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Oklo

A review and critical evaluation of literature

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Abstract

The Oklo natural fossil fission reactors in Gabon, Equatorial Africa, have been studied as a natural analogue for spent nuclear fuel in a geological environment. For these studies, it is important to know what has happened to these reactors since they formed. This review is focussed on existing geological and geochronological information concerning the Oklo reactors and the surrounding ore. A sequence of geological and geochemical events in the Oklo area, as described in the literature, is given. The data and the studies behind this established geochronology are discussed and evaluated. Of the regional geology, special attention is given to the dating of the Francevillian sediments, and the intrusion of a dolerite dyke swarm. The processes that led to the mineralisation at Oklo, the subsequent formation of the nuclear reactors and later migration of fission products are described. Further discussion concerns the studies of the dolerite dyke swarm, since this appears to be one of the most important events related to fission product migration. A close look at the data related to this event shows that further study of the age of the dolerite dykes, and their effect on the uraninite in the Oklo reactors, is needed.

Sammanfattning

De naturliga, fossila kärnreaktorerna i Oklo, Gabon, har studerats som naturliga analoger till utbränt kärnbränsle i en geologisk miljö. Det är viktigt, för dessa studier, att veta vad reaktorerna har utsatts för sedan de bildades. Den här litteraturrapporten sammanställer geologisk och geokronologisk information om Oklo-reaktorerna och den omgivande malmen. De data och studier som beskrivningen av dessa händelseförlopp baserar sig på, diskuteras och utvärderas. Av den regionala geologin ges särskild uppmärksamhet till dateringen av de Francevilliska sedimenten och intrusionen av en svärm av diabasgångar. Processerna som ledde till uranmineraliseringen i Oklo, kärnreaktionerna, samt senare påverkan på reaktorerna beskrivs i kronologisk ordning. En fortsatt diskussion relaterad till denna senare påverkan koncentreras kring intrusionen av diabasgångarna, eftersom denna händelse hade en kraftig effekt på Oklo-reaktorerna. En noggrann utvärdering av de data som berör diabasgångarna visar, att ytterligare studier av diabasgångarnas ålder och deras effekt på Oklo-reaktorerna är nödvändiga.

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1 Introduction

In the 70's, a unique phenomenon was discovered in the Oklo uranium mine: natural uranium depleted in ^{235}U. This phenomenon was interpreted to be due to natural fission chain reactions in separate zones in the uranium deposit, and they were subsequently called the Oklo natural fossil fission reactors. The Oklo reactor zones have been subjected to extensive studies, partly due to the analogue between these zones and spent nuclear fuel repositories.

The Oklo mine is situated in the Francevillian Palaeoproterozoic sedimentary basin in Gabon, Equatorial Africa. The reactor zones have, thus, an impressively long geological history, which provides an excellent opportunity to observe the long-term behaviour of an ancient spent nuclear fuel analogue in a natural environment. The timing and cause for the migration of actinides and fission products (nuclides produced during fission reactions) are of interest for the research concerning the long-term performance assessment of a high level waste repository. The studies have been concentrated in Phase I and Phase II of the EC-financed project "Oklo-Natural Analogue", which was carried out in the 90's.

The geological history of all but one of these reactors ended recently, when the Oklo mine was exhausted. Today only the Bangombé reactor zone is left for future studies.

2 Regional geology

Gabon is situated in West Equatorial Africa, an area affected by the West Congolian orogeny. This orogeny took place at ca. 700 Ma /Cahen et al, 1984/, and left its mark on all preceding rocks. The foreland of the West Congolian belt is represented in Gabon by two Archean massifs, massif du Chaillu and massif of north Gabon, as well as the unmetamorphosed Francevillian sediments and the correlated Ogooué metamorphic rocks. Phanerozoic cover rocks occur to the west and east of this topographic rise /Cahen et al, 1984/.

2.1 The basement

The radiometric age data on the Archean massifs suggest a history of high-grade metamorphism at around 3100 Ma, and a later phase of major magmatic intrusions and metamorphism at ca. 2700 Ma. This phase is considered to be the latest phase of crustal addition in the basement, and ages lower than this (2500–2400 Ma, 2300–2200 Ma and ca. 1950 Ma) are interpreted as reflecting reworking of pre-existing crust /Caen-Vachette et al, 1988; Bonhomme et al, 1978/.

Coinciding with the third reworking at ca. 1950 Ma, magmatic activity occurred in Mayombe in the south-west and in the high grade gneisses of the Ogooué metamorphics in central Gabon /Vachette, 1964/. This occurred approximately at the same time as the Tadilian orogeny according to Cahen et al /1984/. The Ogooué metamorphics occur to the west of a large fault, related to collision and eastward thrusting of these rocks onto the Francevillian series. These events coincide with the Eburnean orogeny as recognised by Ledru /1989/.

2.2 The Francevillian

The relationship between the Francevillian and the Ogooué metamorphics is obscured by the N-S trending thrust, but it is generally believed that these are correlated and of the same age /Gauthier-Lafaye, pers. comm/. A conclusion from this is that the Francevillian sediments should be older than 1950 Ma, when pegmatites intruded the Ogooué metamorphics. However, early work indicate ages of the Francevillian around 1700–1800 Ma. These ages have been found with different isotope systems (K-Ar, Rb-Sr and U-Pb), but were later interpreted as representing late diagenesis /Weber and Bonhomme, 1975/. Age determinations conducted before 1977 have been calculated using old decay constants. Also, sedimentary whole rock samples were analysed.

That the Francevillian is older than 1950 Ma was confirmed by the age of the intercalated N'Goutou syenite in the northern part of the basin, which was dated by Rb-Sr to 2143±143 Ma /Bonhomme et al, 1982/. Also, Sm-Nd isotope systematics on authigenic clays date the early diagenesis of the sediments to 2099 ±115 Ma and 2036±79

/Bros et al, 1992/, which is similar to the Pb-Pb isochrone age of 2265 ± 150 Ma on leachates of diagenetic clays /Gauthier-Lafaye et al, 1996a/. These ages are compatible with the age of the nuclear reactions at Oklo, as discussed later. Thus, sedimentation occurred in the Francevillian basin sometime around 2100 Ma, when also some magmatic activity in the northern part of the basin produced the N'Goutou syenitic complex.

The Francevillian series is divided into five formations: FA to FE, first described by Weber /1969/. The whole series is 1000 – 4000 m thick. This overview follows later publications, e.g. /Bonhomme et al, 1982; Gauthier-Lafaye and Weber, 1989/. The lowermost formation, FA, is 100 – 1000 m thick, and starts with a red, radioactive conglomerate which rests unconformably on the Archaean basement. The rest of FA consists of sandstone and conglomerate typical of fluvial and deltaic environments, and it is this formation that contain all uranium deposits of the basin. The FB formation, resting slightly discordantly over the FA, consists mainly of black shales. FB was deposited during a period of downfaulting, causing this formation to be over 1000 m in the central parts of the basin. Formations FC-FE have more diverse lithology, with shale, chert, dolomite, and volcanic rocks, which become more common at the top of the series. The series is overlain by Mesozoic continental sediments of the Congolian basin.

2.3 The dyke swarm

The Francevillian is cut through by numerous dolerite dykes, all belonging to the same dyke swarm. The majority of the dykes trend E-W, but a few major ones in the central part of the swarm trend in N-S. One of these E-W trending dykes cuts through the Oklo deposit. Attempts to date the dykes have been made by direct K-Ar, /Bonhomme et al, 1978; Weber and Bonhomme, 1975/, Sm-Nd (mineral isochron) and U-Pb (zircon) dating /Sère, 1996/, as well as indirectly by dating uraninite and galena in the ore deposit /Holliger, 1991; Holliger, 1992; Gauthier-Lafaye et al, 1996b/.

The K-Ar measurements yield a range of ages from ca. 980 Ma (on feldspathic fractions as well as whole rock) down to ca 500 Ma (whole rock), a range probably caused by Ar loss. If Ar loss is the only cause for the spread, the oldest age should be the most trustworthy. However, there is also the problem with possible inherited Ar, i.e., Ar that was incorporated into the mineral during crystallisation. Inherited Ar would cause the age to be too high, but this effect is most severe in minerals with low concentration of K. K is a major element in alkali feldspar, but not in pure plagioclase. However, K may occur to some extent (5–10% orthoclase) in the plagioclase solid solution. This is probably the reason why the authors did not consider this effect and concluded that 980 Ma was the age of the dolerite dyke /Bonhomme et al, 1978/. Looking at a larger scale, in stable areas surrounding the Congo basin, 950±50 Ma is recognised as the time for an important major tectono-thermal event, recorded in the Late Precambrian sequences in the stable zones of Equatorial Africa /Cahen, 1982/. It is seen, for example, in an amygdaloidal lava dated at 948±20 Ma.

The Sm-Nd mineral isochron, yielding an age of 783±55 Ma (the age given in the text of Sère /1996/ is 755±83 Ma, but this is a misprint) is a line between two points since

only plagioclase and whole rock data could be used, and the U-Pb zircon age, 746±16, is the intercept between a two-point discordia and the concordia. Older inherited zircon crystals (two points) are discordant, but when the lower intercept is forced through 746 Ma, the upper intercept yields an age relatively close to the age of the basement /Sère, 1996/.

