

U – ISOTOPIC MODEL AS A PROSPECTING TECHNIQUE IN THE MORRO DO FERRO THORIUM DEPOSIT, BRAZIL

DANIEL MARCOS BONOTTO¹

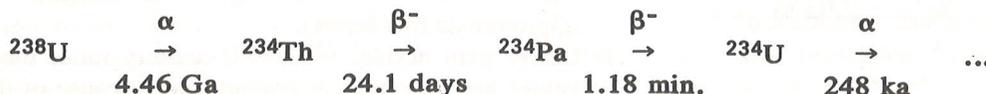
Uranium isotopic analyses were performed for groundwater samples from several boreholes drilled at Morro do Ferro, a thorium and rare earth deposit located on the Poços de Caldas Plateau, Minas Gerais State, Brazil. The data on the isotopic concentrations of dissolved uranium were used to evaluate the applicability of the U-isotopic model developed by Cowart & Osmond (1980) in the area. The $^{234}\text{U}/^{238}\text{U}$ activity ratios for the dissolved U in these groundwaters were between 1 and 2 and the U content varied from 0.01 up to 0.72 ppb. These results show that active uranium dissolution is occurring at Morro do Ferro. The enhanced $^{234}\text{U}/^{238}\text{U}$ activity ratios for the dissolved U are attributed to preferential etching of ^{234}U . The results also suggest that the “normal oxidized” classification of the groundwater samples in terms of their position relative to the U mineralization could be better representative of the studied system instead of the “normal reduced” classification indicated by the model of Cowart & Osmond (1980).

MODELO ISOTÓPICO DO U COMO TÉCNICA DE PROSPECÇÃO NO DEPÓSITO DE TÓRIO DO MORRO DO FERRO, BRASIL – Efetuaram-se análises isotópicas de urânio em amostras de águas subterrâneas de vários furos de sondagem perfurados no Morro do Ferro, um depósito de tório e terras raras localizado no Planato de Caldas, Minas Gerais, Brasil. Utilizaram-se os dados da concentração do urânio dissolvido para avaliar a aplicabilidade na área do modelo isotópico proposto por Cowart & Osmond (1980). As razões de atividade $^{234}\text{U}/^{238}\text{U}$ para o urânio dissolvido nas águas subterrâneas variaram entre 1 e 2 e o teor de urânio dissolvido variou de 0,01 a 0,72 ppb. Esses resultados mostram que ativa dissolução de urânio está ocorrendo no Morro do Ferro. Atribuem-se as enriquecidas razões de atividade $^{234}\text{U}/^{238}\text{U}$ para o urânio dissolvido à lixiviação preferencial de ^{234}U . Os resultados também sugerem que a classificação “normal oxidante” para as águas subterrâneas em termos de sua posição relativamente à mineralização de urânio seria mais apropriada para representar o sistema estudado ao invés da classificação “normal redutora” indicada pelo modelo de Cowart & Osmond (1980).

INTRODUCTION

Uranium-238 is the principal isotope of natural U (99.27% abundance) and is the progenitor of the $(4n + 2)$ series of radioelements. The isotope ^{234}U is radiogenic and the decay chain from ^{238}U through ^{234}U proceeds as follows:

The isotopes ^{238}U and ^{234}U are in secular equilibrium in all minerals and rocks older than one million years. The activity ratio, $^{234}\text{U}/^{238}\text{U}$, is therefore unity in the bulk of a rock matrix. Rock-water interaction, however, frequently results in $^{234}\text{U}/^{238}\text{U}$ activity ratios



¹ Instituto de Geociências e Ciências Exatas – IGCE, Departamento de Física, UNESP, Campus de Rio Claro, Caixa Postal 178, 13500 Rio Claro, SP.

for dissolved uranium which are greater than unity (Cherdyn'tsev, 1969; Osmond & Cowart, 1976). Alternative mechanisms have been suggested for the generation of such enhanced activity ratios. Enhanced chemical solution of ^{234}U due to radiation damage to crystal lattices or to autoxidation from U^{IV} to U^{VI} on decay of the parent ^{238}U has been suggested by Rosholt et al. (1963). This model is supported by the results reported by Chalov & Merkulova (1966). The experiments performed by Kigoshi (1971) and Fleischer & Raabe (1978a, b) suggest that alpha-particle recoil ejection of the ^{234}U precursor, ^{234}Th , into solution (Kigoshi, 1971) may also generate enhanced $^{234}\text{U}/^{238}\text{U}$ activity ratios. Fleischer (1980, 1982) has obtained experimental evidence for chemical dissolution of implantant recoil nuclei (Fleischer, 1975). In this case, the interstitial space was dry during decay and later infiltrated with water that removed the ^{234}U by etching solution.

