 and TR-10-50, Radionuclide Transport report, pp26-27). Radionuclide retention in the host rock immediately adjacent to deposition holes in the event of one of the release scenarios occurring, e.g. shearing or corrosion, could involve various hydrochemical changes that could promote secondary minerals to be formed and in which radionuclides could be co-precipitated. Geochemical analogues for secondary mineral formation and incorporation of trace metals, and for their subsequent remobilisation, need to be considered.

A detailed review by SSM is suggested, focusing on these issues, i.e. heterogeneity and evolution of groundwater compositions, hydrogeochemical processes and secondary mineral evolution, and their potential impacts on radionuclide retardation. Data for background concentrations and hydrogeochemical behaviour of naturally-occurring radionuclides and other relevant analogue trace solutes, and the significance with respect to the retardation model (see pp26-27, TR-10-50), should also be reviewed to assess the conservatism of the simplified model.

3.10. Hydrogeochemistry of radionuclide analogues

General confidence about radionuclide behaviour in the geosphere around a repository can be supported by hydrogeochemical data for trace element solutes that are analogues for radionuclides, and an interpretation of the natural distribution of these solutes between groundwater and the rock minerals. Relevant trace solutes are (i) the natural radionuclides, U, Th, Ra and ^{14}C; (ii) stable elemental homologues for fission products, I, Cs, Sr, Ni; (iii) trace elements that are analogues for transuranics, e.g. the rare earth elements (REEs).

In previous reports (e.g. SR-Can) SKB has considered groundwater data for all or most of these analogue solutes and, with an exception for some anomalous U data, has concluded that the available data are essentially uninterpretable because data are below analytical detection limits, there are artefacts of contamination or perturbation during sampling, and corresponding data for exchangeable element abundances in mineral phases are not available. Although SKB’s position is well-based because it is challenging to get data and to make meaningful interpretations for the hydrogeochemistry of these analogue elements, a lack of such evidence for matrix diffusion, sorption and long-term retention processes is a gap in the scientific support for the geosphere concept underlying the safety case.

An exception to the general lack of interpretable data for analogue elements is the fairly comprehensive study of occasional anomalously high localised concentrations of dissolved U. It is evident from mineral-specific analyses that these dissolved U anomalies are related to U enrichment in some fracture surface coatings (TR-11-01, Vol. 1, p121). SKB concludes that this points to a specific geochemical process in the past that has immobilised U from circulating groundwaters and sustains these anomalous dissolved concentrations but only locally. Various factors may be implicated in the process (e.g. amorphous solid phase co-precipitating U, weakly reducing Eh, dissolved carbonate complexation) but SKB leaves the discussion open as to whether there are significant indicators of how repository-derived U and other radionuclides will behave. It seems to be the case that simple matrix diffusion/sorption is not an adequate conceptual model for U retention, at least (TR-11-01, Vol.1, p135).

Concerning support to SR-Site from natural analogue studies in the wider sense, more information is needed about how the findings of the studies (TR-11-01, Vol. 3, p793) have been incorporated into the conceptualisation and modelling approach for the SR-Site model.

4. Recommendations to SSM

Based on my initial review of the sections of the reports listed in Appendix 1, primarily concerning groundwater chemistry, I recommend to SSM that:

