Oklo, Natural Analogue of the radionuclides migration through the geological barrier

Oklo, Analogue Naturel de la migration de radionucléides dans l'environnement

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Abstract – One of the main part of the CEC project "Oklo-Natural Analogue" is devoted to present time migration studies. This part comprises hydrogeology, groundwater chemistry, isotope geochemistry and modelling. Two sites are being investigated: the less perturbed reactor zone of the Oklo mine (OK84 in the southern mine extension of Okélobondo) at around 400 meters depth and the Bangombé reactor zone, sited in a shallow environment 30 km south of Oklo.

The present contribution aims to define regional hydrogeology and hydrochemistry boundary conditions for the modelling exercise, to assess the present day water-rock interaction in the vicinity of reactor zones, to gather information on the geochemical conditions which allowed the preservation of reactor zones for two billions years, to estimate the uranium migration from the reactor zone in using a natural marker (the depleted $^{235}U/^{238}U$ ratio resulting from the fission) and to compare these data with predictive modelling.

Based on the hydrogeological conceptual modelling, we have sampled waters in recharge areas, discharge areas above and below reactors, and in major local aquifers. 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. Three types of groundwater circulations corresponding to different interaction/transit time within the system can be described. One type is low pH (< 6), poorly buffered, high pCO_2, low Total Dissolved Salt. It corresponds to shallow waters. Confined waters with high pH (> 8), high alkalinities (> 10^{-3} moles.l^{-1}), equilibrium with respect to carbonates and secondary alumino-silicates (kaolinite, low T° adularia and albite, chalcedony) are deep groundwaters from deep pelite horizons or pene-

trating the basement. In between, waters have pH ranging from 6 to 7, alkalinities from 10^{-4} to 10^{-3} moles.l^{-1}, silica content which reflects intense leaching, increase of TDS with increasing pH. These waters which have a tendency to equilibrium with carbonates minerals are intermediate groundwater circulating into the "Complexes" formation.

Water stable isotopes results split the groundwater samples in three groups. One group includes samples from the deeper part of the Okélobondolo mine and a discharge outlet of deep flow lines in Bangombé, which have depleted isotopic contents. Surface and shallow waters which show the highest isotopic content of the set define the second group. Others samples lie along the GMWL between these two end-members. 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 isotopically depleted samples, the recharge occurred under cooler climatic conditions than present one.

In Okélobondo, the deep samples are free of atmospheric tritium and the data we have on the radiocarbon content of TDIC give a mean groundwater transit time estimation above 10,000 years. 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.

We have obtained a complete description and understanding of groundwater flow and chemical evolution in Okélobondo and Bangombé, including major element behaviour, redox control and uranium behaviour in groundwaters. One notes the existence of a good agreement between hydrology and hydrochemistry.

Both markers of water origin and water transit time indicate aquifer systems recharged under steady state conditions and having in this point of view, the same boundary conditions. They indicate as well that it is possible to extend the data from the shallow system (Bangombé) to the deeper one (Okélo).

The hydrochemical study validates the hydrological conceptual models of Okélobondo and Bangombé. The fossil reactor zones are sited in a discharge zone of deep flow paths having a mean transit time of some thousands of years. In Bangombé, however, the local hydrodynamical and chemical conditions are strongly influencing the radionuclides migration. The chemical control for the uranium remobilization is clearly local and associated to the occurrence of organic matter within the reaction zone. In Okélobondo, groundwater chemical conditions at the depth of the reaction zone are favourable to the migration of radionuclides. Reducing horizons are evidenced within the aquifer system down-flow the reaction zone. Geochemical markers show that these horizons could have act as chemical trap of dissolved radionuclides.

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. Based on hydrogeochemical considerations (degree of groundwater evolution, flowpaths, etc.), we have selected samples for uranium isotopic analyses.

Results of a complete cross-section over the Bangombé reactor zone is presented. Groundwater in contact with depleted uraninite is able to record significant uranium isotopic anomalies, and it is possible to evaluate the zone of influence of reaction zones in a steady-state hydrodynamic situation and to compare it with a predictive modelling exercise.

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.

Keywords: Oklo, Natural Analogue, isotopes hydrology, geochemistry.

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RÉSUMÉ – La préservation des réacteurs de fission nucléaire fossiles d'Oklo (Gabon) pendant près de deux milliards d'années dans les formations géologiques du Francevillien inférieur suppose une interaction limitée entre les matériaux des zones de réaction et les eaux souterraines. Cette interaction peut être observée et étudiée sur le système hydrogéologique actuel, ce qui offre la possibilité d'analyser et de modéliser le comportement d'un terme source (combustible nucléaire irradié) par rapport aux circulations aquifères établies dans le champ lointain (barrière géologique). Les objectifs du programme d'hydrogéochimie présenté ici étaient d'établir les conditions aux limites et les paramètres à utiliser dans la modélisation de la remobilisation potentielle de matériau de ces zones par les eaux souterraines puis d'étudier par le suivi d'un traceur naturel spécifique des zones de réaction (le rapport $^{235}U/^{238}U$) cette remobilisation.

