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Continental Weathering as a Possible Origin of Vein-Type Uranium Deposits

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Geochemical and geochronological data presently available, concerning pitchblende vein-type deposits in France, seem altogether too contradictory for use in building a genetic model. Mostly hydrothermal hypotheses have however been suggested, although the previously mentioned idea of formation through continental weathering remains quite relevant; indeed, it may be noted that: a) Uraniferous areas are predominantly connected with granites where geochemical uranium, found as uraninite, can easily be leached through slight weathering, in the absence of any vegetation, along with Si, Al, Na and Ca. b) Apart from Na, these are elements essentially found in mineral deposits of pitchblende with a quartz, clay or calcite matrix. c) Such weathering conditions occurred during the Permian period, about 245 million years ago, at which time these mineralizations may have been laid (geochronology U-Pb); d) Veined pitchblende deposits show much analogy in their mineral associations and sequences to the uraniferous concentrations from superficial sources, as is the case with certain deposits in the United States, formed by the circulation of vadose waters. If such a model proves correct, this type of deposit would be contemporaneous with red-bed series whose presence could then become a valuable guide-line for regional-scale exploration.

I. Introduction

Uranium deposits vary greatly in form and economic importance: while stratified deposits are at present the major source, vein-type deposits are far from negligible. Though of less volume, they are, on the other hand, much more widespread, since they are found in Eastern and Western Europe as well as in North America and Australia. Owing to their frequent relation with intrusive granite, their origin is usually attributed to a hydrothermal process: in fact, geochronological measurements indicate that they are slightly younger than the granite. Numerous problems however remain unsolved as yet, including that of the origin of the uranium. Moreover, a controversy has arisen since it was noted that the origin of many of these deposits was contemporary with continental erosion. We intend, in the present paper, to examine the genetic problem concerning some uraniferous sites in France.

There are many advantages in choosing these particular areas; their regional geology is known since a long time, the deposits have been carefully described, and the presence of a good ten mine workings allows for detailed sampling. Furthermore, important advances have been accomplished in one particular field — namely, that of the geochemistry of uranium in granites — thanks to two or three thousand samples. Moreover, the age of the ore deposits, *i.e.* of Hercynian origin and therefore relatively young, makes an easier study of the usual offset between granitization and uraniferous mineralization processes: all other factors being equal, analytical errors proved to be smaller than those that necessarily appear in measurements performed on very old deposits such as Precambrian ones for instance.

II. French Uranium Deposits

A. *Geology*

An old granitized and metamorphic shield of pre-Cambrian and Caledonian origin was again involved during the Hercynian orogenesis, with renewed metamorphism and granite intrusions ranging from small isolated stocks (with frequent Sn—W mineralization) to huge batholits covering hundreds of square kilometres. Sedimentation which till then was marine, becomes continental: a thick series of conglomerates, sandstones and shales interbedded with coal; this is followed by a progressive peneplanation of the emerged continent occurring during a transient lagoonal and evaporitic Triassic period. The sea then settles in, laying down sediments consisting essentially of carbonates up to the end of the Mesozoic, with perhaps a brief continental interlude during the lower Cretaceous. It then follows a new phase with lagoonal and marine periods alternating; progressively, as from the Oligocene, these give way to a terrestrial and freshwater sedimentation of much less importance than post-Hercynian sedimentation, except for the Alps and Pyrenees areas.

Uranium deposits are of two kinds: vein formations, and stratabound deposits located in Permian continental formations, or, to a less extent, in Oligocene formations (Table 1). The pitchblend-veins, mostly ascribed to the Permian period, occur most of the time in districts related to Hercynian granites, — especially muscovite granite, but also calcoalcaline granites in the Forez and Morvan provinces (Figure 1); these areas have been studied in detail in the following monographs (SARCIA, J. and SARCIA, J. A. 1962; POUGHON 1962; CARRAT 1962; GERSTNER *et al.* 1962; CARIOU 1964; GERMAIN *et al.* 1964; DEHERT *et al.* 1964).

B. *Main Common Characteristics of Mineral Formations* (GEFFROY *and* J. A. SARCIA 1960)

1. Controlling Factors

The determining factors controlling pitchblende concentration are the secondary frac-

Table 1. Chronology for coal deposits, stratabound uranium, two-micas granites and related pitchblende veins

	Age (M.Y.)		Evolution of uraninite-bearing granites: Mortagne granite	Evolution of uraninite-bearing granites: St. Sylvestre granite	Ore deposits: Coal	Ore deposits: Uranium in sedimentary rocks	Ore deposits: Pitchblende veins in granites
Middle	Saxonian	240					Les Brugeauds Le Chardon Fanay
PERMIAN	— — —	250	— — —	— — —	— — —	— — —	— — —
						Rodez Lodève	L'Ecarpière
Lower	Autunian	260	End of deuteric processes				
						Ste Affrique	La Commanderie
— — —	— — —	270	— — —	— — —	— — —	— — —	— — —
	Stephanian	280			Coal	Graissessac Ronchamp	
CARBONIFEROUS		290		End of deuteric processes	deposits		
	Westphalian	300	Intrusion	Intrusion		St Hippolyte	

