MINERALOGICAL MAGAZINE, APRIL 1985, VOL. 49, PP. 281-288

The NAGRA investigation project for the assessment of repositories for high-level radioactive waste in geological formations

WERNER BALDERER
Physics Institute, University of Berne, Switzerland

ABSTRACT. The first results of isotopic investigations as a part of the hydrogeological study of northern Switzerland are presented. This study is part of the global geological research project of the NAGRA (National Co-operative for the Storage of Radioactive Waste) which is intended to provide the scientific knowledge required for the assessment of the feasibility of safe disposal of highly radioactive waste in the geological formations of the area. The aim of the hydrogeological research programme is to assess the natural conditions in the sedimentary sequence and the underlying granite basement, by regional investigation of hydraulic parameters, isotopic composition, and hydrogeochemistry at all existing wells (boreholes and natural thermal springs) and in 11 to 12 deep-drilled boreholes (to final depth of 1500-2500 m).

KEYWORDS: radioactive waste, NAGRA project, hydrogeology, Switzerland.

THE NAGRA (Swiss National Co-operative for the Storage of Radioactive Waste) has a brief to present a feasibility study for the final disposal of high-level radioactive waste by 1985. The feasibility project is based on the so-called multibarrier concept: storage in highly impermeable rocks with low groundwater flow; retardation of eventually escaped radionuclides from the containers by the embedding material and by the sorption properties of the natural rock at the storage site; further long flowpath to the biosphere and dilution by the groundwaters of the different overlying sedimentary aquifers.

The region under investigation is situated in the northern part of Switzerland containing a surface area of about 1200 km^2. In order to assess the geological features of the study region a geophysical, geological, hydrogeological, and hydrogeochemical investigation programme is in progress, including the following topics:

drilling of twelve deep boreholes, transecting the whole sedimentary cover and penetrating the underlying crystalline bedrock up to 1000 m (including core analysis and geophysical logging);

detailed geological mapping of the region next to the drilling sites;

refraction and reflexion seismic survey (vibroseis-method);

gravimetric survey, aeromagnetic survey, and magnetotelluric measurements;

hydrogeological and hydrogeochemical investigations, including the following main objectives:

regional survey of about 111 existing wells and springs assumed to represent outflow points of deep groundwaters,

periodic survey of thirty-three important mineral and thermal springs,

hydrogeological assessment of the deep boreholes, including hydraulic testing, groundwater sampling of all intersected aquifers, and the water-bearing zones in the crystalline bedrock for isotopic and hydrogeochemical analysis,

elaboration and construction of a three-dimensional hydrodynamic flow model (with the following regional extension from the northern part of the Alps in the south to the Black Forest area in the north, and from the Rhine Graben system in the west to the eastern border of Switzerland).

Isotopic investigations on groundwaters from springs, existing wells, and deep drilled boreholes.

In parallel to the hydrogeochemical determinations, most of the existing isotope methods (as shown in Tables I and II) are applied to investigate the mean residence time (distribution) and also the origin and evolution of the groundwaters at springs, wells, and deep boreholes. Beside the direct characterization in terms of 'age' recharge conditions and groundwater hydrogeochemical evolution, the main aim is to reconstruct the regional framework of the hydrodynamic flow pattern (as defined by Toth, 1962) and thus provide a separate tool for the validation of the hydrodynamic flow model.

TABLE I. *Summary of radionuclides and decay products used in the NAGRA research project*

Isotope	$T_{1/2}$ (years)	Primary production reaction (s)	Approximate sample size (L)	Time range of (years)
^{3}H	12.43	^{14}N (n, ^{3}H) ^{12}C	0.1–1	1–50
^{4}He	Stable	$\alpha + 2e \rightarrow {}^{4}He$	0.001–0.1	10^4–10^7
^{14}C	5730	^{14}N (n, p) ^{14}C	60; 0.1*–1*	10^3–5×10^4
^{36}Cl	3.0×10^5	Spallation on ^{40}Ar ^{35}Cl (n, γ) ^{36}Cl	4*	2×10^4–1×10^6
^{39}Ar	269	^{40}Ar (n, 2n) ^{39}Ar	10^4; 10†	30–1200
^{40}Ar	Stable	$^{40}K \rightarrow \beta^+ + {}^{40}Ar$	5	(10^5–10^7)
^{81}Kr	2.1×10^5	^{80}Kr (n, γ) ^{81}Kr (and spallation)	1–10†	5×10^4–8×10^5
^{85}Kr	10.7	Fission of U and Pu	1000	1–40
^{222}Rn	3.8 days	$^{226}Ra \rightarrow \alpha + {}^{222}Rn$	1	0.5–10 days
$^{238}U/^{234}U$	4.5×10^9	$^{238}U \rightarrow \alpha + {}^{234}U + 2\beta$ (via intermediary steps)	10	(5×10^4–10×10^6)

* For accelerator measurement.
† For resonance ionization spectroscopy, in development.
Radioisotopes underlined are discussed in this report.
(After International Atomic Energy Agency, 1983.)

