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Preliminary studies of groundwater flow and migration of uranium isotopes around the Oklo natural reactors (Gabon)

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Abstract

In specific zones of the Oklo uranium deposit, criticality was naturally reached 2 Ga ago. This site thus provides a unique opportunity to show whether the stable nuclear reaction end-products have remained or not in the vicinity of the reactor zones after the termination of nuclear reactions. In addition, the evaluation of the stability of the uraninite matrix over very long periods of time provides information on the possible long-term stability of waste forms such as spent fuel. The Commission of the European Communities initiated in 1991 the Oklo Natural Analogue Programme, a part of which is devoted to present-day migration studies. The Swedish Nuclear Fuel and Waste Management Company (SKB) supports this programme, with special interest in the Bangombe reactor, a shallow reaction zone possibly affected by surficial alteration processes. The Oklo study comprises hydrogeology, groundwater chemistry, isotopic analyses (environmental isotopes, U series, $^{235}U/^{238}U$), and modelling. Two sites are being thoroughly investigated: the less perturbed OK84 reactor zone in Okelobondo (200 m south from Oklo) and the Bangombe reactor zone, 30 km south of Oklo. We focus our study on uranium migration from these reactor zones, using tracers such as the $^{235}U/^{238}U$ isotope ratio. After preliminary field campaigns, a conceptual model was constructed, both for Okelobondo and Bangombe. For this purpose,

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groundwaters have been characterised for three years in different areas around Oklo: Okelobondo groundwaters in mines and boreholes and surface waters, and Bangombe, both in boreholes and surface waters. Detailed investigations were then conducted in order to validate our conceptual models, and finally to enabling us to model U migration from the reaction zones, and to evaluate the performance assessment of deep geological disposal of radioactive wastes. After the presentation of regional and local geology and hydrogeology, we give a complete description and interpretation of groundwater chemical evolution both in Okelobondo and Bangombe. It concerns major-element behaviour, redox control and uranium behaviour. Preliminary conclusions for the conceptual model drawn from hydrogeological studies and from groundwater chemistry agree closely. In addition, we have successfully tested the use of $^{235}U/^{238}U$ isotope ratios in groundwaters to evaluate the migration of uranium from reactor zones.

1. Introduction and aims

Most regulatory authorities require qualitative evaluations of the repository performance for up to 1 Ma. The prediction of this future behaviour is facilitated by comparison with the evolution of natural geological systems, especially those including high amounts of radioactive elements. Such natural systems and natural analogues provide the opportunity to evaluate the most important geochemical processes which should affect the entire repository system over periods of time out of reach in laboratory experiments. They also support the direct and indirect validation of methods, models and data used in performance assessment. Among other internationally funded natural analogue studies, such as Poços de Caldas, Brazil (Chapman et al., 1991) and Cigar Lake, Canada (Cramer and Smellie, 1992), Oklo in Gabon is the only one in which fission products and transuranium elements ever occurred, due to natural nuclear reactions (Bodu et al., 1972, Neuilly et al., 1972). After many studies devoted to descriptions and interpretations in terms of nuclear reactor physics, recently synthesised by Naudet (1991), the interest for applications to nuclear waste disposal problems has been progressively increasing since 1975 (Cowan, 1975; Hagemann and Roth, 1978; Brookins, 1981; Curtis et al., 1983). Many aspects of the evolution of the Oklo reactors are now studied in this respect, through the CEC (Commission of the European Communities) Oklo Natural Analogue Project (Chapuis and Blanc, 1992; von Maravic, 1992) which is coordinated by the French Atomic Energy Commission (CEA) Institute for Protection and Nuclear Safety (IPSN). Our contribution is devoted to the following items: regional hydrogeology and hydrochemistry; present-day water–rock interaction in the vicinity of reactor zones, in order to gather information on the hydrogeological and chemical conditions which allowed the preservation of the ore and reactors for almost 2 Ga; estimation of the migration of uranium from the reactor zone, using the depletion of the $^{235}U/^{238}U$ isotopic ratio resulting from nuclear fission, which provides a natural tracer of uranium migration in surrounding groundwaters; and use of observations, data and models for performance assessment of nuclear waste repositories. Preliminary hydrogeological and hydrogeochemical investigations have been described in a recent CEC report (Bourguet et al., 1992; Gurban et al., 1992; Moulin and Vilarem, 1992; Smellie et al., 1992; Toulhoat et al., 1992).