- In SR-Site reports (plus SDM-Site report), SKB has in general produced a comprehensive compilation of information about groundwater chemistry and has interpreted it so that it can be used in the safety assessment.
- From the perspective of groundwater chemistry, I do not at this initial stage identify any issues that contradict the outcome of SKB's assessment of long-term safety.
- Groundwater chemistry interpretations in the SR-Site reports are mostly rather 'detached' from the data that were acquired from site investigations and that were reported in the site descriptive model reports (SDM-Site). That is a normal outcome of the report hierarchy. SSM needs to be satisfied that all the hydrogeochemical characteristics described in the relevant SDM-Site reports are taken account of in the interpretations used in SR-Site. Heterogeneity of groundwater compositions and its implications for chemical conditions at repository depth and for groundwater movements at various spatial scales and time scales should be adequately represented in SR-Site.
- Much of the hydrochemical data that are inputs to SR-Site is derived from model calculations. For example, initial state water compositions around a repository have been modelled to simulate the effect of dilute water infiltration. Future evolution of water compositions has been simulated by coupled mixing and geochemical reaction modelling (noting that I have reservations about the restricted scope of the water-rock reactions involved). Modelling of the hydraulic and hydrogeochemical processes has many sources of uncertainties. SSM should assess whether uncertainties have been realistically propagated through the modelling and that the modelled data have realistic uncertainties attached to them.
- Both measured and modelled hydrochemical data, in a range of categories, are used in SR-Site as quantitative safety function indicator criteria or as loosely-defined criteria. SSM should satisfy itself that these various criteria are appropriately specified and can be applied consistently to groundwater chemistry data in SR-Site and in any subsequent underground investigations.
- The gradients of increasing groundwater salinity, and patterns of changes in other hydrochemical properties, in rock below repository depth are represented by sparse data so that there are uncertainties that are unlikely to be narrowed by measurements in existing boreholes. SSM needs to understand the uncertainties and be satisfied with SKB's assessment of

possible variations of compositions in these deep locations and of potential implications.

- Hydrochemistry and palaeohydrogeology of pore waters in rock matrix of the fracture domain bedrock units have been interpreted from fairly recent and innovative experimental data. The uncertainties in these data and their significance are active research issues. SSM needs to consider whether the significance attached to the interpretations is reasonable and robust in view of the limited base of research and applications of the data acquisition methods.
- Microbiological data are hardly evident in SR-Site although interpretation of biogeochemical processes potentially lies behind the understanding of potential redox variability and of corrodant abundance. There is a substantial amount of microbiological data in SDM-Site from which clear patterns and interdependence with chemistry of redox-active solutes are not really evident. SSM should be satisfied that the interpretations and any biogeochemical model for future evolution have been developed to an appropriate level by SKB.
- Close integration and exchange of information across interfaces between disciplines in SSM's review project will be desirable in future stages of this review process. From the perspective of the role of groundwater chemistry in SR-Site, the important interfaces are with the EBS system (for long-term water compositions and reactions with minerals in the EBS and near field) and with site-scale hydrogeology (for assessing consistency with flow concepts and models, and for evaluating palaeohydrogeological calibration).

APPENDIX 1

Coverage of SKB reports

Table 1

Reviewed report	Reviewed sections	Comments
TR-11-01 Long-term safety for the final repository for spent nuclear fuel at Forsmark: Main report of the SR-Site project. Volume 1	Section 2.5 Sections 4.3 & 4.4 Sections 4.6 - 4.9 Section 6.2 Sections 8.3.4 & 8.4.5	
TR-11-01 Long-term safety for the final repository for spent nuclear fuel at Forsmark: Main report of the SR-Site project. Volume 2	Sections 10.2.3-10.2.5 Sections 10.3.6-10.3.8 Section 10.3.11 Section 10.3.16 Section 10.4.6-10.4.8 Section 10.4.11 Section 10.5.1 Sections 10.6.3-10.6.4	
TR-11-01 Long-term safety for the final repository for spent nuclear fuel at Forsmark: Main report of the SR-Site project. Volume 3	Sections 11.2.1-11.2.3 Sections 13.4.2-13.4.5 Section 13.5.3 Section 13.8 Sections 15.2 & 15.3 Sections 15.6 - 15.8	
TR-10-52 Data report for the safety assessment SR-Site	Section 6.1 Sections 6.7 & 6.8	
TR-10-48 Geosphere process report for the safety assessment SR-Site	Section 3.1 Sections 5.1 - 5.12	
TR-10-50 Radionuclide transport report for the safety assessment SR-Site	Sections 2.2-2.5	
TR-08-05 Site description of Forsmark at completion of the site investigation phase	Sections 9.1 - 9.6 Section 11.7	
TR-10-58 SR-Site - Hydrogeochemical evolution of the Forsmark site	All sections	
TR-10-39 SR-Site - Sulphide content in the groundwater at Forsmark	All sections	