As the study is not yet completed only a summary of preliminary results of the present investigations is given here.

Description of the main geological features

The study region, an area of about 1200 km^2 situated in northern Switzerland, is shown in fig. 1. It has the following features (Diebold and Sprecher, 1983): the granitic bedrock is covered towards the south by sedimentary deposits up to 2000 m thick but outcrops in the Rhine Valley at the northern border of Switzerland. Fig. 2 is a schematic cross-section from the northern part of the Alps (in the south) through the Helvetic Nappes and the Swiss Plateau (which contains the studied region) to the Rhine and the granitic massif of the Black Forest in the north.

TABLE II. *Summary of stable isotopes used in the NAGRA research project*

		Approximate water sample size (L)
^{2}H in H_2O	± 1	10^{-2}
^{18}O in H_2O	± 0.1	10
in SO_4	± 0.3	1
^{13}C in DIC	± 0.3	1
^{34}S in SO_4	± 0.3	1–10
in H_2S	± 0.3	1–10

DIC = desorbed inorganic carbon.
(After International Atomic Energy Agency (1983), modified after NAGRA, 1983*b*.)

As shown in fig. 2, the crystalline massif, which forms the central part of the Alps, is covered by the overlying sediments further to the north, which are characterized by the following tectonic units (from S. to N.)—the autochthonous and parautochthonous sedimentary complex (mainly Mesozoic sediments forming the so-called Préalps and the Helvetic Nappes) which overthrusts Tertiary sediments of the inclined subalpine Molasse, which itself overthrusts the huge normally stratified complex of the Molasse Basin (clastic sediments of Tertiary age, forming the main part of the morphological picture of the central part of Switzerland, the so-called Molasse-Plateau). In the NE part, the sedimentary series of Jurassic age outcrop in the anticlines and synclines of the so-called folded Jurassic (Kettenjura) and further to the north and to the east by the normally stratified series of the so-called unfolded or tabular Jurassic (Tafeljura).

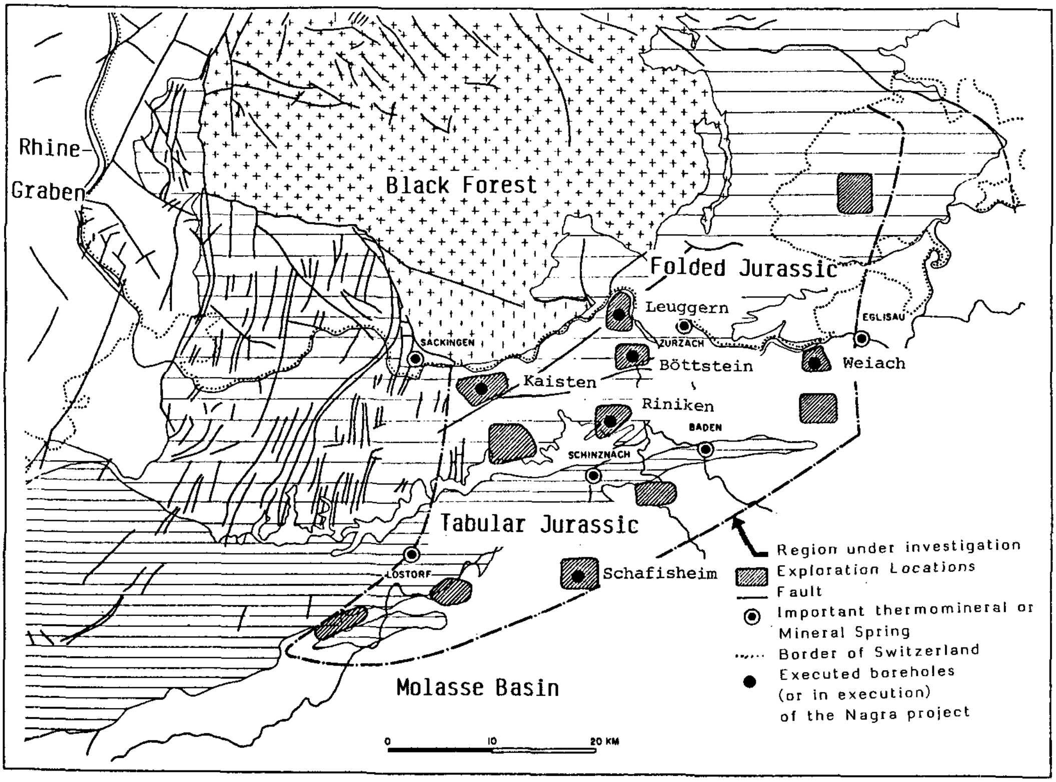

FIG. 1. The region under investigation in the NAGRA research project for northern Switzerland, including the twelve locations for deep exploration boreholes (after NAGRA, 1983*a*).