The dyke through Oklo could be dated indirectly by assuming that major Pb loss occurred from the uraninite at the time of the dyke intrusion, and that the radiogenic Pb crystallised as galena. By using SIMS, in situ measurements of Pb and U isotopes on uraninite and galena were made, which yielded a range of results. Galena model ages spread from ca. 1500 Ma to ca. 400 Ma, with a peak at ca 600–800 Ma /Gauthier-Lafaye et al, 1996b/, in accordance with discordia ages of uraninite (722 Ma and 850 Ma). See Appendix 1 for further discussion.

These approximations help to constrain the time period in which the dyke at Oklo most likely intruded. However, the reason why the ages spread in a time range of over 200 Ma is still unclear. The dykes are generally altered, seen as chloritisation in the microscope /Sère, 1996/. This alteration and chloritisation was also noticed by Weber and Bonhomme /1975/, who observed loss of water during heating of their dolerite samples. Since a pristine dolerite should not contain any significant amount of water, they used the loss of water during heating as a measure of alteration. Microscope studies reveal that the chlorite crystallised later than plagioclase and clinopyroxene, suggesting that the alteration occurred sometime after the final crystallisation of the dyke. At what time and why this happened is not known, but the youngest K-Ar ages suggest that this occurred around 530 Ma.

As a comparison, Mid-Ocean Ridge Basalts (MORB) are often altered, showing chloritisation and albitisation of the plagioclase. This alteration is called spilitisation, and has been argued to be caused by the assimilation of seawater into the crystallising magma. The Francevillian dykes crystallised in a continental setting, thus, assimilation of seawater did not occur. However, it has also been shown that similar type of alteration could happen through hydrothermal alteration, at non-metamorphic conditions, after the crystallisation of the basalt, or at low-grade metamorphic conditions. However, the water/rock ratio is characteristically very high and thus the spilitisation process should be of hydrothermal character rather than metamorphic /Hall, 1987/.

2.4 Phanerozoic events

After the intrusion of the dykes, there was an apparent quiescence for at least ca. 500 Ma. Around 260 Ma ago, uplift (cooling below 130°C) of the basement started, in relation to the opening of the Atlantic ocean. This was recorded by fission tracks in apatite, extracted from basement rocks close to the edge of the Franceville basin. Later dates around 180 Ma ago were recorded in the basement apatites in more central parts of the basin /Sère, 1996/.

Recent weathering, deep and severe due to the tropical climate, has also affected the basin, and the uranium situated in it. Torbernite, a hydrated phosphate of copper and hexavalent uranium, was found to be of the age 76 500 ± 6 800 years /Bros et al, 1997/, with the uranium-series disequilibrium method.

3 Geology of Oklo

The Oklo ore is found in the uppermost unit of the FA, a 7–10 m thick coarsening–upwards deltaic layer that is in contact with the FB black shales. The ore is both structurally controlled and related to redox reactions. The uraninite is closely associated to organic matter. Folds and faults, formed at the time of burial probably due to tectonic events in the Ogooué mobile belt /Gauthier-Lafaye et al, 1996b/, form structures similar to oil traps in which the ore is concentrated. The concentration of the ore varies from 0.1% to 10% (not including the reactor zones). The reactor zones occur as ca. 10 m lensoid bodies of up to 80% uraninite in a clay gangue within the highly concentrated ore.

3.1 Age of the mineralisation and the reactors

Efforts to date the mineralisation yieded ages of 1810±30 /Hagemann et al, 1974/ and 1780 Ma /Lancelot et al, 1975/, which caused some problems recognised by Devillers et al /1975/ who reported an average age of the nuclear reactions of 2000 Ma. Their U-Pb data also suggested an age of the ore of ca. 1800 Ma. This would mean that the nuclear reactions occurred before the deposition of the ore, which seemed unlikely. The method for determining the age of the nuclear reactions /Ruffenach,1979/ appeared indisputable, so Devillers et al /1975/ tried another interpretation of their U-Pb data which yielded a mineralisation age of 2050 Ma. This age, 2050±30 Ma, was later confirmed by Gancarz /1978/, when it was shown that volume diffusion could be the reason for the lower intercept at 375 Ma. This conclusion was challenged by Janeczek and Ewing /1995/, who argued that the major processes causing Pb loss of uraninite in the Oklo region were thermally activated and episodic. In any case, the 2050 Ma age of the mineralisation is in agreement with the more precise age of the nuclear reactions, 1980±60 Ma /Ruffenach, 1978/, and another average age calculation giving 1930 Ma /Devillers and Ménès, 1978/. SIMS studies on single uraninite grains within reactor 10 have yielded ages of 2018±50 and 1968±50 Ma /Gauthier-Lafaye et al, 1996b; Holliger, 1991; Holliger, 1992/, which corresponds to the age of the nuclear reactions.

The mineralisation is therefore said to have occurred at 2050 Ma, just after sedimentation, during early diagenesis. The nuclear chain reactions started around 1980 Ma in highly concentrated parts of the ore; it should be noted that this age is, within error, the same as the age of the mineralisation.

3.2 Mineralisation process

Both bitumen and fluid inclusions have been found in quartz overgrowths, which indicates that quartz diagenesis was contemporanous with oil migration in the basin. Homogenisation temperatures of these fluid inclusions range between 80°C–

400°C, with a prominent peak around 140–180°C /Gauthier-Lafaye and Weber, 1989/. The wide range of temperatures might be due to modification of the fluid inclusions by later events, and the peak at 140–180°C is interpreted as the temperature of the basin during maximum burial, corresponding to ca 4000 m of sediments. Burial temperature had earlier been determined to ca 240°C /Openshaw, 1978/, but this can be reinterpreted as ca 180°C /Gauthier-Lafaye and Weber, 1989/. 180°C as a maximum burial temperature is also supported by later work, giving temperatures in a range of 160°–190°C /Savary and Pagel, 1995/. Later fluid inclusions, related to calcite and barite, recorded temperatures of ca 140°C /Gauthier-Lafaye and Weber, 1989/. These studies show that the Francevillian sediments reached the oil window (when kerogen decays to liquid hydrocarbons) before maximum burial /Gauthier-Lafaye and Weber, 1989/.

The deposit has commonly low grade ore (0.1–1% uraninite) with patches of high grade ore (1–10% uraninite). The uranium mineralisation is clearly related to organic matter, seen best in the low grade ore. In high grade ore, the uranium is often related to red-coloured hematite-rich pockets, rimmed with white clay, in the sandstone, coloured black by organic matter /Gauthier-Lafaye et al, 1978/.

Devillers and Ménès /1978/ used U, Th and Pb concentrations and isotopic compositions to study of the chronology of events that produced the highly concentrated ore, and came to the conclusion that an early placer deposit was in place ca. 250 Ma before the “superconcentrations” formed. It is clear that the Oklo ore is not a placer deposit (see below), and their conclusion is misleading. However, they acknowledge the complexity of Pb, due to at least one ancient and one relatively recent event of Pb mobility.

The generally accepted idea of the mineralisation events was first formulated by Gauthier-Lafaye et al /1978/, and is based on the observation of the two different types of ore together with the relation of bitumen and diagenetic fluids. During maximum burial, oil migrated from the black shales into the upper part of the FA sandstone. At the same time, oxygenated fluids migrated in the sandstone and radioactive conglomerate in the lower part of FA. These fluids caused the precipitation of hematite which give the red conglomerate its colour, and mobilised the uranium that existed in the radioactive conglomerate. After maximum burial, a tectonic event lifted the basin ca 1000 m (fluid inclusions of ca. 140°C). During this stage, hydrofracturing occurred in the sandstone, which was capped by impermeable FB shale. In this secondary porosity, occurring in patches which were typically good oil traps, reducing carbon rich fluids and oxygenated uranium rich fluids met, and uraninite crystallised to form the ore. The different appearance of the high-and low grade ore might suggest that the high grade ore was formed by a secondary oxidation-reduction process, in which the uranium in the low grade ore was mobilised /Gauthier-Lafaye and Weber, 1989/. In later descriptions of the mineralisation events /Gauthier-Lafaye et al, 1996b/, the idea of a secondary oxidation-reduction process is left out, and the high grade ore is generally described as occurring where the hydrofracturing of the sandstone is most intense. These fractures, occurring also in the reactor zones, have been re-oriented. Thus, the latest tectonic event, which gave the deposit its present shape, occurred after mineralisation.

3.3 The reactors

The most obvious characteristic of a fossil fission reactor is the ^{235}U-depletion of uranium. This phenomenon, together with the occurrence of "unnatural", fissiogenic isotope ratios of elements like Zr, Nb, Mo, Ru, REE etc. (collectively called fission products), is the clear proof that fission occurred in the Oklo deposit.

