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STATENS KÄRNKRAFTINSPEKTION
Swedish Nuclear Power Inspectorate

Datum - Date 1988-04-08
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Vår ref - Our ref INTRAVAL (88) 4
Er ref

INTRAVAL Project Secretariat

To:

INTRAVAL participants,
participants at the 1st
INTRAVAL workshop

Barcelona workshop

Please find the following enclosed Appendices as working documents for the Barcelona workshop:

Appendix 1: Tentative agenda (a detailed agenda will be provided at the start of the meeting)

Appendix 2: Test Case 7(a), Pocos de Caldas project. Test Case description, Redox-front and radionuclide movement in an open pit uranium mine.

Appendix 3: Test Case 7(b), Pocos de Caldas. Test Case description, Morro do Ferro migration studies (colloids).

Appendix 4: Test Case 8, Alligator Rivers Analogue Project. General background description.

Appendix 5: Test Case 8, Alligator Rivers Analogue Project. Test Case description, Radionuclide migration in the weathered zone of the Koongarra Uranium deposit.

Appendix 6: Proposed Charter for the Validation Oversight and Integration Commitee (VOIC).

The test case description for Case 4 (Stripa 3D migration experiment) will be distributed shortly.

With best regards,

Kjell Andersson

Encl: Filed in jacket

Kjell Andersson
Secretary to the Co-ordinating Group

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INTRAVAL workshop No. 1 , Barcelona, Spain
April 25-29, 1988

Brief meeting agenda

<u>April 25</u>

9.30- 13.00
15.00-16.30
17.00-19.00

- Introduction
- Cases 1a, 1b, 2, 9, 5

<u>April 26</u>

9.00-13.00

- Cases 3 and 6

15.00 Barcelona bus tour, dinner

<u>April 27</u>

9.00-13.00
15.00-20.00

- Cases 4, 7 and 8
- salt cases
- unsaturated media cases

<u>April 28</u>

10.00-20.00 Tour to Tarragona (or Montserral) with lunch (ENRESA)

<u>April 29</u>

9.00-13.00

- synthetic experiment
- VOIC Committee

4/2

INTRAVAL
Test Case 7(a)

Redox-front and radionuclide movement in an open pit uranium mine.

Draft Preliminary* Test Case Definition

****N.B. Not all the detailed data available are presented in this note, which is intended mainly to allow the INTRAVAL group to assess the feasibility of this work as a test case.***

I. INTRODUCTION

A. Pilot Group Identification

The pilot group will consist of the Poços de Caldas project Technical Committee (N.A. Chapman, BGS; I.G. McKinley, Nagra; J.A.T. Smellie, SGAB; M. Shea, Battelle; E. Penna Franca, Federal University, Rio de Janeiro), plus Professor Ivars Neretnieks of the Royal Institute of Technology, Stockholm.

B. Experiment location

Osamu Utsumi Mine, near Poços de Caldas (Minas Gerais), Brazil.

C. Objectives

To model the past movement of the redox front as it progressed into the rock and to model the associated migration of uranium and other radionuclides.

D. Theories tested

a That intruding oxygenated water will be depleted of oxygen as it encounters reducing minerals in the rock such as ferrous iron (in this case in pyrite).

b That uranium is dissolved in the portion of the rock which has become oxidized and that it will reprecipitate when it migrates into the reducing rock.

c That the associated geochemical reactions can be accounted for, eg the formation of clays.

E. Validation aspects

a Validation of the mechanisms which ensure that the groundwater will be reducing and thus ensure that many of the actinides will have low solubilities and high sorption coefficients.

b Validate the movement of the redox front around a waste-form which is causing radiolysis.

c Validate the precipitation of uranium (and other actinides) at the redox front.

F. Background Information

At Poços de Caldas in the state of Minas Gerais in Brazil the Nuclebras company has operated an open pit Uranium mine since 1975. The bedrock is crystalline and consists mainly of phonolites and nepheline syenites. The deeper portions of the rock contain about 2% by weight of pyrite (FeS_2) and are strongly reducing. The upper portions of the rock have become oxidized by oxygen carried by infiltrating rainwater. Pyrite has become oxidised to ferric oxy-hydroxides of varying degrees of crystallinity. There is a very sharp redox front delineating the upper oxidised rock from the deeper lying reduced rock. The boundary is not horizontal. There are many "fingers" of oxidised rock extending much further downward than the average depth of the front. The fingering is often associated with fractures and fracture zones which have higher permeability than the rock matrix itself. The fractures are sloping at various angles so that the front and fingers are often not moving vertically but at some angle to the vertical. The redox front is rather ragged when observed at a scale of 10-100m.