Further to the north, near the eastern border of Switzerland, which is mainly formed by the Rhine river, the sedimentary cover is eroded and the crystalline bed-rock of the Black Forest massif appears.

The above geological description was the known geology before the NAGRA deep-drilling project. As newly explored by some of the recently drilled NAGRA boreholes, huge Permo-Carboniferous deposits of up to more than 1000 m thickness have been encountered lying above the ordinary crystalline basement within a trench-zone, limited by faults and where the base of the crystalline bed-rock is lowered to 2500 m below ground level (see fig. 3).

This geological situation forms the framework of the results (presented in summarized form) of the regional isotopic and hydrochemical investigations carried out on boreholes and springs, showing the different properties of the groundwaters in the different aquifers.

Summary of the hydrogeochemical and isotopic signatures of groundwaters of the main aquifers in the different geological formations

Besides the aquifers in gravelled Quaternary deposits (found mostly as highly permeable fluvio-glacial sediments accompanying the rivers, whose storage capacity is mainly used for the public water supply) the following aquifers exist in consolidated rocks.

Groundwaters from molasse deposits. The molasse deposits (which have different types of origin and are classified from top to bottom as upper fresh-water molasse, upper marine molasse, lower fresh-water molasse) outcrop in the region of the molasse plateau and therefore, forming the morphology of

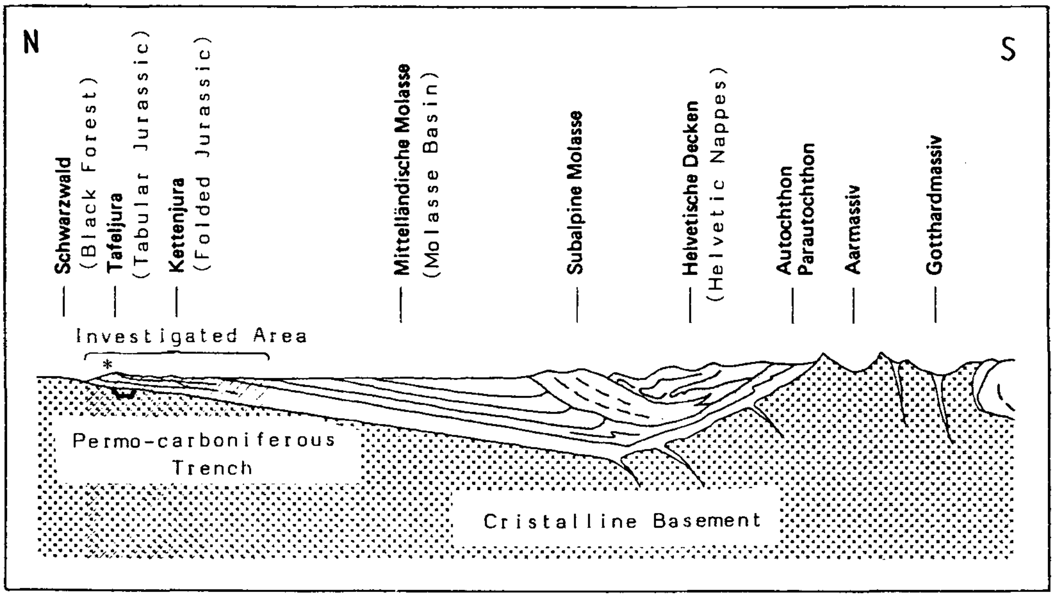

FIG. 2. Geological cross-section through Switzerland from north to south (modified after NAGRA, 1983*a*). * Approximate location of the new explored Permo-Carboniferous trough.

the region (mostly appearing as hills) if not covered by Quaternary deposits. Near the surface, groundwater flows from these molasse deposits through springs which result from lithological-stratigraphical inhomogeneities. These springs mainly yield a Ca–Mg–HCO_3 type water of a mineralization of about 5 to 10 meq./1 (Schmassmann *et al.*, 1984). The residence time of water flowing out at these springs is normally short, of the order of only a few years, and the recharge areas are located in the regions near the springs as deduced from the stable isotope content.