L'étude a été menée simultanément dans deux contextes hydrogéologiques. La mine d'uranium d'Oklo, où la zone de réaction fossile considérée (OK84) est située en profondeur (400 mètres environ sous la surface du sol) sous une

couverture stratigraphique hétérogène. Le modèle hydrogéologique conceptuel construit pour la région d'Oklo place le réacteur fossile OK84 en zone de décharge des eaux souterraines. La profondeur du site et son environnement hydrogéochimique sont, dans ce cas, proche de la situation d'un stockage géologique. On a étudié parallèlement un deuxième site, le site de Bangombé, à 30 km au sud d'Oklo, où la zone de réaction est située près de la surface (une dizaine de mètres sous la surface du sol) toujours en contexte de décharge des eaux souterraines. Sur ce site, des circuits aquifères plus courts et dégagés de toute perturbation liée à l'exploitation minière, peuvent être échantillonnés au plus près de la zone de réaction et permettre une modélisation mieux contrainte de l'effet de l'interaction eau-roche de basse température sur le transport actuel de radionucléides.

L'étude chimique des eaux souterraines met en évidence l'importance du mécanisme de confinement et classe les circulations aquifères en fonction de leur degré d'évolution chimique : 1) des eaux à faible pH (< 6), faibles teneurs ioniques, représentant des circulations superficielles dans les niveaux d'altérites d'Oklo et de Bagombé ; 2) des eaux à fort pH (> 8), forte alcalinité ($> 10^{-3}$ M) et fortes teneurs ioniques (Na^+), ayant atteint l'équilibre par rapport aux minéraux carbonatés et tendant à s'équilibrer avec les alumino-silicates, collectées dans les horizons les plus profonds du système aquifère d'Oklo et témoignant de circulations profondes s'équilibrant en milieu silicaté ; 3) des eaux à pH compris entre 6 et 8, ayant des alcalinités de 10^{-4} à 10^{-3} M, des teneurs ioniques augmentant avec le pH et une tendance à l'équilibre par rapport aux minéraux carbonatés, représentant des circulations de type intermédiaire entre les circulations superficielles et les circulations profondes. Ces eaux sont prélevées à Oklo dans les niveaux du toit de la formation uranifère (niveau des « Complexes »). Certains échantillons, prélevés dans la zone générale de décharge d'Okélobondo intègrent des circulations profondes et des circulations intermédiaires venant des Complexes.

L'étude des isotopes du milieu de l'eau et des sels dissous, qui permet de remonter à l'origine de la recharge et à son temps de transit dans l'aquifère, montre que les eaux les plus confinées ont le temps de transit le plus long et procèdent d'une paléorecharge. L'étude paléohydrologique met en évidence la permanence de la recharge au moins sur les 20 000 dernières années.

Les résultats chimiques et isotopiques valident les modèles hydrogéologiques conceptuels d'Oklo et Bagombé et fixent les conditions aux limites de la modélisation du transport de soluté. La modélisation du transport autour de la zone de réaction de Bagombé doit prendre en compte la dynamique locale des eaux souterraines conditionnée par une recharge récente empruntant des circuits courts et se mélangeant au flux régional. Le contrôle chimique du transport est lui clairement local et associé à la présence de matière orga-

nique dans la zone de réaction ; à Oklo, les flux souterrains au niveau des zones de réaction, sont potentiellement remobilisateurs de radionucléides (conditions oxydantes). Des horizons réducteurs sont mis en évidence dans le système aquifère en aval des zones de réaction. Des indices géochimiques montrent que ces horizons ont pu fonctionner comme pièges à radionucléides transportés en solution.

Le rapport isotopique $^{235}U/^{238}U$ a été considéré comme traceur car l'uranium est relativement soluble dans les eaux peu réductrices d'Oklo et il s'agit de la signature géochimique la plus tangible des zones de réaction. À Bagombé, la distance pour laquelle on détecte la migration actuelle de l'uranium appauvri venant de la zone de réaction dans les eaux souterraines est restreinte à une dizaine de mètres autour de cette zone. L'injection de ces données dans un modèle de simulation du transport de soluté permettra de quantifier ces distances.

Mots clés : Analogue Naturel, géochimie isotopique, isotopes du milieu, hydrogéologie, Oklo.

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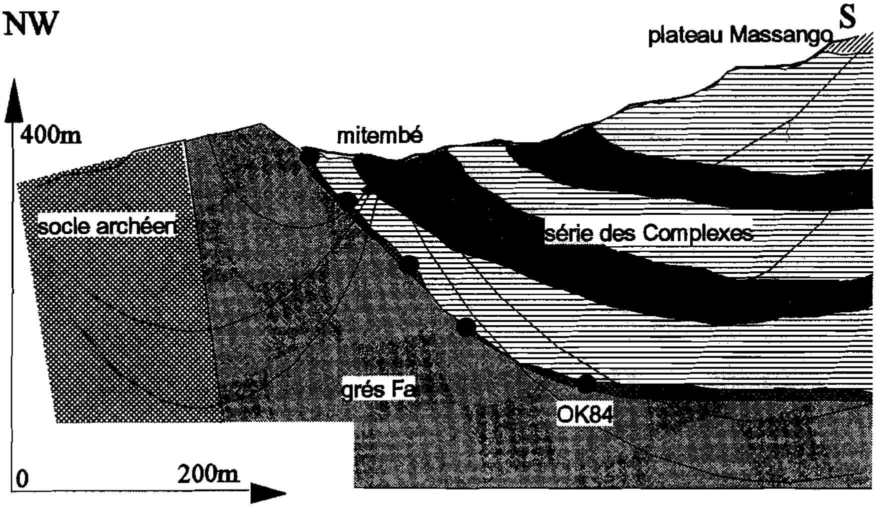
INTRODUCTION AND AIMS