The rock matrix is porous with a porosity of 4-20%. The hydraulic conductivity of the matrix is an order of magnitude lower than that of the overall rock including the fractures.

Pitchblende nodules with typical sizes between 0.5-l cm and nearly spherical in shape, are found in many places just below the redox front in the reduced rock. It is thought that the uranium which is present as U(IV) oxides in the reduced rock has been oxidised to U(VI) which is much more soluble and has been dissolved out of the now oxidised rock and moved with the flowing water or by diffusion into the reducing sections of the rock where it has been reduced to U(IV) by reaction with the pyrite. The 're-reduced' uranium has so low a solubility that it must precipitate.

II. EXPERIMENTAL DESIGN

Not applicable

III. CURRENT STATUS AND EXPERIMENTAL SCHEDULE

A very large amount of data has been collected during the first 18 months of the project. However, systematic collection of the data needed for this study is at an early stage, and although there is sufficient information to make a start, a complete dataset will not be available until early 1989. The International Poços de Caldas project will continue up to the middle of 1989.

IV. EXPERIMENTAL RESULTS

The open pit mine is at present several hundreds of meters wide, nearly 1km long and more than a hundred meters deep in places. The walls of the mine are near vertical and terraced. The floor of the mine is flat. The Nuclebras company has a very detailed set of maps for every 2m depth and also for every 2 to 4m section of the vertical walls. These maps show where the redox front is in great detail, and the concentration profiles of the uranium.

In addition to the Nuclebras data, the international Poços de Caldas project which started in 1986 and which will finish in 1989 has gathered substantial information on rock chemistry, mineralogy, petrography, physical properties, uranium series disequilibration as well as the chemistry of waters at different depths in the boreholes, which extend to about 100m below the present mine bottom. Hydraulic conductivities have been measured in deep boreholes and the rock matrix. Porosities and diffusivities in the rock matrix have also been determined. The compositions of many groundwater samples have been determined and some geochemical modelling has been performed. The content of colloids and their composition has been studied. The microbiological activity in the mine has also been studied.

Hydraulic modelling of groundwater flow patterns and velocities in the area of the mine both prior to excavation as well as with the mine in its present state has been performed by David Noy of BGS, and detailed results are available.

V. PREVIOUS MODELLING

A Hydraulic modelling

This has been performed to describe the flow field prior to excavation and after the excavation.

B Geochemical modelling

Some geochemical modelling has been performed to determine if the waters are in equilibrium with minerals observed.

C Transport modelling

Some modelling has been performed on the movement of the redox front both by diffusion and by flow. Some first attempts at modelling the uranium movement have also been made.

VI. EXPECTATIONS

A Experimentalists view

There is a wealth of data from most areas of interest for the assessment of both redox front movement and uranium and other species movement. There seems to be a good opportunity to obtain a coherent dataset for one and the same location of "all" data needed to understand the migration processes.

B Modellers view

The first attempts at modelling the redox front movement and the movement of the uranium seem encouraging. Although there seems to be a strong need for development to the models to account for channelling and the need to use coupled transport and geochemical codes which are not readily available, even simple codes seem to be able to describe several of the most important processes going on. There is room for use of codes of different complexity and a challenge to develop and adapt codes to describe the observations in ever increasing detail.

The most important objective is, however, to devise conceptual models which are consistent with the main observations and which account for the main reactions: the redox front movement, the migration of uranium, the geochemistry of the processes both qualitatively and (semi)quantitatively.

VII. INFORMATION EXCHANGE

Requests for information should be addressed in the first instance to either Ivars Neretnieks, Royal Institute of Technology, 100 44 Stockholm 70, Sweden (Tel. +46-8-790-8229; Telex 10389 KTHB Stockholm) or Neil Chapman, British Geological Survey, Keyworth, Nottingham, NG12 5GG, UK. (Tel. +44-6077-6111; Telex 378173 BGSKEY G; Telefax +44-6077-4841).