The lens-shaped reactor zones are always found in the high grade ore and are generally around 10 m in diametre. A typical reactor zone has a core of up to 80% uraninite, often euhedral, in a clay gangue consisting mainly of chlorite and illite. The thickness of the core varies between 5 and 20 cm, and it has uranium depleted in ^{235}U. The clay mineralogy in the core bears witness to the corrosion of quartz, and there are structures related to collapse of the host rock /Gauthier-Lafaye et al, 1989/.

The shape of the reactor zones is a result from the nuclear fission reactions due to the heat produced during operation. An estimate of the temperatures achieved can be made by studying the isotopic composition of Lu, which is not a fission product, but is produced by neutron capture. This was done by Holliger et al /1978/ and Holliger and Devillers /1981/ who reported 280±50°C. This is a surprisingly low number; Openshaw /1978/ suggested an operating temperature between 450°C and 600°C, estimated from the density of the fluids in inclusions and the pressure stresses. Later studies on fluid inclusions in quartz in or near the reactor zones indicate temperatures above 200°C (up to 440°C) /Gauthier-Lafaye, 1986/ and ca. 400°C /Savary and Pagel, 1995/. The low temperature estimated from Lu was later said to be due to Lu migration, as well as the uncertainty of the neutron capture cross section of ^{175}Lu /Gauthier-Lafaye et al, 1996b/.

The temperature gradient recorded by the fluids near RZ 10 is 40°C/m, which is interpreted to be a result of conductive cooling /Savary and Pagel, 1995/. However, the clay mantle surrounding the reactor core is said to be formed during the circulation of hydrothermal fluids during the cooling of the reactors /Gauthier-Lafaye et al, 1989; Gauthier-Lafaye et al, 1996b/, and with fluids present, the cooling is then regarded as convective. Fluid inclusions in calcite have recorded temperatures between 100–180°C /Gauthier-Lafaye, 1986/, and are considered to have formed during a late stage of cooling. Late fluids of lower temperature are also said to have been captured in apatite /Savary and Pagel, 1995/.

The structure and petrological composition of the reactors are thus largely results of convecting hydrothermal fluids during the nuclear reactions and subsequent cooling. The hot water dissolved silica, which caused volume decrease and collapse. During cooling, crystallisation of phyllosilicates, calcite and apatite occurred.

There has been some discussion about the effect of radiolysis during the fission. Observations of minium (Pb_3O_4) in RZ10 indicate very oxidising environment. Some fluid inclusions in RZ10 contain both H_2 and O_2, seen as an evidence for radiolysis during the nuclear fission. The radiolysis caused some variation in redox conditions, in areas without organic matter. The prescence of organic matter controlled the redox conditions in the reactor zones, and kept the environment reducing in spite of radiolysis /Savary and Pagel, 1995; Savary and Pagel, 1997/.

The role of organic matter has also been investigated by Nagy et al /1991/, who believe that the organic matter was solidified syngenetic kerogen at the time of the onset of the reactions. This solidified kerogen became liquified during the reactions due to the

circulations of hydrothermal aqueous fluids. These fluids would have been oxidising and caused mobility of uranium. As the temperature rose in the reactors, the bitumen became more solid and encapsulated uranium, which recrystallised as uraninite. This uraninite incorporated fission products at the time of crystallisation. However, in the prescence of organic matter the fluids should have been reducing /Savary and Pagel, 1997/, and oxidation of U and the mobility of organic matter during reactions is still a matter of discussion.

The duration of reactions can be calculated with the help of produced fission products and actinides together with nuclear parameters of the reactors. The two methods of Hagemann et al, /1975/ involved Pu and Tc-Ru, respectively. Both of these methods give time periods of ca 600 000 years (for RZ2). There are, however, problems involving the migration of these elements in relation to uranium, as well as assumptions of constant flux and large uncertainties. Similar numbers are given by Ruffenach et al /1976/, who calculate the duration to ca 800 000 Ma from the percentage of Pu fissions (RZ2). Loss et al /1988/ recognised that there had been complex irradiation and that ^{99}Tc had been mobile, and therefore calculated the duration using Pd fission yields. This results in more precise determination of the percentage of Pu fissions, and the samples analysed yield numbers ranging between 64 000 and 556 000 years, with an average of 220 000 years (reactor zone 9).

From these calculations, the duration of the reactions is said to be 0.1 – 1 Ma. However, it should be noted that the different zones might have operated for different lengths of time, due to different physical parameters.

4 Migration of fission products

The actinides and many of the fission products (Ru, Rh, Pd, Te, Y, Nb, Tc, Bi, REEs, and Zr) produced in nuclear reactions have similar ionic radii as uranium /Gauthier-Lafaye et al, 1996b/. Thus these could be expected to be stable in the uraninite structure, and the migration of these should, therefore, be closely related to the stability of the uraninite structure. To test this prediction, studies of mineralogy and isotope chemistry of uraninite are used to investigate the degree of retention in the uraninite matrix. The percentage of retained fission products has been studied by several authors /Hagemann et al, 1975; Ruffenach, 1978; Loss et al, 1988; Curtis et al, 1989; Hidaka, 1993b; Holliger, 1991; Hidaka, 1994b/, who report varying degrees of retention and migration of the elements mentioned above.

In some cases, almost complete retention has been shown. Other studies show that small scale migrations of actinides, especially Pu, have occurred /Bros et al, 1996; Bros et al, 1993/. Ru, Rh, Pd, Te and Mo have been found in microinclusions in reactor zones 10, 13 and Bangombé. These microinclusions are generally arsenides and sulfides, but have, somewhat misleading, been called “metallic aggregates”. The reason is that the same elements, together with Tc, are found as metallic aggregates in spent nuclear fuel. At Oklo and Bangombé, these inclusions can be up to 0.1 mm in size. The existence of these aggregates indicates that the migration of the platinum metals occurred only on a small scale. The Ru also bears signature of ^{99}Tc, which decayed into ^{99}Ru, from which the ancient migration of ^{99}Tc can be inferred /Hidaka et al, 1993b/.

Other elements are volatile, or do not have ionic radii compatible with the uraninite structure. These are predicted to migrate out of the uraninite matrix. The migration of these elements has been studied by Loss et al, /1984, 1988/, Curtis et al /1989/ and Hidaka et al /1992, 1993a, 1994a/. Results show that Xe, Kr, I, Cd, Br, Rb, Cs, Sr, Ba, Te, Pb, Mo, Sn, and Ag have partly or completely migrated out of the uraninite structure /Gauthier-Lafaye et al, 1996b/. The volatile elements like Cs, Rb, Ba, and Sr have been completely removed from the reactors. Other elements, when migrated, have either been captured by clays, different types of apatite, heavy minerals and, for Zr, oxides.

A summary of the behaviour of fission products and actinides can be found in Gauthier-Lafaye et al /1996b/.

5 Summary and discussion

The age of final stabilisation of the basement, ca. 2.7 Ga, is based on radiometric measurements made in the 60's and 70's. Some of the reported ages should therefore be recalculated with modern constants, however, the result would not change the general conclusion. The age of the basement is, in this context, the same as the last major crustal growth and metamorphism of the precursors, which formed sometime around or before 3.1 Ga. Since the FA formation rests unconformably on these rocks, 2.7 Ga is the maximum age of the FA. The basement has been subject to some later episodes of rejuvenation, of which the latest occurred around 1950 Ma. During this time, several pegmatites intruded in the Ogooué metamorphics, indicating the age of a major tectonic phase occurring in Central Gabon.

By 1950 Ma, the Francevillian sediments (or at least FA and parts of FB) were already deposited but, surprisingly, their isotopic systems do not seem to have recorded the tectonic event at 1950 Ma, which was large enough to reset some isotopic systems in parts of the crystalline basement. The minimum age of FA can determined by the age of the N'Goutou syenitic complex, which crystallised sometime in the time range 2143±143 Ma. The Rb-Sr isotopic system which was used for this age determination is rather easily disturbed, depending on local situation and sampling. One Rb-Sr isochron of the N'Goutou syenite (see Table A2-1, Appendix 2) does in fact yield an age of ca 1800 Ma /Weber and Bonhomme, 1975/, indicating that the Rb-Sr system was disturbed in parts of the complex. The nature of the N'Goutou complex is debated in the literature, but is confirmed as a stratiform complex, intercalated with FB /Gauthier-Lafaye, pers. comm/.

This age, around 2100 Ma, is difficult to extract from whole rock analyses of the sediments. Sediments are known to be difficult to date, due to heterogeneous initial isotopic compositions and closure temperatures. The efforts to date the sediments, both with whole rock and with mineral separates, yielded ages between 1700 and 1800 Ma (see Appendix 2). This could be an indication that an event around that time reset the systems or, alternatively, the systems did not reach their closure temperature until ca 1800 Ma. More sophisticated analyses of diagenetic clay minerals yield ages of early diagenesis around 2100 Ma. This age is similar to the age of the uranium mineralisation outside of the reactor zones, determined to 2050 Ma. The reactor operation started at ca. 1980 Ma, as calculated from nuclear parameters, which themselves have been calculated from fission products found in a few well preserved uraninite grains. In a similar way, the duration of the nuclear reactions has been calculated to 1 to 0.1 Ma.