2. Initial conceptual models at Okelobondo and Bangombe sites

The purpose of this "far-field" part of the Oklo natural analogue study is to identify and then to model the present transfer of groundwater which might mobilise elements produced in the reaction zone. Two sites were proposed for this study: (1) a deep one, Okelobondo (400 m below the surface), that offered the possibility to study the transfer of reactor zone product elements over a 400-m distance, through a very heterogeneous stratigraphy; and (2) a shallow one, Bangombe (15 m below the surface), representing a more homogeneous stratigraphy, but being much more altered. The uranium deposits at Oklo/Okelobondo and Bangombe occur in the basal stratigraphic unit of the Francevillian Sedimentary Series of early Proterozoic age (2150 Ma) which lie unconformably on the Archean basement (2700 Ma). The basal formation (FA) is 500–1000 m thick and consists of interbedded sandstones and conglomerates. The uranium mineralisations at the studied sites are lying at the top of FA formation and have been dated to 2 Ga (Bonhomme et al., 1982). The reactor zones are contained within these uranium deposits. The FA formations overlain by the FB formation are dominated by black shales (Gauthier-Lafaye et al., 1989).

2.1. *Okelobondo*

2.1.1. *Geology*

Overlying the Archean gneiss bedrock, is a monoclinal sedimentary series dipping towards the east, and consisting of detritic rocks, mainly a sandstone–schist–pelite, interbedded with sandstone and so-called "complex" dolomites. A bedrock vertical fault which has an almost north–south orientation constitutes the western border of the sedimentary unit. A representative vertical profile is shown in Fig. 1. Above an upper, very thick, basal layer of FA quartzite sandstone, mineralised at the top, lies the FB series which is divided in: green pelites, pelites 1, complex 1, pelites 2, F1b sandstones, ampelites 1 and 2, complex 2 and pelites 3. Measured isotopic depletions of ^{235}U in the Okelobondo area have confirmed the existence of some reactor zones. The vertical profile shown in Fig. 1 passes through the OK84 area, where petrographic criteria confirm the presence of one reactor.

2.1.2. *Hydrogeological conceptual model*

The geological cross-section and the topographical relief suggest two possible flow paths: (1) a flow path at depth emanating from the eastern recharge zone and discharging along the Mitembe River valley (which is the local drainage system), with the likelihood of interacting with the mineralised layer and reactor zone OK84; and (2) another deep flow path fed from the fault zone west of the Mitembe reaching the same discharge area after passing through the FA sandstones. The conceptual hydrogeological flow pattern proposed made possible a preliminary hydrodynamic modelling of current migration for a first quantification of flow path and transport velocity. The computer code used for two-dimensional (2-D) model calculations is the finite-element METIS code (Goblet, 1989). The first modelling results were an isopotential map and stream line distribution. The calculated velocity distributions were used to calculate trajectories