VII. FUTURE POSSIBILITIES

The international project will continue until mid-1989. Up to this time it is possible to obtain additional data, but this will only be considered in the broader context of the project objectives. After this time only such additional data as can be extracted from the samples which have already been gathered will be available, and this would probably have to be regarded as outside the project framework. There is, however, a substantial collection of cores and hand samples available, and decisions on future activities will be taken towards the end of 1988.

IX. OUTPUT FORMAT

It is envisaged that if this test case is adopted, then the raw geochemical and analytical data ONLY will be supplied to the Project Secretariat by the Pilot Group. This will be in the form of 3-monthly progress reports based on the relevant sections of the Poços de Caldas Project internal reports. Such advice as the INTRAVAL investigators require on the interpretation of these results will be provided as necessary by the Pilot Group.

X. REFERENCES

Barretto. P.C.M., and Fujimori, K. 1986. Natural Analogue Studies: Geology and Mineralogy of Morro Do Ferro, Brazil. *Chem. Geol.,* 55, 297-312.

Eisenbud, M., Krauskopf, K., Penna Franca, E., Lei, W., Ballad, R., Linsalata, P., and Fujimori, K. 1984. Natural analogues for the transuranic actinide elements: an investigation in Minas Gerais, Brazil. *Environ. Geol. Water Sci.,* 6, 1-9.

IPT, 1984. Geology, hydrology, geochemistry and radioactivity of Morro do Ferro: a critical review of the literature. Unpublished report of Instituto de Pesquisas Tecnologicas do Estado de Sao Paulo.

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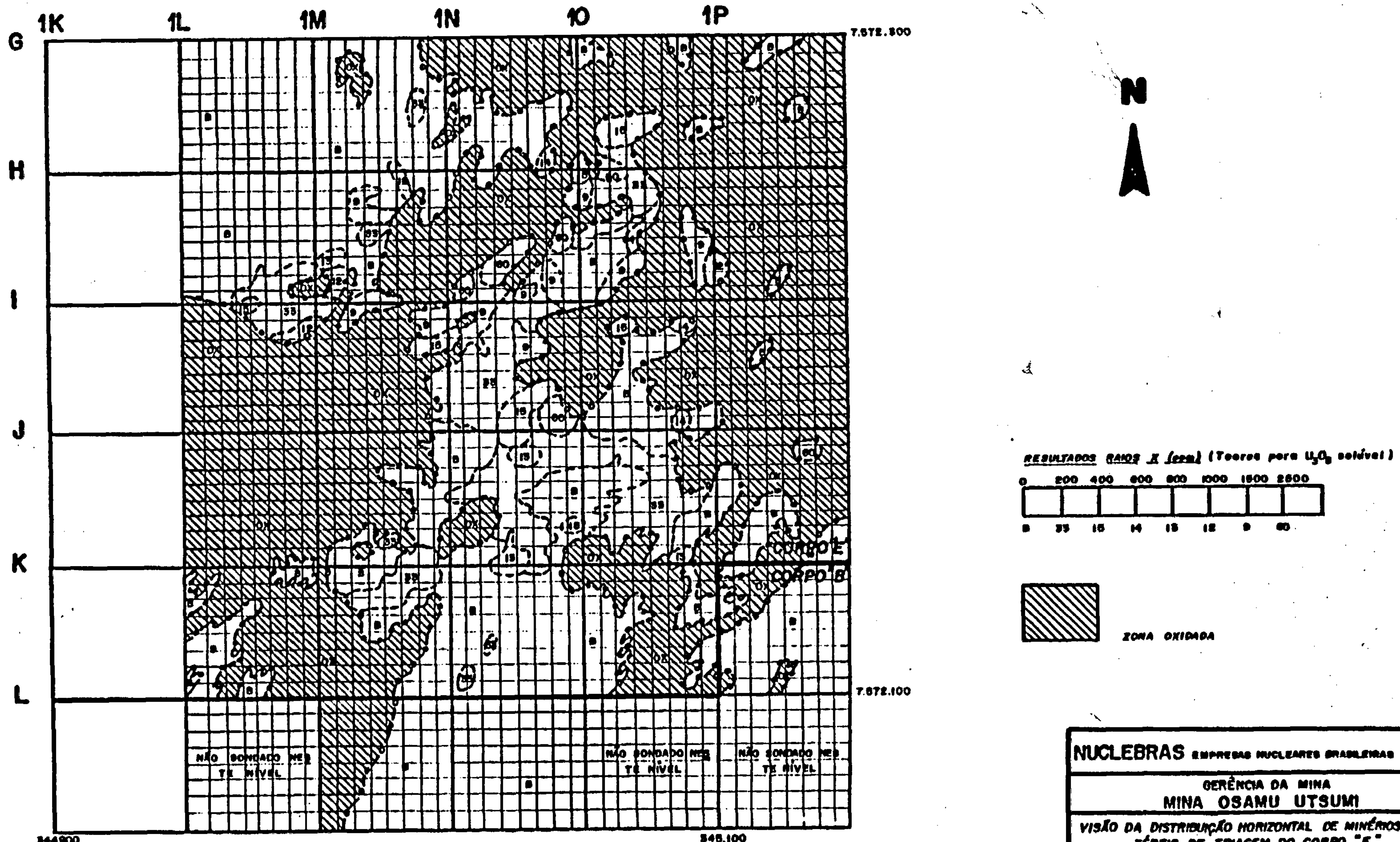
Miekeley, N. and Küchler, I.L. 1988 in press. Interactions between Th and humic compounds in surface waters. Radiochim. Acta.