The ages in the sequence of events beginning with sedimentation of FA and ending with the nuclear reactions, are generally the same, within error. However, logic tells us that the nuclear reaction should not have started before the uranium was in place in the upper part of FA. We also know that the Oklo uranium deposit is not a placer deposit, and therefore formed at some point after the sediment deposition. The detailed process of mineralisation can still be discussed, but is clearly related to both geological structures and redox processes. As a conclusion, the average ages cited are in correct logical order within the time period between 2300 Ma and 1950 Ma.

During the nuclear reactions there were fluids circulating; the temperature was somewhere between 200°C and 400°C. The surrounding sandstone was transformed into phyllosilicates, and while the host rock collapsed, the reactors kept themselves in operation. The redox conditions during the reactions were mainly controlled by the existence of organic matter which kept the fluids reducing, although oxidising products of radiolysis were produced.

What is intriguing is the number of attempts to date the Francevillian sediments that yielded the 1800–1700 Ma ages. In Table A2-1, Appendix 2, some of these ages are recalculated with modern constants. What these ages represent is still unclear; some authors like to call it the final cooling of the reactors, or regional cooling, others call it a very late diagenesis (often correlated with an observed late calcite precipitation). The ages are probably, in some way, related to the last tectonic event in the Ogooué mobile belt, since the N'Djolé micaschists yield the same Rb-Sr isochron age as FB pelites (see Table A2-1, Appendix 2). When the age of FA was still under debate, the correlation with Francevillian sediments and the Ogooué metamorphics was pondered on. An old common Pb age of the Mitendi series, which is believed to be intermediate between the two, yielded an age of 1790 Ma. Unfortunately, the reference given by Weber and Bonhomme /1975/ is by all appearances wrong and the data have not yet been found in the literature by the present author. This could perhaps be another isotopic record of an event that appears to have disturbed the Rb-Sr and K-Ar isotopic systems in the Francevillian basin.

The attempts to date the intrusion of the dyke swarm have been met with several difficulties. K-Ar ages spread almost 500 Ma; the reason is most likely Ar loss, since the dykes show clear signs of alteration. Therefore, the oldest K-Ar age (980 Ma) may give the best estimate of the age of the dyke swarm. Other efforts to date the dyke that cuts through Oklo give spreading ages with maximum between 800 Ma and 600 Ma (see discussion in Appendix 1). No reliable direct dating of the dyke through Oklo is found in the literature, and any age between ca. 1000 Ma and 700 Ma has been used as "the age of the dolerite dyke". These difficulties could arise from a prolonged period of open isotopic systems and elemental migration, due to the regional heating of the Francevillian basin. The elevated temperature would have been caused by crustal thinning and the intrusion of the dyke swarm, associated with regional extension. This period in the Late Proterozoic is a time of tectonic activity through all of the African continent, and what the Franceville basin experienced might be called a pre-Pan-African extension.

Another explanation for the wide range of ages could be later opening of the systems, which was not severe enough to cause complete resetting (see Appendix 2). Perhaps there was one major event when the dykes intruded, around 980 Ma, and then some 250 Ma years later, another opening of the systems due to Pan-African events? These Late Proterozoic events are, however, obscured by the fact that recent uplift and weathering also have affected the isotope systems, the U-Pb system in particular, causing an important loss of radiogenic daughters. This more recent period could also be of some complexity, since uplift (probably related to the opening of the Atlantic ocean) started already at ca. 260 Ma in the outer part of the Franceville basin. That recent weathering has affected the U-Pb system has been shown by the existence of torbernite ($Cu(UO_2)_2(PO_4)_2 \cdot 8–12\ H_2O$) with an age of ca 80 000 years.

The studies of fission products demonstrate the importance of relative migration of the fission products used to estimate fission yield. The calculations of nuclear parameters might involve quite large uncertainties due to uncertainties in capture and fission cross sections. Even though it has been shown that migration has occurred in many cases, there are some cases of very little migration, and these have been used as basis for the calculation of the neutronics, and from there, the age and duration of the reactions. Efforts to quantify migration were made, and elements have been grouped according to how well retained they are in the reactor core and immediate surrounding. Interestingly, some uraninite grains (in reactor 13) have suffered a severe Pb loss, but the fission products seem to be very well retained. This shows that observed Pb mobility should not be confused with mobility of fission products.

Some elements, such as the platinoids, are found as As-S aggregates in the core of the reactors. Even though Ru isotopic ratios show that Tc-rich aggregates formed sometime during the lifetime of Tc, no one has given a reliable answer to the question of when the aggregates we see now were formed. The compositions of the aggregates are similar to the ones found in artificial fuel. At Oklo these aggregates can be up to 100 μm in diameter, while in artificial fuel they are usually only about 1 μm /Janeczek, 1999/. This could perhaps be due another phase of mobility during some event later than the reactions; here it could be wise to remember the association of the aggregates with galena. Pb isotopes measured by SIMS yield $^{207}Pb/^{206}Pb$ similar to galena; however, after correction for a very low $^{235}U/^{238}U$ (0.382%), this value becomes strikingly different, indicating that the aggregates formed just after reactor operation. As a comparison, the correction used when calculated the model age of galena crystals from the same reactor, involves $^{235}U/^{238}U$ of 0.71–0.465%. The validity of this large correction of the As-S aggregate needs further evaluation.

6 References

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Appendix 1

Review of the U-Pb system data obtained during the EC-project "Oklo – Natural Analogue Phase I"

Chemical and isotopic analyses of the Oklo reactor zones and of surrounding environment have shown that substantial amounts of the elements produced during nuclear fission have migrated out of the reactor cores. The observed migration is different for different elements and for different reactor zones, and luckily there are a few exceptions where fission products have been retained enough to allow calculations of nuclear parameters and the age of the fission reactions. Much of this information, obtained during the "Oklo-Natural Analogue Phase I" project, is summarised in a paper by Gauthier-Lafaye et al /1996/. It is in many cases based on data that was presented earlier in various reports e.g., Holliger /1991/ and Holliger and Gauthier-Lafaye /1996/.

In the conclusions of Gauthier-Lafaye et al /1996/, it is stated that there have been two main periods of radionuclide migration at Oklo. The first occurred around the time of the criticality and subsequent cooling (ca. 1950 Ma), shown by the isotopic composition of U, Ru, Zr, Ba and Cs, found in clays of the reactor, "metallic aggregates", phosphates and zircon. Other signs that element migration should have occurred during this time are the elevated temperatures found by studies of fluid inclusions in minerals petrographically determined to have crystallised during the events in question. For example, quartz in fractures and as overgrowths in the sandstone surrounding the reactors contain fluid inclusions that indicate temperatures between 400 and 500°C. Also the reactor clays contain the 2 M polytype of illite, which is said to indicate high temperature. Thus, hot fluids circulated in and around the reactors and the shape and petrological composition of the reactors is a result of the nuclear reactions.

The second period of element migration, stated by Gauthier-Lafaye et al /1996/, occurred during the time of the dolerite dyke intrusions (1000–750 Ma), a conclusion drawn from isotopic studies of radiogenic Pb in uraninite and galena in the ore. Special attention is given to RZ 13, which is located very close to the dolerite dyke that cuts through the Oklo deposit. There is an indication of fluids of ca. 200°C during the Upper Proterozoic /see Savary and Pagel, 1995/, but it is not mentioned by Gauthier-Lafaye et al /1996/. This event in the Upper Proterozoic caused massive migration of radiogenic Pb, possibly together with elements like the platinum metals, Mo and Te. These elements are all found in the typical "metallic aggregate", which as described actually is an aggregate of As-rich Ru-Rh-Pd-Te sulfides. It is found in close association with galena and a (U, Zr)-silicate phase /Gauthier-Lafaye et al, 1996/.

The age of the reactors, 1950±40 Ma, is obtained by using the sample that shows most geochemical stability (SC36, RZ2) considering the elements used for the calculations (U, Th, fissiogenic REE). However, it appears as if this sample is rather unique, and the rest of the reactors show discrepancies between the theoretical number of fissions for an age of 1950 Ma, and the values calculated from analytical data /Table 1 in Gauthier-Lafaye et al, 1996/. This is assumed to be due to migration of fissiogenic REE in

relation to U. It is obvious, however, that the different reactor zones had different nuclear and physical parameters. The impact of this on the age of the reactions for reactors other than RZ2, is not considered further, but a general age for all reactors is set to 1950 Ma.

It is interesting to note that the calculations of neutronic parameters, and the correlation between calculated and theoretical number of fissions, is quite similar between RZ10 and RZ13. RZ10 is taken as an example of a reactor zone which has suffered relatively minor disturbance after the fission reactions, and is also the reactor that has retained most radiogenic Pb. RZ13, on the other hand, chosen as an example of the effect of the dolerite dykes, has suffered a substantial Pb loss. Apparently, radiogenic Pb shows very different behaviour from fission products, and cannot be used as a tracer of general element migration out of the uraninite.