If the deeper formations of the molasse beyond the morphological base line of the valleys (the rivers) are intersected by boreholes a very different type of groundwater is found.

In the deposits of the *upper marine molasse* which is reached by boreholes at depths of about 300 to 600 m (Zurich, Konstanz, Mainau), higher mineralized Na–HCO_3–(Cl)–SO_4 waters with a very high Na content (200 mg/l) and a total mineralization of 8–15 meq./l are frequently encountered (Schmassmann *et al.*, 1984). But as far as residence times can be extrapolated by model calculations from ^3H, ^{39}Ar, ^{14}C, ^{13}C determinations, the probable time range reaches some 10^3 years or as supported by several indications (stable isotopes and noble gas contents) can even reach the range of 10^4 years, which indicates recharge at lower temperature than today's (Andrews *et al.*, 1983; Loosli, 1983; Balderer, 1983).

In the following stratigraphically deeper aquifer in the deposits of the lower *freshwater molasse*, a different type of groundwater, with higher salinity (total mineralization 40–150 meq./l), is observed. The residence time can be deduced from the results of ^{14}C and ^{13}C analysis by means of the current correction models (whose application is limited by the fact that a carbon isotope exchange between water and the aquifer rock matrix had occurred, indicated by the high $\delta^{13}C$ values) and a possible time range of some 10^2 to 10^4 years results (Wigley, 1975, 1976). The stable isotope data indicate a different type of origin, as the corresponding values of δ^2H and $\delta^{18}O$ fall far below the global meteoric water line (Yurtsever and Gat, 1981). A possible explanation of the origin of these waters could be that a brackish-saline water (possibly modified formation water), enriched in $\delta^{18}O$ by evaporation effects, is mixed with a groundwater which has infiltrated under colder climatic conditions, as for example the water of the upper marine molasse aquifer described above.

Groundwaters in Jurassic deposits (*Malm, Dogger, Lias*). For the groundwaters originating from Jurassic deposits, the same distinction is observed between near surface groundwaters from springs and shallow wells originally from morphologically

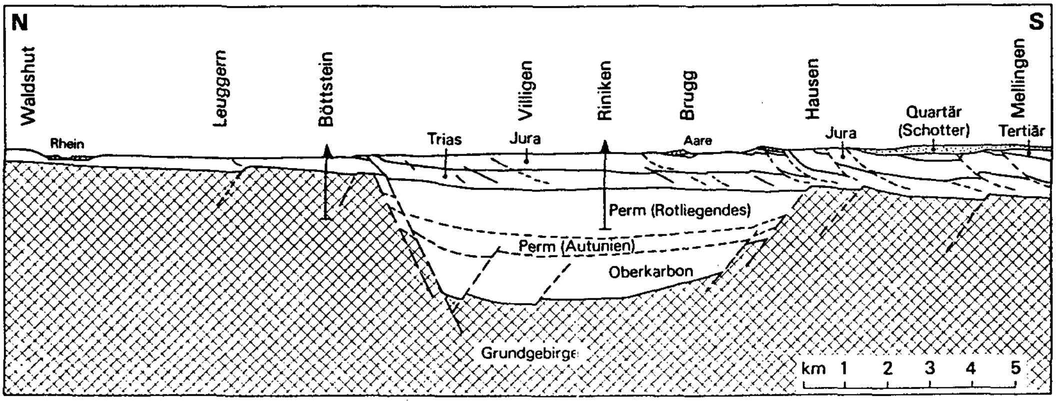

FIG. 3. Detailed cross-section through the region of northern Switzerland with the assumed Permo-Carboniferous trough (maximum depth about 2500 m below ground level) and the location of the NAGRA boreholes Bottstein (Kristal 1) and Riniken (Kristal 3) (After NAGRA, 1984).

defined recharge areas of mainly local flow systems and between groundwaters encountered in these formations in deep boreholes, where direct recharge from outcropping areas can only be possible at a large distance. Therefore, these groundwaters originate from intermediate or even regional flow systems with up to several orders of magnitude higher residence times.

