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Session 1a – Deep Static Geological Systems Analysis

Keynote Lecture – A Review of the History of Groundwater Analysis to Support Residence Time and Pathway Evaluations in Geological Disposal Programmes for Radioactive Waste
Ian Clark (University of Ottawa, Canada)

ABSTRACT

Investigations for geological isolation of radioactive waste have generated substantial research on the origin, age and movement of fluids and gases in the far-field geological formations. The geochemical and isotopic tools now available have evolved through several decades of refinement and analytical development, with application at natural analogue sites and at potential repository sites. Considerable development has also been made on the extraction and analysis of fluids in aquitards and aquicludes. This address highlights the development and use of some traditional tools and new isotopes that are now available for such investigations, presented from the perspective of recent investigations of groundwaters and porewaters at a Canadian site under consideration for a deep geological repository in the Michigan Basin. Here, the use of a host of traditional and novel isotopes and geochemistry provide multiple lines of evidence for long residence times and migrational pathways, showing that long term isolation of radioactive waste in such environments is possible.

Deep Static Geological Systems Analysis – Recent Advances in Isotopic Methods for Residence Time Determinations >50,000y

Atom Trap Trace Analysis of Radioactive Noble-gas Nuclides for Dating

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Long-lived noble-gas isotopes ^{85}Kr (10.8 y), ^{39}Ar (269 y) and ^{81}Kr (229 ky) are ideal tracers for dating environmental samples such as groundwater and ice. Together with ^{14}C, these nuclides can be used to cover the whole range of 10^{0}-10^{6} y. Atom Trap Trace Analysis (ATTA) is an emerging method for the analysis of these isotopes at an isotopic abundance level as low as 10^{-16}. The reliability and accuracy of the ATTA measurements have been examined with an inter-comparison using a group of 12 samples with different ^{85}Kr/Kr ratios in the range of 10^{-13} to 10^{-10}. The ^{85}Kr/Kr ratios were measured independently in three laboratories: a low-level counting (LLC) laboratory in Bern, Switzerland, and two ATTA laboratories, one in Hefei, China, and another in Argonne, USA. The results are in agreement at the precision level of 5%. At present, ATTA can determine the isotopic abundances of ^{85}Kr and ^{81}Kr with typically 5-10% accuracy using krypton gas samples of a few micro-liters (STP). Detection of ^{39}Ar with the abundance at levels of 1 part in 10^{16} has also been demonstrated. However, for practical applications, the capture rates of ^{39}Ar need to be further improved by a factor of 10–100, possibly using isotope pre-enrichment techniques.

Deep Static Geological Systems: Geochemical and Isotopic Tracers of Groundwater Evolution at a Crystalline Rock site in Finland

Shaun Frape

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Many countries in the northern hemisphere are considering crystalline granitic rock as a potential host for a radioactive waste repository. Geological and hydrogeological studies in these environments make use of isotopic tracers to understand groundwater flow, mixing, evolution and potential nuclide transport. A number of research sites worldwide have been studied ranging from working mines to dedicated Underground Research Laboratories (URLs).
The Palmottu Natural Analogue study was conducted between 1996 and 2003 in southwestern Finland. The site geology is dominantly crystalline rock hosting extensive Uranium–Thorium mineralization. A wide variety of geochemical and isotopic tracers were measured in the groundwaters at the site to assess flow and hydrogeochemical processes in Phase I. Later in the program the object was to identify radionuclide mobilization and transportation in Phase II. The upper geological regime at the site hosts a surprisingly active and diverse hydrogeological regime. Below this zone groundwaters are more stagnant and consist of NaCl and Na2SO4 brackish waters. The evolution and source of these solutions was important due to their association with the mineralization and the apparent stability. However, the unique nature of Na2SO4 waters occurring in the crystalline rock environment at this site, especially their possible origin and potential age became a topic that resulted in a number of future studies concerned with potential glacial/permafrost impacts on repository sites in northern locations.