Ion microprobe analyses have been used, by Gauthier-Lafaye et al /1996/, to date the uraninite crystals in and around RZ10. All uraninite analysed has U depleted in ^{235}U, even though most samples come from silicified sandstone ca 1 m below the reactor core. Two uraninite crystals, embedded in organic matter from the reactor core, yield a discordia line with an upper intercept age of 1968±50 Ma /borehole SF42, Figure16 in Gauthier-Lafaye et al, 1996/. Another discordia line is regressed through the data from samples beneath the reactor core, and it yields an upper intercept age of 2018±30 /bore hole SF29, Figure 16, Gauthier-Lafaye et al, 1996/. These ages are the same, within error, to that obtained on normal ore and of the fission reactions. The data from SF29 are also found in Holliger /1991/, who reports an upper intercept age of 2018 Ma, based on seven points. However, eleven points from SF29 are plotted in Figure 13, Holliger /1991/ as well as in Figure 16, Gauthier-Lafaye et al /1996/. Thus, in both papers, four points are excluded from the regression, a fact which is mentioned by Holliger /1991/, but not by Gauthier-Lafaye et al /1996/.

The data from SF29 beneath the core of RZ10, is replotted by the present author in Figure A1-1, this text. In diagram A, Figure A1-1, only the data (seven points) used by Holliger /1991/ were used, and in diagram B, Figure A1-1, all data (eleven points) reported in Gauthier-Lafaye et al /1996/ were used. The program used is Isoplot/Ex 2.00 /Ludwig, 1999/, which also calculates ages based on the discordia-concordia intecepts. The errors on individual data points were taken into account. The 2 σ errors used for the data are 0.1% for $^{235}U/^{238}U$ and 0.3% for Pb isotopic ratios, according to Table 3 in Gauthier-Lafaye /1996/. In Gauthier-Lafaye et al /1996/, the two discordias mentioned (SF42 and SF29) are used as an argument to show that no important alteration event have caused dissolution and recrystallisation of the uranium dioxides. However, in Figure A1-1 (this text), the discordia line in diagram A has a high MSWD (126), an indication that there is a real (non-analytical) scatter or the analytical errors are larger than what is reported. The upper intercept age reached is 2009±91 Ma (2 σ error, not forced through the origin). In diagram B (Figure A1-1), all data points are taken into account and the MSWD is even higher (1624). The upper intercept yields an age of 2083±110 Ma, while the lower intercept differs significantly from 0 (278±120 Ma). These high values of the MSWD are symptoms of a disturbed isotopic system.

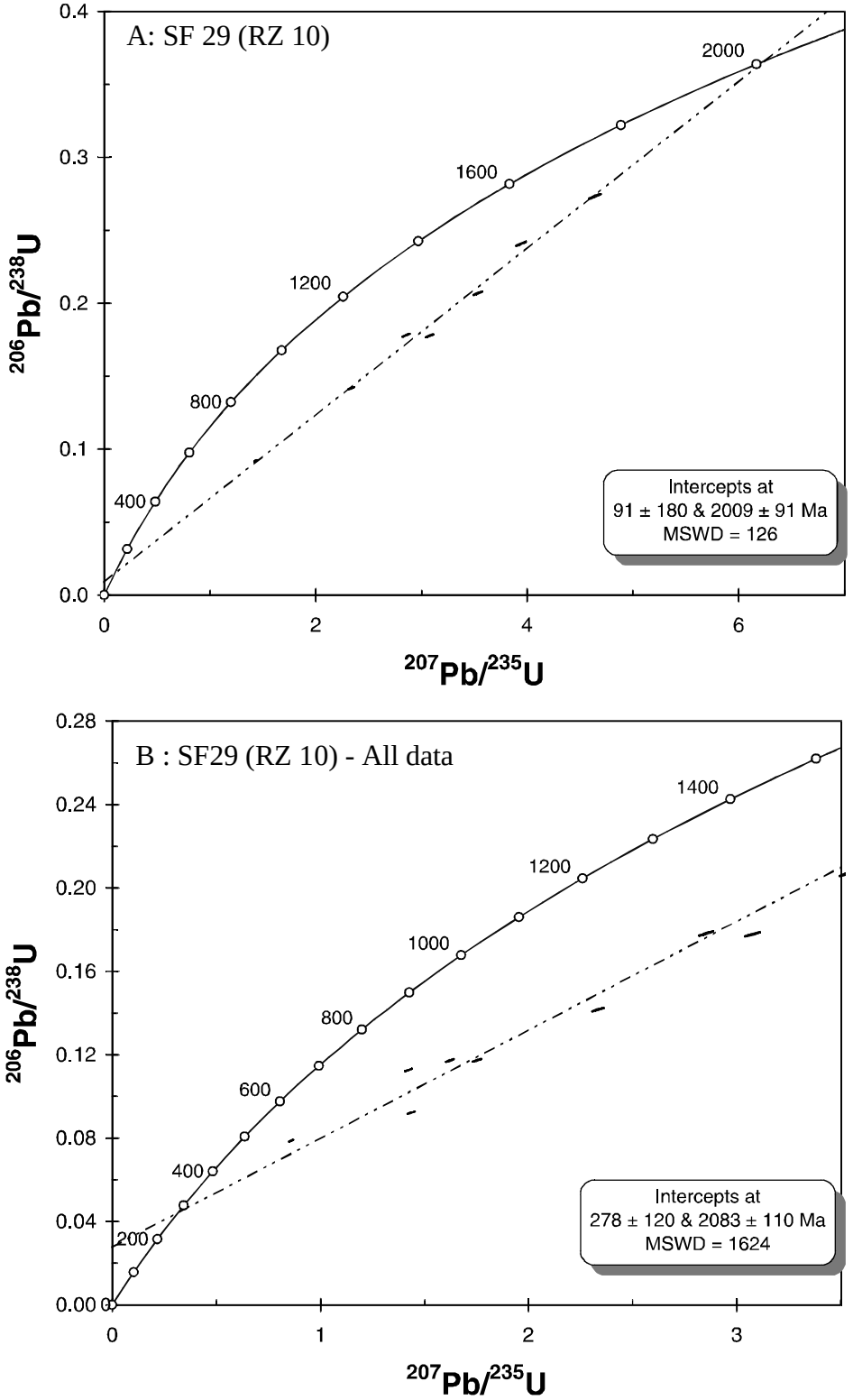

***Figure A1-1**. Replotted data from RZ10, SF29. Isoplot/Ex 2.00 /Ludwig, 1999/ was used for plotting and calculations. Errors are 2σ. A: Data from Table 4 /Gauthier-Lafaye et al 1996/, excluding four points (88.70/1, 89.70/1, 90.66/1, 90.85/1/. This is done according to Figure13 in Holliger /1991/, who mentions that only seven points have been used to calculate the age of 2018±30 Ma. B: All data (eleven points, three out of view) from SF29 in Table 4 in Gauthier-Lafaye et al /1996/.*

From Figure A1-1 (this text), it appears as if the U-Pb systems of some, if not all, of the analysed uraninite crystals in SF29 have been disturbed by some event other than recent lead loss. It thus seems like the conclusion that no important alteration event has occurred in reactor zone 10 is based on selected crystals which have been well preserved, perhaps due to a protective environment (silicified sandstone, organic matter). The reason why the excluded data points would not have been as well preserved is a matter of speculation. There is no explanation of the exclusion of the points in Gauthier-Lafaye et al /1996/ and the explanation in Holliger /1991/, p. 14, is limited to that statement that the age is obtained from "les sept échantillons de la partie gréseuse la plus avale de la zone 10". The U-Pb system of the uraninite crystals found the reactor clay in RZ 10 has been severely disturbed, as can be seen in Figure 12 in Holliger /1991/. 16 data points are reported for this section of bore hole SF29 /Table 4 in Holliger, 1991/ and the discordia regressed through these points yield an upper intercept of 1350 Ma (the age is given without errors). This data, from the reactor clay in bore hole SF29, is not mentioned in Gauthier-Lafaye et al /1996/. It should be noted that the data from RZ10 (SF42 and SF29) is also found in Holliger and Gauthier-Lafaye /1996/, and the data from SF42 is also found in Holliger /1992/.

Gauthier-Lafaye et al /1996/ state, in the introduction of the paper, that the most probable age of the dolerite dyke intrusion at Oklo is ca. 980 Ma. This age is based on two K-Ar dates obtained by Bonhomme et al /1978/ /the authors refer to Bonhomme et al, 1982/ on a dolerite dyke close to Mikouloungou, ca 45 km SE of Oklo. Ion microprobe U-Pb data from RZ13 and ES24 (normal ore) show that uraninite suffered substantial Pb loss around that time: two discordias are plotted /Figure 17 in Gauthier-Lafaye et al, 1996/ yielding upper intercept ages of 850±50 (RZ13) and 722±30 Ma (ES24). Data from RZ9 are also plotted but no discordia could be obtained. Here it is noteworthy that more data have been obtained from RZ13 (SD37) than are reported in Table 4, Gauthier-Lafaye et al /1996/. All U-Pb data obtained on uraninite crystals from SD37 can be found in Table 5, Holliger and Gauthier-Lafaye /1996/. Comparing the two data tables, one can conclude that in Table 4 in Gauthier-Lafaye et al /1996/, 3 samples are excluded (53, 63 and 69.5) and one (70) is reported with a different value of $^{206}Pb/^{238}U$ than in Holliger and Gauthier-Lafaye /1996/. The upper intercept age of the discordia, 850±50 Ma, obtained on the data from SD37, is given both in Gauthier-Lafaye et al /1996/ and Holliger and Gauthier-Lafaye /1996/. However, the excluded data points are not discussed in either of the papers. The data from RZ13 (SD37) is replotted here, in Figure A1-2 (this text), and it becomes apparent that the data points do not form a discordia. An age obtained from these data is, therefore, not meaningful.