Etching is a zero-order rate process and the possibility of $^{234}\text{U}/^{238}\text{U}$ activity ratio by the recoil process at different surface etching rates has been discussed by Andrews & Kay (1978). The chemical etching model was involved to explain the enhanced $^{234}\text{U}/^{238}\text{U}$ ratios observed for oxidizing groundwaters in a sandstone aquifer (Andrews & Kay, 1983). The constancy of $^{234}\text{U}/^{238}\text{U}$ ratios, independent of U concentration, in shallow oxidizing groundwaters in the fracture-flow system in granite at Stripa, Sweden (Andrews et al., 1982) was explained by an initial chemical etching process. Higher activity ratios observed in the reducing zone could have been generated by the recoil process.

Based on uranium concentration and on $^{234}\text{U}/^{238}\text{U}$ activity ratio, Cowart & Osmond (1980)

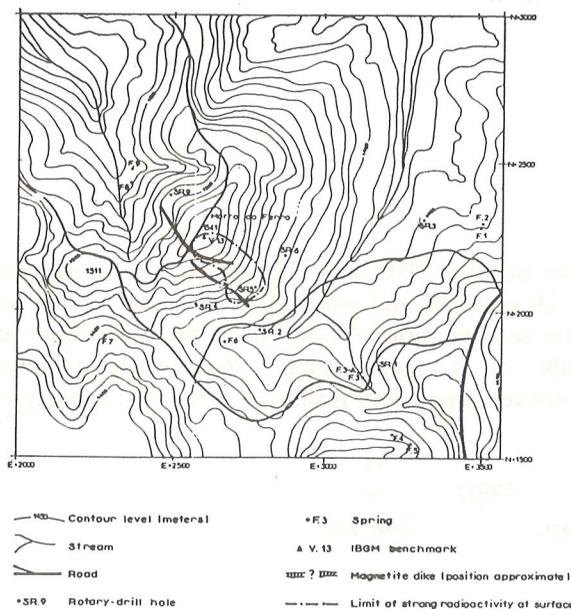


Figure 1. U concentration vs. $^{234}\text{U}/^{238}\text{U}$ activity ratio diagram proposed by Cowart & Osmond (1980).

plotted and classified groundwater samples on a two-dimensional diagram, where several areas of associative significance can be recognized (Fig. 1). In this paper, the data on the isotopic concentrations of dissolved uranium in groundwater samples from Morro do Ferro, Poços de Caldas Plateau, Brazil, were used to evaluate the applicability of this model in the area.

U-SERIES DISEQUILIBRIUM MODEL

According to the model proposed by Cowart & Osmond (1980), the U content and $^{234}\text{U}/^{238}\text{U}$ activity ratio have distinct interpretative meaning and the following isotopic categories were suggested by the authors:

- Average uranium content (1-10 ppb): characteristic of normal oxidized aquifers bathing strata with average uranium mineral content.
- Higher than average uranium content: characteristic of unusually high uranium content in the aquifer strata; the U may be present as primary minerals (in the aquifer grains) or as secondary minerals which have been precipitated within the aquifer.
- Lower than average uranium content: indicative of reducing conditions or strata unusually low in uranium content. Sometimes, a barrier occurs where the circulating groundwater environment changes from oxidizing to reducing conditions and low concentration values occur down-dip from the barrier.
- Normal $^{234}\text{U}/^{238}\text{U}$ activity ratio (1-2): representative of groundwater values world-wide (Osmond & Cowart, 1976).
- Higher than normal $^{234}\text{U}/^{238}\text{U}$ activity ratio: in the case of oxidized groundwaters these ratios are interpreted as to be due to a higher than normal ratio of leachable uranium in aquifer solids (probably from secondary materials) to uranium dissolved in water. They occur especially when the uranium content dissolved in the water is decreasing at the same time that the uranium content of the host strata is increasing (the uranium is precipitating and the condition is named forming accumulation). In the case of reduced groundwaters the probability of high ratios values is enhanced because even normal uranium mineral content of the rock will significantly add to the ^{234}U content of the water by recoil (the diagonal boundary between the normal reduced and the high activity ratio reduced fields of Fig. 1 is designed to approximate this factor).
- Lower than normal $^{234}\text{U}/^{238}\text{U}$ activity ratio: these values are abnormal in groundwater because of the recoil effect and they are interpreted as a temporary (10^5 year time scale) solution of absorbed U, i.e., the remobilization of an accumulation. $^{234}\text{U}/^{238}\text{U}$ activity ratio equals zero denotes ^{234}U below detection.