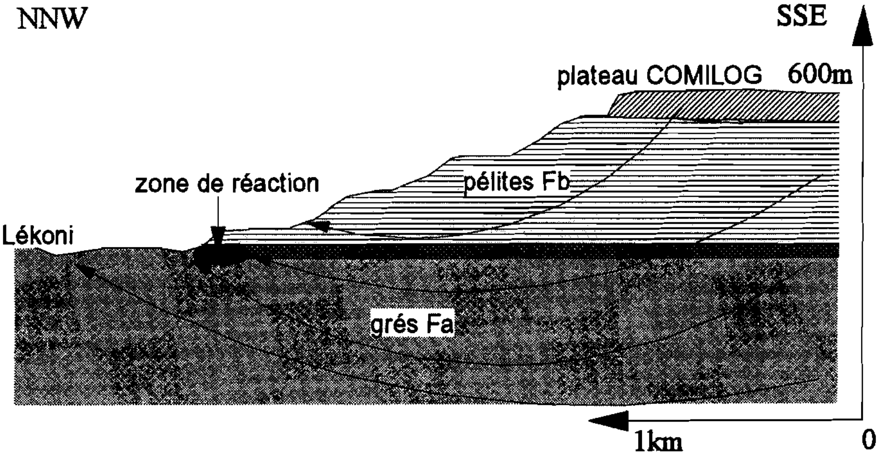
To evaluate quantitatively the long term environmental impact of a geological radioactive waste disposal, geologists invented the Natural Analogue Methodology. Natural systems and processes similar to those that will occur in radioactive waste disposal, are studied as long term natural experiments and used as test-beds for repository performance assessment models.

Among several geological sites where natural analogue studies were developed, Oklo is the only one in which fission products and transuranium elements ever naturally occurred, due to the existence of natural nuclear fission reactors (Bodu *et al.*, 1972). After many studies devoted to nuclear reactor physics (see a synthesis by Naudet, 1991), the interest for applications to nuclear waste disposal problems has progressively increased (Cowan, 1975, Hagemann and Roth, 1978, Brookins, 1981, Curtis *et al.*, 1983). Despite the fact that important quantities of fission products remained within a geological environment over a long period of time, Oklo was never considered as representative of a typical waste disposal site at the global scale. The analogy is rather considered for retention and migration mechanisms in the geosphere of non reprocessed spent fuel components (uraninite, fission products and actinide decay products). Many aspects of the evolution of the Oklo reactors are now studied in this respect through the European Commission Oklo Natural Analogue Project (Von Maravic, 1992, Chapuis and Blanc, 1992) which is



Schematic hydrogeological cross-section over the Okélobondo Site.

Coupe hydrogéologique schématique du site d'Okélobondo.



Schematic hydrogeological cross-section over the Bagombé Site.

Coupe hydrogéologique schématique du site de Bagombé.

FIGURE 1.

(from Gurban *et al.*, 1994)

co-ordinated by the French Atomic Energy Commission (CEA) and groups several research institutes of European Union member states.

The present contribution which aims to illustrate the use of Isotope Hydrology in the field of the geological disposal of radioactive waste, is based on the far-field part of the Oklo Natural Analogue Project. The far-field study was lauched in order to gather information on the hydrogeological and chemical conditions which allowed the preservation of the ore and reactors for almost two billion years and to use these observations and data to test models for performance assessment of nuclear waste repositories.

The reported results on regional hydrogeology and hydrochemistry: present day water-rock interaction, estimation of the uranium migration from reactor zone, using the depleted $^{235}U/^{238}U$ isotopic ratio resulting from nuclear fission which provides a natural tracer of reactor zone material migration in groundwaters, are entirely based on papers from Toulhoat *et al.* (1994) and Louvat *et al.* (1995).

GEOLOGY AND HYDROLOGICAL CONCEPTUAL MODELS

The Oklo uranium mine is located on the territory of the Gabonese Republic, in the southeastern province of Haut-Ogooué. The project investigations are concentrated in two main areas, the Okélobondo mine, southern extension of the Oklo mine, near the city of Mounana, and the Bagombé site near the city of Moanda, 30 kilometers south of Mounana.

The uranium deposits at Oklo/Okélobondo and Bangombé occur in the basal stratigraphic unit of the Francevillian Sedimentary Series of Lower 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 mineralisation at the studied sites is lying at the top of FA formation and has been dated to around 2000 Ma. The reactor zones are contained within these uranium deposits. The FA formation is overlain by the FB formation which is dominated by black shales (Gauthier-Lafaye *et al., 1989).*

The two sites investigated for this study are a deep one, Okélobondo (400 m below the surface) that offered the possibility to study the transfer of reactor zone product elements over a 400 m vertical distance through a very heterogeneous stratigraphy and a shallow one, Bagombé (12 m below the surface), representing a more homogeneous stratigraphy but being much more altered.