TR-10-57 SR-Site: Oxygen ingress in the rock at Forsmark during a glacial cycle	All sections

APPENDIX 2

Suggested needs for complementary information from SKB

1. Clarification is needed of apparent examples of data traceability issues between SDM-Site and SR-Site in the Hydrogeochemical Evolution report for SR-Site (TR-10-58, Salas et al 2010) are: (i) Near-surface waters in the depth range 0-20 m are said to be fresh, i.e. <200 mg/L Cl^- (TR-10-58, p19) which is mostly the case, but Fig 4-1 in R-08-47 (p62) shows at least one sample with about 4000 mg/L (this may be significant because high salinity indicates potential upward flow); (ii) Shallow 'mixed brackish' groundwaters in the depth range 20-200 m are said to have Cl^- in the range 200-2000 mg/L (TR-10-58, p19) whereas Fig 4-1 in R-08-47 shows that Cl^- concentrations are mostly between 3000-6000 mg/L; (iii) The scatter of data points for pH in Fig 6-10 (TR-10-58, p53) is rather different, though generally covering the same range, compared with the scatter shown in Fig 4-6 in R-08-47 (p67); (iv) The number of data points and the range covered (>0.095 mg/L PO_4^{3-}) in Fig 4-4 (TR-10-58, p39) are greater than the data shown in Fig 4-11a in R-08-47 (p73) for which the maximum of the range is <0.07 mg/L.
2. SKB is asked to clarify whether the effect of alkaline alteration on the sorption and diffusion properties of rock has been taken into account in the model of radionuclide transport and retardation (TR-11-01, Vol 2, p337).
3. SKB is asked to clarify the approach taken to estimate the overall uncertainty in composition of initial state water that will enter the deposition holes and resaturate the buffer. This initial state water will derive from fractures with very low transmissivity and possibly will have compositions and chemical buffering intermediate between those of fracture water and pore water (TR-11-01, pp132-133)
4. SKB should provide additional information about the reasoning and evidence that omitting silicate dissolution/precipitation reactions and cation exchange reactions has negligible significance for the interpretation of present-day groundwater conditions and for the hydrogeochemical evolution model. Is there any modelling to show that the omitted reactions are negligible with regard to the safety function indicators? (TR-11-01, p355)
5. Some safety function indicator criteria that relate to groundwater chemistry are loosely-defined, e.g. for O_2 and HS^-, I, H_2, CH_4 and DOC, and K^+ and Fe^{2+}, most of which are specified to be "low". The requirement for 'chemically reducing' conditions is another loosely-defined criterion. SKB should provide more information about how these criteria are applied, i.e. what is a 'low' value and what is an unacceptably 'high' value. The same questions arise for the safety functions for retardation for which no

quantitative criteria have been established, i.e. reducing conditions, ionic strength, and low colloid concentrations. (TR-11-01, Vol 1, pp252-261)