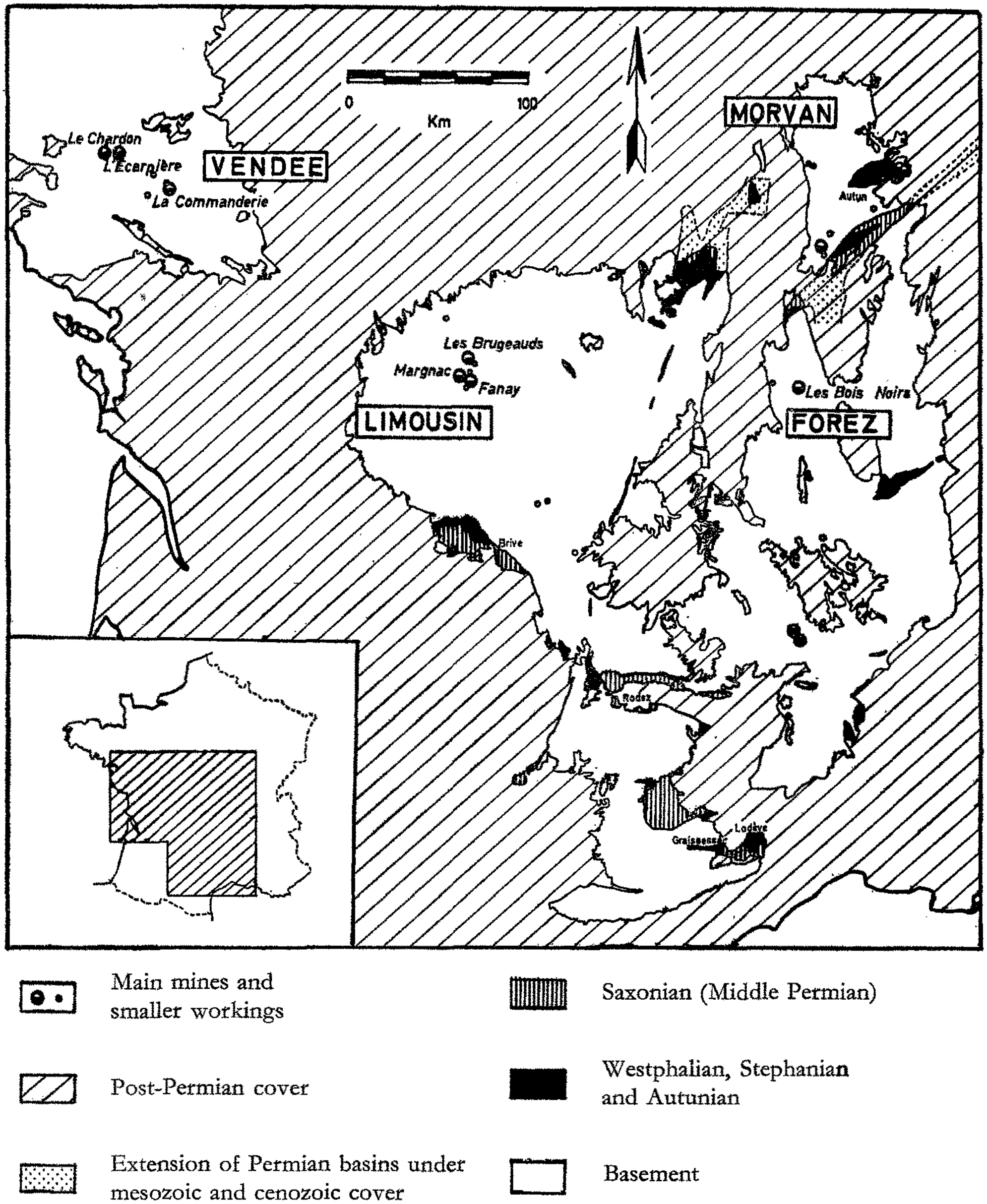


Fig. 1. Main uranium deposits and Carboniferous-Permian sedimentary basins in massif Central and Vendée. After the monography "Les minerais uranifères français" (see biblio.) and geological maps at 1/80000; extension of Permian basins after C. Weber (1972).

tures connected with important tectonic accidents, the presence of rocks with a high proportion of Fe and Mg (such as lamprophyres, dolerites) or of carbon in the sedimentary series. Special attention must be paid to the episyenites, which are column-like formations of dequartzified granite which have thus become particularly porous. When mineralized, these rocks have a strong uranium content: their presence is therefore of prime economic importance.

These pipes often have a near vertical axis; on the other hand, the dissolution of quartz, which makes up 30% of the granite, undoubtedly affects the cohesiveness of that rock. It is quite possible that in certain cases, a process of rock collapse or settling down may have occurred, which would explain the brecciated appearance of some episyenites. Among other effects brought about by this settling down, there is the possibility of a cavity forming, with a subsequent formation of karstic deposits, if that cavity is open to mineralizing solutions.

2. Wallrock Alteration

There is nothing to prove that the removal of silica from granite leading to the formation of episyenites is genetically connected with uraniferous deposits: sterile episyenites do exist, as well as pitchblende concentrations in the absence of episyenites. Conversely, formation of hematite in wallrocks seems a much more constant, though not systematic, feature; all hematized zones are, as a matter of fact, not necessarily uraniferous. Another frequent feature is the discoloration of hematite zones, with a possible occurrence of iron sulfides, when in contact with pitchblende.

3. Mineral Associations

The fact that pitchblende veins are poor in mineral varieties has already been emphasized. Those most frequently found are quartz, calcite and pyrite (or marcasite). Other sulphides occur in varying quantities, depending on the areas: galena, chalcopyrite, sphalerite, more rarely cobalt and nickel sulpho-arsenides, and sometimes bismuth sulphide. The quantity of hematite seems to follow that of calcite; while fluorite sometimes occurs.

On the whole, the sequence is probably the following: pitchblende and quartz, sulphides and finally calcite.

To conclude, it must be noted that French pitchblendes are no exception to the rule concerning paucity in thorium (Nozawa 1960).

4. Genetic Theories Suggested

The fact that pitchblende is secondary to granite appeared fairly soon. For M. Roubault (1955, 1956), the former is the result of a leaching process, undoubtedly deep but nevertheless certain of a source which "seems to be uraninite[1] in the shape of inclusions or of crystals scattered all through crystalline rocks", the localization of which could therefore well explain the origin of these deposits (Roubault and Coppens 1958).

a) *Epithermal Formation Hypothesis.* J. Geffroy and J. A. Sarcia (1958) do not think it is necessary to assume that a dissolution of uraninite occurred, pointing out that these deposits cannot be linked with any ordinary magmatic type, and that, moreover, "it is hard to imagine that these pitchblende-veins make up the upper parts of a classical metallogenic series with a high temperature", these authors associate the formation of uraniferous concentrations with a late granitic activity evidenced, in particular, by a phyllitisation possibly leading up to kaolin deposits. What then could the source of uranium be? By a process of elimination, these authors show that the element U, as well as others such as Si, O, Ca, Mn, F, Sr and Ba, are very easily leached out, even at low temperatures: those are the very elements that occur most constantly in the U vein-type mineralizations, with S and Fe. From that point onwards, one can well imagine the formation of veins through lixiviation of granite "in concentrations that probably depend on ascendant leaching of underlying mylonitized zones"; the elimination of the 5 ppm usually contained in ordinary granite, out of a volume of 0.1 km^3, produces 1000 tons of uranium, which would be the size of a small deposit.

b) *Continental Weathering Hypotheses.* Looking at the problem from an entirely different angle, M. Moreau, A. Poughon, Y. Puibaraud, and H. Sanselme (1966), following the suggestions of G. Bigotte (1964) and J. M. Obellianne (1964) stress a possible genesis by continental change or weathering: they point out that the deposits were formed around 260 M. A. (Durand 1962, 1963) in granites with a relatively high geochemical background, 8 ppm of uranium. On the other hand, mining explorations show that lode-veins rapidly throttle down,

[1] Uranium oxide is called uraninite when cristallized, and pitchblende when colloform.

suggesting a "*per descensum*" filling process. Granites that emerged during the Autunian period could have undergone a deep alteration, liberating the uranium through leaching caused by the development of a stable and abundant vegetation, that is, under biostasic conditions (ERHART 1956): this "on-the-spot decaying process, extracting the oligo-elements and destroying the silicate networks, could have freed the most hydrophilic elements, particularly iron and uranium".