Hydrogeological conceptual model of Okélobondo

The geological cross-section and the topographical relief suggest two possible flow paths: 1) a flow path at depth emanating from the eastern recharge

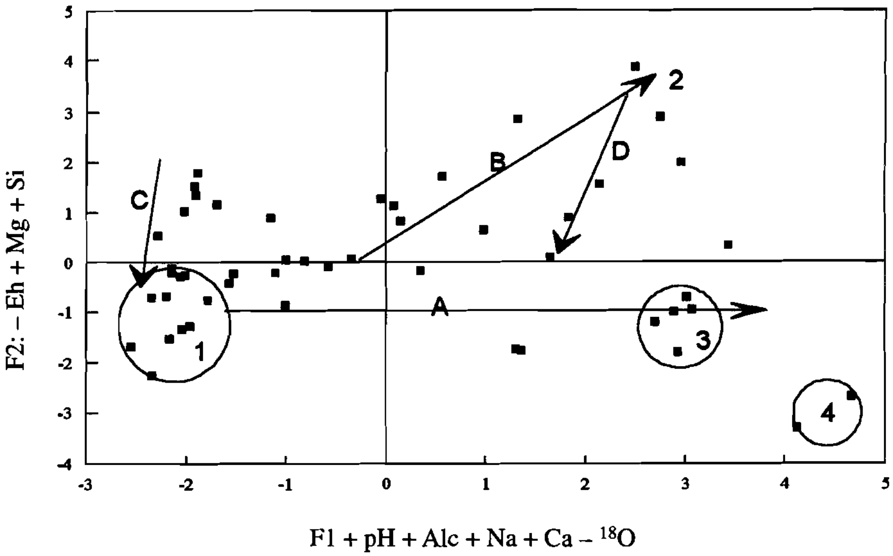


FIGURE 2.

Plot of the 2nd vs 1st factors of the PCA • groups: 1, surface and shallow waters; 2, waters from the Fb formation at Okélobondo; 3 and 4,waters from the deep part of the Okélobondo system (3: FA formation and 4: p1 pelites) • trends: A, confinement; B, evolution within the FB Complexes formation; C and D, redox effect at Bagombé (C) and Okélobondo FB formation (D).

Facteurs 1 et 2 de l'ACP • Groupe 1 : eaux de surface et superficielles ; Groupe 2 : eaux de la formation FB à Okélobondo ; Groupes 3 et 4 : eaux de la partie profonde du système d'Okélobondo (3: formation FA et 4: pélites P1) • Tendance A: confinement; Tendance B : évolution dans la formation des complexes FB ; Tendances C et D : effet redox à Bagombé (C) et dans la formation FB à Okélobondo (D).

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; 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 (Figure 1, from Gurban *et al.*, 1994). The preliminary modelling shows that the OK84 area is located below the discharge area confluence. 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.

Hydrogeological conceptual model of Bangombé

The reactor zone is located at a depth of approximately 12 m, at the foot of a plateau, which corresponds with the local drainage of the area, the Lekoni River. The topographical relief suggests a groundwater flow in a north-east direction, water infiltrating on the plateau (named COMILOG 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 (Figure 1, from Gurban *et al.*, 1994). As in Okélobondo, 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.

EVOLUTION OF GROUNDWATERS

Using the hydrogeological conceptual modelling, groundwaters have been sampled in recharge areas, discharge areas above and below reactors, and in major local aquifers. The access to the groundwaters is good in Bamgombé, in the mine and in the Oklo quarry, and poor in the "Complexes" series (FB formation at Oklo). Nevertheless, it was possible to reconstruct the evolution of the groundwaters, in a way which is consistent with the hydrogeology, using major elements and environmental isotopes.

Based on the statistical treatment of the data (Figure 2), two major guides are 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).

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 (Figure 3). 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 (basal FB) plots in the area corresponding to an equilibrium with both K feldspar and albite (Figure 4). 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 Figure 5, 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 waters (confined waters) are found in the discharge area

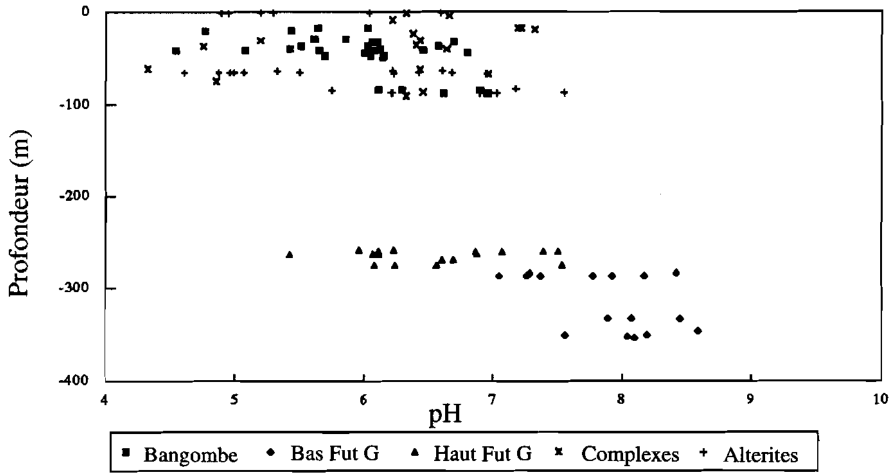


FIGURE 3.