GENERAL DATA

The Morro do Ferro is located nearly in the center of the Poços de Caldas Plateau at 21°47'S and 46°34'W (Fig. 2). The plateau and its circular rim are underlain chiefly by alcaic igneous rocks; tinguaita is the predominant rock type of the plateau, which is thought to have been emplaced as a large ring dike representing the edge of a vast volcanic caldera (Bjornberg, 1959; Ellert, 1959). Feldspars, feldspathoids and pyroxenes are the main minerals of tinguaita and biotite, apatite, zircon, titanite and pyrite occur as accessory minerals (Barbosa, 1979). In the

microlite and monazite also have been identified (Freeborn, 1980; Fujimori, 1982, 1983).

The regional weather pattern is dominated by wet and dry seasons with more than 80% of the precipitation falling between October through March (Lei, 1984). The regional 30-yr annual average precipitation is 170 cm; during 1983 it was 271 cm (Bonotto, 1986).

Several boreholes were drilled at Morro do Ferro to investigate the character of the subsurface flow. One was drilled in the ore body zone (SR-5), four were drilled around the ore body zone (SR-2, SR-4, SR-6 and SR-9) and two were drilled distant from the

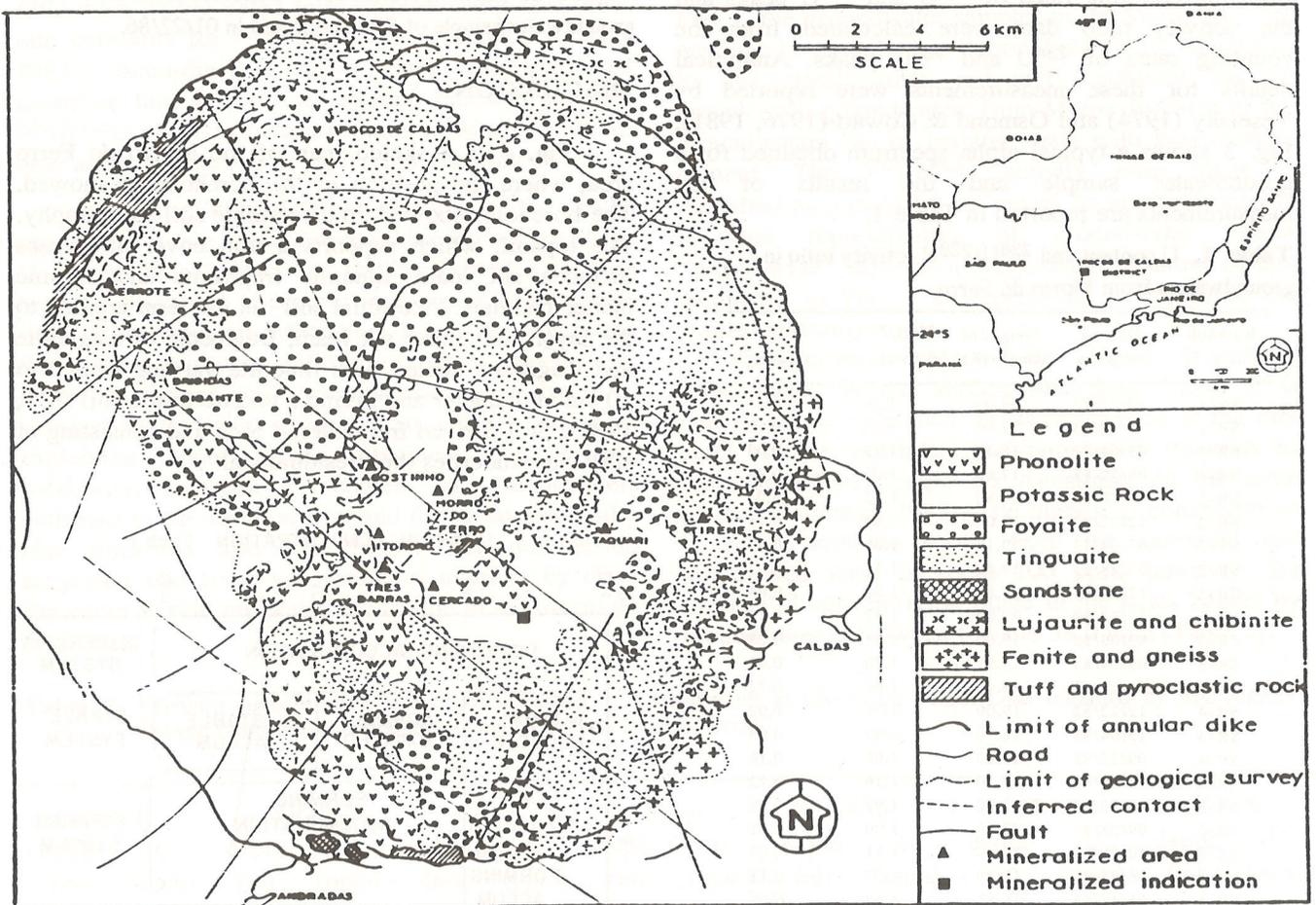


Figure 2. Location and geologic map of the alkaline complex of Poços de Caldas (Oliveira, 1974).

regional context, Morro do Ferro forms part of a mass of tinguaita located at the edge of an area where the rock has been hydrothermally altered.

The Morro do Ferro is about 1 km in diameter, rises to an altitude of 1541 m and a deposit of Th and rare earth elements occurs in the presence of magnetite dikes intercalated in argillaceous layers. Bastnaesite, cerianite and thorogummite were observed as secondary thoriferous minerals and considered as products of weathering of allanite (Wedow, 1967). Cheralite, zircon containing thorium and rare earth elements, rare earth silicate, thorite, thorianite,