Resorting to the idea of a very strong weathering can well be explained by data existing at the time about the way in which U-bearing minerals were distributed in the granite (BOWIE in KLEPPER and WYANT 1955; HAMILTON 1958; ROUBAULT and COPPENS 1957, 1958). So far, it was mostly the local displacement of that element which had been studied, on a sample-scale, without there being, on the whole, any incoming or depositing of metal, from uraniferous inclusions to clay minerals and iron oxides in fissure-joints (ROUBAULT and COPPENS 1960). The origin of uranium was supposed to be in rather stable minerals, such as monazite, sphene and apatite; though the presence of uraniferous pyrite or uraninite had been reported.

III. Recent Geochemical and Geochronological Data and Their Compatibility with Previous Genetic Models

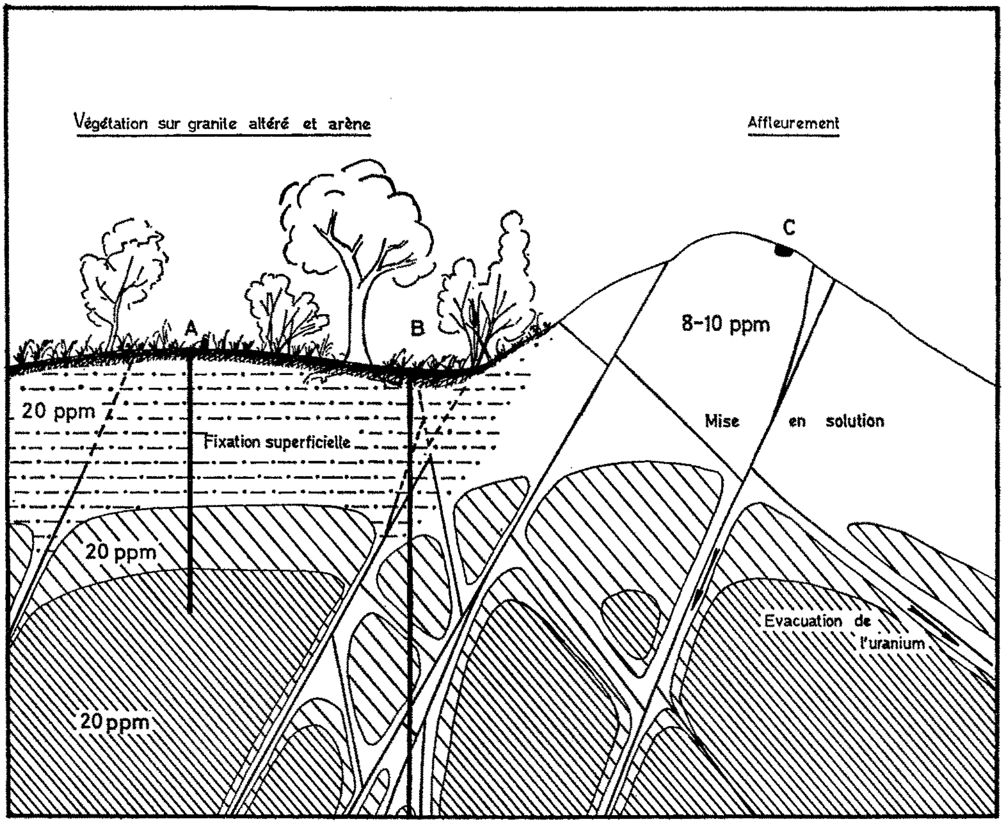
A. *Behaviour of Uranium in Mineralized Granites in Connection with Present-Day Weathering*

A great number of granites containing some uranium or pitchblende concentrations are characterized by the presence of uraninite as an accessory mineral fairly evenly distributed throughout the rock, but preferentially concentrated in quartz and micas. In the neighbourhood of the topographical surface and of fractures along which water may infiltrate, uraninite is systematically destroyed, except in cases where the quartz acts as an efficient protection. As a result, samples drawn from the surface contain approximately 8 to 10 ppm, which is half the real content of the non-modified rock. Hence, it would not be illogical to assume that half the uranium in these granites exists as uraninite (BARBIER, CARRAT and RANCHIN 1967).

The removal of uranium is not, however, the general rule; drillings beneath the first few metres of a weathered granite have shown the constancy in the uranium-content, despite the destruction of uraninite (BARBIER and RANCHIN 1969). The few fluctuations observed were shown to be directly connected with the CO_2 and H_2O contents (about 1 and 2% respectively), i.e. probably with the more or less large quantity of organic matter present; the affinity between uranium and carbon compounds, for that matter, needs no additional proof. The uranium then settles in the intercrystalline fissures or joints, as has already been demonstrated by M. ROUBAULT and R. COPPENS (1960).

This type of fixation by organic products makes it possible to suggest an explanation for contents higher than in normal granite that have been found, here and there, in some outcrops (RANCHIN 1967): one can conclude that these deviations are then not due to the presence of uranium-rich rocks, but rather to local and accidental maintenance of the original background which is higher than in the more or less leached-out superficial rocks. This trapping can only be precarious, since, in a deep drilling also carried out under cover of vegetation, the weathered zones show a marked leaching-out of uranium (BARBIER 1968). This fixation phenomenon must certainly be localized in the neighbourhood of superficial humus-bearing levels.

Whether or not there is a change in the U content of the rock, in practically all cases, the destruction of uraninite is followed by the formation of autunite in the incipient weathering zone, which autunite is then in turn destroyed when the degree of weathering increases (that mineral is but exceptionally met with in outcrops), possibly because of an acidification of the environment. The progressing change in the uranium seems to depend on the presence or absence of plant covering: if it does occur, the fixation takes place in the granite joints; otherwise, there is no fixation but an evacuation into the depths (Figure 2). If the organic matter is destroyed by oxidation (as, for instance, when the vegetation disappears), the uranium is released: this appears to happen in most present-day outcrops which happened to be covered by woods at some early time or other. As for



Geochemical zones:

Leached granite

Unweathered granite

U fixation (by oganic matter mainly)

20 ppm Mean U value for each zone

Oxided granite with autunite

Fig. 2. Schematic section showing U-weathering in an uraninite-bearing granite. Section A: Margnac drillings, Limousin (BARBIER and RANCHIN 1969a). Section B: Blond drilling (BARBIER 1968). Section C: Outcrop sampling (BARBIER et RANCHIN 1969b).

the phosphorus necessary to the formation of autunite, its origin could not be explained as yet; it may well have originated in weathering granite, or it may be of biogenic origin.