Deep Static Geological Systems Analysis – limitations on groundwater dating and the impact of matrix porewaters
Nick Waber (University of Berne, Switzerland)

Session 1b – Deep Static Geological Systems Analysis – Case studies

Horonobe and Mizunami URL sites, Japan (Kenji Amano and Takanori Kunimaru, JAEA, Japan)

Characterisation of the Sellafield site, UK: a case study of deep static geological systems analysis

Adrian Bath (Intellisci, UK)

Boreholes were drilled in an area of 52 km^2 at Sellafield on the coast of northwest England to investigate groundwater conditions in low permeability fractured metavolcanic basement rocks and overlying sedimentary rocks. The basement rocks were being investigated as a potential host for a repository for intermediate-level radioactive wastes until the project was stopped in 1997.

Water samples were collected in deep boreholes to a maximum depth of 1950 m. Groundwater compositions at this site have a range of salinities from fresh/brackish to brine. Various isotopic analyses were carried out for samples from 16 boreholes: ^{3}H, $^{18}O/^{16}O$, $^{2}H/^{1}H$, ^{3}H, ^{14}C, $^{13}C/^{12}C$, $^{36}Cl/Cl$, $^{87}Sr/^{86}Sr$, $^{34}S/^{32}S$, ^{4}He and other inert gas contents. Data for stable water isotopes and ^{36}Cl in selected samples are considered to represent *in situ* compositions, whilst ^{3}H data mostly are a quantitative indicator of the proportion of drilling water mixed in with groundwater and carbon isotopic data are generally unreliable because they are contaminated by breakdown products of organic additives in drilling fluid.

The conceptual hydrogeological model envisaged water masses with different origins, ages and salinities, distributed with varying vertical and lateral transitions according to the regional influences on groundwater movements between inland topography and an offshore basin. Stable water isotopes ($^{18}O/^{16}O$ and $^{2}H/^{1}H$), ^{3}H and ^{14}C (the latter two sets of data having large uncertainties) are consistent with the hydrodynamic model that fresh and brackish water in the sedimentary rocks, typically to 350 m depth in the centre of the area and to a maximum of 600 m elsewhere in the area, was mostly recharged since the end of the last glaciation in northern Europe.

Groundwaters in the basement rocks in the centre of the area are moderately saline down to at least 1000 m. A key question for site characterisation therefore concerned the age, and thus circulation period from recharge to potential discharge, of these groundwaters and the hydrodynamic mixing that accounts for salinity. Their distinctive stable water isotopic ratios indicate cold climate recharge, probably at some time during the last glacial period i.e. between 10-100 ka ago. Background for this age interpretation is provided by stable isotopic compositions of groundwaters in deep English aquifers that have been shown with ^{14}C to be older than 10 ka.

The most likely origin of salinity in groundwaters in basement rocks here is that it is primarily evolved from brine that migrated from sedimentary rocks in the offshore basin. The $^{18}O/^{16}O$ and $^{2}H/^{1}H$ ratios of the brine are relatively higher and thus distinct in relation to the younger and older onshore meteoric water sources. Saline groundwater in the basement is also distinct from the basinal brine in having a higher Br/Cl ratio, so as well as being diluted by mixing with meteoric water has also reacted with the rock which suggests a long residence time.

Variations in $^{36}Cl/Cl$ cannot be explained just by mixing of groundwaters and are interpreted as additionally being due to variable *in situ* ^{36}Cl production rates in different rock formations. ^{36}Cl ratios in saline groundwater sampled from the metavolcanic basement have a mean of 25 x 10^{-15} whereas ratios in groundwaters from the sedimentary cover rocks have a mean of 10 x 10^{-15}. These ^{36}Cl ratios are at or close to equilibrium with *in situ* ^{36}Cl production in the respective formations, according to production rates calculated from U and Th contents of rock based on geophysical logs. This observation was interpreted to constrain the timing of mixing. The distinctive values of $^{36}Cl/Cl$ probably took >1.5 Ma to be attained, implying that the deep brine and highly saline groundwaters have been more or less immobile during

glaciation of the site. This is consistent with some realisations of a density-dependent hydrodynamic model of brine movement, although other realisations with different parameters indicate a greater degree of movement than inferred from the isotopic interpretation.

The characterisation of the Sellafield site involved the integration of geological, hydrogeological, hydrochemical and mineralogical data with interpreted isotopic ages and mixing of groundwater masses in a synthesis of palaeohydrogeology. It produced a conceptual and semi-quantitative model of how the groundwater system has evolved over a considerable length of time in the past comparable to the future timescale of a safety case. This interpretation is the most likely explanation of groundwater movements in response to topographic gradients, salinity/density contrasts, and climate-driven changes to infiltration. A safety case would have evaluated rigorously the uncertainties and considered alternative interpretations.