The data for ES24 (Extension Sud, normal ore) give a somewhat more reliable discordia. It should be noted that these data (ES24) were also published in *Nature* 1991 /Table 2, Nagy et al, 1991/, where the samples are called 8§ and 9§. The interpretation of the major Pb loss (recrystallisation?) is that it happened during the time of the dolerite dyke intrusions. It was stated by Gauthier-Lafaye et al /1996/ that the most probable age for this event is ca. 980 Ma. Discarding the date of 850±50 Ma obtained on RZ13, we are left with the 722 Ma "age" of the normal ore in Extension Sud, and the fact that a substantial Pb loss has occurred in other zones, probably during the same event. This is backed up by radiogenic Pb found in galena /Table 3 in Gauthier-Lafaye et al, 1996 and Table 7 in Holliger and Gauthier-Lafaye, 1996/, which indicates major galena crystallisation sometime between 800 and 600 Ma – this is, however, only a part of the picture.

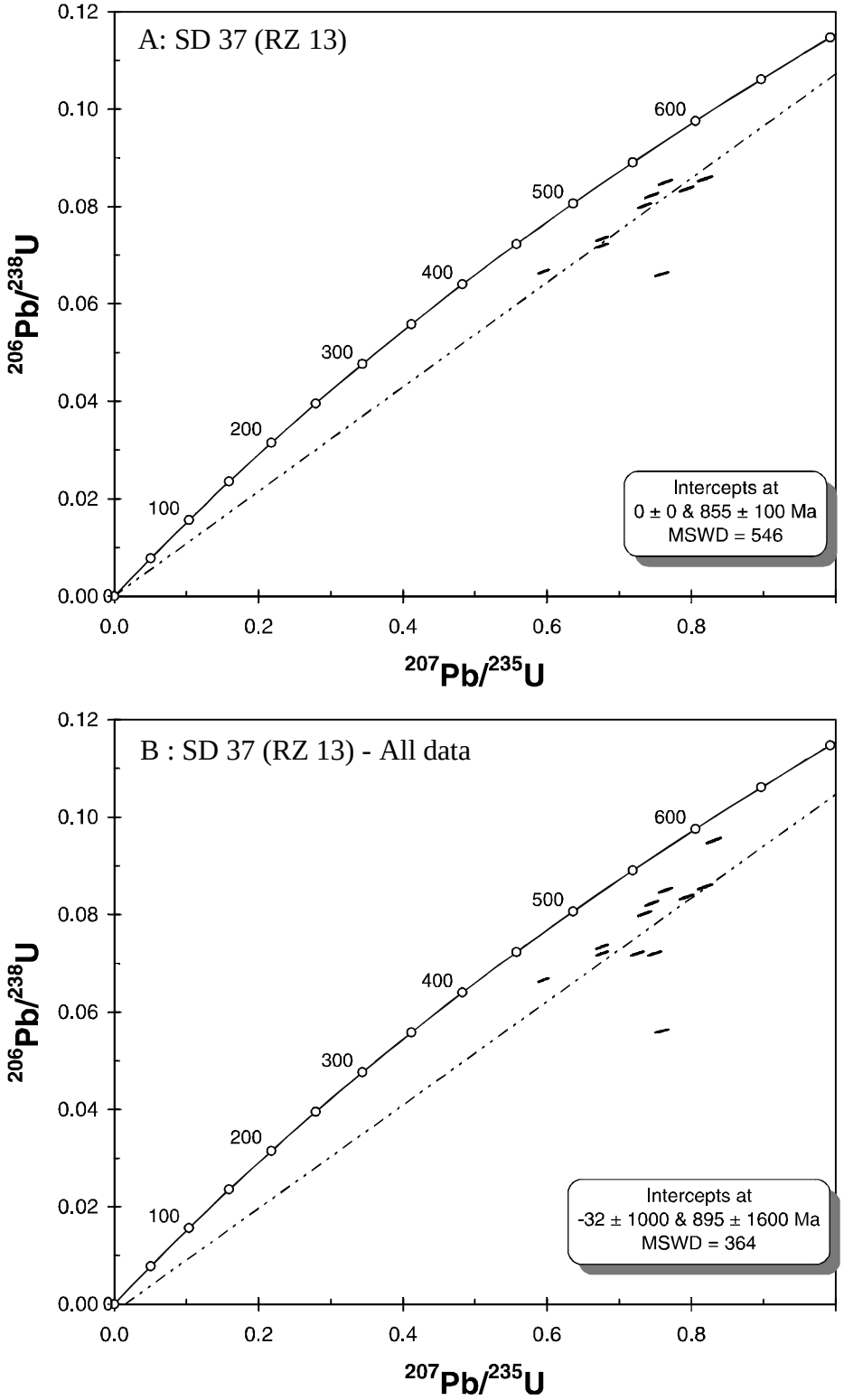

Figure A1-2*. Replotted data from SD37 (RZ13). Isoplot/Ex 2.00 /Ludwig, 1999/ was used for plotting and calculations. Errors are 2σ. A: Data from Table 4 /Gauthier-Lafaye et al 1996/. The discordia was forced through 0 to obtain an upper intercept similar to the one reported by Gauthier-Lafaye et al /1996/ B: All data from SD37, taken from Table 5 in Holliger and Gauthier-Lafaye /1996/.*

The wide range of Pb isotopic compositions of galena is an indication of a complex history of the U-Pb system and not simply a single major lead loss. The galena model ages obtained also depend on the age chosen as the time for primary uraninite crystallisation, which in this case it is set to 1950 Ma.

The discrepancy between the "most probable" age of the dolerite dyke, 980 Ma, and the age of radiogenic Pb loss from uraninite crystals, which appears to have occurred around 700 Ma, is not discussed by Gauthier-Lafaye et al /1996/. Could the crust in the Francevillian basin really have been affected by extensional tectonics during such a long time? It seems highly unlikely. As a comparison, initial rifting of the Atlantic Ocean began approximately 250 Ma ago. Today it is an ocean, with passive margins where rifting started. It seems like more data and further discussion is needed in order to interpret the disturbance of the U-Pb system in the Oklo reactor zones.

The timing of galena crystallisation is related to the timing of formation of aggregates of (S, As)-phases containing fissiogenic platinum metals. These "metallic" aggregates are found in close association with galena in the reactor zones /Gauthier-Lafaye et al, 1996/. There is some confusion regarding the assumed age of these "metallic aggregates". Firstly, the Ru isotopes indicate that the aggregates formed before ^{99}Tc had completely decayed, i.e. a couple of million years after the fission reactions. This is the conclusion from isotopic data presented in Holliger /1992/, where variable ^{99}Ru/^{101}Ru in the aggregates indicate a fractionation between Tc and Ru. Tc-rich aggregates did thus form during or soon after the fission reactions. Secondly, there is an intimate connection between galena and the aggregates, indicating that they would have formed during a period of galena crystallisation. This is further emphasised when the migration of fission products is discussed /Gauthier-Lafaye et al, 1996/ and it is mentioned that the period of dolerite dyke intrusions is responsible for a migration of platinum metals, Mo and Te.

One aggregate analysed for Pb isotopic composition, has a raw ^{207}Pb/^{206}Pb similar to the Pb in galena, as can be seen in Table 3 in Gauthier-Lafaye et al /1996/. However, Gauthier-Lafaye et al /1996/ correct this ratio for a ^{235}U/^{238}U of 0.382%, which is the lowest value these authors report for RZ13. This causes the corrected ^{207}Pb/^{206}Pb to rise dramatically, and thus the calculated model age of the aggregate becomes close to the age of the reactors. However, there is no explanation in the paper of how a certain ^{235}U-depletion has been chosen for correction of Pb isotopic composition in U-free minerals.

Thus, it appears uncertain when the "metallic aggregates", as we find them now, actually formed. Was it close to the age of the reactors, as has been claimed because of the isotopic evidence of fractionation between Tc and Ru, or was it during the episode of major Pb migration? One possible scenario is that Tc and Ru were separated during the early stages of the history of the reactors, and stayed that way until ^{99}Tc had decayed to ^{99}Ru. At a later stage, Ru may have been mobilised from both sources, ^{99}Ru-rich and ^{99}Ru-poor. During the formation of the aggregates, these two isotopic signals may have been heterogeneously mixed, giving rise to a variable isotopic composition of Ru in the aggregates.