The idea of a genesis of the vein-type pitchblende deposits by continental weathering may have immediate implications: there is no need to call on a strong hydrolysis which could only lead to a dispersal of uranium, since even a slight oxidation will set it off in a selective way; on the other hand, an abundant vegetation will inhibit rather the mobility of that metal; while a lack of organic matter will, on the contrary, favour it.

B. *Geochronology of Granites and Uraniferous Mineralizations*

A large series of measurements have made it possible to evaluate the age of granites by the Rb/Sr ratio technique; the results are interpreted by the method of isochrones (Nicolaysen 1961) while the age of the pitchblendes is obtained by the Concordia Curve (Wetherill 1956):

Mortagne granite (Vendée)

300 M.Y: intrusion of the granitic body

260 M.Y.: end of Rb and Sr exchanges — on a sample scale — between the different mineral phases (Sonet 1967).

270 M.Y: formation of pitchblende at La Commanderie (Kosztolanyi 1971) with a reshuffling at 40 M.Y.

245 M.Y: laying down of Le Chardon pitchblende with recent reshufflings around 20–25 M.Y. (Kosztolanyi and Coppens 1970).

255 M.Y: pitchblende at L'Ecarpière, with a reshuffling around 20–25 M.Y. (Kosztolanyi 1971).

Saint-Sylvestre granite (Limousin)

315 M.Y: intrusion of the granite.

290 M.Y: end of intermineral reorganization (Duthou and Vialette 1972).

240 M.Y: pitchblende at the Brugeauds, reshuffling around 15–20 M.Y. (Coppens, Kosztolanyi and Dottin 1969).

245 M.Y: Fanay pitchblende (Kosztolanyi 1971).

These figures are easily interpreted: uraniferous ores cannot have a deuteric or hydrothermal origin related to the end of a granite-formation period, as was stressed by J. L. Duthou and Y. Vialette (1972). The 260 and 290 M. Y. ages given respectively for Mortagne and St. Sylvestre granites indicate the end of the Rb/Sr exchanges in a closed system, on a sample-scale, and can therefore be reasonably taken as the end of the deuteric processes affecting the various mineral phases, if one excludes the possibility of a local modification occurring without change in chemical compositon of certain minerals (among others, the known transformations of biotite into chlorite, calcite, fluorite and iron oxides). Pitchblende is formed later, and this is particularly obvious in St. Sylvestre granite which shows a lapse of about 45 M. Y. An epithermal origin could then be considered, as was done by J. Geffroy and J. A. Sarcia (1958); but the origin of the uranium then becomes a problem, since no evidence of leaching out has been observed in the granite itself, whether or not it has been subjected to tectonic effects, outside the zones of weathering: a cataclasm breaks up the crystals of uraninite but does not induce their disappearance. One does observe, fairly frequently, a slight corrosion of the uraninite (Ranchin 1968) but that affects hardly more than 10% of the crystals, and there is nothing to suggest that it occurred at the same time as the pitchblende deposits (or slightly earlier if there had been any genetic linkage between the two phenomena).

C. *Formation Conditions for Pitchblende*

The measurement of homogenization on liquid inclusions has led J. Leroy and B. Poty (1969) to assume that formation took place at a relatively high temperature — about 400 °C — which together with the suggestions by G. Ranchin (1970) constitutes a new interpretation. These data should not be neglected, since they were obtained through precise physical measurements; nevertheless, they immediately contradict many other data:

— Though the use of minerals as geothermometres is both awkward and controversial, the presence of marcasite formed at the same time as the pitchblende would tend to show a rather low temperature of formation (J. Geffroy and J. A. Sarcia 1960; M. Arnold 1969).

— Beyond 150 °C, experimental syntheses have yielded crystallized uraninite and no pitchblende (Rafalsky 1958), this latter being obtained at 25 °C (Miller 1958).

— A temperature of 400 °C at about 250 M.Y. is surprising for granites which are known to have been subjected then to certain erosion processes, since boulders have been found in the conglomerates (KLEIN 1970); unless one assumes there is an error in the geochronological determinations, or that there occurred some hypothetical volcanic phenomena (or rather that there existed, to be more exact, fumaroles, since no Permian volcanic site has, to date, been found in the Vendée or Limousin provinces).

Wall-rock hematization phenomena must be connected with the circumstances and conditions of ore-formation. Observations made on samples from the Brugeauds mine are as follows:

Hematite preferentially settles in most easily modified minerals of granite: feldspar and biotite; small calcite inclusions, more particularly, completely disappear and holes form, which could either have resulted from the tearing processes used to prepare thin rock slices, or from a natural "boxwork" process. Whatever the case, hematite replaces calcite wherever the latter occured, *i.e.* especially in oligoclase or biotite (containing a greater or lesser quantity of chlorite). It is then in turn destroyed when in the immediate vicinity of pitchblende, in relation with a possible formation (or deposit) of pyrite, as evidenced by some rosette-type residues (J. GEFFROY and J. A. SARCIA 1960).

To conclude, data now existing about the pitchblende deposits are contradictory: if one trusts temperature measurements, this would tend to prove that geochronological measurements are probably erroneous, and vice-versa. Since neither lead to any certain conclusion, the best attitude is, undoubtedly, to accept information only in so far as it can be harmoniously integrated into a model that can explain other facts observed. Since the genetic hypotheses made to date are mainly based on a hydrothermal origin following a long magmatic activity (RANCHIN 1970; CARRAT 1971; RENARD 1971), we think it may be useful to suggest as an alternative a model based on continental weathering: this is borne out by the fact that one finds, soon after the intrusion of granite, both hydrothermal mineralizations and erosion of the Hercynian mountain range (Table 1). On the other hand, as has been shown above, certain facts lead us to reconsider the outline suggested by MOREAU *et al.* (1966).