Groundwater pH vs depth of sampling.

pH des eaux en fonction de la profondeur de prélèvement.

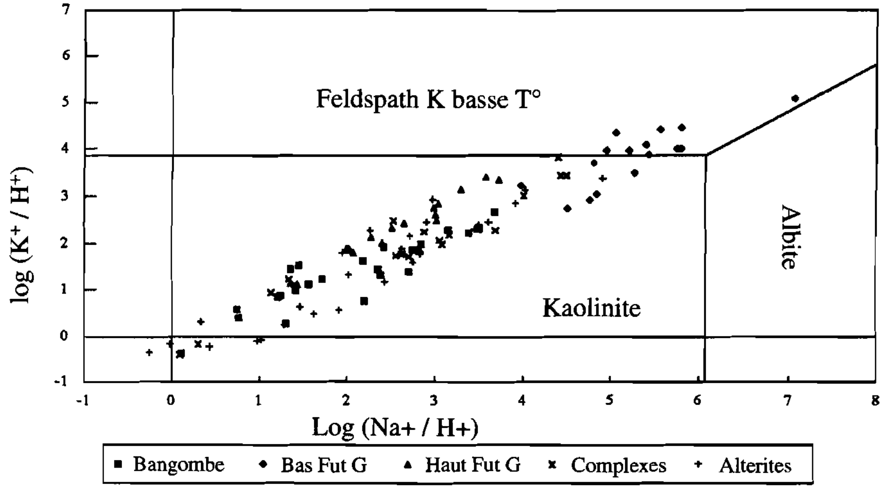


FIGURE 4

Activity diagram with phase boundaries; confined water from FA and P1 pelite formations are referenced as "bas Fut G" on the diagram.

Diagramme d'activité avec limites des phases ; les eaux confinées des formations FA et pélites P1 sont notées « bas Fut G » dans le diagramme.

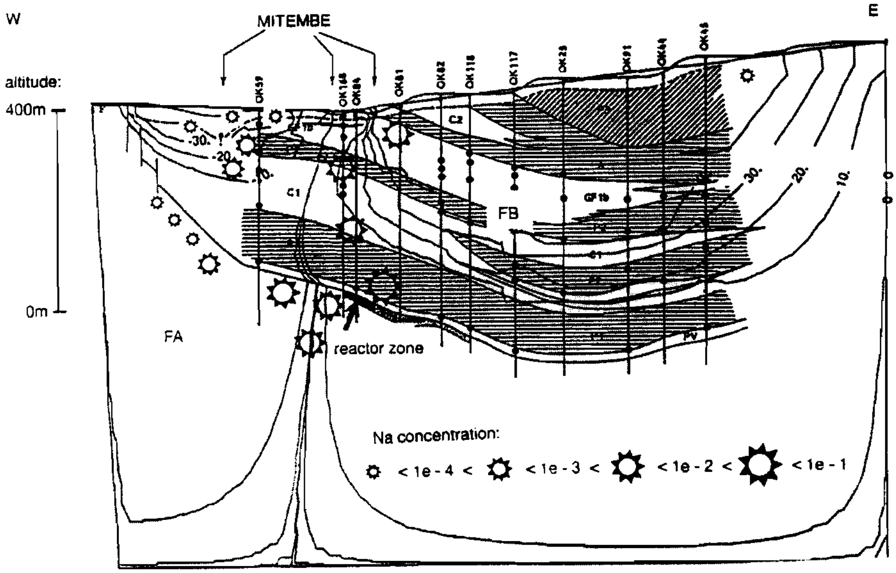


FIGURE 5 (Toulhoat *et al.*, 1994).

Cross-section through OK84. Steam lines are computed with the code METIS using piezometric and permeability measurements. Dissolved Na contents (mole.l^{-1}) are plotted at the level of the sampling points. Waters with high Na content plot on the longuest flow lines.

Coupe de OK84. Les lignes de courant sont calculées par le logiciel METIS à partir des mesures de piézométrie et de perméabilité. Les teneurs en Na dissous (mole.l^{-1}) sont mentionnées au niveau des points de prélèvement. Les eaux à haute teneur en Na se situent sur les lignes de flux les plus longues.

under the Mitembe river, Na-rich waters correspond to the longest stream lines calculated by the computer code.

Redox evolution

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 (Figure 6). The most reducing waters are found in organic matter-bearing aquifers, such as P2 pelites, whereas deep and chemically confined waters such as in deep FA horizon reach rather high Eh values (300 to 400 mV). The 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. This tendency is a rather general observation in many aquifers devoid of sulphides (Beaucaire and Toulhoat,