EVALUATION OF NATURAL ISOTOPES AT RUPRECHTOV NATURAL ANALOGUE STUDY SITE

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At Ruprechtov site in NW Bohemia, Czech Republic, natural uranium deposits were investigated as analogues for uranium migration and immobilisation in argillaceous sediments, being conceivable processes in the post-operational phase of a radioactive waste repository. These investigations were aimed at gaining insight into the behaviour of uranium in a complex natural system.

Ruprechtov site is geologically situated in a Tertiary basin. The study area itself is characterised by a granitic rock body, which partly crops out in the west and in the south, but is covered by kaolin layers of varying thickness in its central part. Wide areas of the basin are overlain by argillised pyroclastic Tertiary sediments enriched in organic matter. Hydrologeological, geochemical and environmental isotope data from groundwater wells at Ruprechtov natural analogue site have been evaluated to characterize the flow pattern and the Carbon chemistry in the system. The observed increase of $\delta^{13}C$ values and decrease of ^{14}C activities in dissolved inorganic C during evolution of the groundwater from its infiltration area down to the so-called 'clay/lignite-horizon' was modelled using simple open- and closed-system models as well as an inverse geochemical model. The results provided some insights into timescales of groundwater flow, but mainly revealed that additional sources of C are active in the system. The occurrence of organic matter degradation by microbial sulphate reduction is confirmed by mineralogical evidence (spherical pyrite) and by an increase of $\delta^{34}S$ values in dissolved SO_4^{2-} in groundwater.

In addition, isotope data from the uranium decay chain have been used to get a deeper understanding of uranium mobilization and immobilization processes at the site. The

investigations included wet chemistry to determine the distribution of U(IV) and U(VI), sequential extraction to characterize different uranium phases, and the determination of $^{234}U/^{238}U$-activity ratios to correlate results between U(IV) and U(VI) distributions and the various uranium phases. Most of the uranium was specified to occur in a very long-term stable, tetravalent phase under reducing conditions in clayey layers.

Opalinus Clay Formation (Switzerland)

Daniel Traber, Nagra, Switzerland

In Switzerland, Opalinus Clay is the preferred host rock for the disposal of high level radioactive waste. It is a c. 110 m thick Mesozoic claystone with a clay mineral content of about 60 wt.-%. Opalinus Clay is part of a c. 200 m thick succession of marine claystones and marls with intercalated thin layers of limestones, calcareous sandstones and iron oolithes.
In the year 2002, Nagra (Swiss National Cooperative for the Disposal of Radioactive Waste) demonstrated the long-term safe disposal of high-level radioactive waste in Switzerland in the framework of siting activities in Northern Switzerland. The presentation will give a general overview of the siting activities with major emphasis on how natural tracers contributed to the project. For example, natural tracer profiles were an important argument for the up-scaling of lab diffusion tests to formation scale and geological relevant time scales. Furthermore, the role of the research activities at the Mont Terri Underground Lab will be addressed.

Session 2a – Shallow Active Systems Analysis – Recent Advances in Isotopic Methods for Residence Time Determinations <50,000y

Groundwater dating in the Water Resources Programme of the IAEA

Takuya Matsumoto (Isotope Hydrology Section, IAEA, Vienna, Austria)

In response to the increasing ability of Member States to analyse stable isotopes on their own over the last decade, the IAEA is expanding its analytical capacity to the analysis of radioisotopes for Member States—including tritium-helium-3 and noble gas isotopes—for groundwater dating. Noble gas isotopes, present in dissolved gases in groundwater, provide valuable information about climatic conditions during recharge, as well as the residence time of water at different time scales and its renewal rate. The analytical facility has been fully operational since 2010 and started running more than 300 samples par year. Our current scientific challenge is characterization of large aquifers with limited hydrogeological information by using multiple age tracers (^{3}He, ^{4}He, ^{81}Kr, ^{14}C and CFCs) to obtain better constraints on aquifer recharge and flow dynamics, and to provide the ability to reliably constrain groundwater models. This effort is accompanied by the development of new gas sampling devices enabling efficient field sampling of gases dissolved in water both for stable noble gas isotopes and for radioactive krypton isotopes (^{81}Kr), and by the implementation of production of laboratory water standards under precisely controlled conditions for noble gas analysis with the provision for supplying them to external laboratories in a near future.