IV. A New Genetic Model: Continental Weathering with Little or no Vegetation

A. *Permian Weathering and the Mobility of Uranium*

1. Weathering During Permian Period

There are two ways of finding out its characteristics; the first consists in direct observation of the fossil soils, while the second method studies a sedimentation series: trying to recreate a natural background or climate from sedimentological data is now a common method. As an exhaustive study of the Permian period is not possible here, one may refer to authoritative works on the sedimentology and weathering involved.

a) *Autunian.* Following a pedological decay of the bedrock, sedimentation accumulates increasingly resistant materials, such as quartz and leptynites; according to L. CARIOU, Y. FUCHS and C. SCEMAMA (1967), this is evidence for a progressive alteration with a formation of soil. The climate is warm, with dry seasons; heights are drier, with a possible development of red alteration, while the reddish sediments turn into buff or greyish beds after reduction by organic matter in the swampy areas. (MILLOT 1964; GARRIC 1965).

b) *Saxonian.* Dried soils become more common, possibly pointing to a more arid climate (GARRIC 1965; PERRIAUX 1961); at the bottom, grounds with a high Fe^{3+}/Fe^{2+} ratio can be found, owing to oxidizing conditions, while hot and dry atmospheric conditions favour the concentration of copper (FUCHS 1969). Sediments often contain detritic feldspars, including the easily attacked plagioclases (MILLOT, PERRIAUX, LUCAS 1961; PERRIAUX 1961): chemical hydrolysis is then very weak. Climate has hardly changed since the Autunian period, "humid and warm, with intermittent dry periods", and "tropophilic forest vegetation in the lower regions and xerophytic bushes on higher grounds or in dry spots". (FALKE 1961; ERHART 1962); G. MILLOT (1964) have completed this description in the following way: "it seems that tops as well as slopes must have been sparsely populated, continually reshaped, constantly rejuvenated, and must have been covered with a meagre vegetation, except for lower grounds that were permanently humid." This is evidently a case of rhexistasia, as H. ERHART (1956) put it. After having alternated, for a long time, with grey sediments, red sediments settle in permanently, since there is no longer any reducing organic matter left.

c) *Possible Effects on the Mobilization of Uranium.* During the Permian period, the Hercynian range is in the process of being broken down. Detritic materials accumulate in the lower levels up to a thickness sometimes reaching 1000 metres. Erosion has therefore been considerable, and the figure of 100 metres which could be suggested concerning erosion within a lapse of time corresponding to the formation of uraniferous ores would not be unreasonable, at least in some places. This figure should probably even be increased, thus confirming the hypothesis of large amounts of uranium having been leached out. In granite containing 10 ppm of uraninite, spread over an area of 400 km^2, the quantity of metal thus released would amount to over a million tons.

Among the characteristics of the Autunian period, special mention must be made of the presence of a thick mantle of vegetation. Observations made on present cuts tend to show that this is not a fact conducive to uranium migration. It is an element quickly fixed by organic matter (undoubtedly also by colloids, iron hydroxides and various other alteration products), a fact which hampers its displacement.

During the Saxonian period, vegetation disappears more or less completely, perhaps because of a drying up of the climate, generally. Erosion occurs essentially through mechanical means, with little hydrolysis, as well as through oxidizing conditions due, at least partly, to the absence of organic matter. The uranium thus released by the destruction of the uraninite is therefore selectively leached out, since it is no longer stabilized. This is a process now being observed on granitic outcrops. The Saxonian period is therefore more favourable to an important leaching out process than is the Autunian.

In actual fact, the essential point does not lie in the matter of chronology, but rather in that of the weathering conditions (MILLOT 1964; GARRIC 1965; FUCHS and PINAUD 1969). It is, for instance, quite possible that during the Autunian, uranium may have been eliminated from the granite or bare heights and may have been fixed in the soil lower down, where there was some vegetation. This "position effect", which would have been noticeable before the Saxonian period, must have subsequently disappeared with the general spreading of oxidizing weathering conditions.

Some direct observation tends to give credit to this scheme: under the Autunian sediments, a paleo-pedological concentration of uranium on clays or iron-rich rocks has in fact been spotted, while the Autunian sediments under the Saxonian deposits have, on the contrary, been leached out (FUCHS 1969).

2. Weathering and Chronology of Uranium Deposits

From the Westphalian to the Autunian periods came a period of relatively stable and abundant vegetation which produces sediments with some uranium-rich concentrations; these formed, or so it would appear, later than the strata in which they were found, though apparently only slightly later (GRIMBERT 1956). Such is the case for the ore deposits of the Westphalian (St. Hippolyte), of the Stephanian (Graissessac, Ronchamp) and especially of the Autunian periods: the Rodez basin, Lodève and Sainte-Affrique (CARLIER 1965; GARRIC 1965; KERVELLA 1965). The Saxonian period, on the other hand, is practically devoid of any such deposits (Table 1).

This difference can easily be explained as a matter of weathering if, along with Y. FUCHS (1969), we accept the fact that fixation of uranium in soils brings about its elimination towards a deposition basin, once the soils are eroded. Such occurrence could not happen during the Saxonian period, because erosion involved weathered rocks which had already, most probably, lost their uranium. What then happened to the metal thus set free? The only possible movement is a migration downwards along the fractures, carried down by rainfalls. These are known to descend quite far since, even in France, oxidized zones occur more than 400 metres below ground level; beneath mountains, the superficial waters have been found to circulate at depths of about 1500 metres, and there is nothing to imply that this is a maximum.

The 500 metres hole drilled in the Monts de Blond (Limousin), is an interesting example; in some of the deep fractures, oxidation has removed the uranium (Barbier, 1968); but other fractures, lined with black, fetid coatings, are on the contrary rich in uranium (200 ppm); this proves that, along with the disappearance in the oxidizing zones, fixation occurs, at least partially, in the reducing zones.

This must be taken into consideration in connection with the age most commonly suggested for pitchblende-ore formation, *i.e.* 240—250 M.Y. This age, according to Afnassyev *et al.* (1964), corresponds to the end of the Permian period; while the Holmes' symposium held in Glasgow (1964), came to the conclusion that it corresponded to the end of the Saxonian; finally, according to Y. Fuchs, F. Leutwein and J. L. Zimmermann (1970), this coincides with the frontier between the Autunian and Saxonian periods. Although this last measurement was obtained by the K-Ar technique — therefore different from the U/Pb technique used for pitchblende — it should be given our full attention, since it involved samples from the Détroit de Rodez (France).

On the basis of geochronological determinations, it would appear that vein-type uraniferous ores are, on the whole, posterior to the Autunian period, and date back to the times when superficial fixation of uranium did not occur, owing to the absence or disappearance of the vegetal mantle. Thus, *sedimentary deposits are followed by the formation of pitchblende veins, in the same way as grey sediments are followed by red strata.* Isotopic dating, geological environment of the uranium deposits, sedimentology, past and present alterations make it possible to elaborate the broad outline of a model in which the different facts observed fit fairly coherently.

In this light, it is not impossible to envisage the existence of Autunian pitchblende veins, since a reddening weathering process is understood to have taken place on high grounds; this might well apply to the Commanderie site, if the age of 270 M.Y. that has been suggested proves to be exact. A different argument leads back to the notion of "fertile granites" on high grounds mentioned by M. Moreau *et al.* (1966). Pitchblende sites would even have a true paleotopographic significance, with the older ones matching the highest positions, while the youngest ones would go with the lower zones where vegetation disappeared relatively late. It should be noted that a culminating position of two-mica granites — at present characterized by negative gravimetric anomalies — is not unlikely.