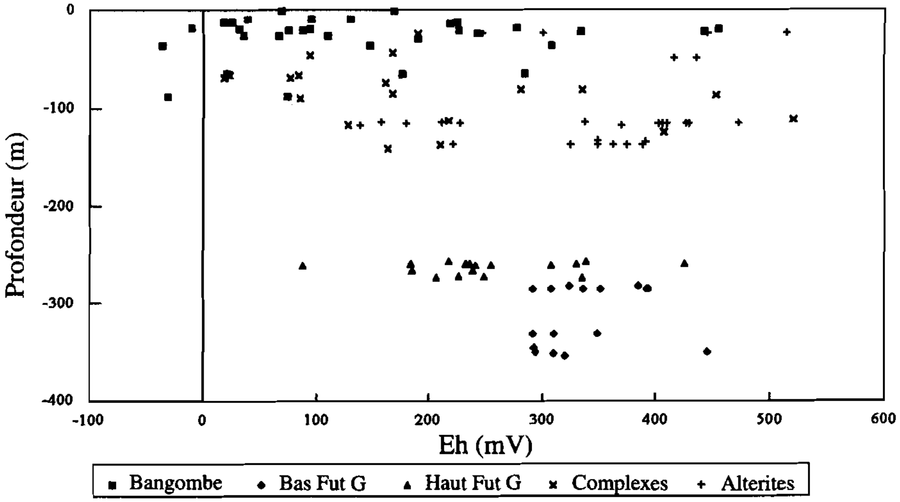


FIGURE 6.

Groundwater Eh vs depth of sampling.

Eh des eaux en fonction de la profondeur de prélèvement.

1987, Grenthe *et al.*, 1992). 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.

Conclusion on groundwater chemistry

Three types of groundwater circulations corresponding to different water-rock interaction/transit time within the system can be described.

— The first type is low pH (< 6), poorly buffered, high pCO_2, low Total Dissolved Salt. It corresponds to shallow waters.

— Confined waters with high pH (> 8), high alkalinities (> 10^{-3} mole.l^{-1}), equilibrium with respect to carbonates and secondary alumino-silicates (kaolinite, low temperature adularia and albite, chalcedony). These waters are deep groundwaters from deep pelite horizons or penetrating the basement.

— In between, waters having pH ranging from 6 to 7, alkalinities from 10^{-4} to 10^{-3} mole.l^{-1}, silica content which reflects intense leaching, increase of TDS with increasing pH. These waters which have a tendency to equilibrium with carbonates minerals are intermediate groundwater circulating into the "Complexes" formation (FB at Oklo-Okélobondo).

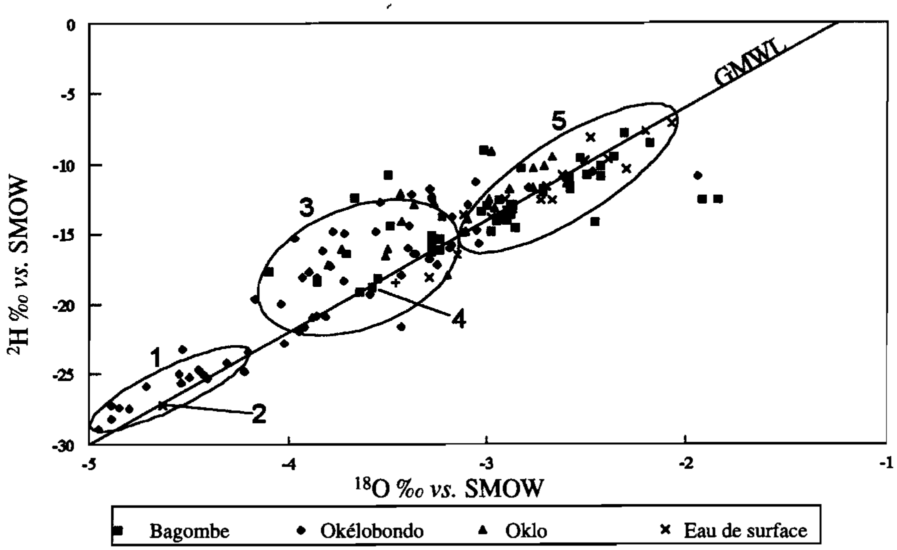


FIGURE 7.

Plot of the stable isotope content of groundwater • groups: 1, samples from the deeper horizon of the Okélobondo system (FA and pl formations); 2, deep discharge at Bagombé; 3, samples from FB formation at Oklo and Okélobondo; 4, BAX01; 5, surface and shallow waters at Okélobondo and Bagombé.

Teneurs en isotopes stables des eaux souterraines • Groupes: 1, eaux des horizons les plus profonds du système d'Okélobondo (formation FA et pélites pl) ; 2, décharge profonde à Bagombé ; 3, eaux de la formation FB à Oklo et Okélobondo; 4, BAX01 ; 5, eaux de surface et superficielles à Okélobondo et Bagombé.

GROUNDWATER ORIGIN AND DYNAMIC

The isotope contents of groundwater recharged under the present conditions are set with respect to the isotopic content of regional atmospheric precipitation. The variation of tritium content in precipitation since its increase from surface thermonuclear tests has been reconstructed on the basis of existing records from the IAEA-WMO network in Equatorial Africa (IAEA, 1992). ^3H content of the present recharge is taken at 5.5 ± 0.8 TU. The stable isotope contents of river and spring water are taken as representative of those of precipitation and identified as the present recharge end-member (Figure 7).