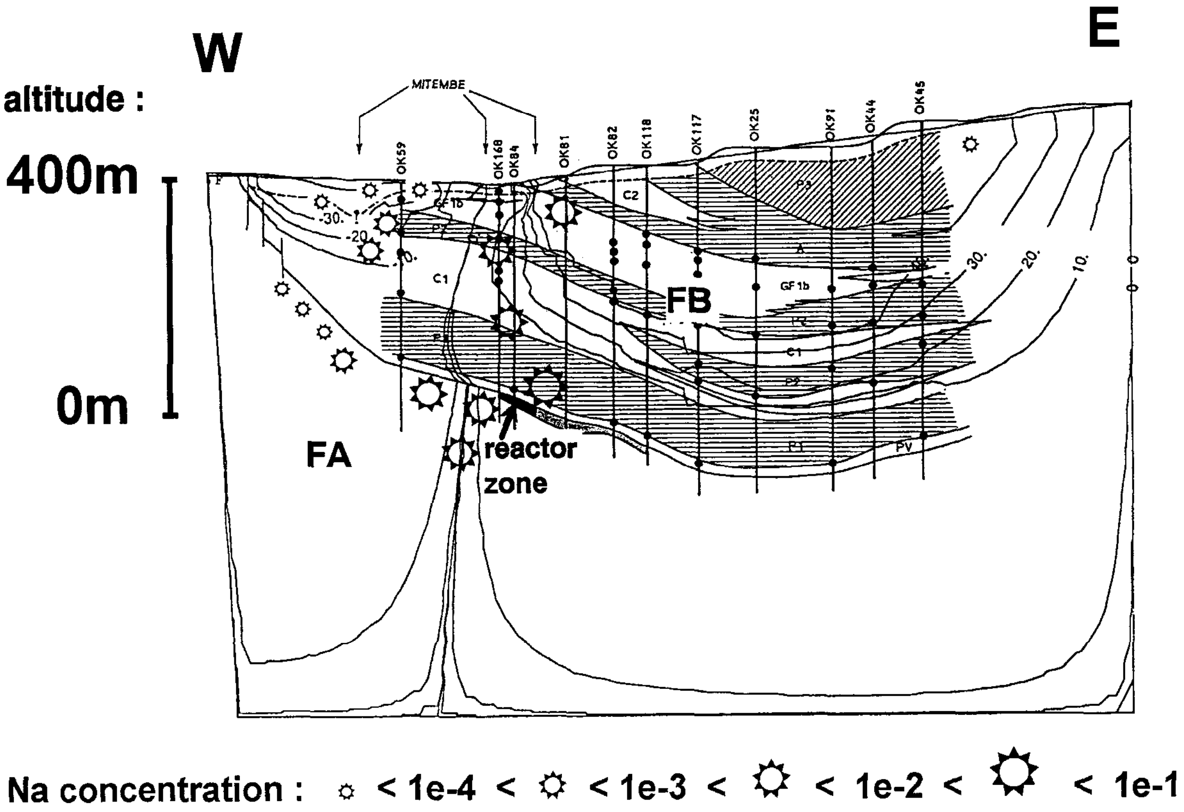

Fig. 1. W–E cross-section through the OK84 Okelobondo reactor zone. Overlying the FA formations and the mineralised horizon, the FB sequence is the following: PV = green pelites; P1 = grey pelites; C1 = Complexes 1; P2 = pelites 2; GF1B = sandstones; A = ampelites; C2 = Complexes 2; P3 = pelites 3. Stream lines are computed with the code METIS, using piezometric and permeability measurements. The local discharge is represented by the Mitembe River. Na concentrations (mol L^{-1}) are plotted. Most waters with high Na concentration plot on the longest stream lines in the general discharge area under the Mitembe River. These deep discharging groundwaters might intersect the OK84 reactor zone.

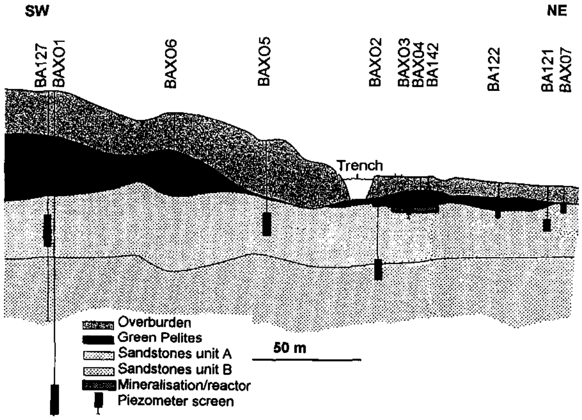

Fig. 2. SW–NE cross-section through the Bangombe reactor zone, showing the new piezometers, which are used to evaluate groundwater flow directions. (Same scale in vertical and horizontal directions).

and travel times for particles released in the model. The preliminary modelling shows that the OK84 area, located below the discharge area confluence, is promising for the continuation of the study of the hydrogeology and transport in the far-field (Fig. 1). The flow is directed upwards which facilitates water ascending from depth to have been in contact with the reaction zone; the area is a mixing zone between two types of deep-derived waters, one originating from the schisto-dolomitic sandstone formations, the other from the Archean bedrock and FA basement sandstones.

2.2. Bangombe

2.2.1. Geology

The Bangombe geology presents a strong analogy with Okelobondo, sharing the same geological stratigraphy (Fig. 2). A subdivision of the sedimentary sequence above the Archean basement in the Bangombe area can be made into a basal sandstone (FA) overlain by a pelitic sequence (FB1). The Bangombe uranium deposit constitutes, as does the Oklo/Okelobondo deposit, a mineralised horizon in the upper part of the FA sandstone. At the Bangombe deposit the pelitic member is in places strongly altered and in some places very thin. A fossil nuclear reaction zone was discovered in 1985. The reactor zone is located at a depth of ~ 12 m, at the foot of the plateau, which corresponds with the local drainage of the area, the Lekoni River.