To conclude, let us note that it is impossible to say whether pitchblendes could have been laid down during the entire Saxonian period or whether their formation coincides with the disappearance of vegetation (agreement, according to Y. Fuchs *et al.* (1970), between the U/Pb dating and the end of the Autunian); the accuracy of geochronological data is insufficient, but the problem is well worth looking into. There are thus three possibilities:

a) Formation of pitchblende vein-type mineralization by erosion of the granites during the complete sedimentation periods of the "red-beds".

In this case, the essential factor is the existence of oxidizing conditions which are here due to the absence (or scarcity) of organic matter.

b) Formation, at the time of disappearance of the vegetal mantle, by liberation of the uranium bound to organic matter in the weathering mantle.

The deposits would then have originated in the destruction of a temporary form of fixation.

c) Weathering of the earlier uraniferous rocks, such as the Autunian carbonaceous sediments. This hypothesis corroborates the theories of G. Bigotte (1964), analogous to hypothesis a., except for the fact that the weathered substance is not granite.

Only hypotheses a. and c. seem likely to involve large quantities of metal; there is, moreover, nothing to exclude the possibility that the three mechanisms may have occurred: with different initial geochemical associations of uranium, erosion would lead to three types of uraniferous deposits, all vein-type, but probably with different mineral associations. However, the precise localization of the main pitchblende deposits in the immediate vicinity of certain granites rich in uraninite seems to exclude a sedimentary phase and thus supports hypotheses a. and b., *i.e.* formation on the spot, without any intermediary migration.

B. *Concentration of Uranium with Respect to other Elements*

In the absence of abundant vegetation, erosion occurs mainly through mechanical means with a rapid elimination of loose debris. In this incipient hydrolysis, and in the case of leucocratic uraninite-containing granites, the elements released are Si, Na and — though to a lesser extent — Ca and Al, as far as the major elements are concerned (BARBIER 1968). Because of the high solubility of sodium, the solutions thus formed can only give rise to siliceous or, more rarely, to silico-aluminous products (clay) with a little calcite.

These are precisely the most frequent gangues of pitchblende deposits: J. GEFFROY and J. A. SARCIA (1960) furthermore point out that siliceous gangues give the impression of having been formed as a gel of silica, uranium oxide and iron sulfide rather than as a succession of deposits of these constituents. In this outline, the presence of iron in large quantities is not easily explained (since this metal is not very mobile in the oxidizing conditions at the surface) unless organic complexes play a part: as a matter of fact, the existence of carbonaceous matter (graphitoids) in pitchblende is well known.

As compared to other trace elements, uranium shows an extremely high mobility during the early stages of weathering: surface granites often lose half their content of uranium while no particular leaching out has been noted for Ba, Sr, Zn or Pb (DE LA ROCHE, LELONG, FRANCOIS 1966; BERNARD and SAMAMA 1968); oxidation is also responsible for the dissociating of the uranium-thorium pair. *A slight weathering found in the absence of important vegetation can therefore explain, in a simple fashion, the separation of uranium.* On the contrary, a well-developed organic life will lead to the fixation of uranium along with other biophilic elements with which uranium will be found associated in fine polymetallic sediments.

To conclude, a partial hydrolysis happening concomitantly with mainly mechanical erosion similar to the one that occurred during the Saxonian period seems quite likely to perform the separation which concentrates in a dissolved phase the components which make up the uraniferous veins. Intervening biochemical processes are apparently necessary to explain the behaviour of iron and the presence of graphitoids.

However, supplementary data on the geochemistry of sulfur and of fluorine would be most welcome to fill in the picture.

C. *Precipitation of uranium: a comparison with Certain Strata-Bound Hematized Deposits*

While downward infiltration of rainwater is an undisputed fact, it does not seem generally admitted as yet that this phenomenon could also give rise to pitchblende veins which are firmly established as having a hydrothermal origin. Yet, the concentration of uranium and the nature of the gangues most frequently met with are easily explained by a process of slight chemical weathering; the formation of uranium oxides at ambiant temperature and in confined conditions is theoretically possible (GARRELS and CHRIST 1965), and has even been obtained experimentally (RAFALSKY 1958); moreover, all pitchblende deposits in fluviatile continental sediments go to prove that this ore is often found in a geological environment belonging to the supergene field. A uraniferous filling of intragranitic fractures under similar conditions is therefore not unlikely.

This type of association is, however, not very convincing in itself, in view of the fact that sedimentary and "hydrothermal" ores often differ as to their mineral associations and related modification; definite resemblances must, however, be pointed out, and will be examined in the following paragraphs.

1. "Shirley Basin" (Wyoming, U.S.A.) Type Deposits

In the strata-bound roll-type U deposits of the United States, D. R. SHAWE and H. C. GRANGER (1968), as against H. H. ADLER (1964), divide mineralization into two main types. The first one of these categories contains deposits surrounded by a ring of rocks showing evidence of reduction: this is the "Colorado plateau" type; in the second the deposits are localized on an oxidized tongue in the mist of formations usually constituted by sandstones. This "Shirley basin" type occurs with a hematization of the sandstone in the vicinity of the pitchblende ore, and this con-

stitutes an analogy with intragranitic mineralizations that justifies a supplementary scrutiny. It must be emphasized that this type of deposit is not exceptional, but is even of common occurrence, since it is met with — apart from the Shirley basin — in Wyoming (Powder River basin) and in South Dakota (Black Hills).
It may be said that the presence of hematite constitutes a characteristic feature, and as much can be said of its behaviour: an early phase of hematization occurred, followed by the replacement by the uranium species, as is proved by the red traces found in the ores; there is always a bleached zone between the pitchblende and the reddened sandstone (Sharp and Gibbons 1964; Mrak 1968; Hart 1966). Concerning the Shirley basin, R. E. Melin (1964) gives the following sequence: hematite, calcite, pyrite, marcasite, pitchblende, with each mineral replacing, at a given point, the mineral previously laid down. This succession in time is accompanied by a succession in space, since pitchblende, calcite and hematite are found between unoxidized and oxidized sandstone.