Ming Zhu Dep. Chem. Eng. KTH. Thesis on the redox front movement.

Smellie, J.A.T., Barroso Magno, L., Chapman, N.A., McKinley, I.G., and Penna Franca, E. 1987. The Poços de Caldas project feasibility study: 1986-1987. In: 'Natural Analogues in Waste Disposal', Come, B.C. and Chapman, N.A. (eds), Graham and Trotman for CEC, 104-115.



NUCLEBRAS EMPRESAS NUCLEARES BRASILEIRAS S.A.

GERÊNCIA DA MINA

MINA OSAMU UTSUMI

VISÃO DA DISTRIBUIÇÃO HORIZONTAL DE MINÉRIOS E ESTÉREIS DE TRIAGEM DO CORPO "E"

FOLHA 01 - NÍVEL 1340 (BASE DO BANCO)

FIG.:08

ESCALA	GEOL. RESP	DES.: V.L. CAMARGO
1:500	HELIO A. SCALVI	DATA: 13/05/82

INTRAVAL
Test Case 7(b)

Morro do Ferro migration studies, Poços de Caldas Project, Brazil.

Draft Preliminary* Test Case Definition

N.B. Not all the detailed data available are presented in this note, which is intended mainly to allow the INTRAVAL group to assess the feasibility of this work as a test case.

I. INTRODUCTION

A. Pilot Group Identification

The pilot group will consist of the Poços de Caldas project Technical Committee (N.A. Chapman, BGS; I.G. McKinley, Nagra; J.A.T. Smellie, SGAB; M. Shea, Battelle; E. Penna Franca, Federal University, Rio de Janeiro).

B. Experiment location

Morro do Ferro, near Poços de Caldas (Minas Gerais), Brazil.

C. Objectives

To describe the geochemistry and physical properties of colloids and their role in transporting Th and Rare Earth elements (REE) in groundwaters. This will involve studying the nature of the colloids, their stability and reactions with the host rock, and their association with Th/REE. This geochemical information, together with systematic data on the groundwater chemistry, will be superimposed on a model of the groundwater flow field in order to assess colloid transport mechanisms.

D. Theories tested

It is not clear that any sound theoretical basis exists to allow description of colloid behaviour and transport in groundwaters. This project is one of the few which are attempting to gather baseline data in the field. Consequently the success of this as a test case within INTRAVAL will depend on testable models being developed during the next two years.

E. Validation aspects

The basis of this analogue study is that groundwaters with a high humic acid content are seeping down through an exceptionally rich Th/REE deposit situated near the surface in the unsaturated zone, are complexing with these elements, and migrating down to the water table, then being transported down hydraulic gradient to re-emerge in a stream some 100m or so down slope. Collection points exist above the water table, and in boreholes at three points along the flow path below the water table.