Examples of different isotopic systems for dating groundwater residence

times <50,000 years

Florian Eichinger (Hydroisotop, Germany)

Isotope analyses and interpretations are a valuable tool to determine groundwater residence times. Additionally, those methods enable to quantify the proportions of certain groundwater components with different residence times (young groundwater components vs. old groundwater components). Various dissolved natural and artificial radio-isotopes as for example ^{37}Ar, ^{39}Ar, ^{85}Kr, ^{3}H and ^{14}C allow due to their different half life times (35 days to 5730 years) the determination of residence times of groundwater in the range of weeks to tens of thousands of years. Nevertheless the interpretation of groundwater residence times based on dissolved radioisotope concentrations has to be done by taking the hydrogeological conditions of the individual groundwatersystems into account.
This presentation will provide an insight into

- the formation and decay of different radioisotopes applied for the determination of groundwater residence times,
- the sampling and analysis of radioisotopes,
- the modelling and interpretation of radioisotope concentrations dissolved in groundwater, and
- concrete case studies.

Session 2b – Shallow Active Systems Analysis – Case Studies

Validity of the groundwater flow model by using isotopic analysis

Tai Sasaki (Japan Nuclear Fuel Limited)

To confirm validity of the groundwater flow analysis model, calculated values of pore water pressure distribution, amount of spring water seeping into exploratory tunnel, and groundwater quality are compared to those of observed. The calculated pore water pressure distribution shows similar trend with that of observed. Thus, it is confirmed that hydrographic classification and relative hydraulic conductivity allotted to each rock type are properly modeled. Also, amount of groundwater discharge to drift is found to be matching between calculated and observed, confirming the validity of allocated hydraulic conductivity to the rocks near the tunnel, which in turn confirms the validity of the allocation of hydraulic conductivity and the model as a whole . In the meantime as to the water quality, groundwater collected from several boreholes are analyzed for oxygen/hydrogen isotopic ratios ($\delta^{18}O$, δD). Some groundwater show small isotopic ratios indicating that they originate at the time of prevailing low temperature during glacial maximum period of 10,000 to 20,000 yBP. Therefore, by using groundwater flow analysis, time required for the water to infiltrate from the ground surface to the depths can be calculated, and these in turn can be used to confirm validities of flow velocities of groundwater and effective flow porosities. It is confirmed that the calculated groundwater flow velocities give about one to twice as high velocities from actual cases.

Hydrological Age-Dating using cosmogenic radionuclides Be-7 and Na-22 in a meso-scale catchment in the Black Forest, Germany

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Since atmospheric tritium levels have nearly reached the natural background, there is a need for further developing existing or additional methods for age-dating of young water. Especially non-gaseous age-dating tracers are needed for hydrological applications in lakes, rivers and springs as well as for surface-groundwater interaction studies. Cosmogenically produced isotopes of sodium and beryllium (^{22}Na, ^{7}Be, half-lives of 2.602 years and 53.29 days, respectively) have been investigated as potential environmental tracers for residence time analysis of surface water and young groundwater. A simple chemical separation scheme for both radionuclides was established and ^{7}Be could be detected in both surface- and groundwater samples in the catchment of river Dreisam located in the Black Forest, Germany. The ions were extracted from 500 l water using an ion exchange resin. The water samples could be dated to minimum ages of about 165 and 323 days for riverine samples and 475 days for a groundwater sample. Measurement was done using a lead covered HPGe-detector. These ages match previously reported ages using stable isotopes and tritium.

Krypton-81 and Groundwater Flow in the Culebra Dolomite Near the Waste Isolation Pilot Plant, New Mexico

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ABSTRACT

The Waste Isolation Pilot Plant (WIPP) in New Mexico is the first geologic repository for disposal of transuranic nuclear waste from defense-related programs of the US Department of Energy. It is constructed within halite beds of the Permian-age Salado Formation. The Culebra Dolomite, confined within Rustler Formation evaporites, is a potential pathway for radionuclide transport from the repository to the accessible environment in the human-disturbed repository scenario. Although extensive subsurface characterization and numerical flow modeling of groundwater has been done in the vicinity of the WIPP, few studies have used natural isotopic tracers to validate the flow models at this site. We

performed measurements of the cosmogenic isotopic tracer ^{81}Kr (half-life 229,000 yr) in two Culebra monitoring wells near the WIPP site, and compared ^{81}Kr model ages with the results of an ensemble of flow models. The ^{81}Kr model ages were ~130,000 and ~330,000 yr for high-transmissivity and low-transmissivity portions of the aquifer, respectively. These model ages are in broad agreement with flow model results, when taking into account diffusive exchange of Kr between the aquifer and the stagnant zones of the confining formations.