2.2.2. Hydrogeological conceptual model

The topographical relief suggests a groundwater flow in a northeast direction, water infiltrating on the plateau (named COMILOG [1] Plateau) is assumed to discharge, due to the strong geological contrast, at the foot of the plateau and further downstream depending on the point of recharge. As in Okelobondo, the proposed conceptual

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hydrogeological flow patterns proposed made it possible to start with a hydrodynamic modelling of current migration. The computer code used for the 2-D model calculations is the USGS (U.S. Geological Survey) SUTRA code. The preliminary modelling shows that the reactor zone, situated in the discharge area of the domain, is interesting for the study of radionuclide transport in the far field: water recharged on the COMILOG Plateau discharges at the mineralisation and further downstream The preliminary results presented above showed the necessity to carry out a drilling programme for collecting new hydrogeochemical data to constrain the conceptual models.

3. Hydrogeological investigations

3.1. Hydrogeological studies at Okelobondo

Since 1988 the Oklo/Okelobondo site has been subject to field studies as part of the CEC–CEA–EMP Oklo Natural Analogue Study. In 1992/1993 an extensive drilling programme was launched in the vicinity of reactor zone OK84 during which three shallow boreholes and three boreholes from a mine gallery were drilled. These new piezometers serve to collect hydrogeological data: hydraulic head, hydraulic conductivity, as well as water samples. Measurements of depth to water level have been made in all completed piezometers, confirming the preliminary hypothesis of a general NE–SW groundwater flow having a discharge area in the Mitembe River bed. The piezometric level is ~ 375–380 m in the outlet area and ~ 435 m in the recharge area. Hydraulic tests have been performed in the new piezometers. Two vertical permeability profiles were performed in new boreholes in Okelobondo South area. An isopotential map along the conceptual section using the interpolated hydraulic heads is difficult to interpret because the whole Oklo/Okelobondo system is very perturbed by mining activity. The hydraulic test results will form the basis of assigning hydraulic conductivities to the different hydraulic units (10^{-8}–10^{-5} m s^{-1}).

3.2. Hydrogeological studies at the Bangombe site

In the autumn of 1992, seven new boreholes (six cored) were drilled together with the reopening and cleaning of four old exploration holes. Packer installations in eleven boreholes, plugging of old exploration holes through the reactor zone and groundwater sampling have been completed. These new piezometers serve to collect hydrogeological data: hydraulic head, hydraulic conductivity, as well as groundwater samples. Measurements of depth to water level have been made in all completed piezometers. Observations made during drilling and in old exploration holes suggest that the free water table is located ~ 10–15 m below the ground surface upstream and ~ 1 m below around the reactor zone. The altered pelitic strata above the water table are consequently unsaturated. Hydraulic testing has been performed during drilling, and after completion of the piezometers; in the latter case often in association with water sampling. An interference test conducted in the outlet area verified a hydraulic connection between the pelite layer and the underlying reactor zone. The effective values of hydraulic conductivity lie between 10^{-8} and 10^{-7} m s^{-1}.

4. Evolution of groundwaters

4.1. Preliminary remarks

Based on the hydrogeological conceptual modelling, we have sampled waters in recharge areas, discharge areas above and below reactors, and in major local aquifers. The access to the groundwaters is good in the mine and in the Oklo quarry, and poor in the FB series, as most boreholes have been lost. Nevertheless, we have been able to reconstruct the evolution of the groundwaters, in a way which is consistent with the hydrogeology, using major elements and environmental isotopes. Two major guides will be considered to evaluate groundwater evolution: (1) progressive acquisition of solutes by water–rock interaction and subsequent re-equilibration with secondary minerals; and (2) redox evolution and redox-sensitive element behaviour (Fe, U).