ore body zone (SR-1 and SR-3) (IPT, 1982).

EXPERIMENTAL

Groundwater samples (15-20 kg) were collected from the boreholes from December 1982 through January 1986. The samples were acidified to pH less than 2.0, uranium was co-precipitated on $\text{Fe}(\text{OH})_3$, the precipitate was recovered and Fe^{3+} was extracted into an equal volume of isopropyl ether. Uranium was purified by anion exchange, extracted with 0.4 M thenoyl-trifluoroacetone diluted in a benzene solution

and deposited on a stainless steel planchet. The planchet was flamed and placed in a counting chamber under modest vacuum.

The U content and $^{234}\text{U}/^{238}\text{U}$ activity ratio were measured by alpha spectrometry using a 0.3 mm depletion depth, 450 mm² area silicon surface barrier detector. The spectra for natural U and ^{232}U tracer added to each sample were recorded on a multichannel analyser. The Decision Level L_c (Currie, 1968) for acceptance of a positive measurement in the ^{238}U , ^{234}U and ^{232}U energy regions was 0.00082 cpm, 0.00154 cpm and 0.00225 cpm, respectively. The concentration data were calculated by isotope dilution from the counting rates of ^{238}U and ^{232}U peaks and the activity ratio data were calculated from the counting rates of ^{234}U and ^{238}U peaks. Analytical details for these measurements were reported by Veselsky (1974) and Osmond & Cowart (1976, 1981). Fig. 3 shows a typical alpha spectrum obtained for a groundwater sample and the results of the measurements are reported in Table 1.

Table 1. U content and $^{234}\text{U}/^{238}\text{U}$ activity ratio in groundwaters from Morro do Ferro.