According to R. E. Melin (op. cit.) these facts can be explained by the progressive neutralization of a quantity of acid water percolating through the sandstones, with the neutralization brought about by the weathering of the rocks through which the water flows. Ahead of the water, the medium is neutral or slightly alcaline, and calcite and hematite are deposited from the solution; as the boundary progresses, calcite and hematite are formed further down; their place is taken by mineral substances which are stable in more acid media, such as pyrite, marcasite and pitchblende. When the solution is practically neutralized by the alkali-metal ions, the sequence is reversed and marcasite, pyrite, calcite and lastly hematite are successively deposited beyond pitchblende.
E. N. Harshmann (1966) interprets there facts in a somewhat different fashion. Unoxidized sandstones contain pyrite, ilmenite, magnetite, calcite and organic matter, which are destroyed in weathered sandstones; these are, however, richer in Se and Fe^{3+} while between the two lie ores containing, more particularly, concentrated mineral carbon, S, Se, Be, Fe^{2+} and U. That author then concludes that the weathering of sandstones is the result of the passing of neutral waters with alcaline and oxidizing tendencies, and that the precipitation is due to the strongly reducing conditions at the leading edge of the sheet of water.
While genetic considerations are being discussed, it must be mentioned that a magmatic-hydrothermal origin constitutes a hardly defensible theory (Harshmann 1968); on the contrary, the interfering of bacterial action is often called on, though the metallogenic efficiency — at least in the present case — of the process is still a controversial point (Warren 1972).

2. Comparison with Intragranitic Veins

The main ores formed are, on the whole, identical in roll-type and vein-type pitchblende deposits: apart from uranium oxide, there are sulfides (mainly of iron), as well as calcite and hematite; the gangue could perhaps be richer in silica in the case of granitic deposits. Furthermore, the succession of ores seems very similar; the hematized remnants, as well as the bleached fringes in contact with pitchblende which are found in the Powder River basin deposits have their counterpart in French deposits (Geffroy and Sarcia 1960). In both cases, hematite and calcite are among the last to precipitate, being preceded by pyrite, which is itself posterior to marcasite (as regards the Limousin sites, cf. M. Arnold 1969). Veins are also enriched in S, Fe^{2+} (pyrite), mineral carbon (calcite) and uranium; the similarity is complete if one refers to selenian minerals reported in certain areas (Agrinier and Geffroy 1965, 1968, 1969).
The analogy is sufficiently close for the filling of the granitic features to be explained by a mechanism similar to the one which gave rise to roll-type deposits: a penetration by superficial water, then a reaction with the sheathing rocks and alteration of the wallrocks (dissolving of the calcite, hematisation), formation of mineral species *in an order which, in time, is the reverse of that observed, in space, at the wallrocks.*

3. Probable Deposition Circumstances

Resemblances noted between the uraniferous mineralization of the "Shirley basin" type and intragranitic pitchblende veins support the theory of formation at low temperature from meteoric water: logically, one should also take into consideration the possibility of deposition through a change in pH conditions. However, contrary to sedimentary deposits, this difference can hardly be generally attributed to a reaction of water with granitic host rocks. Indeed, pyrite and calcite are only found in extremely small quantities, organic matter is completely lacking and weathering of plagioclases (judging from the decrease in sodium content of the granite) is hardly perceptible, even in certain strongly hematized zones.

Further hydrolysis may occur later, through meteorological weathering, but this is in no way connected with hematization. On the other hand, hematization does not systematically seem to go with the formation of autunite (although the uraninite inclusions are destroyed) as in present-day meteorological oxidation, so that its development under oxidizing conditions is not quite certain. But could autunite — which is a highly hydrated mineral species — have remained stable for over 200 M.Y.?

Since the neutralizing and reducing power of granite on the solutions is certainly much weaker than that of pyritic sandstone, with calcite and vegetal products, it can only be effective at the limit of water penetration and in low permeability zones: this could explain the very slight weathering of hematized parts, since iron oxide may be formed by oxidation, but also by neutralization (GARRELS 1965) which, in this case, occurs only at the limit of unaltered granite.

Since the reduction necessary for pitchblende formation and sulfide formation cannot be ascribed mostly to a reaction with granite, its cause must be sought for elsewhere, in a progressive confinement: this would cause an enrichment in alkali-metal ions — hence a neutralization — which would itself bring about a related precipitation of calcite and hematite. Mineralization would then happen in the following sequence:

a) Faulting of the granite, infiltration of slightly acid and oxidizing uraniferous solutions; destruction of the calcite in the granite and slight hydrolysis of the plagioclases: formation of autunite and limonite in the zones of incipient weathering.

b) Superficial erosion of the granite with continuous selective leaching of uranium, stagnation of the waters in depth, due to the levelling of the relief and stabilization of the hydrostatic level. The medium is confined and becomes reducing (biochemical processes, reduction of the organic matter); partial bleaching of the ferric oxides occurs and pitchblende, marcasite and pyrite are deposited.

c) Progressive dehydration and filling up of the cracks; disappearance of autunite and transformation of limonite into hematite; eventually, final laying down of calcite and hematite when the medium slowly becomes alcaline.

D. *Conclusion: "Per Descensum" Genesis of French Vein-Type Pitchblende Deposits*

Having thus followed the migration of uranium from granite at the start of the weathering process to its deposition in depth within the fractures, one can draw a quick sketch of the formation of intra-granitic veins. In the lower Permian, erosion vigorously attack the Hercynian ranges, but plant life is extensive; uranium, which has been freed from granitic bodies rich in uraninite, is trapped in the neighbourhood of the surface at pedological levels; once these have been destroyed, the uranium is removed along with other elements which were also trapped in the superficial formations. The result is a fine sediment rich in uranium and numerous other metals, with an abundance of organic matter.

During the middle Permian, vegetation has largely disappeared and weathering is mainly mechanical, with little chemical attack. The levels at which uranium was fixed have been destroyed and this element becomes one of the very first to be released by weathering, thus being separated from the other metals. After being leached out quite selectively, it migrates towards the deep fractures together with some major elements. Reaction with granite wall-rocks causes hematization, while confinement and reduction lead to the formation of pitchblende, marcasite and other sulfides; progressive alcalinisation finally yields the hematite-calcite terminal phase. This results in the formation of uranium veins which are posterior in time to the stratabound deposits containing organic matter, though there may possibly exist a transition period during which both formations may coexist, one of them being connected with high grounds devoid of vegetation, and the other occurring in accumulation basins.

E. *Variations on and Constants of the Model*

The model suggested may seem over-simplified, inasmuch as it only deals with general facts and does not give an explanation of the variations in the mineral associations, host rocks or in the formation encountered in the uraniferous lode veins. Nevertheless, a fairly widespread type of mineral formation can certainly only be accounted for by a sequence of fairly common processes. On the other hand, a simple model has the advantage of being adaptable to numerous specific cases and, with a few variations, applicable to fairly dissimilar objects.

1. Variations

Of the mother-rock

Though we have used uraninite in granite as an example here, and since the essential requisite is the presence of uranium in a fairly labile form, the rock which contains uranium could just as well be gneiss, a conglomerate or any other silico-aluminic uraninite-containing species. Other potential sources are sandstones and uranium-bearing schists.