4.2. Water–rock interaction

Confinement is the most important mechanism: the dissolution of minerals from host rocks progressively increases pH, alkalinity, and major cations (Na, K, Mg, Ca). A good correlation with pH and depth is found, together with a decrease of tritium as water residence time increases. The progress of water–rock interaction for major silicates is clear: most waters plot in the kaolinite field, confined waters from the FA formation plot on the limit between low-temperature K-feldspar and kaolinite, confined water from P1 pelites (OKH2) plots in the area corresponding to an equilibrium with both K-feldspar and albite. In the last case, most major elements would be controlled by a secondary mineral assemblage (Grimaud et al., 1990), although the presence of these minerals should be checked in aquifer fractures. From Fig. 1, we can observe a very good agreement between the conceptual hydrogeological model and the evolution of waters as shown by Na concentrations: most Na-rich confined waters are found in the discharge area under the Mitembe River; these Na-rich waters correspond to the longest stream lines calculated by the computer code. From Fig. 3, the Ca/Mg ratio appears as a good tracer of groundwater origin, because of the abundance of dolomite in the FB sequence. Silica dissolution and regulation is well illustrated in Fig. 4 in which pH has been selected as an index of water–rock interaction progress (this is possible only in systems devoid of any Cl internal or external input, as the balance between released cations and mobile elements lowers equilibrium pH with silicates (Michard, 1989; Grimaud et al., 1990). One can describe a progressive increase of silica, reaching $8 \cdot 10^{-4}$ mol L^{-1}, which is close to the value at equilibrium with amorphous silica, followed by a sharp decrease, in order to reach the equilibrium value of $2 \cdot 10^{-4}$ mol L^{-1} for chalcedony, a mineral with which most confined aquifers are in equilibrium at low temperature. This evolution of silica concentration is less clear in the intermediate waters which have been collected in the western part of the Fut G in the FA formation. The consequence of this decrease in dissolved silica is the formation of colloidal silica, which tends finally to aggregate to form chalcedony. This is evidenced by the high abundance of newly formed small-size silica colloids in most confined waters, as mentioned above. Such colloids

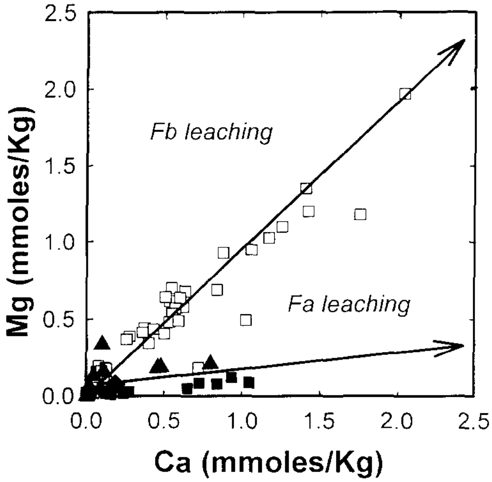

Fig. 3. Mg vs. Ca plot. Most groundwaters from the B aquifers plot on the first diagonal, indicating a leaching of dolomite, which is an ubiquitous mineral in the FB formation, whereas groundwaters from the FA aquifers have much lower Mg/Ca ratios. This is useful to evaluate the actual path of groundwaters through the sedimentary pile. *Hollow squares* = FB aquifers; *filled squares* = FA aquifers; *filled triangles* = Bangombe.

might be very important for radionuclide migration and sorption in the most confined waters. The evolution mechanisms are very similar in Bangombe, where the same tendency towards equilibration is noticed. Due to a more superficial environment, the

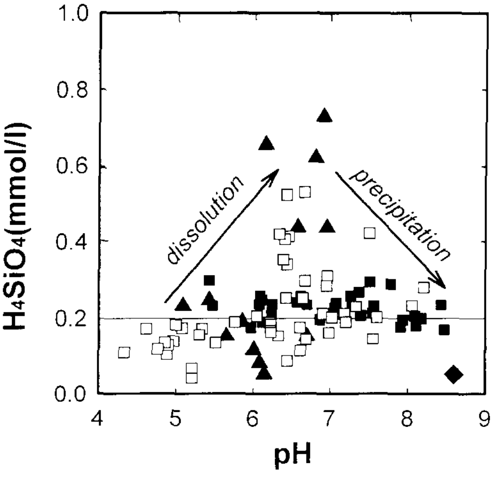

Fig. 4. Increase of dissolved silica concentration followed by a decrease, due to the leaching of primary silicates by superficial groundwaters, then re-equilibration with chalcedony in confined end-members. This process could explain the generation of small size newly formed silica colloids in some higher-pH waters. Chalcedony solubility at 25°C is plotted on the diagram ($2 \cdot 10^{-4}$ mol L^{-1}). *Filled diamond* = deep and confined water from P1 pelite, other *symbols* as in Fig. 3).

ultimate evolution stage (equilibrium with secondary feldspars) is never reached. However, the deepest water (BAXO1) is not so far from equilibrium with K-feldspar.