The chemistry of the groundwaters in four reference zones (two in packer isolated sections of boreholes) is being monitored to establish a basic dataset and any seasonal variability; colloidal material is being extracted from these reference waters and characterised for composition, size and Th/REE content; simple hydraulic testing is being performed to establish the properties of the rock (a mixed but generally porous medium), and a simple conservative tracer test is being considered to calibrate an existing 2-D flow model for the system.

If generic models become available to describe colloid transport in a porous medium, then the results of this study should be useful in attempting validation.

F. Background Information

See Figure 1 for a schematic cross-section of the site.

Geology: The basic geology of Morro do Ferro (e.g. Barretto and Fujimori, 1986) is a fractured volcanic (carbonatite/phonolite) complex containing a massive magnetite dyke system with surrounding magnetite-rich breccia. The whole system has been extensively altered both by hydrothermal activity and weathering processes. The upper tens of metres are a lateritic soil composed of kaolinite, illite, gibbsite, and accessory minerals including jarosite and magnetite. The rock is extensively oxidised, but in borehole MF12 a redox front occurs at 35m, with pyrite occuring in the un-oxidised phonolite below it.

Hydrogeology: Previous hydrological investigations at Morro do Ferro (IPT, 1984; Lei, 1984) which involved the drilling of 9 piezometers to sample the upper part of the saturated zone, showed that the water table is a subdued reflection of the topography. At the top of the hill the water table is at least 80m below the surface, fluctuating by at least 20m between the wet and dry seasons. This represents the water which is stored and slowly released as base flow to the stream. In the valley bottoms the water table is at or near the surface, coinciding with seepages or discrete springs. The groundwater flow is downwards, maintained by a vertical component in the piezometric gradient.

Owing to the necessity to case borehole MF10 down to 62m double packer testing techniques were not feasible. However, a slug test performed when the hole had penetrated the water table gave a bulk hydraulic conductivity of 1.6×10^{-5} ms^{-1}, and a second test performed using a single packer to isolate the lower section of the borehole from 64-74m gave a hydraulic conductivity of $1\times10^{-6} ms^{-1}$. This section of the borehole now forms the reference zone for groundwater sampling. Both of these relatively high values indicate an adequate groundwater supply for the considerable volumes of water required for the hydrochemical characterisation and colloid extraction programmes.

Borehole MF12 was tested using a straddle packer system. Four intervals of c.6m spacing were tested from the end of the casing to the bottom of the hole. From 40-58m the rock has a hydraulic conductivity of $3\text{-}6 \times 10^{-7} ms^{-1}$. Below 58m this falls to $7 \times 10^{-9} ms^{-1}$. Groundwater heads indicate a small upward flow, as would be expected near the valley bottom.

The information obtained has been used to construct a simple 2-D flow model, described in section V.

Geochemistry: An outline mineralogy was mentioned above, but full core analyses of MF12 will be available within the next few months. The main information of interest concerns the hydrochemistry and colloid chemistry.

(a) Hydrochemistry: the reference zones are now being incorporated into a routine analytical programme which is due to be completed in February 1989. Four major samples from each zone, representative of the seasonal variations, will be extensively studied. Throughout this period each zone will be monitored weekly using a sealed flow-through cell containing electrochemical probes for basic properties (Eh, pH, T, cond, etc), and samples monitored for major element chemical variation. The major samples will be used as the source of colloidal material, extracted by ultrafiltration. Preliminary analyses of the waters from MF 10 and 12 indicate them to be mildly reducing with pH around 6, 0.3-0.6μg/l U, 9-20mg/l HCO3, 7-12mg/l SO4, 1.2-1.6mg/l Fe (almost all as Fe II).

(b) Colloid chemistry: high concentrations of Th and REE can be observed in organic-rich surface and unsaturated zone waters. Humic compounds, mainly humic acids seem to be the predominant complexants in these waters; Th, REE's, Fe and Mn are present mostly as colloids (>1000 MWU fraction). Interaction of these species with the rocks are responsible for low concentrations of Th/REE and DOC in the groundwaters. Detailed information on the association of these elements with different size fractions of particulate material are available (e.g. Miekeley and Küchler, in press).

Geophysics: no geophysical data are available.