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Session 3 – Special topic: Accidental Contamination - the Use of Isotopic Indicators to Understand Contaminant Dispersion. Case Studies

Background and current status of understanding at both local (Fukushima NPP) and regional (Fukushima Prefecture) scales

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Japan Atomic Energy Agency (JAEA)

ABSTRACT
The damage to the Fukushima Dai-ichi nuclear power plant (NPP) by the Great East Japan Earthquake and tsunami resulted in discharge of various radioisotopes and, consequently, considerable radioactive contamination was released, both on- and off-site of the NPP [1]. Measurement of radioactivity in the topsoil near the NPP showed discharge of not only caesium but iodine, strontium and plutonium. Short-lived radioisotopes such as I-131 have decayed with the passage of time so that the dose rate is now

dominated by radiocaesium Cs-134 and Cs-137, (releases of strontium and plutonium were small). As such, JAEA is conducting a project on the long-term assessment of Transport of RadioActive Contaminant in the Environment of Fukushima (F-TRACE project), and was initiated in September 2012. In this project, the transport behaviour (fluvial, aeolian etc.), of radiocaesium is monitored at the following environmental study sites: forests, rivers, reservoirs and estuaries; a predictive modelling system for radiocaesium transport is currently under development. Depth profiles of radiocaesium concentration in forest soil suggest that there has been migration of radiocaesium from litter to topsoil and that more than 90% of the radiocaesium was present within the top 5 cm. Concentrations of radiocaesium were below the detection limit of the γ detector (~1Bq L^{-1}) in both river and reservoir water, while concentrations in the sediment were of the order of 10^4-10^5 Bq kg^{-1}, indicating that radiocaesium had strongly adsorbed onto mineral particles that had been deposited in river and reservoir sediment. Further data on the distribution of radioisotopes in Fukushima and the transport process of radioisotopes, especially radiocaesium, through the water system will be presented.

1. *Emergency Operation Center, Ministry of Education, Culture, Sports, Science and Technology and Agriculture, Forestry and Fisheries Research Council, Ministry of Agriculture, Forestry and Fisheries: Summarized Version of the "Results of the Research on Distribution of Radioactive Substances Discharged by the Accident at TEPCO's Fukushima Dai-ichi NPP". http://radioactivity.nsr.go.jp/en/contents/1000/294/24/PressR04%200802s.pdf*

What has been done at Chernobyl – can we learn anything from here?
(IAEA, to be confirmed)

Deriving key geosphere parameters from legacy contaminated sites

Elizaveta Klein, Susie M.L. Hardie, Wolfgang Kickmaier, Ian G. McKinley

MCM Consulting, Baden-Dättwil, Switzerland

A fundamental property of any location selected to host a repository for radioactive waste is its capacity to retard the movement of some key, safety-relevant radionuclides. Site characterisation cannot measure this characteristic directly, but focuses on developing site understanding via a synthesis of field observations of the behaviour of analogue elements and isotopes, integrated with tailored programmes of experimental work on appropriately chosen samples of the geosphere barrier. Models form the essential link between field observations and supporting laboratory (or URL) rock/water/radionuclide characterisation studies. However, residual uncertainties always remain, associated with the extrapolations in time and space than need to be made and the justification for chemical and isotopic analogies used.

An under-used resource that could help to strengthen the safety cases that utilise such information is the knowledge base available from anthropogenically contaminated sites. Worldwide, there are many locations containing significant concentrations of relevant radionuclides, resulting from past waste disposal activities, accidents, nuclear weapon testing, operational releases from nuclear facilities, etc. These have the potential to combine relevant geological settings with the actual radionuclides of interest and, in some cases, timescales of many decades that allow typically slow processes to be quantified. This talk will provide an overview of the range of options available, review some relevant examples where otherwise unobtainable data can be derived and outline work that could increase the utilisation of this potential resource.