6. Anomalous concentrations of NH_4^+ up to 33.2 mM have been reported for Laxemar. SKB should clarify whether this high value is credible and, if so, whether a deep natural source of N, e.g. in NH_4 contained in mica minerals, is possible for Forsmark. (TR-10-58, p102)
7. SKB is asked to clarify why the safety function indicator criterion concerning colloid stability has been simplified to the sum of all cations, i.e. $\sum q[M^{q+}] > 4$ mM in place of considering separately the sum of divalent cations $\sum[M^{2+}]$ and the sum of monovalent cations $\sum[M^+]$. The latter are supported by empirical and theoretical evidence but there is no direct scientific basis for summing all cations as a criterion. (TR-11-01, p252; TR-10-58, p13)
8. SKB is asked to clarify the position on DO data, i.e. the reliability of DO data in the database and the reason for excluding discussion of these.
9. More information is needed to explain how the 'reducing' character is quantified by other redox parameter such as HS^- and Fe^{2+}. Measured Fe^{2+} concentrations generally decrease with depth from 10^{-1} to $<10^{-2}$ mmol/L (Fig 5-6, TR-10-39, p47) so the measured range is rather narrow and subject to various analytical and geochemical factors. SR-Site mentions potential variability of the strength of reducing conditions, but an explanation of what might vary, i.e. Fe^{2+}, HS^-, DOC, is needed, and what will be the significance of those variations in terms of near-field hydrogeochemistry and behaviour of radionuclides.
10. More information is required about the number of HS^- analyses that have been discounted on the basis that they exceed FeS equilibrium, and whether there are independent reasons to reject the HS^- and/or corresponding Fe^{2+} analyses in these cases and to support FeS being the controlling mineral phase.
11. Most of the dissolved organic carbon (DOC) is said to be large molecule organic carbon that is not biogeochemically available to SRB. Is this supported by specific data from the SDM? This is potentially significant because future variations of DOC may be the controlling factor in HS^- production. (TR-11-01, Vol, 2, p361 and Fig. 10-43)
12. Deep saline water at Olkiluoto may be the better analogue on which to base the deep saline end-member composition for Forsmark. SKB is asked for clarification of why the Laxemar saline water composition, rather than that of the Olkiluoto water, was chosen as the end-member. What does modelling indicate concerning sensitivity to saline end-member composition and does it confirm that the additional uncertainties introduced by this are negligible? (TR-10-58, Table 4-2, p37)
13. Has the analytical model used to simulate dilute water infiltration in the initial period after closure been verified and how has it been tested to be adequate for this purpose? (TR-11-01, Vol 2, p339)
14. Why are near-field groundwater compositions in the temperate period modelled up to only 7000 or 5000 years (p343 vs. p347) in the future? It is noted that forward modelling of the 'global warming' variant scenario is extended to 60000 y of temperate conditions. (TR-11-01, Vol. 2, p341)
15. Modelled Eh changes as near-field groundwater is progressively diluted from 'a mean value' of -230 mV to -190 mV. What data have been averaged to obtain these values and whether the change of modelled Eh is primarily dependent on pH change (i.e. because Eh and pH are coupled via

the redox half-reactions) or is reflecting modelled changes of other redox variables? (TR-11-01, Vol. 2, p365)

16. More information is required about what was found in the studies of groundwater chemistry underneath permafrost to indicate that it is not much affected and also about how meaningful and relevant those studies are for long-term evolution at Forsmark. (TR-11-01, Vol. 2, p511)
17. The modelling shows saline water being flushed upwards by the intrusion of glacial melt water so that higher salinity occurs near the ground surface under permafrost. Has saline upconing been taken into account in the hydrochemical evolution modelling? (p493 & p502, Fig. 10-130, TR-00-01, Vol. 2)
18. If melt water infiltrates only for the short period when an advancing or retreating ice front passes over the repository location, how does subglacial groundwater evolve during the rest, i.e. major part, of the glaciation which could be several tens of thousands years?
19. In view of the “large degree of uncertainty in the geochemical modelling results of Salas et al (2010; TR-10-58)”, it would seem to be possible, even if unlikely, that the ranges of simulated groundwater compositions might extend outside the box-and-whisker plots in Figs 10-151 to 10-153. For example, ∑(cations) in Figs. 10-151 and 10-152 goes as low as 10 mM, and full consideration of uncertainties and alternative assumptions might indicate lower values. SKB should be asked to provide more information about uncertainties that have not been included in the hydrogeochemical evolution modelling. (TR-11-01, Figs 10-151 to 10-153)
20. For the O_2 ingress scenario, SKB should provide further information on the groundwater flow model, e.g. variants, sensitivity analyses, to supplement the hydrogeochemical modelling of O_2 attenuation. (TR-10-57)
21. The palaeohydrogeological model has been calibrated by simulating the hydrodynamic mixing of water masses (i.e. the end-member waters) by forward modelling from hypothetical initial conditions up to the present day, and then comparing the modelled compositions with observed compositions. Comparison of modelled and observed data is not shown in TR-11-01. Neither Fig 4-22 or Fig 4-23 seems to show the palaeohydrogeological modelling output as SKB suggests. Both figures, at differing scales, seem too schematic to represent adequately either measured data or modelled output. SKB is asked to clarify what these cross-section figures are showing and to provide additional information on the palaeohydrogeological calibration as used in SR-Site. (TR-11-01, Vol 1, pp 133 & 136, Figs 4-22 & 4-23)
22. More information is requested about how the findings of natural analogue studies have been incorporated into the conceptualisation and modelling approach for SR-Site. (TR-11-01, Vol. 3, p793)