II. EXPERIMENTAL DESIGN

The natural analogue data collected will be in the form of geochemical observations of systematic variations in groundwater and colloid properties over a one-year period. The only experimental aspect is the possibility of an in-situ tracer test between closely spaced (c. 20m separation) boreholes (e.g. MF10 and a new hole down slope in a dipole configuration, with recharge to MF10 and constant pumping from the down gradient hole) using a NaCl spike. This would not be carried out until the middle of 1989 and, if done, would be intended to calibrate the groundwater flow model and provide data on the dispersivity of the rock. The decision on whether to proceed will depend on the nature of the chemical database, and on the availability of colloid transport models to interpret the observations.

A. Parameters measured: Complete characterisation of groundwater hydrochemistry for each reference zone in the boreholes and in the gallery (fig 1); major and trace element content, natural series content, REE's, organic chemistry, electrochemistry, etc. Size distribution of colloidal material in each zone, chemical and physical property analysis, and Th/REE contents.

B. Spatial and temporal scales: see Figure 1 for scale. Timescale of chemical observations is 1 year. Tracer test would probably take about 100+ days.

C. Experimental set-up: not applicable at present.

D. Sampling strategy: see Geochemistry (a) above.

E, F, G. No detailed comments at this stage. No complementary experiments are known of at this time, although colloid data from natural systems are being generated in other analogue programmes (e.g. Alligator Rivers).

III. CURRENT STATUS AND EXPERIMENTAL SCHEDULE

A large amount of background information on the environment and the groundwater conditions at Morro do Ferro is available as a result of the first 18 months of this project, and the earlier studies by Eisenbud et al (1984). However the systematic collection of data with a view to assessing colloid behaviour is only just starting. The first seasonal groundwater sampling will take place in May 1988 and will run until February 1989. The potential tracer test would take place between April and June 1989. A complete dataset will not be available until around October 1989.

IV. EXPERIMENTAL RESULTS

Not applicable at present?

V. PREVIOUS MODELLING

Some simple empirical modelling of Th transport was presented by Eisenbud et al (1984), using estimates of the total quantity of Th available for mobilisation, and data on the Th content of stream waters from the catchment.

Subsequently David Noy of BGS has carried out a 2-D flow model of the profile shown in Fig.1. In the absence of a horizontal scale to this diagram a vertical exageration of 2x was assumed. This gave a profile 370m. long from groundwater divide to stream. The section has been assumed to be homogeneous except for a region of enhanced permeability to a depth of ~30m. close to the stream. This latter feature is suggested by the description of borehole MF12. The permeability of the bulk of the section has been taken to be 5×10^{-7}m/s with a porosity of 0.1, whilst that of the friable weathered zone close to the stream has been taken to be 5×10^{-6}m/s with a porosity of 0.3. The permeability of the bulk of the section is suggested by the hydraulic tests in MF12, but the other figures have been chosen without the guidance of specific field data.

Two separate calculations were performed : the first a steady state calculation with the water table fixed at the dry season level; the second a steady state calculation with the water table fixed at a supposed wet season level. This latter was obtained by raising the level of the water divide some 15m. to the bottom of the mine gallery, and raising the rest of the water table in proportion to its original height above the stream. For each calculation a series of pathlines were traced (Fig. 2) which allow an assessment to be made of the flow rates that can be expected in various parts of the section. In particular, flow rates in the vicinity of the bottom of borehole MF10 were found to be :-

Dry season : 1.16×10^{-7} $m^3/m^2/s$.
Wet season : 1.29×10^{-7} $m^3/m^2/s$.

The pore water velocities, for mass transport calculations, are obtained from these by dividing by the porosity. Using the porosities indicated above, the travel times between MF10 and MF12 were :

Dry season : 1076 days.
Wet season : 949 days.

The effects on the pathlines of the high K zone near the stream can be clearly seen in the pathline plots, but it will also be seen that there is very little difference between the wet and dry season results. This is also reflected in the flow rates given above.

The calculations reported here would seem to suggest that there might be a variation of no more than ~10% in the flow rates at the bottom of MF10 as a result of seasonal changes in the position of the water table. Future work will be directed towards understanding the variation of permeability on the section, since this can be expected to have a greater effect upon the calculated results, and towards observations of the actual variation of the water table, which would allow a more realistic pair of bounding calculations to be performed.