Groundwaters from local flow systems typically occur in springs of the folded Jurassic mountains, and in some cases also at springs in the tabular Jurassic region. These shallow groundwaters are characterized by a low mineralization (total mineralization 3–8 meq./l) of the Ca–HCO_3 type (Dogger, Hauptrogenstein), Ca-Mg-HCO_3 type (Malm, Dogger, Hauptrogenstein), and Ca–Mg-HCO_3-SO_4-type (Dogger, Hauptrogenstein and Lias) [Schmassmann *et al.*, 1984]. Typical residence times of these groundwaters are in the range of the tritium method (Siegenthaler *et al.*, 1970), varying between recent to several years or at most up to about 30 years. (At some springs the outflowing groundwater represents probably also a mixture of local flow systems with some water of deeper circulation systems not containing any measurable tritium.) Recharge areas of these more near-surface circulating waters (as confirmed by the δ^2H and $\delta^{18}O$ data) are mostly in the nearby region of the tabular Jurassic mountains in the vicinity of the studied springs themselves (Siegenthaler and Oeschger, 1980).

Deep groundwaters from the *Upper Jurassic* (Malm) are only known from the north-eastern part of Switzerland (intersected by the NAGRA deep borehole Weiach) and from the southern part of Germany, north of the Swiss border and the lake Constance (region of Singen–Beuren Welschingen). From the hydrochemical and the isotope data and also from geological observations on oil exploration wells, it can be concluded that the groundwater of the Malm aquifers are hydraulically connected with the main aquifers of the molasse deposits especially with the upper marine molasse or to the lower freshwater molasse (as observed in the Weiach borehole).

For the *Middle Jurassic* deposits of the *Dogger* there are no important deep aquifers (caused by the lithological facies) known in the investigated area in Switzerland. But as observed from several boreholes in the region of the Rhine–Graben Zone north of Basle, the same formation can act as a regional aquifer under different lithological facies. The only borehole which yields a groundwater is Neuwiller in France, located near the Swiss border (lying in the southwards continuation of the Rhine-Graben). Its water chemistry is comparable with those of the upper marine molasse. The same formation of the Hauptrogenstein (Dogger) is used for thermal stations in the region of the Rhine Valley Graben in Germany (Bellingen, Steinenstadt, about 20 km north of Basle) but a saline water of Na–Cl-type is found. For the lower *Jurassic* deposits of the *Lias* formation, one spring as encountered at depth in the Weissenstein rail–road-tunnel could present a regional outflow point, with 16.5 meq./l total mineralization characterized as Na–Ca-(Mg)-HCO_3-SO_4 type water, with no detectable tritium but with δ^2H and $\delta^{18}O$ values which are in agreement with the assumed recharge area of the corresponding Jurassic mountain under actual climatic conditions.

Groundwaters from Triassic deposits. For the

Triassic formations, the existing aquifers and groundwaters can also be divided into two groups, one for the aquifers of local recharge and discharge areas, often connected with the appearance as springs above the hydrographic level of the corresponding rivers in the valleys.

The more local types of groundwaters are frequently observed at outflow of the *Keuper* formation (*Upper Triassic*). As its permeability is usually rather low (gypsum-containing marls) only local systems are observed, especially in the folded Jurassic area and at outcrops of the tabular Jura.

The groundwater issuing at these springs has the following features: as the groundwater from Keuper aquifers is often in contact with gypsum-bearing formations these waters are mainly saturated with gypsum and are of the Ca-Mg-SO_4-HCO_3 type which have a total mineralization of 20-35 meq./l (Schmassmann *et al.*, 1984). Mean residence times are according to the measured tritium concentrations of the order of several years and up to about 20 years. The δ^2H and $\delta^{18}O$ values are in agreement with the supposed recharge zones located not very far above the spring origins. Groundwaters from Keuper deposits, which belong to the undisturbed sedimentary areas above the crystalline bedrock and thus of regional extension, have only been encountered by boreholes at two locations (Beznau Gansinger-Dolomite and NAGRA borehole Riniken: Gansinger Dolomite/Schilfsandstein). From the analysis of the groundwater from the two locations the following characterization results: the water from the Riniken (KRISTAL-3) borehole has a total mineralization of about 15 700 mg/l of total dissolved solids and is of Na-Mg-Ca-SO_4-Cl-HCO_3 type. The water from the Beznau borehole (Gansinger-Dolomite formation) has a total mineralization of 14 600 mg/l and is of Na-Ca-Mg-SO_4-Cl-HCO_3 type. According to the ^{14}C and ^{13}C data, and using the current correction models (Reardon and Fritz, 1978; Fontes and Garnier, 1979; Wigley *et al.*, 1978), a model age for the mean residence time is in the range of several thousand years to more than 10^4 years for the Beznau Keuper groundwater and up to 2×10^4 years for the Riniken Keuper groundwater. For the Schilfsandstein groundwater from Riniken, the stable isotope values are higher than the modern values which would normally indicate recharge in warm conditions. It is not known at present how these values should be interpreted with respect to the possible residence time range using the ^{14}C model calculations.