Session 4 – Enhancing Confidence in Safety Assessments

Kunio Ota (NUMO, Japan)

- Integration with other data and information
- Role of QMS

Modeling the evolution of hydrochemical conditions at a site proposed for geological storage of radioactive waste using multidisciplinary information

Presenter: Sven Follin, SF GeoLogic AB, Sweden, www.geologic.nu

Dr Follin is a consultant and scientist in Hydrogeology. He has a participated as principal investigator and groundwater flow modeler in the site investigations and characterizations for a geological repository in crystalline bedrock conducted by the Swedish Nuclear Fuel and Waste Management Company (SKB), Sweden, www.skb.se

Three-dimensional, large-scale models for groundwater flow and solute transport are used for the low-temperature fractured crystalline rock sites in Sweden and Finland that are being considered for the geological disposal of spent nuclear fuel. It has been suggested that comparisons between measured and simulated present-day hydrochemical data provide a means to constrain the complex influences of past climatic events and to improve the ability to understand the palaeo-hydrogeological evolution of the physical system studied.

Here, the presenter demonstrates how the integration of multidisciplinary data and models from one of the sites in Sweden (Forsmark) can aid the appraisal of the hydrochemical conditions at 8000 BC, which is the selected starting point for the palaeo-hydrogeological modeling of the hydrochemical conditions in the Fennoscandian Shield during the Holocene (last 10 ka). Since a firm understanding of the evolution of the hydro-chemical conditions is important for the long-term safety assessment, recognition of the initial hydrochemical conditions is essential for the overall build-up of confidence in the modeling process.

Session 5– Planning Future Investigation Programmes

Planning future investigation programmes

Adrian Bath (Intellisci, UK) and Andrew Parkes (Nuclear Decommissioning Authority, UK)

Investigations for repository siting aim to supply information that is required for engineering and construction assessments and for preparation of operational and post-closure safety cases. Data for the various environmental isotopes, in conjunction with groundwater compositions and mineralogical data, are relevant to the geosphere transport part of a long-term safety case and to general confidence that the long-term evolution of a site's groundwater system is understood. The broad categories of inputs are: (i) the groundwater flow and solute transport model, (ii) understanding of past groundwater conditions (palaeohydrogeology), and (iii)

testing for unfavourable site characteristics.

Strategies and choices for applying the various isotopes will be adapted for different types of site depending on the specific conditions of the potential host formation, i.e. crystalline rock with dominant fracture permeability, clay-rich sedimentary rock with diffusive solute transport, or evaporite with overlying sedimentary cover. Hydrogeological and hydrochemical environments vary widely, and sample availability and sampling methods vary accordingly.

My talk will discuss the logistical issues and technical/scientific challenges of the various isotopic methods, drawing on experience and evaluations by previous speakers at the workshop and by radioactive waste management organisations and nuclear safety regulators.

The main logistical challenge is obtaining groundwater and mineral samples that are confidently representative for *in situ* isotopic compositions and are of sufficient sizes for the respective analyses. Borehole construction and testing/sampling approaches must plan for this challenge, observing the general principle that fewer higher quality samples are more cost effective than large numbers of unreliable samples. Siting investigations may also find that there are difficulties in accessing appropriate analytical services with the necessary specialist scientific expertise. The programming of investigations needs to take account of that and of the timescale in which data and interpretations can be fed back into the ongoing investigation.

Interpretation and modelling of isotopic data to obtain groundwater ages, solute residence times and other palaeohydrogeological information for site suitability assessment and a safety case rely on a background of site-specific research. For some isotopes this is extensive and for others less comprehensive. It will be an expert task to adapt that background to the site of interest. Assumptions and parameters must be fully documented and justified, and the resulting uncertainties should be realistically analysed.

It is suggested that directions of future research might focus on (a) the conjunctive integration of data for various isotopes with hydrochemical data to reduce the ranges of uncertainties in palaeohydrogeological models, and (b) the development of models for groundwater flow and reactive transport that simulate isotopic parameters by coupling hydrodynamics and hydrogeochemistry. However, the value of research and of increasingly sophisticated analytical methods, so that for example smaller samples are required, will be lost unless drilling and testing methods can assure reliable unperturbed groundwater samples.