Borehole No.	Date of sampling	Weight of sample (Kg)	$^{234}\text{U}/^{238}\text{U}$ * activity ratio	U * ($\mu\text{g kg}^{-1}$)
SR-1	12/22/82	19.00	1.50	0.04
SR-1	03/09/83	19.00	1.29	0.10
SR-1	05/04/83	18.10	1.19	0.03
SR-1	09/27/83	17.60	1.09	0.32
SR-1	12/20/83	16.80	1.62	0.04
SR-2	12/22/82	19.00	1.14	0.18
SR-2	05/04/83	19.10	1.06	0.03
SR-2	09/28/83	19.40	1.15	0.67
SR-2	12/20/83	17.60	1.61	0.05
SR-3	12/22/82	19.00	1.42	0.11
SR-3	03/09/83	19.00	1.47	0.28
SR-3	05/04/83	19.90	1.70	0.03
SR-3	09/27/83	19.40	1.01	0.72
SR-4	12/22/82	19.00	1.75	0.02
SR-4	15/04/83	18.70	0.94	0.03
SR-4	09/27/83	17.60	1.08	0.16
SR-4	12/20/83	17.70	1.16	0.22
SR-5	12/22/82	18.00	1.99	0.08
SR-5	09/28/83	17.60	1.00	0.51
SR-5	12/20/83	17.60	1.11	0.07
SR-5	06/03/85	19.80	1.39	0.11
SR-5	06/17/85	19.85	1.35	0.05
SR-5	07/01/85	19.50	2.03	0.01
SR-5	07/15/85	20.30	1.33	0.05
SR-5	07/29/85	20.70	1.20	0.03
SR-5	08/26/85	20.00	1.28	0.01
SR-5	09/05/85	19.50	1.63	0.14
SR-5	11/05/85	15.20	1.11	0.03
SR-5	01/22/86	19.00	1.10	0.30
SR-5	01/23/86	19.55	1.12	0.16
SR-5	01/28/86	19.50	1.32	0.07
SR-6	12/22/82	18.00	1.77	0.08
SR-6	05/04/83	18.80	1.27	0.01
SR-6	09/28/83	17.60	1.27	0.23
SR-6	12/20/83	17.30	0.98	0.06
SR-9	12/22/82	18.00	1.71	0.04
SR-9	05/04/83	17.70	0.97	0.09
SR-9	09/27/83	17.60	1.20	0.30
SR-9	12/20/83	17.40	1.08	0.07

* Uncertainty $\pm 10\%$ corresponding to 1σ standard deviation.

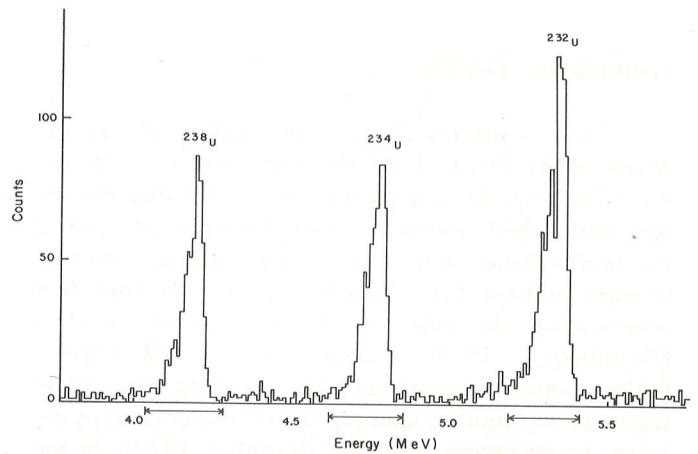


Figure 3. Alpha spectrum for U extracted from the groundwater sample of SR-5 collected in 01/22/86.

DISCUSSION

Fig. 4 is a sketch map of the Morro do Ferro area, where the position of the boreholes is showed. The boreholes showed no discernible soil stratigraphy. The topsoil, which supports a thin cover of grasses includes silt loam, limonite fragments and organic matter residues. "Residual soil" is the term applied to the layer containing silt loam, fragments of magnetite and magnetite coated with limonite varying from 1 to 10 cm in diameter and giving a texture to the soil (IPT, 1982). Soil derived from altered rock and consisting of siltly clay underlies the "residual soil".