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A new look on old data

Early isotopic studies (Rb-Sr, K-Ar, U-Pb) of the Francevillian basin yielded several results indicating that the Francevillian sediments were ca. 1800 Ma old /e.g., Hagemann et al, 1974 and Weber and Bonhomme, 1975/. After further studies, it was established that the Francevillian FA and FB formation were approximately 2100 Ma old. This is mainly based on a Rb-Sr isochron on the volcanic rocks of the N'Goutou syenitic complex /Bonhomme et al, 1982/, and the U-Pb results on normal ore at Oklo /Gancarz, 1978/, and from the reactor zone surroundings /Gauthier-Lafaye et al, 1996/. It is an age in accordance with the age of the nuclear reactions.

The published Rb-Sr and K-Ar dates from 1975 were recalculated, by the present author, with modern constants /Steiger and Jäger 1977/, see Table A2-1. All dates, except the U-Pb, are plotted in Figure A2-1. The U-Pb data obtained mainly on uranium ore from the surroundings of the nuclear reactors is listed in Table A2-2, and is plotted in concordia diagrams in Figure A2-2.

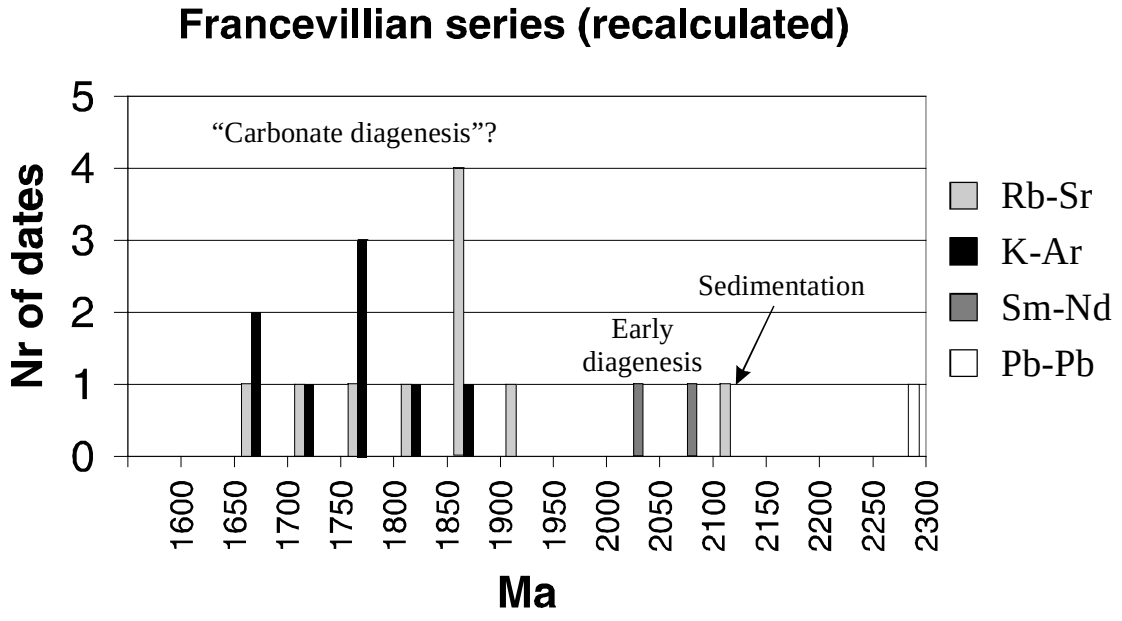

***Figure A2-1**. Histogram of Rb-Sr, K-Ar, Sm-Nd and Pb-Pb dates obtained on samples from the Francevillian volcano-sedimentary series. Based on previously published data listed in Table A2-1.*

Table A2-1. Published dates of the Francevillian volcano-sedimentary series.

Sample	Method	Age	Err	Reference
Pelitic FB-FD-FE	Rb-Sr isochron	1804*	57	Weber and Bonhomme, 1975
N'Goutou syenite	Rb-Sr isochron	2143	143	Bonhomme et al, 1982
N'Goutou syenite	Rb-Sr isochron	1799*	24	Weber and Bonhomme, 1975
Pelite FB (Oklo)	Rb-Sr isochron	1875	29	Bonhomme et al, 1982
Pelite FB (Bangombé)	Rb-Sr isochron	1867	78	Bonhomme et al, 1982
Clays (Oklo reactors)	Rb-Sr isochron	1698	145	Bonhomme et al, 1982
N'Djolé micaschists	Rb-Sr isochron	1874	91	Bonhomme et al, 1982
Illite FA < 0.5	Rb-Sr apparent age	1748		Bonhomme et al, 1982
Illite FA 0.5–2	Rb-Sr apparent age	1889		Bonhomme et al, 1982
Illite FA 2-10	Rb-Sr apparent age	1947		Bonhomme et al, 1982
Illite FA < 0.5	K-Ar	1724		Bonhomme et al, 1982
Illite FA 0.5–2	K-Ar	1796		Bonhomme et al, 1982
Illite FA 2–10	K-Ar	1887		Bonhomme et al, 1982
FD rhyolitic tuff	K-Ar	1755*		Weber and Bonhomme, 1975
FD rhyolitic tuff	K-Ar	1694*		Weber and Bonhomme, 1975
Pelite FA (BA2 955 m)	K-Ar	1812*	85	Weber and Bonhomme, 1975
Pelite FA (Oklo C1)	K-Ar	1696*	48	Weber and Bonhomme, 1975
Pelite FA (Oklo C1)	K-Ar	1761*	51	Weber and Bonhomme, 1975
Clays FA (Oklo ZR2)	K-Ar	616*	14	Weber and Bonhomme, 1975
Clays FA (Oklo ZR2)	K-Ar	2*		Weber and Bonhomme, 1975
Authigenic clays FB	Sm-Nd isochron	2036	79	Bros et al, 1992
Authigenic clays FB	Sm-Nd isochron	2099	115	Bros et al, 1992
Authigenic clays FB	Pb-Pb isochron	2265	150	Gauthier-Lafaye et al, 1996

*Recalculated with constants of Steiger and Jäger /1977/.

A considerable number of dates obtained by the Rb-Sr and K-Ar methods on the Francevillian sediments and volcanic rocks yield ages around 1800 Ma. These ages are found in all levels of the Francevillian series, as well as in schists in N'Djolé in the Ogouué mobile zone, regarded as metamorphic equivalent to the Francevillian. These ages have been connected to the third and last metamorphic event in the Ogooué mobile zone. It is said that this last event would have caused the carbonate-rich diagenesis in the Francevillian basin, which caused the resetting of Rb-Sr and K-Ar isotopic systems. If this is true, this event affected the whole of the Francevillian series, and it appears unlikely that the U-Pb system of uraninite would have remained undisturbed. It is, however, difficult to draw any conclusion about this, since later Pb loss have obscured these "diagenetic" events in the Oklo ore.

Looking at the U-Pb data in Figure A2-2, the overall impression is that there was an intense disturbance of the U-Pb system sometime in the uppermost Proterozoic. It is quite spectacular that no data point can be found near the concordia at the age of the dolerite intrusions (750–980 Ma). Instead, the majority of U-Pb data points plot inside a triangle, whose corners touch the concordia at around 0 Ma, 650 Ma, and 2000 Ma.