4.3. Redox evolution

We have shown that redox is controlled by an equilibrium between Fe(II) (dissolved iron, some times in equilibrium with siderite) and Fe(III), as $Fe(OH)_3$, ferrihydrite (Toulhoat et al., 1992). New data confirm this tendency which is a rather general observation in many aquifers devoid of sulphides (Beaucaire and Toulhoat, 1987; Grenthe et al., 1992). In most aquifer systems, reducing waters are generally found in the deepest and most confined parts. This is not the case here: the Eh-values are scattered, and are not correlated with depth or confinement. The most reducing waters are found in organic-matter-bearing aquifers, such as P2 pelites, whereas deep and chemically confined waters such as in Okelobondo Fut G east reach rather high Eh-values (+300 to +400 mV). This kind of lithological control of Eh is very important for the migration of redox-sensitive elements such as uranium. Consequently, some deep discharging waters carrying dissolved uranium (possibly from the reactor zones) could precipitate uranium when traversing organic-matter-rich formations during their ascent towards the surface. This might explain the uranium anomalies that have been described at the base of the P1 and P2 units (Bourguet et al., 1992). This type of lithological control of Eh is very important for performance assessment, regarding uranium migration and secondary confinement. Uranium concentration never exceed 10^{-7} mol L^{-1}, even close to the mineralisations. Groundwaters are saturated with respect to uraninite (and coffinite) only in reducing waters. Preliminary computations show that in oxidising waters, saturation with respect to schoepite and uranyl silicates is never reached, although some thermodynamically poorly known secondary uranyl silicates might be close to saturation (Gallien, 1994). In Bangombe, the lithological control of redox potential is also effective, but the most reducing waters are found in the immediate vicinity of the reactor (BAX O3), which has been shown to be very rich in organic matter and kerogen-type material (R Bros, pers. commun.). Such waters are also in equilibrium with uraninite and/or coffinite. The local buffering of redox conditions by organic matter, in conjunction with a low permeability, explains the good preservation of the reactor (large amounts of oxidising water should have quickly leached and removed the whole reactor).

4.4. Particles and organic materials

Most particles found in superficial waters are inherited mineral fragments, with a mean size above 1 mm. In more confined waters, a bivariate or more largely dispersed size spectrum appears, with a notable amount of particles below 1 mm, which are generally composed of spheroidal silica particles, and aluminosilicate colloids. Other interesting colloids are iron colloids which are present in large quantities in some mildly reducing groundwaters (Okelobondo Fut G west). They appear as 100–200-nm spheres that tend to aggregate. These colloids are formed by the oxidation of dissolved Fe(II), which precipitates as ferrihydrite. Three hypotheses might account for their formation:

(1) these colloids are really existing in situ and transported in groundwaters; (2) they are formed just before water exits in the gallery after oxidation by atmospheric oxygen; and (3) they form during sampling and sample processing. In reality, contributions from all three mechanisms probably occur. The preservation of redox conditions after the opening of a tunnel is probably very difficult as oxygen readily diffuses into the water-conducting fractures, causing redox fronts. A more precise answer concerning the origin of such colloids might be found by measuring uranium series disequilibrium in particles: no ^{230}Th should be found in those which are formed during sampling (this will be tested in the future). The characteristics of colloids from Bangombe are very similar to those of Okelobondo, i.e. colloids formed by condensation processes in the most confined waters (BAXO1). In Okelobondo, DOC (total organic carbon) ranges between 1 and 3 ppm, whereas in Bangombe, it is more concentrated in waters close to the reactor (BAXO3).