Groundwaters from the Middle Triassic deposits of the *Muschelkalk* are well known (Vuataz, 1982), because this aquifer (especially from the series of dolomitic limestones of the Upper Muschelkalk) is of special importance to several thermal mineral stations; Baden, Schinznach, Losdorf (some have been known since Roman times or at least since the Middle Ages). Also for groundwaters from this Muschelkalk aquifer, a clear distinction has to be made between near-surface circulating, more locally recharged, waters, at most naturally occurring springs, and shallow boreholes in the region of the folded Jurassic and between the groundwaters yielded by deeper boreholes (or from hot thermal springs at tectonic fault-zones) of the normally stratified Muschelkalk deposits in the tabular Jurassic region above the crystalline bedrock and covered by younger sediments.

The near-surface waters of the upper Muschelkalk can be characterized as follows: the groundwaters are under-saturated with respect to gypsum ($S_I = \log IAP/KT = -1.1$ to 0.7) and of Ca-Mg-HCO_3-SO_4 type or if the SO_4^{2-} content becomes predominant over the HCO_3^- content, of Ca-Mg-SO_4-HCO_3 type with a total mineralization of 8-26 meq./l (Schmassmann *et al.*, 1984; Plummer *et al.*, 1976). According to the measured values of δ^2H and $\delta^{18}O$ and the geological-topographical situation of the outflow zones, these wells probably originate from local recharge areas under today's climatic conditions. Also the residence times are normally within the range of the tritium method, e.g. of about 2 to 20 years. In some special geological situations in the folded Jurassic region, a mixture with waters of more regional flow systems (without tritium) seems to occur, according to the low or intermediate tritium content at higher temperature and mineralization. The ^{14}C values as indicated by the $\delta^{13}C$ values are not far from the calculated initial activities which indicates residence times in the range of modern waters but also mixture with waters from regional systems (Siegenthaler, 1972; Siegenthaler and Schotterer, 1977). The groundwaters from the regional, normally stratified Muschelkalk aquifer derived from deep boreholes and at natural occurring hot springs, which are connected to tectonic faults—or overthrust zones, are characterized by a different type of water which is mainly near saturation or saturated with respect to gypsum, anhydrite, calcite, and dolomite, and have also a lower negative standard redox potential and are therefore accompanied by the occurrence of H_2S. Its composition can be characterized as of Ca-Mg-SO_4-HCO_3 type (examples are the waters from Losdorf borehole 3 and 4, and from Weiach, NAGRA deep borehole) or of the Na-Ca-Cl-SO_4 type waters (examples are the hot springs at Baden, the water from Beznau and the NAGRA deep boreholes of Böttstein and Riniken). The mineralization is between 12-50 meq./l for the first type and between 70 (Baden hot springs) and 23 meq./l (at the NAGRA deep-borehole Riniken) for the second type. Model calculation from the measured ^{14}C and ^{13}C values (correction restricted by the incongruent dissolution reaction between sulphate, calcite, and dolomite) results in very low initial ^{14}C activities. According to the models of Wigley-Pearson and Fontes and Garnier the model ages for the residence times are in the range of several hundred to several thousand years for the Ca-Mg-SO_4-HCO_3 type waters and for the highly mineralized saturated or even oversaturated waters with respect to sulphate and carbonate minerals in the range of 10^3 to more than 10^4 years.

For the deposits of the *Lower Triassic deposits* of the *Buntsandstein* only groundwaters derived from boreholes, which are intersecting this thin sandstone aquifer (normal thickness in the investigated area of northern Switzerland between 5 to 25 m) have been studied. Due to its special situation below low permeable limestones and the very low permeable salt and sulphate deposits of the lower Muschelkalk but above and in some cases in direct contact with the crystalline basement or the permo-carboniferous deposits of the permian trench, it acts not as an individual aquifer but is in hydraulic connection with the groundwaters of the underlying rock. For instance, where it is lying on the crystalline basement it yields a groundwater which is very similar to the granite waters (e.g. at the NAGRA deep borehole, Böttstein) with lower mineralization and of Na-SO_4-HCO_3-Cl type but where it is in direct contact to large permian or even Permo-Carboniferous rocks (Rotliegend and Stephanien) it yields a highly mineralized Na-(Ca)-SO_4-Cl type water with a total mineralization between 100 and 223 meq./l (NAGRA deep borehole, Weiach). According to the ^{14}C results the calculated model-ages cover a broad range of residence times, from near modern (more than 30 years) to 4×10^4 years. The large variation of residence times of the groundwaters of the Buntsandstein aquifer at the different locations indicates different circulation systems with different flow direction and therefore also with different recharge zones and recharge conditions.