Groundwater origin

Water stable isotopes results split the groundwater samples in three groups. One group includes samples from the deeper part of the Okélobondo system and a discharge outlet of deep flow lines in Bagombé, which have depleted isotopic contents. Surface and shallow waters which show the highest isotopic content of the set define the second group. Samples from the "Complexes" (FB) system at Okélobondo and the deepest sampling point of Bagombé (BAX01) lie along the GMWL between these two end-members (Figure 7). This difference in water isotopes is interpreted in terms of difference in the origin of the groundwater recharge. For the surface water group, the main recharge is of local origin at present time. For the isotopically depleted samples, the recharge occurred either under cooler climatic conditions than present one or on recharge area higher than those of the surface water group. The present altitude effect on the water stable isotopes content has been investigated over the studied area. A depletion effect in the ^{18}O content of – 0.2 ‰ per 100 meters of elevation has been measured. This value is rather low but comparable to other observations in this region (Fontes and Olivry, 1976). Considering this value, there are no differences in the altitude of the catchment basins within the studied area which can account for the isotopic depletion of the deep groundwaters. This depletion therefore results from recharge processes under cooler climatic conditions than present one.

Groundwater dynamic

In Okélobondo, the deep samples are free of atmospheric tritium. Shallow waters have tritium content between 5 and 6 TU reflecting present recharge conditions and short transit time. Samples coming from boreholes sited into the "Complexes" formation have an intermediate tritium content between 5 TU (shallow waters) and 0 TU (deep situation).

The transit time estimation based on radiocarbon content of TDIC for deep samples, OK3 and OKH3, give respectively 11200 ± 1800 and 21700 ± 1700 years. Groundwaters which belong to the group having water stable isotopes and tritium 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 above packers intervals) show very high tritium content (above 27 TU) with respect to other samples in this zone. This content results from the *in situ* production of tritium generated via the secondary neutron flux produced by the alpha emissions from the uranium mineralization.

In Bagombé, BAXO4 and BAXO3 represent recent local recharge. At the opposite, BAXO1, which is tritium free, represents aquifer circulation extending to the recharge area on the plateau and therefore represents longer transit time. The mean transit time calculated for BAXO1 from TDIC radiocarbon data is 6300 ± 1800 years. The other samples may reflect a mixing of these two types of recharge within the discharge zone.

Paleohydrological information

In site characterization, paleohydrological reconstruction improves the robustness of boundary conditions used in flow-mass transport modelling. However as pointed out by Fontes *et al.* (1993), various processes tend to smooth recorded climate fluctuations in aquifers.Therefore, for low-frequency climate signals, as it is the case in Equatorial region, the use of groundwater as archives of climate is limited. Owing to that, the approach used in this study is rather to verify the consistency of groundwater isotopes evolution with respect to already existing paleoclimate reconstruction for Equatorial Africa.

In Figure 8, estimated transit time based on $^{14}C_{TDIC}$ content of 3 samples representing 3 different flow-lines are plotted *versus* their water stable isotopes content. The longest transit time calculated, the lower ^{18}O content is. These data are compared with mean air temperature on the Chaillu massif, estimated during the last glacial maximum and measured today (Maley and Elenga, 1993). Two air temperature reconstructions for Equatorial Africa, based on statistical treatement of fossil pollen records (Bonnefille *et al.*, 1990; Vincens *et al.*, 1993), have been added in Figure 8 for comparison to the Chaillu situation.

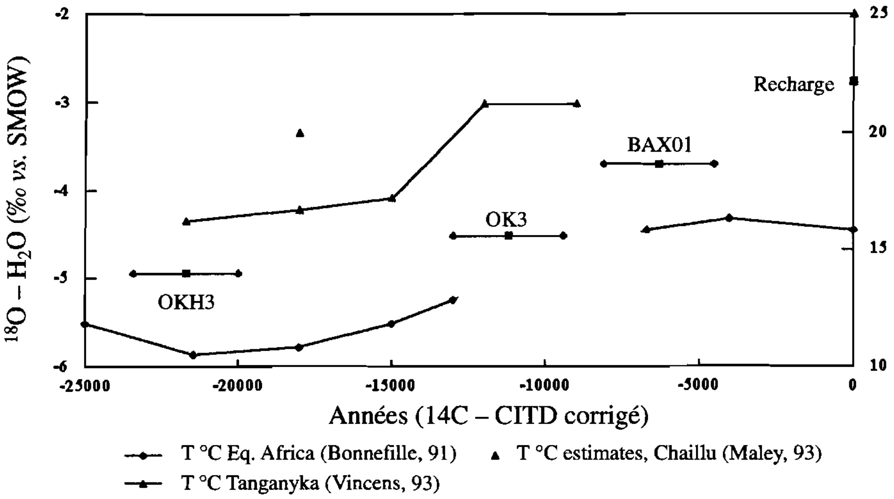


FIGURE 8.

Comparison of groundwater isotope content evolution with time in Okélobondo and Bagombé aquifer systems and mean air temperature reconstruction for Equatorial Africa.

Comparaison de l'évolution au cours du temps des teneurs en isotopes des eaux des systèmes aquifères d'Okélobondo et de Bagombé et de la reconstitution de la température moyenne de l'air pour l'Afrique équatoriale.

The mean isotope/temperature gradient that can be estimated for the Oklo region, since the cooling related to the last glacial maximum, is of 0.5 ‰ / 1 °C and is consistent to what is estimated for other intertropical regions (Stute, these proceedings). Taking into account the aquifer dispersion, the ^{18}O evolution within the aquifer systems of Okélobondo and Bagombé is consistent with temperature evolution over time.