VI. EXPECTATIONS

At present there is no accepted and validated model available for the generation, behaviour and transport of radionuclides in colloid-rich groundwaters. Success with this test case would be critically dependent on there being some appropriate modelling tools developed within the next 18 months. While this test case is by no means a well constrained experiment to which a number of transport codes can be applied with a view to validation, the INTRAVAL group may nonetheless consider it worth attempting.

VII. INFORMATION EXCHANGE

Requests for information should be addressed in the first instance to Neil Chapman, British Geological Survey, Keyworth, Nottingham, NG12 5GG, UK. Tel. +44-6077-6111; Telex 378173 BGSKEY G; Telefax +44-6077-4841.

VII. FUTURE POSSIBILITIES

Very limited; the REE deposit may be mined within the next few years, and funding for additional research work is not assured beyond the end of 1989.

IX. OUTPUT FORMAT

It is envisaged that if this test case is adopted, then the raw geochemical and analytical data ONLY will be supplied to the Project Secretariat by the Pilot Group. This will be in the form of 3-monthly progress reports based on the relevant sections of the Poços de Caldas Project internal reports. Such advice as the INTRAVAL investigators require on the interpretation of these results will be provided as necessary by the Pilot Group, but it is not intended that the Group participates actively in the modelling, other than as advisors.

X. REFERENCES

Barretto. P.C.M., and Fujimori, K. 1986. Natural Analogue Studies: Geology and Mineralogy of Morro Do Ferro, Brazil. *Chem. Geol.,* 55, 297-312.

Eisenbud, M., Krauskopf, K., Penna Franca, E., Lei, W., Ballad, R., Linsalata, P., and Fujimori, K. 1984. Natural analogues for the transuranic actinide elements: an investigation in Minas Gerais, Brazil. *Environ. Geol. Water Sci.,* 6, 1-9.

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Lei, W. 1984. Thorium mobilisation in a terrestrial environment. Unpub. PhD thesis, New York University, New York, 414pps.