Groundwaters in the Permian deposits. The only groundwaters from Permian deposits obtained so far derive from the NAGRA deep borehole Riniken. These waters are highly mineralized (18 g/l) and contain H_2S and CO_2. These waters have δ^2H and $\delta^{18}O$ values which are much heavier than all other observed ground- and recharge-waters. Residence time determinations by ^{14}C results are restricted (because of high mineralization and availability of CO_2 from organic matter) and give results in the range of 10^3 to 5×10^3 years.

Groundwaters from the crystalline basement. As in the investigated area in northern Switzerland the crystalline basement is covered by the different sedimentary layers and only outcrops near the Rhine river, near the northern border (in the region of Sackingen and Laufenberg). All the groundwaters intersected in the different boreholes have signatures of longer flow paths. For this reason, near surface groundwaters, representing more the real recharge type with low mineralization and short residence times, are found only in the outcropping region of Black Forest massif and also in the Alps (known from groundwaters from the Grimsel area). In different boreholes in northern Switzerland the following types of groundwaters obtained from the crystalline basement are known: a Na-SO_4-HCO_3-Cl type, a Na-(Ca)-Cl type and a high mineralized Na-Ca-HCO_3-SO_4-Cl type (Schmassmann *et al.*, 1984). The first chemical type is found in the boreholes of Zurzach (thermal station) and Böttstein. These waters, with a total mineralization of only 14 to 18 mval/l seem to be typical of the geological situation where the crystalline basement is in close contact with the Buntsandstein and Permian deposits are either missing or are of only very low thickness. For these wells, the residence times, according to the ^{14}C and ^{13}C results, are of the order of 6×10^3 to greater than 1.8×10^4 years for Böttstein (for the undisturbed state of hydraulic conditions, according to the samples taken during the drilling period) and of the order of 1.5×10^4 to 2.0×10^4 years for the waters from Zurzack. For those waters a recharge from the Black Forest Massif in the north is the most probable. The second chemical type of Na-Cl or Na-Ca-Cl water is found at Sackingen (thermal station with one spring 'St Magarithenquelle' and a new spring by a borehole 'Badquelle'). For these waters, reflecting mixtures of low mineralized recent waters (as shown by the measurable tritium content) with higher mineralized waters (without tritium), the true end member composition cannot be determined. However, these mixed granite waters have a mineralization of up to 60 meq./l and possibly show influences of the saline highly mineralized groundwaters found in the boreholes with large Permian deposits.

The last type of granite waters has been found at a depth of about 1000 m below the top of the crystalline rock at the NAGRA deep borehole Böttstein (about 1300 m below ground level). As far as it can be deduced from the contaminated sample, the uncontaminated end-member formation water would have a total mineralization of about 200 meq./l. The origin of the highly mineralized water has not yet been resolved.

Conclusions

As shown by this presentation of the first results of isotopic and hydrochemical investigations, it is necessary, as a next step, to understand the real processes taking place during the groundwater flow, not only for the different aquifers in the different geological formations in the region but also to obtain an indirect picture of the spatial distribution of the different hydrodynamic flow systems. For this purpose, the results of hydrodynamic model calculations have to be validated carefully by the interpreted results of hydrochemical and isotopic determinations. Only in the synthesis of these different methods is it possible to define a conceptual picture of the actual hydrodynamic flow systems of relevance to the disposal of highly radioactive waste in the Geological formation of northern Switzerland.

Note. This article does not represent exactly the oral presentation at the Hallimond Lecture meeting in November, 1983. It was preferable to present the actual state of the results of this study (July 1984), and to give the more conceptual and theoretical background of the hydrogeological interpretation in another publication (Balderer, 1984).

Acknowledgements. This report results from the co-ordinated research project between the Low-Level Counting Department of the Physics Institute of the University of Berne and the NAGRA. I specially would like to thank Professor H. Oeschger and Drs J. C. Alder and H. H. Loosli for promoting this research work. I am especially grateful for the valuable discussions with other members of the NAGRA hydrochemistry-group; Drs H. Schmassmann, W. Kanz, and A. Pekdeger. For discussions on isotope interpretation I thank W. Rauert, W. Stichler, Drs J. Beer and M. Andree, Professors J. Ch. Fontes, and P. Fritz, Drs M. Ivanovich and F. J. Pearson, U. Schotterer, Dr U. Siegenthaler; and for discussion on hydrogeology and geology Professor A. Burger, Dr L. Kiraly, Professor H. Jackli, and Drs H. Beck, S. Schlanke, and M. Thury.