5. Environmental isotopes

Water stable isotope results from the last set analysed (October–December 1992) clearly split the Okelobondo groundwater system in three groups. One group includes samples OK1, OK2, OK3, OKH1 and OKH2 (eastern and deeper part of the G gallery) which have depleted isotopic contents. Surface waters which show the highest isotopic content of the set define the second group. Samples from the Complexes system and from the western part of the G gallery lie along the GMWL (Global Meteoric Water Line) between these two end-members (Fig. 5). We interpret this difference in water isotopes in terms of different recharge processes. For the surface water group, the main recharge is of local origin at present time. For the East and deep G gallery samples, the

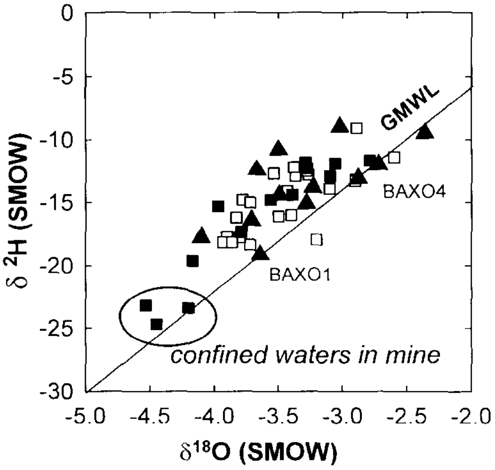

Fig. 5. Environmental isotopes in Okelobondo: δ^2H (‰ vs. SMOW) vs. δ^{18}O (‰ vs. SMOW). The most confined groundwaters of Okelobondo Fut G east plot on the lower left corner of the diagram, which indicates different recharge conditions (same *symbols* as in Fig. 3). *GMWL* = Global Meteoric Water Line.

recharge area is at a higher altitude than the Mounana valley or recharge occurred under cooler climatic conditions than the present one. Although the second hypothesis cannot be disregarded, the hypothesis of an higher altitude in the recharge area and therefore longer transit time for deep samples is more likely. This hypothesis fits with results obtained from radioactive environmental isotopes and dissolved gases. Most of these deep samples are free of atmospheric tritium and the data we have on the radiocarbon content of TDIC (total dissolved inorganic carbon) in OK3 and OKH3 give a mean transit time estimation for groundwater between 10.0 ± 1.7 and 20 ± 2 ka. From sample OKH3, a rough and preliminary calculation of dissolved helium accumulation gives a contact time between groundwater and FA sandstone of 10 ± 1 ka. These waters are defined as "confined" within the hydrogeochemical study. Groundwaters which belong to the group having water isotopic content intermediate between shallow waters and deep waters represent shorter aquifer pathways or mixing between the two end-members. Water samples coming from the OK84 reaction zone (OKH1e and OKH2e, parts of boreholes before measuring chambers) show very high tritium content (> 27 TU) with respect to other samples in this zone. We believe that such an anomalous content results from in situ production of tritium rather than from a recent water contamination. To estimate such production we intend to obtain details of the mineralogy and the water content of this reactor zone. In Bangombe, we can again discriminate between different types of groundwater. BAXO4 and BAXO3 represent local recharge at present. In contrast, BAXO1 represents aquifer circulation extending to the recharge area on the plateau and therefore represents longer transit time. We calculated a mean transit time for BAXO1 from TDIC radiocarbon data of 7.5 ± 0.5 ka. The other samples may reflect a mixing of these two types of recharge in the discharge zone. The helium and methane accumulation in the shallow system seems to evidence an aquifer connection between the reaction zone (BAXO3) and the pelites (BAXO4) above it.

6. Use of uranium isotope ratios as tracers of reactor leaching

Given this set of hydrogeochemical data from the Oklo far-field programme, it was possible to investigate the existence of a natural tracer of nuclear reaction zones in present-day groundwaters. We have considered $^{235}U/^{238}U$ as the best tracer, as uranium is soluble in non-reducing conditions and is the clearest signature of the effect of nuclear reactions in the reactor core uraninite. In addition, ^{235}U and ^{238}U both initiate their respective radioactive decay chains. Therefore, α-recoil or any geochemical perturbation which are encountered using ^{234}U and ^{238}U cannot be considered. Based on hydrogeochemical considerations (degree of groundwater evolution, flowpaths, etc.), we have selected six samples for uranium isotopic analyses. After uranium separation and purification (Toulhoat and Beaucaire, 1987), we have used thermo-ionic mass spectrometry to measure the isotopic ratios. In order to prevent mass fractionation effects which are critical for precise isotopic determinations, we have used a new method ("total consumption method") which integrates all ions produced during ion emission at different current levels (Dubois et al., 1992). From the six samples, two show significant deviations from the natural uranium isotopic composition: OK2 and BA 145); other