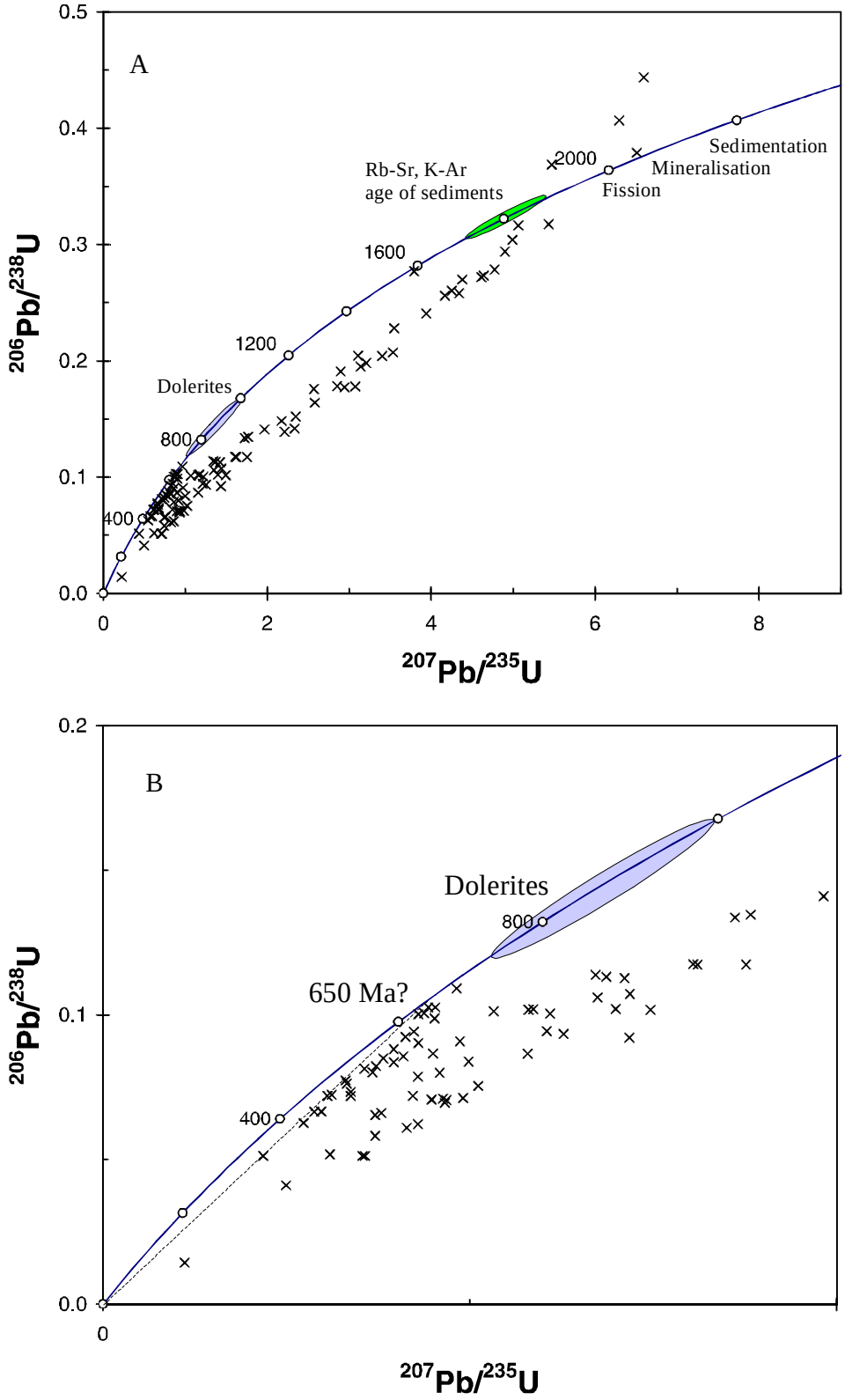

Figure A2-2. *A, All data in Table A2-2. Shaded ellipses indicate the ages of possible disturbances of the isotopic systems in the Franceville basin. B, Close-up on the samples yielding discordias said to be related to a recrystallisation /lead loss around the time of the dolerite dyke intrusions. All samples fall to the right of a discordia with an upper intercept of ca 650 Ma.*

Thus, it appears as if there was a major Pb loss from uraninite at around 650 Ma, followed by a recent Pb loss. This observation might raise the question: was it really the dolerite dyke swarm, or the extensional phase related to it, that caused the massive loss of lead noted in the Francevillian? Perhaps it was a later, Pan-African event around 650 Ma? Alternatively, the age of the dolerite dyke swarm is younger than what has earlier been suggested. In any case, it appears that important lead loss occurred in the Oklo deposit later than ca. 750 Ma.

The observation that the dolerite dykes have been altered /Weber and Bonhomme, 1975/ backs up the possibility for an important Pb loss later than the intrusion of the dolerite dyke swarm. Mafic dykes in sedimentary basins in Congo, interpreted to be of the same age as the dykes in the Francevillian basin, were metamorphosed in the lower green-schist facies sometime around 600 Ma /Vicat et al, 1991; Vicat and Pouclet, 1995/. Thus, the idea that the Pan-African orogeny might have had some effects on the Francevillian basin, is perhaps not too far-fetched.

Table A2-2. Published U-Pb data used in Figure A2-2.

Sample	207/235	206/238	Reference	Comment
KN47-330	3.108	0.2045	Hagemann et al /1974/	TIMS
KN70-1	2.348	0.152		Discordia 1.8 Ga
KN70-2	6.294	0.4065		
KN70-3	6.508	0.379		
KN70-4	1.493	0.1017		
KN 51.3	2.944	0.1772	Lancelot et al /1975/	TIMS
KN 51.4	3.792	0.277		Discordia 1.78 Ga
311.2 [hand]	1.723	0.1337		
KN 168 360	0.2227	0.0143		
KN 168 363	6.594	0.4438		
KN 168 364	5.47	0.3686		
zone 10	0.936	0.0706	Holliger /1988/	SIMS
zone 10	0.932	0.0696		Discordia 1.6 Ga
zone 10	0.859	0.0622		
zone 10	0.982	0.0713		
zone 10	1.023	0.0754		
zone 10	1.256	0.0934		
zone 10	1.437	0.1073		
zone 10	1.609	0.1175		
zone 10	1.398	0.1021		
normal ore	2.172	0.1484	Gancarz /1978/	TIMS
normal ore	3.208	0.1981		Discordia 2.05 Ga
normal ore	1.964	0.1410		Large individual errors, high MSWD
normal ore	5.432	0.3175		
normal ore	5.065	0.3165		
normal ore	2.896	0.1908		
normal ore	4.253	0.2604		
normal ore	1.349	0.1060		
normal ore	4.773	0.2786		
normal ore	2.569	0.1757		

Sample	207/235	206/238	Reference	Comment
SF42 U1/1	3.55	0.228	Gauthier-Lafaye et al /1996/	SIMS
SF42 U1/2	4.61	0.272		Discordia 1.968 Ga
SF42 U1/3	2.58	0.164		RZ 10: 2 uraninite crystals
SF42 U1/4	3.4	0.204		
SF42 U1/5	4.34	0.258		
SF42 U1/6	3.14	0.195		
SF42 U1/7	2.21	0.139		
SF42 U2/8	4.9	0.294		
SF42 U2/9	4.17	0.256		
SF42 U1/10	4.38	0.27		
SF42 U1/11	4.99	0.304		
2	1.7659	0.13458	Nagy et al /1991/	TIMS Edge of RZ9
8	0.665	0.0763		SIMS
8	0.576	0.0665		Discordia 722 Ma
8	0.612	0.072		ES24: same data in Gauthier-Lafaye
8	0.437	0.0512		et al /1996/.
8	0.713	0.0814		
8	0.547	0.0626		
8	0.624	0.0724		
8	0.662	0.0774		
9	0.887	0.1026		
9	0.86	0.1002		
9	0.876	0.1006		
9	0.964	0.1091		
9	0.906	0.1026		
SD37-35	0.819	0.0857	Gauthier-Lafaye et al /1996/	SIMS RZ 13
SD37-45	0.745	0.0823	Holliger 1991	Discordia 850 Ma.
SD37-46	0.676	0.0734		3 points taken out, one changed. The
SD37-50	0.793	0.0836		line was drawn through zero; if not
SD37-55	0.595	0.0666		the discordia will be 1040 Ma.
SD37-58	0.676	0.072		
SD37-68	0.735	0.0802		
SD37-69	0.764	0.085		
SD37-70	0.759	0.0661		
GL3535-1	1.065	0.1013	Gauthier-Lafaye et al /1996/	SIMS RZ9. No discordia.
GL3535-2	0.825	0.0924		
GL3535-3	0.904	0.0988		
GL3535-4	0.86	0.0903		
GL3535-5	0.848	0.0943		
GL3535-6	0.973	0.0909		
GL3535-7	0.9	0.0867		
GL3535-8	0.793	0.0882		

Sample	207/235	206/238	Reference	Comment
SF29-88.70/1	0.859	0.0787	Gauthier-Lafaye et al /1996/	SIMS RZ10.
SF29-89.70/1	1.621	0.1173	Holliger /1991/	Discordia 2.018 Ga
SF29-90.66/1	1.753	0.1174		Marked samples are not part of
SF29-90.85/1	1.422	0.1127		the discordia.
SF29-91.20/1	1.435	0.0922		
SF29-91.66/1	3.942	0.2408		
SF29-91.67/1	4.641	0.2733		
SF29-91.67/2	3.075	0.1778		
SF29-91.67/3	3.536	0.207		
SF29-91.67/4	2.334	0.1417		
SF29-91.68/1	2.853	0.1782		Perhaps not part of the discordia.
SF29-85.25/1	0.997	0.0839	Holliger /1991/	SIMS RZ10.
SF29-85.30/1	0.499	0.0411		Discordia 1.35 Ga
SF29-85.30/2	0.619	0.0518		"Portion argileuse"
SF29-85.77/1	1.158	0.0866		
SF29-85.77/2	1.173	0.102		
SF29-86.12/1	0.927	0.0711		
SF29-86.12/2	0.896	0.0707		
SF29-86.12/3	1.219	0.1004		
SF29-86.42/1	0.742	0.0581		
SF29-86.42/2	0.828	0.061		
SF29-86.42/3	0.845	0.0721		
SF29-87.10/1	1.343	0.1138		
SF29-87.10/2	1.16	0.1018		
SF29-87.10/3	1.372	0.1132		
SF29-87.21.1	1.21	0.0944		
SF29-87.21/2	0.918	0.08		
SF30-5175/2	0.714	0.0512		SIMS RZ 10
SF30-5175/3	0.708	0.0512		uraninite in sandstone
SF30-5175/4	0.742	0.0654		

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