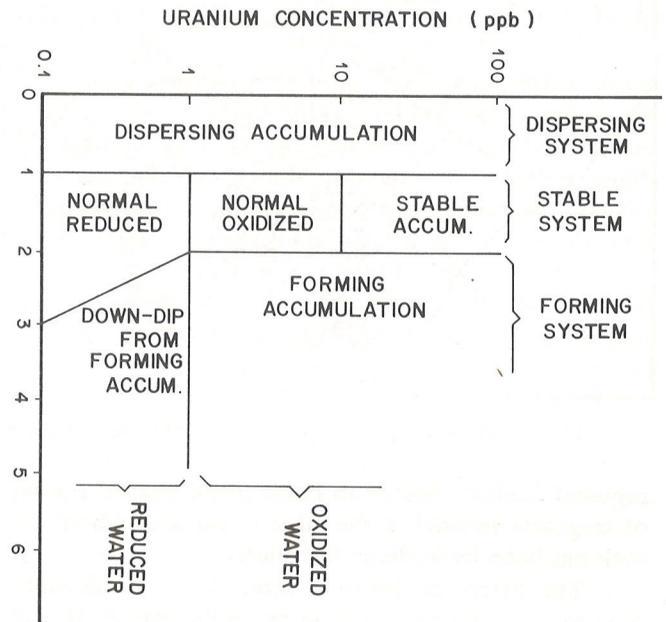


Figure 4. Sketch map of the Morro do Ferro area (IPT, 1982).

Groundwater flows radially out from the center of the mountain at a velocity varying from 1 to 10 cm d^{-1} (IPT, 1982). The maximum path length of groundwater flow is about 400 m (Eisenbud et al., 1979). Lei (1984) computed a removal rate of 10^{-9} yr^{-1} for Th due to groundwater solubilization in Morro do Ferro. An estimate of the residence time for the maximum path length is from 11 to 110 years and that of the etching rate is $0.4 \mu\text{m. yr}^{-1}$.

The results of the $^{234}\text{U}/^{238}\text{U}$ activity ratios reported in Table 1 indicate enhancement of ^{234}U in solution. The effects of alpha-recoil of the intermediate ^{234}Th and differential etching of ^{238}U and ^{234}U were incorporated into a model using zero-order rate constants for ^{238}U and ^{234}U (Andrews & Kay, 1983). According to this model, the evaluated residence time and surface etching rate can generate $^{234}\text{U}/^{238}\text{U}$ enhanced activity ratios in Morro do Ferro only as a consequence of preferential etching of ^{234}U .

Bonotto (1989a) interpreted some of the results reported in Table 1 and discussed the variability of the U content in solution. The results showed a relation between dissolved U content increase and water-table rise due to rainwater infiltration. It was also suggested that during rainwater infiltration, water enriched by humic substances could increase the concentration of uranium to levels in excess of its normal solubility, explaining some values obtained. An inverse correlation between U content of groundwaters collected in the rainy season and the unsaturated silty clay thickness also was found. The relationship suggested that some adsorption of uranium by clays can occur during rainwater infiltration in the Morro do Ferro area.

Other active processes concerning to the U mobilization in the Morro do Ferro area were discussed by Bonotto (1989b). The results of $^{230}\text{Th}/^{234}\text{U}$ and $^{228}\text{Th}/^{232}\text{Th}$ activity ratios measured for materials from the saturated zone of the borehole SR-5 favoured the possibility of occurrence of active cation-exchange between radium and uranium and some adsorption of soluble uranium by Fe and Mn oxides.

If the measured $^{234}\text{U}/^{238}\text{U}$ activity ratio range (0.94–2.03) and U content range (0.01–0.72 ppb) are plotted on the two-dimensional diagram represented in Fig. 1, the groundwater samples in the Morro do Ferro area can be classified as “normal reduced”. According to this classification, reducing conditions can be prevalent in groundwaters from Morro do Ferro or the groundwaters can be bathing strata with low uranium content. The $^{234}\text{U}/^{238}\text{U}$ activity ratio range is classified as normal by the model because it is similar to that representative of groundwater values “world-wide” and the interpretative meaning of this parameter in Fig. 1 is appropriate to represent the studied system.

Uranium in Morro do Ferro is finely disseminated at the surface and deep zones. It is considered as a product of decomposition of the rare earth silicate, coffinite, and radioactive minerals as those found at Campo do Agostinho on the same plateau (Fujimori, 1983). The highest U concentration in some borehole spoils from the saturated and unsaturated zone is about 100 ppm, however, the average U concentration range in the strata bathed by groundwater is between 6.8 and 58.0 ppm (Table 2).