CONCLUSION ON CONCEPTUAL HYDROLOGICAL MODELS

A complete description and understanding of groundwater flow and chemical evolution in Okélobondo and Bagombé, including major element behaviour, redox control and uranium behaviour in groundwaters has been obtained. One notes the existence of a good agreement between hydrology and hydrochemistry.

Both markers of water origin and water transit time indicate aquifer systems recharged under steady state conditions and having, in this point of view, the same boundary conditions. They indicate as well that it is possible to extend the data from the shallow system (Bagombé) to the deeper one (Okélobondo).

The hydrochemical study validates the hydrological conceptual models of Okélobondo and Bagombé. The fossil reactor zones are sited in a discharge zone of deep flow paths having a mean transit time of some thousands of years.

In Bagombé, however, the local hydrodynamical and chemical conditions are strongly influencing the radionuclides migration. The chemical control for the uranium remobilisation is clearly local and associated to the occurence of organic matter within the reaction zone.

In Okélobondo, groundwater chemical conditions at the depth of the reaction zone are favourable to the migration of radionuclides. Reducing horizons are evidenced within the aquifer system down-flow the reaction zone. Geochemical markers show that these horizons could have act as chemical trap of dissolved radionuclides.

U ISOTOPES RATIO AS TRACER 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. $^{235}U/^{238}U$ has been considered 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, alpha recoil or any geochemical perturbation which are encountered using ^{234}U and ^{238}U cannot be considered. Samples selection for uranium isotopic analyses, has been based on hydrogeochemical considerations (degree of groundwater evolution, flowpaths, etc.).

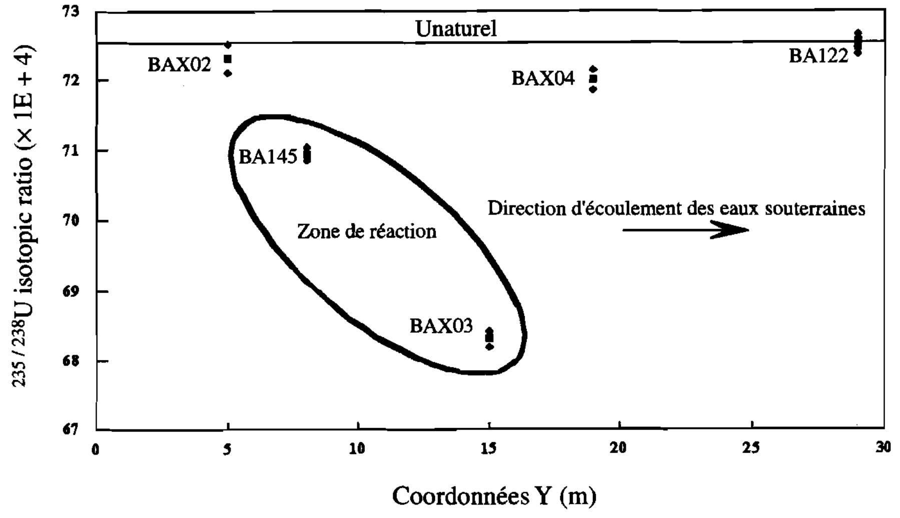


FIGURE 9.

Evolution of the 235/238 ratio of dissolved uranium over a cross section through the Bagombé reaction zone. Waters sampled in boreholes BA145 and BAX03, drilled through the reaction zone show a depleted U isotopic ratio. Up-flow, sample from BAX02 borehole shows a natural U isotopic ratio. Down-flow, the influence of the reaction zone on U isotopic ratio can still be recorded in BAX04 but not in BA122.

Évolution du rapport 235/238 de l'uranium dissous sur une section de la zone de réaction de Bagombé. Les eaux prélevées dans les sondages BA145 et BAX03, forés dans la zone de réaction, montrent un rapport isotopique de l'uranium appauvri. En amont, l'eau du forage BAX02 montre un rapport isotopique naturel. En aval, l'influence de la zone de réaction sur le rapport isotopique de l'uranium en solution peut encore être détectée sur BAX04, mais non sur BA122.

Results of a complete cross-section over the Bangombé reactor zone is presented in Figure 9. Groundwater in contact with depleted uraninite (BAX03 situation) is able to record significant uranium isotopic anomalies, and it is possible to evaluate the zone of influence of reaction zones in a steady-state hydrodynamic situation and to compare it with a predictive modelling exercice (Gurban *et al.*, 1995).

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 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.

IN MEMORIAM

"Predictive modelling of flow-mass transport is, when unconstrained by data, often worse than no model at all": this statement from Jean-Charles Fontes (1994) is nowadays common sense. However, the complete integration of information provided by natural geochemical tracers to constraint modelling has requested from Isotope Hydrologists constant efforts and demonstrations. The recognition of Isotope Hydrology as a necessary step in site characterization for geological radioactive waste disposal results from the strong involvement of few people, and among them Jean-Charles Fontes, to promote it in that particular field of application. The Isotope Hydrology part of the methodological approach used in this study represents, to a large extend, the lessons that I learnt during ten years of scientific collaboration with Jean-Charles Fontes and for which I wished to pay him this tribute.

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