APPENDIX 3

Suggested review topics for SSM

1. Scrutinise SKB's records (see TR-11-01, p92) of expert judgements for the interpretation of groundwater chemistry data and for the selection of representative parameters for use in the safety case. Check for traceability and assess the information on which expert judgements have been based. Check how uncertainties have been estimated and whether these have been propagated through the interpretations and into models.
2. Review the way that the M3 multivariate statistical analysis tool has been used to resolve groundwater chemistry data into proportions of component end-member waters. Check how uncertainties have been propagated through the calculations. Assess the selections of end-member water compositions and identify alternatives. Evaluate the significance of alternative end-member compositions (e.g. higher concentrations for deep saline end-member water, different compositions for altered meteoric end-member water). Check the implications of alternative M3 models in SR-Site.
3. Examine the full data set in SKB's spreadsheet for groundwater chemistry and isotopic compositions. Test the traceability of groundwater chemistry data that are used in SR-Site. Review the information on the structural origins of water samples (e.g. deformation zones, fracture zones, fracture domain bedrock) and check consistency with interpretation and use of data.
4. Review the methods and output chemistry and isotopic data for bedrock matrix pore water. Assess the uncertainties in data. Establish a scientifically robust view of the representativeness and significance of pore water compositions. Evaluate how SKB have reported and used pore water data. Review SKB's interpretation in terms of diffusive exchange between pore water and water in adjacent conductive features.
5. Review the validity of hydrogeochemical conceptual models used in SR-Site in the context of the broad geoscience of comparable investigations. Assess the robustness of assumptions and simplifications used in interpretative and numerical models.
6. Review literature and the outputs from recent investigations concerning groundwater chemistry and hydrogeology in permafrosted and glaciated areas. Assess SKB's conceptual models and assumptions that have been used in the reference scenario and variants for long-term evolution.
7. Sensitivity analyses of SKB's hydrogeochemical modelling which neglects silicate dissolution/precipitation and cation exchange reactions. Use previous SSM modelling of dilute water evolution as the basis. Check the potential implications for long-term evolution modelling, especially for parameters that are safety function indicators.
8. Comparison of heterogeneities in groundwater compositions with the interpreted variations in groundwater pressures. Investigate the evidence for compartmentalisation in the Forsmark groundwater system.
9. Scoping modelling of alternative long-term evolutions of the deep groundwater system. Investigate the effects of different initial and boundary conditions on upconing or flushing of deep saline water. Test SKB's model that suggests dilute water penetrates no deeper than 500 m.
10. Evaluate SKB's data for analogue trace elements (U, Ra, I, Ni, REEs, Cs, ^{14}C) in Forsmark groundwaters and in corresponding rocks and minerals. Review the wide scientific literature and data for crystalline rock systems, including

‘natural flux’ studies. Is it possible to obtain useful information concerning transport and retardation processes for these trace elements and for analogous radionuclides?

11. Review the requirements and possibilities for monitoring of groundwater compositions in tunnels and deposition holes during the construction phase. Specify data requirements. Identify what data will be available for evaluation against safety function indicator criteria, as well as the operational issues in using those data.

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The Swedish Radiation Safety Authority has a comprehensive responsibility to ensure that society is safe from the effects of radiation. The Authority works to achieve radiation safety in a number of areas: nuclear power, medical care as well as commercial products and services. The Authority also works to achieve protection from natural radiation and to increase the level of radiation safety internationally.

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The Authority reports to the Ministry of the Environment and has around 270 employees with competencies in the fields of engineering, natural and behavioural sciences, law, economics and communications. We have received quality, environmental and working environment certification.

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