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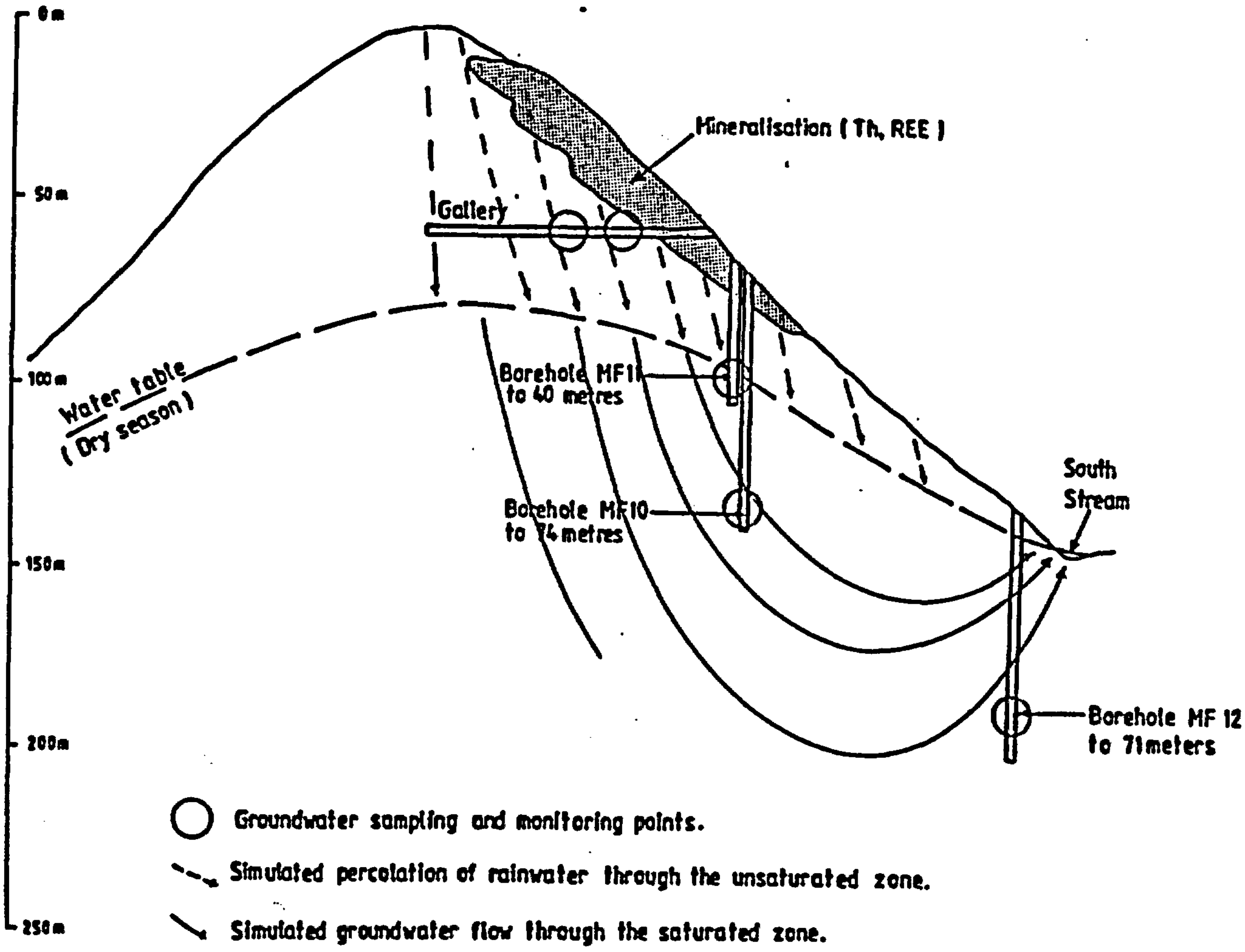
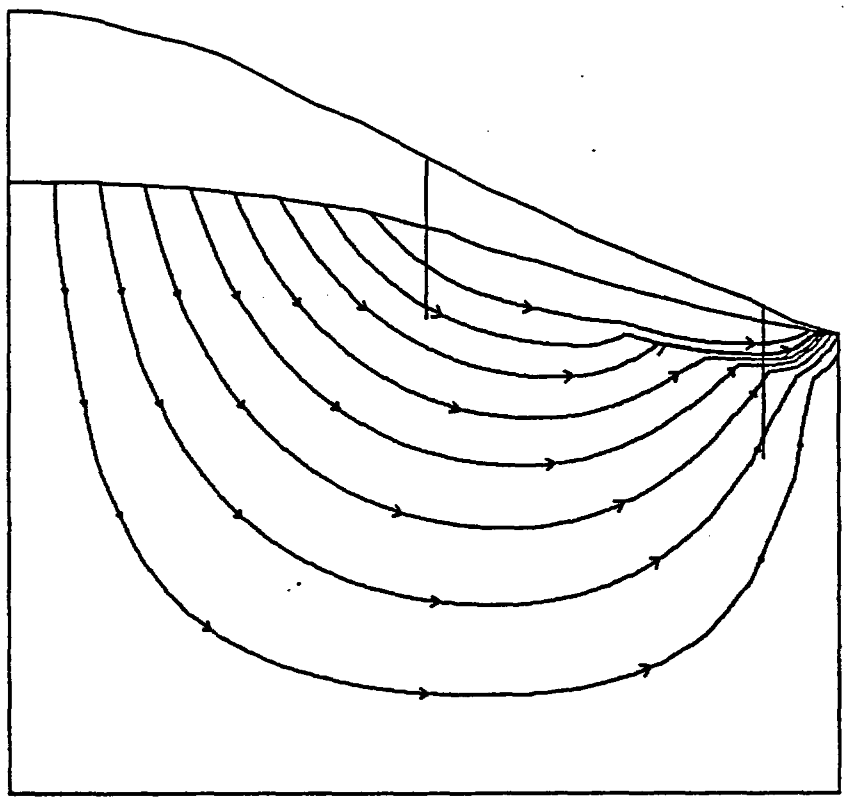

0m
50m
100m
150m
200m
250m
Mineralisation (Th, REE)
Gallery
Water table
(Dry season)
Borehole MF11
to 40 metres
Borehole MF10
to 74 metres
South
Stream
Borehole MF 12
to 71meters
Groundwater sampling and monitoring points.
Simulated percolation of rainwater through the unsaturated zone.
Simulated groundwater flow through the saturated zone.

FIGURE 1

Figure. 2

MORRO DO FERRO DRY SEASON : PATHS



MORRO DO FERRO WET SEASON : PATHS