Table 2. Uranium concentration in the strata bathed by groundwater at Morro do Ferro. Data obtained by Ms. Márcia Carlos, Pontifícia Universidade Católica, Rio de Janeiro, Brasil.

SR-1		SR-2		SR-3		SR-4		SR-5		SR-6		SR-9	
depth (m)	U (ppm)												
16.55	15.0	4.85	75.0	14.40	15.0	31.76	44.0	10.62	42.0	9.85	40.0	28.32	99.0
17.55	15.0	5.90	66.0	15.40	1.0	33.83	53.0	11.68	57.0	11.50	40.0	30.15	46.0
19.60	7.0	6.95	54.0	16.40	10.0	35.96	33.0	12.88	56.0	13.00	50.0	31.82	44.0
20.58	30.0	8.25	38.0	17.40	1.0	36.88	28.0	14.15	15.0	14.50	31.0	33.90	53.0
21.55	14.0	9.50	42.0	mean	6.8	mean	39.5	15.46	13.0	16.00	37.0	36.00	48.0
23.52	7.0	10.50	37.0					16.88	57.0	17.00	36.0	mean	58.0
24.55	9.0	11.50	20.0					18.75	18.0	18.00	39.0		
25.60	4.0	12.50	46.0					21.15	20.0	19.25	43.0		
26.62	13.0	mean	47.2					24.70	104.0	20.75	49.0		
27.65	8.0							26.60	43.0	mean	40.6		
mean	12.2							mean	42.5				

U contents measured by X-ray fluorescence. Uncertainty $\pm 5\%$ corresponding to 1σ standard deviation.

Uraniferous deposits can be broadly defined as the uranium occurrences that exceed about 50 times the average concentration in crustal rocks, i.e., 100 ppm of U_3O_8 (Crawley, 1983). The general pattern of U distribution in the area doesn't characterize its occurrence like a typical uraniferous deposit. However, the U contents in the saturated zone of the aquifer in the Morro do Ferro area are higher than the average U abundance in the upper crust (between 1.67 and 2.68 ppm according to Gabelman, 1977). These evidences suggest that the interpretative meaning of the U concentration in the fence diagram in Fig. 1 is not appropriate to represent the studied system, so that the "normal reduced" classification is not convenient.

Bonotto (1986) reported a value of 10.2 for the pH of the groundwater of SR-9, which is due to the high content of dissolved Ca^{2+} (8 mg l^{-1}). The high Ca is probably related to the dissolution of fluorite, a mineral that has been identified in deep zones of Morro do Ferro. The reported values of pH were 5.4-6.5 for the groundwaters of the other boreholes and the dissolved Ca^{2+} content ranged from 0.01 to 0.62 mg l^{-1} .

The Eh range was from +500 to +700 mV for the groundwater of SR-2, SR-3, SR-4, SR-5 and Sr-6 but was from +300 to +500 mV for the groundwater of SR-1 and SR-9 (Lei, 1984). Thus, the Eh-pH field indicates that the groundwater of Morro do Ferro are oxidizing in character and these evidences also suggest that the "normal reduced" classification is not appropriate to represent the studied system.

According to the terminology proposed by Cowart & Osmond (1980), groundwater samples in the Morro do Ferro area could be better classified as "normal oxidized" in terms of their position relative to the uranium mineralization. However, even in this case it is necessary to remember that the U contents in the strata of the aquifer are higher than the average U content in crustal rocks and that the U occurrence doesn't characterize a typical uraniferous deposit.

CONCLUSION

Active uranium dissolution is occurring at Morro do Ferro, Brazil. Enhanced $^{234}U/^{238}U$ activity ratios for the dissolved U are attributed to preferential etching of ^{234}U . The U-isotopic model proposed by Cowart & Osmond (1980) suggest that the "normal reduced" classification could be applied to the groundwater samples from Morro do Ferro area in terms of their position relative to uranium mineralization. However, additional evidences suggest that the "normal oxidized" classification could be better representative of the studied system. Therefore, although the classification scheme has a reasonable theoretical foundation, it is not considered to be completely applicable to the Morro do Ferro aquifer/

groundwater system. Because of the great complexity of natural systems, it is convenient to use the model with caution on uranium exploration programs.

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