In the host-material

As happens in fractures, certain granitic formations will give rise to local uranium concentration if they can contain stagnant water: such is the case with *episyenites* mentioned at the beginning of this article which simply act as a passive reservoir in the hypothesis considered here. The host rock is, furthermore, not necessarily granitic, and since slightly but definite reactions do occur between the solution and the wallrocks, variations in the accompanying mineral species can be predicted.

In the mineral associations

Mineral substances originate in solutions carrying elements freed by superficial weathering and by exchange with rocks in depth. The nature of the gangues and sulfides, as well as their abundance, will therefore depend on the geological context. This fact is as true on the scale of a single deposit (richness in iron sulfides close to veins of lamprophyre, GEFFROY and SARCIA, 1960) as it is on the scale of a whole district (more abundant calcite and barite in a granite rich in Ca and Ba, BARBIER 1971), or of a metallogenic province (GEFFROY 1964). However, it is, for the time being, not easy to distinguish among elements released by meteorological weathering, and elements arising from topochemical reactions with wallrocks.

It is undoubtedly not a mere coincidence that mineralizations richest in Co-Ni should be associated with the most basic environment in a district essentially made up of schist and gneiss (Nicholson mine, Beaverlodge district, Canada); similarly, the Great Bear lake deposits with Bi, Co, Ni and Ag are found in a volcano-sedimentary series of tuffs and dacito-andesitic effusive rocks (LANG *et al.*, 1962). The Co and Ni content of these rocks can be ten times higher than that of granite and their frequent high Bi content has also been noted (B.R.G.M., unpublished analyses: up to 20 or more ppm). It therefore seems reasonable to assume that uraniferous concentration with Bi-Co-Ni-Ag contents are connected with volcano-sedimentary formations, at least as a working hypothesis.

This does not imply that the metals present in this type of deposit all share the same origin: in the model which has been outlined here, the uranium is freed by meteorological oxidation. Bi, Co, Ni and Ag could originate from a reaction in depth between wallrocks and migrating sheets of liquid during the phase of confinement and neutralization; since we have assumed here that this is a late phase as compared to the formation of pitchblende, this would constitute a simple explanation of the relatively early deposition of pitchblende found in deposits containing Bi, Co, Ni, Ag and U.

In the same line of reasoning, mention must be made of the uraniferous deposit containing chalcopyrite and chalcolite at Les Bois-Noirs (POUGHON 1962), close to the old copper mines of Charrier.

In the deposition time

Weathering conditions as they prevailed during the Permian in Western Europe are not unique: they occurred during the Devonian period as well (old red sandstone), and, in France, during certain periods of the Miocene. Although no pitchblende deposit of Devonian age seems to have been found to this day (but were the necessary mother-rocks then outcropping?) changes in pre-existing deposits occurring at about 20 M.Y. (Miocene) are considered as being possible (CH. KOSZTOLANYI 1971). They are preceded by a period during which continental stratabound deposits are laid down and in which organic matter, however scanty, played an important local part in concentrating the uranium (CARIOU 1964). Weathering during the Miocene may have played, as far as this metal is concerned, as important a part as it did during the middle Permian period, by stopping its removal towards the accumulation basins and by shifting it in depth by the disappearance of organic matter.

2. Prerequisites and Possible Verifications

A certain number of points, constant to all the numerous forms that could arise from this model, must be emphasized:

— The presence of a mother-rock potentially rich in uranium.

— Continental weathering with little or no organic matter.

This last condition, as against those connected with a hydrothermal scheme of formation, excludes a possible genesis at any time prior to 2000 M.Y. (no oxygen in the terrestrial atmosphere, cf. S. M. ROSCOE 1968) and to the marine sedimentation. On the contrary, it implies the *existence of a medium favourable to the laying down of red bed series of which pitchblende would, in a way, constitute a lateral equivalent.* The model which has been suggested thus carries in itself its own means of control, and the clue of the debate between "hydrothermalists" and proponents of a formation "*per descensum*" may lie in stratigraphic or sedimentological studies.
The inferences drawn from this model do not, till now, seem to bring about a refutation. Let us consider, as an example, the uraniferous Beaverlodge District in Canada (close to lake Athabasca); we find that uraninite is given as being in granite and pegmatites (BECK 1968), a fact which agrees with the "favourable mother-rock" condition; the age of the pitchblende veins is at most 1750 M.Y. (KOEPPEL 1968), therefore posterior to the 2000 M.Y. limit, and is furthermore sub-contemporary to a red continental series, the Martin formation (TREMBLAY, 1968). There is thus nothing to forbid looking upon the mineral deposits of that district (which, besides, are quite similar to French deposits in their hematization and their mineralogy) as having also been born of a particular phase of continental erosion.

F. *Consequences for Exploration*

It follows from all this that if the theory suggested is correct, the presence of red sedimentary series lying on an old shield is a factor quite favourable to the presence of uraniferous veins in the basement. These series will most likely occur (especially in the more ancient shields) as scattered remnants lying in former depressions. The mineralizations themselves could be quite distant from them: a sketch made of the main deposits in France shows, at a glance, that there is a distance of over 100 kilometres (Figure 1) between uranium deposits and *present-day* outcroppings of the Permian series. This will therefore only constitute a valuable guideline when used on the scale of a province, i.e. in wide-range prospecting. On a ten-kilometre scale, sharper control will be provided by the presence of rocks with a high uranium content, such as uraninite-bearing granites (BARBIER 1972).
It is however important to note that the existence of red sediments at a given spot does not in itself preclude the possibility of finding uraniferous deposits in the immediate vicinity, as is shown by the Beaverlodge district in Canada and the Morvan district in France.

V. Conclusion

As an alternative to the hypothesis of pitchblende deposit formation by a hydrothermal process, the idea of a genesis by continental weathering has a number of attractive features. As far as is presently known, only geothermometric measurements are not in agreement with it. The rest of the facts — localizing next to uraniferous rock, easy solubilization of the uranium, mineralogy comparable to that of certain roll-type deposits, formation after that of granites and synchronous with important occurrences of continental sedimentation —

all harmonize into a perfectly coherent scheme. Logic goes so far as to give these deposits a true paleotopographical (formation directly beneath heights) and paleoclimatic value (warm and dry climate, scarce vegetation).

A comparison with the geological context of other deposits in Australia, Canada, Eastern Europe and the Soviet Union will no doubt allow for the theory to be proved or disproved. If it should be confirmed, one interesting consequence, from a practical point of view, will be the presence, on a regional scale, of red-bed series lying unconformably on an older basement — a characteristic that *strongly favours the presence of pitchblende vein-type deposits.*

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