Table 1
Isotopic composition of uranium in groundwaters

Reference	X	Y	Z	[U] (mol L^{-1})	$^{235}U/^{238}U$	$\pm 2\sigma$
OK1	295772	9842950	68	$2.32 \cdot 10^{-9}$	0.007253	0.000010
OK2	295565	9843057	114	$1.45 \cdot 10^{-9}$	0.006872	0.000010
OK195-102 m	295940	9843110	308	$1.17 \cdot 10^{-9}$	0.007260	0.000010
BA122-7 m	309318	9828269	355	$5.63 \cdot 10^{-8}$	0.007257	0.000010
BA122-48 m	309318	9828269	314	$7.94 \cdot 10^{-8}$	0.007248	0.000010
BA145-8 m	309299	9828248	355	$8.85 \cdot 10^{-9}$	0.007095	0.000010
Laboratory standard					0.007256	0.000010
Natural U					0.007254	

samples are not significantly different from "normal" uranium (Table 1). Sample BA 145 has been collected in a drill-hole which intersects a known nuclear reaction zone in Bangombe, confirmed by Holliger (1991). Consequently, groundwater in contact with depleted uraninite is able to record significant uranium isotopic anomalies, and the success of our approach is clearly demonstrated. The proportion of uranium originating from the reaction zone is difficult to evaluate, as ^{235}U depletion is not regularly distributed in the reaction zones. In the Bangombe project (Smellie et al., 1992), we plan to evaluate the zone of influence of reaction zones in a steady-state hydrodynamic situation. No influence of ^{235}U depletion is measured in drill-hole BA122 (8-and 48-m depth), which is situated ~ 20 m downstream to BA 145. This preliminary result is in itself very encouraging for further studies which may be applied to repository performance safety assessments. Sample OK2, in which the strongest anomaly has been measured, was sampled in a short drill-hole that penetrates the mineralised zone. From the COMUF files, it can be assessed that the OK175 borehole, which is located 20 m further towards the east, contained some depleted uranium (Bourguet et al., 1992). A further study of this area is planned in order to confirm and to evaluate the extension of this ^{235}U-depleted mineralised area. OK1 and OK195, which are close to the Okelobondo depleted zone OK84, do not show any anomaly which allows us to estimate the spatial distribution of the "influence zone" of OK84 concerning far-field uranium migration. Although preliminary, these results are very encouraging in the scope of our study, as they clearly prove the relevance of our approach. $^{235}U/^{238}U$ depletion in groundwaters is clearly detected in groundwaters that have been in contact with reaction zones, and this will be used as a powerful tracer of uranium migration from reactions zones, independently of the chemical evolution of dissolved uranium (sorption, dissolution, precipitation).

7. Integration of data, final conceptualisation

At this stage of our study, the following available data should permit constraint, to varying degree, of our mathematical models in order to establish the final models including the relevant processes. The next step will be a successive integration of the new data: physical and chemical parameters, followed by the radionuclide transport modelling. Finally, coupled hydrological–geochemical modelling will be performed. The combined results of these modelling efforts can be used in a performance assessment context, indicating which hydraulic processes and features are relevant. Among these are: (1) well-defined hydraulic pathways and barriers; (2) velocity distribution (heterogeneity) and associated travel times; and (3) processes governing speciation of solutes. The understanding of these features and processes together with the hydrogeochemical controls will help to explain the transport of elements associated with the reactor zones, such as the distribution of the $^{235}U/^{238}U$ isotopic ratio, which is the main purpose of this far-field part of the Oklo natural analogue programme.

8. Conclusions

We have obtained a complete description and understanding of groundwater flow and chemical evolution in Okelobondo and Bangombe, including major-element behaviour, redox control and uranium behaviour in groundwaters. One notes the existence of a good agreement between hydrology and hydrochemistry. Concerning U migration from reaction zones, in Okelobondo the particular geological setting could allow migration and subsequent trapping in reduced horizons. In Bangombe, there is an intense buffering of redox conditions in the reactor zone, preventing U migration. This has been tested using the $^{235}U/^{238}U$ isotope ratio in groundwaters as a tracer of reactor zone leaching. The feasibility and importance of this method are clearly demonstrated. The ultimate aim of this study is the integration of hydrology and hydrochemistry in a general coupled model.

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