REFERENCES

Andrews, J. N., Balderer, W., Bath, A. H., Clausen, H. B., Evans, G. V., Florkowski, T., Goldbrunner, J. E., Ivanovich, M., Loosli, H. H., and Zojer, H. (1983) *Environmental isotope studies in two aquifer systems.* IAEA-SM-270/93 IAEA, Vienna.

Balderer, W. (1983) *Bedentung de Isotopen. Methoden bei der hydrogeologischen Charakterisierung potentieller Endlager standorte fur Hochradioaktive Abfalle.* NAGRA Techn. Rept. 83-04 Baden, Switzerland.

——(1984) *To the concept of interpretation of the results of isotope and hydrogeochemical determinations for the definition and characterisation of the natural conditions in geological formations.* Bull. Centre d'Hydrogeologie, No. 5, Universite de Neuchatel, Switzerland.

Diebold, P., and Sprecher, C. (1983) *Seismik-Kampagne 1983/84: Fortfuhrung grossraumiger geophysicalischer Sondierungen.* In *NAGRA informiert* No. 3 and 4. NAGRA, Baden, Switzerland.

Fontes, J. Ch., and Garnier, J. M. (1979) *Determination of the initial ^{14}C activity of the total dissolved carbon: a review of the existing models and a new approach.* Water Resources Research, **15**, No. 2.

International Atomic Energy Agency (1983) *Isotope Techniques in the Hydrogeological Assessment of Potential Sites for the disposal of High-Level Radioactive Wastes.* Techn. Rept. Ser. No. 228, IAEA, Vienna.

Loosli, H. H. (1983) Earth Planet. Sci. Lett. **63**, 51–62.

NAGRA (1983*a*) Techn. Rept. 83-02, Nagra, Baden, Switzerland.

——(1983*b*) Techn. Rept. 83-03, NAGRA, Baden, Switzerland.

——(1984) *NAGRA informierte über Ergebnisse ihrer Forschungen in NAGRA Aktuell*, No. 5, May 1984, NAGRA, Baden, Switzerland.

Plummer, L. N., Jones, B. F., and Truesdell, A. H. (1976) *WATEQF-A FORTRAN IV Version of WATEQ, a computer Program for Calculating Chemical Equilibrium of Natural Waters*, Water Resources Investigations 76-13. U.S.G.S.

Reardon, E. J., and Fritz, P. (1978) *J. Hydrology*, **36**, 201–24.

Schmassmann, H. J., Balderer, W., Kanz, W., and Pekdeger, A. (1984) *Regionale Untersuchungen uber die Beschaffenheit der Tiefengrundwasser.* NAGRA Techn. Rept. NTB 84-21.

Siegenthaler, U. (1972) *Bestimmung der Verweildauer von Grundwasser im Boden mit radioaktiven Umweltisotopen (^{14}C, Tritium).* Gas Wasser Abwasser, No. 9.

—— and Oeschger, H. (1980) *Nature*, **285**, 314–417.

——Oeschger, H., and Tongiorgi, E. (1970) *Tritium and Oxygen-18 in natural water samples from Switzerland.* Isotope Hydrology, IAEA Vienna.

——and Schotterer, U. (1977) *Hydrologische Anwendungen von Isotopenmessungen in der Schweiz.* Gas Wasser Abwasser, No. 7.

Toth, J. (1962) *J. Geophys. Res.* **67**, 4375–87.

Vuataz, F. D. (1982) *Hydrogeologie, Geochimie et Geothermie des eaux Thermales de Suisse et des Regions Limnitrophes, Materiaux pour la geologie de la Suisse.* Hydrologie, No. 29. Commission d'Hydrologie, Organe de la Société Helvétique des Sciences Naturelles. Kummerli & Frey, Bern, Switzerland.

Wigley, T. M. L. (1975) *Carbon-14 Dating of Groundwater from Closed and Open Systems.* Water Resources Research, 11, No. 2.

——(1976) *Nature*, **263**, 219–21.

——Plummer, L. N., and Pearson, F. J. (1978) *Geochim. Cosmochim Acta*, **42**, 1117–39.

Yurtsever, Y., and Gat, J. R. (1981) *Atmospheric Waters.* In *Stable Isotope Hydrology: Deuterium and 18-Oxygen in Water Cycle* (J. R. Gat and R. Gonfiatini, eds.) IAEA, Vienna. Techn. Rept. Ser. 210.